Uniaxial stress-induced ferroelectric phase in $CuFe_{0.95}Al_{0.05}O_2$

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Recently, we have studied a cross-correlated phenomena in multiferroic $CuFe_{1-x}M_xO_2$ (M = Ga, Al) that the application of uniaxial stress induces a new ferroelectric phase (called as FE2 phase), which is different from the well-studied spin-driven ferroelectric phase with helical magnetic ordering and can be mediated by the spin-lattice coupling [1].

For the x=0.05 Al-doped sample, the temperature vs uniaxial stress (*p* // [110]) magnetic phase diagram was obtained, as shown in Fig.1. The ground-state OPD magnetic structure in this phase diagram is a collinear magnetic structure in which the magnetic moment is sinusoidally modulated along the [110] direction with a propagation wave number q~0.2 and is tilted from the [001] direction to the [1-10] direction by an angle θ～50°. To clarify the magnetic structure in the ferroelectric FE2 phase that emerges inside the OPD phase, we performed neutron diffraction experiments using Fonder 4-axis spectrometer.

The single crystal sample (1.2 mm x 1.3 mm x 1.5 mm in size) was mounted in a clamped type-stress cell, and uniaxial stress was applied in the [1-10] direction at room temperature. As seen in the inset of Fig.1, the FE2 phase with a spontaneous polarization value of 170 μC /mm^2 at 2 K emerges below T~12K, suggesting that the effective stress *p* at the low temperature in the clamped type-stress cell seems to be ~170 MPa.

Among the three magnetic domains originating from the trigonal symmetry around the c axis, the (110) domain was confirmed to have almost 100% volume fraction. The magnetic Bragg reflections in the (110) domain were surveyed along two sets of sequences (-1+*q*, *q*, *l*) and (1-*q*,-1+*q*, *l*) with *l* =-8.5,-5.5,-2.5,0.5,3.5,6.5,9.5, as shown in Fig.2.

As a result, the magnetic structure of the FE2 phase turned out to be OPD magnetic structure with a tilting angle of θ~30±5°, meaning that magnetic ordering in the FE2 phase does not itself break the inversion symmetry in the system. However, taking into account of common tilting angle θ~50±5° in the OPD phase under *p*=0 MPa over x=0.035, x=0.05 and x=0.10 sample, a smaller tilting angle θ~30±5° for the FE2 phase found in the x=0.05 sample is likely to be relevant to the appearance of spontaneous electric polarization.

[1] H. Tamatsukuri *et al*., Phys. Rev. B **95**, 174108 (2017).

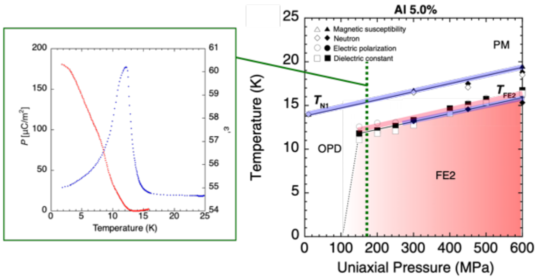

Fig. 1. Magnetic phase diagram of x=0.05 Al-doped sample.

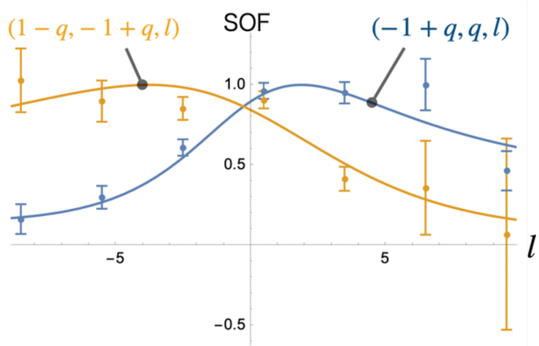

Fig. 2. Index *l*-dependences of Spin Orientation Factor (SOF) experimentally obtained from the integrated intensity of magnetic Bragg reflection along (-1+*q*, *q*, *l*) and (1-*q*,-1+*q*, *l*) scans are plotted, assuming the magnetic moment of 3.6 μ_B. The solid lines are those of SOF calculated based on the OPD magnetic structural model with *θ* ~30±5°.