First-order multi-k phase transitions and magnetoelectric effects in multiferroic Co$_3$TeO$_6$

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A theoretical description of the sequence of magnetic phases in Co$_3$TeO$_6$ is presented. The strongly first-order character of the transition to the commensurate multiferroic ground state, induced by coupled order parameters corresponding to different wave vectors, is related to a large magnetoelastic effect with an exchange energy critically sensitive to the interatomic spacing. The monoclinic magnetic symmetry $C2'$ of the multiferroic phase permits spontaneous polarization and magnetization as well as the linear magnetoelectric effect. The existence of weakly ferromagnetic domains is verified experimentally by second harmonic generation measurements.

At variance with structural transitions which alter the lengths and orientations of the chemical bonds, the spin ordering occurring in magnetic phases has in most cases a negligible effect on the structural lattice. An important exception is represented by the class of magnetostructural transitions occurring in multiferroic compounds in which the magnetic ordering in the multiferroic phase induces simultaneously a change in the atomic structure, which permits the emergence of a spontaneous polarization.[1,2] However, the measured changes of lattice parameters found in the multiferroic phases are generally small (i.e., of the order of 10^{-3}Å) and do not affect the second-order character of the transitions to these phases.[3,4] Here, we describe theoretically the sequence of phases recently reported in Co$_3$TeO$_6$ (Refs. 5 and 6) in which a strongly first-order transition, characterized by substantial discontinuities of the lattice parameters and a remarkable delta-shaped peak of the specific heat, yields a multiferroic ground state displaying magnetoelectric properties. The observed structural changes are related to the coupling between the magnetic order parameters involved at the transition, which correspond to different propagation wave vectors, in contrast with the standard situation found in multiferroic transitions where the coupled order parameters generally pertain to the same k vector.[3,7]

Neutron powder diffraction studies[6,8] show that below the paramagnetic phase described by the space group $G_P = C2/c1'$ the Co$_3$TeO$_6$ undergoes a sequence of three antiferromagnetic phases, summarized in Fig. 1.[9] They are associated with three different k vectors of the centered monoclinic Brillouin zone: $\vec{k}_1 = (0,0.480,0.055)$, $\vec{k}_2 = (0,0,0)$, and $\vec{k}_3 = (0,1/2,1/4)$.

1. The incommensurate Phase I, which emerges at $T_N = 26$ K, shows the coexistence of $\vec{k}_1$ and $\vec{k}_2$. The transition order parameter transforming as the irreducible representation (IR) $\tau_1(\vec{k}_1)$ (see Table I)[10] has four components ($\eta_1 = \rho_1 e^{i\theta_1}$, $\eta_1^* = \rho_1 e^{-i\theta_1}$, $\eta_2 = \rho_2 e^{i\theta_2}$, $\eta_2^* = \rho_2 e^{-i\theta_1}$) which yield the free energy

$$F_1(\rho_1,\rho_2) = \frac{\alpha_1}{2}\left(\rho_1^2 + \rho_2^2\right) + \frac{\beta_1}{4}\left(\rho_1^4 + \rho_2^4\right) + \frac{\beta_2}{2}\rho_1^2\rho_2^2. \quad (1)$$

Minimizing F_1 leads to two possibly stable states having the same point-group symmetry $2/m1'$ as the paramagnetic phase, and corresponding to the equilibrium conditions ($\rho_1 \neq 0, \rho_2 = 0$) and ($\rho_1 = \pm\rho_2$). The point group symmetry $2/m1'$ of phase I and its incommensurate character are preserved when considering a coupling with the order parameter associated with $\vec{k}_2$.

2. The commensurate phase II appearing at $T_1 = 21.1$ K below a second-order transition corresponds to the single wave vector $\vec{k}_2$. It remains stable in a narrow interval of temperature down to $T_2 = 17.4$ K. The one-dimensional IRs Γ_1–Γ_4 at the center of the monoclinic Brillouin zone[10] induce, respectively, the magnetic symmetries $C2/c$ (Γ_1), $C2'/c'$ (Γ_2), $C2/c'$ (Γ_3), and $C2'/c$ (Γ_4). The magnetic structure proposed by Ivanov *et al.*[6] from neutron data coincides with the $C2'/c$ magnetic group which is therefore associated with a one-dimensional order parameter, denoted ζ hereafter, corresponding to the single equilibrium state which minimizes below T_1 the canonical free energy

$$F_2(\zeta) = \frac{\alpha_2}{2}\zeta^2 + \frac{\lambda_1}{4}\zeta^4. \quad (2)$$

In the absence of applied fields the magnetic symmetry of the phase does not allow the emergence of spontaneous polarization or magnetization components.

