Rapid Communications

Nonlinear piezomagnetoelectric effect in $CuFeO_2$

H. Tamatsukuri,* S. Mitsuda, T. Shimizu, M. Fujihala, and H. Yokota
Department of Physics, Faculty of Science, Tokyo University of Science, Tokyo 162-8601, Japan

K. Takehana and Y. Imanaka
National Institute for Materials Science, Nano Physics Group, 3-13 Sakura, Tsukuba, Ibaraki 305-0003, Japan

A. Nakao and K. Munakata
Comprehensive Research Organization for Science and Society, Tokai, Ibaraki 319-1106, Japan

(Received 14 August 2019; revised manuscript received 6 November 2019; published 21 November 2019)

We investigate phenomena correlated with multiple degrees of freedom in a geometrically frustrated magnet, $CuFeO_2$, by applying uniaxial pressure p and magnetic field H. Ferroelectric polarization is induced by a combination of p and H only in a sinusoidally amplitude-modulated magnetic structure phase, in which the sinusoidal magnetic ordering does not break the inversion symmetry. The crystal structure and magnetic structure in this phase seem to be unchanged, even by the application of p and H within the present neutron diffraction experiments. These results indicate that this phenomenon differs from conventional spin-driven ferroelectricity as observed in an H-induced helical magnetic phase of $CuFeO_2$. We propose that the induction of ferroelectric polarization by the combined application of p and H in $CuFeO_2$ can be regarded as a nonlinear piezomagnetoelectric effect.

DOI: 10.1103/PhysRevB.100.201105

Cross-correlated phenomena in solids are not only useful for electronic device applications, as exemplified by the piezoelectric effect, but also have promoted fundamental studies in condensed matter physics to understand their coupling mechanism and underlying novel physical concepts. In particular, recent progress in research on magnetoelectric (ME) effects has revealed a nonlinear cross correlation between ferroic order parameters and nonconjugated external fields [1–3], i.e., a ferroelectric polarization (a spontaneous magnetization) can be controlled by a magnetic field (an electric field). To realize nonlinear ME effects, it is important for an order parameter to have relation to its nonconjugated degrees of freedom. In spin-driven ferroelectric materials, for instance, specific magnetic structures break the inversion symmetry in these systems and generate ferroelectric polarization through a spin-orbit interaction or the exchange striction effect [3].

Geometrically frustrated magnets are the leading candidates for spin-driven ferroelectric materials because they tend to exhibit characteristic magnetic structures that break spatial inversion, such as the spiral arrangement of spins, due to the frustration between spins [3–5]. Besides this tendency, the geometrically frustrated magnets are also widely known to often be spin-lattice coupled systems, in which a magnetic phase transition is accompanied by a lattice distortion to partially relieve the frustration [6–8]. Therefore, several degrees of freedom in the geometrically frustrated magnets, such as charge, spin, orbital, and lattice are potentially related to each other, and novel phenomena correlated with multiple degrees of freedom are expected beyond "cross" correlation.

In this Rapid Communication, we demonstrate that ferroelectric polarization is induced in a geometrically frustrated magnet, $CuFeO_2$, only when both the uniaxial pressure p and magnetic field H are simultaneously applied in a specific sinusoidally amplitude-modulated magnetic structure phase. Taking into account that this sinusoidal magnetic ordering does not break the inversion symmetry in $CuFeO_2$, the present results indicate that this phenomenon differs from the conventional spin-driven ferroelectricity as observed in an H-induced helical magnetic phase of $CuFeO_2$, and therefore could be regarded as a nonlinear piezomagnetoelectric effect.

Delafossite $CuFeO_2$ with the space group $R\bar{3}m$ exhibits both spin-driven ferroelectricity and spin-lattice coupling. A Fe^{3+} ion forms a triangular lattice, which is stacked rhombohedrally along the c axis, and carries a magnetic moment ($S = 5/2$) with antiferromagnetic interactions, which results in geometrical frustration. As shown in Fig. 1, various studies have established a highly rich temperature T vs H magnetic phase diagram for $CuFeO_2$ [9–14]. Each of the magnetic orderings in Fig. 1 has a magnetic modulation wave vector $(q, q, 3/2)$ [15]. As the temperature T decreases without H, the paramagnetic (PM) phase becomes a partially disordered (PD) phase at $T_{N1} = 14$ K [16,17]. In the PD phase, the amplitude of spins almost along the c axis is sinusoidally modulated [$q = 0.196$–0.220 (T dependent)] [17]. On further cooling to $T_{N2} = 11$ K, a collinear four-sublattice (4SL) phase is realized, where the magnetic moments are oriented along the c axis in a two-up, two-down sequence ($q = 1/4$)

