The European Spallation Source Project

The ESS Project

Volume II

New Science and Technology for the 21st Century

The European Spallation Source Project

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The ESS Project

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New Science and Technology for the 21st Century

Foreword

"The scientific case for the European Spallation Source (ESS) has been rigorously investigated, debated and prepared by the ESS collaboration as well as the European Neutron Scattering Association (ENSA). It has been continuously updated to take into account new developments and opportunities across a broad range of science and research in physics, chemistry, materials, engineering, biology and the earth and life sciences. The European Science Foundation (ESF) reviewed the basic scientific case of research neutron sources, and of a new MW-class pulsed source for European science and research, a few years ago and found it sound. Very considerable detailed technical and engineering effort has been invested in the project design studies, and many alternatives have been explored. It is noteworthy that the European neutron user community and European neutron providers have all worked together in preparing the ESS case and support it.

That Europe has enjoyed, and still enjoys, the world lead in the establishment and research use of neutron facilities is unarguable. This lead is centred on the so-called "2nd generation facilities" ILL and ISIS, including their cutting-edge instrumentation. The USA and Japan have recognised the European lead, and the benefits that neutron research brings, and are constructing (USA: SNS) or have approved (Japan: JSNS) 3rd generation facilities which, unless there is a European competitor, will inevitably result in the loss of this lead and in a major loss of talent to these facilities. The Organisation for Economic Co-operation and Development (OECD) Megascience Forum forward look in the late 1990's forecast a severe "neutron-drought" beginning before 2010, caused by the planned closure of facilities around the world, many in Europe, as they became obsolescent. This drought will severely curtail science research using neutrons. The new national facilities in Europe, FRM-2 (Munich) and the planned AUSTRON, and potential upgrades at ILL and ISIS, might diminish the drought. But they cannot provide the "cutting edge" opportunities for Europe available at a 3rd generation MW-class facility such as ESS, with its unique long-pulse and short-pulse target stations at an unprecedented 5 MW beam power level.

The OECD's study identified the need for three 3rd generation facilities - in North America, in the western pacific, and in Europe. The American SNS is under construction and the

Japanese JSNS is approved. The SNS acknowledges that its scientific and technical case borrowed heavily from the earlier work on the ESS project. The ESS scientific and engineering cases have moved on from this starting point and are now benefiting from shared experiences with SNS and JSNS.

The ESF recognises that national funding agencies in Europe face difficult budget decisions from many high quality national and international science projects. Without at this stage offering advice on funding priorities, the ESF commends the science case for the ESS, recognises it as a truly European project and urges that governments and funding agencies work together to take a timely decision."

Enric Banda
Secretary General, European Science Foundation (ESF)

Editorial

This science case document reports a joint effort of the ESS Scientific Advisory Committee (SAC), the European Neutron Scattering Association (ENSA) and scientists from a multitude of scientific disciplines from all over Europe and beyond. The goal of this document was not to encompass neutron applications in all their width and breadth – this has been done at earlier occasions – but rather to assess key scientific problems where the impact of ESS will be most significant. These "scientific flagships" are the essential content of this report. They have been identified and addressed by working groups covering the fields of solid state and particle physics, of materials and soft condensed matter science, of biology and earth science, of chemistry and engineering and the science of liquids and amorphous materials. These working groups evaluated the trends in their respective fields, identified key areas and assessed the impact of ESS. This approach revealed the very broad scope of ESS, it dealt with new concepts and visions and sometimes also with the dreams of these experts. Though there was plenty of scientific imagination involved, predictions for discoveries were not attempted but extrapolations from today´s perspective were worked out in order to illuminate the science in the decades to come. The reality in the age of ESS will almost certainly surpass our imagination by far.

Beyond its invaluable contribution to scientific progress ESS will also become an indispensable tool for Europe´s technological advance. Views into the future were undertaken on the basis of the European research missions of today. These missions were identified from Europe´s framework six program and the foresight themes of different European governments. Such themes comprise for instance functional materials and nanotechnology, microsystems and information technology, traffic and transport, sustainable development, clean technologies and environmental systems, biotechnology and health as well as the preservation of cultural heritage and archaeometry. These topics were assessed by transdisciplinary working groups, who again worked out the impact of ESS in key areas. Finally the complementarity of neutrons with other techniques was thoroughly investigated by a special working group.

Thanks to the inspiration, the excitement and the broad knowledge of all contributors, the document has become and almanac of the neutron science, to be enabled by ESS. It contains a wide variety of ideas, it outlines opportunities and features a deep assessment of the future role of neutrons in the age of ESS in European science and technology.

I like to take the opportunity to thank all participating scientists for their continuous and extraordinary initiative and enthusiasm, clearly visible at the two dedicated SAC workshops

dealing with this science case and also in between, when the input to the workshops had to be prepared. In particular I like to thank the conveners of the science and transdisciplinary groups who shaped the results of all the considerations into the exciting reports printed in this volume.

Jülich, 15.04.2002

Prof. Dr. Dieter Richter
Chairman Scientific Advisory Committee (SAC)

Executive Summary

1. ESS - a European Source of Science

The European Spallation Source (ESS) will be the most powerful third generation neutron source ever designed. Science at ESS will address our world in all its diversity. All the remarkable materials and substances we use, and from which all life is made, are based on combinations of about 100 different types of atoms. Neutrons are one of the most powerful probes for making the arrangement of these atoms visible and for measuring the forces between them. Research at ESS will be performed in areas ranging from materials science to soft matter science, from earth science to particle physics, from chemistry to engineering, from solid state physics to biology and medicine.

2. The ESS will provide new knowledge

Research with neutrons serves first of all to expand our knowledge. The unprecedented improvements in effective intensity delivered by ESS will open new scientific opportunities in many fields of condensed matter science. To give some examples, the ESS will

- *advance our knowledge of the functioning of biological surfaces and interfaces.*
- *unravel complex self organisation behaviour in soft condensed matter.*
- *provide insights into the state of matter under the extreme conditions of the earth's interior, and enable predictions of volcanic eruptions and earth quakes.*
- *give us a new "inside" view of engineering structures.*
- *probe the unsolved mysteries of quantum mechanical tunnelling states in glassy materials.*
- *lead to an understanding of the hydrogen bonds in supramolecular chemistry and pharmaceuticals.*
- *address fundamental questions of particle physics and cosmology, such as the origin of matter antimatter asymmetry and grand unification.*

3. The ESS will advance technologies of tomorrow

The ESS will offer new and improved tools that will enhance our ability to develop and optimise tailor made new materials and substances to achieve better functionalities and higher reliabilities for the benefit of European industry and society. Neutrons at ESS will

- *monitor catalytic processes, they will provide the atomic picture underlying the electrochemistry of batteries and fuel cells and will lay important foundations for a future hydrogen based energy economy.*

- *enable research on materials for advanced information technologies and study for instance nanostructured magnetic clusters, dots and layers and examine highly non linear micro magnetic media.*
- *enable knowledge based design of functional materials such as magneto resistive materials and high temperature superconductors, photoresponsive materials for holographic data storage and magnetic elastomers.*
- *impact structure based drug discovery, structural aspects of aging, food processing and the production of biosensors and biochips.*
- *enable us to access the very large parameter space of nanosystems such as nanostructural alloys and composites, advanced functional polymer materials, nanotubes, nanocrystals and in particular the self organisation of compartmented macromolecules, and allow the in-situ observation of structure formation and processing.*
- *become an important tool for archaeology and conservation of artefacts facilitating the dating of excavation sites, the unveiling of ancient trading patterns, cultural exchanges and manufacturing techniques.*
- *enable better, safer, more economical and environmentally friendly designs of traffic and transport structures through the uptake of new materials, an improved understanding and refinement of the manufacturing processes and a surer foundation for structural integrity assessments.*

4. Neutrons provide undisturbed views of matter and cannot be replaced

Since they are electrically neutral, neutrons are gentle probes that penetrate deep into materials without causing damage. Whether they look at glasses, plastics, metals, proteins, amino acids or magnetic materials, scientists and engineers obtain a direct view on the internal structure, arrangement, magnetism and even movements of atoms.

- *Neutrons see where the atoms are and how they move.*
- *Neutrons see the elementary magnets.*
- *Neutrons see all nuclei whether they are heavy or light.*
- *Neutrons distinguish isotope labelled regions in complex structures.*
- *Neutrons penetrate deep into matter.*
- *Neutron results are easily interpreted because the cross section is quantitative and simple.*

As realised by the ESF standing committee for Physical and Engineering Science (ESF-PESC) due to these properties neutrons will always be an indispensable tool for studying atomic structures and dynamics in condensed matter and cannot be replaced by other techniques. However, the value of neutron results can be considerably enhanced by the use of complementary data obtained by other methods and similarly data obtained by other methods are enhanced by the use of neutron data. There is no single experimental technique that can provide

us with all the information we need to know about materials, different techniques based on different physical processes, provide different information and as the materials under study become ever more complex, it becomes more crucial to study them using multiple complementary experimental techniques.

5. The ESS – the new frontier for neutron science

The history of research with neutrons dates back to the early 1950´s, when the first research reactors were built. Thanks to their unique properties neutrons were used to investigate an ever larger number of research fields. As a consequence the demand for neutrons continues to grow: more neutrons, and better neutrons, tailored to particular uses. Since 30 years the development of research reactors at reasonable costs (and technical risk) has found its end with the ILL, the world's premier research reactor. Now a third generation of neutron sources is emerging. They produce neutrons by shooting highly accelerated protons at a target, causing the nuclei in the target to release neutrons by a process similar to evaporation. Scientists call this spallation. Spallation sources can be run on a pulsed basis. That enables a much more effective exploitation of the neutrons produced.

Among these newly evolving Megawatt spallation sources, ESS will be the premier facility. It will provide for an enhancement in source performance for the different applications by factors between 10 and 100, i.e. much more than has been achieved since the pioneering days of Brockhouse in the early 50's (factor 4). The ESS has been devised to operate using two different pulse sequences to two different targets, so that those researchers using slower neutrons and those using faster neutrons can both look forward to working under optimum conditions. This means that, for example, scientists working on "soft condensed matter" including biology will not have to compromise with colleagues whose subject is hard materials (metals, ceramics, magneto-electronics, chemical and mechanical engineering).

6. The ESS layout and instrumentation is science driven

The crucial ESS parameters, power level, repetition rates and proton pulse length were chosen such as to optimise its scientific opportunities. For that purpose, scientific experts from eight different disciplines analysed the future trends in their fields, identified flagship areas and derived the science demands on ESS. In parallel, instrument experts covering 28 generic instrument types which were grouped into nine categories, evaluated their performance at different possible target stations. The synthesis of the science demands and the instrumentation opportunities at the different target stations led to the choice of a short pulse 50 Hz and a long pulse $16^2/_3$ Hz

target station each operated at 5 MW beam power. Similarly, the instrumentation was prioritised according to the demands of the flagship areas in the different scientific disciplines.

7. The ESS instrumentation opportunities

ESS will offer an unprecedented jump of some two orders of magnitude in the crucial performance parameter, the instantaneous peak flux, compared to the leading continuous or pulsed sources of today. The goal of the ESS project is to combine the vastly enhanced, unique source quality with the most advanced instrumentation concepts. In this way the sensitivity of observing small signals or fast processes in real time (which is the main limitation of neutron scattering in general) is increased by as much as 3 orders of magnitude in some unique core applications and more than two orders of magnitude in the majority of cases. This huge step forward will be reached through the extensive experience accumulated over the past 5 decades at continuous sources and nearly 3 decades of progress with pulsed sources. Innovative concepts will further enhance the efficiency of using the source power in actual experiments. Examples include advanced neutron optical beam extraction schemes and sophisticated multiplexing techniques like repetition rate and wavelength frame multiplication. As a result ESS will not only surpass all other neutron sources, existing or being built, by its higher neutron brightness, but it will further enhance this advantage by a more efficient use of the neutrons produced.

8.The ESS is endorsed enthusiastically by the European users

Currently 4000 – 5000 European researchers are using neutron scattering for their scientific work and their number is expected to grow as the possibilities improve. They have organised themselves within the multidisciplinary European Neutron Scattering Association (ENSA). ENSA has developed a European neutron strategy featuring a network of regional sources and as the centrepiece the ESS. This concept has been unequivocally and enthusiastically endorsed by the neutron communities in the different European countries. 2000 – 3000 users are estimated to frequent ESS every year. More than half of them will be PhD students or postdoctorals, who in addition to carrying out frontline research will be educated in the international environment of ESS, thereby preparing them for the challenges of professional activity in an increasingly global scientific and industrial world.

Chapter 1

Introduction

1. Introduction

A source for many disciplines …

The ESS is the most powerful third generation neutron source ever designed.

This document presents the scientific case for the European Spallation Source (ESS), the most powerful third generation neutron source ever designed. Science at the ESS will address our world in all its diversity. All the remarkable materials and substances we use, and from which all life is made, are based on combinations of about 100 different types of atoms. Neutrons are one of the most powerful probes for making the arrangement of these atoms visible and for measuring the forces between them. Research at the ESS will be performed in areas ranging from materials science to soft matter science, from earth science to particle physics, from chemistry to engineering, from solid state physics to biology and medicine.

The ESS will underpin thousands of research projects.

The ESS will support thousands of individual research projects. It will be a facility for investigating the myriad manifestations of matter, all of which have their own intrinsic fascination. Scientific results obtained at the ESS will underpin many aspects of our future lifestyle and contribute in many ways to our scientific and technological progress. The ESS must be judged on the basis of all the opportunities for scientific discovery and technological development that it will enable. In their rich variety, wide range and multi-disciplinarity, they will make a significant contribution to our knowledge of the natural world, which is the basis of our technology. For the ESS project this multi-disciplinarity is at the same time a strength and a weakness. A strength, because of the width and breath of the scientific results that will be produced. A weakness, because it is much easier to make the case for a single scientific purpose and to assemble a well defined narrow community behind a project, rather than to rally scientists from a multitude of scientific disciplines and to bring together their different cases.

The ESS will make a significant contribution to our knowledge of the natural world.

We cannot hope to predict exactly what the specific challenges will be in the decades to come. In the early 1980's nobody could have predicted that fullerenes or high temperature superconductivity would be discovered within the decade. However we can confidently extrapolate from the present and predict where major advances would be possible if the ESS were available today.

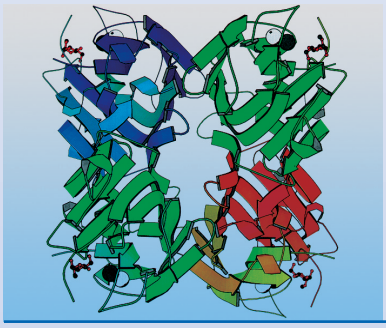
… providing new knowledge …

Research with neutrons serves first of all to expand knowledge.

Research with neutrons serves first of all to expand our knowledge. To give some examples, the ESS will

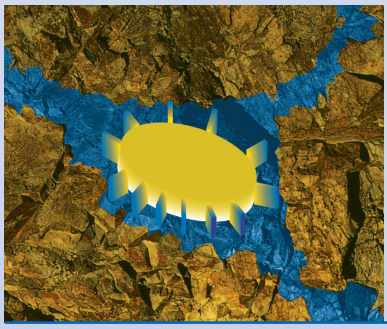
- advance our knowledge of the functioning of biological surfaces and interfaces;
- unravel complex behaviours in soft condensed matter;
- provide an insight into the state of matter under the extreme conditions of the earths interior, facilitating to the knowledge base for future predictions of volcanic eruptions and earth quakes;

Scientific opportunities at ESS



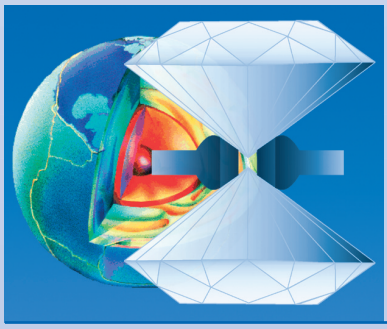
Biology and Biotechnology

Neutrons are particularly sensitive to the dynamics of molecules and single atoms. The relevant instrumentation at the ESS promises large gain factors, up to three orders of magnitude above what is available today. This will allow an unprecedented increase in experimental sensitivity, which, in combination with bio-simulation, will be applied to the study of atomic and molecular structure and dynamics in many fields of biology.



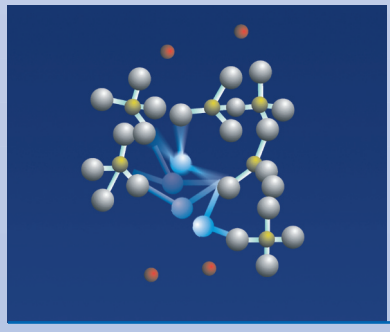
Polymers and Soft Matter

Complexity is one of the most common characteristics of soft condensed matter. The properties are often determined by key components that are dilute. Instrumentation at the ESS will allow the observation of such components under both equilibrium and transient conditions. One example is the exploration of the structure, dynamics and phase behaviour of multicomponent complex fluids in porous media, preparing the way for e.g. tertiary oil production or the remediation of soil contamination.



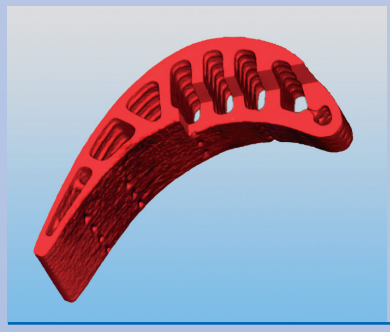
Earth and Environmental Science

Geological activity in the earth's upper mantle is responsible for geo-hazards such as earthquakes and volcanic eruptions. At the ESS, high temperature and high pressure studies of the structure and dynamics of minerals and magmas under earth mantle conditions will lead to significantly improved predictions of earth dynamics and the related geo-hazards.



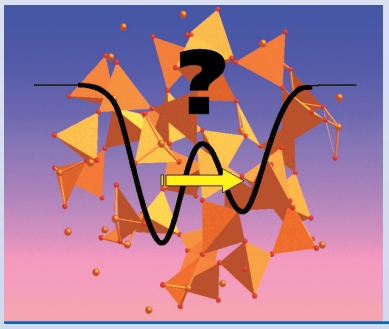
Computer Simulation and Neutron Scattering

Neutron diffraction data is routinely used as the basis for structural models of crystals, glasses and liquids. In the future advanced modelling software will allow the production of dynamical models, e.g. 'movies' showing 'where the atoms are and what the atoms do', on the basis of inelastic neutron scattering data measured over a wide range of momentum and energy transfer at the ESS.



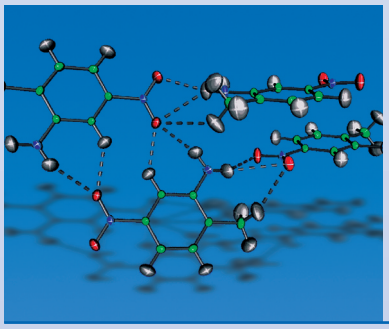
Engineering and Material Science

Structure sensitive imaging will add a new dimension to real scale tomography and radiography. Large field, high resolution images will display the distribution of structures in a material. Real time tomography of hidden objects, such as lubricants or cooling fluids, will become possible.



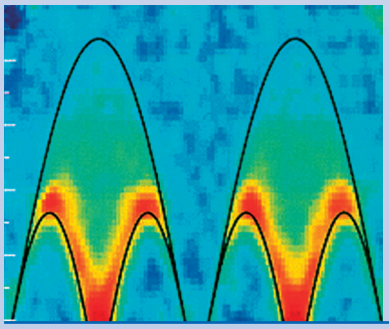
Amorphous and Disordered Materials

One of the major unsolved mysteries in the dynamics of amorphous solids, the origin and nature of the quantum mechanical tunnelling states, will be addressed. These states are ubiquitous in glasses but have so far proved elusive to microscopic measurement due to their extremely low density.



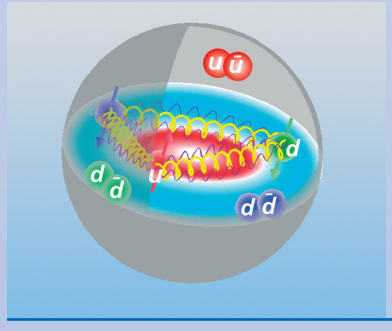
Chemistry and Chemical Structure

The study and understanding of the H-bonding holding together complex molecules, and arrays of molecules, will have an important impact on pharmaceutical materials and supra-molecular chemistry, allowing more rational molecular engineering.



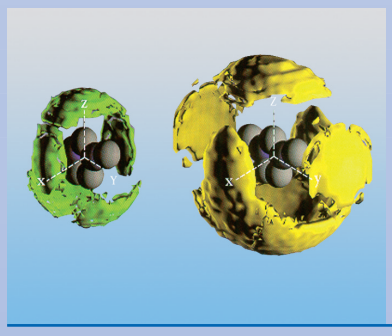
Solid State Physics

Neutrons provide unique access to the magnetic structure and dynamics of solids. Neutron beams at the ESS will provide maps of the magnetic polarisation and spin dynamics of nano-structured systems. Furthermore, the ESS will allow experiments under the extreme conditions required to explore quantum phase transitions.



Particle Physics

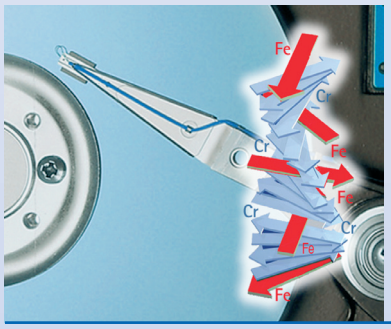
The neutron can be seen as a composite particle consisting of quarks, virtual pions and gluons. Its internal structure determines the decay process, the magnetic moment, and an anticipated electrical dipole moment that would indicate new physics beyond the Standard Model of particle physics. Related measurements can be performed using cold and ultra-cold neutrons. Essential contributions can be expected to the unification of fundamental forces in nature.



Liquids

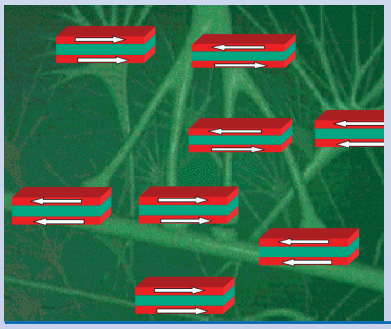
Nowadays, three-dimensional liquid structure refinement can be carried out for liquids of small molecules. A challenge for the ESS will be to extend this to large molecules. ESS will enable the understanding of why some ion combinations or molecular species in solution induce protein folding, while others cause denaturation. ESS will also reveal the structural changes of water surrounding the macromolecule and deliver crucial information on the origin of hydration forces.

ESS contributions to European research missions



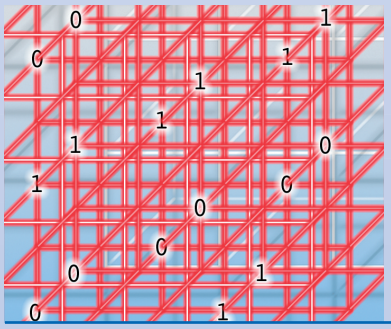
Magnetoelectronics

Magnetic sensors based on the giant magnetoresistance (GMR) effect can be found in hard disc reading heads, position sensors for precision tools and ABS systems. GMR sensors exploit the magnetic field dependence of the electrical resistance in layered magnetic structures, whose details were clarified by neutrons. The ESS will allow experiments on ultrathin and laterally confined films, in order to explore the magnetic structures and interfaces of reading devices as the lateral size of GMR heads shrinks to cope with increasing storage density.



Magnetic Neural Networks

GMR, together with the Exchange Bias (EB) effect that pins the direction of magnetic moments in a certain direction, allows the construction of spin valves, which are essential components of magnetoelectronics. On this basis, smart micro-magnetic-media can be envisaged that could become prototypes for magneto-neural-networks. The ESS will be an invaluable tool for the structural and dynamical evaluation of such systems.

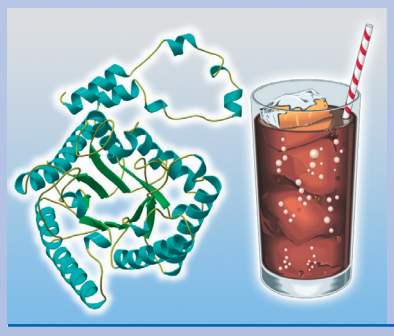


Holographic Laser Discs

Liquid crystalline polymers with photosensitive side groups can undergo pronounced photo-induced structural rearrangements that could be exploited, for instance for three dimensional holographic laser discs with storage capacities of the order of 1000 GB. Structural and dynamical neutron studies at the ESS will help to direct systematic searches for new optimum formulations that meet the demands of a wide variety of applications.

Drug Discovery

Knowledge of the three dimensional structures and dynamics of proteins and nucleic acids, as receptors for drug molecules, opens a structure based path to new drug discovery. For instance, major diseases in aging, such as Alzheimers, are caused by the formation of insoluble amyloid deposits of proteins in the brain and neurofibral tangles in the nerves. A combination of x-ray and neutron crystallographic studies, both of the enzymes that catalyse processing of the amyloid precursor proteins and of the proteins that associate with the plaques, could make an outstanding contribution to the design of therapeutic agents.

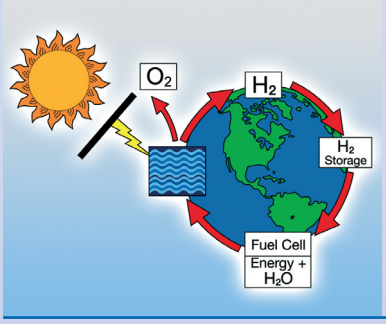


Enzymes in Food Productions

Improved knowledge of the active site structures in enzymes can be used to support their rational redesign. One of the most important enzymes in food production – glucose isomerase – isomerises glucose to fructose. Fructose is used extensively as a sweetener in the food industry, for instance in soft drinks like Coca-Cola. This is a billion euro industry. If the ESS were available today, it would allow a clearly resolved distinction between the magnesium and oxygen atoms in the enzyme and facilitate placement of the bound water molecules and cations that are involved in the enzyme's action.

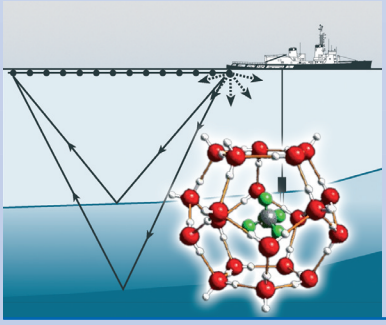
Unveiling Ancient Technologies

Neutron diffraction reveals novel information on archaeological artefacts and helps to unveil long forgotten ancient technologies. One recent example is an analysis of the Copper Age axe of the 5200 year old Iceman (Ötztal). Neutron scattering techniques have only recently been applied to such archaeological artefacts. Many goals are not yet achieved, mainly due to the limitations of present day neutron sources.



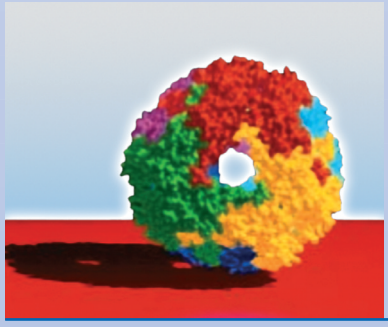
Hydrogen Energy Economy

Hydrogen is an ideally clean carrier of energy. A future hydrogen based energy economy will need substantially better ways of storing hydrogen in a safe, light and affordable manner. Metal hydrides, and ionic compounds of the lighter elements, appear promising candidates. Their relevant structural and dynamical properties can only be clarified by neutron scattering. The ESS will provide the means to study kinetic loading and unloading cycles in-situ, aging processes and associated diffusion mechanisms. This knowledge will be of great importance for rational materials design.



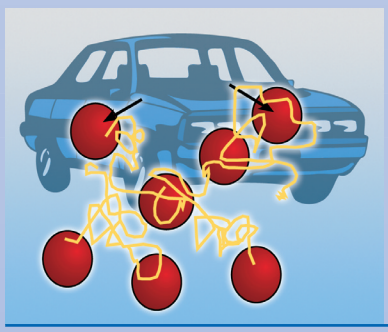
Methane Clathrates: Energy Resource and Marine Hazard

Methane-water clathrates contain the largest proportion of natural gas in the shallow earth (about 7 times the amount available in sedimentary rocks) and constitute an enormous energy resource. However methane release as a consequence of clathrate instabilities causes green house effects and marine geohazards. A full understanding of the crystal chemistry demands structural and dynamical studies under real conditions which are far from the reach of today's neutron sources, but will be within the reach of the ESS.



Templating of Nanostructures

With detailed control of chemistry and processing conditions, it is possible to fabricate complex nano-scale ordered block copolymer systems that can be used as templates for high quality fillers, fabrication of efficient catalysts, medical implants, pharmaceutics, photonic and smart materials, novel nanostructured magnetic devices etc. The rational design of such materials needs to be based on knowledge. The structural complexity, the huge multidimensional parameter space involved, and investigations into the kinetics of structure formation require the high flux of ESS.



Nanomaterials for Transport and Traffic

Rising energy prices and growing environmental awareness are intensifying the search for materials and processes with improved performance. The ESS will have an impact on the development of engine/propulsion technology and novel materials for transport. It will advance our understanding of component failures and of lubrication issues on an atomic/molecular level. One example is the production of light weight nanocomposites, where nanoscale fillers reinforce a polymer matrix.

- give us a new ‘inside’ view of engineering structures;
- probe the unsolved mysteries of quantum mechanical tunnelling states in glassy materials;
- lead to an understanding of the hydrogen bondsin supramolecular chemistry and pharmaceuticals;
- address fundamental questions of particle physics and cosmology, such as the origin of matter-antimatter asymmetry and the validity of grand unification.

The properties of the neutron itself relate to particle physics and cosmology.

A short list of such flagship experiments is displayed in the box “Scientific opportunities at ESS”.

… and new technological solutions.

The ESS will offer new tools to deepen our understanding of both the natural world and the artificial world of materials.

Aside from its impact on basic sciences, neutron scattering at the ESS will also make a direct contribution to the solution of many problems more closely related to everyday life. The ESS will offer new and improved tools that will deepen our understanding of both the natural world and the world of artificial materials, and enhance our ability to use this knowledge effectively, for the benefit of European industry and society.

The ESS will have a strong impact on a large variety of technologies.

Neutrons at the ESS will monitor catalytic processes, they will provide the atomic picture underlying the electrochemistry of batteries and fuel cells and will lay important foundations for a future hydrogen based energy economy. Neutrons at the ESS will be instrumental in the development of improved magnets for magnetic levitation trains. They will facilitate a better understanding of materials failure – one recent outstanding neutron scattering result clarified the failure mechanism of the Inter City Express train accident in Eschede. Neutrons at the ESS will have an impact on nanotechnology - at the Spallation Neutron Source (SNS) in Oak Ridge a multi-million dollar centre for nanophase materials is currently being constructed. Important insights are expected into the principles of nanodevices and spintronics, molecular magnets, magnetic films and surfaces. The development of functional materials involving self-organisation, of supramolecular chemistry featuring hydrogen bonds, and many aspects of biomimetics will be fertilised by the ESS. Biology and biotechnology, e.g. enzymatic catalysis, drug discovery and maybe even our understanding of phenomena such as amyloid diseases causing age related disorders, will benefit from the ESS. (At the SNS special beam lines in this field are under consideration.)

The ESS will be instrumental in drug discovery.

A summary of the impact of the ESS on such research themes is displayed in the box “ESS contributions to European research missions”.

I. ESF/PESC 1996: Prospects for neutron scattering; need for an ESF initiated next step

The behaviour of materials is determined by the arrangement of atoms and the forces between them. Our knowledge of these comes from a range of sophisticated scientific techniques that are sensitive at the atomic and molecular level. Many such techniques are based on scattering from matter by x-rays, light, electrons and neutrons, or more recently on direct imaging by scanning probes. Their power is enhanced by theory and computer modelling. Each method is optimised to probe different aspects of structure and dynamics and there is a great deal of relevant complementarity.

Materials properties are ultimately related to the arrangement of atoms and the forces between them.

The neutron is, in many ways, an ideal probe for the investigation of condensed matter. The neutron interacts with matter mainly through the strong force as a nuclear interaction and the electromagnetic force via its magnetic dipole moment. A list of the most important properties of the neutron and its advantages for the investigation of condensed matter is given in the box on “The main advantages of neutrons”.

The neutron is an ideal probe for investigating many facets of condensed matter.

Neutron scattering has consequently made outstanding contributions to our detailed understanding, at a microscopic level, of technically important materials such as plastics, proteins, polymers, fibres, liquid crystals, ceramics, hard magnets and superconductors, as well as to our understanding of fundamental phenomena such as phase transitions, quantum fluids and spontaneous ordering. A short list of some “Major achievements of neutron scattering” is given in the corresponding box.

Neutrons have made outstanding contributions to condensed matter science.

In 1996 the European Science Foundation (ESF) completed a major evaluation of the scientific prospects for neutron scattering in Europe [1]. This provided a forum where the scientific case for neutrons could be developed, and laid a firm basis for the scientific case for the ESS. A central part of the ESF evaluation was a workshop on the “Scientific Prospects for Neutron Scattering with Present and Future Sources”. The ESF Standing Committee for Physical and Engineering Sciences (PESC) evaluated the results and came to a number of important conclusions:

The European Science Foundation (ESF) performed a major evaluation of neutron scattering science in 1996.

- The use of neutrons continues to evolve, both in traditional and in new fields. Given the enormous impact of new materials in technology, no end to this process can be foreseen. The demand for more sophisticated use of neutrons continues to grow, so that aggressive programmes of instrumentation development are vital to the field.

The demand for neutrons continues to grow.

- Non-neutron tools for matter investigation, such as synchrotron radiation, cannot substitute for the future use of neutron beams. Even in the long term, both neutron scattering and synchrotron radiation research will continue to be indispensable, because the two techniques cannot replace each other (nor be replaced by third methods); indeed they complement and extend each others range and opportunities.

Non-neutron tools cannot substitute neutron beams.

The main advantages of neutrons

The main properties of the neutron that are exploited in scattering studies can be summarised as follows:

- The *energy* of thermal neutrons is similar to the energies of atomic motions. A wide *range of energy scales* may be probed, from the nano-electron volt energies associated with polymer reptation, through molecular vibrations and lattice modes, to electron volt transitions within the electronic structure of materials.

- The *wavelengths* of thermal neutrons are similar to atomic spacing, providing structural information over ten orders of magnitude in scale (10^{-5} to 10^{5}Å). Measurements are thus possible over distance scales ranging from that of the wave function of the hydrogen atom to those of macromolecules.

- Neutrons interact via the strong interaction and hence *see nuclei*, rather than the diffuse electron cloud seen by X-rays. This has major advantages, such as to be able to see light atoms (e.g. hydrogen) in the presence of heavier ones, and to distinguish neighbouring elements more easily. The fact that the scattering cross section of an atom generally varies between isotopes of the same element allows us to exploit isotopic substitution methods to yield structural and dynamical information in even greater detail. It also facilitates the use of contrast *variation*, which enables us to contrast out parts of a complex system, for example the nuclei acid or the protein component of a virus.

- The neutron's *magnetic moment* is ideally suited to the study of microscopic magnetic structures and magnetic fluctuations that underpin macroscopic magnetic phenomena in materials.

- Neutrons only perturb the experimental system *weakly* and its interaction with a nucleus has a simple form. This greatly aids interpretation and often means that neutron scattering provides the most reliable scientific results in areas as diverse as the structure of water or the strain mismatch in superalloys used in turbine blades.

- Neutrons are *non-destructive*, even to complex, delicate biological materials.

- Neutrons are *highly penetrating*, allowing the non-destructive investigation of the interior of materials. This makes them a genuine microscopic *bulk probe*, routinely used in complex environments such as furnaces, cryostats and pressure cells, and enables the study of *bulk processes under realistic conditions.*

- The diversification of neutrons into a number of wider scientific areas continues. Examples are earth sciences, pharmaceuticals sciences, biology and engineering.

Neutrons gain importance in engineering, earth and pharmaceutical sciences and biology.

- The direct impact of neutrons on 'wealth creation' is increasing and will continue to do so. Examples of industrial relevance are in the fields of multilayers, polymers, material science, and engineering.
- Unless appropriate action is taken, sources of neutrons, and hence the supply, are likely to decrease in the next ten years.
- Given the existing and future expected demand in Europe this makes it imperative that: (a) full use is made by European users from the basic and applied science of the present network of medium flux national sources and the highest flux sources – ILL and ISIS and (b) a mechanism is initiated by the ESF to co-ordinate the design and funding request that will allow a truly advanced European Source to be operational within 15 years.

Europe should construct a truly advanced neutron source.

According to this evaluation, the importance of the results obtained using neutron scattering techniques lies not only in their significant – often crucial – contribution to the corpus of scientific knowledge, but equally in their impact on a remarkably wide range of technologically and industrially important areas. Present and future examples that can be cited include biotechnology, drug design, pharmacology, materials processing, environmental technologies, catalysis, energy storage, new materials, energy transmission, transport, data storage and quantum devices, all covering crucially important aspects of modern civilisation.

II. The community of neutron users

It was originally the physics based community that pioneered and developed neutron scattering. However, with the introduction of the user facility concept, neutron science has expanded into all disciplines of condensed matter science, including biology. In 1996 the European Neutron Scattering Association (ENSA) and the European Science Foundation (ESF) conducted a survey of the European neutron scattering community [2]. The distribution over selected European countries and scientific disciplines is illustrated in Figure 1. 4000 scientists in Europe use neutron scattering as one of their major research tools.

4000 European scientists use neutron scattering as one of their major research tools (ENSA-ESF study 1996).

Since the publication of the ENSA/ESF report, there has been a notable expansion of neutron applications in the engineering sciences (*in-situ* tomographic studies, strain scanning etc.) principally as a consequence of substantial investment in dedicated instrumentation. This area is seen as one of considerable growth and increasing industrial importance, for example in support of the aerospace industry.

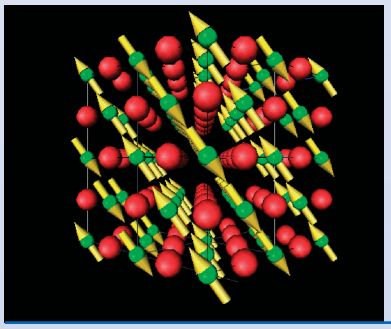
Growth areas in neutron scattering are:

- Engineering

Similarly, the advent of higher resolution and higher count-rate diffractometers which enable the structural characterisation of

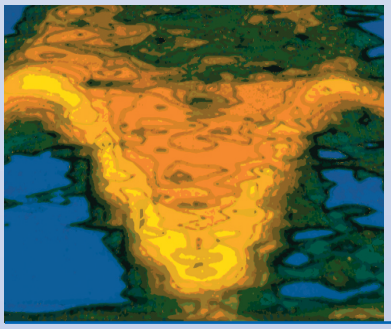
- Earth science

Major achievements of neutron scattering



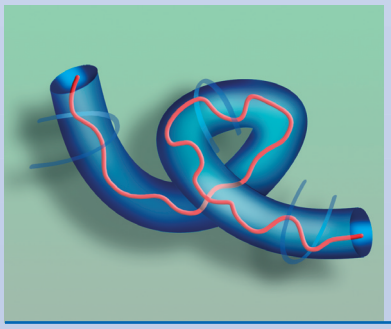
Magnetic Structures

Almost everything we know about magnetic structure – from the early demonstration of anti-ferromagnetism in simple systems (Shull, Nobel prize 1994), to the complex magnetic structures being developed by hard magnets – has come from experiments with neutrons. Similarly, polarised neutron reflectometry provides unique access to the surface and interface magnetism in thin films and multilayers.



Elementary Excitations and Phase Transitions

Similarly, nearly all our knowledge of elementary excitations such as phonons or magnons in crystalline solids, and their relationships to 2nd order phase transitions, stems from inelastic neutron scattering (Brockhouse, Nobel prize 1994).

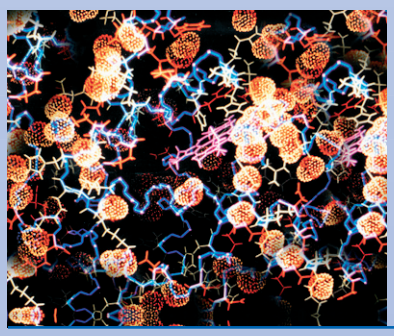


Polymer Conformation and Dynamics

Neutrons have provided the most direct information on polymer conformation and the associated scaling laws, and polymer dynamics such as reptation, corroborating the Nobel prize winning theoretical concepts of Flory (1974) and DeGennes (1991).

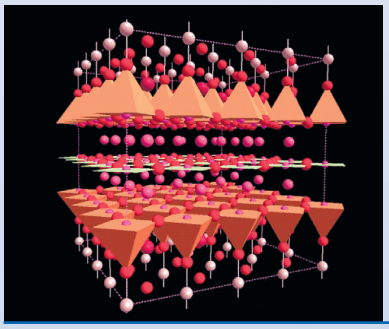
Structure and Dynamics of Liquids

Neutrons have provided much of our basic understanding of the structure and dynamics of liquids. The results have had a major influence on theoretical developments such as memory function formalism, or mode coupling theory for description of the glass transition, and the development of computer simulation techniques now used widely from fundamental physics and chemistry to biology.



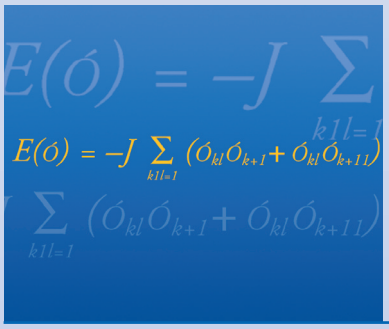
Proton Positions and Motions in Biomolecules

Neutrons have determined water organisation in proteins and other biological systems and function critical hydrogen positions in enzymes. Inelastic neutron scattering led to a characterisation of large amplitude internal motions in small proteins, and of a dynamical transition that is correlated with function. Neutron studies on lipid membranes have provided the basis for our present view of the bilayer as a dynamically rough and extremely soft surface.



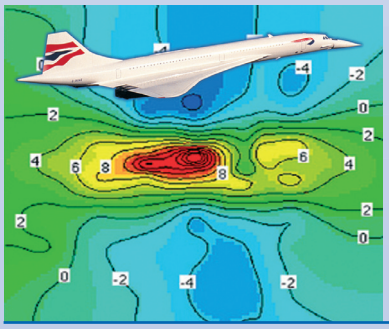
Crystal Structures and Magnetism of High Temperature Superconductors

Neutrons have provided the definitive crystal structures of high temperature superconductors, which serve as the basis for all considerations of the mechanism of superconductivity and have led to production of better quality materials. Neutron spectroscopy has provided unique information on the nature of magnetism in high temperature superconductors, on the interplay between magnetic fluctuations and superconductivity and on the role of the lattice dynamics.



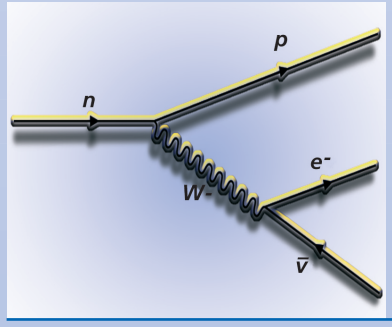
Concepts of Statistical Physics

Neutrons have made major contributions to our understanding of model systems for statistical physics in one, two and three dimensions, including verification of the Haldane conjecture, determination of the properties of the Haldane gap, and the discovery of solitons as the characteristic elementary excitation of strongly non-linear magnetic systems.



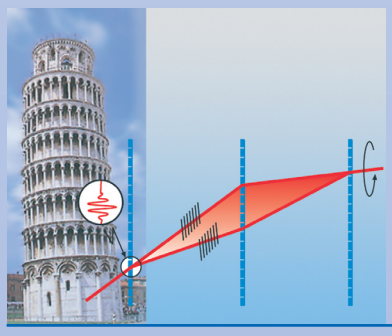
Strain in Engineering Materials

Neutron strain measurement on engineering materials has made an important contribution to our knowledge of residual stresses. These stresses are essential to making reliable estimates of component life times. Important work has been carried out on welded structures, in particular the method is accelerating the introduction of new friction based welding techniques. Often post weld heat treatment is needed to reduce potentially life time threatening residual stresses; neutron diffraction has improved their definition.



Electro-weak Interaction

Neutron decay experiments made essential contributions to the understanding of the „weak interaction“ and the unification of the electromagnetic and weak interaction to the „electro-weak interaction“. Parity non-conservation has been shown for the neutron decay, and neutron decay data also contributed to fixing the number of lepton families as three.



Quantisation of Neutron Waves in the Field of Gravity

Four hundred years after Galilei neutron physicists observed the quantisation of ultra-cold neutrons in the gravity field and the quantisation of thermal neutrons due to confinement.

complex multiphase minerals, soils and clays, is expected to lead to a growth in the use of neutrons by earth scientists, via a transfer of neutron technology from physics, chemistry and materials science.

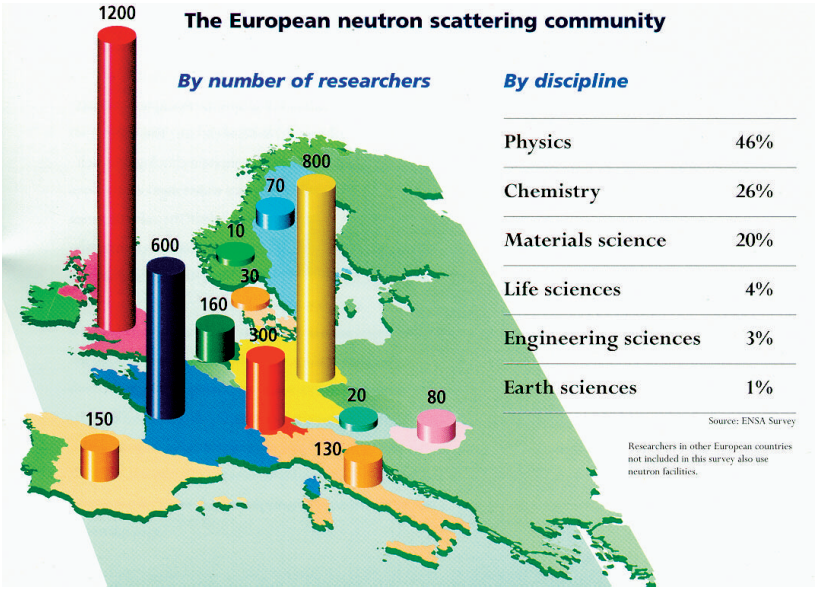


Figure 1: Distribution of neutron users across European countries and scientific disciplines.

- Biology

If we inspect the scientific demand at front rank facilities such as ILL and ISIS, an important observation can be made. While applications in the life sciences are at a level of 4 % on a European average, at ILL 15 % of all beam time is requested in this area. This high level of request at the current highest flux source strongly emphasises the need for particularly high source intensity for those experiments that explore complex biological matter. Neutron scattering is currently at the threshold of the sensitivity necessary for many types of investigation of the structure and dynamics of biological materials. The ESS will lift neutron scattering in life sciences well beyond this threshold, so a further strong increase in applications to biological problems can confidently be predicted. A recent ESF study [3] also underlined the growing importance of neutron science in biology. It concluded that:

Current neutron sources are at the threshold of sensitivity required for the solution of biological problems. The ESS will move well beyond the threshold.

"The neutron approach is unique in providing simultaneously the energy transfers involved and the amplitude of the motions. Neutron studies in general provide information that cannot be obtained by other methods and are strongly complementary to x-ray, electron microscopy and NMR. The use of neutrons in biology, however, has been severely restricted by the lack of beam time due to the shut-down of reactors and the strong demand on the few existing sources that have the necessary instrumentation."

It is estimated that 2000-3000 scientists will carry out experiments at the ESS every year. More than half of them will be PhD students or postdoctoral researchers, who in addition to carrying out frontline research will benefit from the

international environment of the ESS, preparing them for the challenges of professional activity in an increasingly global scientific and industrial world. Beyond the immediate impact on young scientists using neutrons, a decision to build the ESS has great potential for increasing the interest of young people in science and technology. It would express a commitment to science in Europe at a level that challenges investment in the USA and Japan. Training opportunities would be produced in accelerator physics and technology, biology, chemistry, engineering, materials science and physics. There are few investments in a single facility that would have similar impact on so many different scientific fields.

In the international environment of the ESS young scientists will be prepared to meet the challenges of the global scientific and industrial world.

III. Neutron sources world wide; a global OECD strategy

The quality and precision of neutron scattering experiments is primarily limited by the counting rate and, therefore, essentially by the intensity (thermal neutron flux) of the available neutron sources.

Neutron scattering is intensity limited.

Neutrons have 'traditionally' been produced by fission in research reactors optimised for thermal neutron flux. The first such reactors reached criticality in the 1940's with fluxes of 10^{11} to 10^{12} n cm^{-2} s^{-1}. Medium flux reactors like that in Chalk River (Canada) where Brockhouse developed inelastic neutron scattering, for which he was later to win the Nobel prize, became operational in the early 1950's with fluxes up to 4 10^{14} n cm^{-2} s^{-1}. Since then the source intensity has increased only by a factor of 4, an increase achieved almost 30 years ago with the commissioning of the Institute Laue Langevin (ILL) in Grenoble, the world's most powerful neutron scattering research reactor. In the 1990's the United States attempted to surpass the ILL flux by a factor of 5. This Advanced Neutron Source project was abandoned In 1995 because of huge costs (3G$) and technical difficulties.

Since the 1950's the source strength of research reactors has only increased by a factor of four.

Figure 2 shows the increase in flux available from neutron research reactors. After a steep increase in the late 1940's and early 1950's, the flux saturated about 30 years ago.

Accelerator based pulsed sources produce neutrons in a totally different manner. In the spallation process neutrons are evaporated from heavy nuclei by the impact of GeV protons from a high current proton accelerator. For each neutron produced by fission a heat load of 190 MeV has to be dissipated, but for neutrons produced by spallation the heat load is only 30 MeV. The cooling problems which limit high flux reactors are therefore much less severe at spallation sources. Furthermore, spallation sources may be operated in a pulsed mode, well adopted to the requirements of the majority of condensed matter experiments.

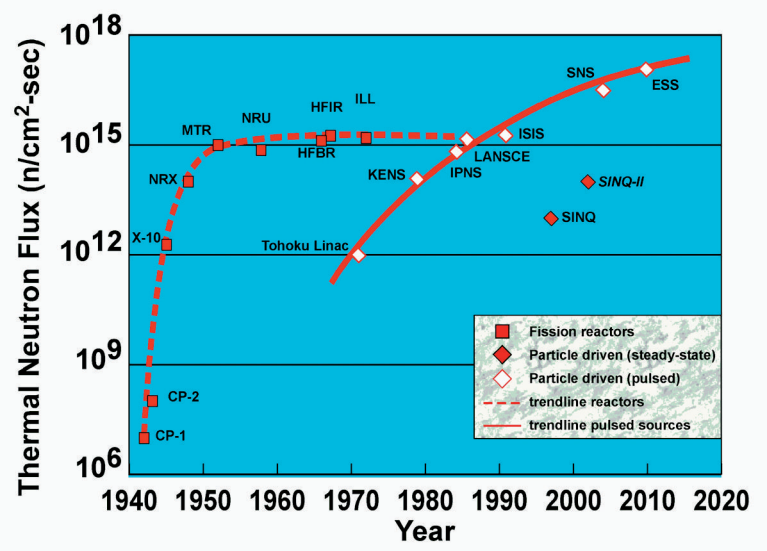


Figure 2: Development of the neutron flux available at reactor (average flux) and spallation sources (peak flux).

Proton accelerators have advanced strongly since the early 1980's. Today accelerator based spallation sources compete with the best research reactors.

In contrast to research reactors, accelerators have improved tremendously since the early 1980's through the development of linear accelerators including superconducting technology, strong focusing synchrotrons, charge exchange injection, radio-frequency quadrupole (RFQ) technology, sophisticated beam dynamics, computer control and particle tracking codes. Accelerator based neutron scattering sources have grown in strength by five orders of magnitude. In recent years, neutron scattering instruments have been developed which can take full advantage of the high intrinsic brightness of these sources using advanced time-of-flight instrumentation. From being a mere curiosity in the 1970's, the neutron beams produced by 100 kW beam power accelerators now rival those of the best reactors in the world.

Third generation MW spallation sources will play a key role in the future of neutron science.

Future third generation MW spallation sources, such as the ESS, will provide up to two orders of magnitude higher peak flux than the best sources of today and will play a key role in future neutron science.

In 1998 the OECD-Megascience forum evaluated the development of neutron facilities for neutron scattering research [4]. It noted that neutron scattering plays, and will continue to play, a crucial role in an extraordinarily diverse range of basic, strategic and applied research. It also noted that there will be a dramatic and inevitable decline in the number of facilities worldwide (Figure 3), and this requires urgent government attention.

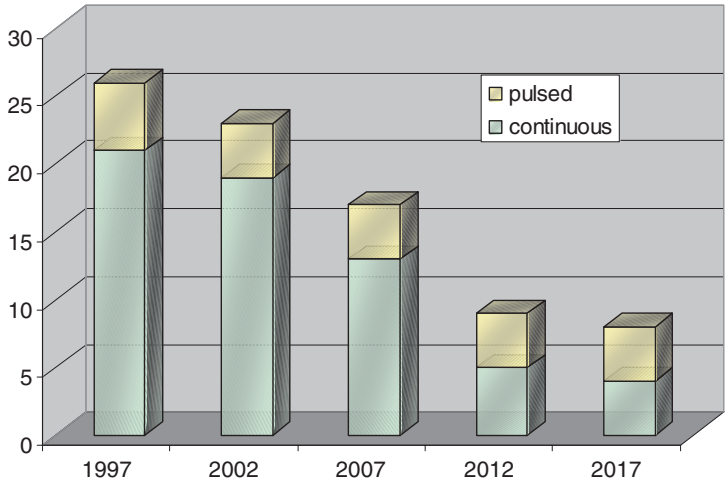


Figure 3: OECD projections for the number of major neutron sources available in the OECD countries and Russia.

The way forward was formulated in terms of a global neutron strategy that was endorsed by the OECD Ministerial Conference in 1999. The OECD recommended to maintain and refurbish a number of smaller sources and to extend and upgrade further the use of ILL and ISIS. But on the top of that an advanced European neutron source should stand alongside complementary third generation neutron sources in America and Asia. Both the US and Japan have adopted these recommendations and are now, in 2002, well advanced with the design and construction of their own advanced regional sources (the SNS and JNS respectively).

The OECD global neutron strategy requires three MW spallation sources in US, Japan and Europe.

IV. The ESS project

The ESS project, established in the early 1990's, is the European quest for excellence in neutron science. Taking into account the developments in reactor technology, and foreseeing the difficulties in this field, the European approach was based on the rapid advances in accelerator technology in the 1980's. This suggested the feasibility of a 5 MW LINAC driving a spallation neutron source. The concept offered an effective increase in performance in most applications of between 10 and 100 over existing neutron sources. This would represent by far the greatest single increase in source performance since the early 1950´s. The implications for neutron scattering science are clearly profound.

The ESS is Europe's quest for excellence in neutron science, moving the intensity frontier forward by one to two orders of magnitude.

By 1997 the scientific case for the ESS was published, together with the technical design, [5,6,7]. Indeed the ESS design had reached such an advanced state that the United States adopted the principal design features as the basis for their own SNS project. Construction of the SNS started in 1999, but unfortunately there has been no parallel advance of the ESS project within Europe.

Since the end of the feasibility study in 1997 several important developments have taken place:

1. It became clear that reactor based neutron sources have reached their ultimate design state with the ILL, i.e. the ESS should not only be the next generation spallation neutron source, but the next generation neutron source of all types.

The ESS will be <u>the</u> next generation neutron source.

2. Scientific research has developed and the use of neutron scattering has spread to new fields.
3. R&D has been carried out on many of the critical technical issues that were identified in the feasibility study.
4. Superconducting (SC) accelerator technology has progressed considerably. The ESS project has collaborated with the CEA on a feasibility study for a large (25 MW) SC proton accelerator feeding up to 5 target stations for different communities and purposes (CONCERT) [8].
5. Both the USA and Japan have started construction of next generation spallation neutron sources and they will define as of 2006 a new competition arena in neutron scattering, taking away the edge of Europe's best sources.

The USA and Japan have started construction of MW spallation sources.

In the Project Proposal Phase of the ESS project, the ESS Council has responded to these points. It has defined as its target to deliver a technically feasible, scientifically challenging and costed ESS project proposal to the European governments in 2002.

It objective is "*to design and construct a European next generation spallation source, that upon completion will be the best neutron source worldwide for all classes of instruments*."

The objective of the ESS is to be the best neutron source worldwide for all classes of instruments.

A Scientific Advisory Committee (SAC) was established not only including a broad assembly of European scientists, but also members representing the SNS and JP science parts. The SAC has reassessed the ESS proposal from 1996. At the ESS-SAC/ENSA Workshop in Engelberg [9] it was very clearly demonstrated that, in order to meet this objective, the ESS would need to have both a short pulse target station (SPTS, 50 Hz, 1.4 μs, 5 MW) and a long pulse target station (LPTS $16^2/_3$ Hz, 2.0 ms, 5 MW).

This requirement provided a challenge for the ESS accelerator task group to design a LINAC which could deliver 2.0 ms long 1.3 GeV pulses, of 110 mA peak current, interleaved between the 50 Hz pulses.

Scientific demands determined the choice of a short and long pulse target station, each operated at 5 MW.

The choice between a normal or superconducting high energy part of the LINAC is by no means trivial for a high current pulsed accelerator. As a consequence the ESS project has deliberately followed two different paths. The first is to revise the 1996 ESS accelerator proposal, taking into account recent R&D work. The second is to investigate a SC version based on the ESS-CEA CONCERT study [8]. The ESS Technical Advisory Committee (TAC) comprising experts from a large number of major facilities all over the world has reviewed them in January 2002. Its assessment is that both designs are capable of delivering the required performance. Details of the

R&D results for the accelerator, target and instruments are given in Volume III of this report. Table 1 lists the main parameters.

Table 1:
Parameters of the ESS facility

*ESS beam parameters**

Particles	Protons
Kinetic energy	1.334 GeV
Beam cross section	Elliptical 6 x 20 cm^2 2D-parabolic beam density distribution
Average current SPTS / LPTS	3.75 mA
Average beam power SPTS / LPTS	5 MW / 5MW
Peak current SPTS / LPTS	62.5A / 112.5 mA
Pulse frequency SPTS / LPTS	50 Hz / $16^2/_3$ Hz
Pulse width on SPTS / LPTS	1.4 μs (2 × 600 ns with 200 ns gap) / 2 ms

* Parameters referring to the SC version of the LINAC

ESS mercury target stations performance parameters

Beam power on targets	5 MW at 50 Hz, 1.4 μs short pulses/ 5 MW at $16^2/_3$ Hz, 2 ms long pulses
Target material	Mercury
Target type	Liquid flow target
Target container	Martensitic steel
Moderators (reference case)	H_2O at ambient temperature, coupled/decoupled Liquid H_2 at 20 K, coupled/decoupled
Reflector (reference case)	Lead, D_2O cooled
Heat deposition in target	2.80 MW at each of the two targets
Local peak power deposition in target material (time average)	ca. 2.5 kW/cm^3
Induced specific radioactivity at saturation for a 15 ton Hg-system	8.0 GBq/g at shutdown; 2.3 GBq/g after 10 days
Specific after heat of the target material	0.67 mW/g at shutdown; 0.13 mW/g after 10days

Neutronic performance of coupled H_2O- moderators at 5 MW beam power

Average thermal neutron flux density for 5 MW on target	3.1×10^{14} neutrons(cm^2 s)
Peak thermal neutron flux density	SPTS: 1.3 x 10^{17} neutrons / (cm^2 s) LPTS: 1.0 x 10^{16} neutrons / (cm^2 s)
Decay time of flux density (dominant mode)	150 μs

With both a short and a long pulse target stations the layout of the facility has changed. The long pulse target station, albeit very similar to the short pulse target station, allows substantial phase space tailoring of the neutron beams by reflector design, choppers and neutron optics, thus providing scope for novel instrumentation.

The two target stations (Table 1) (short pulse target station (SPTS), repetition rate = 50 Hz, 1.4 μs and long pulse target station (LPTS), repetition rate = $16^2/_3$ Hz, 2.0 ms) will share the proton pulses. Figure 4 shows an artist's view of the whole facility. All accelerator facilities (LINAC, 180° achromatic bend, compressor ring and beam transport line) will be buried under concrete and dirt shielding.

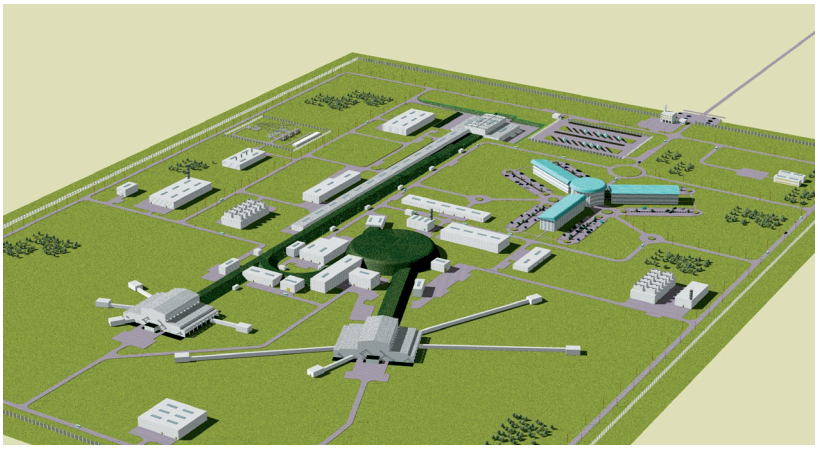


Figure 4: Artist's view of the ESS facility showing the LINAC building with an achromatic 180° bend leading to the compressor ring, from where the beam is distributed to the SPTS. The LPTS is directly connected to the LINAC.

In the first phase the instrumentation development task group of the ESS project assessed the performance of a set of generic instruments, assumed to be of well established design, for the various target station options considered. This evaluation served the dual purpose of supporting the scientific case and providing input for the selection of the target station configurations.

In the second phase innovative concepts of neutron extraction, beam tailoring and instrumentation have been investigated. A common goal is to optimise the instruments so that they benefit fully from the effective pulse lengths, pulse repetition rates, wavelength bands, beam divergences, etc. This can provide gain factors significantly higher than just using of the source power in a more straightforward manner. Pulse shaping, beam delivery optics and multiplexing are typical notions that we will be concerned with. Some of these have not yet been fully realised anywhere, but test results have been used as design assumptions in the evaluation of ESS instrument capabilities (such as using fast disc choppers to shorten pulse lengths). This was justified on the basis of ample experience with similar devices at continuous reactor sources. Others are opportunities to be explored in the future.

Pulse shaping, multiplexing and sophisticated neutron extraction are novel design features at ESS instruments.

In comparison to the best instruments existing today at reactor or spallation sources, the proposed two 5 MW (SPTS and LPTS) target stations will enable a 50-1000 fold increase of sensitivity in most experiments. The box "Instrumentation opportunities at ESS" displays important and novel instrumentation ideas.

Total instrument gains will be 50 – 1000 fold compared to today's best in class instruments.

Figure 5 outlines the phases of the ESS project. In 2001 the ESS design parameters were fixed and the documents presented now (May 2002) constitute the ESS project proposal. Further work on the details of engineering and costing will lead by the end of 2003 to an engineering baseline. A goal has been set to arrive at a decision on funding and constructing ESS by the end of 2003/early 2004. That will include a decision on the site. Preparatory work on site independent requirements has also been carried out and led to several expressions of interests, from both green field sites and sites taking advantage of already existing infrastructure. Such a time schedule will allow first scientific experiments to start in 2011/2012. From the beginning of 2013 the ESS will then move into regular operation mode serving European neutron scatterers.

In 1997 a first design studied demonstrated the feasibility of the ESS.

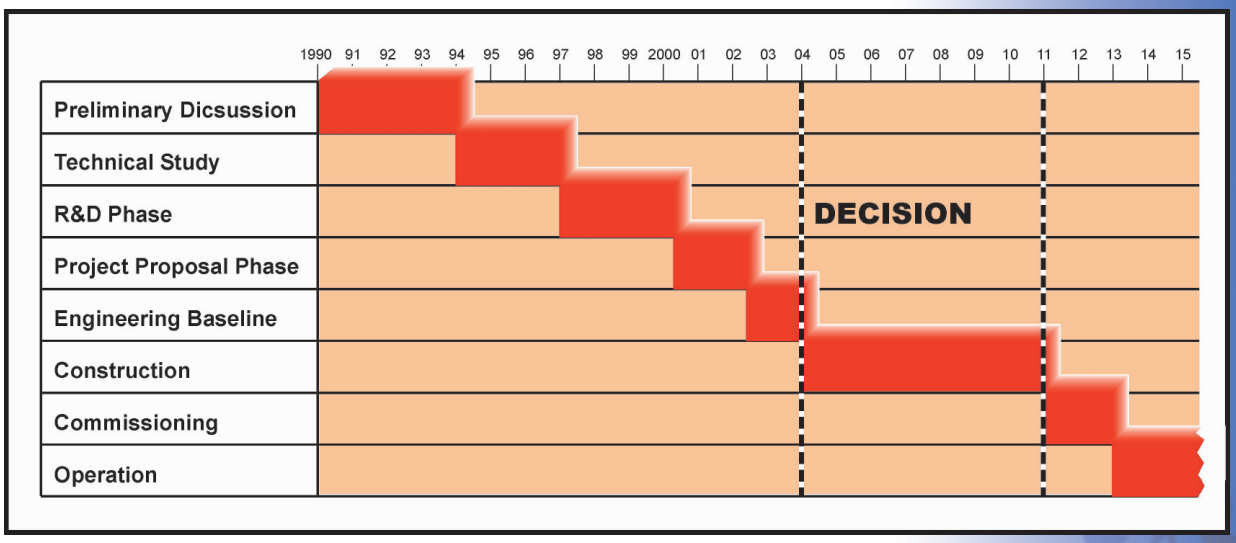


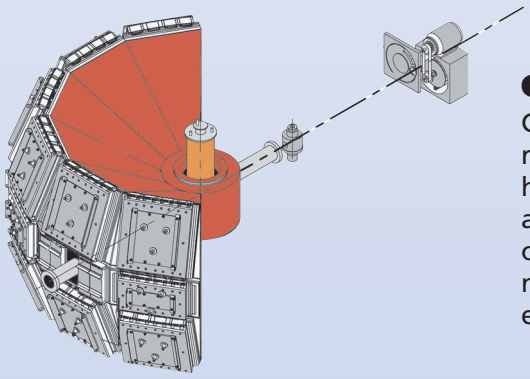
Figure 5: Outline of project phases.

V. Scope, procedure and content

The general scientific case for neutron scattering was described in the Autrans report, published by the European Science Foundation (ESF) and the European Neutron Scattering Association (ENSA) [1]. This report investigated the future potential of neutron science taking into account the other techniques available for the study of condensed matter and bio-materials. The present scientific case for the ESS complements and extends the earlier ESS scientific case, which was published in 1997 in connection with the feasibility study [6]. While the earlier report covered the full breath and width of neutron science, here we have emphasised high profile experiments or "flagship" scientific areas.

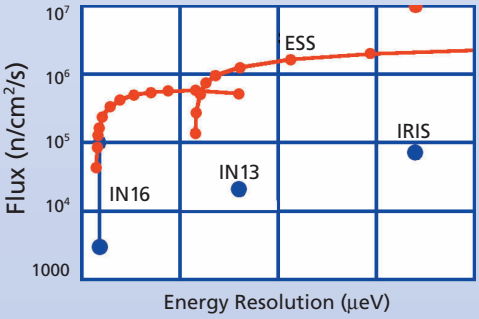
Chapter 2 of this document deals with the ENSA 'neutron landscape', representing the view of European users of

Instrumentation opportunities at ESS



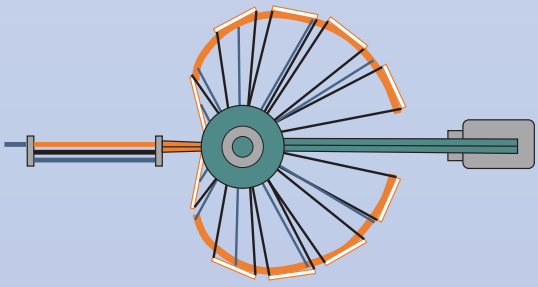
Chopper Spectrometers

Chopper spectrometers provide the ability to collect data over a wide range of energies. Chopper spectrometers at the ESS will cover the high, thermal and cold neutron energy regimes. Use of state-of-the-art arrays of choppers will provide unprecedented flexibility in the optimisation of flux and resolution which, coupled with the high neutron flux from the source, will yield gains in performance over existing sources of up to three orders of magnitude.



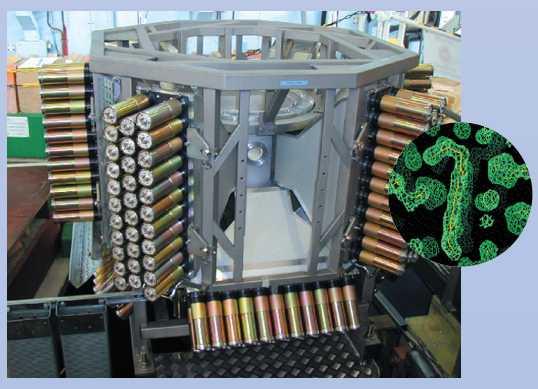
Backscattering Spectrometers

Backscattering spectrometers at the ESS will dramatically outperform existing, equivalent instruments in terms of both flux and range. Arrays of pulse shaping choppers, combined with coupled moderators will allow the tuning of resolution and range to suit specific experimental conditions.



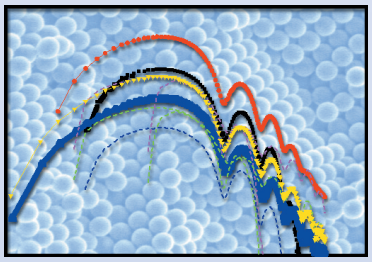
Diffraction for Physical and Chemical Crystallograhy

Powder and single crystal diffractometers at the ESS will combine the high source flux with novel instrumentation techniques to reduce counting times of as little as fractions of a second, allowing the real-time monitoring of reactions and the study of very small sample volumes.



Diffraction for the Life Sciences

The high neutron flux of the ESS will provide new opportunities for structural studies in the life sciences. Two instruments specifically designed for large molecule crystallography are included in the reference instrument suite.

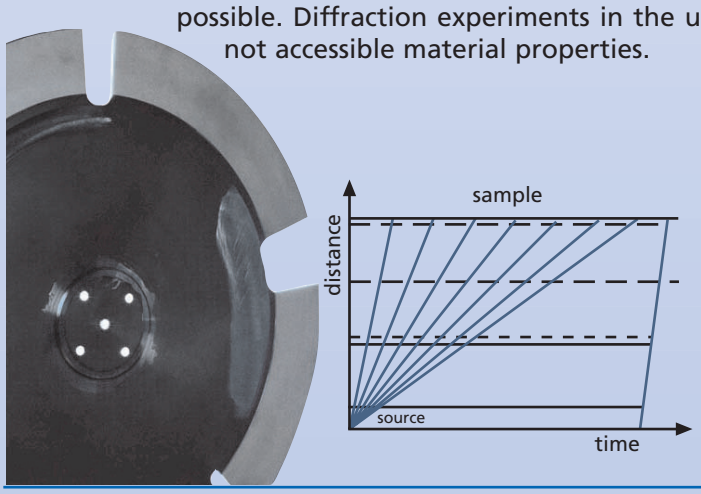


Quantum Leap in Performance

ESS will combine highest source power, innovative lay-out with two complementary target stations, most advanced beam extraction and beam delivery techniques and novel instrument design concepts. The cumulative results make ESS instrument performances up to more than three orders of magnitude superior to that of best to date counterparts. ESS will also largely out-perform other projected next generation spallation sources, for example in small angle scattering (SANS) by at least about an order of magnitude, due to its uniquely high power per pulse at the long pulse target station.

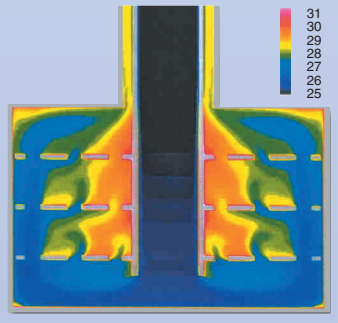
New Techniques for Unprecedented Resolutions

ESS target design opens up the way for using new tools to produce extremely short and sharp pulses to achieve hitherto not feasible resolutions. At the long pulse target station fast mechanical choppers can provide nearly an order of magnitude shorter cold and thermal neutron pulses than otherwise possible. Diffraction experiments in the ultra high resolution range of 10^{-5} will reveal currently not accessible material properties.



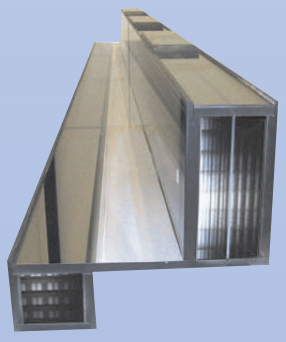
Enhanced Instrument Design Concepts

ESS instruments will take advantage of novel design concepts and techniques, which will allow them to utilize the high source flux with further enhanced efficiency. For example the repetition rate and wavelength frame multiplication methods make possible to extract, if needed, several pulses impinging on the sample from each source pulse, instead of just one. The combination of highest source power and innovative, improved instrument design is the key to make ESS an unprecedented leap forward.



New Quality Multi-spectral Beams

Conventional beam lines deliver optimal neutron spectra in a restricted neutron energy/wavelength range, for example either thermal or cold. A number of ESS beam lines will be equipped with a recently invented beam extraction system and simultaneously provide both thermal and cold neutron spectra. This will open up the way to do very broad dynamic range experiments of unprecedented quality in a single run.



More Efficient Beam Delivery

ESS beam lines will make extensive use of advanced neutron optical beam delivery systems using supermirror coated neutron guides of sophisticated design. Converging neutron guides penetrating the beam shutters will provide much enhanced short wavelength neutron beam intensities on the samples and low loss ballistic neutron guides will transport full thermal and cold beam intensities to 200 – 300 m distances to achieve by now unthinkable resolutions.

neutron facilities. European neutron sources have been classified in three tiers, with the ESS being seen as the centrepiece (top tier) for several decades.

Chapter 3 outlines instrumentation opportunities at ESS, explaining in particular novel approaches in the use of coupled moderators at the long and short pulse target stations. Neutron beam extraction and transport, together with various multiplexing techniques, will play a major role in the optimisation of neutron use at ESS. The instrumentation aspects of ESS were considered by nine expert groups, which met at workshops in Heathrow in February 2001 and in Grenoble in March 2002. A number of topical workshops on particular instrumentation issues were also organised. Initially the instrumentation groups inspected the performance of generic instrumentation at the different ESS target stations. In this way the potential of the source was evaluated. These instrument performances served as the basis for a realistic (conservative) assessment of scientific opportunities at the ESS. The instrument groups then concentrated on prioritised "day one" instrumentation, examining novel concepts in the exploitation of the different target stations. The basic concepts are discussed in *Chapter 3*, while the instrument layouts and performance descriptions are given in Volume IV.

Chapter 4 is devoted to the disciplinary scientific cases for the ESS. It is based on an analysis of future trends in the different scientific fields served by the ESS. We have identified those areas where the ESS will have a significant or decisive impact on scientific problems that cannot be solved today. These are the flagship areas that have already been highlighted. Likely future scientific developments were assessed by eight disciplinary science groups, convened by members of the ESS-SAC. This assessment of the scientific opportunities relied on the evaluation of the performance of generic instrumentation at the ESS. The chairman and several members of the European Neutron Scattering Association (ENSA) were involved in the different science groups.

Chapter 5 deals with the complementarity of neutron scattering and other techniques. This was considered by a group consisting both of neutron users and members with expertise in relevant complementary techniques (synchrotron radiation, NMR, light scattering etc.). The outcome of these considerations underlines earlier findings of the ESF [1] stating that "Synchrotron radiation techniques and radiation sources cannot replace neutron techniques and neutron sources … . Even in the long term both techniques and advanced radiation sources including instrumentation of both categories are indispensable to Europe's lead in science research and technical applications as the two techniques cannot replace each other (nor be replaced by third methods)".

Chapter 6 addresses the impact of ESS on the solution of more practical problems, which are targeted in the development of applied science and technology. Such

problems are the subject of priority research missions in the framework programmes of the EU, or of foresight themes promoted by different European governments. The ESS–SAC selected seven themes for detailed consideration:

- Microsystems and Information Technologies
- Functional Materials
- Health and Biotechnology
- Nanotechnologies
- Cultural Heritage: Artefacts and Materials
- Traffic and Transport
- Sustainable Development; Clean Technologies and Environmental Systems

Finally, in *chapter 7* we outline the inter-relations between the development of the scientific case, the instrument design and performance assessment, and the ESS design. The decision between the different target options was based on the development of the scientific case. The instrumentation opportunities and instrument priorities were set in the second step. These priorities in turn determined the optimisation of the target moderator complexes at the long and short pulse target stations.

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Chapter 2

The ESS in the Context of an Evolving European Neutron Landscape: ENSA's Twenty Year Perspective

2. The ESS in the Context of an Evolving European Neutron Landscape: ENSA's Twenty Year Perspective

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Abstract

The European Neutron Scattering Association has evaluated the infrastructural support for neutron scattering research within Europe over the next two decades. ENSA has concluded that the long term future of European neutron scattering, and the preservation of Europe's lead on the world stage in neutron scattering, and consequently its highly competitive position in condensed matter science and technology, can be secured only through the provision of a European Spallation Source operating as a flagship facility at the hub of a network of fully optimised regional neutron sources. ENSA's recommendations for an optimal neutron landscape, structured as a three tier hierarchy of neutron facilities, is presented and discussed here.

I. Introduction

Europe hosts the largest, most experienced and broadest-based community of neutron beam users.

Neutron scattering is a ubiquitous tool which plays a crucial infrastructural role in underpinning much of condensed matter science and technology within the disciplines of physics, materials science, chemistry, the life sciences, the earth sciences and engineering. Consequently there is little doubt that Europe can legitimately claim a significant strategic advantage in these fields of research. Not only does Europe boast the world's premier neutron scattering sources but also Europe hosts the largest, most experienced and broadest-based community of neutron beam users. Indeed almost 5000 neutron scatterers, over two thirds of the world's total number, reside in Europe and exploit European neutron facilities.

It is therefore tempting to conclude that European neutron scattering science is currently enjoying a "golden age". From a short-term perspective such a view is well justified: the European neutron scattering community can be proud of its achievements, and confident in its world lead. Unfortunately, a medium- to long-term perspective reveals that this lead is not unassailable.

Threats to the European lead in neutron science.

On the one hand Europe in particular faces the impending reality of the much discussed "neutron drought", originally forecast in a 1994 Analytical Report commissioned by the OECD Megascience Forum. This drought is a direct consequence of the continuing expansion of a multidisciplinary neutron scattering community alongside a progressive and inevitable closure of aging neutron research reactors.

On the other hand, a very serious challenge to European scientific and technical supremacy in the field of neutron scattering has been mounted both by the USA and Japan, both of whom are well advanced with their own plans to alleviate their local "neutron droughts" through major financial, scientific and technological investments in third generation advanced neutron spallation sources alongside a commitment to the simultaneous upgrading of their existing world class neutron facilities.

The action being taken by the USA and Japan is in full accord with the explicit recommendations of a 1998 OECD Megascience Forum report, subsequently endorsed by the OECD Ministerial Conference, that first class neutron sources should be upgraded, and that a new third generation MW spallation source should be constructed in each of the North American, Asian and European regions.

Meanwhile, Europe is also undertaking a continuing and vigorous programme of optimisation, development and increasing exploitation of existing second generation neutron beam facilities. However, whilst the USA and Japan will have completed their spallation source projects by 2006, Europe has still not made the political and financial commitment to construct its own third generation MW spallation source. It is clear that without such a commitment the centre of gravity for neutron scattering science of the very highest quality will have shifted from Europe by 2006. Ironically, this is precisely a decade after the European Science Foundation, in a strong endorsement of the European neutron scattering community, declared "*Finding the optimum means to address the continuing needs of this wide and strategically important R&D community group in both the medium and long term within the scientific-technical-strategic and economic opportunities in Europe should be a matter of the highest priority, ensuring the health of many fields of European R&TD and the European lead achieved in the past".*

USA and Japan will have completed their spallation source projects by 2006.

Against this background of a dramatically changing global neutron scattering scene the European Neutron Scattering Association (ENSA) has surveyed the current status and future potential of neutron scattering science and neutron facilities within Europe, with a view to establishing a coherent strategy through which the future of European neutron scattering research can be secured. In so doing, ENSA has adopted a twenty year perspective embodying the analysis and recommendations of the 1998 OECD Megascience Forum, whilst also embracing the philosophy of the EU Research Area Initiative, and in particular including the principles of open access to strategic research infrastructures. A summary of ENSA's twenty year perspective of the emerging European neutron landscape is presented here.

II. The European neutron landscape

The European neutron landscape in 2002.

Considering the European neutron landscape as it is spread before us at the beginning of the 21st Century, it is clear that the two most prominent landmarks are the Insititut Laue Langevin (ILL) in Grenoble and ISIS in Oxfordshire. These two sources, as the world's leading high flux reactor facility and the world's most powerful spallation neutron source respectively, have together set the standards for neutron scattering on a global scale.

Over the last thirty years ILL has established an excellent working model of multinational European collaboration in neutron scattering science. Similarly ISIS, although principally the UK's national facility, is partially supported by European-wide collaboration and funding. According to the 1998 ENSA survey of neutron facilities these complementary and world dominating neutron facilities together account for almost 40 % of all applications of neutron beam techniques to condensed matter science and technology within Europe.

Sited within the wider Europe landscape there are in addition a number of world class medium flux national neutron sources, together accounting for a further 40 % of neutron beam applications, whilst the remaining 20 % of neutron beam allocations are provided by several smaller national, (and even university owned), low flux neutron reactor sources.

All of these national neutron facilities have played crucial roles in the development of European neutron scattering and now they successfully meet specific requirements, several at a European level and some at a local level, for neutron scattering instrumentation, technique and software development, engineering standards, training and ease of access. Some also serve national strategic needs beyond neutron scattering science, for example in medical isotope production, radiography and material irradiation.

Many of these national medium and low flux sources have already developed, or are in the process of developing, extensive multinational collaborations, and many have also adopted policies of open international access, in some cases supported financially by EU programmes. Indeed, extensive co-operation and collaboration between all of the European neutron facilities has engendered a healthy exchange of new concepts in instrumentation, of highly trained and well-motivated personal, and of extensive scientific programmes.

The complementarity of neutron sources and instrumentation across the breadth of Europe has enabled a well informed and well supplied user community to choose those facilities most appropriate and optimally suited to the specific scientific problem in hand. There is no doubt that condensed matter science and technology of the very highest international standard has been, and still is, produced at each one of Europe's neutron facilities, from the smallest national or university research reactors to the most powerful multinational sources.

The European neutron landscape in 2022 and the strategic role and positioning of a third generation European MW Spallation.

However, moving forward twenty years from now we find a European neutron landscape that has changed considerably. It is within this changing, and indeed significantly eroding, landscape that the strategic role and positioning of a third generation European MW Spallation Source must be considered.

According to OECD predictions, in the absence of major upgrades many of Europe's medium and low flux neutron sources which at present are fully integrated features of the European neutron landscape will either have ceased operation or will be approaching the end of their projected lifespan. The national facilities which may be lost over the next twenty years include IRI Delft (The Netherlands), Studsvik (Sweden), Geesthacht and Jülich (Germany), LVR-Prague (Czech Republic), Kjeller (Norway), Swierk (Poland) and Moscow, Ekaterinburg and Gatchina (Russia). The sudden and unexpected closure of the important European neutron scattering centre, the DR3 reactor at Risø (Denmark), in the autumn of 2000 is testimony to the reality of impending neutron drought. DR3 was as much a loss to the wider European community as it was to the Danish neutron scattering community.

A network of regional sources as the first tier in the hierarchy of European neutron sources.

The medium flux facilities which are likely to remain operational for the next twenty years and beyond are BENSC-HMI (Berlin, Germany), IBR-II (Dubna, Russia), LLB (Saclay, France), BNC (Hungary) and SINQ (Villigen, Switzerland). Each of these facilities are, and will remain, powerful world class neutron sources with neutron scattering user communities and collaborations which extend across Europe. It is expected that they will soon be joined by FRM-II (Munich, Germany), a major new 20 MW reactor facility for which an operating license from the German government is eagerly awaited.

Together these five national neutron sources are ideally placed, both technically and geographically, to assume significant and complementary roles within an evolving integrated European-wide network of fully optimised regional facilities. Such a network should provide ease of access to high quality neutron beams and instrumentation. Moreover, it is conceivable that the internationally competitive Austron project, a 0.5-megawatt spallation neutron source planned by Austria, may also reach fruition, joining the European network of regional sources as an important Central Europe component.

Fully optimised ILL and ISIS as the second tier in the hierarchy.

If the network of regional sources is considered as the first tier of a hierarchy of European neutron scattering facilities, then ILL and ISIS together must constitute the second tier. There is little doubt that a suitably refurbished and optimised ILL will retain a dominant position as the most powerful reactor facility in the world, perhaps further enhancing its European dimension through an evolving structure of multinational partnerships. ISIS, though eclipsed in performance in many areas of application by SNS and JSNS, will also remain an extremely powerful European facility, maintaining a major presence on the world stage, aided in particular by the anticipated investment in a second target station, ISIS-II, together with an associated suite of advanced neutron instruments.

In a global context the emerging European neutron landscape is therefore rather bleak. At very best it represents a baseline scenario which will result in an entirely unacceptable decrease in the available neutron instrumentation suite and a corresponding erosion of the breadth and capability of neutron scattering and the cutting edge European condensed matter science and technology which it both drives and supports. It is inevitable that the most challenging problems in condensed matter will be addressed not in Europe, but in the US and Japan where the advanced third generation spallation sources will be operational. It is to these new centres of neutron excellence that Europe's highly trained and energetic neutron scientists will undoubtedly migrate.

The emerging European neutron landscape is rather bleak.

The European Neutron Scattering Association has recognised that in both the quantity and quality of its neutron beam provision the two tier hierarchy of surviving European neutron facilities will, in isolation, fall far short of the increasingly stringent demands and expectations of the European condensed matter science base. Not only will the European lead in neutron scattering be lost, but European competitiveness in the field will also be seriously challenged. Consequently the eighteen national delegates to ENSA have unanimously and emphatically endorsed and embraced the European Spallation Source Project as the only realistic solution for securing the future of a key European scientific and technological strength.

A two tier hierarchy alone cannot meet the demands of the European condensed matter science base.

The need for ESS as the essential third tier of the hierarchy is unanimously endorsed by the eighteen national delegates to ENSA.

ENSA views the European Spallation Source as a truly multinational flagship facility at the hub of a powerful and mutually supportive network of regional neutron sources. As the third and uppermost tier of the hierarchy of neutron facilities ESS will provide Europe with neutron beams and advanced instrumentation of unrivalled quality.

ESS will provide Europe with neutron beams and advanced instrumentation of unrivalled quality.

The role of the ESS in condensed matter science and technology will in many respects be analogous to that of the Hubble telescope in astronomy. The Hubble is changing our perception of the "outer universe", enabling us to see deeper and with greater clarity than ever before, elucidating phenomena that were previously at the limits of detection and revealing new phenomena beyond those limits. The ESS will similarly facilitate neutron scattering studies that will change our perception of the "inner universe", revealing new scientific phenomena and technological functionality through the deeper characterisation of the structural and dynamical properties of matter across all of the scientific disciplines.

ESS as the Hubble telescope of condensed matter science and technology.

Moreover, just as the Hubble telescope functions most efficiently as part of a network of less powerful ground-based observatories, so the efficiency of the ESS will be enhanced by the supporting two tier structure of European neutron facilities, each member of which will continue to play a crucial role in front rank studies of phenomena for which the power of ESS is not essential. Just as at present, the appropriate

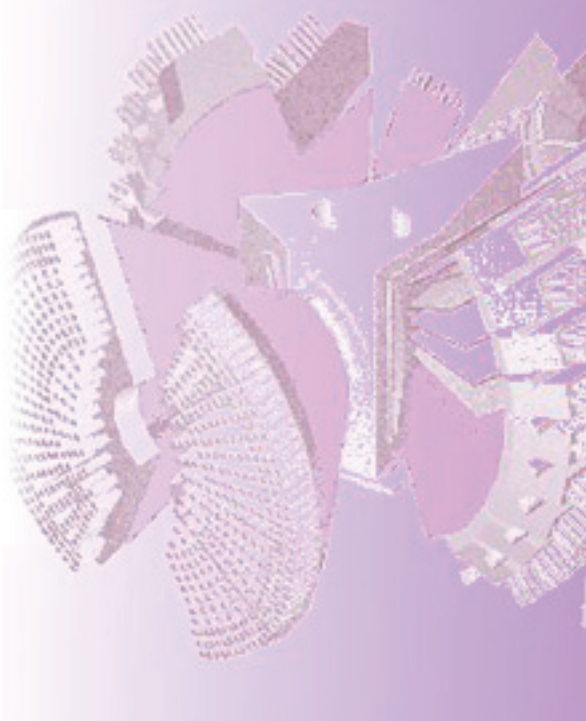
neutron source will be chosen according the particular demands and complexity of a specific problem.

In summary therefore, ENSA, taking a twenty year perspective, considers that the only viable European Neutron landscape is that which embodies a three tier hierarchy of neutron facilities. As the first tier, BENSC-HMI, IBR-II, LLB, SINQ, BNC, FRM-II and possibly Austron, operating as a network of regional facilities, will provide ease of access at both national and regional levels whilst also helping to satisfy the growing demand for neutron beams at a European level. ILL and ISIS, the current European flagship facilities, as the second tier of the hierarchy, will provide internationally competitive and high quality neutron beams with which problems at and just beyond the threshold of current capabilities will be tackled. However it will be the ESS as the new flagship source, and the uppermost tier of the hierarchy, that will open the new and exciting vistas in condensed matter science and technology across all disciplines.

ENSA recommends the adoption of the proposed three tier hierarchy to secure the future of neutron scattering science and technology within Europe.

Chapter 3

Instrumentation Concepts: Advances by Innovation and Building on Experience



3. Instrumentation Concepts: Advances by Innovation and Building on Experience

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Abstract
Tremendous progress in neutron scattering research capabilities has been achieved over the past half century primarily by the development of the performance of the instrumentation, both in terms of new concepts and approaches and advances in components, such as detectors. Over the same period of time the brightness of neutron sources only increased by a modest amount. ESS will offer an unprecedented jump of some two orders of magnitude in a crucial source performance parameter, the instantaneous peak flux during the pulses compared to the leading continuous or pulsed neutron sources existing today, while in terms of time average flux it will equal the most powerful continuous sources. The pulsed character allows for a more efficient use of the total number of neutrons produced, and this efficiency differs from one application to another. The goal of the ESS project is to combine the vastly enhanced, unique source quality with the most advanced instrumentation concepts and techniques to achieve a quantum leap in neutron scattering research opportunities well beyond what could be achieved by concentrating only on enhancing the source performance or only trying to further develop instrumentation. The results of the combined effort as characterised by the sensitivity of observing small signals (which is the main limitation in the use of neutron scattering techniques in general) are found to amount to as much as three orders of magnitude in some unique core applications and more than two orders of magnitude in the majority of neutron scattering work. To achieve this huge step forward, well comparable to the progress expected from the realisation of large free electron lasers in X-ray research, we need to vigorously advance neutron scattering instrumentation techniques together with the source performance. Extensive experience accumulated over the past 5 decades of using continuous reactor sources and nearly 3 decades of progress with pulsed spallation sources provides a solid and sophisticated basis for this effort. The utilisation of innovative concepts will further enhance the efficiency of using source power in the actual experiments. Examples discussed below include enhancement of the efficiency of extracting and transporting neutrons from the source to the sample by advanced neutron optical means and sophisticated, so called multiplexing, techniques which allow us to optimise the efficiency gains by the pulsed character of the source simultaneously for a large number of instruments with very different characteristics. As a result ESS will not only surpass all other neutron sources, existing or being built by its higher neutron brightness achieved by more proton beam energy per pulse, but it will further enhance this advantage by a more efficient use of the neutrons produced.

I. Introduction

Neutron production power grew little in the last 4 decades.

Since the groundbreaking work of B. Brockhouse at Chalk River in the 1950's the thermal flux performance of neutron sources only progressed by now by a mere factor of 4. Actually all this progress was accomplished a long time ago, by 1972 with the commissioning of ILL. The obvious and huge progress since Brockhouse's time is thus not primarily due to advances in source performance in terms of the number of neutrons produced, but to developing ways of using these neutrons more efficiently. This has been achieved by the evolution of neutron scattering instrumentation techniques and the development of specific neutron moderators, the hot and cold sources, in order to enhance the neutron flux at energies above and below the thermal energy range. As the next decisive step in the evolution of neutron sources, ESS will offer a quantum leap in neutron science. It will provide for an enhancement in source performance for the different applications by factors between 10 and 100 compared to the best available today at any existing neutron source of any kind. This progress is a lot more than it has been achieved by now since the pioneering days of Brockhouse. It will essentially be accomplished by improving the efficiency of use, and not primarily by producing more neutrons. Indeed the time average flux of ESS will be comparable to that of ILL, and its pulsed character will allow us to make use of these

Spectacular progress was achieved by advances in instrumentation.

ESS goal: quantum leap in performance compared to all existing neutron sources …

neutrons in a 10 to 100 times more efficient way.

This goal of the ESS project, to provide at least about an order of magnitude enhanced neutron beam performance compared to any existing source, however, cannot be achieved with existing and established techniques. Indeed, on some reactor instruments up to 15 % of the neutron spectrum hits the sample, so a 30 fold increase of the power compared to ISIS alone would be just enough at best to break even with ILL for these kinds of instruments. For ESS we also need to develop new, more efficient approaches both in neutron production and moderation and in instrumentation. Actually these two aspects are closely related, innovation in source performance will call for new instrumentation concepts and new instrument design approaches will allow us to better use the potentials of the source, also by relaxing some design requirements for the target / moderator system, which are hard to meet or unduly detrimental to neutron intensity.

… it can be achieved by combining enhanced power with new approaches for enhanced efficiency.

