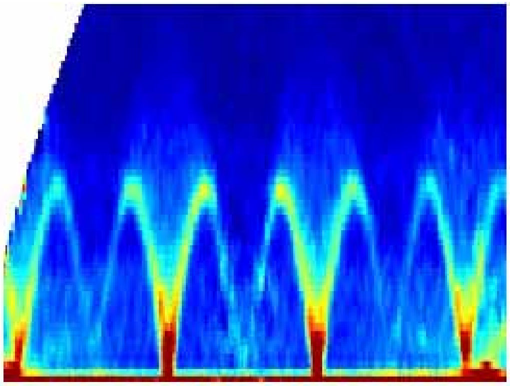

ARCS

A High-Resolution Direct Geometry Chopper Spectrometer at the SNS

SNS

June 3, 2001

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Executive Summary

We propose to construct A high-Resolution, direct-geometry, time-of-flight Chopper Spectrometer, ARCS, on beam line 18 at the Spallation Neutron Source. ARCS will be optimized to provide a high neutron flux at the sample and a large solid angle of detector coverage. To optimize energy resolution and momentum resolution, ARCS will be large. The source-sample distance will be 13.5 m, and the secondary flight path will be 5.5 m for angles to –20° to 40°, and 3.0 m for angles from 40° to 140°. Software will allow the data from the two detector banks to be analyzed simultaneously. The dense array of position-sensitive detectors will detect the direction and velocity of neutrons scattered into 25% of the solid angle around the sample. The spectrometer would be capable of selecting from the full energy spectrum of neutrons provided by an ambient water moderator, making it useful for studies of excitations from a few meV to several hundred meV. A supermirror guide in the incident flight path will boost the performance at the lower end of this range. ARCS will be the most efficient high-energy chopper spectrometer at any spallation neutron source.

The scientific mission for ARCS is to enhance our fundamental understanding of dynamic processes in materials. The primary research topics are: (i) studies of vibrational excitations and their relationship to phase diagrams and equations of state of materials, including materials with correlated electrons, and (ii) studies of spin correlations in magnets, superconductors, and materials close to metal-insulator transitions. The projects described in this proposal are at the forefront of current research in condensed matter physics and materials science.

Inelastic neutron scattering experiments have always been constrained by low neutron flux, necessitating experimental compromises in energy resolution and in the number of spectra that can be measured. With its high detection efficiency and its siting at the SNS, the ARCS spectrometer will go a long way towards eliminating these compromises. ARCS would enable acquisition of inelastic spectra with high energy resolution over a range of physical parameters, such as mapping dynamic processes in solids as a functions of temperature, pressure and composition. Such studies of physical trends, especially with good energy resolution, are largely impractical with today's instrumentation for inelastic neutron scattering. A second major new capability of the ARCS instrument will be measurements of dispersion relations on single crystals. Measurements of the dispersions of elementary excitations such as magnons and phonons have to date been largely the domain of triple-axis spectrometers at reactor sources because instruments at spallation sources suffer from inadequate software and limitations in flux and resolution. The ARCS instrument will solve all these problems, enabling experiments with a sophistication not yet reached with chopper spectrometers.

The development of a new chopper spectrometer and its integration into the SNS facilities will require an extensive design effort. Fortunately, members of this Instrument Development Team (IDT) have already performed much of the work on conceptual design and performance simulations over the past few years. Members of the IDT prepared two detailed proposals for direct geometry chopper spectrometers – the HELIOS instrument for flight path 8 at the Lujan Center at LANL and the VERTEX instrument for flight path 14 the Lujan Center. Additionally, preliminary designs and instrument simulations were completed for a 6 m high-resolution chopper spectrometer for beam line 17 at the SNS, and a 3 m high-flux chopper spectrometer for beam line 18 at the SNS. Members of the ARCS IDT have reached a mature understanding of the scientific capabilities of inelastic chopper spectrometers, their construction timetables, and their costs. The ARCS project will require approximately 2 years of design effort, 3 years of hardware construction, and 5 years of concurrent work on software development.

The Principal Investigator (P.I.) will work closely with the Project Manager (P.M.) on the day-to-day management of the ARCS project. Members of a small and active Executive Committee have been chosen for their breadth of scientific interests in inelastic neutron scattering, their willingness to contribute time and energy, and their expertise with chopper spectrometers. This Committee will advise the P.I. and P.M. on the hardware design, contribute substantially to the software development, and work with the P.M. and the P.I. to control changes to the scope of the project. This Executive Committee will later take responsibility for establishing the scientific program by setting formal procedures for beam time allocation and contractual obligations with SNS.

The ARCS spectrometer will complete an initial triad of inelastic instruments at the SNS. With a high-resolution backscattering spectrometer for low energies, a disk chopper spectrometer for medium energies, and ARCS for high energies, the SNS will be optimized for studies of excitations in condensed matter over the broad range in energy from micro-eV to eV. With such capability there is no doubt that the SNS will be the international center for experimental research in excitations in solids, moving ORNL to the forefront of research in condensed matter physics and materials science.

1. Scientific Justification

Neutron spectroscopy provides unique information about dynamic properties of condensed matter. Time and again this experimental technique has provided essential information for understanding fundamental properties of condensed matter Neutron spectroscopy provided the comprehensive evidence for phonons and the first evidence for spin waves by directly measuring their dispersion relations. Brockhouse was awarded the 1994 Nobel Prize in physics for such work.

A neutron spectrometer is optimized for sensitivity and resolution within a particular range of energy and momentum transfer. At present, no instrument in the U.S. offers state-of-the-art sensitivity and good momentum resolution in the range of energy transfers from 50 to 500 meV, an important range for magnetic and vibrational dynamics. None have the efficiency of the proposed ARCS instrument, whose rapid data acquisition will enable multiple measurements over a range of experimental parameters such as temperature. The resolution and sensitivity of the ARCS instrument will also enable experiments on single crystals; experiments that are marginally practical with present spallation sources owing to inadequate neutron flux. In this Sect. 1 we discuss the scientific areas where progress is impeded owing to inadequate instrumentation. These inelastic scattering studies have been constrained by compromises originating with low flux, low resolution, and low sensitivity. The ARCS instrument, sited at the SNS, will overcome these present limitations and open a new era for inelastic neutron scattering.

1.1 Lattice Dynamics

The past two decades have seen a steady growth in our understanding of alloy phase diagrams. There has been excellent progress in the ab-initio microscopic approach, which provides a free energy function by combining total electronic energy calculations with an entropy from statistical mechanics.[1] Almost every experimental discovery of an important new material such as a high temperature superconductor or a high-strength transition metal aluminide is now followed quickly by a phase diagram calculation. While these phase diagrams are not necessarily precise in detail, they are helpful for further materials development. B. Fultz and his group would use the ARCS spectrometer for investigations in the field of phase stability, specifically on the entropies of solid phases. Experiments to measure phonon DOS curves of different alloy phases almost invariably require the use of polycrystalline samples, for which a direct-geometry time-of-flight spectrometer offers advantages such as the absence of contamination wavelengths and simultaneous data collection over a wide range in $\boldsymbol{Q}$.

1.1.1 Entropy

The free energy, F, of an alloy phase is:

$$F = E - T\left(S_{config} + S_{vib} + \sum_{\xi} S_{\xi}\right) \quad . \tag{1}$$

The configurational entropy, S_{config}, is well known, and methods for calculating S_{config} are well developed. Until recently, however, the vibrational entropy, S_{vib}, has been essentially unknown and assumed or hoped to be small,[1] as have other sources of entropy, S_ξ, in non-magnetic materials. Inelastic neutron scattering has played the major role in proving that changes in vibrational modes are responsible for much of the entropy change of solid-solid phase transitions, sometimes in concert with calorimetric measurements.[2,3,4,5,6,7] The size of the vibrational entropy can be obtained with the phonon density of states (DOS, $g(E)$) of the alloy. In the high temperature limit, the difference in vibrational entropy between two phases, α and β, is:

$$\Delta S_{vibr}^{\alpha-\beta} = 3k_B \int_0^\infty [g^\beta(E) - g^\alpha(E)] \ln(E) dE \tag{2}$$

where the difference of normalized DOS functions avoids problems with the dimensionality of the logarithm.

When the vibrational entropy is known, data on the latent heat of the phase transition can be used to test for other possible sources of entropy. For example, the large entropy of the shape-memory transformation in NiTi is entirely vibrational in origin,[8] whereas there are large contributions from electronic entropy to the latent heat of phase transitions in cerium[9] and uranium[10] metal.

In recent measurements on the α-phase of uranium metal,[10] a large decrease in phonon energies with increasing temperature was observed over the entire temperature range from 50 to 913 K (Fig. 1). Analysis of the vibrational power spectrum showed that the phonon softening originates with continuous softening of a harmonic solid, as opposed to vibrations in anharmonic

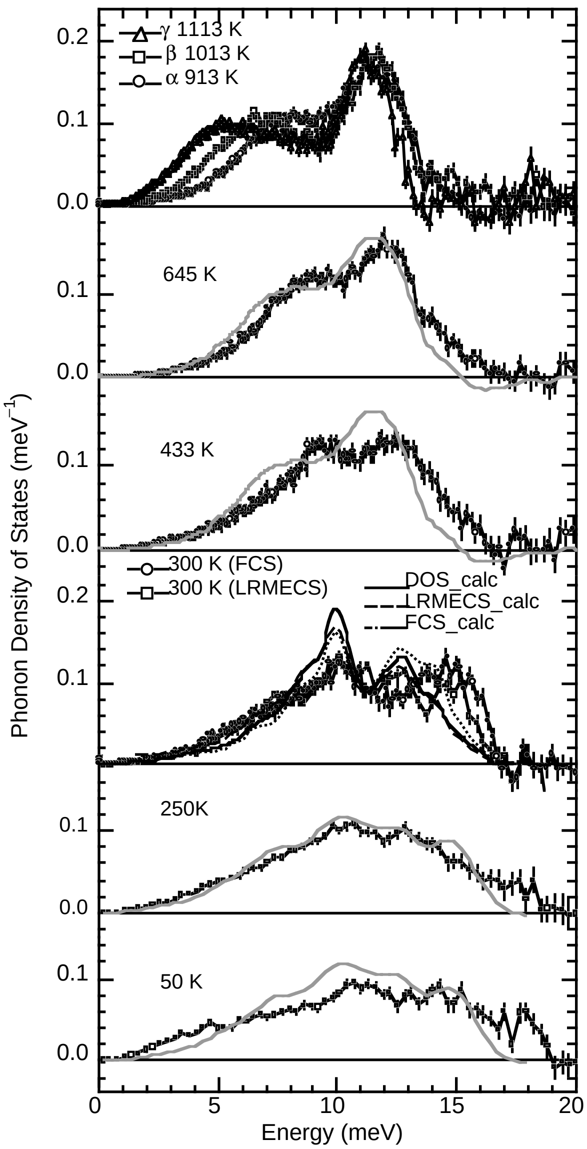

Figure 1: The phonon DOS of uranium measured on FCS at NCNR (T>300 K) and LRMECS at IPNS (T<300 K). The 913 K α-uranium DOS is superimposed on all curves above 300 K and the 300 K data is superimposed on all curves below 300 K. The three solid state phases, orthorhombic (α), tetragonal (β), and body-centered cubic (γ), are compared at the top.

potentials. It follows that thermal excitations of electronic states are altering the force constants. This contradicts the usual assumption that temperature effects on the electronic structure can be neglected when compared to volume effects.

1.1.2 Disordered Solids

It has also been shown that microstructural features of materials affect the vibrational entropy.[7,11,12] Nanocrystalline materials have a significantly larger number of vibrational modes at low energies, promoting thermodynamic stability at finite temperature.[8,9] Interestingly, these modes extend to energies as low as 10 μeV, indicating that they originate from long-range cooperative movements of nanocrystals.

Excitations in disordered materials are a problem for which there has been halting progress over many years. For phonons on disordered lattices, for example, it is possible to set up a Hamiltonian with disorder in masses or force constants, but the spectral density and mode structure often requires large-scale computation. This has impaired detailed comparisons between theory and experiment, and has made it unrealistic to invert the measured data. The software development effort in the ARCS project will address this problem in detail. Large-scale simulations of lattice dynamics will be coupled to simulations of instrument performance, allowing the parameters of the lattice dynamics to be recovered from experimental data.

An alternative way to describe the lattice dynamics in disordered solids is to consider the real space atomic correlations of vibrational modes. For example, if the frequency of local vibration is higher than the phonon frequency it becomes a local mode with no dispersion. When it is within the phonon band, however, a resonant state is created, and the local mode becomes mixed with the extended modes. If the disorder is extensive and there are many overlapping resonant modes, such a description becomes ineffective. It is more informative to apply the Fourier-transformation on $S(\boldsymbol{Q},\omega)$, and determine the dynamic atomic correlation function $g(\boldsymbol{Q}, \omega)$.[13,14,15,16] Since most properties in disordered systems are determined by local atomic structure over relatively short distances, such a description is better suited to relate the structure to properties. T. Egami would pursue this work with the ARCS instrument, which is somewhat like his work on elastic pair distribution functions. The inelastic pair distribution function has a similar requirement to the elastic pair distribution function in that the method requires $S(\boldsymbol{Q}, \omega)$ to be measured reliably to large values of $\boldsymbol{Q}$. Truncation of the range of Q causes a loss of resolution of real-space information.

1.1.3 Equations of State

Although there have been many measurements of macroscopic equations of state of solids, there is, unfortunately, much less experimental data on the temperature and volume dependence of the phonon DOS. The handful of studies on temperature dependencies of phonon dispersions have focused on issues related to diffusion and structural phase transitions, not the phonon thermodynamics. The lack of phonon data as functions of pressure and temperature leaves a gap between ab-initio theories of solid thermodynamics and measurements of *P-V-T* equations of state. Ultimately there should be a rationalization of the macroscopic equations of state in terms of the specific changes in phonons and electrons that underlie them.

The sample environment of the ARCS instrument will include the ability to vary pressure and temperature at the specimen. Collimation of the incident beam and oscillating radial collimators after the specimen will attenuate the stray scattering from the sample cell. The ability of ARCS to measure high-quality phonon DOS on one sample at several temperatures and pressures will enable rapid progress in understanding the *P-V-T* equations of state. Although high pressure measurements of full dispersions of single crystals would be desirable, this is typically impractical. It is realistic, however, to take phonon DOS data with excellent energy resolution and good statistics, and invert them to obtain the force constants of lattice dynamics models. Data quality from present inelastic chopper spectrometers does not justify such inversions, but we have had success in inverting ^{57}Fe phonon partial DOS data from inelastic x-ray nuclear resonant scattering experiments, which have better energy resolution.[17] Data from ARCS should be of sufficient quality for inversion, and inelastic neutron scattering can be used with most elements, unlike nuclear resonant scattering that works with only ^{57}Fe and ^{119}Sn.

1.1.4 Phonons in Correlated-Electron Materials

A characteristic feature of correlated-electron materials is a common energy range for excitations of the charge, spin, and lattice systems. The interaction of these sub-systems can vary strongly with subtle changes in external factors such as doping of charge carriers, temperature, pressure, sample stoichiometry, disorder, etc. For many transition metal oxides, high-temperature superconductors in particular, these factors can produce a microscopically and intrinsically inhomogeneous ground state where charge and spin are loosely ordered into "stripes." The lattice dynamics couple very strongly to charge fluctuations in this ground state, as has been shown recently by inelastic neutron scattering measurements of phonon dispersions in single-crystals[18] (for detailed information) and phonon densities-of-states of powder samples (for parametric studies).[19] Using both powder and single-crystal data, the combined lattice and charge fluctuations appear to be localized on length scales of a few lattice spacings (polaronic) and diffuse excitations are observed in q-space. On the other hand, sharp features do occur in the phonon dispersion at particular wavevectors, indicating a dominant fluctuation mode. This peculiar electron-lattice coupling is crucial to the charge dynamics and may play a central role in high temperature superconductivity. It also represents a challenging experimental problem, requiring measurements of weak signals over large ranges of scattering angles at both high and low resolutions, and the careful analysis of both powder and single-crystal data. We recently established that the chopper instrument MAPS at ISIS can measure the phonon dispersion in a single-crystal of $YBa_2Cu_3O_{6.95}$ with accuracy and counting times comparable to triple-axis instruments.[20] The ARCS spectrometer is ideally suited to study powders and (localized) diffuse excitations in single crystals. The ARCS instrument is also expected to be superior to MAPS in its signal-to-background ratio and advanced software for obtaining scattered intensity in absolute units.

The coupling between the degrees of freedom of the lattice and those of the electrons may also be important in the heavy-fermion and mixed-valent materials. The strong temperature-dependence of the phonon DOS shown in Fig. 1 an example of such coupling – the interatomic potential probably changes with temperature because electronic excited states have a different dependence on atom displacements than do the ground states. Other evidence for this coupling can be found in the isomorphic phase transition from the α to γ phases in Ce metal,[21] and the valence transition at ~40 K in $YbInCu_4$,[22] although the origins of both these phase transitions are unclear. In general, lattice effects are neglected when discussing the correlated-electron behavior in these materials and other related ones, but this viewpoint could be tested in greater detail with the ARCS instrument. In uranium the electron-lattice coupling is strong enough to result in charge-density wave transitions at 23 and 37 K. Although the work described in Fig. 1 showed no distinct change in phonon DOS at the charge density wave transition temperatures, this may have been due to a smearing of the CDW transition in the polycrystalline sample (plausibly originating with internal stresses that develop during thermal contraction[23]). Studies on single crystals, which exhibit sharp CDW transitions, would be better suited for detailed study, and for identifying the particular modes involved in these transitions.