*Present address: Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan; hiromu.tamatsukuri@kek.jp

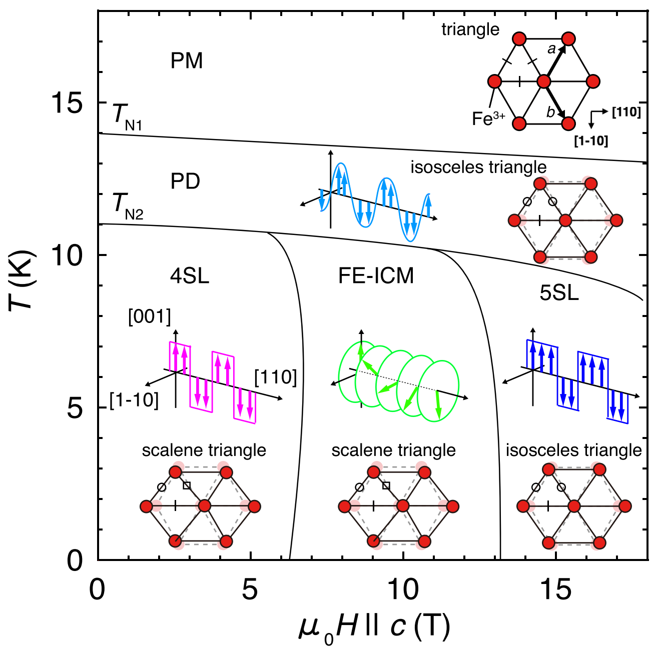

FIG. 1. Schematic $H_{\parallel c}$-T magnetic phase diagram for $CuFeO_2$ under zero applied pressure. Schematic diagrams of magnetic structures and the Fe^{3+} triangular lattices in each phase are also shown.

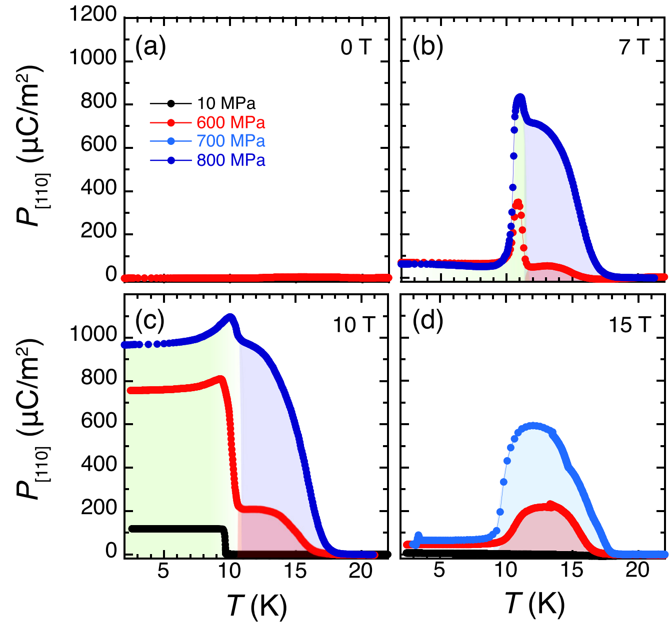

FIG. 2. Temperature dependence of $P_{[110]}$ under applied p at $\mu_0 H \parallel c$ of (a) 0 T, (b) 7 T, (c) 10 T, and (d) 15 T. Data were measured on a heating run after the poling procedure with E_p down to 2 K, except for (d) [35].

