# Experimental report

12/02/2019

**Proposal:** **CRG-2379** **Council:** 10/2016

**Title:** Electric-field control of chiral magnetic dynamical scatterings in multiferroic CuFe1-xGaxO2 (x=0.035)

**Research area:** Materials

**This proposal is a new proposal**


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**Samples:** $CuFe_{0.965}Ga_{0.035}O_2$

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| IN12 | 7 | 7 | 20/06/2018 | 27/06/2018 |


## Abstract:

We propose to perform polarized inelastic neutron scattering (PINS) measurements on spin-driven multiferroic compound $CuFe_{1-x}Ga_xO_2$ (CFGO) with x=0.035 under applied electric field. By the PINS technique, we shall investigate the chirality in spin dynamics. By applying positive or negative electric field on cooling, we shall be able to control the sense of the dynamical magnetic chirality, as well as static magnetic chirality.

**Electric-field control of chiral spin correlations in magnetic excitations in multiferroics CuFe1-xGaxO2**

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Since the discovery of spin-driven multiferroicity in $TbMnO_3$, it has been recognized that a spiral magnetic order can beak spatial inversion symmetry leading to ferroelectricity. In the case of $TbMnO_3$, the symmetry breaking is brought about by a cycloidal-type magnetic order, which has a spin-helicity degree of freedom, that is, clockwise or counter-clockwise arrangement of magnetic moments. A polarized neutron diffraction study on this compound revealed that the spin helicity can be controlled by applying an electric field, which determines sign of the spin-driven electric polarization [2]. Similar experiments were subsequently performed on a number of frustrated magnets with spiral magnetic orders such as $Ni_3V_2O_8$, $CuFe_{1-x}Ga_xO_2$, $MnWO_4$ and etc, demonstrating that in each system, there is a one-to-one correspondence between the spin helicity and the polarity of electric polarization. These observations naturally lead to the prediction that the helicity degree of freedom in spin dynamics can also be controlled by electric field. However, this point remains to be investigated thus far. Here, we have studied magnetic excitations in multiferoic $CuFe_{1-x}Ga_xO_2$ (CFGO) with x=0.035 under electric field to address this issue.

CFGO(x=0.035) has a rhombohedral crystal structure belonging to the space group of R-3m at high temperatures. It exhibits the multiferroics phase, which has a screw-type magnetic order with a modulation wave vector of ($q$,$q$,3/2), below 7 K in zero magnetic field. The spin-driven electric polarization appears along to the [110] direction, which is parallel to the direction of the screw axis of the magnetic structure [3]. In the present study, we performed polarized neutron inelastic scattering measurements on CFGO (x=0.035) using the cold

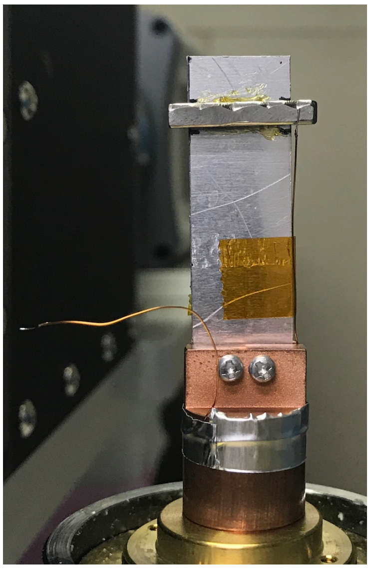

Fig.1 : Picture of CFGO sample used in the present study.

neutron triple-axis neutron spectrometer at the beamline IN12 in ILL. A single crystal of CFGO was grown by the floating zone method. The crystal was cut into a rectangular shape as shown in Fig. 1. A pair of flat surfaces was nearly perpendicular to the [110] direction of the crystal. Pt/Au electrodes were formed on these surfaces to apply electric field along the [110] direction. The distance between the electrodes, namely the thickness of the sample, was 3.35 mm. The sample was loaded into an orange cryostat with the (*h,h,l*) scattering plane. A spin-polarized incident neutron beam was obtained by a supermirror polarizer and a pyloric graphite 002 monochromator. A Heusler analyzer was employed to analyze the energy and spin state of the scattered neutrons. The spectrometer was operated in the fixed-$k_f$ mode with $k_f$=1.30 $A^{-1}$. The polarization direction of the incident neutrons ($\boldsymbol{p}_N$) was controlled to be parallel/antparallel to the scattering vector ($k_f$-$k_i$) by using guide fields, a Helmholtz coil, and two spin flippers.

