The basics of Neutron Spin Echo

Béla Faragó
ILL - Grenoble

Introduction

Abs.

Until 1974 inelastic neutron scattering consisted of producing by some means a neutron beam of known speed and measuring the final speed of the neutrons after the scattering event. The smaller the energy change was, the better the neutron speed had to be defined. As the neutrons come form a reactor with an approximately Maxwell distribution, an infinitely good energy resolution can be achieved only at the expense of infinitely low count rate. This introduces a practical resolution limit around 0.1μeV on back-scattering instuments.
In 1972 F. Mezei discovered the method of Neutron Spin Echo. ~~As we will see in the following~~ This method decouples the energy resolution from intensity loss.
The basics of this method is presented.

Basics

It can be shown [1] that an ensemble of polarized particle with a magnetic moment and 1/2 spin behaves exactly like a classical magnetic moment. Entering a region with a magnetic field perpendicular to its magnetic moment it will undergo Larmor precession. In the case of neutrons:

$$\omega = \gamma B$$

where B is the magnetic field, ω is the frequency of rotation and γ=2.916kHz/Oe is the gyromagnetic ratio of the neutron.
When the magnetic field changes its direction relative to the neutron trajectory two limiting cases have to be distinguished:
1) Adiabatic change, when the change of the magnetic field direction (as seen by the neutron) is slow compared to the Larmor precession. In this case the component of the beam polarization which was parallel to the magnetic field will be maintained and it will follow the field direction.
2) Sudden change, is just the opposite limit. In that case the polarization will not follow the field direction. This limit is used to realize the static (Mezei) flippers.
Now let us consider a polarized neutron beam which enters a B_1 magnetic field region of length l_1. The total precession angle of the neutron will be:

$$\varphi_1 = \frac{\gamma B_1 l_1}{v_1}$$

depending on the velocity (wavelength) of the neutrons. If v has a finite distribution, after a short distance (a few turns of the polarization) the beam will appear to be completely depolarized. If now the beam will go through an other region with opposite field B_2 and length l_2 the total precession angle is:

$$\varphi_{tot} = \frac{\gamma B_1 l_1}{v_1} - \frac{\gamma B_2 l_2}{v_2}$$

In the case of elastic scattering on the sample $v_1=v_2$ and if $B_1l_1 = B_2l_2$ then φ_{tot} will be zero independently of v and we recover the original beam polarization. Let us suppose now that a neutron is scattered with a small ω energy exchange between the B_1 and B_2 region. In that case in leading order in ω :

$$\varphi_{tot} = \frac{\hbar\gamma B l}{mv^3}\omega$$ where m is the mass of the neutron

If we put an analyzer after the second precession field and the angle between the polarization of a neutron and the analyzer direction is φ then the probability that a neutron is transmitted is $\cos\varphi$. We have to take the expectation value of $\langle\cos\varphi\rangle$ over all the scattered neutrons. At a given q the probability of the scattering with ω energy exchange is by definition $S(q,\omega)$. Consequently the beam polarization measured

$$\langle\cos\varphi\rangle = \frac{\int \cos(\frac{\gamma B l}{2mv}\omega)S(q,\omega)d\omega}{\int S(q,\omega)d\omega} = S(q,t)$$