[16,17]. These magnetic phase transitions are accompanied by a spontaneous lattice distortion [18,19]. The equilateral triangular lattice in the PM phase turns to be an isosceles triangular lattice in the PD phase (see Fig. 1), and the space group changes from rhombohedral $R\bar{3}m$ to monoclinic $C2/m$ [18,19]. In the lattice distortion, the hexagonal [110] (monoclinic b) axis elongates and the hexagonal [$1\bar{1}0$] (monoclinic a) axis contracts. Therefore, uniaxial pressure p along the [$1\bar{1}0$] direction can be considered as its conjugate field. In the 4SL phase, a space-group ($C2/m$)-forbidden superlattice reflection ($h + k \neq 2n$ with monoclinic notation) was observed [19]. Although the lower space group in the 4SL phase is not identified, the resultant Fe triangular lattice in this phase is well established as a scalene triangular lattice [19–22]. When $H \parallel c$ is applied below T_{N2}, $CuFeO_2$ undergoes successive magnetic phase transitions. In the range of 7 T $\lesssim \mu_0 H \lesssim$ 12 T, a ferroelectric incommensurate (FE-ICM) phase is realized [14]. The FE-ICM phase has a screw helical magnetic structure with $q = 0.202$–0.208 (H dependent), where the screw axis is parallel to the [110] direction [23]. This helical magnetic structure breaks the spatial inversion symmetry of the system and generates a ferroelectric polarization P, along the [110] direction through the Fe $3d$-O $2p$ hybridization mechanism [24] and/or the extended inverse Dzyaloshinskii-Moriya (or spin-current) mechanism [25,26]. A collinear five-sublattice (5SL) phase appears above $\mu_0 H \simeq 12$ T [17,27]. As in the 4SL phase, the magnetic moments in this phase are oriented to the c axis ($q = 1/5$) [17,27]. The application of H has been reported to restore the scalene triangular lattice in the ground state toward an equilateral triangular lattice; the resultant triangular lattices in the FE-ICM and the 5SL phases are scalene and isosceles triangular lattices, respectively [28,29]. Hereafter, the hexagonal notation is used throughout this Rapid Communication for convenience.

A single crystal of $CuFeO_2$ synthesized by the floating zone technique [30] was cut into a rectangular shape with typical dimensions of $1.64 \times 1.37 \times 1.20$ mm^3, in which three axes are along the [110], [$1\bar{1}0$], and [001] directions. Silver paste was painted onto the [110] surfaces to make electrodes for polarization current measurements. An electric polarization with the [110] electrodes $P_{[110]}$ was deduced by time integration of the polarization current measured with an electrometer (Keithley 6517A). Prior to the measurements, a poling electric field E_p (typically, $|E_p| = 152.4$ kV/m), was applied during the cooling process and then removed. Uniaxial pressure p was applied along the [$1\bar{1}0$] direction. For details of the uniaxial pressure devices and the pressure cell, see Refs. [31,32]. Note that the data for $p = 0$ Pa in this work were measured under slightly applied p ($\leqslant$10 MPa) to produce a single q domain state [15,33]. However, application of slight p does not affect the magnetic phase transition temperatures. Magnetic fields H were applied parallel to the c, [110], and [$1\bar{1}0$] directions using a 15-T superconducting magnet installed at the Tsukuba Magnet Laboratory of the National Institute for Materials Science (NIMS). Neutron-diffraction measurements under applied p and $H \parallel c$ were performed using a time-of-flight neutron diffractometer (BL18 SENJU) installed at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC) [34].

Figures 2(a)–2(d) show the T dependence of $P_{[110]}$ under applied p at selected H. Under zero H, no electric polarization was observed, even under an applied p of 600 MPa. This result is consistent with our previous study [36]. At $\mu_0 H =$ 10 T [Fig. 2(c)], $CuFeO_2$ enters the FE-ICM phase below

9.5 K, which results in the emergence of $P_{[110]}$ without the application of p. When p of 600 MPa is applied at 10 T, $P_{[110]}$ begins to emerge at ~17 K, and then exhibits a steplike additional increase at ~10.5 K. The value of $P_{[110]}$ between 10.5 and 17 K is comparable to those of spin-driven ferroelectrics [2,3]. The same feature is also observed at $p =$ 800 MPa. These results are quite similar to those of Ga-doped $CuFeO_2$, of which the ground state and thermally first excited state are also the FE-ICM and PD phases, respectively [37]. Although the temperature where $P_{[110]}$ emerges is significantly increased (from 8.5 to 17 K) by an applied p of 600 MPa in the Ga-doped $CuFeO_2$, neutron diffraction measurements revealed that the temperature where the transition into the FE-ICM phase occurs was increased by only ~1 K under $p = 600$ MPa; therefore, the PD phase becomes another polar phase (described as a FE2 phase in the literature). Taking this similarity into account, we conclude that another polar phase is induced in the PD phase by the application of both p and H [the red (blue) hatched area]. Note that T_{N1} linearly increases with increasing p; for example, $T_{N1} = 19$ K under an applied p of 600 MPa [36,37]. We confirm that the p dependence of T_{N1} is not changed by the application of H. Hereafter, we refer to the polar phase induced by the combined application of p and H as a FE2 phase, although it may differ from the FE2 phase observed in the Ga-doped $CuFeO_2$ system.

