Experimental report

12/09/2016

Proposal: **4-01-1486** **Council:** 4/2015

Title: Resolving helimagnon excitations in the long-pitch helimagnetic insulator Cu2OSeO3

Research area: Physics

This proposal is a new proposal

Main proposer: **Dmytro INOSOV**

Experimental team: Pavlo PORTNICHENKO
Dmytro INOSOV

Local contacts: Bernhard FRICK
Markus APPEL

Samples: Cu2OSeO3

Instrument	Requested days	Allocated days	From	To
IN16B	7	7	08/06/2016	15/06/2016

Abstract:

In the multiferroic Cu2OSeO3 system, the interplay of interatomic exchange and the Dzyaloshinskii-Moriya (DM) interactions leads to the twisting and canting of magnetic moments and, consequently, to the formation of a spin-spiral ground state. An exotic skyrmion-lattice arrangement can be further stabilized by the application of magnetic field. Our recent time-of-flight (TOF) and triple-axis (TAS) neutron spectroscopy experiments revealed a complicated structure of magnetic excitations in Cu2OSeO3, confirming recent theoretical predictions. However, due to the long pitch of the spin spiral in its zero-field ground state, resolving individual helimagnon branches turned out to be impossible with a conventional cold-neutron spectrometer. Therefore, here we propose to employ the new high-resolution backscattering spectrometer IN16B to map out helimagnon excitations around the (111) or (222) wave vector.

Experimental report No. 4-01-1486

Proposer: Pavlo Portnichenko <pavlo.portnichenko@tu-dresden.de>
Experimental team: P. Portnichenko, D. Inosov (TU Dresden)
Date of experiment: 12-18 May, 2015

Introduction

The noncentrosymmetric multiferroic material Cu_2OSeO_3 (Fig. 1) is the first insulating chiral helimagnet that exhibits a skyrmion lattice phase [1]. Detailed understanding of the microscopic interactions underlying this complex behavior still remains a subject of active theoretical investigation [2, 3]. In our previous experiments we have successfully mapped out the phase diagram of the magnetic states in Cu_2OSeO_3 under the influence of an applied magnetic field using the MIRA, small-axis diffractometer at MLZ [4]. Then, we have also collected INS data on the same compound using both cold- and thermal neutron TAS and TOF spectrometers. Our latest paper [5] present the complete overview of spin excitations throughout the entire Brillouin zone and over a broad energy range. Observed features were successfully described by the previously developed theoretical model of interacting tetrahedra [6]. These data show good agreement with theory, with the exception of the weakest DM interactions leading to very subtle changes in the spectrum that we could not resolve so far. In our PANDA data, the ferromagnon branch appears to be gapless, which is consistent with earlier microwave absorption measurements. The effective magnetic anisotropy causing the gap in the ferromagnetic resonance spectrum was estimated to be ~ 3 GHz [7].

We were expecting, that the low-energy excitations of a noncollinear spin spiral form so-called helimagnons [8], which have been so far observed directly by INS only in the MnSi compound [9]. In this experiment we tried to extend these results and confirm similar behavior in Cu_2OSeO_3. In a simple model parabolic ferromagnon branches stemming from the 1^{st}, 2^{nd}, 3^{rd} and higher-order incommensurate magnetic Bragg reflections hybridize to form multiple flat bands that can be revealed in an energy scan as a series of peaks with an energy splitting determined both by the period of the magnetic helix in real space and by the "stiffness" of the ferromagnon branch. Expected energy splitting is $\Delta E \approx 10.5\,\mu$eV. Since this value is a factor of 10 smaller than in MnSi we used the high energy resolution available at IN16B.