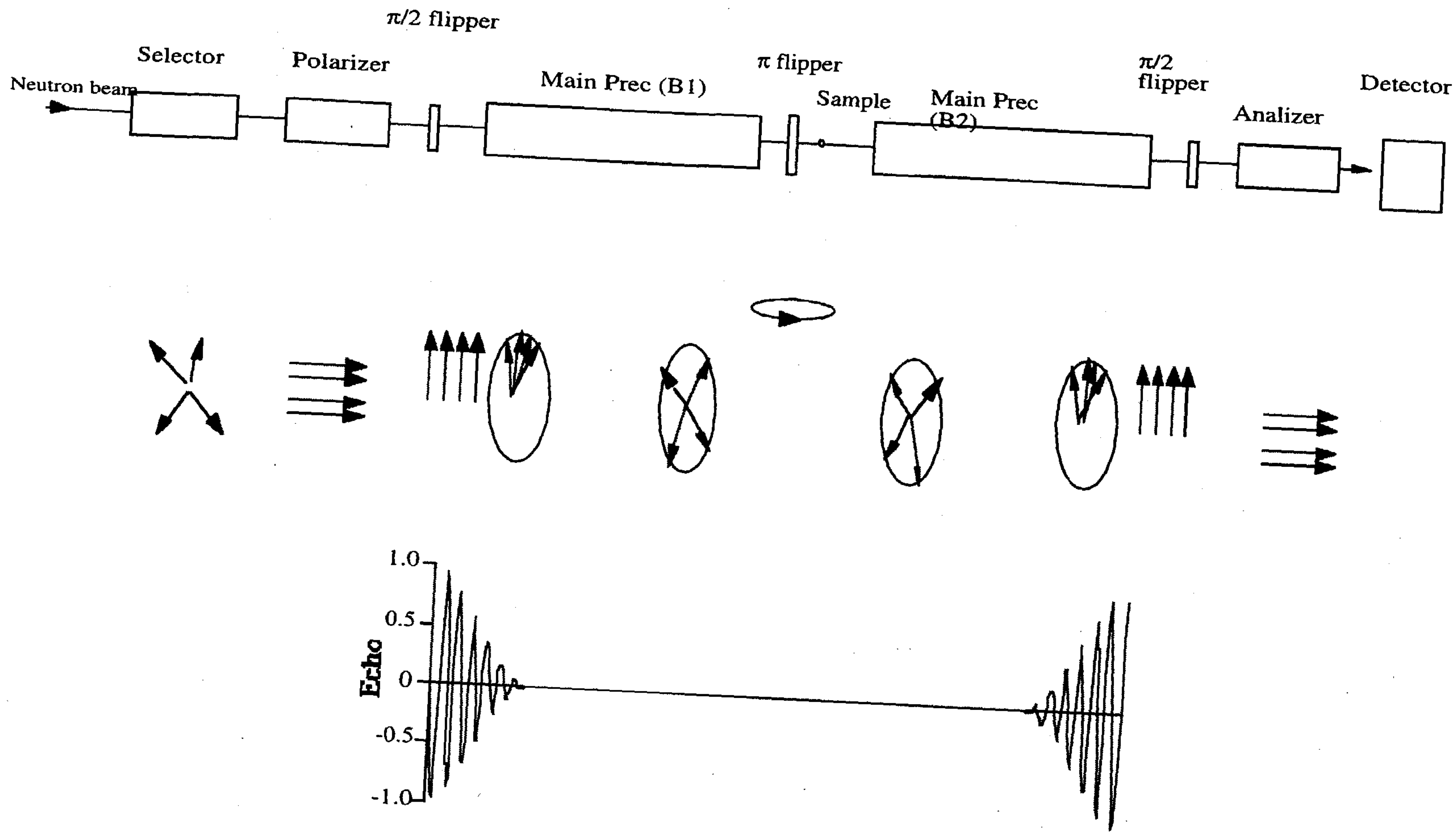
So NSE directly measures the intermediate scattering function where:

$$t = \frac{\hbar\gamma B l}{mv^3}$$

It is important to notice that $t \propto (1/v)^3 \propto \lambda^3$ so the resolution in t increases very rapidly with λ.

Implementation

In practice reversing the magnetic field is difficult as in the middle this would create a zero field point where the beam gets easily depolarized. Instead, a continuous horizontal field is used (conveniently produced by solenoids) and a $\pi/2$ flipper starts the precession by flipping the horizontal polarization perpendicular to the magnetic field. The field reversal is replaced by a π flipper which reverses the precession plane around an axis and at the end a second $\pi/2$ flipper stops the precession and turns the recovered polarization in the direction of the analyzer.


π/2 flipper
Selector
Polarizer
Neutron beam
Main Prec (B1)
π flipper
Sample
Main Prec (B2)
π/2 flipper
Analizer
Detector
1.0
0.5
0
-0.5
-1.0
Echo