1.2 Magnetic Dynamics

Direct-geometry time-of-flight spectrometers are well-suited for probing the magnetic dynamics in materials, offering a wide range of frequencies together with the ability to map out a large region of reciprocal space. Many of the most celebrated successes in applying the direct-geometry time-of-flight method to studies of magnetic dynamics have been in the study of low-dimensional materials. In such systems it is possible to arrange the scattering geometry so the simultaneous variation of momentum and energy transfer along a time of flight trajectory is projected out, thereby providing full coverage of the 1D or 2D reciprocal space. This is not essential however, a recent example is the measurements obtained on HET showing a cut through the very three dimensional dispersion surface of a manganese oxide, one of the so called Colossal Magneto-Resistive (CMR) compounds,[24] shown in Figure 2.

Note that the range of energy transfers accessed in this measurement (0-50 meV) is much lower than that generally thought to be optimal for this class of instrument. The super-mirror guides on the incident flight path will further improve the efficiency of ARCS at incident energies below about 50 meV.

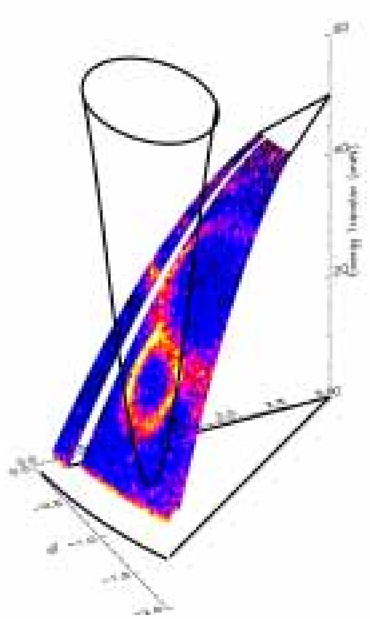

Figure 2: Time of flight spectra obtained on HET from the CMR compound $La_{0.7}Pb_{0.3}MnO_3$ in the ferromagnetic phase at T=10 K. The intersection of the dispersion surface extracted from fits to the data with the neutron trajectories in (**Q**,ω) space is shown schematically.

An example of the use of a direct-geometry time-of-flight spectrometer for studying the magnetic dynamics in a low-dimensional material is a measurement of the spin wave in $FeGe_2$ (a quasi-one-dimensional itinerant antiferromagnet) performed on HET, mapping out the c-axis dispersion up to 400 meV. Figure 3 shows data obtained in ~6 hours using an incident energy of 120 meV at 11 K. In low-dimensional compounds it is possible to characterize a large fraction of the dispersion surface in a single geometry since the variation of $\boldsymbol{Q}$ along a weakly coupled direction in the ($\boldsymbol{Q}$,ω) trajectories does not affect the excitation energy. In the case of $FeGe_2$ the Q_a variation is negligible so the (Q_c,ω) dispersion is mapped out from 40 - 100 meV.[25]

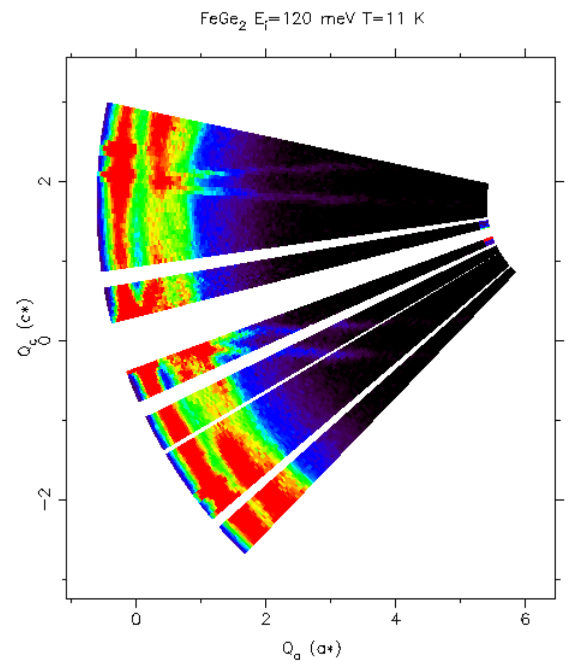

Figure 3: Spin wave and phonon scattering from $FeGe_2$ at 11 K for energy transfers from -10 to 120 meV (left to right) projected onto the (Q_a,Q_c) plane.

Several members of this IDT, and a significant fraction of the condensed matter physics community share an interest in the structure and spin dynamics in correlated electronic materials. The quest for a theoretical description of materials with strong electron-electron interactions is currently at the forefront of condensed matter physics, and the correlation functions measurable by inelastic magnetic neutron scattering provide unique and incisive constraints for theories of the electronic many-body states realized in these materials. Unfortunately there is, however, no instrument in the US that can efficiently probe electronic correlations at energies beyond 50 meV. This has limited the application of neutron spectroscopy to a relatively small class of materials with low temperature anomalies of little technological significance. By increasing the range of energies and reducing the required sample size, ARCS would expand the range of materials that can be studied by neutron spectroscopy. With ARCS we could study spin dynamics in correlated metals with energy scales from 10-500 meV. There is no reason to expect the microscopic information about electronic correlations in these materials to be less interesting than what we learned from neutron spectroscopy in heavy fermion systems where the energy scales are an order of magnitude smaller. High T_c superconductors are important examples of interesting correlated metals, but we may also make progress in understanding seemingly less-exotic 3d metals such as chromium, yttrium, vanadium, or even copper, where the energy scale for magnetic excitations is set by the Fermi energy. Experiments in magnetic multilayers have shown that these materials have considerable ***Q***-dependence in their magnetic susceptibility. New microscopic information about electronic correlations in these systems could have an important impact on our understanding of electronic correlations in the metallic state.

1.2.1 High Temperature Superconductivity

The supreme challenge in the field of strongly-correlated electron systems is an explanation of high temperature superconductivity in the quasi-two-dimensional cuprates. One question in the theory of high temperature superconductors is the relationship between the microscopic spin dynamics and the transport and superconducting properties of the cuprates. To this end, knowledge of the spin correlations over a wide energy range is essential. Until recently, such experiments were confined to excitation energies below ~ 40 meV, larger than the superconducting energy gap but smaller than the intralayer superexchange J ~ 120 meV that sets the energy scale for spin excitations in the undoped antiferromagnetic precursor compounds. In recent pioneering studies using pulsed neutron techniques, these measurements have been extended to higher energies and established the presence of significant spectral weight at energies comparable to J in the $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ families of superconducting cuprates.[26,27,28] The capability of carrying out such high-energy studies with sufficient intensity is currently lacking in the United States, and all of these important experiments were carried out at ISIS or European reactor sources with hot moderators. The ARCS spectrometer would thus fill a critical need in this regard.

Detailed inelastic neutron scattering measurements have also revealed that the doping of La_2CuO_4 replaces antiferromagnetic spin fluctuations by incommensurate spin fluctuations.[29] These features have been ascribed to inhomogeneous charge and spin correlations related to static long-range charge and spin order, as observed in $La_{1.48}Sr_{0.12}Nd_{0.4}CuO_4$.[30] The incommensurate spin fluctuations have recently been observed in single crystals of $YBa_2Cu_3O_{6+x}$ using chopper spectrometers at ISIS,[31] thereby establishing a common feature in high-T_c compounds. This result also demonstrated that chopper instruments can be competitive with reactor-based triple-axis instruments, even when measuring fairly localized features in ***Q***-space. Regular access to a high-intensity spectrometer would enable us to obtain detailed, quantitative measurements of the temperature and doping dependence of the spin dynamics in the cuprate high-T_c materials. This is essential information for testing microscopic theories of high temperature superconductivity.

Crystal field spectroscopy is another promising tool to study the electronic state of the cuprate superconductors. A high-intensity spectrometer is useful for detecting the often weak response from magnetic systems. The ability to investigate electronic transitions and quasielastic scattering at both large and small incident energies makes a machine such as ARCS very useful. For example, an incident energy of 4 meV on HRMECS was used to determine the presence of ~ 1 meV crystal-field level in an Erbium high T_c material.[32] The line width of crystal field excitations has been shown to contain important information about the electronic susceptibility and the pairing state in the cuprates.[33] R. Osborn and collaborators have reported transitions in f-electron systems reaching to the 1 eV range.[34] In S, Kern's investigations of the interactions between the electrons and phonons in the rare-earth phosphates, measurements were performed both small scattering angles (low ***Q***) for the electronic transitions, and large angles (high ***Q***) for the phonons.

A quantitative understanding of high temperature superconductivity in the cuprates will ultimately require broad, systematic studies of transition metal oxides as a function of parameters such as the carrier concentration, dimensionality, the width of the valence band, correlation and charge transfer energies, and disorder. Many of the members of this IDT collaborate actively with solid-state chemists and materials scientists at their institutions and elsewhere who are capable of growing high-quality single crystals of a wide variety of these materials.

1.2.2 One-Dimensional Quantum Magnets

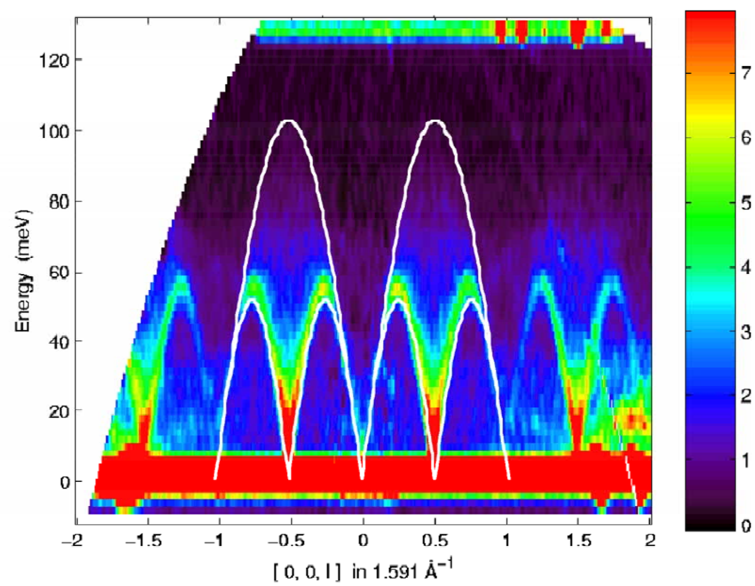

Figure 4: Spin-wave-like excitations in single-crystal $KCuF_3$ measured on MAPS at ISIS using E_0=150 meV at 6 K.

Undoped spin chain compounds are arguably the simplest realizations of quantum many body systems and have for years served as a unique "laboratory" for many-body physics. Some idealized spin chain Hamiltonians can actually be solved exactly; their excitation spectra consist of sharp, dispersive modes and excitation continua. While the spin-wave-like dispersive modes have been studied by neutron scattering since the 1970's, the quantitative exploration of excitation continua has been greatly advanced by direct-geometry time-of-flight spectrometers at pulsed neutron sources. High flux chopper spectrometers have proved to be excellent instruments for investigating such broad, diffuse inelastic scattering, and continue to have a large impact on research in this area. For example, early work on the S=1/2 Heisenberg antiferromagnetic chain system $KCuF_3$[35] employed MARI to study the free spinon continuum spectrum. Direct measurements of the dynamic spin correlation function, $S^{\alpha\alpha}(\boldsymbol{Q},\omega)$, were made possible by orienting the sample with the chain axis parallel to the incident neutron direction, resulting in magnetic scattering with azimuthal symmetry. This allowed integration of the signal over many detectors without loss of information. A complementary approach attempts to survey large regions of reciprocal space simultaneously. This method was used on MARI measure a similar continuum in the spin-Peierls compound $CuGeO_3$.[36] These experiments have tested many aspects of one-dimensional quantum magnetism for the first time, and have also revealed unanticipated features such as a "double gap" structure in alternating spin chains.[37]

The construction of the new MAPS spectrometer at ISIS has made the survey approach much more practical for new materials. Figure 4 shows recent measurements of $KCuF_3$ carried out at MAPS and plotted in an extended zone scheme. Such characterization of broad features of excitations will be much easier to pursue with the new ARCS instrument.

More recently, work on coupled quasi-one dimensional systems embedded in real three-dimensional materials has found that sharp, dispersive modes can coexist with broad continua. In particular work on ordered, coupled S=1/2 chains has verified the existence of a longitudinal mode related to zero point fluctuations.[38] A flexible machine like ARCS would be ideal for studying systems like this with complicated dynamics where broad energy scales are needed.

The gap from one to two dimensions can be bridged by spin-ladders, realized in compounds with two or more directly adjacent, strongly interacting spin chains.[39] Several cuprate spin ladders have been identified, including one that can be doped and becomes superconducting under pressure.[40] Many of these materials are characterized by exchange constants comparable to or larger than the high-T_c compounds,[41] and ARCS would be ideally suited to study their spin dynamics.

1.2.3 Low-Dimensional Conductors

Unusual behavior of a different nature occurs when itinerant electrons are present in low-dimensional structures. In some cases, interactions can lead to a reduction of the electronic energy, which is stabilized by a nested Fermi surface favored in lower-dimensional systems. When the electron-phonon interaction is strong, a soft phonon at the nested wave vector and a collective mode oscillation of the conduction-electron charge density develop in a "Peierls transition" below a critical temperature. This transition is in fact a metal-insulator transition to a charge-density-wave (CDW) ground state because an energy gap forms at the Fermi surface. On the other hand if Coulombic interactions dominate, then a modulation of the conduction electron spin density occurs, resulting in an antiferromagnetic ground state in a "spin density wave (SDW) transition."[42] In real materials, represented by both inorganic and organic examples, a variety of ground states occur including CDW, SDW, and even superconductivity.

The CDW ground state has a unique dynamics. In place of the original vibrational excitations of the lattice, two new branches are found: one that has a linear momentum dependence, and a second with a finite energy at the Γ point.[43] The first mode is associated with phase excitations of the charge modulation, while the second mode corresponds to excitations of the charge modulation. In principle, the phason mode is a Goldstone mode, but for real materials, impurities, which pin the charge density wave to the lattice, gap the mode. The appropriate description for the excitations of the SDW ground state is less clear.[44]

To date there have been very few neutron scattering experiments on quasi-one-dimensional metallic conductors. One example is the organic molecular conductor $TEA(TCNQ)_2$. Using inelastic neutron scattering to investigate the lattice dynamics, Carneiro *et al.* observed a well-defined Kohn anomaly and gaps in the excitation spectrum.[45] From the temperature dependence of the spectrum, the electron-phonon coupling was found to dominate, as expected for this type of system.

Inelastic neutron scattering would be an important tool for studying the dynamics of the magnetism in these low-dimensional conductors. As a starting point, the SDW ground state below ~12 K could be investigated in $(TMTSF)_2PF_6$. More complicated behavior is observed in the related compound $(TMTSF)_2ClO_4$. This system superconducts at low temperatures, but under an increasing applied magnetic field the superconductivity gives way to a series of different SDW states.[46]

As mentioned above, many of these low-dimensional conductors have complicated phase diagrams with a variety of different ground states. In some of the quasi-two-dimensional systems, there is even speculation of a quantum-Hall regime. Two major limitations in studying these materials have been the small sample size of available crystals and the fact that the magnetic moments in these systems are typically between 0.1 and 0.003 μ_B. Improvements in sample growing techniques are producing larger crystals and the greatly enhanced performance of the ARCS spectrometer could open up this area of research.

1.2.4 Magnetism in Actinide Materials

Actinide materials have attracted much attention because their magnetism can be considered intermediate between the local-moment magnetism of most rare-earth magnets and the itinerant magnetism of transition metals. A consequence is that actinide compounds often exhibit 'unusual' magnetic properties, which are being studied by H. Nakotte.

The hybridization of the 5*f* electrons with the electron states of the ligands plays a central role in the magnetism of actinide compounds. Hybridization effects are probably responsible for the quasielastic response seen in many uranium compounds, like UGa_3[47] or URhAl.[48] Compared to Ce Kondo compounds, however, this quasielastic response is much broader in energy and its temperature dependence is unusual. This could be due to the large anisotropy of the hybridization effects, but this behavior could be tested by inelastic neutron scattering measurements on the large single crystals that have become available recently.

For the majority of actinide compounds, their magnetic anisotropy is thought to originate with anisotropic 5*f*-ligand hybridization. This is different from the single-ion crystal-field anisotropy typically found in rare earths. On the other hand, some actinide compounds display local-moment behavior, and one might expect to see crystal-field excitations in the inelastic scattering response. Crystal fields were indeed established in some actinide oxides, like UO_2[49] or PuO_2.[50] Until recently, however, an extensive search for crystal fields in actinide intermetallic compounds has been largely unsuccessful, with the notable exception of UPd_3.[51] There is recent evidence for crystal-field excitations in UPdSn[52] and UPd_2Al_3,[53] but their temperature dependence is not fully consistent with a simple crystal-field picture. Measuring the dispersions of these excitations on single crystals should help us understand whether other or additional mechanisms contribute to these excitations.

