

## Domain rearrangement and spin-spiral-plane flop as sources of magnetoelectric effects in delafossite $\text{CuCrO}_2$

Minoru Soda,<sup>1</sup> Kenta Kimura,<sup>2</sup> Tsuyoshi Kimura,<sup>2</sup> and Kazuma Hirota<sup>1</sup>

<sup>1</sup>Department of Earth and Space Science, Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

<sup>2</sup>Division of Material Physics, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan

(Received 18 December 2009; revised manuscript received 8 February 2010; published 8 March 2010)

Using neutron scattering, we elucidate the origins of magnetoelectric effects observed in a triangular lattice antiferromagnet,  $\text{CuCrO}_2$ , showing a ferroelectricity associated with a spiral-spin order ( $T_{\text{N}1} \sim 24$  K). Below  $T^* \sim 16$  K, we observed a remarkable difference in the magnetic Bragg reflection and the electric polarization  $P$  between their zero-field-cooling (ZFC) and field-cooling procedures, suggesting that the distribution of magnetic domains coupled with electric ones depends on their cooling procedure. We further demonstrate that one of the domains surviving after the ZFC procedure exhibits a sudden flop of the spiral-spin plane by applying a magnetic field, which is ascribed to the flop of  $P$  observed in  $\text{CuCrO}_2$ .

DOI: [10.1103/PhysRevB.81.100406](https://doi.org/10.1103/PhysRevB.81.100406)

PACS number(s): 75.85.+t, 75.25.-j, 75.60.-d, 77.80.Dj

Recently, it has been widely recognized that frustrated magnetism can be a source of large magnetoelectric (ME) coupling.<sup>1,2</sup> The appearance of a noncollinear spiral-spin order induced by a frustration effect breaks the inversion symmetry and makes the system multiferroic in some cases. Most of the spiral-magnetism-induced multiferroics show cycloidal-spiral-spin structures whose magnetic propagation vectors  $\mathbf{q}$  lie in the spiral planes and whose magnetic symmetries allow the systems to be polar and ferroelectric.<sup>3</sup> The microscopic origin of such a ferroelectricity can be successfully explained by the spin current<sup>4</sup> and the inverse Dzyaloshinskii-Moriya (DM) (Ref. 5) mechanisms. In these multiferroics, the application of magnetic field  $H$  causes large ME effects, i.e., a sudden change in electric polarization  $P$ , which has been discussed in terms of a flop of the spiral-spin plane,<sup>6</sup> a microscopic motion of the domain walls,<sup>7</sup> and so on. Very recently, however, spiral-magnetism-induced multiferroics where the above-mentioned mechanisms are not applicable have been found in some triangular lattice antiferromagnets (TLAs).<sup>8-10</sup> Among them,  $\text{CuFeO}_2$  and  $\text{CuCrO}_2$  with a delafossite structure show ferroelectricity induced by *proper*-screw spiral magnetic structures, where  $\mathbf{q}$  is normal to the spiral plane.<sup>11,12</sup> A microscopic theory which relates the variation in the metal-ligand ( $d$ - $p$ ) hybridization with the spin-orbit coupling to  $P$  (Ref. 13) has been applied to explain the *proper*-screw-spiral-induced ferroelectricity.<sup>14</sup>

The compound chosen in this study is one of the multiferroic TLAs,  $\text{CuCrO}_2$ , whose  $\text{Cr}^{3+}$  ions ( $3d^3, S=3/2$ ) form a triangular lattice. (Indices  $hkl$  are described in the reciprocal space of the hexagonal unit cell.) In  $\text{CuCrO}_2$ , ferroelectricity with  $P \parallel \mathbf{q}$  has been observed at temperature  $T$  below  $T_{\text{N}1} \sim 24$  K,<sup>15</sup> where an incommensurate *proper*-screw spiral magnetic order with the spiral plane parallel to the (110) plane develops [left panel of Fig. 1(a)].<sup>12,16</sup> As in other spiral-magnetism-induced multiferroics such as  $\text{TbMnO}_3$ , a first-order ME phase transition occurs in  $\text{CuCrO}_2$ , where the direction of  $P$  is flopped from the [110] to the  $[\bar{1}\bar{1}0]$  direction at  $T$  much below  $T_{\text{N}1}$  by applying  $H \sim 5.3$  T along  $[\bar{1}\bar{1}0]$ .<sup>17</sup> By considering the magnetic symmetry, it is proposed that the flop of  $P$  arises from the flop of the (110) spin-spiral plane into the  $(\bar{1}\bar{1}0)$  plane. However, no direct evidence has