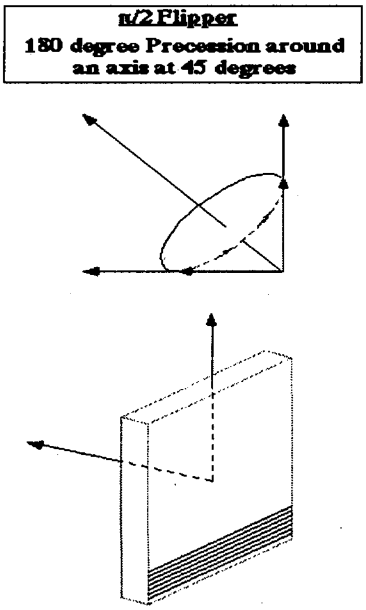
Elements

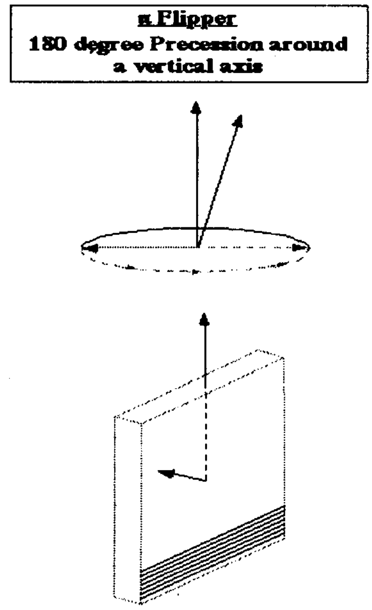
Monochromatisation:
The biggest advantage of NSE is that it decouples monochromatisation form energy resolution. Whenever q resolution is not so important (or it is determined by the angular resolution rather than the wavelength distribution), a neutron velocity selector with high transmission (e.g. $\Delta\lambda/\lambda$=15% FWHM) is the best choice. If necessary the wavelength distribution can be reduced by installing after the analyzer a crystal monochromator (graphite or mica) and the detector in Bragg geometry.

Polarizer:
This is a very essential part of the spectrometer, and generally supermirrors are used. There are two possible choices, reflection or transmission mode. As the polariser-sample distance is in the order of 2-3m the main danger in both cases is the unwanted increase of the incoming beam divergence on the polariser. Due to that relatively big distance here one can loose integer factors in the neutron flux on the sample. Inherently the reflecting supermirrors are more susceptible to increase the beam divergence, and also have the inconvenience that the reflection angle might have to be adjusted to different wavelength if the useful range is large enough. On the other hand the supermirrors of this type were easier to obtain.

π and π/2 flipper:

The flipper action is based on the nonadiabatic limit. Using a flat coil (≈5mm thick) perpendicular to the neutron beam with a relatively strong field inside and arranging such that the horizontal field is small, the polarized neutrons going through the (thin) winding do not have time to follow adiabatically the change in the field direction. All of a sudden the beam polarization is no more parallel to the magnetic field and it will start precess around the field. With the appropriate choice of the field strength and direction the neutrons will precess exactly π or π/2 by the time they fly through the flipper.





Main precession coils

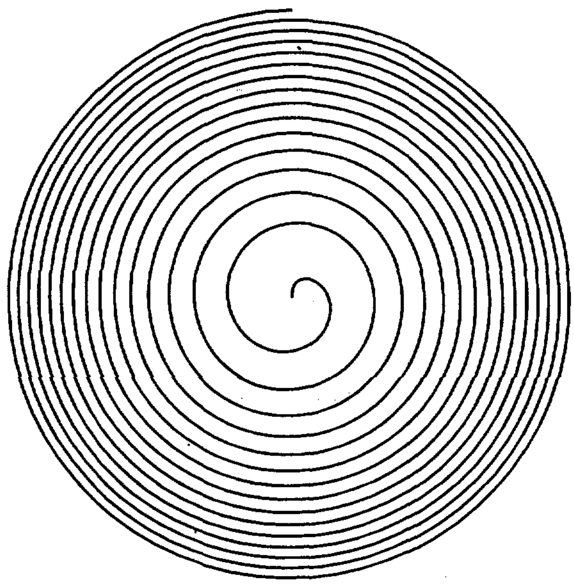
We have seen that the Fourier time is proportional to the field integral. If we want a good resolution, we need high field and long integration path. Both will have a drawback. Even the most symmetric solution for the main precession coil, a solenoid, will produce an inhomogenous field for finite beam size. The underlying physical reason is that in the middle of the precession coil we have a strong field and at the flippers, as was explained above, we need a small horizontal component. Since Maxwell we know that div(B) is zero, consequently the transition region will introduce inhomogeneities.
Making long precession coils is not necessarily optimum because that increases the distance between the source (polariser exit) and the sample as well as the sample-detector distance. The counting rate will decrease with $R^{-2}*R^{-2}$.

