

# Spiral-plane Flop and Rearrangement of Magnetic Domain in Multiferroic Material CuCrO<sub>2</sub>

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Recently, a ferroelectricity induced by a noncollinear spin arrangement has been extensively investigated both experimentally and theoretically. Materials exhibiting such a ferroelectricity are known for “multiferroics,” in which a cycloidal magnetic ordering and a proper-screw magnetic ordering have been reported. In the cycloidal magnetic ordering type such as TbMnO<sub>3</sub>, the microscopic origin of the ferroelectricity can be explained by a spin-current model or the inverse Dzyaloshinskii-Moriya (DM) mechanism. As to the proper-screw magnetic ordering type such as CuCrO<sub>2</sub>, on the other hand, Arima proposed an idea that a d-p hybridization due to the spin-orbit coupling induces the electric polarization  $P$  for such a noncollinear magnetic ordering in a low-symmetry crystal.[1]

The compound chosen in this study is one of the multiferroic TLAs, CuCrO<sub>2</sub>, whose Cr<sup>3+</sup> ions ( $S=3/2$ ) form a triangular lattice. In CuCrO<sub>2</sub>, ferroelectricity with  $P \parallel q$  has been observed at temperature  $T$  below  $T_N \sim 24$  K, where an incommensurate proper-screw spiral magnetic order. As in other spiral-magnetism-induced multiferroics such as TbMnO<sub>3</sub>, a first-order magnetoelectric (ME) phase transition occurs in CuCrO<sub>2</sub>, where the direction of  $P$  is flopped from the [110] to the [1-10] direction by applying  $H \sim 5.3$  T along [1-10].[2] In this study, we use a neutron scattering technique to examine the origins of the ME effects observed in CuCrO<sub>2</sub>.

We revealed that the application of magnetic fields causes a transition from the proper-screw magnetic ordered state into a cycloidal one, as shown in Fig. 1. The transition is associated with a ferroelectric po-

larization flop, i.e., gigantic magnetoelectric effects. Such a transition between two ferroelectric phases with different-types of spiral structures has never been observed in known multiferroics. In addition, we found that the ferroelectric character of the cycloidal phase in magnetic fields cannot be explained by the above well established mechanisms. This means that the cycloidal-induced ferroelectricity of CuCrO<sub>2</sub> is distinct from that in known spiral-magnetism-induced multiferroics. Furthermore, we have found that the distribution of the magnetic domains is strongly affected by a magnetic-field-cooling procedure below a characteristic temperature  $T^* \sim 16$  K, which contributes to the ferroelectric polarization. The direct experimental evidence that the ferroelectric polarization is controlled with the magnetic domains has never been reported.

[1] T. Arima, J. Phys. Soc. Jpn. 76, 073702 (2007).

[2] K. Kimura et al., Phys. Rev. Lett. 103, 107201 (2009).

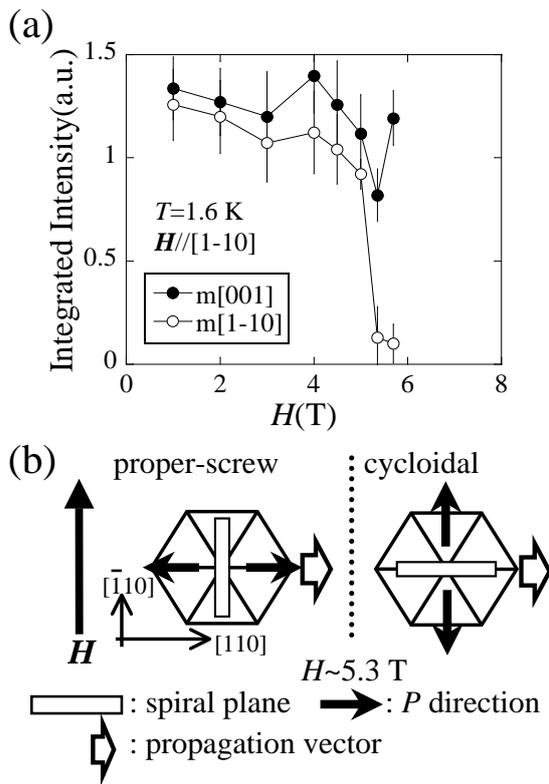


Fig. 1. Fig. 1(a) H-dependence of the integrated intensity of magnetic reflections. (b) Spiral-plane flop from proper-screw magnetic structure to cycloidal one with increasing H.