been provided for the proposal. Due to a sixfold symmetry in the triangular lattice plane, the *proper*-screw spiral structure is triply degenerated (by neglecting doubly degenerated spin chirality), i.e., three magnetic domains exist [domains A–C in Fig. 1(b)]. In the  $d$ - $p$  hybridization model, the direction of  $P$  of each domain is perpendicular to the spiral-spin plane. The existence of such multiple domains makes  $\text{CuCrO}_2$  unique in terms of its ME response to both magnetic and electric fields among spiral-magnetism-induced multiferroics.<sup>15,17</sup> In this Rapid Communication, we use a neutron-scattering technique to examine the origins of the ME effects observed in  $\text{CuCrO}_2$ . It was found that the distribution of the domains is strongly affected by a magnetic-field-cooling (FC) procedure below a characteristic temperature  $T^* \sim 16$  K. We also directly demonstrate that the flop of  $P$  is attributed to a flop of the spiral-spin plane, which corresponds to a transition from a *proper*-screw spiral to a cycloidal-spiral structure.

A single crystal of  $\text{CuCrO}_2$  with dimensions  $\sim 5 \times 3 \times 0.8$  mm<sup>3</sup> was grown by a flux method, as reported previously.<sup>15</sup> Nonpolarized and spin-polarized-neutron-diffraction studies under magnetic fields  $H$  were carried out for the single crystal using the triple axis spectrometer PONTA installed at the JRR-3 reactor of the Japan Atomic Energy Agency. An experimental setup for the spin-polarized neutron measurements is drawn in Fig. 1(c) schematically. The polarization vector  $\mathbf{p}_\text{N}$  of the incident neutrons was controlled by a spin flipper. The flipping ratio of the polarized neutron beam, which was optimized at each  $H$ , was 12–21. The neutron scatterings with the spin flipper on and off correspond to the spin-flip scattering and the nonspin-flip scattering. The crystal was oriented with the [110] and [001] axes in the scattering plane. By a superconducting magnet, magnetic fields were applied along a direction vertical to the scattering plane ( $H \parallel [\bar{1}\bar{1}0]$ ). The direction of  $\mathbf{p}_\text{N}$  was set along  $[\bar{1}\bar{1}0]$  or  $[\bar{1}\bar{1}0]$ . The peak profile of the  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection was measured along the [110] direction. Before the measurements, the crystal was cooled from 35 K ( $>T_{\text{N}1}$ ) to  $\sim 1.6$  K at  $H=0$  or 5.7 T, which we refer to the zero-field-cooling (ZFC) and FC (5.7 T) processes, respectively. Although the system has an incommensurate *proper*-screw

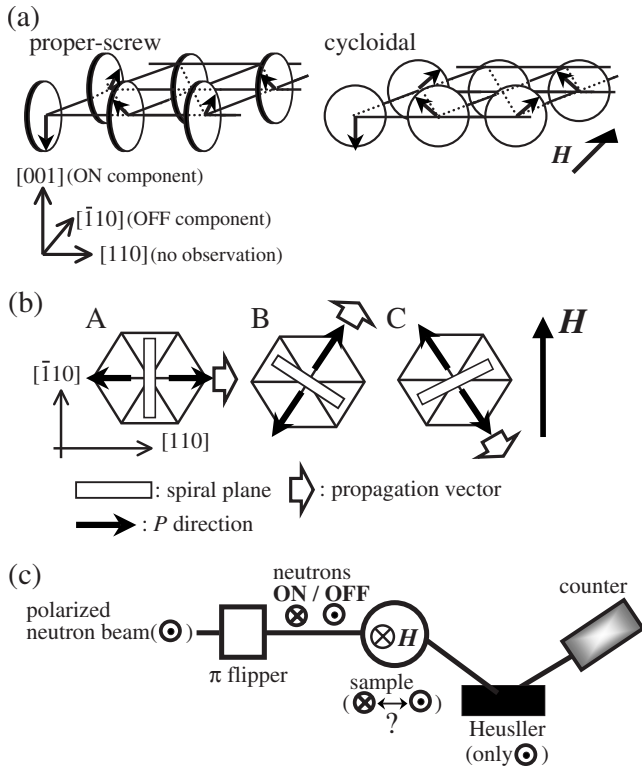


FIG. 1. (a) Schematic magnetic structures of CuCrO<sub>2</sub>. The directions of the observed spin components are also drawn. (b) Schematic magnetic domains of the proper-screw magnetic structure. The  $P$  direction is perpendicular to the spiral plane. (c) Schematic spin-polarized-neutron study geometry.

