

## Nonmagnetic impurity effect in ferroelectric phase of $\text{CuFeO}_2$

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In the past several years, a magnetic oxide  $\text{CuFeO}_2$  (CFO) has been intensively investigated as a new class of magneto-electric (ME) multiferroics, whose ferroelectricity is originated from a proper-screw type magnetic ordering. The recent experimental studies have revealed that the ferroelectric helimagnetic phase, which originally shows up as a magnetic-field-induced phase in pure CFO, also shows up as a ground state in slightly diluted  $\text{CuFe}_{1-x}\text{Al}_x\text{O}_2$  (CFAO) and  $\text{CuFe}_{1-x}\text{Ga}_x\text{O}_2$  (CFGO). Although the origin of the ferroelectricity must be common to CFO, CFAO and CFGO, the reported values of the observed macroscopic electric polarization,  $P$ , are rather different from each other. In particular, the previously reported value of  $P$  in CFAO ( $\sim 50\mu\text{C}/\text{m}^2$ ) is remarkably smaller than the values in CFO and CFGO ( $200 \sim 400\mu\text{C}/\text{m}^2$ ).

In the present study, we have thus performed polarized neutron diffraction and in-situ pyroelectric measurements on CFAO, in order to answer the question: "What determines the magnitude of  $P$  in CFO, CFAO and CFGO systems?". The polarized neutron diffraction measurements on CFAO ( $x = 0.015$ ) were carried out at the triple-axis neutron spectrometer PONTA installed by University of Tokyo at JRR-3 in the Japan Atomic Energy Agency (JAEA). Incident polarized neutron beam with energy 34.06 meV was obtained by a Heussler (111) monochromator. The flipping ratio of the polarized neutron beam was  $\sim 14$ . The experimental settings and procedures including pyroelectric measurement were the same as those in the previous polarized neutron diffraction measurement on CFAO ( $x = 0.02$ ).[3]

Since our previous work have revealed a one-to-one correspondence between the

spin helicity and the polarity of the induced ferroelectric polarization, in the present work, we have deduced asymmetry in the fractions of the left-handed and right-handed helical magnetic ordering from the results of the polarized neutron measurements. We refer to this asymmetry of the spin helicity as  $D$ .

Figures 1(a-1) and 1(a-2) show the measured values of  $D$  and  $P$  as functions of  $E_p$ . We found that  $P$  and  $D$  in CFAO were rather insensitive to  $E_p$ . However, we also found that CFAO can achieve  $P$  of  $\sim 250\mu\text{C}/\text{m}^2$ , which is comparable to the values of  $P$  in CFO and CFGO, as shown in the inset of Fig. 1(b). This implies that the magnitude of the local electric polarization in CFO system does not reduced by the Al-substitution, but the sensitivity of  $P$  to  $E_p$  is reduced.

### References

- [1] T. Kimura *et al.*: PRB **73** 220401(2006).
- [2] T. Nakajima *et al.*: JPSJ **76** (2007)043709.
- [3] T. Nakajima *et al.*: PRB **77** 052401 (2007).

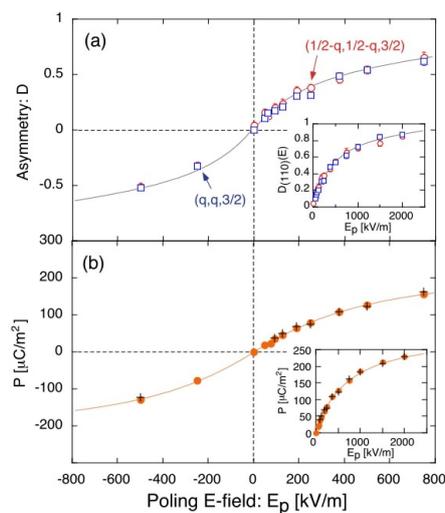


Fig. 1. [(a)-(b)]  $E_p$  dependences of (a) $P$  and (b) $D$  in CFAO ( $x = 0.015$ ) at  $T = 2.0\text{K}$ .