Experimental report

23/03/2021

Proposal: **TEST-2965** **Council:** 4/2018

Title: Spin waves in GeCo2O4

Research area:

This proposal is a new proposal

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Rafik BALLOU

Local contacts: Jacques OLLIVIER

Samples: GeCo2O4

Instrument	Requested days	Allocated days	From	To
IN5	5	5	21/09/2018	26/09/2018

Abstract:

EXPERIMENTAL REPORT

EXPERIMENTS N° TEST-2965, TEST-2935 INSTRUMENT: IN5

DATES OF EXPERIMENT 21/09/2018 → 26/09/2018, 6/09/2018 → 11/09/2018

TITLE:

Spinwaves in the spinel compounds $GeNi_2O_4$ and $GeCo_2O_4$

LOCAL CONTACT: Jacques OLLIVIER

EXPERIMENTAL TEAM: (names and affiliation)

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Abstract

We propose to measure the spinwaves spectrum of the spinel compounds $GeNi_2O_4$ and $GeCo_2O_4$ on the time-of-flight spectrometer IN5. We thus intend to determine the Hamiltonian of these materials, including magnetic interactions up to the 3rd neighbors and possible anisotropy terms. This should give more insight into frustration mechanisms at play and the possibility of an original hedgehog-like triple-k magnetic structure that we have predicted through calculations.

Summary of the results:

Spinel compounds of generic formula AB_2O_4 crystallize in the cubic space group Fd-3m. The B sites can accommodate a magnetic ion, which form a pyrochlore lattice, the archetype of frustrated network consisting of tetrahedral joined by their vertices. The Ge spinels compounds GeM_2O_4 (M=Co, Ni, Fe) are particularly interesting as their complex ordering is expected to arise from competing interactions beyond the third neighbours [1,5] (see Figure 1 (b)).

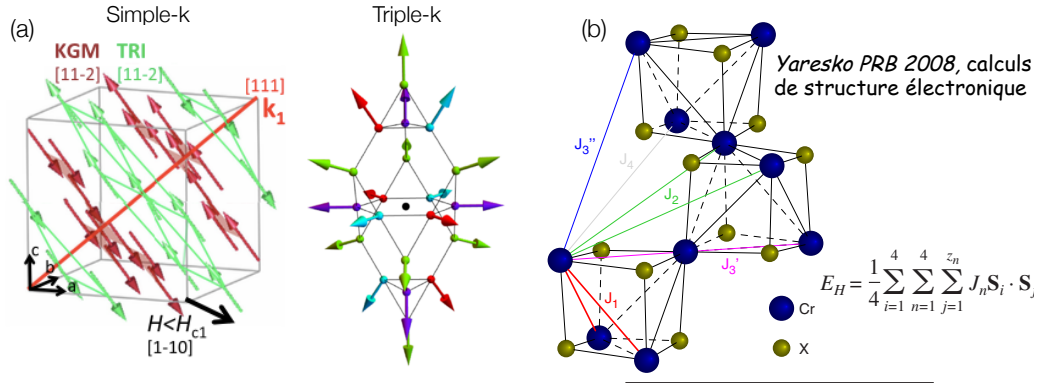

Figure 1: (a) Right: simple-k picture proposed from single-crystal neutron diffraction in $GeCo_2O_4$ [4]. Left: calculated hedgehog triple-k magnetic structure. (b) Illustration of the magnetic interactions up to the 4th neighbors (from [5]).

The magnetic order of the Ni and Co compounds has been investigated by neutron diffraction and is characterized by a k=(1/2, 1/2, 1/2) propagation vector [1]. The AFM long-range order occurs at T_N = 23.5 K for the Co compound and in two steps, at T_{N1} = 12 K and T_{N2} = 11.5 K, for the Ni compound. In a simple-k picture, the magnetic structure can be described as alternating kagome (KGM) and triangular (TRI) ferromagnetic (FM) planes, perpendicular to the <111> direction associated with the propagation vector, and antiferromagnetically coupled to the nearest planes of the same kind [1,2,3,4] (see Figure 1 (a)). However, the magnetic order could as well be described by several Fourier components associated with different members of the star of the k vector.