3. At $T_2 = 17.4$ K a commensurate phase III arises in which the neutron diffraction pattern corresponding to $\vec{k}_2$ persists coexisting with magnetic peaks associated with the commensurate wave vector $\vec{k}_3$. Both sets of reflections are observed in the whole range of stability of the phase down to 1.6 K (Ref. 6). $\vec{k}_3$ is in general a position inside the Brillouin zone, corresponding to a four-dimensional IR of G_P, denoted $\tau_1(\vec{k}_3)$, whose matrices are listed in Table I. Keeping for the four order-parameter components the same notation ($\eta_i = \rho_i e^{\pm i\theta_i}, i = 1,2$) as for phase I the transition free energy reads

$$F_3(\rho_1,\rho_2,\theta_1,\theta_2) = F_1(\rho_1,\rho_2) + \frac{\beta_3}{4}\left(\rho_1^4\cos 4\theta_1 + \rho_2^4\cos 4\theta_2\right)$$
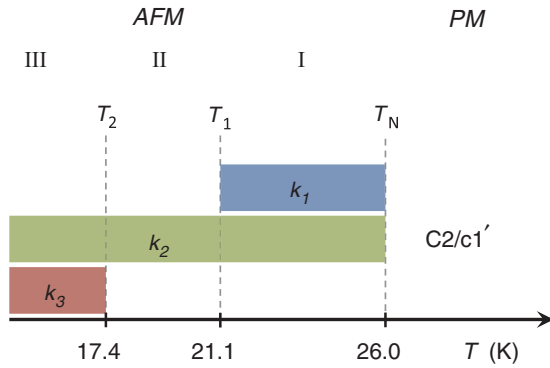$$+ \frac{\beta_4}{2}\rho_1^2\rho_2^2\sin 2(\theta_1 + \theta_2) + \cdots, \quad (3)$$

FIG. 1. (Color online) Sequence of phases observed in Co_3TeO_6 from Ref. 6.

which differs from F_1 by the β_3 and β_4 lock-in invariants. Phase III results from the coupling of the order parameters $\eta_i(\vec{k}_3)$ and $\zeta(\vec{k}_2)$ corresponding to the total free energy

$$F_T = F_3(\rho_i, \theta_i) + F_2(\zeta) - \frac{\delta}{2}\zeta^2(\rho_1^2 + \rho_2^2), \qquad (4)$$

where the δ term represents the lowest-degree coupling between the two order parameters. Table II lists the symmetries and equilibrium conditions of the seven possibly stable magnetic phases resulting from the minimization of F_T. The phase with magnetic symmetry $C2'$ and a 16-fold multiplication of the primitive paramagnetic unit cell, shown in Fig. 2(a), coincides unambiguously with the reported neutron diffraction observations[6] in phase III. It corresponds to the equilibrium values of the order parameters $\zeta \neq 0, \rho_1 = \rho_2 = \rho_e, \theta_1 = \theta_2 = \theta_e$ which allow the emergence of a spontaneous polarization P_y and a spontaneous weak magnetization $\vec{M} = (M_x, M_z)$, with, respectively, two ferroelectric and two weakly ferromagnetic domains [Fig. 2(b)].

Figure 3 shows the distribution of magnetic domains in the monoclinic xz plane. The image was gained by optical second harmonic generation (SHG) as described by the authors of Ref. 5. The orientation of the domain walls along an arbitrary direction in the xz plane further supports the $2'$ symmetry. This symmetry is also consistent with the presence of the χ_{xxx} and χ_{zzz} components of the SHG susceptibility tensor.[5] Note, however, that it differs from the symmetry m assumed in Ref. 5. In Ref. 5 only i-tensor components were considered

as the origin of the SHG signal since in magnetically induced ferroelectrics like $MnWO_4$ the SHG signal is always linearly coupled to the spontaneous polarization. In contrast, the SHG signal leading to Fig. 3 reveals that SHG in CTO is related to c-tensor components reproducing the weakly ferromagnetic order. SHG with $\chi_{xxx} \neq 0$, $\chi_{yyy} = 0$, and $\chi_{zzz} \neq 0$ as c-type susceptibilities leads to the magnetic symmetry $2'$. Since this is a very recent result, a discussion of our SHG data in Ref. 11 is still based on the assumption of SHG coupling to the electric polarization which lead to results inconsistent with the ones reported in this work.