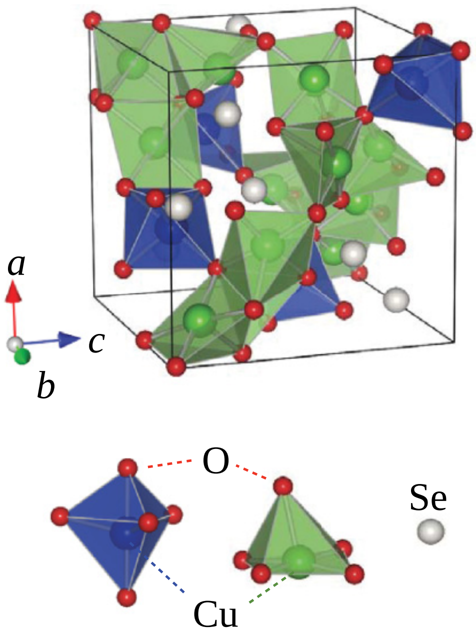

Fig. 1: The crystal structure of Cu_2OSeO_3 from Ref. 1.

Experimental configuration

Measurements were performed on a pre-aligned array of Cu_2OSeO_3 single crystals with a T_C of 58 K that was initially mounted on an Al-holder with its $(1\bar{1}0)$-axis vertical. Using the published lattice parameters $a = b = c = 8.925$ Å, $\alpha = \beta = \gamma = 90°$ we aligned on the (111) reflections. Although intensity of the helimagnon is several times higher in the vicinity of the (222) reflections, with the available Si(111) analyzer we were limited in $|\mathbf{Q}|$. The wave vector of the scattered neutrons $k_f = 1$ Å was fixed. As we are interested in the vicinity of the (111) reflection we calculated $2\Theta \sim 75°$ for the (111) Bragg reflection. According to the instrument geometry, position sensitive detector (PSD) tubes #7 and #8 are on the $2\Theta \sim 71.9°$ and 79.7° respectively, thus we expect to find maximum of the Bragg reflection intensity between these tubes. By rotating the sample stick insert we foud maximum intensity to be in the tube #7.

After counting for 20h we still could not see any useful signal in the inelastic chancel. We decided do reduce the background by masking the cryostat. As we are mostly interested in the central part of the detector we masked top and the bottom part of the cryostat. It reduced the number of neutrons which reach the detector after scattering from the analyzer, and has not affected on the Bragg intensity.

However this effort did not help. Even with the Cd mask we still observed significant amount of neutrons counted by the PSD tubes, which are actually

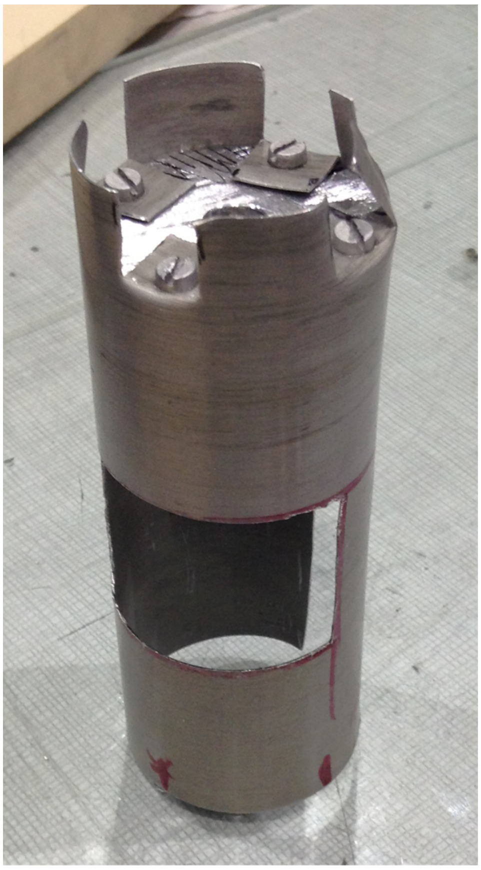

Fig. 2: Cd mask that was used to reduce the background.

masked. This could mean that the voltage on the PSD tubes is to high and we are counting a lot of gammas. We found that the count rate went down from $\sim$ 11cnts/sec (1580 V) to $\sim$ 5cnts/sec (1530 V). However this background was still too high. We also observed significant intensity in the low $|\mathbf{Q}|$ region, so we decided to check if it comes from the double-scattering from the sample on the cryostat walls. After we removed the sample, we realized that most of the background signal is coming from the cryostat. We decided to install an additional Cd mask on the sample, as this might reduce the background from the cryostat. We prepared a mask in a way that only tiny widow with the opening angle towards direct beam and PSD tube #7 was left (Fig. 2). With this configuration we continued until the end of the experiment. For the last two days we removed the sample and measured only empty cryostat with the installed mask.