magnetic structure, as shown in Fig. 1(a) at  $H=0$ ,<sup>12</sup> the magnetic reflection is described as  $(\frac{1}{3}, \frac{1}{3}, 0)$  in the present study because the difference from  $(\frac{1}{3}, \frac{1}{3}, 0)$  is negligible. As mentioned above, the three magnetic domains [A–C in Fig. 1(b)] may coexist below  $T_{N1}$  in the absence of magnetic and electric fields. In the present experimental setup, however, only the A domain is observable at  $H=0$  because the neutron intensities of the B and C domains with the incommensurate propagation vector are zero at  $(\frac{1}{3}, \frac{1}{3}, 0)$ .<sup>12</sup> To compare the neutron-scattering results with the ME properties, measurements of  $P$  with an electric field  $E$  were also independently performed after the cooling processes similar to those for the neutron-scattering measurements. In the previous study, we confirmed that  $E$  affects the ratio of the spin-chirality domains but not that of the A–C domains.<sup>12</sup>

First, the  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection at  $H=0$  was measured by nonpolarized neutrons after ZFC and FC (5.7 T). The inset of Fig. 2(a) shows peak profiles of the magnetic reflection at 1.6 K after these two cooling processes. A clear peak is observed after ZFC while it disappears after FC (5.7 T). This demonstrates that the A domain exists after ZFC but vanishes after FC (5.7 T). The main panel of Fig. 2(a) shows the  $T$  dependence of the integrated intensity of the  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection measured with increasing  $T$  after these two cooling processes. The integrated intensity after the ZFC process (closed circles) gradually decreases with increasing  $T$  and becomes zero above  $T_{N1}$  while that after the FC (5.7 T) process (open circles) emerges with increasing  $T$  from 1.6 K

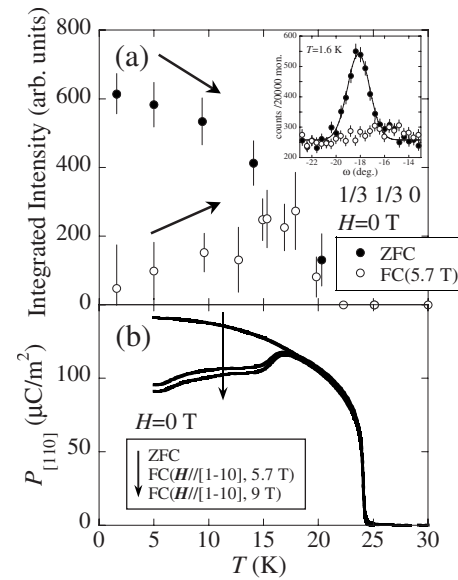


FIG. 2. (a)  $T$  dependence of the integrated intensities of  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection measured by the nonpolarized-neutron-diffraction study. The inset shows the magnetic profile measured at 1.6 K. (b)  $T$  dependence of  $P$  along [110].

and develops with warming up to  $T^* \sim 16$  K. The integrated intensity after FC (5.7 T) reaches a maximum at  $T^*$  and coincides with that after ZFC above  $T^*$ . This indicates that the A domain once removed by FC gradually develops with increasing  $T$  and its volume becomes equal to that after ZFC above  $T^*$ . A similar difference between ZFC and FC (5.7 T) data was also observed in the ME property. Figure 2(b) shows the  $T$ -dependences of  $P$  along [110] at  $H=0$  after ZFC and FC (5.7 and 9 T). At low  $T$ ,  $P$  after FC is smaller than that after ZFC [ $\sim 140 \mu\text{C}/\text{m}^2$  for ZFC and  $\sim 100 \mu\text{C}/\text{m}^2$  for FC (5.7 T) at 5 K]. The difference between the ZFC and the FC data diminishes with increasing  $T$  and disappears above  $T^*$  where the FC data show a distinct anomaly.