"Fresnel coils"

How to correct the unavoidable inhomogeneities due to the finite size beam? Let us consider the field integral difference between the trajectory on the symmetry axis and one parallel at a distance r through the solenoid. In leading order this gives:

$$\Delta\int B dz \cong \frac{r^2}{8}\int \frac{1}{B(z)}\left(\frac{\partial B(z)}{\partial z}\right)^2 dz$$

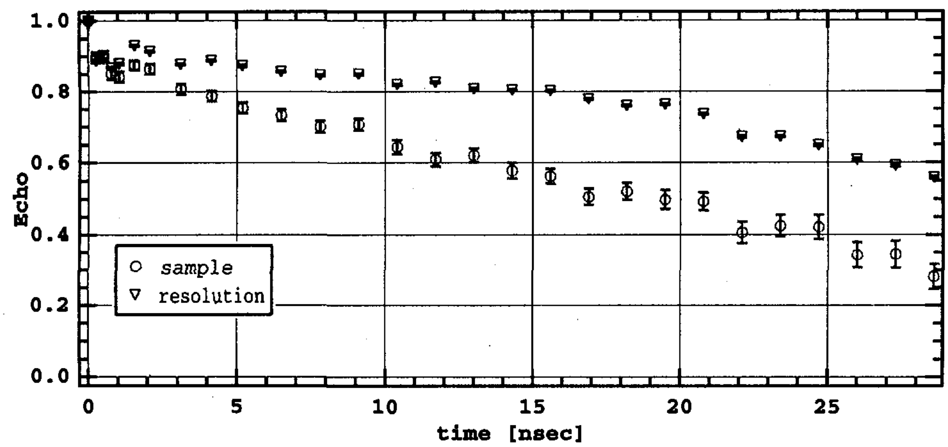
It can be shown also that a current loop placed in a strong field region changes the field

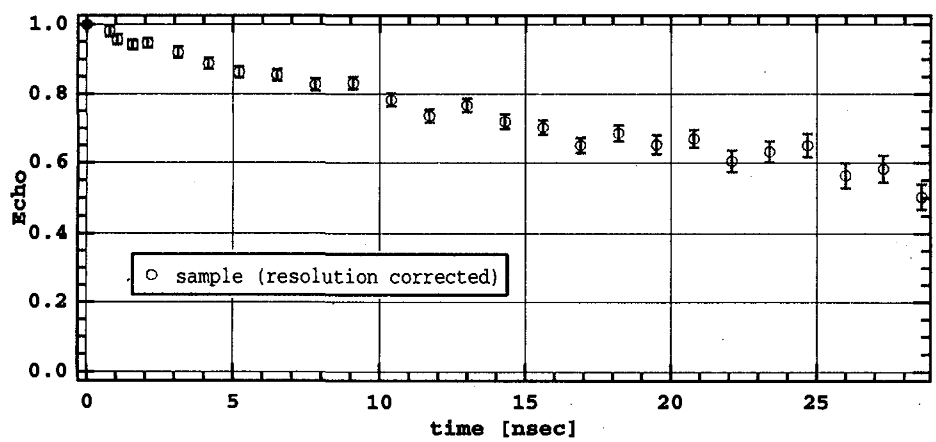


integral by 4πI, if the trajectory passes inside the loop and by zero if it passes outside. To correct the above calculated r^2 dependence we just have to place properly arranged current loops in the beam. The so called Fresnel coils just do that.[1]
They are usually made by printed circuit technology preparing two spirals where the radius changes with $r \propto \sqrt{\varphi}$ placed back to back with a thinnest possible isolator (30micron kapton) to minimize neutron absorption.

Resolution:

Why field inhomogeneities are bad? In fact all we are interested in is the field integral. We have to use finite beam size if we want to have finite counting rates. With finite beam size we have different neutron trajectories arriving on the sample and coming to the detector. If the field integral for different trajectories are non equal then the final precession angle $\varphi_{tot} = \frac{\gamma B_1 l_1}{v_1} - \frac{\gamma B_2 l_2}{v_2}$ will be different for different trajectories and the detected polarization even for an elastic scatterer will be reduced to $\langle \cos\varphi \rangle$. The echo measured on the sample will be further reduced due to the energy exchange. Using an elastic scatterer we can measure the influence of the inhomogeneities and divid ir out. (in other words while in energy space the instrumental response has to be deconvoluted from the measured curve to obtain the sample response, in the Fourier transformed space the deconvolution becomes a simple division)
Nevertheless if the field integral for different trajectories is too different, then $\langle \cos\varphi \rangle$ quickly becomes zero and no matter how precisely we measure it, division by zero is usually problematic!

