Neutron Spin-Echo Instruments

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Neutron Spin Echo (NSE) spectroscopy decouples energy resolution from beam characteristics like monochromatisation or collimation and combines the highest energy resolution from all inelastic neutron scattering techniques with the high intensity advantage of a beam which is only 10-20% monochromatic. As a consequence of that, when it comes to pulsed neutron sources, the most relevant parameter is the integrated neutron flux and pulse width characteristics have only minor importance on the NSE performance. The recent break-through experiment at IN15 demonstrated the feasibility of NSE at pulsed sources. NSE is a Fourier method and yields the intermediate scattering function S(Q,t) directly. High resolution means high values of t. This may be achieved by using long wavelength (as for any TOF instrument the resolution depends on λ^3) and by employing large field integrals. Since at a reactor it uses 10-20% broad bands of neutrons from a velocity selector, however, the possible intensity gain due to the pulse structure is limited in an analogous way as that for a SANS machine. Therefore it needs the combination of cold neutrons with a broad wavelength band, which implies that the optimal choice would be a low repetition rate, full power target with a coupled cold moderator. This calls for the 16Hz long pulse option, which combines the highest integrated neutron flux with a low repetition rate. When it comes to choosing between the two short pulse options, it becomes clear that NSE can only benefit from the higher integrated neutron flux of the 50Hz source and the second choice would be the one with a cold coupled moderator at 50Hz. A dilute 10Hz target would then be the worst choice.

Introduction

Neutron spin precession in magnetic fields measures velocity changes individually.

Neutron Spin Echo (NSE) reaches the highest energy resolution from all neutron spectroscopic techniques by decoupling the value of the minimal neutron velocity changes that may be detected from the width of the velocity distribution of the incoming neutrons [1]. This is achieved by using the neutron spin as a kind of stop-watch carried by each neutron individually. Inelastic scattering leads to a difference in spin precession of neutrons flying through a precession path before the sample and a symmetric path after the sample, and this affects the final beam polarisation, which is converted to an intensity signal by an analyser in front of the detector. In the original generic IN11 spectrometer type the precession paths are magnetic field regions generated by a pair of solenoids (Fig.1).

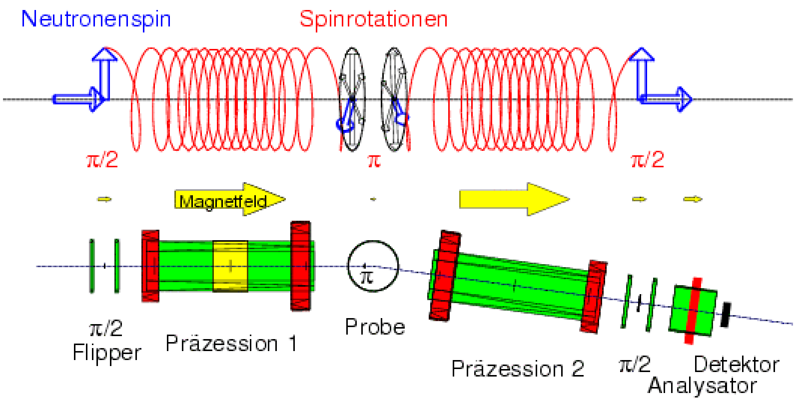


Fig. 1: NSE principle.

NSE is a Fourier method

NSE spectroscopy yields directly the Fourier-transform S(Q,t) of the scattering function rather than S(Q,ω).
For quasi-elastic scattering (ω=0) the detected intensity is given by the relation:

$$I(Q) = \eta\ (\ S(Q) + \delta \int \cos(\omega\,\gamma\,\lambda^3 J) S(Q,\omega)d\omega) \qquad (1)$$

Fourier Time = t µ l³ x B x pathlength

where η and δ are instrument constants for sensitivity and polarisation efficiency respectively and $J = \int |B|\,dl$ is the magnetic field integral in each precession region. The Fourier time is given by: t = $\gamma\lambda^3 J$ with γ = 1.86 x 10^{20} s/(Tm^4), i.e. t = 186 ns for J=1 Tm and λ =1 nm. As a Fourier method –with respect to the energy spectrum-, NSE suffers from the known properties of all Fourier methods and small spectral contributions are buried under the noise caused by the total scattering intensity S(Q). However, the method is ideally suited for the investigation of quasi-elastic scattering and relaxation type motions, which dominate S(Q) and are best analysed in the time domain, where a detailed line shape analysis is of crucial importance and resolution "deconvolution" consists of a simple division.

