Identifying Landau order parameters and their transformation properties for complex multiferroics: The case of Mn_2GeO_4

A. B. Harris

Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

(Received 18 January 2017; revised manuscript received 15 May 2017; published 16 August 2017)

For the case of Mn_2GeO_4 we characterize the complex magnetic phase that exists at temperatures below 5.5 K by order parameters for both the commensurate and the incommensurate ordering. For the incommensurate ordering we are forced to consider the transformation properties which interrelate magnetic modes at different noncollinear members of the star of the incommensurate wave vector. The known transformation properties of the underlying magnetic wave functions are used to deduce the transformation properties of the order parameters. These results are applied to construct the high-order invariants in the free energy which have been used elsewhere to describe the characteristics of switching between different domains. The properties of different domains are discussed in detail.

DOI: 10.1103/PhysRevB.96.054422

I. INTRODUCTION

A. Background

The last dozen years have seen an explosion in the study of multiferroic systems in which incommensurate magnetic order induces ferroelectricity, following the pioneering work of Refs. [1–3]. In 2005 a microscopic model was developed [4] based on the idea of a "spin current." This mechanism has been widely cited in terms of a picture in which the spin structure is characterized as being a magnetic spiral. Shortly thereafter, in studies on $Ni_3V_2O_8$ [5] and on $TbMnO_3$ [6], a phenomenological Landau theory was developed which invoked a trilinear magnetoelectric interaction. The virtue of this theory was that it showed for the first time exactly how the crystal structure controlled the direction of the spontaneous polarization, independent of whether the microscopic mechanism was that of the spin current model or of some other model, e.g., one based on ionic displacements [7]. Perhaps motivated by the spin current model, Mostovoy [8] subsequently introduced an idealized model based on the spin structure being a perfect spiral. This model has been extremely popular. However, there are two issues that, in contrast to the phenomenological theory, it does not address properly. The first is that in most cases the spiral magnetic order results from the existence of two order parameters describing order along the two axes of the spiral. For $Ni_3V_2O_8$, as the temperature is lowered, first one ordering appears and then, at a lower-temperature transition, the second order parameter grows continuously from zero. Thus, when the second order parameter is very small, one has an elliptical helix whose ratio of major to minor axes is large (so that it really does not look like an ideal helix). According to the phenomenological approach [5,6,9] the induced polarization is proportional to the product of the two order parameters, one of which develops continuously from zero. This result has been quantitatively verified by a neutron-scattering experiment [10]. The second issue is that the spin-current mechanism [4] or Mostovoy construction [8] could not explain the magnetoelectric transition in $RbFe(MoO_4)_2$ [11], in contrast to the success of the Landau theory [11]. The failure of the Mostovoy construction [8] and the spin-current model [4] was subsequently explained by identifying a "missing" interaction [12]. In most situations missing interactions can be ignored and the Mostovoy picture is a useful way to determine the orientation of the induced electric polarization. However, as is shown elsewhere [13], to treat the coupling of domains in Mn_2GeO_4 (MGO) requires analyzing several higher-order terms in the free energy. Since the application of Landau theory [9] is not trivial in this case, the present paper will explain in detail how to identify order parameters in a system as complicated as MGO. We then use the known way the magnetization distribution transforms to deduce how the order parameters transform under the symmetry operations of the space group. Having this information we construct the invariant terms in the free energy in the phase which exists for MGO for $T < 5.5$ K (see Fig. 1).

Since this discussion is somewhat complicated, we start, in Sec. II, by giving a brief review of Landau theory and how various order parameters can appear as the temperature is lowered. In Sec. III we describe the magnetic modes at the relevant wave vectors (namely zero wave vector and the star of **k** for MGO) as obtained from a well-established computer resource (ISODISTORT) [14], whose results we have verified by hand. Here, in what we regard as a textbook example, we show how order parameters are introduced as amplitudes of the magnetic modes which have the allowed symmetry for the wave vectors in question. In Sec. IV we determine the transformation properties of the order parameters. Since each order parameter specifies a distribution of magnetization, the transformation properties of the order parameters follow from the known way the associated distribution of magnetization transforms. Since it is necessary to treat all the wave vectors in the star of **k**, we develop the transformations that relate order parameters at different wave vectors of the star of **k**. The advantage of the introduction of order parameters with their associated transformation properties is that they carry all the symmetry information of each mode. Although there is some arbitrariness in defining the transformation from one vector of the star to another, we use this information in Sec. V to construct the possible invariants which make up the free energy. It is these third- and fifth-order couplings which govern the domain dynamics, as described in Ref. [13]. In Sec. V we also discuss in detail how the various observables vary from one domain to the next.

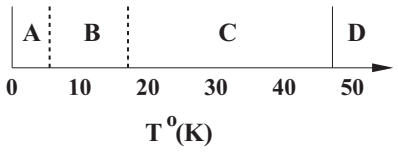

FIG. 1. Magnetoelectric phase diagram of MGO [15]. First-order (continuous) transitions are indicated by dashed (full) lines. Phase D has no long-range magnetic or electric order. Only in phase A (below 5.5 K) does the system exhibit incommensurate magnetic order together with a spontaneous electric polarization along the z axis. Phase A also exhibits commensurate magnetic order of irreps X_1 and X_3. Phases B and C (not considered in the present paper) display commensurate magnetic order.

B. Ultrabrief synopsis

For a reader who can devote only a few minutes to this paper, I suggest to (1) look at Eqs. (9) and (10) to see how the order parameters are introduced; (2) read the first paragraph of Sec. IV which defines transformations of the order parameters based on Eq. (14); (3) look at Eqs. (22), (35), (45), and (54) to get the flavor of the transformations; and (4) consider in Sec. V B the construction of invariants (which couple domains or induce ferroelectricity) used in Ref. [13] to explain domain dynamics.

II. BRIEF REVIEW OF LANDAU THEORY

Here we give a brief review of the basic structure of Landau theory. For a more detailed review of Landau theory as applied to multiferroics, see Ref. [9]. We first consider the free energy F associated with the development of magnetic ordering at a continuous phase transition at temperature $T = T_c$ for a system of Ising spins. When there is one magnetic ion per unit cell, then the free energy as a function of its magnetic moment vector $\mathbf{M}$ is of the form

$$F = a(T - T_c)M^2 + uM^4 + \cdots , \tag{1}$$

where a and u are positive constants. Note that time-reversal symmetry only permits terms of even order in M. By minimizing the free energy with respect to M one obtains the usual mean-field result that $M = 0$ for $T > T_c$ and $M(T) \propto (T_c - T)^{1/2}$ for $T < T_c$. More generally, when there are several (p) magnetic sites in the unit cell, the free energy will be a generalization of Eq. (1) such that the term in M^2 is replaced by a quadratic form in the variables M_i, where M_i is the magnetic moment at site i. This quadratic form is diagonalized by a transformation to normal modes (whose amplitudes are M_α) which is analogous to the introduction of normal modes for phonons. As for phonons, the modes carry symmetry labels which identify their symmetry. In the usual language, the modes are referred to as irreducible representations (irreps). Then the generalization of Eq. (1) is equivalent to (with $T_1 > T_2 > T_3 \ldots T_p$)

$$F = \sum_{\alpha=1}^{p} a_\alpha (T - T_\alpha)M_\alpha^2 + u_\alpha M_\alpha^4 + \cdots . \tag{2}$$

In the interest of simplicity we do not consider the possibility that $T_\alpha = T_\beta$ for $\alpha \neq \beta$. (In technical language this assumption is that all irreps are one dimensional.) Taken literally, Eq. (2) would imply that as the temperature is reduced, first M_1 appears continuously, then at a lower temperature M_2 appears continuously, and so forth. (Usually, condensing M_1 inhibits condensing any other M_α which has the same symmetry as M_1.) In MGO, there are four different symmetry zero-wave-vector modes with symmetry labels [15] X_1, X_2, X_3, and X_4. As is apparent from Fig. 1, the continuous scenario just mentioned is not followed (viz. order in the lowest-temperature phase does not develop via sequential continuous transitions [16]). Nevertheless, the symmetry of the lowest-temperature phase is the same as if the irreps X_1 and X_3 had appeared in the scenario of Eq. (2). The irrep X_3 is a mode with a net magnetic moment along the crystal c axis and X_1 is an antiferromagnetic mode with moments perpendicular to the crystal c axis. We emphasize that in this paper our main aim is to describe the symmetry properties of the lowest-temperature phase of MGO [15,17,18]. To construct a plausible Landau free energy to explain the extremely complex phase transition [16] below which not only X_1 and X_3 but also the incommensurate modes (which we discuss next) exist is beyond the scope of the present study. Also, note that for the purpose of the present paper we do not keep track of the detailed structure of the modes X_n. *Only the symmetry properties of the wave functions determine, say, whether or not an electric polarization develops.* Since our main aim here is to construct the symmetry-allowed potentials for the free energy, we do not need to explicitly consider the details of the wave functions.

Finally, we consider the description of the incommensurate modes at the wave vector star [15] $\pm\mathbf{k}_A = (0.136, 0.211, 0)$ and $\pm\mathbf{k}_B = (0.136, -0.211, 0)$. As we have just discussed, we only need to keep track of the symmetry of the allowed modes. Since the wave vector is invariant under the mirror m_z that takes z into $-z$, the allowed modes will be either even or odd under m_z [19]. Thus the symmetry properties of the modes can be identified independent of the actual interactions, and indeed without even knowing the details of the spin structure. Also, one should note that we do not rely on the condition that the magnetic structure consist of an assembly of magnetic helicies. While helical structures often give rise to ferroelectricity, the converse, namely that ferroelectric incommensurate magnets must be helices, is clearly not the case. An amusing example of structures which have the same symmetry, but which have very different appearance, is (a) orientationally ordered solid ortho-H_2 (consisting of rodlike molecules) [20] and (b) orientationally ordered nearly spherical C_{60} [21] molecules, both of which have $Pa3$ space-group symmetry. In other words, knowing only the space-group symmetry of a magnetic structure is not a guarantee that one can identify, even qualitatively, the actual structure.

III. CRYSTAL SYMMETRY AND MODES

A. Crystal symmetry

For these discussions we record the symmetry operations of the orthorhombic space group for MGO, namely $Pnma$ = No. 62 in Ref. [22], where x, y, and z refer to the a, b, and c

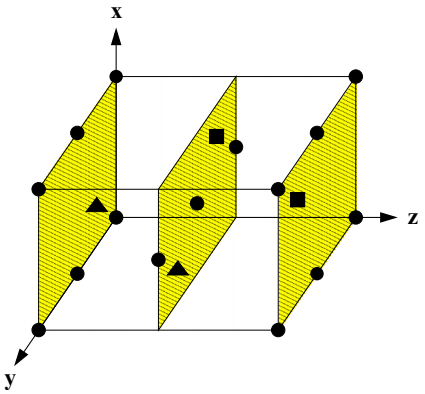

FIG. 2. Magnetic (Mn) ions in the unit cell of MGO. Filled circles (triangles or squares) represent *ch* (*pl*) sites. For $\epsilon = 0$ all the ions would be in the three planes $z = 0$, $z = 1/2$, and $z = 1$. Because ϵ is positive but very small, the Mn ions represented by a triangle are displaced (by ϵ) to the right of the plane and those represented by a square are displaced (by ϵ) to the left of the plane.