Although the above neutron-scattering result confirms that the A domain is removed by the FC procedure, no information on the B and the C domains can be obtained from the result. According to the anisotropy of the magnetic susceptibility perpendicular and parallel to the spiral plane of the proper-screw structure,<sup>15</sup> it is expected that the B and the C domains become more stable than the A domain by FC. Thus, the region occupied by the A domain after ZFC is likely to be replaced with the B or the C domains after FC. This assumption well explains the result of  $P$  shown in Fig. 2(b). According to the  $d$ - $p$  hybridization model,<sup>14</sup> the directions of the magnetoelectrically induced  $P$  of the three magnetic domains are restricted, as shown in Fig. 1(b).<sup>12,15</sup> Here we define  $P_{\text{full}}$  as the electric polarization along [110] for the crystal occupied only by the A domain. Then if the A, B, and C domains are equally distributed in volume after ZFC, the magnitude of  $P$  along [110] can be estimated at  $\frac{2}{3}P_{\text{full}}$  by a simple geometrical calculation. When it is considered that only the B and C domains occupy the crystal after FC, as stated above,  $P$  along [110] after FC can be expected as  $\frac{1}{2}P_{\text{full}}$ . The difference of  $P$  between ZFC and FC ( $P_{\text{ZFC}}:P_{\text{FC}} = \frac{2}{3}:\frac{1}{2}$ ) estimated from this assumption is consis-

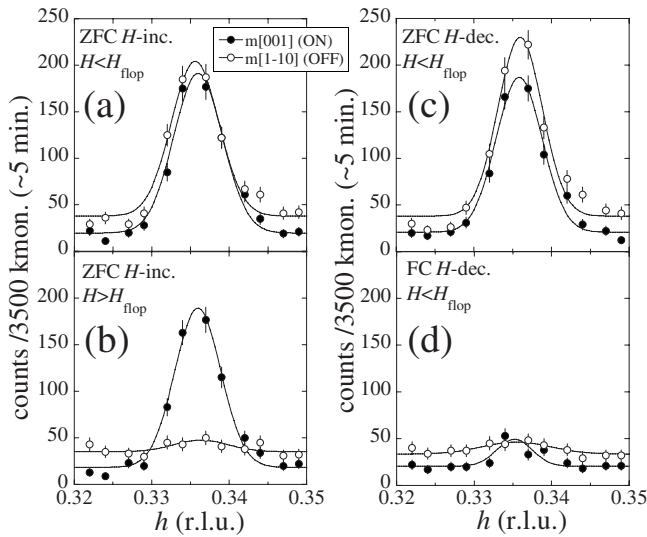


FIG. 3. (a) and (b)  $Q$ -scan profiles of  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection measured at  $H=2$  T ( $<H_{\text{flop}}$ ) and  $H=5.7$  T ( $>H_{\text{flop}}$ ) with increasing  $H$  after ZFC. (c) and (d)  $Q$ -scan profile of  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection measured at  $H=2$  T ( $<H_{\text{flop}}$ ) with decreasing  $H$  after ZFC and FC (5.7 T).

tent with that observed. Thus, FC procedure removes the A domain and rearranges the domain structures, which affects the ME properties of  $\text{CuCrO}_2$ . It is also worth mentioning the existence of a characteristic temperature  $T^*$ , above which the intensity of the  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection and  $P$  along  $[110]$  after FC become equal to those after ZFC. This result indicates that the frozen domain structure consisting of only the B and the C domains after FC is completely melted by thermal energy above  $T^*$ .

To further explore the effect of applied  $H$  along  $[1\bar{1}0]$  ( $H_{[1\bar{1}0]}$ ) on the magnetic domains coupled with ferroelectric domains, which are frozen at low  $T$ , we performed spin-polarized neutron measurements under  $H_{[1\bar{1}0]}$ . In general, only a magnetic moment perpendicular to the scattering vector contributes to a magnetic reflection. For polarized neutrons with the polarization vector  $\mathbf{p}_N$ , furthermore, a magnetic moment parallel to  $\mathbf{p}_N$  produces a nonspin-flip scattering, and a moment perpendicular to  $\mathbf{p}_N$  does a spin-flip scattering. The relation between the observed scattering and the spin components in the present study is drawn schematically in Figs. 1(a) and 1(c). The  $[001]$  component of magnetic moment ( $m_{[001]}$ ) contributes to the spin-flip scattering while the  $[1\bar{1}0]$  component ( $m_{[1\bar{1}0]}$ ) to the nonspin-flip scattering. In the present study, the  $[110]$  component cannot be observed because the scattering vector is parallel to  $[110]$ .

