Uniaxial-pressure control of ferroelectricity in a spin-driven magneto-electric multiferroic CuFeO2

Hiromu Tamatsukuri, Setsuo Mitsuda, Ryuta Henmi
Tokyo University of Science, Department of Physics

A triangular lattice antiferromagnet $CuFeO_2$ exhibits a spin-driven ferroelectric phase (a FE-ICM phase), in which a noncollinear helical magnetic structure breaks inversion symmetry in this system, by substituting a few percent of nonmagnetic Ga^{3+} for magnetic Fe^{3+}. In addition, $CuFeO_2$ is also a strong spin-lattice coupling system, whose magnetic phase transitions are accompanied by crystal lattice distortions owing to the geometrical spin frustration. Therefore, this compound is one of the model materials for investigating novel magneto-electric cross-correlated phenomena induced by anisotropic uniaxial pressure p.

Recently, we found that the application of p along the $[1\bar{1}0]$ direction, which is conjugate to the spontaneous lattice distortion, induces another ferroelectric phase (a FE2 phase) in $CuFe_{1-x}Ga_xO_2$ (CFGO) with x = 0.035. In previous neutron diffraction experiment under applied p of 400 and 600 MPa (NSL-00000329), we revealed that this ferroelectric transition was accompanied by the magnetic phase transition from the OPD phase, as shown in Fig. 1(b). In this study, we have investigated how a phase-boundary line between the OPD and the PD phases starting from the point (p = 0 MPa, T = 13.5 K) meets with a phase-boundary line between the OPD and the FE2 phases in the p-T magnetic phase diagram of the x=0.035 sample, as an extension of previous study.

The neutron-diffraction measurements under applied p were carried out at the two-axis diffractometer E4 installed at the Berlin Neutron Scattering Center in the Helmholtz Zentrum Berlin for Materials and Energy. The wavelength of incident neutron was 2.44 Å. Since the direction of p is parallel to the $[1\bar{1}0]$ direction, the scattering plane is the (HHL) plane.

A magnetic phase transition temperature from the OPD to the PD or the FE2 phases, T_{N2}^{high}, can be determined by temperature dependence of a magnetic wave propagation wave number q; q on cooling starts to vary at T_{N2}^{high} as shown in Fig. 1(a). As a result, we have determined the phase boundary line in the p-T magnetic phase diagram of the x=0.035 sample, as indicated by a red line in Fig. 1(b). We have found that a temperature of ferroelectric polarization emergence, T_{FE2}, corresponds to T_{N2}^{high} above p = 350 MPa, while in a range of 200 MPa $\leq p \leq$ 350 MPa, it does not.

From this experiment, the magnetic structure in the FE2 phase is expected to be the sinusoidal type, which does not break the inversion symmetry in this system as in the PD phase. Since the scattering plane is restricted to the (HHL) plane, however, we can still consider the possibility that there are some tiny modifications from the sinusoidal magnetic structure induced by applied p, and this modified magnetic structure results in the inversion symmetry breaking. To understand the origin of ferroelectricity in the FE2 phase, it is a necessary task to perform more detail magnetic structure analysis in the FE2 phase using magnetic reflections in wider reciprocal lattice space beyond the (H, H, L) plane.

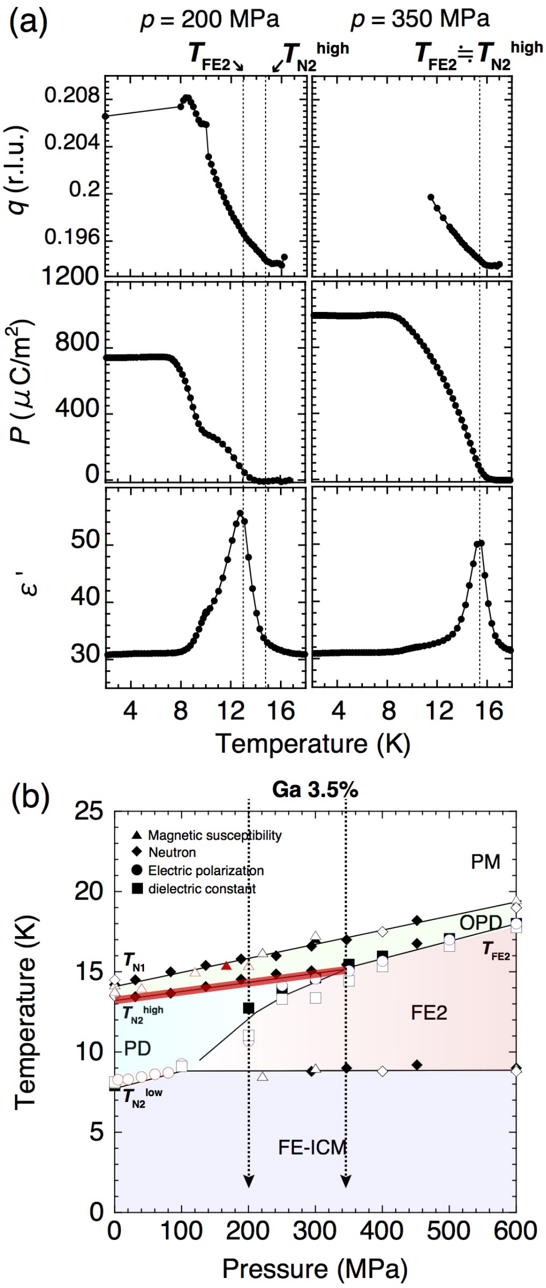

Fig. 1. (a) Temperature dependence of q, P[110] and dielectric constant under p of 200 MPa (left panel) and 350 MPa (right panel). (b) A p-T magnetic phase diagram of a x=0.035 sample. A red line indicates a phase boundary obtained by this experiment.