Experimental report

24/01/2024

Proposal: **4-01-1791**

Council: 4/2023

Title: Investigating the spin-lattice coupling in the Ge spinel compound $GeCo_2O_4$

Research area: Physics

This proposal is a new proposal

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Samples: $GeCo_2O_4$

Instrument	Requested days	Allocated days	From	To
IN12	7	0		
THALES	7	7	03/11/2023	10/11/2023

Abstract:

We propose to investigate the spin-lattice coupling in the spinel compound $GeCo_2O_4$ by studying the possible presence of hybrid excitation modes. Indeed a previous inelastic study of this compound reveal an acoustic phonon mode crossing multiple magnon branches in an energy range between 3 and 6 meV. For this study, we require the use of polarisation analysis in order to properly separate the phonon and magnon contributions in the complex excitation spectrum, as well as check for a sizable nuclear-magnetic interference term in the polarisation matrix.

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THALES (7 days from 2023/11/03 to 2023/11/10)

Introduction

GeM_2O_4 (M = Fe, Co and Ni) are cubic spinels crystallizing in the space group Fd-3m at room temperature. The magnetic ions are located on a network of corner-sharing tetrahedra, namely a pyrochlore lattice, favoring magnetic frustration and complex magnetic ground states [1-3]. In this work, we focus on $GeCo_2O_4$, which orders antiferromagnetically at T_N = 23.5 K with a cubic to tetragonal structural transition [2,3]. The low temperature magnetic structure is characterized by a propagation vector $\boldsymbol{k}$ = (1/2 1/2 1/2) and corresponds to alternating kagome and triangular ferromagnetic planes perpendicular to the <111> direction, with spins aligned along the [1 1 -2] direction [3]. Moreover, elastic constant studies using ultrasound velocity measurements also reported acoustic anomalies (in particular, in the *C44* elastic constant related to the transverse acoustic phonons) at the magnetic transition, and possibly associated to the structural distortion, thus suggesting a coupling between the magnetic excitations and the acoustic phonons [4]. The goal of our experiment is to investigate the presence of a possible spin-lattice coupling in $GeCo_2O_4$ and shed light on the microscopic mechanisms leading to the magnetic ordering in this compound.

Experimental details

We have performed longitudinal polarization analysis on THALES with fixed final energy E_f = 4.7 meV (k_f = 1.5 Å^{-1}). The large single crystal of $GeCo_2O_4$ was oriented in order to have (H H L) as the horizontal scattering plane. Our previous measurements on cold time-of-flight spectrometer IN5 showed a complex excitation spectrum near (2 2 2) (Fig. 4), where acoustic phonons are crossing gapped spin waves above 3 meV (Fig. 4). A spin-lattice hybrid mode may occur at the crossing between the phonon mode and a spin wave branch and may induce a gap between the two modes at this point, as previously observed in $YMnO_3$ [5,6]. We are therefore investigating this possible hybridization scenario using polarized neutron in $GeCo_2O_4$. The purpose was to separate the nuclear and magnetic contributions around this region of reciprocal space and energy transfer and possibly reveal a spin-lattice hybrid mode.

Results

The spin-flip (SF) and non-spin-flip (NSF) channels were measured with the neutrons polarized along x (parallel to the scattering vector $\boldsymbol{Q}$), using the outgoing flipper (F2). In the regions of our scans, the magnetic intensity is very strong, so the SF channel is dominating over the NSF channel. The NSF intensities were therefore corrected by removing the leak from the SF channel, by considering a flipping ratio R = 15, measured on the incoherent scattering of a quartz standard for the same experimental configuration.