Spin wave excitations are another important topic of research in actinide magnetism. Below the magnetic ordering temperature, there is strong evidence for spin waves in uranium compounds such as UFe_2[54] and UNiGe.[55] In the case of UNiGe, the excitation persists to temperatures far above T_N. The 5*f*-ligand hybridization contributes to the spin-wave stiffness, and this stiffness has a strong temperature dependence, presumably due to two-magnon interactions. Evidence exists for the formation of a gap in the spin-wave spectrum at $\boldsymbol{q} = 0$, suggesting possible 3-dimensional analogs to the one-dimensional quantum magnetism of Sect. 1.2.2. Studies on single crystals with the ARCS instrument can provide the detailed information on spin waves and their dispersion that are needed to understand how magnons propagate in systems with essentially delocalized 5*f* states.

1.2.5 Heavy Fermion Magnetism and Superconductivity

There are exciting developments in the area of heavy fermion systems to which experiments at ARCS could make important contributions. R. Osborn will use ARCS in studies of the dynamic susceptibility of disordered heavy fermions, usually containing uranium ions that exhibit unusual power-law correlations at low temperature. Pulsed neutron spectroscopy at IPNS and ISIS have already revealed a novel scaling of the magnetic response which is universal in ω/T over a very wide dynamic range of frequencies and temperatures.[56] This is believed to result from quantum critical scattering associated with the proximity to a T=0 phase transition. A key question to be resolved is the dimensionality of the correlations. At high temperatures (>30K), the scattering is single-ion in nature but there is evidence of a crossover at low temperatures to a quantum-disordered regime showing weak antiferromagnetic correlations. The scaling properties are reminiscent of marginal Fermi liquids, proposed to explain the unusual normal-state properties of high-temperature superconductors, which also undergo quantum phase transitions as a function of doping. We intend to pursue the systematics of this scaling as a function of composition and other thermodynamic variables. Pulsed neutron time-of-flight spectrometers have been especially valuable in determining this scaling behavior because of the wide range of accessible energies and angles. The angular range is also necessary in subtracting the phonon scattering reliably.[57]

Inelastic neutron scattering has also proved powerful for testing the connection between magnetic fluctuations and superconductivity in heavy-fermion compounds. In UPd_2Al_3, it was found that the inelastic scattering changes abruptly at the superconducting temperature.[58] The inelastic neutron scattering data strongly suggest a pairing mechanism mediated by spin fluctuations.[59]

1.2.6 Metal Insulator Transition in Oxides

Oxides of 3d metals other than copper also show intriguing behavior dominated by electronic correlations. R. Osborn has started a program to study the magnetic correlations close to the metal-insulator transition in $La_{1-x}Sr_xTiO_3$, a three-dimensional electron analogue to the well known hole-doped cuprate $La_{2-x}Sr_xCuO_4$. These compounds undergo a Mott-Hubbard transition at $x \sim 0.05$, which is accompanied by a divergence of the d-electron effective mass according to specific heat and magnetic susceptibility.[60] This implies a narrowing of the energy scale of d-electron magnetic fluctuations into the range accessible by ARCS. Early studies indicate that the response is enhanced close to the transition, but this is difficult to determine unambiguously because of the limited sensitivity of current instrumentation. Optimized for sensitivity in the relevant energy range, ARCS will enable single crystal investigations of the transition to the Mott-insulating antiferromagnetic phase. G. Aeppli and collaborators are studying manganites of composition $La_{1-x}Pb_xMnO_3$ which are fully spin polarized metals by virtue of the double-exchange mechanism. The same microscopic physics also underlies the recently discovered "colossal magnetoresistance" effect that has potential for technological applications. While these materials have been known for more than 40 years, an experimental characterization of the excitations of the double-exchange Hamiltonian has only recently been achieved through studies at ISIS.[24] The manganites show rich phase diagrams with competing charge-localized and metallic states whose spin dynamics is only beginning to be studied. The flexibility and dynamic range of ARCS will undoubtedly prove useful in this area as well.

Whereas various 3d metal oxides are currently under intense investigation in laboratories around the world, compounds based on 4d/5d transition metals are potentially very interesting materials that are only now beginning to attract serious attention of condensed matter physicists. The large size of the outer electron cloud in the 4d/5d series results in greatly increased hybridization and potentially new physics. For this reason we call such materials "extended electron" compounds. Examples of recent interest include $SrRuO_3$, which is a "badly metallic" ferromagnet (i.e. with electronic properties similar to those of the normal state of high Tc superconductors, fullerenes, and organic metals), and the derivative compound Sr_2RuO_4, now suspected to be a p-wave superconductor.[61] There is much to be learned by studying the magnetic fluctuation spectrum of such materials, and in some cases perhaps even optical phonons. There are several technically challenging aspects of such measurements that may be well suited to ARCS. The rapidly diminishing form factors force measurements with high energy transfers at small wavevectors. Typically only small single crystals or powders are available. The itinerant nature of the magnetism in many of these materials also can result in excitation continua. The effective dimensionality of the ruthenates, as well as that of the cuprates, can be systematically changed in the Ruddlesden-Popper scheme which interpolates quasi-continuously between two and three dimensional electronic structure by changing the number of directly adjacent transition metal oxide layers, resulting in compounds such as Sr_2RuO_4, $Sr_3Ru_2O_7$, etc. The ruthenates and other analogous and isomorphous compounds in this series will be studied by IDT members.

1.3 Chemical Physics and Other Applications

Inter- and intramolecular vibrations of chemical substances are often studied with inverted-geometry backscatter spectrometers having large-area crystal analyzers. Such instruments offer an excellent combination of energy resolution and intensity. Their drawback is that they provide little information on the momentum transfer of the scattering. Momentum resolution is not a goal of many measurements where the scattering is incoherent or the sample is isotropic. It is therefore expected that most of the chemical physics community will continue to perform experiments on backscatter and filter-difference spectrometers. With its high sensitivity, however, the ARCS instrument could be useful for some studies of intermolecular vibrations. The momentum information provided by the ARCS instrument could be used for obtaining spatial information on the molecular motions as described in Sect. 1.3.1. and 1.3.2.

ARCS could prove useful for other scattering experiments where energy resolution is important. There has been recent interest in how local structural distortions in crystalline materials (with dimensions comparable to the size of the unit cell) alter electrical and magnetic properties. Some such studies of elastic diffuse scattering require an instrument with good E resolution and high sensitivity, but not particularly good Q resolution.

1.3.1 Chemical Physics

Internal vibrations of molecular crystals exhibit very little dispersion owing to the weakness of intermolecular forces compared to intramolecular forces. Furthermore, the specificity and sensitivity associated with scattering by hydrogen is one of the main strengths of inelastic neutron scattering in comparison with optical methods of spectroscopy for organic samples. The present emphasis on incoherent scattering from hydrogen has caused most research on vibrations of molecular crystals to be performed with inverse geometry spectrometers.

Even for incoherent scattering, however, a potentially useful feature of the ARCS instrument could be its flexible use of high energy incident neutrons, which provides control of the Q of the scattering process. Varying Q for inelastic scattering is helpful for identifying features in scattering spectra that originate with fundamental modes or overtone modes of vibration. Specifically, binary overtones and combinations, including combinations known as phonon side bands involving internal plus external modes, become enhanced relative to fundamental ("one-phonon") excitations at higher Q because they depend on Q^4 rather than Q^2. This leads to spectral congestion at higher frequency in an inverse geometry instrument. The phonon-wings actually steal intensity from the fundamental transitions. Furthermore, the Debye-Waller factor decreases as Q increases. It should be possible to use ARCS to obtain data from which one can deconvolve spectra into their components to obtain the true single-phonon hydrogen-weighted density of states function and to obtain higher resolution spectra.

With its high sensitivity the ARCS instrument could be useful for some studies of intermolecular vibrations that are not possible with an inverse-geometry instrument. The momentum information provided by the ARCS instrument could be used for obtaining spatial information on the molecular motions for each vibration. This requires coherent scattering and either oriented single crystals, or cubic structures. An example of this type of experiment is given in the work of Heid and coworkers on the vibrations of C_{60}.[62] In this work it was shown that it was possible to discriminate between modes belonging to the same symmetry class on the basis of their eigenvectors, a property that can be used as a stringent test of theoretical models. Eigenvectors predicted by the ab-initio theory were found to be in very satisfactory agreement with the experimental data while those obtained from the phenomenological model turn out to be less reliable. The high sensitivity of the ARCS instrument could alter the historical balance of instrument usage for work in chemical physics.

1.3.2 Deep Inelastic Neutron Scattering Studies of Hydrogen

Deep inelastic neutron scattering (DINS) is the only experimental technique that provides direct access to the single particle atomic momentum distribution.[63] Thus, DINS provides a powerful probe of the local environment and single particle properties, particularly in quantum systems such as helium and hydrogen. For example, liquid helium has occupied a central role as a model system for the study of strongly interacting fluids,[64,65,66] critical phenomena,[67,68] and even the birth of the Universe.[69] One of the most interesting properties is the appearance of a superfluid phase that is a macroscopic manifestation of Bose condensation,[70] a topic of considerable recent interest with its observation in dilute gases[71,72]and excitonic systems.[73] However, liquid He offers far richer behaviors than weakly interacting dilute gas, and DINS has been central in exploring the role of the condensate in these fascinating behaviors.[74,75,76,77,78,79]

The momentum distribution of hydrogen is dominated by zero-point quantum motion at modest temperatures. The momentum distribution therefore provides a unique microscopic perspective on the dynamics of hydrogen[80] in environments including hydrogen bonds, metal hydrides, and novel environments such as for hydrogen adsorbed on buckeyballs and nanotubes. In addition, DINS measurements can also provide detailed information on the orientational potentials experienced by the hydrogen molecule in important chemical and biological systems. The high intensity and large angular coverage expected for ARCS will enable detailed studies of the momentum distribution and its effects on external parameters, such as pH.

Deep inelastic neutron scattering (DINS) measurements are having a major impact on the understanding of H and H_2 adsorption on surfaces. For example, gases adsorbed in bundles of carbon nanotubes have received considerable attention. Much of the interest centers on phenomena intrinsic to quasi-one-dimensional phases of the hydrogen.[81,82] There is also a significant technological interest in hydrogen storage in novel carbon materials, prompted by plans for feasible, cost-effective and environmentally-friendly fuel cell technology. Issues both basic and applied have stimulated much research on the adsorption of H_2 in carbon nanotubes.[83,84,85,86,87,88,89] Figure 5 shows a recent measurement of H_2 adsorbed on carbon single-wall nanotubes. The scattering consists of several peaks, whose shape mirrors the atomic momentum distribution. The peaks correspond to transitions between different rotational energy levels, and occur at the recoil energy plus the rotational energy. The width of the peaks in Q provides information on the localization, which is higher than the bulk value, but lower than for H_2 on graphene (Grafoil).[90] More interestingly, the rotational transition energies from the ground state are significantly different from those of free molecules, indicating that the molecules experience a significant orientational potential when adsorbed. These measurements, coupled with model calculations, indicate the existence of significantly hindered rotational motion of H_2 trapped in the interstitial channels of a nanotube bundle. Currently the small amount of material available, and the low count rates for these measurements limit such studies. ARCS would eliminate these restrictions and make DINS studies a routine part of the development of new materials for energy storage.

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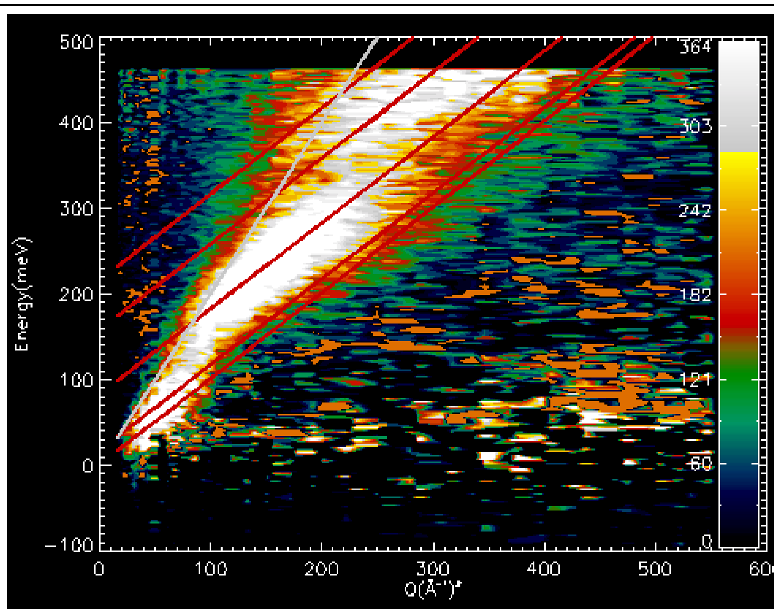

Figure 5: A contour plot of scattering measurements at an incident neutron energy of 500meV with the background removed. The color bar gives shows the relation between color and intensity. The red lines are the recoil energy plus the energy of the rotational transitions as labeled on the graph. The white line is the predicted recoil energy for atomic H.

peaks in *Q* provides information on the localization, which is higher than the bulk value, but lower than for H_2 on graphene (Grafoil).[100] More interestingly, the rotational transition energies from the ground state are significantly different from those of free molecules, indicating that the molecules experience a significant orientational potential when adsorbed. These measurements, coupled with model calculations, indicate the existence of significantly hindered rotational motion of H_2 trapped in the interstitial channels of a nanotube bundle. Currently the small amount of material available, and the low count rates for these measurements limit such studies. ARCS would eliminate these restrictions and make DINS studies a routine part of the development of new materials for energy storage.

1.3.3 Diffuse Scattering Measurements of Local Structure

Diffraction experiments, which measure static atomic structures and magnetic structures, are one of the main fields of research for neutron scattering in materials and condensed matter sciences. In general, the sharp peaks from Bragg scattering give information about the long-range structure of the average lattice, whereas the elastic diffuse scattering arises from local deviations from the average structure. Examples of such local deviations include chemical short-range order, static atomic displacements off of regular crystallographic sites, and local fluctuations in the magnetic structure. Although ARCS is not designed as a diffractometer, its ability to measure elastic diffuse scattering would offer a powerful and unique capability.

For studies of elastic diffuse scattering, *Q* resolution is less important because the scattering has no sharp features in *Q*. Inelastic processes such as phonon and magnon scattering contribute to the background of diffraction patterns measured with conventional time-of-flight neutron diffractometers. The energy resolution of ARCS will isolate the true elastic scattering, permitting more quantitative determinations of the local structure than is possible with a conventional neutron diffractometer without the energy selectivity of a Fermi chopper. Energy selectivity is a critical requirement because the intensity of the elastic diffuse scattering is typically very weak compared to inelastic scattering sources that also contribute intensity between the Bragg peaks. ARCS will permit isolation of the elastic diffuse scattering, $S(Q,0)$. Furthermore, because local structure often affects dynamical processes in solids, having the full $S(Q,E)$ makes it possible to see the effect of local structure on the dynamics of the solid.

J. L. Robertson has measured the chemical short-range order (SRO) and static atom displacements in a single crystal of Fe-Ni Invar. The neutron diffraction data shown in Fig. 6 have broad features between the Bragg peaks. This measurement was performed at a reactor source without a polarized neutron beam. It was time-consuming, and also required additional measurements with synchrotron radiation for quantitative interpretation.[101] The work eventually isolated spin-spin and spin-chemical correlation functions in addition to the SRO and static displacements. The incident intensity and large-area detector coverage of ARCS would greatly facilitate such measurements of the elastic diffuse scattering.

1.3.4 Characterization of Novel Materials

The vast majorities of novel materials are developed by solid-state chemists through guided trial-and-error using susceptibility, specific heat, and transport measurements as diagnostic tools. Because current neutron scattering instrumentation requires large amounts of material, this powerful sample characterization tool is typically only applied by specialists as a means of explaining previously-discovered anomalies. The high sensitivity option of ARCS will enable rapid surveys of excitations in new materials that are available only in small quantities. We expect that this will make neutron spectroscopy an important tool for the initial characterization of novel materials, thus bringing more detailed information about a larger amount of different materials to a broader community of materials scientists.

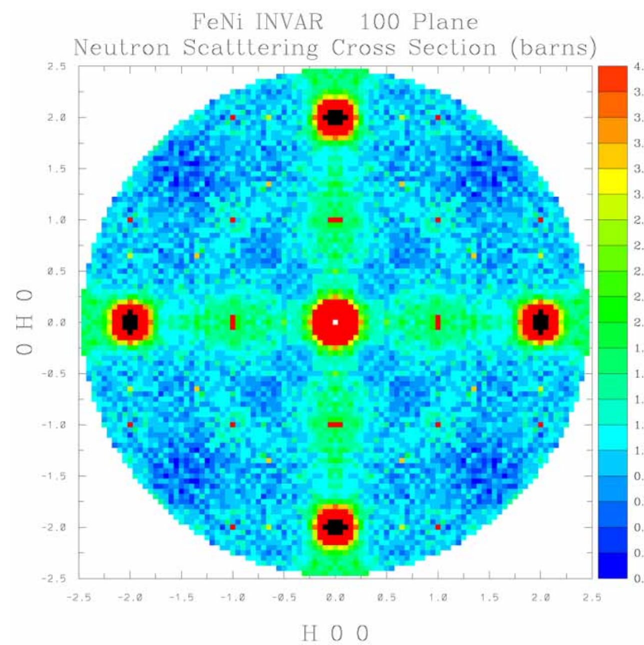

Figure 6: Neutron diffraction pattern from a single crystal of Fe - 36.8 at.% Ni obtained using the HB3 3-axis spectrometer at ORNL.