When $\mu_0 H = 15$ T and $p \geqslant 600$ MPa are applied [Fig. 2(d)], the FE2 phase also appears in the temperature region corresponding to the (original) PD phase. Of significant note, $P_{[110]}$ vanishes in the 5SL phase [35]. At $\mu_0 H = 7$ T [Fig. 2(b)], $P_{[110]}$ induced by the combined application of p and H can also be observed in the temperature region that corresponds to the PD phase, besides $P_{[110]}$ in the "tip" region of the FE-ICM phase between 10 and 11.5 K. Note that $P_{[110]} \sim 100\ \mu C/m^2$ at 7 T can be observed even at 2 K. This is because the FE-ICM phase partly coexists with the 4SL phase due to the application of p, which has been well documented [31,37].

Figure 3(a) shows the H dependence of $P_{[110]}$ at 13.6 K under applied p. The p-induced $P_{[110]}$ shows a ferroelectric nature, as evidenced by the polarity reversal with dependence on the sign of E_p. Unlike the T dependence, on the other hand, $P_{[110]}$ is gradually induced by the application of H, rather than emerging as a consequence of an explicit phase transition. The magnetic field where $P_{[110]}$ becomes evident shifts to a lower value with increasing p. In contrast, for the case of $H \parallel [110]$ or $[1\bar{1}0]$ [see Fig. 3(b)], $P_{[110]}$ is not observed even under applied p, regardless of the same (original) PD magnetic ordering as in $H \parallel c$. These results clearly indicate that for the emergence of the FE2 phase, it is necessary to apply H parallel to the magnetic moment.

To determine whether or not the crystal and/or the magnetic structure in the PD phase is changed by the application of p and H, time-of-flight neutron diffraction experiments were performed under applied p and H. Because of the complex geometry of experimental components in these measurements, such as a pressure device composed of Al and ZrO_2 pistons and a superconducting magnet, attenuation and absorption of the scattered neutrons could not be corrected for; therefore, the accuracy of the integrated intensities is rather limited in

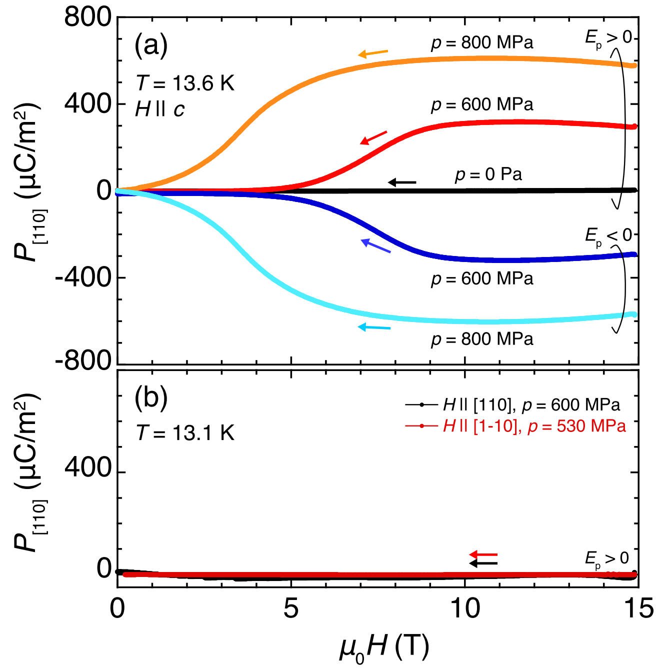

FIG. 3. Magnetic field dependence of $P_{[110]}$ under applied p. Magnetic fields were applied along (a) the c axis, and along (b) the [110] and $[1\bar{1}0]$ axes. Data for (a) and (b) were measured with a decreasing H process after the poling procedure with E_p at 15 T down to 13.6 and 13.1 K, respectively.

spite of the small error bars. However, as shown in Fig. 4, the integrated intensities of both the nuclear and magnetic reflections exhibit almost no change under applied H [38]. Therefore, the change in the crystal and the magnetic structures by the application of p and H is almost indistinguishable within the accuracy and resolution of the present experiments. This result indicates that the modification of the PD crystal/magnetic structures by application of p and H is quite small, if it exists. Thus, this result strongly suggests that $P_{[110]}$ in the FE2 phase is not purely spin driven, as in the FE-ICM phase.