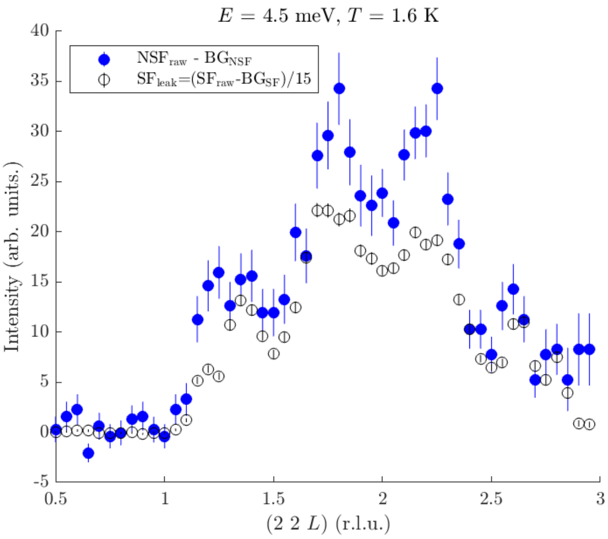

Fig. 1. Constant-E scan, the backgrounds are obtained by fitting SF and NSF raw intensities to gaussians, and the SF leak is calculated considering R =15.

An example of this correction is shown in Fig. 1. Both SF and NSF raw intensities are fitted to several gaussians to subtract the background from both channels. Then, the SF leak is calculated by dividing the SF intensities (with background subtracted) by R = 15, and subtracted to the NSF intensities. Constant-E scans along (2 2 L) are shown in Fig. 2, with the corrected intensities as described above.

