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Neutron spin-echo on magnetic single crystals in the quantum limit

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Abstract

As interest in low-temperature physics increases, whether for the study of frustrated magnets or for quantum effects, neutron spin-echo will become increasingly important, because of the high-energy resolution achievable. The behaviour of quasielastic scattering for low temperatures relative to the energy scale of interest is investigated. In addition, we note that momentum-transfer selective magnetic scattering may be susceptible to parasitic echoes in certain experimental configurations.

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1. Introduction

Studies of quantum systems and effects must often be carried out at low temperatures. To investigate low energy transfer ranges, one obvious technique is via neutron spin-echo, and this is of interest in studies as diverse as low-energy dynamics, quantum phase transitions and frustrated magnets. One example is our recent study of the quasielastic scattering in the antiferromagnetic-superconducting phase of UPd_2Al_3 [1]. Some other examples are covered in the review by Ehlers [2]. In this paper, we discuss two experimental aspects that were important in the analysis of the data, and which may prove useful to other workers in the field. The first issue is the spin-echo signal when the energy scale of interest is *not* significantly smaller than the sample temperature (subsequently called the quantum limit). The second issue affects magnetic samples when the neutron wavelength spread is limited.

2. Neutron spin-echo at low temperatures

In a neutron spin-echo measurement, the measured quantity is the polarisation at the detector for a given Fourier (de-correlation) time t [3], given by the cosine Fourier transform of the scattering function $S(\mathbf{Q},\omega)$, for momentum transfer $\mathbf{Q}$ and energy transfer $\hbar\omega$:

$$P_{\mathrm{NSE}}(t)=\frac{\int\frac{k'}{k}S(\mathbf{Q},\omega)\cos(\omega t)\,\mathrm{d}\omega}{\int\frac{k'}{k}S(\mathbf{Q},\omega)\,\mathrm{d}\omega},\tag{1}$$

where k' and k are the scattered and incident wavevectors. It is well-known in neutron scattering [4] that $S(\mathbf{Q},\omega)$ may be written as

$$S(\mathbf{Q},\omega)\propto\frac{\chi''(\mathbf{Q},\omega)}{1-\exp\left(-\frac{\hbar\omega}{k_{\mathrm{B}}T}\right)},\tag{2}$$

where $\chi''(\mathbf{Q},\omega)$ is the imaginary part of the response function and the denominator is the detailed balance factor. For quasielastic scattering, this may take the form

$$S(\mathbf{Q},\omega)\propto\frac{\Gamma}{\Gamma^2+\omega^2}\frac{\omega}{1-\exp\left(-\frac{\hbar\omega}{k_{\mathrm{B}}T}\right)},\tag{3}$$

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where $\Gamma(\mathbf{Q})$ is the full-width half-maximum of the quasielastic response in the energy domain (assumed here to be smaller than the incident neutron energy). Mezei [3] evaluated this in the classical limit $k_BT \gg \hbar\omega$, so that $(1-e^{-\hbar\omega/k_BT}) \to \hbar\omega/k_BT$. In this limit $P_{NSE} = e^{-\Gamma t}$.

This result is pleasing as it links the energy-space linewidth to the characteristic decay time directly. However, the definition of the classical limit is somewhat problematic, as the (formal) integrals in Eq. (1) are over all energies. In practice, the observable energy transfers are limited by the incident neutron energy and the extent of χ''. We also note that at high-energy transfers, the approximations linking neutron time-of-flight to energy transfer may break down, and the detector may function nonlinearly.

Below this limit, the energy lineshape is no longer Lorentzian (Fig. 1) and so in the time domain the response is no longer a simple exponential. The detailed balance factor must be fully accounted for. In this regime, the analytical form of $\int (k'/k)S(\mathbf{Q},\omega)\cos(\omega t)\,d\omega$ is not obvious. We have therefore used numerical methods to calculate the observed P_{NSE} for a typical spin-echo measurement.