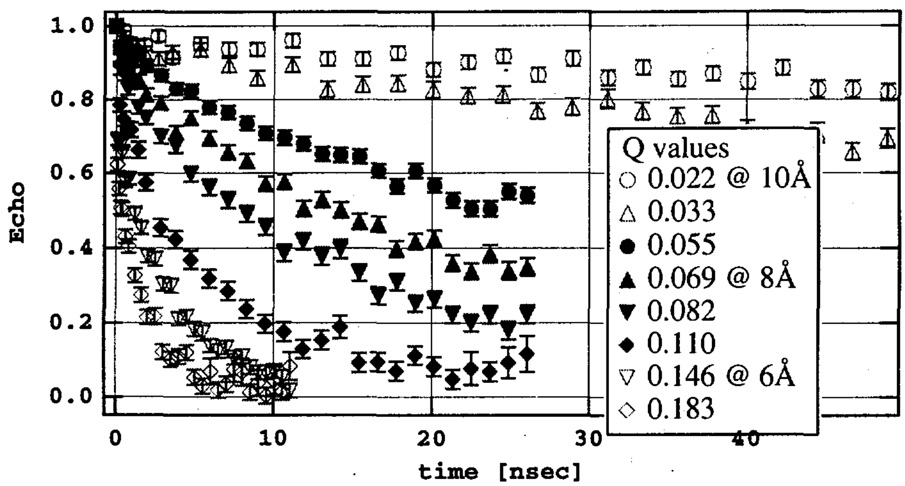




To put this into context lets have a look at some realistic numbers:
On IN11 we have a maximum field integral of $\approx 3*10^5$ Gausscm. With 8Å wavelength this gives a Fourier time of 28nsec. An exponential decrease with a time constant of 28nsec correspond in energy space to a Lorentzian line width of 23 nev. With the incoming neutron energy (8Å) his is a relative energy exchange of the order of 10^{5}. The 360 degree phase difference between two trajectories corresponsd to 17 Gausscm difference in field integral. This means a requested relative precision of better than $6*10^{-5}$ for all trajectories!
So what is the theoretical limit to the achievable resolution? There is no easy answer. In principle with sufficiently precise in-beam correction coils (like the Fresnels but including higher order term corrections) any resolution should be reachable. In practice we could certainly reached already an overall correction better than 10^{-4} .

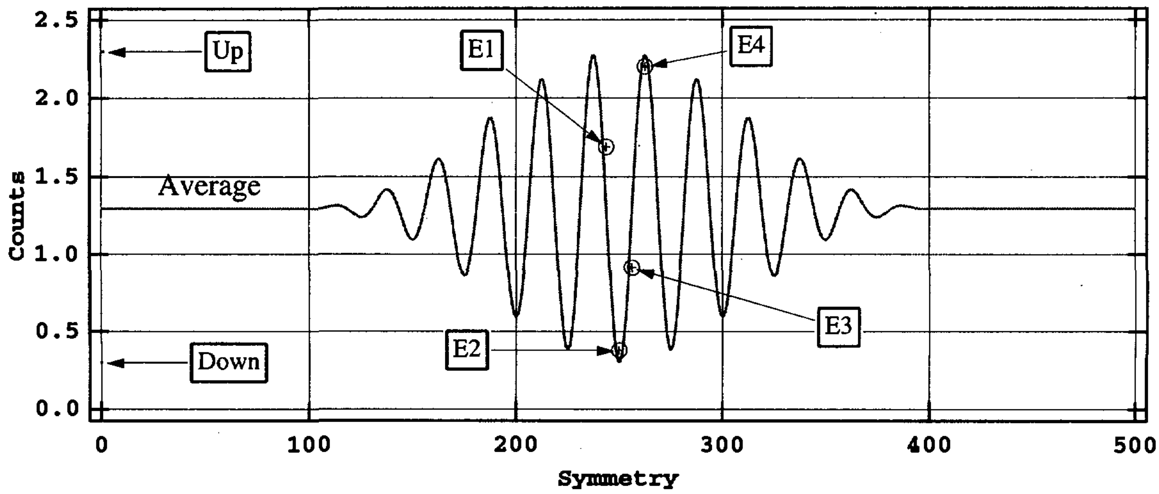
Measurement sequence

Now we will detail how an experiment is really done. Once the aims of the experiments are clear one has to decide what wavelength to use. Long wavelength gives better resolution but lower flux. In the example below we illustrate for a given sample (Polystyrene solution in deuterated cyclohexane) how much one can gain by using different wavelengths to match the requested resolution. Going from 10Å to 8Å and 6Å in each step we gain about a factor five in intensity.



Then it has to be decided at which precession field (Fourier time) the echo pints has to be measured. Depending on the problem studied, it can be adequate to take equidistant points on a logarithmic scale.