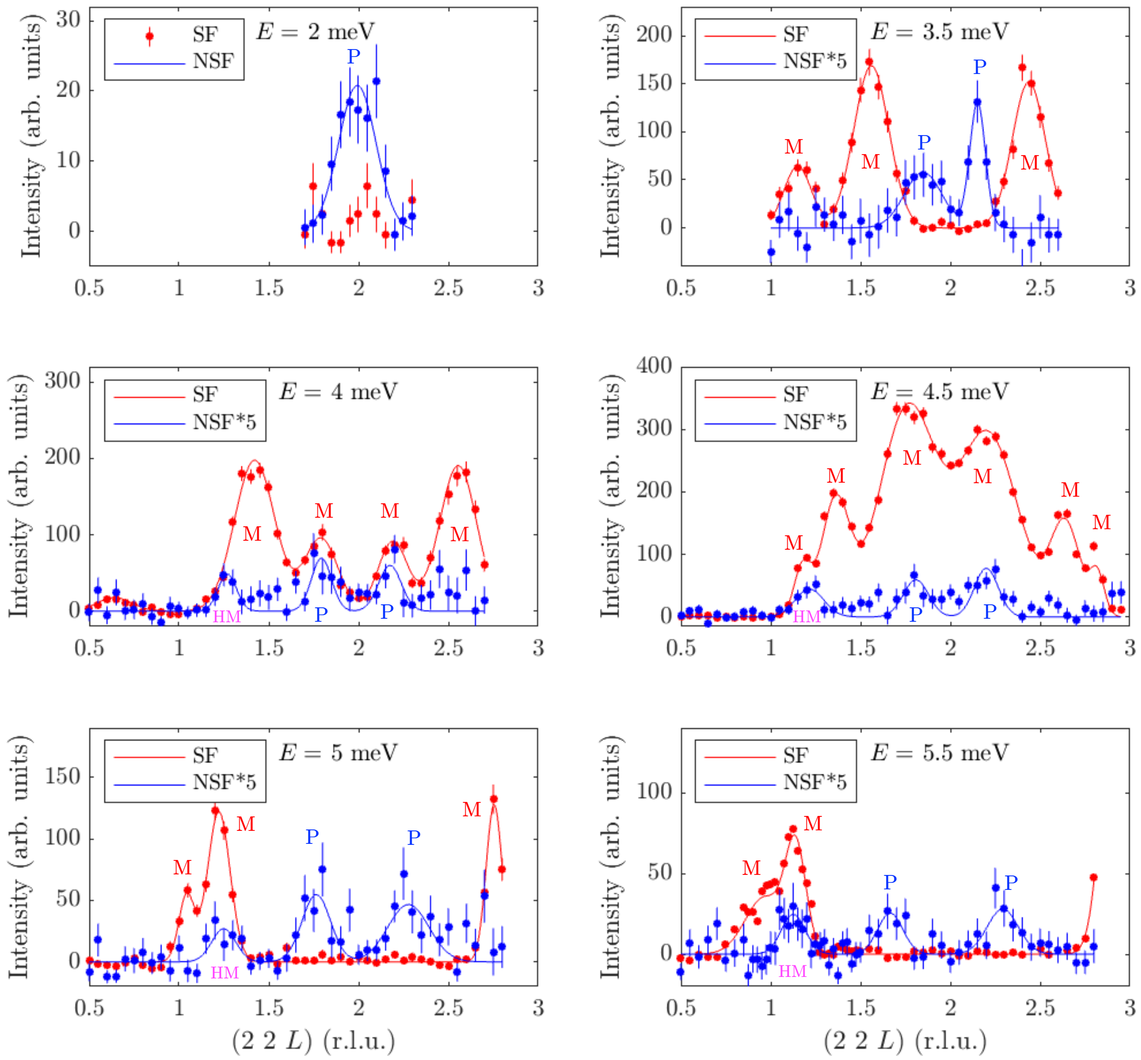

Fig. 2. Constant-E scans showing SF (red) and corrected NSF (blue) intensities (data points). The NSF intensities are rescaled for a visual comparison with the SF intensities. Fits to gaussians are shown in continuous line. The labels indicate the nature of the modes: (M) magnon, (P) phonon, (HM) hybrid mode.

As expected from our previous IN5 measurements, the phonon mode is seen around L = 1.8 in the NSF channel, centered at same position of a magnon mode for E = 4 meV and E = 4.5 meV in the SF channel. More strikingly, an additional mode is observed around L = 1.2 from E = 4 meV to E = 5.5 meV in the NSF channel.

This novel mode was followed at different temperatures along (2 2 L) and E = 4.5 meV. As shown in Fig. 3, it disappears above T_N = 23.5 K, which indicates its link to the magnetic ordering. We note that strong paramagnetic signal is still observed in the SF channel until 100 K. Also, we were geometrically limited to

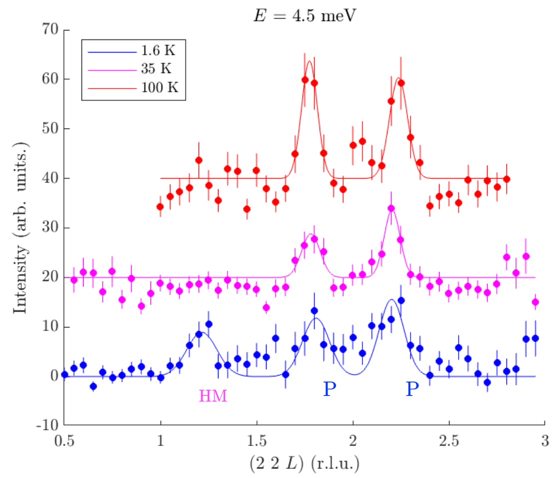

Fig. 3. Temperature dependence of the NSF corrected intensities for (2 2 L) scan at E = 4.5 meV. The labels indicate the nature of the modes: (P) phonon, (HM) hybrid mode.

go to higher-Q, and could not clearly observe the symmetric mode with respect to (2 2 2), i.e. (2 2 2.8) for (2 2 1.2). Still, at 4.5 meV, the presence of this mode is indicated by the rise of intensity near $L = 3$ (as shown in the blue curve in Fig. 3), with a potential shift due to the instrumental resolution.

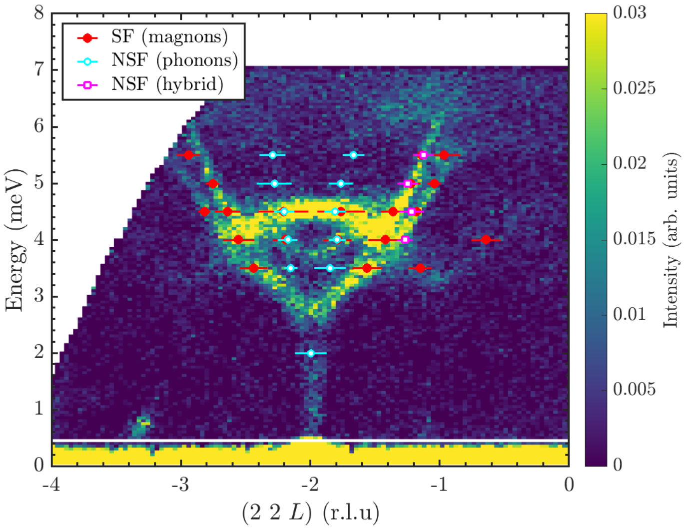

Fig. 4. The centers of the fitted gaussians from the constant E-scans are mapped on slices measured on IN5. The error bars indicate the full-width half maximum of the fitted gaussians.