Since the transitions to phases I and III result from the coupling of two order parameters corresponding to distinct k vectors they display necessarily a first-order character[12] following the triggering mechanism proposed by Holakovsky[13] in which one order parameter triggers the onset of another order parameter across the first-order discontinuity. The mechanism requires taking into account a sixth-degree invariant of the "triggered" order parameter $[\frac{\gamma}{6}(\rho_1^2 + \rho_2^2)^3]$ in the total free energy F_T. Under the conditions $\delta > (\frac{|\beta_1 + \beta_2|\lambda_1}{2})^{1/2}$ and $\lambda_1 - \frac{4\delta^2}{|\beta_1 + \beta_2|} < -4(\frac{\alpha_2\gamma}{3})^{1/2}$, the phase corresponding to the sole "triggering" order parameter ζ becomes unstable with respect to a phase in which both order parameters ζ and $\eta \sim (\rho_i, \theta_i)$, are nonzero. In this phase ζ is frozen and η determines the symmetry-breaking process. The transition can be shown to occur discontinuously at a higher temperature than the transition temperature at which a phase with $\zeta = 0, \rho_i \neq 0$ would appear.[13] Accordingly, the triggering process, which is activated in the region of phase coexistence preceding the transition at T_2, is due to a large *negative* value of the interaction term for the coupling between the two order parameters, which determines the value of the coupling coefficient δ. Note that the first-order character of the transition to the multiferroic phase at T_2 is confirmed by the strong discontinuities (of the order of 10^{-2} Å) observed in the lattice parameters[6] and by a remarkably sharp peak of the specific heat.[5,6,14] In contrast, the transition from the paramagnetic to the antiferromagnetic phase I is weakly first order with almost negligible lattice discontinuities and a standard specific heat anomaly.[5,6] The transition from phase I to phase II which involves a single order parameter has typical second-order transition features with no noticeable discontinuity of the lattice parameters.[5,6]

TABLE I. Generators of the active irreducible representations of the paramagnetic space-group $C2/c1'$ associated with the wave vectors $\vec{k}_1$, $\vec{k}_2$, and $\vec{k}_3$ in Co_3TeO_6. Columns matrices represent diagonal matrices. T is the time-reversal operator. $\epsilon = \exp(ik_1^z c)$, $\omega = \exp(ik_1^y b/2)$.

	$(2_y\|0,0,\frac{c}{2})$	$(\bar{1}\|0,0,0)$	T	$(1\|\frac{a}{2},\frac{b}{2},0)$	$(1\|0,0,c)$
$\tau_1(\vec{k}_1)$	$\begin{bmatrix} & & & 1 \\ & & 1 & \\ 1 & & & \\ & 1 & & \end{bmatrix}$	$\begin{bmatrix} & 1 & & \\ 1 & & & \\ & & & \epsilon^* \\ & & \epsilon & \end{bmatrix}$	$\begin{bmatrix} -1 & & & \\ & -1 & & \\ & & -1 & \\ & & & -1 \end{bmatrix}$	$\begin{bmatrix} \omega & & & \\ & \omega^* & & \\ & & \omega & \\ & & & \omega^* \end{bmatrix}$	$\begin{bmatrix} \epsilon & & & \\ & \epsilon^* & & \\ & & \epsilon^* & \\ & & & \epsilon \end{bmatrix}$
$\Gamma_4(\vec{k}_2)$	$-\mathbf{1}$	$-\mathbf{1}$	$-\mathbf{1}$	$\mathbf{1}$	$\mathbf{1}$
$\tau_1(\vec{k}_3)$	$\begin{bmatrix} & & & 1 \\ & & 1 & \\ 1 & & & \\ & 1 & & \end{bmatrix}$	$\begin{bmatrix} & 1 & & \\ 1 & & & \\ & & -i & \\ & & & i \end{bmatrix}$	$\begin{bmatrix} -1 & & & \\ & -1 & & \\ & & -1 & \\ & & & -1 \end{bmatrix}$	$\begin{bmatrix} i & & & \\ & -i & & \\ & & i & \\ & & & -i \end{bmatrix}$	$\begin{bmatrix} i & & & \\ & -i & & \\ & & -i & \\ & & & i \end{bmatrix}$

TABLE II. Seven possible choices (a) of the magnetic space groups (b) derived from the minimization of $F_T(\zeta, \rho_i, \theta_i)$ in Eq. (4). (c) Equilibrium values of the order parameters. (d) Basic translations of the conventional monoclinic or triclinic unit cells. (e) Multiplicity of the volume of the primitive paramagnetic unit cell. (f) Origin of the coordinates.

