Neutron Resonance Spin Echo Spectroscopy on Split Modes

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Abstract. We present an analytical formalism investigating effects of multiple excitations with different dispersion parameters in the context of neutron resonance spin echo spectroscopy. This allows identifying conditions to obtain high-resolution information on the energy split of two dispersive modes which may differ or not in their local energy gradient. Comparison to experimental data is provided by recent measurements on magnon excitations in RbMnF\textsubscript{3}. Potential future applications are investigations of hybridized phonon or magnon modes or mixed excitations.

1. Introduction

Neutron resonance spin-echo (NRSE) spectroscopy is a unique experimental tool to measure life times of dispersive elementary excitations over extended regions in the Brillouin zone with resolution in the $\mu$eV range. It is thus natural to explore the application of the technique for resolving modes with a separation in energy which is less than the resolution of standard spectrometers, e.g. triple-axis spectrometers (TAS). However, in this case, the spin-echo conditions cannot be satisfied for both modes simultaneously and it is important to understand the consequence for the echo-amplitude on a quantitative level. Based on an extension of Popovici’s matrix approach for the TAS resolution function [1], we have developed the formalism which can be used for data correction for NRSE-TAS experiments [2, 3]. In this paper we report on the generalization of our earlier treatment now including instrumental parameters not matching the dispersion properties, allowing the local gradient $\nabla q_1(\omega)$ to have an arbitrary direction in wave vector-energy space and apply it to scattering from a second mode contributing to the echo amplitude.

The significance of this work is to identify conditions which allow obtaining high-resolution information on the energy split of two overlapping dispersive modes which may differ or not in their local energy gradient. As a special case mixtures of spin-flip and non-spin-flip scattering and their spin-echo time dependent contribution to the echo signal are included. This is important in life time studies of magnons.
We first consider a simplified model for the echo amplitude in section 2 while the complete model is discussed in section 3. A comparison to NRSE experimental data is presented in section 4.

2. Simplified model for the time-dependence of the echo amplitude

In this section we introduce a simplified model for the echo amplitude as a function of spin echo time which assumes that the spin echo conditions are met for both modes simultaneously. Although this is strictly not possible, the model can be useful in practical cases when the violation of the echo condition is very small or one deals with relatively small spin echo times (compared to quasielastic NSE) which is actually often the case in NRSE spectroscopy leading to negligible depolarization of the detuned mode.

In a minimal model the echo amplitude $A_{E}$ may be described in terms of the Fourier transform of two Lorentzians. Explicitly we can write:

$$A_{E} = \frac{1}{N} \int R(\omega - \omega_{N})S(\omega - \omega_{3})e^{-(\omega-\omega_{3})^{2}/(4h^{2})}d\omega,$$

where $N$ is a normalization factor which ensures that the echo amplitude $A_{E}=1$ at spin echo time $\tau=0$. We allow for the energy resolution of the background TAS as an additional weighting function

$$R(\omega - \omega_{N}) = \exp \left(-\frac{1}{2} \frac{h^{2}(\omega - \omega_{N})^{2}}{\Delta E_{TAS}^{2}} \right),$$

where $\Delta E_{TAS}$ is the FWHM of the TAS energy resolution function which is centered at energy $h\omega_{N}$ and therefore has its maximum at energy $h\omega_{N}$.

Assuming that the structure factor does not vary within the resolution ellipsoid we can factorize the scattering function $S(Q, \omega)$, i.e. the $Q$-dependent part will only contribute to absolute intensity and can be neglected for our purposes. In this minimal model there is no provision for dispersion at all and neither TAS resolution in $Q$-space nor spin-echo related resolution effects are considered. We do however allow for two excitations in that we parameterize

$$S(\omega - \omega_{3}) = A_{1} \frac{\Gamma_{1}}{\Gamma_{1}^{2} + (\omega - \omega_{3})^{2}} + A_{2} \frac{\Gamma_{2}}{\Gamma_{2}^{2} + (\omega - \omega_{3} - \Delta \Omega)^{2}},$$

Here $A_{1,2}$ are amplitudes which determine the relative intensity weight of the two Lorentzians with line widths given as HWHM $\Gamma_{1,2}$. $\Delta \Omega$ is the energy separation of the two modes. We have explicitly introduced $\omega_{3}$ expressing the fact that we consider the scattering function relative to the first excitation mode which has the energy $h\omega_{3}$. Strictly speaking Eq. (3) violates the detailed balance condition for $S(Q, \omega)$. Here we implicitly assume $\omega > 0$ and zero temperature which allows neglecting the anti-Stokes term.

For inelastic scattering the spin echo phase (total Larmor precession phase $\phi$ at the detector) will have a constant term $N\tau\omega_{N}$. This term depends on instrument parameters and is fixed by the spin echo condition. As a matter of fact $h\omega_{N}$ is just the mode energy $h\omega_{3}$ if the apparatus is tuned to this mode.