Here, we phenomenologically discuss ferroelectricity induced in $CuFeO_2$ by the combined application of p and H. Induction of electrical polarization using two external fields, a strain σ and H, is known as the piezomagnetoelectric (PME) effect, which is described as $P_i = \pi_{ijkl} H_j \sigma_{kl}$, where π_{ijkl} is the piezomagnetoelectric tensor [39,40]. In the 1990s, detection of the PME effect had been somewhat poor because the PME coefficient (or a matrix element of π_{ijkl} tensor) is small. However, reports on the PME effect [41] and related phenomena [42–44] have been growing due to recent approaches where modern experimental techniques have been employed and multiferroic materials have been targeted. However, the phenomenon observed in the present work should not be the PME effect. Indeed, the H dependence of the p-induced $P_{[110]}$ [Fig. 3(a)] and the p dependence of $P_{[110]}$ at fixed T and H [Figs. 2(b)–2(d)] are not linear. Moreover, π_{ijkl} must vanish when the magnetic point group contains $\bar{1}$ (space inversion) or $1'$ (time inversion) [39,40]. The magnetic point groups in the PD and 4SL phases of $CuFeO_2$, and moreover that in the FE-ICM phase, are $2/m1'$, $2/m1'$, and $21'$, respectively

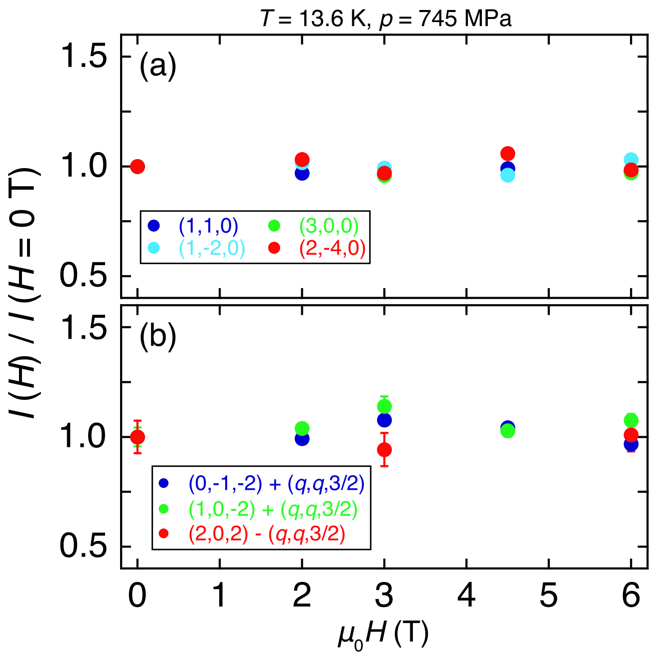

FIG. 4. Relative integrated intensities $I(H)/I(H = 0\ \mathrm{T})$ of neutron diffraction from (a) nuclear reflections and (b) magnetic reflections as a function of $\mu_0 H \parallel c$. Measurements were performed at 13.6 K under an applied p of 745 MPa.

[45], which makes π_{ijkl} vanish, although it is still a possibility that the $2/m1'$ point group in the PD phase is modified by the application of p and H, which results in an active PME effect. Therefore, to classify $P_{[110]}$ induced in $CuFeO_2$ by the combined application of p and H, we propose that the phenomenon in this study is a nonlinear PME effect, by analogy with spin-driven ferroelectricity as a nonlinear ME effect [46].