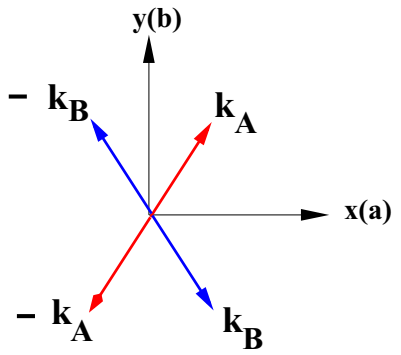

FIG. 3. Star of the wave vector $\mathbf{k}_A = (0.136,0.211,0)$ [15].

crystal axes:

$$\begin{aligned}
&\mathcal{E} = (x,y,z), \quad \mathcal{I} = (\overline{x},\overline{y},\overline{z}),\\
&m_x = \left(\overline{x}+\tfrac{1}{2},y+\tfrac{1}{2},z+\tfrac{1}{2}\right), \quad 2_x = \left(x+\tfrac{1}{2},\overline{y}+\tfrac{1}{2},\overline{z}+\tfrac{1}{2}\right),\\
&m_y = \left(x,\overline{y}+\tfrac{1}{2},z\right), \quad 2_y = \left(\overline{x},y+\tfrac{1}{2},\overline{z}\right),\\
&m_z = \left(x+\tfrac{1}{2},y,\overline{z}+\tfrac{1}{2}\right), \quad 2_z = \left(\overline{x}+\tfrac{1}{2},\overline{y},z+\tfrac{1}{2}\right).
\end{aligned} \tag{3}$$

We will also need to refer to the inverse operations:

$$\begin{aligned}
&m_x^{-1} = \left(\overline{x}+\tfrac{1}{2},y-\tfrac{1}{2},z-\tfrac{1}{2}\right), \quad 2_x^{-1} = \left(x-\tfrac{1}{2},\overline{y}+\tfrac{1}{2},\overline{z}+\tfrac{1}{2}\right),\\
&m_y^{-1} = \left(x,\overline{y}+\tfrac{1}{2},z\right), \quad 2_y^{-1} = \left(\overline{x},y-\tfrac{1}{2},\overline{z}\right),\\
&m_z^{-1} = \left(x-\tfrac{1}{2},y,\overline{z}+\tfrac{1}{2}\right), \quad 2_z^{-1} = \left(\overline{x}+\tfrac{1}{2},\overline{y},z-\tfrac{1}{2}\right).
\end{aligned} \tag{4}$$

The eight sites in the unit cell (shown in Fig. 2) are [15]

$$\begin{aligned}
&\boldsymbol{\tau}_1 = (0,0,0), \quad \boldsymbol{\tau}_2 = (1/2,0,1/2),\\
&\boldsymbol{\tau}_3 = (1/2,1/2,1/2), \quad \boldsymbol{\tau}_4 = (0,1/2,0),\\
&\boldsymbol{\tau}_5 = (a,1/4,\epsilon), \quad \boldsymbol{\tau}_6 = (a+1/2,1/4,1/2-\epsilon),\\
&\boldsymbol{\tau}_7 = (1-a,3/4,-\epsilon), \quad \boldsymbol{\tau}_8 = (1/2-a,3/4,1/2+\epsilon),
\end{aligned} \tag{5}$$

where $\epsilon \approx 0.0$ and $a \approx 0.275$. Sites $\boldsymbol{\tau}_1$–$\boldsymbol{\tau}_4$ ($\boldsymbol{\tau}_5$–$\boldsymbol{\tau}_8$) are known as "*ch*" ("*pl*") sites [15].

B. Zero wave-vector modes

Two zero wave-vector irreducible representations (irreps) are active: X_1 and X_3. Their parity under the mirror operations is given in Table I. The actual wave functions for these modes, given in the Supplemental Material to Ref. [15], are not needed for our symmetry analysis. As mentioned, the phase with X_1 is a type of antiferromagnetic ordering and X_3 ordering has a net magnetic moment along the z axis.

TABLE I. Symmetry properties of the two active zero-wave-vector magnetic order parameters X_n for the irrep Γ_n. Both these order parameters (OPs) are odd under time reversal.

OP	m_x	m_y	m_z
X_1	$+1$	$+1$	$+1$
X_3	-1	-1	$+1$

C. Incommensurate modes

In Appendix A, based on results from ISODISTORT [14] we show that the magnetization throughout a domain of wave vector $\mathbf{k}_A$ for the irrep $D^{(\sigma=1)}$ must be of the form

$$\begin{aligned}
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_1) &= a_\alpha e^{-i[\mathbf{N}+\boldsymbol{\tau}_1]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_2) &= \mu_\alpha a_\alpha e^{-i[\mathbf{N}+\boldsymbol{\tau}_2]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_3) &= b_\alpha e^{-i[\mathbf{N}+\boldsymbol{\tau}_3]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_4) &= \mu_\alpha b_\alpha e^{-i[\mathbf{N}+\boldsymbol{\tau}_4]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_5) &= z_\alpha e^{-i[\mathbf{N}+\boldsymbol{\tau}_5]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_6) &= \mu_\alpha z_\alpha e^{-i[\mathbf{N}+\boldsymbol{\tau}_6]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_7) &= z_\alpha^* e^{-i[\mathbf{N}+\boldsymbol{\tau}_7]\cdot\mathbf{k}_A+i\phi} + \text{c.c.}\\
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_8) &= \mu_\alpha z_\alpha^* e^{-i[\mathbf{N}+\boldsymbol{\tau}_8]\cdot\mathbf{k}_A+i\phi} + \text{c.c.},
\end{aligned} \tag{6}$$

where $\mathbf{N} \equiv (N_x,N_y,N_z)$ specifies the integer coordinates (we always use rlu) of the unit cell, $\mu_x = \mu_y = -\mu_z = -1$, the superscripts on M label the wave vector and the irrep, and the subscript is the component label, $\alpha = x$, y, or z (see Fig. 3). The following assume values not fixed by symmetry: a_α and b_α which are real valued, z_α which is complex valued, and ϕ which is an overall global phase. As we verify later, this irrep is even under m_z [19]. To illustrate, we show, in Fig. 4, the magnetic structure of one of the two magnetic helices which form the structure of MGO. But we emphasize that the analysis which follows relies only on the symmetry of the magnetic structure of MGO and not on the fact that its structure consists of magnetic helices. Accordingly, we do not reproduce here the structural parameters which are given in the Supplemental Material of Ref. [15].

For economy in notation we write Eq. (6) as

$$\begin{aligned}
M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_\mathbf{n}) = e^{i\phi}[&a_\alpha,\mu_\alpha a_\alpha,b_\alpha,\mu_\alpha b_\alpha;z_\alpha,\mu_\alpha z_\alpha,z_\alpha^*,\mu_\alpha z_\alpha^*]_n\\
&\times e^{-i[\mathbf{N}+\tau_\mathbf{n})\cdot\mathbf{k}_\mathbf{A}} + \text{c.c.}
\end{aligned} \tag{7}$$

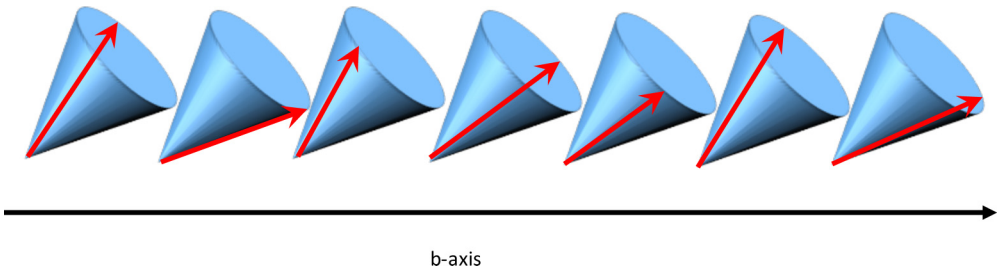

FIG. 4. Magnetic structure of one of the two helices in the unit cell associated with wave vector $\mathbf{k}_A$.

In this notation the magnetization for irrep $D^{(\sigma=2)}$ is given by

$$M_\alpha^{(A,2)}(\mathbf{N}+\boldsymbol{\tau}_\mathbf{n}) = e^{i\phi}[c_\alpha,-\mu_\alpha c_\alpha,d_\alpha,-\mu_\alpha d_\alpha;w_\alpha, \\ -\mu_\alpha w_\alpha,w_\alpha^*,-\mu_\alpha w_\alpha^*]_n e^{-i[\mathbf{N}+\tau_\mathbf{n})\cdot\mathbf{k_A}} + \text{c.c.} \tag{8}$$

As we verify later, this irrep is odd under m_z [19]. Note that we use different constants for irreps 1 and 2. We do this to emphasize the fact that the wave functions for different symmetries are not related, just as atomic s and p functions are not related to each other by symmetry. Analogous results for wave vector $\mathbf{k}_B$ are obtained below.

D. Definition of order parameters

If incommensurate magnetic ordering appears below a continuous phase transition, then the temperature dependence of the wave function when the ordering initially develops gives rise mainly to a change of scale of the coefficients. Accordingly, we describe the wave function as a temperature-dependent amplitude times a normalized wave function, as in Eq. (9), below. In so doing, we incorporate the phase factor $\exp(i\phi)$ into the amplitude, thus giving rise to a complex-valued amplitude which we identify as the order parameter, here denoted $\mathbf{Q}_X^{(\sigma)}$, for the irrep σ at wave vector $\mathbf{k}_X$, where $X=\pm A$ or $X=\pm B$ (and $\mathbf{k}_{-X}$ denotes $-\mathbf{k}_X$). Of course, it is an approximation to assume that the temperature dependence of the wave function merely induces a temperature dependence of the order parameter. However, since this approximation correctly describes the symmetry of the phase, it is often useful and can form the basis for a renormalization group treatment of the critical behavior [23]. Furthermore, as shown in Appendix B, corrections due to the additional temperature dependence of the wave function can be generated within the Landau formulation of the order parameter which we describe here. Note that in traversing the phase diagram the symmetry can only change if a phase boundary is crossed.

Here we are interested in describing the symmetry of a system which can have both incommensurate irreps simultaneously present. This means that order parameters for the two irreps can simultaneously be nonzero. In what follows each mode is characterized by its complex-valued order parameter $\mathbf{Q}_\mathbf{X}^{(\sigma)}$ which has its own magnitude and phase. Thus we write the contribution to the α component of the magnetization from irrep 1 at wave vector $\mathbf{k}_A$ to be

$$M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_n) = \mathbf{Q}_A^{(1)}[a_\alpha,\mu_\alpha a_\alpha,b_\alpha,\mu_\alpha b_\alpha;z_\alpha,\mu_\alpha z_\alpha,z_\alpha^*,\mu_\alpha z_\alpha^*]_n e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]} + \text{c.c.} \\ \equiv \mathbf{Q}_A^{(1)}\Psi_{\alpha,n}^{(A,1)} e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]} + \text{c.c.} \tag{9}$$

and that from irrep 2 at wave vector $\mathbf{k}_A$ to be

$$M_\alpha^{(A,2)}(\mathbf{N}+\boldsymbol{\tau}_n) = \mathbf{Q}_A^{(2)}[c_\alpha,-\mu_\alpha c_\alpha,d_\alpha,-\mu_\alpha d_\alpha;w_\alpha,-\mu_\alpha w_\alpha,w_\alpha^*,-\mu_\alpha w_\alpha^*]_n e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]} + \text{c.c.} \\ \equiv \mathbf{Q}_A^{(2)}\Psi_{\alpha,n}^{(A,2)} e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]} + \text{c.c.}, \tag{10}$$

where we require the wave functions $\boldsymbol{\Psi}^{(A,m)}$ to be normalized:

$$\sum_\alpha \left[2a_\alpha^2+2b_\alpha^2+4|z_\alpha|^2\right] = \sum_\alpha \left[2c_\alpha^2+2d_\alpha^2+4|w_\alpha|^2\right] = 1. \tag{11}$$

We can equally well write the equation for $M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_n)$ as

$$M_\alpha^{(A,1)}(\mathbf{N}+\boldsymbol{\tau}_n) = \mathbf{Q}_A^{(1)*}[a_\alpha,\mu_\alpha a_\alpha,b_\alpha,\mu_\alpha b_\alpha;z_\alpha^*,\mu_\alpha z_\alpha^*,z_\alpha,\mu_\alpha z_\alpha]_n e^{-i(-\mathbf{k}_A)\cdot[\mathbf{N}+\boldsymbol{\tau}_n]} + \text{c.c.} \tag{12}$$

This leads us to write

$$\mathbf{Q}_{-X}^{(\sigma)} = \mathbf{Q}_X^{(\sigma)*}, \quad \Psi_{\alpha,n}^{(-X,\sigma)} = \Psi_{\alpha,n}^{(X,\sigma)*}, \tag{13}$$

where the subscript $-X$ refers to the wave vector $-\mathbf{k}_X$. We assume that the $\Psi_{\alpha n}^{(A,\sigma)}$ have been determined by fitting experimental data, as in Ref. [15]. However, as stated earlier, their actual values do not affect the symmetry analysis. Below we will obtain the transformation properties of the order parameters and thereby determine the wave functions for $\pm\mathbf{k}_B$ in terms of those for $\pm\mathbf{k}_A$.

IV. TRANSFORMATION PROPERTIES OF THE ORDER PARAMETERS

We define the transformation of order parameters under an operator $\mathcal{O}$, by considering the effect of $\mathcal{O}$ on the distribution of magnetization over the system. We write the fundamental equation, which enables us to deduce the transformation properties of the order parameters, as

$$\mathbf{M}(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathcal{O}^S\mathbf{M}([\mathcal{O}^R]^{-1}[N_x,N_y,N_z;\boldsymbol{\tau}_n]), \quad (14)$$

where $\mathcal{O}^S$ is the part of the operator $\mathcal{O}$ that operates on spin and $\mathcal{O}^R$ is the part of the operator $\mathcal{O}$ that operates on the position of the spin. This equation says that the transformed magnetic moment, indicated by a prime, which, before transformation, was at $[\mathcal{O}^R]^{-1}[\mathbf{N}+\boldsymbol{\tau}_n]$, has been transformed by $\mathcal{O}^S$ and is placed at its final location at $\mathbf{N}+\boldsymbol{\tau}_n$. In this section we will consider the transformations under the three perpendicular mirror planes, since these operations can be taken to be the generators of the point group. In Eqs. (9) and (10) we have already defined the wave functions for the wave vector $\mathbf{k}_A$ and in Eq. (13) for the wave vector $-\mathbf{k}_A$. In Sec. IV C we will obtain those for wave vectors $\pm\mathbf{k}_B$.