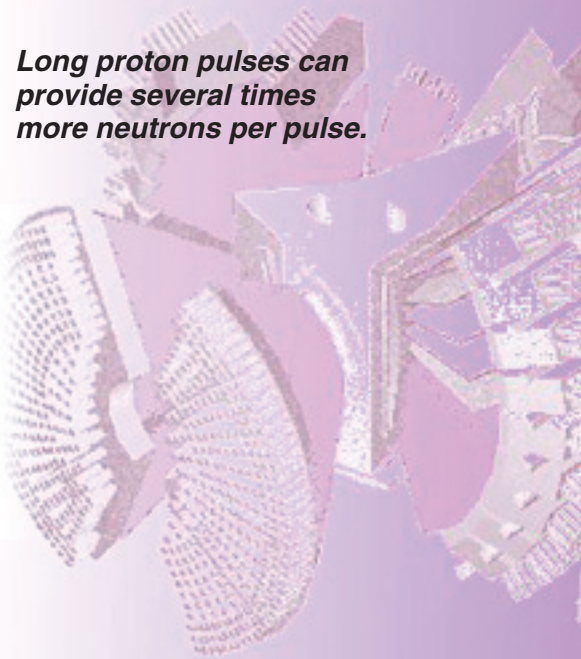
As in the past, a key feature of enhancing the efficiency of neutron sources is to improve the neutron moderators. Current spallation neutron instruments face neutron moderators optimised for producing short neutron pulses by limiting the time allowed for neutron thermalization with the help of neutron absorbers placed around and/or inside the moderators. The enhanced efficiency of neutron use at ESS will also have to include implementing more efficient, so called coupled moderators. The first two of this kind have recently been installed at Lujan Center in Los Alamos, now making this source about 2 times as bright as ISIS at half the accelerator beam power. The higher moderation efficiency of these moderators is accompanied by longer moderation times, with significant neutron intensities emitted for up to 3 – 4 ms after the beginning of the pulse. As of today, there is no experience available with the use of such long pulses.

New type of moderator produces more thermalized neutrons in longer pulses.

Another option to enhance neutron production efficiency, in particular in view of the long moderation times in coupled moderators, is to avoid the compression of the ms long linear accelerator pulses to μs length short pulses by proton storage rings. The ESS design goal of 100 kJ total proton energy per short pulse is certainly at the technological limit both in ring accelerator design and material strength. By making economy of the pulse compression rings, up to some 500 kJ beam energy per pulse becomes feasible in 2 – 3 ms long linac pulses. The ESS long pulse target station will receive 300 kJ energy in 2 ms long pulses. Again, as of today, we have no practical experience with the production and use of such long pulses.

Long proton pulses can provide several times more neutrons per pulse.

The great achievements of spallation sources by now, in particular ISIS, offers a solid base for planning ESS. On the other hand, to achieve ESS goals we also need to complement the established techniques by novel approaches for more efficient neutron moderation and production, such as coupled moderators and long pulses. It turns out, that the powerful instrumentation techniques developed at steady state



reactor sources provide a number of technical opportunities for the use of these options.

First ESS instrument performance evaluation was based on established technology. Enhanced approaches keep emerging.

By now one of the main goals of the Instrumentation Task Group was to assess expected instrument performance on ESS. The results are contained in the reports of the 9 instrument groups [1]. In order to be on the safe side, this task was deliberately based on prudent extrapolation from established approaches and conservative estimates of the performance of the novel kind of moderators, namely the coupled ones and those on the long pulse target station. Nevertheless, it is fair to think that in nearly 10 years from now innovations and new ideas to emerge in the meantime will also play a major role for the instruments that will really be built.

This first instrument performance evaluation served as input data for establishing the scientific potentials of ESS and to help to decide the basic ESS parameters on the basis of the scientific case, the expectations of the broad scientific community of users. This effort led in the first place to the definition of the accelerator parameters and the dual target station, 50 Hz, 5 MW short pulse and $16^2/_3$ Hz, 5 MW long pulse general layout. While deliberately staying on the conservative side when evaluating the expected performance of ESS experimental facilities, it is a central goal of the ESS project team to systematically explore all possibilities to further enhance this performance by the use of novel and more efficient approaches. It also includes the optimisation of moderator design and layout, to devise most efficient beam extraction and beam delivery system to transmit neutrons from the source moderators to the samples, to conceive instrument design concepts that allow the instruments to take best advantage of the crucial source parameters, such as pulse length and pulse repetition rate and to develop enhanced performance, more efficient and more economic instrumentation tools and components. The goal of this report is to describe a number of novel general instrumentation concepts and paradigms that open ways for ESS to set new standards in neutron scattering research not only by its higher accelerator power but also by the enhanced efficiency of turning this power into scientific opportunities at the instruments.

II. Moderator performances

Current ESS reference moderator performance data are based on previous project evaluations.

The instrument assessment effort of the Instrumentation Task was based on a set of assumed moderator parameters, which have been established in collaboration with the Target and Moderator Task Group in December 2000 [2]. In what follows we will refer to this moderator data base as "Dec. 2000" compilation. At that time no detailed neutronics calculations of the ESS target-moderator system were available, and the compilation was based on calculations made at SNS, Los Alamos and in Japan, and also on performance estimates of existing sources. The most complete and most recent of these calculations, those made at SNS, were given the most weight.

Neutronics calculation studies of a variety target configurations by the ESS Target and Moderator Task Group have shown in the meantime, that these "Dec. 2000" estimates are rather close to what can be ultimately expected from moderator systems with proven feasibility (within less than 50 %, both in terms of neutron intensities and pulse lengths). On the basis of this additional validation, these moderator parameters are being continued to be used as ESS reference until the end of 2003, when the final ESS moderator performance calculations will become available, which will take into account all engineering details of the target / moderator system as it will be built. The process of the definition of the final target / moderator system design is pursued in close collaboration between the Target and Moderator and Instrumentation tasks.

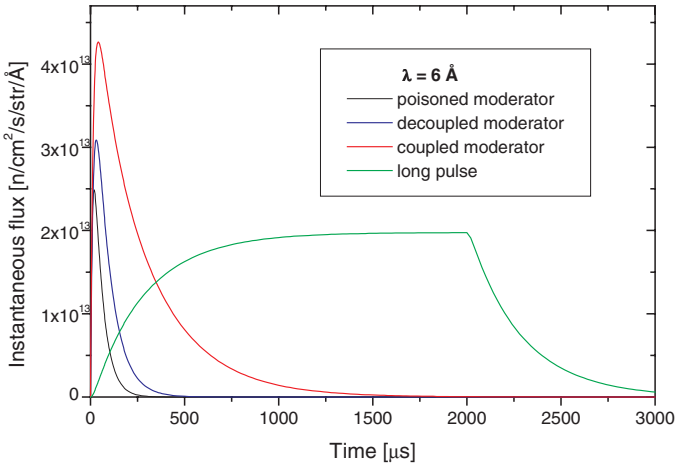


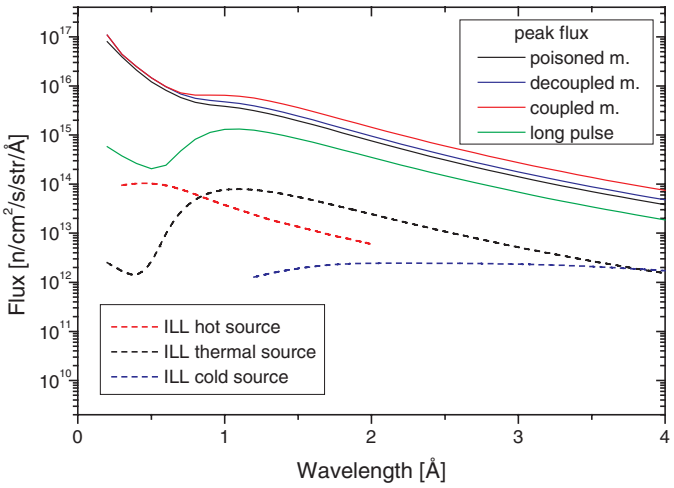
Figure 1: Example of ESS reference neutron pulses in the beam lines for the various cold (liquid H_2) moderators considered. [2]. For the three short pulses the proton pulse length is negligible (< 2 μs). The steady state flux of ILL is 6.10^{11} in the same units and at the same wavelength. The proton beam energy is assumed to 100 kJ per short pulse and 300 kJ per long pulse.

For short proton pulses three different types of moderators are envisaged (poisoned de-coupled, de-coupled and coupled), which provide different neutron pulse lengths (Figure 1). For ms long proton pulses the neutron and proton pulse lengths are effectively the same (not the pulse shape though) and therefore only the brightest (coupled) moderator needs to be considered.

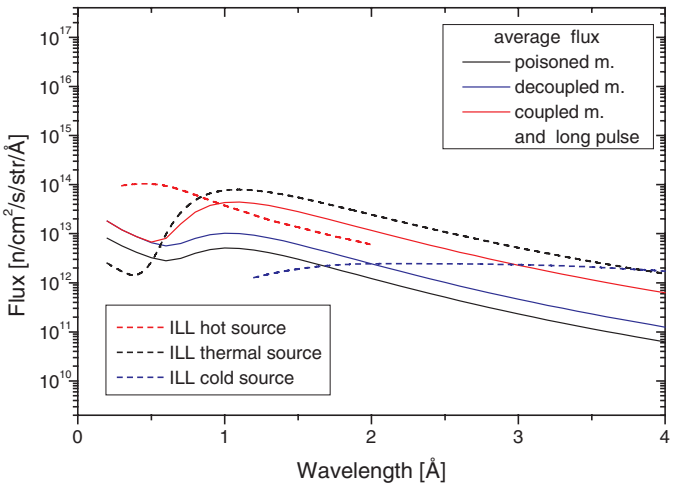
One important difference between the current ESS reference design and SNS is that ESS assumes the use of Pb reflector around the target and moderators and SNS a composite reflector, consisting of Be in the inside and Ni or Fe outside. Pure Pb reflector provides for higher time average flux from all moderators, however the pulse lengths, primarily those of the thermal moderators also become larger. The final decision on the ESS choice of reflector therefore will have to be made after careful consideration of the advantages and

Optimal target/moderator design options for ESS are being evaluated in close collaboration between the Target and Moderator and Instrumentation teams.

disadvantages for the reference suite instruments on both target stations.



The peak instantaneous source brightness of ESS will be about two orders of magnitude higher than that of ILL and ISIS.



The time average brightness of the coupled moderators at ESS will be comparable to that of ILL.

Figure 2: Peak (top) and time average (bottom) flux of reference ESS thermal neutron (ambient water) moderators as a function of wavelength [2] compared to the steady state flux of the various ILL moderators [3]. For the short pulses 5 MW time average proton beam power is assumed at 50 Hz pulse frequency and for the long pulse the same 5 MW power at $16^2/_3$ Hz.

Comparison to existing sources, namely to ISIS and ILL was a basic goal of the performance evaluation by the Instrument Task Group. Concerning ISIS, this is based on the assumption that the ESS de-coupled moderators will provide 30 times the flux at similar line shape to the corresponding ISIS decoupled moderators. ISIS does not have coupled moderators (which are now foreseen for a projected second target station at ISIS), so the time average flux of the brightest (coupled) ESS moderators is expected to amount to about 120 times that of the ISIS un-poisoned moderators today, at 2-3 times longer pulse lengths. A comparison to ILL is part of the “Dec. 2000”

compilation. It is based on the flux data published in the ILL "yellow book" [3], which refer to neutron fluxes measured in the various ILL beam tubes and guides at considerable distance from the reactor core. Due to the Liouville theorem the flux is independent of the position it is measured at, as long as there is no beam attenuation due to interaction with materials, such as beam windows or neutron mirrors. The time average neutron flux at 5 MW ESS power was estimated in "Dec. 2000" to be equal to that of ILL for coupled cold (liquid H_2) moderators and about 50 % that of ILL for coupled thermal (ambient H_20) moderators (Figure 2). In contrast, the peak flux of the ESS poisoned moderators will achieve 30 – 60 times the steady state flux of ILL, and that of the coupled ones some 60 – 120 times, depending on neutron wavelength. For the various experimental applications, the effective useful flux of ESS compared to a continuous source will thus range between the ratios of the average and the peak fluxes, i.e. from 0.5 to 120 in the case of comparing to ILL.

New design options will be considered to enhance the ESS time average hot neutron flux.

The hot (epithermal) neutron flux is a special case. While in this, under-moderated regime the peak flux of ESS is more than 100 times superior to the steady state flux at ILL, the time average flux is some 10 times less. The former assures great performance for pulsed sources when good hot neutron wavelength resolution is required, e.g. in powder diffraction. In contrast, the latter only offers modest capabilities in single peak analysis on single crystals in such trademark neutron polarisation analysis work (e.g. CRYOPAD). Implementing a hot moderator on ESS would alleviate this deficiency, however, the alternative suggestion of installing a beam-line viewing directly the Pb reflector, rather than a moderator, appears to be a much superior (and by now untried) solution [4], which needs to be explored by neutronics calculations in the near future.

The number of moderators at a target station is an important design parameter.

The moderator performances of "Dec. 2000" assume that each moderator is located at the most favourable position next to the target. This position is well defined in space, in particular within some 5 cm in the direction of the proton beam hitting the target. Therefore only two moderators on a target station can occupy the brightest position, one below and one above the target. Placing 4 moderators on a target station necessarily reduces the flux of all 4 moderators by amounts between approximately 10 – 40 %. It is a design choice how the reduction is distributed between two moderators behind each other, e.g. trying to maximise the flux of one of them (usually the upstream one with respect to the proton beam direction, at the expense of the downstream one). Furthermore, the performance of moderators on the long pulse target station was assessed in "Dec. 2000" by assuming the same target-moderator design deemed optimal for short pulses. Thus the long pulse moderator flux is certainly underestimated, possibly by as much as a factor of 1.5 or more. By now the thickness of the H_2 moderator has been identified as a parameter to be further optimised.

Planned optimisation of the long pulse target station is expected to bring substantial further gains.

Another conservative assumption was made in "Dec. 2000" in

the absence of methane or similar moderators. Liquid H_2 has a rather low proton density per cm^3, and it is therefore rather inefficient as de-coupled or poisoned de-coupled moderator, where the thickness of the moderator is limited to less than 5 cm in order to keep small the contribution of neutron flight path uncertainty to the pulse length. Potential flux gains for decoupled and poisoned moderators by methane or similar could eventually reach a factor of 2 (while no essential gain is expected for coupled moderators). However, radiation at ESS levels rapidly destroys methane, and this is a tremendous, unsolved technical obstacle. Thus as of today methane or similar moderators cannot be envisaged as feasible options. This might, however, change in the future, e.g. by the development of techniques to make solid methane pellets circulated by a cryogenic fluid.

If difficult feasibility issues can be solved, methane or similar moderators will become a great option for decoupled moderators.

III. Enhanced beam extraction and beam delivery

Current practice at pulsed spallation sources for delivering the highest flux of hot and thermal neutrons with wavelengths $\lambda < 2$ Å is to just make the sample to have a direct, unobstructed view of the moderator. From the closest reasonable moderator to sample distance on ESS, some 12 m, this will only provide 0.6° delivered beam divergence capability both horizontally and vertically. In comparison, modern curved, focussing crystal monochromator systems developed for steady state sources, can efficiently deliver 2 - 10° incoming beam divergences to the sample. While crystal analysers are very advantageously used in inverse geometry time-of-flight (TOF) spectrometers, crystal monochromators are in general not adequate for pulsed source instruments.

Making use of direct view beam delivery means that the neutron flux on the sample will decrease with the distance L between moderator and sample as L^{-2}. Therefore, in order to achieve a certain incoming beam velocity resolution, one currently tries to use the shortest L compatible with the desired resolution. For the choice of moderator this implies to prefer the one with the shortest feasible pulse length and therefore the lowest brightness (Figures 1 and 2). This doctrine changes radically by the introduction of supermirror coated neutron guides for the extraction and delivery of incoming neutrons energies up to 900 meV (or down to wavelengths as short as 0.3 Å).

Converging supermirror guides allow us to transport short wavelength neutron beams to larger distances ...

The current upper limit of beam divergence delivered by supermirror guides in both horizontal and vertical directions is practically set by the performance of the best (commercially) available supermirrors to

$$\delta\alpha \cong 0.6^\circ \times \lambda \text{ [Å]} \qquad (1)$$

i.e. to 2 times the supermirror cut-off angle in both directions. This means state of the art supermirror guides can deliver higher flux on the sample than the direct view of the moderators for all wavelengths $\lambda > 1$ Å. Adequately designed

... and to take advantage of the brighter neutron moderators without loss of resolution.

converging guides make sure that the guide performs at least as well as the direct view for shorter wavelengths too and in addition there is some additional gain compared to eq. (1) for distances L < 30 m [5]. The real revolutionary feature of this kind of beam delivery is, however, that the beam intensity now becomes very little dependent on the source to sample distance L. This opens up two new design options for advanced instruments:

a) Achieve higher resolution without much intensity loss by keeping the same moderator and making L longer.

b) Enhance the beam intensity on the sample at equal resolution by switching to a longer pulse, higher brilliance moderator.

This is later illustrated in Figure 3.

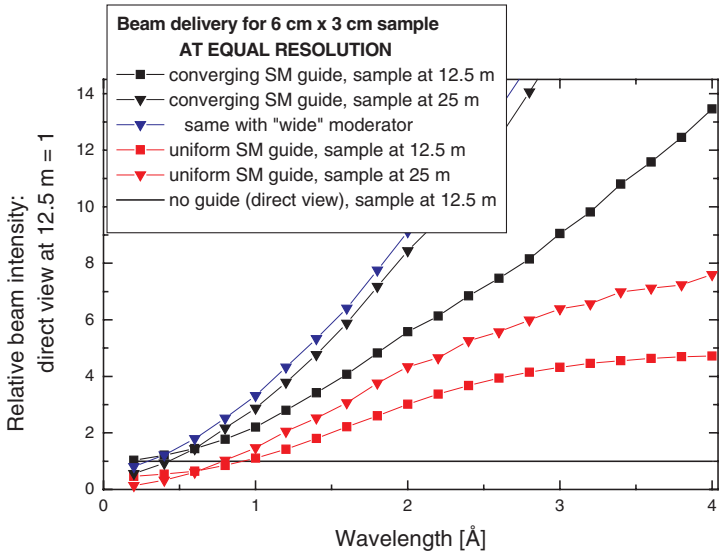


Figure 3: Beam extraction and delivery options for thermal and hot neutrons. At L = 25 m moderator to sample distance an unpoisoned, decoupled moderator offers at least equal resolution compared to its poisoned counterpart assumed for the L = 12.5 m options. In addition, the larger distance allows us to use a "wide" moderator (here 20 cm width instead of the standard 12 cm) without loss in angular resolution. The main advantage of the converging guides compared to the straight ones is that they do not obscure the direct view of the moderator, a crucial feature at short wavelengths ("shadow effect", Ref. [5]).The converging guides start at 2 m from the moderator, the straight ones at 5 m (in order to limit the shadow effect). Results of simulation calculations assuming boron as moderator poison and decoupler.

The larger source to sample distance can also improve the angular resolution of the incoming beam, and consequently the corresponding wavenumber resolution $\delta q_\perp$. For direct view $\delta q_\perp$ is inversely proportional to the wavelength, while for long enough usual uniform guides it is independent of the wavelength and distance, and corresponds to that for direct view at L = 12 m and λ = 1 Å. For converging guides $\delta q_\perp$

equals to that for direct view at the shortest wavelengths and gradually drops to that of the uniform guide towards long wavelengths. For a converging guide the enhanced L can, alternatively, taken advantage of for further improvement of the flux on the sample at short wavelengths by enhancing the moderator surface in the direction parallel to the target surface (which at ESS corresponds to the horizontal dimension). The "wide" moderator option in Figure 3 shows this potential, which is being investigated by the ESS Target and Moderator team from the point of view of neutronics performance. Here it is tentatively assumed that a 20 cm wide moderator will maintain 90 % of the average surface brilliance of the 12 cm wide moderator. Thus we arrive at the conclusion, that compared to the currently common "direct view of poisoned moderator" approach for short wavelength work the supermirror guide beam extraction combined with more brilliant, longer pulse, larger surface moderators offers higher or much higher flux on the sample without loss of resolution in wavelength or $q_{\perp}$ for all wavelength > 0.3 Å!

This novel beam extraction concept can provide substantial gains in neutron flux on the sample for all wavelengths > 0.3 Å.

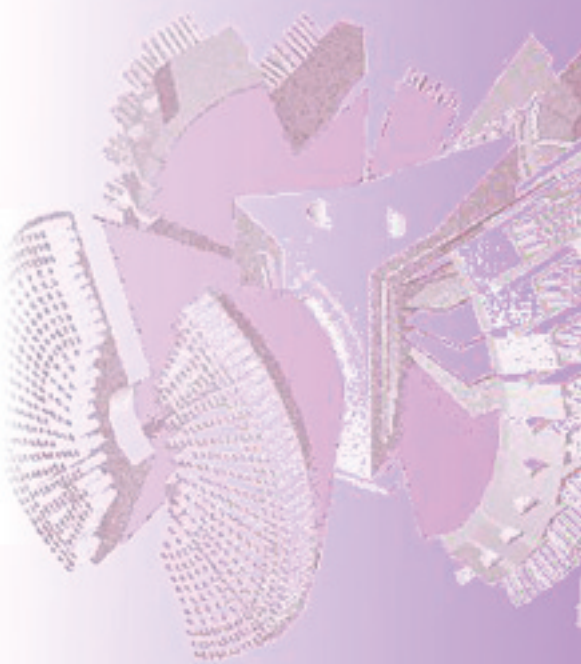
The enhanced beam delivery capability of simple supermirror neutron guides for cold neutrons and over large distances is mitigated by the reflection losses due to the increasing number of reflections. However, more sophisticated guide design approaches, primarily the so called ballistic guide concept (see e.g. in [5]) now allows us to transport beams with divergences up to the limit given by eq. (1) over distances up to several hundred meters with moderate losses in the range of 30 % or less.

Neutrons with wavelengths > 1 Å can be transported with little losses over 200 – 300 m by advanced neutron guides.

In sum, the utilisation of supermirror coated guides turns around one of the key instrument design paradigms for pulsed sources. Compared to the current "use the shortest possible moderator pulse to keep the moderator–sample distance as short as possible" approach the "use the highest possible flux (hence longer pulse) moderator and freely optimise the source–sample distance" concept can offer superior instrument performance for all neutron wavelengths above typically 0.3 Å, i.e. practically for all neutron scattering applications with the exception of eV spectroscopy.

The source – sample distance now becomes a free parameter in instrument design optimisations.

Supermirrors also offer another unprecedented new opportunity in beam extraction, actually both for pulsed and continuous sources, which has only recently been put forward [6]. By now neutron beam-lines deliver a given spectrum of neutrons depending on the moderator (at reactors also called "source") temperature: namely "cold", "thermal" and "hot" neutrons. The new concept, multi-spectral beam extraction aims at combining at least two of these spectra in a single beam-line, primarily – in view of the preceding considerations – in a single neutron guide. This will offer decisive advantages in many experiments which require a broad incoming wavelength band, and could eliminate the need of performing two separate experiments on two different instruments. Arguably multi-spectral beam extraction will double the utility of some beam-lines and instruments. Protein crystallography can be invoked as just one example, where the most useful



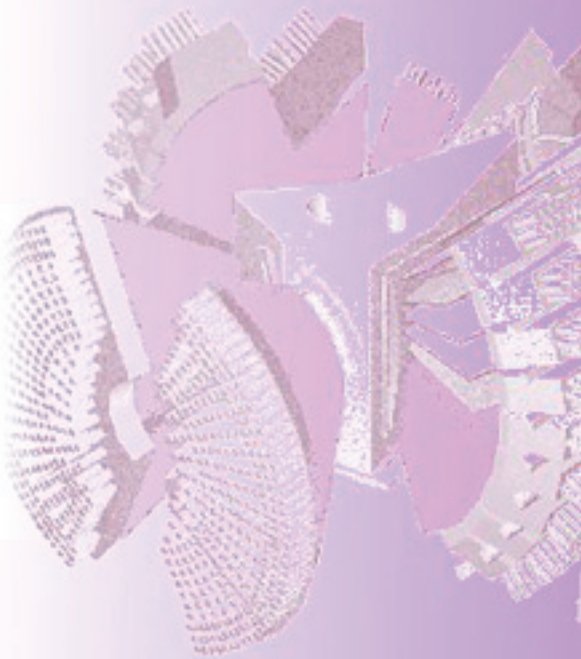
neutron wavelength range extends from 1.5 to 4 Å, which range is not ideally covered by either a cold or a thermal moderator.

It has to be mentioned that the design and performance of a so called "composite" H_2 – H_2O moderator has been investigated at SNS, the results were, however, found not favourable and the option has been abandoned. One problem to start with is that composite moderators perform with reduced efficiency around the centre of gravity of each components. The "cool" (e.g. about 100 K methane) moderators can well fill the gap between the thermal and cold spectra, while displaying a much narrower band of optimal flux than multi-spectral beams (and their feasibility at MW power levels is not yet demonstrated).

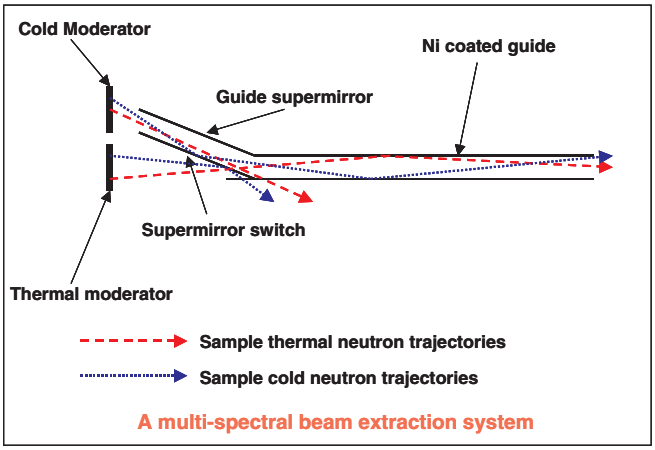
Supermirror neutron optics also allows us to combine the advantages of cold and thermal neutron spectra within a single beam.

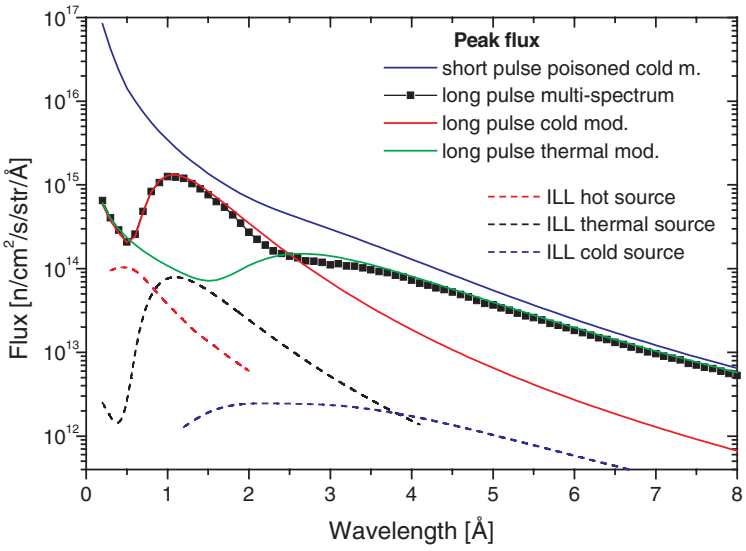
One proposed realisation of multi-spectral beam extraction for a spallation source is based on using a (stationary) supermirror plate deposited on a thin, transparent Si substrate, as illustrated in Figure 4. This supermirror plate acts as a wavelength dependent switch between the two moderators placed side by side. Such a thermal – cold moderator pair can be readily implemented on a pulsed spallation source such as ESS. Indeed, following the pioneering work of Watanabe et. al, the best design option for H_2 moderators involves surrounding the H_2 can by a bulky water premoderator. For multi-spectral beam extraction this premoderator will have to be made larger on one side of the cold moderator and the ensemble has to be placed in a manner that the extended premoderator surface can also be viewed through the beam ports. The total width of this pair of moderators will have to be about 20 – 22 cm for ideal performance. The bottom part of the figure shows the simulated neutron spectrum for the ESS long pulse target station in a large cross section (6 cm x 9 cm) Ni guide following the multi-spectral beam extraction optics. The beam at the end of this guide can be compressed, if needed, by a converging supermirror coated guide "funnel" into a $2\ \text{cm} \times 3\ \text{cm}$ maximum divergence beam (eq. (1)) at the sample at any distance $L > 20$ m. It is also feasible to use converging guide geometries over the whole multi-spectral beam extraction system.

Note that in this example for $\lambda < 1.5$ Å the neutrons come from the thermal moderator only, for $\lambda > 4$ Å predominantly from the cold moderator and from both moderators in-between. In this intermediate range of "mixing" the angular distribution of the fraction of the beam coming from one or the other moderator is strongly non-uniform, the total beam with contributions of both moderators mixed together is, however, as uniform as usual in a guide, both spatially across the cross section and in its angular distribution. This uniformity is important for the experiments to use multi-spectral beams, and actually it can be shown that it is a direct consequence of the Liouville theorem. In fact, we have to do here with one of the by now numerous examples of experimental techniques in particle physics which misleadingly appear to be forbidden just by the



Liouville theorem. Indeed, a neutron at a given position with a given wavelength and direction can only come from one or the other moderator (except for a beam partition by partially reflecting, partially transmitting optical elements). However, an extended volume of the phase space of particle parameters can be uniformly filled by several sources in a mosaic like fashion. These kinds of approaches are fundamental in modern accelerator design, just to mention one example.





Multi-spectral beams open up new experimental opportunities by efficiently using very broad wavelength bands.

Figure 4: The principle of multi-spectral beam extraction based on a supermirror spectral switch (top) and peak flux obtained by such a beam extraction system at the ESS long pulse target station in the Ni coated neutron guide (black squares) as compared to other ESS and ILL moderators. In the simulation calculation commercial quality "m = 3" supermirrors and 0.5 mm thick Si substrate was assumed. The length of the supermirror switch is 6.5 m, it starts at 1.5 m from the moderators, the Ni guide is 6 cm wide.

IV. Pulsed neutron beams: big advantages with tough strings attached

The overwhelming reason to prefer pulsed sources to continuous ones for highest performance is the vastly enhanced efficiency of making use of the produced amount of

Pulsed sources allow for much more efficient use of the neutrons produced ...

neutrons in the vast majority of neutron scattering experiments (while the pulsed structure is virtually of no advantage for irradiation work). On a continuous source beam monochromatisation is synonymous to throwing away all those neutrons in the Maxwellian source spectrum which do not possess the required velocity with the required precision, which in the majority of the cases amounts to only making use of 1-3 % of the spectrum. On pulsed sources the neutrons with different velocities arrive at different times to the instrument, so they can be distinguished from each other and consequently used at the same time. This allows us to utilise a large fraction, up to 60 – 90 % of the spectrum. This tremendous gain in efficiency, as most good things, does not come free. The pulsed nature of the beam imposes a whole set of boundary conditions, which lead to substantial compromises in instrument design options and can partially (or in rare cases even completely) erode the very gain in efficiency one seeks to achieve. As we plan to make ESS to benefit all areas of neutron scattering science, we will face new challenges to minimise this erosion of performance in a number of new situations. Some key approaches to achieve this will be considered in the next chapter, after having reviewed here the detrimental boundary conditions in need of remedy.

… but they also impose tough accelerator, target and instrument design compromises to be addressed.

a) Repetition rate

At a given time average power the repetition rate of the pulsed source is a parameter largely neutral to the ultimate source performance – unless it is too high. Indeed, if there is not enough time T left between pulses to allow the full desired wavelength band $\Delta\lambda$ to be accepted at the desired moderator to sample (or detector) distance L, i.e. the

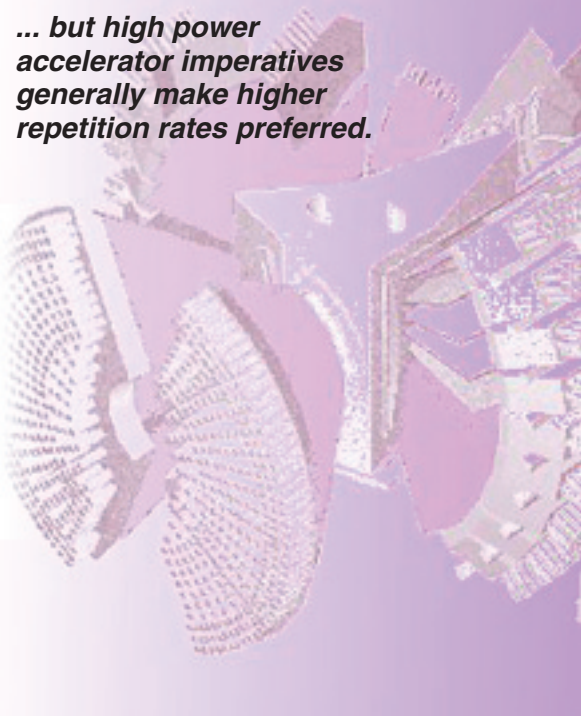
For many instruments pulse repetition rates below ~20 Hz only offer full efficiency ...

$$T[ms] > L[m] \times \Delta\lambda[Å] / 3.96 \qquad (2)$$

relation is not fulfilled, the source brightness is not fully taken advantage of. Thus a low repetition rate, practically something like 20 Hz or less is the guarantee for efficient use of a pulsed source in most applications.

It has to be emphasised that this consideration is only valid if the total proton beam power of the source can be kept constant, independently of the choice of the repetition rate. In reality, however, the beam energy per pulse is the parameter actually determinant for the technical complexity, feasibility and price tag of the accelerator system. At constant energy per pulse condition, however, the optimal repetition rate becomes very strongly dependent on the type of the instrument and it can be as high as 1000 Hz. Such a high value is, on the other hand, wasteful for other instruments, which cannot efficiently deal with more than 20 - 30 pulses per second, i.e. their performance does not increase if additional power comes in the form of higher repetition rate. In this sense 50 – 100 Hz seems to be a rather reasonable compromise, between instrumental needs and accelerator design imperatives to minimise the beam energy delivered per pulse,

... but high power accelerator imperatives generally make higher repetition rates preferred.



in particular for short pulses. In contrast, for long proton pulses in the ms range the energy per pulse could reasonably reach values well in access of 500 kJ/pulse and thus a low (< 20 Hz) repetition rate is a good option up to some 10 MW time average power.

With long accelerator pulses < 20 Hz repetition rate becomes feasible even for > 5 MW power.

b) Wavelength resolution

The source neutron pulse length δt and the moderator to sample (or detector) distance determine the incoming wavelength resolution on a usual spallation source instrument:

$$\delta\lambda[\text{Å}] = 3.96 \times \delta t[\text{ms}] / L[\text{m}] \qquad (3)$$

If $\delta\lambda$ is less than necessary, the pulsed source instrument will lose in efficiency relative to a continuous source machine. If one does not consider background noise issues (e.g. the necessity on continuous sources to filter higher order reflections when using crystal monochromators, etc.) and beam losses due to absorption, finite reflectivities, etc. a pulsed source instrument can deliver a full neutron intensity gain corresponding to its peak flux compared to a similar type of continuous source machine if and only if all of the 3 conditions are met:

i) The required incoming beam divergence is less in both directions than the supermirror guide limit, eq. (1).

ii) The source repetition rate is low enough to satisfy the condition (2) and high enough to make the effective data collection time fill most of the elapsed time.

iii) $\delta\lambda$ as given by eq. (3) matches the $\delta\lambda$ freely chosen on the continuous source instrument.

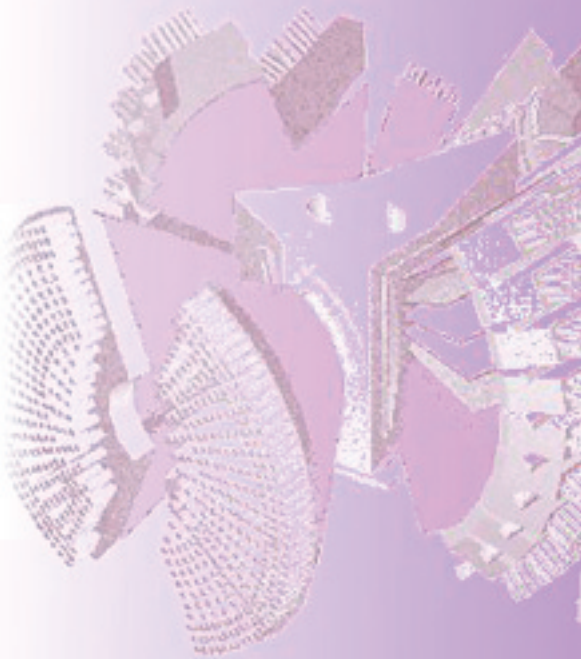
Pulsed source instrument design challenges include

- ***avoiding too good resolution***
- ***avoiding too low pulse repetition rates***
- ***avoiding too high pulse repetition rates***

at the same time for a variety of very different instruments.

These three points are actually quite some strings attached and can prove to be contradictory, for example L chosen to fulfil eq. (3) happens to be much too large for condition (2) to be satisfied (e.g. this is usual in high resolution powder diffraction) or the minimum value of L as determined by shielding and other instrument design imperatives leads to both too narrow wavelength band $\Delta\lambda$ and too good wavelength resolution $\delta\lambda$.

The canonical example for the latter is small angle scattering, SANS. Here the required good angular resolution can only be achieved without counterproductive limitation of the sample diameter to much less than the usual 0.5 – 1 cm if L ~ 40 m (D22 at ILL). At 50 Hz repetition rate and for short pulses this implies $\delta\lambda$ ~ 0.6% (at λ = 4 Å), i.e. some 20 times too small compared to D22 and $\Delta\lambda$ ~ 2 Å, i.e. some 3 – 4 times less than the potentially useful part of the Maxwellian spectrum of a cold coupled moderator. In this crucial example these two factors substantially lower the intensity gain of ESS compared to ILL, namely detailed Monte Carlo simulations show that instead of the peak flux ratio of 60 it becomes 3 - 4 at best. For the ESS $16^2/_3$ Hz long pulse target option the situation is more favourable: with $\delta\lambda$ ~ 5 % and $\Delta\lambda$ ~ 6 Å an intensity gain



of about a factor 10 remains from the 25 fold peak flux advantage.

An even tougher example than SANS is thermal neutron spectroscopy, as represented by the TOF instrument IN4 at ILL. Around 1 Å wavelength the divergence loss at direct view of the moderator compared to a focussing crystal monochromator is about 20 fold (even with substantial crystal reflectivity losses assumed), and it is compounded with a typically 8 fold loss by too low pulse repetition rate (50 Hz vs. some 400 Hz). All this is not compensated by the 100 fold peak flux advantage of ESS.

These kind of efficiency losses are ultimately due to the fact that the pulsed source repetition rate and the neutron pulse lengths just cannot be simultaneously optimal for all kinds of instruments of conventional design. In what follows we will discuss how novel (by now not implemented) instrument design techniques can mitigate the problem. The need for innovative source and instrument design is a centrally essential feature of the ESS project: ESS is not just a next generation pulsed spallation source. Spallation sources as we know them today are efficient complements to the current mainstay tools for neutron scattering, continuous reactor sources. By now we know, that there is no way to enhance the neutron performance of reactor sources at reasonable costs beyond that of ILL. Thus the real challenge of ESS is that it has to transform the spallation technique from one complementary to reactor sources to one superior across the board. ESS will be the next generation neutron source compared to both existing reactors and existing or planned spallation sources. It is probably a fair estimate that at most 30 % of moderator and instrument design approaches needed to achieve this goal are established by now.

V. New multiplexing and pulse shaping approaches

Multiplexing and pulse shaping allows us to better satisfy a larger variety of instrument requirements.

In order to alleviate the contradictions between demands on source parameters set by accelerator physics, target and moderator design needs and the variety of instrumental requirements, we need to develop techniques which make the instrument benefit from effective source parameters, namely repetition rate and neutron pulse lengths, different from the actual ones. Suppressing source pulses, up to actually 4 out of 5, is a well established method in order to reduce the effective pulse repetition rate. We will consider here the potentials offered by some other, not yet experimentally tested techniques, which are a lot more efficient than throwing away a large fraction of the source pulses.

Pulse shaping by fast choppers make unprecedented short and sharp pulses and high resolutions available.

The use of fast pulse shaping choppers close to the source (which can practically mean not less than some 6 m distance) in order to shorten the moderator pulse length for high resolution applications has already been considered in the generic instrument evaluations (see the report on indirect geometry spectrometers in Ref. [1]), although it has not been experimentally realised yet. It will allow us to achieve shorter

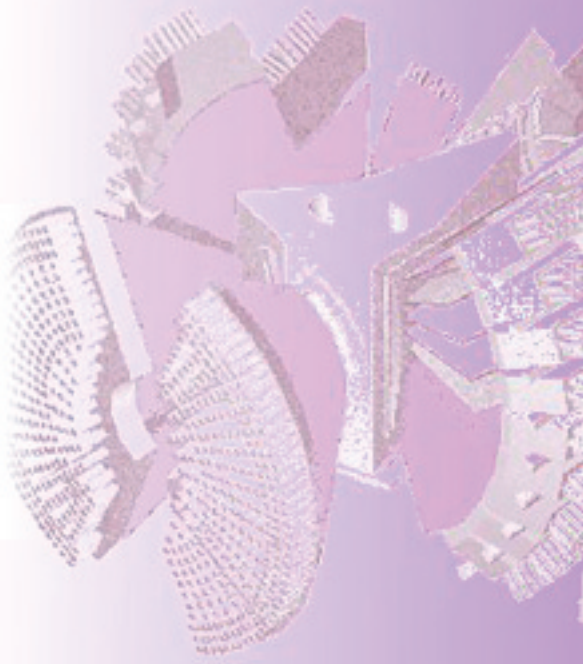
pulse lengths than provided by poisoned moderators. Additional, not less important advantages are that the short pulses cut out of coupled moderator pulses benefit from the higher peak flux of these moderators compared to the poisoned ones (Figure 1) and by this way one could eventually reduce the number of required moderators on one target station. The great inconvenience of the technique is that it normally drastically reduces the available wavelength band (e.g. to ~ 0.2 Å when used with cold coupled moderators). New techniques to alleviate this drawback will be discussed below.

Multiplexing approaches can be roughly defined by trying to make use of multiple beam parameter domains, which would conventionally exclude each other. A most beautiful example is the pioneering proposal of the MUSICAL spectrometer by Alefeld [7], in which the beams of several different crystal monochromators are distinguished from each other by time delays induced by judiciously choosing the positions of the various crystals. We will now consider a number of related schemes, aimed at enhancing the efficiency of use of pulsed sources by getting around restrictions implied by relations (1) – (3) above. Paradoxically, we will not really need to consider Alefeld's original idea here: the combination of pulse shaping choppers and very long flight paths bridged by advanced, low loss "ballistic" neutron guides now offers a simpler approach to very high resolution backscattering spectroscopy, as shown in Ref. [1].

a) Repetition rate multiplication

Repetition rate multiplication can enhance the effective flux on the sample by several times.

The example of a IN4 type instrument above illustrates that even 50 Hz pulse repetition rate is a substantial disadvantage in direct geometry TOF spectroscopy, where 100 Hz to 500 Hz pulse frequency is the rule at continuous sources. This higher frequency is determined by the time needed to analyse the scattered beam, and at lower repetition rates no data are collected for most of the time. This drawback can be removed by extracting a number of pulses with different monochromatic neutron velocities from the same source pulse [8]. For example in the IN500 project at Los Alamos up to 240 pulses/s can be delivered to the sample from 20 source pulses per second. In practice this means that instead of one experiment made, as usual, with a single wavelength, a number of experiments with different incoming neutron wavelengths are accomplished simultaneously, and the information needs to be combined in the data evaluation process. This can be more efficiently accomplished the smaller is the difference between the neutron wavelengths in subsequent pulses (e.g. 0.265 Å at 240 Hz multiplied rate on IN500). This is best achieved by long source to sample distances, in which case the most intense and longest coupled moderator pulses are the best choice for matching the required resolution. Numerical evaluation of a few specific examples confirmed [8] that multiple pulses with different wavelengths can deliver comparably useful information, thus e.g. in case of an IN4 type instrument much of the count rate



loss due to the low repetition rate can be recovered. Thus in contrast to the conventional situation discussed above, repetition rate multiplication will allow ESS to deliver superior neutron flux even in this case (and at better angular resolution, without the focussing crystals).

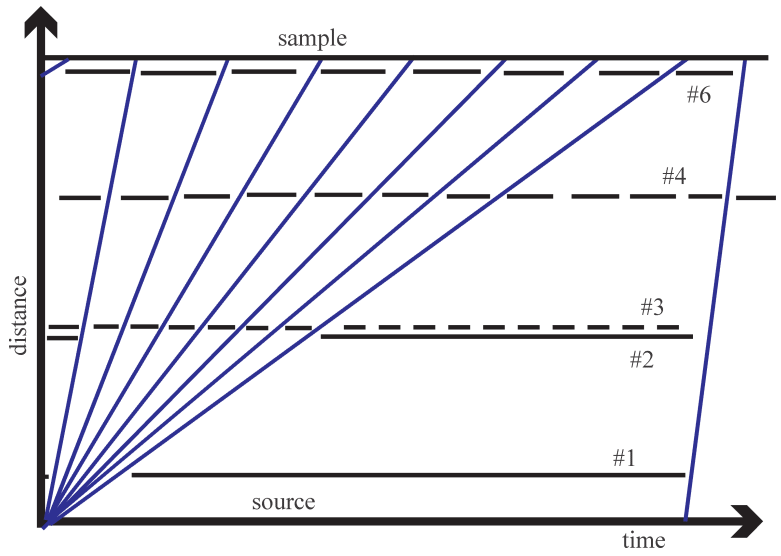


Figure 5: Principle of Repetition Rate Multiplication as realised by a disc chopper system. For an example, the source to sample distance in the IN500 project is 63 m, the time between source pulses 50 ms, and the maximal speed of the disc choppers used is 14400 RPM. Choppers #3 and #6 consist of two counter-rotating discs each. The role of chopper #5 (not shown), mounted close to #6, is to reduce the basic 240 Hz pulse repetition rate on the sample, if necessary, by optionally only letting through every 2^{nd}, 3^{rd}, 4^{th} or 6^{th} pulse. Note that the effective source pulse length for all pulses on the sample is determined by the timing between the sharp rising edge of the source pulse and the closing time of chopper #3. By properly choosing the chopper parameters no neutrons with $\lambda < 80$ Å can make their way through the chopper system, other than the ones selected as shown here [8].

Repetition rate multiplication can be readily realised by the use of disc choppers of the type well established at continuous source cold neutron spectrometers, such as IN5 at ILL or NEAT at HMI. At the same time it also implies source pulse shaping, as explained in the caption of Figure 5.

b) Wavelength frame multiplication

Wavelength frame multiplication can extend the wavelength band without the need of changing the frame definition chopper timing ...

The use of a broad wavelength band allows one to achieve a wide dynamic range in wavenumber q. Data can be taken with various delays after the pulse emitting the neutrons, i.e. the wavelength band $\Delta\lambda$ allowed by (2) can be rather freely positioned around various average wavelengths. This positioning is accomplished by frame definition choppers (choppers #1 and #2 in Figure 5 serve this function) and in a series of data collection periods one can cover any reasonable wavelength band. It remains, however, desirable to also be able to collect data in a broad wavelength band quasi simultaneously, for example with a sample whose state changes rapidly with time. An example of how to reach this is

illustrated in Figure 6 [9].

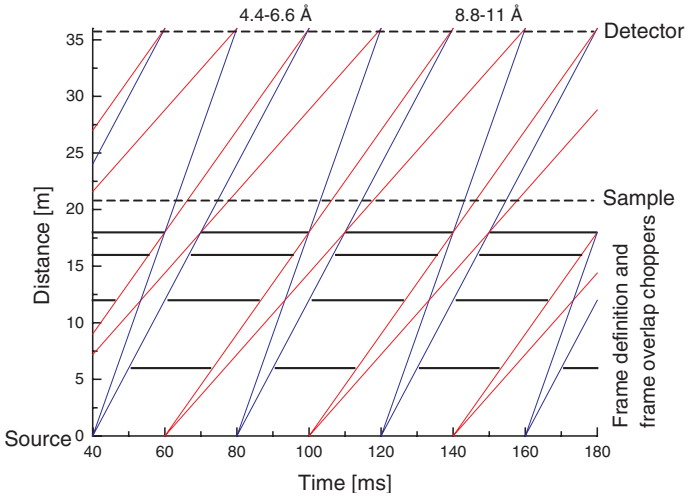


Figure 6: An example of wavelength frame multiplication for a 36 m long instrument at a 50 Hz source. At the junction of subsequent frames at the detector the source pulse length and the chopper opening/closing time uncertainties produce overlap of the penumbra. More than 17 ms from the 20 ms frame duration between source pulses are free from overlap or higher order leakage neutrons with wavelength < 80 Å [9].

The set of frame definition choppers shown makes sure, that in subsequent periods T between two source pulses different wavelength bands alternate on arrival at the sample or detector. For example with three alternating wavelength bands (three fold frame multiplication) a SANS machine on the 50 Hz source can cover the same or broader band than a conventionally operating instrument on a 10 Hz source (Figure 7).

Another, more important application of wavelength frame multiplication is related to the use of pulse definition choppers, which can cut the length of coupled moderator pulses or long pulses to actually shorter duration (10 – 20 μs) than that of the poisoned moderator pulses. For example, this approach has been found to allow us to reach sub μeV resolution in backscattering spectroscopy at some 200 m source to sample distances (as mentioned above) which resolution previously was deemed to only be achievable at good intensity by the MUSICAL approach. This kind of pulse shaping is particularly easily performed with long pulses, but the source pulse length limits the wavelength band, according to relation (2). With the pulse shaping chopper at 6 m from the moderator and 2 ms pulse length, the band width is 1.333 Å.

… and to make broad wavelength range accessible to chopper shaped, very short and sharp pulses to achieve very high resolution.

The frame multiplication scheme shown in Figure 8 allows us to append in a gap free manner a number of 1.3 Å wavelength bands one after the other (2 are shown in the figure) in order to fill the whole time frame between two subsequent source pulses. For example the 5.33 Å band obtained by 4 fold frame multiplication corresponds to the full bandwidth of the $16^2/_3$ Hz

source at 45 m source to detector distance.

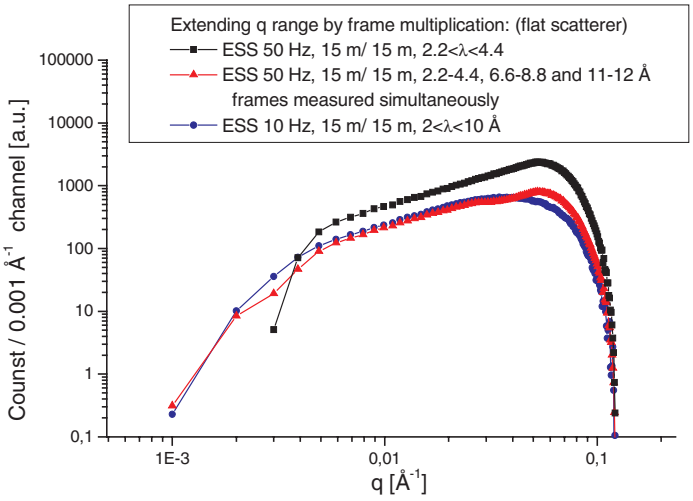
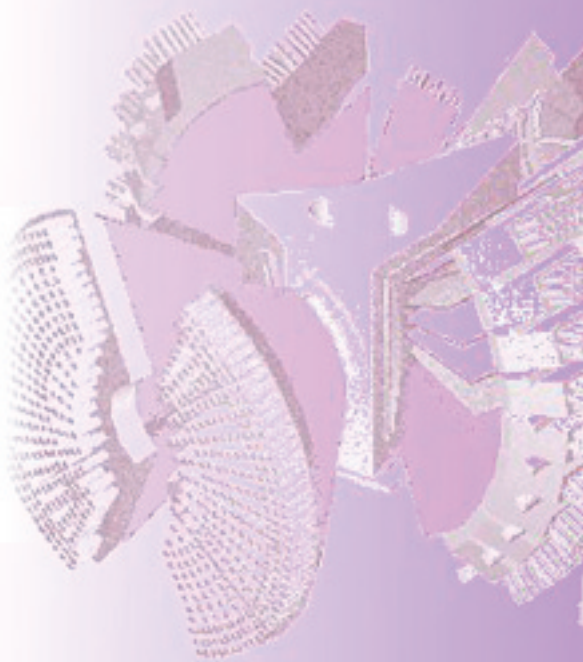


Figure 7: Example of extending the q range of a SANS instrument on the 50 Hz target station by wavelength frame multiplication (red triangles) compared to single frame 50 Hz data collection (black squares) and single frame operation at 10 Hz with equal pulses (blue dots).

This "frame multiplication for pulse shaping chopper" allows us to cut out very short and sharp, symmetrically shaped pulses from long pulses, as an alternative choice to the asymmetric, exponentially decaying short source pulses from poisoned moderators in virtually any application. To achieve the same resolution these symmetric pulses can be about 50 % larger in FWHM than the exponentially shaped source pulses. With the long pulse peak flux nearly equal to the poisoned moderator peak flux (Figure 1 and 4), the long pulses actually offer equal intensity at equal wavelength resolution (or much shorter pulses at wavelengths > 2 Å for achieving unprecedented high resolutions). This will open up, especially combined with multi-spectral beam extraction (Figure 4) the option to use the long pulse target station for all high wavelength resolution applications (e.g. very high resolution powder diffraction) for wavelengths > 0.8 Å eventually alleviating the demand for a high number of different types of moderators on the 50 Hz short pulse station. Note that it is possible to emulate the wavelength dependent pulse width of poisoned moderators by adequate "eye-of-the-needle", very high speed disc chopper systems [9].



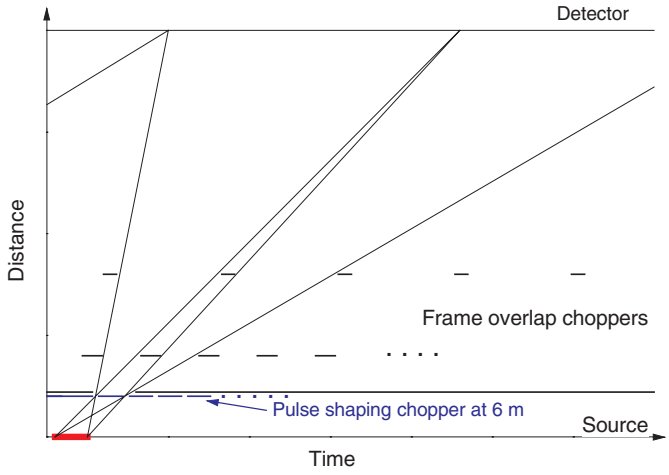


Figure 8: Principle of wavelength frame multiplication used to extend the strongly reduced wavelength band conventionally allowed for by a single opening of a pulse shaping chopper outside the bulk shielding. For 2 ms long pulses this technique enables us to simultaneously and continuously cover typically required wavelength bands of 3 – 8 Å with short pulses of adjustable length on a long pulse source [9].

c) Multiple beams and detectors

Similarly to making several pulses impinging on the sample one after the other, one can conceive to make several converging incoming beams placed next to each other. Such converging beams can be produced at little technical risk by using a series of collimating beam frames or slits, starting at a rather large cross section virtual source created outside the source bulk shielding. Such a virtual source can be conveniently achieved by using supermirror optics, for example in the manner the central section of a ballistic guide can be fully illuminated by the diverging "reflector" section, even if its dimensions are bigger than that of the moderator [6]. It is harder to implement such optics on existing source beam tubes, not designed for this purpose. The following two examples show the importance of this kind of multiplexing in space.

Fast switching between a set of incoming beams can speed up data collection in a broad range of q.

Most reflectometers on continuous sources use a monochromatic beam with scanning the grazing impact angle. This offers the advantage of optimising sequential data collection by distributing the available time in a most efficient manner between the points in the scan. With the TOF technique on a spallation source the data collection efficiency is necessarily less optimal, since in a more or less broad simultaneously measured band every point gets the same share of time. The advantage on the other hand is, that a simultaneous scan can eventually be completed in a shorter time for time dependent studies, since one saves the time needed to physically change the angle between beam and sample (which approach is the faster really depends on the experiment, case by case.) To achieve a broad q range in a fixed angle TOF experiment one needs the broadest possible wavelength band, i.e. the lowest possible source frequency, which for the short pulse option ultimately means to reduce

the source flux by eliminating most of the 50 Hz source pulses. The alternative way to broad band rapid data collection is to implement a number of beams impinging on the sample at different angles, e.g. at 0.5, 1 and 2°, as tested or planned at several places (LANSCE, HMI, SNS, ...). Since for a typical reflectivity curve only a few % of the total measuring time is required to obtain more than enough statistics at the smaller angles, this method allows us to retain the full power of the 50 Hz source and cover the q range of a 10 Hz source more than 4 times faster (Ref. [1]). Switching from one beam to another by opening and closing beam slits can be done with little time lost, it could even be accomplished between two subsequent source pulses.

Coverage of a broad q range is also an issue in SANS. On continuous sources this is most often achieved in a very efficient way by moving the detector more or less close to the sample. Again, this allows us to optimally divide the measuring time between the different detector positions, and if the time needed to move the detector is much shorter than the necessary data collection times, this is the most efficient approach on all sources. An alternative approach is to place several detectors at different distances from the sample, as already implemented at several facilities (Dubna, ISIS, ...). The method can work particularly well for pulsed sources, since the gaps in scattering angle between the detectors is bridged in the q space by the use of a more or less broad wavelength band. Figure 9 illustrates the far superior data collection rate and q range one can obtain by this technique on the $16^2/_3$ Hz, 5 MW long pulse source compared to both ILL and the 10 Hz, 1 MW option.

Multiple detectors vastly extend the simultaneously explored q range in high speed SANS data collection.

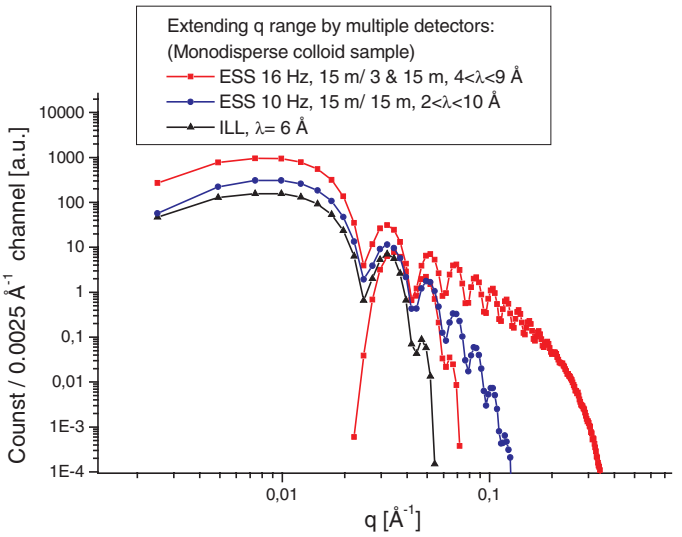
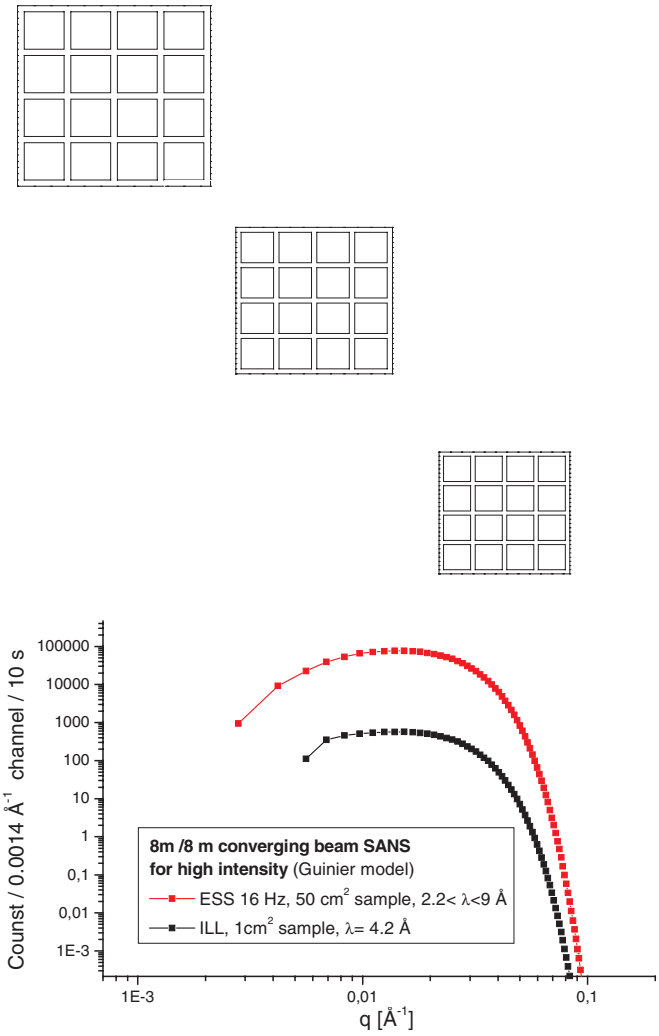


Figure 9: Monte Carlo simulated small angle scattering spectra from a dilute monodisperse colloid sample for 15 m – 15 m long SANS instruments, with an additional detector at 3 m from the sample for the ESS $16^2/_3$ Hz instrument (red squares). The detectors are of 1 m x 1 m dimension, with the additional detector at 3 m having a 23 cm x 23 cm hole in the middle. The two red curves have a convenient overlap, in spite of the gap in angular coverage between the two detectors. The data also illustrates the order of magnitude intensity gain compared to ILL (black triangles) at good resolution.

The final example of multiplexing is to enhance the data rates in SANS by making a large number of identically collimated beams converge on the same spot on the area detector. It has been proposed to realise this by a set of diaphragms with multiple holes (e.g. at Los Alamos), as indicated in Figure 10. The real significance of this approach is that it will allow us to enhance the data collection rate at ESS by another order of magnitude compared to the most powerful SANS machines today – at least for flat samples larger than 20 – 30 cm^2. This will make possible to take meaningful SANS spectra in less than a second time and thus open up fully new opportunities in time dependent studies.



Multiple converging incoming beam combined with the superior neutron power of ESS can increase the speed of SANS data collection by two orders of magnitude.

Figure 10: Neutron absorbing frames to build a converging collimator, which essentially just amounts to repeating the same incoming beam configuration several times next to each other. The simulated SANS data (bottom) confirm the some 100 fold enhanced data collection rate at ESS compared to a single beam, identical angular resolution experiment at ILL.

The same techniques of converging beams can also help us to achieve better angular resolution and reduce the lower limit of the accessible q range to some $2 - 3 \times 10^{-4}$ $Å^{-1}$. Here the use of a series of static frames will be made difficult by the different curvature of different wavelength neutron trajectories

under gravity. Focussing mirrors should offer a better approach if the problem of high background due to non-specular reflections can be solved. In any case, to enhance the resolution capability of SANS instruments to lower q values than customary today, the higher flux of ESS will need to be combined with advances in instrument design.

VI. Summary of ESS instrument performances

ESS instruments will benefit from both highest neutron power and enhanced instrument design.

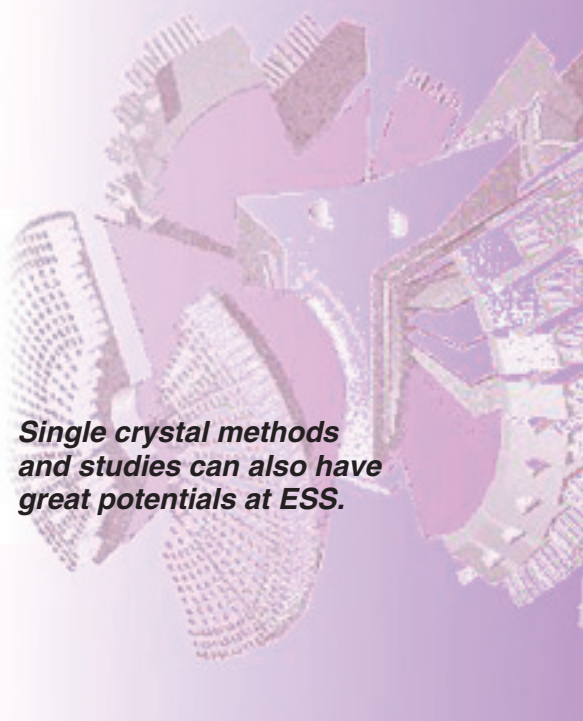
The table below, summarises the instrument performance gain factors presented in more details in the topical Instrument Group reports compiled in Ref. [1]. A few similar instruments are represented just by one of them and 4, not included in the reports, have been added (TAS, D7, MUSICAL and fundamental physics beam). A further deviation in the table compared to Ref. [1] is including the converging collimator for the high intensity SANS, see Figure 10 above. The neutron intensity gains due to the enhanced flux of ESS have been evaluated separately from additional gains in data collection rate due to foreseeable improvements in instrumentation methods and techniques. The source gain factors tell us how much of the increased performance will only be available by realising ESS. The total gain factors, i.e. source plus progress in instrumentation, on the other hand, are those which will determine what kind of new scientific opportunities ESS will offer. The highest number in the table, well above 1000 in the case of cold neutron spectroscopy, is one of the best studied and established figure, and it illustrates the point in an instructive manner. Compared to the extremely popular and powerful TOF spectrometer IN5 at ILL (which was roughly equivalent to the more recently designed NEAT at HMI), the source gain factor was estimated to be around 50, which is somewhat less than the peak flux ratio in order to take into account the need to use repetition rate multiplication to recover losses due to the rather low source repetition rate of 50 Hz. Supermirror optical beam delivery has been shown to provide for an additional gain factor of 6 – 7 [6] compared with Ni coated guides, a factor of 4 can be gained at least in detector solid angle, and improved chopper design offers another factor of 1.5 – 2 at equal resolution (optimised chopper positions, counter-rotating choppers, trapezoidal pulses). This safely adds up to more than the 1600 fold improvement quoted in the table, of which about a factor of 9 - 10 is being realised by the current reconstruction of IN5. Knowing from experience all the new science that could be achieved by IN5 in the past, one can expect an explosion of qualitatively new opportunities this tremendous increase of power will make happen.

Conservatively estimated 50 – 1000 fold increase of sensitivity in most experiments will open up new fields of scientific opportunities.

These gains are enabled by the on average 20 fold increase of the effective neutron flux compared to the best today.

Not only the highest, but the lowest numbers in the table also merit some attention. Single peak single crystal work with hot neutrons suffers from the already mentioned low time average flux around 0.5 Å wavelength. The pulsed time structure of the source offers some fringe benefits even if one uses crystal monochromators, by making easier the identification of spurious effects, such as taking care of the problem of higher order reflections without the use of filters. But the real solution

Single crystal methods and studies can also have great potentials at ESS.



here is to work out an equivalent of the hot source for ESS, as discussed above. For the very similar case of triple axis spectroscopy the often invoked speculative possibility of very low background between source pulses, if realised, might become an additional and very important fringe benefit. TOF spectrometers with large detector solid angle coverage, and consequently wide open detector geometry, are less likely to achieve very low backgrounds needed to bring extremely week signals to evidence. For this reason, even with their very limited data collection rates, TAS instruments might well have a role to play at pulsed spallation sources with time average fluxes approaching that of ILL.

Table 1:

Expected performance of generic instruments on the various target station options for ESS. ● : first choice or one of essentially equivalent first choice options. O : second choice: about a factor of 2 inferior in data collection rate to the first choice. The numbers correspond to instruments implemented on the best (one of the bests) target options. Blue numbers: compared to ILL beams and best existing instruments at ILL. Black numbers: compared to ISIS beams and best existing ISIS instruments. Table compiled on the basis of Ref. [1].

Instrument	50 Hz 5 MW	$16^2/_3$ Hz 5 MW	Source gain	Total gain
High energy chopper	●		30	30
Thermal chopper	●		30	240
Cold chopper	●	O	50	1600
Variable, cold chopper	O	●	20	800
Backscattering 0.8 μeV	●		25	50
Backscattering 17 μeV	●		150	600
Molecular spectrometer (TOSCA)	●		50	100
Electron Volt Spectrometer	●		30	300
High resolution NSE		●	10	100
Wide angle NSE		●	9	300
Triple-Axis	●	●	0.5-1	1-4
High Resolution Single X	●		>>10	>>10
Chemical Single X	●		>>10	>>10
High Resolution Protein	●		>20	>20
Low Resolution Protein	●	●	3-5	3-5
Single Peak incl. Cryopad	●	●	0.3-3	0.3-3
High Resolution Powder	●	●	50	150
High Q Powder	●		60	120
Magnetic Powder	●	O	60	60
High Resolution Reflectometer	●	O	20	40
High Intensity Reflectometer	O	●	15	40
Liquids Diffractometer	●		20	20
High Intensity SANS		●	8	100
High λ Resolution SANS	●		150	300
Engineering Diffractometer	●		30	90
Fundamental Physics	●	●	1	NA
Diffuse scattering (D7)	O	●	15	300
Backscattering (Musical)		●	40	40
Average (geometrical)			>19	>47

VII. Technical risks and challenges

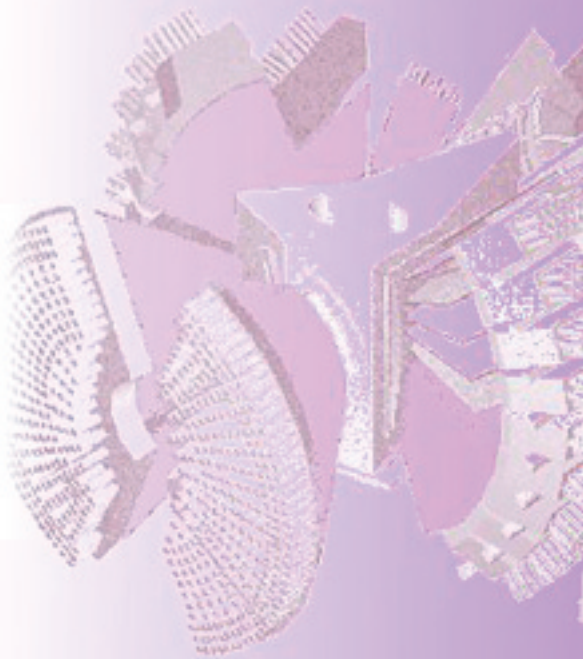
The reference instrumentation techniques assumed in the evaluation of the expected performance of a set of generic instruments at ESS (as summarised in the topical reports by the various Instrument Groups in Ref. [1]), were chosen to be fairly conservative, and with a few exceptions well established at current spallation sources. The most notable exception is the use of fast disc choppers to cut out very short pulses from the longish (> 0.2 ms) coupled cold moderator pulses in order to achieve sub μeV resolution over 200 m flight path in backscattering spectroscopy. The more unusual approaches discussed in the present paper have not yet been implemented, but they are largely based on using components well established on reactor sources, e.g. fast disc chopper systems consisting of 6 – 8 phased rotors. Background noise, in particular the direct fast neutron bursts from the proton pulses will certainly represent some technical challenge, as does the need to increase the maximum acceptable count rate of detectors. Nevertheless, the technical risks involved in the instrument performance assessment are really small.

Established spallation and reactor instrument techniques combined offer a solid basis for instrument design and innovation.

In this context the issue of neutron detectors deserves particular clarification. On the basis of current experience at pulsed spallation neutron sources we can estimate that the highest instantaneous counting rate at ESS will be in the range of 10^5 counts/s/cm^2 when a Bragg peak from a sizable sample hits the detector during the pulse. This high counting rate will be sustained for a few tens of μs. The time average counting rate over 1 m^2 position sensitive detector area will reach 10^8 counts/s. These numbers are about an order of magnitude higher than what can be handled today. Nevertheless, we can be fully confident, that in 10 years from now we will have the required high speed detectors at our disposal. Many neutron scattering laboratories, including some ESS partner institutions, pursue vigorous programs in developing higher performance detectors. The results of this research are quite likely to make available the detectors needed by ESS in due time. In a few years time ESS should join this effort by its own team. One has also to keep in mind, that the biggest impact of ESS will be at research areas where the current neutron beam intensities are simply not sufficient to produce well observable signals. Examples are inelastic scattering studies of many problems of microscopic dynamics and the investigation of very small samples. In these cases current detector speeds will be fully adequate.

It can be confidently assumed that the speed of neutron detectors will advance in the next 10 years to meet the new requirements set by the unprecedented neutron flux at ESS.

The innovative approaches discussed above are those novel general concepts proposed to be relevant for the design of many instruments. Hence they are part of the general approach at ESS to enhance the efficiency of the use of neutrons and they also serve as input for design decisions on the ESS layout as a whole, including accelerator and target / moderator systems. The hardware components at the basis of these concepts are all well established and in use at existing facilities, either reactors or spallation sources. So the technical risks are primarily involved in novel kind of use of



available components. This risk is kept low by an extensive effort of computer simulation studies and by building and testing of prototypes by ESS partner institutions.

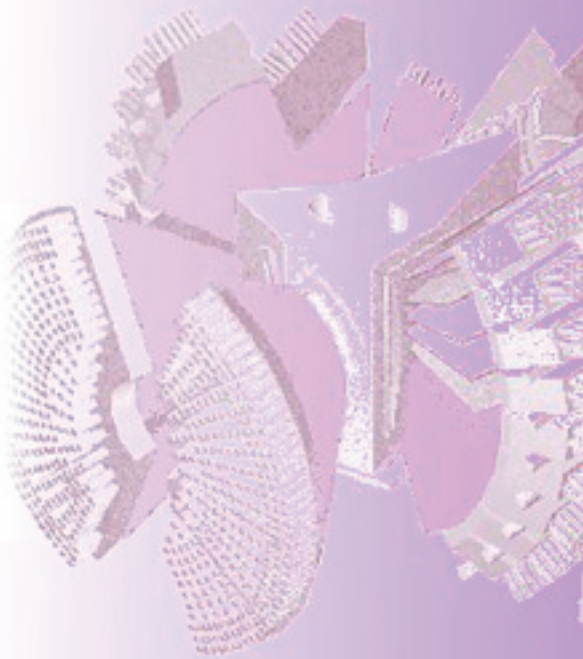
There are many more specific innovations suggested by now which can become the basis of conceiving individual new instruments or enhance performance in a particular type of experiment. For example one fascinating group of ideas is to accelerate neutrons, which would allow us to enhance the flux density (so called phase space transformation) that is otherwise kept constant in static, time independent environment by virtue of the Liouville theorem. For example a promising approach of neutron acceleration by magnetic fields has been proposed [10]. Another idea, the use of Larmor precession techniques for the analysis of elastic scattering processes also made much progress recently [11]. The ESS instrument development approach reserves a particular space for innovations of this kind too, whether already in the making or to emerge in the future. At this stage 2 of the about 20 instruments planned to be built first at ESS are assumed to be yet unspecified and unknown, novel kind of machines. New ideas to emerge might make possible to perform experiments at ESS in 10 – 15 years from now we cannot imagine today. It is a particular challenge to encourage and recognise such developments in due time. This is a calculated risk, but not taking this risk is the worst risks of all.

ESS instrument development plans leave room for novel approaches not yet imagined.

VIII. Summary

The overriding goal of the ESS project is to provide decisive advances in research opportunities for a broad user community in all fields of exploration related to condensed matter: physics, chemistry, geology, engineering, life sciences, environment, cultural heritage. This goal will be achieved by combining the benefits of advances in all areas of science and technology relevant to building ESS: accelerators, target and moderator design and instrumentation. ESS is a global approach, where the technical potentials in one area are fully taken into account as opportunities and boundary conditions in another one. Literally speaking, proton ion source design imperatives are kept in mind in defining the instrument concepts and vice versa. The innovative configuration of a short and a long pulse target station is a good example. It is the accelerator design response to specific requirements of a group of neutron scattering instruments to have more neutrons per pulse with less emphasis on the pulse duration. Particular attention is paid to extract and transport neutrons from the source to the instruments with enhanced efficiency by using advanced neutron optical systems. Employing multiplexing instrument design techniques will allow ESS to best satisfy the different needs of different neutron scattering instruments. An example is the broad variety of pulse repetition rates that will be possible at ESS: $16^2/_3$ Hz on the long pulse target station, 50 Hz on the short pulse one and up to 200 Hz effective rate by using a multiplexing instrument design. As a

ESS will be a quantum leap in performance by combining highest power with innovative approaches to assure highest efficiency in using that power.



result of this approach of global optimisation ESS will not only be the most powerful spallation source in terms of proton beam power, but it will also benefit from enhanced efficiency in terms of turning the proton power into neutrons on the sample, compared to current state of the art used at existing sources and at facilities that are being built. As a result ESS will offer a quantum leap of 2 – 3 orders of magnitude superior sensitivity to what we know today in the majority of neutron scattering studies of condensed matter.

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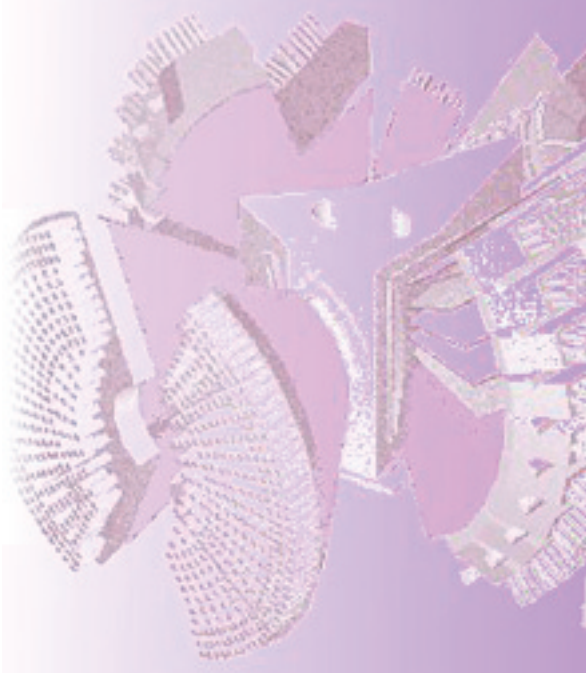

Chapter 4

The Scientific Potential of ESS

4. The Scientific Potential of ESS

The ESS is the next decisive step in the evolution of neutron sources and will offer a revolution in neutron science. As the world leading facility the ESS will open novel scientific opportunities and allow new experiments which are presently not feasible. This chapter is devoted to considerations on the scientific potential of the ESS in the different science disciplines which are accessible by ESS. The following reports focus on future trends in the different disciplines emphasising high profile flagship areas at and beyond thresholds.

In order to assess the future development in the different scientific areas eight science groups have been created which were convened by members of the Scientific Advisory Committee (SAC) of ESS. Table 1 displays the disciplines and conveners.

Eight science groups have been created which were convened by members of the SAC.

Table 1:
Science Groups and their Conveners

Science Group	*Convener/s*	
Solid State Physics	A. Furrer C. Vettier	PSI Villigen, Switzerland ILL, France
Material Science and Engineering	H. Zabel	University of Bochum, Germany
Biology and Biotechnology	J. Helliwell	University of Manchester, United Kingdom
Soft Condensed Matter	J. Colmenero D. Richter	Univ. of the Basque Country & DIPC, Spain FZ Jülich, Germany
Chemistry, Structure, Kinetics and Dynamics	H. Jobic W. David	CNRS/Univ. Lyon 1, France ISIS, United Kingdom
Earth Science, Environmental Science and Cultural Heritage	R. Rinaldi	University of Perugia, Italy
Liquids and Glasses	R. McGreevy	University of Uppsala, Sweden
Fundamental Physics	H. Rauch	Atomic Institute of the Austrian University, Austria

The scientific potential of the ESS depends on the technical parameters and configuration of the facility. To value the impact of the ESS on ambitious challenges of the different disciplines the instrumentation opportunities of ESS are of great importance. Therefore the scientific demands on the instrument suite derived from the flagship areas were discussed in the frame of instrumentation opportunities at the ESS, which were assessed by instrument specialists representing the different fields of instrumentation. Table 2 displays the nine instrument groups and the respective conveners, which were assembled in order to study the performance of generic instrumentation at the two target stations of ESS.

Scientific demands were discussed in the frame of instrumentation opportunities.

Table 2:
Instrument Groups and their Conveners

Instrument Group	*Convener*	
Powder Diffraction	P. Radaelli	ISIS, United Kingdom
Direct Geometry Spectrometers	R. Eccleston	ISIS, United Kingdom
Indirect Geometry Spectrometers	K. Andersen	ISIS, United Kingdom
Neutron Spin Echo Spectrometer	M. Monkenbusch	FZ Jülich, Germany
SANS	R. Heenan	ISIS, United Kingdom
Reflectometry	H. Fritzsche	HMI Berlin, Germany
Single Crystal Diffractometer and Protein Crystallography	C. Wilson	ISIS, United Kingdom
Structure Factor Determination	A. Soper	ISIS, United Kingdom
Engineering	P.J. Withers	Manchester Material Science Centre, U.K.

Since the instrument experts were reliant on an assessment of the scientific demands on the ESS and at the same time the scientists needed a consolidated knowledge of the instrumentation opportunities and the potential opened up by the ESS, an intense exchange of expertise and collaboration between both groups was vital. This was for instance realised by joined workshops like the ESS SAC / ENSA workshop on "Scientific Trends in Condensed Matter Research and Instrumentation Opportunities at ESS" which was held in May 2001 in Engelberg, Switzerland. About 80 scientists from all fields of neutron science were assembled there by a combined effort of the Scientific Advisory Committee (SAC) of the ESS and the European Neutron Scattering Association (ENSA), in order to define the final ESS layout on scientific grounds and to assess trends in condensed matter science related to neutron research. One major goal of the workshop was to predict new opportunities, in particular important and decisive areas of science where ESS is expected to impact strongly in solving problems which cannot be accessed presently. In a similar way scientists focussing on the neutron itself in the area of fundamental physics, discussed the main issues in their field and derived their demands on the facility.

An intense exchange of expertise and collaboration between the science and instrument groups was vital.

To embrace the increasing importance of neutron scattering in science fields like Biology or Earth Science the SAC initiated two topical workshops, namely 'Perspectives of neutron scattering for the earth sciences with ESS' in Cambridge, U.K. and 'Flexibility and functions of proteins' in Heidelberg, Germany (both in January 2002). The outcome of these workshops was relevant to arrive at an accurate determination of future trends and new opportunities at the ESS in the emerging science fields of neutron scattering.

To embrace the increasing importance of neutron scattering in emerging science fields the SAC initiated two topical workshops.

The Engelberg workshop led to a new approach to the scientific case for the ESS by focussing on threshold areas and flagship experiments. The outcome of the concerted endeavours during the workshop in Engelberg was the basis for the disciplinary science case reports presented in this chapter[1].

[1] These science reports present a forward look into the scientific opportunities offered by ESS. They are not scientific papers and therefore there is no attempt to add references to refer to the large amount of work behind these scientific areas. References are generally only used in order to relate to specific examples or figures.

4.1 Solid State Physics

R. Currat[1], R. Cywinski[2], C. Fermon[3], A. Furrer[4], B. Keimer[5], G.H. Lander[6], D.F. McMorrow[7], H.R. Ott[8], C. Vettier[1]

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[8] Laboratorium für Festkörperphysik, ETH Zürich, CH-8093 Zürich, Switzerland

Abstract
Solid state physics encompasses fundamental research that has underpinned much of the technological progress in the last 50 years. Recent trends include the emphasis on complexity, including organic materials, and reduced dimensionality down to the scale of quantum dots. In the large variety of instruments used in solid-state physics, scattering has a special place as it gives information on **spatial** correlations. Within scattering techniques, neutrons are unique as they are able to provide, simultaneously, information on **both** the **static** and **dynamical** correlations. We discuss these advantages for neutrons, stress the materials-driven nature of this approach, and present a selection (by no means complete) of flagship experiments that will be possible only at the ESS. We conclude with a discussion of the best instruments and pulse structure for frontier experiments at the ESS.

I. Introduction

This report identifies future research frontiers in solid state physics. It starts by reviewing briefly the general capabilities of neutron scattering methods for the study of phenomena in condensed matter physics, with particular emphasis on the capabilities provided by powerful modern spallation neutron sources such as the ESS. Although complementary methods such as the other scattering probe, synchrotron X-rays, and local probes like NMR, EPR and Mössbauer, provide important information, our deliberations have confirmed the unique opportunities afforded by neutrons in general and by ESS in particular. The report highlights examples of flagship experiments, and addresses the impact that the ESS can have in these frontier areas. Recommendations for instruments and target options are presented.

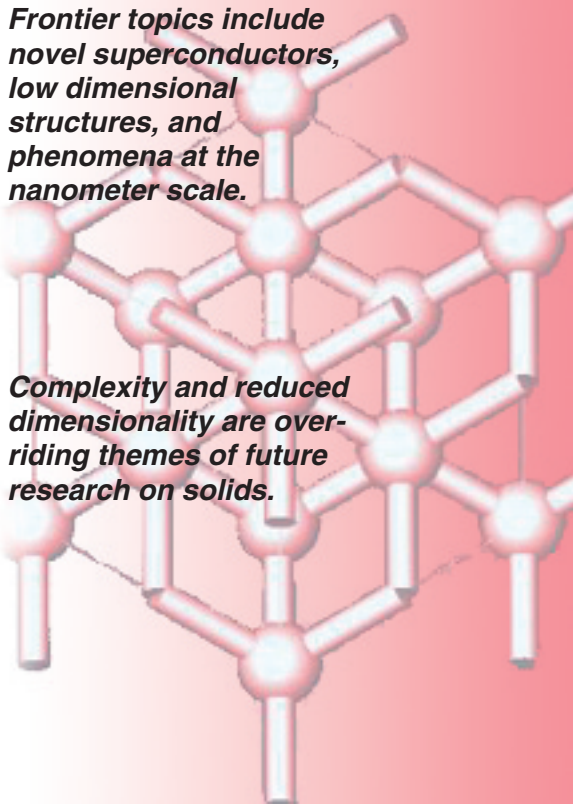
Advances in solid state physics are at the root of most technologies shaping today's world. Neutrons are key to our understanding of solids. The ESS will have a large impact on cutting-edge research in solid state physics.

Research in the solid state continues to have a high impact, both in basic physics as well as with respect to technological applications. Recent examples include high-T_c and other unconventional superconductors, low dimensional semiconductor structures, magnetic thin films, and materials with giant magneto-resistance. A great deal of basic research is now technology driven and much is oriented towards nanometer-scale systems. The properties of nano-patterns and self-assembled quantum dots are of great interest from both a theoretical and experimental perspective.

Frontier topics include novel superconductors, low dimensional structures, and phenomena at the nanometer scale.

The future challenge in basic solid state physics is the exploration and understanding of the collective behaviour of large numbers of interacting particles. Although future trends are notoriously difficult to predict, two important directions emerge. Firstly the tendency to higher complexity, specifically materials which have physical properties determined by competing interactions, and secondly the trend to reduced dimensionality, both by synthesizing materials with low

Complexity and reduced dimensionality are over-riding themes of future research on solids.



dimensional structural elements and by reducing the physical size of objects to surfaces and interfaces, single atom wires and dots. One basic interest in solid state physics is to establish the ground state of relevant systems. This may be done by exploring possible excitations out of the ground state, neutrons are a versatile, and often unique probe with which to accomplish this goal.

The table summarises some of the research areas that are expected to be of major interest in ten years time.

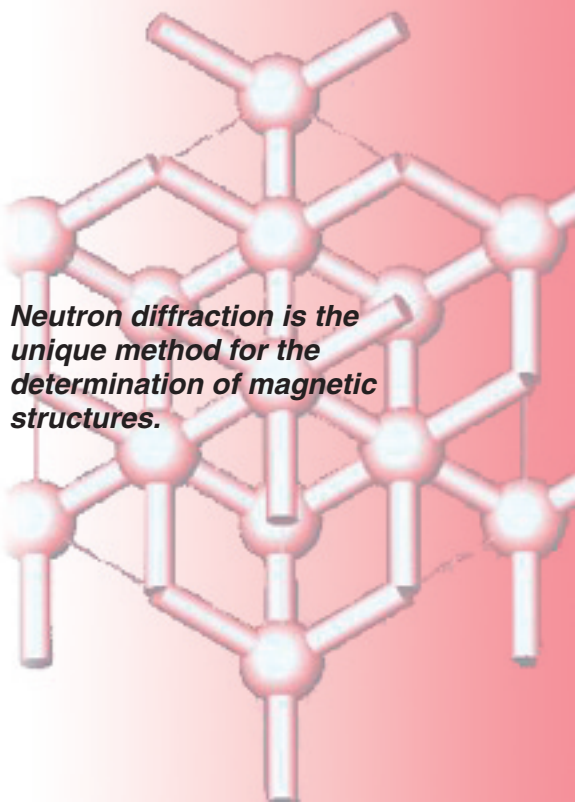
Table 1:
Frontier Research Areas in Solid State Physics

Dimensionality	Complexity	Structures and lattice effects	Non-equilibrium and time-dependent phenomena	New Materials
Quantum dot arrays Transport and magnetic properties in 1-d systems Domains walls, domains correlations, grain boundaries Surfaces and thin films	Interplay of spin, orbital and charge degree of freedom Coupled excitations Strongly interacting electron systems Flux line lattices Phase transitions, quantum critical points	Frustration Disorder, interfacial roughness Proximity effects Lattice modes Confinement	Fast response to external probes and fields Magnetic fluctuations and relaxations Tunnelling	Molecular magnets Interfaces/hybrid structures Self-organising molecular systems Novel magnets and superconductors Organic materials

II. The role of neutrons

In solid state physics, the degrees of freedom and interactions necessitate the use of a large variety of experimental methods. Bulk measurements of thermal and transport properties are invariably the first step, but several techniques are needed which are sensitive to the atomic environment and electronic structure, and which provide information on the spin state of the electron. Electron correlations in the solid state exist over vast ranges of spatial and temporal (energy) length scales. Local probes such as STM, and atomic spectroscopic studies using electromagnetic radiation provide crucial information. In addition, scattering methods (neutrons, x-rays and electrons) provide inter-atomic information on spatial correlations. However, the neutron has a unique combination of properties that make it indispensable for many problems in solid state physics. The de Broglie wavelength of thermal neutrons is on the same scale as inter-atomic spacings, allowing diffraction experiments to be conducted to locate the positions of atoms. Because of their mass, neutron have a rather low kinetic energy; they can be moderated and neutron beams are produced with energy in the range 0.1 meV to 10 eV, well matched to solid state excitations. (This is to be contrasted with x-rays, which at comparable wavelengths are much more energetic, in the keV range.) Because of their magnetic moment (spin), neutrons are sensitive to magnetic moments arising from electronic and nuclear magnetic

Because of a unique combination of properties, neutrons are a powerful and indispensable probe of solids.



Neutron diffraction is the unique method for the determination of magnetic structures.

moments. Because they are uncharged, neutrons penetrate deep into materials. Because they are weakly interacting (in contrast to electrons), measured scattering cross-sections can be compared directly to theory.

Neutrons have played a pivotal role in the investigations of phase transitions and co-operative phenomena, magnetism, structure (static and dynamics), as well as in many other fields. Particularly intriguing is the connection between phase transitions and theoretical concepts, such as symmetry breaking, order parameter, universality class, scaling and critical behaviour. In the past, the interplay between theoretical concepts and experimental observations concerning phase transitions has been extremely successful and many significant contributions have been made using neutron scattering.

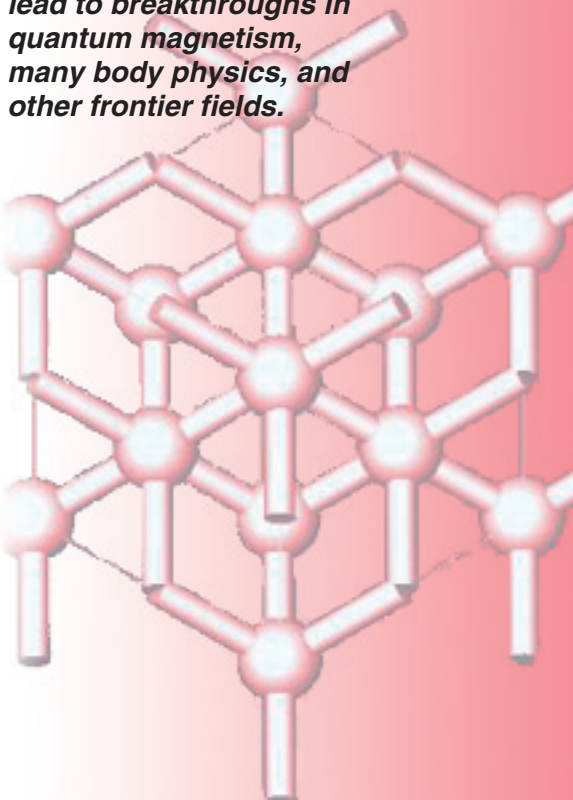
The interplay between neutron experiments and theory has driven the development of many new concepts in solid state physics.

A vast panoply of neutron techniques have contributed to this work. Neutron diffraction (from powders and single crystals) is a basic, but essential, technique, providing information on chemical and magnetic structures. Neutron reflectometry using polarised neutrons has given us a clearer picture of the growth and the physics of magnetic thin films and superlattices. Inelastic neutron scattering is the only probe that provides a complete picture of both structural and magnetic dynamics in solids. Emerging techniques include analysis of three dimensional polarisation, and the direct mapping of the full dynamical susceptibility over the entire Brillouin zone. All of these techniques suffer from the intrinsic low brilliance of neutron sources; as a result, the materials studied must have a sizeable volume and/or sizeable scattering density. The (lateral and vertical) spatial resolution is restricted to a few 100 μm while the temporal resolution is in the 0.1 sec range. The advent of the ESS will offer entirely new capabilities to explore spatial and temporal properties of condensed matter with μm and ms resolution, respectively.

Low intensity continues to be the major limitation of neutron research. Due to its high flux, the ESS will open up entirely new opportunities.

Here we emphasise a few topics where neutron scattering techniques are expected to play a major role. In ***magnetism***, significant advances are expected in synthesizing molecular and organic magnets, i.e. solids built from structurally well-defined clusters containing magnetic ions in a complex environment. These are of both fundamental importance, and with respect to potential application in magnetic storage devices. New developments are also expected in exploring novel magnetic phases and their dynamics in low-dimensional systems. The study of ***phase transitions*** will continue to be a major field of research with neutron based techniques. Systems of high complexity, exhibiting extreme many body effects (e.g. unconventional superconductivity) and low dimensional features are known and expected to undergo a large variety of phase transitions. Their exploration using neutron techniques will provide crucial insights into the microscopic mechanisms causing these phenomena. Of high current and most likely future interest is the relationship between spin polarisation and transport of conduction electrons in specially tailored materials, ***spintronics***. High

Research at the ESS could lead to breakthroughs in quantum magnetism, many body physics, and other frontier fields.



intensity neutron beams will play a central role in elucidating the spin polarisation and dynamics of these electrons.

III. Future opportunities – flagship areas

The ESS will lead to breakthroughs in three distinct ways:

a) to allow scientists to address new problems, and to ask new questions.

b) to provide new tools to tackle problems at the research frontiers.

c) to offer high quality experimental data for unambiguous discrimination between theoretical models.

In the following, we present selected flagship areas which are representative of the topics listed above.

