Experimental report

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Proposal: **5-51-514** **Council:** 4/2016

Title: Investigation of high-pressure phases of Multiferroic Delafossite $CuFeO_2$ by using CryoPAD and Hybrid-Anvil-Cell

Research area: Physics

This proposal is a new proposal

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Samples: $CuFeO_2$

Instrument	Requested days	Allocated days	From	To
D3 CPA	5	5	12/12/2016	17/12/2016

Abstract:

We propose to perform neutron polarimetry analysis under high-pressure for multiferroic $CuFeO_2$ using CryoPAD in combination with hybrid-anvil-cell developed by the user. The ferroelectric polarization in $CuFeO_2$ is induced hydrostatic pressure as well as magnetic field, and impurity doping. Our recent high pressure experiment with unpolarized neutrons demonstrated drastic change in magnetic orderings from non-polar to polar phases in $CuFeO_2$, which is unique behaviour in multiferroics.(N. Terada, P. Manuel et. al, PRB 89 220403(R) (2014)) For understanding the pressure-induced phase transitions in $CuFeO_2$, it is essential to determine magnetic structure in the high-pressure phases by neutron polarimetry analysis.

Experimental report for “Investigation of high-pressure phases of Multiferroic Delafossite $CuFeO_2$ by using CryoPAD and Hybrid-Anvil-Cell”

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Abstract: In this experiment, we have succeeded in measuring full polarization matrices under high pressure condition for the first time, by using the CryoPAD apparatus on D3 beamline and our developed Hybrid-Anvil-Cell (HAC). This experiment also determined the complex spiral magnetic structures in these pressure-induced phases, each of which possesses a different spin orientations and symmetry. The results presented demonstrate that the SNP measurements are feasible in combination with high pressure conditions using this new setup, and that this method is a useful approach to studying novel pressure-induced physical phenomena associated with complex magnetic ordering.

Experiment: The neutron polarimetry experiments were carried out using a CRYOPAD apparatus[1, 2] on the D3 beam line. Single crystal samples of $CuFeO_2$, grown by the floating zone technique, were cut into rectangular shapes with dimensions of 1.1 × 1.1 × 0.2 mm^3 and 0.4 × 0.5 × 0.2 mm^3 for experiments at *P* = 0.2 and 2.0 GPa, and P = 4.0 GPa, respectively. The former and latter crystals have mosaic widths, 0.40 ± 0.02 and 0.50±0.05, respectively. The crystal qualities were kept even under pressure up to 4.0 GPa, by using glycerin as the pressure transmission medium. The cut samples were mounted in the HAC with the monoclinic *a*-axis (hexagonal [1 -10]) vertical, in order to provide access to the monoclinic (0, *K*, *L*) (hexagonal (*H, H, L*)) reflections. The incident neutrons are polarized and monochromatized at the Heusler monochrometer. The incident wavelength 0.85 Å was employed. The final neutron spin was analyzed with ^{3}He filter cells. The data has been corrected for the exponential decay of the ^{3}He polarization. A sapphire anvil with a 4.2 mm diameter culet, supported by CuBe, and a nonmagnetic diamond composite (with a SiC binder) were used up to 2.0 GPa, while the supported sapphire anvil of 2.7 mm diameter culet and WC with a nonmagnetic Ni binder were employed during the 4.0 GPa experiments. These materials were confirmed to be nonmagnetic by magnetization measurements. Aluminum gaskets (Al2017) with 2.0 or 1.0 mm diameter hole were used for the P = 0.2 and 2.0 GPa, and P = 4.0 GPa experiments, respectively. The HAC was inserted into a Orange cryostat.

Results: As shown in Figs. 1a and 1b, we observed magnetic reflections corresponding to the ICM2 phase below T =10.0 K at P =2.0 GPa, which was