(a)	(b)	(c)	(d)	(e)	(f)
1	$P\bar{1}'$	$\zeta \neq 0, \rho_1 \neq 0, \rho_2 = 0, \theta_1 = 0$		4	$(0, 1/2, 0)$
2	$P\bar{1}'$	$\zeta \neq 0, \rho_1 \neq 0, \rho_2 = 0, \theta_1 = \pi/4$	$(-1, 0, 0), (1/2, 3/2, 1), (1/2, 1/2, -1)$	4	$(1/4, 3/4, 0)$
3	$P1$	$\zeta \neq 0, \rho_1 \neq 0, \rho_2 = 0$		4	$(0, 0, 0)$
4	$C2'$	$\zeta \neq 0, \rho_1 = \rho_2, \theta_1 = \theta_2$	$(0, 0, 4), (0, 2, 0), (-1, 0, 0)$	16	$(0, 5/8, 5/4)$
5	$P\bar{1}'$	$\zeta \neq 0, \rho_1 \neq 0, \rho_2 \neq 0, \theta_1 = 0, \theta_2 = -\pi/4$		8	$(0, 1/2, 0)$
6	$P\bar{1}'$	$\zeta \neq 0, \rho_1 \neq 0, \rho_2 \neq 0, \theta_1 = \pi/2, \theta_2 = -\pi/4$	$(0, 2, 0), (1, 0, 0), (0, 1, -2)$	8	$(1/4, 1/4, -1/2)$
7	$P1$	$\zeta \neq 0, \rho_1 \neq 0, \rho_2 \neq 0$		8	$(0, 0, 0)$

The dielectric contribution to the free energy $F_D = -\nu P_y \zeta^2 [\rho_1^2 \rho_2^2 \cos 2(\theta_1 + \theta_2)] + \frac{P_y^2}{2\epsilon_{yy}^0}$ yields the equilibrium value of P_y below T_2:

$$P_y = \nu \epsilon_{yy}^0 \zeta^2 \rho_e^4 \cos 4\theta_e. \tag{5}$$

At $T = T_2$, P_y undergoes an upward discontinuity, imposed by the first-order character of the transition. On further cooling it increases as $\approx (T_2 - T)^2$ since the ζ order parameter is frozen in phase III. A similar temperature dependence holds for the spontaneous magnetization components M_x and M_z. From the spontaneous magnetic contribution to the free energy in phase III $F_M = \mu_u M_u \zeta \rho_1^2 \rho_2^2 \cos 2(\theta_1 + \theta_2) + \frac{M_u^2}{2\chi_{uu}^0}$ (with $u = x, z$), one gets

$$M_u = -\chi_{uu}^0 \mu_u \zeta \rho_e^4 \cos 4\theta_e \tag{6}$$

as the equilibrium value below T_2. The application of magnetic fields along x or z yields a renormalization of the transition temperature according to

$$T_2(H_u) = T_2(0) - \chi_{uu}^0 \mu_u \alpha_0^{-1} \zeta^2 H_u^2. \tag{7}$$

Where $\alpha = \alpha_0[T - T_2(0)]$ is the coefficient of ρ_e^2 in F_T. For $\mu_u > 0$ the transition temperature is lowered under the application of a magnetic field and $T_2(0) - T_2(H_u)$ increases quadratically with H_u, as observed in Co_3TeO_6 under the H_z field. The magnetic susceptibility components $\chi_{uu} = \frac{M_u}{H_u}$ are obtained by minimizing the field-induced contribution to the free energy $F_M(H_u) = \mu_u M_u H_u \zeta^2 \rho_e^2 + \frac{M_u^2}{2\chi_{uu}^0} - H_u M_u$. This reveals