2.1. Calculation details

Calculations were made assuming that $S(\mathbf{Q},\omega)$ had the form given in Eq. (3). P_{NSE} was then calculated by numerical integration over an energy range of -10 to $+3$ meV ($+3$ meV being a typical incident neutron energy on the IN11 spectrometer at the Institut Laue-Langevin). The cut-off at -10 meV was chosen arbitrarily to be significantly greater than the Γ in the given problem. Increasing the cut-off makes no practical difference to the curves calculated here for $\Gamma = 0.02$ meV. The integral was calculated using a trapezoidal method with energy steps of size (0.0001 meV). For comparison purposes, $S(\mathbf{Q},\omega)$ is also shown in Fig. 1 calculated using the same parameters. The choice of Γ determines the time window of interest. Fig. 1 shows the temperature dependence of P_{NSE} with Γ fixed at 0.02 meV. Even at the highest temperatures, the calculated curve does not agree completely with the simple exponential at small Fourier times, as expected when one considers that $S(\mathbf{Q},\omega)$ is never completely Lorentzian in form.

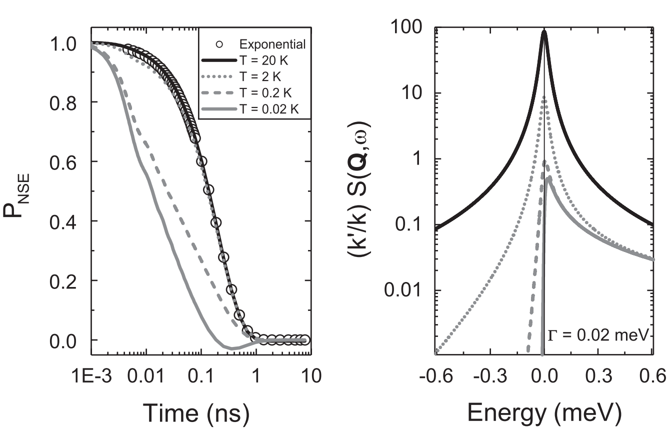

Fig. 1. Left: numerical integration of Eq. (1) for $\Gamma = 0.02$ meV (0.23 K) at a range of temperatures with incident energy 3 meV, as compared to the appropriate simple exponential. Below 0.02 K, there is no observable change in P_{NSE}. Right: $(k'/k)S(\mathbf{Q},\omega)$ from Eq. (3) as a function of energy transfer with $\Gamma = 0.02$ meV. The line $\omega = 0$ is marked. Note the shift in peak position.

Below the high-temperature limit, the curve-shape changes markedly, although all of the dynamics disappear at the same Fourier time. Even at 2 K, which might be considered a 'high' temperature for dynamics with a characteristic energy of 0.02 meV ($=0.23$ K), there is some deviation from the simple exponential.

Changing Γ shifts the curve in the time-domain, and moves the limit between the classical and quantum regimes. The shape of the curve is then dependent on the position of the temperature on this scale. We have found that the system is classical for temperatures two orders of magnitude greater than the system energy scale. Conversely, the time response does not change for temperatures lower than one order of magnitude below the energy scale.

At the lowest temperatures, the curve is negative at times, corresponding to a π phase flip. This is brought about by oscillations, also seen at smaller Fourier times, which are thought to be related to the shift away from 0 meV of the quasielastic peak in the quantum limit [3].

3. Parasitic echoes from magnetic samples

3.1. Observation

In a spin-echo measurement, an oscillation with a particular period is obtained by altering the magnetic field integral, written as ΔB, through which the neutron spin precesses, in prescribed steps. The oscillation's periodicity is fixed by the step size chosen for ΔB. In Ref. [1] an unexpected second modulation was seen in echo groups measured at echo times <36 ps (see Fig. 2). This additional wavelength is approximately half that of the ordinary echo group, and the intensity varies strongly with the echo time (Fig. 3b). The presence of this second modulation distorts data gathered by the usual 4-point method, but this problem is easily detected if several periods of an echogroup are measured (Fig. 3a).