Resolution does not depend on pulse width!

The relevant parameter for the NSE resolution is the maximum value of the Fourier time, t = t_{max}, at which a useful measurement can be performed. As pointed out above, this "resolution" parameter depends only on λ^3 and on the magnetic field integral. The Fourier time does NOT depend on the bandwidth of the incoming neutrons and for this reason all existing NSE spectrometers use a 10-20% FWHM monochromatic beam for an improved data acquisition rate. All these spectrometers operate at steady sources. From the above it becomes clear that when it comes to the operation at a spallation source, the Fourier time will NOT depend on the pulse width, which determines

the width of the wavelength band “seen” at some instant by a detector element.
A reasonably accurate determination of the polarisation change due to inelastic scattering requires a large number of counts (>several 10^4) and for this reason the method calls for the highest incoming flux and the most efficient solid angle detection.

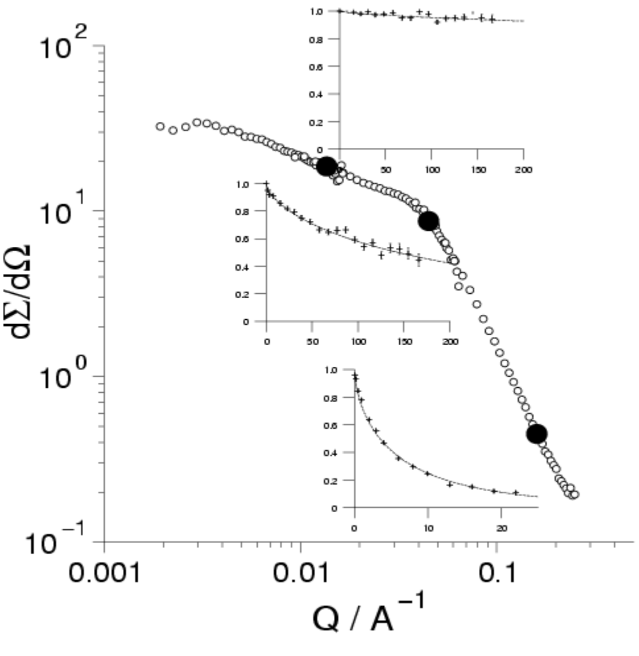
Intensity is crucial!

(Q, w /t) – Range

As far the dynamics in the small angle scattering (SANS) region is concerned (soft-condensed matter systems, ferromagnetic critical dynamics etc) NSE offers a ω-resolution, which is reasonably well adapted to the dynamics investigated. Other problems like glass dynamics and magnetic fluctuations are also in the scope of NSE spectroscopy. As pointed out above, the NSE resolution is rather to be expressed in terms of the maximum achievable Fourier-time, t_{max}. In order to allow for unified comparisons this Fourier time can be converted to an energy transfer: $\Delta\omega = 1/t_{max}$. In the low Q (SANS) regime the current state-of-the-art is still limited by the longest Fourier-time rather than by the proper Q-resolution of the instruments, since typically the relaxation rates behave proportional to $Q^{2..4}$.

Resolution is t_{max}

For comparison:
$Dw = 1/t_{max}$



Longer Fourier times t_{max} needed for low Q !

Fig. 2: NSE relaxation curves corresponding to different locations on the SANS intensity of a bicontinuous microemulsion. Note the considerable slowing down at low Q reaching the state-of-the art limit (IN15) already at Q=0.15 nm^{-1} as displayed by the inserts showing S(Q,t)/S(Q) versus t in ns obtained at IN15,IN15,FZJ-NSE.

An unambiguous determination of small decays in relaxation curves at t_{max}, like the one seen in the upper part of Fig.2, requires a low statistical error (i.e. high intensity) and a high stability of the spectrometer, particularly when it comes to decay rates $\Gamma(Q)$ which are much smaller than $1/t_{max}$ ($\Gamma(Q)<1/t_{max}$). The λ^3-dependence of Fourier times t together with Q~1/λ helps to cope with this general physical trend.