A. Transformation by inversion $\mathcal{I}$

Perhaps the simplest operation is spatial inversion $\mathcal{I}$. Since $\mathcal{I}^{-1}=\mathcal{I}$, Eq. (14) is

$$\begin{aligned}\mathbf{M}(N_x,N_y,N_z;\boldsymbol{\tau}_n)' &= \mathcal{I}^S\mathbf{M}([\mathcal{I}^R][N_x,N_y,N_z;\boldsymbol{\tau}_n])\\ &= \mathbf{M}([\mathcal{I}^R][N_x,N_y,N_z;\boldsymbol{\tau}_n]),\end{aligned} \quad (15)$$

where we used the fact that the magnetic moment is a pseudovector to write the second version of the above equation. Thus, for wave vector $\mathbf{k}_A$ and irrep σ,

$$\begin{aligned}\mathbf{M}_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' &= Q_A^{(\sigma)}\mathcal{I}^R\left[\Psi_{\alpha n}^{(A,\sigma)}e^{-i\mathbf{k}_A\cdot(\mathbf{N}+\boldsymbol{\tau}_n)}\right]+\text{c.c.}\\ &= Q_A^{(\sigma)}\left[\mathcal{I}^R\Psi_{\alpha n}^{(A,\sigma)}\right]e^{-i[-\mathbf{k}_A\cdot(\mathbf{N}+\boldsymbol{\tau}_n)]}+\text{c.c.},\end{aligned} \quad (16)$$

where we used $\mathcal{I}\boldsymbol{\tau}_n=-\boldsymbol{\tau}_n$. To evaluate the right-hand side of Eq. (16) we need to identify $\mathcal{I}\boldsymbol{\tau}_n=\boldsymbol{\tau}_{\overline{n}}$, where $\overline{n}$ is implied by the last column of Table II. Thus

$$\mathcal{I}^R\Psi_{\alpha n}^{(A,\sigma)} = \Psi_{\alpha\overline{n}}^{(A,\sigma)} = \left[\Psi_{\alpha n}^{(A,\sigma)}\right]^*, \quad (17)$$

TABLE II. Transformation of $\boldsymbol{\tau}_n$ by $\mathcal{I}$ to the value $\boldsymbol{\tau}_{\overline{n}}$. Since the vector $\boldsymbol{\tau}$ is not changed by adding a lattice vector to it, the third column is equivalent to the fourth column.

n	$\boldsymbol{\tau}_n$	$\mathcal{I}\boldsymbol{\tau}_n$	$\Rightarrow$	$\boldsymbol{\tau}_{\overline{n}}$
1	$(0,0,0)$	$(0,0,0)$	$(0,0,0)$	$\boldsymbol{\tau}_1$
2	$(\frac{1}{2},0,\frac{1}{2})$	$(-\frac{1}{2},0,-\frac{1}{2})$	$(\frac{1}{2},0,\frac{1}{2})$	$\boldsymbol{\tau}_2$
3	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$(-\frac{1}{2},-\frac{1}{2},-\frac{1}{2})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$\boldsymbol{\tau}_3$
4	$(0,\frac{1}{2},0)$	$(0,-\frac{1}{2},0)$	$(0,\frac{1}{2},0)$	$\boldsymbol{\tau}_4$
5	$(a,\frac{1}{4},\epsilon)$	$(-a,-\frac{1}{4},-\epsilon)$	$(1-a,\frac{3}{4},-\epsilon)$	$\boldsymbol{\tau}_7$
6	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2}-\epsilon)$	$(-a-\frac{1}{2},-\frac{1}{4},-\frac{1}{2}+\epsilon)$	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2}+\epsilon)$	$\boldsymbol{\tau}_8$
7	$(1-a,\frac{3}{4},-\epsilon)$	$(a-1,-\frac{3}{4},\epsilon)$	$(a,\frac{1}{4},\epsilon)$	$\boldsymbol{\tau}_5$
8	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2}+\epsilon)$	$(a-\frac{1}{2},-\frac{3}{4},-\frac{1}{2}-\epsilon)$	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2}-\epsilon)$	$\boldsymbol{\tau}_6$

where we used Eqs. (9) and (10) to write $\Psi_{\alpha\overline{n}}^{(A,\sigma)}=[\Psi_{\alpha n}^{(A,\sigma)}]^*$. Thus,

$$\mathbf{M}_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathbf{Q}_A^{(\sigma)}\left[\Psi_{\alpha n}^{(A,\sigma)}\right]^*[e^{-i[-\mathbf{k}_A\cdot(\mathbf{N}+\boldsymbol{\tau}_n)}]+\text{c.c.} \quad (18)$$

This is of the form

$$\mathbf{M}_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathbf{Q}_{-A}^{(\sigma)\prime}\Psi_{\alpha n}^{(-A,\sigma)}[e^{-i[-\mathbf{k}_A\cdot(\mathbf{N}+\boldsymbol{\tau}_n)}]+\text{c.c.} \quad (19)$$

This part of the argument requires some discussion. The right-hand side of Eq. (19) consists of three factors which we refer to as Q, Ψ, and exp. The Q factor is simple: no symmetry operation can change the irrep, so the superscript σ is fixed. The effect of the symmetry operation (here $\mathcal{I}$) on the wave vector subscript of Q is obvious. The scripts on the Ψ factor are those we would expect result from complex conjugation. The exp factor is exactly what appears in the analog of Eq. (12). Any leftover phase factors (there are none here) are assigned to the Q factor. Thus

$$\mathcal{I}\mathbf{Q}_{-A}^{(\sigma)} \equiv \mathbf{Q}_{-A}^{(\sigma)\prime} = \mathbf{Q}_A^{(\sigma)}, \quad \Psi_{\alpha n}^{(-A,\sigma)} = \left[\Psi_{\alpha n}^{(A,\sigma)}\right]^*, \quad (20)$$

consistent with Eq. (13). The analogous result holds for $\mathbf{k}_B$. So in all we have

$$\mathcal{I}\mathbf{Q}_X^{(\sigma)} = \mathbf{Q}_X^{(\sigma)*} = \mathbf{Q}_{-X}^{(\sigma)}. \quad (21)$$

We may consider the order parameter to be a four-component column vector $\vec{\mathbf{Q}}_\sigma$ with components $(Q_\sigma)_1=Q_A^{(\sigma)}$, $(Q_\sigma)_2=Q_B^{(\sigma)}$, $(Q_\sigma)_3=Q_A^{(\sigma)*}\equiv Q_{-A}^{(\sigma)}$, $(Q_\sigma)_4=Q_B^{(\sigma)*}\equiv Q_{-B}^{(\sigma)}$. We summarize the results of Eq. (21) for the effect of inversion by writing

$$\begin{aligned}\mathcal{I}\vec{\mathbf{Q}}_\sigma \equiv \mathcal{I}\begin{bmatrix}\mathbf{Q}_A^{(\sigma)}\\ \mathbf{Q}_B^{(\sigma)}\\ \mathbf{Q}_{-A}^{(\sigma)}\\ \mathbf{Q}_{-B}^{(\sigma)}\end{bmatrix} &= \begin{bmatrix}0&0&1&0\\0&0&0&1\\1&0&0&0\\0&1&0&0\end{bmatrix}\begin{bmatrix}\mathbf{Q}_A^{(\sigma)}\\ \mathbf{Q}_B^{(\sigma)}\\ \mathbf{Q}_{-A}^{(\sigma)}\\ \mathbf{Q}_{-B}^{(\sigma)}\end{bmatrix}\\ &\equiv \mathcal{M}_\sigma(\mathcal{I})\vec{\mathbf{Q}}_\sigma.\end{aligned} \quad (22)$$

In general, if $\mathcal{O}$ is an operator, then we write

$$\mathcal{O}(Q_\sigma)_n = \sum_m[\mathcal{M}_\sigma(\mathcal{O})]_{nm}(Q_\sigma)_m. \quad (23)$$

B. Transformation by m_z

Since m_z leaves the wave vector $\mathbf{k}$ invariant, we consider it next. We start by considering the case when $\mathbf{k}=\mathbf{k}_A$. Thus we apply Eq. (14) when $\mathbf{k}=\mathbf{k}_A$ and $\mathcal{O}=m_z$:

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \lambda_\alpha M_\alpha([m_z{}^R]^{-1}[N_x,N_y,N_z;\boldsymbol{\tau}_n]), \quad (24)$$

where, since $\mathbf{M}$ is a pseudovector, $\lambda_\alpha=(-1,-1,+1)$. Thereby we find that

$$\begin{aligned}&M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)'\\ &\quad= \lambda_\alpha\left(\left[m_z^R\right]^{-1}\Psi_{\alpha n}^{(A,1)}\right)e^{-i\mathbf{k}_A\cdot[m_z^R]^{-1}[\mathbf{N}+\boldsymbol{\tau}_n]}\mathbf{Q}_A^{(1)}+\text{c.c.}\end{aligned} \quad (25)$$

To evaluate the exponential for $k_z=0$, note that acting on a vector of the form $(v_x,v_y,0)$, we have $[m_z^R]^{-1}(v_x,v_y,0)=$

TABLE III. As Table II. Transformation of $\boldsymbol{\tau}_n$ by m_z into $\tau_{\overline{n}}$.

n	$\boldsymbol{\tau}_n$	$\left[m_z^R\right]^{-1}\boldsymbol{\tau}_n$	$\Rightarrow$	$\boldsymbol{\tau}_{\overline{n}}$
1	$(0,0,0)$	$(-\frac{1}{2},0,\frac{1}{2})$	$(\frac{1}{2},0,\frac{1}{2})$	$\boldsymbol{\tau}_2$
2	$(\frac{1}{2},0,\frac{1}{2})$	$(0,0,0)$	$(0,0,0)$	$\boldsymbol{\tau}_1$
3	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$(0,\frac{1}{2},0)$	$(0,\frac{1}{2},0)$	$\boldsymbol{\tau}_4$
4	$(0,\frac{1}{2},0)$	$(-\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$\boldsymbol{\tau}_3$
5	$(a,\frac{1}{4},0)$	$(a-\frac{1}{2},\frac{1}{4},\frac{1}{2})$	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2})$	$\boldsymbol{\tau}_6$
6	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2})$	$(a,\frac{1}{4},0)$	$(a,\frac{1}{4},0)$	$\boldsymbol{\tau}_5$
7	$(1-a,\frac{3}{4},0)$	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2})$	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2})$	$\boldsymbol{\tau}_8$
8	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2})$	$(-a,\frac{3}{4},0)$	$(1-a,\frac{3}{4},0)$	$\boldsymbol{\tau}_7$

$(v_x-1/2,v_y,0)$. So

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \lambda_\alpha\left(\left[m_z^R\right]^{-1}\Psi_{\alpha n}^{(A,1)}\right)e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]}e^{ik_x/2}\mathbf{Q}_A^{(1)}+\text{c.c.} \quad (26)$$

Now we consider $[m_z^R]^{-1}\Psi_{\alpha n}^{(A,1)}$. In Table III we see that

$$\left[m_z^R\right]^{-1}\Psi_{\alpha n}^{(A,1)} = \Psi_{\alpha\overline{n}}^{(A,1)}, \quad (27)$$

where $\overline{n}=n-1$ if n is even and $\overline{n}=n+1$ if n is odd. But, since $1/\mu_\alpha=\mu_\alpha$, we have, from Eq. (9), that

$$\Psi_{\alpha\overline{n}}^{(A,1)} = \mu_\alpha\Psi_{\alpha n}^{(A,1)}. \quad (28)$$

Note that $\mu_\alpha\lambda_\alpha=1$, so that the final result is

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = e^{ik_x/2}\Psi_{\alpha n}^{(A,1)}e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]}\mathbf{Q}_A^{(1)}+\text{c.c.} \quad (29)$$

This is of the form

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathbf{Q}_A^{(1)'}\Psi_{\alpha n}^{(A,1)}e^{-i\mathbf{k}_A\cdot[\mathbf{N}+\boldsymbol{\tau}_n]}+\text{c.c.} \quad (30)$$

In other words,

$$\mathbf{Q}_A^{(1)'} = m_z\mathbf{Q}_A^{(1)} = e^{ik_x/2}\mathbf{Q}_A^{(1)}. \quad (31)$$

For $\sigma=2$ we have

$$m_z\mathbf{Q}_A^{(2)} = -e^{ik_x/2}\mathbf{Q}_A^{(2)}. \quad (32)$$

The difference in sign for $m_z\mathbf{Q}_A^{(2)}$ occurs because here, instead of Eq. (28), one has

$$\Psi_{\alpha\overline{n}}^{(A,2)} = -\mu_\alpha\Psi_{\alpha n}^{(A,2)}, \quad (33)$$

as follows from Eq. (10). The results of Eqs. (31) and (32) give rise to the top row of $\mathcal{M}_\sigma(m_z)$ in Eq. (35), below.