2. IDT Organization

2.1 Management

A large construction project, built at the SNS but managed through a university with personnel at national laboratories, is a managerial challenge.

2.1.1. Personnel

Figure 7 shows the proposed organizational structure for the ARCS project. DOE BES will interact directly with the P.I., Brent Fultz, who will be responsible for executing the project and for timely reporting on progress. An Executive Committee will also inform DOE BES of significant developments or report on issues and progress as requested. The person in the central position of Project Manager, Doug Abernathy, will have an appointment as a Visiting Associate at Caltech. This flexible appointment is planned without a salary, but a Caltech salary is possible if this is desired later. It is expected that the Project Manager will be located primarily at the Argonne National Laboratory to utilize the support staff and the design expertise in the neutron Instrument Systems group under Kent Crawford. Nevertheless, the Visiting Associate appointment will allow the Project Manager to operate the purchasing and accounting systems at Caltech. For these interactions and for the award management by the P.I., an administrator will be required at Caltech. The project communications plan provides a videoconferencing facility will in the P.I's Caltech office that links to Argonne and Oak Ridge. Communication between Caltech and Argonne has been occurring on a near-daily basis.

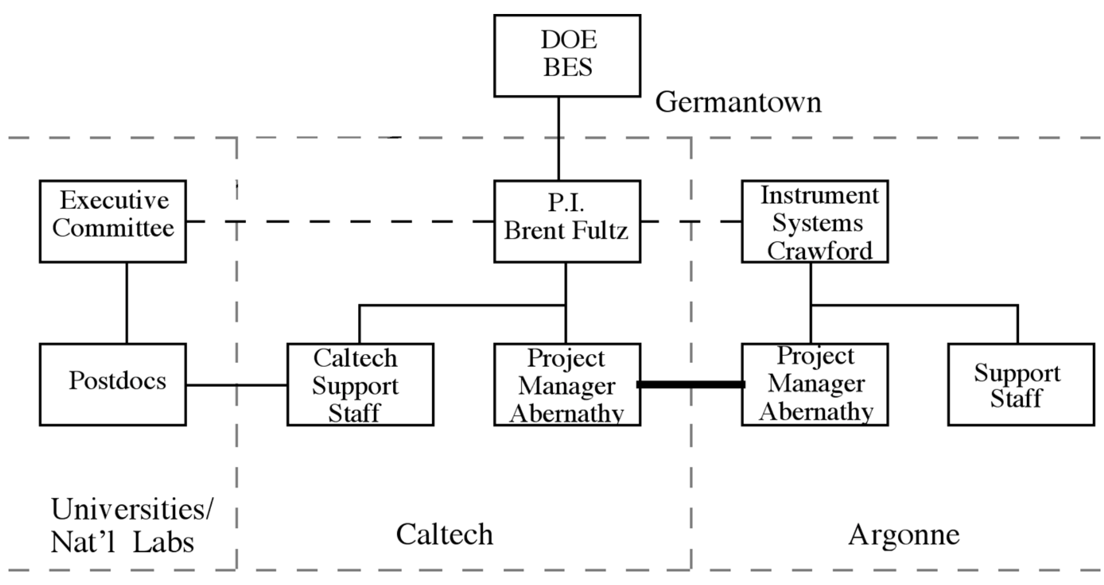

Figure 7: Organizational chart of the ARCS project management. Light dashed lines bound geographical locations.

After the design stage is complete and construction begins at the SNS site, a member of the ARCS team must have a regular presence at Oak Ridge. In the first stages of heavy construction, this person could be a staff engineer at ORNL, working under contract with ANL. This person will execute the testing of components, assembly of the ARCS instrument, and system testing where possible. Close contact with the Project Manager and P.I. will occur through videoconferencing facilities. In addition, this person will need to interact with the Caltech business systems to acknowledge receivables, for example.

As early as two years into the project, the ARCS project manager will move to Oak Ridge, eventually making the transition to instrument scientist. The instrument scientist will participate in the final configuring and commissioning of ARCS. He or she will become familiar with the instrument before operations begin at the SNS. It is expected that DOE BES agrees to support this instrument scientist position through the SNS.

2.1.2. Role of the Executive Committee

The Executive Committee will assess all major design issues and oversee the project execution so that the ARCS instrument will benefit a broad science program as quickly as possible. The Executive Committee is organized and selected to be small and engaged, with a membership listed below. The Committee will approve the detailed instrument design and the work breakdown structure that will be prepared as part of the design effort. At least some of the members of the Committee (perhaps those who are most active in the early stages of the project), together with the P.I. and the Project Manager, will serve as the change control board to assess proposed changes to the scope of the ARCS project. The broad-based science orientation of the Executive Committee makes it the best organization for directing the development of the higher-level software that will be required for interpreting data from the ARCS instrument. Involvement in the software development will likely be the largest effort by nenbers of the Executive Committee.

The members of the Executive Committee will seek outside sources of funding for some aspects of the ARCS project. This could, for example, include the writing of a proposal to the NSF or a private foundation to fund a high-field magnet or a dilution refrigerator for the sample volume of ARCS. The Executive Committee will also seek funding for scientific software development. Brent Fultz, Oscar Bruno, and others are presently organizing a white paper proposal to the NSF in the area of the mathematics of materials, and some of this effort will be focused on scattering calculations. The software and sample environments for ARCS will be planned in consultation with the IDTs for the CNCS and backscatter spectrometers. (The anticipated sharing of sample environment facilities was an important reason for selecting a horizontal geometry for the ARCS spectrometer.) In the later stages of the project, it is expected that the Executive Committee will oversee the development of the user program for the ARCS spectrometer. Members of the Executive Committee will negotiate with DOE and SNS management for the fraction of instrument time allocated directly to the IDT, and the Committee will then allocate beamtime requests originating from within the IDT.

2.1.3. Financial Management

Figure 8 shows the financial structure of the ARCS project. The P.I. is responsible for all funding and cost reporting to DOE. The salary of the Project Manager and his engineering staff will be provided through a subcontract to the Argonne National Laboratory. It is expected that the P.I. will be consulted for any Argonne internal employee evaluation of the Project Manager.

In practice all purchases, contracts, and awards will occur through the Business Services Office of Caltech. Bill Cooper, Caltech's Director of Purchasing Services has proposed how his office will interact with the Project Manager (see his memo in an appendix). The Project Manager will have the day-to-day contact with this Caltech office. In the May 1 meeting between the P.I., Project Manager, and Caltech Purchasing Services, it was resolved that it is not necessary for the Project Manager to present in person at Caltech, except for a few days near the beginning of the project. The Project Manager, or an administrative staff person at Argonne, can be qualified to serve as a Caltech buyer for purchases of less than $5,000.

The Project Manager will control the construction at Oak Ridge and the funds to do so, even though these functions are performed out of Argonne and Caltech, respectively. In the time before the Project Manager moves to Oak Ridge, a staff person at Oak Ridge will need to confirm receivables and help reconcile invoices with purchase orders.

There is zero overhead for equipment purchases through Caltech, and subcontracts are charged Caltech overhead (presently 58 %) on the first 25 k$ of each multi-year subcontract.

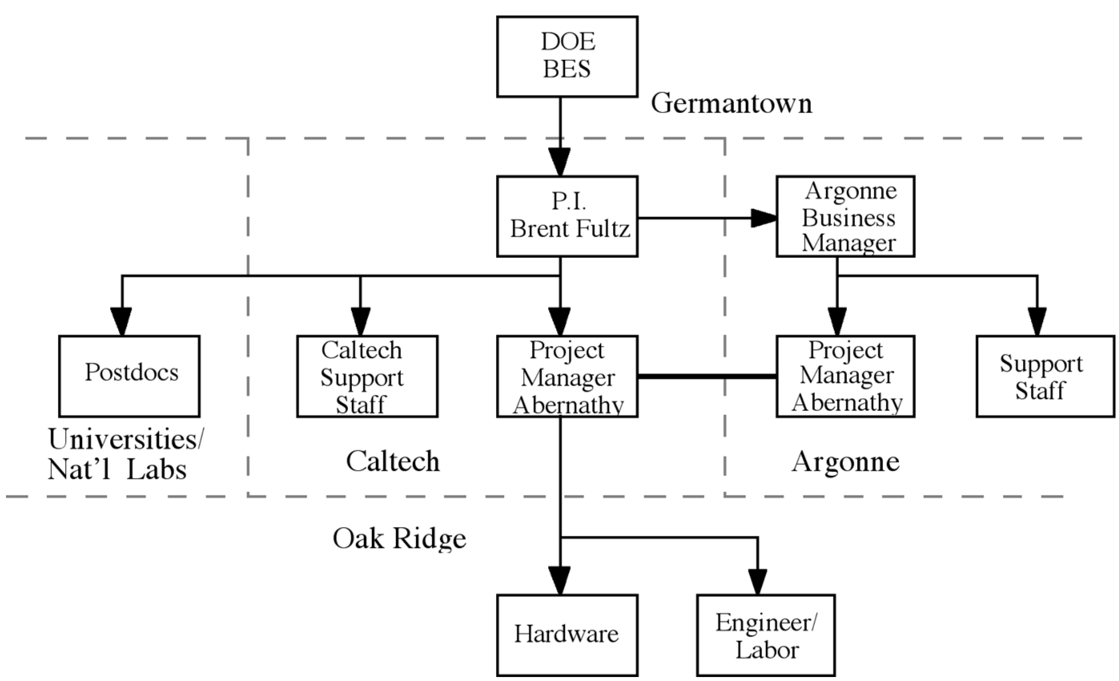

Figure 8: Funding channels for the ARCS project.

2.2 Instrument Development Team

2.2.1 Executive Committee Membership

Brent Fultz (California Institute of Technology) [Principal Investigator]:

Area of Expertise: Long-term interest in thermodynamics and kinetic processes in solids.

Relevant Experience: Ph.D. 1982 from U.C. Berkeley, faculty at Caltech since 1985. Fultz's group showed that vibrational entropy is important for the thermodynamics of alloy phase stability. In the last few years they have been measuring phonon spectra of materials by inelastic neutron scattering with all relevant inelastic spectrometers in the U.S. From phonon spectra, reasons for differences in vibrational entropy between alloy phases can often be identified. Recent measurements on cerium and uranium have shown large effects of electronic excitations on phase stability, the role of crystal field (Schottky) entropy, and thermal dependencies of the electronic structure in the case of uranium.

Doug Abernathy (Oak Ridge National Laboratory, Spallation Neutron Source) [Project Manager]:

Area of Expertise: Scattering instrument design, construction and operation. Surface scattering and x-ray photon correlation spectroscopy.

Relevant Experience: Senior Technical Associate at AT&T Bell Laboratories (1985-1987), working with Dr. G. Aeppli. Designed operational equipment and controls for an eV resolution backscattering monochromator and analyzer for the HFBR. Ph.D. (1993) Massachusetts Institute of Technology, studying the structure and phase behavior of the Si(113) surface using a specially constructed x-ray surface diffraction UHV chamber. Joined the European Synchrotron Radiation Facility (1993-1998) as an instrument scientist for the Troika beamline, studying surface diffraction and the production and use of coherent x-rays. Currently working for the Instrument Systems of the Spallation Neutron Source at ANL (1998-) to develop concepts for neutron spectrometers.

Ward Beyermann (University of California, Riverside):

Area of Expertise: Correlated-electron physics and biomaterials, thermodynamic and transport properties, neutron scattering, measurements at low temperatures and in high magnetic fields.

Relevant Experience: Ph.D. (1988) from the University of California, Los Angeles. Faculty at University of California, Riverside since 1990. My group has investigated the thermodynamics, transport, magnetic excitations, and structure of actinide and lanthanide intermetallic materials. Many of these measurements are conducted at low temperatures and in high magnetic fields up to 60 T. We have also conducted inelastic and elastic neutron scattering experiments at NCNR, IPNS, and the Lujan Center. Some of the phenomena that we are interested in include: quantum critical fluctuations and disorder in systems that exhibit non-Fermi liquid properties, many-body quadrupolar interactions in intermetallics, the influence of large magnetic fields on correlated-electron behavior, and vibrational excitations in Fullerenes, nanotubes, and biomaterials.

Robert J. McQueeney (Los Alamos National Laboratory):

Area of Expertise: Condensed matter physics, correlated electronic systems, lattice and spin excitations, neutron and x-ray scattering, pulsed neutron scattering instrumentation.

Relevant Experience: Ph.D. (1996) from the University of Pennsylvania. Visiting Scientist at Caltech (1998). Joined staff at Los Alamos National Laboratory, Manuel Lujan Jr. Neutron Scattering Center (1998). Instrument scientist for the PHAROS chopper spectrometer. Principal investigator for the VERTEX chopper spectrometer project. Significant user of many neutron and x-ray facilities: Lujan Center, IPNS, HFIR, HFBR, ISIS, NSLS, and APS. Software programming experience for analysis and reduction of neutron scattering data and calculation of relevant correlation functions.

Steve Nagler (Oak Ridge National Laboratory):

Area of Expertise: condensed matter physics, low-dimensional and quantum magnetism, phase transitions.

Relevant Experience: Ph.D. (1982) from the University of Toronto with neutron scattering experiments at Chalk River. Visiting Scientist at IBM Research (Yorktown Heights) 1982-84. Professor of Physics at University of Florida (1984-1996). Joined ORNL in 1995 as a senior research scientist. At UF S. E. Nagler built an x-ray lab based on a rotating anode and a large Huber 4 circle (same as used at most synchrotron scattering beamlines), and was a founding member of the MRCAT consortium to build beamlines at the Advanced Photon Source. Extensive use of major scattering facilities, including x-ray synchrotron sources (SSRL and NSLS), continuous neutron sources (ILL, HFBR, and HFIR) and the pulsed neutron source ISIS, particularly the chopper spectrometers MARI and HET.

Ray Osborn (Argonne National Laboratory):

Area of Expertise: Condensed matter physics, neutron scattering studies of strongly correlated systems, rare earth and actinide magnetism, transition metal oxides. Running pulsed neutron spectrometers and user program.

Relevant Experience: Ph.D. (1989) University of Southampton UK. Scientist at ISIS pulsed neutron facility, UK, (1985-1992), responsible for the HET chopper spectrometer and user program. Scientist in Materials Science Division of Argonne National Laboratory (1992-) running a scientific program in strongly correlated electron systems (spin dynamics and crystal fields in heavy fermion compounds, quantum critical scattering in non-Fermi liquid compounds), and CMR compounds (magnetic correlations in manganites). Responsible for LRMECS spectrometer at IPNS and user program. Have extensive experience of using a wide range of inelastic neutron spectrometers at different sources, including triple-axis spectrometers, reactor and pulsed time-of-flight spectrometers (direct- and indirect-geometry), polarized neutrons, on both single crystal and polycrystalline materials.

2.2.2 Instrument Development Team Members

The membership of the IDT does not include all scientists who have expressed interest in using the ARCS instrument. Such a list would be nearly double the length of the list below. The persons listed below as IDT members are those most willing to attend a national workshop and contribute to the discussion on the instrument and its science program. Most have contributed to Sect. 1 of this proposal, and many were at the April 30 meeting at Caltech. This group and others has been receiving regular communications about the ARCS concepts and the preparation of this proposal, for example.

D. Abernathy, Argonne National Laboratory
W. Beyermann, University of California, Riverside
C. Broholm, Johns Hopkins University
O. Bruno, California Institute of Technology
T. Egami, University of Pennsylvania
B. Fultz, California Institute of Technology
B. Hudson, Syracuse University
C-K. Loong, Argonne National Laboratory
M. Manley, Los Alamos National Laboratory
R. McQueeney, Los Alamos National Laboratory
H. Nakotte, University of New Mexico, Las Cruces
F. Mezei, Los Alamos National Laboratory
S. Nagler, Oak Ridge National Laboratory
R. Osborn, Argonne National Laboratory
J. L. Robertson, Oak Ridge National Laboratory
P. Sokol, Pennsylvania State University
J. Tranquada, Brookhaven National Laboratory

2.3 User Program

Because the energy resolution of ARCS will be similar to that of thermal-beam triple-axis spectrometers, there will be some overlap of these user communities. However, ARCS will offer dramatically improved efficiency for measurements of dynamics over wider ranges of frequency and momentum space. ARCS offers enormously improved sensitivity to inelastic scattering for energy transfers between 50 meV and 500 meV. Second, the large solid angle detector bank will enable experiments on systems where the scattering is distributed over a wide range of energies and momenta, as is the case for many novel magnetic systems that do not possess well-defined dispersion surfaces. Even for more traditional mapping of dispersion surfaces the phase space gains can be substantial, since large regions of $(\boldsymbol{Q},\omega)$ must sometimes be surveyed to locate all dispersive modes.

Building a successful user program around ARCS will require an instrument scientist. This person must be involved actively with the later stages of construction, meaning that a search for this person should begin no later than two years after the project is begun. Ideally he or she should join the project as soon as the SNS budget permits to enable active participation in the design of the instrument. We expect that Dr. Doug Abernathy, the ARCS Project Manager, will make the transition to role of instrument scientist for ARCS. Dr. Abernathy's appointment as instrument scientist would ensure continuity from the design and construction phase of the project through to commissioning and establishment of the fully functioning user program. It is assumed that the SNS will then bear the responsibility of operating ARCS.