As described above, important ingredients to induce the phenomenon are the sinusoidal magnetic structure and the application of $H \parallel c$, of which the direction corresponds to the parallel direction of the magnetic moments in the sinusoidal magnetic ordering. When $H \parallel c$ is applied in the PD phase, an explicit magnetic phase transition does not occur, whereas the amplitude of the moments is slightly modified by the application of $H \parallel c$, as evidenced by $2q$ reflections on the forbidden $(h, k, 0)$ plane [10]. Since the amplitude of the moments is sinusoidally modulated in the PD phase, modification of the short moments should be larger than that of the longer moments under applied $H \parallel c$. This situation means that H modification of the moments differs at each of the Fe sites. On the other hand, the application of $H \perp c$ is considered to incline the magnetic moments toward the direction of H, rather than to change the length of them. Therefore, this "site-dependent H modification of the moments" can arise only in the PD phase under applied $H \parallel c$. Although we could not clarify at the present stage how the application of p induces $P_{[110]}$ in combination with the site-dependent H modification of the moments, we point out the possibility that the site-dependent modification of the moments plays an important role in the induction of $P_{[110]}$ in $CuFeO_2$ by the combined application of p and H.

Finally, we consider the relationship between the p-induced FE2 phases in pure $CuFeO_2$ with H and in Ga-doped $CuFeO_2$ without H. As in our previous study [36], the FE2 phase does not emerge in pure $CuFeO_2$ without H, even under an applied p of 800 MPa in the PD phase. In some geometrically frustrated magnets, it is reported that the nonmagnetic impurities affect the system as an effective random field [47–49]. Taking these studies into account, the results of Ga-doped $CuFeO_2$ without H suggest that the nonmagnetic impurities site-randomly affect this system as a magnetic field to some content, which yields a situation similar to the site-dependent H modification of the moments in pure $CuFeO_2$ under applied $H \parallel c$. This suggestion would also be corroborated by the emergence of the FE-ICM phase either by the application of $H \parallel c$ [14] or by the introduction of nonmagnetic impurities [50,51].

In summary, we have searched for other phenomena correlated with multiple degrees of freedom in the geometrically frustrated magnet $CuFeO_2$, by applying uniaxial pressure p and a magnetic field H. Ferroelectric polarization is induced in the original PD phase by the application of p together with H. The crystal and magnetic structures seem to be unchanged by the application of p and H, which suggests that this phenomenon differs from conventional spin-driven ferroelectricity, as in the FE-ICM phase. We propose that the induction of ferroelectric polarization in $CuFeO_2$ by the combined application of p and H can be regarded as a nonlinear PME effect.

The neutron experiments were performed at J-PARC/MLF under user Programs No. 2015A0090 and No. 2017A0173. This work was supported by the NIMS Joint Research Hub Program and partly by Grants-in-Aid for Scientific Research (C) (No. 26400369) from the Japan Society for the Promotion of Science.

[1] T. Kimura, T. Goto, H. Shintani, K. Ishizaka, T. Arima, and Y. Tokura, Nature (London) **426**, 55 (2003).
[2] S. W. Cheong and M. Mostovoy, Nat. Mater. **6**, 13 (2007).
[3] Y. Tokura, S. Seki, and N. Nagaosa, Rep. Prog. Phys. **77**, 076501 (2014).
[4] T. Kimura and Y. Tokura, J. Phys.: Condens. Matter **20**, 434204 (2008).
[5] T. Arima, J. Phys. Soc. Jpn. **80**, 052001 (2011).
[6] Y. Yamashita and K. Ueda, Phys. Rev. Lett. **85**, 4960 (2000).
[7] F. Becca and F. Mila, Phys. Rev. Lett. **89**, 037204 (2002).
[8] D. L. Bergman, R. Shindou, G. A. Fiete, and L. Balents, Phys. Rev. B **74**, 134409 (2006).
[9] S. Mitsuda, M. Mase, T. Uno, H. Kitazawa, and H. A. Katori, J. Phys. Soc. Jpn. **69**, 33 (2000).
[10] S. Mitsuda, M. Mase, K. Prokes, H. Kitazawa, and H. A. Katori, J. Phys. Soc. Jpn. **69**, 3513 (2000).
[11] O. A. Petrenko, M. R. Lees, G. Balakrishnan, S. de Brion, and G. Chouteau, J. Phys.: Condens. Matter **17**, 2741 (2005).