We now use the above results to obtain analogous results for wave vectors $\mathbf{k}_B$. Since the value of $\mathbf{k}$ does not appear explicitly, the wave functions of $\mathbf{k}_B=(k_x,-k_y,0)$ are of the form

$$\boldsymbol{\Psi}^{(B,1)} = [a'_\alpha,\mu_\alpha a'_\alpha,b'_\alpha,\mu_\alpha b'_\alpha,z'_\alpha,\mu_\alpha z'_\alpha,z^*_\alpha,\mu_\alpha z'^*_\alpha] \quad (34)$$

and similarly for $\boldsymbol{\Psi}^{(B,2)}$. The relation between (a',b',z') and (a,b,z), given in Table IV will be derived later. Thus Eq. (31) holds when $\mathbf{k}_A\equiv(k_x,k_y)$ is replaced by $\mathbf{k}_B\equiv(k_x,-k_y)$ and

TABLE IV. Wave functions $\Psi_{\alpha n}^{(A,\sigma)}$ and $\Psi_{\alpha n}^{(B,\sigma)}$. Here $\lambda'_\alpha=(-1,1,-1)$. In lines 2–5 of this table we do not take account of the fact that the wave function for $\mathbf{k}_A$ and $\mathbf{k}_B$ are related by a symmetry operation. In lines 6 and 7, $\Psi_{\alpha n}^{B,\sigma}$ is constructed by applying m_y to $\Psi_{\alpha n}^{(A,\sigma)}$ [see Eqs. (42) and (43)].

$n=$	1	2	3	4	5	6	7	8
$\Psi_{\alpha n}^{(A,1)}=$	a_α	$\mu_\alpha a_\alpha$	b_α	$\mu_\alpha b_\alpha$	z_α	$\mu_\alpha z_\alpha$	z^*_α	$\mu_\alpha z^*_\alpha$
$\Psi_{\alpha n}^{(A,2)}=$	c_α	$-\mu_\alpha c_\alpha$	d_α	$-\mu_\alpha d_\alpha$	w_α	$-\mu_\alpha w_\alpha$	w^*_α	$-\mu_\alpha w^*_\alpha$
$\Psi_{\alpha n}^{(B,1)}=$	a'_α	$\mu_\alpha a'_\alpha$	b'_α	$\mu_\alpha b'_\alpha$	z'_α	$\mu_\alpha z'_\alpha$	z'^*_α	$\mu_\alpha z'^*_\alpha$
$\Psi_{\alpha n}^{(B,2)}=$	c'_α	$-\mu_\alpha c'_\alpha$	d'_α	$-\mu_\alpha d'_\alpha$	w'_α	$-\mu_\alpha w'_\alpha$	w'^*_α	$-\mu_\alpha w'^*_\alpha$
$\Psi_{\alpha n}^{(B,1)}/\lambda'_\alpha=$	$\mu_\alpha b_\alpha$	b_α	$\mu_\alpha a_\alpha$	a_α	z_α	$\mu_\alpha z_\alpha$	z^*_α	$\mu_\alpha z^*_\alpha$
$\Psi_{\alpha n}^{(B,2)}/\lambda'_\alpha=$	$\mu_\alpha d_\alpha$	$-d_\alpha$	$\mu_\alpha c_\alpha$	$-c_\alpha$	$-w_\alpha$	$\mu_\alpha w_\alpha$	$-w^*_\alpha$	$\mu_\alpha w^*_\alpha$

we obtain

$$m_z\vec{\mathbf{Q}}_\sigma = m_z\begin{bmatrix}\mathbf{Q}_{\mathbf{A}}^{(\sigma)}\\ \mathbf{Q}_B^{(\sigma)}\\ \mathbf{Q}_{-A}^{(\sigma)}\\ \mathbf{Q}_{-B}^{(\sigma)}\end{bmatrix} = (-)^{\sigma+1}\begin{bmatrix}e^{ik_x/2}&0&0&0\\0&e^{ik_x/2}&0&0\\0&0&e^{-ik_x/2}&0\\0&0&0&e^{-ik_x/2}\end{bmatrix}\begin{bmatrix}\mathbf{Q}_{\mathbf{A}}^{(\sigma)}\\ \mathbf{Q}_B^{(\sigma)}\\ \mathbf{Q}_{-A}^{(\sigma)}\\ \mathbf{Q}_{-B}^{(\sigma)}\end{bmatrix} = \mathcal{M}_\sigma(m_z)\vec{\mathbf{Q}}_\sigma. \quad (35)$$

Here we noted that $\mathbf{Q}_{-X}^{(\sigma)}=[\mathbf{Q}_X^{(\sigma)}]^*$ to obtain the lower half of the matrix.

C. Transformation by m_y

To identify the modes for wave vector $\mathbf{k}_B$ from those of wave vector $\mathbf{k}_A$, we transform the wave functions for $\mathbf{k}_A$ into those for $\mathbf{k}_B=(k_x,-k_y,0)$. Although symmetry allows the parameters of the wave function (e.g., a, b, etc.) to be arbitrary, once they are fixed for wave vector $\mathbf{k}_A$, they are implicitly fixed (to within a phase factor: see Appendix C) for wave vector $\mathbf{k}_B$. Under transformation by m_y we write Eq. (14) as

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \lambda'_\alpha M_\alpha\left(\left[m_y^R\right]^{-1}[N_x,N_y,N_z;\boldsymbol{\tau}_n]\right), \quad (36)$$

where, since $\mathbf{M}$ is a pseudovector, $\lambda'_\alpha=(-1,+1,-1)$. Thereby, for irrep σ we find that

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \lambda'_\alpha\left(\left[m_y^R\right]^{-1}\Psi_{\alpha n}^{(A,\sigma)}\right)e^{-i\mathbf{k}_A\cdot[m_y^R]^{-1}[\mathbf{N}+\boldsymbol{\tau}_n]}\mathbf{Q}_A^{(\sigma)}+\text{c.c.} \quad (37)$$

To evaluate the exponential for $k_z=0$, note that acting on a vector of the form $(v_x,v_y,0)$ we have $[m_y^R]^{-1}(v_x,v_y,0)=(v_x,1/2-v_y,0)$. So

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \lambda'_\alpha\left(\left[m_y^R\right]^{-1}\Psi_{\alpha n}^{(A,\sigma)}\right)e^{-i[k_x(N_x+\tau_{nx})+k_y(-N_y-\tau_{ny}+1/2)]}\mathbf{Q}_A^{(\sigma)} + \text{c.c.} \quad (38)$$

TABLE V. As Table II. Transformation of $\boldsymbol{\tau}_n$ by m_y into $\tau_{\overline{n}}$.

n	$\boldsymbol{\tau}_n$	$[m_y^R]^{-1}\boldsymbol{\tau}_n$	$\Rightarrow$	$\boldsymbol{\tau}_{\overline{n}}$
1	$(0,0,0)$	$(0,\frac{1}{2},0)$	$(0,\frac{1}{2},0)$	$\boldsymbol{\tau}_4$
2	$(\frac{1}{2},0,\frac{1}{2})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$\boldsymbol{\tau}_3$
3	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$(\frac{1}{2},0,\frac{1}{2})$	$(\frac{1}{2},0,\frac{1}{2})$	$\boldsymbol{\tau}_2$
4	$(0,\frac{1}{2},0)$	$(0,0,0)$	$(0,0,0)$	$\boldsymbol{\tau}_1$
5	$(a,\frac{1}{4},0)$	$(a,\frac{1}{4},0)$	$(a,\frac{1}{4},0)$	$\boldsymbol{\tau}_5$
6	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2})$	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2})$	$(a+\frac{1}{2},\frac{1}{4},\frac{1}{2})$	$\boldsymbol{\tau}_6$
7	$(1-a,\frac{3}{4},0)$	$(1-a,-\frac{1}{4},0)$	$(1-a,\frac{3}{4},0$	$\boldsymbol{\tau}_7$
8	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2}-a)$	$(\frac{1}{2}-a,-\frac{1}{4},\frac{1}{2}-a)$	$(\frac{1}{2}-a,\frac{3}{4},\frac{1}{2}-a)$	$\boldsymbol{\tau}_8$

Now we consider $[m_y^R]^{-1}\Psi_{\alpha n}^{(A,\sigma)}$. In Table V we see that

$$\left[m_y^R\right]^{-1}\Psi_{\alpha n}^{(A,\sigma)} = \Psi_{\alpha\overline{n}}^{(A,\sigma)}, \tag{39}$$

where $\overline{n} = 5-n$ if $n<5$ and $\overline{n}=n$ for $n>4$. Then

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \lambda'_\alpha e^{-ik_y/2}\Psi_{\alpha\overline{n}}^{(A,\sigma)} e^{-i[k_x(N_x+\tau_{nx})+(-k_y)(N_y+\tau_{ny})]}\mathbf{Q}_A^{(\sigma)} + \text{c.c.} \tag{40}$$

This is of the form

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathbf{Q}_B^{(\sigma)'}\Psi_{\alpha n}^{(B,\sigma)} e^{-i[k_x(N_x+\tau_{nx})+(-k_y)(N_y+\tau_{ny})]} + \text{c.c.} \tag{41}$$

We choose the signs of the wave functions for $\mathbf{k}_B$ such that

$$\Psi_{\alpha n}^{(B,1)} = \lambda'_\alpha\Psi_{\alpha\overline{n}}^{(A,1)} = \lambda'_\alpha[\mu_\alpha b_\alpha, b_\alpha, \mu_\alpha a_\alpha, a_\alpha; z_\alpha, \mu_\alpha z_\alpha, z_\alpha^*, \mu_\alpha z_\alpha^*]_n, \tag{42}$$

$$\Psi_{\alpha n}^{(B,2)} = -\lambda'_\alpha\Psi_{\alpha\overline{n}}^{(A,2)} = \lambda'_\alpha[\mu_\alpha d_\alpha, -d_\alpha, \mu_\alpha c_\alpha, -c_\alpha; -w_\alpha, \mu_\alpha w_\alpha, -w_\alpha^*, \mu_\alpha w_\alpha^*]_n. \tag{43}$$

As expected, $\Psi_{\alpha n}^{(B,1)}$ is of the form of Eq. (34), but now we have an explicit evaluation of $\Psi_{\alpha n}^{(B,\sigma)}$, given in Table IV. With these definitions the transformed value of the order parameter $\mathbf{Q}_B^{(\sigma)}$ is

$$\mathbf{Q}_B^{(\sigma)'} = m_y\mathbf{Q}_B^{(\sigma)} = (-1)^{\sigma+1}\mathbf{Q}_A^{(\sigma)}e^{-ik_y/2}. \tag{44}$$

This result fixes the second row of $\mathcal{M}_\sigma(m_y)$ in Eq. (45), below. The result is that

$$\begin{aligned} m_y\vec{\mathbf{Q}}_\sigma &= m_y\begin{bmatrix}\mathbf{Q}_A^{(\sigma)}\\ \mathbf{Q}_B^{(\sigma)}\\ \mathbf{Q}_{-A}^{(\sigma)}\\ \mathbf{Q}_{-B}^{(\sigma)}\end{bmatrix} \\ &= (-1)^{\sigma+1}\begin{bmatrix}0 & e^{ik_y/2} & 0 & 0\\ e^{-ik_y/2} & 0 & 0 & 0\\ 0 & 0 & 0 & e^{-ik_y/2}\\ 0 & 0 & e^{ik_y/2} & 0\end{bmatrix}\begin{bmatrix}\mathbf{Q}_A^{(\sigma)}\\ \mathbf{Q}_B^{(\sigma)}\\ \mathbf{Q}_{-A}^{(\sigma)}\\ \mathbf{Q}_{-B}^{(\sigma)}\end{bmatrix} \\ &= \mathcal{M}_\sigma(m_y)\vec{\mathbf{Q}}_\sigma. \end{aligned} \tag{45}$$

The other rows of $\mathcal{M}_\sigma(m_y)$ can be deduced from the second row by changing the sign of k_y or by complex conjugation.

We repeat our previous warning about the phase. We could have defined $\Psi_{\alpha n}^{(B,\sigma)}$ to be the negative of its value in either or both of Eqs. (42) or (43). Such a definition would introduce a σ-dependent phase factor into Eq. (45). This possibility is analyzed in Appendix C, where we show such a phase factor implies a choice of sign for the order parameters, but does not affect the invariant potentials determined in Sec. V.