....and intensity !

In the NSE-SANS regime the Q-resolution is usually of minor importance. The situation is somewhat different in the large Q-regime, when it comes to investigating the dynamics at or around structure peaks, e.g. in a glass-forming polymer. Here Q-resolutions better than the 10-20%, imposed by the typical wavelength distribution of reactor based instruments, are often needed. In exceptional cases as oriented lamellar phases (microemulsions, block-copolymers) or macrocrystals of colloids a Q-resolution better than 10% would also be beneficial in the SANS regime.

As it will pointed out below, a welcome feature of the pulsed structure of spallation sources is the improved monochromatisation of the beam detected at any instant by the detectors, which would lead to an improved Q-resolution with respect to the one of reactor based NSE instruments.

The design goals for novel NSE spectrometers at the ESS would be to overpass the existing instruments both in resolution, Q-range and luminosity. The maximum Fourier time in the SANS region should reach the 10^{-6} s domain, which up to now was only covered by dynamic light scattering techniques.

ESS+development→
t_{max} = 1000 ns
1:1000000
0.07 nm^{-1} < Q < 40 nm^{-1}

Best-in-class Instruments

Currently 6 high resolution instruments of the generic IN11 type exist in the world (IN11$_{\text{ILL}}$ [1], IN15$_{\text{ILL}}$ [2], FZJ-NSE$_{\text{FZ-Jülich}}$ [3] = NIST-NSE$_{\text{NIST,USA}}$, C2-2-NSE$_{\text{Tokai,Japan}}$, MESS$_{\text{LLB}}$). A large detection solid angle of 30° has been realised in the IN11 add-on IN11-C [4] with a severe drawback in resolution. A novel instrument class, realised by the SPAN spectrometer at HMI-Berlin, allows for simultaneous measurements in a very wide solid angle reaching 0.27 strad with a resolution, which should be made comparable to that of high resolution instruments like IN11 [5]. The zero-field NSE technique has been realised at the LLB in Saclay (G1bis) [6] and an improved version (RESEDA) of this instrument will be operated at the new Munich reactor FRM-II.

IN11
IN15
FZJ-NSE
NIST-NSE
C2-2-Tokai
MESS
IN11C
SPAN
G1bis
RESEDA

“Normal” and resonance NSE add-ons to triple axis (TAS) spectrometers have been installed or are being developed [7, 8]. These spectrometers use the TAS method to filter out all contributions to S(Q) expect the inelastic one that should be investigated and allow for measuring intrinsic linewidths of elementary excitations like phonons with an energy resolution in the range of 1 µeV. The NSE technique is also used/proposed to detect other than velocity changes coded into the precession angle like direction changes for Ultralow-(S)ANS [9]. Furthermore, a modified NSE method can be used in high resolution diffraction [9]. These more general applications of Larmor precession to neutron instrumentation should better be discussed in the context of the hosting instruments, i.e. TAS, SANS and single crystal diffractometers than in the frame of NSE spectrometers.

Triple-axis add-ons
Diffraction add-ons
→hosting instruments

IN11, the first NSE spectrometer ever built [1] has a high neutron flux on the sample. The incoming wavelength of IN11 can be varied from 0.4 up to 1.2 nm leading to large dynamic (1:20000) and Q-ranges (from 0.1 up to 27 nm^{-1}). The total signal of three single detectors is used for the detection of neutrons.

IN11: 1:20000 dyn. Range
Q <= 27nm^{-}

A substantial improvement in NSE spectroscopy was marked by the IN15 spectrometer [2]. The long incident wavelengths ($0.8 = \lambda = 2.4$ nm) of this spectrometer allow for reaching long Fourier times up to 350 ns. The dynamic range of IN15 is ~1:13500 and the Q-range spans from 0.01 up to 1.5 nm^{-1}. Detection is done by an area detector, which covers a solid angle of ~ 5 10^{-3} strad. IN15 was also conceived with a TOF option and is the test rig for NSE at spallation sources.

