Neutron-spin-echo from polarizing samples

J. Kindervater\textsuperscript{1}, W. H{"a}ußler\textsuperscript{2,1}, A. Tischendorf\textsuperscript{1} and P. B{"o}ni\textsuperscript{1}
\textsuperscript{1}Technische Universit{"a}t M{"u}nchen, Physik Departement E21, 85748 Garching, Germany
\textsuperscript{2}Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM II), 85748 Garching, Germany
E-mail: jonas.kindervater@frm2.tum.de

Abstract. The dependence of the neutron spin polarization on the difference of the magnetic field integrals along the precession fields before and after the sample gives rise to a spin-echo signal. In neutron spin-echo (NSE) experiments with non-polarizing samples, a single spin-echo group is observed (or two in case of some magnetic sample systems). However, in NSE experiments using polarizing samples, three different groups appear: i) the spin rotation (SR) group, ii) the antiparallel echo (APE) group and iii) the parallel echo (PE) group. The echo groups contain information on the life time of the excitations from which the intermediate scattering function $S(Q,t)$ can be derived. In this article, we discuss the basic strategies for NSE measurements using the 2-, 4-, and 20-point method. Finally, we report on quasi elastic NSE experiments on a polarizing magnetic sample MnSi, which stabilizes a single-handed spin spiral. Here, the overlap of the spin rotation and spin-echo group for small spin-echo times requires a measurement of the NSE signal over a wide range of phase angles.

1. Introduction

Neutron Spin Echo (NSE) spectroscopy \cite{1} as a quasi elastic neutron scattering technique uses the neutron spin to detect small changes in kinetic energy of the neutrons before and after the sample. The NSE-technique yields directly the intermediate scattering function $S(Q,t)$ being the Fourier transform of the scattering function $S(Q,\omega)$. NSE allows to measure the dynamics of excitations with sub-\textmu eV resolution. A measure for the resolution is given by the spin echo time $\tau = (\gamma m_n^2 \bar{\lambda}_0^3 BL)/(2\pi h)$, where $\gamma$ is the gyromagnetic ratio, $m_n$ the mass and $\bar{\lambda}_0$ the average wavelength of the neutrons. $\tau$ is proportional to the magnetic field integral $BL$. In this paper we report on the effects of polarizing samples on the NSE-signal at small $\tau$ when the spin echo groups interfere with a spin rotation (SR) group. Therefore, to determine $S(Q,t)$ from a NSE-measurement, the influence of the sample on the polarization of the beam has to be taken into account.

Section 2 gives a short introduction to the NSE-technique. In section 3, three standard methods for analysing NSE-data are discussed. In part 4 we show how to treat NSE-data from polarizing samples in the limit of small $\tau$.

2. Neutron Spin Echo Spectroscopy

Fig. 1 displays a schematic of the NSE spectrometer RESEDA (Ref. \cite{2}) located at the end position of neutron guide NL5 at the Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM II). The wavelength band of the incident polarized beam is determined by a velocity selector. While passing the precession region in the first coil, the neutron spin accumulates a spin phase
Figure 1: Principal components of RESEDA. The zero field regions are produced by means of double $\mu$-metal shielding.

$\phi_1$ that is given by $\phi_1(\lambda) = \gamma B_1 L_1 \lambda / (\hbar m_n)$. In the second spectrometer arm the field $B_2$ is anti parallel to $B_1$. Therefore the spin of the neutrons precesses backwards and the net spin phase is $\phi = \phi_2 - \phi_1$. The final polarization $P$ of the neutron beam is directly proportional to $S(Q, t)$. The constant magnetic fields as used in NSE can be replaced in the Neutron Resonance Spin Echo (NRSE) technique by RF-spin flippers that define the boundaries of regions with zero field as shown in Fig. 1 (Ref. [3, 4]). The dependence of the neutron spin polarization on the difference of the magnetic field integrals along the precession fields before and after the sample gives rise to a spin echo signal. For both, NSE and NRSE in case of a monochromatic beam, the intensity at the detector, i.e. the so called spin echo group, is given by

$$I(\lambda) = I_{av}(P \cos(\phi) + 1) = A(\cos(\phi) + 1) + I_{BG},$$  \hspace{1cm} (1)

where $I_{av}$ is the average intensity, $P$ the polarization, $\phi(\lambda)$ the spin phase and $I_{BG}$ the background. In the ideal case all neutrons of the beam contribute to the spin echo signal and precess the same way along the magnetic field. Commonly this is described by a fully polarized echo i.e. $P = 1$. A real spin echo measurement is influenced by both depolarization effects and unpolarised background, which can not be distinguished. Therefore a real measurement can be described either by a reduced polarization $P < 1$ or introducing a background $I_{BG} > 0$. In spin echo the normalized intermediate scattering function $S(Q, \tau)$ is measured. This can either be determined from the polarization $P$ or the normalized amplitude $A/A_0$, where $A_0$ is the amplitude for elastic scattering.

