Experimental report

04/10/2017

Proposal: **4-01-1492** **Council:** 4/2016

Title: Soft mode spin waves in frustratedmutliferroic Cu3Bi(SeO3)2O2Cl

Research area: Physics

This proposal is a new proposal

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Samples: Cu3Bi(SeO3)2O2Cl

Instrument	Requested days	Allocated days	From	To
IN3	2	2	12/07/2016	14/07/2016
IN12	7	0		
THALES	0	6	01/09/2016 07/11/2016	05/09/2016 09/11/2016

Abstract:

This proposal aims to continue our study of the spin wave spectrum in the magnetically frustrated multiferroic, Cu3Bi(SeO3)2O2Cl (francistie). Francisite is interesting due to its unique magnetic and structural phase transitions which work to form functional properties. A buckled kagome lattice of competing ferromagnetic and antiferromagnetic sites is stabilised below 27 K by a microscopic Dzyaloshinskii-Moriya interaction. This fragile state is easily perturbed by weak external fields initiating a metamagnetic spin-flip transition. The magnetic phase is also accompanied by a ferroelectric phase onset by a structural distortion at 127 K. The combination of a frustrated multiferroic phase with metamagnetic switching capabilities is an intriguing physical question with clear application based possibilities. The question of the precise nature of the ground state may be answered through mapping the fundamental excitation spectrum. We have already observed spin waves in the (a*, c*) plane but predict soft modes characteristic of strong frustration in the (b*, c*) plane. The measurement of the (b*, c*) plane is therefore essential for our final classification of the ground state.

Soft mode spin waves in frustrated mutliferroic $Cu_3Bi(SeO_3)_2O_2Cl$

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$Cu_3Bi(SeO_3)O_2Cl$, or francisite, is a buckled kagome compound made from two unique Cu spin ½ sites (Cu1 and Cu2) and displaying unusual frustration mechanisms [1]. In fact, there is a strong competition between nearest-neighbor ferromagnetic and next-nearest-neighbor antiferromagnetic interactions. With only isotropic exchange interactions, and despite dominant ferromagnetic couplings, this would lead to a classical degenerate ground state that should survive quantum fluctuations. However, the first experimental studies of this material have shown that it actually orders in a canted magnetic structure below 25 K and that the magnetic macroscopic behavior is highly anisotropic. A theoretical work has predicted that the essential ingredient stabilizing this magnetic order is a small Dzyaloshinskii-Moriya interaction [2].

In our neutron scattering experiments performed at the ILL we have found that the magnetic behavior is further complicated by the fact that this material additionally undergoes a structural distortion at 115 K distorting from a *Pmmn* to *Pcmn* structure accompanied by a doubling of the unit cell along the *c* axis. Taking this into account, we were able to refine the magnetic structure within the new atomic symmetry, revealing an increased canting of the Cu1 spins. The refined magnetic structure is shown in Fig. 1.

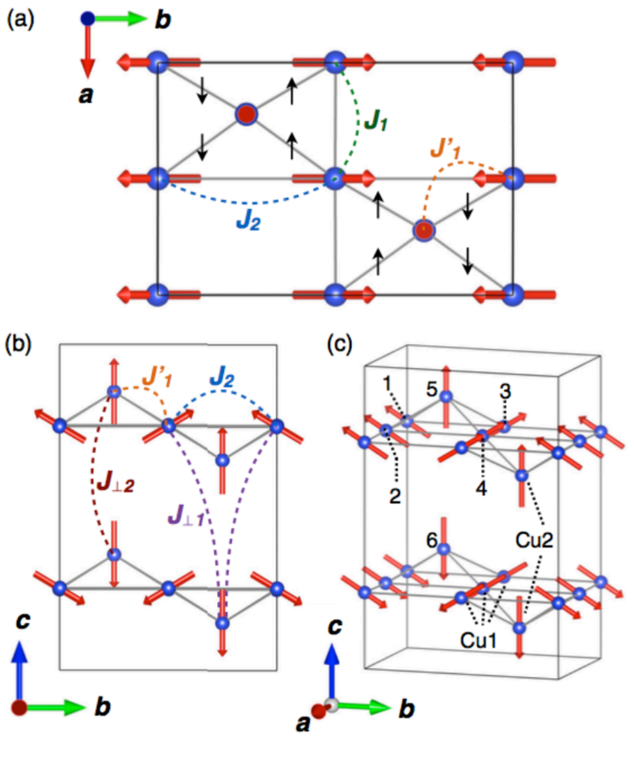

Fig. 1: Refined canted magnetic structure at 2 K with *Pcmn* space gorup. The Cu1 and Cu2 spins are shown in the *ab* plane (a), in the *bc* plane (b), and in a three dimensional representation (c). The spins are indexed according to Eq. (1) in Ref. [3]. The exchange interactions in the plane and between the planes are shown [(a) and (b)], as well as the dominant component of the DM interaction between the nearest-neighbor Cu1 and Cu2 spins (black arrows) in panel (a). This figure has been adapted from Ref. [3].