Dynamics of superlattices, thin films, wires and dots

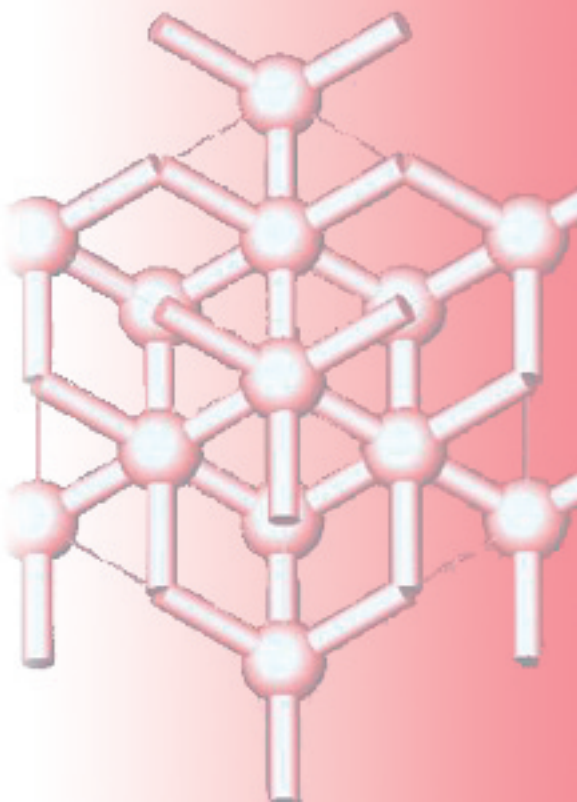
Following the discovery of giant magneto-resistance in 1988, the physics of micro- and nano-structured magnetic materials has become a field of intense activity. Thin films and superlattices, as well as wires and dots are now extensively studied for their fundamental properties and their potential applications in systems like sensors and magnetic random access memory devices (MRAMS). Understanding the dynamics of these systems will continue to be a key challenge. In contrast to Brillouin light scattering and ferromagnetic resonance techniques, neutron scattering gives access to the whole Brillouin zone.

Neutron beams at the ESS will provide maps of the magnetic polarisation and dynamics of nano-structured materials and devices.

Brillouin light scattering has provided the first dispersion curves in magnetic dots, but is limited to 30 GHz. Until now, there is no theory able to explain the excitations measured in dots, even in simple NiFe square dots. An experimental input at higher frequencies appears to be essential in understanding the dynamics of these systems. A reflectometer at the ESS will offer the capability of measuring spin wave spectra in very thin films, wires and dots, and will certainly have an important impact on the field of nano-magnetism.

In contrast to alternative techniques, neutrons provide access to a wide range of energies and momenta.

The observation of magnetic inelastic scattering from superlattices is presently at the limits of neutron technology. An interesting experiment has been performed recently at the ILL on a Dy/Y superlattice, where the effects of folding on the inelastic response function due to the superlattice periodicity have been observed. Such neutron experiments allow the exchange coupling parameters, both within a single layer and between the layers, to be deduced. However, this will require a considerable increase in intensity, coupled with better resolution, especially if technologically important films such as transition-metal superlattices are to be examined and understood.



Molecular magnets

A typical example of a molecular magnet is Mn_{12} acetate with total spin quantum number S = 10, giving rise to thousands of excited spin states which can only be disentangled by high resolution neutron spectroscopy. For instance, the lowest lying group of spin states comprises (2S + 1) = 21 levels of energies $\eta\omega \leq 1.2$ meV as shown in Figure 1 below. Mn_{12} acetate exhibits quantum tunnelling between these spin states which can be tuned in a controlled manner by an applied magnetic field. This opens the way to a novel class of information storage systems on the molecular level. Unfortunately, the quantum tunnelling in Mn_{12} acetate is restricted to temperatures of a few Kelvin. The search is on for materials that would preserve the virtues of Mn_{12} acetate at liquid nitrogen temperature.

Molecular magnets could serve in atomic-scale information storage systems. Neutron scattering is a unique probe of their excitation spectra, whose accuracy will be tremendously enhanced by the ESS.

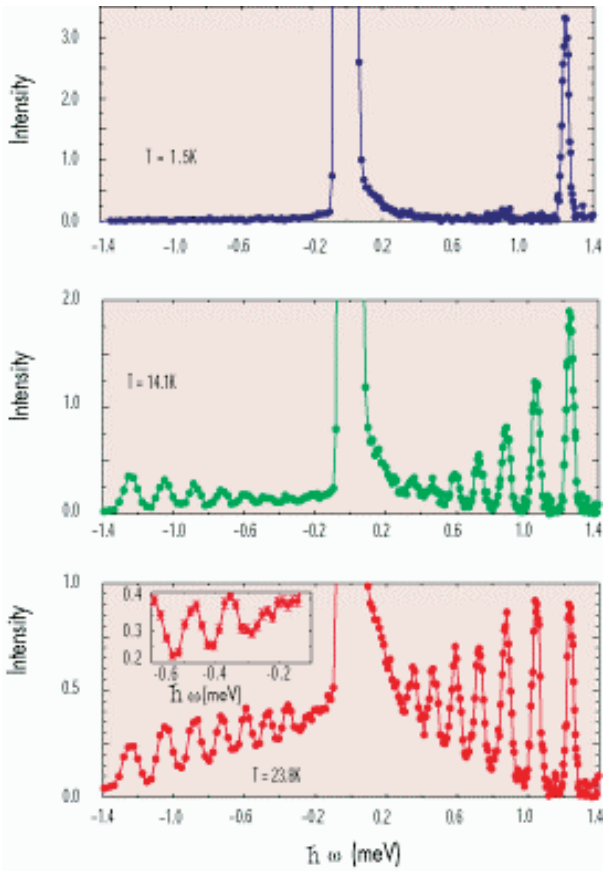


Figure 1: Energy spectra observed at three different temperatures in Mn_{12} acetate (Courtesy of I. Mirebeau).

Spin density waves in organic materials

Among the low-dimensional electronic systems, the charge transfer Bechgaard salts $(TMTTF)_2X$ and $(TMTSF)_2X$ (X = PF_6, AsF_6, SbF_6, SCN) show the richest phase diagrams with almost all known electronic phases: a metal, a paramagnetic insulator, spin and charge density wave states, a spin-Peierls state and finally an unconventional superconducting state.

The ESS may enable the first direct observation of the tiny magnetic moments which are central to current theories of organic conductors.

Other salts in the same family have been shown to exhibit the quantum Hall effect. The phase diagrams have been mapped out mostly based on transport and specific heat data as well as NMR results.

For the spin density wave (SDW) phases, detailed NMR predictions exist concerning the wavevector of the modulation and the amplitude of the ordered moment. Yet, so far, direct neutron evidence for a SDW superlattice peak is missing, due to a combination of the weak magnetic moment (~ $0.08\mu_B$), the unfavourable magnetic form factor and small sample sizes. Experiments of this type will become possible with the intensity available at the ESS, allowing a direct determination of the SDW amplitudes and periodicities, and thereby opening up an entirely new area of research.

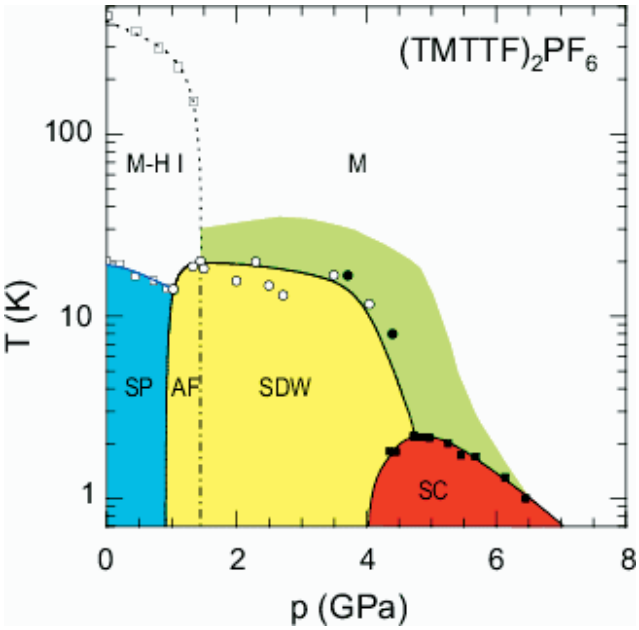


Figure 2: Phase diagram of the Bechgaard salt $(TMTTF)_2PF_6$ deduced from resistivity (Courtesy of D. Jaccard).

Revealing exotic interactions

Theoretically predicted (but hitherto unobserved) interactions in solids will be discovered and exploited at the ESS.

The properties of magnetic materials are usually described in terms of bilinear spin interactions. Neutron spectroscopy with its dipole selection rule $|\Delta M| = 1$ has been the technique of choice to measure the magnetic excitation spectrum and thereby, to allow the direct determination of the magnetic exchange coupling constants. However, there are numerous examples such as molecular magnets, high T_c cuprates and f-electrons compounds where higher-order interactions (e.g. quadrupolar, octupolar, three- and four-body exchange) are relevant, but their sizes could so far not be determined directly. In principle, neutron scattering allows the direct observation of higher-order term transitions. However, the associated transition matrix elements are typically two orders of magnitude smaller than for dipolar scattering. Such novel experiments would be made possible by the ESS.

Coupled excitations

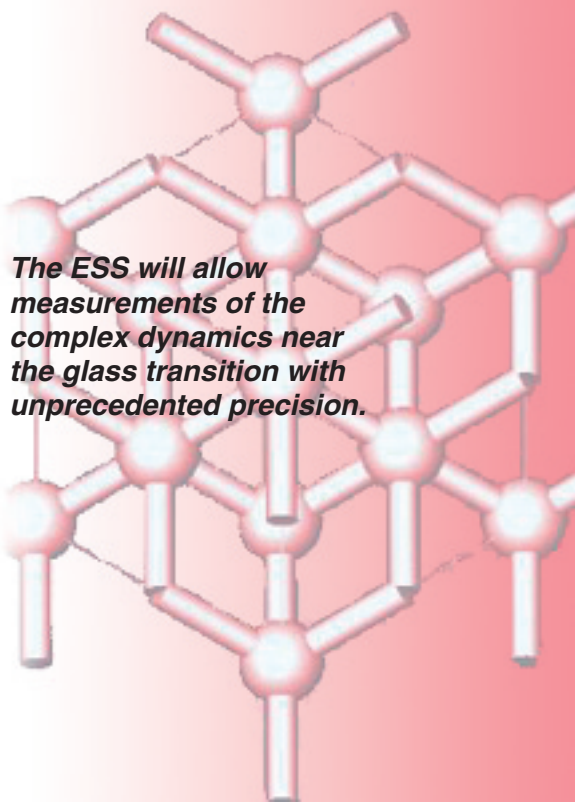
Inelastic neutron scattering at the ESS will open a new window on the electronic structure of metals and superconductors.

Excitation phenomena in solids can be classified into single particle continua and collective modes. Because of intensity constraints, neutron scattering experiments have been almost exclusively limited to collective modes. Recent investigations of two-spinon continua in insulating one- and two-dimensional quantum magnets are pushing the limits of current sources. At a chopper spectrometer at the ESS with wide reciprocal space coverage, detailed maps of single particle Stoner continua in metals and superconductors will be obtained up to energies of the order of the Fermi energy. In systems where correlation effects are strong (which are currently at the forefront of condensed matter science) it will be possible to extract a wealth of information on the band dispersions, Fermi liquid parameters, superconducting coherence effect, etc., that is currently inaccessible. The high neutron flux at the ESS will also enable high resolution measurements of the intrinsic lifetimes of collective modes over the entire Brillouin zone. Although predictions for the lifetimes of magnetic and lattice vibrational excitations (for instance, due to electron-phonon scattering) have been available for many years and are becoming ever more accurate, they could thus far be tested only in a few special cases.

Physics of defects at the dilute limit

Defects are ubiquitous in solids, and high intensity neutron beams at the ESS will provide incisive information about their microscopic structure.

The increasing sophistication of “first principles” theoretical calculations of the fundamental electronic structure, total energies and atomic short range order, places stringent demands upon the accuracy of experimental measurements of the extended atomic and magnetic defects around impurity atoms in metals, alloys and compounds. Experimentally, information on this problem can be obtained only from diffuse neutron scattering experiments, with polarisation analysis. The associated cross sections are extremely small and counting times are often prohibitively long. Moreover, such experiments should ideally be carried out at extreme dilution to circumvent the often intractable problem of non-linear superposition of overlapping defects. These experiments are crucial for a full solution of the defect problem and experimental corroboration of the most sophisticated of our “first principles” band theoretical calculations, but is not feasible at present neutron sources.



Spin glass dynamics

The ESS will allow measurements of the complex dynamics near the glass transition with unprecedented precision.

It has been said that the deepest and most interesting unsolved problem in solid state physics is probably the nature of glass and the glass transition. Indeed the status of the glass transition as a true thermodynamic transition is still questioned. Spin glasses provide a simple analogue of the structural glasses yet the *accurate* measurement of relaxation processes in spin glass systems is at the very limits of what is feasible at present using spin echo facilities. A wider Fourier time, coupled with significantly improved counting statistics, is required to determine the precise functional form of the relaxational dynamics, both for comparison with structural

glasses and to discriminate between the proposed theoretical models. In addition the measurements should also be performed over a wide range of magnetic dilution, the lower ranges of which are entirely inaccessible at present. The implications of such studies are profound, as many of the spin glass relaxational models are finding applications in areas as diverse as virus mutation, protein folding and the travelling salesman problem.

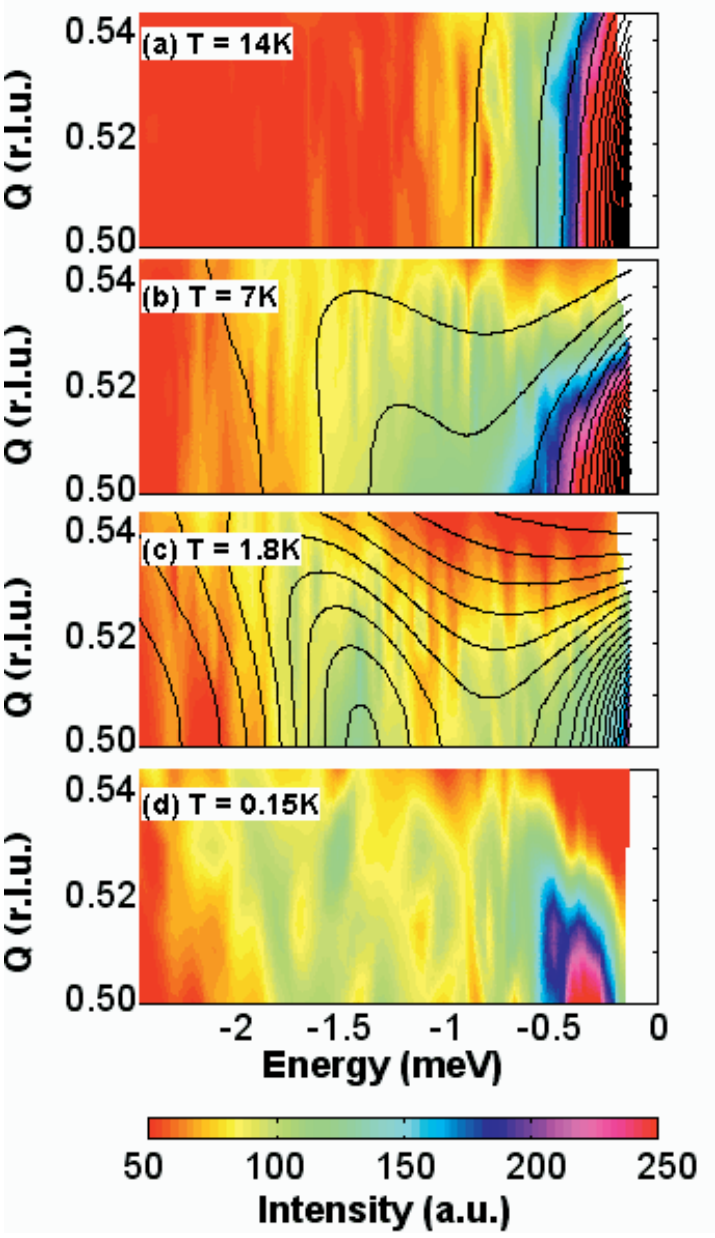


Figure 3: Temperature dependence of the magnetic response of UPd_2Al_3 near the superconducting phase transition. Above the phase transition, there is coexistence of quasi-elastic scattering and dispersive modes. In the superconducting state the quasielastic scattering is replaced by a low-lying inelastic mode (data taken from (Courtesy of N. Bernhoeft).

Quantum phase transitions

Of special interest are situations where either the competition between different interactions prevents the system from readily adopting a well-defined ground state, or where a restriction in spatial dimensionality does not allow for long-range ordering phenomena. Interactions of similar, medium or large magnitude may lead to very complicated phase diagrams. Cases where all the interactions are of similar strength and weak are very challenging. Here the situation may occur that the phase transition only sets in at T = 0K, in the quantum critical regime. A related example would be the important and technically challenging experiment on a material such as the recently discovered ferromagnetic superconductor UGe_2. Inelastic scattering experiments need to be performed at low temperature (< 0.5 K) and at pressures of up to 3 GPa.

Experiments at the ESS will set new benchmarks for extreme conditions of external pressure and magnetic field, thus providing important insights into zero-temperature phase transitions driven by quantum fluctuations.

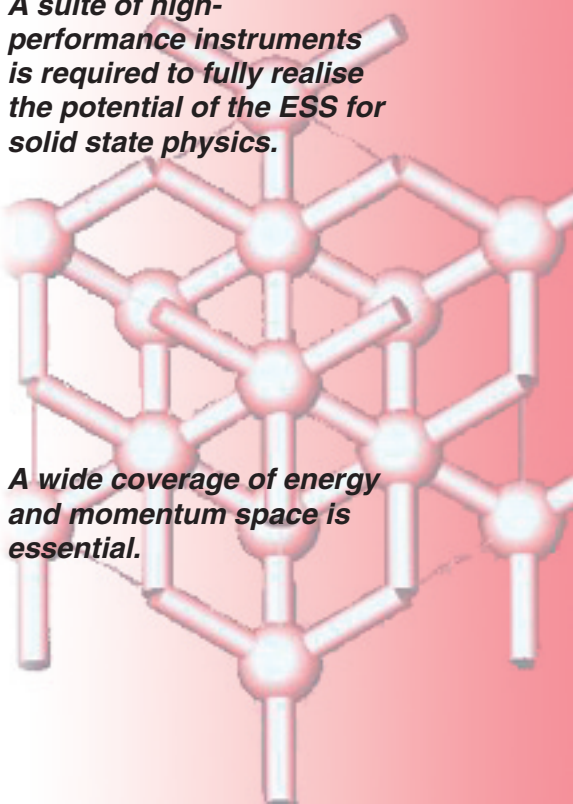
The key requirement is to map out the inelastic response function over a wide range of momentum space and energy as a function of temperature, external pressure and applied magnetic field. While neutron scattering can make unique and essential contributions to our understanding of the mechanisms underlying these phenomena, the sample volumes that can be subjected to these extreme conditions are necessarily small. At current neutron sources, inelastic scattering studies are hence restricted to a few special cases. Detailed investigations of the dynamic aspects of magnetic field and/or pressure induced quantum phase transitions require the ESS.

It should be stressed that in such experiments the positions in momentum space where the maxima will occur are unknown, so that a wide “mapping” technique is required. Many new materials call for this approach; unavailable at present, but a planned development at the ESS.

IV. Instrument requirements at ESS

The requirements for instrumentation in solid state physics are based upon two principle demands: that of probing (Q,ω) space for excitations, and that of providing a detailed structural (atomic and magnetic) characterisation of the sample. Whilst the former demand can only be met by a suite of inelastic scattering spectrometers, the latter must be met by a range of rather disparate total- and elastic scattering instruments, namely powder and single crystal diffractometers, diffuse scattering instruments and reflectometers. In all cases polarised incident neutrons and polarisation analysis are either essential or a distinct advantage, and the instruments should be capable of accepting extreme sample environments (e.g. pressure, magnetic field, temperature etc.).

A suite of high-performance instruments is required to fully realise the potential of the ESS for solid state physics.



The key scientific topics we have highlighted demand a coverage of Q space from $0.1 \leq Q \leq 12 Å^{-1}$ and an energy transfer range of six orders of magnitude, from μeV to eV. This can be achieved through an instrument suite consisting of a

A wide coverage of energy and momentum space is essential.

backscattering spectrometer, a variable resolution cold chopper spectrometer, thermal and high energy chopper spectrometers, and a constant-Q spectrometer. The resulting coverage of Q-ω space is illustrated in the diagram below.

A single crystal diffractometer is an essential component of the suite for determination for the structural and magnetic order and for crystallographic studies of multilayer systems. Polarised incident neutrons and high magnetic fields at the sample position will enable magnetic spin density determination.

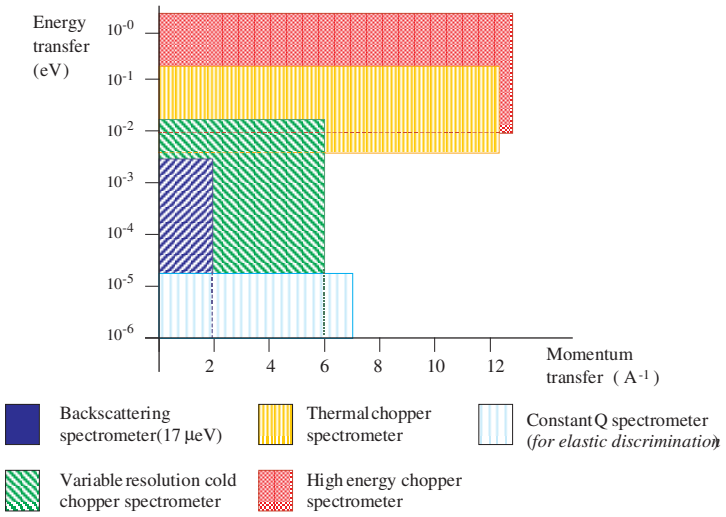
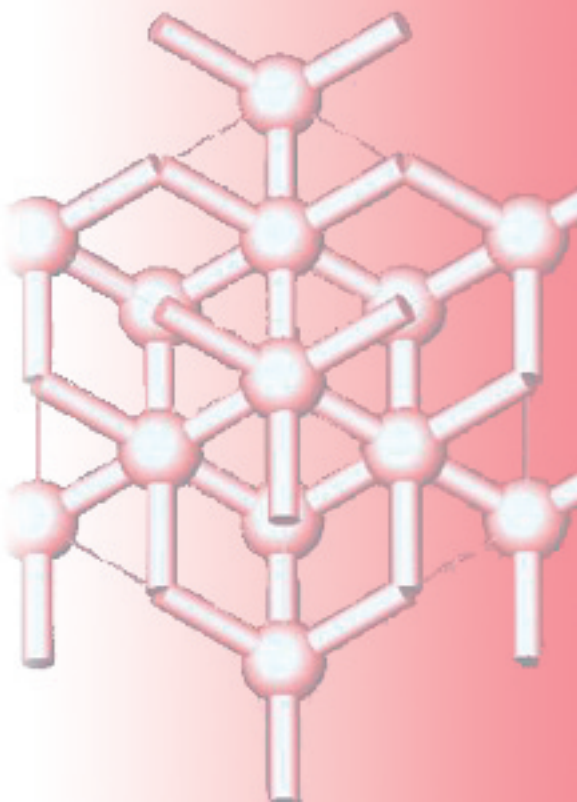


Figure 4: Required coverage in the Q-ω space.

Solid state physics places several conflicting demands on powder diffractometry. Firstly the growing complexity of magnetic structures (e.g. spirals and spin density waves) studied by neutron diffraction requires high resolution (Δ d/d > 0.1%) across a Q-range from a lower limit of at least 0.3 $Å^{-1}$ to 12 $Å^{-1}$. The efficient mapping of the evolution of magnetic and structural phases in parameter space defined by pressure, temperature and magnetic field is best met by a diffractometer following the GEM design at ISIS.

The study of the structures of artificial films, multilayers and mesoscopic structures requires a reflectometer optimised for intensity rather than resolution. Polarisation analysis and surface capabilities are essential. A dynamic range of 10^8 up to a Q of 0.512 $Å^{-1}$ represents a real advance in reflectometry and should be considered to be a design goal.



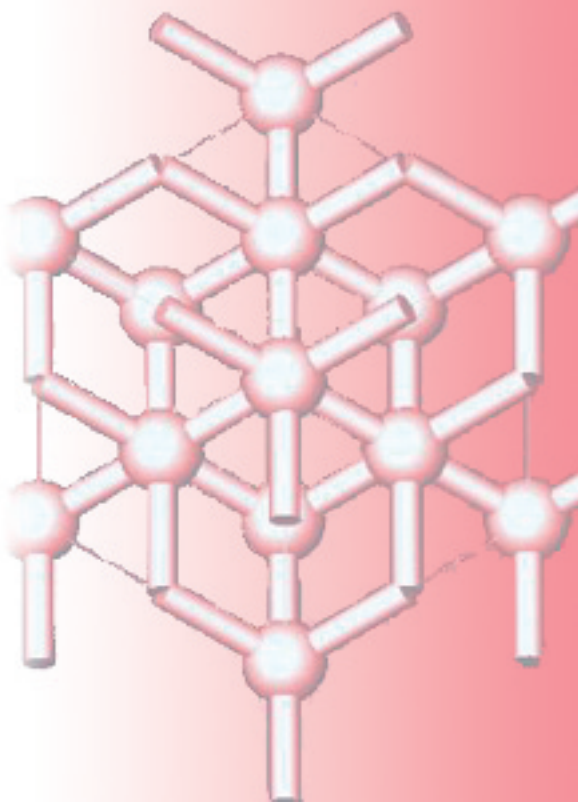
Our conclusions are summarised below:

Priorities for instruments:

First Priority:
Chopper Medium Resolution
Chopper High Resolution
Cold Chopper
High Intensity Reflectometer + Energy Analysis
Magnetic Powder Diffractometer

Second Priority:
High Resolution Powder Diffractometer
Chemical Single Crystal
High Resolution NSE
Medium Resolution Backscattering
Diffuse Scattering Diffractometer
Constant Q

The instruments needed for solid state physics are best placed at the 50 Hz 5 MW and $16^2/_3$ Hz 5 MW target stations.



Achievements of neutrons in solid state physics

- Almost everything which is known about magnetic structures of electrons– from the early demonstration by Shull of antiferromagnetism in simple systems, to the complex magnetic structures being developed for new magnetic materials – has come from experiments with neutrons. Complex magnetic orderings which include spirals, fans, and cycloids, and asymmetric magnetisation have been unravelled by neutron diffraction experiments.

- Pioneering experiments have been made on nuclear spin ordering in Cu and Ag, which are antiferromagnets below 70 nK and 600 pK respectively. These observations of one of the weakest interactions in a solid is a significant step towards defining the ultimate ground state of electronically non-magnetic materials.

- Polarised neutron reflectometry of surface and interface magnetism in thin films and multilayers is providing technologically relevant information. Important examples include the observations that the magnetic order can propagate through non-magnetic layers and that giant magnetoresistance is not necessarily associated with antiferromagnetic coupling. Magnetic roughness can be separated from the chemical roughness.

- Neutron diffraction provided the first microscopic evidence for flux line lattices in conventional superconductors and played a major role, especially at higher magnetic fields where no other technique can image flux lines, in accounting for the large dissipation in the high-T_c materials.

- Neutrons have provided the definitive crystal structures in high temperature superconductors, which have served as the basis of all considerations of the superconducting mechanism and have led to production of better quality samples. Neutrons have precisely located the positions of the oxygen atoms, where the charge carrying holes reside. Of particular importance has been the demonstration of the charge transfer concept of hole doping with oxidation, and that the superconducting temperature was related to the evolution of structural order. Neutron spectroscopy has provided unique information on the nature of magnetism in high temperature superconductors, on the interplay between magnetic fluctuations and superconductivity and on the role of the lattice dynamics.

- Inelastic neutron scattering has provided unique information on the interaction and anisotropy energies which determine the Hamiltonian operator. Extensive measurements on magnons and crystal field excitations over a wide range of frequency and momentum space have revealed e.g. the interaction of the spin waves with the single particle excitations (Stoner continuum) in iron. Similarly, measurements of phonons and their density of states have improved our understanding for example of the role of phonons in the martensitic phase transition in the IVb metals.

- Major contributions to our understanding of model systems for statistical physics in one, two and three dimensions, include the verification of the Haldane conjecture, determining the properties of the Haldane gap, and the discovery of solitons, as the characteristic elementary excitation of strongly non-linear magnetic systems. This also includes the observation of solitons in solids and the demonstration of all the expected, but previously not observed, properties.

- Outstanding results from space and time-dependent studies of correlated electron systems include proof of the importance of correlations for the spin dynamics and the antiferromagnetic character of the spin correlations. The interplay of electronic degrees of freedom (charge, spin and orbital order of electrons) has been amply evidenced.

- One of the most fundamental questions in condensed matter physics concerns the liquid and solid phases of ^{3}He and ^{4}He. Experiments with neutrons have provided unique and important information on, for example, the Fermi liquid parameters of ^{3}He, the Bose condensation of liquid ^{4}He and the magnetic structure of solid ^{3}He. The importance of understanding these quantum systems was highlighted by the award of the 1996 Nobel prize in physics for the discovery of superfluidity in ^{3}He by Lee, Osheroff and Richardson.

4.2 Materials Science and Engineering

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Abstract
Materials science and engineering provide the keys to future technologies, economic wealth and sustainable growth. They are also the keys to mastering many of the challenges for the next generation, such as the development of new energy sources and the reduction of pollution. Because of these many different aspects, materials scientists and engineers use a large number of experimental techniques. Neutron scattering has always been an important tool for the provision of structural information on the atomic scale and for the understanding of dynamical properties of solids and liquids. However, in the early days of neutron scattering this technique was an exquisite tool in the hands of a few specialists. Because of its sustained success in providing unique answers to materials science problems, neutron scattering has become increasingly popular among materials scientists and likewise engineers. Today the advance of numerous materials science topics relies heavily on the availability of strong neutron sources. The strongest neutron sources available at the present time are often at the limit of their capabilities. Presently the data acquisition is often too slow for in-situ and real-time studies of dynamic changes and process monitoring. Modern technologies demand information from smaller sampling regions, sometimes buried in larger component volumes or environmental chambers, samples in complex environments, samples in real time evolution and from samples in extreme fields. The next generation of high power pulsed neutron sources will dramatically alter the nature of the experiments which are possible, allowing for the first time investigations of materials in real time, with realistic dimensions and in real conditions. Improved data acquisition will enable pump-probe type experiments, to study the spin structure in magnetic nanostructures for information technology devices, and to provide high resolution three dimensional maps of stresses and textures of engineering components during fatigue cycling. In addition, the time structure, which is unique to pulsed sources, will be exploited, for example in chemically sensitive radio- and tomographic imaging. Therefore, the ESS will be an extremely powerful tool for the analysis of advanced materials, which are of scientific, commercial and practical interest. In this respect the ESS will be highly significant for maintaining the competitive edge of materials science within the European Union as compared with that in other parts of the world.

In order to capitalise on the opportunities the ESS offers for neutron research in the area of Materials Science and Engineering, several instruments are required, including an engineering diffractometer for stress-strain analysis, a high resolution and focussing small angle scattering instrument for in-situ kinetic measurements of microstructure and pore growth, a polarised neutron reflectometer for the analysis of magnetic nanostructures, a tomography and radiography instrument for imaging large industrial components, and a high resolution/variable resolution cold chopper spectrometer for the analysis of the internal dynamics of atoms and molecules.

I. Introduction

Materials science development during the past 50 years.

The road to our present day use of a vast variety of novel materials and engineering processes is marked by an outstanding array of discoveries, inventions, and theoretical insights. The start of modern materials research dates back about 50 years. Since then the transistor has been invented, superconductivity explained, high temperature superconductors and the quantum hall effect have been discovered, semiconductor lasers and magnetoresistive reading heads for hard disks have been developed, and finite element modelling has revolutionised structural and process design. New structural materials have become available such as high temperature and corrosive resistant steels, light weight foam metals, carbon fibre reinforced materials, shape memory alloys, superalloys, photo-voltaic materials, energy conversion and storage materials. Material processing techniques have

seen similar advances through new friction based solid state welding techniques, surface hardening treatments, and corrosive resistant coatings. At the same time, the understanding of more traditional building materials such as concrete has dramatically improved. These are but a few examples to illustrate the remarkable progress materials scientists have achieved in recent years.

II. The role of neutron scattering

Neutron scattering contributes to materials science and engineering.

Neutron scattering has established itself as one of the most important tools for the analysis of materials. Metals, ceramics, and their composites, semiconductors, superconductors, nanophase materials, liquids, polymers, paints, lubricants, concrete, coal, wood, bones and biomaterials as well as many other materials have been analysed with great success using neutron scattering. The results are accessible through publications and data banks. Engineering developments and applications draw heavily from this body of work.

Neutron scattering answers questions in materials science and engineering.

In contrast to other experimental techniques, neutron scattering provides access not only to the positional order of the atoms but also to their dynamics. Depending on the material chosen, the motion of atoms in thermal equilibrium ranges from local vibration of atoms in solids, diffusion of atoms in liquids and solids, creeping motion in polymers, rotation of molecular sidegroups, and tunnelling of atoms through potential barriers. As a result of these investigations, the elastic properties of materials are very well understood, the danger from hydrogen embrittlement of steels can be predicted, and the dependence of the lifetime for metal – semiconductor junctions on the operation temperature can be determined.

Examples of the contributions of neutron scattering to materials science and engineering problems.

In many situations neutron scattering is an indispensable tool, since no other method provides information within the same space – time window. Without neutron scattering our knowledge of the vibrational properties of atoms, the diffusional dynamics of liquids and polymers, the jump vectors of interstitials in host lattices, and the rotational dynamics of molecules would be much more limited. Without neutron scattering we would not have information on oxygen ordering in high temperature superconductors or on hydrogen vibrational potentials in metals. Furthermore, neutrons have a magnetic moment that interacts with the microscopic moments in solids and liquids and with magnetic field distributions in superconductors. Without neutron scattering we would not know about the ordering of magnetic moments in ferro- and antiferromagnetic materials, flux ordering and melting in superconductors, about the vortex states in low dimensional magnetic systems, and spin disorder in frustrated magnetic alloys.

Dynamical properties of liquids and solids.

Oxygen and hydrogen ordering.

Ferro- and antiferromagnetic structures.

Non-destructive analysis of engineering components.

Neutrons can also penetrate many centimetres through engineering materials, allowing non-destructive studies of large components and samples in complex environmental or processing chambers. Neutrons are thus particularly well

suited to non-destructive studies of components in their as-fabricated and in-service condition such as engine parts. At the same time, neutrons take a volume average such that results are relevant for the properties of real materials.

Volume average

Whenever new materials become available, neutron scattering plays a key role in providing a microscopic understanding of their structural, dynamic and magnetic properties. Such understanding is vital for the development of these materials in technological applications and for their implementation into the production process. This is shown by the improved insight that has been gained through neutron scattering into high-T_c compounds, into Giant Magneto Resistance (GMR) and Colossal Magneto Resistance (CMR) materials, into fullerenes, and other materials of present interest. This will undoubtedly also be the case for any new class of materials that are discovered.

Novel materials

Neutron diffraction also provides important insights into new and established processing methods, from the stresses caused by new joining techniques to the molecular alignment of polymer molecules during plastic manufacture.

Processing and monitoring

Although neutron source particle fluxes are lower than those of the new generation of high brilliance synchrotron sources, neutrons nevertheless remain the preferred tool in many areas of materials science, because of the quality of the information each particle delivers. For example, neutron scattering is preferable in many cases over x-ray scattering because of a lower background and a cleaner signal, particular for magnetic scattering. Furthermore, neutron scattering provides absolute structure factors and total magnetic moments for direct comparison with theory. Only neutron scattering provides a strong contrast by isotope substitution, which is best known for the case of hydrogen and deuterium, but is also extremely useful in other fields such as the study of binary metallic alloys. Furthermore neutrons provide a probe of bulk properties deep within samples, for instance in engineering strain measurement.

Intensity limitation

Neutron advantage

Contrast variation with isotopes.

III. New opportunities with neutrons at the ESS

With the ESS gain factors exceeding two orders of magnitude are projected. The increased flux and intrinsic time structure will allow the development of new materials science investigations with innovative instrumentation inconceivable on existing neutron sources. With the provision of these improvements, a huge impact on materials science is expected:

More intensity means faster data rate and new opportunities.

- Time resolved experiments, with second to milli-second resolution;

- Higher spatial resolution to the important sub-millimetre region for monitoring residual stress, high pressure and high temperature experiments, and for detecting of interfacial diffusion and reactions at interfaces of metal and

organic multilayers;

- In-situ real time experiments, for example nucleation and recrystallisation of undercooled liquid alloys, the ageing and fatigue of alloys under cycling conditions;

- Analysis of the spin structure and spin fluctuations of laterally patterned arrays or magnetic nanostructures;

- Routine determination of 3-D maps of stress and texture within engineering components;

- Neutron tomography for the production of 3-D images of machine parts under working conditions in real time, and with structural sensitivity;

- Pulsed radiography to study fast time dependent phenomena with isotope sensitivity.

Chief areas of materials science and engineering and new opportunities at the ESS.

The sustainable growth of society can only be achieved if new materials and material combinations are explored on all length scales and time frames, and under real conditions. The following examples are identified as important areas that can be foreseen to play a vital role within the next decade and beyond. These areas will benefit tremendously from neutron scattering experiments at the ESS, but represent only a small number of the potential applications of neutrons to materials science and engineering.

Lubrication

Neutrons provide insight into the functioning of lubricants via the analysis of the macroscopic flow and the dynamics on a molecular level.

Ineffective lubrication leads to premature wear and failure of mechanical parts, which causes an estimated damage in the USA of about 6 % of the gross national product [1]. It has been demonstrated that neutrons can unravel the structure and dynamics of lubricants in moving engineering parts [2,3]. For the first time the dynamics on all length scales, embracing macroscopic flow to molecular diffusion under real loads, can be studied with neutron scattering, opening a new approach for the understanding the lubricant flow dynamics, which will undoubtedly lead to the development of new lubricants for extreme conditions. In particular, with neutron scattering it is possible to determine the velocity gradient of liquids under shear load and to distinguish between Newtonian and non-Newtonian liquid flow. Moreover, with neutron spectroscopy the macroscopic velocity gradient can be related to the excitation of internal rotational and librational modes of the molecules. Presently the lubricant layer in these measurements has a minimum thickness of about 0.3 mm, far more than realistic scales of industrial interest. Studies of reduced film thickness relevant to real applications (~ 10 μm) will only become feasible at the ESS with the improved intensity available. Moreover, the intrinsic time structure of the ESS is ideally suited for time-of-flight spectroscopy studies of the flow dynamics and internal excitations.

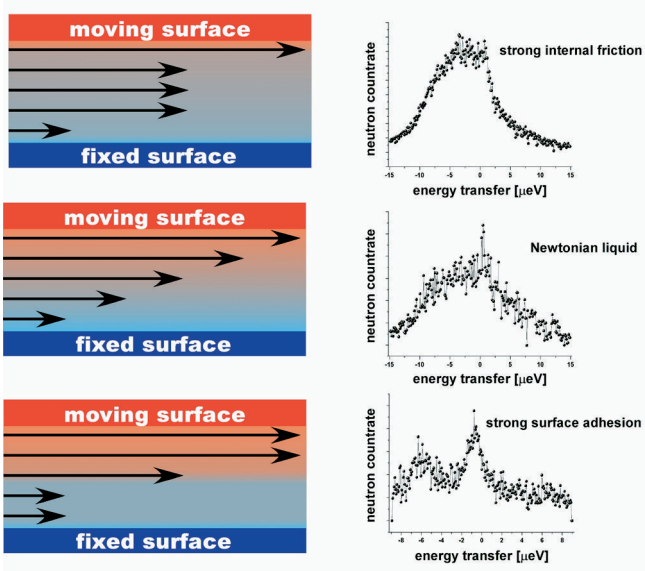


Figure 1: Schematic velocity profiles for liquids under shear load are shown to the left. The middle left panel shows the classical Newtonian profile with a linear velocity gradient between a fixed and a moving plate. Lubricants often deviate from this behaviour. Either the lubricant may stick well to the surface of the plates, while the interior layers are weakly coupled, yielding a velocity profile as shown in the top panel; or the interaction between the liquid layers is strong, leaving only a thin liquid layer with an intermediate velocity. Neutron scattering experiments test the velocity profiles and provide simultaneously information on the internal dynamics of the molecules and their frictional losses. The right panel shows neutron scattering spectra for a lubricant flow between a disk at rest and a disk with constant angular velocity. The top spectrum is for a liquid with strong internal friction, the middle for a liquid with a Newtonian velocity profile, and the bottom for a liquid with strong adhesion to the surface. (courtesy of Wollf, Magerl, Hock, Frick, Zabel [3] and ILL report 2002).

Mechanism of deformation and damage

Neutron scattering provides unique micro-mechanical data from deformed materials under real conditions.

Describing materials deformation and understanding the mechanisms involved are a vital part of engineering science. Neutron scattering provides unique micro-mechanical data. The neutron sampling size is particularly well matched to the scale of engineering stress-strain concepts. However, going beyond model systems and timescales to real fatigue cycling conditions and components requires data acquisition rates, which are inaccessible today. ESS will enable the assessment of real scale components on realistic time scales. For example, new solid state joining techniques require more accurate information about the generation of residual stresses that will add to in-service stresses foreshortening life. Furthermore, finite element models have become the main method for design and assessment of engineering structures [4]. These models cannot be developed reliably without accurate information to validate them. Neutron diffraction is the only technique that can do this, as it provides data from deep inside most engineering materials. Model validation has become vital to modern industry, as timescales for technique and component design process, from drawing board to final uptake, become shorter.

Neutron diffraction can provide information from deep inside most engineering materials.

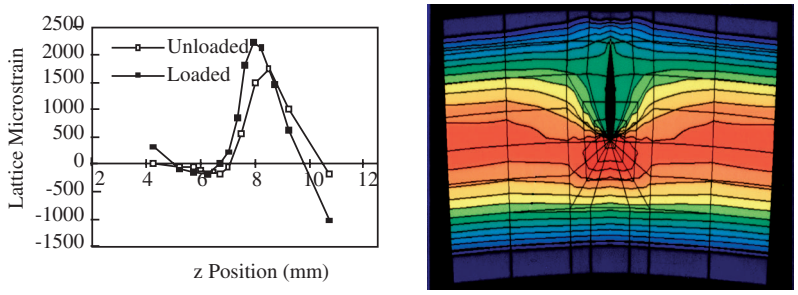


Figure 2: Crack tests are performed on engineering metal bars that are subjected to fatigue cycling with a varying load. The left figure shows the elastic crack opening strain along the line of a crack in an aluminium bar as measured by neutron diffraction; note how the crack tip stress is significant even when the crack opening load is removed. The right panel reproduces the associated finite element model, showing how prior residual stresses cause the crack to close near the top surface upon unloading; this closure holds the crack tip open (red: tensile, blue: compressive stress). In the future this type of experiments can be carried out in real time under real fatigue cycling conditions with the pulsed spallation source. (courtesy of Korsunsky, Fitzpatrick and Withers [5]).

Energy storage and conversion devices

Society is becoming increasingly dependent on energy storage and conversion devices, such as batteries, fuel cells and solar cells. New rechargeable cathode and anode battery materials should provide a higher energy density, and be environmentally friendly and cheap, in order to facilitate large-scale applications of renewable energy.

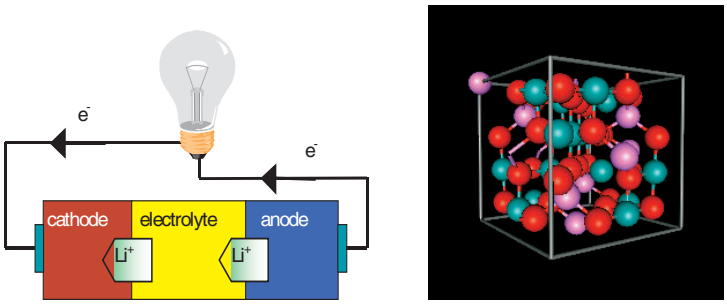


Figure 3: Schematics of a Li - ion battery to the left. Neutrons provide the essential information on the position and mobility of the Li^+ ions in environmentally more friendly and high energy density $LiMn_2O_4$ batteries. Neutron scattering results have recently proven the coupling between structural transitions and ordered ionic charge localisation in the $LiMn_2O_4$ spinel structure, shown to the right [6,7]. The Li ions are shown in purple, the Mn ions in blue-gray, and the oxygen ions in red. At the ESS the charge and discharge of the battery could be monitored on the atomic scale under real operating conditions, which will help to improve rechargeable battery lifetimes.

Optimisation of such materials and devices relies on in-situ characterisation under operating conditions for improving the

performance and durability of such devices. ESS will take structural and dynamical information from tailored laboratory model systems to real time scales and conditions. In the drive to replace $LiCoO_2$ with more environmentally friendly and high energy density $LiMn_2O_4$ batteries, neutrons provide information on the movement of the Li ions and associated structural changes. Such studies take advantage of the neutron´s inherent ability to monitor light atoms, even in the presence of heavy atoms.

Information technology

Greater demands are being placed on new magneto-electronic devices for faster data handling and higher density non-volatile data storage. The device applications (MRAM, GMR heads, spin transistor) depend on control over the switching behaviour, hysteretic losses, and bit life times [8].

Spin valves for reading heads and for non-volatile magnetic access information storage devices.

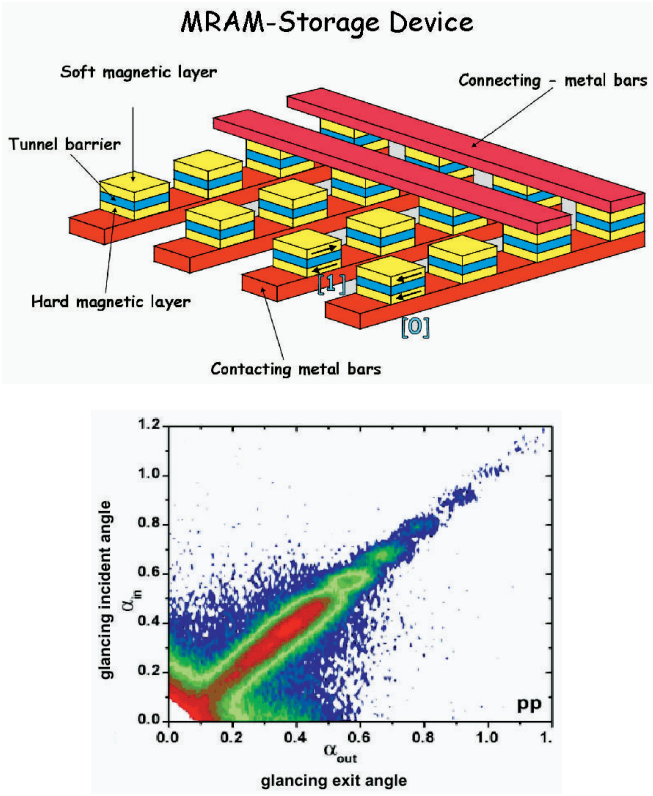


Figure 4: The upper panel shows a design of a non-volatile MRAM. The device relies on the switching of magnetic domains with current pulses in the read and write lines. Spin valve structures are used for controlling the magneto-resistance, that depends on the relative orientation of the magnetisation in the top and bottom ferromagnetic electrodes. Neutron scattering is necessary to analyse the spin structure deep inside spin valve systems. One essential part of a spin valve system is a ferro-/antiferromagnetic contact for providing a shift of the magnetic hysteresis of one ferromagnetic layer as compared to the other. In the lower panel a map of the polarised neutron reflectivity is shown from an antiferromagnetic CoO-film in contact with a ferromagnetic Co-film at remanence. The specular scattering (diagonal stretching from the lower left to the upper right) and the off-specular diffuse scattering provide hints to the spin structure at the interface during reversal (courtesy of Radu, Voroblev, Major, Humblot, to be published and ILL report 2002).

These properties have their origin in the magnetic domain structure of ultra-small artificially shaped mesas. Advanced high intensity neutron scattering techniques are required to provide the magnetic information on a nanometer length scale [9]. Presently magnetisation profiles can be gained only from laterally extended films. The ESS will contribute to the understanding of spin structures and fluctuations in magnetic dot arrays embedded in heterostructures in a way that is not achievable with current neutron sources.

In contrast to other experimental techniques which provide high resolution images of the magnetic domain structure, neutron scattering will contribute to the understanding of spin structures at interfaces during real magnetisation reversal processes. This is particularly useful since the magnetic neutron scattering cross section is well known and neutrons are sensitive to ferromagnetic as well as antiferromagnetic spin structures. Flux limitations at present neutron sources do not allow studies of magnetic heterostructures with lateral sizes smaller than 5 x 5 mm^2. With the ESS, the lateral size of a magnetic array pattern could be reduced to the realistic dimensions of magnetic storage devices.

Process monitoring and optimisation

Imaging of embedded objects

Because of the non-destructive deep penetration of neutrons, they are particularly well suited for imaging embedded features inaccessible by any other means [10]. Combined with the inherent ability of time-of-flight methods to discriminate between different structural components, including magnetic phases, ESS will add a new dimension to real scale tomography and radiography. Relatively little neutron tomography has been undertaken to date because even radiographs take a significant time to acquire.

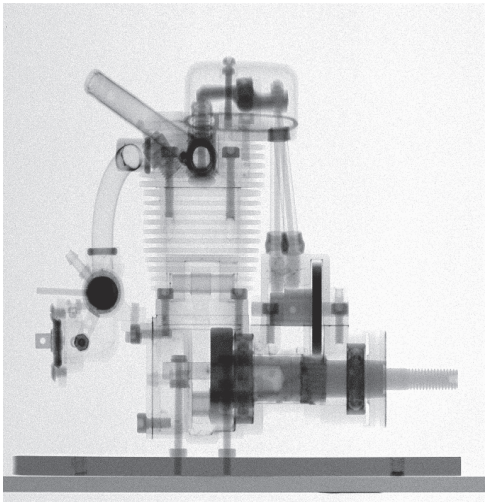


Figure 5: Neutron tomographic imaging of an engine part. Neutrons not only provide a high contrast from hidden parts inside of engineering components but also a chemical contrast due to crystalline phase sensitivity. At the ESS the data acquisition rate will be high enough to monitor a running engine.

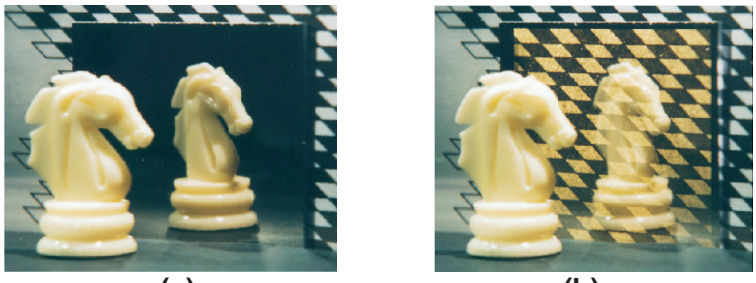
The new source will enable large field (200 x 200mm) sub-millimetre resolution images to be obtained in seconds and thus tomography experiments will become feasible for the first time. Furthermore the pulsed nature will allow the development of new areas of science, such as structural mapping neutron tomography. This technique uses the Bragg edge cut-off to discriminate between different structural phases and requires a pulsed white neutron beam.

Tomography and radiography of hidden objects become available in real time.

Monitoring protons

The presence of hydrogen in the atomic lattice can switch the optical and magnetic properties of materials. Neutrons can monitor where the hydrogen atoms go and how they change the structure. With the ESS not only the steady state structure can be determined, but it will also become possible to monitor hydrogen penetration and diffusion in real scale fuel cells at operating temperatures. Yttrium metal coatings have recently been discovered, which can be switched between reflecting and transparent by the charging and discharging of hydrogen [11]. Figure 6 displays the coating in the reflecting state and after hydrogen charging in the transparent state. Neutron scattering has played a crucial role in determining the location of the hydrogen atoms and in understanding the basic mechanism of the switching behaviour [12,13]. New compounds have recently been discovered which promise faster switching times, as well as easy and safe hydrogen handling through electrochemical cycling. In the future hydrogen will be crucial for energy storage materials and for energy conversion devices. Neutron scattering will continue to play a vital role for the understanding and optimisation of materials and of devices for future hydrogen based technologies.

Neutrons tell where the hydrogen atoms go and what they do inside of the host lattice.



(a) **(b)**

Figure 6: Two states of a Yttrium film are shown by their optical properties before and after hydrogen loading. The 500 nm thick yttrium film is covered with a 20 nm thick Pd protection layer and placed behind a white knight and before a chess-board. In the α phase the film shows metallic reflectivity (a), while in the trihydride γ phase the film becomes transparent for the visible light (b) (courtesy of Griessen [11]).

IV. Instrumentation

In order to capitalise on the opportunities the ESS offers for neutron research in the area of Materials Science and Engineering, the following instrument suite has been defined as necessary, in an approximate order of importance:

Instrument suite for materials science and engineering.

- Engineering diffractometer for stress-strain analysis, able to receive large samples and complex loading environments.
- High resolution and focussing small angle scattering (SANS) for in-situ kinetic measurements of microstructure and pore growth.
- Polarised neutron reflectometer with high Q-resolution and with high intensity for the analysis of magnetic heterostructures and nanopatterns.
- Tomography and radiography instrument for the absorption and time-of-flight imaging of large industrial components. The time-of-flight method will allow the use of Bragg-edge cut-off techniques to identify different components and phases within the engineering materials.
- High resolution powder diffractometer for the analysis of crystal structures and phases. High resolution backscattering spectrometer for the investigation of hydrogen motion and for the flow dynamics of lubricants.
- Variable resolution cold chopper spectrometer for the analysis of hydrogen diffusion, molecular rotation and confirmation, and for the internal dynamics of lubricants.
- Diffuse scattering diffractometer with full polarisation analysis for the investigation of disordered and hard magnetic materials.

V. Summary

Materials Science and Engineering encompasses many sub-disciplines, and also overlaps, in part, with solid state physics, with chemical structures and kinetics, with soft matter, and with liquids and glasses, described in other parts of this document. Unique to Materials Science and Engineering is, however, the bridging function from the fundamental understanding of materials properties to device applications and development of engineering components. Neutron scattering has played a recognised key role in this field during the past 50 years. With its increased flux and intrinsic time structure the ESS is expected to have a huge impact on materials science and engineering developments in the future because of:

- the higher flux that enables real time experiments, providing movies of structural and dynamical developments of materials;
- the higher resolution in respect to time, real space and reciprocal space that supplies a much improved data base for finite element models of engineering parts;
- the real operating conditions of pressure, temperature, and liquid or gas environments that materials can be tested in;
- the real scale components that are accessible to neutron beam studies.

Thus neutron scattering at the ESS in combination with computer simulations will have a very large impact on studies of materials and engineering components in real time, on real scale, and under real conditions.

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Achievements of neutrons in materials science and engineering

- Inelastic neutron scattering has provided the complete phonon dispersion curves for all classes of materials, including metals, alloys, semiconductors, insulators, compounds and crystals of noble gas atoms. Because of this huge data set, a detailed knowledge of inter-atomic potentials has been established and the effect of ionic, covalent, and metallic bonding on the elastic and thermal properties of materials is well understood.

- The characteristic shear and bending modes of soft layered materials could for the first time be determined with inelastic neutron scattering, such as for graphite and its intercalation compounds, for clay minerals, and for transition metal dichalcogenide materials. This class of layered materials plays an important role in very diverse fields of applications, ranging from lubricants in car engines to stoppage layers in environmental waste dumps. The transition metal dichalcogenides are also well known as low dimensional and anisotropic superconductors.

- Martensitic phase transitions transform a particular material from one crystal structure to another at a characteristic temperature. Often the memory of the former shape is preserved and can be recovered by back transformation. Alloys which exhibit memory effects are classified as shape memory alloys. Neutron scattering has provided fundamental insights into the mechanism of this type of diffusionless structural phase transition by studies of the structure and their phonon dispersion as they change from one structure to another. Often the softening of particular phonon modes could be made responsible for the lattice instability.

- Ferroelectric transitions form another subgroup of structural phase transitions, leading to a spontaneous electrical polarisation of the crystal. Certain ions are being displaced from high symmetry centro-symmetric positions in the high temperature paraelectric phase to positions of lower symmetry in the low temperature ferroelectric phase. The displacement changes the structure and induces a local electric dipole moment. Neutron scattering was essential for the understanding of this type of transition by mapping the temperature dependence of the lattice vibrations. Depending on the symmetry of the lattice vibration, ferro- and antiferroelectric transitions could be identified. Ferroelectric materials are used as ultrasonic transducers, in piezo – lighters, in high-precision positioning tools, and they are promising materials in non-volatile data storage devices.

- Superionic conductors are a class of solids which exhibit exceptionally high ionic conductivity. The conductivity reaches values which are more usually encountered in ionic liquids. These materials therefore have potentially important technological applications, for example in solid state batteries. Using neutron diffraction, the structure of superionics were first correctly described, including the random distribution of the cations in a rigid body centered host frame of anions. Furthermore, with neutron scattering experiments it was possible to determine the single ion mobility and to relate this to the ionic conductivity in an electric field. Thus, neutron diffraction and spectroscopy has played a crucial role for the present day understanding of this important class of materials and for their future development in batteries and fuel cell electrolytes.

- Hydrogen in metals has been of sustained interest to material scientists and physicists because of its intriguing structural, thermodynamic and electronic properties. Best known are, however, the damaging mechanical properties of hydrogen, causing embrittlement in structural materials. Neutron scattering was the prime tool for identifying the interstitial sites occupied by the hydrogen atoms in the metal host, their jump vectors, and their mobility. In addition, with neutron spectroscopy the local site symmetry can be determined as well as the potential parameters for hydrogen as a three-dimensional anisotropic oscillator. Compounds have been identified via neutron diffraction in which

the hydrogen atoms reside closer than in a H_2 bond and for which the volume density is higher than in liquid hydrogen.

- Neutron scattering has been essential in providing structural and dynamical information on fullerenes and how single buckyballs interact with their solvent.

- Neutron scattering has contributed to the understanding of coals from different geographic areas. In particular with small angle neutron scattering pore sizes, pore structure, and pore density could be determined. Neutrons are most suitable for these kind of measurements because of their highly penetrating power. Similar experiments have been carried out on sedimentary rocks, showing that the pore-rock interface has fractal properties over three orders of magnitude, the largest range over which fractal behaviour has ever been observed in a natural system. Other neutron studies have focused on pore characteristics of artificially sintered metals, alloys, and ceramics, and have thus contributed to a fundamental understanding of the relation between materials strength and microstructure.

- Neutron strain measurement on engineering materials has made an important contribution to our knowledge of residual stress. These stresses are essential to making reliable estimates of component life times. Important work has been carried out on welded structures, in particular the method is accelerating the introduction of new friction based welding techniques. Often post weld heat treatment is needed to reduce potentially life time threatening residual stresses; neutron diffraction has improved their definition. It has also played an important role in the optimisation of processes which introduce life elongating residual stresses such as peening and cold hole expansion. In particular it is helping to define peening conditions for the new laser shock peening process. Finally the method has contributed greatly to our knowledge about the micromechanics of composites by providing information about each of the individual phases leading to optimised composite properties. The high penetration power of neutrons has again been critical for assessing sub-surface strain gradients, chemical reactions or phase transformations in materials under industrially-relevant conditions, and for characterising bulk microstructures.

- In–situ neutron scattering has vitally contributed to our present day understanding of the hardening process of concrete. Again the high penetration power and the sensitivity to hydrogen make neutrons a unique tool for the investigation of one of the most important structural materials.

- High resolution neutron diffraction has recently been used for the first time to investigate in-situ the precipitation dynamics of oxygen in Czochralski-grown Si-crystals up to 1200°C. Internal gettering of oxygen is the basis for creating high quality denuded zones in Si-wafers, a prerequisite for the production of modern Very Large Scale Integrated (VLSI) devices.

4.3 Chemical Structure, Kinetics and Dynamics

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Abstract
Our informed understanding of the materials world around us is based upon a detailed knowledge of the structure and dynamics of materials on the atomic and molecular level. This contribution reports on molecular and supramolecular structures with sizes from tenths to hundreds of nanometers and dynamic processes studied by inelastic and quasi-elastic spectroscopy in the field of chemistry.

Neutron and X-ray scattering are complementary processes: X-rays scatter from the atomic electrons while the neutrons probe nuclei. This important difference means that neutrons have the ability to accurately locate light elements in the surrounding of heavy atoms. Since the chemistry of a mixed-metal oxide is determined principally by the location of the light oxygen atoms (key examples include high temperature superconductors and colossal magnetoresistance materials), neutron diffraction is the technique of choice for such measurements. Scattering from the nucleus in non-magnetic systems avoids any effect due to charge transfer and thus gives valuable information on the chemical bond. Isotopic effects and contrast between neighbouring elements are also an advantage as compared to X-rays. Neutrons scatter relatively weakly, but the cross-section is well understood and the high penetration depth of neutrons allows measurements under extreme conditions of pressure and temperature, or in-beam chemical reaction measurements on large components. The ESS will give substantial intensity gains over current sources, removing much of the flux-limited problems of current neutron instrumentation. Furthermore, the time-of-flight technique at the ESS, combined with short pulses and long paths, will allow high resolution and high flux to solve more difficult structural problems. The high flux and fixed scattering geometry inherent at ESS will facilitate a new generation of complex sample environments for in-situ experiments.

Chemists respond to the present demands for higher performance materials, cleaner environments and improved efficiency in use of chemicals in a wide variety of ways. These include the use of smart materials that respond to their environment, the use of thin films to build devices and the exploitation of pharmaceuticals and other agents such as catalysts that are active in much smaller quantities than previously used. These developments require the extension of analytical tools to study chemistry and chemicals in small quantities, in complex mixtures and under the conditions of imposed external environments of stress, temperature, chemical environment and other fields or constraints. One of these tools is vibrational spectroscopy, where neutrons have unique properties compared with other techniques. With the larger neutron flux available at the ESS, it will be possible to follow in-situ catalytic reactions. One will be able to record vibrational spectra above room temperature with a spectrometer covering a wide range of energy transfers, at low momentum transfers. The reaction pathways will be tracked down by observing the reaction intermediates. With such an instrument, it will be possible to measure spectra in aqueous solutions, which is the natural medium of biological molecules. Chemistry also involves local and diffusive transport processes which give rise to incoherent (single particle) and coherent (collective) quasielastic scattering. High-resolution neutron spectroscopy yields the microscopic information in space and time. The increased flux at ESS will extend such studies to lower concentrations, to systems with large inherent background, to more complex motions and parameter dependent studies.

I. Introduction

Neutrons probe nuclei and give a better contrast for light elements, isotopic substitutions or neighbour elements in the periodic table.

Neutron diffraction gives a unique structural fingerprint of the crystalline state. Light atoms are detected with high precision even in the presence of heavy atoms such as transition metals and actinides. In materials science, archetypal examples include hydrogen storage materials such as metal hydrides [1], and mixed metals oxides such as high temperature superconductors [2] and battery materials [3]. In organic chemistry, the precise location of hydrogen atoms free from charge transfer effects contributes to our detailed understanding of hydrogen bonding from simple model peptide systems to supramolecular chemistry. The possibility of isotopic replacement, and in particular H/D substitution, can be used to great advantage in elucidating specific structural details (Figure 1).

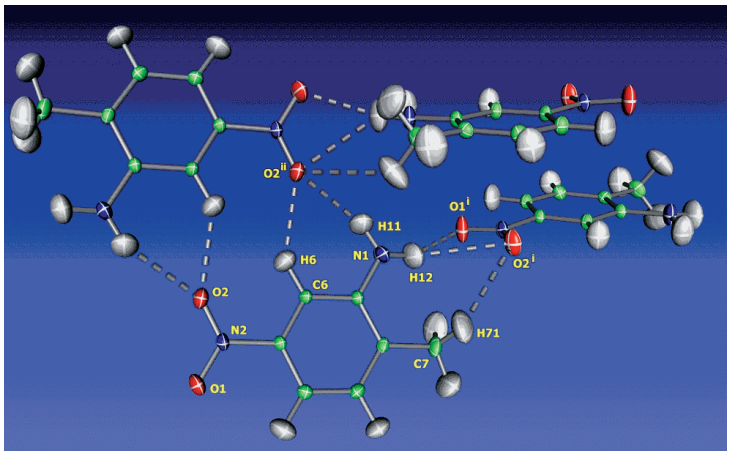

Figure 1: Hydrogen bonding schemes in nitroanilines with non-linear optical activity.

II. The impact of high flux sources

Neutron penetration depth allows in-situ experiments to follow bulk reactions in complex environment.

With increasing neutron fluxes, and particularly with the advent of the ESS, neutron diffraction experiments typically involve the collection of a large series of diffraction profiles. These time-resolved, parametric experiments enable structural trends to be analysed as a function of physical parameters such as temperature [4], pressure [5] and magnetic field. In turn, this leads to a fuller understanding of phase diagrams and structural transitions, and to deeper insights into structure-property relationships. Time-resolved neutron diffraction studies are also a very powerful means of following chemical reactions. Neutron powder diffraction allows bulk analysis of materials in “real-life” industrial configurations yielding important crystallographic, thermodynamic and kinetic information about reaction behaviour. Recent studies include investigations into concrete ageing, silicate compounds, hydrothermal syntheses, self-propagating chemical reactions, amorphisation, hydride formation and decomposition and the charge/discharge behaviour of batteries (Figure 2).

III. Future opportunities and flagship areas

Energy storage and conversion

Environmental problems, such as the green house effect, lead to research on new solutions for energy management. Fuel cells will probably be the cleanest and most versatile power source of this century. However many scientific problems remain to be solved: efficient catalytic processes at electrode surfaces, ionic diffusion in solid state electrolytes, chemical reaction kinetic optimisation. In all these cases, neutrons will be useful for probing both structural aspects and chemical mechanisms.

Clean and efficient energy storage and conversion

Materials for cryo-coolers used in space, tritium storage units, fuel cells and alkaline batteries are now metallic hydrides. From a fundamental point of view, competition between magnetic and hydrogen ordering in rare earth-transition metal

hydrides has been the subject of much recent research. The recent discovery of switchable mirrors also gives an insight into the physics than can be studied through these compounds. In the surrounding of heavy metals, neutron diffraction is a unique tool for locating the proton in metallic hydrides: during the absorption process, the electron is transferred from the proton to the conduction band and only neutrons, probing the nucleus, can give accurate structural data. Moreover, for forthcoming applications, most of them related to clean energy storage and energy conversion, time resolved experiments would give valuable information on the chemical and kinetic processes involved during charge/discharge cycles, allowing optimisation of these materials.

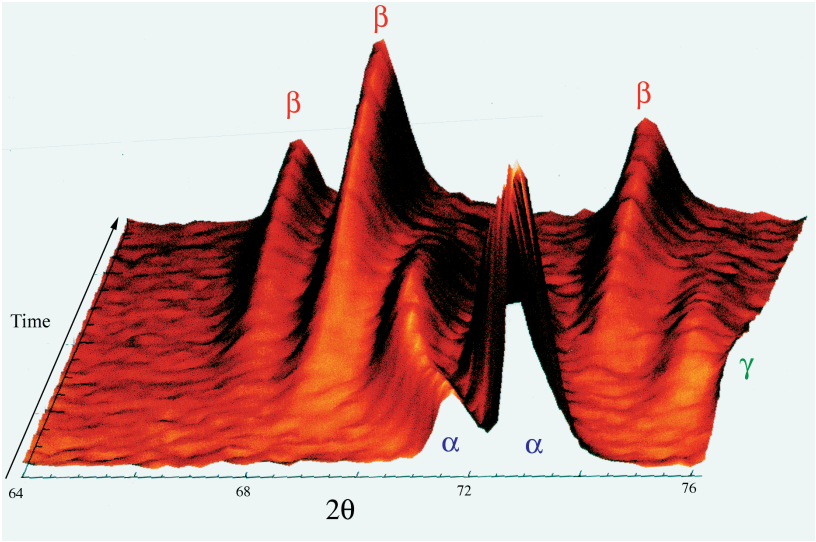

Figure 2: Three dimensional view of the neutron diffraction patterns of a metal hydride electrode during an electrochemical charge in 10 hours. The different phases involved in the reaction are given on the plot [3].

An increase of flux is essential for studying faster processes such as very fast discharge phenomena or short circuit batteries. Materials used in modern batteries often include H and Li as charge carriers. The cations are distributed over various sites in the crystal structure and show high mobility. Their detailed analysis is most important in order to understand the conduction pathway and requires neutron diffraction experiments. In addition, the dynamics can be studied with quasi-elastic neutron scattering. However, the accuracy of crystallographic data is often affected by the complexity of the experimental environments. A better resolution, associated with a larger Q range, will be necessary for solving new problems.

In-situ studies of catalysts

A catalytic cycle involves adsorption, diffusion, and reaction steps. Neutron

In catalysis, one can study the surface species that result from molecular adsorption, dissociation, or chemical reaction with neutron spectroscopy, even for some systems that

cannot be studied by diffraction. The technique is well suited for determining the different adsorption sites for atomic hydrogen on metal or sulfide catalysts [6], for identifying the active species in catalytic reactions, and for understanding deactivation (Figure 3). One limitation is the quantity of catalyst that nowadays has to be prepared (tens of grams). A neutron flux one or two orders of magnitude larger would also allow us to study the adsorption of non-hydrogenous compounds, such as CO, SO_2 and NO_x. Kinetic studies would then also be possible.

scattering techniques can play a major role in all these aspects of catalysis.

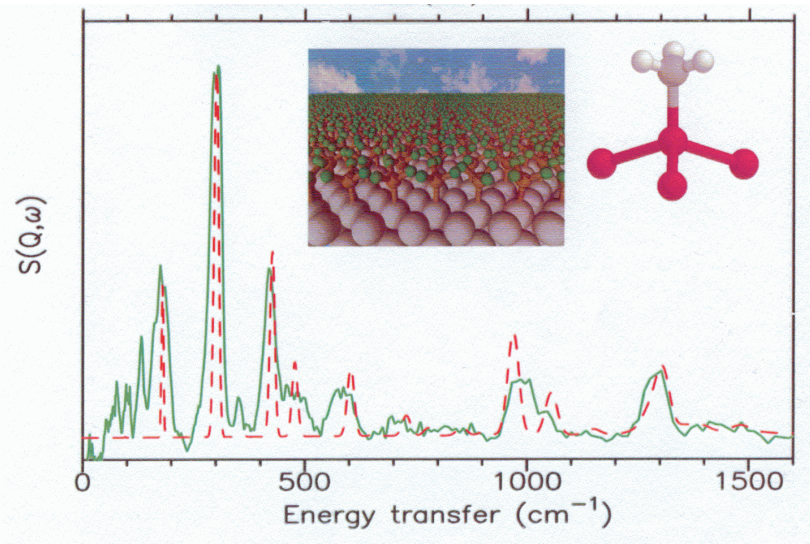

Figure 3: Observed (solid line, recorded at 20 K) and calculated (dashed line) spectra of an industrial palladium catalyst after reaction. The surface is covered with methyl groups, which explains the deactivation [7].

In order to make catalytic processes cleaner and more efficient, one must identify the active species and the reaction intermediates. To follow in-situ catalytic reactions, one needs a spectrometer that can measure the whole vibrational spectrum at small momentum transfer. There are numerous catalytic reactions that would benefit from such an instrument in hydrogenation, oxidation or desulfurisation. For example in the conversion of n-butane to maleic anhydride one could find out if the intermediate is an olefin or an alkoxide. This would permit the building of a kinetic model for the reaction.

Improving catalysts by spectroscopic studies of reagents and intermediates.

Hydrogen bonding and proton dynamics in advanced materials

Analysis of the structure of molecules of biological relevance is important for understanding their different functions. To study the conformational flexibility of these molecules, knowledge of the intra- and inter-molecular forces is required. To achieve this task, vibrational data can be used in combination with quantum mechanical calculations. Hydrogen vibrational dynamics can only be accessed by neutron scattering [8]. A better energy resolution is needed to separate the numerous modes of such complex molecules. So far, measurements have been performed in the solid

Neutron scattering is a powerful means of analysing the proton dynamics of molecules of biological and pharmaceutical interest.

phase; aqueous solutions would be a more natural medium. The analysis of organic molecules interacting with the surface of a substrate, as in biominerals and drug supports, offers new opportunities. The details of the bonding interaction would lead to a basic understanding of the biomimetic processes of formation in biomineralisation.

Advanced molecular materials

There has been a recent increase in exciting work in the development of molecular materials with useful and tuneable physical properties such as magnetism, superconductivity, non-linear optical activity, polymorphism, etc. This area is likely to expand dramatically in the next decades. Much of this work is focused on understanding the intermolecular interactions holding 3-D arrays of molecules together, which are often weak hydrogen bonding interactions. The directionality of these interactions is crucial, and the accurate definition of hydrogen atom positions by single crystal neutron diffraction is vital. For example, in pharmaceutical materials the understanding of polymorphism can often rely on small energy differences between molecular configurations, while in supramolecular chemistry accurate quantification of weakly bonded motifs will allow for more rational crystal engineering allowing chemists to tailor properties by designing structures. ESS will advance this expanding area of molecular science by allowing routine characterisation of all atoms in such structures. Specifically, ESS will allow such studies to be carried out on smaller crystals, will offer faster structural characterisation and allow more systematic examinations of the phase space of candidate molecular materials. This maps most appropriately onto the needs of the science.

Hydrogen bonds play a fundamental role in the structure and reactivity of chemical and biological systems.

Proton transfer along a hydrogen bond is the simplest example of a chemical reaction, a covalent bond is broken and the hybridisation of the acceptor and donor atoms is exchanged. The potential energy barrier to proton transfer, separating the two wells which correspond to the stable positions of the hydrogen atom, is therefore high and the mechanism for transfer entails tunnelling through the barrier. However, because of the modulation of the electronic state of the molecular skeleton during proton transfer, molecular vibrations also participate, thus promoting or hindering proton transfer. New theoretical methods are being developed to handle coupled tunnelling and vibrational dynamics. Neutron scattering is a uniquely powerful tool for precisely locating hydrogen atoms in these systems and then measuring their dynamics. The tunnelling dynamics in hydrogen bonds can be probed directly by quasi-elastic scattering and molecular vibrations are measured by inelastic scattering.

Diffusion in porous materials

Molecular diffusion can be followed, on a microscopic scale, with neutrons.

Molecular diffusion in porous materials, such as zeolites, is important in catalysis or separation processes. In addition to their fundamental character in elucidating confinement effects, the aim of these studies is to create new diffusion models valid for complex systems. When the size of the molecule is comparable to the pore size this leads to diffusion

limitations, and diffusion coefficients are 3 to 12 orders of magnitude lower than in the gas phase. Various experimental and theoretical techniques (Figure 4) are used to determine diffusion coefficients. In several cases, it has been found that quasi-elastic neutron scattering is the only technique which is able to derive reliable intracrystalline diffusivities [10]. The neutron spin echo technique allows us to probe much longer time scales, this has been demonstrated for the diffusion of deuterated molecules in zeolites.

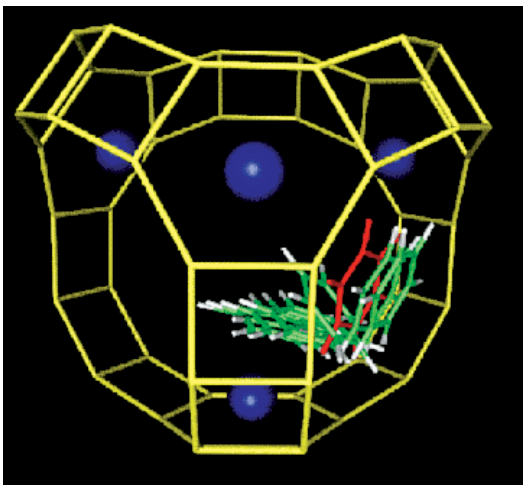

Figure 4: Minimum energy path for benzene in NaY zeolite, between a cation site (Na in blue) and a window site (the molecule at the transition state is in red) [9].

Rotational tunnelling

Rotational tunnelling is a uniquely sensitive probe of fundamental and system properties. Neutrons yield complete microscopic information.

Rotational tunnelling is one of the simplest quantum dynamical processes. High resolution neutron spectroscopy has significantly promoted our understanding of the properties and importance of quantum motion in solids. This is largely due to the extreme (exponential) sensitivity of tunnelling to the intermolecular potentials in combination with the fact that it can be unambiguously identified. Rotational tunnelling, combined with theoretical chemistry programs, is especially suited to obtaining precise rotational potentials. They allow the determination of the intermolecular interaction potentials [11] via the use of pressure, of disorder in molecular alloys and glassy systems, of the influence of time dependent perturbation of the environment (coupling with phonons) and of deviations from single particle dynamics by coupling to other degrees of freedom. This last field, multidimensional tunnelling processes, will be one of the most exciting in the future. Rotation-translation coupling is already established [12]. The coherent counterclockwise rotation of a methyl group and its centre of mass in a four-fold environment imposes a surprising fourfold proton density distribution of a three-fold rotor (Figure 5) confirmed by neutron diffraction.

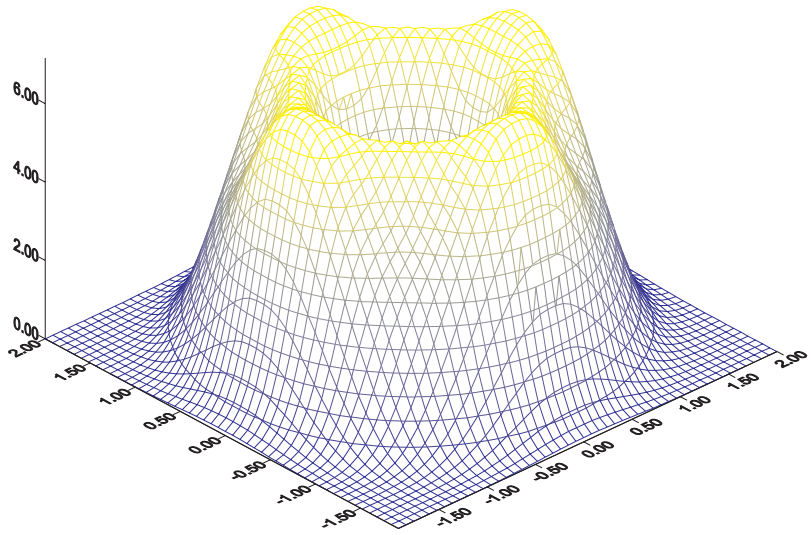

Figure 5: Four-fold proton density distribution of a three-fold rotor due to rotation-translation coupling in a four-fold environment [12].

Tunnel splittings of excited rotational states, well dispersed molecules isolated in a matrix, extended coherent surfaces, single crystals, disordered materials, time-dependent effects such as spin conversion, new forms of multidimensional tunnelling and new non-hydrogenous rotors will become routinely accessible to rotational tunnelling spectroscopy with ESS.

Electrochemistry at surfaces

Neutron reflection experiments are relatively new, but are now being applied to a wide range of chemical studies. A good example is the application of this technique to electrochemistry. The interesting chemistry happens at interfaces (electrodes) and a wide-range of different chemical species are present. Neutron reflection is an excellent tool for the determination of the distribution of various ions and molecules near an interface and for the determination of the composition and structure of deposited layers. The underlying scientific problems arise from important technologies such as those of energy storage, analytical and microanalytical devices and biological sensors. At present relatively few experiments have been conducted [13,14] but considerable progress can be foreseen. The uniform surface areas of samples that are available for study is often very limited and a few mm^2 is more common than the few cm^2 usual for reflection experiments with current instrumentation and sources. Higher flux instrumentation will allow experiments on such, more realistic, samples. The changes that occur in electrochemical devices will also be followed in real time, or by application of cyclic data acquisition phased with external potentials or currents.

Neutron reflection is a powerful tool for the study of interfaces, such as those in electrochemical systems. The high flux at ESS will reduce the required sample area and make it possible to study many new systems.

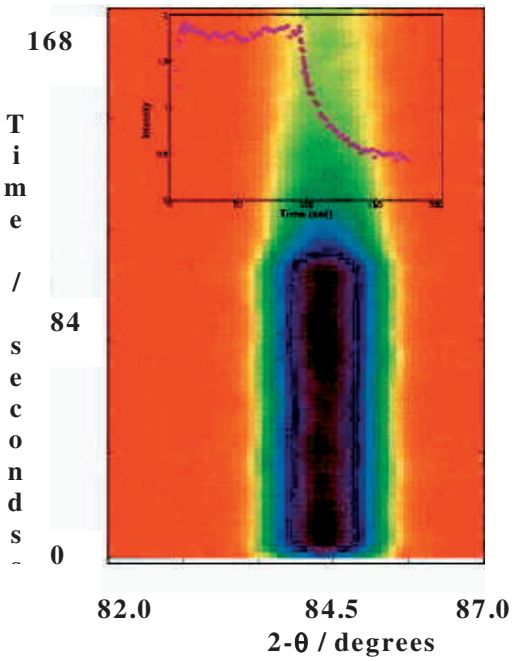

Figure 6: Diffraction from a dispersion of $Ni(OH)_2$ particles in D_2O showing the change in intensity of the 004 peak in one direction when flow starts. Each time slice is two seconds [16].

Stroboscopic data acquisition offers many possibilities for the study of dynamic processes under the influences of external fields such as flow, stress, electrical and magnetic forces.

An example of an experiment that is of interest to the chemical industry is one in which the alignment of plate-like particles under flow is studied [15]. This is shown in the Figure 6. Particles with ~ 90 nm diameter and approximate aspect ratio of 5:1 were prepared as model experimental systems. Each particle is a single crystal. The alignment can be followed by diffraction of neutrons from the particles in the dispersion. This can be followed dynamically by use of a cyclic data acquisition procedure. Experiments are limited by the small signal in relation to the background. At present only relatively large volumes can be studied. This type of data, in conjunction with small-angle neutron scattering, has already been used to identify phase changes under shear. This type of experiment, including magnetic and electric orientation, could become common if higher flux were available to study small samples that could be subject to more uniform fields.

Polymer synthesis

SANS can be used to follow polymerisation reactions in real time; to determine reaction mechanisms and the influences of synthesis conditions on the structure of materials.

Small-angle neutron scattering has become a pre-eminent tool for the characterisation of polymers, colloidal particles and a variety of mesophases. Work to follow the synthesis of these materials in-situ will become more common [17]. This has been difficult up to now because of the poor time resolution available and the need to study samples which are dilute in the component of interest, and which contain many other different molecules. It is to be expected that investigations aimed at deepening the understanding of polymerisation mechanisms will develop. For example, these could study the location of initiator in emulsion polymerisation reactions or the conformation of the polymer molecules as they form. SANS has been widely used to look at the

morphology of the resulting latex particles [18] but only a little work has been viable with present flux [19].

Chemical kinetics

Time resolved studies of chemical kinetics with stroboscopic data acquisition.

ESS, with a high flux and instruments optimised for resolution for particular experiments, will allow a much wider range of kinetic experiments. Cyclic data acquisition permits a time resolution of 1 ms. At present, experiments have been performed on pulsed flow and ultrasonic excitation of a crystal. For example, studies of reorientation dynamics in concentrated colloids have already been made. Future applications would include cyclic electrochemical processes. This would give information about the distribution of ions in both solid and liquid phases: this would be studied by neutron reflection, SANS and neutron diffraction.

IV. Instrumentation requirements

Relation to ESS-instrumentation

Chemistry has wide-ranging requirements for neutron instrumentation. These include elastic scattering, quasi-elastic scattering and inelastic scattering.

Powder and single crystal diffractions, small angle scattering and reflectometry.

In determining more complex structures, or looking at more subtle microstructure, high resolution powder diffraction is crucial to provide a sufficiently large number of well-defined Bragg peaks. With ESS fluxes, a high-resolution machine will be able to study small samples, such as isotopically enriched materials or compounds synthesised in small quantities by novel synthetic routes.

Parametric studies provide a wealth of information on structure-property relationships. A powder diffractometer for rapid time-resolved experiments should have good detector stability, short acquisition time, large Q range, best achievable resolution and a versatile sample environment. Data accumulation times should be of the order of 10 ms.

To increase pressure beyond the present limit (about 50 GPa) it is necessary to reduce sample sizes to about 0.01 mm^3. This type of study obviously needs a powder diffractometer optimised for extreme environments with high flux and large beam time to get valuable results. The possibility of combining HP (> 50 GPa) and HT (> 2000 K) is a challenging goal.

A high throughput small-molecule single crystal diffractometer will offer high quality data from sub-mm^3 samples of both organic and inorganic materials in around 1 hour. With sufficiently large crystals, parametric studies as a function of temperature and/or pressure should be routine. The capability for single crystal experiments on larger molecules will also provide detailed insights into the behaviour of complex supramolecular assemblies and large inorganic systems.

Small-angle scattering will be important, with a wide range of momentum transfer in a single configuration to follow dynamic

processes. An ability to choose the resolution appropriate to the problem is desirable. A Q range of at least 0.001 to 0.5 A^{-1} should be accessible.

Inelastic and quasi-elastic scattering.

Neutron reflection, for studying interfaces, will need a high flux and a wide dynamic range to cover reflectivities as small as 10^{-8}. A range of Q_{max}/Q_{min} of 30 in a single configuration is required.

Vibrational spectroscopy for in-situ catalysis or biological molecules in aqueous solutions requires measurements of spectra at low momentum transfer values, $Q < 2Å^{-1}$, up to 400 meV, at intermediate resolution. A high resolution instrument, $\Delta E/E \approx 1\%$, is also required.

Transport processes, intramolecular vibrations, hydrogen bond dynamics and rotational tunnelling are all influenced or perturbed or coupled to lattice vibrations. A direct geometry high performance time-of-flight instrument, for the energy regime up to 100 meV, is necessary for controlling and exploiting the phonon density of states.

In the quasi-elastic domain, spectrometers with well-defined line shapes are needed. Only an instrument able to access large Q values at intermediate resolution will allow us to differentiate between different models of motion. Polarisation analysis would be useful to separate coherent and incoherent scattering, e.g. to eliminate Bragg peaks or to study collective phenomena.

Backscattering spectrometers should be available with the best possible energy resolution for very slow motions or strong potentials, and with large momentum transfers.

A neutron spin echo spectrometer with a Fourier time limit of 1 μs is required for following slow diffusion.

Apart from the general requirements for improvement in flux, and in resolution in energy and momentum transfer, that will benefit most experiments, there are some interesting possibilities for a new, pulsed neutron source. Instruments could be built that have some of the available configurations optimised for cyclic data acquisition with appropriate short sample to detector distances and high spatial resolution on the detector.

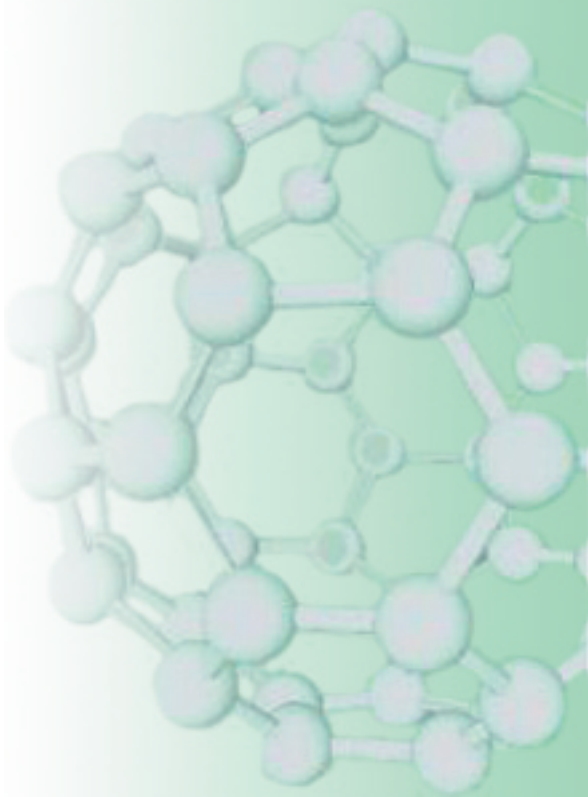

Table with instrument priorities

50 Hz short pulse target station	$16^2/_3$ Hz long pulse target station
High resolution powder	High intensity SANS
Thermal chopper	Variable cold chopper
Cold chopper	High intensity reflectometer
High energy chopper	High resolution neutron spin echo
Chemical single crystal	Focusing low Q SANS
High resolution reflectometer	
Magnetic powder	
High resolution backscattering	
Single pulse diffractometer	

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Achievements of neutrons in chemistry

- In molecular compounds, neutron diffraction has provided the most definitive answers to structural problems, especially (i) accurate positional parameters of all atoms; (ii) accurate determination of thermal motion; (iii) structures in which hydrogen bonding plays a major role (e.g. ferroelectrics, hydrides, and hydrates, including the role of water in biomolecular systems).

- Neutron powder diffraction, through the Rietveld technique, has produced the most accurate, reliable, and complete refined structures from powder data. In combination with X-ray powder diffraction, complex superconductors and host-guest interactions in zeolites have been studied.

- The simultaneous determination of the structure and dynamics of a chemical species under synthesis has enabled unstable reaction trajectories to be characterised in great detail.

- Neutron spectroscopy has assisted crystallography in locating hydrogens away from the points of high symmetry where they had been previously determined by X-ray crystallography.

- Neutron spectroscopy has been key to an understanding of the nature of hydrogen in materials. For example, the state of the majority of adsorbed hydrogen on the hydrodesulphurisation catalyst MoS_2 was determined to be molecular. On the other hand, atomic hydrogen was observed by neutron spectroscopy on RuS_2. This different behaviour towards a hydrogen atmosphere revealed that RuS_2 has a pseudometallic comportment whereas for MoS_2 redox or acid base properties are involved.

- Neutron spectroscopy has contributed to our understanding of industrial processes. Deactivation of an industrial Pd catalyst has been explained by the presence of a layer of methyl groups on the surface, which prevents the interaction of larger organic molecules with the metal.

- The local diffusion of molecules through porous solids, such as zeolites, can be followed by neutrons where other methods either fail or are inappropriate. Only with quasi-elastic neutron scattering has it been possible to measure simultaneously self and transport diffusivities.

- High intensity neutron powder diffraction has enabled in-situ diffraction measurements to be performed on real systems. Nickel-Metal hydrides batteries have been studied and the results have helped to overcome cycle life problem by showing how the appearance of an intermediate phase reduces the constraints during the cycling process. The rate-limiting factor for high charge-discharge rates of secondary batteries was shown to be the kinetics of the metal to hydride phase transition rather than the diffusion of hydrogen.

- In combination with X-rays, neutron diffraction has probed charge distributions that are essential for understanding some of the most profound details of a crystal structure, for example the effects of charge transfer, magnetostriction, and the onset of superconductivity.

- Neutron diffraction has been used to study not just crystal structure but also disorder and phase transition behaviour in molecular systems and, in particular, in the fullerenes and their derivatives.

4.4 Soft Condensed Matter

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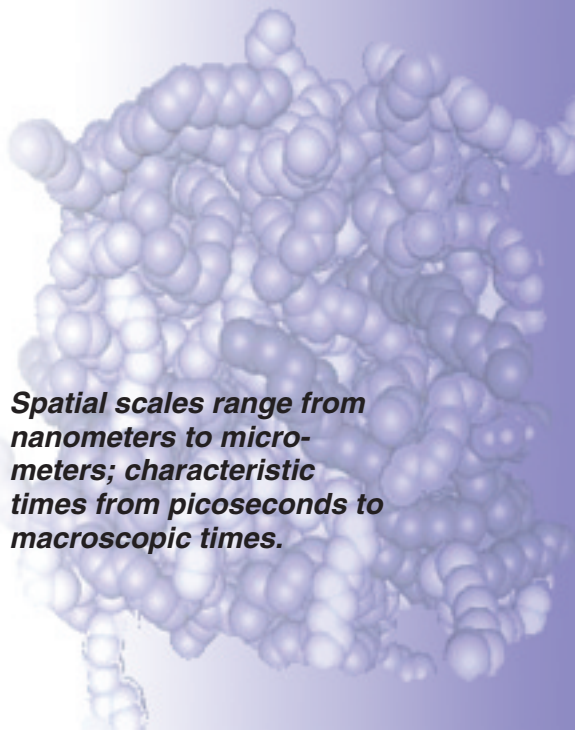
Abstract
Neutron scattering techniques play a unique role in the study of both the structural and dynamical properties of the wide range of substances categorised as "soft matter". Among the advantages presented by these techniques, two are of crucial relevance in the soft matter field: the suitability of the length and time scales accessed by neutrons, and the capability to manipulate the contrast by specific deuteration of any constituent of the system. *Neutron scattering is the only tool* for unravelling the molecular morphology and motions in soft matter systems at the different relevant length scales. On the other hand, the understanding of structural properties and dynamics at a molecular level is the key for advancing in this field: the envisaged trends move towards the study of increasingly complex, often multi-component materials tailor made for industrial applications. The combination of neutron scattering techniques, advanced chemistry and molecular modelling will be essential. Experiments on very dilute components, or on very small amount of matter (e.g. particular topological points, at the interfaces ...) are demanded. Moreover, in-situ studies will investigate time dependent and transient phenomena, non-equilibrium situations and so on. Such experiments will become possible by the orders of magnitude increase in sensitivity offered by the next generation neutron sources. In particular, the availability of a $16^2/_3$ Hz long pulse target station would allow optimisation of most of the instruments devoted to soft-matter studies, such as small angle neutron scattering instruments, reflectometers and high resolution neutron spin echo spectrometers.

I. Introduction

Soft matter includes a large variety of materials in daily use, and with a wide range of properties based on common physico-chemical origins.

The concept of "soft matter" subsumes a large class of molecular materials, including e.g. polymers, thermotropic liquid crystals, micellar solutions, microemulsions and colloidal suspensions, and also includes biological materials, e.g. membranes and vesicles. These substances have a wide range of applications such as structural and packaging materials, foams and adhesives, detergents and cosmetics, paints, food additives, lubricants and fuel additives, rubber in tyres etc., and our daily life would be unimaginable without them. In spite of the various forms of these materials, many of their very different properties have common physicochemical origins such as a large number of internal degrees of freedom, weak interactions between the structural elements and a delicate balance between entropic and enthalpic contributions to the free energy. These properties lead to large thermal fluctuations, a wide variety of forms, sensitivity of the equilibrium structures to external boundary conditions, macroscopic softness and various metastable states.



Spatial scales range from nanometers to micro-meters; characteristic times from picoseconds to macroscopic times.

The structural units of soft matter systems are large molecules or aggregates of molecules showing different structural and dynamical properties depending on the length scale of observation. This implies a need to cover large ranges in the experimental space/time windows for a complete

understanding of their characteristic features. Furthermore, aggregation in these systems may lead to a large internal interfacial region, which can then make a dominant contribution to the overall properties.

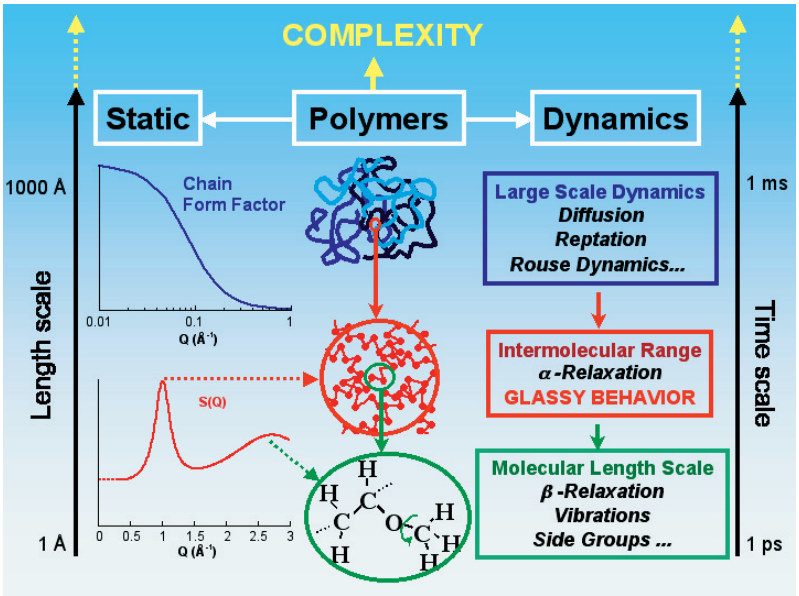


Figure 1: The different static and dynamic scales relevant for soft matter systems are schematically shown in the simplest case of a polymer.