### 3. Standard data analysis strategies

There are various methods used to calculate the polarization of neutrons at the detector. In the following, we discuss the statistical errors that are involved in conducting three different strategies.

For our further discussion we consider an ideal spin echo instrument for which the intensity and therefore the polarization are given by Eq. (1). For the velocity selector we adopt a Gaussian wavelength distribution with $\Delta \lambda / \lambda_0 = 15\%$ corresponding to a relative standard deviation $\sigma_\lambda = \sigma / \lambda_0 = 0.0637$ and with a mean wavelength $\lambda_0 = 5\AA$:

$$f(\lambda, \lambda_0) = \frac{1}{\sigma \sqrt{2\pi}} \exp \left[ -\frac{(\lambda - \lambda_0)^2}{2\sigma^2} \right] \hspace{1cm} (2)$$
The intensity of the spin echo group is given by folding Eq. (1) with the wavelength distribution Eq. (2), i.e.

$$I(\phi(\lambda_0)) = \int_{-\infty}^{\infty} f(\lambda, \lambda_0)(A \cos(\phi(\lambda)) + A + I_{BG})d\lambda = A \left( \cos(\phi) \exp \left[ -\frac{\phi^2}{2\sigma^2} \right] + 1 \right) + I_{BG},$$

(3)

Here, $\phi$ designates the net spin phase for the average wavelength $\lambda_0$. Fig. (2) shows the spin echo signal given by Eq. (3). It is a cosine function that is damped by a Gaussian with a relative standard deviation $\sigma^{-1}$. In the following we discuss two models for each strategy based on the assumption that i) a total number of 1000 neutrons per unit time is scattered by the sample into the polariser before the detector and ii) a background of 500 neutrons per unit time is added to the signal. For example, if a $n$-point echo is measured, for each of the $n$ measured points of the echo signal an intensity of $1000/n$ reaches the analyser before the detector.

3.1. 2-Point-Echo method

The polarization of a neutron beam can be determined by measuring the intensities of the spin up $I_{up}$ and spin down $I_{down}$ neutrons at the detector, i.e. by conducting two measurements:

$$P_1 = \frac{I_{up} - I_{down}}{I_{up} + I_{down}}.$$  

(4)

$I_{up}$ and $I_{down}$ can also be interpreted as two points of the cosine function of the spin echo group shown as green circles in Fig. 2 that are separated by a phase of $\phi = \pi$. Therefore, we call this measurement a 2-point-echo. From Eq. (1) the amplitude is given by $A = P \cdot I_{av}$. Introducing the average counts $I_{av} = (I_{up} + I_{down})/2$ there are altogether three possibilities to determine the amplitude $A$.

- $A_1 = (I_{up} - I_{down})/2$
- $A_2 = I_{av} - I_{down}$
- $A_3 = I_{up} - I_{av}$.

For general NSE measurements, where for example 16 spin echo times are measured, one can consider $I_{av}$ as independent from $\tau$, therefore the statistical error of $I_{av}$ is decreased by a factor of 4. The statistical errors for $A_i$ as determined by using Gaussian error-propagation are given in Tab. 1. A major disadvantage of the 2-point-echo Method is the loss of the phase information leading to large errors in $P$ if the spin echo group accidentally shifts.
3.2. 4-point-echo Method

An alternative strategy is to take four points in steps of $\pi/2$ starting at $-\pi/2$ (red crosses in Fig. 2) allowing to analytically calculate the amplitude of the cosine function. This strategy is used by various groups at the FRM II and the ILL, because it has the advantage of determining $A$ and the phase of the spin echo group. The four points are given by

$$I_n = A(\cos(\phi + \frac{\pi}{2}(n-1)) + 1) + I_{BG}$$

with $n = 0 \ldots 3$. From these points, the amplitude is given by

$$A = \frac{1}{2} \sqrt{(I_0 - I_2)^2 + (I_1 - I_3)^2}.$$  \hfill (6)

The statistical error of the amplitude as calculated by Gaussian error-propagation is given in Tab. 1. The uncertainty is increased from 0.032 to 0.045 when compared with the optimum 2-point-echo Method.