On the triple-axis spectrometers at ILL (ThALES, IN12 and IN3), we have performed inelastic neutron scattering measurements in order to identify the relevant ingredients of the Hamiltonian from the spin wave spectrum. In disagreement with the previously considered model of Rousochatzakis *et al*. [2], a global gap of the excitations is observed that cannot be solely explained by the Dzyaloshinskii-Moriya interactions but requires additional anisotropic exchange interactions. This disagreement can be seen when comparing the simulated dispersion maps of Figs. 2 (a) and (b) with the experiment in Figs. 2 (c) and (d). Here we find that while many of the key features of in the measured spin wave maps, including the dispersion and the spectral weight of the excitations, are captured in the simulations produced by the Hamiltonian of Rousochatzakis *et al*. [2], there is a failure to reproduce the observed spin wave gap of ~1.5 meV.

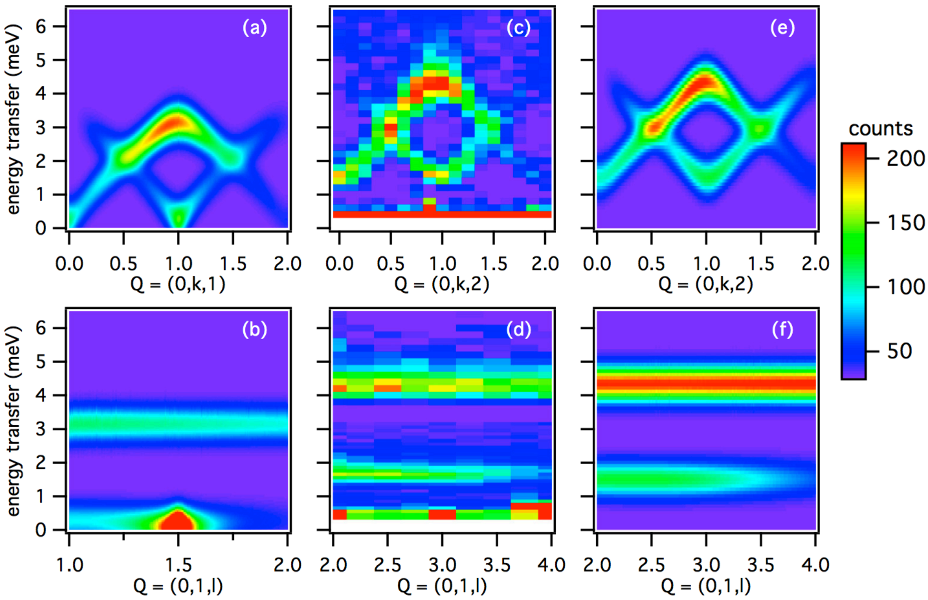

Fig. 2: Spin wave dispersions for CBSCl in the (b^*,c^*) scattering plane. Simulated dispersion calculated for Q=(0,k,1) (a) and Q = (0,1,l) (b) using the proposed Hamiltonian of Rousochatzakis *et al*. [2]. Measured spin wave dispersion at 2 K for Q = (0,k,2) (c) and Q = (0,1,l), mapped from several ThALES energy scans (d). Simulated spin wave dispersion using modified Hamiltonian incorporating symmetric anisotropic exchange for Q = (0,k,2) (e) and Q = (0,1,l) (f). Note that the (c)–(f) maps are indexed in the distorted structure implying a doubling of the c lattice parameter. This means there is a doubling of the l index compared to the (a), (b) spectra calculated in the undistorted structure. This figure has been adapted from Ref. [3].

To reproduce the spin wave gap in our simulations we have attempted many modifications to the Hamiltonian including adjusting the interaction strengths, altering the DM interaction vector, and introducing anisotropy. We also considered the consequences on the magnetic Hamiltonian of the structural distortion occurring at 115 K from the *Pmmn* to *Pcmn* space groups with a doubling of the unit cell along c. Successful attempts to reproduce the observed magnetic structure and the associated spin wave dispersion were finally achieved by introducing an adapted anisotropic exchange to the simulations. The simulated dispersion from the modified Hamiltonian is seen in Fig. 2 (e) and (f). Here we find excellent agreement to the experimental results. The results of our inelastic neutron study on ThALES therefore show that a refinement of the Hamiltonian for $Cu_3Bi(SeO_3)O_2Cl$ is necessary and reveals that a non-negligible symmetric exchange anisotropy is required, on top of the weak Dzyaloshinski-Moriya interaction, to stabilize the magnetic phase and induce the observed spin wave gap. The results of this study have been published as an article in Physical Review B and are listed under Ref. [3].

REFERENCES

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