II. The role of neutrons

Among the experimental techniques used for the investigation of the structure and dynamics of soft matter, neutron scattering (NS) plays a unique role for several reasons:

Space-time resolution at proper scales, variation of contrast and high penetrability, make neutrons a unique tool for studying the structural and dynamic properties of soft matter at a molecular level.

i) The suitability of the length and time scales accessed, especially by Small Angle Neutron Scattering (SANS) and Neutron Spin Echo (NSE), allows the exploration of large scale properties (for instance, the conformation of a large macromolecule, its diffusion in the embedding medium and its entropy driven dynamics) as well as features characteristic of the local scales (e.g. the inter- and intra-chain correlations in a glass forming polymer and their time evolution, the rotational motions of methyl groups, vibrations ...).

ii) By variation of the contrast between the structural units or molecular groups, complex systems may be studied selectively. In particular, the large contrast achieved by isotopic substitution of Hydrogen (one of the main components of soft matter) by Deuterium constitutes the most powerful tool for deciphering complex structures and dynamic processes in these materials.

iii) Neutron reflectometry constitutes a unique technique for the investigation of surfaces and interfaces in soft matter.

iv) The high penetration of neutrons in matter allows the study of the influence of external fields or parameters, e.g., the evolution of the system under processing conditions.

v) The space-time resolution of these techniques reveals the molecular motions leading to the viscoelastic and mechanical properties of soft matter systems. This knowledge is valuable for the design of tailor made materials.

Seminal experiments unravelled chain structure and dynamics in polymer melts.

The unique power of neutron scattering for revealing essential features in the field of soft matter can be exemplified by two pioneering experiments that can already be considered as "classic". The first one is the experimental proof of the random coil shape of polymer chains in the melt or in the glassy state as proposed in the 50's by Flory. This confirmation was only possible in the 70's (R.G. Kirste, W.A. Kruse and J. Schelten, Makromol. Chem. **162**, 299 (1973)) with the development of SANS. Since in the bulk a given macromolecule is surrounded by similar units, only by using contrast variation and deuterating single molecules could Flory's proposition be demonstrated. This measurement of the single chain form factor by SANS was one of the first applications of NS to polymer science. The dynamic counterpart of this experiment could only be solved 25 years later. Neutron spin echo (NSE) investigations on the long time chain dynamics recently allowed the confirmation of de Gennes predictions on the mechanism of reptation in polymers (P. Schleger, B. Farago, C. Lartigue, A. Kollmar, and D. Richter, Phys. Rev. Lett. **81**, 124 (1998)).

III. Future opportunities

General future trend: tailor made multi-component materials for industrial applications.
This implies increasing complexity!

Soft condensed matter systems in the future will increase in complexity both in structure as well as in the number and specific roles of their components, e.g. multi-component soft and soft / hard materials tailor made for industrial applications. Such complexity will cover a wide range of length and time scales, posing challenging problems to basic science. Desirable systems show complex interaction potentials with several minima, generating different structures according to the mechanical and thermal history. The understanding of the interplay of geometry and topology, and the characterisation of interfacial features, are of utmost importance for the future developments and design of novel materials. Finally, the structural changes induced by external fields such as shear play a crucial role in the outcome of industrial processing. These issues have to be dealt with as a necessary precondition for achieving controlled improvement in the fabrication of future materials.

The interplay between neutron scattering, advanced chemistry and computer simulations will be of great importance.

Neutron scattering in combination with advanced chemistry is the necessary tool for facing the new challenges in the field of soft matter. Here the focus is on linking chemical architecture to microscopic and macroscopic properties. The interplay between computer simulation and neutron scattering also promises to become particularly effective because of the common ability of neutron scattering and computer simulation to home in on a key structural unit.

High intensity neutron

Future trends will require a wide variety of experiments,

including investigations on dilute components, or on very small amounts of matter such as particular topological points or at interfaces. Sometimes these experiments involve polarisation analysis, short time measurements or in-situ studies. In all these cases, very high intensities of the neutron beam are required.

beams are essential for future developments on soft matter.

Assuming the availability of a high flux neutron source with the characteristics of ESS some flagship areas in this field can already be envisaged and are listed below.

Molecular rheology

The application and industrial processing of many soft condensed matter systems strongly depends on their rheological properties, which are determined by the interactions and motions of the constituent structural units such as chain molecules, aggregates, colloidal particles, surfactants, etc. Their understanding is one of the great challenges of basic soft condensed matter and would certainly facilitate the molecular design of new materials. As an example we consider the rheology of polymer melts which is currently described in terms of the reptation model. A very long polymer chain in a melt suffers local restriction of its motion by topological entanglements with other chains along its length. The polymer chain can be envisaged being confined in a "tube" formed by the neighbouring chains. The snakelike motion along the tube (reptation) is the main mechanism controlling the dynamics of a highly entangled linear chain.

A rheology based on molecular understanding would allow the design of tailor made materials.

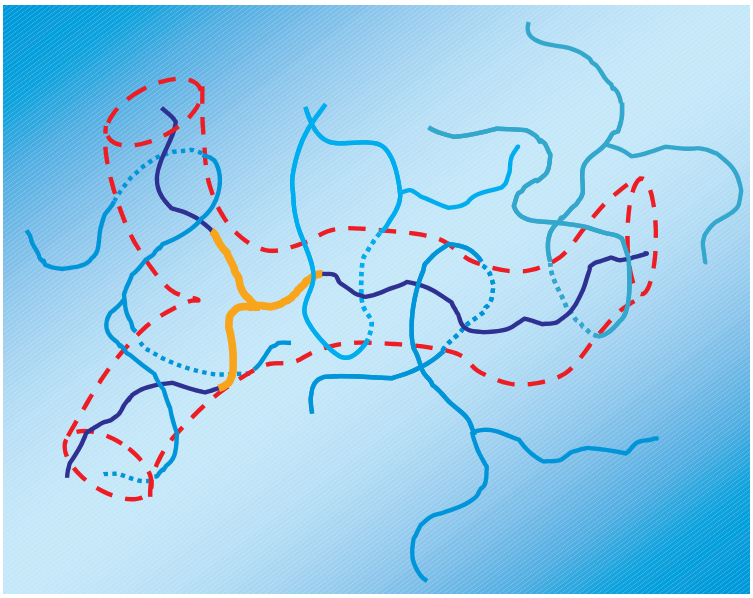


Figure 2: Schematic representation of the reptation mechanism of a branched polymer. The topological constraints imposed by the surrounding chains are modelled by the imaginary forked red tube. The relaxation of the internal modes and the diffusion of the polymer have to take place inside and along the tube prior to free diffusion. The yellow portion in the selected macromolecule represents the labelled branching point which dynamics would reveal the detailed molecular mechanism for reptation in this system.

The time is now ripe for the investigation of more complex topologies such as branched polymers (stars, H-polymers, combs, trees). Only a small number of branches in a polymer molecule may substantially alter the industrial processing although they have little effect on the solid-state properties of the final material. Neutrons are uniquely suited to achieve this goal. Hydrogen atoms would be replaced with deuterium in specific topological points of the molecule and the dynamics of these points followed by NSE techniques. The high dilution of the labelled topological points makes such experiments impossible with currently available neutron fluxes.

Buried interfaces

A large reduction in illuminated area gives access to new classes of buried interface.

Neutron reflectivity gives unique structural and compositional information about buried interfaces. At present, only a restricted range of materials are sufficiently transparent to permit the experiment. By allowing a reduction of illuminated area of about two orders of magnitude, next generation neutron sources will relax the conditions for transparency to an extent that will give neutron reflection access to almost any interface. Two areas of particular importance are the liquid / liquid interface, where adsorbed polymers or amphiphiles play a crucial role in determining the stability of emulsions, and biolubrication, where the delicate control of environmental factors (pH, ion concentration, etc.) is used to manipulate the conformation of polyelectrolytes at the lubricated interface.

A key experiment here is the response of the layer structure to compression and / or shear under different solution conditions, an experiment that would become feasible at significantly higher neutron fluxes.

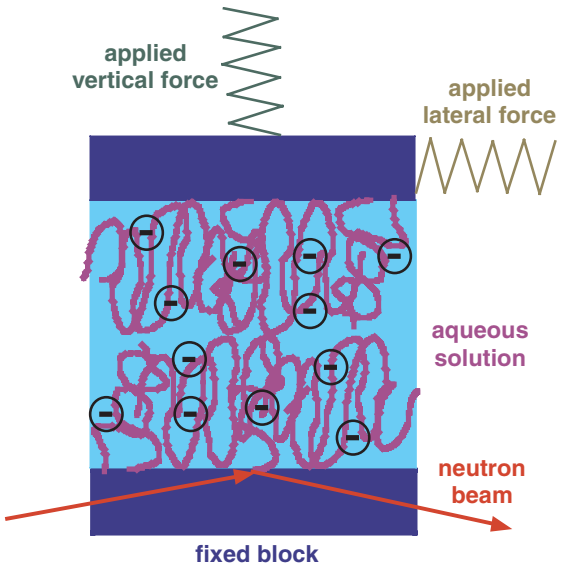


Figure 3: The study of biolubrication by neutron reflection. The high intensity neutron beam will allow the illuminated area of interface to be reduced to a size (less than 10 mm^2) that should be manageable in conjunction with a force balance. This will allow the direct study of the conformation of adsorbed polyelectrolyte by reflection while various forces are applied to the system.

Self-assembly and structure formation

Combination of nanometer scale processing and molecular self-assembly is a scientific and technological challenge.

The control of molecular self-organisation by means of chemical and physical stimuli is the key to successful creation and control of structures on the nano- or micro-meter scale. The use of self-assembly ranges from the development of new formulations for pharmaceuticals and pigments to the morphological control of adhesion of bio-materials and to the development of molecular electronics. Combination of nano-meter scale processing and molecular self-assembly is a scientific and technological challenge where neutron scattering will make key contributions. Specific labelling and the large penetration depth of neutrons are both essential in revealing the detailed structure and mutual interactions. This knowledge is necessary in order to understand and to control the intricate interplay between the components in colloidal formulations - in basic science as well as in e.g. pharmaceutical applications. In molecular electronics the precise control of surface structure is of crucial importance for the performance. High intensity neutron sources can investigate the molecular organisation on a surface-size relevant to electronic components.

With the next generation neutron sources, it will be possible to follow the formation of self-organised systems with the necessary time resolution. The process of nucleation and growth in particle formation and crystallisation can be followed in-situ, thus allowing us to find effective ways of controlling this process by suitable polymers. The formation and change of structures that are induced by shear or temperature quenches can be studied in-situ. These are both of fundamental interest and highly relevant to the processing and application of polymer components. Another example concerns materials whose structures spontaneously respond to external changes, such as temperature or pH (smart hydrogels), where again molecular self-assembly is the driving force.

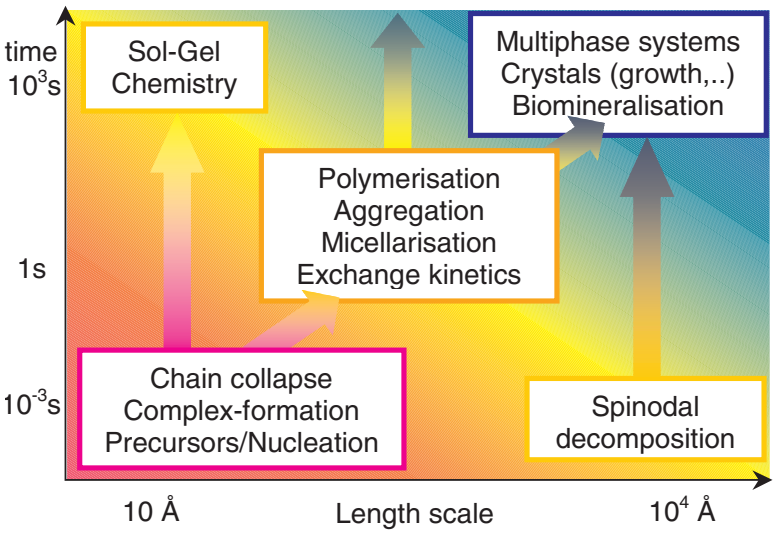


Figure 4: Kinetic map for structure formation in soft condensed matter systems.

Window to biology

Statistical concepts for understanding common features of different systems are the basis of soft matter physics. This approach can be translated to gain insight into the functionality of biological systems.

The methods and concepts of soft condensed matter physics in many cases provide the key to understanding the functioning of biological systems. These methods have particular power when the problem to be addressed is not one of specificity at the atomic level, i.e. it is valid for a class rather than a single system. Neutrons are effective in this area because (i) they probe a length scale appropriate for structure formation in many biological systems (1-100 nm) and (ii) isotopic substitution can be used to highlight the feature of interest. The possibility of accessing these phenomena is at present marginal, but the ESS will allow the implementation of a programme of systematic study in this field.

Conformational transitions in biomacromolecules

Biological macromolecules undergo a wide range of conformational transitions which adapt their functionality to the changes in microenvironment, e.g. helix-coil, coil-globule transitions, and the process of adsorption. Up to now most experimental efforts have focussed on equilibrium conformations, whereas kinetics is usually the controlling factor, especially in conformational changes in the actual biological cell. Neutron scattering has been an excellent tool for following equilibrium conformations and ESS will extend its range into the realm of real time biophysical processes.

Figure 5: Typical computer simulation snapshots of conformations of a semiflexible elastic filament (modelling a DNA macromolecule) in the course of the collapse transition with the formation of a toroidal globule.

The conformation of biomacromolecules can also be changed by the use of additional components such as surfactants and synthetic polyelectrolytes. This may lead to partial folding or unfolding, redistribution of the location of hydrophilic and hydrophobic regions, or change the properties of adsorption on a membrane. These interactions can also lead to the

creation of new objects, e.g. capsule formation around proteins or DNA, which can be of pharmaceutical importance for controlled and targeted drug transportation and release. The ability to distinguish different components by isotopic labelling makes neutron scattering uniquely powerful for studying the equilibrium conformation of these complexes, as well as the kinetics of the processes involved.

Minerals in biology

Inorganic materials are an important component of biological systems, e.g. calcium carbonate and calcium phosphate crystals in skeletal structure. These crystals are formed in the presence of biological macromolecules which constitute an important part of the emerging structures. The methods that living systems have developed to generate and handle these materials are often very intricate and are only just starting to be understood. What is clear, however, is that the synthesis, transport, aggregation and final deposition of these nanocrystals involves a complicated sequence of interactions with biomacromolecules. In that, neutron scattering is particularly sensitive to the macromolecular component of the composite structure. It provides a unique tool to follow the evolution of the microstructure of these materials, utilising especially contrast variation of the aqueous environment. This applies also to biomimetic crystals grown in vitro in the presence of macromolecules or other large organic molecules.

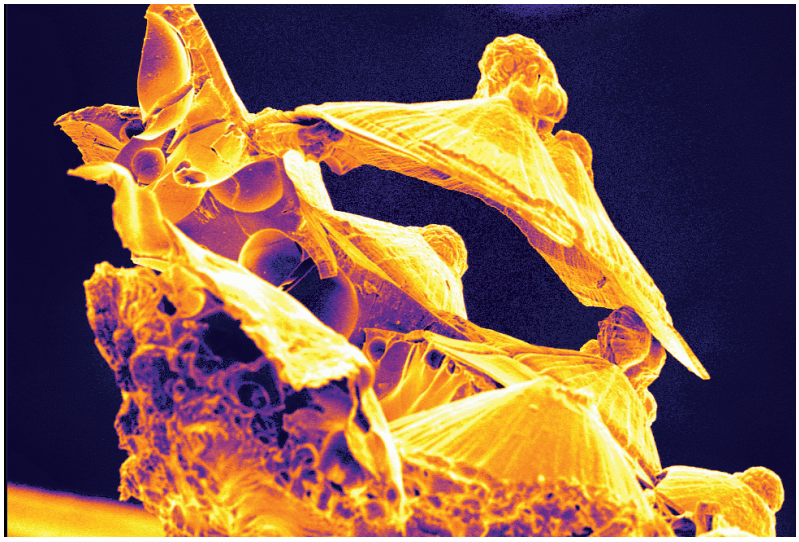


Figure 6: Fragments that are obtained by precipitating $CaCO_3$ in the presence of specific polycarboxylates. Without additive $CaCO_3$ usually crystallises in the form of rhombohedric crystals. The particles remind one of the complexity of structures occurring in the course of biomineralisation (size: 0.67x0.50mm) (J. Rieger, W. Heckmann, BASF Aktiengesellschaft, Ludwigshafen).

Self assembly and plant growth

The growth of plants is important in agricultural research and development. The strong influence of the environment (temperature, moisture, water, and their variation with time) demonstrates that it controls the self assembly of molecules

and macromolecules in the course of plant growth. For example, the plant cell builds out of its initial membrane a primary wall made of polysaccharides (cellulose and pectin based copolymers). The resulting cell then grows a secondary wall of hydrophobic polyphenols (lignins). The evolution of the interface between these two layers, which is crucial to the development of the plant, can be studied by using isotopic labelling to swell the hydrophilic layer with heavy water. Measurements can be done on model biomimetic interfaces using neutron reflectivity or on natural systems using SANS from a concentrated suspension of cell walls. The sensitivity offered by ESS will allow exploration of the key processes of triggering of the lignin stage and subsequent transport of the hydrophobic polyphenols towards the wall.

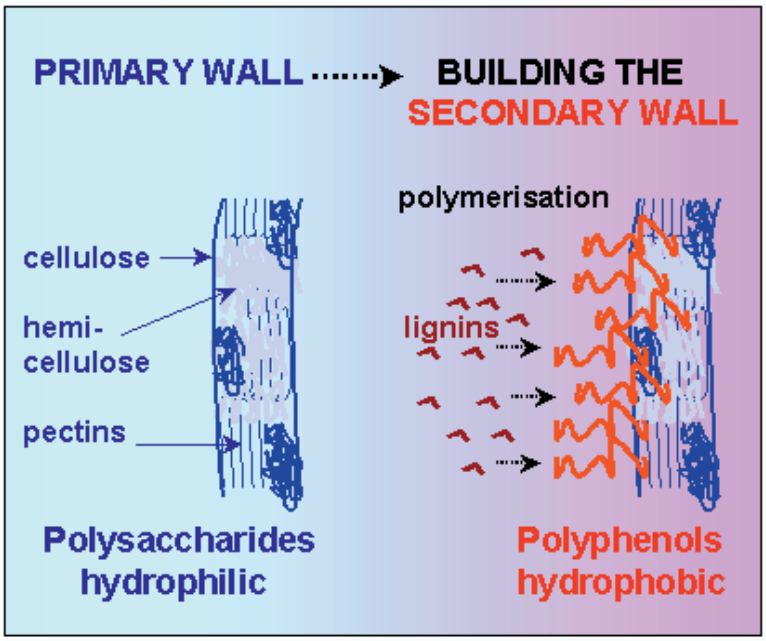


Figure 7: Schematic representation of the composition of the walls in the plant cell and how the secondary wall grows by polymerisation of lignins on the primary wall. Crucial questions like how the profile of the walls evolves with time, or whether interpenetration takes place could be addressed by neutron reflectivity and/or SANS. However, only the high intensity available with the ESS will make such kind of experiments feasible (Proposal by Catala et al., INRA).

New materials produced by external constraints

Combining phase transitions and external fields generates novel materials.

External fields can be applied close to phase boundaries to generate new materials. For example, blending two polymers above their separation temperature via shear and/or chemical reaction will produce an anisotropic structure, much finer than classical blends, which can be quenched to produce the desired material. Studying the successive stages following the onset of shear allows us both to select the best stage for application, and to understand the process. For example, as shown in Figure 5, flow enhances fluctuations of concentration perpendicular to flow and, due to the vicinity of the phase transition, these fluctuations diverge into alternately rich and

poor layers. Separation before shear would give a much coarser grained mixture.

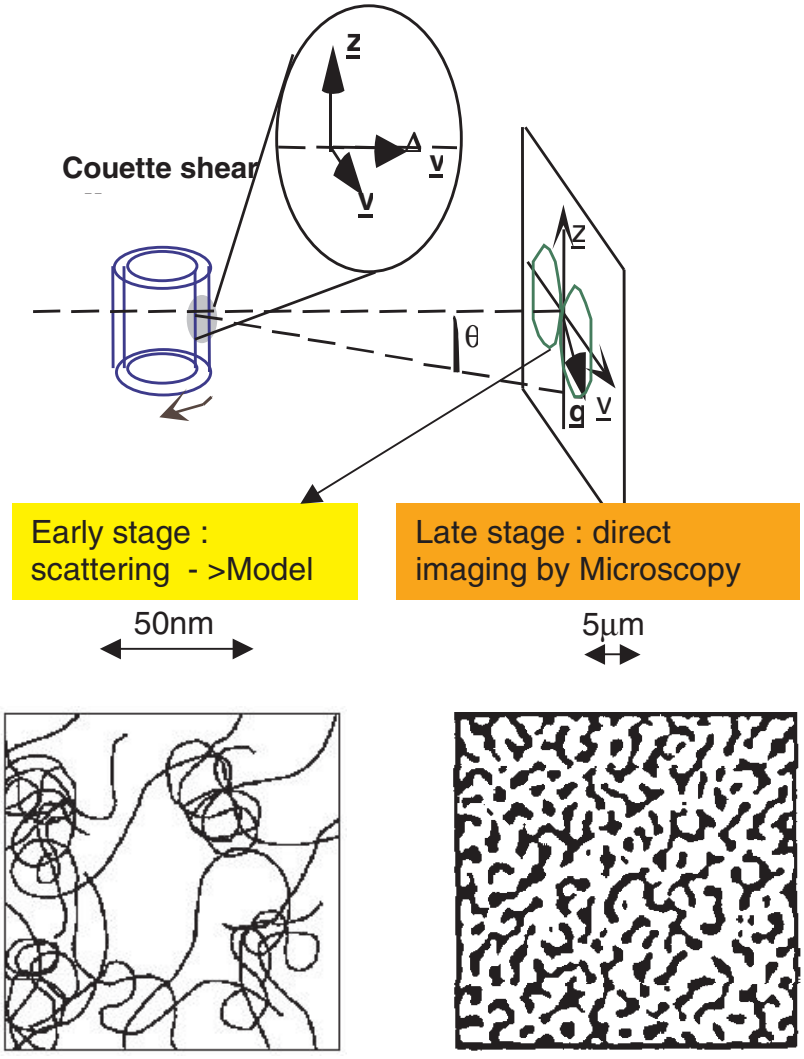


Figure 8: Snapshots of the evolution of a polymer-deuterated solvent mixture close to phase transition under shear: a) early stages: enhancement of concentration fluctuations, accessible only by SANS; b) later stages (microscopy): separation occurs also at larger scale (mm), the system is a multi-scale material.

These experiments really require a powerful neutron beam. The control of the final stage system needs extremely precise tuning because of the complex temporal evolution. In turn, this demands fast, in-situ and simultaneous observations over a full range of length scales in conjunction with measurements with other techniques. The latter are also essential for both industrial and scientific reasons: measurement of the stress, light scattering, or birefringence or microscope measurements are easy through a SANS shear cell. For example, in the ultimate stages a strong change in stress might result from phenomena at the micron scale. Without multiscale observation with light and neutrons, understanding is hopelessly complicated.

Soft-hard nanocomposites

Hybrids of crystalline inorganic particulates and polymers have been developed to combine the advantages of both classes of materials and widen their range of application. Enhanced mechanical, dielectric, fire-resistance, heat deflection, permeation-barrier properties have been reported or can be expected. The properties of particulate-filled composites are generally determined by the properties of the individual components, composition, structure (spatial distribution), particle-particle interaction and particle-matrix interaction. Apart from the processing conditions (shear forces), the extent to which particles agglomerate depends on the balance between the attractive and repulsive forces between the particles as well as between them and the matrix. Neutron scattering is a unique tool for studying the structure, interactions and dynamics of such complex hybrid materials. Partial structure factors in colloidal mixtures and colloid-polymer mixtures are for example the key quantities for a decisive test of the newly emerging theoretical description of the effects of interactions in these systems. However, the measurement of partial structure factors requires much higher neutron flux than presently available. Moreover, the length scales found in many of the application-oriented studies require a substantial extension of current instrumentation.

The temporal evolution of non-equilibrium structures is a key for understanding processing and performance of hybrid systems.

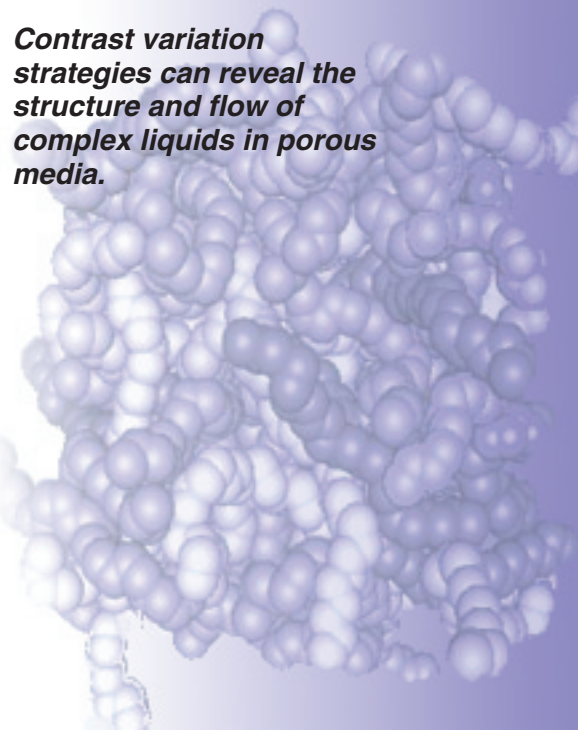
Key topics in such complex mixtures are kinetics and non-equilibrium aspects such as aggregation, gelation and/or phase separation. The temporal evolution of the structure of systems that undergo aggregation and eventually a sol-gel transition is a very interesting problem that is also directly related to recent progress in material sciences (novel non-metallic materials, ceramics processing, food science and technology etc.). However, this requires time-resolved SANS experiments in the second and millisecond time scale. This can only be achieved at much higher neutron flux than currently available, possibly combined with stroboscopic sampling and an extension of the accessible range of length scales by at least one order of magnitude.

Complex liquids in porous media

Understanding the behaviour of complex liquids in porous media is a particular challenge for the science of soft matter. By complex liquids we mean multicomponent systems of polymers, colloids, micelles and surfactants, whose characteristic length scales are frequently identical to those of porous materials. Complex liquids in porous media are of great practical significance. They are found in oil production where water comes up against petroliferous rocks, in processes separating materials through membranes, in remediating contaminated soils, in cleaning powders and pastes, and even in cosmetics.

Contrast variation strategies can reveal the structure and flow of complex liquids in porous media.

The coincidence of the characteristic length scales of the liquid and the geometrical constraints due to the pores has a profound influence on the thermodynamics, the phase behaviour and the transport properties of the complex liquid.



For example, a network of small pipes could be partially plugged by the oil droplets of a microemulsion. The ability to predict the behaviour of oil/surfactant, polymer/surfactant, polymer/protein or protein/surfactant systems in micro-environments based on a scientific understanding would enable effective control of many industrial processes.

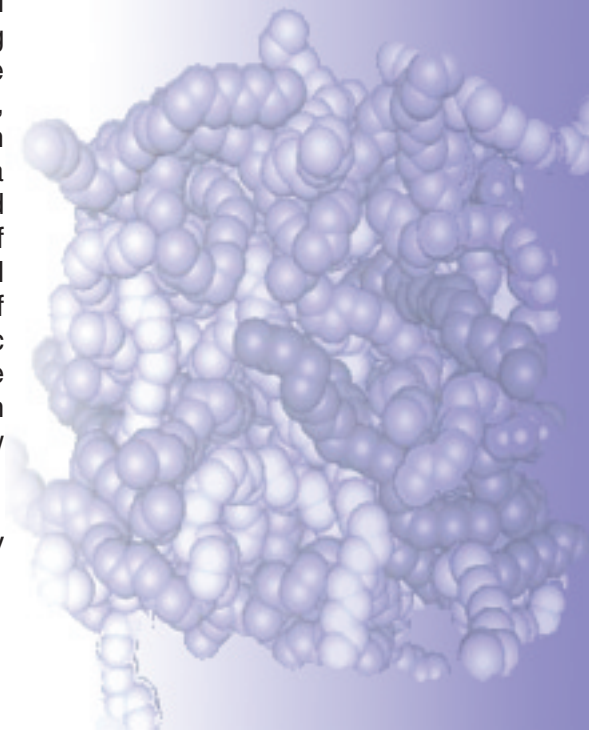
Without adjusting the mean contrast of the liquid to that of the porous material it is practically impossible to obtain direct information on the morphology and structure of liquids in the porous medium. In order to perform successful experiments a precise background control is indispensable. Furthermore contrast variation strategies involving several components need to be envisaged. This is only feasible with at least one order of magnitude higher fluxes than those available.

Probing molecular dynamics in non-crystalline matter

Dynamics of collective modes in non-crystalline materials provide a link between the motion of atoms and the mechanical properties.

The unique capability of neutrons to tell "where the atoms are and what they are doing" (Nobel prize citation 1994, Brockhouse and Shull) derives from the fact that inelastic neutron scattering can reveal the motion of atoms on the time and length scale of the atomic order and motion in crystalline material. The periodicity of the lattice allows us to draw conclusions on aspects of the atomic motion on virtually any length scale, including collective motion over distances much larger than the elementary cells, such as propagating phonon modes.

Currently much technological and fundamental research interest has shifted from crystalline materials to non-crystalline matter. In soft materials the structure, macroscopic mechanical properties, and phase changes are determined to a high degree by the motion of atoms, i.e. the molecular dynamics. This is quite understandable in the absence of a well defined crystalline order because the quasi infinite number of different local atomic configurations will include situations in which atomic rearrangements can easily take place by overcoming low barriers. The collective motion of atoms, correlated over distances of several atomic spacings, will be of particular importance for determining the material functions. Neutrons remain the unique probe for delivering information in the correct space and time ranges to explore and understand this intermediate scale dynamics. However, without the crystalline periodicity we have to observe all length scales between atomic and mesoscopic distances, which is a much more difficult task. For this reason we will have to record and analyse with precision signals which are 2-3 orders of magnitude smaller than those in crystalline materials. This will become possible for the first time with the 3 orders of magnitude gain in sensitivity ESS will provide for inelastic scattering spectroscopy with cold neutrons. Motions on the intermediate, typically nanometer, scale are naturally much slower than on the atomic time scale of 10^{-13} s, so slow neutrons are best suited for revealing them.



The interplay between neutron scattering, which is the only

experimental probe available to explore space and time behaviour at the intermediate microscopic/mesoscopic scale boundary, and advancing molecular dynamics model calculations will allow us to tell "where the atoms are and what they are doing" for the first time, in atom by atom detail.

IV. Instrument requirements at ESS

The instruments at ESS relevant for soft matter research can be put into three categories:

Soft condensed matter requires as top priority high intensity SANS, low Q SANS, high intensity reflectometer and high resolution NSE, at the long pulse target station.

i) Top priority
- High intensity small angle neutron scattering (SANS)
- Focussing low Q SANS
- High intensity reflectometer
- High resolution neutron spin echo

All flagship areas require large increases of intensity over previous sources in order to be successful. The $16^2/_3$ Hz long pulse target station will offer the required intensities with gain factors between 10 and 100 for our highest priority instruments. Resolution in wavelength is not crucial for the high intensity SANS and the reflectometer.

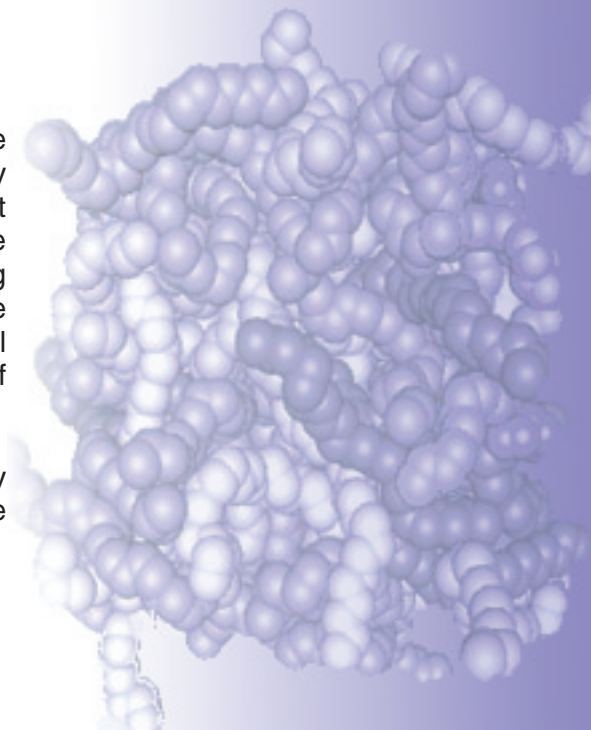
ii) Second priority
- Variable chopper cold time of flight
- Wide angle NSE
- Polarised diffuse scattering instrument of the D7 type

These instruments are also best placed at the long pulse $16^2/_3$ Hz target station, where they will have gains of up to 1000 over the best of their class currently available. This will create new opportunities for the study of local dynamic processes such as primary and secondary relaxations in polymers, soft vibrations and fast collective motions, all of which to a large extent determine the mechanical properties of polymers and composite systems. The D7 type instrument will allow the investigation of partial correlations and a more selective approach to local order in disordered materials.

iii) Third priority
- 1.5 μeV backscattering instrument
- Liquids diffractometer

The backscattering instrument is complementary to the wide angle NSE, but should be extended to a larger Q-range by providing Si(311) crystals. Whilst NSE gives time dependent relaxation functions directly, backscattering measures in the Fourier domain. For incoherent scattering backscattering nevertheless has the advantage that it does not pay the penalty of polarisation loss. The liquid diffractometer will supplement the D7-type instrument for the determination of local structures.

In summary, soft condensed matter research will definitely profit from the high intensity and large dynamic range of the long pulse target station.



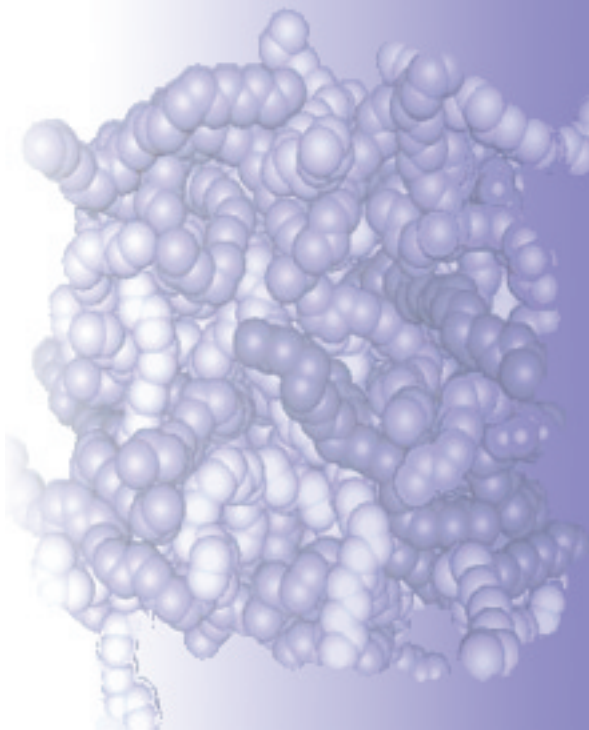
V. Summary

The science of soft condensed matter constitutes a field of large breadth and richness of phenomena, many with close links to technological applications. As a consequence of their unique properties, neutrons must play a key role for the exploration of this field. The decisive advantages of the neutron are the simultaneous accessibility to the proper length and time scales, together with the possibility of changing the scattering contrast.

Future developments in soft matter science will move towards the study of increasingly complex, often multicomponent systems, where time dependent phenomena in real time experiments, non-equilibrium situations and transient phenomena will be the focus.

Neutron scattering, in combination with computer simulations, will have a very large impact on all these future scientific endeavours. Finally there remains the possibility that the principles of soft condensed matter science will also have an impact on the better understanding of a number of biological functions.

All the future trends require a strong increase in the available neutron intensity. The prime instruments for soft condensed matter research all will be highly improved with the power of the long pulse target station.



Achievements of neutrons in soft condensed matter science

- Neutrons have provided direct information on the polymer conformation in melts and glasses corrobating the Gaussian coil structure based on the Nobel prize winning concepts of Flory (Nobel prize 1974).

- Spin Echo neutron spectroscopy has revealed the salient features of polymer chain dynamics covering simultaneously the proper length and time scales. In particular neutrons have provided direct evidence for the reptation mechanism proposed by Edwards and de Gennes (Nobel prize 1991).

- The pertinent scaling laws governing polymer solutions and phase transitions in polymer systems (blockcopolymers, blends) have been elucidated by small angle neutron scattering (SANS).

- SANS measurements on the molecular deformations in rubbers have clarified the basic mechanisms of rubber elasticity and have established scale dependent deformation mechanisms.

- Quasielastic neutron scattering has established that the relaxation of the short range order in glass forming polymer melts follows the temperature dependence set by the viscosity. Furthermore it was shown that the α-relaxation must be viewed as a sublinear diffusion process.

- Neutron scattering led to a quantitative understanding of methyl group dynamics in glassy polymers from quantum tunnelling to stochastic motion.

- Neutron reflectivity in combination with contrast variation methods elucidated the mechanism of interdiffusion between compatible and non compatible polymer films.

- The conformation of liquid crystalline polymers in the melt and the different liquid crystalline phases has been established by SANS using contrast variation.

- Neutrons have clarified the detailed structure of surfactant films at the air-liquid and liquid-liquid and air-solid interface. In particular the partial distributions of head groups, water and hydrocarbons could be established.

- The mushroom structure of amphiphilic blockcopolymers anchored in the surfactant films of a microemulsion was established by double contrast variation. In such systems the logarithmic renormalisation of the bending elasticity was directly measured.

- The mechanism of film formation of coalescent latex particles has been unveiled by in-situ real time SANS experiments.

4.5 Liquids and Glasses

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Abstract
Neutron scattering is a key experimental technique in the study of the atomic structure and dynamics of liquids and glasses. Neutrons at ESS will play a central role in studies using multiple complementary techniques, e.g. X-rays, light scattering and NMR, each providing information on specific aspects of the structure or dynamics of complex disordered materials. The data obtained will be simultaneously analysed with sophisticated modelling techniques or used as a stringent test of computer simulations. Such a coherent approach will not only enable a radical step forward in our understanding of the basic physical processes in disordered materials, but also in our ability to understand, control and eventually exploit the atomic scale structure and dynamics for the production of materials with optimised properties for technological and other applications.

Several 'flagship' areas of research can already be identified where major progress will be possible at ESS. Isotopic substitution and polarisation analysis will become routinely used in inelastic scattering experiments to enable the determination of partial dynamical structure factors. The ability to measure such data accurately over a very wide range of momentum ($\hbar Q$) and energy ($\hbar\omega$) transfer, coupled with ever increasing computer power, will enable visualisation of 'where the atoms are and what the atoms do' through the production of structural and dynamical models in real space and time. Inelastic small angle neutron scattering, or 'neutron Brillouin scattering', will provide key information on, for example, the transition from classical hydrodynamics to generalised hydrodynamics in liquids, or from extended to localised modes in glasses, or on the origin of the 'Boson peak'.

Extensive studies of the structural effects of different ions and molecules at different concentrations will be carried out in solutions, either aqueous or non-aqueous. There will be similar studies to explain the composition dependence of phase stability or ionic conductivity in complex materials for battery electrodes or electrolytes, fuel cells or sensors. Isotopic substitution, particularly H/D substitution, will play an important role here. Studies will be made under extreme conditions of temperature and pressure, to mimic conditions deep in the core of the earth or other planets. Some of these may use pulse techniques, or be 'single shot' experiments, taking advantage of the time structure of the ESS neutron beam. Microscopic structural models will be extended to mesoscale, and eventually connected to macroscopic properties.

A wide range ($0.1 < Q < 600$ nm^{-1}) liquids/glass diffractometer will be required for structural studies, together with a small angle scattering instrument whose range overlaps and then extends to lower Q. A diffractometer optimised specifically for total scattering studies of structural disorder in crystalline materials should also be considered. This is likely to be a growth area in the next decade. For dynamical studies a full set of spectrometers is required to provide as wide as possible coverage of (Q,ω) space. Particular emphasis should be placed on the small Q, high ω, region that can only be accessed using detectors at small angles. While some of this region can be accessed nowadays using X-rays it must be emphasised that, except in the case of single component systems (i.e. elements), the information obtained is complementary and not competitive.

I. Introduction

Disordered materials are common in daily life; water, windows, optical fibres, batteries, sensors.

Disordered materials play a central part in our daily life. Liquid water covers two thirds of the surface of the earth and is the major component of our bodies. The core of the earth is a liquid at high temperature and pressure. Glasses are in our windows and in optical fibres for communications. They are eaten as candy and used as stable coatings on medicines. Ionic conductors are in batteries in cars, mobile telephones and computers and in many types of sensor. Yet our understanding of such materials, especially in relation to more ordered crystalline materials, is still very limited.

During the 20th century scientific studies of condensed matter were dominated by ideas based on well ordered crystalline materials. The intrinsic symmetry of crystals can be used to

simplify theoretical descriptions to such an extent that they can be applied to 'real' systems. However the same ideas and methods cannot be directly applied to liquids and glasses, because of the lack of symmetry, and even in the case of highly disordered crystals they can be inappropriate or misleading. The study of disordered materials requires techniques that are optimised differently from those for crystalline materials. The information obtained from an experiment is continuous and different at all points covered by the experiment (there is no redundancy of information due to symmetry) so quantitatively precise measurements must be made at as many 'points' as possible.

'Model' studies are now being replaced by work on complex 'real' systems, often with applications. Many parameters must be varied. There is a real need for third generation neutron sources.

Until the beginning of the 1990's most research on disordered systems concentrated on 'model' systems from which we gained a general understanding of some of the basic physical processes determining their behaviour and properties. During the 1990's developments in neutron sources and instrumentation, and in methods of computer simulation and modelling, have led to a strong trend towards more complex 'real' systems, especially those with technological applications. Since most materials are not well ordered crystals, this work is increasingly important. The scientific aim is to extend our knowledge of the basic structure and dynamics of disordered materials, and their relationship with the interatomic interactions, to the point where we understand how this controls function, i.e. the macroscopic properties of interest. Usually this requires investigations over a much wider range of 'parameter space' than is feasible at present, including 'extreme conditions'. The critical information will be obtained from the derivatives of S(Q) and S(Q,ω) with respect to these parameters (e.g. composition for multi-component systems, isotope variation, T, P, E, B ...) measured over the widest possible Q and ω ranges and with more element specific information. The demand for information, produced by this trend towards complexity, will not be met by any single experimental technique. It will require all of the developments of third generation neutron sources such as ESS, third generation synchrotrons, computers and so on. When they are put together, these will offer tremendous new research possibilities.

II. The role of neutrons

The particular properties of the neutron make it a key probe for the study of liquids and glasses:

The neutron is a key probe for the study of liquids and glasses:

- ***large (Q,ω) range,***
- ***direct link to computer simulation,***
- ***isotopic substitution,***
- ***high penetrating power,***
- ***magnetic moment.***

- The ability to cover a large area of momentum/energy transfer space (Q,ω), i.e. length and time scales, well adapted to the length scale of the atomic structure and the frequencies of elementary excitations in liquids and glasses. Because liquids and glasses lack symmetry it is important to cover as large an area as possible.
- Due to the simplicity of the coupling function (it is a constant), the scattering can be measured on an absolute scale and directly related to the results of computer simulation/modelling, which considerably improves the possibilities for data interpretation. Hence the importance

of information from neutron scattering increases as computing power increases.

- The isotope dependence of the scattering cross-section can be used to mark an element in a complex material and thus give, in a unique way, detailed and element specific information on both structure and dynamics.
- The high penetrating power of the neutral particle enables studies of materials in containers or complex environments. This is a considerable advantage, for example in the investigation of liquids at high temperature.
- A magnetic moment, but no charge, enables detailed studies of magnetic structure and dynamics and allows the use of polarisation analysis to isolate information on the correlations of single atoms.

Other techniques, e.g. X-ray scattering, light scattering and NMR, can provide specific information and over a wider range of either Q or ω, **but not both**. Even when the range overlaps the information obtained is nearly always additional to that from neutrons, not the same. As the trend is towards the study of more and more complex systems we envisage that neutrons at ESS will be used routinely as the central component in a study using multiple complementary techniques. Data will either be analysed simultaneously using sophisticated modelling techniques, or used as stringent tests of computer simulations. Such a coherent approach to the studies of liquids and glasses will not only provide detailed information from experiments, but also enable a detailed interpretation of that information.

Neutrons at ESS will be used routinely as the central component in studies using multiple complementary techniques (X-rays, light scattering, NMR etc.) with data being analysed simultaneously using sophisticated modelling techniques, or used as a stringent test of computer simulations.

III. Scientific directions and opportunities at ESS

Fundamental studies of the atomic dynamics of disordered matter

The determination of partial structure factors, $S_{ab}(Q)$, and transformation to give the partial radial distribution functions, $g_{ab}(r)$, was a major past achievement of neutron scattering. Nowadays this enables a detailed understanding of the atomic structure of disordered matter. ESS will give us the opportunity to take a similar essential step towards understanding the atomic dynamics. This is considerably more difficult because it requires an accurate determination of the dynamic structure factor, $S(Q,\omega)$, over a very wide (Q,ω) range, with the additional complications of isotopic substitution and/or polarisation analysis, to obtain information on the dynamics of individual atom types in multi-component systems, $S_{ab}(Q,\omega)$ and $S_a(Q,\omega)$. Experiments that provide such complete information are not feasible at present sources.

In order to understand the dynamics of liquids and glasses, at the same level we now understand the structure, will require a third generation neutron source.

This will open the way for a completely new approach to understanding the dynamics of disordered systems. After double Fourier transformation of $S_{ab}(Q,\omega)$ to obtain $G_{ab}(r,t)$, which is not possible now because of the limited range and statistical accuracy that can be obtained, we will be able to interpret the dynamics in real space and time. In addition there

Analysis will be by direct comparison with molecular dynamics simulations or by iterative refinement of dynamical models.

will be the possibility for a direct comparison with molecular dynamics simulations, or for 'visualisation' via the iterative refinement of dynamical models, as is routinely done nowadays for structural models.

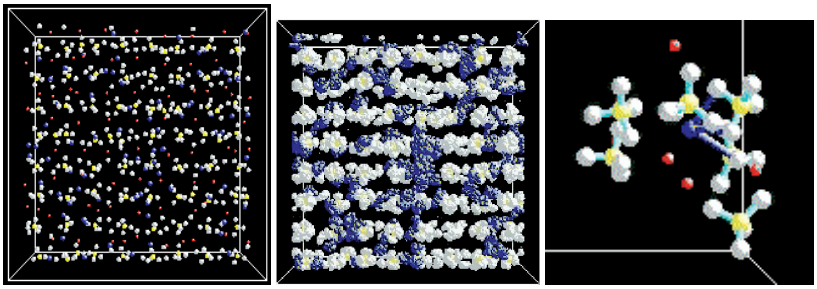


Figure 1: Different views from a 'movie' showing the correlation between proton transport and molecular rotation in the proton conductor $CsDSO_4$. ESS will allow this type of dynamical model to be produced directly from experimental data [P. Zetterström, A.V. Belushkin, R.L. McGreevy and L.A. Shuvalov, Solid State Ionics **116,** 321 (1999)].

The dynamics can be followed from the well understood region where pure hydrodynamics applies into the poorly understood region of generalised hydrodynamics.

The collective excitations (equivalent to phonons in harmonic crystals) carry essential information on the inter-atomic interactions. For such investigations the Q range below the main peak in the static structure factor is most important; here the static and dynamic properties are sensitive to the complicated, attractive part of the interaction. The ESS will give us the chance to investigate this region of dynamical space, which is very difficult to access, with sufficient intensity and resolution and over a sufficient energy range - in favourable cases even down into the Q region where pure hydrodynamics applies. This is one of the two firm and basic dynamical theories applicable to disordered systems. We will then have the possibility to study the transition from this continuum theory to generalised hydrodynamics where the parameters of the theory become functions of Q and ω (in an unknown manner). Studies of this kind also have to be extended to the partial dynamical structure factors, at present only possible in special cases. The partial collective dynamics, which depend on the element in focus, are expected to be rather different from the sum over all partials contained in the total dynamic structure factor.

Specialised inelastic small angle neutron scattering instruments will be required.

Another step towards a full understanding of the details of the dynamics will be the determination of the shapes and widths of the inelastic ("Brillouin") and the central ("Rayleigh") line. These two quantities contain essential information on the processes leading to the decay of collective atomic motions. Up to now collective excitations in classical liquids have never been observed as maxima or shoulders above $Q \sim 3\ nm^{-1}$, except for liquids with metallic pair interactions (screened Coulomb). In amorphous systems they contain in addition information on the mixing of modes, which from theoretical investigations are expected to exist as pure modes (plane wave like) only in the very low Q region. Extremely reliable and specialised inelastic small angle scattering spectrometers

are needed for this kind of investigation, sometimes known as neutron Brillouin scattering.

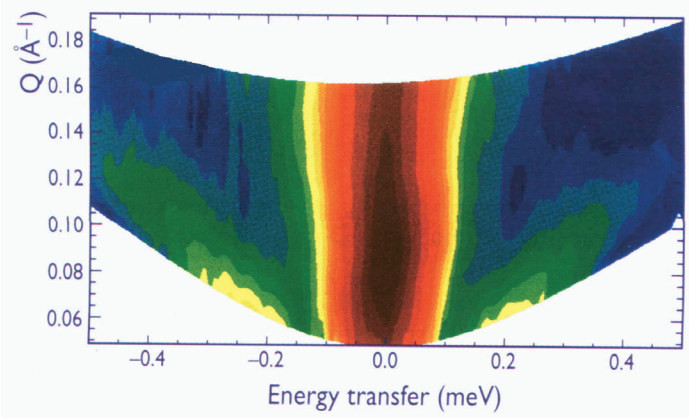


Figure 2: Dynamical structure factor S(Q,ω) for liquid Kr at T = 300 K and P = 800 bar, measured using neutron Brillouin scattering [P. Verkerk, Institut Laue Langevin Annual Report 16 (1996)].

Two level systems, an unsolved "mystery" of the dynamics of glasses, can be studied at ESS.

ESS will also allow us to address one of the unsolved "mysteries" in the dynamics of amorphous solids and defect crystals: two level systems (TLS) or tunnelling states. These excitations seem to have a broad distribution at rather low energies. Their density is very low (10^{-4} to 10^{-5}) and thus inaccessible with present neutron sources, but they strongly influence the thermal properties, especially below 1 K, the heat conduction around 10K (most likely causing the plateau region characteristic of amorphous solids), and other material properties. To study TLS we will need a very strong neutron source, isotopic substitution (to determine what is tunnelling), very high resolution (< 100 μeV) and low temperatures.

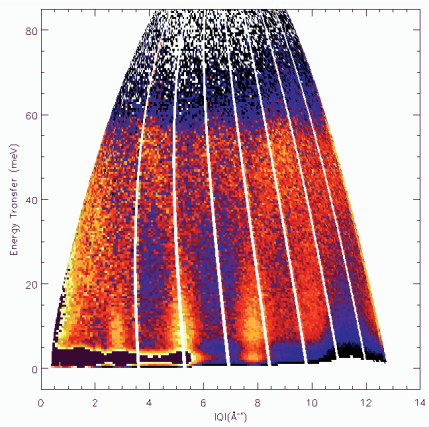


Figure 3: S(Q,ω) for vitreous SiO_2 measured on the MARI spectrometer at the ISIS spallation neutron source, illustrating the wide range of (Q,ω) space that can be covered. Important information is still missing in the region on the left hand side of the diagram [M. Nakamura, M. Arai, T. Otomo, Y. Inamura, S.M. Bennington, J. Non Cryst. Solids **377,** 293-295 (2001)].

Chemistry and life sciences

Absolute diffraction intensities and isotopic substitution are the key

In the past 25 years neutron diffraction investigations of the atomic structure of chemical liquids have assumed an increasingly pivotal role. A broad range of experimental and

theoretical techniques has been applied extensively to these systems, including several types of spectroscopy, X-ray and neutron diffraction, EXAFS (X-ray absorption fine structure), and both classical and "first principles" computer simulations. Neutrons often provide the fundamental benchmark data with which to compare and sometimes normalise other data, the interpretation of which is difficult due to unknown coupling functions. This is due to two particular properties of the neutron. Its simple, point-like, interaction with the scattering nucleus enables absolute diffraction intensities to be extracted, and different isotopes of some of the most relevant atoms (especially hydrogen) have sufficient contrast in their respective neutron scattering amplitudes to permit isotopic labelling of those atoms within a complex fluid.

features of neutron scattering for the study of solutions.

The earliest examples of this work were studies by Enderby and co-workers of the hydration of ions in aqueous solution in the 1970's. They used the technique of isotope labelling to map out the coordination structure around individual ions. For the first time it became possible to provide reliable quantitative answers to questions such as how many water molecules there are coordinating particular ions. Other techniques, such as X-ray diffraction and computer simulation, had failed to provide unambiguous answers. The same neutron data also provided early information about the likely orientation of water molecules around specific anions and cations. However, the fundamental question of how ions are organised in aqueous solution is still unresolved. Are they "charge ordered" as in a molten salt?

Ion-ion correlations in solution are still not well understood.

During the 1980's it was established that hydrogen/deuterium isotope substitution could be used to provide detailed information about water structure, initially in the pure state, but also during the 1990's, in both solutions and liquid mixtures. It was demonstrated that the neutron could be used to map out the fundamental correlations of the hydrogen atom on one molecule with atoms on neighbouring molecules, thus providing a direct and quantitative probe of, among other things, the hydrogen bond.

Neutrons can be used to study hydrogen bonding in pure water and solutions.

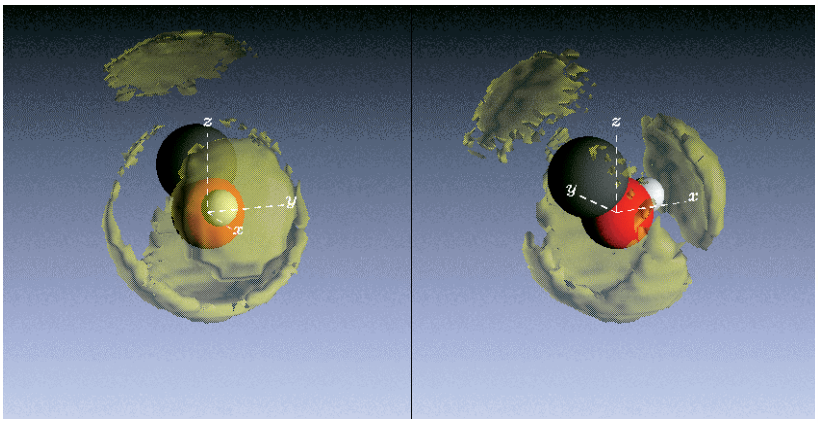


Figure 4: Isodensity surface plot showing the coordination of methanol molecules around a central molecule [A.K. Soper, Physica B **276-278**, 12 (2000); T. Yamaguchi, K. Hidaka, and A.K. Soper, Mol Phys. **96**, 1159 (1999)].

The next step has been made only recently. Neutron data with suitable isotope contrasts, perhaps also combined with data from other techniques such as X-ray scattering and NMR, can now be used to build realistic 3-dimensional models of liquids of interest in much the same way as has been done for the refinement of crystal structures for many years. This process of liquid structure refinement enables the experimenter to map out in unprecedented detail the way that water molecules, ions and other molecular entities are arranged. These methods have been applied to aqueous and non-aqueous systems alike, and a variety of molecular liquids have been tackled successfully.

Nowadays realistic three dimensional models of atoms and molecules in liquids can be built based on neutron diffraction and other complementary data.

Against this background the ESS presents an exciting opportunity to extend this work in different directions. What is now clear is that each ion or other molecular entity in solution has a subtly different effect on the solvent structure. Some ions, such as sodium, cause profound structural changes to the solvent itself, but others apparently have little effect. The bizarre fact is that some ion combinations or molecular species in solution can induce protein folding, while others cause protein denaturation. There is now little doubt that an important factor in this behaviour arises from the effect that the ions or other molecular species have on the structure of the water surrounding the macromolecule. It should be emphasised here that structural information at the level of detail that is attainable with neutron diffraction on these systems could not be achieved by other techniques, although those other methods can often provide important *complementary* data. The ESS will enhance this capacity even further by allowing the aqueous environment of larger molecular entities, of interest to the chemistry and life sciences communities, to be examined for the first time.

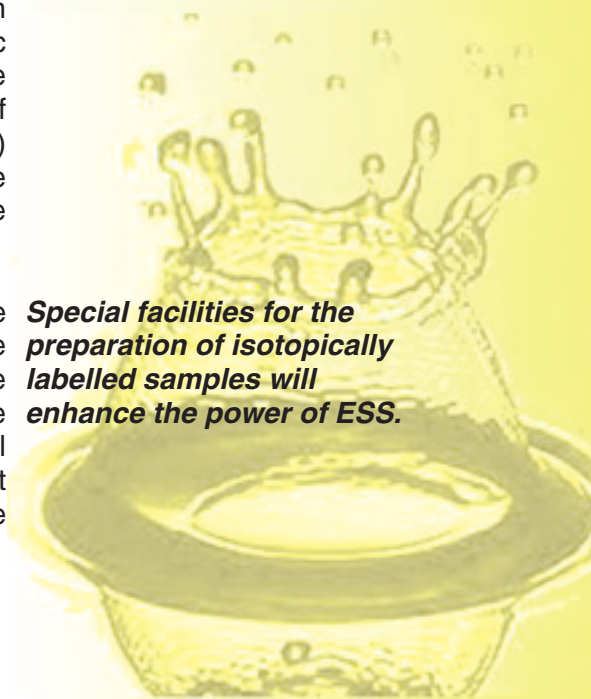
ESS will give an exciting opportunity to study how ions and dissolved molecules affect the structure of the water around them and have an influence on biological function, for example protein folding.

The type of study envisaged here is therefore a wide-ranging study of the effect of ions and other molecular entities on solvent structure. The parameter space to be explored is enormous, involving at least 100 different combinations of ion pairs in solution at several key concentrations. There is a large number of other molecular entities which need to be studied at the same time, both on their own in solution and in the presence of ions. Only by performing this systematic neutron study of enough of the relevant solutions will the characteristic structural trends due to different ions and molecules be identified. With current facilities the complete exploration of the dissolved species phase space (type and concentration) would take very many years to complete, particularly at the lowest solute concentrations where at present some experiments are hardly feasible.

An enormous parameter space must be covered in a systematic study. This is not feasible with current facilities.

A further stage of this work would be the exploration of the aqueous environment of large molecules in solution in the presence of different ions and molecular species, via isotope labelling of suitably synthesised macromolecules. The isotope contrast available for such work is likely to be low, and will require extremely stable instrumentation as well as the highest neutron fluxes available. It will also require dedicated sample

Special facilities for the preparation of isotopically labelled samples will enhance the power of ESS.



preparation facilities, including the production of isotopically labelled molecules.

More complex non-aqueous fluids can also be studied, or molecules as they interact.

The methods described above are not restricted to aqueous systems and a variety of other molecular systems can be tackled. Many chemical processes take place in much more complex non-aqueous fluids. Some examples are room temperature electrolytes, catalytic fluids used in polymerisation processes and “Lewis acids”. All of these systems can be explored at the atomic level with the methods described, in suitable cases before, during and after a chemical or physical change has taken place (if a real time experiment is possible). A feature of this kind of work would be the development of a chemical reaction cell, with the ability to probe particular samples with a variety of radiations and techniques in addition to neutron scattering. The data obtained will provide the essential information for a detailed understanding of how molecules interact in solution.

Materials science

ESS can play a key role in providing information at the atomic level to aid materials design.

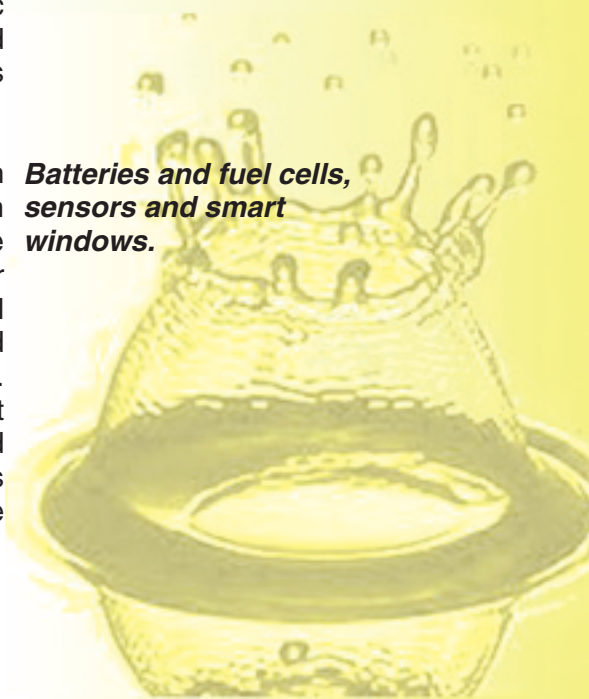
Materials science is increasingly concerned with a detailed understanding of the structure and dynamics of materials on a microscopic level and its relation to macroscopic properties and function, with the eventual aim of being able to design new or optimised materials on an atomic basis. Here we have chosen three of the many possible examples concerning disordered materials, where the application of neutrons from ESS will play a key role. Brief demonstration measurements are possible now, but the large number of components means that the wide ranging studies required for a detailed understanding can only be carried out at ESS.

Ultra-soft or ultra–hard magnetic metallic glasses or nanocrystalline materials.

Ultra-soft or ultra-hard magnetic metallic glasses can be prepared with direct technological applications (e.g. transformer cores, miniature induction coils in cars, various types of sensor) or used as precursors to obtain similar nanocrystalline materials. For optimisation we need to understand the structure, both atomic and magnetic, of systems with typically at least three atomic species. This will require the combination of neutron scattering (possibly including polarisation analysis to separate the magnetic contribution to the scattering), X-ray scattering, EXAFS and computer modelling for measurements on multiple samples covering a three component phase diagram.

Batteries and fuel cells, sensors and smart windows.

Ion conductors have an enormous range of applications in devices for energy storage (e.g. batteries), energy production (e.g. fuel cells), sensors and smart windows. These are increasingly important in modern society, with the demand for high efficiency, small size and weight, environmental friendliness and safe operation. Conflicting requirements lead to complex solutions, often without any real understanding. For example one needs to understand the apparent competition between favourable ionic conductivity and mechanical properties in polymer electrolytes, or the effects caused by local atomic correlations in hydrogen storage



materials. This requires structural measurements of multicomponent systems and dynamical measurements of the diffusion of dilute ionic species or the relaxation of polymers over a wide time scale.

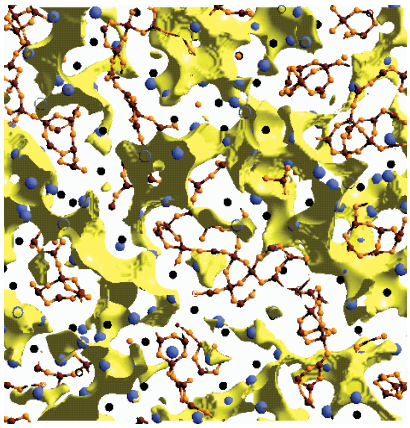


Figure 5: Model of conduction pathways in a Ag based fast ion conducting glass, determined from the combination of neutron and X-ray diffraction data and reverse Monte Carlo modelling [St. Adams and J. Swenson, Phys. Rev. Lett. **84**, 4144 (2000), Phys. Rev. B **63**, 054201 (2001)].

Waveguides, optical fibres, optical amplification.

The requirements for optical fibres, waveguides, optical amplification etc. can place extremely stringent requirements on the precise glass composition on an atomic scale. To control the transmission in a particular wavelength band, for example, we may need to understand the factors that determine the bonding of individual rare earth ions, or clusters of such ions, in a multi-component glass structure. These are usually at low concentrations and hence best studied with EXAFS. However the longer range correlations and the structure of the host glass must be determined by the combination of neutron and X-ray diffraction. Similar studies are required for the characterisation of glasses used for long term radioactive waste storage.

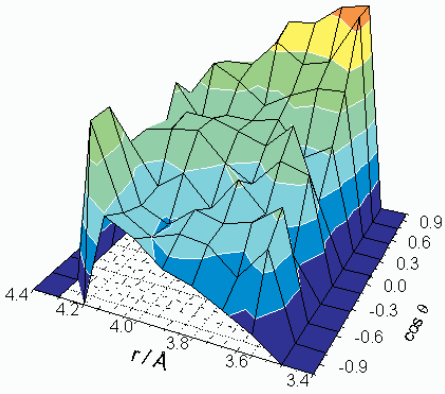


Figure 6: Angular correlation of neighbouring magnetic moments as a function of their separation (i.e. local spin-lattice correlations) for the CMR material $La_{0.8}Sr_{0.2}MnO_3$, determined by total neutron scattering [A. Mellergård, R.L. McGreevy and S. Eriksson, J. Phys.: Cond. Matter **12**, 4975 (2000)].

Techniques similar to those used to study the structures of liquids and glasses can also be used to study structural disorder and local structural correlations in crystalline materials. These are sometimes known as 'total scattering' studies to distinguish them from the elastic scattering studies of conventional crystallography. This is a relatively new field and as yet no diffractometer has been built which is specifically optimised for such work. The requirements are stringent; better resolution than a typical liquids diffractometer, but also better count rate and control of background since the diffuse scattering can be much weaker. ESS would offer considerable opportunities in this area. There are many possible applications to technologically important materials such as high temperature superconductors, colossal magneto-resistance (CMR) materials and ionic conductors.

Similar methods can be used to study disorder in crystalline materials, for example high temperature superconductors and colossal magneto-resistance materials.

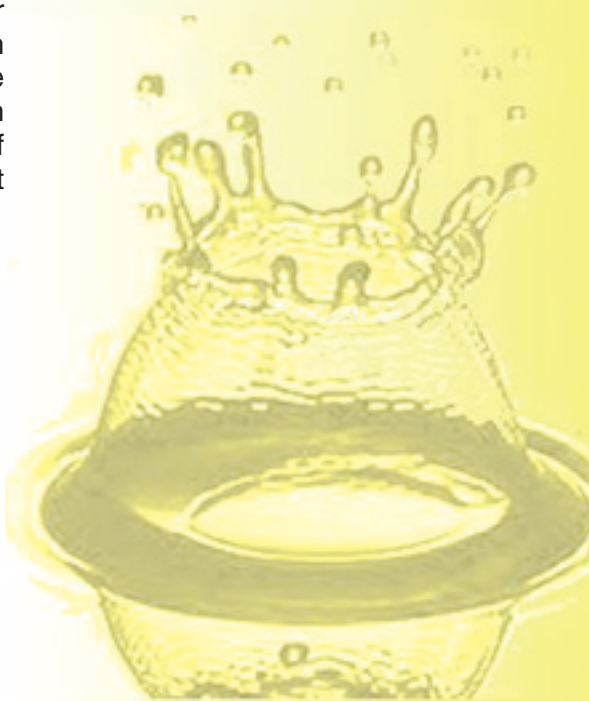
Extreme conditions, kinetics, melting and freezing, crystallisation and order-disorder transitions

Structural and dynamical investigations under extreme conditions will become possible at ESS due to the increased count rate, allowing for smaller samples (as used for high pressure studies) and faster data acquisition if required. When the extreme conditions are only present for a very short time, e.g. extreme temperature and pressure in a shockwave experiment, the diffracted neutrons from individual pulses could be used. In addition the instrument design of a time-of-flight diffractometer on a pulsed spallation source has intrinsic advantages for the layout of complicated sample environment, particularly pressure cells. Very high pressures, produced either statically or dynamically (shockwaves), are needed in order to understand for example the fluids which exist in the core of the earth and other planets. Even metallic hydrogen might become observable this way.

Systems under extreme conditions, such as very high temperature and pressure, can be studied, possibly in shockwave experiments.

The preparation of bulk amorphous materials, e.g. by annealing a quenched, crystalline high-pressure phase, can be investigated by neutron diffraction, which has the advantage that it averages over the bulk sample. Alternatively the crystallisation processes of amorphous precursors can be followed *in-situ* in real time by neutron diffraction. Powder pattern line profile broadening analysis, which requires a high resolution diffractometer, can provide information on the evolution of grain size. Time resolved small angle neutron scattering can provide information about the formation of grains. However the amorphisation/crystallisation rates that can be followed are limited by the power of current sources.

Crystallisation and amorphisation can be studied kinetically.



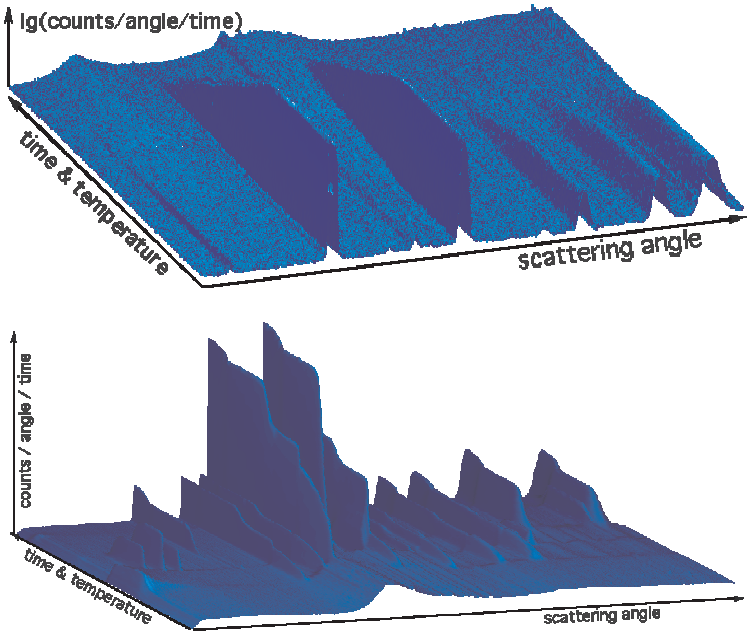


Figure 7: Amorphisation of crystalline GaSb and crystallisation of amorphous YFe studied by neutron thermo-diffractometry. [V.E. Antonov, O.I. Barkalov, M. Calvo-Dahlborg, U. Dahlborg, V.F. Fedotov, A.I. Harkunov, T. Hansen, E.G. Ponyatovsky and M. Winzenick, High Pressure Research **17**, 261 (2000); S. Kilcoyne, P. Manuel, and C. Ritter, J. Phys.: Cond. Matter **13**, 5241 (2001)].

With ESS the investigation of phase transitions (e.g. the liquid-glass transition) will be a matter of interest not only under equilibrium conditions, but also under non-equilibrium conditions in real time, i.e. the kinetics. Topics of study include phase separations in supercritical systems - in particular the solidification of super-cooled liquids - solution of solutes in a solvent, migration of liquids in porous media, ion migration in glasses, or glass formation using sol-gel processes. In some cases, e.g. fluid systems or migration in an electric field, the process to be studied is reversible and reproducible in a cyclic manner, allowing for repetitive, stroboscopic, data acquisition in order to achieve the required count rate. Performing such experiments at ESS will be an advantage where cycling is difficult to sustain over longer times. One can even envisage complex measurements where the time/wavelength structure of the pulse is utilised; the time resolution that can be achieved is then intrinsically limited by the path length of the neutron in the sample (of order 10 μs for thermal neutrons).

Strobosocopic measurements of kinetic processes will be possible down to 10 μs, an intrinsic limit.

In 'single shot' kinetic experiments a gain in intensity means directly a gain in time resolution. The limit on ESS will be set by the frequency of 50 Hz, i.e. 20 ms. However, even with ESS we are unlikely to reach such a short time scale when studying the structure of disordered systems, which require a much higher count rate than in the case of powder diffraction of crystalline materials.

'One shot' measurements of kinetic processes are limited to 20 ms by the source.

IV. Instrumentation requirements

High quality research on liquids and glasses requires instruments that are optimised for this purpose. In addition specific, fast, software must be considered as an integral part of every instrument.

Specialised software must be an integral part of every instrument.

Diffraction studies of liquids and glasses, or total scattering studies of disorder in crystals, require very high precision in intensity measurement, so they should be separated from 'conventional' crystallographic studies, even if the instruments are similar in some of their specifications. Only this will assure the meticulous control of background and stability that is required, but is less important for most purely crystallographic experiments. The liquids/glass diffractometer should have as wide a range as possible ($0.1 < Q < 600 nm^{-1}$); a complementary SANS instrument should overlap and extend to lower Q.

High precision measurement of diffracted intensity is crucial.

A complete set of inelastic neutron scattering instruments is required covering the entire energy range from neV to eV: spin-echo, backscattering, cold, thermal and hot time-of-flight and resonance detection. One key point for inelastic scattering studies of liquids and glasses is the provision of small angle scattering detectors, with evacuated flight paths to reduce background scattering. Such detectors are vital if full advantage is to be taken of the possibilities of ESS.

A complete set of inelastic scattering instruments with small angle scattering detectors is required.

Achievements in liquids and glasses using neutrons

- Structure of liquids and solutions. In early work isotope substitution was used to give information on the detailed structures of binary liquid metals and molten salts. This led to, for example, an understanding of charge ordering in ionic liquids. Isotope difference studies of ionic (aqueous) solutions allowed the solvation geometry to be determined directly, showing that tables of solvation numbers in text books were often totally wrong. The same methodology is now routinely applied to solutions of more complex molecules, polymers etc.

- Structure of metallic glasses. Almost all our knowledge of the structures of metallic alloy glasses comes from neutron diffraction with isotope substitution. This has led to important ideas on the role of concentration fluctuations in structural ordering. The only direct information on the magnetic structure of magnetic glasses has been obtained by neutron diffraction, using isotope substitution or polarisation analysis.

- Structure of network glasses. Precise neutron diffraction data over a wide Q range provides the benchmark for developing and testing structural models of glasses. Neutron diffraction, in combination with complementary techniques (e.g. X-ray diffraction, EXAFS, NMR) has confirmed many important structural models, such as Zachariasen's random network model or the modified random network model for alkali silicates. Neutron data have also been crucial in our understanding of the microscopic mechanism of ion conduction in glasses, including the mixed alkali effect.

- Disorder in crystalline materials. The techniques that have been developed for neutron diffraction studies of liquids and glasses have more recently been extended to studies of disorder (including magnetic disorder) in crystalline materials. Many of the materials studied have technological applications (ionic conductors, CMR materials, high T_c superconductors, ferroelectrics) where the properties of interest depend on the disorder.

- Dynamics of simple liquids. Inelastic neutron scattering has provided much of our basic understanding of both single particle motions and collective modes in simple liquids, neither of which could be explained by theory developed for crystals. The results of 'neutron Brillouin scattering' experiments now drive the development of extended hydrodynamic or modified kinetic theories. More complex behaviour (e.g. 'fast sound') has been identified in multi-component liquids.

- The glass transition and mode coupling theory. Neutron scattering has been the most important technique in studies of the glass transition, particularly in testing the predictions of mode coupling theory and indeed in driving its development. The ideas and techniques that have been developed from work on simple liquids have been extended to routine use in polymer studies and in biophysics.

- Theory and computer modelling/simulation. The simplicity of the neutron-nucleus interaction means that structural and dynamical data can be measured precisely, normalised to an absolute scale, and related directly to the predictions of theory or computer modelling/simulation. Neutron measurements (mainly on simple liquids) have therefore had a major influence on theoretical developments, such as memory function formalism or mode coupling theory, and have similarly played a central role in the development of computer simulation techniques (e.g. molecular dynamics). These are now used widely in physics, chemistry, materials science, life sciences etc. The potentials used in simulations are almost always validated by comparison with neutron scattering data.

4.6 Biology and Biotechnology

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Abstract
Neutrons have a unique role to play in determining the structure and dynamics of biological macromolecules and their complexes. The similar scattering signal from deuterium, carbon, nitrogen and oxygen allows the full determination of the positions and dynamics of the atoms “of life”. In addition the negative scattering length of hydrogen allows the well-known contrast variation method to be applied to dissect the component parts of multimacromolecular complexes. In the post genomic era structure determining techniques are reaching towards high throughput and a high number of proteins to be investigated. Thus, the ESS will offer major gains in neutron capability over the current technical frontier with reactor source technology whereby smaller samples, smaller quantities and lower concentrations all become viable. Thus, the major structure and dynamics techniques of protein crystallography, small angle neutron scattering, inelastic scattering and membrane reflectometry will all benefit in a major way. The considerably reduced measuring times will allow native rather than artificial membranes to be probed by reflectometry, including membrane bound proteins at the surface of actual cells. From the membrane biophysical studies via such native state reflectometry new nano-composites can be envisaged and designed. Structural biology, as well as biotechnology, will benefit from the powerful ability of neutrons to contribute to the location of hydrogen atoms and water molecules in biological systems. Thus it will contribute to the production of missing complementary data relevant for molecular modelling and to the strategy of rational drug design, in synergy with other biophysical approaches.

I. Introduction

We are in the post genomic era (Figure 1).

The present major driving forces of life science at the molecular and cellular scale are functional genomics and proteomics. Information on the specific functions of most if not all proteins encoded in human and other genomes is desirable. Major obstacles are the vast complexity of the individual proteins and the even more delicate interaction of different proteins and other biomolecules to form (transient) functional complexes. Neutrons are a unique non-destructive tool for probing precious biological molecular samples [e.g. see ref. 1].

Biology strives to understand molecular recognition.

Neutrons provide physiologically relevant, and yet precise, structural data.

Crystallography, SANS and inelastic scattering of biological molecules constitute major components of the future needs at ESS for 3-D structure analysis. In addition reflectometry on native systems will allow 2-D systems to be studied. Hence in all these methods the role of neutrons as a non-destructive and non-absorbing probe, with contrast variation involving H/D exchange, place neutron scattering as a unique approach. X-ray scattering and, in a different range of accuracy as well as time scale, NMR, provide, of course, important results in structure-function biology. However neutron studies more efficiently locate functional important water molecules and hydrogen atoms, especially at temperatures which are near physiological, rather than at cryo-temperature by SR X-ray protein crystallography. The structural precision of neutron room temperature data is routinely better than NMR atomic position data.

Combining neutrons, as a non-destructive probe of matter, with other techniques.

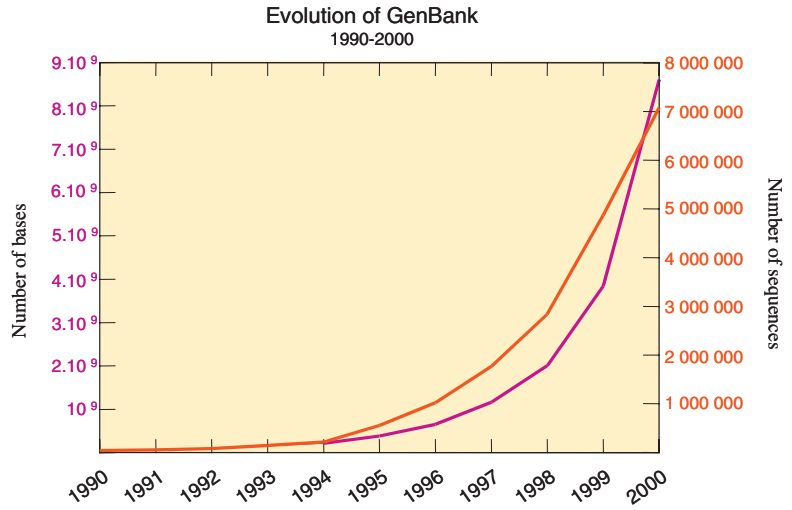


Figure 1: We are in the post genomic age of vast quantities of gene sequence information being available.

II. The role of neutrons

Neutrons are well suited to reveal water and hydrogen.

Hydrogen and water are involved in all the molecular processes of life. Very seldom is information on these aspects taken into account and in any case this experimental information is mostly incomplete. *If only neutron capabilities in protein crystallography could reach the throughput of X-ray work then the impact of neutrons would radically alter this situation.* In fact the research and development has largely been done with some very fine scientific examples e.g. on the detailed dissection of enzyme mechanism involving hydrogen location. Since many enzyme reactions involve hydrogen there is great potential for wide application if the technical capability can be found. Moreover the role of water in molecular recognition is pivotal as, for example, the lubricant of protein ligand interactions or the bonding mediator. Structural definition of bound water by X-ray diffraction is very sensitive to mobility especially at room temperature. Neutrons have the advantage that the scattering of deuterium is equal to that of the water oxygen. The incorporation then of the full structural detail of bound water can radically alter the modelling of proteins *in silico* for the improved discovery of new compounds, for example as leads in drug design. There are two major hurdles for wide application of neutron protein crystallography; firstly the size of crystals routinely available and secondly a molecular weight ceiling of about 30 kDa; the molecular weight histogram in the yeast genome for example peaks at 30 kDa and so at least half of all proteins in the genome are out of range of current neutron protein crystallography capabilities even if big crystals can be grown. This situation should change radically with ESS.

Molecular recognition involves water

The small crystal challenge

A molecular weight ceiling

In the modern trend towards high throughput, as well as high resolution structure determination, many proteins will not readily crystallise. The field of solution scattering can offer the chance to help determine the fold of a protein in solution. Efforts have started in this direction and show promise but it is strongly felt that the use of deuterated specific amino acids can, with small angle neutron scattering (SANS), provide the

Not all proteins crystallise

Helping protein fold discovery

much needed breakthrough in this field. Major gains in neutron flux to reduce especially the quantity and concentration of protein required will be of interest for higher throughput fold discovery. SANS at ESS could do this.

Dissecting complexes via contrast variation.

Neutrons provide unique possibilities in solution scattering studies of biological macromolecules, primarily thanks to contrast variation by H/D exchange. For multi-component systems (e.g. the ribosome (Figure 2), nucleoproteins or lipoproteins) information about the distribution of the components has been obtained by variation of the D_2O content in solution. Moreover, specific deuteration permits to highlight and analyse selected parts of macromolecular structures *in-situ*.

Neutrons are crucial to biophysics.

The results provided by neutron scattering are highly complementary to other analysis techniques (small angle X-ray scattering (SAXS) and diffraction, electron microscopy (EM) and analytical ultra-centrifugation (AUC)). Moreover, structural models built on the basis of neutron scattering data are able to incorporate information from different high and low resolution techniques and possibly also reconcile concurrent models.

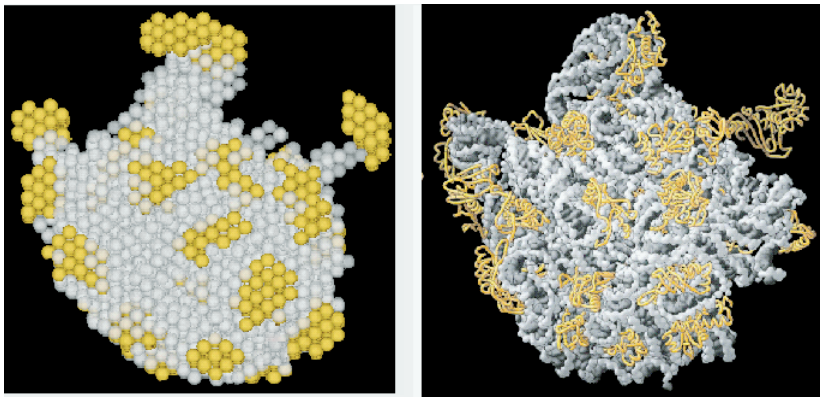


Figure 2: Comparison between the 50S subunit in the map of the 70S *E.coli* ribosome obtained from solution scattering [2,3] (left, resolution 3 nm), and the crystallographic model of the 50S ribosomal subunit *H.marismortui* (right, resolution 0.24 nm, Figure adapted from [4]. Yellow, ribosomal proteins, grey, ribosomal RNA.

Macromolecular dynamics are poorly understood.

Neutrons can reveal molecular vibrations.

Despite its acknowledged importance for biological function and activity, the dynamics dimension in molecular structural biology remains difficult to characterise and poorly understood. Neutron scattering is perfectly and uniquely suited to the space-time window of biological macromolecules. Protein function and activity, including enzyme catalysis, ligand binding, receptor action, electron and proton transfer, are strongly dependent on internal dynamics and conformational fluctuations or rearrangements such as postulated in the induced-fit hypothesis that long ago replaced the lock and key view for enzyme substrate interactions. Dynamics experiments probe the forces that underpin a molecular structure and its fluctuations, and in

chemistry they provide a stringent test for a structural or interaction model. Static or time-averaged structural information alone is not sufficient for a full understanding of specific drug binding to a protein or DNA, for example, and the molecular dynamics dimension (at the quantum chemistry and molecular mechanics levels) has to be taken into account in a "docking" experiment or calculation. Although here too it will be essential for our understanding of their function, there is practically no information available on internal dynamics at a higher level of molecular organisation in cells, protein-protein assemblies (e.g. large chaperones, multi-subunit proteases complexes, multi-enzyme complexes) or protein-nucleic acid assemblies (e.g. ribosomes, chromatin, transcription factor DNA complexes). In an original approach bridging molecular and cell biology, neutron experiments have recently provided data on the dynamics of proteins, *in vivo*, within their cellular environment (Zaccai pers. Comm.). Neutron scattering in fact provides unique opportunities to probe the natural cellular environment, which because of its molecular crowding properties, is very different from the usual conditions of laboratory biochemistry. The field of neutron applications to study biological molecular dynamics is wide open, with hydrogen-deuterium labelling allowing to focus on the dynamics of amino acid groups within a protein, or protein domains within a complex. Sample requirements, however, are at present unrealistic for the technique to accomplish a definitive impact. The ESS should provide a factor of at least 100 gain due to the source and optimised instrumentation.

The dynamics of molecular complexes remains understudied.

The cell is a crowded environment

ESS is a route to the real cell

Membrane biophysics studies on the molecular scale are crucial for understanding the self-organisation processes which underlie many functional aspects of the membrane, in particular membrane transport, molecular recognition on surfaces and adhesion between cells and substrates. While these problems have been studied in the past, largely in model systems, there is now a strong tendency towards studying far more complex *native* membranes. New preparation techniques allow native membranes (e.g. plasma membranes from eukaryotic cells) to be deposited onto solid substrates while maintaining their functional integrity. With faster data acquisition ESS should allow, via neutron reflectometry (NR), the study of membrane behaviour *in-situ* including membrane transport and membrane damage by invasive toxins.

Functional membranes will be studied

III. Future opportunities

In high throughput structural biology research the best sample size is rarely above 100 x 100 x 100 μm. It is essential for neutron protein crystallography to find source, instrument and sample (deuteration) combinations to face this challenge. Since we know from genomics that the average molecular weight of, for example, yeast gene products is 30 kDa, there are many projects that would become amenable for study by neutron protein crystallography methods if the sample size requirement could be relaxed.

Small biocrystals are usually the reality

ESS should allow the study of smaller samples

Large complex neutron crystallography at high resolution is a new frontier.

There is a barrier to the application of high resolution neutron structural study posed by molecular weight, which determines the unit cell volume, of large biological complexes. Such weakly scattering crystals cannot be studied currently (Figure 3). If, however, we combine the ESS source and instrument improvements, and improved knowledge of the protein preparation and crystallogenesis for the growth of large crystals, the unit cell size capability could reach 250 Å. Thus, for example, the small subunit of the ribosome could be amenable to study by neutron protein crystallography methods.

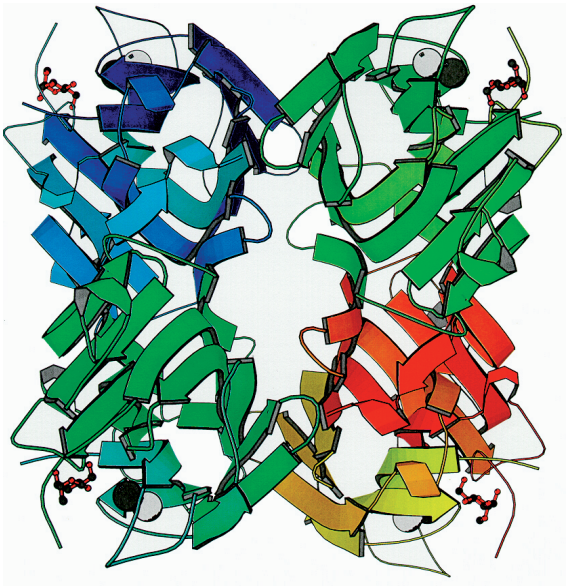


Figure 3: The determination of the hydrogen atoms for this tetrameric complex of concanavalin A with glucose is out of range of both SR X-ray and neutron techniques. The crystals are not stable in cryoprotectant and room temperature SR undulator diffraction (ESRF Quadrigia) is only at 1.9 Å resolution. On the LADI ILL instrument with a D_2O soaked crystal and a crystal size of 4x3x2mm the neutron diffraction resolution is 3.5 Å, insufficient for atomic analysis. An optimised ESS 'Large Molecular Weight' nPX instrument could bring the 50 kDa in the asymmetric unit crystal in range to find the sugar recognition hydrogen atoms (as deuteriums) [5,6].

Extending the resolution of SANS.

Recently, a new method has been developed to analyse wide angle X-ray solution scattering data up to 5 Å resolution in terms of the approximate positions of dummy amino acid residues. With neutron scattering, this approach can be extended to gain additional information about the positions of *individual* residues, either by the preparation of proteins with specifically deuterated residues or, for native samples, by making use of the change in contrast of residues during H/D exchange (e.g. after placing a hydrogenated protein in D_2O). This requires a neutron source with high flux and high dynamic range. This should contribute to the determination of the protein fold in solution from experiments on native samples and it would be an approach relevant for high throughput fold definition for proteins which do not easily crystallise e.g. detergent solubilised membrane proteins.

SANS is also used to determine the kinetics, stoichiometry and organisation of large macromolecular complexes (viruses, molecular machines such as chaperones, etc). According to its flux specification, ESS will reduce the quantities needed for these studies by at least ten. SANS will be used to study complexes identified in ever greater numbers by proteomic analysis. The products of structural genomics (i.e. atomic resolution structures) will be combined with the SANS results to deconvolute the complexes into their functional components (Figure 4).

SANS can help to define the make-up of molecular machines.

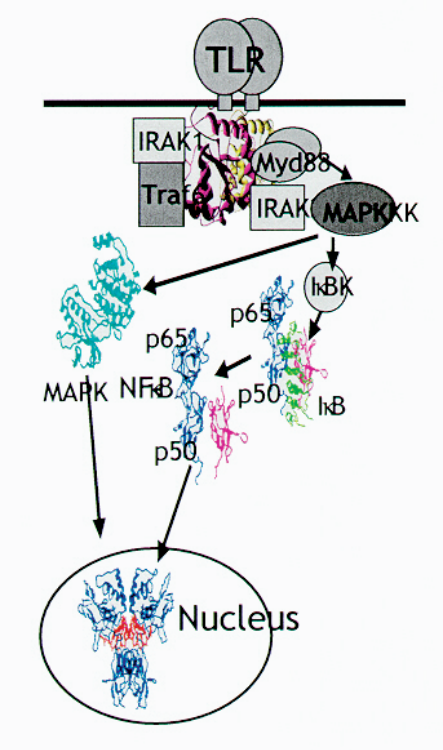


Figure 4: Schematic diagram of a typical eukaryotic cell signalling pathway. When the B cell receptor TLR2 binds its ligand it recruits a cascade of proteins in the cell cytoplasm: Myd88, IRAKs 1 and 2 and Traf6. This leads to the activation of MAPKKK and the phosphorylation and activation of the MAPkinase cascade and the IkBK signalsome. The resulting phosphorylation of IkB induces its ubiquination and degradation. Degradation of IkB results in the release of active p50/p65 components of NF-kB. NF-kB then translocates to the nucleus and transactivates immunomodulatory genes. Similarly, MAPkinase (MAPK) can translocate to the nucleus and activate transcription factors such as Elk, AP-1 and ATF-2. The resolution of structures for the constituent molecules (and their complexes) varies from zero to atomic. Neutron scattering and crystallography can help to fill in the gaps in this knowledge.

The long measuring times at current source flux levels restrict reflectometry and scattering studies to non-labile systems. Thus native membranes are not generally amenable to study. Major gains in peak and average fluxes open up such techniques to native, biologically relevant systems. This will widen the applicability of the knowledge gained to biotechnologies such as biosensors and nano biostructures.

Neutron reflectometry is a key to membrane-based biosensor development.

Taken together the above methods help create a cell model in silico; a virtual cell. The seamlessness and speed with which modern biologists can interrogate vast databases of genomic, proteomic and structural data was inconceivable to most in the field 15 years ago. How will we continue to improve the computer-user interface so that life scientists can maximise their use of these expanding reserves of knowledge? How can we ensure that all experimental data are being used, not just published? The integration of all existing databases into a virtual cell framework may accomplish this. The virtual cell will be a graphical, interactive, "functional" representation of a given cell (be it bacterial or even perhaps mammalian). The biologist will design investigations based on the response of the cell to imposed stimuli; the response will be the result of calculations made by the cell on the basis of literature data accessed from existing databanks. Neutrons should be significant contributors to the knowledge data base not only at the atomic level but more generally at the cellular and thus functional level.

The virtual cell

The role of neutrons in the virtual cell project.

Current and developing global research missions

How will the opportunities with neutrons at ESS map onto internationally agreed tasks and trends in the biosciences as well as open up new fields? In biology the post-genomic era, created by the speed and efficiency of gene sequencing, provides a huge stimulus to and the radical need for development of the structure determining techniques.

The 'business case' for ESS

New fields

It is a paradigm that structure is based on function. However this paradigm has been recast largely by neutron inelastic scattering techniques to include dynamics. Thus structure and dynamics determines function. The major gains in capability of ESS for inelastic scattering should greatly widen the applicability to many more systems.

Concerted action projects exist in structural genomics at world level. Notable cases include projects (i) in USA e.g. the Structural Genomic Program supported by the National Institute of Health (NIH) for high throughput structure determination of proteins from complete genomes (pathogens, arcahebacteria, …) and from human, mouse and other higher organisms (ii) in Europe on human proteins and also on pathogens and yeast and (iii) in Japan on various genomes including human and mouse proteins supported in particular by the Riken, the MITI and the Ministry of Education. Greatly expanded provision of synchrotron radiation beamlines for structural biology in the USA, Europe and Japan have been made for these projects and a general expansion of the field. Japan has a large new NMR park for structural genomics in Yokohama. The unique role of neutrons as a non-destructive probe of the structure and dynamics of biological macromolecules have been assessed by the USA and Japan who are now building state-of-the-art spallation neutron sources at the megawatt power level. The ESS reaches beyond even those power levels thus making a technical and scientific capability that is compelling.

Global concerted actions map well onto ESS frontier capabilities.

The USA and Japan have approved the construction of state-of-the-art megawatt level spallation neutron sources.

Finally the structure and dynamics results obtained will be applicable for industrial and biotechnology exploitation (see below).