3.2. Explanation

In paramagnetic neutron spin-echo the neutron polarisation component parallel to the scattering vector is flipped at the sample (Fig. 4a), and can be broken down into a component that is effectively π flipped and a part that is anti-parallel relative to the incident polarisation. The π flipped part unwinds in the scattered arm of the spectrometer to give an echo group at the detector as the field integral is changed. The other part does not meet

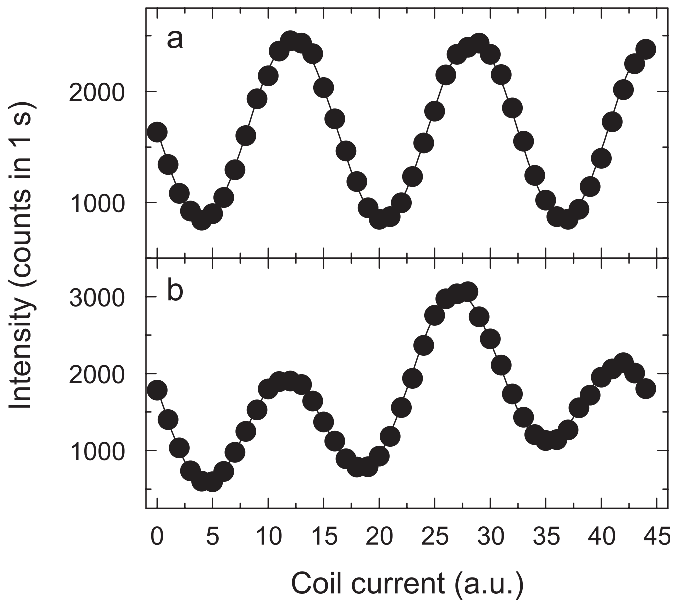

Fig. 2. Sample echo groups measured in a single crystal of UPd_2Al_3 at the magnetic Bragg peak (0 0 0.5), on IN11 (ILL). The coil current corresponds to a change in ΔB. Each point was measured for 5 s, and then normalised to 1 s. Error bars are smaller than the point size. The x-axis is calibrated for the particular Fourier time to give ~3 periods in 45 points: (a) is an echo group measured at 78 ps (one sine wave); and (b) is an echo group measured at 4.7 ps (two sine waves).

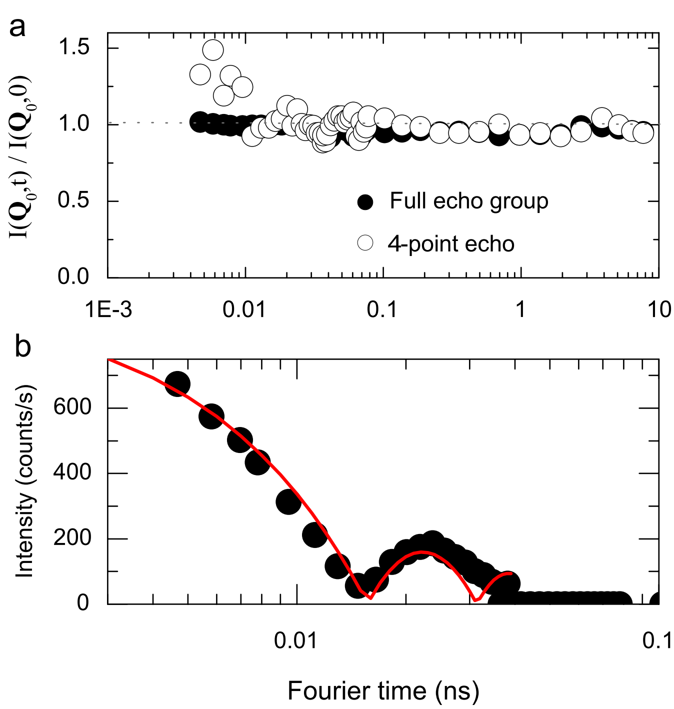

Fig. 3. (a) The momentum and time-dependent intermediate scattering function $I(\mathbf{Q}_0, t)$, normalised to $I(\mathbf{Q}_0, 0)$, of a magnetic Bragg peak in UPd_2Al_3 as a function of Fourier time at 50 mK using the 4-point echo method (open circles) and the multiple-period echo method with the spurious modulation ignored (closed circles). (b) The intensity in counts per second of the second modulation, observed at the magnetic Bragg peak position (0 0 0.5) as a function of Fourier time. The line is described in the text.

