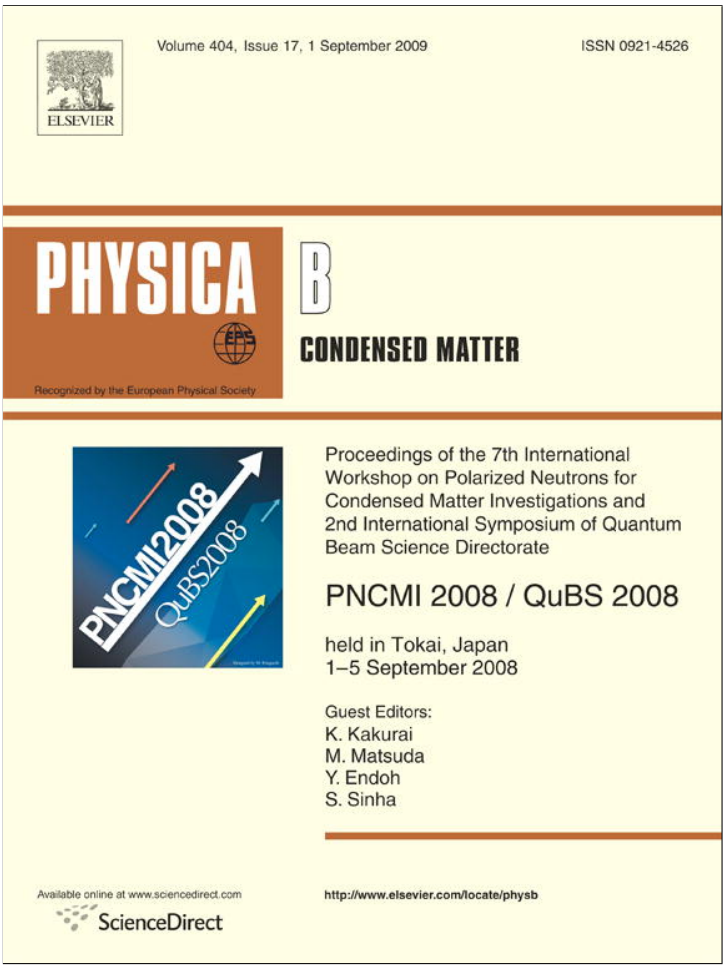

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Challenges in neutron spin echo spectroscopy

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ABSTRACT

With the new brilliant neutron sources and the developments of novel optical elements, neutron spin echo (NSE) spectroscopy evolves to tackle new problems and scientific fields. The new developments pave the way to complex experimental set-ups such as the intensity modulated variant of NSE (IMNSE), a powerful technique which was introduced some 20 years ago but found limited use up to now. With the new compact supermirror or He^3 polarizers IMNSE becomes attractive for a broad range of applications in magnetism, soft matter and biology. A novel development along this line is the polarimetric NSE technique, which combines IMNSE and the zero-field polarimeter Cryopad to access components of the scattered polarization that are transverse to the incoming polarization. Polarimetric NSE is the method of choice for studying chiral fluctuations, as illustrated by new results on the reference helimagnet MnSi.

1. Introduction

In neutron scattering novel concepts and experimental methods are driven by scientific challenges as well as by the science of the neutron and the capabilities offered by the manipulation of the neutron spin. The precession of the neutron magnetic moments in a magnetic field is used in Larmor labeling, Larmor diffraction and of course in NSE to circumvent the Liouville theorem and break the resolution limits in neutron scattering.

Neutron spin echo (NSE) spectroscopy was introduced by Mezei in the early 1970s [1] and revolutionized the thinking about polarized neutrons, which at that time was dominated by the historical longitudinal polarization analysis experiment of Moon et al. [2]. Neutron spin echo is exclusively based on the transverse components of the polarization and demonstrated that neutron polarization is a vector with more information than its projection along the magnetic field, which defines the quantization axis of the system. NSE uses the Larmor precession of neutron spins in a magnetic field to directly measure the energy transfer at the sample and decouples the resolution from beam characteristics like monochromatization or beam collimation. Besides Larmor precession, polarized neutrons give unique information on the arrangement of the magnetic moments in a sample. Polarization analysis is an integral part of neutron spin echo spectroscopy but the situation is straightforward only for "normal" magnetic cross sections [3]. Chiral or nuclear–magnetic interference terms need more sophisticated methods.