Biological membranes are worthy of a special mention. The extreme sensitivity of neutron reflection makes it uniquely suitable for the study of labile biological structures [7,8,9]. The internal reflection at the solid/liquid interface combined with contrast variation allows the exact determination of the membrane structure and of the crucial polymer layer separating the biological membrane and the solid support. This will provide deep insight into the role of the soft polymer cushion for maintaining membrane integrity and function, crucial knowledge for the design of advanced biosensors. Moreover, using 2-D detection and in-plane Bragg scattering, there is the chance to study at molecular resolution the association and self-assembly of functional clusters in the plane of the membrane. This knowledge is crucial for the understanding of how proteins and lipids temporarily associate in a functional membrane. As a flagship experiment in this field we propose the study of native membranes and whole cultured cell layers on polymer cushioned solid substrates (Figure 5). This will permit the measurement of the cellular membrane response on the action of external stimuli (drugs, stress, pressure ...) at molecular resolution. It will also shed light on the extremely poorly understood interaction between different membrane constituents under conditions of membrane transport, ligand receptor binding and cell adhesion.

Advanced biosensors

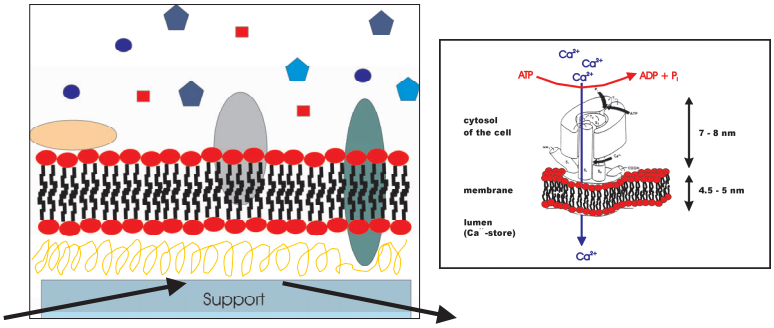


Figure 5: Neutron reflectometry. ESS fluxes will allow the study of native plasma membranes extracted from living cells deposited to planar substrates. A specially designed polymer cushion [9] between solid substrate and membrane will provide the soft interface required for keeping the membrane spanning proteins (ion channels, receptors, transporters) in their active state during the experiments.

Industry: implications in biotechnology and industry

Membrane science for biosensors and health.

The major advances in the fields of cell culture, softening and biocompatibilisation of solid surfaces, protein reconstitution techniques and nanostructuring methods can now provide native membrane samples of different protein and lipid composition in geometries that are uniquely suited for neutron studies. Membrane based biochips will become the key for

advanced biosensors, as well as screening and bioseparation devices. In particular, the combination of nanostructured semiconductor surfaces with native membranes via a soft polymer cushion represents a crucial technology for many applications in diagnostics and also in proteomics. Research and development in these fields amounts presently to $20 billion world-wide with an annual growth of 25 %.The enabling technologies that will emerge in the next years from this research will provide the tools for finding molecular markers for the early stage detection of illness and for the unravelling of the human proteome. Neutron data, if neutron developments cope with the need of the methods, on such complex systems can become a prerequisite in the design of even more advanced combinations of biological matter with solid surfaces for biochips including biosensors.

Rational drug design

The role of structural data in drug discovery in the pharmaceutical industry will increase when it is much more routine that hydrogen atoms, bound water deuteriums and the dynamics information can be incorporated. Thus the discovery of new pharmaceuticals and of enhanced efficiency compounds will accelerate. Also, because neutrons are non-destructive, unlike X-rays, room temperature structure and dynamics data can be provided.

Neutrons provide structure and dynamics data at physiologically relevant temperature.

IV. Instrument requirements at ESS

Target stations

50 Hz target is a popular choice for biology.

The 5 MW ESS targets are world beating. The 50 Hz short pulse target station is considered to be the most favourable choice for protein crystallography, dynamics, neutron reflection and in-plane Bragg scattering.

A SANS station on the long pulse target station should provide high flux and high dynamic range which would permit the use of smaller amounts and concentrations of material. The values of Q_{min} down to $5x10^{-4}$ and Q_{max} up to 1 $Å^{-1}$ are needed for detailing large complexes in solution and for protein fold definition, respectively.

Instruments

Three neutron protein crystallography instruments are proposed from the outset.

In neutron protein crystallography we forecast two main frontiers. The first is to considerably reduce the crystal size, including to reduce the data collection time from weeks to days, and the second frontier is to considerably extend the molecular weight capability to study complexes at high resolution (Figure 3, [5,6]). In the first case a brighter neutron source, well-focussed beams and smaller-pixel detectors will be in the design. To meet the second challenge we have to continue to harness the expertise of the crystallogenesis community to produce big crystals. Thus larger beams and bigger-pixel detectors are needed. This is a different instrument altogether. Also we should harness longer wavelengths to enhance the scattering-efficiency-with-

wavelength-effect as well. A methane moderator tailored to wavelengths 1.5 to 5 Å is being investigated. A third instrument should be developed at ESS for contrast variation at low resolution to dissect the intermolecular interactions in membrane crystals so as to better understand such crystallogenesis cases. There is a need here because the best current source, ILL, still has long measuring times (months). ESS will bring a benefit but perhaps demand will be low. Perhaps this third instrument could be shared with a high resolution SANS beamline.

High throughput SANS needs high flux.

In SANS we forecast an increase in the number of experiments that will be undertaken, not least because of genomics and initiatives in high throughput samples production, but also where experiments in solution will be at a premium (over techniques like crystallography where crystallisation is a recognised bottleneck to high throughput). The range of SANS experiments, tailored to molecular weight ranges that will be encountered, will be wide. Also the harnessing of as high an intensity as possible, to reduce the quantity of sample and the concentration needed, will be very important. Thus the long pulse target station in particular offers extra opportunities for realising such intensity gains for SANS.

Molecular dynamics served by the 50 Hz target station.

In dynamics experiments the instrument and target options need to match the time-space windows of interest. These are from the picosecond to the nanosecond or longer timescales, and from 1 to 10 Å length scales. Gains with respect to present ILL instruments will be > 100 at ESS. The 50 Hz target station is appropriate for these instruments. Instrumental priorities are for the variable resolution cold neutron chopper, multichopper spectrometers, the 1.5 μeV backscattering instrument and an 8 μeV back scattering instrument with large Q range (up to 5 $Å^{-1}$, similar to IN13). As a benchmark the ESS 8 μeV instrument will be > 500 x IN13 performance at ILL. These instruments would cover the elastic, quasi-elastic and inelastic resolutions implicated in the dynamics of biological systems of various sizes, from diffusing water molecules to domain motions in large complexes. Neutron spin echo (NSE) may in principle present interesting applications for studying slow coherent diffusive motions, but an optimistic estimate is that a gain of a factor of at least 100 on present ESS instrument conceptual designs would be required. The potential to use NSE to study the dynamics of small drug molecules or hormones in the context of their biological interactions should nevertheless be studied and assessed.

Matching the energies of life.

Observing function in the membrane plane.

The membrane and cell surface projects identified in sections I-III are largely not feasible with today's reflectometers because of the flux which requires accumulation times that are long for labile biological structures. The planned ESS reflectometer with its more than an order of magnitude higher flux will for the first time enable such studies. The desirable Q-range for these types of studies is up to 0.5 $Å^{-1}$ while resolution is not essential. Crucial will be the availability of

2-D detection and the option to measure Bragg scattering in the membrane plane.

Samples production is very important.

Sample production is as important as the source and the instrument for biology, especially in order to properly exploit the unique ability of neutrons to distinguish between major biological components. We must be able to produce and to label the constituents of our systems, be this individual atoms within a protein, proteins within a complex or complexes within a cell. Current, commonly-used, overexpression systems must be optimised for growth in deuterated media or in minimal media supplemented with deuterated amino acids (or sugars, co-factors etc). This must include not just the work-horse of protein production, *E. coli*, but also yeast, insect and mammalian cell systems. The future of molecular life sciences demands that we are able to successfully overexpress, purify and characterise fully post-translationally modified proteins. Further, if we are to achieve broad coverage of the cellular world in the virtual cell program, identified in section II, we must be able to characterise these cells in their entirety.

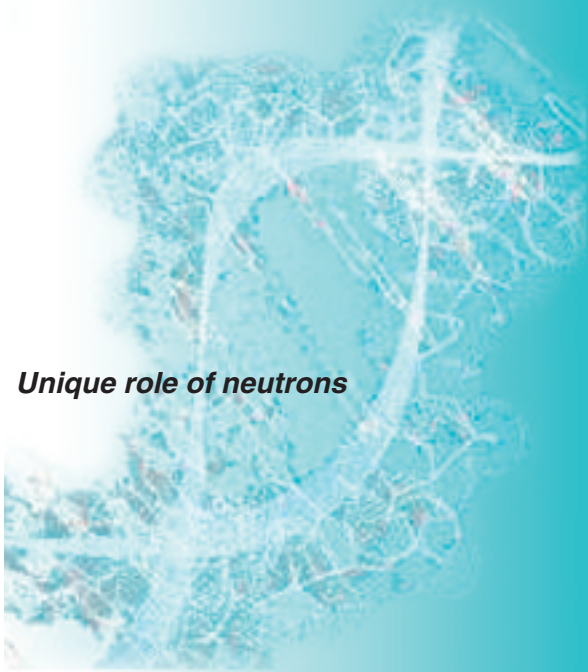
Labelling biomolecules

Multi-dimensional studies

Successful characterisation of biological systems depends on data from several orthogonal, complimentary studies. Increasingly, in order to ensure the reproducibility of conditions under which a system is studied and correspondence of the data so obtained, these studies are being carried out simultaneously. This is also efficient in terms of experimental time - a key factor in high-throughput post-genomic studies. Thus, in order to make a significant and rapid contribution to the databanks of the virtual cell (or even those in use currently) SANS can be coupled to capillary electrophoresis. Proteins within a cell extract will be separated electrophoretically in quartz capillaries as they flow slowly past an orthogonal, non-destructive neutron beam. Their meso-resolution structures will be restored automatically (using programs which will have evolved from DAMMIN, GASBOR etc [2,3]) and related directly to the electrophoretic band (and the identity of the protein obtained from MALDI-TOF analysis of digestion fragments thereof).

The combination of neutron reflectometry with either fluorescence microscopy (single molecule), plasmon spectroscopy or infrared spectroscopy can provide much additional information, in particular for the assessment of biomolecule interaction from the bulk with the membrane.
Neutron reflectometry is a natural partner for surface plasmon resonance (SPR) studies, although currently the timescales for these experiments are rather different. Nonetheless, SPR gives crucial information on the on- and off-rates of molecular interactions (even within deposited model membranes). The coupling of NR and SPR would permit the through-bilayer and in-plane structural characterisation of the equilibrium system (once binding saturation or de-saturation is reached). The design of a new reflectometer on a high intensity ESS should

Unique role of neutrons



be integrated with other biologically relevant and complementary techniques in a single setup.

Complementary aspects

There are of course other structure definition techniques such as X-ray crystallography and scattering, NMR, electron microscopy, mass spectroscopy, atomic force microscopy and light scattering. Data obtained with these techniques present key data to describe biological molecules and their complexes at different levels of detail. Neutrons provide a unique role in diffraction and inelastic scattering as a probe of biological structures because of the near equal scattering lengths of deuterium, carbon, oxygen and nitrogen and for dynamic measurements because the momenta of neutrons are matched to atomic vibrations. The additional possibility for harnessing the negative scattering length of hydrogen of course makes the unique contrast variation approach feasible. Thus the results provided by small angle neutron scattering are highly complementary to other analysis techniques (X-ray scattering and diffraction, electron microscopy, analytical ultracentrifugation).

Adding movement to static structures.

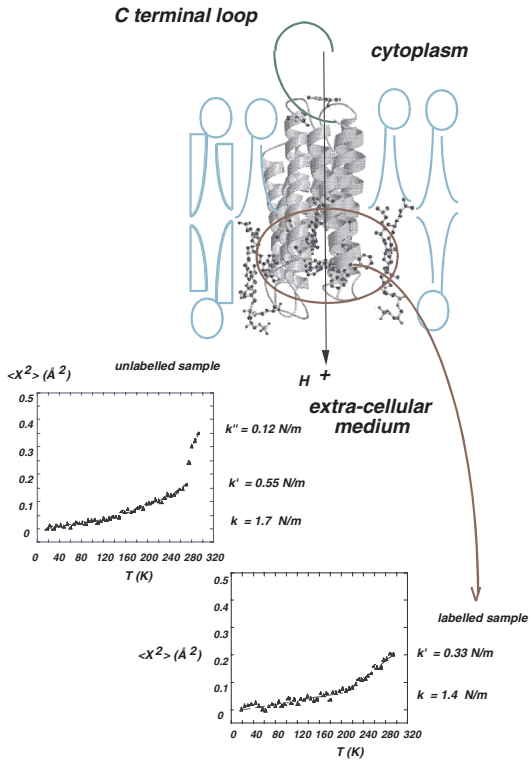


Figure 6: Dynamics-function relation in bacteriorhodopsin, the light driven proton pump in purple membranes, characterised by neutron scattering and specific isotope labelling of amino acids in the active core of the protein. An effective force constant, <k'>, for each part of the structure was calculated from the mean square fluctuations measured as a function of temperature. The protein, globally, is quite soft at physiological temperature with a <k'> value of 0.12 N/m. The core, however, is significantly stiffer with a <k'> value of 0.33 N/m, suggesting it ensures a valve function in the pump mechanism [10]. For a review see [11].

The structural models built on the basis of neutron scattering data and all other presently available methods allow to incorporate information from different high and low resolution techniques and possibly also reconcile divergent models. As a demonstrative example we can cite the determination by inelastic neutron scattering of the role of vibrational states in the function of the bacteriorhodopsin membrane proton pump (Figure 6), for which a high resolution structure is known.

V. Summary

Biology research at the structural level is expanding enormously. Vast numbers of structures will bring encyclopeadic power to help understand biological function. The 'toolbox' of methods involves complementary structural probes (X-rays, neutrons, electrons and NMR).

Many biological mechanisms involve hydrogen transport or transfer and/or the role of bound water molecules.

Neutrons are pre-eminent in their capabilities to yield a complete structure of a biological macromolecule i.e. including **all** of the ordered atoms in a sample, including hydrogens. This is without radiation damage and thus can be at physiologically relevant temperatures. Small but significant differences in structure at the cryotemperatures routinely employed with SR X-ray crystallography are seen versus room temperature. Multi-temperature structure results are also then a necessity. Could neutrons yield these results routinely? The low flux of neutron research reactors, even the ILL, and the very low flux of the currently most powerful spallation source ISIS prevents this. At ISIS the flux is so low that very little biological macromolecular structure work is undertaken. In Europe biology use at all neutron sources is a mere 4 % of the total whereas at ILL it is 12 % - 14 %. There is therefore a flux threshold effect. A 5 MW ESS offers substantive gains in measured signal to background via time-of–flight methods. Biology's share of neutron facility provision would get a big boost from a 5 MW ESS with gains of up to 30 (depending on unit cell size) for neutron protein crystallography over the ILL.

A further unique opportunity of neutrons is selective labelling and contrast variation using hydrogen/deuterium exchange. These techniques are extremely effective in SANS and reflectometry to localise specific fragments in complex structures (e.g. nucleoproteins and protein-membrane complexes). A 5 MW ESS, yielding a gain of more than an order of magnitude for reflectometry and up to two orders for SANS, would allow one to perform qualitatively novel experiments (e.g. labelling of individual residues in proteins, analysis of macromolecules, which are only soluble at very low concentrations, and detailed structure of native biological membranes from off-specular reflectivity).

A special place for neutron capability is in its exquisite sensitivity to dynamics of atoms. The pioneering works at ILL

on bacteriorhodopsin for example show that it is structure and dynamics that determines function. The 5MW ESS instrument gain factors are large. Thus such gain factors offer extension to larger time domain vibrations. Hence, as biosimulation methods are extending their simulation times towards the microsecond domain, the reality checks' of experimental data would come only from ESS inelastic neutron scattering results.

Overall, if biology, and biotechnology, is to fulfill its promise of being the subject of the 21st century then Facilities provision for the determination of the structure and dynamics of biological macromolecules must be extended. Neutrons offer a unique role and the R&D preparation for ESS is very mature. ISIS, building on the Japanese source work, provides the platform technically. Biology at ILL is showing what can be accomplished with a regular stream of examples: enzyme crystal structures with key hydrogen positions, bound water structure in protein ligand recognition studies, first time-resolved SANS of the chaperonin structural intermediates, the dynamical basis of the proton transfer mechanism in bacteriorhodopsin, reflectometry studies of biomembranes, to name a few. ESS would greatly extend this output.

Because it will provide drastically improved specifications, ESS can and should be an actor in the development of structural proteomics in Europe. In this intensive context, it will bring, with SANS, complementarity and unique tools for the characterisation of samples. Furthermore, it will contribute to the comprehension of the dynamic of biological systems and to the determination of structures at all levels of resolution including atomic resolution, to the identification and location of light ions, water and hydrogens atoms, in the case of molecules and integrated complexes (proteomes, transcriptomes, viruses, with a special mention to membrane proteins). This will be achieved by taking advantage of the progress realised in the field of recombinant proteins in bacteria, eukaryotes and cell free systems for expression of labelled molecules.

We are grateful to Dr. Patricia Bassereau and Dr. Jon Cooper for their careful reading and helpful suggestions on the text at the ESS Dourdan Workshop March 2002.

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Achievements of neutrons in biology and biotechnology

- The positioning of all the 21 proteins in the 30S subunit of the ribosome of E. coli, as well as a map of protein-RNA distribution in the entire 70S ribosome using small angle neutron scattering (SANS).
- The organisation of nucleic acid and protein in spherical viruses using SANS and low resolution neutron protein crystallography.
- Characterisation of nucleic acid protein interactions fundamental for gene regulation and expression i.e. in nucleosomes, aminoacyl-tRNA synthetase, DNA dependent RNA polymerase using SANS.
- Characterisation of intermediates in the functional cycle of the chaperone protein complex GroEL/GroES using SANS.
- Determination of function-critical hydrogen positions in enzymes, for example aspartic proteinase using crystallography.
- Determination of solvent organisation in proteins using crystallography (such as concanavalinA and myoglobin) and other biomolecular systems such as DNA and cellulose using fibre diffraction. Dynamics of labile hydrogens in different parts of lysozyme and other proteins. Orientations of ring systems and methyl rotors.
- Characterisation of single and multilayers (adsorbed proteins) at the air/water and oil/water interface using reflectometry.
- Detailed structure of the lipid bilayer using reflectometry and biological membranes using diffraction.
- Determination of the protein-detergent distribution in membrane protein crystals, and the localisation of lipid in a lipoprotein using low resolution crystallography.
- Characterisation of a dynamical transition associated with the solvent environment in myoglobin and other proteins using inelastic scattering.
- Characterisation of dynamics correlated with function in purple membrane using inelastic scattering. Structural information on this photoactive membrane, including hydration properties, localisation of the retinal chromophore in bacteriorhodopsin (the membrane protein), the positions of glycolipids and the structure of trapped intermediates in the photocycle using diffraction and selective deuteration.
- Characterisation of dynamics-stability relations in proteins from extremophile organisms (adaptation to extreme environments) using inelastic scattering.

4.7 Mineral Sciences, Earth Sciences, Environment and Cultural Heritage

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Abstract

The materials under study in Mineral Sciences, and Earth Sciences in general, pertain to all aggregation states of matter; solid, glass, molten, liquid and gas. Hence the methods of investigation are those adopted in all other branches of science dealing with such states. In the case of the Earth Sciences, however, we must consider a further degree of complexity introduced by the unrestricted nature of the chemical and physical parameters, i.e. chemical composition, temperature and pressure, underlying the formation and transformations of these objects.

Neutron scattering has only recently been added to the methods of investigation used in the Earth Sciences, mainly thanks to the latest generation of diffractometers and spectrometers at modern neutron sources which allow the accurate determination of subtle, yet very important, structural details in minerals as a function of temperature and pressure.

Still, many areas of Mineral Science research, and related fields, remain out of the reach of present-day neutron instrumentation. We have selected a few representative examples, and in turn a few keynote or "flagship" experiments that could be tackled with an ESS-type instrumental set-up, leading to a significant advance in this field of research. The three broad themes identified are:

1. In-situ measurements of structure-property relationships in mineral phases under geological conditions.
2. Structure, reactivity and physical properties of multi-component melts and fluids under geological conditions.
3. Texture and stress analysis of polymineralic rocks; modelling of tectonic processes and rock anisotropies.

Within these broad themes, four *"flagship experiments"* are offered as actual examples, namely:

1. - a) In-situ diffraction and spectroscopic studies of molecular components in methane clathrates.
- b) Spin dynamics of iron-containing deep Earth phases.
2. - Space- and time-resolved tomography of volatile-containing crystallising magmas.
3. - Time-resolved simultaneous structure, texture and stress analysis of polymineralic systems under variable P/T conditions.

I. Introduction

The Earth Sciences can profit greatly from neutron scattering studies using the latest and future generation neutron sources.

The use of neutron scattering by the Earth Sciences community has a relatively short history, but it is now clear that the potential of neutron scattering methods for the solution of Earth Sciences problems, including many environmental problems, is enormous. Many of the problems encountered in Earth Sciences have, until recently, simply been too complicated for earlier neutron sources and instrumentation. Only with the advent of the latest generation powder diffractometers at modern spallation sources such as ISIS, has it become possible to study the crystal structures of minerals as a function of temperature and pressure with sufficient accuracy to be really useful in solving subtle problems such as cation ordering. However, there are many areas in the Earth and environmental sciences for which the present sources and instrumentation are still inadequate. Examples include measurements of the structural changes in minerals at very high pressures and simultaneous high temperatures, locations of light elements in complex structures, strain measurements and scanning of polycrystalline aggregates under non-ambient conditions, and

studies of the dynamical properties (neutron spectroscopy), also at non-ambient conditions. This information will enable modelling of fundamental processes in the Earth, ranging from large scale phenomena such as deep-focus earthquakes and volcanic activity, through to the transport (and disposal) of pollutants in the Earth's crust and stone preservation in monuments.

Typical examples include hydrogen containing phases.

Recent experience with state-of-the-art sources, such as ISIS and ILL, has shown that the outlook for applications of neutron scattering in Earth Sciences is extremely promising. There are many features of neutron scattering that find ready application to the study of natural materials. One clear example is that many natural materials contain hydrogen. Hydrogen is virtually invisible to X-rays, but it will scatter neutrons reasonably strongly, both coherently and incoherently. This means that neutron scattering from hydrogen can act as a probe of both single-atom dynamics and collective excitations. Hydrogen is central to so many problems in geology and environmental science that there are countless important applications of neutron scattering in these areas. As the facilities for collecting high-quality data are further developed, so our ability to resolve these scientific issues increases.

In-situ studies of dynamic processes and phenomena.

The ESS will enable us to tackle many long standing issues related to geological and environmental processes. The ability to construct sample environments that will reproduce the temperature and pressure conditions of the deep Earth, and the increase in neutron intensity allowing reduced interaction volumes and shorter data collection periods, will allow us to perform many *"in-situ"* studies of mineral behaviour, which will greatly increase our understanding of the behaviour of the constituent materials of the Earth. The ability to probe the structures and motions of relatively complex minerals will provide many new insights and allow us to understand the numerous interactions that govern the behaviour of Earth materials in their natural environment. We anticipate being able to study both solids and fluids by neutron techniques, as well as for the first time the interfaces between these two states of aggregation.

II. Points of merit for neutron scattering in Mineral, Earth and Environmental Sciences

Hydrogen in minerals

"Water" in minerals and rocks is of paramount importance in determining their properties.

Neutrons, as opposed to X-rays, are efficiently scattered by hydrogen ^{1}H and deuterium ^{2}H atoms. Many minerals contain hydrogen, often in the form of bound or free hydroxide ions or in the form of bound or free water molecules, within either structurally active sites or interstitial cavities in the crystal structure. Water in minerals and rocks is extremely important in regulating a large variety of behaviours and properties of interest to the Earth Sciences spanning from the atomic to the continental scale. Because hydrogen has an extremely large

cross section for incoherent scattering, neutron scattering can be used to study the motions of individual hydrogen atoms. Slow motions of the hydrogen atoms, such as diffusion or reorientational motions, can be probed by quasi-elastic scattering. Fast motions can be probed by high-energy spectroscopy. On the other hand, since deuterium has a reasonable cross section for coherent scattering and no appreciable cross section for incoherent scattering, deuterated samples can be used in diffraction studies for the location of hydrogen sites in crystal structures and their modifications under inner earth conditions [1].

Scattering cross section

Thermal motions, site occupancies, short- and long-range order information are more easily detectable by neutron scattering.

The fact that the scattering cross section for neutrons does not change with scattering vector, whereas with X-rays it falls off approximately as the inverse of the atomic radius, means that neutron scattering allows us to collect diffraction data to large scattering vectors. This is useful for a number of reasons. (i) When investigating complex crystal structures and crystal chemistries, evident in many minerals, it allows for a significant increase in the amount of information available in a diffraction pattern. (ii) A wide coverage of scattering vector is essential for information about thermal motion. (iii) To extract information about site occupancies, and to decouple this information from the thermal motion, it is again essential to have data over a wide range of scattering vectors. Furthermore, in crystals with considerable disorder, or in amorphous materials or liquids, there is a lot of information about short-range order contained within the total scattering, S(q). The Fourier transform of S(q) provides information about the pair distribution function, g(r), the resolution of which will depend directly on the range of the scattering vector in the data. Thus one can obtain better data for g(r) from neutron scattering than from X-ray scattering, although it is most profitable, sometime essential, to combine data from both techniques, especially where complex systems are concerned.

Iso-electronic (and quasi-iso-electronic) species

Scattering contrast is not electron-dependent.

The contrast between the neutron scattering cross sections of mineralogically common atoms or cations which have equal or similar numbers of electrons, such as the following groups of ions; Ti^{4+}-Ca^{2+}-K^{+} K^{+}-Cl^{-} Na^{+}-Mg^{2+}-Al^{3+}-Si^{4+} Fe^{2+}-Mn^{2+} O^{2-}-F^{-}, as well as the corresponding atomic species and also the rare earth elements, allows neutron diffraction to be used for the direct determination of their site occupancies and order-disorder distributions. Untangling the ordering of these cations using X-rays can only be achieved indirectly by the analysis of bond lengths, but these are not definitive since bond lengths are affected by factors other than the specific site occupancy. Neutron diffraction provides the direct determination of site occupancies for these frequently coexisting cations in minerals. Furthermore, although synchrotron X-ray resonant scattering can certainly be used to enhance scattering contrast in favourable cases, it cannot be performed systematically since it is dependent on available edges and bonding features.

In-situ experiments

"Natural occurrence" for a mineral often means high temperature and high pressure.

To study the behaviour of minerals requires the reproduction of their "natural" environment and thus the need for simultaneous high temperatures and high pressures. "*In-situ*" studies are most appropriate to obtain a thorough knowledge of the relations between thermo-baric variables and structural properties such as phase transitions, cation partitioning, bond valence, electronic structure, etc. Traditionally high pressures have been easier to work with using X-ray diffraction and diamond anvil cells, but the use of time-of-flight neutron techniques has recently allowed considerable progress in high pressure mineralogy. The low attenuation of neutron beams by many materials can effectively make extreme sample environments (HT, HP, Reaction Cells, differential loading frames etc.) easier to handle for neutron scattering than for other experimental techniques.

Examples of frontier applications of neutron diffraction are nowadays mostly in the field of *in-situ* studies where mineral structures are investigated while the sample is kept at high temperature (Figure 1) [2], [3] and/or high pressure (Figure 2) [4].

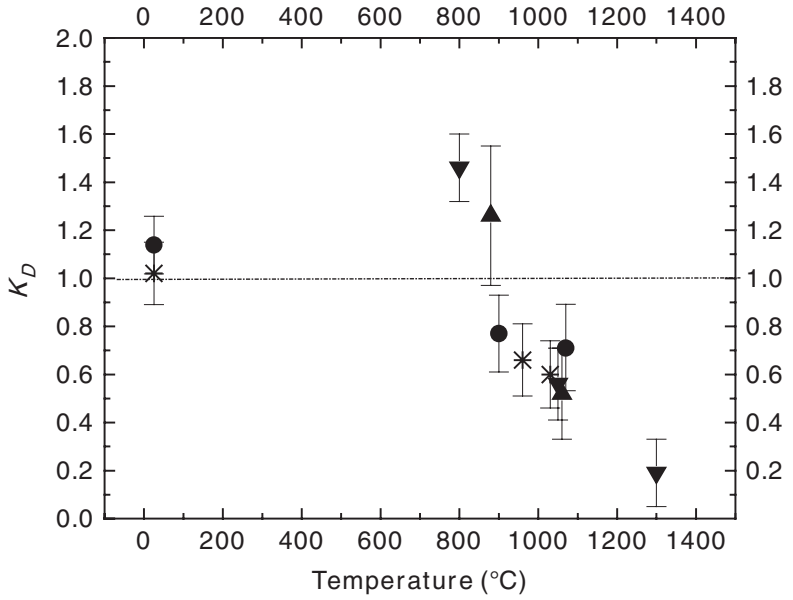

Figure 1: Example of data obtained by high temperature *in-situ* neutron diffraction on single crystals. The plot shows the variation of ordering with temperature between Fe and Mg in Fa12 and Fa10 natural olivines**.** *Up triangles*: Fa12 ISIS-SXD data (880, 1060°C); *stars* Fa12 ISIS-SXD data (25, 960, 1030°C); *down triangles* Fa12 ISIS-SXD data (800, 1050, 1300°C); *filled circles* Fa10 ILL-D10 data (25, 900, 1070°C); *line at* K_D=1 marks total disorder; *points above 1* indicate Fe^{2+} segregation into site M1; *points below 1* indicate Fe^{2+} segregation into M2. The crystals undergo a peculiar, previously undetected, ordering reversal with temperature which is non-quenchable (Rinaldi et al. [2]).

With the ESS, the foreseeable increment in neutron flux (a factor of 30 over ISIS) and detector efficiency are expected to provide a much wider scope for these studies, extending the

capabilities of pressure/temperature devices by a factor of twenty fold at a conservative estimate, hence opening a whole new area of Earth Science studies (Figure 2).

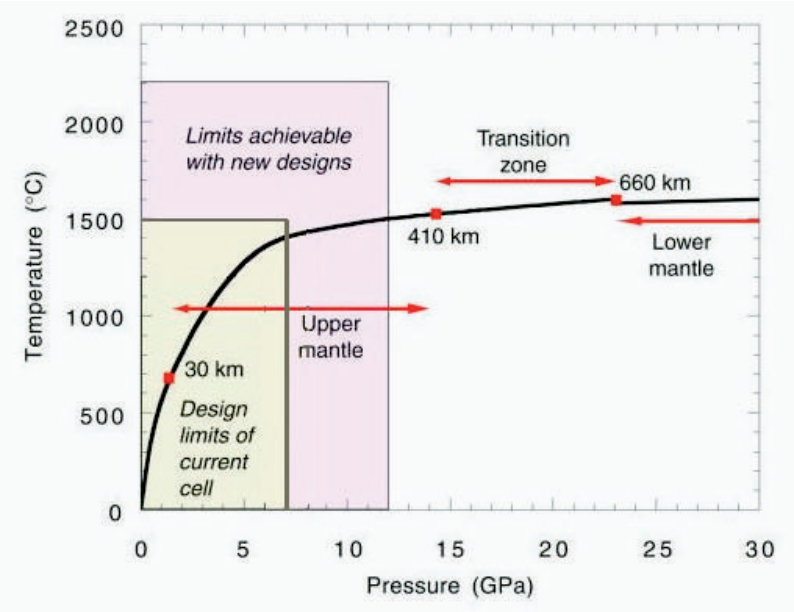

Figure 2: Pressures and Temperatures achievable with present-day and future designs cells usable at Neutron Facilities.

Spectroscopy and modes

Fundamental structural properties of minerals can be investigated by INS.

Neutron scattering is extremely good for studying the dynamical properties of materials. Unlike spectroscopy with electromagnetic radiation, inelastic neutron scattering is not subject to tight selection rules on mode symmetries and wave vectors. For this reason neutron scattering can be used to determine phonon dispersion curves and phonon densities of states, providing both a fundamental understanding and the prediction of mineral behaviour and phase transformations of minerals under the pressures and temperatures of the Earth's interior.

The extension of *"in-situ"* techniques to inelastic and quasi-elastic scattering appears very promising. Such measurements, although requiring highly sophisticated means of data interpretation, offer a unique opportunity for solving details of the dynamics (atomic and protonic dynamics, soft modes, etc.) and allow better modelling and interpretation of fundamental thermodynamic parameters. Limiting factors are mainly associated with the availability of large enough and homogeneous natural single crystals of the phases of interest. Powder inelastic neutron scattering could also provide a viable complementary route, especially when associated with non-ambient techniques.

The limitations in INS studies of minerals are essentially correlated with the lack of large pure samples as both single crystals and powdered pure specimens. The limited scope of the studies carried out so far on minerals reflect this drawback in the use of present-day neutron sources. Here again, an

increment in neutron flux, as expected from an ESS-type source, would serve the purpose of extending INS to smaller purer samples of many more mineral species and phases [5].

Magnetic properties

Neutrons are especially suited for studies of the magnetic properties and magnetic structure of minerals.

Neutron scattering is the best probe of the microscopic ordering of magnetic moments, and can be used to determine magnetic structures, collective magnetic excitations, and crystal field energy levels. The magnetic structures and transitions of Fe minerals present in high pressure environments in the deep Earth is of paramount importance for elucidating their physical properties and behaviour. Although magnetic X-ray scattering can certainly be performed with synchrotron radiation, it is in practice limited to resonant species (i.e. Fe and a few rare earth elements), therefore the use of the ESS neutron source will allow much better measurements, especially under pressure.

Direct Imaging and stroboscopic techniques

In-situ physical properties and dynamics of magma melts; direct imaging of internal fabrics in minerals, rocks and historical artefacts in bulk.

Neutron penetration and the time-structure of a pulsed source can be advantageously exploited for time-resolved neutron absorption measurements to determine the viscosity and density of magma-type melts at high pressure and temperature. Neutron imaging experiments at pressures up to 5-10 GPa and temperatures of 1300-1500°C in a cm scale cell would yield precise in-situ measurements that could also be extended to the study of reaction fronts in silicate crystallisation [6]. Decomposition and exsolution occur in minerals (and rocks) when cooled from the melt. The resulting domain structures (and textures), can be important geothermal indicators. The pulsed nature of a spallation source is ideally suited for following the kinetics of these phenomena *in-situ* by using stroboscopic methods. Such a study, on the spinodal decomposition of the system AgBr-AgCl [7], has been performed, with difficulty, at a steady state reactor source, but would be much easier with the ESS, in particular allowing us to tackle the much more complicated mineral systems. Time scales from *ms* to *h* would be accessible. Examples could include the exsolution in pyroxenes (e.g. pigeonite and augite), the kinetics of cation ordering and the development of (incommensurate) superstructures (e.g. plagioclases). More readily available measurements would be those on the static inner fabric of materials and artefacts, beyond the reach of less penetrating probes, for applications in many fields including archaeology and the preservation of cultural heritage.

Mineral surfaces

Study of protonation reactions responsible for mineral alterations and surface transformations.

The breakdown, weathering and transformation of minerals on the Earth involves the migration of hydrogen through the mineral surface and into the subsurface of the crystals, thus changing the physical properties of the minerals in the surface region. As these reactions occur at the mineral/mineral, mineral/fluid or mineral/biota interface, the study of such

protonation reactions is fundamental to our understanding of weathering and mineral breakdown. At present X-rays are used in reflectivity mode to investigate mineral surfaces but, as previously mentioned, neutrons are far superior to X-rays for the investigation of protons.

Texture – stress – structure analysis

Using the penetrating and resolving power of neutrons to extract the geological history and behaviour of rocks.

Texture, defined as preferred orientation in a crystalline material, carries a fingerprint of the rock's history. The complexity of geological texture analysis results mainly from the overprinting of different textures upon several mineral components from different periods of geological activity. Quantitative texture analyses provide fundamental information for the modelling of rock anisotropies and reconstruction of tectonic events.

The high penetration capability of neutrons and the availability of wide beams allow the investigation of large specimens which produce global volume textures with high grain statistics, even on coarse-grained materials. Using position-sensitive detectors and time-of-flight techniques, texture can be analysed from reflection-rich diffraction patterns of polymineralic rocks containing low symmetry mineral constituents [8].

Residual stress analysis of geological material is crucial because natural effects on rocks are orders of magnitude smaller than in technological materials and drilling gives rise to stress relaxation. Furthermore, transient stresses and strains can be directly observed through in-situ measurements at various pressures and temperatures.

A future prospect at a new high flux neutron source is the performance of simultaneous phase, structure, texture, and stress analyses. A recently developed method of refinement of the orientation distribution function makes use of an iterative procedure (called WIMV) to correct spectra for anisotropy. Cycles of Rietveld- and WIMV-like algorithms ensure the full determination of all the parameters involved in diffraction patterns. Around 1000 patterns are needed at different sample orientations. At least one curved position sensitive detector helps minimise acquisition times. This method already works at neutron centres (ILL for instance), and also using x-rays (http://lpec.univ-lemans.fr/texture/texture.htm). Time of flight neutron diffraction gives the benefit of multiple detectors, as proposed for the HIPPO line at Los Alamos. The ESS would allow us to tackle the more complicated natural systems. Furthermore, increasing the number of detectors around a given experimental set-up would allow us to carry out dynamic studies. We could follow phase transitions versus temperature, or textural transitions with pressure or re-crystallisation processes. How do phases re-crystallise under magnetic, thermal or pressure fields? How does texture develop? What can we learn about such processes from experiments on natural and man-made materials?

Non-destructiveness in bulk

In general, the non-destructive nature of many neutron scattering experiments makes the technique well suited for handling large, undisturbed samples and/or rare and unique objects. These can be natural or man-made and encompass areas as diverse as sediment layers, fossils, meteorites, and historical artefacts. There is a strong need to measure large samples in the Earth Sciences, where grain growth is important. Increasing the volume is the only way to acquire statistical reliability in terms of the number of grains. Grains of the order of one mm^3 are not rare in rocks, and spherical samples of 30 mm diameter are sometimes required.

Non-destructiveness in bulk.

III. Further prospects of advancement in the Earth Sciences (and related fields) with the ESS

Some of the most significant issues in the Earth Sciences are those related to the prediction of earthquakes and volcanic eruptions. The reliability of the relevant models largely depends on knowledge of the physical and chemical properties of the materials involved (oceanic crust, upper mantle, continental crust). First and foremost amongst these properties is the role of water in these materials and in the related magmas.

More examples of frontier applications with an ESS-class neutron source are again in the field of in-situ studies, where mineral structures and material behaviours are investigated while samples are kept at high temperature and/or high pressure.

To draw an effective parallel, one may consider the problem of weather prediction based upon atmospheric models. It is quite evident that the present day prediction of up to five days was not even foreseeable two decades ago. As regards the prediction of earthquakes due to plate subduction, if and when it becomes possible it will be entirely dependent on the accuracy of the models that are currently being developed.

At present we can expect to obtain considerable knowledge in this direction, in part by the use of neutron scattering techniques to study the structure and properties of minerals under mantle conditions. The main obstacle is the comparatively low flux of existing neutron sources.

Given the availability of an ESS-type neutron source, three areas of key research activity may be envisaged which would provide a leap forward in this direction for the Earth Sciences:

Flagship Research Programmes

1. *In-situ measurement of structure-property relations in mineral phases under high pressure and temperature conditions representative of the Earth's interior*.
   This mineral physics project would be of great interest to many fields of research in the areas of mantle rheology, subduction modelling, seismology, tectonophysics, etc.

2. *The study of the structure, reactivity and physical properties of multi-component melts and fluids under pressure and temperature conditions representative of the Earth's interior*.
   This petrology and mineral chemistry project would be of

great interest for magmatology, volcanology (including ancient and present-day volcanic activity), rocks and minerals genetics, and many other related fields.

3. *Texture – stress and structure analysis of polymineralic rocks for the reconstruction of tectonic processes and modelling of rock anisotropies.*
   This project concerns the characterisation and interpretation of the textural and mechanical properties of complex polyphasic materials, and their evolution during geological processes. These processes are often analogous to those occurring during HP/HT industrial processing of materials. This project is of interest for geology, geophysics, petrology, mineralogy and materials science.

Some examples of novel representative experiments

Representative experiments

Within the framework of these widely varying fields of research, a number of representative novel experiments can be proposed. Within the research fields described, four flagship experiments have been selected to show the future experimental possibilities.

Study of the pressure-induced spin dynamics and spin-collapse in ($Fe_XMg_{1-X}O$) and Fe_2SiO_4

High pressure, Fe spin-dynamics in the Earth's lower mantle.

As a first step, this includes the determination of the magnetic and crystal structure under pressure up to 100 GPa (lower Mantle). Even more advanced would be a study of magnons under pressures up to 140 GPa (Mantle-Core boundary). Rationale: The pressure-induced spin collapse in 3d ions has long been proposed as a mechanism for adaptation of simple crystal structures to high pressures. Violations of Hund's rule (spin maximisation) could reduce the 'volume' of an Fe^{2+} ion by around 25 %. However, recent experimental evidence (Mössbauer spectroscopy [9], [10]) has shown that this occurs at around 100 GPa. The experiments did not allow a detailed understanding of the nature of the phase transition. The problem is also very challenging from a theoretical point of view, as the methods currently used (DFT with generalised gradient approximation) only give semi-quantitative results.

Hence, what is required for this experiment is a study in which firstly the crystal and magnetic structures of FeO, ($Fe_XMg_{1-X}O$) and Fe_2SiO_4 are investigated under pressures up to 100 GPa, using a high neutron flux magnetic powder diffractometer. Secondly a study of inelastic magnetic scattering on a single crystal at the same pressures, using a HET like instrument (high energy chopper and cold chopper spectrometers) to obtain the transition energies between spin levels and (most demanding) a constant Q instrument with very high flux to measure magnons at high pressures.

In-situ high P/high T and high P/low T neutron spectroscopic investigation of the molecular dynamics of volatile species (H_2O, OH, CO_2) in minerals and nanoporous compounds through inelastic and total scattering

Molecular dynamics, vibrational spectroscopy, volatiles.

Inner surface molecular interactions require studies at variable T and P (0-1500 K, up to 1 GPa). High temperatures are required for deep Earth's materials, and low temperatures are required for Earth's crust and planetary surface materials. In addition, total scattering at high P/high T could be used to study minerals under geological conditions. Currently available fluxes and resolutions limit "molecular neutron spectroscopy" to energy transfers of about 1500 cm^{-1} and to low temperatures. This prohibits the full utilisation of neutron molecular spectroscopy as a complementary tool to Raman and IR-spectroscopy. The need is for spectroscopic measurements of the molecular dynamics of H_2O in nanoporous, hydrous, and nominally anhydrous (NAM) mineral compounds including studies of surface hydration and reactions.

To understand the molecule-inner surface interaction typically encountered in nanoporous solids, experiments in a heatable/coolable high pressure cell (0-1500°C, up to 1 GPa) are required. Very low temperatures are important to understand the transition into the quantum regime (tunnelling), high pressures are important to be able to tune the strength of the host-guest interaction.

High P/high T and high P/low T structural behaviour in minerals

HP/HT structures, mantle-core phases, water in minerals, phase transitions, gas hydrates.

Current technology has a foreseeable upper pressure limit of about 20 Gpa, achievable with a Paris-Edinburgh type cell (PE) with heating. At present the maximum operating pressure is 7 GPa and simultaneous P/T measurements can be carried out up to 1500°C (Figure 2). An increase of the T limit to 2200°C can be envisaged. Reduction of the sample volume, improvements in pressure cell technology, and the higher neutron flux of ESS would allow access to higher pressures, possibly above the 150 GPa range.

A few pertinent examples are given below

a) In-situ studies of mantle hydrous and nominally anhydrous Mg-silicates (alphabet phases, wadsleyite, spinel) at pressures and temperatures of the transition zone (~ 15 GPa; ~1500°C) would help to characterise the nature of the seismic discontinuity known to occur at a depth of 410 Km, and the water budget of subduction zones [11] [12]. The structural behaviour of protons in hydrous phases at high P/T governs upper mantle melting, volcanic and earthquake activity, although little is known about the effect of pressure and temperature (20 GPa, 1300 K) on the stability or the equilibrium amounts of "water" incorporated in these phases. This is presently impossible since the low H content of such minerals precludes neutron diffraction

experiments with existing sources. A gain of 10 in flux and a further gain of 5 to 10 with improved instruments (diffractometers, detectors, data processing) will make it possible.

b) Cation ordering at high P and T. The thermodynamic consequences of cation order-disorder as a function of P, T and time, must be investigated in order to understand the geophysical and geochemical mechanisms involved. Important phases such as the pyroxenes, olivines and spinels are just beginning to be investigated. A large amount of work is required to cover the chemical and physical variants of the corresponding natural phases and the kinetics of phase transitions encountered.

c) Accurate determination of the structure of pressure-stabilised micro-porous compounds (gas hydrates) and of their physico-chemical properties. The kinetics of their phase transformations are needed to elucidate many aspects of these poorly understood significant components of shallow geological environments in both oceanic and continental sediments (Figure 3). Gas clathrates have been postulated to be of societal relevance in at least three ways: resource, climate, and hazard.

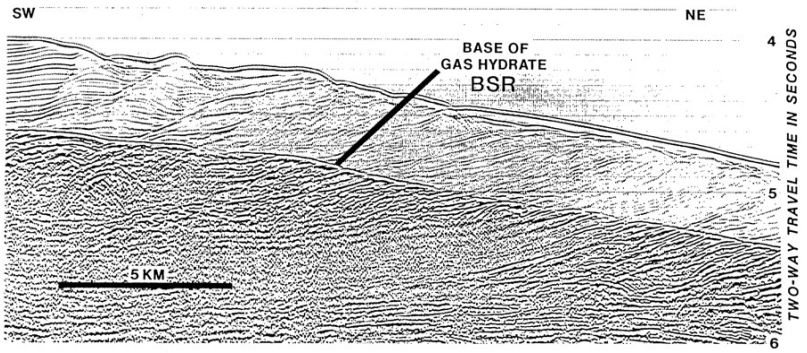

Figure 3: A 12-fold multichannel seismic reflection profile from the crest and eastern flank of the Blake Outer Ridge. The strong BSR (bottom-simulating reflection) is inferred to represent the base of the gas hydrate stability zone. (Kvenvolden, 1998 [13]).

Kvenvolden [13] reports on the immediate importance of submarine geo-hazard aspects in considerations of human activities and installations subject to the instability of deep water oceanic sediments (communications cables, ocean drilling rigs, etc.) which may be affected by slope failures, debris flows, slumps, slides and possible tsunamis and also, perhaps, ancient and historical events of global warming associated with the release of green-house effect gases. The need is for accurate phase diagrams and stability under variable P/T conditions and saline concentrations. Structural studies can also elucidate the mechanisms responsible for the seismic reflectivity attributed to these compounds.

In-situ diffraction and spectroscopic studies of molecular components in methane clathrates

Methane clathrates

This experiment indicates the high level of accuracy required to investigate fine structural and vibrational details in complex compounds.

Gas hydrates have hydrogen bonded rigid cage structures of water molecules entrapping gas molecules. The molecule, for example methane, has a different symmetry point group with respect to the cage symmetry and rapid flipping of the molecule over several configurational states possibly occurs. This is reflected both in the spectroscopically observable vibrational modes and in the long-range disorder of the molecules observable by diffraction, where anharmonic motion may be also observed.

Both experiments need to be of high quality to yield fine structural details, and they must be carried out under low T/high P conditions, although in this case the P range is easily accessible. The experiment requires high resolution single crystal and powder diffraction for the structural part, and resolution-enhanced TOSCA-like molecular spectroscopy for the vibrational part. A 50 Hz, short pulse, 5 MW source is advised.

Time-resolved neutron radiography and tomography of the behaviour of fluids and melts at HT/HP conditions

Absorption, tomography, radiography, rheology, melts, magmas, solutions.

Imaging based on neutron transmission measurements is potentially an excellent tool to study macroscopic changes down to length scales of 5 micrometers with a time resolution of 1/10th of a second. One example is falling sphere experiments at high pressures. Currently, synchrotron-based falling-sphere experiments for the in-situ determination of the viscosity and density of melts is the only way to determine viscosities at very high pressures. These experiments suffer from problems with sample size and constraints due to the high-pressure cell. From our own experience, we know that neutron imaging measurements can be much more accurate, especially as convection can be monitored by doping with highly absorbing compounds, such as Gd-oxide. A set-up which would allow a falling sphere experiment at pressures up to 5-10 Kbars / 1500°C in a cell of about 10 cm height would allow very precise measurements. In addition to a homogeneously illuminated area of about 10 x 10 cm^2, this would also require the development of a position sensitive detector with 5 micrometers resolution (current state of the art is about 250 micrometers). In these dynamic experiments the pulse structure and high peak flux of ESS can be fully exploited by synchronisation with the camera shutter, an advantage over a continuous source.

Neutron tomography is also a promising technique for investigating the internal structure of multi-component systems in a non-invasive manner [14]. Time resolved neutron imaging could also be used study reaction fronts, such as occur during crystallisation of silicates or alloys as well as magma mixing

and mingling properties in order to model natural systems in magma chambers involving the need for high temperature and moderate pressures.

New techniques such as resonance absorption for precise temperature measurements and transmission Bragg edge detection for partially crystalline melts would also have to be further developed to improve characterisation of samples in-situ.

Similar to those regarding silicate melts are many problems involving mineral-water interface reactions still unsolved. Experiments on aqueous solutions in either reflectance mode (surface) or transmission mode (bulk) would yield better models for surface capacitance and cation adsorption energetics, for instance.

Space- and time-resolved absorption measurements of volatile-containing crystallising magmas

Magma rheology and dynamics.

Neutron imaging and tomography are used to study the rheology and the processes (magma mixing, convection, gas segregation, reaction fronts, crystal growth, etc.) occurring during the high-temperature crystallisation of silicate magmas. Measuring the P, T, X dependence of the viscosity and density of silicate melts would provide crucially important data for petrology and, particularly volcanology issues. Use of sealed vessels would allow gas fugacities (H_2O, CO_2, F, etc.) to be controlled. Degassing experiments (e.g. speed with which bubbles grow during vesiculation) should also be possible. Other mineral physics/chemistry questions to be addressed would be, for example, mechanisms of nucleation and growth, phase competition during growth as a function of melt composition, inhibition of crystal growth by specific elements or molecules, etc.

A parallel very intense beam is needed on an area of 10 x 10 cm^2 for recording the image of the autoclave vessel. Present space resolution, of the order of 1 mm, should be improved by one order of magnitude. New high resolution position sensitive detectors are needed to resolve fine details of the evolving system. With the logarithmic dependency of the viscosity on temperature one can fine-tune the time-scale of the experiment by selecting the appropriate temperature. A 50 Hz, short pulse, 5 MW source is necessary for time-resolved measurements.

Strain partitioning upon deformation of polymineralic rocks

In-situ measurements of stress and strain partitioning in rock deformation.

One of the major aims in the Earth Sciences is to refine our understanding of the structure and composition of the Earth's interior using seismological data. The interpretation of this data is heavily reliant upon laboratory measurements of the elastic properties of the relevant rock types [15]. These rock-types are essentially polymineralic aggregates with a range in composition and microstructure that is far too large for it to be

feasible to determine the properties of every rock-type individually.

Consequently, it makes sense to seek a method that will permit the elastic properties of polymineralic materials to be specified in terms of the elastic properties of the component phases. In order to formulate mechanical equations of state for polymineralic materials that are based on some function of the properties of the component phases, it is necessary to establish the relative contribution of each phase to the aggregate's properties. However, in rock deformation experiments it is usually only aggregate properties that can be measured. There is a glaring dearth of experimental evidence as to what happens to the strain partitioning between the component phases in a composite in the elastic regime, and also when one or more of the phases starts to yield plastically. Once yielding occurs there is the possibility of load transfer between the phases, and the extent to which this occurs exerts a profound influence on aggregate properties (Figure 4).

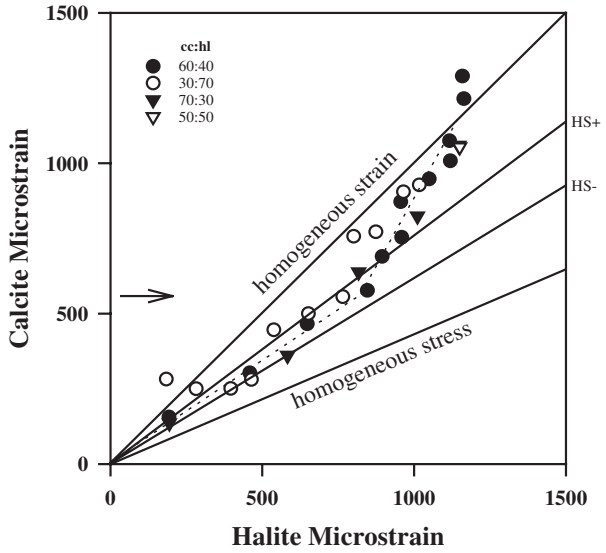

Figure 4: Calcite and halite axial elastic strains at different applied loads [16]. Also shown are the predicted phase strains assuming homogeneous stress, homogeneous strain, and as given by the upper (HS+) and lower (HS-) Hashin-Shtrikman bounds on the composite properties. The dashed line shows the trend in the data (calcite ≥ 50%) and the arrow shows the calcite strain at the elastic limit of calcite.
The elastic limit of the halite was at a halite strain of about 350μstrain. The elastic strain partitioning between the two phases was unaffected by the yielding of the halite. However, above a calcite elastic strain of about 550μstrain, the strain partitioning between the two phases started to tend towards homogeneous elastic strain, a condition that was attained by a total (elastic+plastic) strain of about 1%. The curve describing the elastic strain partitioning between the calcite and halite was independent of composition. The change in elastic strain partitioning at a calcite strain of 550μstrain corresponds to the point at which the elastic limit of calcite is attained.

Neutron diffraction experiments at spallation sources, conducted on samples held under differential load in the

neutron beam line, offer a solution to these two problems. By determining the change in lattice parameters of each component phase as a function of applied load, the elastic strain of each phase, and hence its contribution to the total deformation, may be ascertained [17]. These experiments are only feasible due to the penetrating and polychromatic nature of the neutrons produced at spallation sources. Future developments foreseeable at the ESS would be an improved strain resolution from lower counting times, the development of high temperature equipment, and smaller beam sizes enabling greater spatial resolution. Such developments could be a large step beyond that available at ENGIN-X at ISIS.

Deformation mechanisms in polymineralic rocks

Comparative texture studies of complex systems, time evolution of rock texture.

Due to the high penetration capability of the neutrons, volume texture investigations can be performed on rather large natural specimens up to about 10 cm^3. Such volumes are necessary to ensure sufficient grain statistics for all mineral constituents in polymineralic rocks, especially with coarse grain sizes (e.g. about 1 mm) [18].

Large sets of geological samples from different locations must be investigated in order to study mineral specific textural changes under different deformation conditions [19], or for different minerals under similar deformation conditions in order to understand the underlying deformation mechanisms [20]. A high intensity source and a large beam are needed to perform textural investigations on large samples sets within a reasonable time scale. Time-of-flight techniques are essential because pole figure measurements can be performed without any sample scanning, and because simultaneous structure and texture refinements become possible.

Influence of stress and development of texture upon deforming geomaterials

Stress and texture

Large scale deformation of crustal and mantle materials generates the development of microstructures, involving twinning, phase transitions, and mineral structural and textural transformations. To explore the influence of differential stress and strain partitioning on plastically deforming polymineralic materials, and the development of textures, requires HT/HP conditions to simulate geological processes.

Texture analysis without sample rotation requires large banks of detectors (HIPPO-type instrument) and accurate detection of lattice variations in low-symmetry materials requires high-resolution diffractometry (resolution-enhanced ENGIN-X instrument, with ample sample space). Preferably, in certain cases the two experiments should be performed simultaneously, in order to follow the complete evolution of the sample. A 50 Hz, short pulse, 5 MW source is most suitable.

IV. Cultural heritage materials

Archaeological artefacts, conservation, non-destructive analysis.

Phase, microstructure and texture analysis of natural materials such as stone, as well as ceramics and metal objects, is still relatively new but the potential applications of such powerful techniques span many fields of interest within archaeological research, from standard fingerprinting to complex conservation problems. Diffraction techniques are important for helping to date excavation sites, to establish trading patterns, to determine cultural exchange between regions, to elucidate historic and regional abundance of traded goods and to help identify the original source of raw materials. Phase and microstructure characterisation of ancient objects by diffraction methods can hint to manufacturing techniques. Diffraction studies may address the phases of the source materials or alteration and corrosion phases produced by exposure (e.g. patina, black crusts, etc.).

Owing to the non-destructive character of neutron scattering techniques, and to the large interaction volume, the applicability to relatively large, intact and potentially precious archaeological objects, is easily predicted. There will be many new applications in the fields of study and conservation of historical artefacts. An example of intervention guided by such studies is given in Figure 5.

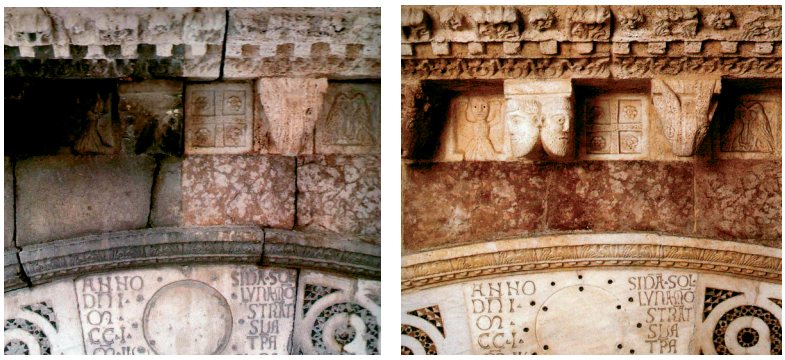

Figure 5: Foligno Cathedral; limestones, marbles and travertine – before and after restoration (Courtesy of B. Moroni and G. Poli, Perugia).

TOF neutron diffraction can provide complementary information to X-ray diffraction, especially when non-destructiveness is an important issue, e.g. if objects must not be damaged by cutting, drilling, scraping etc. No preparation of objects is needed as data can be collected from large and intact objects of almost any shape. The experimental set-up is simple and free of sample movement [21].

Correlations between phases, or ratios of phase fractions, may be used to characterise or classify an artefact (Figure 6). During firing of ceramics the starting materials undergo solid state reactions depending on the firing temperature, duration and atmosphere [22]. Ancient or pre-historic ceramics fired at moderate temperatures often exhibit very complex diffraction patterns due to a wide variety of mineral phases, among them clay minerals and sheet silicates which need high intensity and resolution for identification and quantification.

Materials may change their microstructure due to mechanical treatments utilised in the production of artefacts. Hence diffraction 'fingerprinting' may also provide valuable information about the manufacturing processes. Texture is a case study of its own, and may for example be an important characteristic of a mechanically treated archaeological metallic object. The potential of neutron diffraction is further enhanced by the ability to investigate the phase abundance, texture or grain distribution of metal objects, for example bronze objects and coins [23]. Neutron diffraction is particularly powerful for sandwich situations, e.g. metal sheet stacking, coins with coatings or objects locked inside containers. Another aspect is to determine whether a coin is authentic or a fake, as well as to distinguish between differently struck coins.

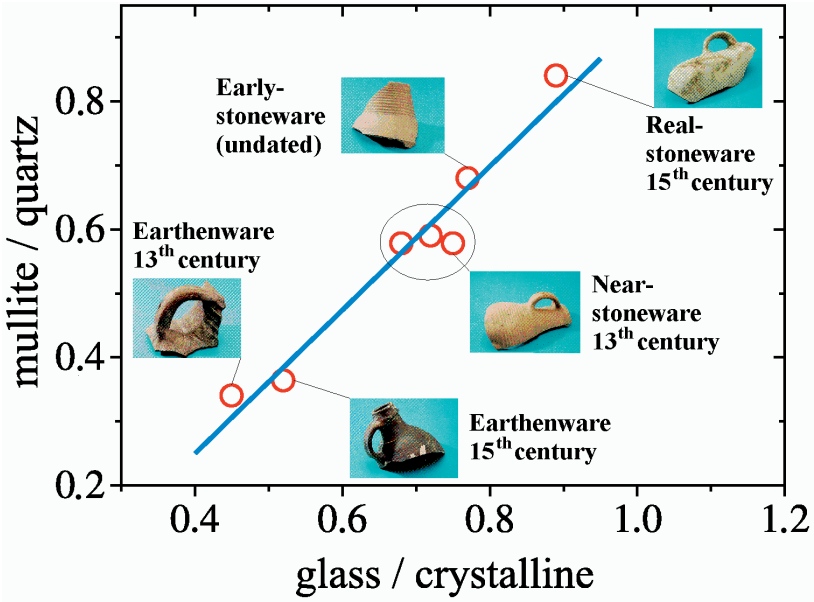

Figure 6: Classification of Medieval German pottery from Siegburg using ratios of phase fractions mullite/quartz and glass/crystalline that were obtained non-destructively by TOF neutron diffraction.

In the non-diffractive mode, information on the inner fabric of large scale materials and artefacts (a few μm to several cm), which is beyond the reach of X-rays, can be obtained by making use of recently developed neutron detectors which lend themselves to *neutron imaging* and *tomographic* reconstruction. Applications of this technique to archaeological artefacts are already envisaged; the availability of improved instrumentation, especially in terms of detector capabilities, would definitely represent a major improvement for this area of research.

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Achievements of neutrons in mineral, Earth and environmental sciences

- Accurate and precise crystal structure determinations of minerals (including hydrogen bond properties) under ambient conditions and also under extreme conditions by the simultaneous application of elevated pressure and temperature (in-situ). Measurement of accurate equation-of-state data, phase stability and crystal structure behaviour under geologically relevant conditions to help our understanding of global dynamics, from catastrophic events to phenomena that span 10's of millions of years. Recent examples of geologically relevant minerals include: brucite, micas, olivines and spinels. Very recent new achievements in this field have been obtained thanks to the newly developed PEARL instrument and Paris-Edinburgh cell at ISIS, which provide excellent diffraction data at simultaneous pressures and temperatures of 7 GPa and 1700 K.

- Cation ordering of isoelectronic and quasi-isoelectronic species in crustal and mantle silicates and oxides by dynamical in-situ studies of order/disorder processes and transitions, often unveiling the non-quenchable nature of such phenomena. These experiments provide thermodynamic and kinetic data required for modelling mineral behaviour, mineral reactions and geospeedometry. The most significant examples are olivines, spinels, cristobalite and leucite.

- Low- and high-temperature hydration/dehydration processes in hydrous and nominally anhydrous minerals studied by neutron diffraction has provided the mineral structural basis for an understanding of geo-dynamic phenomena. These reversible transitions impact significantly upon a range of Earth science issues including the role of phosphates, zeolites and clay minerals in the remediation of contaminated land, the role of sulphates and clay minerals in crustal failure, and the behaviour of dense hydrous Mg-silicate phases and nominally anhydrous minerals in mantle/subduction zone derived earthquakes.

- Neutron spectroscopy has been pivotal in the evaluation of vibrational properties near and at phase transitions, measurements of soft phonon modes and the dynamics of hydrous components in minerals. The measurements of phonon dispersion curves to investigate interatomic potentials in geologically relevant phases (quartz, forsterite, enstatite, calcite, leucite, pyrope, etc.) have resolved issues associated with structural instabilities. Measurements of phonon density of states of framework minerals has enhanced thermodynamical modelling of phase transitions (i.e. cristobalite).

- Total neutron scattering measurements have for the first time been used to account for long-range crystallographic order providing a complete description of the temperature-dependent behaviour of framework silicates. Results for quartz show that a classical soft-mode treatment of its properties is too simplistic and that the structural behaviour is best described in terms of its ability to respond to low-frequency, high-amplitude vibrational modes.

- Strain/stress measurements have been performed in-situ at elevated pressures and temperatures on natural rocks (e.g. sandstone, quartzite and marble) with strains of the order of 10^{-4} using the EPSILON instrument at Dubna. The partitioning of elastic strains between the component minerals in polymineralic rocks has been determined using ENGIN at ISIS providing significant insights into the controls and influences of rock composition and microstructure on the properties of the Earth's crust and mantle.

- In-situ observation of the evolution of both thermal and residual strains from within the center of large solid samples of natural rocks has recently been performed, providing a link between the thermal properties of rocks to the thermal response of the component minerals. Thermal cracking commences at 180ºC when the thermal strain deficit along the

a-axes of quartz grains induces a thermal stress that is close to the bulk tensile strength of the rock.

- Texture analysis of bulk, single-phase mineral samples such as pyrite, chalcopyrite, quartz, hematite and calcite, and very recently, on polymineralic rocks such as gneiss mylonite, granulite and eclogite. These types of investigations yield information on the overall kinetics of rock deformation relating to large-scale geological processes.

- Non-destructive phase identification and quantitative phase analysis of archaeological objects (i.e. Bronze age ceramics, Attic ceramics, Etruscan bronzes, Medieval German stoneware) has been obtained using time-of-flight diffraction at ROTAX, ISIS.

- Small and wide angle neutron scattering experiments probing the structure of silicate and aluminosilicate glasses have provided information regarding the structure and dynamics of extrusive magmas and the dynamics of intrusive and mantle magmas. Additionally, the structural environment around toxic and radioactive metals is now generating accurate models and realistic materials designed for the long term containment of harmful metals.

- Neutron diffraction has recently been applied to the interaction of fluids with minerals that are associated with cements, oil and construction industries, waste containment and environmental remediation. Fluid interactions with vermiculites and montmorillonites, pillared clays, and zeolites are being studied in-situ under pressures and temperatures relevant to hydrostatic geological conditions (equating to burial depths of 10 km) and with chemistries relevant to environmental applications such as barrier materials and nuclear waste containment.

4.8 Fundamental Neutron Physics

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Abstract
Neutrons appear both as composite particles and as quantum waves. Both features have been investigated with thermal, cold and ultracold neutrons at many neutron sources. The higher intensity and the pulse structure of the ESS provide new possibilities for fundamental neutron physics experiments. The questions of small right-handed contributions to our left-handed world, why is there much more matter than antimatter in the universe and how neutrons interact with each other can be tackled. Non-classical neutron states can be produced and used for novel fundamental quantum optics investigations. Intensity gains of ultracold neutrons of the order of 1000 can be anticipated at a new ultracold neutron target station where the use of a spallation process for the production of neutrons becomes especially obvious. The decision of the ESS Council to install two target stations, one short pulse and one long pulse station, permits new types of crossed beam experiments. This also provides the possibility of crossed beam experiments in connection with the proposed UCN station just located at this crossing area.

I. Introduction

Neutrons are known as a powerful tool for particle and nuclear physics and they are ideal probes for quantum optics investigations. The European Spallation Source is, therefore, of intense interest for fundamental studies in these fields.

Investigations of the neutron's properties give information about the elementary forces of nature - strong, weak, electromagnetic and gravitational.

During the past 25 years, our world-view of nature has changed dramatically, on aspects ranging from the constituents of elementary particles to the status of the universe. Neutron physics has made major contributions to this evolutionary process of understanding. On the grand scale, cosmology has evolved into an exact science and neutron physics has contributed to the understanding of element formation and of phase transitions in the history of our universe. Various data extracted from measurements of neutron beta decay have been used extensively to fix the number of particle families at three. On the very small scale, neutron experiments have made substantial contributions to our understanding of strong, electroweak and gravitational interactions. Neutron interferometry and neutron spin-echo experiments have shown how non-classical states of neutrons can be created and used for highly sensitive investigations in condensed matter and fundamental physics research.

Quantum optics with neutrons opens new fields.

Many crucial questions remain to be answered and the increased flux from ESS will enable major progress in a range of areas. For example, unique experiments can be performed which will help (a) to determine the basic structure of the fundamental interactions acting in nature, (b) to elucidate the history of the universe and to predict its future, and (c) to study fundamental questions of quantum and measurement theory. The community in this field comprises about 300 scientists, with many young people starting new work.

II. Flagship experiments

The following generic experiments will become feasible at ESS. They use ultra-cold, cold and hot neutrons from the source. The results of these experiments are intended to raise the highest scientific interest, and they can be published in journals with the highest impact factor, but they are also rather risky.

The question of the origin of handedness of nature

The exotic decay of the neutron into a hydrogen atom and an antineutrino can help us to find phenomena beyond the Standard Model.

In nuclear decay experiments it was recognised in the late 1950's that one of the four fundamental forces - the weak force - is, as far as we have been able to discern so far, exclusively left-handed. Most Grand Unified Theories, however, start with a left-right symmetric universe, and explain the evident left-handedness of nature through a spontaneous symmetry breaking caused by a phase transition of the vacuum. This scenario, if true, would mean that today neutrinos should carry a small right-handed component. Although limits on the right-handed currents have been derived from free neutron and muon decay experiments, what is really needed is a clear-cut "yes" or "no" experiment. Such an experiment, planned for ESS, is the two-body β-decay of unpolarised neutrons into hydrogen atoms and antineutrinos which occurs with a relative probability of $4.2 \cdot 10^{-6}$ compared to the usual β-decay.

Usual decay mode: $n \rightarrow p + e^- + \bar{\nu}_e$

exotic decay mode: $n \rightarrow H + \bar{\nu}_e$

What is so interesting about this decay is that one of the four hydrogen hyperfine states cannot be populated at all if the neutrinos are completely left-handed. A non-zero population of this substate would, therefore, be a direct measure of a right-handed component.

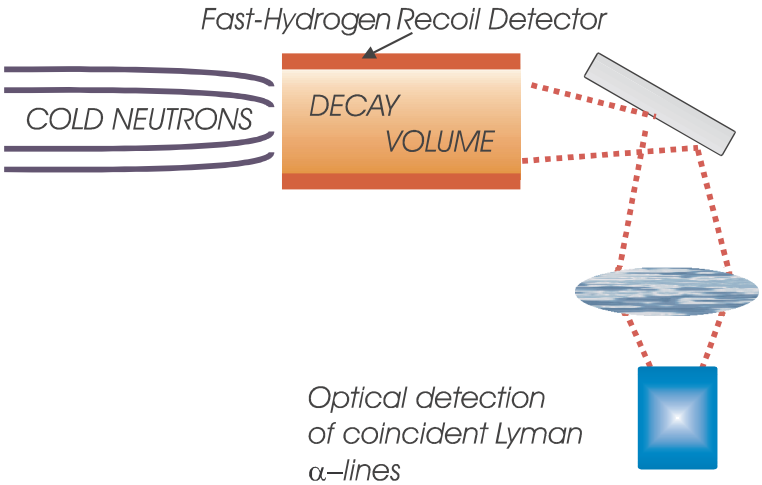


Figure 1: Scheme for the measurement of the neutron decay into a hydrogen atom.

This experiment has severe background suppression requirements for which the pulsed structure of ESS, allied with its intensity, is well suited. Thus, with ESS it may be possible to prove for the first time that nature does not possess an intrinsic handedness and that there is exciting new physics beyond today's Standard Model of particle physics.

In a second stage the experiment has to be done with polarised neutrons where the transition probabilities between the hyperfine levels can be changed drastically.

An intense pulsed cold neutron beam is required for these experiments, which are not possible with current neutron sources.

The origin of the baryon asymmetry of the universe

The big bang theory presumes that equal amounts of matter and antimatter were created in the primordial explosion. In the subsequent process of annihillation of matter and antimatter only very few heavy particles ("baryons") and an equal number of antiparticles from this early period could survive. Our mere existence contradicts this expectation; there remained about 10^8 times more baryons in the universe than predicted and almost no antibaryons have survived. So far, the only viable solution of this problem is the violation of charge-parity symmetry (CP) which, on all reasonable expectations, is equivalent to a violation of time symmetry (T) that could have led to a small excess of particles before the annihilation stage.

Further measurements of the electric dipole moment of the neutron can help our understanding of matter-antimatter asymmetry in the universe.

Violation of the CP-symmetry has been observed in the decay of kaons. However, this single positive result is not sufficient to verify the above conjecture, nor to identify the origin of CP- or T-violation. Grand Unified Theories (GUT) require T-violating amplitudes that are orders of magnitude larger than can be accommodated by the present Standard Model. Therefore, another generation of experiments is needed to obtain decisive answers.

The most direct access to these questions lies in the detailed investigation of neutron decay and in measurements of its electric dipole moment. Electric dipole moment measurements started in the fifties and increased their sensitivity by one order of magnitude every seven years. They are based on searches for a deviation equal to $\pm d \cdot E$ from the well-known angular frequency of

$$\hbar\omega = 2|\boldsymbol{\mu}|\mathbf{B} \pm \mathbf{d}\cdot\mathbf{E}$$

of a neutron spin in a magnetic field B and a parallel or antiparallel electric field E. Current theories of the baryon asymmetry of the universe are related to an EDM of about 10^{-28} e cm, a limit that is accessible with the ESS. The current upper limit is $6 \cdot 10^{-26}$ e cm.

These experiments are most effectively done with ultra-cold

neutrons (UCN) where recent developments on new UCN sources predict orders of magnitude intensity gains. The possible arrangement of such a UCN station between the two proposed ESS target stations is shown in Figure 2. The UCN station would take the full power (10 MW) of the linac for about 1 s every 10 minutes, i.e. less than 1 % of the beam power.

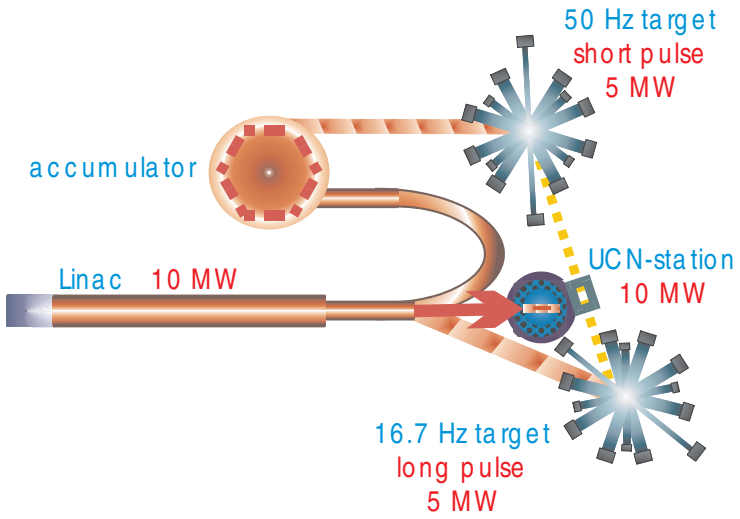


Figure 2: General layout of ESS with a dedicated UCN target station (Serebrov system; not in scale - the UCN target station is an ESS option, but not integral part of the project).

A separate UCN station served by 1 % of the proton beam power and the long pulse option would give new perspectives for research with ultracold neutrons.

The question of charge independence of nuclear forces

The strong or nuclear force is governed by the fundamental quark-quark interaction described by Quantum Chromo-Dynamics (QCD). It is believed that the strong nuclear force is essentially the same for protons and neutrons or, more generally, for up and down quarks. In this respect, the nuclear part of the singlet scattering length should be the same for the proton-proton and the neutron-neutron systems, and it should be similar to the neutron-proton interaction. The neutron-proton scattering length is the only precisely known quantity whereas the nuclear part in the proton-proton system is masked by the Coulomb interaction and the neutron-neutron scattering length has only been extracted indirectly from several three-body interaction processes. The best way to check, whether the deviations in the singlet scattering lengths extracted from these experiments really signal a breakdown of isospin invariance, is a direct scattering measurement of the neutron-neutron scattering at very low energy.

$$n + n \rightarrow n + n$$

$$a^s_{np} = a^s_{pp} = a^s_{nn} \; ?$$

$$a^t_{nn} \equiv 0 \; ?$$

With a pulsed ESS for the first time a direct neutron-neutron scattering experiment becomes feasible.

In a second stage a dedicated experiment using polarised neutron beams could subject the hypothesis of the flavour independence of the quark gluon interaction to a precision test at the baryon level.

As the interaction rate for a neutron-neutron scattering experiment scales with the square of the neutron flux density, the high peak intensity of ESS has huge advantages. The planned ESS-experiments make use (a) of the time structure, by allowing the fast neutrons of one pulse to hit the slower ones of a preceding pulse and (b) coincidences in time and space for the counts for each scattered pair of neutrons.

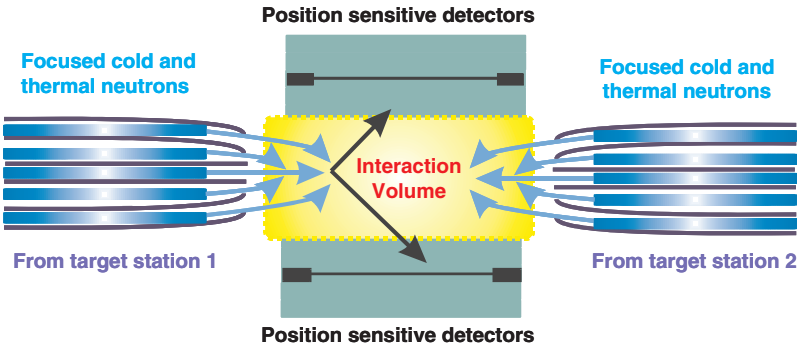


Figure 3: Sketch of the proposed neutron-neutron scattering experiments.

Well focused dense cold neutron beams from both target stations interacting at the crossing area would be optimal for such experiments. The interaction volume should be also accessible from UCN's from the UCN station.

Neutron quantum optics

The phase of a neutron wave has become a measurable quantity since the invention of neutron interferometry. Basic tests of quantum mechanics have been performed in the past and it has been shown how neutrons can be used as a powerful tool in quantum optics. Non-classical neutron states, which are extremely fragile against any dissipation, have been created in neutron interferometry and neutron spin-echo experiments. Major interest concerns the verification of topological phases which are determined by the geometrical form rather than by the strength of the interaction. A complete quantum state reconstruction will become feasible by a simultaneous coherence function and momentum post-selection measurement procedure.

Non-classical states of neutrons can be produced and used in neutron interferometry and neutron spin-echo systems.

The coupling of the neutron magnetic moment to oscillating magnetic fields permits multi-photon exchange and dressed neutron states, while the quantisation of neutron states inside microscopic structures facilitates new possibilities in basic and advanced materials research. Pulsed beams can be trapped between perfect crystal plates forming narrow band neutron resonators that can be developed further as neutron accumulator systems. Inside travelling magnetic fields an

Neutron resonators, accumulators, and new bunching systems become feasible.

advanced method of beam tailoring becomes feasible permitting intensity gains by another factor of ten. These new possibilities have to be exploited as a step towards advanced quantum optical devices serving as resonators and phase space transformers and compressors.

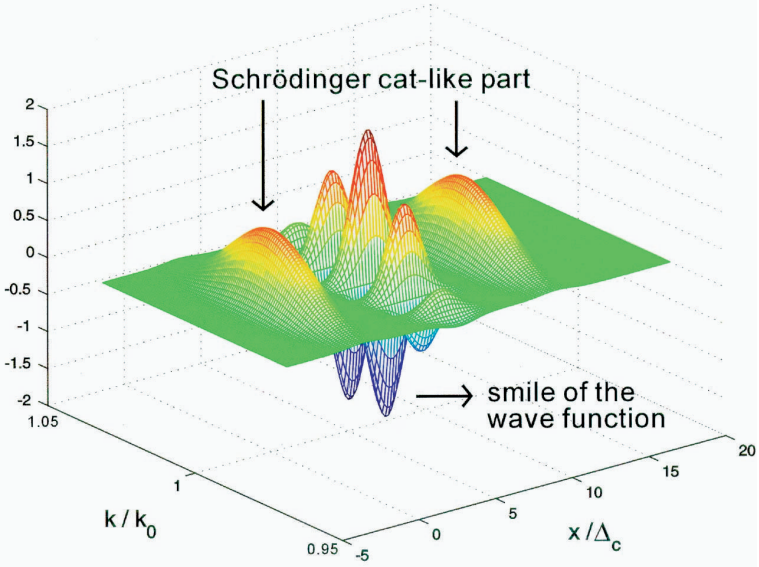


Figure 4: Wigner representation of a non-classical neutron state as it exists in neutron interferometry and neutron spin-echo arrangements.

The long pulse option of ESS can surpass the existing possibilities. A cold neutron beam line adaptable for travelling magnetic fields, and in a vibration free and thermally isolated and controlled experimental area, would be desirable for these experiments.

III. Various other scientific achievements anticipated at ESS

So far the flagship experiments for ESS have been discussed. There is a rich variety of other topics in the field of fundamental neutron investigations of which we mention only a few.

Neutron decay experiments, in particular measurements of the neutron lifetime and angular correlation coefficients, determine certain free parameters of the Standard Model complementary to high energy physics research. The V_{ud} parameter of the quark mixing matrix for the d-u transition in neutron decay plays a key role in testing the unitarity of this matrix, which yields information on possible physics beyond the Standard Model. The experiments determine the strength and structure of the weak quark current and provide the possibility of observing new processes generated by scalar and tensor components, with or without T violating terms or right handed currents.

Research on fundamental phenomena is rather popular among young students and researchers.

Today all weak semileptonic phenomena with significance for

cosmology, astrophysics and particle physics must be calculated from neutron decay data. Certain neutron decay experiments can make use of the pulse structure of ESS for background suppression.

Another topic of high interest is the investigation of the weak interaction between nucleons. This may be carried out by means of coherent spin rotation of transversely polarised neutrons or by differential absorption of a longitudinally polarised neutron beam interacting with unpolarised nuclei of hydrogen or helium.

- The proposed ultracold neutron factory will in addition host two long-term projects: the search for an electric dipole moment of the neutron and measurements on free neutron decay. Ultra-cold and very cold neutrons will be used for elastic and inelastic surface reflections and as probes for nano-structured materials. Quantum gravitational states have been measured and weak gravity effects become accessible. New bunching, cooling and trapping systems will be developed.

- Neutron quantum optical experiments will become feasible where the time structure of the beam can be used to produce a steady beam with an intensity governed by the peak flux of ESS. Topological phenomena could be tackled in a new way. The transition from a quantum to a mixed state could be studied in detail contributing to our understanding of a quantum measurement. Quantum Zeno-effect experiments will show how a quantum state can be frozen when a continuous measurement is performed.

IV. Issue of target station and beam lines

For Fundamental Physics with ultracold neutrons the following additional target is needed:

The proposed ultracold neutron target station has unique features and opens new fields of research.

(a) UCN-station accepting the whole beam power for about 1 % of the time (1 second on, 10 minutes off). It should be located near to the crossing area of beam from both target stations.

For Fundamental Physics with neutrons the following beam lines and experimental areas are needed:

(b) Beam lines for producing a high density neutron gas from the cold moderators of the $16^2/_3$ Hz and 50 Hz target stations.
(c) A beam line for neutron optics at a thermal guide associated with an experimental area with special environmental conditions (vibration-free, air-conditioned, humidity-controlled etc.).

Achievements of neutrons in fundamental physics

- Neutron decay experiments with cold and ultra-cold neutrons contributed considerably to the understanding of weak interaction and to the unification of weak interaction and electromagnetic interaction to the so-called “electro-weak interaction”. All five decay parameters have been measured and the number of lepton families has been fixed to three. Measurements concerning the decay of polarised neutrons demonstrated a complete parity non-conservation in these decay processes which indicates a complete left-handed matter world.

- Neutron fission experiments gave accurate numbers for the fission yields of various fission materials and showed the existence of ternary fission. In this connection useful contributions to the understanding of nuclear transmutation effects have been delivered.

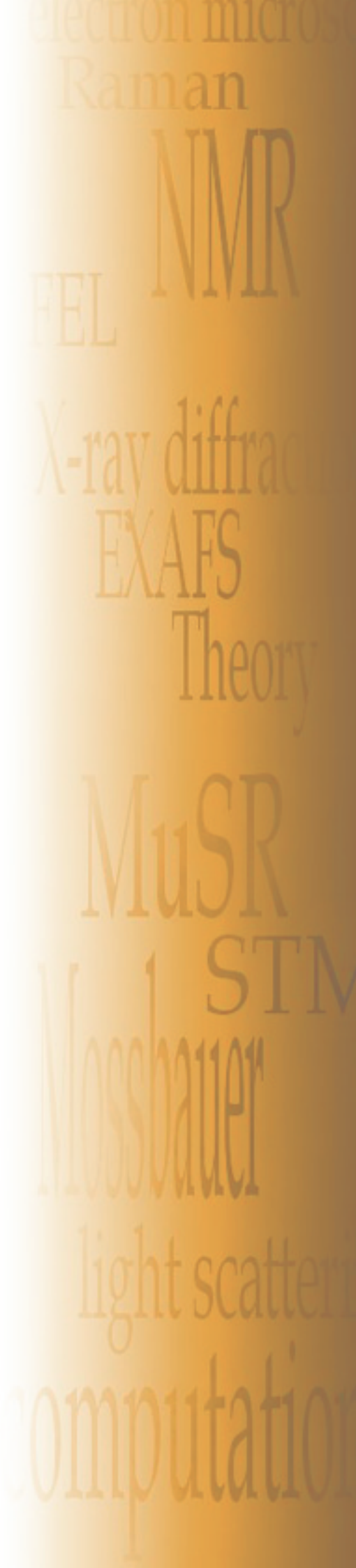
- The search for an electric dipol moment of the neutron yielded new limits for the existence of physical laws beyond the Standard Model of Particle Physics. Although the existing limit is very small (about 10^{-25} e.cm) this kind of measurements provide a possible access to “New Physics”.

- Neutron interferometry provides widely separated coherent beams and permitted the realisation of many quantum physics “Gedanken”-experiments. The 4π-symmetry of spinor wave functions, the spin-superposition law, the magnetic Josephson-effect and the coupling to the Earth gravitational field has been tested on a macroscopic scale. Several Aharonov-Bohm phases and various geometrical (topological) phases have been measured for the first time.

- The observation of the quantisation of neutron states within the Earth gravitational field and of thermal neutrons due to confinement effects opened the field of peV spectroscopy.

Chapter 5

Neutron Scattering and Complementary Experimental Techniques



5. Neutron Scattering and Complementary Experimental Techniques

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Abstract
The neutron will always be an indispensable tool for studying atomic structure and dynamics in condensed matter because of its particular unique properties. However the value of neutron data can be considerably enhanced by the use of complementary data obtained with other methods, and similarly data obtained with other methods are enhanced by the use of neutron data. There is no single experimental technique that can provide us with all the information we need to know about materials. Different techniques, based on different physical processes, provide different information, and as the materials under study become ever more complex, it becomes more crucial to study them using multiple, complementary experimental techniques.

I. Introduction

Modern society places tremendous demands on scientific research. For example, we should develop new materials that are lighter, stronger, smaller, cheaper and environmentally friendlier. We should also understand the reasons for these improved properties on a basic (microscopic) scientific level, both as part of the natural human drive for knowledge and in order to design even better materials in the future. The solutions to such problems become increasingly complex and the demand on the techniques used to study them becomes increasingly great. The approach needs to become 'problem based' rather than 'technique based', so the trend is towards the use of multiple complementary experimental techniques, each of which gives us a small piece of the puzzle. Advanced techniques of computer simulation and modelling can be used to help put together the pieces.

Future scientific approaches will be 'problem based' and not 'technique based'.

Figure 1: Different experimental techniques provide different views of the same 'reality'- partial, transformed, distorted – most often all three.

Developments in experimental techniques over the past decades have been amazing, and will continue to be so in coming decades. X-ray tubes have given way to 3rd generation synchrotrons with many orders of magnitude greater brilliance, and this trend will continue with the x-ray free electron laser (XFEL). Optical lasers can now be used to investigate matter on the time scales of chemical reactions. Developments in neutron scattering have been less dramatic in terms of such 'power numbers', yet an ever greater number of scientists

Experimental techniques continue to develop rapidly. Each provides a part of the 'puzzle'.

uses neutron scattering to study an ever wider range of scientific problems. Why? Because on the one hand neutrons offer unique characteristics as an experimental probe, and on the other hand complementarity means that advances in the use of one probe, e.g. x-rays, produce an increased demand for information that can only be provided by another probe, e.g. neutrons. The combination of information means that the result is very much greater than the sum of the individual parts. There are also cases where more than one technique can provide the required information. Other criteria are then used, such as which is fastest, cheapest, or most accessible. Neutron scattering is not usually then the winner! However it should be stressed again that different techniques, because they are based on different physical processes, in general provide different, and complementary, information.

Competition is mainly financial or social, not scientific.

There is indeed competition between techniques, but it is financial or social rather than truly scientific. The financial competition is clear, particularly for large international facilities. Governments and funding agencies would obviously prefer a simple solution, where a single 'super machine' can provide all the required answers. This can lead to 'over optimistic' claims about the possible future performance of some techniques. Such claims are unfortunate, since in the long run they will benefit neither the scientific community at large, nor the individual community making them.

Neutron scatterers are 'naturally mobile' and can take advantage of complementarity.

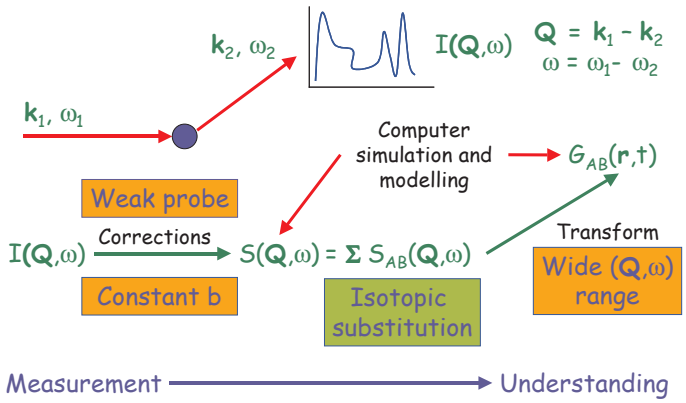
Competition at some level is always valuable, since it provides a natural social drive for progress. However there is also a social tendency for researchers to use the technique with which they are familiar, or which happens to be available 'locally', rather than to use the technique that is optimum from a purely scientific perspective. It can be noted that neutron scattering differs from most other techniques in that it can only be carried out at large national or international facilities, there being no small scale 'home laboratory' version. Those already using neutron scattering therefore tend to be enthusiastic about using additional, complementary techniques, wherever they may be found.

In this chapter we will attempt to give a balanced view of the type of information that is available from neutron scattering and how it complements, and is complemented by, other techniques. There are obviously a large number of techniques that could be discussed, and an enormous number of combinations, so it is impossible to be complete. We have chosen a 'mixed' approach. Firstly we describe briefly the basic idea of a neutron scattering experiment, and the specific advantages of the neutron as a probe. Then there are a number of short sections related to specific (groups of) techniques: NMR, FEL/XFEL, μSR, microscopy and computing. Finally we give an overview of complementarity in a particular subject area. We have chosen life sciences, since it is a small field within neutron scattering at present, but one where major advances are expected at the ESS. Interspersed with these sections are a number of specific examples that illustrate the power of complementarity.

II. Advantages (and disadvantages) of neutron scattering

In an ideal scattering experiment in condensed matter science we would like to be able to use the momentum change, **Q**, and energy change, ω, of the scattering particle to measure a set of functions $S_{AB}(\mathbf{Q},\omega)$ over a very large area in the measurement, (**Q**,ω), space. This measured information can then be transformed into the space where we understand, that is real space and time, (**r**,t). The transformed function, $G_{AB}(\mathbf{r},t)$, tells us about the relative positions and motions of atoms type A and B, i.e. 'where the atoms are and what the atoms do'. However real scattering experiments are not ideal in several ways:

- Only a limited (**Q**,ω) area is covered.
- The measured I(**Q**,ω) must be corrected for experimental effects to obtain the required function, S(**Q**,ω).
- S(**Q**,ω) is a sum of the different $S_{AB}(\mathbf{Q},\omega)$ corresponding to the different types of atoms in the material.
- There may be some known or unknown coupling function, e.g. I(**Q**,ω)=C(**Q**,ω)*S(**Q**,ω),



Neutrons tell you 'where the atoms are and what the atoms do'.

Figure 2: The particular advantages of the neutron as a scattering probe.

In these respects neutron scattering has particular and rather unique advantages, as illustrated in Figure 2.

- I(**Q**,ω) can be measured over a very large area of (**Q**,ω) space – much larger than for any other technique (see Figure 4).
- The neutron is a weak probe so the measured intensity can readily be corrected for experimental effects to obtain the scattering cross section on an absolute scale.
- The neutron scattering length, b, is a constant for a particular element, i.e. independent of **Q** and ω, and is isotope dependent. This enables the corrected I(**Q**,ω) to be directly related to S(**Q**,ω), and the use of isotopic substitution (contrast variation) where several measurements are performed to provide information on different $S_{AB}(\mathbf{Q},\omega)$. The corrected data can also be directly related to the results of computer simulation or modelling.
- The neutron has a magnetic moment, but no charge, so the same arguments apply to measuring the quantities of

interest for understanding magnetic structure and dynamics.

Of course neutron scattering has some specific disadvantages, the most obvious of which are that it can only be carried out at a small number of large facilities and that it is flux limited. ESS will have a small impact on the first of these, but a large impact on the second.

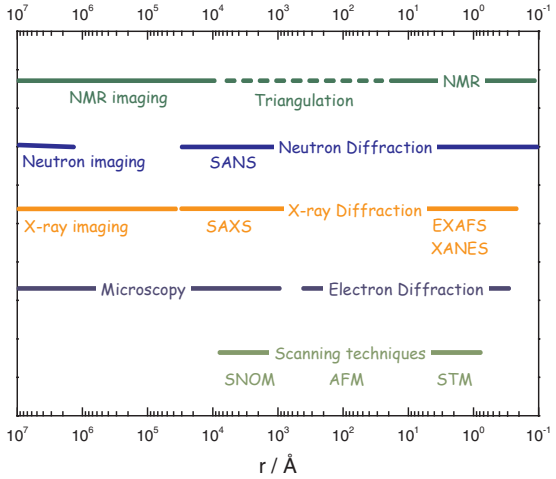
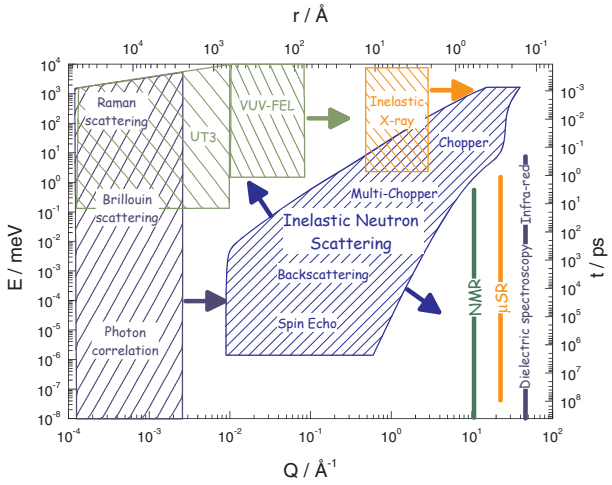


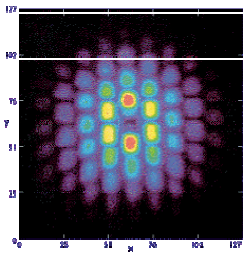
Figure 3: The range of neutron diffraction and other experimental techniques. While there is considerable overlap in terms of the distance scales, the information obtained is often complementary because of the different element specificity. Furthermore, magnetic structure determination is nearly entirely a domain of neutron scattering. Techniques in the lower half of the diagram are typically only applied to very small samples, which can be an advantage or a disadvantage.