D. Transformation by m_x

We now consider transformation by m_x. We write Eq. (14) for irrep σ as

$$M_\alpha(\mathbf{N}+\boldsymbol{\tau}_n)' = m_x^S M_\alpha^{(A,\sigma)}\left(\left[m_x^R\right]^{-1}[N_x,N_y,N_z;\boldsymbol{\tau}_n]\right). \tag{46}$$

Since $\mathbf{M}$ is a pseudovector we set $m_x^S = \lambda''_\alpha$, with $\lambda'' = (1,-1,-1)$. Then

$$M_\alpha(\mathbf{N}+\boldsymbol{\tau}_n)' = \lambda''_\alpha\left[\left[m_x^R\right]^{-1}\Psi_{\alpha n}^{(A,\sigma)}\right]e^{-ik_A\cdot[m_x^R]^{-1}[\mathbf{N}+\boldsymbol{\tau}_n]}\mathbf{Q}_A^{(\sigma)} + \text{c.c.} \tag{47}$$

To evaluate the exponential for $k_z=0$, note that acting on a vector of the form $(v_x,v_y,0)$, we have $[m_x^R]^{-1}(v_x,v_y,0) = (1/2-v_x, v_y-1/2, -1/2)$. So

$$\begin{aligned} M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' &= \lambda''_\alpha\mathbf{Q}_A^{(\sigma)}\Psi_{\alpha,\overline{n}}^{(A,\sigma)}e^{-i[k_x(-N_x)+k_yN_y]-i[k_x(-\tau_{n,x}+1/2)+k_y(\tau_{n,y}-1/2)]} + \text{c.c.} \\ &= \lambda''_\alpha\mathbf{Q}_A^{(\sigma)}\Psi_{\alpha,\overline{n}}^{(A,\sigma)}e^{-i[(-k_x)N_x+k_yN_y+(k_y\tau_{ny}-k_x\tau_{nx})+(k_x-k_y)/2]} + \text{c.c.}, \end{aligned} \tag{48}$$

where now $\boldsymbol{\tau}_{\overline{n}} = m_x^R\boldsymbol{\tau}_n$, so that

$$\overline{1}=3,\quad \overline{2}=4,\quad \overline{3}=1,\quad \overline{4}=2,\quad \overline{5}=8,\quad \overline{6}=7,\quad \overline{7}=6,\quad \overline{8}=5. \tag{49}$$

Equation (48) is of the form

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathbf{Q}_{-B}^{(\sigma)'}e^{-i[-\mathbf{k}_B\cdot(\mathbf{N}+\boldsymbol{\tau}_n)}\Psi_{\alpha n}^{(-B,\sigma)} + \text{c.c.} \tag{50}$$

Thus we have

$$\mathbf{Q}_{-B}^{(\sigma)'} = m_x\mathbf{Q}_{-B}^{(\sigma)} = \rho_\sigma\mathbf{Q}_A^{(\sigma)}e^{-i(k_x-k_y)/2},\quad \Psi_{\alpha n}^{(-B,\sigma)} = \rho_\sigma\Psi_{\alpha\overline{n}}^{(A,\sigma)}\lambda''_\alpha. \tag{51}$$

The reason we have included the factor $\rho_\sigma=\pm1$ is because $\Psi^{(B,\sigma)}$ was already fixed by Eqs. (42) and (43). Accordingly, here we have to choose the sign of ρ_σ to be consistent with our previous definition of $\Psi^{(B,\sigma)}$. Using Eq. (49) and taking $\Psi_{\alpha n}^{(A,\sigma)}$ from

Table IV we find that Eq. (51) gives

$$\rho_1\lambda''_\alpha\big[\Psi^{(A,1)}_{\alpha\bar{n}}\big]^* = \Psi^{(B,1)}_{\alpha n} = \Psi^{(-B,1)*}_{\alpha n} = \rho_1\lambda''_\alpha[b_\alpha,\mu_\alpha b_\alpha,a_\alpha,\mu_\alpha a_\alpha;\mu_\alpha z_\alpha,z_\alpha,\mu_\alpha z^*_\alpha,z^*_\alpha], \tag{52}$$

$$\rho_2\lambda''_\alpha\big[\Psi^{(A,2)}_{\alpha\bar{n}}\big]^* = \Psi^{(B,2)}_{\alpha n} = \Psi^{(-B,2)*}_{\alpha n} = \rho_2\lambda''_\alpha[d_\alpha,-\mu_\alpha d_\alpha,c_\alpha,-\mu_\alpha c_\alpha;-\mu_\alpha w_\alpha,w_\alpha,-\mu_\alpha w^*_\alpha,w^*_\alpha]. \tag{53}$$

Comparing Eqs. (42) and (52) we require that $\rho_1\lambda''_\alpha = \lambda'_\alpha\mu_\alpha$ which gives $\rho_1 = +1$. Comparing Eqs. (43) and (53) we require that $\rho_2\lambda''_\alpha = \lambda'_\alpha\mu_\alpha$ which gives $\rho_2 = +1$. The final result is that in terms of the order parameter vector $\vec{\mathbf{Q}}_\sigma$ introduced in Eq. (22), we have $m_x\vec{\mathbf{Q}}_\sigma = \mathcal{M}_\sigma(m_x)\vec{\mathbf{Q}}_\sigma$, with

$$m_x\vec{\mathbf{Q}}_\sigma = m_x\begin{bmatrix}\mathbf{Q}^{(\sigma)}_{\mathbf{A}}\\ \mathbf{Q}^{(\sigma)}_{B}\\ \mathbf{Q}^{(\sigma)}_{-A}\\ \mathbf{Q}^{(\sigma)}_{-B}\end{bmatrix} = \begin{bmatrix}0 & 0 & 0 & e^{i(k_x+k_y)/2}\\ 0 & 0 & e^{i(k_x-k_y/2)} & 0\\ 0 & e^{-i(k_x+k_y)/2} & 0 & 0\\ e^{-i(k_x-k_y)/2} & 0 & 0 & 0\end{bmatrix}\cdot\begin{bmatrix}\mathbf{Q}^{(\sigma)}_{\mathbf{A}}\\ \mathbf{Q}^{(\sigma)}_{B}\\ \mathbf{Q}^{(\sigma)}_{-A}\\ \mathbf{Q}^{(\sigma)}_{-B}\end{bmatrix} = \mathcal{M}_\sigma(m_x)\vec{\mathbf{Q}}_\sigma. \tag{54}$$

In Eq. (51) we have explicitly calculated the (4,1) matrix element of the matrix $\mathcal{M}_\sigma(m_x)$. The other matrix elements can be obtained by suitably changing the sign(s) of the components of the wave vector(s).

E. Transformation by 2_z

We write for irrep $\sigma = 1$ and wave vector $\mathbf{k}_A$ under transformation by 2_z

$$\begin{aligned}M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' &= \lambda'''_\alpha M_\alpha\big(\big[2^R_z\big]^{-1}[N_x,N_y,N_z;\boldsymbol{\tau}_n]\big)\\ &= \lambda'''_\alpha\big(\big[2^R_z\big]^{-1}\Psi^{(A,1)}_{\alpha n}\big)e^{-i\mathbf{k}_A\cdot[2^R_z]^{-1}[\mathbf{N}+\boldsymbol{\tau}_n]}\mathbf{Q}^{(1)}_A + \text{c.c.}\\ &= \lambda'''_\alpha\Psi^{(A,1)}_{\alpha\bar{n}}e^{-ik_x[-N_x-\tau_{nx}+1/2]-ik_y[-N_y-\tau_{ny}]}\mathbf{Q}^{(1)}_A + \text{c.c.}\\ &= \lambda'''_\alpha\Psi^{(A,1)}_{\alpha\bar{n}}e^{-i(-k_x)[N_x+\tau_{nx}-1/2]-i(-k_y)[N_y+\tau_{ny}]}\mathbf{Q}^{(1)}_A + \text{c.c.},\end{aligned} \tag{55}$$

where $\bar{1} = 2$, $3 = 4$, $\bar{5} = 8$, $6 = 7$, and the inverse relations also hold, so that $\Psi^{(A,1)}_{\alpha\bar{n}} = \mu_\alpha\Psi^{(A,1)*}_{\alpha n}$. Also $\lambda'''_\alpha = (-1,-1,+1)$, so that $\lambda'''_\alpha = \mu_\alpha$. Thus Eq. (55) is of the form

$$M_\alpha(N_x,N_y,N_z;\boldsymbol{\tau}_n)' = \mathbf{Q}^{(1)\,\prime}_{-A}\Psi^{(-A,1)}_{\alpha n}e^{-i[(-k_x)(N_x+\tau_{nx})+(-k_y)(N_y+\tau_{ny})]}, \tag{56}$$

with

$$\mathbf{Q}^{(1)\,\prime}_{-A} = 2_z\mathbf{Q}^{(1)}_{-A} = \mathbf{Q}^{(1)}_A e^{-ik_x/2}, \quad \Psi^{(-A,1)}_{\alpha n} = \lambda'''_\alpha\Psi^{(A,1)}_{\alpha\bar{n}} = \Psi^{(A,1)*}_{\alpha n}, \tag{57}$$

in agreement with Eq. (13). Since $\mathbf{Q}^{(1)}_{-B}$ has the same value of k_x, the above result implies that

$$\mathbf{Q}^{(1)\,\prime}_{-B} = 2_z\mathbf{Q}^{(1)}_{-B} = \mathbf{Q}^{(1)}_B e^{-ik_x/2}. \tag{58}$$

The transformation properties of $\mathbf{Q}^{(1)}_X$ are obtained from those of $\mathbf{Q}^{(1)}_{-X}$ by changing the sign of $\mathbf{k}$. A similar analysis for irrep 2 (but with $\Psi^{(A,2)}_{\alpha\bar{n}} = -\mu_\alpha\Psi^{(A,2)*}_{\alpha n}$) leads to the final result that $2_z\vec{\mathbf{Q}}_\sigma = \mathcal{M}(2_z)\vec{\mathbf{Q}}_\sigma$, with

$$\mathcal{M}_\sigma(2_z) = (-1)^{\sigma+1}\begin{bmatrix}0 & 0 & e^{ik_x/2} & 0\\ 0 & 0 & 0 & e^{ik_x/2}\\ e^{-ik_x/2} & 0 & 0 & 0\\ 0 & e^{-ik_x/2} & 0 & 0\end{bmatrix}. \tag{59}$$

F. Transformation by other operations

Here we record the result for translation. For instance, apply Eq. (14) to the magnetization when the transformation operator is a translation $\mathbf{T}$ through a lattice vector:

$$[\mathbf{M}(\mathbf{N}+\boldsymbol{\tau}_n)]' = \mathbf{T}^S\mathbf{M}([\mathbf{T}^R]^{-1}[\mathbf{N}+\boldsymbol{\tau}_n]). \tag{60}$$

This gives, for translation $\mathbf{T}$,

$$[\mathbf{M}(\mathbf{N}+\boldsymbol{\tau}_n)]' = [\mathbf{M}(\mathbf{N}-\mathbf{T}+\boldsymbol{\tau}_n)] = e^{i\mathbf{k}\cdot\mathbf{T}}[\mathbf{M}(\mathbf{N}+\boldsymbol{\tau}_n)]. \tag{61}$$

When the translation $\mathbf{T}$ is through an integer number of lattice constants in the three lattice directions, N_x, N_y, N_z, we write

$$\mathcal{M}_\sigma(\mathbf{T}_{N_x,N_y,N_z}) = \begin{bmatrix} e^{i(k_xN_x+k_yN_y)} & 0 & 0 & 0 \\ 0 & e^{i(k_xN_x-k_yN_y)} & 0 & 0 \\ 0 & 0 & e^{i(-k_xN_x-k_yN_y)} & 0 \\ 0 & 0 & 0 & e^{i(-k_xN_x+k_yN_y)} \end{bmatrix}. \tag{62}$$

G. Composition rules

If $\mathcal{O}^{(1)}$ and $\mathcal{O}^{(2)}$ are two operators, then we might write that

$$\begin{aligned} \mathcal{O}^{(1)}\mathcal{O}^{(2)}(\vec{\mathbf{Q}}_\sigma)_n &\equiv \mathcal{O}^{(1)}[\mathcal{O}^{(2)}(\vec{\mathbf{Q}}_\sigma)_n] \\ &= \mathcal{O}^{(1)}\left[\sum_m (\mathcal{M}_\sigma(\mathcal{O}^{(2)}))_{nm}(\vec{\mathbf{Q}}_\sigma)_m\right] \\ &= \sum_m \sum_s (\mathcal{M}_\sigma(\mathcal{O}^{(2)}))_{nm}(\mathcal{M}_\sigma(\mathcal{O}^{(1)}))_{ms}(\vec{\mathbf{Q}}_\sigma)_s \\ &\equiv \sum_s \mathcal{M}_\sigma(\mathcal{O}^{(1)}\mathcal{O}^{(2)})_{ns}(\vec{\mathbf{Q}}_\sigma)_s, \end{aligned} \tag{63}$$

from which we might conclude that

$$\mathcal{M}_\sigma(\mathcal{O}^{(1)}\mathcal{O}^{(2)}) = \mathcal{M}_\sigma(\mathcal{O}^{(2)})\mathcal{M}_\sigma(\mathcal{O}^{(1)}). \tag{64}$$