Already in the early 1970s it was shown that a zero-field area around the sample is required for accessing components of the final polarization transverse to the incoming polarization [4–6]. This zero-field chamber approach however found limited use, mostly in the direct beam until the development of Cryopad at ILL [7,8]. Cryopad is based on Meissner shields and may be equally used on a three-axis spectrometer or a diffractometer providing access to any scattering angle. The notion of spherical neutron polarimetry (SNP) emerged with Cryopad being able to do the most generalized polarization analysis experiments and to measure any theoretically possible pair-correlation function at any point of the reciprocal space.

An investigation of chiral or nuclear–magnetic interference terms by NSE requires the combination of Larmor precession areas with a zero-field chamber. It is possible to solve these apparently contradictory requirements by combining Cryopad and a variant of intensity modulated NSE [9]. A similar configuration was already proposed in the mid 1980s [10] for the direct beam. The combination of Cryopad with NSE was first demonstrated on the wide angle NSE spectrometer SPAN in HZB [11] and then implemented on IN15 at ILL (Grenoble). In this paper we introduce some modified NSE sequences, the polarimetric NSE method and some preliminary results obtained on a well characterized helical magnet, an MnSi single-crystal [12].

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2. Spin echo sequences

An extensive description of the different neutron spin echo configurations was given by Mezei in the proceedings of PNCMI 2000 [13]. The starting point is the classic neutron spin echo sequence:

polarizer−[$\pi/2$ flip−precession−π flip (sample)−precession] −$\pi/2$ flip−analyzer

with the part in brackets corresponding to the NMR spin echo (SE) sequence, where the second $\pi/2$ flipper is not required. In NSE, when the sample area must be decoupled from the echo measurement it is possible to stop the precessions at the sample. This is achieved by replacing the π flip at the sample by two $\pi/2$ flips before and after the sample, respectively, which leads to the ferromagnetic NSE sequence:

polarizer−[$\pi/2$−precession−$\pi/2$−(sample) −$\pi/2$−precession]−$\pi/2$−analyzer

here again the sequence in brackets corresponds to the NMR stimulated spin echo (SSE) configuration [14], which is the basis of magnetic resonance imaging (MRI). The ferromagnetic NSE subsequence [$\pi/2$–precession–$\pi/2$ flipper] codes the precession phase and stores this information in the beam (or the system of nuclear spin in SSE), which may be recovered at any time by a similar subsequence. In SSE there are no rules for the time elapsed between sub-sequences and often complex schemes with a cascade of flips and even with a superposition of SE and SSEs are used [15].

In neutron spin echo, the situation is different. Neutrons are not steady as the nuclei of an NMR sample but propagate along the spectrometer. In the reference frame of the neutron the π and $\pi/2$ flips are experienced as a time sequence very much like in NMR. In the frame of the laboratory, however, the flippers are steady and the flips take place at different positions along the neutron trajectory. In other words a given time sequence seen by the neutron corresponds to a certain arrangement of a neutron spectrometer. Neutron scattering is an intensity limited technique, and spectrometers tend to be as compact as possible. For this reason the time lap and distance between two ferromagnetic spin echo sub-sequences must be reduced to a minimum.

In the case of polarimetric NSE the area around the sample is taken by Cryopad with its nutators, Meißner and μ metal shields. Spherical polarimetry requires a well defined polarized neutron beam but the broad monochromatization (∼15%) of NSE spectrometers depolarizes the beam at the sample. The neutron spins are distributed on the precession plane that is perpendicular to the magnetic field. For this reason it is important to redefine the beam polarization at the sample using a re-polarizing device in front of Cryopad as illustrated by Fig. 1.

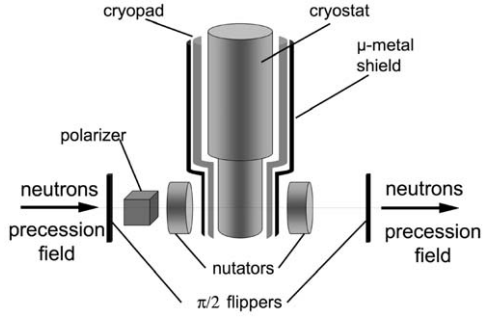

Fig. 1. Schematic view of the arrangement around the sample position with the $\pi/2$-flippers, additional polarizer, nutators and Cryopad.