IN15: t_{max} = 350ns

The FZJ-NSE [3] is the latest development of the IN11-type instruments. It uses novel magnetic field compensation techniques

FZJ-NSE:
compensation,
no “tuning”

and computed setup for increased stability. The spectrometer can be easily operated at large scattering angles and it is equipped with a multidetector similar to that of IN15. The maximum magnetic field integral is about twice that of IN15. The maximum Fourier time, however, is limited by the corrections needed to cover the total solid angle of the detector and by the restrictions of beam delivery from a multilayer bandpass mirror ($\lambda \sim 0.8$ nm).

Needs:
- ***Long wavelengths***
- ***Broad band / frame***
- ***Field integrals > 1Tm***
- ***Large solid angles***

Combination of new techniques for magnetic field corrections with the use of a broad wavelength band including long wavelengths, improvement of the corrections for large solid angle operation and use of superconducting main solenoids for very large magnetic field integrals and long Fourier times will lead to the next generation instrument of IN11 type. Such a spectrometer will combine the virtues of the above state-of-the art instruments and further extend them.

→ generic IN11 type

Another development option, besides extending the maximum Fourier time, consists into increasing the detector solid angle over which NSE measurements can be performed simultaneously and thus increase the NSE counting rate. The NSE homogeneity requirements for IN11 generic instruments, where magnetic fields are created by long solenoids give however an upper limit for the detector solid angle. The novel magnetic field configuration of the spectrometer SPAN at BENSC allows for significantly increasing the detector solid angle. The novelty consists in the precession field, which has a real cylindrical symmetry and all scattering angles, typically from -150^{o} to 150^{o}, are accessible at the same time. The coils and power supplies of SPAN lead to a reduced energy resolution, amounting ~1/3 that of IN11. The high symmetry and homogeneity of the magnetic field configuration however allows for much higher resolution and the real limit of the set-up should be beyond 2-3 times the actual one [5]. SPAN is characterised by a very broad wavelength band from 0.25 up to 1.0 nm leading to large dynamic (1:30000) and Q-ranges (from 0.2 up to 48 nm^{-1}). Neutrons are detected by three groups of detectors and at the present stage the overall opening of detectors equipped with analysers reaches 30^{0} and a solid angle of 0.02 strad.

→ SPAN type

→ NRSE

The resonance NSE technique (NRSE) has virtually equal requirements for the delivered beam (i.e. target and moderator characteristics) as NSE. Therefore it should not be considered separately. However, the highest Fourier times –at viable intensity– and the highest detector solid angles are reached by NSE spectrometers. We will therefore compare state-of-the-art and best-in-class properties of current and future instruments only in terms of NSE spectrometers.

→ NSE at pulsed sources

The first successful tests at IN15 [10] showed that it is technically possible to combine NSE and TOF and that this combination brings considerable advantages: a high flexibility in choosing the Q resolution and a wider simultaneously covered NSE dynamic range. After this significant breakthrough, NSE can be implemented at pulsed neutron sources at its present state of the art. Intensity gains or losses with respect to existing spectrometers are then determined by the luminosity of the source and the wavelength band covered by a measurement. Furthermore, in the frame of ESS, novel instrumentation concepts should be developed, that would open possibilities beyond the current limits of NSE spectroscopy. A development of the SPAN-generic design to combine energy resolution comparable to that of IN11 with a maximum sold angle is part of the long time scale ILL Millenium programm. Similar development should also take place in the frame of ESS. Extending the Fourier times and resolution beyond the actual limits, set by IN15, should open new possibilties in the (Q,t)-window. This development is of crucial importance for soft-condensed matter and biological systems and the first NSE instrument at ESS should be a highest-as-possible resolution machine.

Target Station and Moderator

For the choice of target station (i.e. repetition frequency, pulse width and average power) and moderator (i.e. spectral distribution and pulse width) the following criteria apply for NSE spectrometers:

- High intensity, is important for all measurements but it is crucial at long wavelengths
- The width of the pulse should lead to less than 10% relative wavelength uncertainty at the detector for an improved Q-resolution, with respect to reactor based instruments
- The bandwidth of useable neutrons within one frame should be as large as possible.

The first criterion calls for a cold coupled moderator. Fig.3 (taken from the ESS-moderator picture gallery) shows that the average flux from a cold moderator at a 5MW target is virtually equivalent to that of the ILL cold source, independently of repetition frequency and pulse length.

→ coupled cold moderator

→ long pulse (2ms) OK!