Perhaps the most pressing problem in building a vibrant user program for the ARCS spectrometer, and for the SNS in general, is the development of the neutron scattering community in the United States. Much academic research has migrated away from neutron scattering and towards photon scattering as national synchrotron facilities have come to strength over the past decades. This transition was a gradual one, and a gradual reversal is not well matched to the leap in capabilities for neutron science that will be provided by the SNS. Faculty in physics, chemistry and materials science will be motivated towards neutron science once the SNS is available, but this transition could be expedited through some planning now. Although this issue extends well beyond the present proposal, the ARCS project would include two components to help in this transition. First, the project will provide for some focused research by members of the Executive Committee. The focus is on the software development needed to obtain scientific results from the ARCS instrument, but this work must be performed with realistic assumptions using real data. Since this software effort requires deliverables, the work is better suited to postdoctoral fellows than graduate students. Graduate students will necessarily be alerted to this university-based effort, however, and it is expected that they will be lured into the scientific excitement of the science components of the work.

A second way to expand the user base is to pull in researchers from adjacent fields. Specific tasks of software development for the ARCS project require specialists in theory, scientific software, and applied mathematics. The persons contacted so far by the P.I. have embraced the excitement of being involved in the large national science effort of the next decade. Contacts between members of the ARCS Executive Committee and theorists in the condensed matter sciences are preliminary but promising. The prospect of using computational results directly in the full experiment simulations (Sect. 3.5.2) is appealing to theorists as a means of influencing experimental work, for example. It is expected that a workshop to discuss such issues and test their sociology will occur early in the ARCS project. Specialists in software engineering and applied mathematics bring a sophistication and professionalism to the computational aspects of large software projects that is not typical of condensed matter researchers. Oscar Bruno, Professor of Applied Mathematics at Caltech, has had a longstanding interest in scattering phenomena, and he has joined the ARCS Instrument Development Team.

3. Instrument Description

ARCS will be installed on flightpath 18 in the SNS experiment hall viewing the ambient water moderator. Figure 9 shows the layout of the instrument. The design was guided by the desire to maximize the flux at the sample and maximize the solid angle of detector coverage. This must be achieved with an energy resolution appropriate for the scientific program described in Sect. 1 (typically 1–3% of E_i). The instrument is designed for single crystal measurements from the outset. This implies a goniometer for crystal rotation and a detector array of linear position-sensitive detectors with few gaps in angular coverage.

The detectors are arrayed into two banks of multiple tiers. The low angle bank from –20 to +40 degrees is at a radius of 5.5 m from the sample. The 1.2 m PSDs are arranged into five tiers vertically. The detector tubes in the upper and lower tiers will be tilted off vertical to approximate the tangent to a sphere around the sample. The high angle bank extends from +40 to 140 degrees at a radius of 3.0 m from the sample. It comprises three tiers of 1.0 m PSDs. The detector banks extend out of the scattering plane by ±30 degrees. The total solid angle covered by all detectors is approximately π steradians, with about half the coverage by the low angle bank and half by the high angle bank. We intend to use SNS standard designs for detectors, detector electronics, and the data acquisition system. We also assume that we will be able to benefit from engineering work on instruments at the SNS that are further along the development cycle, such as shielding and vacuum calculations.

We have not yet considered coordination of the siting of the ARCS instrument with other spectrometers at the SNS. Several issues will have to be addressed.

- Floor space close to the target is premium real estate. For this reason the incident flight path of ARCS is longer than optimal. The performance at higher incident energies could be improved by moving the spectrometer as close as possible to the target.
- The radiation shielding for the incident flight path will be a structure with components shared with the adjacent instrument on flight path 17.
- The sample environment inserts will be subject to the same safety concerns as other cryogenic and furnace systems in use at the SNS.
- The vacuum vessel, being quite large, requires careful engineering to ensure it can be operated safely.

3.1 Incident Flight Path

ARCS will be located on flight path 18 viewing an ambient water moderator. We have not yet optimized the depth of poisoning of the water moderator. At present this depth is set at the midpoint of the moderator (2.5cm depth), which is appropriate for longer flight path instruments with maximum flux in the thermal energy range.

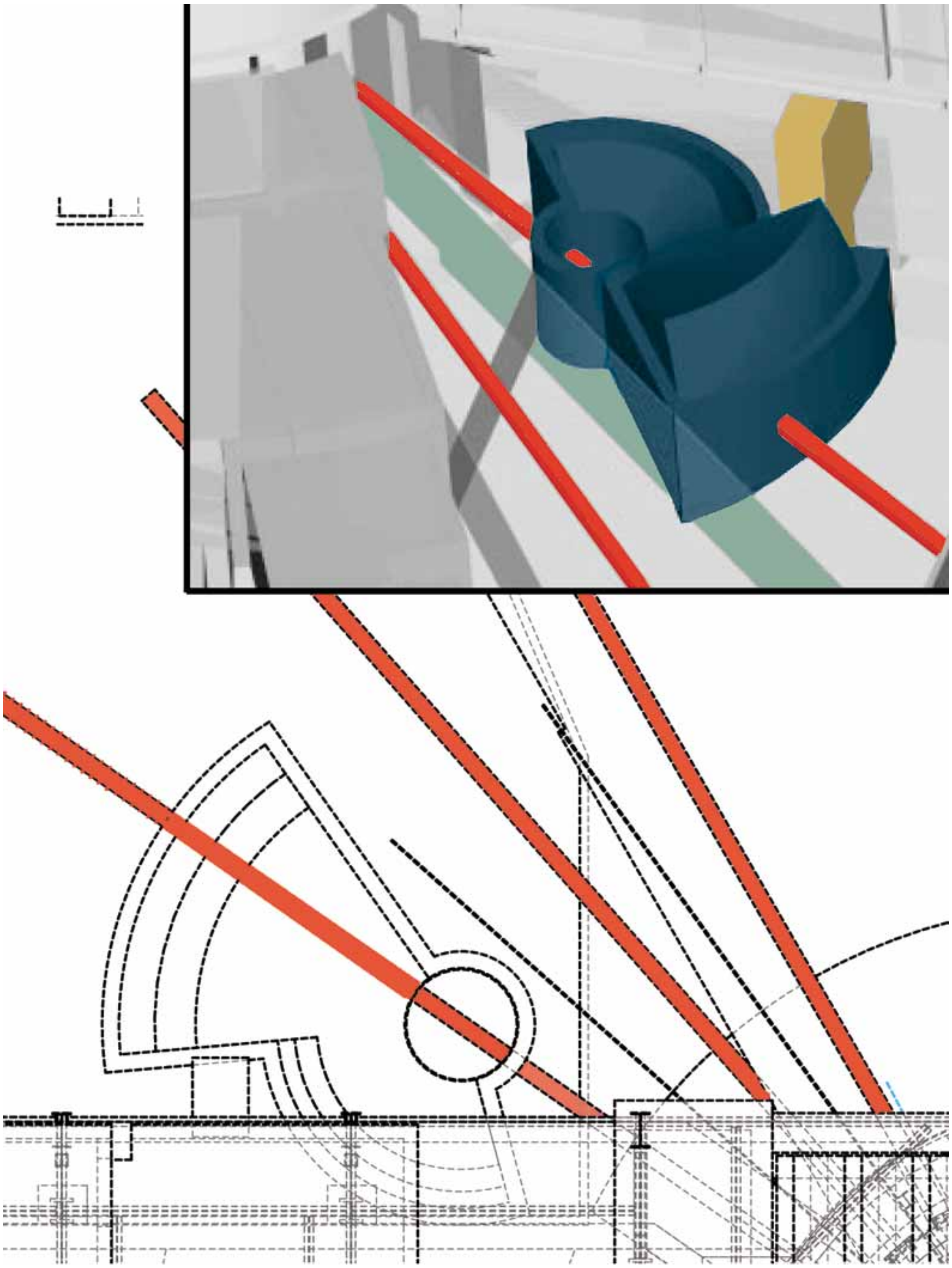

Figure 9: Layout of ARCS on Flight Path 18. Lower figure is top view. Flight paths are red, structural post in brown. Top: solid model of spectrometer.

The main components of the beam-line outside the biological shielding are:

1) The background-suppressing T-zero chopper
2) Spurion-suppressing disk chopper
3) The monochromating Fermi chopper
4) Adjustable slits and Soller collimators
5) Supermirror guide

One of the important advantages of direct-geometry chopper spectrometers on pulsed neutron sources is the flexibility they provide for optimizing the incident energy and resolution of the measurements. The incident energy can be varied from 10 meV to 1 eV by changing the Fermi chopper phase, speed, and, occasionally, the slit package. ARCS will take full advantage of this flexibility with at least three Fermi chopper slit packages to allow the measurement of energy transfers from 1 meV to 500 meV.

3.1.1 Biological Shielding

Because of the high power of the SNS source, massive shielding will be required for all instruments, but the shielding design must allow access to the beamline components. For typical instruments, the SNS Shielding Group is presently calculating the composition and thickness of the necessary shielding as a function of the distance from the target monolith. Detailed calculations by this group will be part of the ARCS design effort to ensure that the instrument will meet all SNS requirements for radiation levels in the target building. These calculations will likely be performed after shielding components have been designed and already ordered for other instruments in the SNS target building.

3.1.2 T_0 Chopper

The purpose of the T-zero (T_0) chopper, which will be located outside the biological shielding at about 9 m from the moderator, is to suppress the prompt pulse of fast neutrons produced when the proton beam strikes the target. A thick blade of the alloy Inconel is rotated at the source frequency of 60 Hz and phased to block the beamline when this background radiation is produced. There is a maximum energy for which the beamline is not blocked, depending on the blade width, rotation speed, radius of the blade, and position along the beamline. For ARCS this maximum energy is 670 meV. If an experiment required a higher incident energy than 670 meV, the T_0 chopper will be able to rotate at twice the source frequency (120 Hz) to pass neutrons of energies higher than 2 eV.

The ARCS spectrometer will benefit from the prototyping efforts underway in the SNS chopper development group at Argonne National Laboratory. A 60 Hz T_0 chopper will be installed in the GPPD instrument at IPNS, to help identify design flaws that are apparent only after extended usage. It is expected that a second prototype able to operate at 120 Hz will be tested in 2001.

3.1.3 Disk Chopper

The high rotational frequency of the Fermi chopper causes it to open many times between neutron pulses. One consequence is that neutrons of different velocities could pass through the Fermi chopper at its different openings. These spurious neutron pulses will be suppressed with a disk chopper rotating at a lower frequency. It is not clear if disk choppers will be sufficiently opaque to the most energetic neutrons, so we may use a rough premonochromator consisting of a coarse Fermi chopper, much as used for T_0 choppers on the HRMECS and LRMECS spectrometers at IPNS. By spinning at a lower multiple of the source frequency than the Fermi chopper, only neutrons of the correct energy will pass to the sample in the desired data acquisition time frame. Simulations will be undertaken to determine if this type of chopper will improve the pulse shape by cutting more completely the tail of the moderator pulse, or if a simpler disk chopper is sufficient.

3.1.4 Fermi Chopper

The Fermi chopper will be located 11.6 m from the moderator. Its purpose is to produce a monochromatic burst of neutrons whose energy is determined by the chopper phase. The intensity and resolution of the neutron pulse incident on the sample are governed by both the chopper rotation frequency, which can be any integer multiple of the source frequency up to a maximum of 600 Hz, and the dimensions of the curved collimation slits within the chopper. The relevant dimensions are the slit width, radius of curvature, and external radius of the chopper body. Although precise optimization of the intensity and resolution requires the ability to vary all these parameters, a set of three or four chopper slit packages should provide adequate performance over the entire range of accessible energy transfers, including the flexibility to relax the energy resolution in favor of intensity. This is a common practice at ISIS, for example.

3.1.5 Adjustable Slits and Soller collimators

We will install a number of sets of adjustable slits to reduce backgrounds for samples that are smaller than the overall beam size. The locations for these slits remain to be determined. The slits will have both horizontal and vertical adjustments. Since all locations will be inaccessible when safety interlocks are in place, the slits will have stepper-motor drives for remote operation.

A system of slits and Soller collimators placed between the Fermi chopper and the sample will allow the incident beam divergence to be tailored to each experiment. There may be situations where the divergence delivered by the guide system is too large, particularly at lower energies, and slits will be adjusted to control where neutrons reach the guiding surfaces. Soller collimators after the Fermi chopper will absorb any neutrons that are too divergent, and eliminate scatter from the Fermi chopper itself.

3.1.6 Supermirror guide

Especially for a long incident flight path, most experiments are restricted by low flux rather than by angular divergence. At energies below about 100 meV, a supermirror guide in the incident flight path serves to increase the flux at the expense of beam divergence. In principle the guide should extend as far back to the source as possible. The ARCS instrument will have three sections of guide totaling 11.25 m of the 13.6 m incident flight path — 2.0 m within the shutter itself, 4.5 m from the shutter to the T_0 chopper, and 4.75 m from the disk chopper to the Fermi chopper. The cost estimate is for a state-of-the-art supermirror (m=3.6) guide.

Care will be taken in designing the guide mounting scheme so that background from the target will be minimized. The multilayers of the guide will be deposited on thin glass substrates held within steel jackets, allowing maximum shielding close to the reflecting layers. Neutron-absorbing fixed apertures of B_4C will be placed as needed to suppress the neutrons scattered where the beam hits the shield jacket. The guide sections will be evacuated for efficient transport of lower-energy neutrons.

3.2 Vacuum Vessel

The vacuum vessel serves three purposes. It provides an evacuated secondary flight path, its shielding provides a low background environment for the detector bank, and its vacuum provides thermal insulation for some sample environments. The inner volume of the vessel will be approximately 300 m^3, a massive structure that will require careful engineering for safe and reliable operation.

At present we anticipate that the detectors will be mounted in vacuum within the vessel. This eliminates the thin aluminum windows that must withstand many cycles of differential pressure between zero and one atmosphere. This is a safety issue because the evacuated structure stores 30 megajoules of mechanical energy. Separation of the vessel structural integrity from the detector and window mounting also means that detector coverage can be almost continuous in angle. This is a prime consideration for single crystal spectroscopy. The SNS detector and data acquisition groups are developing preamplifier and digitization electronics needed to operate linear-position-sensitive ^3He tubes in vacuum, and we assume the ARCS project will use these developments. The plan is to digitize the signals from 8- or 16-detector packs in vacuum, reducing the need for complex vacuum feedthroughs of analog detector signals. Feedthroughs will be by fiber optic technology. Vacuum sensors will shut off the detector high voltage if the pressure in the vessel gets into a range where arcing could occur.

The design of the detector and sample tank will allow for the future use of large cryomagnets and possible polarizing elements. It should be possible to build the vacuum vessel of non-magnetic materials within 1 or 2 meters of the sample to avoid interactions with fringing fields of magnets in the sample region. The most promising technology for producing high-energy polarized neutrons are polarized He filters, which are sensitive to magnetic fields. The spectrometer design will be consistent with future installation of incident beam and analyzing filters so they can be installed once their design and usage becomes mature.

The best practice for inelastic neutron scattering measurements with chopper spectrometers is to eliminate any windows between the sample area and the detector flightpath (as has been done for the MAPS spectrometer at ISIS, for example). To eliminate fatigue problems from frequent cycling of the vacuum in the main part of the vessel, and to save time in restoring a cryogenic vacuum at the sample (the internal neutron schielding, "crispy mix," is somewhat hygroscopic), a method for isolating the sample area vacuum from the rest of the tank is needed. Sample changes will occur while the main vessel remains under vacuum, but the sample space will be opened to the main vacuum after the sample space is evacuated.

3.2.1 Shielding Requirements

To improve dynamic range and sensitivity, the ARCS detector banks will be shielded from neutrons that would otherwise enter from outside the instrument. The vacuum vessel will be surrounded by at least a 30 cm shell of shielding in thin-walled steel containers filled with a mixture of wax and borax ($Na_2B_4O_7$) for moderation and absorption. Experiments will be conducted at IPNS to determine the thickness and composition of the shielding, perhaps using shielding components as specimens in the LRMECS instrument. In anticipation of future proton current upgrades and taking into account previous incidents where shielding requirements were underestimated, we will tend to be generous with shielding material. There will be shielding on all sides of the vacuum vessel, with special attention being paid to shielding around the incident beam path and to complete coverage of the inside of the vessel with boron-loaded epoxy (crispy mix).

3.2.2 Vacuum Pumping System

The pumping system will have the following features.

1. It will be completely automated and equipped with interlocks for safety and to protect the investment in the spectrometer components.
2. It will be able to bring the entire vacuum vessel from ambient pressure to base pressure in 1 hour.
3. It will achieve a base pressure of 10^{-6} mbar in the sample and detector vacuum chamber.
4. It will avoid cryopumping of water and other material onto a sample held at 3.6 K.
5. It should not require service or regeneration during the period of a typical SNS beam cycle.

Rapid pump-down of the entire vessel will be provided by a roots pump backed by a conventional rotary pump. Turbo-molecular pumps will be used to reach a pressure of 10^{-6} mbar in the main vacuum. To avoid cryopumping onto the sample, the sample volume will also be pumped by a cryo-pump (poly-cold). Finally, a dry air venting system will be used to maintain a dry atmosphere in the vessel when it is not under vacuum.