$$\chi_{uu} = \chi_{uu}^0 (1 - \mu_u \zeta^2 \rho_e^2). \tag{8}$$

Therefore the discontinuous *jump* of the order parameter $\rho_e(T_2)$ coincides with a *drop* of $\chi_{uu}(T_2)$ the magnitude of which decreases with increasing field up to a threshold field corresponding to $\rho_e(H_u^c) = \frac{1}{\zeta \sqrt{\mu_u}}$ above which χ_{uu} undergoes an *upward* discontinuity at $T_2(H_u)$. This behavior is verified experimentally[5] for H_z with a threshold field $H_z^c \approx 12$ T. The slight decrease with temperature observed for χ_{zz} *below* T_2 indicates a slight increase of the order parameter within
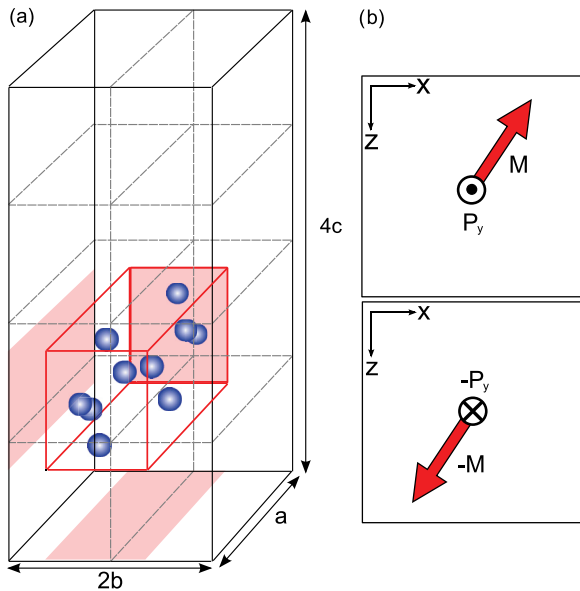

FIG. 2. (Color online) (a) Paramagnetic unit cell embedded into the 16-fold unit cell of the multiferroic phase III of Co_3TeO_6. (b) Ferroelectric and weak ferromagnetic domains in phase III. In the text we use Cartesian coordinates instead of the monoclinic coordinates according to $x \sim a$, $y \sim b$, $z \sim c$.
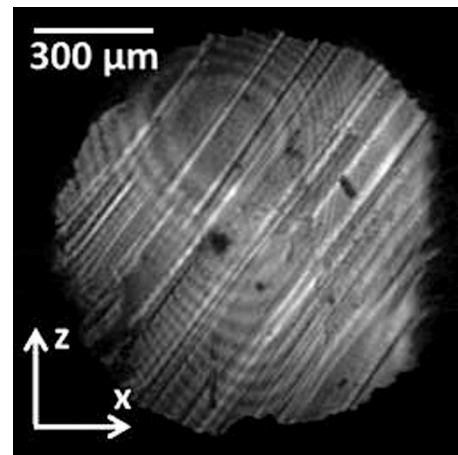

FIG. 3. SHG image of a polished single-crystal platelet (thickness $50 \,\mu$m) of Co_3TeO_6 at $T = 5$ K. Diagonal parallel stripes correspond to magnetic domain walls, whereas the gradual ringlike oscillation of brightness is caused by interference of the laser light in the sample. The incident light at $E = 1.29$ eV was propagating parallel to the crystallographic y axis and polarized parallel to the direction of the domain walls, while the SHG light was polarized perpendicular to the domain walls.

the multiferroic phase, with an almost step-like dependence on temperature across the transition. As a consequence of this Heaviside-like behavior the specific heat C which is proportional to the order-parameter derivative displays a delta-shaped like behavior across T_2 (Refs. [5], [6], and [14]). This remarkable property of the first-order multiferroic transition in Co_3TeO_6, which has been previously observed at the first-order ferromagnetic transition in Fe_2P (Refs. [15] and [16]), is reminiscent of structural transitions having a reconstructive mechanism[12] as observed, for example, at the fcc-hcp transition in cobalt.[17] However, in Co_3TeO_6 the symmetry-breaking mechanism involved at T_2 is not of the reconstructive type since the $C2'$ symmetry of phase III is group-subgroup related to the $C2'/c$ symmetry of phase II. Therefore the strong discontinuities reported for the lattice parameters[6] at T_2 should correspond to a strong magnetoelastic coupling with an exchange energy critically sensitive to the interatomic spacing. This is in agreement with the large spin lattice coupling deduced by Her *et al.*[14] from the magnetic hysteresis curves and with the direct exchange pathway corresponding to the shorter Co–Co distances existing in phase III (see Fig. 12 in Ref. [6]) for the neighboring atoms Co(5)–Co(5), Co(2)–Co(5), Co(3)–Co(3), and Co(4)–Co(4). The strong magnetoelastic effect is favored by the coupling between the order parameters ζ and η_i. Here, the already existing antiferromagnetic order parameter ζ triggers the emergence of the order parameter η_i which is reflected by the transition discontinuity.