→ repetition rate < 20Hz

→ 5MW average power !

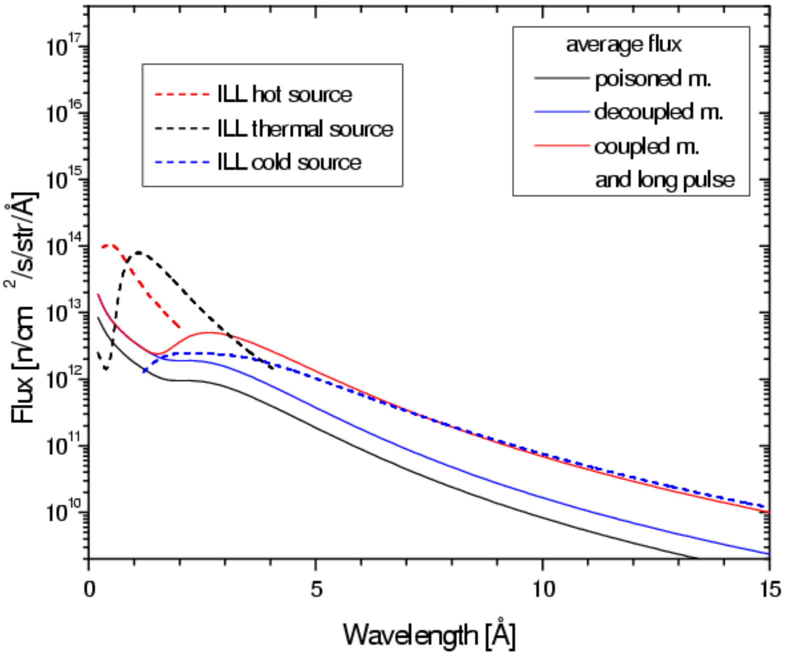


Fig. 3: spectral flux expected for different ESS-moderators in comparison with that of the ILL.

Average cold flux: <ESS> = <ILL>

Any further gains (beyond ILL) should come from the effective simultaneous use of a larger wavelength band than that of a steady source instrument. The absolute width $\Delta\lambda=\lambda_{max}-\lambda_{min}$ of the wavelength band arriving at a detector positioned at a distance L from the moderator depends only on L and on the repetition frequency of the source ν:

Bandwidth/frame → gain

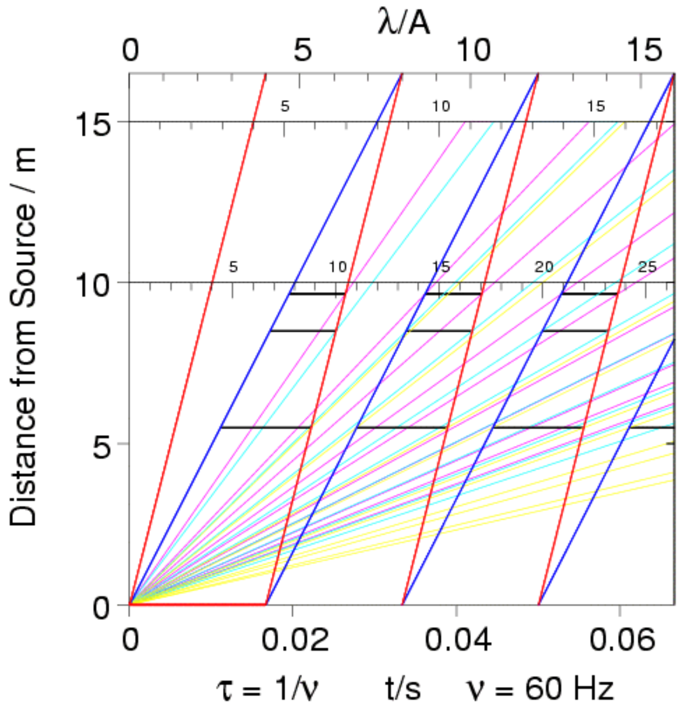
Bandwidth/frame ~ 1/L

$$\Delta l = (h/m_n)/(nL) = (3.96\times10^{-7}\,m^2/s)/(nL) \qquad (2)$$

Analogously, wavelength uncertainty δλ depends only on the pulse width of the source Δτ and on the distance L:

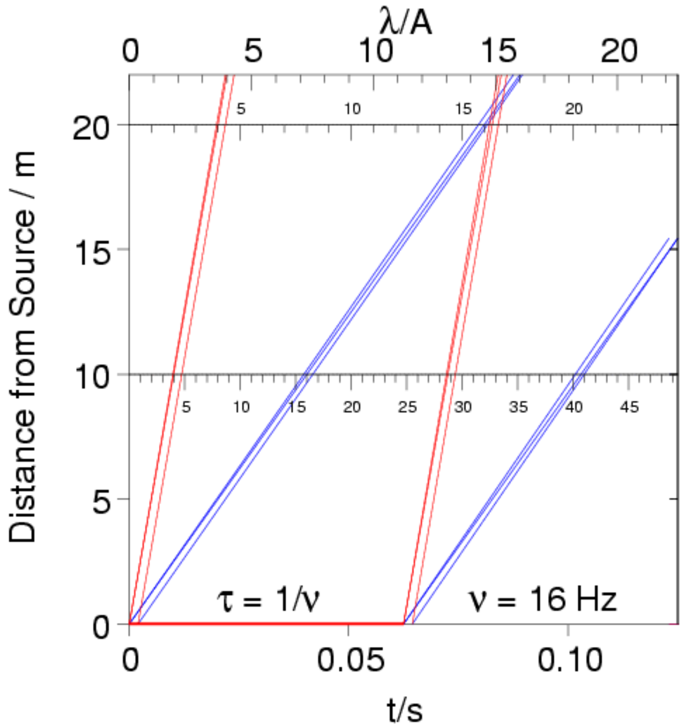
$$dl = (h/m_n)\,\Delta t/L = (3.96\times10^{-7}\,m^2/s)\Delta t/L \qquad (3)$$

The resulting wavelength widths for different situations may be read off from Figures 4 and 5.



50(60)Hz
→ 3+choppers needed

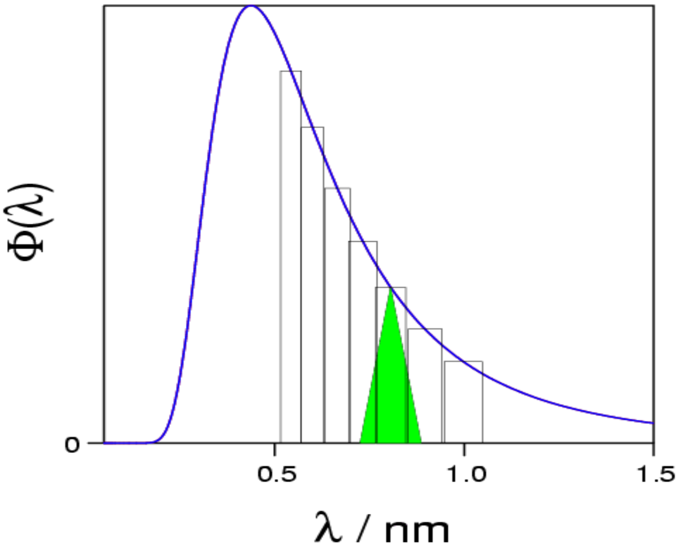
Fig. 4: Path-time diagram for a fast repetition frequency target, min. 3 choppers are needed to prevent wavelength mixing from different pulses. The bandwidth at L=17m is about Δλ=0.4 nm.



16Hz
→ 1 chopper (at 6m)

Fig. 5: Path-time diagram illustrating the situation expected at the long pulse target. The wavelength uncertainty due to the pulse width is about 0.04 nm and the free bandwidth exceeds 1 nm at L=20m allowing for constructions with even larger moderator-detector distances. One frame overlap chopper at about 5m is sufficient.

An unbiased gain factor may be obtained by comparing the time needed to perform a pulsed source experiment with a wavelength band from λ_{min} to λ_{max} with that for a corresponding sequence of experiments using different wavelength bands each with the same relative width w, given by the velocity selector, at a reactor as illustrated by Fig. 6.



Gain factor g_0 ~$\ln(l_{min}/l_{max})/w$

w = selector width at reactor

Fig. 6: Matching selector bandwidth experiments (green triangle) to a time-of-flight sequence encountered in a spallation experiment.