We have performed calculations, combining a Fourier space analysis to find the propagation vector(s) and a real space spin configuration minimization at zero temperature, in order to elucidate the phase diagram of these interesting materials. These calculations were performed with magnetic exchange interactions up to the 3rd neighbours. Under some conditions, a triple-k structure is stabilized which features a lattice of 3-dimensional topological hedgehog-like spin configurations (see Figure 1 (a)), not discernable in neutron diffraction from the simple-k structure with k=(1/2, 1/2, 1/2) described above. We think that this remarkable triple-k structure could be stabilized in particular in $GeNi_2O_4$ since it retains the cubic structure at low temperature contrary to the Co sample that presents a structural distortion [2,4]. In the latter, the frustration could indeed be partly released through magnetostructural effects leading to the 1-k magnetic structure already reported [4].

To get a further insight into the Hamiltonian of these materials, we have measured the spinwave excitations on two large single-crystals of $GeNi_2O_4$ and $GeCo_2O_4$ using the time-of-flight spectrometer IN5. The chosen horizontal scattering plane was (hh0), (00l) and the samples were measured at several wavelengths (2, 3, 4.35 Å) at the base temperature of an orange cryostat and in the paramagnetic state just above the T_N.

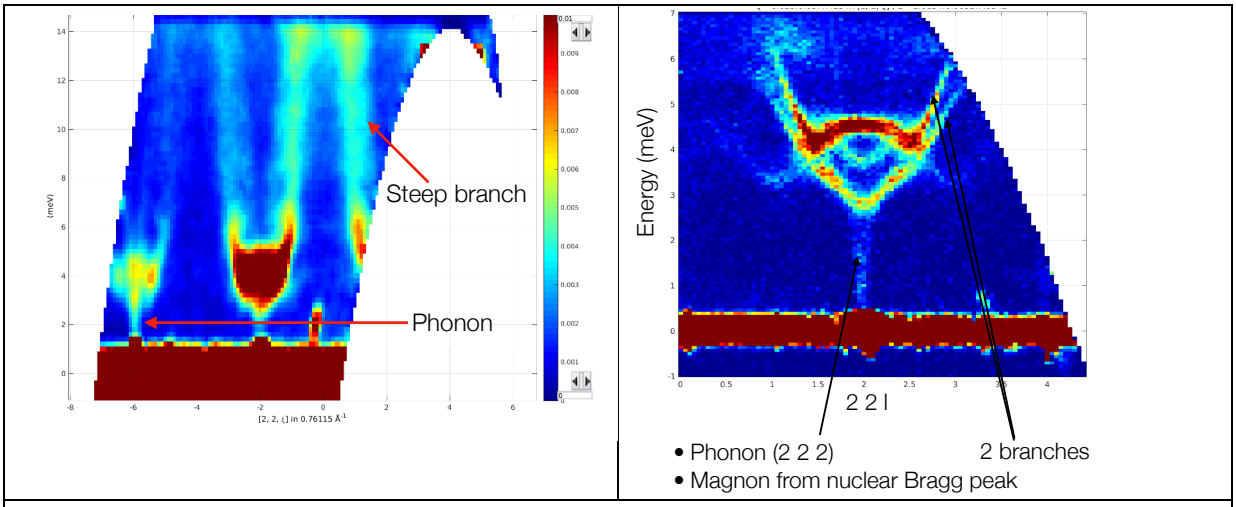

Figure 2: Single-crystal inelastic neutron scattering with Q-scan along (2 2 l) in **$GeCo_2O_4$** measured on IN5 with λ=2 (left) and 3 (right) Å at 1.5 K in the (hh0)-(00l) scattering plane.

We have found nicely and complex dispersive low energy modes (see Figures 2 and 3), with energy gaps (1 and 3 meV for $GeNi_2O_4$ and $GeCo_2O_4$ resp.). For the Ni compound, the spinwaves are anisotropic. For the Co compound, they are more isotropic. Moreover, starting from these excitations rise weaker excitations that strongly disperse, as shown in the figure 2. We assume that they have a magnetic origin but they could also be phonons entangled with the magnetic ones due to the strong magnetoelastic effects at play in the Co. We are working on the data modeling by spin wave calculations with the software developed by Sylvain Petit at the LLB but there are some obstacles: 1) the ambiguity between the simple-k and triple-k structure. 2) For Co, we were not able to see the top of the branches of the strong dispersive mode on IN5. We therefore need complementary high-energy measurements to complete these rich sets of data and conclude on the Hamiltonian of these materials, in particular on the possibility of a hedgehog-like magnetic structure for the Ni compound, and of magnetoelastic excitations for the Co one.