One reason this result is wrong is that the first equation of Eq. (63) interprets $\mathcal{O}^{(1)}\mathcal{O}^{(2)}(\vec{\mathbf{Q}}_\sigma)_n$ to mean $\mathcal{O}^{(1)}[\mathcal{O}^{(2)}(\vec{\mathbf{Q}}_\sigma)_n]$, whereas Eq. (64) interprets it to mean $[\mathcal{O}^{(1)}\mathcal{O}^{(2)}](\vec{\mathbf{Q}}_\sigma)_n$. Another problem is that up to now, the operators $\mathcal{O}^{(n)}$ operate on order parameters and *not* on each other. This situation is discussed in detail by Wigner [24]. Instead we assert that

$$\mathcal{M}_\sigma(\mathcal{O}^{(1)}\mathcal{O}^{(2)}) = \mathcal{M}_\sigma(\mathcal{O}^{(1)})\mathcal{M}_\sigma(\mathcal{O}^{(2)}). \tag{65}$$

As an example of Eq. (65) consider the relation from Eq. (3) that $2_z = m_y m_x \neq m_x m_y$. Then, according to Eq. (65) we should have

$$\mathcal{M}_\sigma(2_z) = \mathcal{M}_\sigma(m_y)\mathcal{M}_\sigma(m_x) \neq \mathcal{M}_\sigma(m_x)\mathcal{M}_\sigma(m_y), \tag{66}$$

which the reader can verify using Eqs. (59), (45), and (54).

V. LANDAU FREE ENERGY

A. Minimal (uncoupled) model for order parameters

We start by describing the symmetry of the model when the order parameters X_1 and X_3 at zero wave vector and $\mathbf{Q}_X^{(\sigma)}$ at wave vector $\mathbf{k}_X$ are not coupled to one another. We can imagine that ordering has developed via consecutive continuous transitions, as might happen for a suitable set of parameters having the same symmetry as MGO, but quite different in detail. Although this is not the experimental scenario, it will provide a correct description of the symmetries of the phase. Thus we imagine X_1 and X_3 to be governed by a free energy

$$F_{1,3} = a_1(T - T_1)X_1^2 + u_1X_1^4 + a_3(T - T_3)X_3^2 + u_3X_3^4, \tag{67}$$

and similarly the incommensurate order parameters to be governed by a free energy, the simplest form of which is

$$\begin{aligned} F_X = {} & a_{X,1}(T - T_{X1})\left(\left|\mathbf{Q}_A^{(1)}\right|^2 + \left|\mathbf{Q}_B^{(1)}\right|^2\right) \\ & + a_{X,2}(T - T_{X2})\left(\left|\mathbf{Q}_A^{(2)}\right|^2 + \left|\mathbf{Q}_B^{(2)}\right|^2\right) + \mathcal{O}(|\mathbf{Q}|^4). \end{aligned} \tag{68}$$

We point out that within such a simple theory and barring an unphysical accidental degeneracy, $\mathbf{Q}_X^{(\sigma=1)}$ and $\mathbf{Q}_X^{(\sigma=2)}$ would not have the *same* wave vector [as Eq. (68) assumes] because the exchange interactions are never exactly isotropic in an orthorhombic crystal. However, if the equilibrium value of the two wave vectors are almost equal in a simple approximation, then there are terms in the Landau free energy which lock the two wave vectors into equality [25], and we assume this to be the case here. Then the nature of the ordered phase is dictated by the form of the quartic and higher-order terms of the Landau free energy. Consider, for example, the quartic terms. In the space of $\mathbf{Q}_A$ and $\mathbf{Q}_B$ there are isotropic terms

$$\Delta F = \sum_\sigma u_\sigma\left[\left|\mathbf{Q}_A^{(\sigma)}\right|^2 + \left|\mathbf{Q}_B^{(\sigma)}\right|^2\right]^2. \tag{69}$$

This term would allow for an arbitrary superposition of both wave vectors, $\mathbf{k}_A$ and $\mathbf{k}_B$ within a single domain. However, it has been shown [13] that each domain contains only a single wave vector. That indicates that the free energy includes the term

$$\Delta F = \sum_{\sigma\sigma'} B_{\sigma\sigma'}\left|\mathbf{Q}_A^{(\sigma)}\right|^2\left|\mathbf{Q}_B^{(\sigma')}\right|^2, \tag{70}$$

which strongly disfavors having two wave vectors simultaneously present when $B_{\sigma\sigma'}$ is large and positive. Finally, we point out that we expect terms in the free energy to prevent irreps from having the same phase. At positions where one irrep is maximal, there is usually less phase space into which the other irrep can condense. This argument is reflected by the term [25]

$$\Delta F = A\sum_{X+A,B}\left(\mathbf{Q}_X^{(1)}\left[\mathbf{Q}_X^{(2)}\right]^* + \left[\mathbf{Q}_X^{(1)}\right]^*\mathbf{Q}_X^{(2)}\right)^2, \tag{71}$$

with $A > 0$. This term is proportional to $\cos^2(\Delta\phi)$, where $\Delta\phi$ is the phase difference between the complex-valued order parameters of the two irreps. We expect A to be large and positive, so that the cases $\Delta\phi = \pm\pi/2$ are strongly but equally favored. Thus we write

$$Q_X^{(1)}Q_X^{(2)*} - Q_X^{(1)*}Q_X^{(2)} = 2i\zeta_X\left|Q_X^{(1)}Q_X^{(2)}\right|, \tag{72}$$

where $\zeta_X = \pm 1$ is the phase of $Q_X^{(1)}$ relative to that of $Q_X^{(2)}$ in units of $\pi/2$. (This is the definition of the helicity ζ.)

B. Coupling terms in the free energy

Before proceeding to higher order we emphasize that we only want to enumerate the lowest order terms which have each possible allowed symmetry. To construct such higher than quadratic order terms which are allowed by symmetry, we formulate the following rules. Rule 1: we do not allow a term which includes a factor which itself transforms like unity, such as X_n^2 or $|\mathbf{Q}_X^{(\sigma)}|^2$, because the term without this factor should

TABLE VI. Symmetry of the building blocks.

	Operator	Transforms like
$F_1 =$	X_1X_3	m_xm_y
$F_2 =$	$i\left[\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} - \mathbf{Q}_A^{(1)*}\mathbf{Q}_A^{(2)} - \mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} + \mathbf{Q}_B^{(1)*}\mathbf{Q}_B^{(2)}\right]$	m_z
$F_3 =$	$i\left[\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} - \mathbf{Q}_A^{(1)*}\mathbf{Q}_A^{(2)} + \mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} - \mathbf{Q}_B^{(1)*}\mathbf{Q}_B^{(2)}\right]$	$m_xm_ym_z$
$F_4 =$	$\left\|\mathbf{Q}_A^{(1)}\right\|^2 - \left\|\mathbf{Q}_B^{(1)}\right\|^2$	m_xm_y
$F_5 =$	$\left\|\mathbf{Q}_A^{(2)}\right\|^2 - \left\|\mathbf{Q}_B^{(2)}\right\|^2$	m_xm_y
$F_6 =$	P_z	m_z

already be in our list of allowed terms. Such terms do not lead to a different symmetry. They only make a quantitative change in the response of the system. Rule 2: Due to translational invariance, an allowed term must conserve wave vector. In view of rule 1, rule 2 implies that the incommensurate order parameters can only occur in the combination $\mathbf{Q}_X^{(\sigma)}[\mathbf{Q}_X^{(\sigma')}]^*$ [26]. Rule 3: due to time reversal invariance any term must contain an even number of magnetic order parameters. In view of the previous rules, X_1 and X_3 can only occur in the product X_1X_3. To summarize: the allowed building blocks for invariants are (a) X_1X_3, (b) $i(\mathbf{Q}_X^{(1)}[\mathbf{Q}_X^{(2)}]^* - [\mathbf{Q}_X^{(1)}]^*\mathbf{Q}_X^{(2)})$, (c) $|\mathbf{Q}_A^{(\sigma)}|^2 - |\mathbf{Q}_B^{(\sigma)}|^2$, (d) components of the electric polarization $\mathbf{P}$ or magnetization $\mathbf{M}$. Similar terms in which $\mathbf{P}$ is replaced by the applied electric field or $\mathbf{M}$ is replaced by the applied magnetic field are also possible, but are not considered here. Note that the term $(\mathbf{Q}_X^{(1)}[\mathbf{Q}_X^{(2)}]^* + [\mathbf{Q}_X^{(1)}]^*\mathbf{Q}_X^{(2)})$ is excluded by the potential of Eq. (71). In Table VI we list the symmetry of the various building blocks. To verify these results use Eqs. (35), (45), and (54) to write

$$\begin{aligned} m_x\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} &= \mathbf{Q}_B^{(1)*}\mathbf{Q}_B^{(2)}, \quad m_x\mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} = \mathbf{Q}_A^{(1)*}\mathbf{Q}_A^{(2)}, \\ m_y\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} &= -\mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*}, \quad m_y\mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} = -\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*}, \\ m_z\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} &= -\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*}, \quad m_z\mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} = -\mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*}. \end{aligned} \tag{73}$$

In Appendix C we consider the effect of introducing an arbitrary choice of phase factors into either or both Eqs. (42) and (43). There we show that these choices of phases do not lead to a modification of Eq. (73). One can check that, in agreement with Table VI,

$$\begin{aligned} m_xF_2 = m_yF_2 = -m_zF_2 = F_2, \\ m_xF_3 = m_yF_3 = m_zF_3 = -F_3. \end{aligned} \tag{74}$$

The simplest invariant is

$$\begin{aligned} U &= aF_2F_6 \\ &= ia\left[\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} - \mathbf{Q}_A^{(1)*}\mathbf{Q}_A^{(2)} - \mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} + \mathbf{Q}_B^{(1)*}\mathbf{Q}_B^{(2)}\right]P_z \\ &= -2a\left[\left|\mathbf{Q}_A^{(1)}\right|\left|\mathbf{Q}_A^{(2)}\right|\zeta_AP_z - \left|\mathbf{Q}_B^{(1)}\right|\left|\mathbf{Q}_B^{(2)}\right|\zeta_BP_z\right]. \end{aligned} \tag{75}$$

This is the usual trilinear magnetoelectric interaction [5,6] written in the last line in terms of the helicities ζ_X. Next we have the invariants

$$V_1 = b_1F_1F_4 = b_1X_1X_3\left[\left|\mathbf{Q}_A^{(1)}\right|^2 - \left|\mathbf{Q}_B^{(1)}\right|^2\right], \tag{76}$$

$$V_2 = b_2F_1F_5 = b_2X_1X_3\left[\left|\mathbf{Q}_A^{(2)}\right|^2 - \left|\mathbf{Q}_B^{(2)}\right|^2\right]. \tag{77}$$

In principle we could also list F_4F_5. But when each domain only has a single wave vector, this term is not interesting. Finally we have $W \equiv F_1F_3F_6$:

$$\begin{aligned} W &= icX_1X_3P_z\left[\mathbf{Q}_A^{(1)}\mathbf{Q}_A^{(2)*} - \mathbf{Q}_A^{(1)*}\mathbf{Q}_A^{(2)}\right. \\ &\quad \left. + \mathbf{Q}_B^{(1)}\mathbf{Q}_B^{(2)*} - \mathbf{Q}_B^{(1)*}\mathbf{Q}_B^{(2)}\right] \\ &= -2cX_1X_3\left[\left|\mathbf{Q}_A^{(1)}\right|\left|\mathbf{Q}_A^{(2)}\right|\zeta_AP_z + \left|\mathbf{Q}_B^{(1)}\right|\left|\mathbf{Q}_B^{(2)}\right|\zeta_BP_z\right]. \end{aligned} \tag{78}$$

Again, we omit the terms $F_3F_4F_6$ and $F_3F_5F_6$ because when each domain only has a single wave vector, these interactions are not interesting. The consequences of the potentials U, V, and W for the switching behavior of MGO are discussed in detail in Ref. [13].