The resulting gain factor is:

$$g_0 = (\Phi_{ESS}/\Phi_{ILL})\ln(l_{\max}/l_{\min})/w \qquad (4)$$

The flux ratio is assumed to be 1 for the 50Hz target and the long pulse target (see Fig. 1), whereas the 10Hz target with it's "diluted" pulse sequence is assigned a flux ratio of 0.2. Furthermore we assume the reference velocity selector width to be 10%, i.e. w=0.1. It should be noted that for a wide wavelength span the distribution of intensity does not perfectly comply with the experimental needs because the counting time is necessarily equal for all sub-bands. Fortunately many SANS samples exhibit increasing intensity with lowering Q (i.e. at longer wavelengths for the same scattering angle) which may partly compensate the spectral intensity drop at long wavelengths. In addition the λ^3 proportionality of the Fourier time fits to the dynamical slowing down with some power of Q. Nevertheless, a gain factor g_0 derived from large wavelength ratios (>2-3) will probably be an overestimation.

50Hz: gain = 2.7-7.4

10Hz: gain = 14 x 0.2 = 2.8

16Hz Long-Pulse: gain = 14 !

ln*(4)/0.1 = 13.9*

For the same distance from the source, a 50Hz target will reduce the waveband width Δλ by a factor of 3 with respect to the 16Hz option, which is the choice par excellence for NSE. This will lead to a consequent reduction of the gain factor. In order to partly counterbalance this drawback the 50Hz target will force to a very compressed design heading at a source-detector distance of L < 18m, which is about the minimum conceivable. This will yield to a free wavelength-band of 0.44 nm with a plain gain factor of 7.4 if the range between 0.4 nm< λ<0.84 nm is considered, and this factor reduces to 2.7 in the more interesting range (as far as large Fourier times are concerned) between 1.4 nm < λ < 1.84 nm.

g_0 > 10 may be questionable

50Hz → L < 18m needed

On the other hand the 16Hz long pulse source would allow for building longer instruments, which would also improve the background. The band width at 20m from the source would be Δλ =1.2 nm and the gain factor would reach almost 14 in the range 0.4 nm < λ < 1.6 nm and would reduce to 6.9 in the range 1.2 nm < λ < 2.4 nm. For the same distance between the source and the detector, the 16Hz long pulse option delivers a gain factor typically twice as high as that of the 50Hz.

Technical Issues

NSE instruments at pulsed spallation sources will look very similar to those at continuous sources. There will be some extra pressure – depending on the repetition rate- to reduce the length if possible. Data collection has to cope with the assignment of incoming neutrons to varying Fourier times t and Q values according to the λ determined from the time-of-flight. Only the flippers and the phase coil (and possibly some current sheets, which would correct for gravitational effects on the paths) should be wavelength dependent in a way that requires broadband operation. The recent results obtained at IN15 showed that current ramping synchronised to the pulse frequency as indicated in Fig. 7 is sufficient for combining NSE and TOF [10] and well defined echo-signals were observed over the totality of a large wavelength frame from λ=0.6 nm up to1.8 nm.

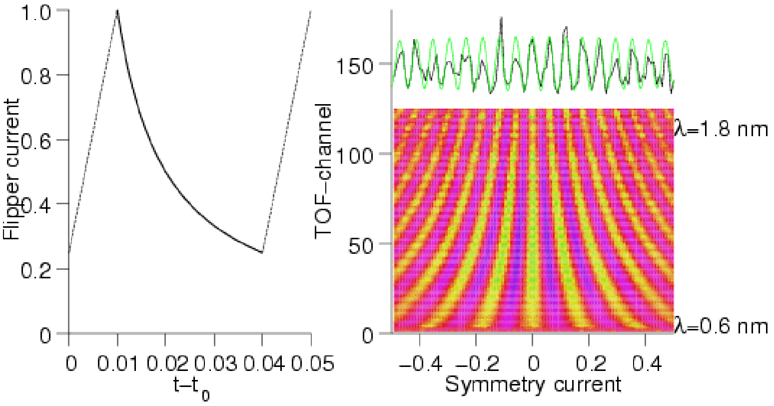
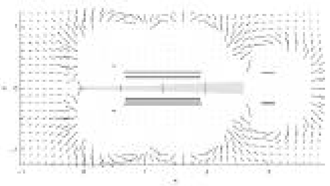


Fig. 7: Dynamical flipper ramping allows for broadband operation.