3.3 Detectors

The detector array on ARCS will consist of 975 1.2 m linear position sensitive detectors (PSDs) in the low-angle bank and 518 1 m PSDs in the high-angle bank. The lengths of the detectors will be divided into pixels of 2.5 cm length, for a total of 65,832 individual detector elements. Each pixel in the low-angle bank will subtend an angle of 0.25°, and the high-angle pixels will subtend 0.5°. We are considering the installation of thin detector tubes at the smallest angles, perhaps doubling the angular resolution below 5 degrees. This fine angular resolution translates into excellent Q-resolution for magnetic scattering experiments, but its importance needs further discussion in the context of other effects on Q resolution, such as incident divergence.

For powder samples, software will rebin the detector data into Debye-Scherrer rings. Angles from –30° to +140° in the horizontal plane and ±30 degrees vertically will be accessed with even spacing of the detectors to the extent possible while maintaining the structural integrity of the vacuum vessel. The detector array and associated electronics are the single most expensive component of ARCS, but they are required for a world-class instrument. The wide-angle detector coverage translates directly into spectrometer efficiency, and the combination of low angles for studies of magnetic dispersions and high angles for phonon measurements are as essential for separating these signals as they are for supporting the two components of the scientific program.

3.4 Computer Hardware and Data Acquisition

The data acquisition and analysis systems will be built from commodity components following the standard under development at the SNS. Software for ARCS will benefit from (and provide benefits to) similar systems employed on CNCS. Software for data visualization is being developed by the SNS in a generic form, making it suitable for time of flight data obtained at any facility. By adhering to the emerging, HDF-based NeXus data format, portability of ARCS data to this (or any other) analysis package will be ensured.

The data acquisition hardware will be a modular system for performing the necessary event timing and positioning. An ethernet connection to the host UNIX computer will be sufficient for the data rates anticipated on ARCS (the full detector images are large, but for typical inelastic experiments counting times are long, so a delay of a few seconds is tolerable). The user interface to the data acquisition system will be through a web browser, decoupling the location and operating system of the spectrometer from those of the user. The sample environment will also be controlled via a web browser interface though the use of Labview and IEEE-488 or RS-232 interfaces. Password-based security, specific to each experiment, will ensure safe and secure remote operation. Only systems that can be modified without compromising safe operation will be remotely controllable, although all relevant machine parameters should be viewable. These attributes for the data acquisition system are already among the specifications for the SNS data acquisition system, and the ARCS project will take advantage of these developments by the SNS.

3.5 Software

The effort required for software development is frequently underestimated in the planning of scientific projects, often causing software development to be reduced in scope. There can be no scope reduction for the lower-level software, because neglect means that the instrument will not operate. Likewise, mid-level software for experiment control and data acquisition is rarely descoped in the development of a working instrument. The neglected software is that needed for higher-level functions. The typical justification is that higher-level software serves only to improve the user interface, and does not affect the functionality of the instrument. Unfortunately, the members of the IDT have yet to see a case where this was true for an inelastic neutron scattering instrument. In some cases, inelastic instruments are built without the capability of transforming data into coordinates of energy and momentum transfer. Drawing an analogy to diffractometers for elastic scattering, this is equivalent to having a diffractometer that is unable to provide users with diffraction patterns. The ARCS software must be finished simultaneously with the hardware if the instrument is to be productive scientifically.

We assume the lower-level software development for the ARCS project will be developed by the SNS. The SNS Instrument Systems group has a focused effort to develop common standards for data acquisition, storage, and perhaps real-time visualization. Higher-level software specific to the instrument must be developed as part of the ARCS project. Planning for this software has, in fact, already influenced the hardware design. For example, a single horizontal 5.5 m secondary flight path from –30 to +140° will occupy the space for two instruments in the SNS target building, and is therefore impossible. Instead of the inconvenience and incompatibilities of a vertical instrument, we are proposing a horizontal instrument with two horizontal flight paths of differing lengths. It will therefore be necessary to combine data from two detector banks, each with a different characteristic resolution. This is readily possible with appropriate software. We are aware that previous instrument configurations with two flightpaths were unsuccessful owing to inadequate software.

The higher-level software functions of the ARCS instrument will be developed by postdoctoral fellows in collaboration with a software engineer at the Center for Advanced Computing Research (CACR) at Caltech. The experience of CACR with other scientific software development efforts at Caltech has shown that scientists must write the actual code, since they are the ones who understand its function. Scientists tend to write code that is expedient and often efficient. Unfortunately, code written by most scientists does not translate directly into software objects with properties that permit seamless integration into larger structures. A software engineer must work with scientific programmers and enforce adherence to standards of object properties, data structures, and documentation, for example. Such an engineer will be alert to how code implementations can lead to changes in the software plan, and he or she will help identify ways to alter the code or identify changes needed in the software plan. Such information from a software professional is needed so the IDT can understand the issues for change requests in the software effort.

The P.I. has had enough contact with the Center for Advanced Computing Research to know that appropriate software engineering staff are available to support the ARCS software effort. After further identification of personnel, the software engineering effort will begin with a meeting at Caltech between the ARCS Executive Committee, interested SNS personnel (perhaps including members of other instrument IDT's), and CACR personnel. This kickoff meeting of the software project should set the broad scope of the software engineering effort much as the April 30 meeting identified the issues and set the parameters for the hardware design. This kickoff meeting will be followed by a workshop of the ARCS Executive Committee to begin work on the design of a software roadmap, which will guide the development of a work breakdown structure for the software project. The detailed software design may require a year of effort. If it is done properly, the actual code generation and integration into the instrument effort should be much more straightforward, and results will be more predictable and maintainable in the future.

Not all of the required software functionalities have yet been identified. In what follows we describe two important and largely independent thrusts are needed for the higher level software, denoted "S($\boldsymbol{Q}$,ω)" and "full experiment simulation."

3.5.1 S(Q,ω)

Each neutron detected in a detector pixel at (ϕ,θ) is identified by its arrival time, t. Software must rebin these spectra in instrument coordinates, $S_{\phi,\theta}(t)$, into spectra in the physical coordinates of momentum and energy, S($\boldsymbol{Q}$,ω). This requires keeping track of resolution volume, statistical uncertainty, detector efficiency, and crystal coordinate transforms. This is a challenge because each ARCS measurement will comprise independent time spectra from each of 65,832 detector pixels, and each of these pixel spectra will consist of thousands of microsecond time bins. Front-end rebinning software should be fast and convenient so it can serve as the main window for an experimenter to access the data for rapid visualization. The experimenter needs rapid access to incoming data during an experiment to assess the progress and quality of the measurements. Such real-time information is crucial so that beamtime is not wasted on measurements that show no results, or wasted by performing measurements of excessive quality when the beamtime could be used more effectively for other work.

The ARCS spectrometer system must provide users with some form of the "scattering law," S($\boldsymbol{Q}$,ω), with coordinates of $Å^{-1}$ and meV. Data transformation into these physical coordinates of momentum and energy is a minimum that must be provided to each user when he or she has completed an experiment. With two flight paths it is also necessary for the software to provide some correction for instrumental resolution and artifacts. For single crystal measurements, several IDT members have used the approximate S($\boldsymbol{Q}$,ω) that is provided by the software package "Mslice," in use at ISIS and developed by Radu Coldea under the supervision of Steve Nagler of the present IDT. The data from a single setting of the spectrometer can be projected, sliced, and viewed to identify the important features of the dynamical response of the sample. The Mslice package requires further development to accommodate the ARCS geometry and instrument resolution. Adapting the software to the ARCS instrument will also require the development of experimental calibration procedures, from which S($\boldsymbol{Q}$,ω) can be obtained in absolute units. Other specifications for the "S($\boldsymbol{Q}$,ω) software" may include corrections for instrument background, multiple scattering, and multiple excitations. This software could be further extended to isolate intensities originating with different scattering mechanisms by exploiting their energy or momentum dependencies.

After the software roadmap is completed, Steve Nagler and Ward Beyermann will supervise the effort to develop software that delivers S($\boldsymbol{Q}$,ω) to the experimenter.

3.5.2 Full Experiment Simulation

We propose to develop an entirely new approach to handling data from inelastic neutron scattering experiments — a full simulation of the paths of neutrons from the moderator, into the instrument, through the sample, and into the detector banks. Monte Carlo simulations such as McStas are already essential tools in the design of inelastic scattering instruments, and much of the design optimization of the ARCS instrument is being performed with McStas simulations. What we propose that is new is to include the dynamic behavior of the sample in these simulations, and testing these simulations against measured data. By running multiple cases, feedback into the dynamics model will be provided through the quality of fit of the simulations to the data.

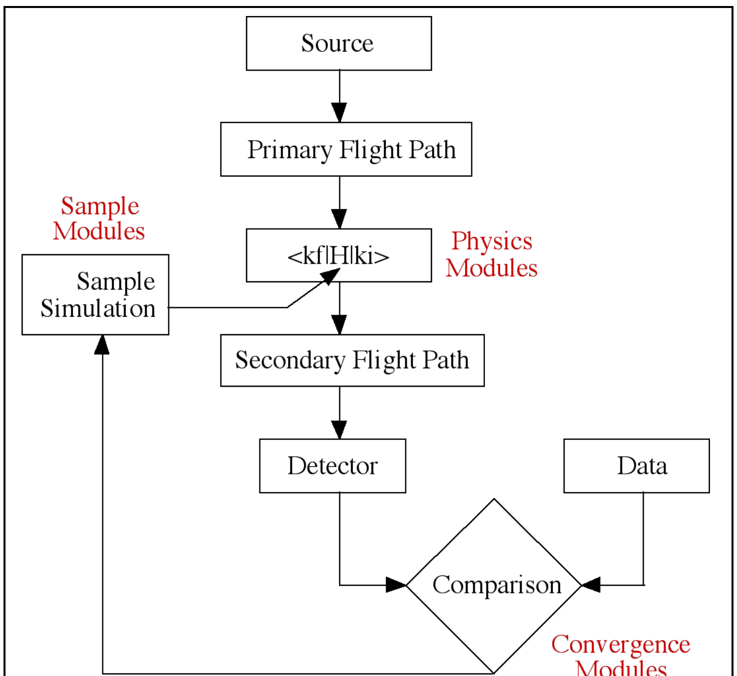

Figure 10: Flow chart for full experiment simulations. The simulations are refined by iterative comparison with the experimental data, but not all steps may need to be repeated for each iteration.

The concept of a full experiment simulation is illustrated in Fig. 10. It shows modules for sample dynamics that generate excitations in solids, and independent scattering physics modules that take these excitations and use them with coherent scattering lengths and sample orientation to predict the detector signals from coherent magnetic scattering, for example. Tests for convergence in the presence of statistical scatter and competing processes can be useful for understanding the reliability of the scientific results. These modules, highlighted in red in Fig. 10, are new concepts that need to be developed as part of the ARCS project.

Some features of full experiment simulations are:

a. A highly accurate understanding of instrument characteristics will be attained over time, allowing more reliable interpretations of measured spectra. The instrument simulation modules will be used in the analyses of all experiments, instrument design, and instrument calibration and maintenance. The instrument resolution, background, spurions, and sensitivity will become increasingly well understood in ways that enable quantitative predictions for new experiments or new modes of operation. Full experiment simulation is a natural way to perform simultaneous analysis of the full data set from two detector banks. It is expected that some, perhaps most, beamtime proposals and experiment plans will include simulational tests of experiment viability and optimization.

b. The elementary dynamic processes of some systems are understood, but cannot be expressed as $S(\boldsymbol{Q},\omega)$ in a simple way. This is true, for example, of excitations in disordered systems. Model Hamiltonians can be written easily, but important physical features such as the localization of excitations, for example, have only approximate solutions. A simulation of the dynamics is often the most exact approach. Full experiment simulations will allow the measured data to be compared directly to the model Hamiltonian, and will optimize parameters of the model.

c. Full experiment simulations offer a type of connection between theory and experiment that is not typical of present research in condensed matter physics and materials science. By placing calculated behavior into the sample module, theoreticians can test models directly against experimental data, or test whether a model could provide measurable data. There should be no limit to the sophistication of the calculations, or the computing platform required to perform them, provided they fit into the sample module so fluxes of incident and outgoing neutrons can be calculated. Full experiment simulations are a vehicle for theoreticians and experimentalists to collaborate in sophisticated new studies of excitations in condensed matter.

We are assuming that the present Monte Carlo software for instrument simulations, McStas (perhaps Vitess) is sufficiently robust and sufficiently well maintained to provide a reliable framework for the simulation of ARCS and other instruments.* It is interesting that the development of Vitess required approximately 6 postdoc-years, including the graphical user interface. The development of scattering physics modules, convergence modules, and sample modules is expected to be a larger effort, which we have scaled accordingly.

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* Since this software plays such a central role for SNS instrument design, we are assuming that the SNS will take some role as custodian of McStas or another package of open source software for instrument simulation.

Several types of Hamiltonians for sample dynamics are used today for calculational support of inelastic neutron scattering experiments, and some are mainstays of data analysis. It is expected that modules will be developed to handle all common scattering processes such as momentum-resolved coherent magnetic scattering from polarized spin dispersions in single crystals, the incoherent multiphonon expansion for anisotropic polycrystals, quasielastic scattering from a fluctuation spectrum of magnetic moments, etc. These modules may be regarded as intrinsic characteristics of the neutron scattering experiments. There are numerous types of sample dynamics modules that could provide inputs to these scattering modules. Some sample dynamics models are practically standard for data analysis, but others need to be discussed thoroughly in the software planning stage so their importance can be assessed.

Once the detailed software design is complete, the full experiment simulations will be developed under the oversight by Fultz, McQueeney and Osborn. Full experiment simulations cannot serve all types of experiments, especially those for which the structure or dynamics are unknown. The "S($\boldsymbol{Q}$,ω) approach" therefore remains an essential software effort.

3.5.3 Software Planning and Testing with Data

Meetings to plan the higher level software will begin immediately. Even though the SNS does not yet have institutional standards for higher level software, work to develop a software plan, followed by a set of specifications, can begin early. The software work breakdown structure or software roadmap will probably prove more difficult than for the hardware, since this is less charted territory. The development of the work breakdown structure for a large software project, if at the level of detail of defining the software objects and their properties, is a large fraction of the work itself. The actual coding may not begin until late 2002.

Some coding will begin early, and will be done expediently without standards for software engineering. For example, simulations of idealized experiments, performed with small extensions to McStas, will be used for optimizing the instrument design. For example, we need to test the value of a dense array of detectors at low angles for studies of single crystal samples with typical mosaic spreads. These simulations need to be performed before settling on the details of the detector array. We do not expect this code to be offered to users for analysis of single crystal data, although the experience of testing McStas with software modules may help us with the eventual design of sample modules, scattering modules, and may show the viability of working with McStas itself for full experiment simulations. Some of these simulations will be performed with commercial software packages. The package MATLAB, for example, has a suite of extensions that can generate portable C/C++ code from working MATLAB applications, and we need to learn how this software-generated code could be integrated into the ARCS software.

Finally, for the purpose of developing software, and for developing a user program at the SNS, it is important for members of the IDT to have experience with the analysis of data of direct relevance to them. This is particularly true for inelastic scattering studies on single crystals using chopper spectrometers, experiments for which the U.S. community has had relatively little experience. Such measurements are usually not practical with our operating inelastic chopper spectrometer, LRMECS at IPNS. When the PHAROS instrument is working at Los Alamos, such measurements should be possible. The expedient solution, however, is to support travel of the IDT members to the ISIS facility, where measurements could be performed on the MAPS or MARI instruments. It is hoped that DOE BES will help with the institutional arrangements, perhaps building on the present postdoc exchange program.

We expect that the higher-level software development for ARCS will be adaptable to the cold neutron chopper spectrometer. Paul Sokol and Brent Fultz have tentative plans for a workshop to discuss software and sample environments for the two instruments. For example, McStas simulations of the CNCS instrument should be able to incorporate the scattering physics and some sample dynamics modules developed under the ARCS program.

3.6 Sample environment

Sample environments will be provided by units that mount on a rotating stage atop the ARCS vacuum vessel. The stage will have a rotating vacuum seal large enough to enclose a bolt circle that we hope will be standard at the SNS. The different systems for sample environment to be mounted with this bolt circle are:

1. Thimble, a thin-walled aluminum containment to separate the spectrometer vacuum from a cylindrical specimen region at ambient pressure. Specialized sample environments provided by users will fit within this thimble.
2. Low temperature heliplex system for experimentation from 3.6 K to 350 K. This unit is a closed-cycle refrigerator with enhanced low temperature capability provided by a Joule-Thompson heat exchanger.
3. Displex/heater unit for a temperature range of <30–650 K.
4. High temperature furnace unit capable of temperatures from 600–2073 K.

These units are described in detail below. An over-riding consideration in choosing these units was their ease-of-use. Especially in the early years of ARCS, all users will be inexperienced with its characteristics. Further risks to experiments from complexities of cryostat control, for example, are important to avoid. Displex units also provide for more convenient computer interfacing, and promise a greater degree of automatic control of data acquisition.

3.6.1 Sample Rotation Stage

Much of the science to be done on ARCS involves single crystal samples. For these experiments it is necessary to orient a particular direction in the reciprocal lattice of the sample, $\boldsymbol{q}$, with respect to the wavevector transfer, $\boldsymbol{Q} = \boldsymbol{q}_f - \boldsymbol{q}_i$, probed by the spectrometer. Figure 11 shows a case where the scattering of the (red) neutron beam involves a change in wavevector of $\boldsymbol{Q}$, which lies on a cone around the center of the sample. In many cases, a vector in the sample, $\boldsymbol{q}$, can be made to intersect this cone by a simple rotation by ω about a vertical axis. This degree of freedom is particularly simple to implement with a rotation stage. On the other hand, to probe several directions of $\boldsymbol{q}$ simultaneously, such as mapping full dispersions, additional degrees of freedom for sample tilt will be required.