We should also note the existence of the invariant

$$\begin{aligned} Y &= \left[eX_1X_3 + \sum_\sigma f_\sigma\left(\left|\mathbf{Q}_A^{(\sigma)}\right|^2 - \left|\mathbf{Q}_B^{(\sigma)}\right|^2\right)\right]P_xP_y \\ &\equiv \left[\chi_E^{-1}\right]_{xy}P_xP_y, \end{aligned} \tag{79}$$

where χ_E is the electric susceptibility tensor. The question is how does Y vary from one domain to the next? As explained below Eq. (80), X_1X_3 is the same within all domains. However, it is clear from Eq. (79) that domains of $\mathbf{k}_A$ and those of $\mathbf{k}_B$ will have different values for Y. It is not clear that this difference is large enough to be experimentally accessible.

C. Equilibrium phases

In this section we minimize the free energy including coupling terms and thereby determine the various equilibrium domains that are possible. This discussion is *not* equivalent to discussing the switching between equilibrium states. To illustrate the difference between these two analyses consider the following two scenarios. In scenario I, one simply cools into the lowest temperature phase and then asks if there is any correlation between the orientation of the net magnetization and that of the net ferroelectric polarization: i.e., in any arbitrarily selected domain are these two collinear vectors always parallel or always antiparallel to one another? The experimental answer is "no" [15]: in some domains the two vectors are parallel and in other domains they are antiparallel. In scenario II, one asks a different question: if the magnetic field is used to reverse the direction of the magnetization in a domain, will that always cause the direction of the ferroelectric

polarization in that domain to reverse? The experimental answer to that question is "yes" [13].

Here we only consider the equilibrium properties and we rely on the experimental observation that each domain contains only one of the two possible wave vectors [13]. So we have therefore two choices for domains: they have either wave vector $\pm\mathbf{k}_A$ or wave vector $\pm\mathbf{k}_B$. We assume that the magnitudes of the order parameters $\mathbf{Q}_X^{(\sigma)}$ are fixed by the terms in the free energy which only depend on these variables, and that domains A and B are related by $|\mathbf{Q}_A^{(\sigma)}| = |\mathbf{Q}_B^{(\sigma)}|$. We assume that the *magnitudes* of $\mathbf{X}_1$ and $\mathbf{X}_3$ (but not their algebraic signs) are similarly fixed and are the same within all domains. For a domain of wave vector $\mathbf{k}_X$ we now determine the number of equilibrium domains consistent with the minimization of the interaction free energy for a domain of wave vector $\mathbf{k}_X$, which we write as

$$F_X = U + V_1 + V_2 + W + \zeta_X^2 P_z^2/(2\chi_E), \tag{80}$$

where χ_E is the electric susceptibility and for later convenience we included a factor $\zeta_X^2 = 1$. Note that X_1 and X_3 only appear in V_1, V_2, and W and only in the combination X_1X_3. Since the magnitudes of X_1 and X_3 are assumed fixed, it is clear that we will only be able to determine $\eta \equiv X_1X_3/|X_1X_3|$ in terms of the parameters of F_X. [This means that η depends on the coefficients b_1 and b_2 in V_1 and V_2, as in Eq. (86), below.] Accordingly, we will have domains in which the sign and magnitude of X_1X_3 is determined so as to minimize the free energy, but the algebraic sign of X_3, the magnetization, can be chosen arbitrarily while keeping the sign of the product X_1X_3 fixed. Similarly, since P_z and ζ_X only appear in U and W in the combination $P_z\zeta_X$, we can only determine the combination $C_X = P_z\zeta_X$ in terms of the parameters of F_X. If we had only the U term, the situation would be simple: C_X would be proportional to a: for a positive, positive helicity would give rise to positive P_z in the $\mathbf{k}_A$ domain and to negative P_z in the $\mathbf{k}_B$ domain. Here, we will include the presumably perturbative effect of the W term [see Eq. (87), below]. As mentioned above, for a given wave vector we have four possible domains corresponding to the choices of signs of $\eta = X_1X_3/|X_1X_3|$ and of the helicity ζ_X (or, equivalently, of the sign of P_z).

For a domain of wave vector $\mathbf{k}_X$ we have the following free energy:

$$\begin{aligned} F_X &= U + V_1 + V_2 + W \\ &= -2a\tau_X C_X|\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}| + \eta\tau_x[b_1|\mathbf{Q}^{(1)}|^2 + b_2|\mathbf{Q}^{(2)}|^2]|X_1X_3| \\ &\quad - 2c\eta C_X|X_1X_3||\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}| + \frac{1}{2\chi_E}C_x^2, \end{aligned} \tag{81}$$

where $\tau_A = -\tau_B = 1$ and we noted that $|Q_A^{(\sigma)}| = |Q_B^{(\sigma)}| \equiv |Q^{(\sigma)}|$. First we minimize with respect to P_z, i.e., with respect to $C_X = P_z\zeta_X$, which leads to

$$\chi_E^{-1}C_X = 2(a\tau_X + c\eta|X_1X_3|)|\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}|, \tag{82}$$

with $\eta = \pm 1$ in the correction term yet to be determined. The equilibrium free energy as a function of η becomes (using $\zeta_X^2 = 1$)

$$\begin{aligned} F_X = &-2\chi_E(a\tau_X + c\eta|X_1X_3|)^2|\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}|^2 \\ &+ \eta\tau_X[b_1|\mathbf{Q}^{(1)}|^2 + b_2|\mathbf{Q}^{(2)}|^2]|X_1X_3|. \end{aligned} \tag{83}$$

Finally, we now minimize this with respect to η. We write

$$F_X = -2\chi_E(a^2 + c^2|X_1X_3|^2)|\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}|^2 + \eta\tau_X|X_1X_3|\mathcal{R}, \tag{84}$$

where

$$\mathcal{R} = b_1|\mathbf{Q}^{(1)}|^2 + b_2|\mathbf{Q}^{(2)}|^2 - 4\chi_E ca|\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}|^2. \tag{85}$$

Minimization with respect to η leads to

$$\eta = -\tau_X\mathcal{R}/|\mathcal{R}|. \tag{86}$$

Note that $\mathcal{R}$ is the same for all domains. Thus in a domain of wave vector $\mathbf{k}_X$

$$\begin{aligned} \chi_E^{-1}C_X &\equiv \chi_E^{-1}P_z\zeta_X \\ &= 2\tau_X(a - c\mathcal{R}|X_1X_3/\mathcal{R}|)|\mathbf{Q}^{(1)}\mathbf{Q}^{(2)}|. \end{aligned} \tag{87}$$

All domains have the same magnitude of P_z, but its sign varies from domain to domain.

To summarize: minimization of $F_X = U + V_1 + V_2 + W$ fixes (1) the value of $P_z\zeta_X$ and (2) the sign of X_1X_3 in terms of the parameters of F_X. But, in addition to the wave vector, the variables X_3 and P_z are broken symmetry variables whose signs vary from one domain to the next. Each domain is characterized by (1) the axis of the wave vector $\hat{\mathbf{k}}_A$ or $\hat{\mathbf{k}}_B$, (2) the sign of P_z [or equivalently according to Eq. (82), the sign of the helicity ζ_X], and (3) the sign of the net magnetization X_3 along $\hat{c}$. But all domains are symmetry related to one another.

VI. CONCLUSIONS

Many of our results for the system Mn_2GeO_4 (MGO) have useful analogs for other noncollinear magnetic incommensurate systems. For instance, note the way we simplified the output of ISODISTORT in Appendix A. Second, the case of MGO illustrates how one introduces order parameters as the amplitudes of the magnetic "modes." This formulation is reminiscent of the description of lattice vibration in terms of normal-mode amplitudes and the symmetry analysis that follows here parallels that of phonon modes. A significant advantage of introducing order parameters is that they conveniently carry with them the symmetry properties of the modes. In Sec. IV we use the known way the magnetization transforms to deduce the way the order parameters transform under the various symmetry operations. In the usual scenario involving magnetic order parameters, the symmetry is trivial. Here in a more complex setting, the analysis is more involved and one has to keep track of what is called here the "wave function." In Sec. V the transformation properties of the order parameters are used to construct the array of higher-order potentials which govern the interaction between the various degrees of freedom which result in having eight possible symmetry-related domains involving choices of wave vector, electric polarization, and net magnetic moment. These higher-order potentials have been shown in Ref. [13] to explain the intricate switching which takes place between these domains under suitable application of electric or magnetic fields. To try explain this switching without introducing order parameters, i.e., by arguing in terms of magnetic wave functions, would be a nightmare.

An essentially equivalent version of this paper is at arXiv:1701.04976 [29].

TABLE VII. Structure of the six modes for $\mathbf{k} = \mathbf{k}_A \equiv (0.138,0.211,0)$ [14,27,28], irrep $D^{(1)}$, for *ch* sites $\boldsymbol{\tau}_1 - \boldsymbol{\tau}_4$ from ISODISTORT for space group $Pnma$ = No. 62 in Ref. [22]. The magnetic moment distribution is $\mathbf{M}(\mathbf{R}+\boldsymbol{\tau}_n) = e^{-i\mathbf{k}\cdot(\mathbf{R}+\boldsymbol{\tau}_n)}\mathbf{m}(\boldsymbol{\tau}_n)$, where the first column lists the real-valued amplitudes which give $\mathbf{m}(\boldsymbol{\tau}_n)$, where the $\mathbf{m}(\boldsymbol{\tau}_n)$ are listed in the form $(R,\theta) \equiv R\exp(i\theta)$. An arbitrary overall phase, the same for *ch* and *pl* sites, is not included in these tables.

AMP		m_x	m_y	m_z		m_x	m_y	m_z
Y_1	$\boldsymbol{\tau}_1$	(1, 0)	(0, 0)	(0, 0)	$\boldsymbol{\tau}_2$	(1, 180)	(0, 0)	(0, 0)
Y_2	$\boldsymbol{\tau}_3$	(1, 0)	(0, 0)	(0, 0)	$\boldsymbol{\tau}_4$	(1, 180)	(0, 0)	(0, 0)
Y_3	$\boldsymbol{\tau}_1$	(0, 0)	(1, 0)	(0, 0)	$\boldsymbol{\tau}_2$	(0, 0)	(1, 180)	(0, 0)
Y_4	$\boldsymbol{\tau}_3$	(0, 0)	(1, 180)[a]	(0, 0)	$\boldsymbol{\tau}_4$	(0, 0)	(1, 0)[a]	(0, 0)
Y_5	$\boldsymbol{\tau}_1$	(0, 0)	(0, 0)	(1, 0)	$\boldsymbol{\tau}_2$	(0, 0)	(0, 0)	(1, 0)
Y_6	$\boldsymbol{\tau}_3$	(0, 0)	(0, 0)	(1, 180)[a]	$\boldsymbol{\tau}_4$	(0, 0)	(0, 0)	(1, 180)[a]

[a]We will reparametrize $Y_6 \to -Y_6$ and $Y_4 \to -Y_4$, so that these phases are regularized.

ACKNOWLEDGMENTS

I thank M. Kenzelmann for inviting me to collaborate in the study of Mn_2GeO_4 for many clarifying discussions of the results of Ref. [13], and for supplying Fig. 4. I also acknowledge discussions concerning irreps with J. White and using ISODISTORT with B. Campbell.

APPENDIX A: MODES FOR k_A

Modes for irreps $D^{(1)}$ and $D^{(2)}$

In Tables VII and VIII we show the modes for irrep $D^{(1)}$ for $\mathbf{k}_A = (k_x,k_y,0)$. Tables IX and X give the analogous data for irrep $D^{(2)}$. From these tables (taken from ISODISTORT) we obtain the magnetization distribution throughout a domain for irrep σ as

$$\mathbf{M}(\mathbf{N}+\boldsymbol{\tau}_n) = \mathbf{m}^{(\sigma)}(\boldsymbol{\tau}_n)e^{i\chi - i\mathbf{k}\cdot(\mathbf{N}+\boldsymbol{\tau}_n)} + \text{c.c.} \qquad \text{(A1)}$$

We allow the modes to have an arbitrary overall phase factor, $\exp(i\chi)$, because the origin of the incommensurate excitation

TABLE IX. As Table VII. Mode structure for $\mathbf{k} = (0.138,0.211,0)$ for irrep $D^{(2)}$.