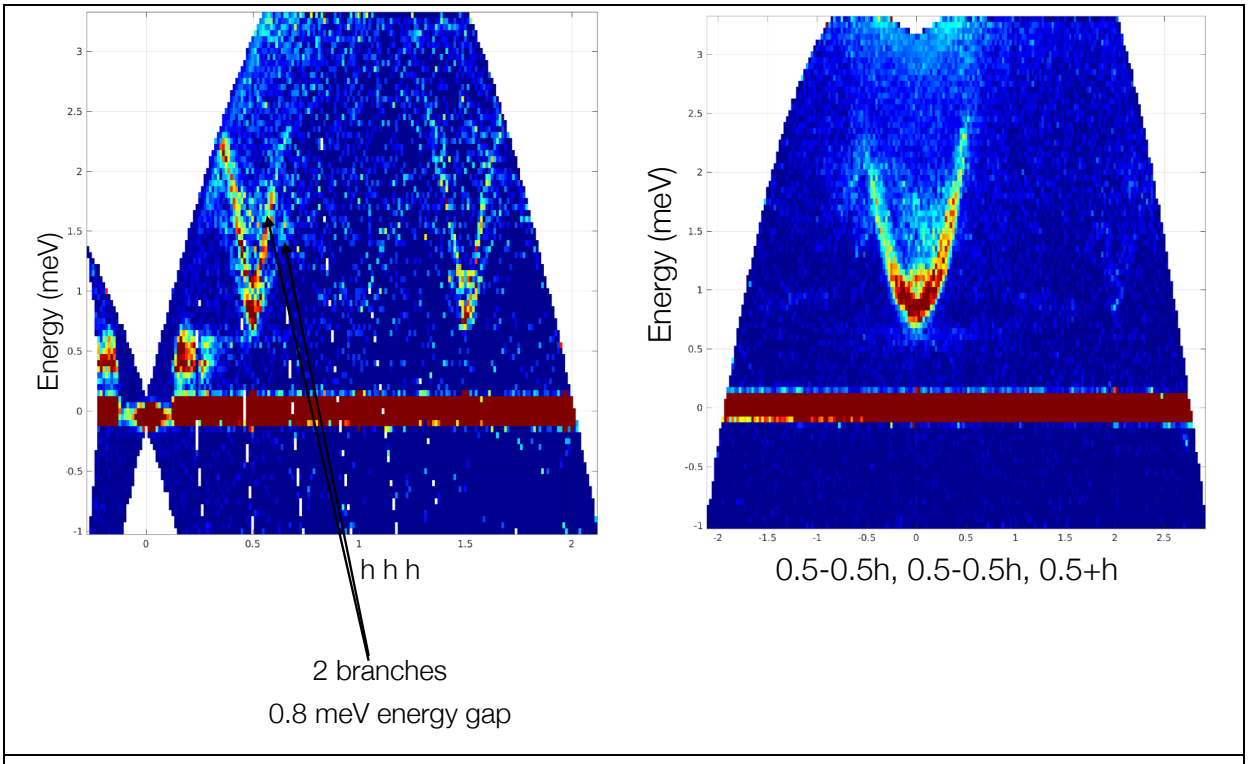

Figure 3: Single-crystal inelastic neutron scattering in **$GeNi_2O_4$** measured on IN5 with λ=4.35 Å at 1.5 K in the (hh0)-(00l) scattering plane, showing the anisotropy of the spinwaves.

References:

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[2] M. Matsuda, J.-H. Chung, S. Park, *et al*., Europhys. Lett. **82**, 37006 (2008).
[3] M. Matsuda, T. Hoshi, H. Aruga Katori, M. Kosaka, and H. Takagi, J. Phys. Soc. Jpn. **80**, 034708 (2011).
[4] X. Fabrèges, E. Ressouche, F. Duc *et al*., Phys. Rev. B **95**, 014428 (2017).
[5] A. N. Yaresko, Phys. Rev. B **77**, 115106 (2008).