3.3. 20-point-echo Method

The 20 points of the spin echo group are selected in the interval $-9/2\pi \leq \phi \leq 5\pi$ in steps of $\pi/2$ around the spin echo point (blue triangles in Fig. 2). The amplitude of the oscillation decreases with increasing distance from the spin echo point $\phi = 0$. Therefore the statistical error is strongly affected by the width of the wavelength band that defines the width of the Gaussian envelope of the signal. To calculate the statistical error of $A$ we divide the 20-points of the echo into 5 groups of 4 echo points. Each of the groups is analysed according to Eq. 6. The total statistical error is then determined by quadratic summation.

$$\Delta A = \frac{1}{5A} \sqrt{\sum_{i=1}^{4} \Delta A_i}$$

The statistical error of $A$ is slightly increased from 0.045 to 0.053 when compared with the 4-point-echo Method (Tab. 1).

3.4. Comparison of the methods

An overview of the statistical errors of the different methods is given in Tab 1. Considering only the statistical error and the measuring time the best way to measure the polarization is the 2-point-echo Method. Considering that the phase may vary, the 4-point-echo Method becomes the best choice. The benefit of a well known envelope in the 20-point-echo Method is accompanied by a larger statistical error.

If a background of 500 neutrons per unit time is assumed, the statistical error increases by approximately 25% which is a similar effect as a loss of polarization. To compensate the effect of the background, the measurement time has to be increased by a factor of 1.5.

Following the discussion from section 3.1, our standard method for data analysis is as follows: First, all echos are fitted within the 4-point-echo Method with free fitting parameters, which are $A$, $I_{av}$, and $\phi$. If all $I_{av}$ are identical within the errors, the average of $I_{av}$ is calculated and fixed. In a second data treatment the data are now fitted with $A$ and $\phi$ as free parameters. If $\phi$ turns out to be constant too, it can also be fixed in a third data treatment.

4. Neutron spin echo from polarizing samples

Magnetic samples without a centre of symmetry like the B20 compound MnSi may act as a polariser [5] leading to three spin echo like groups. Fig. 3 shows the spin echo signal versus
Table 1: Statistical error of the amplitude (described in section 3). $I_{\text{BG}}$ are the counts added as background to the NSE-signal of the sample. No background: $I_{\text{BG}} = 0$, Background: $I_{\text{BG}} = 500$.

|        | $A_1$ | $A_2$ | $A_3$ | $A_{4}$ | $A_{10}$ |
|--------|-------|-------|-------|---------|----------|
| $I_{\text{BG}} = 0$ |       |       |       |         |          |
| $\Delta A/A$ | 0.032 | 0.046 | 0.077 | 0.045   | 0.053    |
| $I_{\text{BG}} = 500$ |       |       |       |         |          |
| $\Delta A/A$ | 0.039 | 0.065 | 0.089 | 0.056   | 0.064    |

Figure 3: The single-handed magnetic spiral in non-centrosymmetric MnSi polarizes the neutron beam during diffraction from the magnetic satellites. A complete NSE-scan exhibits two spin echo groups (APE and PE) and a spin rotation group (SR). $I_2$ designates the current in the second spectrometer arm.

In the centre there is the SR group and on the left and right hand side the anti parallel echo (APE) group and the parallel echo (PE) group, respectively. The APE group is observed in classical NSE-experiments on non-magnetic materials where the magnetic precession fields are directed anti parallel. The PE group is typically found in magnetically scattering samples where the sample magnetization component transverse to the polarization leads to spin-flip scattering and therefore occurs at parallel field adjustment. The rotation group occurs because the sample polarizes the beam. The spins of the repolarised beam start to precess in the the second NSE coil after scattering. After a few rotations the polarization of the beam is lost due to the broad wavelength distribution. In contrast, the polarization of the APE and PE groups recurs due
to the fact that the absolute value of the magnetic field integrals in both spectrometer arms become equal. In order to record the whole NSE signal, the current of the second NSE coil has to be scanned over a wide range. In magnetic systems, NSE measurements are ideally performed on the PE group where nuclear background scattering does not influence the results. The SR group is independent of the spin dynamics of the sample and does not depend on $\tau$.