For each wavelength and each precession field value, the flippers have to be tuned to make the right π or $\pi/2$ flips as was explained above. The vertical component in the flipper depends nominally only on the wavelength, but the horizontal component must be adjusted with the help of correction coils, and these values depend on the wavelength and on the stray field of the precession coils. This is rather time consuming even if it is fully automatized. The field integral on the two side of the π flipper must be equal to a precision of 10^{-5} . This is done by using an additional winding on the first precession coil, the symmetry coil. Scanning the current in this coil will go through the exact symmetry point producing the typical echo group as shown below



The periodicity of the damped oscillation is determined by the average wavelength and the envelope is the Fourier transform of the wavelength distribution. These depend only on the monochromator (velocity selector) so do not carry information on the sample. (With one notable exception, when the sample is crystalline and remonochromatize the beam. In that case the echo envelope becomes much more extended)

With the echo we measure how much of the initial polarization we recover. For elastic scatterer on an ideal instrument we recover it fully. During the scattering process the initial beam polarization can change. One reason for this can be when the sample contains hydrogen which has a high spin incoherent cross section. Spin incoherent scattering changes the initial polarization from P to -1/3P. The scattered intensity is thus composed of the sum of coherent and incoherent scattering decreasing, the up/down ratio. The relative weight of these can, and will change as a function of scattering angle (or wave vector q). The measurement sequence thus starts with the measurment of spin-up and spin down for the given scattering angle and sample condition. This is accomplished by setting a guide field to maintain beam polarization and counting with π flipper on and π flipper off. This will give us two points marked as Up and Down on the figure above. It would be inefficient to measure always the whole echo group as we are interested only in its maximum amplitude. At most it is equal to (Up-Down)/2. On an elastic scatterer we can quickly find where the center of the echo group is as a function of the current in the symmetry coil. (Note that this current will be a function of the scattering angle because changing the direction of the detector arm will change the field overlap of the two solenoids and also the magnetic contribution of the earth field will change.) As we also know the periodicity of the sinusoidal echo group it is more efficient to measure four points placed around the center with $\pi/2$ steps. This will give us

$E1 = Aver+Echo*\sin(\phi)$
$E2 = Aver-Echo*\cos(\phi)$
$E3 = Aver-Echo*\sin(\phi)$
$E4 = Aver+Echo*\cos(\phi)$

From the four measurement Aver,Echo and ϕ (the phase) can be determined, and finally

$$I(q,t)/I(q,0) = 2*E/(Up-Down)$$

If we are sure that the phase is zero, then it is sufficient to measure E2 and E4. However in general to monitor any possible phase drift, (due to current instability, external perturbation, ... or accidental displacement of a coil.....) usually all the four points are measured but giving more weight to E2 and E4. This four point measurement has to be repeated at all the precession angles, but the Up and Down counts are measured only once at the beginning.

Further optimization can be done as follows

Because (Up-Down)/2 = Average and (E1+E2+E3+E4)/4 = Average, furthermore this is the same for all precession currents at a given scattering angle, we have a very precise measurement of the Average. Counting Down and E2 longer than the other points, we gain a lot in statistical accuracy.

Signal and Background

No general recipe can be given, beside that both should be measured and the appropriate correction done! Nevertheless lets discuss a few typical cases and for the sake of simplicity lets take an ideal instrument (all flippers, polariser,analyzer 100% efficiency, perfect field integrals)

Coherent, (spin) incoherent scattering:

As was mentioned incoherent scattering changes the beam polarization to -1/3. Let's consider the case when the scattered intensity is composed of $I= I_{coh} + I_{incoh}$. Then we will have

$Up = I_{coh}+1/3I_{incoh}$
$Down = 2/3I_{incoh}$
$Echo = I_{coh}*f_{coh}(t) -1/3I_{incoh}*f_{incoh}(t)$

where $f_{coh}(t),f_{incoh}(t)$ denotes the time dependence (dynamics) of each contribution and at t=0 both are equal to one.

Our usual data treatment will give

$$I(q,t)/I(q,0) = [I_{coh}*f_{coh}(t) -1/3I_{incoh}*f_{incoh}(t)]/[I_{coh}-1/3I_{incoh}]$$

This means that from Up and Down we can determine the relative weight of each contribution but the separate time dependence is impossible to extract (except if suitable model exist and even then it is hard to achieve the necessary statistical accuracy) . Fortunately in many cases it is possible to minimize the incoherent contribution by using deuterated samples.(Note the incoherent echo signal is further reduced by the factor 1/3) In other cases it is possible to suppose that the dynamics of $f_{coh}(t)$ and $f_{incoh}(t)$ is the same or nearly identical and then

$$I(q,t)/I(q,0) = [I_{coh}*f_{coh}(t) -1/3I_{incoh}*f_{incoh}(t)]/[I_{coh}-1/3I_{incoh}] = f(t)$$