The  $H_{[1\bar{1}0]}$  dependence of the  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection was measured at 1.6 K after both the ZFC and FC (5.7 T) processes. First, we discuss the ZFC results. Figure 3(a) shows  $Q$ -scan profiles of the magnetic reflection with the spin-flip (closed circles) and nonspin-flip (open circles) scattering at a relatively low  $H$  ( $=2$  T) applied at 1.6 K after ZFC. The intensity of the spin-flip scattering is almost the same as that of the nonspin-flip scattering, indicating that  $m_{[1\bar{1}0]}$  and  $m_{[001]}$  are nearly equal. This result is consistent with the incommensurate proper-screw structure [Fig. 1(a)] of the A domain.<sup>12</sup>

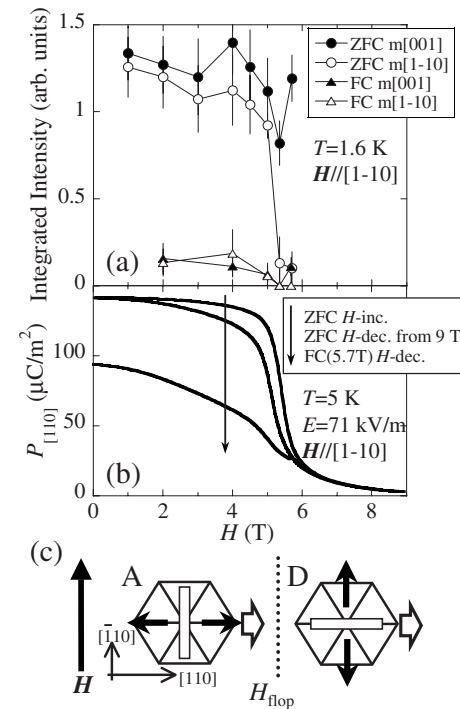


FIG. 4. (a)  $H$  dependence of the integrated intensities of  $\frac{1}{3}\frac{1}{3}0$  magnetic reflections. Close and open marks correspond to  $m_{[001]}$  and  $m_{[1\bar{1}0]}$ , respectively. Circle and triangle marks correspond to ZFC and FC, respectively. (b)  $H$  dependence of  $P$  along  $[110]$ . (c) Spiral-plane flop from A domain to D domain with increasing  $H$ .

As shown in Fig. 3(b), however, the application of high  $H$  ( $=5.7$  T) causes a distinct change in the nonspin-flip scattering. The disappearance of the nonspin-flip scattering means that  $m_{[1\bar{1}0]}$  is zero at 5.7 T applied after ZFC. By contrast, neither the  $(\frac{1}{3}, \frac{1}{3}, 0)$  propagation vector nor the intensity of  $m_{[001]}$  changes at  $H=5.7$  T. These results suggest that a magnetic structural change with keeping the propagation vector occurs between 2 and 5.7 T, as discussed later.

To examine the magnetic structural change in more detail, we show in Fig. 4(a) the  $H_{[1\bar{1}0]}$  dependence of the integrated intensity of  $\frac{1}{3}\frac{1}{3}0$  magnetic reflection measured at 1.6 K after ZFC. The closed and open circles denote  $m_{[001]}$  and  $m_{[1\bar{1}0]}$ , respectively. Both  $m_{[001]}$  and  $m_{[1\bar{1}0]}$  are almost constant and equal below  $\sim 5$  T, indicating that the system has the proper-screw magnetic structure below  $\sim 5$  T. With further increasing  $H$ ,  $m_{[1\bar{1}0]}$  is suddenly suppressed at  $H_{\text{flop}}=5.3$  T and becomes almost zero above  $H_{\text{flop}}$  while the intensity of  $m_{[001]}$  has no such change. Since  $m_{[001]}$  remains at  $\mathbf{q} \sim (\frac{1}{3}, \frac{1}{3}, 0)$ , the transition at  $H_{\text{flop}}$  corresponds to the spin-flop one where  $m_{[1\bar{1}0]}$  parallel to  $H$  changes to  $m_{[110]}$  perpendicular to  $H$ . This means that above  $H_{\text{flop}} \sim 5.3$  T, the system has both  $m_{[001]}$  and  $m_{[110]}$  with  $\mathbf{q} \sim (\frac{1}{3}, \frac{1}{3}, 0)$ , i.e., the cycloidal magnetic structure shown in the right panel of Fig. 1(a). These results indicate that the proper-screw magnetic structure ( $=$ A domain) becomes the cycloidal one at  $H_{\text{flop}} \sim 5.3$  T with increasing  $H$  along  $[1\bar{1}0]$ , as shown in Fig. 4(c). We define this cycloidal magnetic structure with  $\mathbf{q}$  along  $[110]$  as D domain.