Results

We have investigated the spectrum of magnetic excitations in Cu_2OSeO_3 in the vicinity of the (111) reflection using backscattering spectrometer IN16b. We have shown that within the error bar the signals with and without the sample look identical. We conclude that the available flux is too low to observe magnetic intensity at a given Q point in our sample, and therefore we can not resolve individual branches of the helimagnon. We treated each half of the spectrum individually (each half corresponds to the half period of the doppler drive movement) since we noticed that they have significantly different background (Fig. 3). Only region of the spectra where we see evidence of the signal is $\sim 8\,\mu$eV peak on the energy loss side (Fig. 3, Top). However absence of the similar peak on the energy gain side seriously questions its magnetic origin.

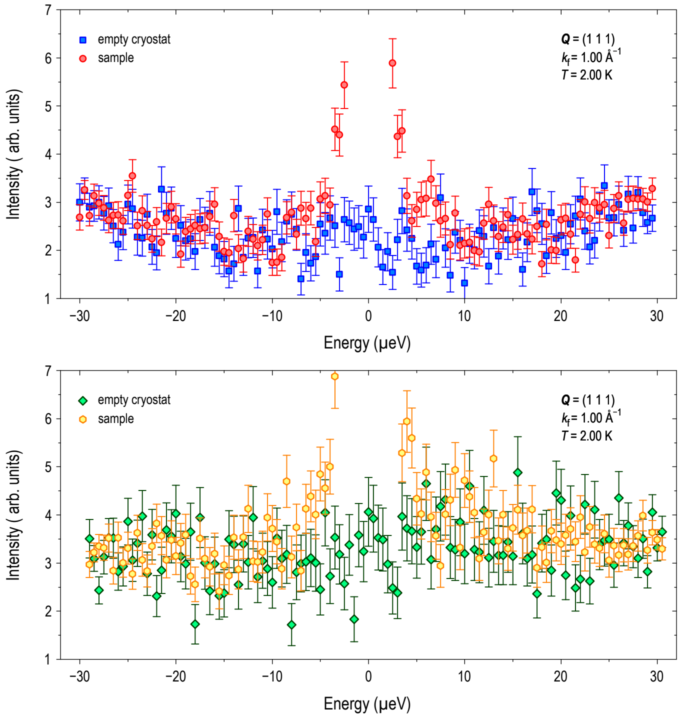

Fig. 3: Top: first half of the energy spectrum and its comparison with the signal from the empty cryostat. Bottom: second half of the spectrum.

[1] S. Seki *et al.*, Science **336**, 198 (2012).
[2] J. H. Yang *et al.*, Phys. Rev. Lett. **109**, 107203 (2012).
[3] V. A. Chizhikov & V. E. Dmitrienko, arXiv:1305.5382 (2013).
[4] S. Seki *et al.*, Phys. Rev. B **85**, 220406(R) (2012).
[5] P. Y. Portnichenko *et al.*, Nature Comm. **7**, 10725 (2016).
[6] J. Romhanyi *et al.*, Phys. Rev. B **90**, 140404(R) (2014).
[7] M. I. Kobets *et al.*, Low Temp. Phys. **36**, 176 (2010).
[8] M. Janoschek *et al.*, Phys. Rev. B **81**, 214436 (2010).
[9] M. Kugler *et al.*, Phys. Rev. Lett. **115**, 097203 (2015).