[12] G. Quirion, M. L. Plumer, O. A. Petrenko, G. Balakrishnan, and C. Proust, Phys. Rev. B **80**, 064420 (2009).
[13] T. T. A. Lummen, C. Strohm, H. Rakoto, and P. H. M. van Loosdrecht, Phys. Rev. B **81**, 224420 (2010).
[14] T. Kimura, J. C. Lashley, and A. P. Ramirez, Phys. Rev. B **73**, 220401(R) (2006).
[15] Owing to the trigonal symmetry along the c axis of the $R\bar{3}m$ space group, the other two magnetic modulation vectors of $(-2q, q, 3/2)$ and $(q, -2q, 3/2)$ are equivalent to $(q, q, 3/2)$. This leads to a formation of three structural/magnetic domains, called a q domain in this Rapid Communication.
[16] M. Mekata, N. Yaguchi, T. Takagi, T. Sugino, S. Mitsuda, H. Yoshizawa, N. Hosoito, and T. Shinjo, J. Phys. Soc. Jpn. **62**, 4474 (1993).
[17] S. Mitsuda, N. Kasahara, T. Uno, and M. Mase, J. Phys. Soc. Jpn. **67**, 4026 (1998).
[18] F. Ye, Y. Ren, Q. Huang, J. A. Fernandez-Baca, P. Dai, J. W. Lynn, and T. Kimura, Phys. Rev. B **73**, 220404(R) (2006).
[19] N. Terada, S. Mitsuda, H. Ohsumi, and K. Tajima, J. Phys. Soc. Jpn. **75**, 023602 (2006).
[20] N. Terada, S. Mitsuda, Y. Tanaka, Y. Tabata, K. Katsumata, and A. Kikkawa, J. Phys. Soc. Jpn. **77**, 054701 (2008).
[21] S. Kimura, T. Fujita, N. Nishihagi, H. Yamaguchi, T. Kashiwagi, M. Hagiwara, N. Terada, Y. Sawai, and K. Kindo, Phys. Rev. B **84**, 104449 (2011).
[22] T. Nakajima, A. Suno, S. Mitsuda, N. Terada, S. Kimura, K. Kaneko, and H. Yamauchi, Phys. Rev. B **84**, 184401 (2011).
[23] T. Nakajima, S. Mitsuda, S. Kanetsuki, K. Prokes, A. Podlesnyak, H. Kimura, and Y. Noda, J. Phys. Soc. Jpn. **76**, 043709 (2007).
[24] T. Arima, J. Phys. Soc. Jpn. **76**, 073702 (2007).
[25] T. A. Kaplan and S. D. Mahanti, Phys. Rev. B **83**, 174432 (2011).
[26] J. T. Zhang, C. Ji, J. L. Wang, W. S. Xia, B. X. Guo, X. M. Lu, and J. S. Zhu, Phys. Rev. B **96**, 235136 (2017).
[27] T. Fukuda, H. Nojiri, M. Motokawa, T. Asano, M. Mekata, and Y. Ajiro, J. Phys. Chem. Solids **60**, 1249 (1999).
[28] N. Terada, Y. Tanaka, Y. Tabata, K. Katsumata, A. Kikkawa, and S. Mitsuda, J. Phys. Soc. Jpn. **75**, 113702 (2006).
[29] N. Terada, Y. Narumi, Y. Sawai, K. Katsumata, U. Staub, Y. Tanaka, A. Kikkawa, T. Fukui, K. Kindo, T. Yamamoto, R. Kanmuri, M. Hagiwara, H. Toyokawa, T. Ishikawa, and H. Kitamura, Phys. Rev. B **75**, 224411 (2007).
[30] T. R. Zhao, M. Hasegawa, and H. Takei, J. Cryst. Growth **166**, 408 (1996).
[31] T. Nakajima, S. Mitsuda, K. Takahashi, K. Yoshitomi, K. Masuda, C. Kaneko, Y. Honma, S. Kobayashi, H. Kitazawa, M. Kosaka, N. Aso, Y. Uwatoko, N. Terada, S. Wakimoto, M. Takeda, and K. Kakurai, J. Phys. Soc. Jpn. **81**, 094710 (2012).
[32] T. Nakajima, Y. Tokunaga, V. Kocsis, Y. Taguchi, Y. Tokura, and T.-h. Arima, Phys. Rev. Lett. **114**, 067201 (2015).
[33] T. Nakajima, S. Mitsuda, T. Haku, K. Shibata, K. Yoshitomi, Y. Noda, N. Aso, Y. Uwatoko, and N. Terada, J. Phys. Soc. Jpn. **80**, 014714 (2011).