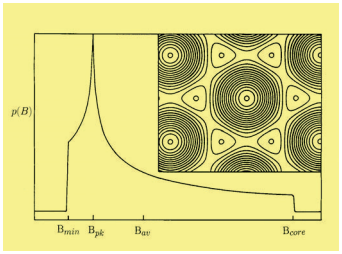


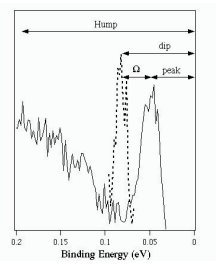
Inelastic neutron scattering gives information that covers a large area in space and time. Other techniques mainly cover different areas.

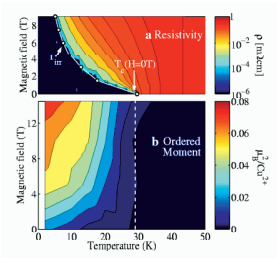
Figure 4: The range of inelastic neutron scattering and other experimental techniques. The complementarity is obvious. With ESS and the FEL/XFEL the ranges will extend in the directions indicated, filling in the 'missing areas'. Note that the diagram covers typical atomic length and time scales.
Techniques that do not directly provide distance information are indicated only as bars along the time axis. The time scale only refers to equilibrium phenomena. Non-equilibirium effects, such as those studied in 'pump-probe' experiments at very short times (fs) will always remain the domain of photon based techniques. At ESS 'pump-probe' experiments will be possible down to µs, covering the longer length and time scales important in soft condensed matter and biological systems.

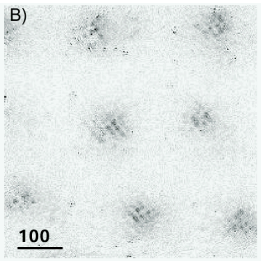
High temperature superconductivity











- ***Small angle neutron scattering***
- ***Muon spin rotation***
- ***ARPES***
- ***Inelastic neutron scattering***
- ***Magneto-transport***
- ***Neutron diffraction***
- ***Scanning tunnelling microscopy***

High T_c superconductors are extreme type II superconductors where studies of the vortex structures and dynamics in the mixed state are important for both fundamental understanding and potential technological applications. Theory predicts that the vortex cores may be magnetic, but while great progress has been made in understanding the flux line lattice structure, searching for the magnetic signature from the vortex core pushes present techniques to their limit.

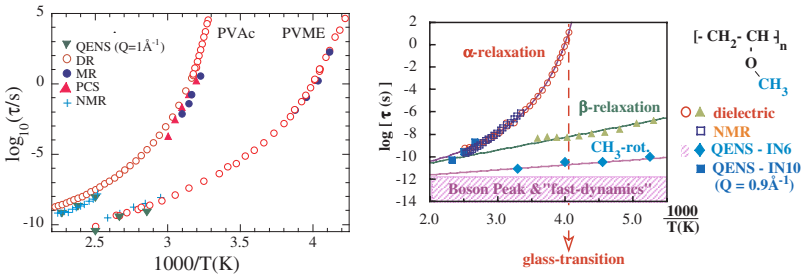
Top left: The flux line lattice in $YBa_2Cu_3O_7$, as measured by small angle neutron scattering.
Top centre: Muons can be used to measure the internal field probability distribution resulting from the flux-line lattice, e.g. in $Bi_2Sr_2CaCu_2O_{8+\delta}$. Such measurements enable monitoring of flux-lattice melting.
Top right: Angle resolved photoemission spectroscopy (ARPES) measures the energy spectrum of quasi-particles at any point in reciprocal space. In $Bi_2Sr_2CaCu_2O_{8+\delta}$ the low T spectral function exhibits a 'peak-dip-hump' structure. The 'peak-dip' energy difference is believed to be due to the interaction of the electrons with a collective mode of energy Ω. It has been suggested that this mode corresponds to the magnetic resonance peak observed by inelastic neutron scattering in several high T_c superconductors below T_c. The energy of the 'peak-dip' difference for $Bi_2Sr_2CaCu_2O_{8+\delta}$ matches that of the magnetic resonance, implying a tight connection between the electronic and magnetic properties [1].
Bottom left: Neutron diffraction on $La_{1.9}Sr_{0.1}CuO_4$ has provided the first compelling evidence that the vortices are indeed magnetic. Below the zero-field critical temperature, an applied field has the effect of inducing antiferromagnetic, incommensurate magnetic order, at the same time as creating the vortex state. At low temperature, the induced order increases first linearly with field, and then starts to saturate, in agreement with theoretical prediction. The upper panel shows magneto-transport measurements parallel to the CuO_2 planes; the colours indicate the electrical resistivity. In a magnetic field, vortices are thought to form at temperatures where the resistivity falls below its value at $T_c(H=0)$. Phase coherent superconductivity, characterised by zero resistance, sets in at the much lower 'irreversibility' temperature, $T_{irr}(H)$, marked by the white circles. In the lower panel the square of the ordered spin moment per Cu^{2+} ion, proportional to the neutron scattering signal, is shown as a function of temperature and applied magnetic field. It first becomes significant below the zero-field superconducting transition temperature $T_c(H=0)$ and increases with decreasing temperature and increasing field [2].
Bottom right: Spectacular confirmation of the Q-space image of the antiferromagnetic vortex state, as provided by neutron diffraction, has been given by real space STM images of the vortex state in $Bi_2Sr_2CaCu_2O_{8+\delta}$. Here STM probes the electronic local density of states, which is periodically modulated in and around the vortex cores (seven vortices are visible). The period is half that found for the magnetic order with neutrons, as expected for coupled spin and charge density wave order parameters [3].

III. NMR and neutrons

NMR is a powerful technique but conceptually much more difficult than neutron scattering.

NMR and neutron scattering techniques have in common the possibility to observe both structure and dynamics, although in many cases the space and time scales are different. The atomic specificity may also be different, but both techniques are favourable for studying light atoms, particularly hydrogen. However NMR techniques do not reveal the correlations in space and time that are present in the dynamic structure factor measured with neutrons. With the advent of very high field NMR, many more nuclei of low gyromagnetic ratio and low molecular concentration and/or abundance can be studied. Multiple pulse and multiple quantum techniques allow, in favourable cases, detailed study of the local structure both of individual large molecules in solution or of the solid state. However, compared to diffraction techniques that are conceptually simpler, NMR techniques may suffer severe limitations for particular systems and require care and experience in setting up the experiment and critically interpreting the result. Even if modern commercial instruments are already provided with software for a large range of advanced techniques, it cannot be taken for granted that naive attempts to reproduce the experiments of experts but on new, even if in principal similar, problems will succeed. Experiments have to be tailored to individual systems, taking into account specificities such as their relaxation times in the laboratory or rotatory frame.

Relaxation, mode coupling and the glass transition



Relaxation in complex systems has become an enormous field of study in the past decade. The ideas of mode coupling theory, originally developed for simple liquids, have been applied to polymers. The glass transition has been studied in bio-molecular systems. The common aspect that brings these topics together is the importance of relaxation processes over an enormously wide timescale (up to fifteen orders of magnitude), which clearly necessitates the use of many complementary experimental techniques. Quasi-elastic neutron scattering and neutron spin-echo spectroscopy are particularly powerful in that they can simultaneously provide information on both time and distance scales and hence, especially when used in combination with H/D substitution (if appropriate), help to separate some of the different contributions to the relaxation.

Left: The characteristic time for segmental relaxation of two polymers, poly(vinyl acetate) (PVAc) and poly(vinyl methyl ether) (PVME) as "seen" by different relaxation techniques (Courtesy of A. Arbe and J. Colmenero).

Right: Relaxation map of PVME (Courtesy of A. Arbe and J. Colmenero).

- ***Dielectric relaxation (DR)***
- ***Quasi-elastic neutron scattering (QENS)***
- ***Mechanical relaxation (MR)***
- ***Photon correlation spectroscopy (PCS)***
- ***NMR***

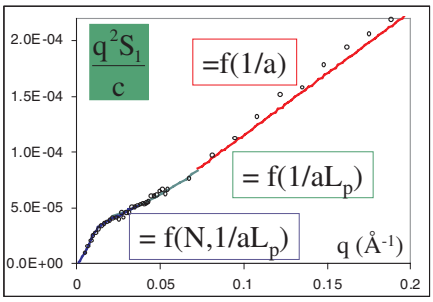
The secondary and ternary structures of proteins and protein complexes can be obtained from 2-D NMR in solution, using the Overhauser effect (NOESY maps) to measure the distance between well identified residues. This triangulation may be very tedious if a low resolution crystallographic structure is not available. For cases where single crystals are impossible to grow, NMR focuses on the small structural differences induced by solution in water, by the mobility of some parts or the binding of a substrate.

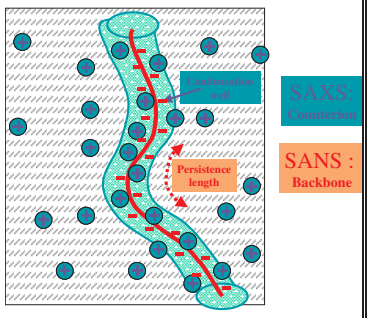
Protein structure determination in solution.

Understanding the catalytic efficiency of solid zeolites may involve the measurement of inter-atomic distances with an accuracy of a few hundredths of an Å. REDOR techniques allow this precision in cases where there is an effectively isolated pair of nuclei. The application to more coupled systems requires a combination of NMR and diffraction techniques.

Catalyst efficiency.

Poly-electrolytes





- ***Small angle neutron scattering***
- ***Small angle x-ray scattering***

Small angle neutron scattering (SANS) and small angle x-ray scattering (SAXS) can be used in a complementary way to study charged systems in soft matter. One example is poly-electrolytes, which are polymers bearing charged groups. They include both synthetic polymers of industrial interest (colloid stabilisation, high viscosity additives, super-swelling gels) and natural polymers (DNA, polysaccharides). In solution in water, electrostatic interactions control both the conformation of the polymer backbone, which is rod-like due to repulsion between charged monomers, and the distribution of counter-ions, which are usually positive metallic cations, monovalent, e.g. Li^+, Na^+, K^+, or divalent, e.g. Ca^{++}.

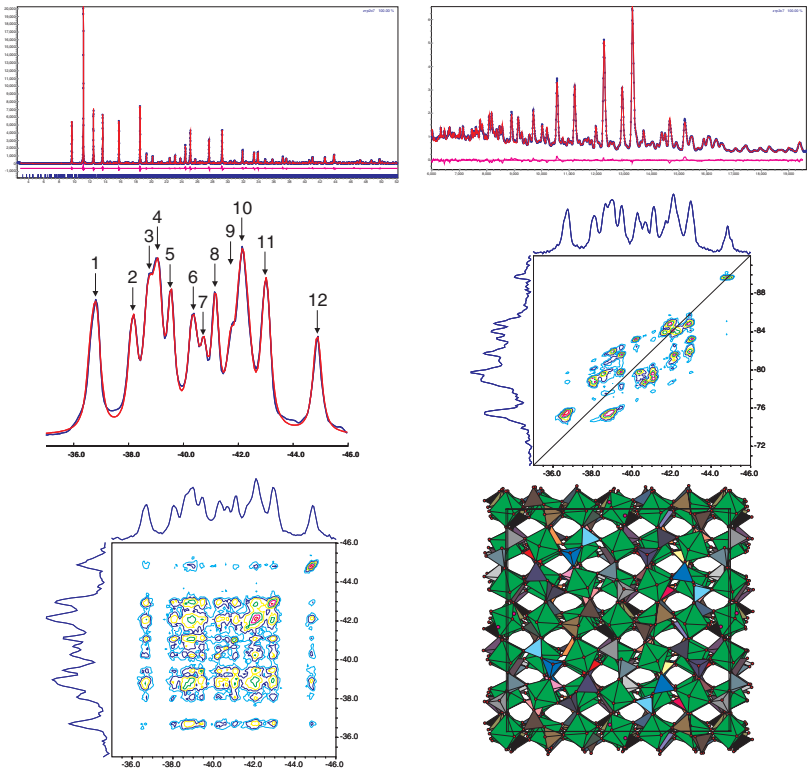
Using a mixture of normal (hydrogenous) and deuterated chains, SANS allows direct measurement of the chain conformation, while the counter-ions can be neglected (*left*). For the same sample, SAXS will be sensitive to the high contrast between water and the counter-ions, giving information on the fraction of counter-ions trapped in the high potential near the backbone, and on the correlations between them (*right*). This can be extended to more complicated polymer architectures, such as star-shapes. In the case of Ca^{++} anomalous x-ray scattering can also be used, so these techniques can be extended to systems related to biology.

The rotational dynamics of individual chemical groups in plastic crystals or polymers can control the energy dissipation which is favourable for toughness, but unfavourable for insulators submitted to high frequency electromagnetic fields. This can be studied by isotopic labelling with a quadrupolar nucleus, such as deuterium, and 2-D exchange spectroscopy. The experiment samples the rotational autocorrelation function

Molecular dynamics in soft matter.

in a different time window than that probed by the incoherent dynamic structure factor measured by neutron scattering. Because of the Q selectivity the latter has the advantage of being sensitive to rotations and translations and the coupling between them. 4-D exchange experiments can be designed to probe the instantaneous heterogeneity in glasses and the lifetime of these very slow density fluctuations. Such heterogeneities may be also probed by neutrons at different length and time scales, again exploiting the Q-dependence.

Zero thermal expansion ceramics



ZrP_2O_7 is a ceramic material that is of interest because it has low thermal expansion over a wide temperature range, a property that is intrinsically linked to its particular crystalline structure. This is one of the most complex structures ever solved by powder diffraction, but it required the combination of five sets of data from neutron and x-ray diffraction and NMR (Courtesy of J.S.O. Evans).

High resolution (synchrotron) x-ray diffraction gives information on the basic structural framework and accurate lattice parameters (*top left*). Neutron diffraction is needed to give more precise information on the O positions and thermal parameters, which are of particular importance (*top right*). However the refinements were not entirely satisfactory and the choice of space group was ambiguous. Three sets of NMR data, 1D ^{31}P NMR (*centre left*), 2-D ^{31}P NMR (*centre right*) and 2-D Double Quantum (Post C7) NMR (*bottom left*), were needed to unambiguously determine the space group, leading to an accurate solution and refinement of the crystal structure (*bottom right*).

- ***x-ray powder diffraction***
- ***Neutron powder diffraction***

- ***1-D NMR***
- ***2-D NMR***

Static inhomogeneities on the mesoscopic scale, such as arise from spinodal decomposition in polymer blends or constrained phase separation in block copolymers, can be identified by NMR. This involves selective excitation of magnetisation in one phase, followed by diffusion to the nuclei

Mesoscale inhomegeneities.

in the other phase, which can be followed using solid state high resolution techniques. Interpretation of the time evolution in terms of morphology requires the spin diffusion coefficient and the shape of the domains to be known. Characterisation by neutron diffraction (where the contrast can be enhanced by isotopic substitution) is more direct and allows a calibration of the spin diffusion coefficient, which cannot be directly measured.

Imaging and tomography.

Neutron and x-ray tomography complement the progress of NMR imaging in medicine (based on 2-D selection through magnetic field gradients, and 3-D reconstruction). This progress now allows resolution in the μm range, of interest for bulk measurements in non-transparent systems at the lower Q range accessible to small angle neutron scattering (for example imaging of different rubbers in tires).

Dynamic polarisation.

^{1}H is a strong incoherent scatterer of neutrons, but a weak coherent scatterer. Isotopic H/D substitution is a standard technique for increasing the coherent scattering, and decreasing the incoherent. An alternative method is nuclear polarisation of ^{1}H, which allows one to change the scattering *in-situ* and continuously. It can be used in several ways. High (> 50%) ^{1}H polarisations have been used to unravel structural details of macromolecules in solution and of hydrogen rich single crystals. Alternatively, the scattering from paramagnetic centres can be strongly and selectively increased by surrounding them with polarised ^{1}H domains. The combination of simultaneous neutron scattering and spin-selective NMR pulse techniques is a promising yet so far unexplored field for which a pulsed source such as the ESS would be particularly suitable, enabling the study of relaxation times in the ms to s range.

IV. The FEL/XFEL and neutrons

The FEL and x-FEL offer exciting new possibilities. The ESS will be highly complementary.

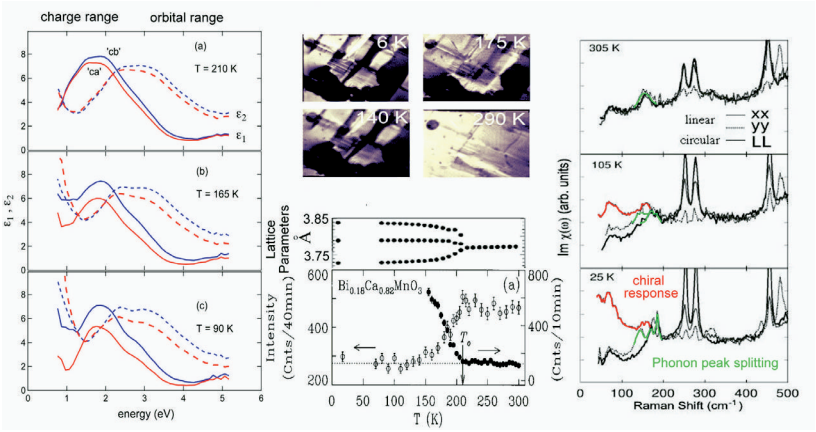
Recent progress in the development of VIS, UV, and VUV free-electron lasers (FEL) has produced exciting new possibilities for studies in the solid state, soft condensed matter and life sciences. New development projects (DESY x-FEL@TESLA; Stanford x-FEL) aim to extend the spectral range of the FEL from the VUV to the hard x-ray range. By 2010 one can expect that the scientific communities using these new types of device will have grown significantly. Some competition has grown up between the x-FEL and ESS projects because they are seeking funding at the same time. Each facility will certainly offer separate possibilities to explore scientifically 'uncharted waters'. However the total area that can be covered is undoubtedly much larger if both facilities are used together in a complementary fashion.

Development of optical and neutron methods will provide seamless coverage of a large (Q,ω) region.

Many different optical techniques have been used in the past to obtain information on atomic and electronic structures and dynamics. Elastic and inelastic techniques such as reflectometry, ellipsometry, Raman and Brillouin scattering and photon correlation spectroscopy, are well established. However, even this more classical field is presently under strong development because of industrial demands (VUV-

lithography) and due to the introduction of new high power light sources such as the FEL. One example is the UT3-system for inelastic light scattering in the deep UV and upper VUV spectral range. It can thus be expected that the 'optical' techniques at low momentum transfer will move to higher momentum transfers and (almost seamlessly) connect to the frequency/momentum range of inelastic neutron scattering (see Figure 4).

Mixed valence manganites



The possibility of applications in the magnetic recording industry has increased interest in colossal magneto-resistance materials in recent years. These are one example of the more generally interesting class of strongly correlated systems, where the relationships between charge, lattice and spin degrees of freedom, both locally and at long range, are of central importance. Here we illustrate the complementarity between different optical techniques and neutron scattering in a study of $Bi_{1-x}Ca_xMnO_3$ ($x \sim 0.75$), which shows a metal-insulator transition that is accompanied by charge ordering. Note how the different techniques are used to give information on scales ranging from macroscopic to atomic. As neutrons couple directly to mass and spin, and photons couple preferentially to and through charge degrees of freedom, these techniques are also complementary on a very fundamental level.

Left: Ellipsometry is used to determine the anisotropy in the dielectric function, $\varepsilon = \varepsilon_1 + i\varepsilon_2$, due to ordering effects in the charge and orbital channels. ε_1 and ε_2 are denoted by solid and dashed lines, respectively. Two measurement configurations, represented by red and blue lines, are rotated by 90° with respect to each other to probe the optical anisotropy. This starts to develop in an energy range that is governed by the O-2p to Mn-e_g levels and thus represents orbital ordering of the Mn-e_g levels. The complete charge ordering develops with decreasing temperature, as shown by high anisotropy in the charge channel [4].
Top centre: The anisotropy can be imaged by polarised microscopy and the domain sizes can be determined [5]. Each image shows an area $500 \times 800\ \mu m^2$.
Right: Inelastic light scattering from a single domain provides information on structural changes at the transition by, e.g. peak splitting of the phonon mode around 200 cm^{-1}, and reveals the existence of a quasi-elastic fluctuational chiral reponse breaking time-reversal symmetry in the charge-ordered state[8].
Bottom centre: Neutron scattering provides complementary atomic scale information about a transition from a cubic to orthorhombic structure (upper panel) that explains the phonon peak splitting. No fluctuational ferromagnetic moment is found in the charge-ordered state (lower panel) (open circles, $|q| = 0.17$ Å^{-1}, $E = 1$ meV), but an antiferromagnetic background (filled circles, $q = (1/2,1/2,0)$, $E = 1.7$ meV) suggesting that the fluctuational moments as seen by inelastic light scattering are due to closed loop motions of charge degrees of freedom [6].

- ***Ellipsometry***
- ***Microscopy***
- ***Raman spectroscopy***
- ***Neutron diffraction***
- ***Inelastic neutron scattering***

Investigating the same problem in different ways.

The FEL/x-FEL and the ESS are complementary in the sense that they can provide information from opposite ends of the same scientific problem. As an example, one of the most exciting opportunities the FEL offers is an excellent time resolution (50 – 100 fs) for pump and probe experiments in synchronisation with optical lasers, allowing the atom specific analysis of relaxation processes, without however offering atomic resolution of the corresponding length scales. Moreover, the FEL beam diameter is optimised to be below 50 μm and has typically a penetration depth of about 10 - 100 nm in the VUV, which allows the investigation of small samples and thin films, but not large and thick materials. On the other hand the ESS allows the investigation of large sample volumes and structures buried deep within one sample. Furthermore, the VUV-FEL only has a narrow energy range in which water does not absorb the photons (the' water window') to allow studies of biological systems in an aqueous environment, whereas neutrons are well suited for such a study over a wide energy and momentum range.

Complementarity, not competition.

One area of apparent competition might be in the high energy, high momentum transfer region (top right corner of Figure 4). This used to be the clear preserve of inelastic neutron scattering, but developments of (triple-axis) x-ray inelastic scattering have now produced a large region of overlap. In specific cases, for example the measurement of phonon dispersion curves at high energies, the neutron will always 'struggle' because of its lower velocity, so in the overlap (Q,ω) region photons would now be the probe of choice. However for comparable measurements in liquids and glasses, the different element specificity of the two techniques makes them highly complementary for all but the simplest (elemental) systems. Developments at the x-FEL and the ESS will enhance this complementarity even further.

High throughput studies are complemented by highly focused studies.

Another area of apparent competition could be in 'high throughput' studies. The x-FEL might allow, for example, 'one shot' structure determinations of single protein molecules, thus providing the possibility for even more structure determinations than are carried out today. However, it must be remembered that the ESS will be a unique facility in Europe (and the world). Even if neutrons at the ESS are required to answer specific questions on only a fraction of one percent of all these structures, this will still mean that the relevant instruments are continually overloaded. Neutrons may be slow, but they are able to reveal specific and unique information.

V. Muons and neutrons

μSR: three techniques in one.

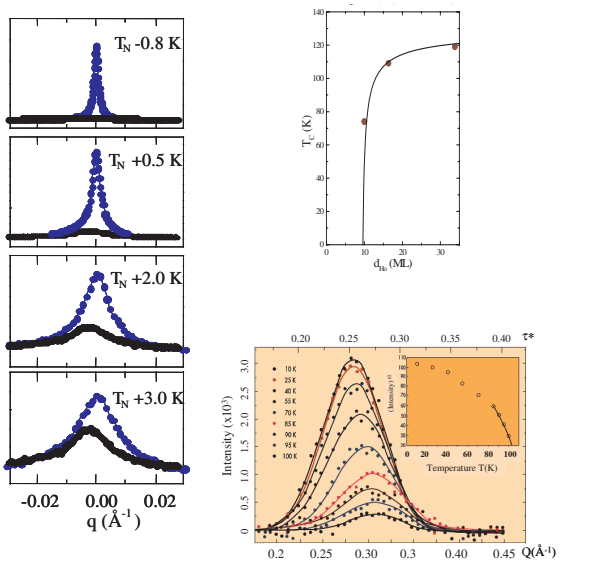
μSR is a universal acronym for muon beam studies; **m**uon **s**pin **r**otation, **r**elaxation and **r**esonance. Rotation describes the dephasing of muon spins by local fields within the sample in the presence of a field transverse to the muon spin direction. Relaxation defines the time-dependent loss of polarisation of the muon spins by internal fields either in zero applied field or in a field applied parallel to the initial muon

spin direction. Resonance is observed in the presence of an applied RF field. The precessional or relaxational behaviour of the muon spin is determined by measuring a time differential histogram of the positrons that are preferentially emitted along the muon spin direction.

Muons have a special place as a complementary probe to neutrons in a document on the ESS. Like neutron scattering, μSR can only be carried out at large scale facilities. In two cases those facilities are also neutron facilities, where there is considerable overlap of the scientists and scientific problems that each probe addresses.

A muon facility could be built at the ESS.

Magnetic thin films



- ***Resonant x-ray magnetic scattering***
- ***Polarised neutron diffraction***

The first neutron diffraction study of a magnetic structure, anti-ferromagnetic ordering in MnO, was published in 1948, while the earliest papers on the use of photons to study magnetism appeared in the 1970's. Nowadays, x-ray magnetic scattering has become a routinely used technique, showing a great deal of complementarity with neutrons. While neutrons are still the only technique for studying powder samples, or magnetic excitations using inelastic scattering, x-ray scattering offers many new possibilities, for example for separating spin and orbital moments.

Ho is a model system for investigating thin film magnetism. The atomic moment is the highest for all elements, giving a strong signal for neutron experiments, it is antiferromagnetic over a wide temperature range, with the magnetic and lattice (charge) scattering well separated, and the resonance energies are well placed for x-ray studies. Currently neutron studies are in practice flux limited to 16 monolayer samples, but x-ray studies can go down to 10 monolayers using the considerable signal enhancement provided by resonant magnetic x-ray scattering at the Ho M_5 edge (Courtesy of V. Leiner, W. Weschke, C. Schüßler-Langeheine, H. Ott and H. Zabel).

Left: The Ho(000 τ) peak of a 16 monolayer thick Ho(0001) film on a sapphire substrate, with a Y(0001) buffer layer. Blue points: specular radial scan. Black points: slightly off-specular scan showing only the diffuse background. The Bragg peak has a true Gaussian shape below the Néel temperature; above it is Lorentzian from the diffuse scattering.
Right: A similar series of scans for a 10 monolayer film
Top centre: The scaling of the Néel temperature with film thickness.
Bottom centre: Radial neutron scans of the (00.0+1ι*) magnetic satellite peak for a 16 monolayer film. In the inset the magnetic-order parameter is plotted as a function of temperature.

Measurement of ultra-small magnetic moments.

The greatest strength of the μSR technique is that it is a highly sensitive probe of the distribution and dynamics of extremely small magnetic fields at interstitial lattice sites within a sample, both in zero and applied magnetic field. This is in contrast to NMR, which probes internal fields at regular lattice sites. The fields generated by nuclear moments are easily measured by μSR, whilst the dynamical response (10^{-10} - 10^{-4} s) links the time domains covered in static and common ac bulk measurements and neutron spectroscopy The field dynamics measured by the muon can be related either to intrinsic internal field fluctuations, or to the apparent fluctuations caused by a muon diffusing through a lattice. The latter case is of interest since the muon itself behaves like a light isotope of hydrogen (M_μ = 1/9 amu). By now there is a growing list of applications in which the information gained by the complementary use of muons and neutrons provides greater insights than either technique alone can offer.

Magnetic structure determination.

μSR is not itself a technique for structure determination, but it is complementary to neutron magnetic structure determination methods. The muon's ability to measure ultra-small internal fields has indicated the presence of ordered moments of 10^{-3} μ_B and smaller. Identification of such ordering processes, for example in the high T_c cuprates, heavy fermion systems and organic magnets, has prompted neutron diffraction experiments at the very limits of what is currently feasible (10^{-2}). Such studies will become even more important as the ESS stretches the boundaries of sensitivity and provides the possibility of seeking out novel mechanisms for moment localisation and magnetic order. μSR has also been used to resolve ambiguities in powder diffraction data, for example in rare earth nickelates.

Magnetic fluctuations.

Although the neutron is generally the technique of choice in studies of magnetic spin fluctuations, providing both spatial and temporal information, there are several examples of combined μSR and neutron studies of excitation spectra that have provided additional insights. For example, in β-Mn and YMn_2, spin fluctuation compounds on the limits of moment stability, μSR has identified the spin liquid to spin glass cross-over, undetected by neutron spectroscopy, and enabled mapping of the complete evolution of the spin fluctuation spectra with temperature. Neutron inelastic scattering then enabled the spatial extent of the dynamic spin correlations to be extracted.

Spin glasses.

The spin glass transition is in many ways a relatively simple analogue of the complex structural glass transition. Neutron spin echo is the ideal tool for probing both the spatial extent and dynamics of the spin correlations above T_c, but even with ESS such precise measurements will be difficult and time consuming. μSR provides the opportunity of, on the one hand, screening for suitable model spin glass systems for study by NSE, and on the other of considerably extending the available dynamic range. Similar complementarity is now being exploited in studies of the dynamics of magnetic nanoparticles and recording media.

New developments in the field of μSR include 'cold muon' spectroscopy. Muons of energy a few MeV are cryogenically cooled to a few keV, allowing the typical penetration depth within the sample to be 'tuned' from a few mm to a few Å. They can therefore be used to both complement and extend neutron reflectometry techniques. The cold muon technique has been demonstrated at PSI, although cryogenic moderation of muons is an inefficient process and beams are correspondingly weak.

'Cold' muon spectroscopy: a new technique to complement neutron reflectivity.

Pulsed RF techniques, which provide the opportunity to reorient muon spins within the sample (in many ways similar to spin echo NMR), are under development. These will be very complementary to the recently developed technique of 3-D neutron polarimetry, which is the most advanced method available for magnetic structure determination.

Advanced methods for magnetic structure determination.

VI. Computers and neutrons

Neutron scattering and computational techniques are highly complementary, for one simple reason. From a model of atomic positions, possibly as a function of time, it is straightforward to calculate the expected neutron scattering spectrum and to compare it to experimental results, on an absolute scale. There are no unknown, or difficult to calculate, coupling functions. Neutron scattering therefore provides the best method for testing and benchmarking computational methods, e.g. Monte Carlo (MC) and molecular dynamics (MD) simulations.

Neutron spectra are easily calculated from computer models.

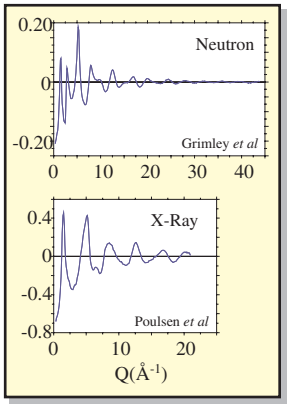
The development of computational techniques, and of course the development of computing power, has been so great over the past 40 years that it might be questioned whether experiment still has a role to play. MD, based on parameterised potentials, can simulate up to multi-million atom models or μs time scales. *Ab-initio* methods enable the calculation of electronic structures or simulation of the structures and dynamics of both atoms and electrons without an input potential. However, while simulations provide tremendous insight into generic types of behaviour, we are not yet at the stage where they can predict the properties of specific materials quantitatively.

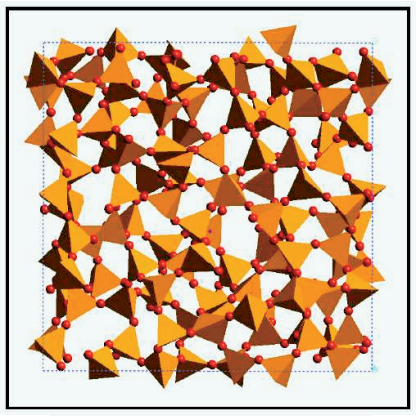
Computing power has developed enormously.

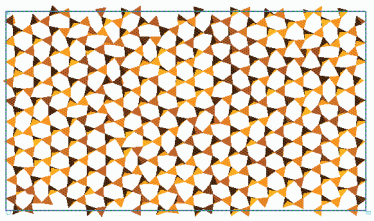
In the next decades we can foresee a situation where the interplay between experiment and simulation increases, and hence the gap between them narrows. In structural genomics there is a trend towards the development of predictive techniques based on large databases of experimental information. This will certainly extend to other areas, and the databases will expand to include many different types of complementary information. The experimental data will either be used to refine improved, transferable, interatomic potentials for MD, or to test refined *ab-initio* methodology. Alternatively it will be possible to directly refine dynamical models based on e.g. inelastic neutron scattering data, in the same way that structural models are nowadays routinely refined on the basis of diffraction data. Such modelling methods also offer a route for combination of many types of complementary experimental data.

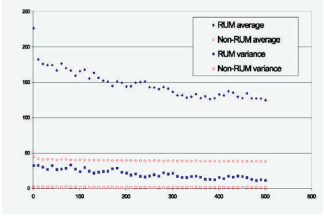
Databases of experimental information, including neutron scattering data.

Silicate crystals and glasses









- ***Neutron diffraction***
- ***x-ray diffraction***
- ***Computer modelling***

- ***Inelastic neutron scattering***
- ***Molecular dynamics simulation***

Silicate minerals make up a large part of the earth. Silicate glasses have been produced for thousands of years. Yet there are still many unanswered questions about the basic properties of these materials, which are both of fundamental interest and related to the physical properties that make them so important.

Nowadays, the combination of wide Q range neutron diffraction and (hard) x-ray diffraction data (*top left*) [7] and computer modelling can be used to produce detailed structural models of silicate glasses (*top right*) [8], despite the lack of long range crystalline order.

'Rigid unit modes', involving correlated rotations of many tetrahedral SiO_4 groups, are important in the dynamics of both crystalline and glassy polymorphs (*bottom left*) [9]. Information can be obtained by inelastic neutron scattering but, because these modes lack the well known 'characteristic' signatures of phonons (or possibly single crystals are not available), the interpretation is dependent on an understanding obtained from real space analysis of molecular dynamics simulations (*bottom right*) [10]. Low energy rigid unit modes have been identified in simulations of silica glass, which may be related to the tunnelling modes that have been used to explain the low temperature heat capacity of (all) glasses. However, because of the low density of such modes, direct experimental confirmation will require the ESS.

Simulation range is well matched to ESS capability.

The developments that can be readily predicted over the next decade mean that ESS will be well matched to the computational capabilities. The wide (Q,ω) range illustrated in Figure 4 corresponds to the (r,t) range that will by then be routinely accessible by MD simulations – for example simulations of polymer structure and dynamics which can simultaneously relate molecular group motions on the sub-ps timescale with whole chain relaxations and motions on the ns-μs timescale. ESS will allow measurement over this whole range with improved statistical accuracy and resolution.

Another aspect of computational developments that should not be overlooked for ESS is the great potential for the use of

'expert system' software to allow the most effective use of the valuable beam time. Detailed pre-experiment simulations will suggest the most effective experiment plan, which can then be automatically updated and modified as the experiment progresses. High level data analysis can also be automated, leaving the use free to concentrate on interpretation of the results rather than manipulation of the data. This is also important because a high proportion of the users of the ESS will not be 'neutron experts'; often they will be experts in other techniques who require the complementary information provided by neutrons.

'Expert system' software for effective use of beam time at the ESS.

VI. Electron microscopy and neutrons

The ESS will allow an extension of current small angle neutron scattering (SANS) towards even smaller angles by using focusing techniques (Foc-SANS). This will increase the complementarity with microscopy, e.g. electron microscopy (EM) and atomic force microscopy (AFM), which we discuss here in the field of soft matter and biology.

Focussing SANS at the ESS will complement microscopy.

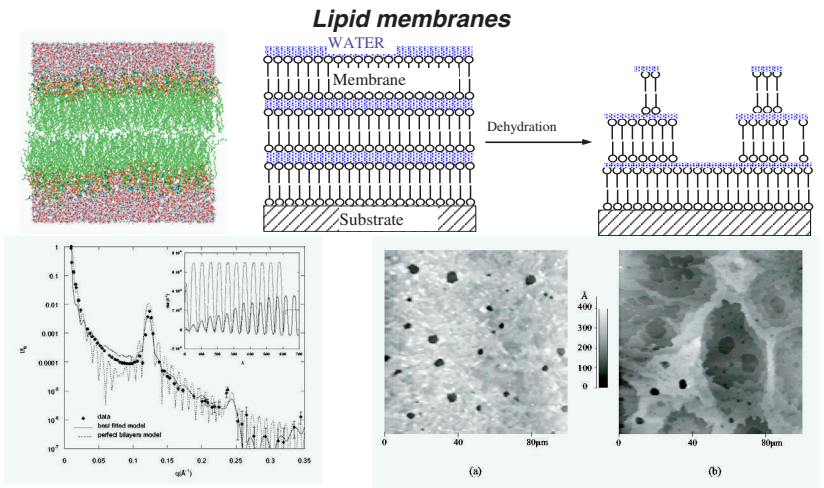
One well known difficulty for EM has been the characterisation of solutions, since this can only be done after evaporation of the solvent. The same is true for AFM, which is increasingly used as a characterisation method in the growing field of new macromolecular and supra-molecular synthesis. The shape of individual solutes must not be dependent on the strong changes induced by elimination of the solvent. For example, the shape of a polymer chain (self avoiding walk) is determined by monomer-solvent interactions. However, good results can be obtained for more compact shapes. Similarly, evaporation may modify the correlations between solutes. Real solutions or dispersions are more easily studied with SANS, which can provide information both on the shape of individual elementary macromolecules and on arrangements of aggregates and clusters (or "j-mers" in the quaternary structure of proteins). Some more continuous structures can be studied by EM, but here again complementary SANS will be useful to prevent artifacts and undoubtedly strengthen the interpretation. Some recent EM techniques are very powerful for fluid systems, such as cryo-fracture which allows observation of the surface resulting from a fracture inside a bulk sample. Self-assembled systems, for example, are a common field of study.

Studies of solutes and aggregates in solution.

For solid samples (no solvent), EM can of course be much more directly applied, but the preparation of samples is a delicate and lengthy process that may again alter the material. The 'direct' images obtained are exciting and appealing. However care must be taken in their analysis, even if the interpretation seems obvious. Sometimes many pictures are necessary to obtain a correct statistical weighing of different phenomena. SANS can provide help even at this level, because the neutron measurement is relatively fast and simple (once the beam is available). A few SANS spectra will complement a long series of images by providing a quantitative and accurate Fourier transform over an ensemble average. In a second step, many samples can be studied,

In-situ studies of solids under deformation.

possibly with *in-situ* fast variation of external conditions (kinetics) and complex sample environment. For example, EM cryofracture could be carried out on a surfactant micro-emulsion frozen under shear. Foc-SANS would provide complementary data *in-situ*, under deformation. Of course it is then easy to match this information with the smaller size range of conventional SANS, 1000 Å and below, where EM in soft matter is difficult.



- ***Molecular dynamics simulation***
- ***x-ray reflectometry***
- ***Neutron reflectometry***
- ***Atomic force microscopy***

Membrane lipids constitute about 50 % of the mass of most animal cell plasma membranes. All are amphipathic molecules containing both hydrophilic and hydrophobic surfaces. They can take the shape of (*top left*) a bilayer (bimolecular sheets with the hydrophobic tails sandwiched between the hydrophilic head groups) or a micelle (a spherical shape with tails pointing inward). Membrane lipids have many functions including the responsibility of what goes in and out of the cell.

Stacks of multi-lamellar lipid membranes are unstable against dehydration (*top right*). Reflectometry provides information on the structure perpendicular to the layers. x-ray data can extend to high Q values and hence provide accurate information on the overall layer thickness, roughness etc. However neutron data (*bottom left*), using isotopic (H/D) substitution, provides better contrast between the water and the membrane and hence on the periodicity within the layer. AFM (*bottom right*) allows lateral structure determination and helps to explain some features of the neutron and x-ray profiles (Courtesy of L. Perrino, J. Dayan, F. Graner, E. Amalric and G. Fragneto).

Distinguishing metal and mineral components from organic components.

Neutrons have the same complementarity here as when compared to x-rays, since EM is also sensitive to electronic density whereas SANS measures nuclear density. SANS therefore allows studies of metallic or mineral components and organic ones. One can take the example of platinum particles stabilised by a polymer. After drying, TEM shows that most of the particles are surrounded by polymer. However in solution aggregates of such wrapped particles may exist and would be detected by SANS. If the dispersion is vaporised to form a film this is not visible. Once this characterisation has been done, study of the polymer around the particle surface can be carried out using deuteration. Deformation under the electron beam is possible for thin films, whereas SANS can study *in-situ* deformation of thick films for which the synthesis and mechanical characteristics are different.

AFM has already been mentioned above since the complementarity with SANS presents some similarities. AFM can provide information over some of the range of conventional SANS, though SANS (and the extension to wide angle neutron scattering) can give information at sizes smaller than the AFM lower limit related to tip effects. Similar considerations may apply to the complementarity between AFM and neutron surface measurements. Neutron reflectivity probes the profile orthogonal to the surface, while off-specular techniques (grazing angle-SANS) give information on the lateral structure, which is more closely related to the information from AFM.

AFM complements specular and off-specular neutron reflectivity.

VIII. Complementary techniques in the life sciences

Research in the Life Sciences exploits a wide range of multidisciplinary techniques that provide complementary information on biological processes at the atomic, molecular and cellular levels of detail. The tremendous progress in genomic research and engineering has led to concerted initiatives that aim to determine how genomic codes are translated and transformed into the complex systems and networks that compose a living cell. Structural genomics programmes aim to determine the high resolution structures of all proteins and nucleic acids (DNA/RNA) that are accessible through the main high-throughput techniques, such as x-ray crystallography and solution NMR. As this vast structural database begins to accumulate, demand will rapidly grow for access to other techniques that can help us to understand how these macromolecules work and how they combine and interact with lipids, membranes and other components in the cell.

Structural genomics will increase demand for complementary techniques.

Neutron diffraction and spectroscopy make significant contributions across a wide range of biological research. This is primarily because of their ability to detect hydrogen and to distinguish between its isotopes, H and D. Applications range through (i) pinpointing individual hydrogen positions in proteins, (ii) probing the structure and dynamics of proteins, nucleic acids and membranes, (iii) characterising the interaction of higher order protein/protein and protein/RNA/DNA complexes. These studies use neutrons to address questions that have not - or cannot - be answered by other techniques and the results have often been profound. It is clear that novel techniques and applications will emerge at ESS and applications in life sciences can be expected to flourish.

Neutrons and isotopic H/D substitution show where hydrogen atoms are and what hydrogen atoms do.

Deuteration

At low resolution, H_20/D_20 contrast variation techniques are used to sequentially highlight and map specifically labelled or chemically distinct components of large protein-protein and protein-nucleic acid complexes. In diffraction, provision of fully (per)deuterated samples reduces the high intrinsic background scattering by a factor of 10 and (dramatically) enhances the visibility of deuterium. In dynamics applications, specific labelling of amino acids or fragments provides an

Deuteration will be an integral part of experimental design.

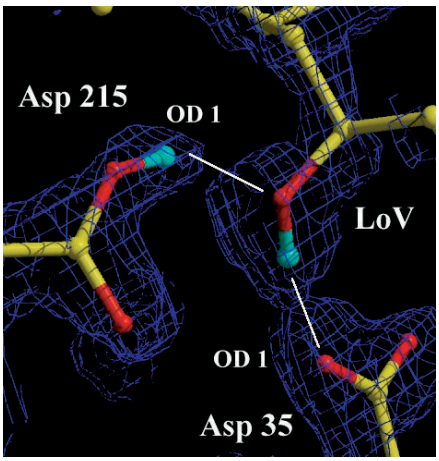
elegant means of probing local dynamics. The provision of specifically D-labelled bio-molecules thus has critical impact upon many neutron applications and will be an integral part of the experimental design at ESS, resulting not only in much higher throughput of experiments but will also new, more sophisticated and more powerful approaches to the solution of complex biological problems.

The most dominant and widely used techniques for high resolution structure solution are x-ray single crystal diffraction and Nuclear Magnetic Resonance (NMR), which account for ~ 14000 and ~ 2600 structures, respectively, in current data bases. Other important techniques, including neutron scattering, cryo-electron microscopy and electron diffraction, have quantitatively lower output but make specific and often profound contributions to the field.

Structural genomics use high-throughput techniques.

- ***Single crystal x-ray diffraction***
- ***Single crystal neutron diffraction***
- ***NMR***

Understanding enzyme mechanism helps drug design



Some enzymes rely upon the transfer of hydrogen atoms in order to work. Once the catalytic mechanism is known, drugs can be rationally designed that fit and block the transfer mechanism at the active site. Aspartic proteinases, which cut and process other proteins, have been implicated in a number of human diseases and targeted for drug design. However, the available x-ray structures of aspartic proteinase/drug complexes showed where the drugs bound at the active site, but could not show where critical catalytic hydrogen atoms resided nor how they interacted.

Neutron diffraction of an aspartic proteinase bound to a potential inhibitor has directly revealed for the first time the key catalytic hydrogen positions at the active site of the enzyme. This clear definition of the charge status of the active site groups is essential for modelling the catalytic mechanism that is used for rational drug design against all members of this enzyme family e.g. the HIV proteinase which is the target of a number of clinically prescribed AIDS drugs.

The neutron structure indicated that the hydrogens involved in drug binding have special properties (low barrier hydrogen bonds). Complementary 1D-NMR techniques were used to verify this and now allow rapid screening to identify whether inhibitor drugs are bound.

Figure: The neutron atomic models (yellow and red) of the catalytic protein groups (Asp 215 and Asp 35) and the drug inhibitor (LoV) at the active site of Endothia pepsin show extra neutron density (blue net) that defines the hydrogen positions (turquoise) [11].

X-ray crystallography

X-ray crystallography is the most important technique, but hydrogen atoms are difficult to see in x-ray structures.

X-ray crystallography is the most powerful and widely used technique for the determination of the structure of proteins, nucleic acids (DNA/RNA) and their complexes, providing that good quality crystals can be obtained. Precise structural parameters are derived, representing a single conformational state of the system. However, proteins are not static, even in single crystals, and diffraction resolution is limited by 'disorder' due to the intrinsic flexibility and local dynamics of the protein, compounded by crystal disorder and radiation damage to the sample, and only rarely extends to atomic resolution (< 1.2 Å). Hydrogen atoms - which constitute ~50% of biological material - typically cannot be seen at all.

Neutron diffraction

Neutron diffraction shows functional hydrogen positions in crystals.

Neutron diffraction enables the positions of individual hydrogen/deuterium atoms and exchangeable waters to be revealed in biological structures at modest resolution (2 Å), provided that large crystals can be obtained. The low intensity of currently available neutron beams restricts applications to proteins of < 50 kDa. Neutron protein crystallography is therefore used to address specific questions concerning enzymatic mechanism, ligand-binding interactions and solvent effects that cannot be answered by x-ray analysis alone.

Neutrons allow detailed analysis of water structure.

Water is vital to almost all biological processes. In neutron diffraction, the visibility of D_2O water molecules is strongly enhanced, enabling water structure and hydrogen bonding geometry to be investigated in detail at protein surfaces. For example, in Concanavalin A (a protein that binds to complex carbohydrates), neutrons provide additional information to x-rays on water structure that is necessary for proper thermodynamic and modelling studies of the sugar-binding interactions. Similarly, in γ-crystallins (eye lens proteins implicated in the formation of cataracts), neutron data supplement high resolution (1.2 Å) x-ray studies to help understand critical water interactions at the surface of the protein.

NMR

NMR is the method of choice for solutions.

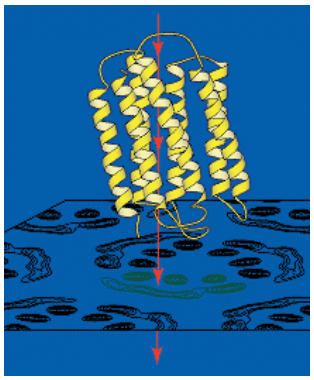
NMR is the method of choice for structure determination in solution, and when proteins are difficult to crystallise (e.g. prion proteins), but is limited to relatively small structures (< 30-40 kDa). The average structural parameters derived from the NMR ensemble are less precise than those obtained by crystallography. However, NMR provides direct information on hydrogen positions and can be used to study protein flexibility and the dynamics of enzymatic reactions. NMR spectroscopy probes time scales that are completely complementary to neutron spectroscopy and the two combine to provide a more complete description of macromolecular dynamics.

Neutrons see disordered components in crystals.

At lower resolution (< 8 Å) H_2O/D_2O contrast variation can be used to locate lipids, detergents and other disordered groups in crystals that often cannot be seen by x-ray analysis alone. For example, membrane spanning regions of the protein Porin

from E.coli are hydrophobic and the protein had to be solubilised and crystallised using detergents before the crystal structure could be determined. The x-ray structure showed the protein in detail, but neutrons were required to locate the associated detergent regions in the crystal. These insights are important, because although membrane proteins account for up to 40 % of the genome and represent 85 % of target proteins for pharmaceutical research, only ~ 30 such structures are known. Understanding the crystallisation behaviour of detergent solubilised membrane proteins is likely to be of primary importance in the future.

Proteins are dynamic, not static



Proteins and nucleic acids are dynamic and naturally bend, flex, twist, rotate and vibrate in performing their functions. Their molecular dynamics can be investigated over a wide range of time and length scales using strongly complementary methods, such as NMR and FTIR. Neutron spectroscopy accesses molecular dynamics in the ns to ps range, appropriate for study of the weak forces that stabilise biological macromolecules and contribute to thermal motions, and is unique in providing simultaneously both the energy transfers involved and the magnitude of the amplitudes.

The conversion of sunlight into energy supports almost all life on earth. Specialised proteins, such as Bacteriorhodopsin (bR) of Halobacterium salinarium, act as light-driven proton ion pumps, creating electrical gradients across cell membranes that transfer energy in much the same way electrical batteries work. The mechanism of the bR system has been studied intensely for decades and numerous techniques have contributed pieces of the puzzle. The 2-D crystal structure was solved by electron diffraction and contains seven trans-membrane helices that surround a light sensitive molecule, retinal. Exposure to sunlight triggers structural changes in the protein and forms a proton gradient. Neutron diffraction established that the water molecules essential to proton transfer where localised in the centre of the bR ring, and located two functionally important glycolipids that surround the protein in its natural membrane environment. Elastic incoherent neutron scattering (EINS) experiments revealed areas of differential dynamical behaviour within the protein and identified a dynamic transition that is likely correlated with function. High resolution x-ray crystallography studies have characterised structural deformations during key intermediary steps of the photo-cycle that are probably coupled to the molecular mechanism of proton transport, but the picture is still incomplete. Subtle new structural features have been found, specific lipids have been shown to influence key steps of the photo-cycle and new experiments are being performed to examine their implications.

Figure: The seven helices of bacteriorhodopsin are shown as they project on the crystal lattice. The red arrow shows the direction of motion of the proton pumped from the inside of the cell to the outside when the protein is illuminated with light.

- ***NMR***
- ***FTIR***
- ***Inelastic neutron scattering***

Cryo-electron microscopy

How do bio-molecules interact?

As the number of protein and nucleic acid structures increases, attention will focus on how they interact and combine to form functional complexes within the cell. The study of larger complexes is beyond the capabilities of NMR and is a challenge for x-ray crystallography, though the results are spectacular when suitable crystals can be obtained! When they cannot, techniques such as cryo-electron microscopy provide highly complementary information at > 10 Å resolution for structures > 250 kDa, but only small angle scattering offers the potential to study these interactions directly in the solution state.

Small angle scattering

Protein/protein or protein/lipid/nucleic acid complexes.

Small angle scattering provides structural information on the interaction of biological macromolecules in solution and on conformational changes under near physiological conditions. Small angle neutron scattering, SANS, provides complementary information to its x-ray equivalent, SAXS, by using H_20/D_20 contrast variation and deuterium labelling techniques to sequentially highlight and map chemically distinct or D-labelled components of larger protein/protein or protein/lipid/nucleic acid complexes (e.g. viruses, ribosome). The structures derived may be interpreted at higher resolution by docking atomic resolution structures into the maps. Next generation SANS instruments at ESS will open new opportunities for dynamic studies to allow conformational changes and assembly/disassembly processes to be followed on biologically relevant timescales. New computational techniques are being combined with region-specific D-labelling of proteins to provide detailed shape functions at medium resolution, ~ 6 Å, that may also be useful as initial phasing models to solve x-ray crystal structures.

Neutron reflectometry

Probing interfaces in biomembranes.

Neutron reflectometry is emerging as a powerful technique for characterising the structure and organisational changes in phospholipid membranes. Whereas complementary techniques such as x-ray reflectometry and atomic force microscopy provide topographic images and detail, neutron reflectometry using specific labelling and contrast variation allows the internal layer structure to be dissected and examined directly. Contrast variation techniques have been used, for example, to study how the peptide penetration is able to traverse bio-membranes without perturbing them, making it an ideal carrier for drug delivery into cells. There is tremendous potential for developing this approach to examine the interaction of (labelled) drugs, peptides and other biomolecules with model and cellular membrane systems.

IX. Conclusion

Without ESS, Europe will be less competitive.

The decision on whether to build a large international facility such as the ESS, which would provide unique scientific possibilities, should be based on the scientific and technological advantages. When the ESS is built it will undoubtedly produce world leading scientific research in its own right. However the results can also undoubtedly be even greater if the ESS is operated in such a way as to ensure that full advantage is taken of the high degree of complementarity that neutron scattering has with other techniques, as has been illustrated above. The ESS, as part of the European Research Area networked with facilities serving other experimental techniques, will maintain Europe's cutting edge in many crucial areas of condensed matter science.

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Chapter 6

Contribution of ESS to Europe's Priority Research Themes

6. Contribution of ESS to Europe's Priority Research Themes

Societal issues interrelate increasingly with the advance in knowledge.

Research with neutrons serves first of all to expand our knowledge on non living and living matter. The promises of ESS in the scientific disciplines have been outlined in terms of scientific flagships in the chapter 4. With the expected transformation towards a knowledge based society, basic knowledge interrelates increasingly with societal issues like the advance of technology, sustainable development, health, aging etc.

In the future, especially with the aid of ESS, neutron scattering will become more than ever before a tool for applied science and will make direct contributions to solving problems more closely related to every day life.

Neutrons will contribute to many applied fields of science.

We expect that important progress will be made for society in the fields of new materials, energy storage, energy transfer, catalysis, "clean" technologies, computers, environment, production processes, the design of pharmaceuticals, biotechnology etc.

Present day achievements:

Precursors of such future societal benefits are already visible in some of today's achievements, where neutron tools have prominently contributed towards the solution of practical problems. We will briefly outline a few:

- ***solvent free dry cleaning.***

  A method of dry cleaning which completely dispenses with solvents and functions on the basis of supercritical carbon dioxide was recently developed at Oak Ridge with the aid of neutron scattering. The method uses CO_2-soluble polymeric soaps or surfactants for the dry cleaning process.

  The invention of this method was a great success and is already being used in 40 states of the United States. The scientists involved received the Presidential Green Chemistry Challenge Award from the US president in 1997.

Figure 1: Presidential Green Chemistry Challenge Award awarded for the development of solvent free detergent in dry cleaning.

- A second example concerns experiments recently performed by Degussa. Degussa uses palladium catalysts to convert ketones into alcohol by oxidation. It was found that after a certain time the catalyst was no longer effective. In-situ neutron scattering experiments at ISIS enabled the cause to be identified. It could be seen from the vibrational spectra that methyl groups had become attached to the surface of the palladium, bringing the catalyst to a standstill. After this cause had been identified it was possible to remedy the situation.

- diagnosis of catalyser disfunction.

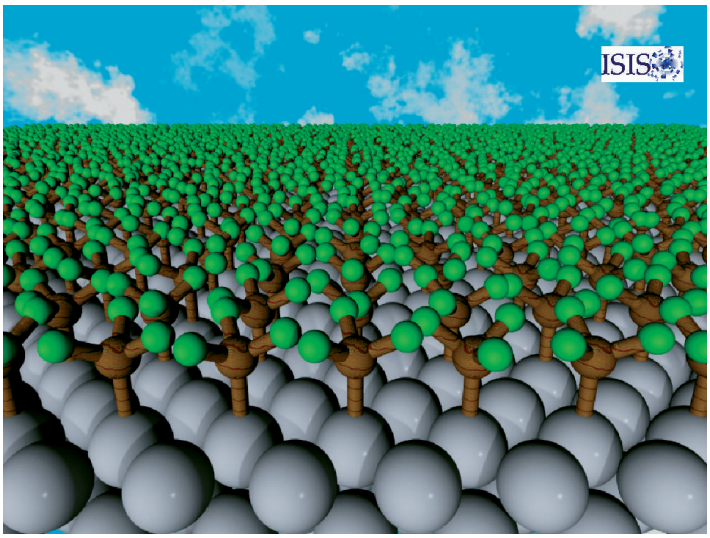

Figure 2: Artist's view of Pd-catalyser poisoning by methyl groups. A "Methyl-wood" is growing.

- A final example deals with recently developed additives for diesel fuel controlling the growth of wax crystals upon cooling. By selfassembly these polymeric additives form small aggregates consisting of a crystalline nucleus with hairs on both sides. These hairs keep the aggregates in suspension.

  When the diesel oil is cooled, the additives function as nucleators for wax crystals and prevent larger crystals from precipitating when the temperature drops. Their discovery, their functional mechanisms and their optimisation were achieved on the basis of small-angle neutron scattering experiments.

- taylored antifreeze for diesel fuel.

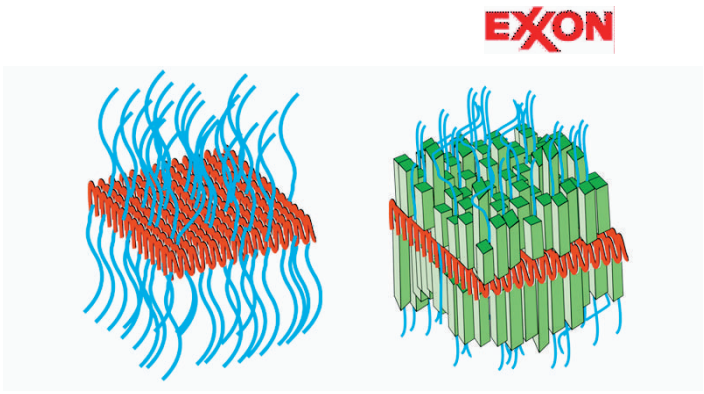

Figure 3: Polymeric templates for wax crystal modification in diesel fuels. The green columns symbolise the growing wax crystals.

Societal needs define foresight and priority missions.

Societal needs are expressed in foresight themes set up by the European governments and priority research missions like those foreseen in the framework six program of the European Union. This chapter is dedicated to evaluate the promises of ESS in the frame of such defined societal needs. Since we cannot predict the future, we ask, where ESS would lead to major advances, if it were available today.

In order to place ESS into the environment of priority research missions, a transdisciplinary approach was mandatory.

Selection of priority missions.

The evaluation process leading to the results presented in this chapter, was performed in several steps involving the ESS-SAC, the disciplinary science groups mentioned in chapter 4 and finally transdisciplinary mission oriented working groups. In a first step ESS-SAC assessed the European priority research themes of the frame work 6 program in relation to the corresponding priority missions established in the different European countries. On that basis general trends were evaluated and the most important topics with the greatest overlap were identified. In each case it was asked how much and how decisive ESS could contribute to these themes. Finally, seven research missions which are widely supported by the EU as well as by many European countries evolved. These priority research themes are:

- Microsystems and Information Technologies
- Functional Materials
- Health and Biotechnology
- Nanotechnologies
- Cultural Heritage: Artefacts and Materials
- Traffic and Transport
- Sustainable Development; Clean Technologies and Environmental Systems

Transdisciplinarity of priority research themes.

The priority research themes in general are transdisciplinary and encompass different scientific disciplines. Consequently in a further step the different disciplinary science groups were asked to assess the contribution of their discipline to the

different priority themes. Two aspects were to be considered:

1. The ESS contribution should overcome existing thresholds. We were looking in particular for ESS experiments which are beyond present capabilities and which will open up new opportunities.

2. These ESS contributions to priority research missions do not necessarily coincide with scientific flagship areas considered in the disciplinary science cases but emphasise quite different aspects: e.g. the development of polymeric detergents, which work in hypercritical CO_2 may be not so much a scientific breakthrough but they will help a lot to establish clean technologies in the dry cleaning business and so on.

Interrelation between curiosity driven and applied research.

Nevertheless there exists a strong interrelation between the more basic science flagship areas worked out in chapter 4 and the priority research themes discussed here. As an example Table 1 displays the connections between the soft matter flagships of chapter 4 and the priority themes. Three stars signify very important, two stars important and one star some relevance of the respective flagship area for a priority theme. Significant cross correlations are observed.

Table 1:
Cross correlations between soft matter flagships and European priority research themes
(***very important, **important, *relevant).

SOFT CONDENSED MATTER — Flagship Areas / European Priority Research Themes	Molecular Rheology	Buried Interfaces	Self-Assembly and Structural Formation	Window to Biology	New Products by External Constraints	Soft-Hard Nanocomposites	Complex Liquids in Porous Media	Probing Molecular Dynamics in Non-Crystalline Matter
Microsystems and Information Technology			***					
Functional Materials	*	***	*	*	*	**	*	*
Health and Biotechnology				***			*	
Nanotechnologies		*	***		*	**		*
Cultural Heritage: Artefacts and Materials	-	-	-	-	-	-	-	-
Traffic and Transport	**	*					*	
Sustainable Development; Clean Technologies and Environm. Systems		*	*					*

Table 2 generalises this approach and displays the transdisciplinary aspect of the ESS contributions the priority to research missions. It presents to what extend the different scientific disciplines, as they will be present at ESS, are involved in the different missions.

Table 2:
Involvement of the scientific disciplines in European priority research missions
(***very important, **medium, *low).

Science Groups / Priority Research Themes	Biology & Biotechnology	Chemical Structure, Kinetics and Dynamics	Earth Science	Liquids & Glasses	Materials Science & Engineering	Soft Condensed Matter	Solid State Physics
Microsystems and Information Technologies		*	*		***	*	**
Functional Materials		**	*	**	***	***	***
Health and Biotechnology	***	*		*	*	*	
Nanotechnologies		*	*	**	***	***	**
Cultural Heritage: Artefacts and Materials		*	***		*		
Traffic and Transport		*		**	***	**	*
Sustainable Development; Clean Technologies and Environmental Systems	*	***	***	**	***	**	*

In a second SAC-ENSA workshop which took place in Dourdan in March 2002 transdisciplinary science groups were convened. Their mission was to arrive at a sound assessment of ESS's impact on the different priority research themes. Their topics and conveners are displayed in Table 3. These working groups operated on the basis of information, mostly in form of examples, which was provided beforehand by the disciplinary science groups. The reports in this chapter present the outcome of these considerations.

Table 3:
Priority research themes and the respective convener/s.

Priority Research Theme	Convener/s	
Microsystems and Information Technologies	H. Zabel C. Vettier C. Fermon	Univ. of Bochum, Germany Institut Laue Langevin, France CEA Saclay, France
Functional Materials	B. Cywinski	Univ. of Leeds, United Kingdom
Health and Biotechnology	J. Helliwell	Univ. of Manchester, United Kingdom
Nanotechnologies	J. Colmenero A.R. Khokhlov	Univ. of the Basque Country & DIPC, Spain Moscow State University, Russia
Cultural Heritage: Artefacts and Materials	R. Rinaldi	Univ. of Perugia, Italy
Traffic and Transport	P. Withers A. Magerl	Univ. of Manchester, United Kingdom Univ. of Erlangen-Nürnberg, Germany
Sustainable Development; Clean Technologies and Environmental Systems	H. Jobic	CNRS/Univ. Lyon 1, France

Neutrons elucidate aspects of the origin of the universe.

Finally, beyond utilitarian aspects, the ESS also has a cultural mission, cultural in the sense that it will help to widen our horizon and our knowledge about where we come from and where we might go. The physics of the neutron itself bears important messages on the origin of the universe the 'big bang' and the fundamental forces of nature. In order to illustrate this aspect, the last report in this chapter gives a short outline of the connection between neutron physics and fundamental questions of cosmology.

6.1 Microsystems and Information Technologies

C. Fermon[1], J.B. Suck[2], B. Toperverg[3], C. Vettier[4], H. Zabel[5]

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[2]Technical University Chemnitz, Institute of Physics, Materials Research and Liquids, D-09107 Chemnitz, Germany
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Neutrons provide crucial knowledge at the atomic and molecular scale on materials used for advanced information technologies (IT). Such materials include optical and optoelectronic devices, materials for microelectronics, for spintronics, for high density storage media, and for fast and non-volatile random access memory. The material analysis with neutrons from a spallation source is essential for technological advances which improve the ease of communication world wide, the speed of information exchange, and the international security standards. The ESS will enable research in areas, which are not accessible at the present time. In particular it will allow:

- Investigations of nano-structured magnetic dot arrays and of buried ultrathin layers;
- Studies of the spin dynamics in molecular clusters and in nano-fabricated magnetic clusters;
- Characterisation of the magnetic roughness at interfaces between different materials;
- Exploration of the magnetic phase diagram of metal alloys for ultrathin films;
- Examination of artificially designed and highly non-linear micro-magnetic media.

The ESS will be the world leading neutron facility for the analysis of the next generation of IT materials and for expanding our knowledge in science and technologies.

I. Introduction

Information technology is not only concerned with software development, but also with data storage and retrieval.

Information technology (IT) is often identified with software development and testing; with high-speed, wireless communication, multimedia, and internet networks; with user interfaces, text retrieval, standards conformance, and with developing cryptographic methods for protecting the integrity, confidentiality, and authenticity of information resources. Another aspect of information technology is the hardware required for information storage, for data recording and reading, for information management, transmission, and display. Advanced materials and novel components are to be developed for future electronic devices, for higher density storage media, and for fast and non-volatile random access memory.

In order to arrive at novel components and microsystems for future information technologies, the fundamental properties of the materials involved have to be explored with many different experimental tools. Neutrons have always played an essential role for the analysis of magnetic materials and for mapping out their phase diagrams. In the future the application of neutron methods to ultrathin magnetic films and heterostructures will be challenged by the size of the samples. Today only laterally extended films can be investigated with neutron methods. However, future technologies demand more complex structures which are laterally confined and which have been nanostructured by lithographic methods. To overcome the difficulties imposed by sample size and weak cross section, a higher intensity neutron source is required. Therefore, the ESS is an essential tool for the understanding of spin structures and fluctuations in confined magnetic systems not achievable with current neutron sources.

II. Past achievements and potentials for neutron applications in the IT sector

Neutrons bear a magnetic moment.

One of the most prominent contributions of neutron research has been the investigation of magnetic materials. Neutrons bear a magnetic moment and thus they represent tiny magnets. Similar to the way a compass needle reacts to the geometry and the strength of the earth magnetic field, the magnetic moment of the neutron has been used with great success to investigate magnetic properties of a large variety of materials on an atomic scale. Thus, our present day knowledge of magnetic structures and magnetic interactions are mainly drawn from neutron scattering experiments. An example is shown in Figure 1: antiferromagnetic manganese-oxide. This was in fact the first antiferromagnetic structure determination via neutron scattering, carried out by the Nobel laureate Cliff Shull. Magnetic materials find applications in a wide range of Industrial products such as electric motors, transducers, magnetic cranes, bending magnets for accelerators, and undulator magnets for synchrotron sources. Another area of applications are magnetic sensors in cars, in particular in ABS systems, in reading heads of hard disks, and positioning sensors for precision cutting tools.

Neutron research has contributed extensively to our understanding of magnetic materials.

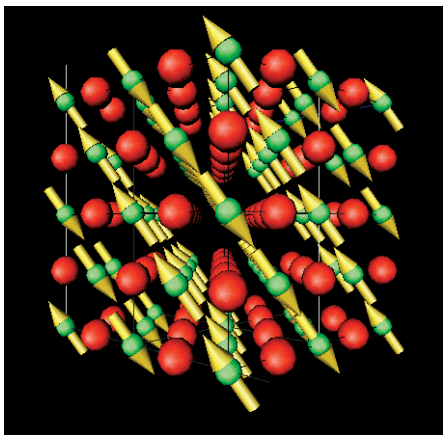

Figure 1: Crystal structure and magnetic structure of manganese-oxide, showing an antiparallel alignment of magnetic moments on alternate manganese lattice planes. Antiferromagnetic crystals are important for producing an exchange bias effect.

Recent neutron research focuses on systems with reduces size of increased complexity.

Recent neutron research of magnetic materials has focused on systems with reduced size such as thin films, magnetic clusters, molecular nano-magnets, and on magnetic heterostructures with increased complexity. These systems find, in part, applications as sensors and in information storage devices. Because of the weak absorption neutrons are also highly sensitive to ultrathin layers buried deep under overlayers, as is often the case in semiconductor hetero-structures. The high transmission power of neutrons is also routinely used for the radiography of devices with highest reliability requirements. As these devices become smaller and smaller, an increased neutron flux is needed for reliability tests of microsystems. Thus, the future challenges of neutron research in the IT sector can only be met with higher intensity

from the ESS. Some recent activities of neutron research in the specified areas are highlighted below to illustrate the point.

Molecular nano-magnets

Molecular nano-magnets are fascinating magnetic clusters as revealed by neutron work.

Molecular nano-magnets constitute one of the possible routes toward pushing down the size limits for information storage because of their small size and large magnetic moment. They consist of an assembly of a small number of paramagnetic ions, each bearing a magnetic moment and a spin (like a spinning top) embedded in large molecules, such as Mn_{12}-acetate. The structure of this molecule is shown in Figure 2. In this system, the arrangement of the manganese atoms in the molecule has been determined using neutron diffraction; the different manganese atoms are coupled by the interaction of the atomic magnets, which leads to a parallel alignment of their magnetic moments. From the magnetic point of view, the 12 manganese atoms can be considered as a single large pseudo-spin system (S = 10). Such molecules with S = 10 behave as super-paramagnetic particles, at the borderline between classical and quantum behaviour. The understanding and exploitation of their properties require the knowledge of their preferred direction within the crystal lattice. Indeed, local interactions between the manganese atoms and the surrounding atoms lead to the occurrence of a set of excited states for the global spin with discrete energy levels. These quantum states correspond to various orientations of the pseudo-spin; depending on the balance between thermal energy or heat and the energy scale of these reorientation states. The pseudo-spin may at low temperatures be frozen into a preferred orientation or it may randomly jump from one direction to another at higher temperatures.

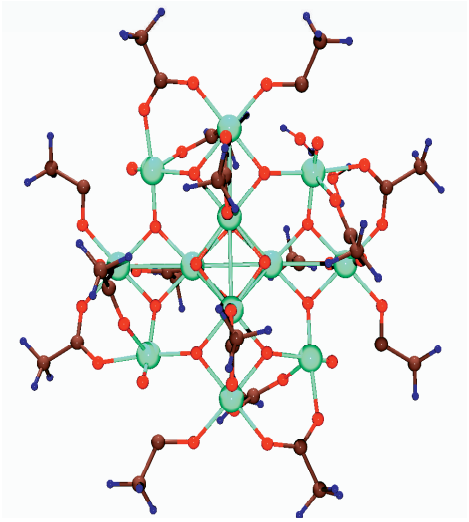

Figure 2: Manganese-acetate forms a large magnet molecule. At very low temperatures the magnetic moments of the 12 manganese atoms (shown in cyan) align in parallel yielding a large moment. Neutron research has revealed the ground state of this delicate magnetic system and its excitations. The search is on for magnetic molecules which sustain higher temperatures and exhibit a high moment at room temperature for information storage.

Neutron spectroscopy

Neutron spectroscopy is the only tool, which allows the full determination of these magnetic levels. In the case of Mn_{12}-acetate, it was found that the energy barrier preventing an orientational change of the magnetisation is only about 10 K. This is obviously too low for practical applications and the search is on for higher energy barriers, ideally near room temperature. This can be achieved by changing the chemical environment of the transition metal atoms in order to increase the interactions. Therefore, the combination of chemistry and neutron scattering is the key to the optimisation of such nano-magnets. More powerful neutron sources such as the ESS will make it possible to investigate the properties of these molecular nano-magnets arranged on various substrates in order to form two- or even one-dimensional arrays.

The combination of chemistry and neutron scattering methods is the key to optimised nano-magnets.

The Giant Magnetoresistance (GMR)

The Giant Magnetoresistance (GMR) was found only 15 years ago and applications can today be found in reading heads of hard disks, as position sensors in precision tools, and in ABS systems. The GMR effect requires a stack of at least three layers, a non-magnetic metal layer sandwiched between two ferromagnetic layers, as schematically shown in Figure 3.

Giant magneto-resistance and exchange bias effect are at the basis of smart micro-magnetic media.

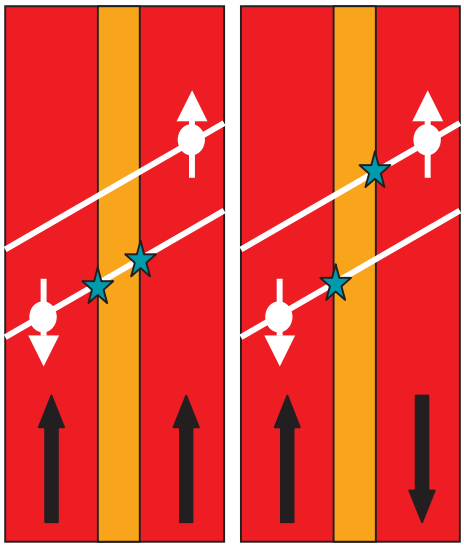

Figure 3: Transport of spin polarised electrons through a trilayer is shown, which consists of two ferromagnetic sheets sandwiching a non-magnetic metal. For parallel alignment of the ferromagnetic films (black arrows in left panel) one spin state of the electrons (up) can travel through the trilayer, whereas the other one (down) is blocked. In case of an antiparallel alignment of the ferromagnetic films both spin states of the electrons are scattered strongly at the respective interfaces with opposite spin directions, providing a higher resistance than in the parallel state. The change of the resistance with the orientation of the ferromagnetic layers is called ‘giant magneto-resistance’ (GMR).

Electrons, which travel in this stack, sense with their magnetic moment (spin) whether the magnetisation directions of the ferromagnetic layers are parallel or antiparallel. For parallel

alignment the resistance is low and for antiparallel alignment it is high. In zero field the GMR sensor has a high resistance due to the antiparallel orientation of the ferromagnetic layers. With increasing field both layers rotate into the field direction thereby lowering their resistance.

The Exchange Bias (EB) effect

The exchange bias effect relies on the interaction between ferromagnetic and antiferromagnetic layers across their common interface.

The Exchange Bias (EB) effect, discovered in the middle of last century, is an additional effect, which in combination with the GMR effect has revolutionised the IT developments in recent years. The EB effect requires the interaction between a ferromagnetic and an antiferromagnetic layer across their common interface, resulting in a characteristic shift of the magnetic hysteresis. The EB effect is used for pinning the magnetisation direction of one ferromagnetic layer in a preferred direction, while the second ferromagnetic layer can rotate freely in response to an external field. Thus the basic principles of the GMR and the EB effects can be combined in so-called spin valve structures for controlling the magneto-resistance. A spin valve structure is schematically shown in Figure 4. Depending on the relative orientation of the magnetisation in the top and bottom ferromagnetic layers, the spin current can be switched from high to low, denoting the logic “0” and “1”. Arrays of spin-valve structures are required for building the next generation of high performance non-volatile magnetic random access memory (MRAM) devices. The first prototypes have already been presented. Neutron scattering is essential for analysing and understanding the magnetic structure in spin valve systems. Unlike most other methods, neutrons yield detailed information on the relative orientation of different ferromagnetic layers deep inside of magnetoelectronic devices, such as the MRAM.

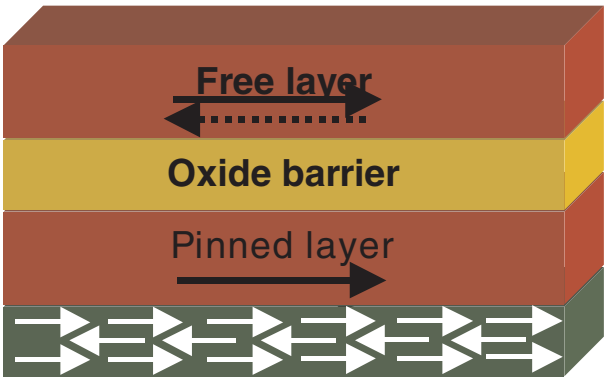

Figure 4: The magnetisation of a ferromagnetic layer in contact with an antiferromagnetic layer becomes pinned in a preferred orientation, while the second layer is free to rotate in an external magnetic field. Electrons, which travel through such a trilayer are sensitive to the relative orientation of the ferromagnetic layers. These magnetic heterostructures are called spin valves since the resistance can be changed similar to a valve.

In spite of the obvious success of GMR sensors and spin-valve structures, a thorough theoretical understanding is still lagging behind. The solution of this complex problem requires the joint use of reliable quantitative tools available through modern experimental methods. Among them neutrons play a pivotal role with the ability to shed light on the most cumbersome questions on magnetic arrangement on the micro- and mesoscopic scale. Experiments with polarised neutrons (at the Institut Laue-Langevin in Grenoble) have allowed to reconstruct three-dimensional images of ferro- and antiferromagnetic ordering and to follow an evolution of the submicrometer domain structure under applied magnetic field. This is a very essential step forward in understanding the GMR and EB effects and for providing the necessary scientific background for future magnetoelectronic devices.

Neutron scattering elucidates the functioning of spin values in MRAM devices.

Three-dimensional images of ferro- and antiferromagnetic ordering reveal the domain structure of complex interfaces.

Smart micromagnetic media

Smart micromagnetic media may be visualised by novel and innovative configurations of micromagnetic elements, perhaps forming a prototype of magneto-neural networks, as indicated schematically in Figure 5. Then one will be faced with problems of stable configurations, excitations, propagation of signals, intellectual capacity and other inherent features of such systems. They cannot be well predicted at present. However, in contrast to natural neural networks, the structural and dynamical properties of artificial magnetic micro-media can be studied with neutrons at sufficiently intense neutron sources, such as the ESS. In the past neutron research has helped substantially to unravel the basic physical principles of spin-glasses and random field problems, thereby pushing forward the knowledge in the field of neural networks. It is expected that neutron investigations of artificially designed and highly non-linear micro-magnetic media may again be become one of the strategic scientific directions in the forthcoming decade.

ESS will clarify structural and dynamical properties of artificial micro-magnetic media.

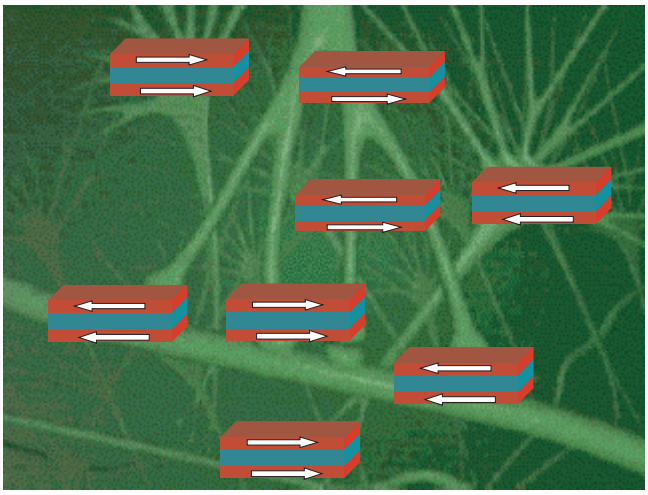

Figure 5: Smart magnetic media may consist of an array of coupled GMR elements, which form an artificial network similar to natural neural networks.

III. Future developments of microsystems and information technologies

Structural and dynamic investigations of magnetic ultrathin films, heterostructures and nanostructured media require the flux of ESS.

One of the most important advantages of neutron scattering is the well understood cross section for neutrons with the material to be studied. This is true for non-magnetic as well as for magnetic materials. Furthermore, the interaction between neutrons and the sample is weak, allowing a data analysis via simple and efficient approximations. The weak interaction is usually compensated for by a reasonable large sample. However, advancements of thin film technologies and IT components demand the investigation of samples with much reduced dimensions. For instance, the magnetic structure investigation of a 2 nm thick Iron film requires counting times of about 24 hours at the most powerful neutron sources available today. If the counting times become too long, background intensity builds up and the quality of the data is no more sufficient to extract meaningful information. Furthermore, investigations of dynamical properties such as spin waves and fast magnetic switching in nanostructured films are out of reach with present day neutron sources and instrumentation. Therefore for structural and dynamical investigations of ultrathin films, heterostructures, magnetic dot arrays (an example is shown in Figure 6), and systems in nanostructured media the neutron flux from the ESS will be essential and is not achievable otherwise.

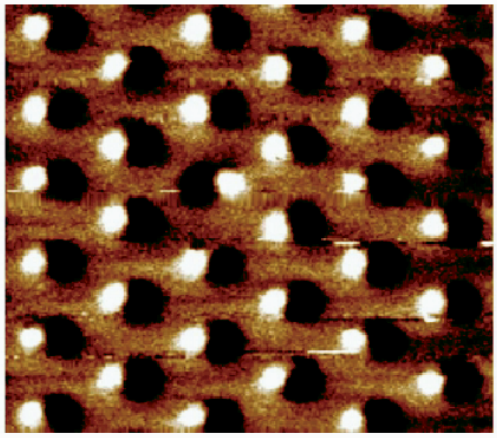

Figure 6: Shown is an array of magnetic islands on a substrate. The islands were imaged with a magnetic force microscope showing that they are aligned in the same direction like an array of compass needles on a nanometer scale. The magnetisation reversal and the internal dynamics of the magnetic moments within the islands needs to be investigated by neutron spectroscopy, which is not possible with currently available neutron sources.

Ultrathin oxide layers for microelectronics and spin electronics

Neutrons shed light on hidden oxide layers semiconductor devices.

Miniaturisation of components is one of the main challenges in microelectronics. Transistors should be smaller for higher integration and for speed enhancement. Oxide layers with a thickness of only 4 atoms should be built for field effective transistors with an extremely high reproducibility on large surfaces. Thickness variations over macroscopic distances are not tolerable since they deteriorate functionality and reproducibility. The characterisation of these very thin layers

by non-destructive methods is impossible up to now when they are deeply buried in more complex systems. Neutrons appear to be the best tool to study this kind of buried barrier due to their deep penetration even at grazing incidence (almost parallel to the layer) and due to their sensitivity to interface changes. With a high flux source like ESS it would be possible to measure the spatial distribution of the thickness and chemical composition of these ultrathin layers on large wafers within a couple of minutes.

Dynamics of thin films, wires and arrays of dots

Technically used frequencies are no longer far away from internal spin dynamics. Neutrons have to show the limits.

Understanding fast dynamics in small systems now becomes essential because the calculation and the transmission rate in computers have exceed the threshold of 1 GHz. At these high rates, the response of materials becomes local and has to be observed by local probes sensitive to the dynamics. Neutrons are by far the best probe for studying the dynamics of solids. But for intensity reasons it has up to now only been applied to bulk systems. One of the main advantages of the ESS will be the application of this probe to micro- and nanoscale systems. For instance, piezo-electric and ferroelectric thin films are used for strain- and deformation sensors and are being developed for non-volatile data storage (ferroelectric memories). Knowledge on the vibrational properties of these devices, retrieved from inelastic neutron scattering experiments, is required in order to determine their response even for picosecond excitations. Another example are magnetic storage and spin electronic devices, where the fast switching is limited by magnetic vibrations (spin waves). In fact the magnetisation reversal is mingled together with the inherent spin waves of the magnetic structure. In case of submicron magnetic dots as shown in Figure 6, these vibrations can take only discrete values. Complementary experimental techniques available today for the study of these excitations are strongly limited as concerns their frequency range and their probing depth beneath protective covers. The development of neutron scattering for the investigation of small system is therefore of utmost importance.

Hard magnets for microsystems

Hard magnets find applications in micro-electronics and microsystems.

Integration of efficient micromotors, microgenerators, micro-switches, and micro-sensors is important for a large number of applications, such as mobile phones, battery free watches, medical pumps, etc. For example, the watch industry aims at a wrist watch built on one chip, which contains a micro-sized power generator via the arm movement, an energy storage cell, and a micro-motor for driving the small and large hands, as well as a magnetic sensor for feed back and control. The battery driven quartz watch as shown in Figure 7 will be outdated soon.

Today, a main limitation for the application of magnetic microsystems is the implementation of hard magnets into thin films. This includes the development of low temperature deposition techniques for hard magnetic films and for their

integration in submicron electronic circuitry. Hard magnets with perpendicular anisotropy are also required for the injection of spins in semiconductors for spin-sensitive electronic devices. In all these cases, neutron scattering is the method of choice, because it gives the most detailed and acute information on magnetic materials. However, as the characteristic length scales shrink, it is absolutely necessary to compensate the reduced scattering volume by an increased neutron flux, which is only possible through the ESS. Furthermore, through the development of a new generation of instruments, magnetic structures of patterned hard magnets on substrates will be possible, giving a key tool for process optimisation.

Figure 7: In all quartz watches integrated circuits are used to make the quartz crystal oscillate, to divide the quartz frequency down to one pulse per second, and to drive the display. In the future the battery will be replaced by a micro sized power generator and an energy storage cell, a micro motor for driving the small and large hands, as well as a magnetic sensor for feed back and control. Advanced deposition techniques for hard magnetic films integrated in submicron electronic circuitry in combination with neutron scattering for the investigation of magnetic structures and interfaces are necessary to invent the next generation wrist watches.

Magnetic roughness of interfaces

Magnetic roughness: a barrier to overcome in efficient spin and electron transport.

The performance of magnetoelectronic and spin-electronic devices is hampered by the presence of structural and magnetic roughness at the interface separating two different materials. For the transport of electrons interfacial roughness leads to scattering, resulting in an increase of the electrical resistance. In addition, upon scattering on magnetic imperfections the electron spin may flip, reducing the spin transport from one material to the next. The structural roughness of interfaces is well understood and can nowadays be characterised on an atomic scale. However, the magnetic roughness, which may consist of spin disorder or magnetic domain walls is much more complex to analyse.

Interdiffusion of two different materials at the interface is usually considered as a gentle type of imperfection. However, if two different types of magnetic materials are involved, this may lead to a strong spin frustration. A striking example is

shown in Figure 8. Magnetic roughness is detrimental to polarisation of electrons close to an interface and to the transport of spin polarised current across an interface. It may lower the performance of magnetoelectronic devices similar to a rough surface, which lowers the reflectance of light. Neutron reflectivity is a highly sensitive probe for the analysis of magnetically rough interfaces, because the neutron spin will flip at these interfaces in a similar fashion as the electron spin does. Because of the weak cross section, the neutron spin flip scattering is, however, easier to analyse than electron spin flip scattering and therefore quantitative tests of theoretical predictions become feasible. Advancements in thin film deposition techniques rely on experimental parameters in the quest to improve the interfacial roughness. With decreasing lateral size of magnetic nanostructures, the structural and magnetic roughness of interfaces needs to be investigated with higher flux neutron sources. In order to meet this challenge, the neutron flux from ESS is required.

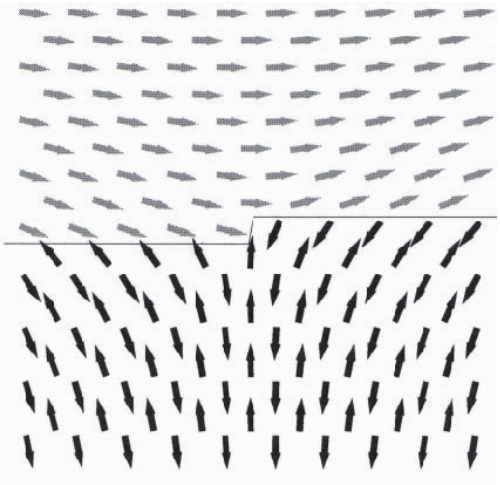

Figure 8: Magnetic roughness is an important issue in magnetoelectronics. A single step at the interface between a ferro- and anti-ferromagnetic material can completely change the orientation of the magnetic moments (arrows) close to the interface and change their functionality. Shown is a model calculation of the spin structure in iron (grey arrows) and in chromium. Without the step the arrows would align horizontally. With the step the spin structure becomes twisted.

Magnetic phase diagrams

Magnetic phase diagrams of ferromagnetic alloys look different in bulk and in thin films. Neutrons research helps to establish them for future applications.

The development of engineering magnets rests on the knowledge about magnetic phase diagrams of binary and ternary alloys, which may serve as a tool box. Phase diagrams display all possible structural and magnetic phases as a function of alloy concentration, including their local magnetic moments and their transition temperatures. Magnetic phase diagrams of bulk alloys are accessible through handbooks and data banks. Neutron research had a huge impact on the compilation of this body of work. In thin magnetic films the phases diagrams are altered due to strain, structure, and finite size effects. The phase boundaries are usually shifted and often magnetic phases can be stabilised which would not exist in the bulk. An example for the FeNi phase diagram is shown

in Figure 9. The essential parameters of thin film magnetic phase diagrams are the film thickness, the interaction with the substrate, and the residual stress. Neutron scattering is the best tool for providing magnetic moments of thin films on an absolute scale. In contrast to macroscopic techniques such as SQUID magnetometry, neutron scattering does not average over the whole thickness of a film. Instead it reveals the magnetisation profile in the direction perpendicular to the film and is therefore sensitive to rough or non-magnetic interfaces. Neutron research was of highest importance for the results presented in Figure 9. As the thickness of magnetic alloy films used in magnetoelectronic devices shrinks, there is a strong demand for analysing their magnetic phase diagram. At the same time, the thinner samples require a higher flux. Therefore this task can only adequately be met by the next generation high flux neutron source such as the ESS.

Magnetic phase diagrams are needed for applications in magneto-electronic devices.

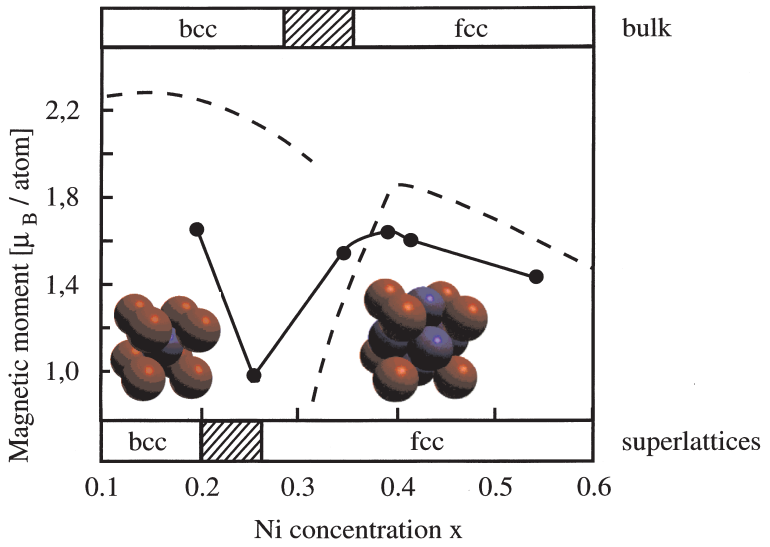

Figure 9: The structural and magnetic phase diagram of metal alloys is different in the bulk and in thin films. Shown is an example for the phase diagram of the Iron-Nickel alloy. In the bulk a structural phase transition from a body centred cubic (bcc) to a face centred cubic (fcc) structure occurs at an alloy composition of 30 % Ni and 70 % Fe. The transition is accompanied by a severe loss of magnetic moment. In thin films the phase boundary is shifted to lower Nickel concentrations. Depending on the thickness and the substrate the phase transition can be completely suppressed. Determining the phase diagram of magnetic alloys is an important task for the benefit of future IT developments. A very high neutron flux from the ESS is required to meet this challenge.

IV. Instrumentation requirements at the ESS

In order to develop the crucial knowledge on the atomic and molecular scale on magnetic nanostructures and micro-systems for future information technologies, the following instrument suite is required:

Instrument suite for the investigation of magnetic nanostructures and microsystems.

- Polarised neutron reflectometer with high resolution and with high intensity for the analysis of magnetic nano-structures and nanopatterns;

- High resolution and focussing small angle scattering under conditions of total reflection (Ref-SANS) for in-situ studies of magnetisation reversal processes of magnetic dot arrays;
- Variable resolution cold chopper spectrometer for the analysis of magnetic excitations in magnetic clusters;
- Diffuse scattering diffractometer with full polarisation analysis for the investigation of disordered and hard magnetic materials.

6.2 Functional Materials

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Abstract
Functional materials underpin society. Developing and improving these materials in a directed and systematic fashion is entirely dependent upon gaining a deep understanding of their key fundamental properties at the atomic and molecular level. Neutron scattering is already showing the way not only to the optimisation of functional properties of materials but also to the optimal processing of the materials themselves. The remarkable capabilities of the ESS to provide substantially higher intensities and greater resolution will not only accelerate this process but also launch new strategies based upon systematic multi-parametric studies and, for the first time, upon characterisation of dynamic and diffusional properties in real time processing.

I. Introduction

Functional materials shape our technological society.

Our high technology society is entirely dependent upon materials that have been developed and optimised, sometimes incrementally over many years, for specific functions. Indeed for most of man's history functional materials have been discovered quite by chance, and slowly improved by trial and error in a laborious and evolutionary fashion.

Neutrons are ideal for studying phenomena deep inside large technological systems.

However, as technology advances at an ever greater pace this serendipitous approach to discovering and developing material functionality becomes increasingly inefficient. The growing complexity and diversity of both the materials and their functionality demands a more systematic and parametric approach. This in turn demands even more powerful and precise methods for characterising and optimising structure-function relations, of finding where atoms and molecules are so that key substitutions can be made, for studying and understanding the dynamical processes that often define the material's properties, and for developing new processing routes to novel materials and economical processing routes to existing materials.

ESS will allow these studies to be extended to multi-parameter space.

Neutron scattering is playing an increasingly important role in this crucial area of technology and there is no doubt that the ESS will make a major and revolutionary contribution. Neutrons are a very penetrating probe with which the structure and dynamics of materials can be studied on technological length scales under real working conditions. The ESS will enable us to realise the full potential of such studies, allowing detailed systematic and parametric investigations of material properties as combined functions of composition, temperature, pressure and magnetic field. Such multi-parametric studies, which will be possible on time scales that cannot be realised at present, will lead to a more

Faster processing routes to better materials are promised.

comprehensive understanding and exploitation of the physical and chemical processes that underpin material functionality.

Even a cursory examination of those scientific and technological journals devoted to “functional materials” will reveal that the term is almost all encompassing. The definition “functional materials” can include superconductors or battery materials, nanostructured materials and quantum dots or giant magnetoresistive multilayers, biomimetic and biocompatible systems or nimonic alloys and so on. The list of advanced materials and their functions is almost endless. However many of these functional materials have been adequately addressed under other sections of this report. Here we shall consider a few specific, but extremely important, examples, focussing upon both their preparation and characterisation.