Figure 2(c) shows an elastic scattering profile along the (*h,h*,3/2) line. Prior to the measurements the sample was cooled under electric field of +298.5 kV/m. Blue and red symbols denote the observed intensity measured when the spin state of the incident neutrons was the up (u) and down (d) state, respectively. Hereafter, we refer to the former and the latter as $I_{ud}$ and $I_{du}$, respectively. Note that because $\boldsymbol{p}_N$ was set to be parallel to the scattering vector, the neutron spin state was always flipped through the magnetic scattering process. We observed two magnetic peaks at (*q,q*,3/2) and (1/2-*q*,1/2-*q*,3/2), which arose from the screw-type magnetic order. For both the magnetic reflections, we observed asymmetry between $I_{ud}$ and $I_{du}$, which indicates that there was asymmetry in spin helicity in the system. Ideally, for the (*q,q*,3/2) [(1/2-*q*,1/2-*q*,3/2)] reflection, $I_{du}$ ($I_{ud}$) should be as small as the background signal when the system is in a single-domain state in terms of spin helicity. However, both the $I_{ud}$ and $I_{du}$ were actually finite for these reflections. This means that the applied electric field was not strong enough to completely remove the minority domains in which the spin-driven electric polarization is antiparallel to the electric field.

Figs. 2(a) and 2(b) show the constant-*E* cuts with the energy transfer of 0.8, and 0.5 meV, respectively. We observed nearly linear spin-wave dispersion emerging from the magnetic Bragg peak at E=0. Interestingly, the asymmetry between $I_{ud}$ and $I_{du}$ was also observed in the excitation spectra. This demonstrates that the spin-wave excitation also has the same spin helicity as that in the static magnetic structure. We also measured elastic and inelastic scattering profiles after cooling the sample under a negative electric field (Figs. 2(d), 2(e) and 2(f)), revealing that the asymmetry between $I_{ud}$ and $I_{du}$ was reversed not only in the elastic scattering profile, but also in the inelastic scattering profile. These results demonstrate that in spin-driven multiferroics, the spin-helicity degree of freedom in spin dynamics can also be controlled by electric field. Further analysis is still ongoing to reveal the electric field dependence of the whole magnetic excitation spectra.

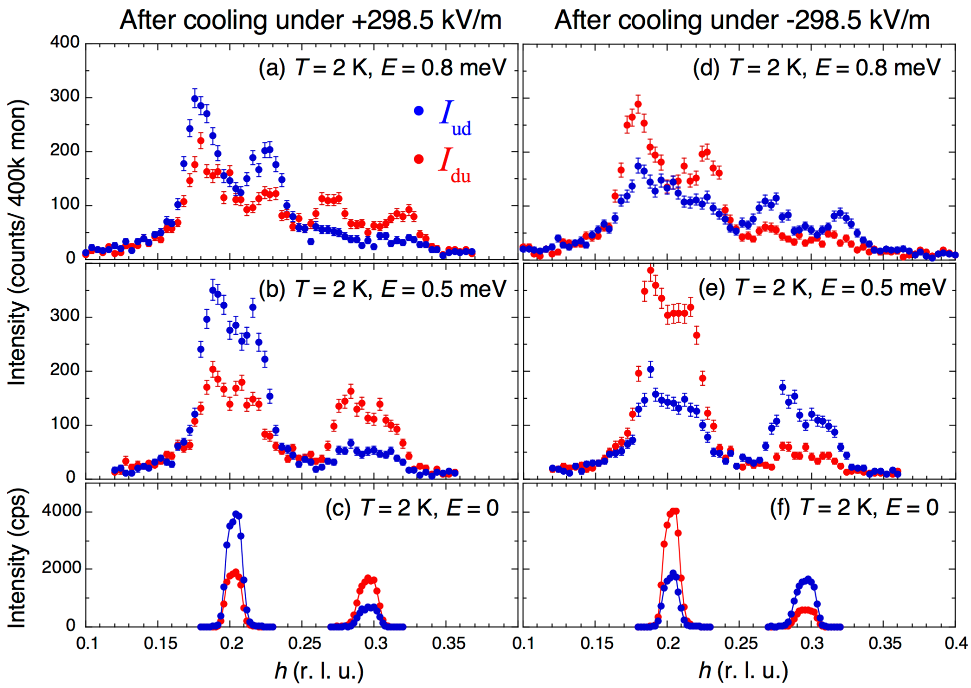


Fig.2 : Elastic and inelastic scattering profiles along the (h,h,3/2) line measured [(a)-(c)] after cooling the sample under electric field of +298.5 kV/m, and [(d)-(f)] those for negative electric field.

Reference:


[1] T. Kimura *et al*., Nature **426**, 55 (2003).

[2] Y. Yamasaki *et al.,* Phys. Rev. Lett. **98**, 147204 (2007).

[3] T. Nakajima *et al*., Phys. Rev. B **79**, 214423 (2009).