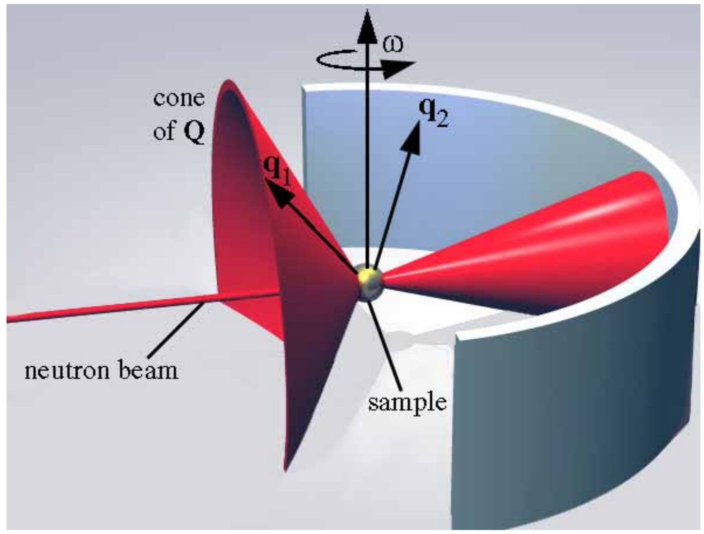

Figure 11: One degree of freedom allows $\boldsymbol{q}_1$ to intersect **Q** by rotation of ω. Another degree of freedom is required for both $\boldsymbol{q}_1$ and $\boldsymbol{q}_2$ to simultaneously intersect **Q**. (Here $\boldsymbol{q}_\mathrm{i}$ points along the incident beam, $\boldsymbol{q}_\mathrm{f}$ lies along the red cone at right.)

The rotation stage for the vertical rotation will include the flange for mounting the sample environment units atop the vacuum vessel. This requires the design of a rotating vacuum feedthrough. The diameter of the SNS standard bolt circle will set the size of the sample environment units. The sample rotation stage will be based on a commercial rotation table. A stepping motor and associated controller connected to the data acquisition software will control the angular setting of the rotation stage to within an accuracy of approximately 0.01 degrees. Mounting of this rotation stage on ARCS, with a flexible bellows coupling to the chamber, will be investigated. Having all sample motions on the spectrometer itself seems most practical for the standardization of data collection and subsequent analysis.

3.6.2 Thimble for Specialized Sample Environments

While most sample environments will use the vacuum of the ARCS vacuum vessel to provide an evacuated scattering volume and thermal insulation, some special conditions require a separate sample vacuum. Examples of such cases are

1. When the ARCS vacuum vessel does not provide adequate thermal insulation, as for example in the case of a dilution refrigerator.
2. When there is a risk that the sample may disintegrate and contaminate the main vacuum vessel, as in the case of the high temperature furnace.
3. When a specialized sample environment is needed that cannot be mounted directly on the rotating vacuum flange.

For these cases and others there will be an ambient pressure thimble that can be mounted on the rotating vacuum flange to provide ambient pressure and temperature in the sample region of the spectrometer. The thimble will be made of aluminum tube stock and have a thin aluminum window at beam height to minimize scattering. The thickness of this window will be in the range of 1–2 mm as set by strength and safety considerations. All parts of the thimble outside of beam height will be coated with neutron-absorbing material such as boron-loaded epoxy on the side facing the vacuum vessel.

3.6.3 Oscillating Radial Collimator

Important parts of the scientific projects to be pursued on ARCS involve sample environments that necessarily place significant amounts of material in the incident beam path. Examples include the high-temperature furnace and various high-pressure cells to be used on the instrument. Owing to their different flight paths, neutrons that scatter elastically from material that is not at the sample position will in general arrive at the detector bank at different times than neutrons which scatter elastically from the sample. The result can be intense spurions that are difficult to distinguish from inelastic scattering from the sample itself.

To suppress these spurions and inelastic background from sample cells, ARCS will be equipped with an oscillating radial collimator. The collimator will be located in the detector vacuum vessel in such a way that it can be used in combination with any sample environment including the ambient pressure thimble. The collimator will cover the full angular range of the detector bank and the blade spacing will be chosen so as to define a cylindrical scattering volume at the sample with a diameter of order 1 cm. With an inner diameter of approximately 2 m and a blade length of 1 m, this requires a collimation of approximately 1° per channel. Several companies have delivered such collimators in the past. We are basing our price estimate on the cost for the radial collimator purchased by John Copley of NIST from Euro Collimator in 1997. One innovation would be the necessity of using a neutron absorbing material such as boron, which remains efficient up to neutron energies of order 500 meV. Given the large blade spacing we believe this problem can be solved without reducing the transmission of the device below 90 %.

The radial collimator will be mounted on a rotation stage to provide the oscillating motion that prevents permanent shading of specific directions. Not only the collimator blades, but also the rotation stage and the frame from which the blades are suspended, will be coated with neutron-absorbing material to avoid spurious scattering. Provisions will be made to automatically lower or raise the collimator out of the beam path when it is not needed.

3.6.4 Low-Temperature Closed-Cycle Refrigerator (3.6–350 K)

Since low temperatures are required for much of the scientific program, it is important to acquire a displex unit. Our preference is to obtain a unit with extended low temperature capability. One approach is taken by APD Cryogenics in their commercial Heliplex design. It uses a closed-cycle refrigerator in combination with a Joule-Thompson heat exchanger to achieve base temperatures below 3.6 K. A temperature controller dedicated to this device will be purchased.

3.6.5 High-Temperature Closed-Cycle Refrigerator (<30–650 K)

Ross W. Erwin of NIST has modified a closed-cycle refrigerator to enable it to reach temperatures as high as 650 K. The trick is to use a sapphire spacer between the cooling head and heating stage. The thermal conductivity of sapphire is temperature-dependent so that it provides better heat conduction at low temperatures than at higher temperatures. The NIST group is agreeable to providing us with drawings and technical advice. We plan to build our own unit, however, which is based on a closed-cycle refrigerator unit from APD or Leybold, a heating stage from Air Products, and a sapphire link between them. Although the closed-cycle refrigerator has a base temperature of 6.5 K, we expect that with the sapphire spacer the base temperature should be between 25 and 30 K. A temperature controller dedicated to this refrigerator will be purchased.

3.6.6 Furnace (600–2073 K)

We have evaluated the performance and reliability of several high temperature furnaces that have been used previously for neutron scattering work. The clear choice is the "ILL" furnace manufactured by AS Scientific Products, Abingdon, England, and shown in Figure 12. This unit uses a cylindrical Nb heating element around the sample, which can be as large as 4.5 cm diameter and 10 cm high. Cylindrical Nb radiation shields surround the heating element, and the water-cooled housing has thin Al windows for 360° neutron access around the sample. Neutron access is ±20° out of the horizontal scattering plane. This unit has relatively low background, but the background should be essentially negligible with incident beam collimation and the use of radial collimators in the ARCS vacuum vessel. The unit is reasonably priced, about 70 k$ with vacuum system, power supply, interlocks, and miscellaneous spare parts.

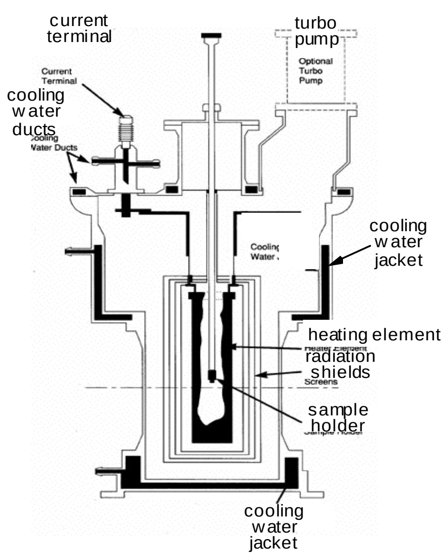

Figure 12: The ILL high temperature furnace. The thin Nb heating element and Nb radiation shields are in the neutron beam, but we have had little trouble with spurious scattering in reactor experiments with collimated beams at ORNL.

An important advantage of this ILL furnace is that it is a mature product, with an integrated vacuum system, temperature controller, power supply, and safety interlocks. It will likely be used with other instruments at the SNS.

3.6.7 Issues with Pressure Cells

Pressure experiments with a samples having masses of tens of grams and larger are often performed by mounting the sample in a canister that can be compressed in a hydraulic press, and then locked in a compressed position. Upon removal from the hydraulic press, the pressure is maintained in this compact "pressure cell", which can be moved into the specimen region of the ARCS spectrometer, for example. An obvious feature of the pressure cell is that it is made of high-strength materials such as sapphire or steel, and typically contains more such material when higher pressures are required. For pressures of 10 kbar or higher, spurious scattering from the pressure cell can overwhelm the scattering from the sample itself, especially inelastic scattering from the sample. This problem can be alleviated considerably by collimation. The incident beam is collimated to have a width comparable to the sample, and the scattered beam is collimated with the oscillating radial collimators.

3.7 Projected System Performance

3.7.1 Complementarity of ARCS, CNCS, and the Backscattering Instrument

The ARCS spectrometer will complete a initial triad of inelastic scattering spectrometers at the SNS. In general, the low-energy range will be handled by the long flightpath back-scatter spectrometer, the medium-energy range will be covered by the cold neutron disk chopper spectrometer CNCS, and ARCS will perform measurements at higher energy and momentum transfers. Figure 13 shows how the three instruments cover the space of energy and momentum transfer. For ARCS, the kinematics for inelastic scattering has been used to determine the maximum and minimum *Q,* given the range of scattering angles from 3º to 140º and incident energies from 10 meV to 1000 meV. A minimum energy transfer equal to 0.02 E_i and a maximum of 0.7 E_i were also assumed. Figure 13 shows that ARCS will be the instrument providing access to large Q and ω. There is an overlap of capabilities of CNCS and ARCS at energies below 10 or 20 meV. This is highly desirable, especially because it includes the important energy range of many thermal excitations. Some overlap of instrument capabilities is also important when experiments are planned over a broad energy range by using the two instruments. It should be possible for some experiments to be performed on either ARCS or CNCS, facilitating scheduling of the user program.

This section presents estimates of the performance of the proposed ARCS instrument. Analytical methods for calculating the resolution in ω and Q and the flux on the specimen were used because they allow many possible instrument configurations to be evaluated. Although there are a number of simplifying assumptions, the performance numbers given here should provide a reasonable estimate of the capabilities of ARCS. To the maximum extent possible, ARCS will be optimized for high flux, high resolution in ω, and high resolution in Q. Comparisons with other instruments have not yet been made, but it should be noted that the SNS source will be roughly a factor of 12 times as intense as the ISIS source.

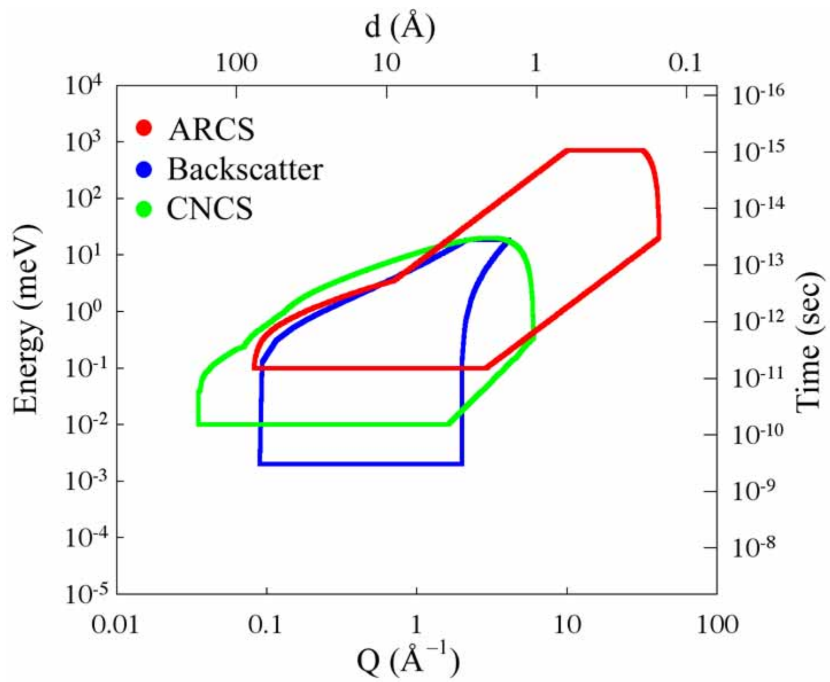

Figure 13: Energy and momentum resolution coverage of the three instruments in the initial SNS inelastic instrument suite.

With its large detector coverage, neutron guides (whose benefits are not modeled here), sample motions for single crystals, and advanced software, there is no doubt that ARCS will be a world-class Fermi chopper spectrometer.

3.7.2 Intensity and Energy Resolution

For a chopper spectrometer, the flux at the sample for a particular energy resolution is of course sensitive to the lengths of the primary flightpath (moderator to sample) and secondary flightpath (sample to detector). The solid angle coverage of the low-angle bank is about π/2 sr, and another π/2 sr of coverage is provided by the high-angle bank. These two detector banks provide different resolutions in energy and momentum, however. Energy resolution is best for the low-angle 5.5 m bank with its longer flightpaths. It is expected that experiments on magnetic dispersions will optimize the chopper operation to obtain the best balance of resolution and flux in this low-angle bank. The high angle banks will provide phonon data, or phonon corrections for interpreting magnetic scattering data.

Figures 14 and 15 show the performance of the two detector banks of the ARCS instrument at 250 meV and 63 meV. Two models for the performance of chopper spectrometers have been used. One model, due to Jack Carpenter,[102] assumes the contributions to energy broadening from moderator pulse width, Fermi chopper open time, and sweep time for the Fermi chopper with respect to the moderator are independent and add in quadrature. A second model, derived by Toby Perring,[103] accounts for the correlations between these contributions caused by the rotation of the Fermi chopper. In our implementation of both models the sample is assumed to be small, and the moderator flux and pulse width as a function of energy were obtained from detailed Monte Carlo calculations of the SNS ambient water moderator with mid-line poisoning (2.5 cm depth).

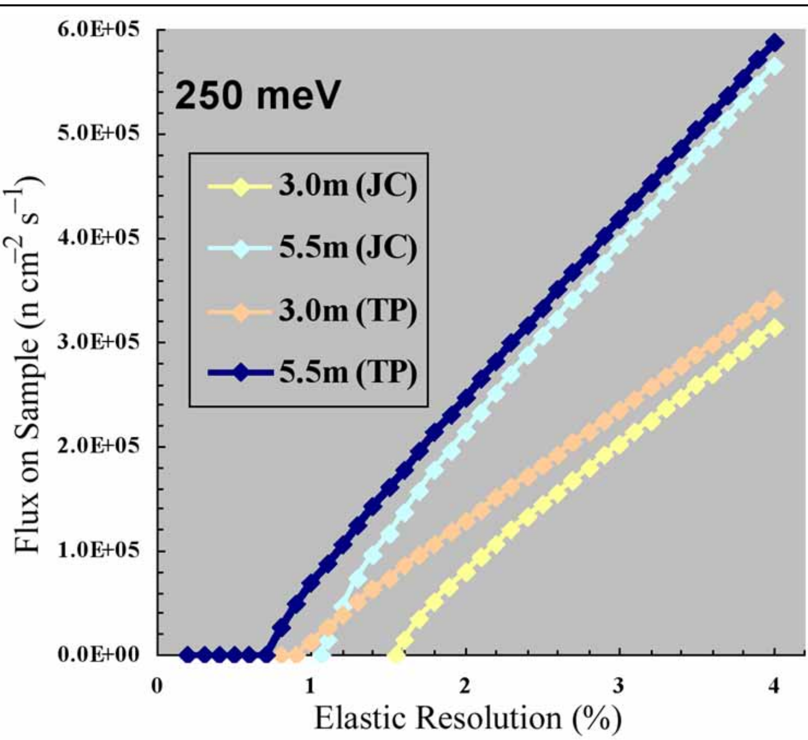

Figure 14: Flux on sample versus energy resolution for ARCS detector banks at 250 meV, using models of Jack Carpenter (JC) and Toby Perring (TP).