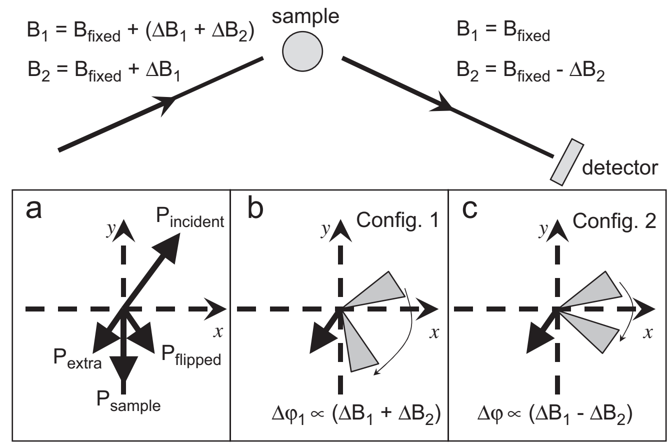

Fig. 4. The upper part shows two spectrometer configurations; ΔB indicates that the magnetic field integral along arms (1, 2) is changed during a measurement: (a) the neutron polarisation precession plane ($y \| \mathbf{Q}$) as seen at the sample. (b) and (c) are the precession plane at the detector for the two different configurations. The grey cones indicate the spread in $\mathbf{P}_{extra}$ at the detector.

the echo condition and is washed out if the frequency distribution of the scattered beam is large enough.

If the scattered beam is tightly monochromated, as from a Bragg peak, or if the detector is set to select a narrow frequency band, the spread of precession angles is limited and the un-flipped portion reaches the detector with an overall non-zero polarisation (see Fig. 4). In the extreme case, the frequency band is a delta function and hence there is a fixed precession angle for a given field integral. This un-flipped portion may vary as the field integral is changed, giving rise to a modulation in the observed signal. In Config. 1 (Fig. 4), this modulation is the same as that of the π-flipped portion of the scattered polarisation. In Config. 2, if $\Delta B_1 = \Delta B_2$ there is no variation, but if the field integral variation is different in the two arms, a second periodicity appears in the echogroup. In our experiment, IN11 was operated in Config. 1 for $t \geqslant 36$ ps, and in Config. 2 for $t < 36$ ps, with $\Delta B_1 = 2.5\Delta B_2$, explaining the sudden disappearance of the second periodicity on changing the setup (Fig. 3b). The ratio of the two observed periodicities $(\Delta B_1 - \Delta B_2/\Delta B_1 + \Delta B_2)$ is 0.429, as compared to the experimentally observed value, 0.43.

The amplitude of this second periodicity is determined by the spread of precession angles at the detector. If the distribution is a δ-function, the second periodicity will have the same amplitude as the expected signal. If the spread of precession angles, $\Delta\phi$, is smaller than 2π, the amplitude will vary as the absolute value of $\int \cos(\phi) D(\phi)\,\mathrm{d}\phi$, where $D(\phi)$ is the neutron wavelength profile translated into the precession angle domain and centered about $\phi = 0$. As the precession angle profile precesses around the circle in the experiment, this centering is necessary to assess the polarisation change correctly.

The line in Fig. 3b was obtained assuming a top hat profile for the neutron wavelength distribution. Its width is the only fitting parameter. The wavelength used was 5.52 Å (as determined from the 2θ angle of the magnetic Bragg peaks), leaving a vertical scaling factor. The wavelength spread for the line in Fig. 3 is 0.068 ± 0.001 Å (as compared to 0.08 Å estimated from the Bragg peak width). This wavelength spread determines the position of the minimum, and corresponds to complete cancellation when the range of precession angles at the detector is close to 2π. The intensity increase observed cannot be reproduced if an extensive neutron wavelength profile (e.g. Lorentzian or Gaussian wavelength profile) is used. Instead, the peaks in the intensity dependence are smoothed out as the range of precession angles covered is larger.

This effect arises when the field integral is varied unequally on the incident and scattered arms when studying a *magnetic* signal, and the effect is most pronounced if the velocity distribution is restricted. It may be signalled by an apparent $I(\mathbf{Q},t)/I(\mathbf{Q},0)>1$ when using the 4-point measurement method. If one wishes to use this technique, it is therefore advisable to make sure that the field integral changes are equally balanced on both arms.

4. Conclusions

The effects of operating in the quantum limit on neutron spin-echo measurements have been explored for quasielastic scattering. This method may also be applied to inelastic scattering as well. The existence of parasitic echoes from magnetic samples is also accounted for and a way to avoid these echoes is described.

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