II. Materials processing and beyond

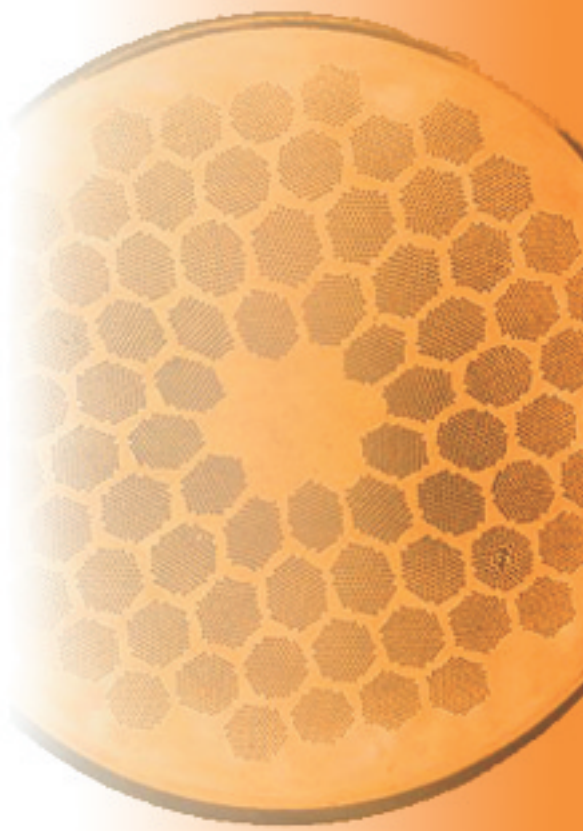
Development of an atomic understanding of materials processing.

The production of functional materials may be relatively simple involving, for example, the co-melting of the constituents of a metallic alloy, or it may be extremely complex, as is the case with biomimetic self-organisation. In both cases, however, the processing routes to high performance functional materials are often imprecise and routes which are apparently optimal have often been obtained through trial and error, and do not necessarily reflect the most efficient or cost effective methods of preparation.

Neutron diffraction can be used to monitor processing in-beam and under extremes of temperature or pressure, on samples of a technologically significant size, thereby allowing the efficacy and efficiency of accepted processing routes to be investigated.

One particular example of great commercial significance is that of the processing of superconducting Nb_3Sn wires for high field superconducting magnet production. The process involves first swaging bundled Nb and Sn filaments in a copper matrix, then winding the magnet coils and finally annealing the magnetic coils according to a predetermined profile to achieve the appropriate Nb_3Sn superconducting phase. The precise annealing profiles are not well known, and failure to complete adequately the process leads directly to failure of the magnet coil.

Neutron diffraction studies have already provided the first direct insights into the transformation process in material of commercial volume (Figure 1), and call into question the validity of long-accepted processing procedures. Extension of these techniques from conventional superconductors to high temperature superconducting cuprate cables is of enormous technological and economic importance. For example, the development and application of such high-T_c power cables in the United States is prompted by estimated savings of $8B per annum.



Complex routes to new functional materials.

Whilst the essential structural transformations during processing may already, in some cases, be studied using the most advanced neutron instrumentation currently available, the ESS affords not only greater intensity but also higher resolution which will additionally permit significantly more complex multi-phase materials to be studied and allowing the evolution with time and temperature of crucial microstructural parameters to be extracted.

In addition the ESS offers a new dimension to such studies, the prospect of monitoring atomic diffusion through quasielastic studies carried out during in-situ processing. Faster processing routes to better materials are promised, not just in the case of the multi-million euro superconductor industry, but also for the production of many other functional materials ranging from steels to dental implants, and multilayer metallic alloys to nanowires.

Similar parametric processing also has direct relevance to the preparation of novel functional materials under extremes of pressure and temperature. Such materials include the newly discovered polymerised fullerenes, super-hard solid CO_2, new sintered ceramics, carbides, molecular sieves, bulk amorphous metallic alloys and zeolites.

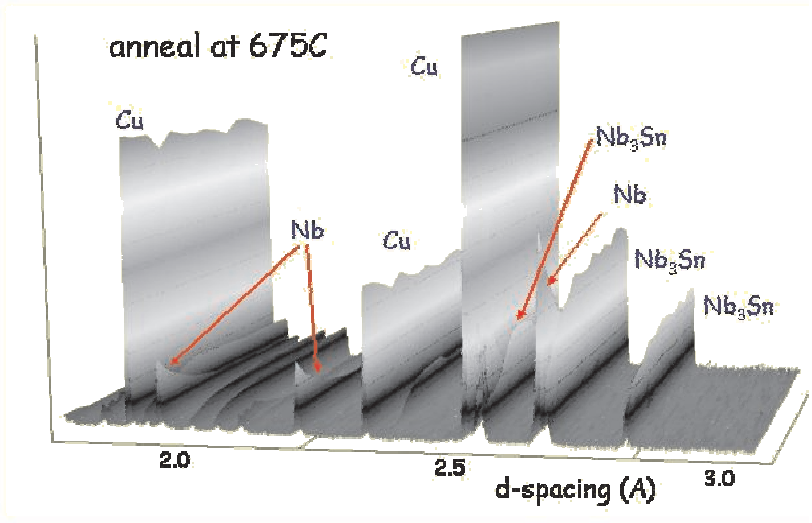


Figure 1: The in-beam processing of commercial modified jelly-roll Nb_3Sn superconducting wires showing the transformation from precursor to final product.

III. From fundamental properties to applied functionality

Achieving an understanding of the mechanisms underlying functionality in materials.

The strongly correlated behaviour, competing electronic interactions, and quantum and classical phase transitions that underpin a great deal of complex physical phenomena, and fascinate the fundamental solid state physicists, also lead to complex functional properties.

In this context the materials that have perhaps been most widely studied by neutron scattering over the last decade fall into this class. They are the high temperature superconducting cuprates and the colossal magnetoresistive (CMR) materials. Whilst the former materials will revolutionise power transmission, ultra-fast electronic circuitry, levitating transport systems, and MRI by offering non-dissipative current transport and magnetic flux exclusion, the CMR materials exhibit remarkably large changes in electrical resistance simply by fine tuning an external applied magnetic field, thereby lending themselves to applications in ultra-sensitive sensors and recording devices.

High T_c superconductors and CMR manganites as the paradigm for emerging strategies in the development of functional materials.

Numerous experimental techniques have been brought to bear on these materials, including resonant and non-resonant x-ray scattering, electron microscopy, muon spin resonance and a variety of magnetic, optical and transport measurement, in a remarkable example of multi-disciplinary synergy. However, it is not an exaggeration to claim that a large part of our current understanding has been provided by neutron diffraction and inelastic neutron scattering.

It is precisely such understanding that is the key to new and emerging strategies in the discovery and optimisation of new functional materials, strategies which could not be further removed from the traditional serendipitous approach to materials development. The high T_c cuprate superconductors and the CMR manganites have become the paradigm for these new strategies.

Thanks to neutron scattering we are beginning to understand the behaviour of these important functional materials, and many others, at the atomic level. As a result, when our understanding is complete it may be possible to deconstruct these materials and then reconstruct them to provide new or improved functionalities.

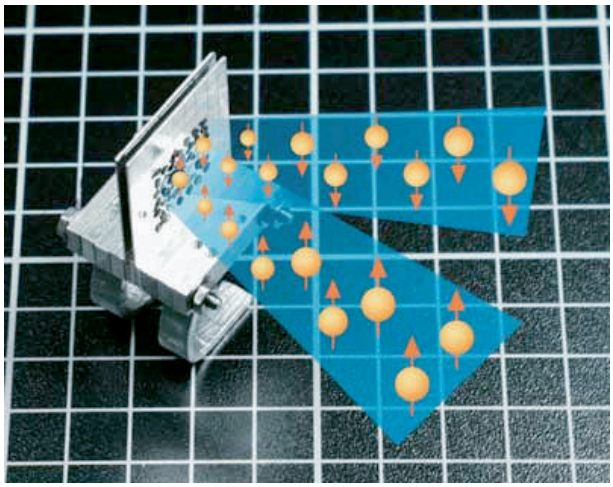


Figure 2: Spin-flip neutron scattering is a powerful probe of the ground states and excitations of correlated electron materials such as high temperature superconductors and magnetoresistive oxides.

However a complete understanding can be achieved only by fully exploring the fundamental structures and excitations of such materials in multi-parameter (electric and magnetic field, pressure, temperature and composition) space. A full mapping of multi-parameter space is, however, well beyond the capabilities of even the latest and best-optimised instrumentation at ISIS and the ILL.

The exploration of structural and dynamic materials properties in multiparameter space is essential.

It is inevitable that many new materials with new functionalities, perhaps arising from their proximity to a classical or a quantum phase transition, will be discovered. It will be not just desirable but essential to study their fundamental properties in extended multiparameter space in order to enhance and exploit their functionality. It is therefore equally inevitably that it will be the ESS which will provide the cutting edge facilities to perform such cutting edge science.

IV. Molecular electronics

The systematic and parametric approach to materials characterisation that has proved crucial to our understanding of "hard" functional materials such as the superconductors and responsive magnetic oxides is equally important for the rapidly increasing number of functions "soft" materials based upon molecular, organic and polymeric architectures.

Molecular electronics.

One such exciting new field is that of molecular electronics, which has been opened through the use of organic compounds as active materials in field effect transistors. In the past few years, aromatic molecules such as pentacene have been shown to have carrier mobilities almost as high as for silicon. Superconductivity has been observed at 117 K in C_{60} co-crystallised with $CHBr_3$.

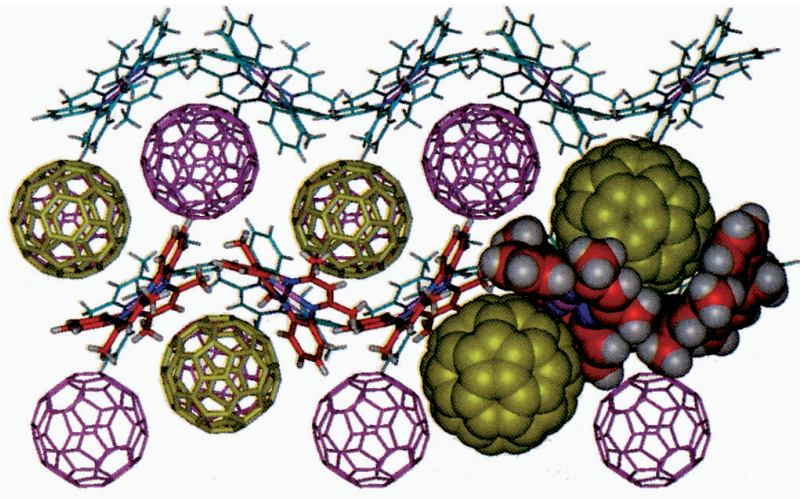


Figure 3: Attractive C_{60} supramolecular architectures with potential applications in micromolecular electronics.

Since doping of these organic materials is achieved by charge injection in an electric field, a proper understanding of the

carrier transport mechanism requires measurements of the dynamical properties (phonons) of thin molecular films between electrodes, a task that demands considerably higher neutron fluxes than are available today.

There are many other related supramolecular arrays, particularly those based upon C_{60}, which provide structural frameworks, morphologies and electronic properties that may well lend themselves to developments in advanced micromolecular electronic applications. However as complexity increases, the demands placed on the methods of structural and excitonic characterisation rapidly grow to the point of exceeding present day capabilities. Here the ESS will play an immeasurably important role.

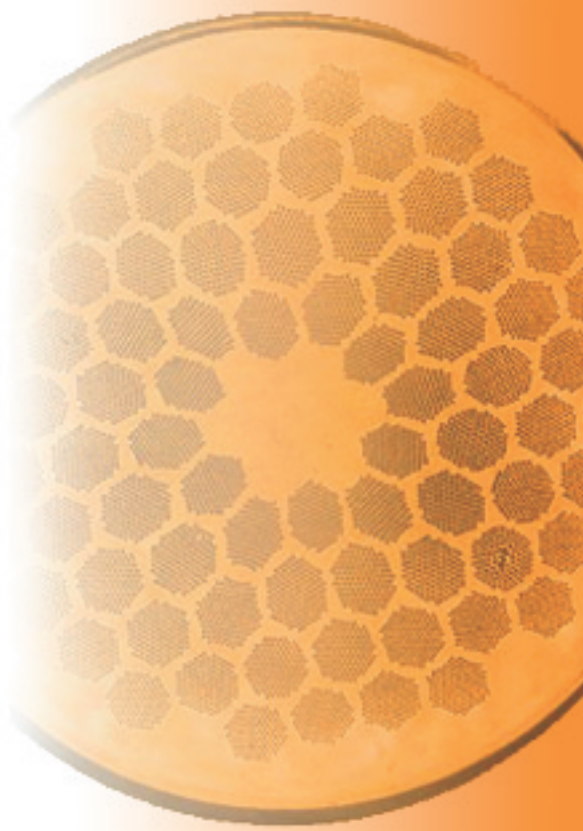
V. Magnetic elastomers

Smart materials.

A particularly important focus of fundamental and technological research is the study of "smart" or “intelligent” soft materials, i.e. those materials capable of following a small change of external conditions in a predetermined way. A new generation of such materials has been obtained using novel composites consisting of small magnetic particles, usually in the nanometer range, dispersed in a highly elastic polymeric matrix. Combination of magnetic and elastic properties leads to the unique ability of such materials to change their shape and mechanical properties in an applied magnetic field in a reversible manner. Since magnetic field is a convenient stimulus in terms of signal control, it is of great importance to develop and study such flexible polymeric systems with potential applications in mind. Giant deformational effects, high elasticity and a quick response to magnetic fields open new opportunities for using such materials in a wide range of applications, including magnetic coupling, peristaltic pumps, manipulators and "artificial muscles".

The magnetoelastic behaviour of composites depends upon the distribution of magnetic particles within the matrix. In uniform magnetic field the particles form chain like structures. This structure can be fixed by the chemical procedure and the resulting magnetic composite becomes anisotropic in terms of mechanical and magnetic properties.

The optimisation of the properties of magnetic elastomers requires microscopic understanding of the coupling of magnetic nanoparticles to the polymer matrix. Neutron scattering is an efficient tool for such studies, because of the excellent contrast of the hydrogen-rich polymer matrix and the hydrogen-free magnetic particles. The ESS will enable the study of not only the net effects of magnetic field on the distribution of magnetic particles within the composite material, but also of the microscopic dynamics of the response of the magnetic elastomer. This will provide tools for the design of new material structures and methods of synthesis which are optimum for a given application.



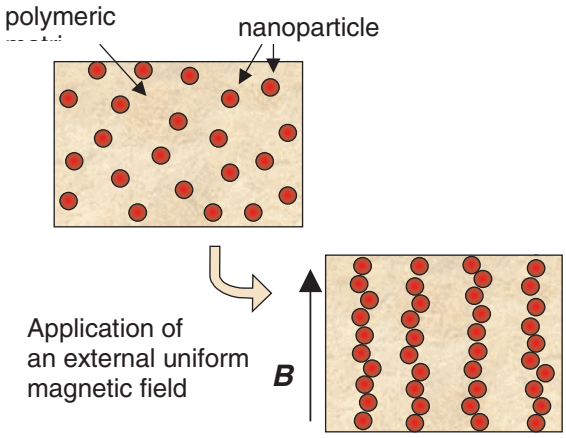


Figure 4: A schematic representation of the effect of an external magnetic field on the distribution of magnetic particles within composite materials.

VI. Photoresponsive Materials

Functional polymers.

Photoresponsive materials change their properties as a reaction to incident light. Important examples are glasses which change their transparency as a function of illumination intensities, and polyelectrolyte gels which undergo a collapse transition under the influence of electromagnetic radiation of certain wavelengths.

Recently, a novel type of photoresponsive materials for optical data recording, processing and display has been designed. These materials are based on comblike liquid-crystalline (LC) polymers with mesogenic and photosensitive-photochromic side groups undergoing pronounced photo-induced structural rearrangements. The photosensitive LC polymers can form stable films, fibers, and coatings with the local properties controlled by optical methods.

Holographic recording devices.

One application of such novel photosensitive materials is in 3-D holographic laser disks. The estimated capacity of such disks is about 1000 GB, 1500 times the average value for compact disks currently in use.

The microscopic processes which define the remarkable properties of the new photosensitive materials, under considerable scrutiny. Neutron scattering is already an essential tool in the study of such materials, but the additional possibility of structural and dynamical neutron studies at extremely high resolution will help to direct a systematic search for new optimum formulations meeting the demands of a wide variety of practical applications of these LC photosensitive polymers.

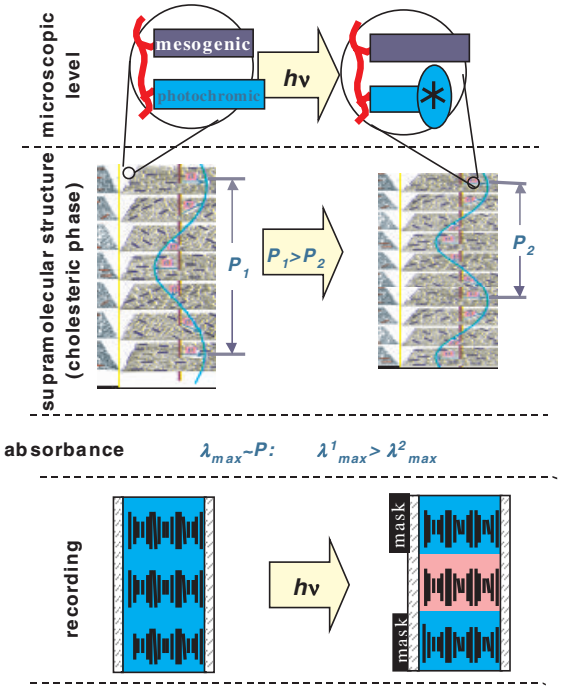


Figure 5: Scheme illustrating the change of macroscopic optical properties of planar-oriented film of a photochromic cholesteric LC copolymer containing both mesogenic and chiral photochromic side groups. Under the action of UV-radiation photochromic side groups of the polymer undergo isomerization transition, which lead to the change of pitch P of the helical supramolecular structure of the polymer. Since the wavelength max of the maximum absorbance is directly proportional to the pitch P it is possible to design an optical recording process with the local optical properties of the polymer film controlled by light irradiation.

VII. Towards increasing complexity – functional machines

From simple materials to the extremes of complexity.

The first functional materials were simple elements and alloys Copper, iron, bronze, and organic materials such as wool and wood. Industrialisation and developing technology brought with it the need for more specialised complex materials, catalysists, superconductors, magnetic materials and semiconductors. These are all largely single component and used in relatively simple structural form, but we are now seeing the growth in applications of advanced multi-component materials, such as the molecular electronic devices, magnetic elastomers and photoresponsive polymers discussed above. As advanced as these materials might be, it is likely that the future will rest with the elegant “Functional machines” constructed as complex multi-component assemblies, whose individual components work together to perform predetermined functions.

Real molecular motors and machines.

A particular example the biological motor F1-ATPase which may be integrated with nano-electro-mechanical systems to create a new class of nano-scale hybrid functional machine. ATPase is used by mitochondria to synthesise ATP from ADP, phosphate, and proton gradients. ATP, is the primary

energy source of our bodies. During synthesis The F1 portion of ATPase has a sub-unit that turns with a rotation that can be reversed by separating the F1 sub-unit from the rest of the protein and feeding the sub-unit ATP. F1-ATPase can generate > 100 pN, has a measured rotational velocity of 3 r.p.s. under load, and a diameter of less than 12 nm, suggesting that F1-ATPase could manipulate currently manufacturable nanomechanical structures. Since the human body produces large quantities of ATP, NASA has suggested that an implantable sensor could be placed in an astronaut's body operated by F1-ATPase requiring no other power source, providing medically important data on an astronaut's health indefinitely.

Self-powering sensors.

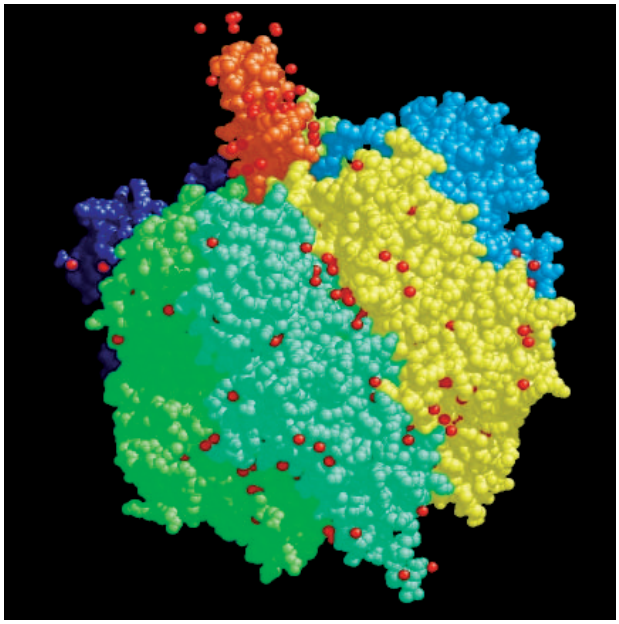


Figure 6: F_1-ATPase in which the orange sub-unit rotates. The remaining six sub-units oscillate sequentially as ATP is hydrolysed.

The design and synthesis of building blocks that can assemble as components to form such real working functional units requires a deep underlying understanding of molecular sub-structures and interactions, both at the surface and in the bulk. However the rewards for such understanding, which can be provided by advanced neutron scattering techniques, are enormous. In the shorter term it is expected that controlled transport across membranes, sensing of analytes, photosynthetic devices and coatings, and responsive display systems are al achievable. In the longer term, microstructured chemical “factories”, pumps and actuators complete with cog-like gearing mechanisms and functions will be possible.

Molecular cogs and gears.

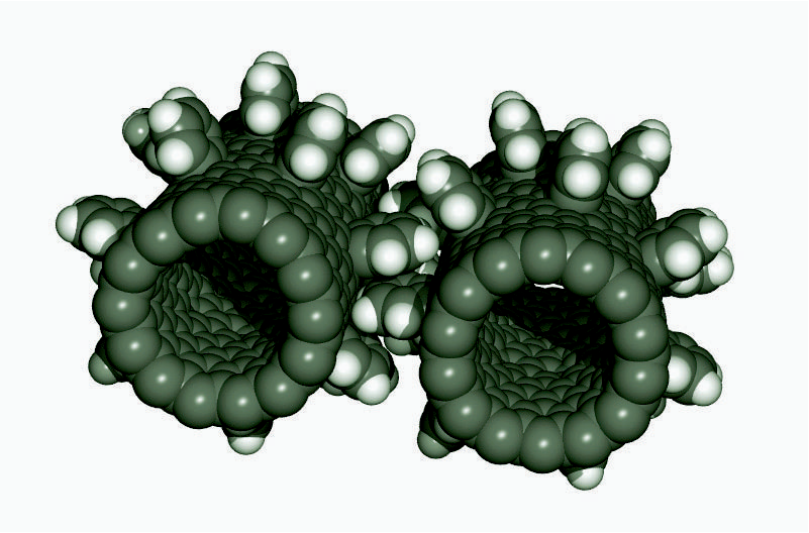


Figure 7: Towards real molecular motors - Molecular cog-and-wheel structures.

VIII. Instrument requirements in the field of functional materials

The need for a more detailed microscopic understanding of advanced functional materials grows with their complexity and functions. The study of fundamental physical properties and processing methods requires primarily sophisticated structure determining techniques, like a high intensity SANS instrument and a high resolution powder diffractometer but also magnetic structure determination will be important. With increasing complexity the demands placed on methods to study dynamics rapidly grow, requiring the full spectrum of high intensity inelastic spectrometers, covering the energy range from a high energy and cold chopper instrument to a high resolution NSE spectrometer.

6.3 Health and Biotechnology

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Abstract
The life sciences revolution, which now includes the new genomic knowledge, is making applications in daily life, world economy and health. In combination with other major methods of obtaining biochemical and structural information such as synchrotron radiation, bioinformatics, biosimulation, NMR, various microscopies and mass spectrometry, neutrons will contribute in many fields of biology concerning human health and therapies, biotechnology and food science. The funding agencies' priority research missions where the ESS would have the largest impact include: structure based drug discovery; structural aspects of aging; food processing and agriculture; production of biosensors and biochips. In all these areas the ESS will allow one to address crucial questions by performing principally new experiments impossible with current neutron sources.

I. Introduction

The life sciences revolution involves molecular level detail.

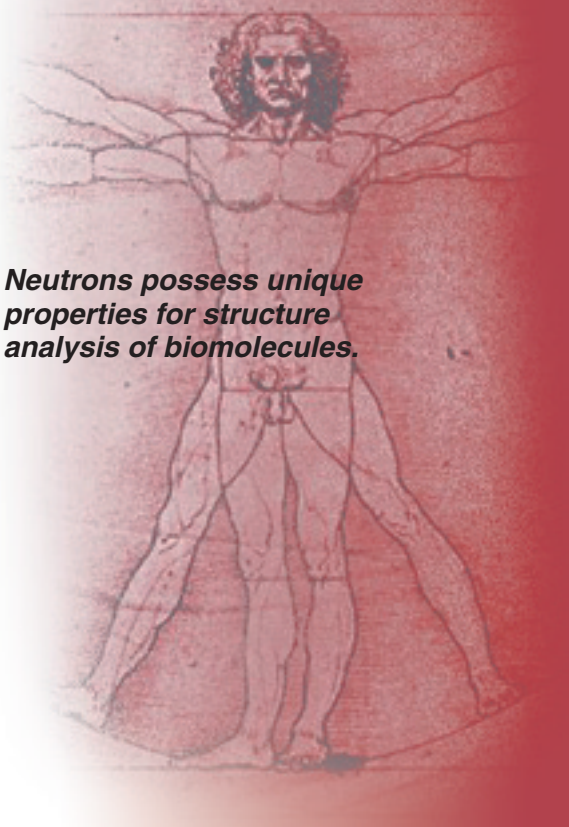
The end of the last century was marked by an explosion of progress in the Life Sciences and applications in daily life, world economy and health, finally including the revolution introduced by the knowledge of genome sequences from all types of organisms from human to bacteria and viruses. The fields of functional and structural genomics were born. Manipulation of single biomolecules and their complexes is resulting in the emergence of nanobiotechnologies, biosensors, membranes with associated new functions and mechanical properties, biomotors, etc. One of the key practical aspects related to these developments is based on the world-wide effort for the production of recombinant proteins and their complexes, including labelled molecules for studies by a variety of different biophysical techniques (in particular, neutrons and NMR), using new expression systems and robots, in connection with the recent availability of large cDNA collections.

Expanded cDNA collections open the prospect of selectable, vast, protein expression.

The ESS will bring about a ten to thousand fold increase of the available neutron intensity at key instruments important for investigations into biology. The specificity and uniqueness of neutrons for studies of biological molecular structure and dynamics is well established namely:

Neutrons possess unique properties for structure analysis of biomolecules.

- neutrons constitute a highly penetrating and non-destructive radiation;
- neutrons provide the ability to "see" hydrogen and to differentiate between light atoms in structures and to locate water molecules;
- neutrons allow to selectively identify and characterise components within complexes and in solution interactions;
- neutrons measure both amplitudes and frequencies of atomic motions and thereby are able to characterise molecular dynamics.



A special mention should be given to the role of neutrons in the challenge of structural proteomics of membrane proteins. Neutrons will provide powerful tools for their structural and dynamic characterisation *in-situ*, in the complex membrane environment by a variety of techniques including reflectometry, diffraction and inelastic scattering.

Due to the unique properties of neutrons, they are able to provide new insights in many areas of applied life sciences research. We now give below a selection of examples for which the ESS would allow major progress.

II. Drug discovery

Structure based drug design.

The knowledge of the 3-D structure and dynamics of proteins and nucleic acids as receptors for drug molecules, opens up an approach for new drug discovery known as Structure Based Drug Design. There are already examples of drugs discovered for the treatment of HIV and influenza virus. In the past, the process of drug discovery was much more laborious and/or uncertain. A famous example of "chance" discovery was penicillin in 1928 by Fleming. Society rightly expects a science-knowledge based approach these days to drug design; neutrons can efficiently contribute to the field.

Receptor site details both in topology and electric charge are needed.

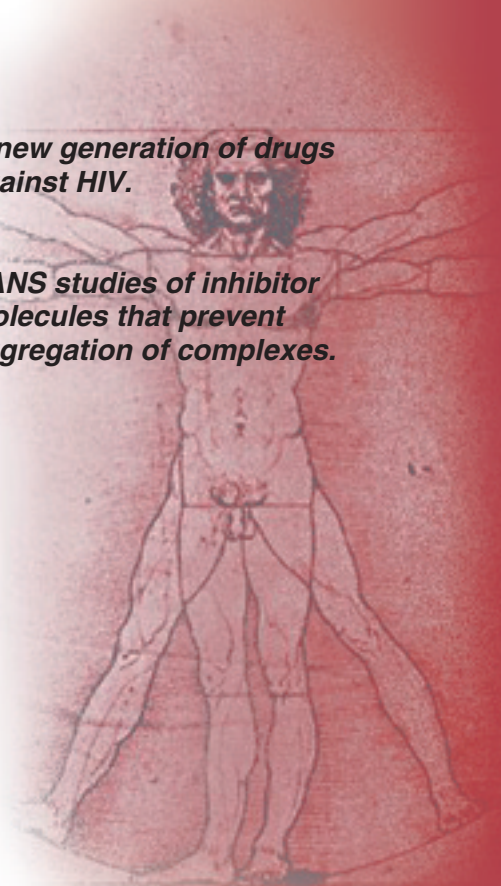
The 3-D structure characterisation of proteins and nucleic acids as receptors is challenging. The positioning of hydrogens and location of water molecules are generally the domain of neutron crystallography i.e. for which hydrogen and its heavy isotope deuterium make much more considerable contributions to the total scattering than for x-rays. The complete picture of a receptor site surface, both in topology and electric charge, is provided by a combination of x-rays and neutrons. As biological catalysts, enzymes are a very important class of proteins and, in most cases, they utilise the breakage and formation of hydrogen bonds as part of their mechanism of action. A recent example of a joint x-ray and neutron approach is the elucidation of the enzyme mechanism of a homologue of HIV aspartic proteinase (Figure 1). These results can help in the next round of drug design against HIV.

A new generation of drugs against HIV.

SANS studies of inhibitor molecules that prevent aggregation of complexes.

Valuable information for drug design at the quaternary structure level – e.g. whether specific association of proteins into larger complexes is prevented via an inhibitor bound to one or other member of the complex, as well as the structure determination of non-crystallisable proteins - will be provided by small angle neutron scattering (SANS).

The dynamics data of atoms at the receptor sites of proteins can be obtained using selective deuteration with the aid of inelastic neutron scattering. At present such studies are limited by the amounts of material required and to timescales shorter than nanoseconds. Higher effective intensity at ESS will largely ameliorate these limitations.



Finally, many drug targets involve proteins embedded in the cell membrane, the outer surface of which is the first point of contact of an invading virus. Structural information on these 'cell surface' receptors in their natural environment is very difficult to obtain but lateral and vertical structural information at the 10 Å level may be attained using 2-D reflectometry.

Stopping the invading virus at the cell surface.

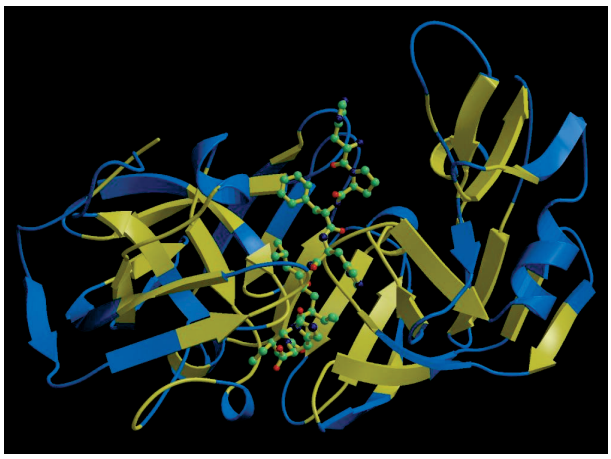


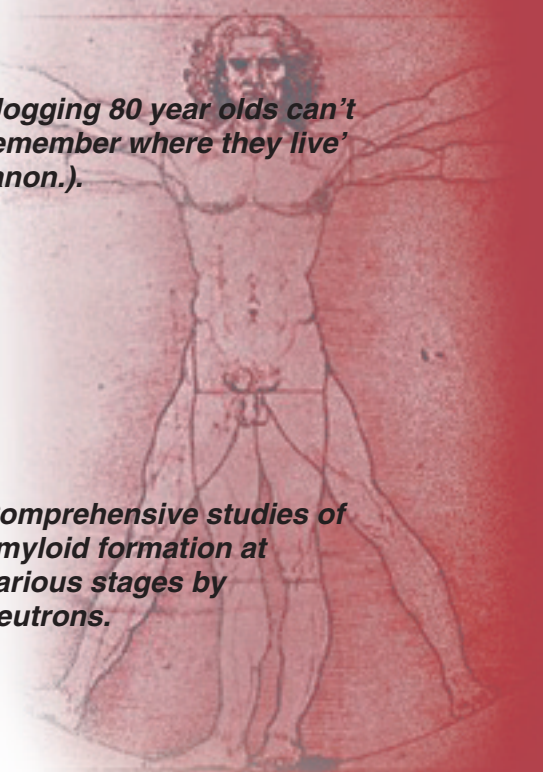
Figure 1: The high resolution neutron structure of the aspartic proteinase endothiapepsin with an inhibitor bound in the active site has provided much information relevant both to studies of the catalytic mechanism and to drug design. Prior to neutron studies the positions of protons on the catalytic aspartates had not been determined. This work represents the largest protein structure (35 kDa) studied to date by neutron crystallography at high resolution (2 Å). Most importantly the data give the protonation states of the catalytic aspartates. This has an important bearing on mechanistic proposals for this class of proteinase and for design of drugs which mimic the enzyme-bound transition state.

III. Aging

Structural studies of proteins involved with aging raise many prospects for the treatment of age-related disorders. One example is Alzheimer's disease which is the most common cause of dementia in older people. The progression of Alzheimer's disease involves the destruction of cells that control memory and eventually many other areas of the brain become affected until the patient dies. The majority of patients develop the illness beyond the age of 65 although 10 % of victims develop the disease before reaching this age. Alzheimer's disease is one of a wide range of so-called amyloid diseases which are caused by the formation of insoluble deposits of proteins. Alzheimer's disease, is caused by the formation of neuritic plaques in the brain and neurofibrillary tangles in the nerves. The plaques have been shown by electron microscopy to contain long protein fibrils. NMR and fibre diffraction show that these consist of polypeptide in the twisted β-pleated sheet conformation.

A critical problem of aging is Alzheimer's disease.

'Jogging 80 year olds can't remember where they live' (anon.).



Neutron scattering techniques have been very successful in the clarification of aggregation and self-assembly mechanisms of compartmented synthetic polymers and it may be assumed that they also could contribute significantly to elucidate amyloid formation. SANS allows one to study both the kinetics

Comprehensive studies of amyloid formation at various stages by neutrons.

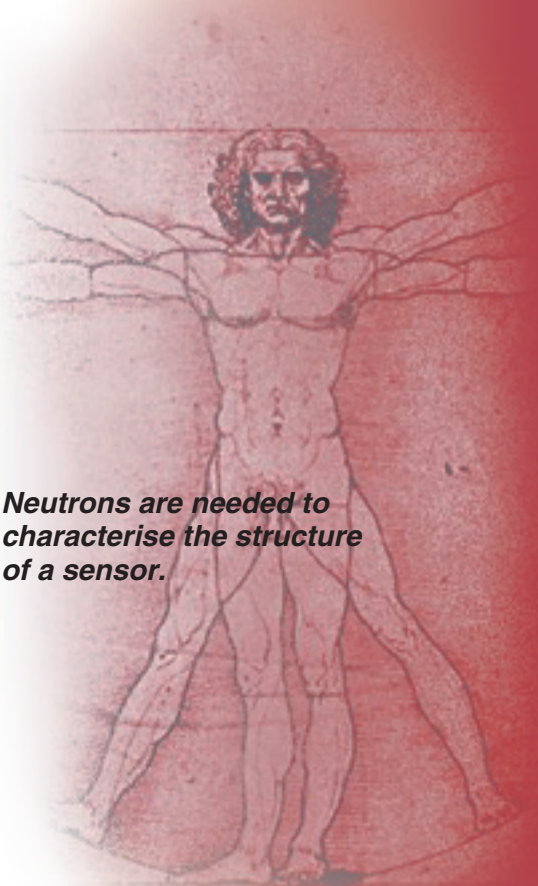
of aggregation and the associated structure at the most important initial stages. Neutron crystallographic studies both of the enzymes which catalyse the processing of the amyloid precursor proteins and of the proteins which associate with the plaques, thereby stabilising them and masking them from breakdown by proteinases, could have an enormous contribution to the design of therapeutic agents. Given the x-ray structures, neutrons will reveal hydrogen sites, hydrogen bonding structures as well as the hydration state. For example, it has recently been shown that an aspartic proteinase (β-secretase) is involved in processing the amyloid precursor protein in Alzheimer's disease and there is major interest in designing drugs which could inhibit this enzyme as a potential therapy. Neutron structural studies of a related aspartic proteinase (Figure 1) has defined the charge status of active site groups and this information is needed for rational drug design against all members of this enzyme family e.g the HIV proteinase which is the target of a number of clinically prescribed AIDS drugs. There is also scope for neutron fibre diffraction to probe the protein deposits formed in Alzheimer's disease. In summary, studies of this and other amyloid diseases will help to design drugs that inhibit plaque formation.

Design of therapeutic agents.

IV. Biosensors and biochips

In the last decade, huge investments have been made on the development of biosensors. These devices detect and respond to biological or chemical substances. They have many biomedical applications, *in vivo* and *in vitro,* as well as in disease detection, cellular repair, DNA recognition, drug delivery or the detection of physiological parameters. They will also be used in the environmental domain for the detection of pollutants. With the progress in miniaturisation achieved in the silicon industry, nanoscale biosensors are now on the market as well as "bio-arrays". DNA chips are capable of running a huge number of recognition experiments in a single chip, and likewise for protein-chips, an incredible number of ligand-receptor pairs can be probed in a single experiment (Figures 2 and 3). Current protein-chips consist of soluble proteins or peptides grafted onto a solid substrate. One of the challenges is to generalise this technique to membrane-based biochips, extending it to membrane proteins and consequently to ion pumps or channels, transporters etc. Optical techniques such as fluorescence and near-field microscopy or atomic force microscopy (AFM), or even semi-conductor inspired methods are classically used for detection in bio-chips.

Biosensors have many biotechnology and health applications including detection of pollutants.



In principle neutron reflection techniques offer an extreme sensitivity towards both the lateral structure as well as the perpendicular profile of a labile planar membrane carrying biopolymers. The low incident flux on the existing neutron facilities has permitted the study only of simple model membrane systems: pure lipid bilayers, or those with incorporated peptides or proteins (penetratin, alamethicin, bacteriorhodopsin, cytochrome C). Even in these simple situations, the experiments are restricted by the large sample

Neutrons are needed to characterise the structure of a sensor.

size required (few cm^2). This requirement for defect-free samples on a large area, makes it difficult to study native biological membranes. Kinetics and off-specular experiments on biological membranes are completely impossible with current neutron sources and instruments.

ESS will profoundly alter the situation. The specular reflection at the solid/liquid interface combined with contrast variation will allow the exact determination of the membrane structure and of the crucial polymer layer separating the biological membrane and the solid support.

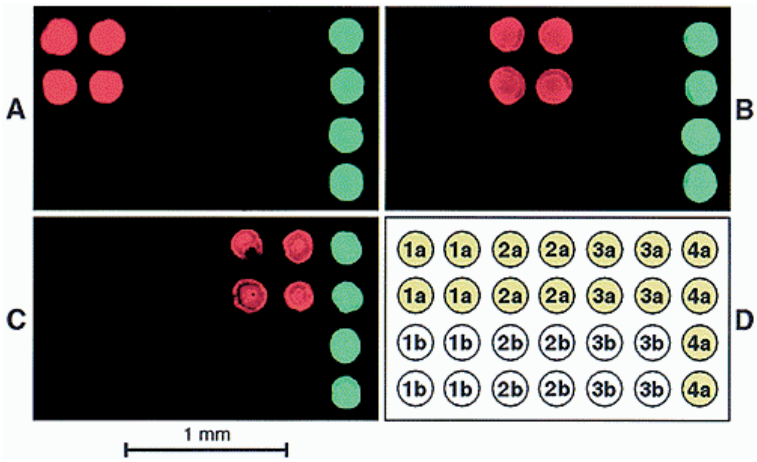


Figure 2: Protein array produced by protein printing on a glass slide (from MacBeath G. et al, J. Am. Chem. Soc. 121 (1999) 7967). Fluorescent-conjugated proteins specifically bind on their corresponding ligand decorated spot. The high number of spots ($1000/cm^2$) allows 10000 experiments at once.

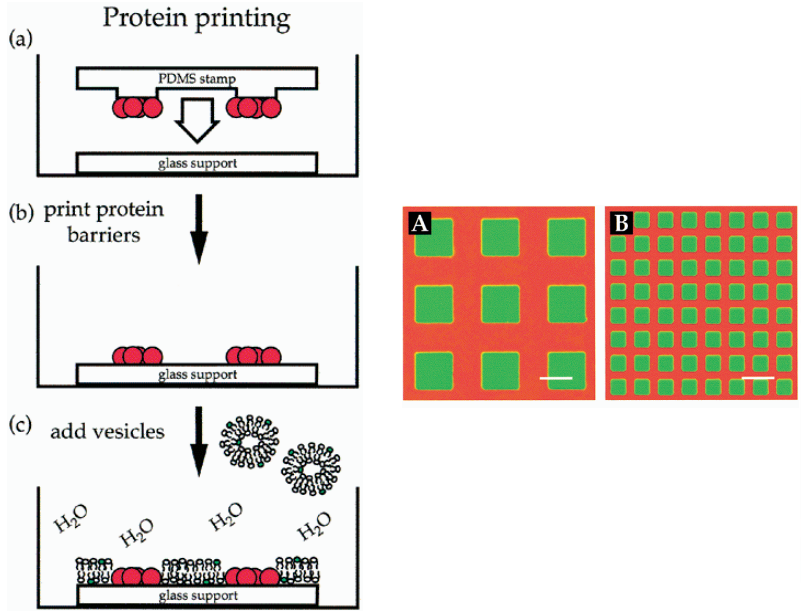


Figure 3: Model protein patterning produced by protein printing (a and b) and further membrane deposition (c) (from Kung L.A. et al, Langmuir, 16 (2000) 6773). Finally, in A and B, different achievable spots sizes are displayed (bars: 10 microns). The protein (bovine serum albumin) is labeled in red and lipid membranes in green.

Moreover, using off specular detection and in plane Bragg scattering, ESS will enable to study with molecular resolution the association and selfassembly of functional complexes within the membrane and their response to external stimuli. Kinetic experiments will allow to follow the function of a biochip. Such experiment will provide crucial information about how the different membrane constituents interact, how ligand receptor binding takes place and how cell adhesion is affected by external conditions.

Kinetic characterisation with current neutron sources is not possible.

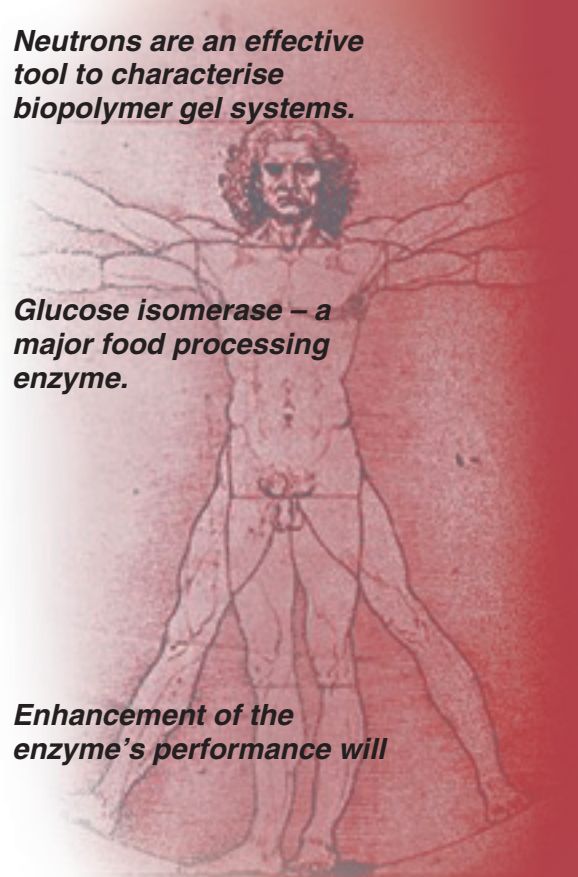
V. Food and agriculture

One of the most important applied directions of modern science is a systematic search for compounds and materials to be used in improving quality of consumer products, food processing and increasing yields of agricultural production. The properties of these compounds have to be thoroughly characterised on a nanometer scale and therefore structural analysis methods, including neutrons, are in many cases indispensable.

Biopolymer gels are used as food additives.

Polymer and biological gels are widely used in our common life and the importance of gels in industrial applications grows steadily. Temperature and pH sensitive gels are actively investigated and used for the production of intelligent materials such as switchable catalysts. Gels based on natural polymers e.g. cellulose, carageenans or hyaluronic acid, are most attractive for the food industry, as well as for the cosmetics and medical sector. In particular, thanks to their ability to induce thickening or gelation of solutions, carrageenans are widely used as food additives. In complexes with oppositely charged surfactants, natural polyelectrolyte gels form highly ordered nanostructures, also interesting for nanobiotechnology. At this stage the preparation of these materials is largely empirical and the characterisation is poor. A better understanding of the parameters that play a role in the stability and phase behaviour of particular classes of natural gels is thus of great practical interest. Due to the possibility of selective deuteration, elastic and inelastic neutron scattering are unique tools to study natural gels and their complexes with biologically active compounds including proteins and individual amino acids employed as surfactants.

Neutrons are an effective tool to characterise biopolymer gel systems.



Glucose isomerase – a major food processing enzyme.

One of the most important enzymes in food production is D-xylose ketol-isomerase enzyme (glucose isomerase) which is used industrially on a large scale to isomerise glucose to fructose. Fructose is used extensively as a sweetener in the food industry (e.g. for soft drinks like Coca Cola). To increase enzyme performance (activity and thermal stability), enzyme suppliers have examined glucose isomerase from a number of natural and genetically modified biological sources. The latter often display better properties so that improved knowledge of the active site structure and mechanism of enzymatic operation is of great interest to industrial enzyme manufacturers. x-ray diffraction provides the structure of the enzyme up to 1.9 Å resolution but does not allow for a clear distinction between water and magnesium due to the similar

Enhancement of the enzyme's performance will

number of electrons in oxygen and magnesium. Furthermore, it is difficult to place the water and cations that seem to be implemented in the enzyme activity. Neutron diffraction studies of this enzyme will be used to resolve the water positions. These studies would certainly benefit from enhanced diffraction resolution (i.e. currently around 2.5 Å at ILL LADI) towards 1.5 Å.

yield substantial saving.

Another example of a food-processing enzyme which is used as a fungal rennet in cheese manufacture is endothiapepsin. This process requires the specific cleavage of a single Phe-Met bond in the bovine milk protein κ-casein which is normally cleaved specifically by the aspartic proteinase calf-chymosin. However there is much interest in engineering the fungal aspartic proteinases such as endothiapepsin to have similar specificity and pH dependence as chymosin. Understanding the basis for an enzyme's pH dependence requires a full knowledge of its electrostatics which is something that neutron diffraction is well suited to. In addition, an understanding of the electrostatics of this group of enzymes will contribute greatly to engineering mutants appropriate for biotechnological applications.

Enzymes in cheese production.

The above are just a few selected examples treated in detail. In general, neutrons are also of great importance for the studies of many systems related to agricultural applications. Examples include characterisation of protein-nucleic acid interactions in plant viruses, analysis of the enzyme mechanisms involved in nitrogen fixation, phosphate metabolism, brewing, baking, antibiotic resistance, structural studies of the targets for pesticides e.g. the glycine decarboxylase complex, investigations of molecular and cellular adaptation to extreme environments, i.e. salt, acidity and temperature resistance in plants.

There are many further applications in agriculture.

VI. ESS relevance and instrument priorities

The entire suite of neutron-related methods (high and low resolution neutron crystallography, SANS, inelastic scattering, reflectometry) provides a comprehensive structural picture of many industrially important biological macromolecules.

ESS will remove bottlenecks in current methods.

The large size of the crystals required for present neutron analysis (several mm^3) excludes many promising candidates, and the ESS can decisively ease this bottleneck. Moreover, it is possible to bring into view more challenging problems using ESS, e.g. larger proteins. Thus many more systems will become amenable to structure based drug design. The instruments required are especially the ESS "smaller crystal protein diffractometer" and the "larger molecular weight protein crystal diffractometer".

Two ESS neutron protein crystallography instruments are needed. The ESS instruments offer gains of ~ 20.

ESS high intensity SANS on the long pulse target station will offer considerable gains over the current state-of-the-art. Here, smaller quantities and weaker concentrations of materials can be studied in shorter collection times. It will

The high intensity SANS instrument on ESS offers a total gain of 100 over current state-of-the-art.

allow systematic approaches to characterise the homogeneity (or aggregation state) of the samples, their oligomeric and complexed states - important information in a structural proteomic context. Time-resolved studies of mixing components can also be possible for the first time in many cases with ESS. ESS would have an enormous impact on the studies of biopolymer gels allowing e.g. kinetic experiments to analyse the pH-dependent structure transitions, light-induced changes of the gels containing photosensitive proteins, etc.

Dynamics information on the recpetor-ligand interaction can be obtained using selective deuteration utilizing inelastic neutron scattering. These experiments are not feasible now. ESS will considerably extend the sensitivity that is necessary, in order to perform the required dynamic contrast experiments.

The ESS inelastic scattering instruments offer total gains of 2-3 orders of magnitude.

In the various reflectometry applications ESS will decisively extend sensitivity to allow off-specular and time-resolved experiments - even native systems will be studied with implications for the design of new biochips.

ESS reflectometers offer total gains of 40.

6.4 Nanotechnologies

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Abstract
Nanotechnology is based on specific properties of materials which are determined by their structure on a nanometer scale. Such nanosized objects determine our life in the form of protein complexes, as viruses, as colloidal particles in water and as aerosols. They find use as dispersion colours and as adhesives. In industry they play an important role in the formulation of pigments and in the production of catalysts. Designing and controlling composite materials on the nanoscale is promoted by self-assembly and self-organisation of compartmented macromolecules. Complexity is one of the main features of nanosystems such as nanostructural alloys and composites, advanced functional polymeric materials, ultrathin films, quantum dots, nanotubes, nanocrystals, etc. Their structure can be unravelled by neutron scattering probing distance scales spanning the entire nanoscale regime. The ESS will enable us to access to the very large parameter space of such systems and allow the in-situ observation of structure formation and processing.

I. Introduction

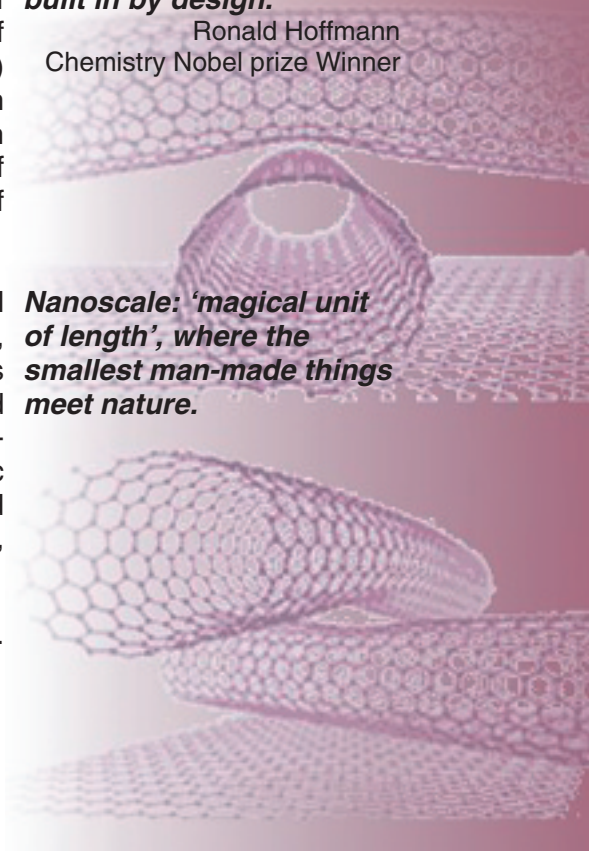
Nanoscience is concerned with discovering, understanding, characterising and fabricating materials and systems with novel properties, phenomena and processes that occur primarily because of their small size. Structures having dimensions of 1 to 100 nm (10^{-9} to 10^{-7} m) can induce important property changes compared to those exhibited by larger structures. New behaviours associated with the nanoscale are not merely the result of orders-of-magnitude reduction in size, but are caused by the emergence of genuinely new phenomena. These include the effects of confinement on electronic structure, the dominance of interfacial and surface phenomena (in contrast to bulk effects) with increasing surface-to-volume ratio, and purely quantum effects. The spatial scale of nanoscience is placed in between the scale of atoms dominated by atomic properties and that of the bulk materials. It has been termed a 'magical unit of length', where the smallest man-made things meet nature.

"Nanotechnology is the way of ingeniously controlling the building of small and large structures, with intricate properties; it is the way of the future, a way of precise, controlled building, with incidentally, environmental benignness built in by design."
Ronald Hoffmann
Chemistry Nobel prize Winner

Nanoscience is positioned at the crossroad of several traditional academic disciplines like physics, chemistry, biology, computational science and engineering and deals with complex materials generally involving soft and hard components. Nanoscience covers topics such as self-organising and self-assembling organic and inorganic materials, nanostructured alloys and composites, advanced functional polymeric materials, ultrathin films, quantum dots, nanotubes, nanocrystals, etc.

Nanoscale: 'magical unit of length', where the smallest man-made things meet nature.

Thus, complexity is one of the main features of nanosystems.



In fact, nanotechnology was spawned by the "bottom-up" science, which deals with how complex systems are built from simple atomic-level constituents.

Apart from the scientific interest, a major driving force behind the research in nanosystems is the prospect of wider technological applications. High-strength carbon nanotubes only a few billionths of a meter in diameter; nanoscale reinforced ordered polymer composites; magnetic storage disks that could hold 100.000 times more data than current disks; molecular machines; faster computers and more efficient catalysts and pharmaceuticals are some of the technological developments already envisaged.

"If I were asked for an area of science and engineering that will most likely produce the breakthroughs of tomorrow I would point to nanoscale science and engineering."

Neal Lane
Science Advisor to President Clinton

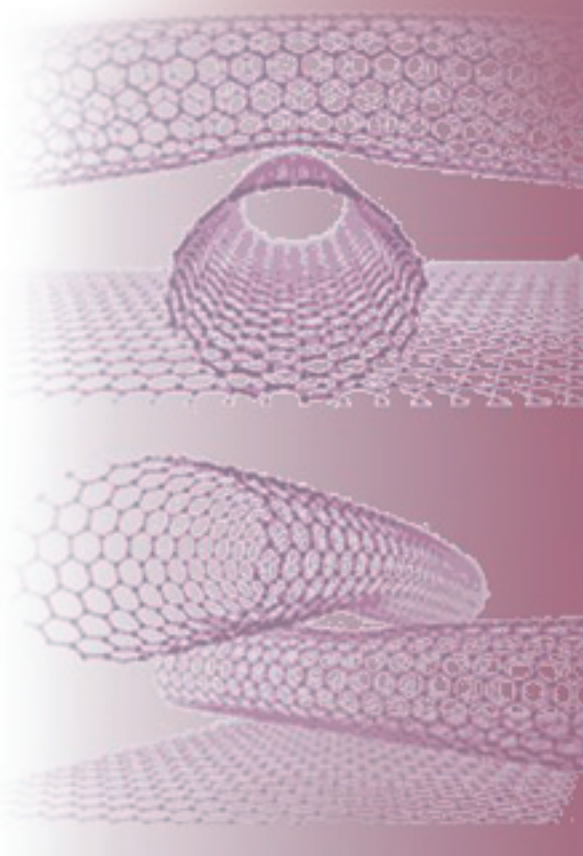
Nanotechnology is not only a priority area of the EU but also for many other countries including Japan and the USA. In 2001, the White House Office of Science & Technology Policy identified nanotechnology as one of 11 R&D areas that are "important national efforts requiring co-ordinated investments across several agencies".

II. Potential of neutron research

In new branches of materials research such as nanoscience, basic and technological aspects are particularly strongly interwoven. This tendency may be exemplified by the field of 'soft matter science'. Among the identified disciplinary flagship areas 'buried interfaces', 'self assembly and structure formation', 'soft-hard nanocomposites' and 'probing molecular dynamics in non-crystalline matter' have a direct impact on nanotechnology. As discussed in that context, neutron scattering offers unique capabilities for exploration, characterisation and finally for understanding and control.

General future trend: tailor made multicomponent materials for industrial applications. Neutron scattering employing Hydrogen/Deuterium labelling enables the unravelling of complex structures.

Nanoscale systems and devices involve complex materials usually containing different nanophases. Their structure can be unravelled by neutron scattering and in particular by small angle neutron scattering (SANS). Neutrons enable us to probe structure on distance scales spanning the entire nanoscale regime: atoms to macromolecules. In addition, the large cross-section difference for hydrogen and deuterium, enables H/D labelling studies of complex "soft", biological and "soft-hard" nanosystems. This isotopic labelling can also be used to highlight particular interfaces of nanosystems involving "soft" components. Moreover, many nano-devices contain magnetic phases. Neutron scattering has proven to be the unique tool for the investigation of magnetic structure of matter, both static and dynamic (fluctuations) as well.



Furthermore, understanding the mechanisms of nanoparticles and nanosystems formation and, in particular, the processes of self-assembly are of utmost importance for developing new products and technologies. Enhanced flux neutron scattering allows these processes to be followed in real time. This is of particular relevance in the case of "soft" nanosystems as, for instance, organic nanoparticles and self-assembling polymer

templates. Moreover, the ability of neutrons to penetrate macroscopic flow devices will allow exploration - in real time – of actual industrial processing mechanisms.

The nanoscale experiments described above involve small samples, complex molecules, time-dependent aspects of synthesis or self-organisation and weak interactions. All of them require a high neutron flux to be feasible. Most experiments we can envisage are at the limit of present day capabilities or they are just impossible. Thus, the future development of the field strongly demands high flux sources such as ESS.

Examples for present achievements of neutron methods in nanoscience comprise to a large extent equilibrium studies on nanostructured bulk phases and on liquid-liquid, liquid-solid and liquid-air interfaces if they can be made large enough. Neutrons have explored the self-assembly of block copolymers and the properties of nanofillers in elastomers; they have clarified the self-organisation of surfactants forming nanoreactors in solution for the production of nanoparticles. Neutrons have touched on nanosized magnetic structures on substrates or thin film magnetism including the giant magneto-resistance (GMR) effect.

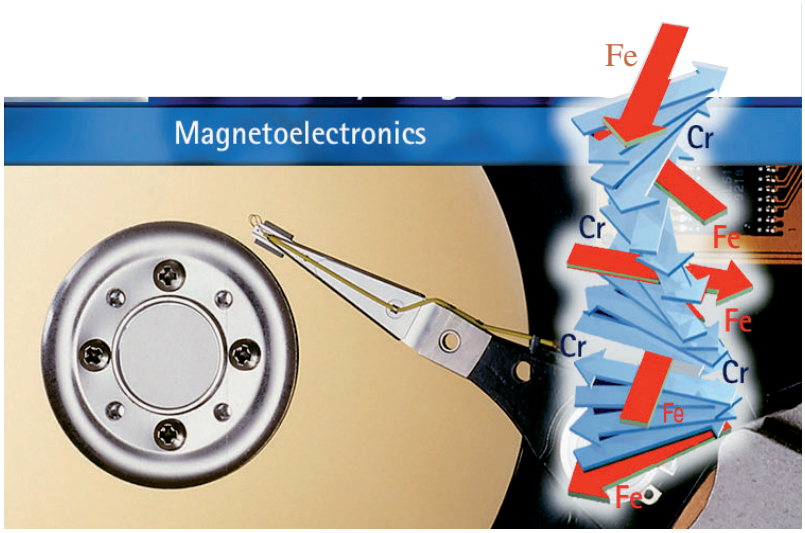


Figure 1: Giant magneto-resistance (GMR) reading head of a hard disk storage media. The arrows display the spin structure as revealed by neutron scattering.

III. Novel opportunities through ESS

Some of the challenges in nanoscale science and technology which could be solved by the new neutron capabilities provided by ESS are:

- The structure, interaction, dynamics, magnetisation kinetics and spin dependent transport in magnetic nanosystems.
- Effects of reduced dimensionality on collective phenomena in nanosized matter.
- Measurements of the correlation lengths (both static and

dynamic) associated with spontaneous electronic phase separation and competing ground states, in highly correlated electronic systems in nanosized materials.

- Identification of molecular-level processes occurring at liquid-solid interfaces, in order to understand, which nanomaterials are stable and why processes differ for macro- and nanomaterials.
- Identification of the difference between activated and inactivated states of catalysts (how the catalyst is poisoned) using monolayer-sensitivity inelastic neutron scattering.
- Direct, in-situ measurement of nanoscale phase separation kinetics (polymer blends, metallic alloys).
- Identification of the components and interactions of multiprotein complexes, to enable harnessing these “Molecular Machines” for functional nanostructures and nanotechnology.
- Direct, in-situ measurement of self-assembling building blocks or templates from polymers.
- Time-resolved studies of the formation processes of nanoparticles.
- Time-resolved studies of polymer guided crystallisation in biomineralisation processes.
- In-situ processing of polymeric nanocomposites.

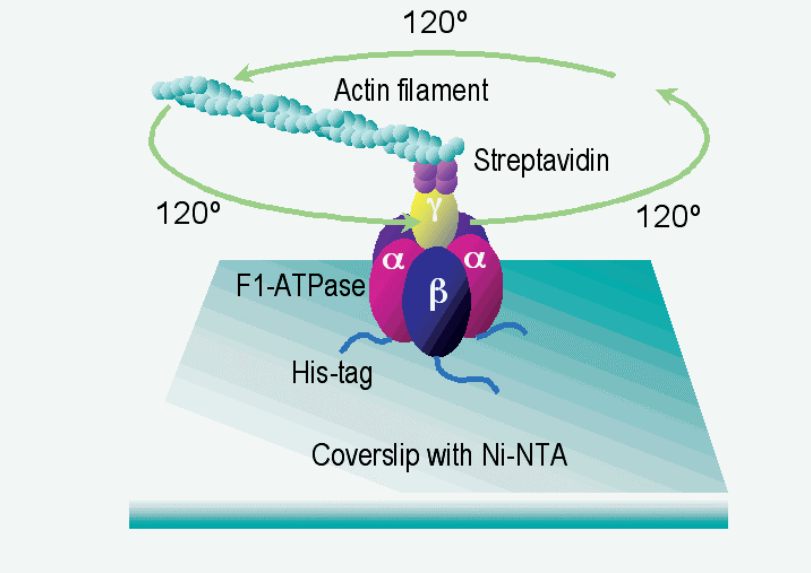


Figure 2: Nanoscale machines enable cells to carry out basic functions. The F1-ATPase complex, depicted in the diagram above, enables cells to produce the biochemical fuel it needs. SANS could follow conformational changes of the protein while functioning.

In order to avoid overlap with priority research missions like microsystems and information technology, functional materials and biotechnology, in the following we focus on flagship topics related to soft nanosystems.

IV. Examples

Controlled formation of organic nanoparticles

The importance of nanoparticles, i.e. particles with dimensions in the range of about 10 nm to a few hundred nanometers is obvious. They determine our life in the form of protein complexes, as viruses, as colloidal particles in drinking water, surface water and sea water, and as aerosols. They find use as dispersion colours and as adhesives. In industry they play an important role in the formulation of pigments and in the production of catalysts. Numerous attempts are being made to deliver nanoparticulate forms of pharmaceutically active compounds specifically to the desired site of action in the body. Finally, the use of nanoparticles as quantum dots with special properties for electronic components is also mentioned. Beyond these practical aspects there is active scientific interest in nanoparticles owing to their special properties, which lie between the properties of molecules and those of bulk material.

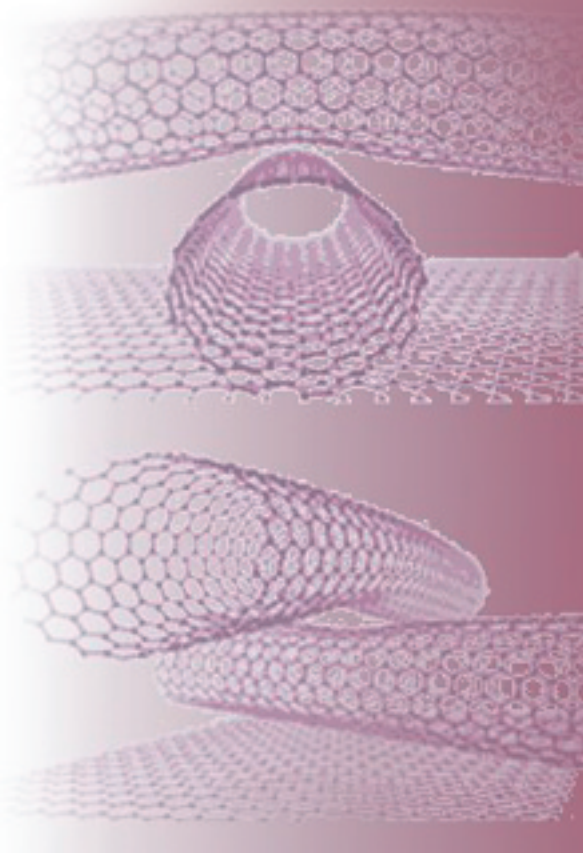
Nanoparticles determine our life in the form of protein complexes, as viruses, as colloidal particles in drinking water, surface water and sea water, and as aerosols.

Control of nanoparticles' size and composition leads to new properties for catalysis, electronic components and delivery of drugs.

Many active organic materials are poorly soluble in water, or even insoluble. Aqueous forms of application thus require special formulation techniques in order to be able to utilise or optimise the physiological (pharmaceuticals, cosmetics, plant protection, nutrition) or technical (varnishes, printing inks, toners) action. The most interesting properties of nanodispersed active organic materials include the impressive increase in solubility, the improvement in biological resorption, and the modification of optical, electrooptical, and other physical properties which are achievable only with particle sizes in the middle or lower nanometer range. In this context attention is drawn to the recent increase in research activities which have as their objective the continuous, automatic preparation of nanodispersed systems by precipitation from molecular solution. This undertaking is complicated by the complexity of many precipitation processes, see Figure 3, which extends far beyond the often-used picture of nucleation and growth.

Historically a strongly phenomenological approach to formation of nanoparticles is encountered; attempts are made to deduce a mechanism of formation from the final structure of the particles, where a purely descriptive treatment is considered satisfactory. But meanwhile it has become obvious that it is necessary to understand at a basic, i.e., molecular, level how a nanoparticulate system is formed [D. Horn, J. Rieger, Organic Nanoparticles in the Aqueous Phase - Theory, Experiment, and Use, Angew. Chem. Int. Ed. **40** (2001) 4330]. This knowledge is of prime importance as only with a knowledge of the mechanistic aspects of particle formation can the process be manipulated specifically in the sense of control.

SANS is a unique technique to characterise nanoparticulate formulations since most systems consist of at least three components (water, active ingredients, stabilising polymers or surfactants). Insight into the structural features is gained by



contrast variation - either by D_2O/H_2O exchange or by selectively deuterating one of the components. High flux neutron sources will open the opportunity for time-resolved studies. Only then will it be possible to really understand the formation processes in nanoparticulate systems and to develop new ideas and products based on this knowledge.

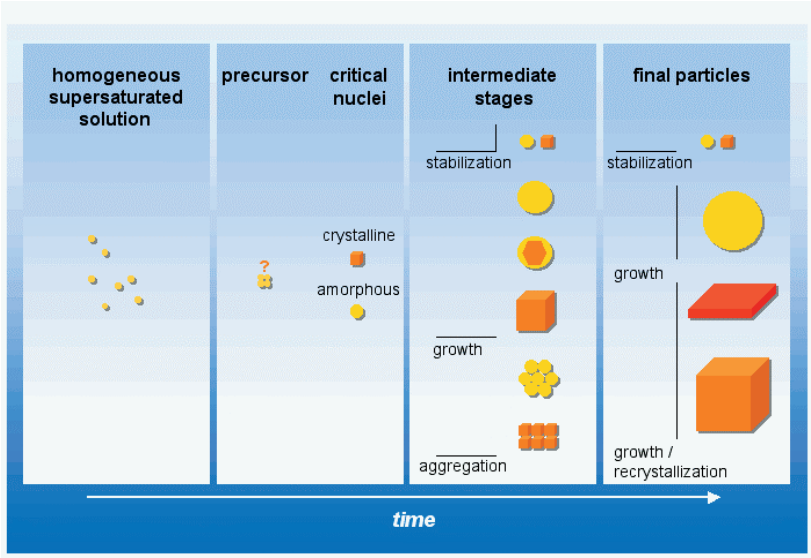


Figure 3: Structure formation during particle formation processes from supersaturated systems. By knowledge of the molecular mechanisms it will be possible to control the process by design and by the addition of suitable polymers or surfactants.

Polymer templates in nanotechnology

Self-assembling block copolymers offer an unprecedented variety of morphologies and their control by appropriate nanoengineering. Such amphiphilic block copolymers have been used recently, in order to produce ceramic polymer hybrids, where the hydrophilic parts are integrated into the ceramic material [P. F. W. Simon et al. Chem Mater. **13**, 3464 (2001)].

Self-assembled block copolymers can be used in order to template a manifold of nanostructures. Applications in biology include systems to direct cell or tissue response for biocompatibility or bio-sensor applications.

The self-assembling process rests on the fine balance of competing interactions between the different polymeric parts and exhibit similarities to complex biological systems. In the bulk, diblock copolymers self-assemble into four different structures: spheres, cylinders, bicontinuous gyroid structures and lamellae. Together with a silicon precursor a variety of nanoscale structures may be achieved. Via the control of polymer chemistry and processing conditions the nano objects are predetermined in size, shape and composition. Mesoporous materials for separation technologies and highly efficient catalysts are obtained. Similarly, diblock copolymer structures may be used to create templates for wax control in diesel fuels or pour-point depressors in crude oil. The polymeric templates may be extended to biology, including systems to direct cell or tissue response for biocompatibility or bio-sensor applications. Applications in medical science include implants, prostheses, drug delivery and diagnostics.

The future potential of the self-assembly approach for the creation of new nanomaterials lies in the versatility of polymer chemistry in connection with for instance sol-gel chemistry, which may be exploited for synthesis. Figure 4 displays the way to complexity in going from diblock copolymers to triblocks and ultimately to biological-like compartmented macromolecules. Already on the basis of triblock copolymers a huge variety of morphologies may be created. More than a hundred morphologies with a greatly enhanced parameter space were identified. The properties of these phases may be tailored by appropriate polymer block selection and the addition of judicious amounts of the corresponding homopolymers.

A precise control of the feature sizes is achieved via the length of the blocks. The number of building blocks along the chain thus elevates the complexity of the resulting structures by large amounts. An understanding of all these structures is still a matter of intense research. The pathway of future research may go along to even more complex structures and finally relate to the compartmented structures of biomolecules with all their rich variety (see Figure 4).

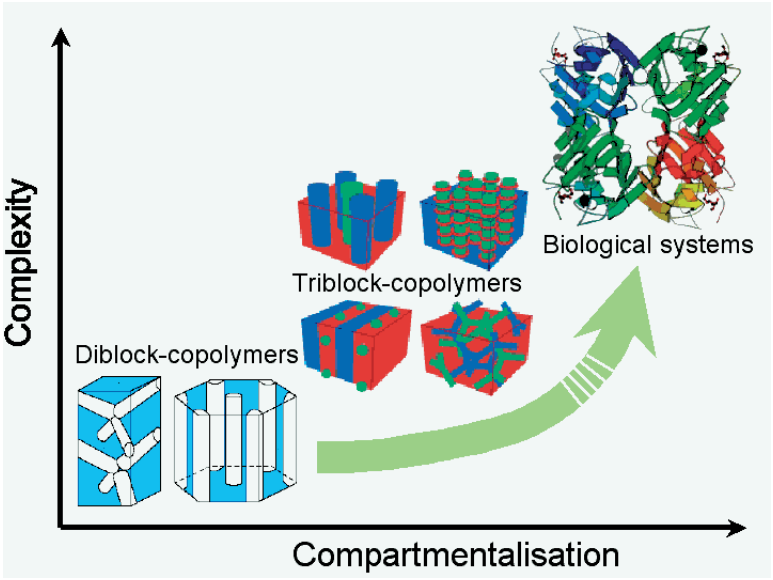


Figure 4: Increasing complexity from compartmented synthetic polymers to life.

The structural complexity of nanocomposites as well as their formation kinetics is only accessed by high flux neutron sources like ESS.

This manifold of structures challenges the use of such polymer systems as new structure-directing agents for novel nanostructured materials. Depending on the hard component to be introduced, templating of magnetic devices, smart materials, photonic crystals, membrane structures for gas separation or fuel cell applications are conceivable.

A rational design of such materials will need a detailed characterisation of all components. Small angle neutron scattering and neutron reflectometry using selective labelling, contrast variation and possible polarisation analysis will offer

unparalleled means of investigating the self-assembled structures. In concert with other techniques like synchrotron radiation, NMR and imaging methods the information necessary for a thorough understanding will be obtained. The potential impact of neutron sources for the elucidation of these structures is strongly emphasised in the recent Nanotechnology report [www.er.doe.gov/bes/nanoscale.pdf] from the US Department of Energy. The structural complexity together with the multidimensional parameter space will require extensive sets of experimental data only achievable with a high flux neutron source like ESS. Furthermore any investigation into the kinetics of structure formation will need fast experiments depending on the high flux of ESS.

Nanocomposites: Nanoparticle reinforced polymers

Requirements for increased fuel economy in motor vehicles demand the use of new lightweight materials – typically plastics that can replace metal. Nanocomposites are a new class of materials which are presently studied extensively. They consist of traditional polymers reinforced by nanometer scale particles dispersed throughout (see Figure 5). These reinforced polymers may present an economical solution to metal replacement. They are likely to find also use in non-automotive applications such as pipes and fittings for the construction industry; refrigerator liners; business, medical and consumer equipment housings; recreational vehicles and appliances.

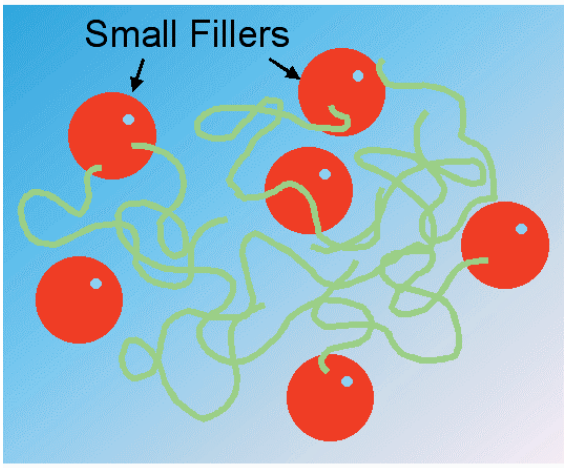
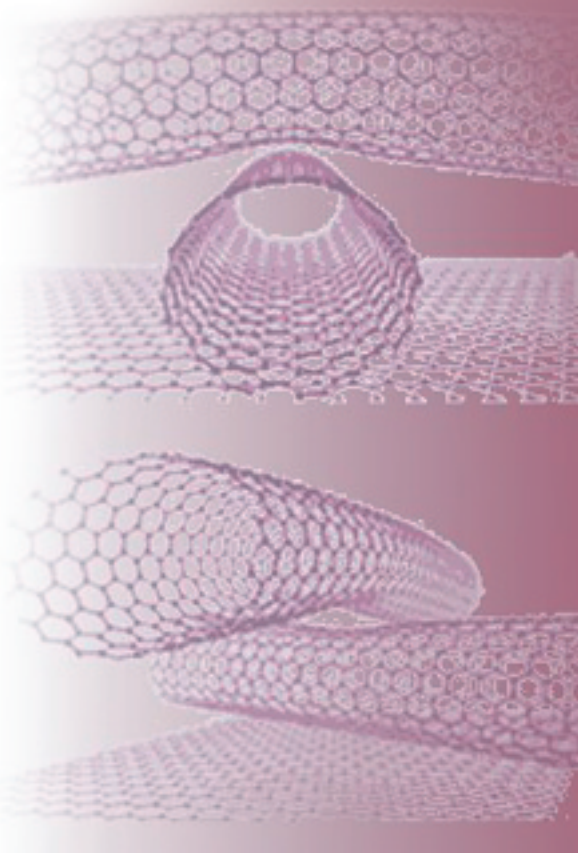


Figure 5: Nanoparticle reinforced polymer materials.

Rational nanocomposite fabrication and optimisation will be greatly aided by ESS.



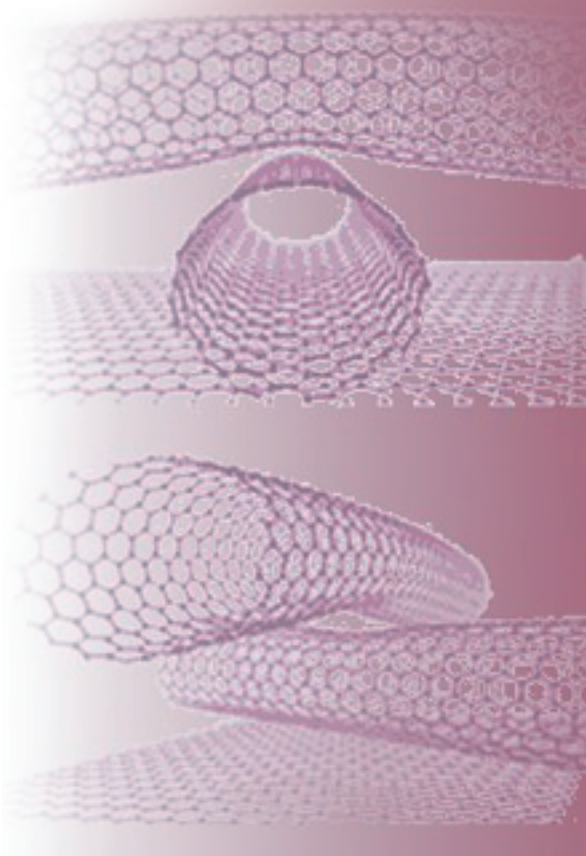
In principle, nanocomposites can be easily extruded or molded to near-final shape, providing stiffness and strength approaching that of metals at reduced weight. Corrosion resistance, noise dampening, parts consolidation, and recycleability would be improved. However, producing nanocomposites requires the development of methods for dispersing the particles throughout the plastic, as well as means to efficiently manufacture parts from such composites.

Using SANS at the ESS the degree of dispersion heterogeneities, orientations, etc. may be followed in-situ during extrusion and optimised. The non-linear mechanical properties are poorly understood but of great industrial interest.

The particle displacements and interactions during deformation may be followed by SANS. Both are very important for mechanical properties. They can be controlled to a large extent by adsorbing or grafting a layer of polymer on the filler particle surface. Using deuteration, the layer of polymer can be observed within the nanocomposite film. It is also possible to observe the conformation of a polymer chain inside the filled film. One therefore has access to the state of chains close to particles, or in the bulk space in between the particles.

V. Instrument requirements

Neutron scattering research in the field of nanotechnology primarily requires very high intensity SANS instruments and reflectometers in order to reveal nanoscale structures in small samples and also to allow for real-time experiments. Focussing SANS will be needed to explore the largest structures. A full spectrum of high intensity spectrometers is needed to probe the wide time domain from 10^{-14} to 10^{-7} s, in order to reveal dynamic processes as different as binding in catalysis or displacements of nanoscale particles.



6.5 Cultural Heritage: Artefacts and Materials

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Abstract

The discovery of ancient artefacts and artworks that bear witness to our cultural heritage typically raises a variety of questions: from the correct determination of their historical and cultural time-frame, the location and method of production, to the choice of treatments and environmental conditions for restoration and preservation. A large variety of chemical, physical and microstructural techniques are currently employed to characterise objects of cultural significance, indeed the same techniques that are generally applied to studies in the mineralogical and material sciences, and which deal with the characterisation of solid, generally inorganic matter such as; mineral, stone, ceramic, glass, metal, and their derivates.

Neutrons, as opposed to x-rays, are the best probe for examining the interior of thick samples. Neutron analysis, which is intrinsically non-invasive, is both unique and complementary to more conventional techniques. When sampling is not possible, neutron methods provide chemical, phase specific, and microstructural information from undisturbed large volumes. Furthermore, comparison with artificially produced materials, such as metals and alloys, can also be effectively exploited in order to obtain indirect information on the manufacturing techniques of the objects under investigation.

Specifically, TOF neutron diffraction at the ESS will provide invaluable information on objects from museums and galleries that must not be damaged by cutting, drilling, scraping etc. Data can be collected from large, intact objects of almost any shape, and the experimental set-up is both simple and free from sample movements. The many-fold increment in signal and resolution afforded by this new source, will allow element sensitive small volume phase identification and quantification, detailed crystal structure analysis of the constituent phases, and direct imaging in two- three- and four- (diffraction) dimensions by imaging and tomography making use of energy-tuning techniques. These methods could certainly provide a clearer picture of the technological, commercial and, more generally, historical and archaeological aspects of the sample. With a view towards preservation, they would provide invaluable information regarding the choice of restoration and conservation procedures.

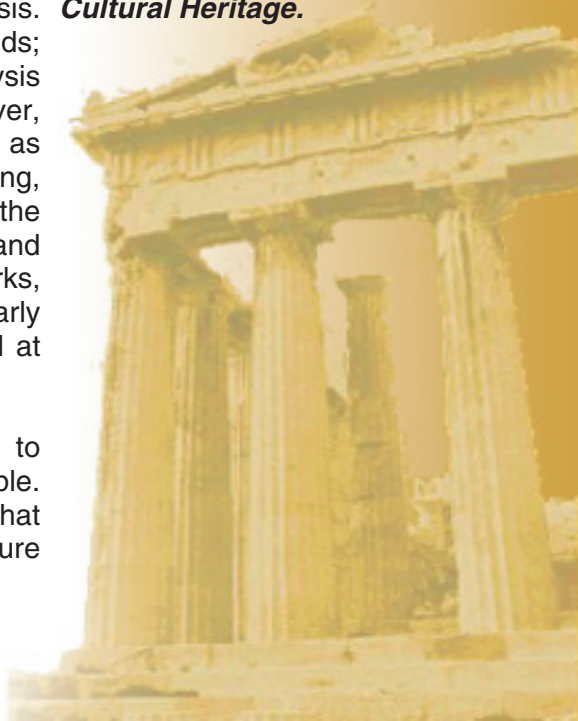
As with the mineral and Earth sciences, the potential of neutron scattering is only recently being realised in the fields of preservation of cultural heritage and archaeometry. With the availability of an ESS-class neutron source there is much to look forward to with the opening of new avenues in this field of study.

I. Introduction

The contribution of neutron scattering to Cultural Heritage.

Most frequently, archaeometric determinations of artefacts are obtained by methods such as electron microbeam analysis and imaging, x-ray fluorescence and neutron activation analysis. These are predominantly chemical probe methods; complementary information can be obtained by phase analysis through diffraction methods (x-ray, electron or neutron). However, most of these methods are invasive in one form or another as they require destructive sampling techniques such as coring, transversal sectioning, or even powdering some portion of the sample. When dealing with objects of cultural heritage and historical significance (prehistorical artefacts, priceless artworks, palaeontological material) sample destruction or damage is clearly unacceptable. Consequently, much current research is aimed at developing non-destructive, diagnostic techniques.

Moreover, extrapolation of results taken from microsamples to represent large objects or bulk samples is strongly questionable. What is required is a non-invasive diagnostic technique that provides fundamental information on composition and structure



from anywhere within an antique object (i.e. penetrating deeply into the sample as well as probing a large part of its volume) which is not currently available.

II. Neutrons in archaeometry and conservation

Microstructure, phase identification/quantification and texture analysis of archaeological objects by neutron diffraction has only recently been made available, but the potential application of such powerful techniques spans many fields of interest within conservation, archaeological and natural history research, ranging from routine fingerprinting to complex conservation problems. The standard diagnostic tools used today for ceramics, glass, paintings, and metals are not suitable for the characterisation of inhomogeneities at both the microscopic and macroscopic scale, that would provide information on thermal profiles, element distribution, mixing, and mechanical properties developed during manufacture. For instance, segregation, dendritic heterogeneity, degree of hardening and twinning, all represent fundamental aspects required for the determination of the historical and cultural background of archaeological findings, to correctly reconstruct their history and to evaluate sample deterioration.

Non-destructiveness in bulk

Neutrons are non-invasive.

The non-destructive nature of neutron scattering experiments makes the technique well suited for handling large, undisturbed samples and rare, unique objects, both natural and man-made, that encompasses materials as diverse as, for instance: sediment layers, rocks, fossils, bones, ceramic, glass, bronze and other metals. The importance of good grain statistics demands measurements from large samples. In stone diagnostics in particular, where grains on the order of one mm^3 are not rare, again large samples are often required. Furthermore, a generalised interpretation of analytical results from small portions of a large artwork be it stone, ceramic, glass or metal, can be highly misleading. Neutrons offer a non-destructive diagnostic technique providing fundamental information on the composition and structure of an antique object, extending deeply into its interior as well as on a large part of its volume.

Direct imaging, radiography, tomography

In-situ physical properties by direct imaging.

Neutron penetration can be advantageously exploited for neutron imaging to determine the inner features of materials and artefacts, such as composition, density and phase distributions, beyond the reach of less penetrating probes, with specific applications in archaeology and preservation.

At present most neutron imaging is performed either by simple neutron radiography, which exploits the absorption contrast of different elements in the object to obtain two dimensional projections, or by neutron-induced gamma activation, which also allows chemical analysis by measuring the decay time of the activated species.

These techniques can be used to investigate painting materials as performed at HMI Berlin for the correct attribution of some of Rembrandt's vs. Rembrandt's school works from the Berlin-Dahlem Museum (Preussischer Kulturbesitz).

Paintings and the new dimension afforded by higher flux at the ESS.

It is expected that the neutron flux supplied by the ESS will enhance the quality of imaging of artworks through the use of energy-tuned narrow neutron beams, so that the resonant absorption of specific nuclei and the prompt emission of gamma rays by short lived isotopes may be used for a 2-dimensional (imaging) or 3-dimensional (tomography) analysis of the successive paint layers as well as of bulkier materials. A further dimension will also be provided by the neutron diffraction signal.

III. Stone materials

Surface reactivity and water content

The specific problem of stone degradation (i.e. from historical buildings) needs detailed characterisation of the constituent materials and textures, including porosity and fluid contents. The methods and techniques are identical to those used in the investigation of modern building materials.

Reactive H_2O, CO_2 and SO_4 must be neutralised prior to preservation.

Water, carbon dioxide and sulphate ions are largely responsible for the degradation of natural building stones even when present in very low concentrations; the detailed study of such systems on an atomic scale can solely be addressed by the unique capabilities of the ESS. The breakdown, weathering and transformation of minerals generally involves the migration of hydrogen through the mineral surface and into the subsurface of the crystals. This implies volume increase (flaking), may induce oxidation and can be coupled with other ion exchange reactions or the freeze-thaw cycling. All of these processes change the physical properties of the near-surface region of the minerals. As these weathering reactions occur at the mineral/fluid or mineral/biota interface, a fundamental understanding of mineral surface reactivity requires the application of surface sensitive spectroscopy and diffraction.

Commonly, x-rays are used in the reflective mode to investigate mineral surfaces, however, neutrons are far superior to x-rays for direct probing of protons and deuterons. Furthermore, rapid high-resolution, high-intensity neutron spectroscopy, reflectivity and diffraction are required to study the dynamics and to generate models of mineral hydration, molecular binding and ion exchange involving iso- and quasi-iso-electronic species (e.g. Ti^{4+}-Ca^{2+}-K^{+}, K^{+}-Cl^{-}, Na^{+}-Mg^{2+}-Al^{3+}-Si^{4+}, or Fe^{2+}-Mn^{2+}).

Neutron spectroscopy at ESS to study molecular dynamics and mineral hydration in stones.

Prior to any surface treatment of degrading stones and sculptures in monuments and buildings, a bulk, quantitative assessment of the residual H, OH and H_2O content must be performed. This analysis must be non-destructive and capable of handling/exploring large dense samples while simultaneously detecting weak and/or very subtle signals. Only the ESS will enable studies on very low concentrations present in such samples.

Diffraction methods in archaeometry and restoration

Dating excavation sites and unveiling trading patterns, cultural exchanges and manufacturing techniques.

Within archaeological research phase, microstructure and texture analysis of artefacts and stone materials using neutrons is also relatively new but have a great potential in the future. Diffraction techniques are important for helping to date excavation sites, to establish trading patterns, to determine cultural exchange between regions, to elucidate historic and regional abundance, trading networks and to help identify the original source of raw materials. Phase and microstructural characterisation of ancient objects by diffraction methods can provide suggestions as to the specific manufacturing techniques that were being used. Diffraction studies may also address the issues of source materials or alteration/corrosion phases produced by changes in the environment (e.g. patina, black crusts, etc.).

From stone fingerprinting to conservation problems.

Owing to the non-destructive character of neutron techniques, their applicability to relatively large, intact, and precious archaeological objects is obvious. Additionally, the large interaction volume and rapid data collection at ESS will provide a range of new applications in the study and conservation of historical artefacts. TOF neutron diffraction provides new and unique information to that from x-ray diffraction. No preparation of the objects is needed and the experimental set-up is simple and free of sample movements. Ultimately, restoration and conservation problems relating to artefacts such as that reported in Figure 1 can be very effectively addressed through this wide variety of neutron scattering techniques.

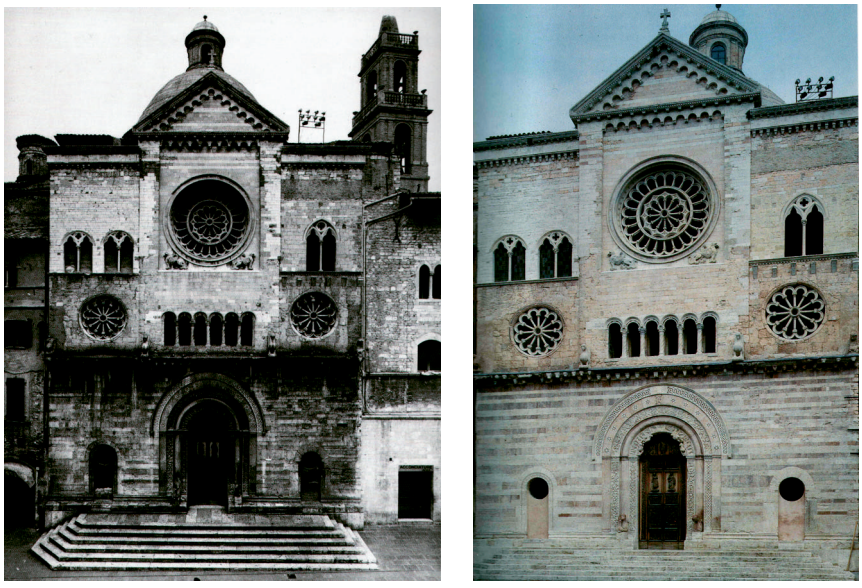


Figure 1: Foligno Cathedral before and after restoration. Conservation problems still need to be addressed (photo courtesy of B. Moroni and G. Poli, Perugia).

IV. Ceramics

Provenance of ceramics through mineral identification.

Correlations between phases, or ratios of phase proportions, may be used to characterise or classify an artefact. During ceramics firing the starting materials undergo solid state reactions the character of which depends upon the firing temperature, duration and atmosphere. Ancient or pre-historic ceramics fired at moderate temperatures, often exhibit very complex diffraction patterns due to a wide variety of mineral phases, among them clay minerals and sheet silicates which need high intensity and resolution for identification and quantification.

One example of the application of fingerprinting is that of medieval German ceramics from Siegburg and Brühl, two prominent sites for stoneware development and production in the Middle Ages, where the presence of cristobalite is characteristic of Brühl pottery. Further classification of pottery fragments by mineral phase fractions measured using TOF neutron diffraction is shown in Figure 2.

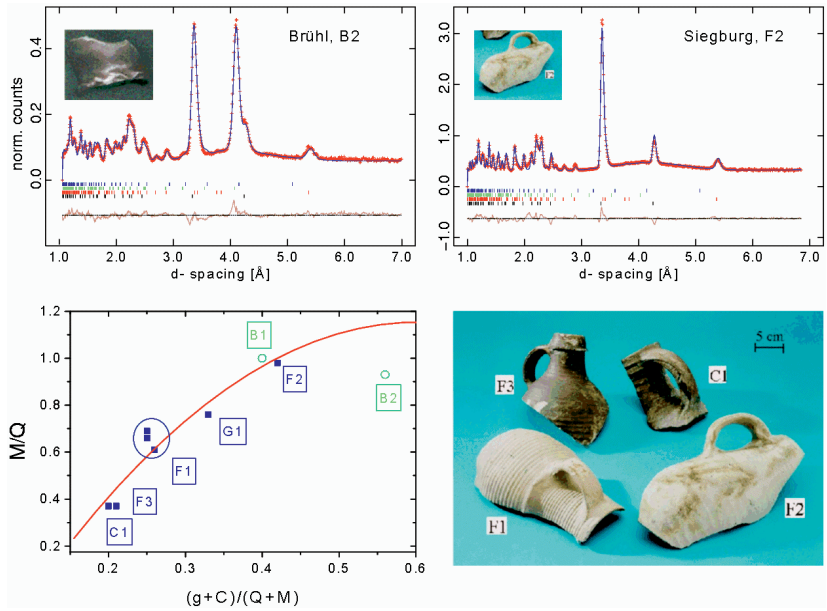


Figure 2: TOF-diffraction patterns taken at ROTAX, ISIS, on Brühl and Siegburg ceramics (top) and classification of pottery fragments (right) from earthenware (C1, F3) via early stoneware (F1, G1) to real stoneware (F2) using mineral phase fractions of quartz (Q), mullite (M), cristobalite (C) and glass (g).

V. Metal artefacts

Characterisation, authentication, and interpretation of manufacturing processes of metal artefacts

Applications to Bronze and Iron Age artefacts.

Materials change their microscopic structure as a result of mechanical or thermal treatments during manufacturing. This implies that a structural analysis by neutron diffraction may give valuable information about ancient production processes. This has been demonstrated by TOF neutron diffraction analysis in the case of an Etruscan bronze *olpe* (see Figure 3). The compositions of the bottom and the wall of the object were determined to be typical for classical bronzes with 90% copper and 10% tin.

Interestingly, a significant amount of lead was found in the handle, and a repair patch at the base of the vessel revealed a considerable degree of corrosion as indicated by the detection of a substantial amount of cuprite (copper oxide). Furthermore, from detailed analysis of the diffraction peak profiles it was possible to distinguish the underlying original bronze from the patch material which is almost pure copper.