There can be also some scattering from the sample holder, solvet, etc. Some cases can be treated in a simplified way. For example in polymer solution scattering from deuterated solvent gives coherent scattering, but it is too inelastic to be seen by NSE, which means its relaxation time is out of the time window. In this case it will influence Up-Down but gives no echo. It is sufficient to measure Up and Down of the solvent and eventually verify at one precession field that there is no echo signal. Similarly sample holders usually give only some elastic scattering. Again Up,Down and a few echo points in time are sufficient to establish its contribution.

NSE vs standard inelastic instruments

NSE not only has very high energy resolution but also has a very wide dynamical range. With some tricks it can be as high as tmax/tmin = 1000 . With the combination of several wavelength this can be further extended by a factor 10-100. We can measur e energy exchanges up to 100μeV which not only overlaps with back scattering but also with TimeOfFlight like IN5 or IN6. When to choose which instrument?
The fundamental difference is that NSE measures in Fourier time while TOF in omega space. Consider a sample which has a strong (95% of the intensity) elastic line, and well defined but weak (5%) excitations at finite energy. While this is well separated in omega space on NSE the Fourier transform will give a small (5%) cosine oscillation around 0.95. Statistically this is very unfavorable. Furthermore the rough wavelength distribution (around 15% FWHM) of NSE will smear out the oscillations ($t \sim\lambda^3$). For that type of experiments TOS is better suited.

On the other hand when all (or most of) the intensity is quasielastic, NSE can give more precise information on the time dependence. The main reason is that the deconvolution of the instrumental resolution is replaced by a simple division which leaves the data less influenced by numerical manipulation.
Studying fully incoherent scatterers is also not very favorable for NSE. Already incoherent scattering in general gives rather weak intensity, secondly the -1/3 factor in polarization decreases the statistical accuracy. These drawbacks are somewhat lifted by now on the multidetector spin echo instruments like In11C and SPAN in Berlin. (see below)

Variants

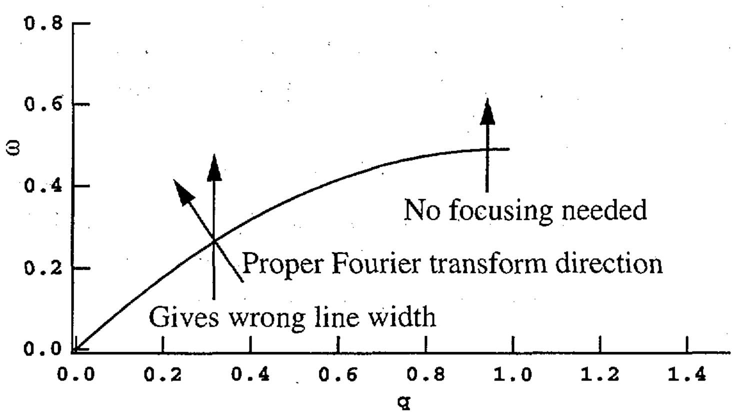
inelastic NSE

One could ask the question whether the method could be used in the case of well defined excitations? The answer is yes, but with some modification. By choosing $B_1l_1 \neq B_2l_2$ the same Fourier back-transformation happens but around the excitation energy ω_0 [1]:

$$\omega_0 = \frac{m}{2\hbar} v_1^2 \left(1 - \left(\frac{B_1 l_1}{B_2 l_2} \right)^{\frac{3}{2}} \right) \qquad \frac{\int \cos\left(\frac{\gamma B_1 l_1}{2mv_1} (\omega - \omega_0)\right) S(q,\omega) d\omega}{\int S(q,(\omega-\omega_0)) d\omega} = S(q,t)$$

v_1 is the velocity of the incoming neutron and v_2 is after the scattering

The problem here is that around ω=0 $S(q,\omega)$ usually has a strong peak (either quasielastic scattering, diffuse elastic scattering and/or incoherent scattering or just instrumental background) The solution is to use a triple axis spectrometer geometry which acts as a filter, pre-cutting an interval around ω_0 with a coarse resolution compared to NSE. There is a further requirement when ω is q dependent. (like a phonon branch) We have to make the spin precession dependent on the incoming and outgoing direction. Interested readers should look up the details in ref[1]. Here we just give an easy to understand picture. Quasielastic NSE measures the Fourier transform from ω to time space. This is a Fourier transform in the 1D space. Now we switch to the (at least) 2D q-ω space. If ω is q independent (at least in the vicinity we are measuring, e.g. close to the Brillouin zone boundary) with asymmetric field integral we make the Fourier transform around a finite ω_0 and we obtain the line width. Now if we do the same e.g. on an acoustical branch we will Fourier transform along a vertical line. To measure the real physical line



width, we need to do the Fourier-transformation along a line perpendicular to the dispersion relation. This refocalisation of the echo works if the spin precession becomes dependent on the neutron direction.