With the variable transform $\omega = \omega_{3} + \Delta \omega$, where $\Delta \omega$ is the energy deviation from the mode energy of the first Lorentzian in Eq. (3), the echo amplitude reduces to

$$A_{E} = \frac{1}{N} \int R(\Delta \omega - \omega_{N} + \omega_{3})S(\Delta \omega)e^{-(\Delta \omega)^{2}/(4h^{2})}d\Delta \omega.$$
Fig. 1 shows the scattering function (Eq. (3)), the scattering function multiplied and convoluted with the TAS energy resolution (Eq. (2)) as well as the corresponding Fourier transforms. An approximate analytic expression of the echo amplitude in case the line widths are identical is given by an exponential decay modulated by a cosine term
\[ A_e = \left| A_{m} \right| + \left| A_{m} \right| \cos \left( \frac{2 \pi \tau}{T} \right) \exp \left( - \frac{\tau}{\hbar} \right), \] (5)
where the period \( T \) of the modulation is inversely proportional to the energy difference
\[ T = \frac{4 \pi \hbar}{\Delta \Omega}, \] (6)
and the amplitude of the modulation is determined by the intensity ratio of the modes \( A_1/A_2 \)
\[ A_{m} = \frac{A_1 - A_2}{A_1 + A_2}, \quad A_1' = A_1 R(- \omega_f), \quad A_2' = A_2 R(\Delta \Omega - \omega_f). \] (7)

We note that in this minimal NRSE model the amplitude parameters should not only reflect the scattering probability but also contain the information about the fraction of the signal which gives rise to a polarized signal at the detector. Two limiting cases are immediately obvious: If the spin-echo conditions are strongly violated for the second mode there will be an increase of depolarized background independent of spin-echo time and \( A_2 \) can be set to zero. Provided that the signal from the second mode is completely depolarized at the smallest experimentally accessible spin-echo time [4] the only effect to be considered is that the initial echo amplitude at \( \tau=0 \) will be less than 1. The other limiting case is when the spin-echo conditions are perfectly satisfied for both modes and thus the modulation of the signal extends to large spin-echo times. In the latter case the amplitudes \( A_{1,2} \) are determined by the structure factor and the energy resolution function only. Any intermediate case requires going beyond the simple model.

**3. Extended model for the time-dependence of the echo amplitude**

Next we outline the main generalizations required to attain a full model which explicitly takes into account the decaying contribution of the detuned mode. An earlier treatment of the resolution function [3] assumed that the spin-echo conditions are perfectly satisfied and that the TAS resolution ellipsoid coincides with the centre of the dispersion. We have now derived corresponding analytical expressions giving up this restriction. In addition we allow for instrumental parameters not satisfying...
the spin-echo conditions for inelastic, dispersive NSE, and two dispersion surfaces with different local gradients in wave-vector-energy space. As a starting point we always express the echo amplitude using the Larmor phase \( \phi \) as a function of the incident and final wave vectors \( k_i, f \).

\[
A_E = \frac{1}{N} \int R(k_i, k_f) S(Q, \omega) e^{i\phi(k_i, k_f)} d^3k_i d^3k_f.
\]  

(8)

Larmor phase, dispersion relation and TAS resolution function are expanded to second order which allows using a covariance matrix formulation of the resolution problem.

3.1. Violated spin-echo conditions

For brevity we outline the result for the functional form of the echo amplitude assuming a single dispersion branch but violated spin-echo conditions. A full derivation will be given elsewhere [5]. We find that instrumental parameters introduce terms in the Larmor precession phase which are linear in the wave-vector variables \( k_i, k_f \). This implies a Gaussian decay of the echo amplitude on top of the usual second-order resolution contribution

\[
A_E = \left[ \frac{\det \bar{L}(\tau^\prime = 0)}{\det \bar{L}(\tau^\prime)} \right] \exp \left( -\frac{1}{2} \xi \kappa^2 \right).
\]  

(9)

Here \( \tau^\prime \) is a reduced spin-echo time, i.e. the usual spin-echo time but modified by an additional factor introduced by the detuning, and \( \kappa \) is essentially a linear transform of the inverse of the complex resolution matrix \( \bar{L} \) [2] providing an effective width for the Gaussian. Detuning curves of individual instrument parameters (Fig. 2) are readily obtained from the present formalism and provide an estimate on the sensitivity of the experiment with respect to instrumental detuning which may be due to limited knowledge of the dispersion parameters.

![Detuning curves of individual instrument parameters](image)

Fig. 2: Detuning of the tilt angles of the RF coils \( \theta_1, 2 \) and the frequencies \( f_1, 2 \) of the RF field in first and second spectrometer arm evaluated at spin echo time \( \tau = 15 \) ps.