When we decreased  $H$  from 5.7 T, the recovery of  $m_{[1\bar{1}0]}$

was observed below  $H_{\text{flop}}$ , as shown in Fig. 3(c). This indicates that the magnetic structural change induced by  $H_{[1\bar{1}0]}$  is reversible. The results after FC (5.7 T) differ considerably from those after ZFC. As shown in Figs. 3(d) and 4(a), the intensities of both  $m_{[001]}$  and  $m_{[1\bar{1}0]}$  are almost zero at all the magnetic fields. This result indicates that the domains with  $\mathbf{q}$  parallel to  $[110]$ , such as the A domain, never appear after the FC procedure.

A remarkable ME effect showing a sudden flop of  $P$  from  $[110]$  to  $[1\bar{1}0]$  has been observed at  $H_{\text{flop}}$  applied along  $[1\bar{1}0]$  in  $\text{CuCrO}_2$ .<sup>17</sup> In Fig. 4(b), we show  $P$ s along  $[110]$  as a function of  $H$  applied along  $[1\bar{1}0]$  at 5 K, which was measured after ZFC or FC (5.7 T). Only in the ZFC data, the  $H$ -dependence of  $P$  shows a sudden drop above  $H_{\text{flop}}$ . The field-cooling procedure suppresses  $P$  along  $[110]$  below  $H_{\text{flop}}$ . The  $H$  profiles of  $P$  measured after the respective cooling procedures are similar to those of the  $m_{[1\bar{1}0]}$  component shown in Fig. 4(a). The sudden change of the spin-polarized neutron intensity at  $H_{\text{flop}}$  contributes to the flop of  $P$  from  $[110]$  to  $[1\bar{1}0]$ , as discussed later.

The above results of both the neutron scattering and the electric polarization after the ZFC procedure can be explained by considering that the A domain transforms into the D domain with the spiral plane perpendicular to  $[1\bar{1}0]$  at  $H_{\text{flop}}$ , as illustrated in Fig. 4(c). This transition between the A and the D domains corresponds to the spiral-plane flop without the  $\mathbf{q}$  change. The magnetic structure of the spin-flopped D domain is cycloidal-spiral one with  $\mathbf{q}$  along  $[110]$  and the spiral axis parallel to  $[1\bar{1}0]$ , as shown in Fig. 1(a). In the spin-current model, it is expected that this cycloidal magnetic structure has  $P$  along  $[001]$ .<sup>4,15</sup> However, no  $P$  is observed along  $[001]$  in the cycloidal magnetic phase ( $H > H_{\text{flop}}$ ), instead  $P$  appears along the  $[1\bar{1}0]$  direction.<sup>17</sup> Such a violation of the spin-current model predictions has never been observed in known cycloidal-spiral-induced multiferroics. This means that the magnetoelectrically induced  $P$  along  $[1\bar{1}0]$  in the cycloidal phase of  $\text{CuCrO}_2$  is unique and cannot be explained by these well established mechanisms. Arima proposed an idea that a  $d$ - $p$  hybridization due to the spin-

orbit coupling induces  $P$  for the proper-screw magnetic structure in the delafossite TLAs,<sup>14</sup> and Seki *et al.*<sup>10</sup> and Kimura *et al.*<sup>15</sup> recently discussed a cycloidal-structure-induced ferroelectricity by a similar symmetry consideration. By considering the  $d$ - $p$  hybridization model, it is expected that the A domain has a macroscopic  $P$  along  $[110]$ , as stated above.<sup>12,14</sup> On the other hand, the magnetic symmetry of the D domain is  $m$  with the mirror plane normal to  $[110]$  and allows  $P$  in any directions normal to  $[110]$  in the  $d$ - $p$  hybridization model. Thus, we consider that the flop of  $P$  is attributed to a spiral plane flop in the A domain, which corresponds to a transition from a proper-screw-spiral structure with  $P//[110]$  into a cycloidal-spiral one with  $P//[1\bar{1}0]$ . It is also worth noting that after the FC procedure no spiral-plane flop is observed because only the A domain surviving after ZFC exhibits the spiral-plane flop.