The respective monoclinic symmetries of phases II and III permit a variety of magnetoelectric effects under applied magnetic or electric fields. For instance, in phase II applying a magnetic field H_y induces contributions by P_x and P_z to the polarization which vary as $P_{x,z} \approx \zeta H_y$. In phase III one has $P_{x,z} \approx \zeta \rho_e^2 H_y$, that is, at constant temperature P_x and P_z increase linearly with H_y in both phases, while at constant field they increase with temperature as $(T_1 - T)^{1/2}$ in phase I and as $(T_2 - T)$ in phase III. Conversely, applying the magnetic field H_x or H_z in phase II induces a polarization polarization $P_y \approx \zeta H_{x,z}$. In phase III an additional contribution to the spontaneous polarization P_y, namely $\Delta P_y \approx \zeta \rho_e^2 H_{x,z}$, is generated. Reversed magnetoelectric effects should also be observed in phases II and III with the onset of an induced magnetization M_y depending linearly on electric fields E_x or E_z. Additional contributions to the M_x and M_z components of the spontaneous magnetization are generated under an electric field E_y. Accordingly, our theoretical analysis suggests that magnetic-field-induced polarization components $P_x(H_x)$, $P_x(H_z)$, and $P_z(H_x)$ are not allowed. Their observation was reported earlier[5] and should be related to a misorientation of the sample admixing a y component to the x axis. The possibility for such a misorientation was indeed pointed out by the authors of Ref. [5].

In summary, the observed properties of the sequence of three phases reported in Co_3TeO_6 have been described theoretically. The most striking feature of this phase sequence is the coexistence of propagation vectors in the incommensurate and commensurate multiferroic phases, the Brillouin zone center $\vec{k}_2$ vector, which persists in all three phases, coupling successively with the incommensurate $\vec{k}_1$ vector in phase I, and with its lock-in commensurate variant $\vec{k}_3$ in phase III. Another remarkable property is the strongly first-order character of the multiferroic transition which relates both to the triggering mechanism coupling the $\vec{k}_2$ and $\vec{k}_3$ related order parameters, and to a strong magnetoelastic coupling. The sharp peak of the specific heat is shown to be consistent with the almost constant value of the spontaneous magnetization in the multiferroic state. Furthermore, a number of experimental observations of Ref. [5] have been scrutinized: The monoclinic symmetry of phase III is $2'$ (instead of the m as previously proposed[5]). This permits a spontaneous weak magnetization (M_x, M_z), which is found to be in agreement with the domain structures observed in SHG measurements. It also allows a spontaneous polarization P_y which has not been investigated before whereas contributions $P_{x,z} \neq 0$ are no longer expected. The magnetoelectric effects that should exist in Co_3TeO_6 have been worked out theoretically. A verification of these predictions, supported by the application of both magnetic and electric fields, is necessary for confirming the validity of the theoretical description presented in this article.

At last we emphasize that our theoretical analysis and conclusions differ in an essential manner from the symmetry analysis proposed in Ref. [11] for two reasons. (i) Scrutinization of the data reported in Ref. [5] leads to a revision of the magnetic symmetry and the direction of electric polarization. Since this is a very recent result it could be taken into account in the present work but not in Ref. [11]. (ii) In Ref. [11] mainly one-dimensional order parameters associated with the wave vector $\vec{k}_2$ are considered. In contrast, we analyze the symmetries and physical properties of phases I and III as resulting from the coupling of order parameters associated, respectively, with the wave vectors $(\vec{k}_1, \vec{k}_2)$ and $(\vec{k}_2, \vec{k}_3)$, in agreement with the neutron diffraction data reported in Ref. [6]. In particular the field-induced component $P_z(H_x)$ discussed in Ref. [11] is shown to be absent in our description, whereas we predict the existence of a single spontaneous (zero-field) polarization component P_y.

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