3.2. Second dispersion surface
For the functional form of the echo amplitude assuming two dispersion branches, one perfectly tuned, the other violating the spin-echo conditions we obtain for the echo amplitude

\[ A_E = \frac{1}{N} \left| \int S_1(\Delta \omega) e^{-i \tau,1,\Delta \omega} d\Delta \omega + \exp \left( T L T^{-1} \right) \int S_2(\Delta \omega) e^{-i \tau,2,\Delta \omega} d\Delta \omega \right|. \]  

(10)

This result is very similar to Eq. (4) since to good approximation \( \tau,1, \approx \tau,2, \). However the integral over the second Lorentzian has now an additional complex prefactor

\[ f_p = \exp \left( T L T^{-1} \right) \]  

(11)

allowing to estimate to what extent the second dispersion branch contributes to the echo amplitude. Here the column vector \( T \) is linear in the spin-echo time and thus the prefactor \( f_p \) leads to a Gaussian damping of the modulation. In the limit of satisfied spin echo conditions \( T \) vanishes.

Fig. 3 shows the model scattering function multiplied and convoluted with the TAS energy resolution as well as the corresponding Fourier transforms now explicitly considering the spin-echo-time-dependent prefactor \( f_p \).

We note that a special case of the detuning problem is realized investigating magnon excitations with non-spin-flip scattering while the second precession field region has a polarity adapted to spin-flip scattering. This case is covered assuming that the RF frequency \( f_2 \) is detuned to \(-f_2\). All other parameters stay tuned.

4. Comparison to experimental data

The case of a second dispersion branch within the background spectrometer resolution has been experimentally realized in recently performed NRSE measurements at TRISP, FRM II, with a particular sample of the simple cubic Heisenberg antiferromagnet RbMnF\(_3\) consisting of two grains of comparable size. This provides an ideal ground for checking the model against experimental data since the second dispersion surface is identical to the first and its position in wave-vector-energy-space is easily obtained from the experiment. The spin wave dispersion of magnons in RbMnF\(_3\) is known in sufficient detail [6]. The relative orientation of the two crystallites has been obtained by standard procedures maximizing the intensity of two Bragg peaks for each of the two grains. Applying an extension of the original UB matrix formalism [7] to inelastic scattering following Lumsden et al. [8]
we obtain the peak positions of the inelastic signal in good agreement with experimental data collected
on the thermal TAS PUMA, FRM II (see Fig. 4 for an example).

![Graph showing experimental peak positions compared to calculated positions.]

**Fig. 4:** Experimental peak positions (data points) arising from the double dispersion surface compared to calculated peak positions indicated by the vertical lines. Note that these two scans are at wave vectors [0.75 0.75 -0.25] (left panel) and [0.75 0.75 +0.25] (right panel) where a clear separation of the two modes is observed. There are however positions in Q-space where the split is unnoticeable within standard TAS resolution.

The experimental NRSE data for the zone boundary magnon at \( \mathbf{Q} = [0.5 0.5 -1] \) and \( E = 8.46 \) meV has been fit to the simple model Eq. (5) (see Fig. 5). The energy split from the fit was found to be 0.420(19) meV at a sample temperature \( T_S = 3.18 \) K and 0.414(6) meV at \( T_S = 7.75 \) K. This is in reasonable agreement with the calculated energy split of 0.464 meV. Also shown in Fig. 5 is the time dependence applying the full model Eq. (10) found to be in agreement with the NRSE data within statistical error. In order to fit the energy split using the full model, more data points are required than available. This inadequacy is due to the fact that the original aim of the spin-echo experiment was actually not to determine the energy split, but rather the line width of the mode.

![Graph showing NRSE data and fit.]

**Fig. 5:** Experimental NRSE data measured on the zone boundary magnon \( \mathbf{Q} = [0.5 0.5 -1], E = 8.46 \) meV, \( T_S = 3.18 \) K. The dashed blue line is a fit of Eq. (5) to the data. The solid black line is the echo amplitude calculated according to Eq. (10).

Finally we note that in the particular experiment on RbMnF\(_3\) magnons our model allows identifying those regions in the Brillouin zone where the line-width data is affected by the second grain. Two conditions must be met for spurious effects to be significant: The energy separation of the two dispersion surfaces must be less than the FWHM of the background TAS resolution and the magnitude of the complex prefactor must be less than 50% at the smallest experimentally accessible spin echo time used in the data analysis. It is found that a precise measurement of the line width is unaffected by the second grain in most of the Brillouin zone except for small regions close to the zone boundary.
5. Conclusion and Outlook
The present analytical formalism allows investigating effects of multiple excitations with different dispersion parameters in the context of NRSE spectroscopy. The significance of this work is to identify conditions which allow obtaining high-resolution information on the energy split of two overlapping dispersive modes which in general may differ in their local energy gradient. Potential future applications may be seen for investigations of hybridized phonon or magnon modes or mixed excitations. More experimental work is clearly needed to establish the method when applied to split modes.

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