[34] T. Ohhara, R. Kiyanagi, K. Oikawa, K. Kaneko, T. Kawasaki, I. Tamura, A. Nakao, T. Hanashima, K. Munakata, T. Moyoshi, T. Kuroda, H. Kimura, T. Sakakura, C.-H. Lee, M. Takahashi, K.-i. Ohshima, T. Kiyotani, Y. Noda, and M. Arai, J. Appl. Crystallogr. **49**, 120 (2016).
[35] It should be noted that $P_{[110]} \simeq 40$ (65) μC/m^2 seems to exist also in the 5SL phase under an applied p of 600 (800) MPa, respectively. This is because data under applied $p \geqslant 600$ MPa below 14 K were measured on a cooling run after the $E_{\rm p}$ poling is conducted down to ~14 K. Therefore, we cannot deny in this experiment a possibility that owing to the application of p, the 5SL phase as well as the PD phase become another polar phase. This point is desired to be clarified in a future study.
[36] H. Tamatsukuri, S. Aoki, S. Mitsuda, T. Nakajima, T. Nakamura, T. Itabashi, S. Hosaka, S. Ito, Y. Yamasaki, H. Nakao, K. Prokes, and K. Kiefer, Phys. Rev. B **94**, 174402 (2016).
[37] H. Tamatsukuri, S. Mitsuda, T. Nakamura, K. Takata, T. Nakajima, K. Prokes, F. Yokaichiya, and K. Kiefer, Phys. Rev. B **95**, 174108 (2017).
[38] Under zero H, we basically confirmed that the magnetic structures in $CuFeO_2$ (and also Ga-doped $CuFeO_2$) were not changed by applied p up to 600 MPa, although these measurements were restricted within a (H, H, L) plane [36,37].
[39] H. Grimmer, Acta Crystallogr. Sect. A **48**, 266 (1992).
[40] J. P. Rivera and H. Schmid, Ferroelectrics **161**, 91 (1994).
[41] V. D. Fil, M. P. Kolodyazhnaya, G. A. Zvyagina, I. V. Bilych, and K. R. Zhekov, Phys. Rev. B **96**, 180407(R) (2017).
[42] T. Nakajima, Y. Tokunaga, Y. Taguchi, Y. Tokura, and T.-h. Arima, Phys. Rev. Lett. **115**, 197205 (2015).
[43] Y. Shiomi, H. Watanabe, H. Masuda, H. Takahashi, Y. Yanase, and S. Ishiwata, Phys. Rev. Lett. **122**, 127207 (2019).
[44] T. N. Gaydamak, I. A. Gudim, G. A. Zvyagina, I. V. Bilych, N. G. Burma, K. R. Zhekov, and V. D. Fil, Phys. Rev. B **92**, 214428 (2015).
[45] N. Terada, N. Qureshi, L. C. Chapon, and T. Osakabe, Nat. Commun. **9**, 4368 (2018).
[46] This means that our results should be explained by an additional coupled term in a free energy in this system, as in the spin-driven ferroelectric materials [52], not higher-order terms of $\pi_{ijkl} H_j \sigma_{kl}$.
[47] J. van Duijn, B. D. Gaulin, M. A. Lumsden, J. P. Castellan, and W. J. L. Buyers, Phys. Rev. Lett. **92**, 077202 (2004).
[48] M. Mekata, T. Tatsumi, T. Nakashima, K. Adachi, and Y. Ajiro, J. Phys. Soc. Jpn. **56**, 4544 (1987).
[49] T. Nakajima, S. Mitsuda, K. Kitagawa, N. Terada, T. Komiya, and Y. Noda, J. Phys.: Condens. Matter **19**, 145216 (2007).
[50] S. Seki, Y. Yamasaki, Y. Shiomi, S. Iguchi, Y. Onose, and Y. Tokura, Phys. Rev. B **75**, 100403(R) (2007).
[51] N. Terada, T. Nakajima, S. Mitsuda, H. Kitazawa, K. Kaneko, and N. Metoki, Phys. Rev. B **78**, 014101 (2008).
[52] M. Mostovoy, Phys. Rev. Lett. **96**, 067601 (2006).