In conclusion, we have carried out neutron-scattering measurements to clarify the origins of magnetoelectric effects observed in spiral-spin ordered phases for  $\text{CuCrO}_2$ . We found that the distribution of the domains can be controlled by a cooling process, which contributes to the ferroelectric polarization. We also demonstrate that the flop of the ferroelectric polarization observed in one of the magnetic domains arises from the flop of spin-spiral plane, which corresponds to a transition from a proper screw into a cycloidal-spiral-spin structure. The ferroelectricity of the flopped cycloidal phase is unique and cannot be explained by the conventional mechanisms for cycloid-spiral-induced ferroelectrics. Though coupled magnetic and ferroelectric domains have been investigated for several multiferroics,<sup>18,19</sup> a wider variety of possible domains [at least 3(magnetic  $\mathbf{q}$ )  $\times$  2(spin chiral)  $\times$  2(spin flop)] are characteristic of  $\text{CuCrO}_2$  and are responsible for its soft magnetoelastic response to magnetic and electric fields.

We are grateful to T. Arima, H. Yamaguchi, and M. Hagiwara for fruitful discussions. Work at the JRR-3 was supported by ISSP of the University of Tokyo. This work was supported by KAKENHI (Grants No. 19052002, No. 19052001, No. 2001004, No. 20740171, and No. 20674005) and Global COE program (Program No. G10).

<sup>1</sup>S.-W. Cheong and M. Mostovoy, *Nature Mater.* **6**, 13 (2007).

<sup>2</sup>T. Kimura, *Annu. Rev. Mater. Res.* **37**, 387 (2007).

<sup>3</sup>M. Mostovoy, *Phys. Rev. Lett.* **96**, 067601 (2006).

<sup>4</sup>H. Katsura *et al.*, *Phys. Rev. Lett.* **95**, 057205 (2005).

<sup>5</sup>I. A. Sergienko and E. Dagotto, *Phys. Rev. B* **73**, 094434 (2006).

<sup>6</sup>N. Aliouane *et al.*, *Phys. Rev. Lett.* **102**, 207205 (2009).

<sup>7</sup>F. Kagawa *et al.*, *Phys. Rev. Lett.* **102**, 057604 (2009).

<sup>8</sup>M. Kenzelmann *et al.*, *Phys. Rev. Lett.* **98**, 267205 (2007).

<sup>9</sup>T. Kimura *et al.*, *Phys. Rev. B* **73**, 220401(R) (2006).

<sup>10</sup>S. Seki *et al.*, *Phys. Rev. Lett.* **101**, 067204 (2008).

<sup>11</sup>T. Nakajima *et al.*, *J. Phys. Soc. Jpn.* **76**, 043709 (2007).

<sup>12</sup>M. Soda *et al.*, *J. Phys. Soc. Jpn.* **78**, 124703 (2009).

<sup>13</sup>C. Jia *et al.*, *Phys. Rev. B* **74**, 224444 (2006); *Phys. Rev. B* **76**, 144424 (2007).

<sup>14</sup>T. Arima, *J. Phys. Soc. Jpn.* **76**, 073702 (2007).

<sup>15</sup>K. Kimura *et al.*, *Phys. Rev. B* **78**, 140401(R) (2008).

<sup>16</sup>M. Poienar *et al.*, *Phys. Rev. B* **79**, 014412 (2009).

<sup>17</sup>K. Kimura *et al.*, *Phys. Rev. Lett.* **103**, 107201 (2009).

<sup>18</sup>M. Fiebig *et al.*, *Nature (London)* **419**, 818 (2002).

<sup>19</sup>I. Cabrera *et al.*, *Phys. Rev. Lett.* **103**, 087201 (2009).