4.1. Theoretical spin echo from a polarizing sample

From now on we focus on a special case: We assume that the sample system acts as a polariser, polarizing along an axis in the precession plane. The intensity from such a sample system is given by

$$I_{\text{final}} = I_S \left( \frac{1 + \cos(\phi_1(\lambda))}{2} \right) \left( \frac{1 + \cos(\phi_2(\lambda))}{2} \right).$$

(8)

Here, $I_S$ is the mean intensity scattered from the sample into the detector. $\phi_1$ and $\phi_2$ describe the phase of the spin precession after the primary and secondary spectrometer arm, respectively. The intensity at the detector is given by integrating Eq. (8) over the Gaussian wavelength distribution yielding

$$I = \frac{I_S}{4} + \frac{I_S}{4} \cos(\phi_1) \exp \left[ -\frac{\phi_1^2}{2\frac{\lambda_0^2}{\sigma^2}} \right]$$

$$+ \frac{I_S}{4} \cos(\phi_2) \exp \left[ -\frac{\phi_2^2}{2\frac{\lambda_0^2}{\sigma^2}} \right]$$

$$+ \frac{I_S}{4} \cdot \frac{1}{2} \left[ \cos(\phi_1 - \phi_2) \exp \left[ -\frac{(\phi_1 - \phi_2)^2}{2\frac{\lambda_0^2}{\sigma^2}} \right] + \cos(\phi_1 + \phi_2) \exp \left[ -\frac{(\phi_1 + \phi_2)^2}{2\frac{\lambda_0^2}{\sigma^2}} \right] \right].$$

(9)

Here, $\phi_i (i = 1, 2)$ designate the spin phase of neutrons with the average wavelength $\lambda_0$ after the primary and secondary arms. As discussed above the magnetic field is only changed in the secondary arm, i.e. $\phi_1 = \text{const}$. Therefore, the first line of equation (9) is a constant contribution to the intensity. There are three echo like groups: The SR group (line 2) and the two spin echo groups (line 3). The factor of $\frac{1}{2}$ in line 3 shows that the polarization of the APE and PE group limit the maximum available polarization to 50% of the incident polarization.

4.2. Analysis of data from polarizing samples

For long spin echo times, the SR and spin echo groups are well separated (Fig. 4). In this case, the 4-point-echo method is the most appropriate technique to extract the amplitude $A$ from the spin echo groups. Note that the SR group provides the intrinsic polarization of the NSE-spectrometer in combination with the sample. Therefore, depolarization effects by the sample may be directly accounted for. However, with decreasing $\tau$, the SR and spin echo groups start overlapping and the 4-point-echo method cannot be applied any more. Therefore, more spin echo points are required to disentangle the SR and spin echo groups. As discussed above, the SR group is independent of $\tau$. Hence, the data can be analysed in the following way: First, the data sets for all $\tau$ are fitted using Eq. (9) with all parameters free primarily the amplitude and the average intensity of the SR, PE and APE group. In a second step, the average amplitude $A_{\text{SR}}$ of the SR group and $I_{\text{SR}}^{\text{av}}$ are determined. If $A_{\text{SR}}$ and $I_{\text{SR}}^{\text{av}}$ for all measurements are equal within the statistical error, these two parameters are fixed and all data sets are re-analysed and the amplitude of the spin echo groups is extracted.
5. Summary

By means of the spin echo method, the intermediate scattering function $S(Q, t)$ can be directly determined from the dependence of the polarization of the scattered neutrons on the spin echo time. Depending on the sample under investigation, various strategies for analysing the data can be applied. If the spin echo groups are clearly separated from the SR group, the 4-point-echo method may be the most efficient method to extract the amplitude of the polarization. The statistical error can be minimized by fitting multiple data sets thus fixing the value of the average intensity more accurately. In addition an excellent phase stability is obtained. NSE-experiments from polarizing samples lead to the appearance of a spin rotation group that may overlap at short spin echo times with the spin echo groups. Here, the number of the measured echo points has to be significantly increased. We have developed an appropriate strategy to analyse such data and successfully applied the technique to analyse the quasielastic scattering from polarizing MnSi.

References

[1] Mezei F 1972 Zeitschrift für Physik A Hadrons and Nuclei 255(2) 146–160 ISSN 0939-7922 10.1007/BF01394523 URL http://dx.doi.org/10.1007/BF01394523

[2] Häußler W, Gohla-Neudecker B, Schwikowski R, Streibl D and Böni P 2007 Physica B: Condensed Matter 397 112–114 ISSN 0921-4526 URL http://www.sciencedirect.com/science/article/pii/S0921452607001408

[3] Gähler R and Golub R 1987 Zeitschrift fr Physik B Condensed Matter 65(3) 269–273 ISSN 0722-3277 10.1007/BF01303712 URL http://dx.doi.org/10.1007/BF01303712

[4] Golub R and Gähler R 1987 Physics Letters A 123 43 – 48 ISSN 0375-9601 URL http://www.sciencedirect.com/science/article/pii/0375960187907602

[5] Shirane G, Cowley R, Majkrzak C, Sokoloff J B, Pagonis B, Perry C H and Ishikawa Y 1983 Phys. Rev. B 28 6251–6255