Magnetic scattering

The application of NSE to magnetic scattering has special features. One is that ferromagnetic samples can hardly be studied. The random orientation of magnetic domains introduces Larmor precessions around random axis and unknown precession angles normally resulting in depolarisation of the beam and the loss of the echo signal. In some cases the application of a strong external field to align all the domains in the same direction can help for one component of the polarization to survive [1]

The second case is paramagnetic scattering. If the scattering vector (q) is in the **y** direction, then in the sample only the spin components which are in the **xz** plane (perpendicular to q) will contribute to the magnetic scattering. As the precession plane of the neutrons in the usual NSE geometry is the **xy**, the neutrons which arrive spin parallel to q will undergo spin flip scattering, while those arriving perpendicular to q will have 50% probability of spin flip and 50% probability of non spin flip scattering. Accordingly the scattered beam polarization can be decomposed as

$$\frac{1}{2}\begin{pmatrix}-P_x\\-P_y\\0\end{pmatrix}+\frac{1}{2}\begin{pmatrix}P_x\\-P_y\\0\end{pmatrix}.$$

We can realize that the second term is equivalent to a π flipped beam around the **x** axis. Consequently without a π flipper the second term will give an echo with 50% amplitude of the magnetic scattering, while using a π flipper the first term results in a negative echo of 50% amplitude plus a full echo from the eventual nuclear scattering as well. This means that without π flipper ONLY the magnetic scattering gives an echo signal, which eliminates the need of time consuming background measurements to separate the nuclear contribution.

Antiferromagnetic samples do not depolarize the beam, so in most cases they are identical to paramagnetic samples. However if we measure a single-crystal it might happen to be a single domain and depending on the spin direction one can have full echo without π flipper or we might need a π flipper to see any echo (spins parallel to q).

ZFNSE

This method was introduced by Gähler & Golub [2]. We will just give a quick explanation how to understand the basic principle.
The magnetic field profile along the axis of a classical NSE schematically is the following: Nearly zero horizontal field at the first $\pi/2$ flipper, strong (B_0) field in the first solenoid, nearly zero at the π flipper, again strong field in the second precession region, finally nearly zero again at the second $\pi/2$ flipper. In B_0 the neutron precesses with the Larmor frequency $\omega_0=\gamma B_0$. Let us choose now a rotating frame reference which rotates with ω_0. In this frame the neutron seems not to precess in the solenoids which means it sees $B'_0=0$. However now in the originally low field region it will rotate with ω_0 which indicates the presence of a strong B_0 field. The small field of our static flipper will appear to rotate also with ω_0. Let us realize this configuration now in the lab frame. Inside the π and $\pi/2$ flippers we must have a strong static field B_0 and perpendicular to that a small field which rotates with $\omega_0=\gamma B_0$. We need a well localized strong magnetic field perpendicular to the neutron beam, and inside a smaller coil perpendicular both to the neutron beam and to the strong field. This small coil has to be driven by a radiofrequency generator which will produce the necessary rotating field. In between the these flippers a strictly zero field region has to be insured to avoid "spurious" spin precession.

Multidetector variants

At the time of writing, four instruments are developed to extend the "classical" NSE to use multidetectors, and thus reducing the data acquisition time. In all cases the problem is that useful information can be collect in all detectors only if the echo condition can be fulfilled simultaneously for all of them. This is not a straightforward exercise as the field integrals must be equal with a precision of 10^{-5}! This strict condition can be somewhat relaxed, if we can have enough information in each individual detector to make the phase correction on a one-by-one basis. This is not always possible if the scattering is weak. In that case on an elastic scatterer the relative phase differences between detectors can be measured and imposed during the data treatment.

[1] Mezei,F. (editor) Neutron Spin Echo Lecture Notes in Physics 128 Springer-Verlag 1979
[2] R.Gähler, R.Golub Z.Phys, B65 (1987) 269.

Aknowledgement:
The author had the chance to learn most of his knowledge from F. Mezei.