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A resolution model for mode multiplets probed with neutron resonance spin-echo spectroscopy

Felix Groitl, Klaus Kiefer, Klaus Habicht *

Helmholtz Zentrum Berlin, Hahn-Meitner-Platz 1, D-14109 Berlin, Germany

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ABSTRACT

A resolution model, which takes into account a violation of the spin-echo conditions for inelastic scattering as it is appropriate for high resolution spin-echo measurements of mode multiplets, is experimentally tested. Phase-sensitive measurements were performed while a dispersion surface is moved through the resolution ellipsoid of a triple-axis background spectrometer. The results are found to be in agreement with the model predictions. A tunable, artificially split dispersion was realized with a double crystal setup mounted on a piezoelectric device allowing for rotation and tilt of the first crystal, while the second one was rigidly fixed. Elastic measurements with the instrument configured in Larmor diffraction geometry were used to probe the crystal orientation with high sensitivity. First inelastic measurements are in agreement with a simplified model indicating persistence of the echo modulation over the entire spin-echo time range probed by the experiment.

1. Introduction

Nowadays neutron resonance spin-echo (NRSE) spectroscopy on triple-axis spectrometers (TAS) is a well established neutron scattering technique for measuring the lifetimes of elementary excitations over extended regions of the Brillouin zone [1–3]. The high resolution of the method potentially allows for separating excitations in energy which are unresolved by standard neutron scattering techniques. Possible applications are hybridized magnon–phonon modes which are prominent in the emerging class of multiferroics [4,5] or excitations with small energy separations, which are found in orbital Peierls systems [6]. Prior to application of the method to complex systems with interesting physical properties is a basic understanding of the potential of the method. Despite the high resolution of the spin-echo technique, exact phonon focussing required for inelastic, dispersive elementary excitations cannot be achieved for two or more modes simultaneously hence this would introduce unavoidable depolarization effects. Thus it is of considerable interest to understand under what experimental conditions the signature of split modes, a modulation of the echo amplitude, can be identified. A model which accounts for the violation of the spin-echo conditions for inelastic scattering is currently developed [7]. Although the second order expansion of the total Larmor precession phase can conveniently be treated within a matrix formulation [8], the full model is complex and experimental verification is desirable. Here we report on selected experimental tests to check this model. Both echo-phase and echo-amplitude sensitive measurements were performed on inelastic excitations. Accompanying elastic measurements with a mosaic-sensitive Larmor-diffraction type of NRSE setup on split elastic peaks demonstrate echo modulation in the spin-echo *length* domain and allow for an independent accurate calibration of the parameters which determine the dispersion splitting.

2. Resolution models

2.1. Simple models for modulated echo amplitudes

In the simplest approach we neglect depolarization effects due to the violation of the spin-echo conditions. We assume that the echo amplitude A_E is the Fourier transform of two peak functions with identical width, but different peak amplitudes A_1, A_2. The different amplitudes take into account intrinsic effects and the fact that the scattering function is weighted by the resolution function of the background spectrometer. The amplitude difference defines a contrast $C=1-A_M$ with

$$A_M = \left|\frac{A_1-A_2}{A_1+A_2}\right|. \quad (1)$$

In Ref. [7] we give an approximate analytic expression of the echo amplitude as a function of the spin-echo time τ for the case of two Lorentzians with line width Γ. An exponential decay modulated by a cosine squared term adequately describes the modulation. Here we present a model for the elastic case applicable to Gaussian mosaic distributions of two Bragg peaks rotated against

* Corresponding author. Tel.: +49 30 806242807; fax: +49 30 806243094.
E-mail address: habicht@helmholtz-berlin.de (K. Habicht).

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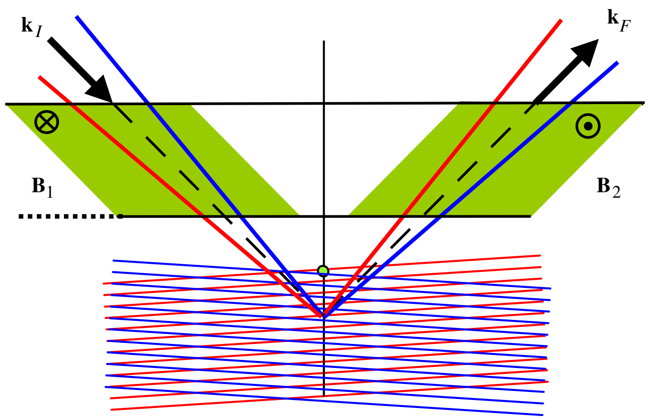