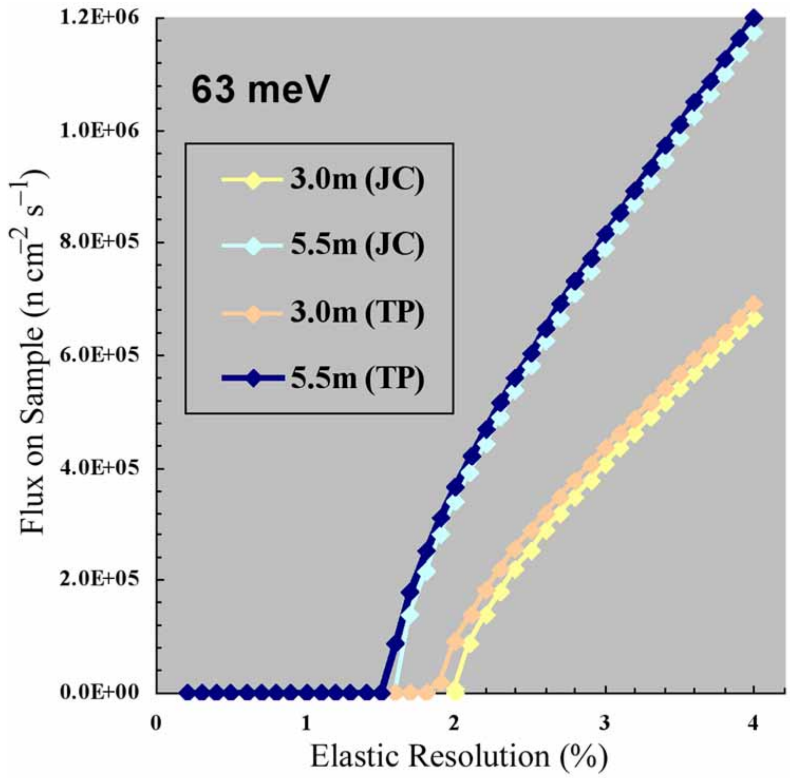

Figure 15: Flux on sample versus energy resolution for ARCS bank at 63 meV, using models of Jack Carpenter (JC) and Toby Perring (TP).

It is evident that both models give similar results, with the major discrepancy at higher incident energies and smaller energy resolutions. We presume this is due to a beneficial correlation from the tilt of the moderator face with respect to the beamline that is included in the Perring model. In any case, the longer flightpath significantly improves the available flux for a given energy resolution. Unfortunately, the target building constrains this 5.5 m flightpath to low angles.

Figure 16 provides a figure-of-merit for several instrument configurations analyzed by Doug Abernathy.* This figure-of-merit was developed by taking results such as those in Fig. 14 and 15 for the various possible instrument configurations with differing flight paths and detector coverage, and weighting them by the solid angles of the detector banks. This figure-of-merit of Fig. 16 gives a total sensitivity for a fixed value of energy resolution. The proposed ARCS instrument, model D, offers the highest performance of all those considered. Although some of the instrument designs perform as well as the proposed instrument for particular angles and energies, the proposed instrument offers essentially the best performance of energy resolution and intensity at any scattering angle.

A summary of the intensity available with ARCS for a fixed resolution of 3% is given in Fig. 17, using the Perring model. There are two curves corresponding to the two secondary flightpaths. Of course, for any single measurement only one Fermi chopper slit package will run, so the low-angle bank will always provide better energy resolution than

* The proposed ARCS spectrometer is essentially “Model D” described in the document “Possibilities for High-Energy Chopper Spectrometers at the SNS” by Doug Abernathy, April 23, 2001. The document also discusses design issues for the flightpaths 17 and 18 of the SNS target building.

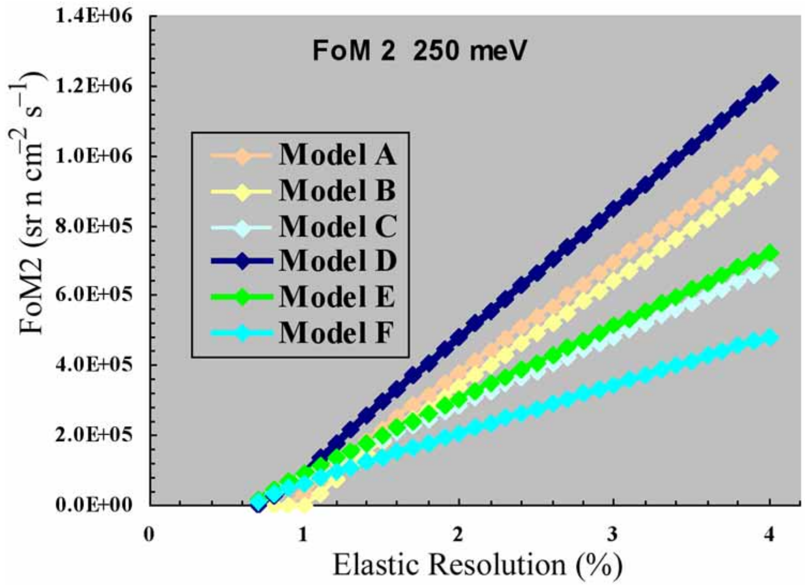

Figure 16: Figure-of-merit of flux on sample for each bank times the solid angle of the bank for equal energy resolutions. Comparison is for six instrument configurations. The proposed ARCS instrument is Model D.

the high-angle bank. The software effort in this proposal will allow the experimenter to model this behavior and optimize the arrangement for his or her experiment.

3.7.3 *Q* Resolution

Specifications for *Q* resolution emerged as a topic of lively discussion since the Caltech meeting of the ARCS IDT on April 30, 2001.

Optimization of an inelastic instrument for momentum resolution is quite different from optimization for energy resolution. Momentum resolution has been less of a focus for the design of chopper spectrometers, however, because historical trends have emphasized measurements on polycrystalline samples. The goal of measuring dispersive excitations in single crystals requires a detailed consideration of *Q* resolution, however. Four important sources of broadening in ***Q*** are:

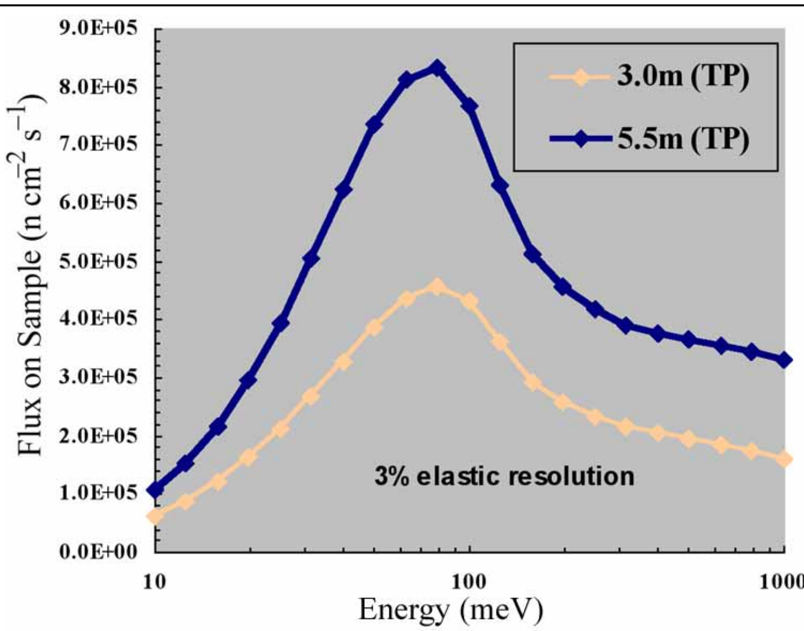

Figure 17: Flux on sample for each bank with 3% elastic resolution as a function of incident energy. Gains from the neutron guide are not included.

1) The finite size of the moderator, even over a long primary flight path, provides a significant angular divergence of neutrons on the specimen. This divergence is about 0.4° without a guide, and will be larger for thermal neutrons that have been reflected along the guide. Unless the excitations in the specimen select among these incident $\boldsymbol{q}_i$, an unlikely situation for most studies using inelastic scattering, the divergence of the incident beam may prove the largest source of broadening of $\boldsymbol{Q}$. Although incident divergence can be improved with apertures and Soller collimators, the loss of intensity may prove unpopular with users. In many experiments we expect the Q-resolution to be dominated by incident divergence.

2) The sample itself can cause a significant angular broadening of the scattered intensities. A large sample having a size comparable to the detector pixel size is one such source of broadening. A 0.2° mosaic spread of a good crystal will cause an angular spread of 3.5 cm at 10 m. We should assume that high quality crystals will be available, however.

3) The energy resolution leads to a coupled smearing in $\boldsymbol{Q}$. Since the energy resolution is quite good for all flight paths, this source of Q-broadening is not the biggest problem for single crystal work, especially in the forward direction.

4) The sinθ-dependence of Q conspires with finite detector pixel size to give an optimal secondary flight path of infinite length in the forward direction. A ratio of $\Delta Q/Q$ of 1.0 % is not possible for scattering angles, 2θ, smaller than 14° even if a 10 m secondary flight path is used with 2.54 cm detectors. Optimization at small angles leads to long instruments such as used in small angle neutron elastic scattering measurements.

The overall Q-resolution will be a convolution of these effects 1 through 4, and the individual broadenings add in quadrature if Gaussian broadenings are assumed. While there is improvement in eliminating an individual source of broadening, there is diminishing return by working to make one effect much smaller than the others. In particular, the incident divergence of 0.4° is matched by a secondary flight path of 3 m if 2.54 cm detector pixels are used. Since diagonal paths along the pixel array are somewhat longer, the 5.5 m flight path is probably about as long as can be justified for Q-resolution. It is interesting that although the relatively short secondary flight paths of ARCS are the limiting parameters for ω resolution, these lengths are acceptable for Q-resolution, given the divergence of the incident beam.

Figure 18 shows the calculated momentum resolution, ΔQ, as a function of Q for various pixels of the ARCS detector array as the incident energy varies from 10 to 1000 meV, with the energy resolution fixed at 3%. The calculations were performed with the Carpenter model, which sums in quadrature the momentum broadenings from the moderator and chopper pulse widths and the angular widths of the pixels and moderator. Figure 18 presents the sum in quadrature of contributions parallel and perpendicular to the direction of incident beam. This approximation should be an upper limit to the resolution because it ignores various correlations among the different contributions. As expected, the

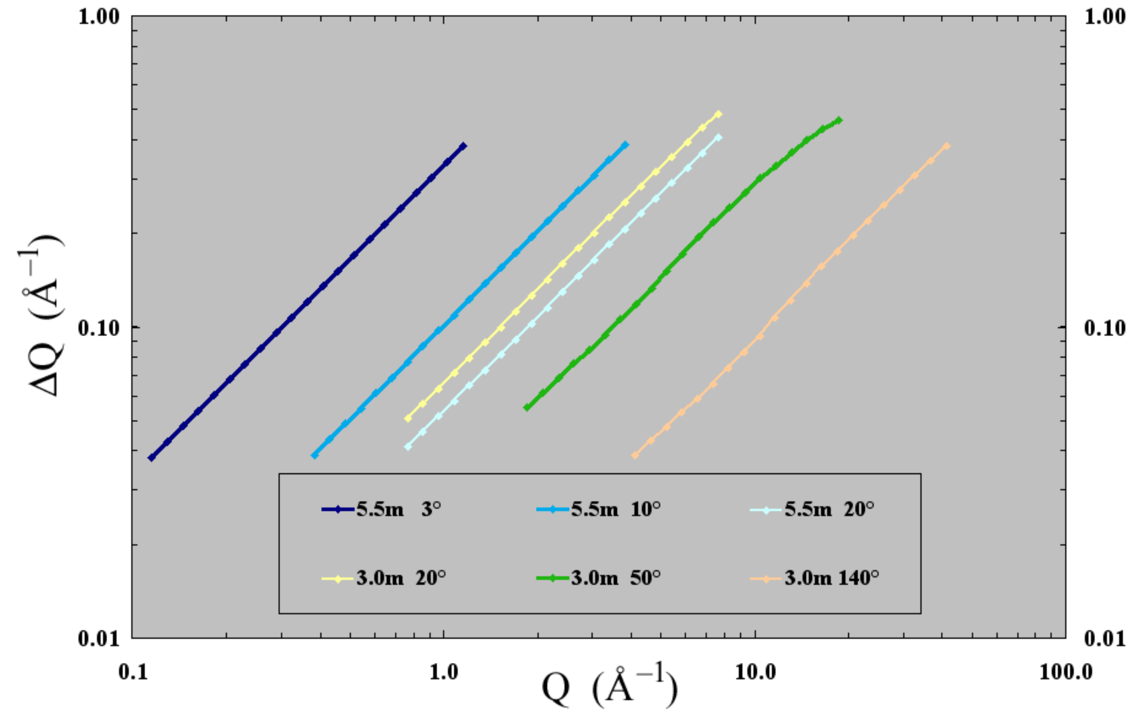

Figure 18: Q-resolution for elastic scattering for various pixels as the incident energy varies from 10 meV to 1000 meV, and $\Delta E/E$ of 3 %.

fractional resolution of the momentum transfer, $\Delta Q/Q$, is best at the largest angles of scattering. The calculated $\Delta Q/Q$ varies from 30 % at 3° to 1 % at 140°. At each scattering angle, $\Delta Q/Q$ is essentially the same for all incident energies. As a fraction of the magnitude of the incident wavevector, each pixel has an approximately constant resolution that varies from about 1.7% to 2.4%. The transition from a 5.5 m flightpath to a 3.0 m flightpath (here at 20°) increases the $\Delta Q/Q$ by about 25%.

Although this analysis of momentum resolution was for elastic scattering, its predictions are conservative for neutron energy loss processes. When $E_f < E_i$, angular errors in $\boldsymbol{q}_f$ have a smaller effect on Q (q_f is smaller in $\boldsymbol{Q} = \boldsymbol{q}_f - \boldsymbol{q}_i$). Nevertheless, a detailed analysis of momentum and energy resolution must take into account the shape of the instrument resolution function in 4-dimensional $(\boldsymbol{Q},\omega)$ space and how this resolution function cuts the dispersion surface under study. Investigations by analytical and Monte Carlo techniques are underway to quantify these effects and assess their impact on the optimization of ARCS for typical experiments in the scientific program.

4. Summary and Overview

Building on several years of interactions on other instrument proposals, an Instrument Development Team, IDT, has been organized to propose the construction of A high Resolution direct-geometry Chopper Spectrometer, ARCS, at the Spallation Neutron Source in Oak Ridge, Tennessee. A Principal Investigator and Project Manager have been chosen, an energetic and engaged Executive Committee has been formed, and the IDT membership includes persons with enough enthusiasm to travel to workshops on instrument development.

The ARCS IDT has coalesced around the science program described in Sect. 1. It covers most research in physics and materials science that measures momentum and energy transfers between condensed matter and neutrons with energies greater than 10 meV. The research topics fall into approximate categories of magnetic scattering and phonon scattering. Both phenomena often occur simultaneously, however, and they can be coupled in correlated-electron systems, for example.

About ten concepts for inelastic neutron spectrometers were evaluated with analytic models. The meeting of the ARCS IDT in Pasadena on April 30, 2001 settled on an instrument with superb intensity, a large solid angle of detector coverage, good resolution in E, and good resolution in Q. Given the long primary flight paths for instruments at the SNS, the E resolution improves with longer secondary flight paths. This length is constrained by space and money, and the space constraint is severe for high angles of scattering. The proposed instrument therefore has two secondary flight paths of differing lengths.

The proposed "two-flight-path-instrument" concept has been the subject of extensive and spirited discussion within the IDT. Our present thinking is that shortening the 5.5 m flight path at low angles to 3 m is an unnecessary compromise, although it would reduce the cost of the instrument by 3 M$. Since the secondary flight path cannot be 5.5 m at high angles, the only option for the high-angle bank is to eliminate it. Elimination gives a cost savings of 1 M$, but reduces considerably the scope of the science program. Although a 5.5 m vertical design was discussed, this concept has problems with detector coverage out of the scattering plane, and has special problems with its sample environment. All these issues are still under discussion, but the IDT has largely settled on the instrument shown in Fig. 9. Changes in concept are still possible, however.

Most of the instrument technologies pose little risk. Chopper and guide technologies are proven, as are the position-sensitive ^{3}He detectors and their analog components. The IDT is relying on the SNS experimental systems group to develop PSD banks that will operate within the instrument vacuum and use digital signal extraction by fiber optic cables. Perhaps the greatest risks are the costs of the bulk shielding around the primary flight path, and the installation costs at Oak Ridge. These costs cannot be assessed accurately without more detailed design work, but ARCS will be a beneficiary of detailed design work for other instruments at the SNS. Contingency funds have not been included in the budget. Given the present uncertainties, a realistic budget with contingency funds could be as high as 20 M$. A more detailed budget is possible only after the engineering design is complete and a resource-loaded work breakdown structure has been approved by the ARCS Executive Committee. Although it is possible that the scope of the project may be reduced, or it may be decided to complete the instrument in stages, at present we believe we can build a superb and fully-operating instrument for the budget requested.

Higher-level software for data analysis is a central part of the ARCS project. This software effort is necessary for the instrument to be useful scientifically, and is indispensable for a spectrometer with two secondary flight paths. Software for data analysis and full experiment simulations will be the focus of the university-based effort by the Executive Committee. This software effort offers a new connection to the theoretical community, and possible leverage of ARCS funds from national programs in computational and simulational science. The IDT recognizes that software projects are notorious for going out of control. The scope of the ARCS software effort will be developed carefully in the first year of the project, and must be guided by a professional software engineer.

To date, nearly all inelastic neutron scattering experiments have suffered from low flux. With its high detection efficiency and high flux from an ambient water moderator at the SNS, the ARCS spectrometer will break free from this flux constraint. The ARCS instrument will enable the acquisition of high-quality inelastic spectra over a range of physical parameters such as temperature, pressure and composition, and will permit rapid measurements of multiple dispersion relations on single crystals. With its flux, detector coverage, resolution, and software, the ARCS instrument will overcome numerous historical problems with inelastic neutron scattering. ARCS promises a new level of sophistication for experiments on condensed matter and materials.

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