Additional polarizer and analyzer elements around the sample were first introduced for the intensity modulated variant of NSE (IMNSE) in the mid 1980s [9]. Schematic drawings of an IMNSE arrangement and of a polarimetric NSE setup may be found in these proceedings [16]. IMNSE may be used for samples that completely depolarize the beam. The very first realization of this method lead to the measurement of the emergence of magnons in Fe just below the Curie temperature. The deduced magnon energies of some μeV are probably the smallest energies for magnons ever observed [9].

In polarimetric NSE a re-polarizer is needed in front of the sample to define the incoming beam polarization vector. A component of the scattered polarization vector is then selected by Cryopad and the other two components are depolarized by Larmor precession in the outgoing nutator [17]. This discriminating effect is destroyed in IMNSE when a re-polarizer is installed after the sample because in this case the depolarized components would also give an echo on top of the component selected by Cryopad. It is therefore crucial not to use a re-polarizer after the sample in polarimetric NSE on the contrary to the configuration suggested in Ref. [13].

With the polarizer in front of the sample the amplitude of the modulation becomes 1/2 of the corresponding "normal" paramagnetic spin echo sequence [11]. Assuming an overall transmission of 30% (60% for one spin component) for the additional re-polarizer, the intensity losses reach ∼70%.

The losses increase if the re-polarizing elements have a low angular acceptance and/or require large space, which explains why the original IMNSE experiment remained a unique event without any follow ups for almost two decades.

The situation is now changing with the development of novel neutron optical devices based on supermirrors or He^3 filters. It is now possible to design polarizers which combine high transmission, high polarization and a very compact design. These can be accommodated within some centimeters in space and do not require big modifications of the host instruments. The polarimetric NSE measurements shown here were made with a 10 cm long compact solid state re-polarizer [18], which gave a flipping ratio of ∼45 at a wavelength of 9 Å.

3. Experimental results

The first polarimetric NSE measurements took place at the wide angle NSE spectrometer SPAN at BENSC [11]. The aim was to study with this new method the chiral fluctuations in MnSi close to T_C. The time domain covered by SPAN however was not adapted to this investigation. The next step was to implement the polarimetric NSE on the high resolution NSE spectrometer IN15 (ILL) and start a series of measurements to unambiguously identify and analyze the chiral fluctuations in MnSi close to the helical transition point.

MnSi crystallizes in the $P2_13(T_4)$ structure with the Mn atoms occupying the 4a site with $x = 0.138$. The lack of inversion symmetry of the structure leads to the chiral Dzyaloshiski–Moriya interaction and a helical magnetic structure. The experiments were done on a well characterized [12] single crystalline sample of MnSi with a thickness of 2 mm and a diameter of 20 mm, cut from a large single-crystal grown at Ames Laboratory. The lattice constant is 4.558 Å. The incoming wavelength was 8 Å with a monochromatization of 15% FWHM.

The measurements were carried out at one of the four equivalent magnetic reflections $000 + \vec{\tau}_{111}$, where $\vec{\tau}_{111} = (0.036, 0.036, 0.036)/\sqrt{3}\,\text{Å}^{-1}$. The position sensitive detector overed an angular range of $\sim 3 \times 3^\circ$ around one of the Bragg peaks and the Q resolution was $\sim 0.005\,\text{Å}^{-1}$ FWHM.

As usual in polarimetry we choose the set of orthogonal polarization axes with $\hat{x}$ parallel to $\vec{Q}$, $\hat{z}$ conventionally perpendicular to the scattering plane and $\hat{y}$ completing the right-handed cartesian set [19]. The established measurement strategy is to have the incident and scattered polarization vectors along x, $-x$, y and z and determine the polarization matrix elements.

$$P_{\alpha\beta} = \frac{\tilde{P}_{\alpha\beta}P'_{\alpha} + P^{\dagger}_{\beta}}{|\vec{P}'|} \quad (\alpha,\beta) \in \{x,y,z\} \tag{1}$$

where $\vec{P}'$ is the incident polarization vector, $\tilde{P}$ the polarization tensor and $\vec{P}^{\dagger}$ the polarization created by the scattering process, which in the case of MnSi comes from the chiral term. The MnSi sample was oriented so that $\vec{x}//(1,1,1)$ (i.e. $\vec{Q}$). For this reflection the polarization matrix is

$$\mathbf{P_{MnSi}} = \begin{pmatrix} -1 & \eta\xi & \eta\xi \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{2}$$

where the non-diagonal terms result directly from the chiral character of the helix and provide direct access to the degree of chirality in MnSi. ξ determines the chirality of the helix (+1 for right-handed and -1 for left-handed) and η measures the fraction of the dominant domain ($\eta = 1$ for a single-handed state and $\eta = 0$ for a disordered state e.g. paramagnet or for equally populated chiral domains). For the sake of comparison the polarization tensor for an ideal paramagnet and for nuclear (coherent) scattering are

$$\mathbf{P_{para}} = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \mathbf{P_{nuclear}} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \tag{3}$$