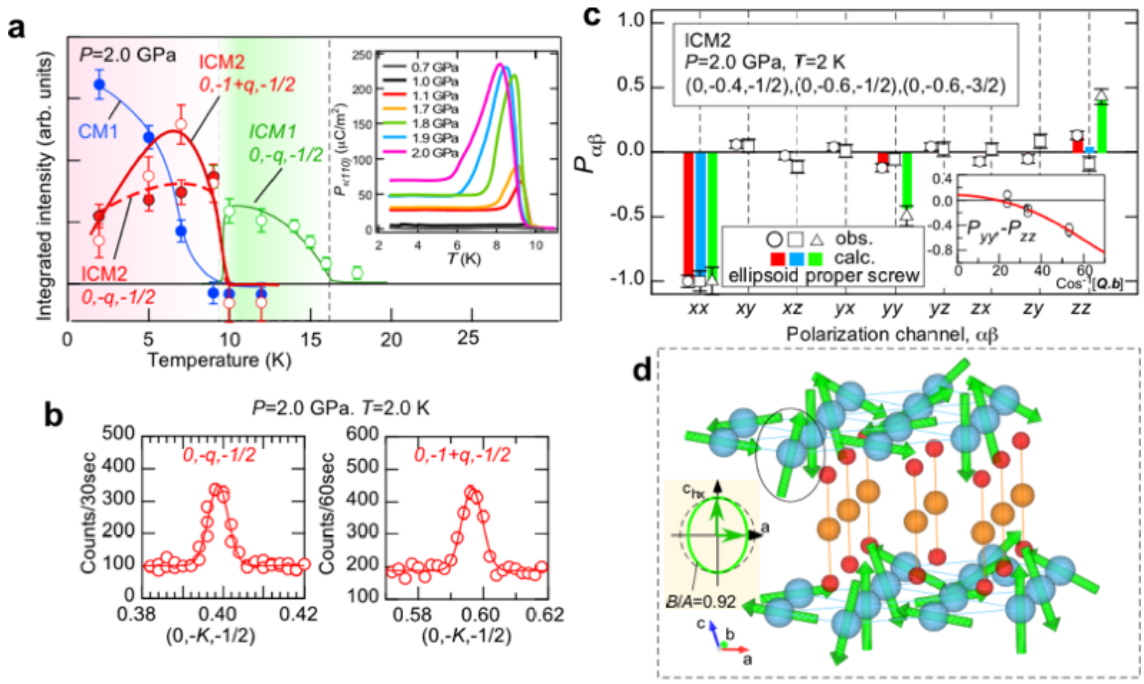

Fig. 1 (a) Temperature dependence of the integrated intensities of magnetic reflections and (b) typical diffraction profiles line for the spin flip channel in the x direction at P = 2.0 GPa. The inset in (a) shows the temperature dependence of electric polarization along the hexagonal [110]-direction (the monoclinic b-axis) in typical pressures up to 2.0 GPa under poling electric field 286 kV/m. (c) A comparison between observed and calculated polarization matrix elements for *0,-0.2,-1/2, 0,-0.3,-1/2* and *0,-0.3,-3/2* reflections at T = 2 K and P = 2.0 GPa. The inset in (c) shows the fitting results for the refinement of the ellipsoidal ratio and the tilting angle θ. (d) An illustration of the magnetic structure determined under these conditions: an ellipsoidal proper screw structure with 0.92 ± 0.05.

coexistent with the CM1 phase below 7 K, because this pressure value was close to the phase boundary. The $P_{\alpha\beta}$ values were determined for the reflections at (0,−*q*,−1), (0,−1+*q*,−1) and (0,−1+*q*,−3), and are presented in Fig. 1c. The observed matrix elements, P_{yz} and P_{zy}, were zero for all reflections measured within the experimental accuracy, a result that differs from the case of SDW in the ICM1 phase. Therefore, one of the ellipsoidal spiral axes (the minor axis), was parallel to the *z*-direction (the monoclinic *a*-axis), which relates to the conditions. The other axis (the major axis) was perpendicular to the a(z)-axis. We refined the direction of the major axis (the canting angle θ from the c_{hx}-axis toward the *b*-axis) and the ellipsoidal ratio for the P_{yy} and P_{zz} elements sensitive to these parameters. The refined direction of the major axis was along the c_{hx} -axis, $\theta = 4 \pm 4^{\circ}$, and the ellipsoidicity parameter was 0.92 ± 0.05 . P_{yx} and P_{zx} were zero within the experimental accuracy, meaning that the helicity domains (both right-handed and left-handed) of the spiral structure were equally populated, which is consistent with no electric polarization observed under zero electric field. We thus found that the magnetic structure in the ICM2 phase is the proper screw structure exhibiting slight ellipsoidicity (Fig. 1d).

Conclusion:

As mentioned above, we have determined the magnetic structure in the pressure-induced ferroelectric phases in multiferroic $CuFeO_2$ by using the nonmagnetic HAC and CRYOPAD combination. Magnetic structures in the other phases for ICM3 and ICM4 phases were also determined in the present experiment. Although we cannot write the details of these phases due to the page limitation in the experimental report, we have already submitted a paper.