The centers of the fitted gaussians from Fig. 2 are mapped on IN5 data in Fig. 4, where the error bars indicate the full-width half maximum of the gaussians, to give an idea of the instrumental resolution. In the SF channel, the mixture of magnon modes is hard to distinguish below 4.5 meV. While in the NSF channel, the acoustic phonon modes are clearly observed. The dispersion of the hybrid mode is difficult to understand, as it seems to follow the upper magnon branch above 5 meV and the lower one below 4.5 meV. More scans at intermediate energies would be necessary to figure out the dispersion of this hybrid mode and understand the underlying mechanism.

Given the weak intensity of the phonon along (2 2 L) and therefore the hybrid mode, we have tried some scans along (H H 2) and (H H H). We note that the statistics were lower for these scans given less counting time. Fig. 5 shows that the hybrid mode may be seen along (H H 2) at 4.5 meV. However, the NSF intensities are not stronger along these directions.

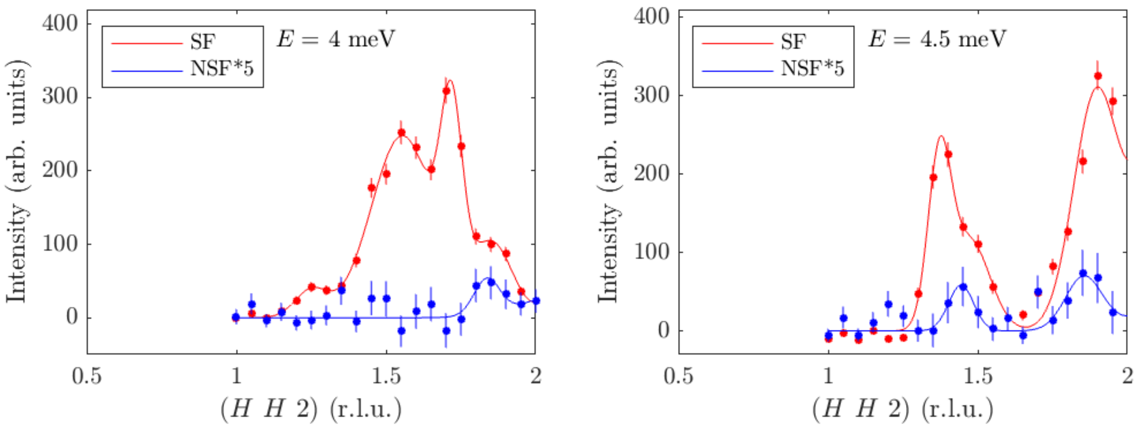

Fig. 5. Constant-E scans along (H H 2)

To conclude our polarized neutron data revealed the presence of an unexpected NSF signal that follows the dispersion of a magnetic excitations. This NSF signal disappears at T_N suggesting its link to the magnetic ordering. To go further, it would be interesting to probe this NSF signal at another reciprocal space position such as close to (4 4 0), where the phonon is strong with still a strong magnetic signal (as measured with unpolarized neutrons on IN5 previously). Given the hybrid character of the observed mode, its intensity would be expected to be stronger, which may make its observation easier for future experiments. A closer look of the nuclear-magnetic interference terms around this mode is also necessary along with a complete temperature dependence to correlate the apparition of this signal to the onset of the magnetic ordering.

[1] S. Diaz et al., Phys. Rev. B 74, 092404 (2006)
[2] P. T. Barton et al. Phys. Rev. B 90, 064105 (2014)
[3] X. Fabrèges et al., Phys. Rev. B 95, 014428 (2017)
[4] T. Watanabe et al., Phys. Rev. B 84, 020409(R) (2011)
[5] S. Petit et al., Phys. Rev. Lett. 99, 266604 (2007)
[6] S. Pailhès et al., Phys. Rev. B 79, 134409 (2009)