The onset of the helimagnetic phase in MnSi is marked by an intensity jump of more than one order of magnitude within less than 0.1 K. The jump confirms the existence of the first order phase transition seen on the specific heat measurements [20] and with very high accuracy defines the transition temperature $T_C = 29.05 \pm 0.05$ K. The data above and below T_C follow power laws similar to those in Ref. [12] as expected for a second order phase transition. However, this description fails in the close vicinity of T_C and the jump cannot be accounted for by any power law.

With the Cryopad and the polarimetric NSE configuration on IN15 we were able to measure separately the relaxation of the diagonal and of the crossed chiral terms of the polarization matrix given by Eq. (2). The corresponding intermediate scattering functions $I(q,t)$ were obtained by normalizing the spectra against the resolution measured below T_C, typically at 25 K. Fig. 2 shows the relaxation of the crossed term $\mathbf{P}_{zx}$ (i.e. $\vec{P}'\|\hat{z}, \vec{P}\| - \hat{x}$)). The polarimetric NSE configuration enables for the first time an unambiguous separation of the magnetic chiral fluctuations from the "trivial" diagonal part. All NSE configurations, including IMNSE, cannot determine the inelasticity of the non-diagonal terms of the polarization matrix independently from the diagonal ones.

The diagonal and the crossed terms give similar NSE spectra, which follow a simple exponential decay and were fitted by the function $I(q,t) = (1-a)\exp(-t/t_0) + a$, with a being the elastic part and $I(q,\ t\to 0) = 1$. A striking feature of the spectra is that the characteristic time t_0 does not vary with temperature, whereas the elastic background, evolves from ~20% to 100% within 0.2 K following very much the fast increase of the intensity seen in Fig. 2 and masking the decay of $I(Q,t)$ at T_C. A detailed presentation of these results is given in [21].

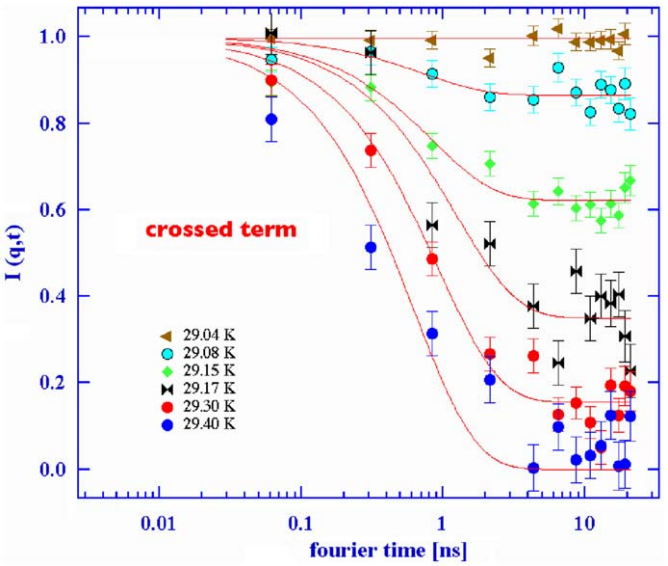

Fig. 2. Intermediate scattering function of the non-diagonal term $\mathbf{P}_{zx}$ (i.e. $\vec{P}'\|\hat{z}, \vec{P}\| - \hat{x}$) measured in polarimetric NSE setup just above the helical transition of MnSi. The lines are fits of an exponential decay superimposed on an elastic background.

4. Conclusion

The first measurements of polarimetric NSE on MnSi show that the polarimetric NSE variant of IMNSE is opens up new opportunities for polarized neutron spectroscopy. We are confident that polarimetric neutron spin echo will be further developed to become the method of choice for a deeper understanding of chiral and other complex magnetic phase transitions. With the development in polarizing neutron optical devices and the onset of the next generation neutron sources, neutron spin echo spectroscopy will enjoy higher fluxes which will also lead to increased flexibility. Even though NSE might never see the richness of echo sequences used in NMR, it seems realistic to conceive that in the near future complex NSE configurations, like IMNSE, may also be used for weakly scattering samples.

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