Fig. 1. Sketch of the mosaicity-sensitive Larmor diffraction geometry. The lattice planes of the two crystals (thin blue and red lines) are tilted w.r.t. each other. Neutrons acquire a different total Larmor precession angle as their paths (thick blue and red lines) depends on which crystal they scattered from. The NRSE coils are parallel and the effective magnetic field regions (green) before and after the sample have an antiparallel orientation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

each other by an angle $\Delta\Omega$ in the scattering plane (as applied in Section 3.2.1). The corresponding mosaicity-sensitive Larmor diffraction geometry is depicted in Fig. 1. We can approximate the echo amplitude of the exact Fourier transform with a Gaussian modulated with a cosine term:

$$A_E = \left| A_M + (1-A_M)\cos\left(2\pi\frac{\delta}{\Delta}\right)\right| \exp\left(-4\ln 2\frac{\delta^2}{w_S^2}\right), \quad (2)$$

where δ is the spin-echo length, identical to the van-Hove correlation length. The period of the modulation Δ is inversely proportional to the magnitude of the reciprocal lattice vector Q_B of the Bragg peak considered and the rotation angle $\Delta\omega$

$$\Delta = \frac{4\pi}{Q_B \Delta\omega}. \quad (3)$$

The FWHM w_S of the distribution function in correlation length may be obtained from the FWHM of the sample mosaic η_S:

$$w_S = \frac{2\pi}{Q_B \eta_S}. \quad (4)$$

In an NRSE setup with a mosaicity-sensitive Larmor diffraction geometry the spin-echo length δ is given by

$$\delta = 4\pi\frac{m}{\hbar}\frac{L\tan\theta}{k_I} f_{eff}\frac{1}{Q_B}, \quad (5)$$

where L is the distance between the RF coils operated with effective frequencies f_{eff} which are tilted by angles θ in opposite directions. k_I is the magnitude of the neutron wave vector.

Note that in Eq. (2) we take the modulus of the whole prefactor. This is crucial for fitting the elastic data of Fig. 4 as well as the inelastic data of Fig. 4. For the fit in Ref. [7] we removed the sign of the cosine by using cosine squared to obtain an adequate fit of the data since we were dealing with a small modulation A_M.

In the inelastic case for higher spin-echo times violation of the spin-echo conditions generally leads to depolarization effects and eventually the decay of detuned modes resulting in a time dependence of the contrast has to be considered.

2.2. Extended model

Previously, depolarization effects arising from a curvature of the dispersion surface, mosaic samples and field integral variations due to neutron trajectories for a given tilted precession field instrument geometry have been treated by a matrix formalism [8,9]. Two basic assumptions made here, i.e. (1) satisfied spin-echo conditions and (2) gradient of the dispersion always being parallel to the wave vector of the excitation, were given up leading to a more general model which is currently developed and tested. A full description of the formalism is beyond the scope of this paper. Here we merely summarize important cases captured by the formalism. Linear terms (other than energy transfer) are included in the total Larmor precession phase. It is worth to emphasize that a rocking scan on a dispersive excitation allows an experimental determination of the slope of the dispersion for transverse modes in a phase-sensitive NRSE measurement. Detuning of any instrumental parameter has now been traced up to second order and predicts an additional phase of the spin-echo signal than a first-order treatment. A Gaussian damping of the modulation is obtained if one mode satisfies the echo conditions while a second mode is detuned. Thus the amplitude of the modulation of the signal is a function of the spin echo time. On the other hand the apparatus can be tuned in a way that the spin-echo conditions for two modes are violated by comparable detuning. Here, a Gaussian decay of both modes is to be expected, but the modulation contrast remains essentially time independent. It is possible to extend the model to more than two excitations, although no more than 3 modes are practical for NRSE experiments. The overall aim is to predict these effects quantitatively and to provide a fit model for experimental data on multiple modes. Confidence in the details and the predictive power of the formalism is provided by experiments on simple, well understood model systems. In the below sections, we will discuss two of these experiments.

3. Experiments

3.1. Phase-sensitive NRSE measurements on a single dispersion

If the spin-echo parameters remain fixed and the sample is rotated in an inelastic NRSE experiment on a TAS, the dispersion surface is effectively moved through the fixed resolution ellipsoid of the background spectrometer. In other words, the spin echo probes different portions of the dispersion, and the result is a decay of the polarization, as the apparatus is no longer tuned to the excitation, and a phase shift. Since the intensity also changes with moving the dispersion of the transverse mode, the only way to experimentally access the phase is to record a full spin-echo curve for each rotation angle of the sample. We performed this experiment using the transverse acoustic phonon (2 0.1 0) in elemental Pb with an energy $E=0.97$ meV at a fixed incident wave vector $k_I=1.7$ Å^{-1} with the NRSE option available at the cold neutron TAS V2/FLEX at the BER-II of HZB, Berlin. The sample was kept at a temperature of $T=100$ K to enhance the intrinsic lifetime. At $T>100$ K the intrinsic lifetime declines due to phonon–phonon interaction, while at $T<100$ K the inelastic signal is reduced by the Bose factor.

The data shown in Fig. 2 were collected at spin-echo time $\tau=20$ ps. The total accumulated Larmor phase was calculated as a function of the rocking angle using the extended model. The experimental data are in full agreement with our model and display the non-linear behaviour of the phase as predicted. In a simplified approach (restricted to first-order expansion of the Larmor phase) the linear coefficient of the phase dependence allows us to extract the slope of the dispersion $d\omega/dq$. By a linear fit we obtain $d\omega/dq=6.42(14)$ meV Å, which is significantly lower than the value of 7.43 meV Å extracted from a full force-constant parameterization of the dispersion including three-body interactions [10] fed into our model. This demonstrates that higher order terms in the Larmor phase are generally significant and important to consider.