Dynamical flipper operation by ramped currents is tested

The narrow space for the instruments imposed by the requirement to keep L short and the necessity of massive shielding is not compatible with extended stray fields. Magnetisation of iron shields must be avoided, which calls for well compensated "stray-field-free" designs for the instruments positioned close to the source, which should be of IN11 type. A high resolution SANS - NSE spectrometer, which would benefit the most from broad band operation should be placed typically at a (detector)-distance of 18-20m from the source. Such a spectrometer should be relatively compact and should mainly operate in the SANS range. For that reason it should easily placed in the "crowded" region next to the shielding of the source.

No stray field design will be required



For NSE measurements at high Q a SPAN-type spectrometer with improved energy resolution would be needed. Such a spectrometer should be placed at a larger distance from the source, typically at 40m. This large distance is dictated by the space requirements (diameter of 8-9m) of the spectrometer and should also allow for reducing magnetic cross talk with other instruments. From the equations above it is clear that at 40m from the source the wavelength band will be restricted by a factor of two with respect to that delivered at 20m, which will consequently reduce the gain factors. With the 16Hz long pulse source the gain factor amounts to 9.2 for $0.4\ \mathrm{nm} < \lambda < 1.0\ \mathrm{nm}$ and 4.7 for $1.0\ \mathrm{nm} < \lambda < 1.6\ \mathrm{nm}$, i.e. a factor 1.5 less than the gains obtained at 20 m from the source. When it comes to the 50 Hz target the wavelength band spans only 2 Å and the gain factors amount to 4 for $0.4\ \mathrm{nm} < \lambda < 6\ \mathrm{nm}$ and 2.2 for $0.8\ \mathrm{nm} < \lambda < 1.0\ \mathrm{nm}$, i.e. roughly a factor 2-3 less than at 18m from the source. However, the large solid angle (up to 0.27 strad) over which simultaneous NSE measurements will be possible will largely

SPAN design
L = 40m

16Hz Long-Pulse:
gain = 9.2

50Hz
gain = 4!

compensate the drawback in gain factors. The long distance would also lead to an improved monochromatisation and Q-resolution, which would be a welcome feature of this configuration. Indeed no wide angle NSE spectrometer with good Q-resolution exists at the moment although some NSE experiments at high scattering angles require good Q-resolution.

Finally, compared to the existing instruments additional gains should be expected due to the application of advanced neutron optics like improved neutron guides and optimised analyser/polariser -techniques. Especially the necessarily shorter distances of guide and free flight sections will yield to intensity gains compared to existing installations. Despite the fact that these are "soft" gains they will still contribute to the real achieved gains.

Additional "soft" gains may contribute a factor of 10

References

[1] F. Mezei (Ed.), "Neutron Spin Echo Proceedings", Lecture Notes in Physics 128 , Springer, Berlin (1979)

[2] P. Schleger, G. Ehlers, A. Kollmar, B. Alefeld, J. F. Barthelemy, H. Casalta, B. Farago, P. Giraud, C. Hayes, C. Lartigue, F. Mezei and D. Richter, Physica B, 266 (1999) 49-55

[3] M. Monkenbusch, R. Schätzler and D. Richter, Nucl. Instr. and Methods A399, 301 (1997)

[4] B. Farago, Physica B, 241-243 (1998) 113

[5] C. Pappas, F. Mezei and G. Ehlers, B. Farago, C. Pappas and F. Mezei, ILL Millenium Workshop, 6-7 April 2001, Grenoble France

[6] M. Köppe , M. Bleuel , R. Gähler, R. Golub , P. Hank , T. Keller , S. Longeville, U. Rauch , J. Wuttke Physica B 266 (1999) 75

[7] TASSE (NSE-TAS combination) C.M.E. Zeyen, J. Phys. Chem. Solids 60 (1999) 1573

[8] T. Keller, R. Golub, F. Mezei, and R. Gähler, Physica B 241-243 (1998) 101

[9] M.Th.Rekveldt, T.Keller, W.H.Kraan, Physica B 297 (2001)18

[10] B. Farago et al., ILL Annual Report 1999.