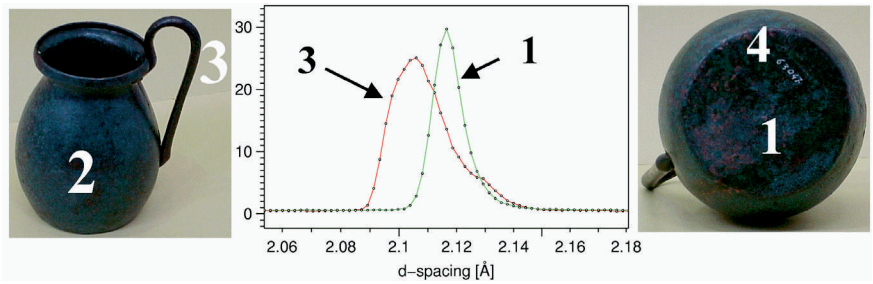


Figure 3: Side (left) and bottom (right) view of an Etruscan olpe (400 BC, Museum of Chiusi, Italy) different parts of which have been analysed by quantitative multiphase analysis using TOF neutron diffraction (ISIS, ROTAX): (1) bottom wall, (2) lateral wall, (3) handle and (4) repair patch. Different peak positions indicate different tin contents while different peak widths indicate different microstructures (centre).

More information could be extracted from Rietveld refinement analysis of the peak profiles (Figure 3) which were compared to the results from modern reference samples that have been produced under controlled conditions. Raw casting of the jug's handle is indicated by broad and structured bronze peaks, whereas the much narrower peaks of the wall suggest partial recrystallisation by mechanical and thermal treatment. These results are important because the manufacturing techniques of such small vessels are not yet entirely understood.

Looking into the interior of materials.

Neutron diffraction is particularly powerful for the analysis of the interior of materials, such as stackings of metal sheet, coins with coatings, or objects located inside sealed containers. There is further potential of neutron diffraction for investigating the volume textures and grain distributions of metal objects. Texture is a case study of its own and is an important characteristic for mechanically treated archaeological artefacts. The grain distributions in coins for example, could be used to discriminate authentic objects from forgeries or fakes, or to distinguish between differently worked coins.

Manufacturing history by texture analysis in Copper Age artefacts.

A recent example concerning the interpretation of metal textures is the analysis of Copper Age axes performed by neutron diffraction. Among others, the unique copper axe found together with the mummified body of the 5200 years old Iceman (Ötztal Mountains, Eastern Alps) was analysed (Figure 4). This is the only prehistoric axe ever found with the original handle and bindings. Full, non-destructive texture analysis has proven that textural information can be successfully extracted from the diffraction data irrespective of the shape of the object, and that the specific manufacturing history of each axe can be derived.

Figure 4: The copper axe of the Iceman (3200 BC) with its original handle and bindings.

Figure 5 shows the pole figures of a copper axe from Castelrotto (Italy), showing clear signs of thermal recrystallisation.

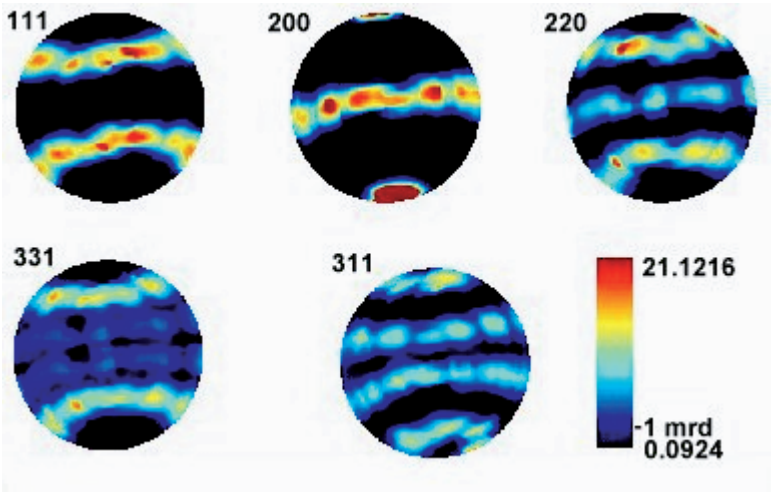


Figure 5: Experimental pole figures of a copper axe obtained by neutron diffraction at ILL.

VI. Forecast of novel opportunities at ESS

Neutron diffraction represents a very promising archaeometric tool and the knowledge of its potential in the examination of artefacts of nearly all shapes and materials in a truly non-destructive manner is still at a very early stage of exploitation by the relevant science community. The examples of neutron studies on artefacts and archaeological objects presented here are all very recent and some of the achievements are still far from the goals envisaged, mainly due to limitations of present neutron sources and instrumentations.

The unique capabilities offered by the ESS as regards intensity and the corresponding advancements in time-of-flight instruments will allow the simultaneous detection of weak and/or very subtle signals and will enable combined analyses of phase identification, phase fraction, microstructure and texture on both, very tiny and large objects. Referring to the examples given here, the ESS will allow neutron studies beyond current thresholds in the following areas:

The ESS enables both simultaneous and multi technique analysis.

- Phase and microstructural characterisation of stones and ceramics by investigating large suites of site-specific objects and, for comparison purposes, also of well-defined reference samples in a reasonable time and finally on a routine basis.
- Combined texture and microstructure analysis of metallic objects, e.g. the Iceman axe, in order to get complementary information on the manufacturing conditions by analyses of grain size and strain as well as preferred orientation of crystallites, again also on comparative samples obtained by different manufacturing processes.
- Texture analysis of precious and large or heavy objects in a complete stationary experimental set-up which is possible only at TOF-instruments with a wide three-dimensional detector arrangement surrounding the object.

VII. Instrument requirements in the field of stones, ceramics, glasses, metals and paintings

The investigation of artefacts and materials in the field of Cultural Heritage with respect to phase and microstructure investigations of stones, ceramics and metals needs both a High Resolution and High Intensity Powder Diffractometer as well as an Engineering Diffractometer for large objects and a High Q and/or Liquids Diffractometer for glasses. The Tomography/Radiography set-up is needed for the analysis of paintings and other artefacts. Surface studies will need a High Intensity Reflectometer and dynamical studies of small amounts of H_2O, CO_2 and SO_4 in stones have to be performed on a high intensity TOF-spectrometer.

6.6 Traffic and Transport

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Abstract
The world has become a smaller place as a combination of improved communications and transport. Projecting from the lessons of the past we can see ways in which we contribute to solve problems in traffic and transport by technological progress if ESS were available today. It will lead to better, safer, more economical and environmentally friendly designs through the uptake of new materials, an improved understanding and refinement of the manufacturing processes and a surer foundation for structural integrity assessments. The developments are divided into five main sections; engine/propulsion technology, transport structures, structural integrity and safety, lubrication, guidance & transport management technology. In all these areas, it is believed that the ESS will make a significant contribution.

I. Introduction

Transport has changed immeasurably over the last 20 years and contributes greatly to our quality of life. Many expect to travel on demand for business and make multiple holidays overseas for pleasure. This contributes to our wealth as well as a better understanding of each others cultures. The last 20 years have seen:

- a large increase in the number of cars, trucks, and aeroplanes,
- an increase in traffic congestion,
- a move away from public to private transport,
- an increase in atmospheric pollution and global warming.

For many the joy may be in travelling more than in arriving, nevertheless our quality of life is heavily dependent on arriving safely and on time. With increasing numbers of people and goods world-wide to travel we must meet important challenges:

- improved safety in transport,
- decreased travel times,
- lower transportation costs for both people and freight,
- reduced congestion at higher carriage and convenience levels,
- environmentally tolerant transport system.

"Progress in transport and traffic is reliant on improved and new materials and processes."

Rising energy prices and growing environmental awareness are intensifying the quest for materials with improved performance. High capacity, high energy efficiency, and safe traffic systems are required together with a reduction in their environmental impact.

Neutron scattering can help to unravel structural issues by standard techniques like powder diffraction for phase determination, texture and strain analysis or small angle scattering. In many of these fields major steps forward will be

made through ESS just because of the increased flux which translates directly into increased sensitivity allowing one to probe more realistic conditions. Also, this allows for a better spatial resolution and for systematic studies, needed to approach application relevant technological information on real size components.

In addition to issues on structural materials, functional units e.g. fuel management, traction control, active suspension, anti blocking system, or traffic management rely on device materials like semiconductors, lasers, piezoelectric crystals or telecommunication units, will play an increasingly important role and need to be engineered to demand. Neutron scattering can contribute at many levels in developing appropriate devices.

We have identified 5 main sections, where ESS could make a difference.

- Engine/propulsion technology
- Transport structures
- Structural integrity and safety
- Lubrication
- Guidance & transport management

II. Engine/propulsion technology

Higher efficiency in propulsion systems needs higher operating temperatures.

Fuel-efficient propulsion systems demand an increase in engine operating temperatures exploiting new materials and new manufacturing processes. Fast turbine powered ships will reduce freight costs, while fast trains will improve the linkage of major cities across Europe. New technologies are being considered like magnetic suspension trains and next generation aeroplanes. Also the year 2001 has seen the first tourist in space.

"Stronger, stiffer, hotter, lighter, …"

- *New superalloys:* superalloys provide the backbone of turbine engines providing remarkable strength up to 1.200°C and resistance against corrosion. Performance enhancement is primarily concerned with their micro-structural control, including the role of point defects, grain boundaries, precipitations, cavities, dispersions, or micro-cracks. At present SANS which provides information about these all features, is flux and resolution limited, ESS would enable microstructures to be monitored in real-time during heat treatment or in-service, or for spatial variations in microstructure to be mapped.

- *Ceramic engines/combustors:* due to the high temperature capabilities and high wear resistance, a ceramic built engine could run far more efficiently than today's engines. Present designs demand ductility and toughness. Advanced ceramics on the other hand are very strong but brittle. Neutron tomography enables engineers to relate defects to strength of materials leading to new design criteria for ceramic engines. However, because it relies on taking 100 s of radiographs to get a 3-D image, tomographs

taking 100 s of radiographs to get a 3-D image, tomographs can take a long time to acquire (tens of minutes to hours). ESS will radically shorten the time to acquire tomographic images.

- *Metal + ceramic composites:* the combination of ceramic fibres within a titanium alloy offers the designer the potential to save weight in highly loaded rotating structures, enabling integrally bladed ring structures (blings) to be considered as an alternative to traditional bladed discs. Only neutron diffraction can provide information about the stresses within these complex structures; ESS will allow real full-scale engineering structures to be examined.

Metal-ceramic composites offer prospects for weight saving in highly loaded structures.

Further key materials are hard magnets, catalytic converters, recyclable plastics and neutron scattering at the ESS will contribute to the development of advanced components.

Figure 1: The internal stresses within composite bladed ring prototypes such as this one can only be studied by neutron diffraction, however currently this can only be done over the latter stages of production due to the low flux of existing neutron sources.

III. Transport structures

Economic air travel is leading to the design of the A380, the largest passenger plane to date. Such large structures are demanding new materials processing techniques. One day, just as shipping has moved away from riveted designs so too will aerospace with innovative welded structures. Composite structures that have led to marked improvements in crashworthiness in racing cars may be transferred to family cars.

- *Riveted structures:* at $1 a rivet a 100,000 rivets are costly and a potential source of cracking in aircraft. Welded structures based on laser welding or friction stir welding need to be developed with the help of neutron strain measurements to limit stress and distortion, but currently strain mapping has insufficient resolution and is too time consuming.

"It is unlikely that tomorrows aeroplanes will be riveted – just as we have done for ships we must find better ways to join aeroplane structures."

- *Surface treatments:* coatings, paints, hardening and peening treatments are used for both lifetime and aesthetic reasons. Neutron diffraction and reflectivity can be used to accelerate their optimisation, but low fluxes limit studies of the former to fairly thick coatings – with the ESS this will not be the case, reflectometry on the other hand will benefit from higher flux when examining time dependent phenomena such as wetting.
- *New joining methods:* new friction based joining methods are cheaper and more flexible than existing fusion welding methods opening up new designs and leading to weight reduction and performance enhancement (see case studies below). The high resolutions and faster strain mapping capabilities provided by the ESS will accelerate process introduction and lead to safer designs through a knowledge of residual stress.

IV. Structural integrity and safety

Failure in transportation leads to the loss of many thousands of lives annually. The understanding of component failure is an inexact science. Failure is often stress driven and exploits underlying defects.

Figure 2: In many cases unexpected component or structural failure is costly or inconvenient, in the public transport sector (as well as the nuclear sector) it is commercially and socially unacceptable. Design of future very large bodied aircraft would benefit from better information about process and service stresses provided by new sources such as ESS.

- *Composite materials and laminated structures:* are being introduced into aerospace structures yet little is known at present about how they damage on impact and how this accumulates – neutron tomography allows this damage to be imaged and monitored as a function of bird strike for example but without ESS is too slow and of poor spatial resolution.

"Recent problems in the aerospace and rail industries have highlighted the premium on the safe design and operation of our transport systems."

- *Life extension*: many strategies are available for prolongating the life of components (cold hole expansion, ameliorative peening, repair strategies, etc). Neutrons see damage and stress levels evolve during service conditions and the ESS will enable these studies to be extended to more realistic in-service conditions and shorter time-scales.

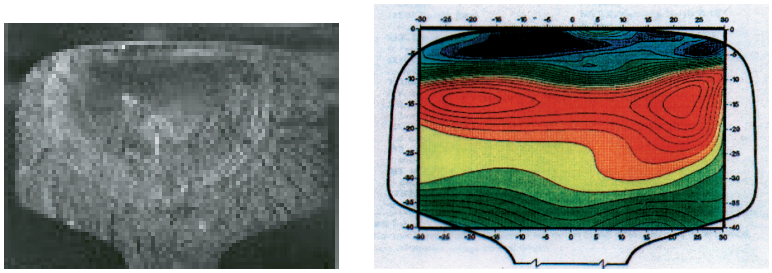

Figure 3: Tache Ovale is one form of rail failure and is caused by cracking from internal defects (left) under tensile stresses (see neutron map - right (red is tensile)).

V. Lubrication

Only recently has inelastic neutron scattering been employed to study lubricants under shear embracing the enormously wide time scales from the slow macroscopic flow to the rapid motion of sub-molecular units which occur on the nanoseconds level. Of particular importance for such investigations is the unique feature of neutrons to see the liquid dynamics with high precision even in heavy and complex environments characteristic for application relevant conditions.

"Friction between moving parts results in a significant loss of energy which is estimated to accumulate to 6 % of the gross national product of the USA."

- *Oils/fluids:* today's experiments employ unrealistically thick lubricating layers for sufficient signal. This constraint will be lifted with the increased flux of ESS and lubrication layers with a thickness typical for engineering gaps will come within reach. Such studies will provide significant information not available otherwise to optimise lubrication for reduced friction and wear and the effects of additives or coatings to the lubricant flow can be followed for application-relevant conditions.

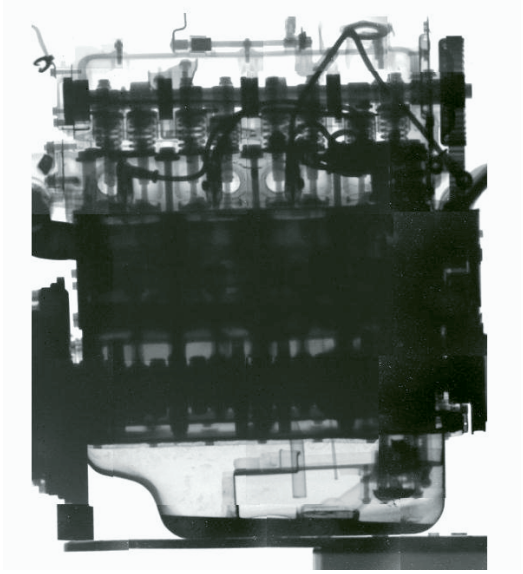

Figure 4: A FIAT 4 stroke engine rendered semi transparent by neutron radiography – with the ESS we will be able to monitor the performance of oils within engines.

VI. Guidance and transport management

The future will see an increase in the tools available to the travel from guidance systems to navigate unknown places to traffic management systems that reduce car accidents by controlling speeds, making drivers aware of other road users and automatically paying road tolls. Many rely on the development of new device materials involving Si, active polymers, glass, actuators, etc.

- *Magnetic sensors* based on thin film technology are widely used in automotive for ABS, position detectors, engine control and servo-systems. Polarised neutron reflectometry is a unique and powerful tool for thin film characterisation – the ESS will provide much higher polarised neutron fluxes enabling a wider range of devices to be studied.
- *Traction control, actuators and airbags* have all led to improvements in traffic safety and many of the functional materials at their heart would have benefited from the ESS for example by leading to improved electronic devices and sensors through control of structure.

"The amount of traffic will continue to increase – only better traffic management & guidance can prevent crippling congestion and reduced safety."

VII. Case studies

Friction joining

The next generation of higher temperature aeroengine alloys are particularly difficult to join by conventional welding techniques. New solid state joining techniques, such as inertia or linear friction welding, are a solution but require more accurate information about the generation of residual stresses so that their life can be reliably predicted. Linear friction welding for joining blades to discs offers significant weight reductions and will also enable to join dissimilar alloys and metals and at some stage even metals to ceramics.

Friction joining of aerospace superalloys.

A high flux neutron diffraction source will play a major role in this development, because only neutron diffraction enables in-depth measurements of the residual stresses in such full-scale components. Shorter counting times then envisaged at today's neutron sources are necessary in order to carry out systematic studies of welding parameters and post weld heat treatments.

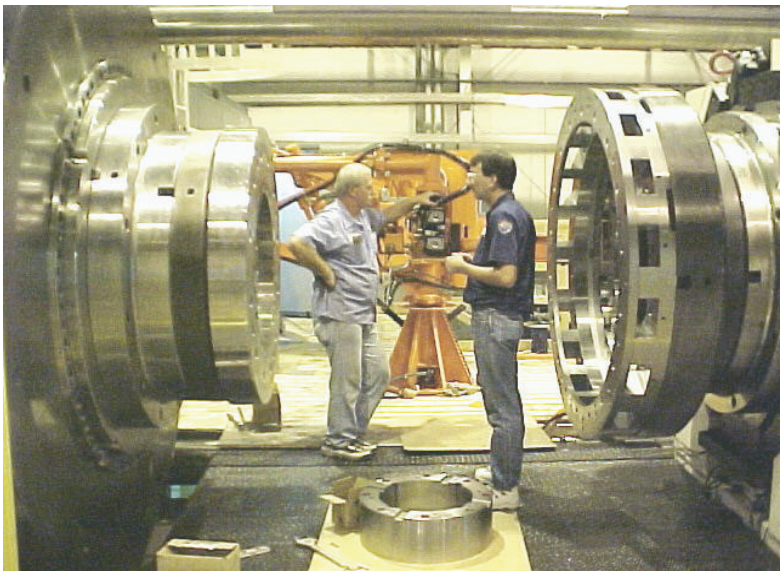

Figure 5: Inertia friction welding rig for joining turbine disks (above), linear friction welding will be used to weld blades to disks to form blisks giving lighter weight and improved performance over existing slotted blade/disk assemblies (below).

Failure mechanism of railway wheel

Neutron strain measurements of train disaster.

Recent railway disasters caused by material failure of wheels and rails illustrate that a better understanding of contact fatigue failure is needed. The life-time of railway wheels is limited by the accumulation of plastic strains and the formation of micro-cracks due to cyclic loading. Residual stresses are the result of local micro-structural changes especially of local cold work hardening. The locations of cold work hardening depend on the manufacturing engineering and they are re-distributed by interaction between rail and wheels.

Figure 6: Recent railway disaster in Germany.

Neutron Strain measurements on a sector of an Intercity Express broken railway wheel show that residual stresses play a vital role on failure mechanism on such a components [M. Grosse, U. Sthur, M. Ceretti, L. Köszegi, Journal of Neutron Research **9**, 489-493 (2001) and LLB Scientific Report 1999-2000]. Only neutrons enable measurements on such big components. The high flux at the ESS will enable one to look at complete wheels rather than cut sections as shown below.

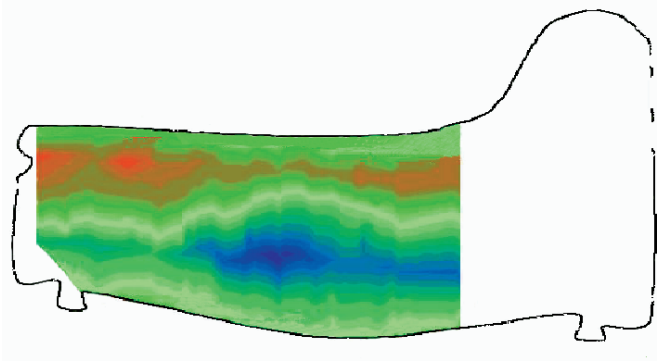

Figure 7: Compression zones (red) and dilatation zones (blue) in a wheel ring of the ICE train. Contact to the rail is at the upper surface. It is the tensile stresses that cause the failure recorded for the ICE.

Wide chord fan blades

The large fan blades necessary for the Airbus A380 engines require Warren girder or honeycomb supports with Ti skins. The manufacture of these structures is very complex. An understanding of the stress state at the fan root would complement existing lifing estimates, while a knowledge of the integrity of the internal structure would help us to understand the damage micromechanics after a bird strike. All this could be obtained non destructively on the ESS. This would be achieved by the improved spatial resolution of tomographic imaging and strain measurement over that currently achievable.

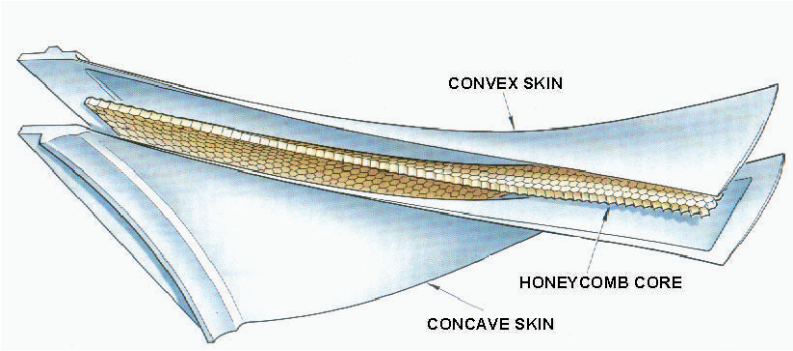

Figure 8: Stresses at the root and between the internal structure and the skin of wide chord fan blades can only be measured by neutron diffraction.

VIII. Instrument requirements

The instruments required are:

- Engineering diffractometer: *for high resolution fast strain mapping of large 3-D components and structures.*
- Tomography/radiography: *for high resolution defect and structure mapping of large structure.*
- High intensity small angle scattering: *for microstructural and defect characterisation in metals, polymers and ceramics. With ESS it may be possible to map gradients in these microstructural parameters either in space or time.*
- High resolution back scattering: determination of *full 3-dimensional macroscopic flow pattern of lubricants by Doppler effect.*
- Cold chopper*: simultaneous observation of slow microscopic molecule dynamics and macroscopic flow dynamics of liquids under shear*.
- Thermal chopper: *observation of faster molecule dynamics of liquids under shear*.
- Powder diffraction: *for structural characterisation and texture determination, for the study of the transitions of functional materials and devices and for the study of magnetic materials.*
- High resolution NSE: *to characterise motions and loss mechanisms in polymer blends relevant for the development of tires.*
- High intensity reflectometer: *to characterise the structure for coatings and thin layers including magnetic layers, as used in devices.*

6.7 Sustainable Development, Clean Technologies and Environmental Systems

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Abstract
Sustainable development will guarantee safe, affordable and clean energy for future generations. In order to arrive at this goal, significant improvements of present energy storage and conversion processes are needed. Lower emission levels of pollutants can be achieved using hydrogen as an energy carrier and through the development of high performance batteries and fuel cells. Clathrates represent a large resource of carbon hydrates but their utilisation depends on a better knowledge of their physical properties. Catalysis is essential for the development of clean and efficient chemical processes. Removing impurities in fuels, or reducing the level of exhaust emissions from motor vehicles, can be facilitated with new catalysts. Finally, burial of nuclear wastes must be achieved in a safe and affordable way.
The large flux and new capabilities provided by the ESS will allow a range of important new experiments. In-situ experiments in real materials and devices will be performed at shorter time scales in order to follow faster processes. Examples are structural phase transformations giving insight to aging properties, ion diffusion in electrode materials and fuel cell membranes, identification of intermediate products in working catalysts, and examination of spent nuclear fuel.

I. Introduction

Renewable energy.

Environmental impact.

Hydrogen storage.

Fuel cells and batteries.

Clathrates.

Catalysts.

Safe waste disposal.

The steadily increasing energy requirements of the world imply a concomitant increase of pollutants and greenhouse gases unless renewable and environmentally benign energy sources are used. In addition, portable energy sources will be needed to ensure mobility and autonomy, and will require batteries or fuel cells. Renewable energies are likely to be produced centrally but then transportation, energy conversion and storage have to be developed. Hydrogen appears to be the best choice for an energy carrier of the future. This gas can be used directly in fuel cells to produce electricity. Clathrates form the largest deposit of hydrocarbons known to date. They are environmentally relevant for issues like the green house effect and they could contribute to energy resources. Materials production in chemical plants can best be performed using clean and efficient processes with respect to waste products and energy consumption. Many of these processes rely on catalysts that need considerable fine-tuning of process parameters. Safe storage of nuclear waste on a long time scale requires chemical stability of the host matrix materials, which can be monitored by neutron diffraction on minute quantities.

II. Contributions from ESS

Neutron scattering at the level provided by the ESS will make possible the characterisation of materials and processes in their complex environments. Examples include experimental efforts, in order

- to monitor *in-situ* structural changes occurring in energy storage and conversion devices (batteries and fuel cells)

involving light elements like H and Li;

- to study the microscopic diffusion of protons in hydrides or exchange membranes and oxygen ions in fuel cell electrolytes;
- to investigate physical and structural properties of gas clathrates;
- to localise active sites in catalytic materials;
- to follow in-situ catalytic reactions with inelastic scattering;
- to characterise radioactive materials related to nuclear waste materials available in very small amounts;
- to understand the formation and growth of aerosol particles using small angle scattering (climate change);
- to follow reactions occurring in supercritical CO_2 by small angle and inelastic scattering;
- to study the texture and stress-strain behaviour of rocks to help predict earthquakes and volcanic eruptions [1].

Impact of neutron scattering.

This list, which is not exhaustive, shows that the high flux and new instrumentation provided by ESS will allow neutron scattering to make major contributions in the key areas which are energy storage and conversion, and clean chemical processes. Only with the ESS can one envisage in-situ characterisation of these systems in their complex environments. Some examples will now be detailed.

III. Examples for the impact of ESS

Batteries

New rechargeable battery materials should provide a high energy density, be safe for the environment and cheap, in order to facilitate large-scale application of renewable energy. To achieve these goals, many research efforts are devoted to modern rechargeable battery materials, mostly nickel-metal hydride (Ni-MH) and Lithium ion (Li-ion) ones.

Batteries for autonomy and mobility.

Metallic hydrides (MH) are extensively studied with regards to their ability to store reversibly large amount of hydrogen. Electrochemical storage has been successfully developed to produce rechargeable Ni-MH batteries.

Lithium ion batteries are also most promising. The active materials are layered inorganic compounds for the cathodes and graphitic carbons for the anodes. These materials are intercalation compounds, capable of reversible insertion of Li ions.

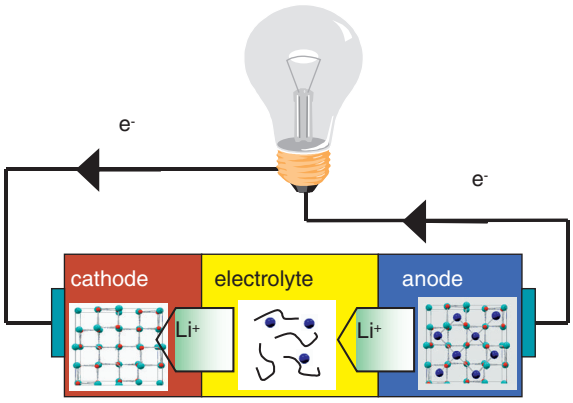
Phase transitions in Li intercalated materials.

New compounds are being developed continuously such as magnesium-based materials for Ni-MH batteries or titanium oxide and boron phosphate for Li-ion batteries.

Electrolytes become increasingly complex. The discovery that the blending of polymer electrolytes with nano-sized particles enhances conductivity [2] opens many new directions for optimisation of electrolytes. In this field neutron scattering has

Improving diffusion within electrolytes.

made a significant contribution to the understanding of the process [3,4] and this, in turn, will lead to higher performance.



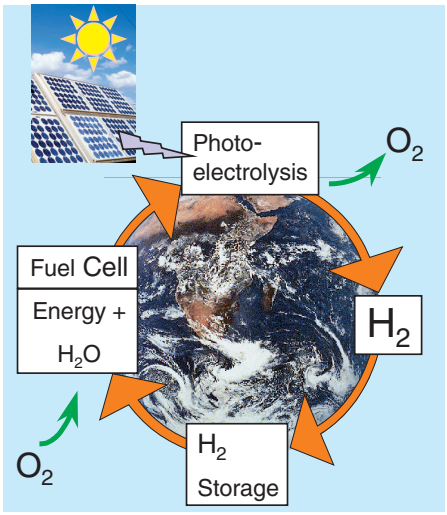
In-situ investigations of electrode materials to design improved batteries.

Figure 1: Schematics of a rechargeable Li-ion battery. At the ESS the charge and discharge of the battery could be monitored under real operating conditions, which will help improving battery lifetimes.

The key feature for rechargeable batteries is the long-term stability under thousands of cycles. The high flux of ESS and the favourable characteristics of neutrons would make possible the *in-situ* monitoring of the structural changes of the electrode materials under real charge-discharge conditions.

Hydrogen storage

Hydrogen is considered to be an ideally clean carrier of energy. In order to realise a future hydrogen based energy economy, better hydrogen storage is required in a compact, light, safe and affordable manner.



Hydrogen energy cycle.

Figure 2: The conversion of sunlight and water into hydrogen and oxygen, hydrogen storage, and utilisation of hydrogen in fuel cells, form a clean energy cycle. Neutron scattering uniquely shows the location and motion of hydrogen in the new materials that will have to be discovered to realise the hydrogen energy cycle.

There is currently considerable promise for hydrogen storage in compounds based on V, Ti and Mg and nanostructured

Metal hydrides.

carbon [5] and blends of such materials. Also some of the lightest elements in the periodic table like Li, B, Na or Al form stable ionic compounds (alanates) with hydrogen that may be applicable as reversible hydrogen storage material.

Carbon nanostructures and alanates.

Knowledge of the hydride structural properties is obviously needed and cannot be achieved without neutron diffraction.

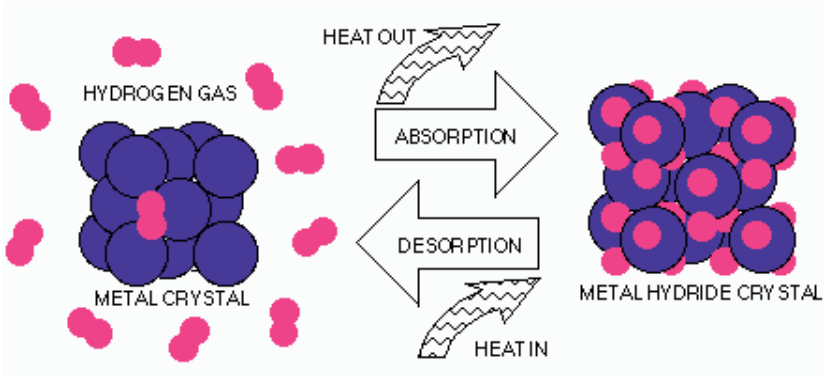


Figure 3: Metal hydride electrode observed by in-situ neutron diffraction. High rate charge-discharge cycling induces phase transformation and internal strains due to lattice parameter mismatches.

The diffusion path and coefficient of hydrogen, which determines the kinetic of the reaction, can be obtained from quasielastic neutron spectroscopy. Moreover, due to the reversible properties needed for these materials, aging properties must be carefully evaluated through *in-situ* study of intensive cycling processes.

Aging properties.

Fuel cells

A fuel cell can produce electricity with high efficiency from hydrogen gas and oxygen, with water as the only waste product. At the core of fuel cell technology is the development of proton or oxygen ion conducting electrolyte membranes.

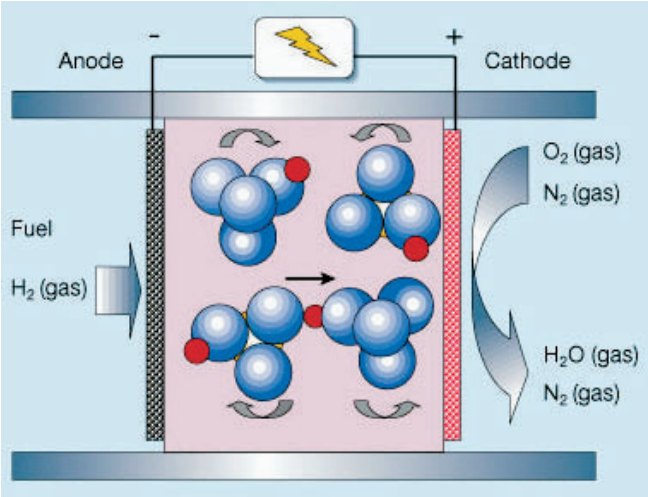
The most successful 'wet' electrolyte so far is NAFION®, a polymer that contains many SO_3 side groups to which H_3O^+ can be attached [6]. The diffusion of H^+ or H_3O^+ through the material is aided by the motion of the polymer side groups and backbone, as is the case in lithium ion electrolytes. Recently it was discovered that solid acids that show superprotonic conduction can be utilised as fuel cell membrane [7]. These 'dry' materials do not need water for the transport of protons and are therefore not limited to temperatures below 100°C.

Superprotonic conductors as electrolyte membranes are a key element of fuel cell technology.

Neutron scattering at ESS will make it possible to determine the structure and dynamics of the (molecular) sites that hydrogen visits, and to study the diffusive motions of hydrogen. Such studies will be possible *in-situ* and with varying parameters like humidity, oxygen content of the air, and the presence of polluting gases like CO in realistic concentrations. In the case of solid oxide fuel cells (SOFC), only high flux neutron beams could give information about the

ESS will allow in-situ real time observations of proton and oxygen transport.

diffusion of the oxygen ion within the electrolyte at temperatures around 900°C.



Conduction of H^+ or O^{2-} ions.

Figure 4: Principle of a fuel cell. After the hydrogen gas is catalytically dissociated at the anode the proton enters the membrane material consisting in this case of the superprotonic conductor $CsHSO_4$.

Clathrates

Gas clathrates are widespread geological materials relevant for:

- Energy resources: they are estimated to host the largest proportion of natural gas, mostly methane, in the shallow earth (about 7 times the resources available in gas deposits hosted in sedimentary rocks and shallow sea sediments).

Methane in ocean sediments.

- Climatic changes: they act as buffers for CH_4 in the oceans; sea-level changes affect the release of gas in the atmosphere via pressure-controlled phase transitions and producing short term green-house effects.
- Marine geo-hazards: phase transitions induced by slight changes of ocean temperature and salinity in the clathrate layer may cause instabilities in the oceanic platform sedimentary deposits with effects ranging from disruption of communication cables to triggering tsunamis.

Environmental impact of clathrates.

Crystallographically, clathrates have atomic structures made up by water cages entrapping guest gas molecules. Such structures pose relevant physico-chemical problems, which need very fine investigative tools for the correct interpretation of their behaviour under different pressure and temperature conditions. Detailed investigations of the crystal chemistry and dynamics of gas hydrates are needed to clarify the relationships between microscopic and macroscopic properties. Vibrational dynamics of the entrapped molecular species and the anharmonic behaviour of the atoms in the structure are far from reach with present-day neutron sources. Flux and resolution need to be substantially enhanced in order to obtain significant diffraction and spectroscopic data from these compounds, and such studies will only begin seriously at the ESS.

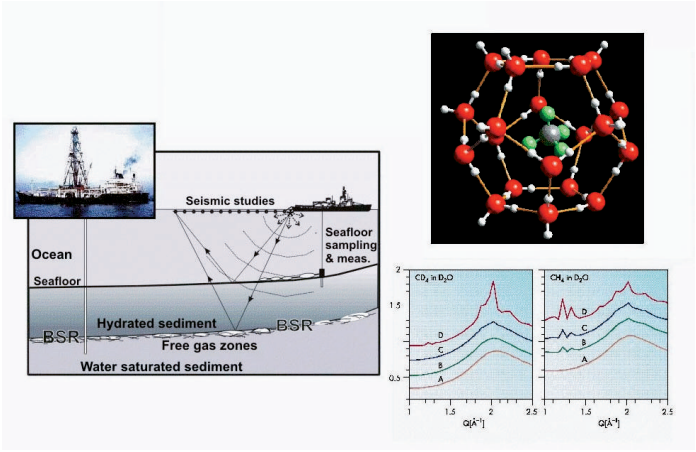


Figure 5: Methane clathrates are widespread in ocean sediments.

Catalytic processes

The location of active centers, e.g. protons (acid sites) or metals (redox sites) in crystalline oxides is crucial in catalysis. These sites must be identified in order to understand the reactivity and selectivity of the reactions occurring in these materials. For example, the siting of titanium in the molecular sieve Ti-silicalite (TS-1), which is an oxidation catalyst, has been recently the subject of several powder neutron diffraction studies. With the available instrumentation, it has been found extremely difficult to determine the multiple Ti site substitution, out of the 12 possible ones (Figure 6). The site distribution varies according to the different studies [8-10]. Clearly, the task of finding the Ti sites is complicated by its small concentration and by silicon vacancies, but it illustrates the need for a more intense neutron source in order to locate unambiguously the active centers.

Understanding catalysts through the observation of their active centers and the processes thereon.

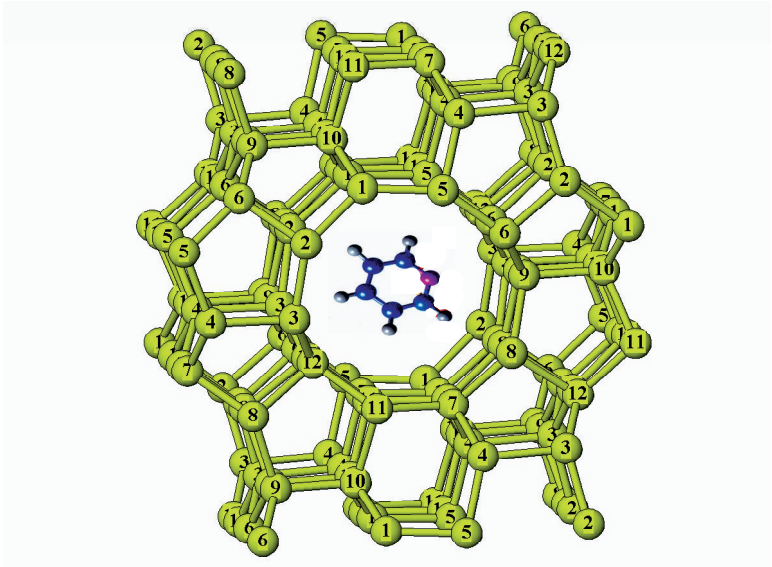


Figure 6: With the high flux and new instrumentation that will be available at ESS, it will be possible to locate more precisely the active sites in catalysts, and to follow in-situ catalytic reactions in order to identify the reaction intermediates.

The action of catalysts is often followed by the vibrational fingerprints of the reactive species. At present, experiments in vibrational spectroscopy must be done at low temperatures to reduce multiphonon contributions, but real catalysts need to be examined at room (or high) temperature. Multiphonon contributions should be reduced by using low momentum transfers. For the first time, ESS will allow in-situ catalytic reactions to be followed and reaction pathways to be identified. For example, the oxidation of n-butane to maleic anhydride is the subject of extended research, both for industrial and fundamental reasons, as this reaction is very complex, with eight abstractions of H atoms, three insertions of O atoms, and subsequent electron transfers. Whereas other spectroscopic techniques like Raman are sensitive only to the bulk of the active catalyst, it would be possible at the ESS to identify the different intermediates involved in the reaction, in particular to determine whether the reaction proceeds via an olefinic (1) or alkoxyde (2) pathway (Figure 7).

ESS will enable to follow in-situ catalytic reactions.

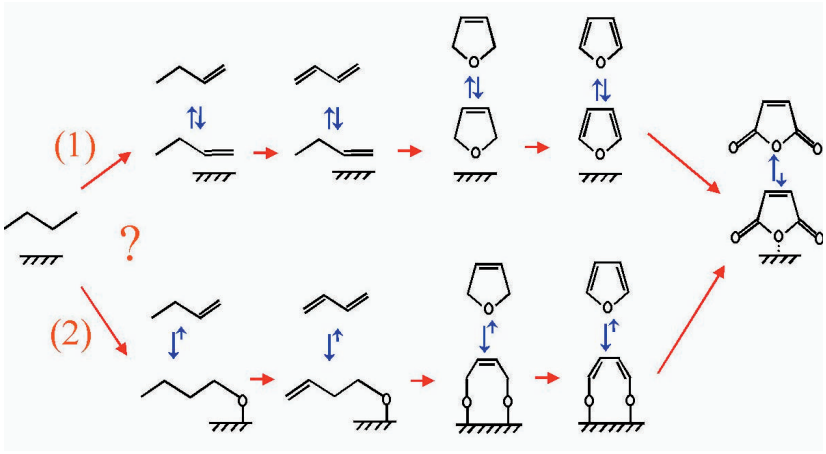


Figure 7: Two possible pathways proposed in the literature for the mild oxidation of n-butane to maleic anhydride on phosphorus vanadium oxide catalysts.

Nuclear waste

Nuclear waste presents a legacy that will only decay over many generations. Various policies are presently followed in different countries for the disposal of waste. All are predicated on a knowledge base of irradiated material that is far from satisfactory or complete, especially considering the heterogeneous nature of the waste. Many different tools, but not yet neutrons, are used in the characterisation of nuclear waste. The main reason neutrons are not used being the difficulty in examining sizeable samples of such irradiated materials at user facilities. However, the brilliance of the ESS will allow powder diffraction experiments to be performed on samples as small as a few mg, thus allowing a new tool to be brought to bear on this important problem. The time-of-flight method has a distinct advantage as neutron induced fission can be separated from the diffraction pattern.

Safety issues.

Questions that need to be addressed are the phases present as a function of distance from the barrier (either the cladding

in the case of direct burial, or the container in case of reprocessed fuel), and, in particular, the metal to oxygen ratio of this material. Although x-rays can (and do) establish the crystal structure, they *cannot* determine the metal to oxygen ratio, and both corrosion and reactivity potentials depend on this ratio. Following this ratio as a function of distance from the cladding (or barrier) characterises the waste in a way unavailable today, and might well become a crucial aspect of quality control and monitoring of actinide waste. Small angle scattering may also play a role in understanding the porosity and the nature of the interfacial region.

Long term stability.

After the fuel is processed and the actinides removed the remaining material is often stored in glass form. Here again, neutron studies can play an important role in characterising the encapsulated material and understanding the binding of the actinides into the glass, which is critical for the long-term storage of the radioactive waste. Once again, the brilliance of the ESS will allow *real* materials to be studied.

IV. Instrument requirements at ESS

The instrumentation required covers many aspects of neutron scattering: elastic, quasi-elastic, inelastic, and small-angle. This includes:

- High resolution powder, chemical single crystal, magnetic, and single pulse diffractometers.
- High energy, thermal, and cold chopper spectrometers.
- Back-scattering spectrometers.
- High intensity SANS.
- High resolution neutron spin-echo.

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6.8 Neutrons and the Origin of the Universe

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Abstract
Neutron physics relates to a number of hot topics from the field of particle physics and cosmology. Experiments with neutrons are testing the very early stage of the universe. Examples are neutron interferometry experiments, a search for neutron-antineutron oscillations, a search for a nonvanishing electric dipole moment and measurements of neutron decay parameters.

I. Introduction

Not much is known about the early stage of our universe. We think that all started dense and hot with the so called "big bang"; since then the universe has been expanding and has been cooling to the situation we find today. Most theories about this early stage of the universe expect a high degree of symmetry, e.g. no difference between left and right or between particles like fermions and bosons. This situation of high symmetry is very different from what we find today, where only left-handed neutrinos can be found and differences between particles are obvious. We explain this difference between now and before with the assumption that the universe evolved through different phase transitions, where symmetry breaking occurred at different stages.

Phase transitions in the early universe:

- ***Planck***
- ***GUT transitions***
- ***Inflation***
- ***Electroweak transition***
- ***Nucleon freeze-out***
- ***Atomic freeze out***
- ***Galactic freeze out***

Hence, our understanding of the development of the universe is based on the knowledge about phase transitions (see Figure 1), a subject that has been studied in all its variety with neutron experiments vigorously over the past 30 years. Discussions about cosmology start at the Planck scale about 10^{-44} s after the start of the universe. At 10^{-36} s we have grand unified theory phase transitions, the electroweak phase transition takes place after about 10^{-10} s, the nuclear freeze-out after about 1 s and the atomic and galactic freeze-out after 10^{5} y.

II. Very early stage: Planck scale, grand unified theories

Planck scale

We discuss different stages of our universe and start at the Planck scale, the length scale of quantum fluctuation of space time geometry. Discouraging for experimentalists in the past was the fact, that the Planck length is 10^{-35} m. This is the scale where one expect to see something of interest which is twenty orders of magnitude smaller than the neutron diameter of ~ 10^{-15} m. Even the Large Hadron Collider (LHC) at C.E.R.N. will just be able to probe distances at a 10^{-19} m level. Fortunately, new theoretical efforts are under way. New theories on string theory and multidimensions cause gravity to reach Planck length well above 10^{-35} m. „Extra dimensions are wrapped into big cycles of at least 10^{-14} m radius and perhaps

"Extra dimensions are wrapped into big cycles of at least 10^{-14}m radius and perhaps as enormous as a millimeter."

Arkani-Hamed

as enormous as a millimeter“. If large extra dimensions existed, we would find new physics at the Large Hadron Collider at C.E.R.N. like string vibrations and mini black holes.

Deviations from Newton’s gravity law in the 1 µm to 100 µm range should also be detectable with suited experiments e.g. with neutron interferometry or spectroscopy on the sub-peV scale. In any case, as we have no means to reach the Planck scale of 10^{19} GeV or 10^{-35} m, this symmetry must be inferred from low energy measurements, be this at 100 GeV in present day’s “high energy” experiments, be this at 10^{-20} eV in present days “low energy” experiments”.

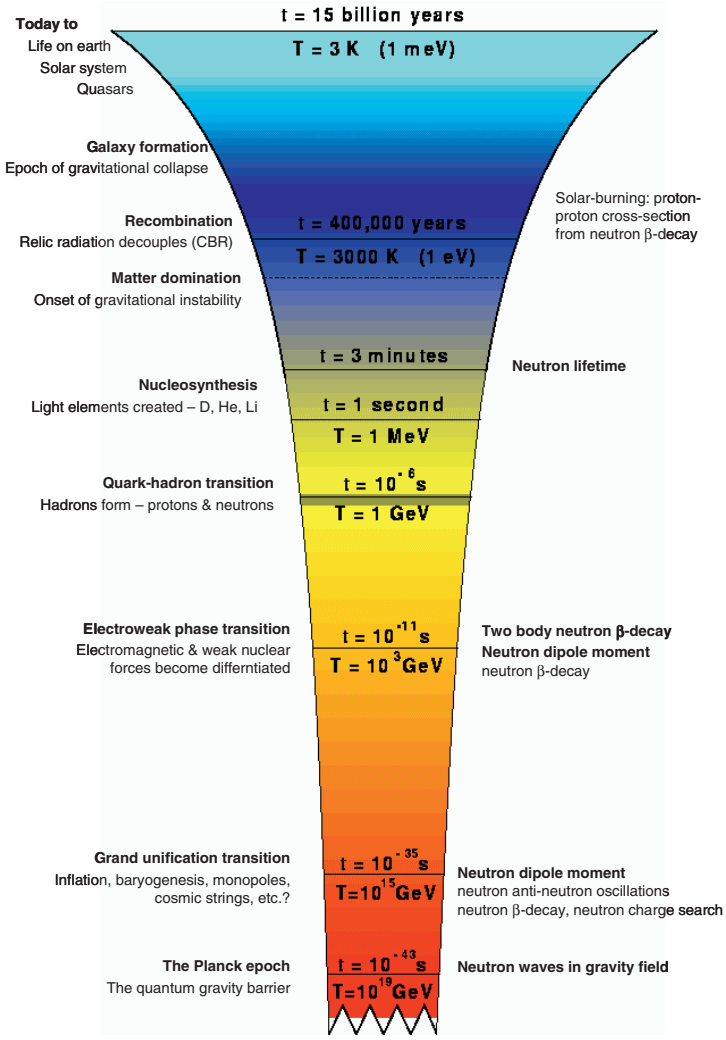


Figure 1: Development of the universe. Left: phase transitions and evolutionary steps. Right: neutron contributions to their clarification.

Grand unified theories

The basic idea of grand unified theories (GUT) is that the three forces – the strong, the weak and the electromagnetic force – are actually components of one single unified force. An underlying symmetry relates one component to the others so that all is described by a single force law. The same symmetry

implicates that particles like electrons, quarks and neutrinos are basically the same. On the other hand we know that these particles nowadays behave very differently. Thus, the theory is constructed such that the symmetry is spontaneously broken in the present universe. The detailed mechanism of symmetry breaking is related to the analogous mechanism in condensed matter phase transitions, a complicated interplay between large numbers of electrons and atomic nuclei. Thus, particle physicists profit from models developed in condensed matter physics.

Grand unified theories also bring new features to neutron physics which have drastic new implications. To give an example, many grand unified theories foresee a conversion of a free neutron into its antiparticle, an antineutron. But we have no indication for neutron-antineutron-oscillation from experiments so far. The present experimental limit for neutron anti-neutron oscillations is $\tau > 10^8$ s. With the search for a nonvanishing electric dipole moment neutron physics also directly test grand unified theories. The experimental limits are close to the predictions from supersymmetry theory.

In grand unified theories, there are serious cosmological problems associated with problems like magnetic monopoles or domain walls. The inflationary universe model provides a simple and elegant solution to these problems.

Experiments with neutrons testing this very early stage of the universe are indicated on the right hand side of Figure 1. Examples as mentioned are neutron interferometry and peV-spectroscopy for gravity tests; a search for neutron-antineutron oscillations, a search for a nonvanishing electric dipole moment and measurements of neutron decay parameters for tests of grand-unified-theories.

III. The matter-antimatter asymmetry and the neutron dipole moment

At this stage, a serious problem becomes evident: Where is the anti-matter?

With the electroweak phase transition, we leave the field of speculation and enter a region we can access directly with laboratory experiments on earth. Matter is built from two types of fundamental fermions, called quarks and leptons, appearing in three particle families (see Table 1). The Quarks occur in several varieties or flavours labelled u = up, d = down, c = charm, s = strange, t = top and b = bottom. The baryons condensate out of the quark-gluon plasma and the strong interaction glues the quarks together to neutrons and protons. Neutrons are built from two d-quarks and one u-quark, whereas protons are built from two u-quarks and one d-quark: ddu = n (neutron), duu = p (proton).

Table 1:
Elementary particles: Leptons and Quarks in three families.

First family	Second family	Third family
Quarks		
u	c	t
d	s	b
Leptons		
ν_e	ν_μ	ν_τ
e	μ	τ

At this stage, a serious problem becomes evident: Where remains the anti-matter? Our big bang theory presumes so far that equal amounts of matter and antimatter were created in the primordial explosion. In the subsequent process of annihilation of matter and antimatter only very few heavy particles ("baryons") and an equal number of antiparticles from this early period could survive. Our mere existence contradicts this expectation; there remained about 10^8 times more baryons in the universe than predicted and almost no antibaryons have survived. So far, the only viable solution of this problem is the violation of charge-parity symmetry (CP) which, on all reasonable expectations, is equivalent to a violation of time symmetry (T) that could have led to a small excess of particles before the annihilation stage. The most direct access to these questions lies in the detailed investigation of neutron decay parameters or in measurements of its electric dipole moment. Electric dipole moment (EDM). The existence of an EDM violates time reversal and also charge parity (CP) symmetry.

The most direct access to charge parity symmetry breaking lies in the detailed investigation of neutron decay parameters or in measurements of its electric dipole moment.

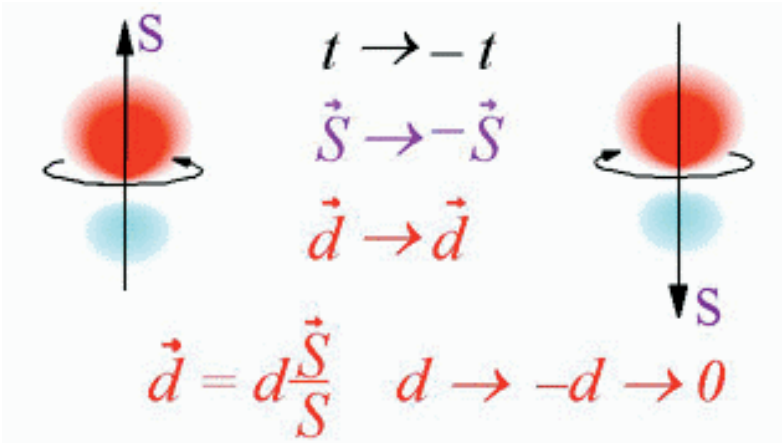


Figure 2: Time reversal operation for a neutron: under time reversal the direction of the spins changes, since it is an axial vector, while the charge distribution does not change. The electric dipole moment, which is a vector, has to be parallel to the spin, since it is the only available vector in the rest frame of the particle. Therefore, if time reversal is a good symmetry, the electric dipole moment must be zero.

EDM measurements started in the fifties and increased their sensitivity by one order of magnitude every seven years. Current theories of the baryon asymmetry of the universe is related to an EDM of about 10^{-28} e cm, a limit that is accessible with one flagship experiment for the ESS. The current upper limit is $6 \cdot 10^{-26}$ e cm.

IV. The left-handedness of the universe and the neutron β-decay

Parity is maximally violated in weak interaction. Only left-handed fermions take part in the interaction. Most Grand Unified Theories, however, start with a left-right symmetric universe, and explain the evident left-handedness of nature through a spontaneous symmetry breaking caused by a phase transition of the vacuum, a scenario, which, if true, would mean that the neutrinos today should carry a small right-handed component. Although limits on the right-handed currents have been derived from free neutron and muon decay experiments, what is really needed is a clear-cut "yes" or "no" experiment. Such an flagship-experiment, planned for ESS, is the two-body β-decay of unpolarised neutrons into hydrogen atoms and antineutrinos. What is so interesting about this decay is that one of the four hydrogen hyperfine states cannot be populated at all if the neutrinos are completely left-handed. A non-zero population of this sub-state would, therefore, be a direct measure of a right-handed component.

What is really needed is a clear-cut "yes" or "no" experiment. Such an flagship-experiment, planned for ESS, is the two-body β-decay of unpolarised neutrons into hydrogen atoms and antineutrinos.

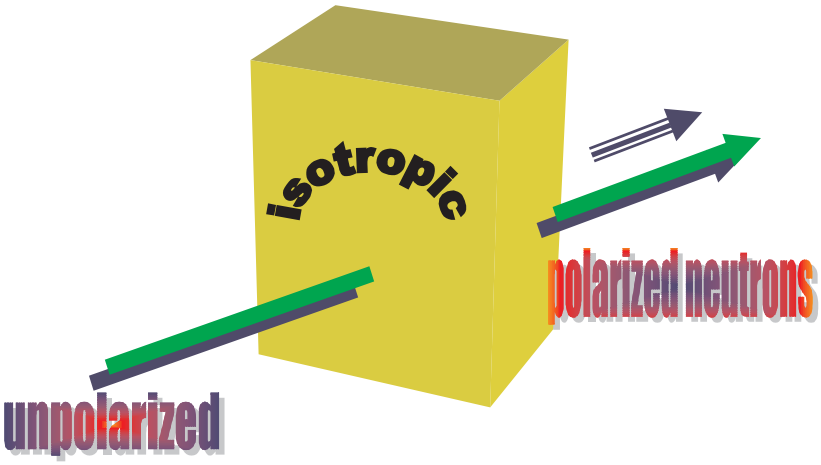


Figure 3: Polarisation of a neutron beam passing isotropic matter. The polarisation arises as a consequence of the left handedness of the weak interaction.

The most beautiful manifestation of parity violation occur in low energy neutron physics. An originally unpolarised beam of slow neutrons passing a material which is isotropic in all its usual material properties will show eventually a non-negligible polarization of up to several percent along its direction of propagation. Even a non specialist can clearly see that

something is wrong because all our senses of symmetry are violated. Of course, this effect is due to the fact that there is nothing like an isotropic material, because, under the weak interaction, all matter is left-handed.

Neutron decay experiments are quite competitive with the direct searches for right-handed components in high energy collider experiments. Under certain assumptions, best limits for one parameter, the 'left-right mixing angle', comes from neutron physics. The best limits on the mass of a 'right-handed W-boson', however, comes from neutrinoless double-β-decay.

V. Neutron life time and primordial "big bang" nucleo synthesis

About a second after the weak interaction drops out of equilibrium, we can follow what a gas of interacting baryons (protons and neutrons) does when the universe expands and cools. At this time nuclei can survive and neutrons and protons are converted into deuteron, triton and He. 100 s later, Big Bang nucleosynthesis made helium with traces of deuterium and Lithium. The protons left over remain as hydrogen. The primordial mass fraction of He is ~ 0.25 % and agrees well with the observed helium abundance in our universe. The theory of primordial Big Bang nucleosynthesis is considered to be very powerful since all inputs entering the calculations are known from laboratory experiments. In the past 15 years, finer details of the results have been taken into consideration. For example, the more types of relativistic particles exist, the greater is the energy density and as a consequence, the faster is the cosmological expansion rate, yielding a higher neutron to proton ratio. A higher neutron to proton rate ends up in more helium. On the other hand, a short neutron lifetime lowers the neutron to proton ratio thus yielding less helium. As a consequence, neutron lifetime measurements together with the observed helium abundance have made a definite prediction about the number of particle families. The formal statistical result provided a reasonable fit to three particle families but making a fourth neutrino family exceedingly unlikely. After this result, particle accelerators were beginning to probe to the cosmological level of sensitivity. At C.E.R.N., the existence of three particle families has been brilliantly verified with a measurement of the decay width of the Z-boson.

The power of primordial Big bang nucleosynthesis is that all inputs entering the calculations are known from laboratory experiments.

Neutron lifetime measurements together with the observed helium abundance have made a definite prediction about the number of particle families.

More information can be obtained from neutron β-decay. A neutron decays with a half-life of about 10 min. into a proton, an electron and an electron antineutrino. In this decay a d-quark is converted into an up quark via W exchange. Observables are the lifetime and spin and momenta of electron and proton. As mentioned before, neutrons are built from two d-quarks and one u-quark, whereas protons are built from two u-quarks and one d-quark. Thus, ordinary matter is made exclusively from up and down quarks, whereas the other quark flavours are observed in collider experiments.

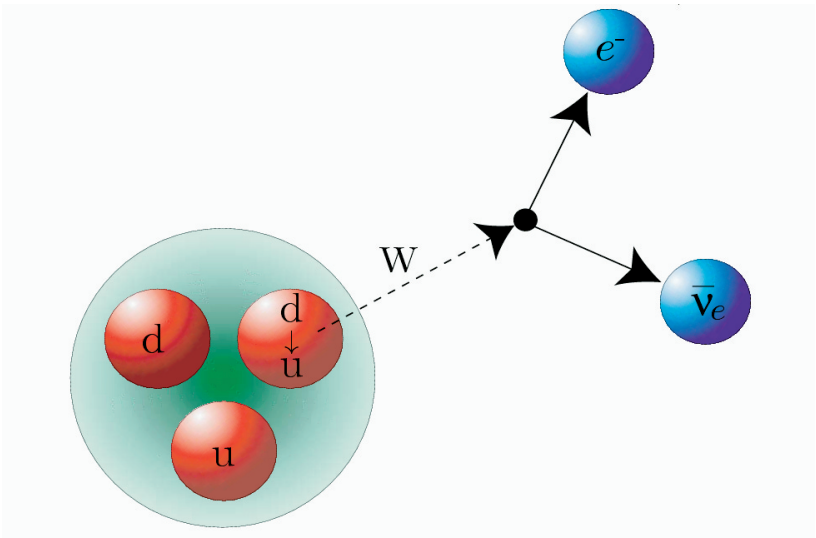


Figure 4: Neutron β-decay, where a d-quark is converted into an up-quark via the exchange of a W-boson.

In the neutron decay, the decaying down-quark does carry small contributions from strange and bottom quarks. On the other hand, a decaying bottom quark has a small contribution from a down and a strange quark. Therefore, the quarks being involved in the process of weak interaction do mix and the mixing is expressed in the so-called CKM-matrix. The Standard Model of elementary particle physics requests that the mixing ends up in a zero-sum, in other words, every quark gives as much as it takes in this mixing (the CKM-quark-mixing matrix has to be unitary). Now, the first decisive element of this matrix has been derived from neutron decay data in such a way that the unitarity test can be performed based solely on particle physics data. Much to our surprise, with the neutron-decay data the zero-sum reveals a significant deviation Δ. So far, the first entry to this matrix has been taken from nuclear physics also indicating a slight problem with the unitarity of the CKM-matrix.

Finally, Table 2 summarises the standard model parameters as well as observables which relate to new physics beyond the standard model, and shows how neutron physics is involved. Neutron physics is not only involved in the demystification of the origin of the universe but also carries very significant messages for cosmology, astrophysics and particle physics. Today all semileptonic cross sections used in these fields are based on the neutron decay data.

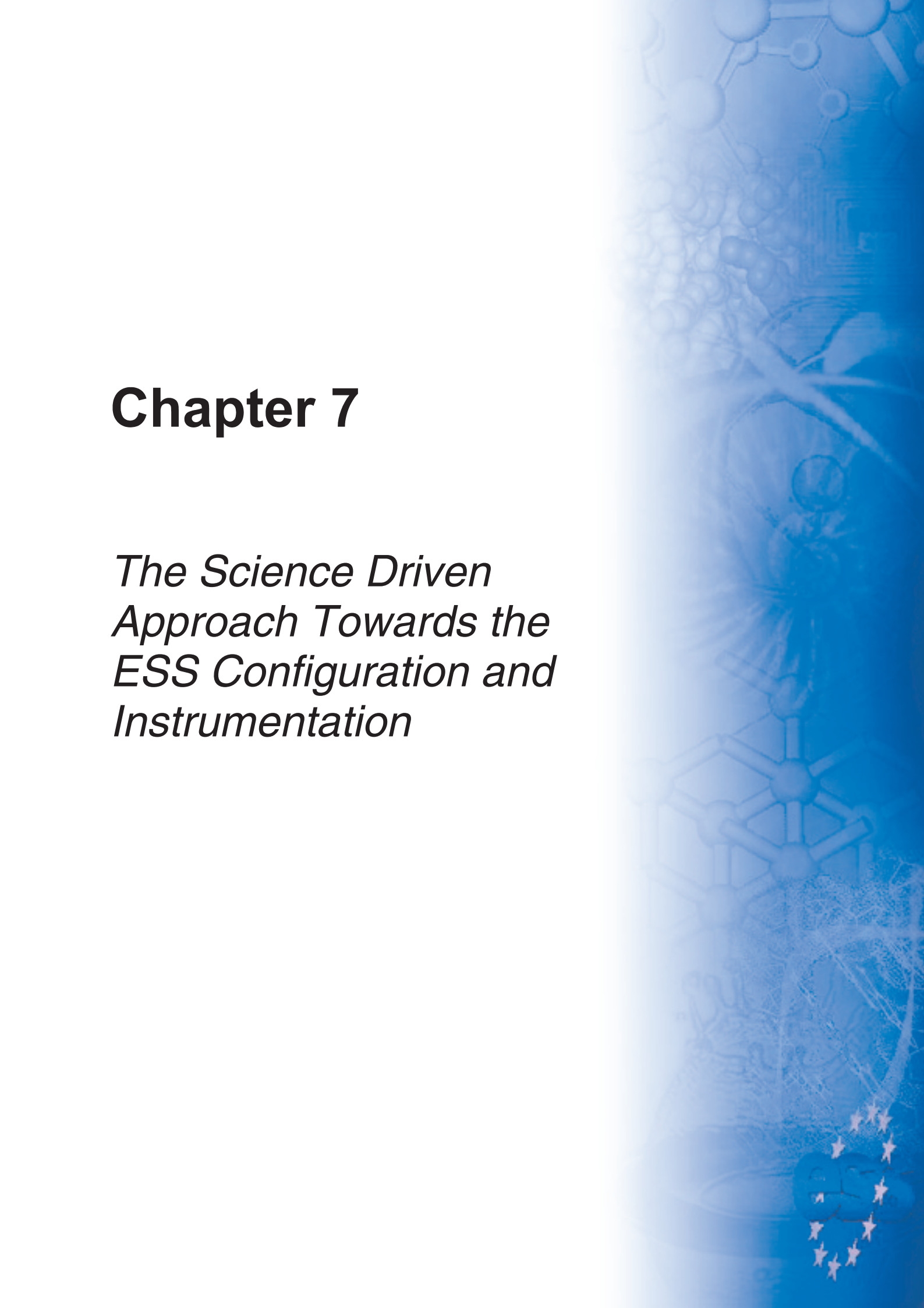
Today all semileptonic cross sections used in cosmology, astrophysics and particle physics are based upon neutron decay data.

Table 2:
Observables and derived quantities related to new physics or Standard Model parameters.

Observables and derived quantities related to new physics	Standard Model parameters
• gravitational to inertial mass ratio • neutron interferometry • magnetic monopole moment • electric dipole moment • neutron-antineutron oscillation time • mass and phase of right-handed W-boson • time-reversal violating amplitude in neutron decay • neutron charge (charge quantisation violating)	• weak-magnetism amplitude in neutron decay • ratio of axial to vector coupling in neutron decay • neutrino-nucleon cross-sections • number of neutrino families • first quark-mixing matrix element (test of unitarity) • proton-proton weak cross-sections (solar burning) • electric polarisability • parity violating correlations in neutron-nucleon and neutron-nuclear interactions

Chapter 7

The Science Driven Approach Towards the ESS Configuration and Instrumentation



7. The Science Driven Approach Towards the ESS Configuration and Instrumentation

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Abstract
In order to arrive at an ESS layout which is science driven, decisions on the configuration and instrumentation needed to be based on science demands. The compilation and evaluation of requirements from the various communities exploiting neutron scattering in their respective field of science and technology was one major task of the Scientific Advisory Committee. On the basis of these science demands recommendations on the technical layout and the instrument suite of ESS were deduced and submitted to the ESS Council. This chapter describes the approach taken by the Scientific Advisory Committee to ensure that Europe's third generation neutron source will serve the needs of users in all fields of science relevant for neutrons in the most effective way possible.

I. Introduction

The ESS is a science driven facility.

In order to assure that ESS develops towards a true European Source of Science, it is of utmost importance, to design and instrument ESS according to scientific demands in all fields of condensed matter science including biology. This is the paradigm of ESS. In order to foster this goal, in 2000 the ESS council created a Scientific Advisory Committee (SAC) with members from all disciplines relevant for ESS and the scientific leaders from the megawatt spallation source projects abroad. In close consultation with the European Neutron Scattering Association (ENSA) these scientists were appointed and charged with the task to elaborate the science case and to advise the Council on the technical layout and the instrumentation of ESS.

In order to investigate the scientific opportunities of the ESS, the SAC has convened eight science working groups with the mission to explore the likely lines of development in neutron related scientific fields. From the assessment of future trends and the identification of major scientific challenges – the so called flagships – the science demands on ESS were derived. With respect to them the different ESS design options needed to be benchmarked. This relates to the accelerator (pulse structure, repetition rate, power, etc.), the target stations and moderators which provide different opportunities for instrumentation and the instruments itself.

The science based parameters have a major impact on the whole of the ESS project design.

This chapter does not describe all contributions of the SAC to processes of decision-making on various technical parameters and all aspects of the involvement of the SAC in the work of the different task groups. It rather illuminates the approach taken by the SAC, in order to arrive at science based recommendations on the target stations, the accelerator layout and a list of "Day One" instruments for the ESS. On the basis of these recommendations the ESS Council decided the configuration of ESS and its prioritised instrumentation.

II. Target station selection

The consideration of a long pulse target station was one of several new developments in the ESS project since 1997.

Since the 1997 ESS proposal, the ESS project has developed in several new directions encompassing the old ESS with major new features. As one of these new developments in addition to the short pulse target stations the ESS has investigated the scientific prospects for a long pulse target station. The following target stations were considered:

(i) a 50 Hz short pulse (1.4 μs) 5 MW target station.
(ii) a 10 Hz short pulse (1.4 μs) 1 MW target station.
(iii) a $16^2/_3$ Hz long pulse (2 ms) 5 MW target station.

In order to make a rational target station assessment, the performance of key instruments at the different target stations needed to be known. In order to achieve this goal, the ESS instrument task convened nine instrumentation expert groups. These groups were charged to explore the performance of generic instruments at the different target moderator ensembles. The work was based on neutron spectral distributions calculated by the target group.

At a SAC meeting in San Sebastian in March 2001 a first exchange of information between the science and instrumentation group conveners took place. The joined activities culminated in an ESS-SAC/ENSA workshop in May 2001 in Engelberg, Switzerland. The major task of this workshop on 'Scientific Trends in Condensed Matter Research and Instrumentation Opportunities at the ESS' was to set priorities for ESS design options based on scientific requirements and in particular to arrive at a selection of the target stations.

At the workshop the instrument groups provided a performance evaluation of generic instruments at the different target stations which is published as a separate ESS report. The key information was summarised in the form of instrument performance sheets, similar to those displayed in volume IV. Table 1 gives an overview about the main results. It displays the expected performance of generic instruments at the various target stations at ESS. First choices from the point of view of the instrument groups are given as black dots, second choices by open dots. The instrument performance is compared to present "best of their class" instruments at the worlds premier reactor (ILL) and premier spallation source (ISIS). Gain factors are given which distinguish source gains and further gains due to better instrument design. The numbers correspond to instruments implemented on the best (or one of the best) target options. Blue numbers compare to ILL beams while black numbers compare to beams at ISIS. The average gain factors at the bottom are geometrical averages over all instruments. A summary of the instrument group considerations including thoughts on future innovative instrumentation is given in chapter 3 and volume IV.

Table 1:
Expected performance of generic instruments on the various target station options for ESS. ● : first choice or one of essentially equivalent first choice options. O : second choice: about a factor of 2 inferior in data collection rate to the first choice. The numbers correspond to instruments implemented on the best (one of the bests) target options. Blue numbers: compared to ILL beams and best existing instruments at ILL. Black numbers: compared to ISIS beams and best existing ISIS instruments (see chapter 3 by F. Mezei).

Instrument	50 Hz 5 MW	10 Hz 1 MW	$16^2/_3$ Hz 5 MW	Source gain	Total gain
High energy chopper	●			30	30
Thermal chopper	●			30	240
Cold chopper	●	O	O	50	1600
Variable, Cold chopper	O		●	20	800
Backscattering 0.8 μeV	●			25	50
Backscattering 17 μeV	●			150	600
Molecular Spectrometer (TOSCA)	●			50	100
Electron Volt Spectrometer	●			30	300
High Resolution NSE			●	10	100
Wide Angle NSE			●	9	300
Triple-Axis	●		●	0.5-1	1-4
High Resolution Single X	●			>>10	>>10
Chemical Single X	●			>>10	>>10
High Resolution Protein	●			>20	>20
Low Resolution Protein	●		●	3-5	3-5
Single Peak incl. Cryopad	●		●	0.3-3	0.3-3
High Resolution Powder	●	●	●	50	150
High Q Powder	●	O		60	120
Magnetic Powder	●	●	O	60	60
High Resolution Reflectometer	●	O	O	20	40
High Intensity Reflectometer	O		●	15	40
Liquids Diffractometer	●			20	20
High Intensity SANS			●	8	100
High λ Resolution SANS	●	●		150	300
Engineering Diffractometer	●			30	90
Fundamental Physics	●		●	1	NA
Diffuse scattering (D7)	O		●	15	300
Backscattering (Musical)			●	40	40
Average (geometrical)				>19	>47

With the instrument performance data at hand and the analysis of the science trends and subsequent science demands available, the science groups set their priorities for different instruments at different target stations. For instruments important for their field of science, the groups had the option to make first and second choices of target stations. Table 2 presents the outcome of the target station evaluation, first choices are indicated by (A), second by (B).

Table 2:
Result of the target station evaluation (A: first choice, B: second choice).

			A	B	A + B
50 Hz	5 MW	SP	70	12	82
10 Hz	1 MW	SP	3	8	11
$16^2/_3$ Hz	5 MW	LP	31	2	33

Obviously, the 50 Hz, 5 MW short pulse target station drew the most first choices with 70 (A), while the $16^2/_3$ Hz, 5 MW long pulse target station came very clearly as second with 31 first choices. Compared to that, the 10 Hz, 1 MW short pulse target station received only 3 first choices and was qualified as less preferable.

Thus, the ensemble of scientists covering the different research fields in neutron science came to a very clear conclusion. They recommended to build ESS as a 10 MW source with two target stations: a $16^2/_3$ Hz long pulse target station which is directly fed by the LINAC and a 50 Hz, 5 MW short pulse target station which is placed after the accumulator rings.

The scientists recommend to build the ESS with a 50 Hz short pulse and a $16^2/_3$ Hz long pulse target station.

After the ESS-SAC/ENSA workshop the SAC assessed and evaluated the outcome of the workshop. It was realised that the 50 Hz short pulse target station had the highest requests from today's perspective. On the other hand it was recognised that the $16^2/_3$ Hz long pulse target station has the larger scientific potential for innovation, while the 10 Hz short pulse target station was of little interest. Therefore, the SAC advised the ESS Council:

"The ESS project should incorporate a 50 Hz short pulse target station as well as a $16^2/_3$ Hz long pulse target station both at a level of 5 MW proton beam energy with equal priority. This recommendation implies a 10 MW proton LINAC serving the two stations."

In its meeting in June, 2001 the ESS Council followed the recommendation of SAC and fixed the neutron parameters of ESS accordingly.

III. Selection of "Day One" instruments

After the target station selection was accomplished the next task required a rational approach to select a first suite of instruments called "Day One" instrumentation. This instrument selection took place at a SAC meeting in November 2001 in Grenoble. In order to define the scientific demands for ESS instrumentation the different science groups were asked to provide a priority list of instruments based on the flagship areas discussed in chapter 4. For that purpose each group was allotted with 20 points to be distributed among flagship areas and generic instruments. Table 3 displays the

accumulated results from all science groups. The distribution of points establishes a prioritisation for the instruments.

Table 3:
Instrument priorities from the disciplinary science groups.

Science Groups / Instruments	Solid State Physics	Material Science and Engineering	Biology and Biotechnology	Soft Condensed Matter	Chemical Structure, Kinetics and Dynamics	Earth Science Environmental Science	Liquids and Glasses	Σ
High Energy Chopper	1				2		1	**4**
Thermal Chopper	5				1		3	**9**
Cold Chopper	5				1			**6**
Variable, Cold Chopper				1	1		3	**5**
High Resolution Backscattering	1	4	1		1			**7**
Medium Resolution Backscattering	1		3		1		2	**7**
Molecular Spectroscopy (TOSCA)					1	3		**4**
eV Spectroscopy								**-**
High Resolution NSE	1			3	1			**5**
Wide Angle NSE				2				**2**
Triple Axis								**-**
High Resolution Single Crystal						2		**2**
Chemical Single Crystal	1				1			**2**
Low Resolution Protein			1					**1**
Small Crystal Protein			1.6					**1.6**
Larger Protein, Big Crystal			3.4					**3.4**
Single Peak incl. Cryopad								**-**
High Resolution Powder	1	4			2	5	1	**13**
High Q Powder					2	1	2	**5**
Magnetic Powder	1				1	2		**4**
High Resolution Reflectometer		4	3		1			**8**
High Intensity Reflectometer	2		1	4	1			**8**
Liquids Diffractometer							5	**5**
High Intensity SANS			5	6	2		2	**15**
High λ Resolution SANS			0.8		1			**1.8**
Engineering Diffractometer		4				4		**8**
Diffuse Scattering (D7)	1						1	**2**
Tomography/Radiography		4				3		**7**
Focussing low q SANS			0.2	4				**4.2**

Figure 1 illustrates the results for the short pulse target station (red bars), Figure 2 displays the outcome for the long pulse target station (red bars). Selecting the instruments with a score higher than the mean value, represented by the red horizontal lines in Figure 1 and Figure 2, a first selection of an instrument suite with five instruments at the long pulse and eight instruments at the short pulse station was obtained.

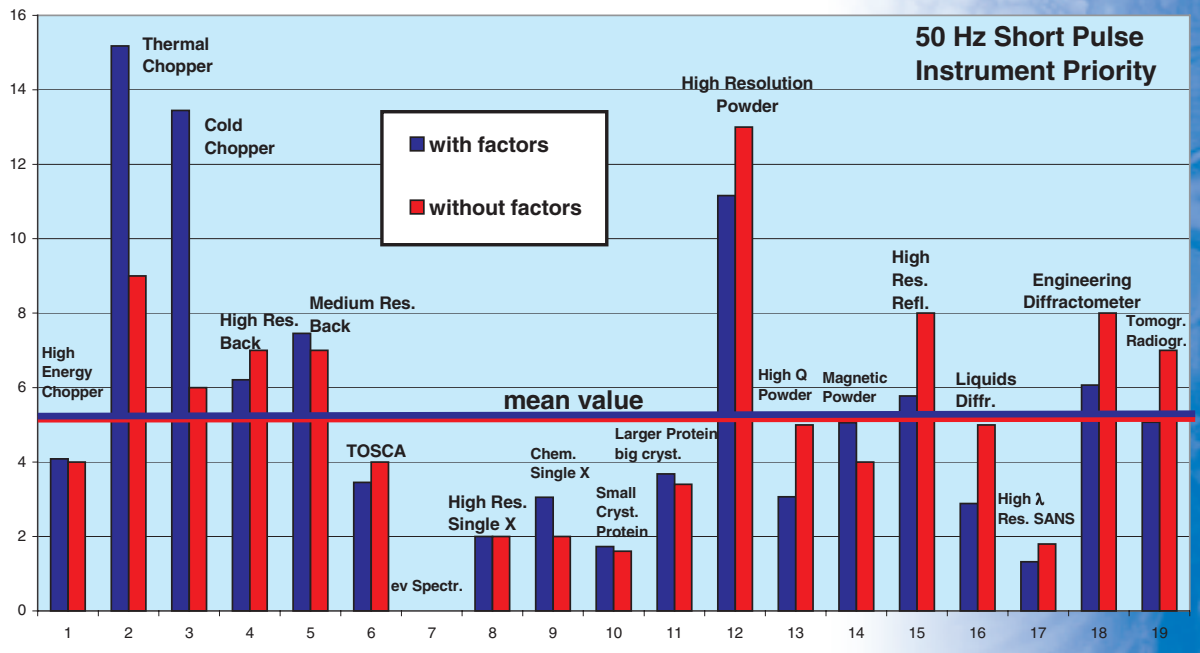

Figure 1: Instrument priorities at the short pulse target station with and without scaling factors.

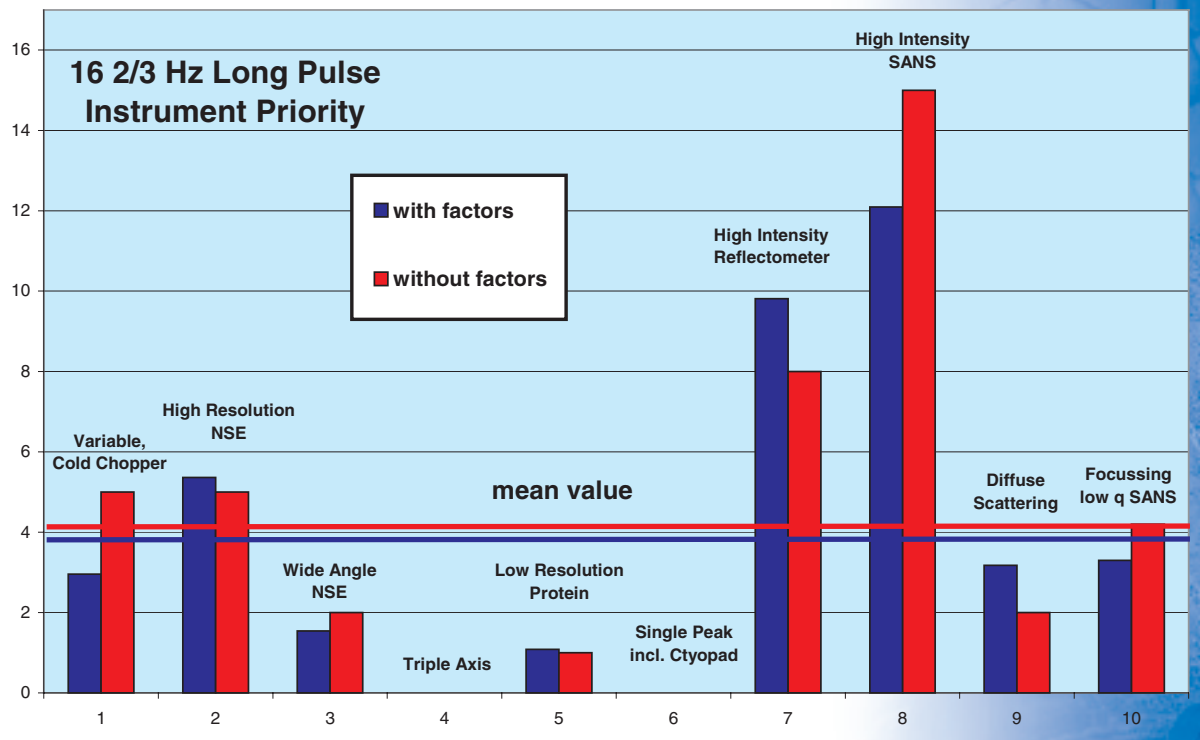

Figure 2: Instrument priorities at the long pulse target station with and without scaling factors.

This first assessment treated the different scientific disciplines in an equal fashion neglecting thereby the significantly different sizes of the respective communities. In a second approach these differences were factored in. Starting point was the statistics of requested beam time at the ILL in Genoble for fall 2000 and spring 2001. These demands were grouped into disciplines. The relative magnitudes defined factors which were used to weigh the demand figures from the different science groups. The blue bars in Figure 1 and Figure 2 illustrate the results of this procedure. Now, for the instruments at the long pulse target station, only three instruments have scores higher than the average (blue horizontal line). However, the list of six instruments with the highest scores, extracted from Figure 2, contains again the five instruments one obtains without scaling factors plus the Diffuse Scattering instrument.

Comparing the results, it was realised that - though the sequence of scores has changed - the table of instruments remains practically the same. This is true for both target stations and shows, that the method of instrument selection is very robust.

The outcome of this instrument prioritisation was taken as a solid basis for further considerations. A final choice was made after taking into account also other aspects. For instance the demands of rapidly growing science fields like biology or to some extend also chemistry were taken into account separately. Also novel instrumentation opportunities like single pulse diffraction were considered additionally. These considerations led to a consolidated “Day One” instrumentation list for ESS which is displayed in Table 4.

Table 4:
Consolidated “Day One” instrumentation list.

50 Hz short pulse target station	**$16^{2}/_{3}$ Hz long pulse target station**
High Resolution Powder	High Intensity SANS
Thermal Chopper	High Intensity Reflectometer
High Resolution Reflectometer	Variable Cold Chopper
Engineering Diffractometer	High Resolution NSE
High Resolution Backscattering	Focussing low q SANS
Tomography / Radiography	D7 – type
Cold Chopper	+ two novel instruments
Chemical Single Crystal	+ one instrument test facility
Small Crystal Protein	
Magnetic Powder / Liquids Diffractometer	
High Energy Chopper	
Single Pulse Diffractometer (novel)	

The instrument task group took this “Day One” instrumentation list as a starting point for further consideration of instruments and for an optimisation of the moderator – target - instrument complex.

At the 2nd SAC workshop on “Scientific and Technological Challenges in the 21st Century - The Contribution of ESS”, held in March 2002, cross disciplinary expert groups evaluated possible contributions of the ESS to Europe’s societal needs. Such research missions have been identified by the European Union or different European governments as “Priority Research Themes”. The reports on the contribution of ESS to these missions can be found in chapter 6.

The experts were also asked to define the instrument requirements from the standpoint of these more applied science fields. Table 5 displays the outcome following the same scheme as discussed with respect to the disciplinary prioritisation. Figures 3 and 4 compare the outcome of this evaluation with the previous one. The red and blue bars compare the instrument priorities based on the needs of applied science to those derived from the demands from more basic science. While at the long pulse target station the major difference is an even higher score of the SANS and the Reflectometer instruments, which were also selected by the disciplinary science groups with highest priority, at the short pulse target station the differences are more subtle. However, there is no significant change of the instruments with the highest scores.

The definition of instrument priorities is still evolving.

The evaluation procedure described above resulted in a quite unequivocal determination of instrument priorities with respect to the list of generic instruments. However, the ongoing work of instrument scientists and potential users, to assess future needs and opportunities, leads to an evolution of instrument ideas and priorities. In particular a number of new instruments have been proposed for further consideration. For instance, at the end of the 2nd SAC workshop the expert group assessing the impact of ESS on future trends in Nanotechnology proposed a *Point Focus Reflectometer,* while the Microsystems – group underlined their interest in a *combined SANS and Reflectometer (RefSANS)* – instrument.

At the Workshop on “Perspectives of Neutron Scattering for the Earth Sciences with the ESS”, held in Cambridge, UK, in January 2002, the experts came to the conclusion that a dedicated high pressure / high temperature instrument would be of great importance for scientists working in the Earth and related sciences fields. The case for such an instrument has been submitted to the SAC and will be considered.

These examples demonstrate that the definition of the science demands on the instrument suite is still evolving. They also underline the importance of an open list of “Day One” instruments which allows the consideration of instrument requirements as they may emerge from future work of experts assessing the trends and the respective role of neutrons in their field of science and technology.

Table 5:
Instrument priorities from the cross disciplinary science groups.

Mission Group / Instruments	Sustainable Developm., Clean Technologies and Environm. Systems	Nanotechnologies	Health and Biotechnology	Functional Materials	Cultural Heritage: Artefacts and Materials	Traffic and Transport	Microsystems and Inform. Technologoies	Σ
High Energy Chopper	3			1				**4**
Thermal Chopper	3	2.2		1		1		**7.2**
Cold Chopper				2		1		**3**
Variable, Cold Chopper		2.2						**2.2**
High Resolution Backscattering	1	1.2		1		2		**5.2**
Medium Resolution Backscattering	1		1	1				**3**
Molecular Spectroscopy (TOSCA)	2				2		1.55	**5.55**
eV Spectroscopy								**-**
High Resolution NSE	2	1.2		1		1		**5.2**
Wide Angle NSE				1				**1**
Triple Axis							1.5	**1.5**
High resolution Single Crystal							1.55	**1.55**
Chemical Single Crystal							1.55	**1.55**
Low Resolution Protein								**-**
Small Crystal Protein			5					**5**
Larger Protein, Big Crystal			3					**3**
Single Peak incl. Cryopad								**-**
High Resolution Powder	4			3	6	2		**15**
High Q Powder	2			2	2			**6**
Magnetic Powder				2			1.55	**3.55**
High Resolution Reflectometer			1				4.6	**5.6**
High Intensity Reflectometer	1	4.4	5	1	1	1	4.6	**18**
Liquids Diffractometer					1			**1**
High Intensity SANS	1	6.6	5	3		2	1.55	**19.15**
High λ Resolution SANS								**-**
Engineering Diffractometer					5	6		**11**
Diffuse Scattering (D7)							1.55	**1.55**
Tomography/Radiography					3	4		**7**
Focussing low q SANS		2.2		1				**3.2**

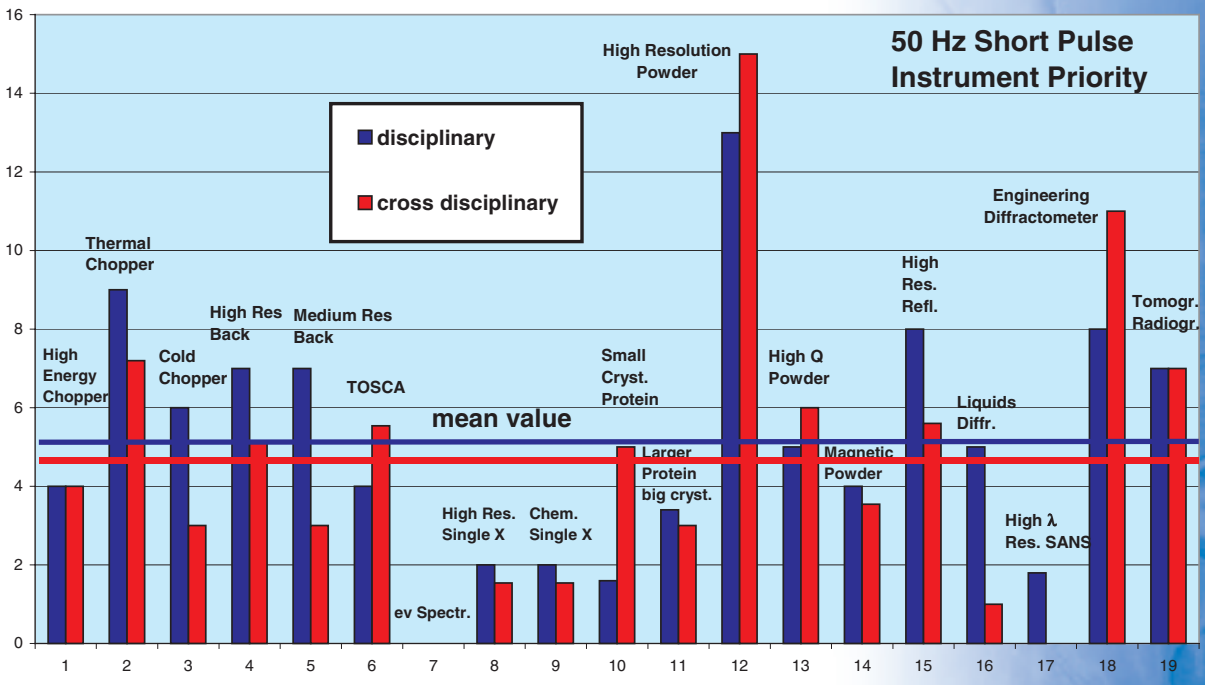

Figure 3: Instrument priorities from the disciplinary and cross disciplinary groups at the short pulse target station.

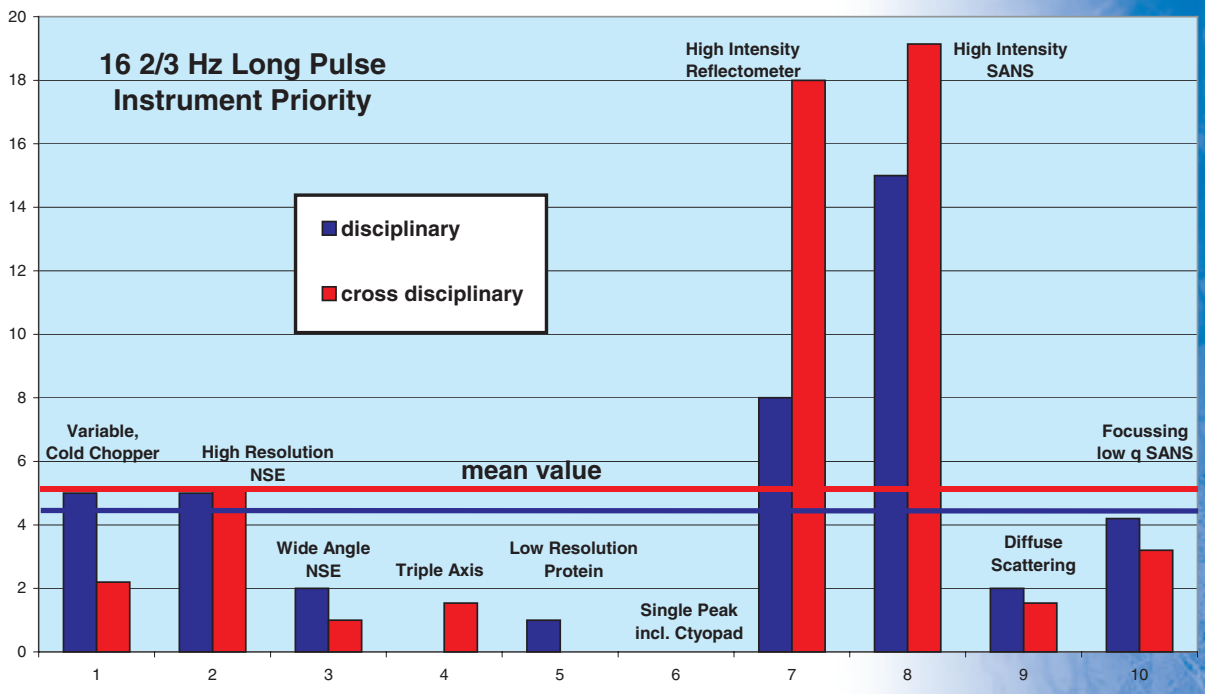

Figure 4: Instrument priorities from the disciplinary and cross disciplinary groups at the long pulse target station.

Appendix

- Scientific Groups
- Mission Oriented Groups
- Complementarity Group
- Instrumentation Groups

Appendix

Scientific Groups *(*group convener/s)*

Solid State Physics

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G.H. Lander, IfT Karlsruhe, Germany
D.F. McMorrow, Risø Roskilde, Denmark
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Material Science and Engineering

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A. Magerl, Univ. Erlangen-Nürnberg, Germany
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P.J. Withers, Univ. Manchester, United Kingdom
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Chemical Structure, Kinetics and Dynamics

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Soft Condensed Matter

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K. Mortensen, Risø Roskilde, Denmark
D. Richter, FZ Jülich, Germany*
J. Rieger, BASF Ludwigshafen, Germany
P. Schurtenberger, Univ. Fribourg, Switzerland
R.K. Thomas, Univ. Oxford, United Kingdom

Liquids and Glasses

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R.L. McGreevy, Univ. Uppsala, Sweden*
A.K. Soper, RAL Abingdon, United Kingdom
J.-B. Suck, Univ. Chemnitz, Germany
R. Vacher, Univ. Montpellier, France

Biology and Biotechnology

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O. Byron, Univ. Glasgow, United Kingdom
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D. Svergun, EMBL Hamburg, Germany
J.-C. Thierry, IGBMC-CNRS Strasbourg, France
J. Zaccai Grenoble, ILL Grenoble, France

Mineral Sciences, Earth Sciences, Environment and Cultural Heritage

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R. Rinaldi, Univ. Perugia, Italy*
W. Schäfer, Univ. Bonn and FZ Jülich, Germany
P.F. Schofield, Natural History Museum London, UK
B. Winkler, Univ. Frankfurt, Germany

Fundamental Physics

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J. Byrne, Univ. Sussex, United Kingdom
D. Dubbers, Univ. Heidelberg, Germany
W.E. Fischer, PSI Villigen, Switzerland
M. Pendlebury, Univ. Sussex, United Kingdom
H. Rauch, Atomic Inst. of the Austrian Univ., Wien, Austria*
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M. Monkenbusch, FZ Jülich, Germany
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Cultural Heritage: Artefacts and Materials

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Sustainable Development, Clean Technologies and Environmental Systems

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T.C. Hansen, ILL Grenoble, France
H. Jobic, CNRS Villeurbanne, France*
G.H. Lander, IfT Karlsruhe, Germany
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R.L. McGreevy, Univ. Uppsala, Sweden*
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A. Steuwer, Univ. Manchester, United Kingdom
P.J. Withers, Univ. Manchester, United Kingdom*

Particle Physics

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