AMP		m_x	m_y	m_z		m_x	m_y	m_z
Y_1	$\boldsymbol{\tau}_1$	(1, 270)	(0, 0)	(0, 0)	$\boldsymbol{\tau}_2$	(1, 270)	(0, 0)	(0, 0)
Y_2	$\boldsymbol{\tau}_3$	(1, 270)	(0, 0)	(0, 0)	$\boldsymbol{\tau}_4$	(1, 270)	(0, 0)	(0, 0)
Y_3	$\boldsymbol{\tau}_1$	(0, 0)	(1, 270)	(0, 0)	$\boldsymbol{\tau}_2$	(0, 0)	(1, 270)	(0, 0)
Y_4	$\boldsymbol{\tau}_3$	(0, 0)	(1, 90)	(0, 0)	$\boldsymbol{\tau}_4$	(0, 0)	(1, 90)	(0, 0)
Y_5	$\boldsymbol{\tau}_1$	(0, 0)	(0, 0)	(1, 270)	$\boldsymbol{\tau}_2$	(0, 0)	(0, 0)	(1, 90)
Y_6	$\boldsymbol{\tau}_3$	(0, 0)	(0, 0)	(1, 90)	$\boldsymbol{\tau}_4$	(0, 0)	(0, 0)	(1, 270)

is arbitrary. We now write the results of ISODISTORT given in Tables VII and VIII in a simpler, but equivalent form. For irrep D$^{(1)}$ for the *ch* ($\boldsymbol{\tau}_1 - \boldsymbol{\tau}_4$) sites we make the cosmetic replacement

$$m_\alpha^{(1)}(\boldsymbol{\tau}_1) = a_\alpha, \quad m_\alpha^{(1)}(\boldsymbol{\tau}_2) = \mu_\alpha a_\alpha,$$
$$m_\alpha^{(1)}(\boldsymbol{\tau}_3) = b_\alpha, \quad m_\alpha^{(1)}(\boldsymbol{\tau}_4) = \mu_\alpha b_\alpha, \qquad \text{(A2)}$$

where $(\mu_x,\mu_y,\mu_z) = (-1,-1,+1)$. For the *pl* sites, the situation is more complicated. Table VIII gives the terms from ISODISTORT which depend on Z_3 and Z_4 as

$$m_x^{(1)}(\boldsymbol{\tau}_5) = (Z_3 - iZ_4)e^{i\phi}, \quad m_x^{(1)}(\boldsymbol{\tau}_6) = -(Z_3 - iZ_4)e^{i\phi},$$
$$m_x^{(1)}(\boldsymbol{\tau}_7) = (Z_3 + iZ_4)e^{-i\phi}, \quad m_x^{(1)}(\boldsymbol{\tau}_8) = -(Z_3 + iZ_4)e^{-i\phi}. \qquad \text{(A3)}$$

Note that this is a parametrization in terms of three parameters. However, this is an overparametrization: if ϕ is arbitrarily varied, m_x can remain unchanged by a suitable rotation in the complex (Z_3,Z_4) plane. Accordingly, we reproduce these results via a two-parameter representation in terms of the complex-valued variable $z_x = (Z_3 - iZ_4)e^{i\phi}$, so that

$$m_x^{(1)}(\boldsymbol{\tau}_5) = z_x, \quad m_x^{(1)}(\boldsymbol{\tau}_6) = -z_x,$$
$$m_x^{(1)}(\boldsymbol{\tau}_7) = z_x^*, \quad m_x^{(1)}(\boldsymbol{\tau}_8) = -z_x^*. \qquad \text{(A4)}$$

TABLE VIII. As Table VII. Mode structure for $\mathbf{k} = (0.138,0.211,0)$, irrep $D^{(1)}$, for *pl* sites. ISODISTORT sets $\phi = 327.55$, but as we discuss, this value has no significance.

AMP		m_x	m_y	m_z		m_x	m_y	m_z
Z_1	$\boldsymbol{\tau}_5$	(0, 0)	$(1,\phi)$	(0, 0)	$\boldsymbol{\tau}_6$	(0, 0)	$(1,\phi-180)$	(0, 0)
	$\boldsymbol{\tau}_7$	(0, 0)	$(1,360-\phi)$	(0, 0)	$\boldsymbol{\tau}_8$	(0, 0)	$(1,540-\phi)$	(0, 0)
Z_2	$\boldsymbol{\tau}_5$	(0, 0)	$(1,\phi-90)$	(0, 0)	$\boldsymbol{\tau}_6$	(0, 0)	$(1,\phi-270)$	(0, 0)
	$\boldsymbol{\tau}_7$	(0, 0)	$(1,450-\phi)$	(0, 0)	$\boldsymbol{\tau}_8$	(0, 0)	$(1,630-\phi)$	(0, 0)
Z_3	$\boldsymbol{\tau}_5$	$(1,\phi)$	(0, 0)	(0, 0)	$\boldsymbol{\tau}_6$	$(1,\phi-180)$	(0, 0)	(0, 0)
	$\boldsymbol{\tau}_7$	$(1,360-\phi)$	(0, 0)	(0, 0)	$\boldsymbol{\tau}_8$	$(1,540-\phi)$	(0, 0)	(0, 0)
Z_4	$\boldsymbol{\tau}_5$	$(1,\phi-90)$	(0, 0)	(0, 0)	$\boldsymbol{\tau}_6$	$(1,\phi-270)$	(0, 0)	(0, 0)
	$\boldsymbol{\tau}_7$	$(1,450-\phi)$	(0, 0)	(0, 0)	$\boldsymbol{\tau}_8$	$(1,630-\phi)$	(0, 0)	(0, 0)
Z_5	$\boldsymbol{\tau}_5$	(0, 0)	(0, 0)	$(1,\phi)$	$\boldsymbol{\tau}_6$	(0, 0)	(0, 0)	$(1,\phi)$
	$\boldsymbol{\tau}_7$	(0, 0)	(0, 0)	$(1,360-\phi)$	$\boldsymbol{\tau}_8$	(0, 0)	(0, 0)	$(1,360-\phi)$
Z_6	$\boldsymbol{\tau}_5$	(0, 0)	(0, 0)	$(1,\phi-90)$	$\boldsymbol{\tau}_6$	(0, 0)	(0, 0)	$(1,\phi-90)$
	$\boldsymbol{\tau}_7$	(0, 0)	(0, 0)	$(1,450-\phi)$	$\boldsymbol{\tau}_8$	(0, 0)	(0, 0)	$(1,450-\phi)$

TABLE X. As Table VII. Mode structure for $\mathbf{k} = (0.138, 0.211, 0)$ for irrep $D^{(2)}$.

AMP		m_x	m_y	m_z		m_x	m_y	m_z
Z_1	$\boldsymbol{\tau}_5$	$(0, 0)$	$(1, \phi)$	$(0, 0)$	$\boldsymbol{\tau}_6$	$(0, 0)$	$(1, \phi)$	$(0, 0)$
	$\boldsymbol{\tau}_7$	$(0, 0)$	$(1, 540 - \phi)$	$(0, 0)$	$\boldsymbol{\tau}_8$	$(0, 0)$	$(1, 540 - \phi)$	$(0, 0)$
Z_2	$\boldsymbol{\tau}_5$	$(0, 0)$	$(1, \phi - 90)$	$(0, 0)$	$\boldsymbol{\tau}_6$	$(0, 0)$	$(1, \phi - 90)$	$(0, 0)$
	$\boldsymbol{\tau}_7$	$(0, 0)$	$(1, 630 - \phi)$	$(0, 0)$	$\boldsymbol{\tau}_8$	$(0, 0)$	$(1, 630 - \phi)$	$(0, 0)$
Z_3	$\boldsymbol{\tau}_5$	$(1, \phi)$	$(0, 0)$	$(0, 0)$	$\boldsymbol{\tau}_6$	$(1, \phi)$	$(0, 0)$	$(0, 0)$
	$\boldsymbol{\tau}_7$	$(1, 540 - \phi)$	$(0, 0)$	$(0, 0)$	$\boldsymbol{\tau}_8$	$(1, 540 - \phi)$	$(0, 0)$	$(0, 0)$
Z_4	$\boldsymbol{\tau}_5$	$(1, \phi - 90)$	$(0, 0)$	$(0, 0)$	$\boldsymbol{\tau}_6$	$(1, \phi - 90)$	$(0, 0)$	$(0, 0)$
	$\boldsymbol{\tau}_7$	$(1, 630 - \phi)$	$(0, 0)$	$(0, 0)$	$\boldsymbol{\tau}_8$	$(1, 630 - \phi)$	$(0, 0)$	$(0, 0)$
Z_5	$\boldsymbol{\tau}_5$	$(0, 0)$	$(0, 0)$	$(1, \phi)$	$\boldsymbol{\tau}_6$	$(0, 0)$	$(0, 0)$	$(1, \phi) - 180$
	$\boldsymbol{\tau}_7$	$(0, 0)$	$(0, 0)$	$(1, 540 - \phi)$	$\boldsymbol{\tau}_8$	$(0, 0)$	$(0, 0)$	$(1, 360 - \phi)$
Z_6	$\boldsymbol{\tau}_5$	$(0, 0)$	$(0, 0)$	$(1, \phi - 90)$	$\boldsymbol{\tau}_6$	$(0, 0)$	$(0, 0)$	$(1, \phi - 270)$
	$\boldsymbol{\tau}_7$	$(0, 0)$	$(0, 0)$	$(1, 630 - \phi)$	$\boldsymbol{\tau}_8$	$(0, 0)$	$(0, 0)$	$(1, 450 - \phi)$

Similarly, we can reproduce the results of Table VIII from ISODISTORT for m_y on the pl sites by setting $z_y = (Z_1 - iZ_2)e^{i\phi}$ in which case

$$\begin{aligned} m_y^{(1)}(\boldsymbol{\tau}_5) &= (Z_1 - iZ_2)e^{i\phi} = z_y, \\ m_y^{(1)}(\boldsymbol{\tau}_6) &= -(Z_1 - iZ_2)e^{i\phi} = -z_y, \\ m_y^{(1)}(\boldsymbol{\tau}_7) &= (Z_1 + iZ_2)e^{-i\phi} = z_y^*, \\ m_y^{(1)}(\boldsymbol{\tau}_8) &= -(Z_1 + iZ_2)e^{-i\phi} = -z_y^*, \end{aligned} \tag{A5}$$

and for M_z on the pl sites by setting $z_z = (Z_5 - iZ_6)e^{i\phi}$ in which case

$$\begin{aligned} m_z^{(1)}(\boldsymbol{\tau}_5) &= (Z_5 - iZ_6)e^{i\phi} = z_z, \\ m_z^{(1)}(\boldsymbol{\tau}_6) &= (Z_5 - iZ_6)e^{i\phi} = z_z, \\ m_z^{(1)}(\boldsymbol{\tau}_7) &= (Z_5 + iZ_6)e^{-i\phi} = z_z^*, \\ m_z^{(1)}(\boldsymbol{\tau}_8) &= (Z_5 + iZ_6)e^{-i\phi} = z_z^*, \end{aligned} \tag{A6}$$

Similar identifications are made for irrep $D^{(2)}$ and we obtain Eqs. (7) and (8).

APPENDIX B: TEMPERATURE DEPENDENCE OF MODES

Look at Eq. (6). There one sees that each mode involves 12 real parameters (four for each spin component). Thus, there are actually 11 additional modes having the same symmetry as the mode we focus upon. Accordingly, we introduce the corresponding mode amplitudes Q_n, with $n = 1,12$, where the free energy at quadratic order due to the irrep in question in the disordered phase is (with $a_n > 0$)

$$F = \sum_{n=1}^{12} a_n (T - T_n)|Q_n|^2, \tag{B1}$$

where T_1 is the largest T_n, so that the mode labeled "1" is the one that first condenses as the temperature is lowered. To study the mean-field temperature dependence for T just below T_1 we go to higher order:

$$F = -a_1(T_1 - T)|Q_1|^2 - \sum_{n>1}(T_n - T)|Q_n|^2 + u|Q_1|^4 + V, \tag{B2}$$

where $T_1 < T$, $T_n > T$ for all $n > 1$, and $u > 0$. Thereby we find the standard result: $\langle Q_1 \rangle = 0$, for $T > T_1$ and for $T < T_1$,

$$|\langle Q_1 \rangle| = [a_1/(2u)]^{1/2}[T_1 - T]^{1/2}, \tag{B3}$$

where $\langle\ \rangle$ denotes an equilibrium value. In the present case, the terms which modify the critical wave function associated with Q_1 are all even order in the order parameters. The quadratic terms are diagonal by construction of the normal modes. So the leading term which give corrections to the wave function of the critical mode is of the form

$$V = \sum_{n>1} |Q_1|^2 [c_n Q_1 Q_n^* + c_n^* Q_1^* Q_n], \tag{B4}$$

where c_n need not be real valued. For $T < T_1$ the effect of this term is approximately the same as that of

$$V = 2\sum_{n>1} |\langle Q_1 \rangle|^2 [c_n Q_1 Q_n^* + c_n^* Q_1^* Q_n]. \tag{B5}$$

Thus we see that at quartic order there is a mixing of modes governed by the temperature-dependent prefactor proportional to the equilibrium value, $|\langle Q_1 \rangle|^2$, which in mean-field theory is proportional to $T_1 - T$. Of course, this mixing only takes place within the space of modes having the same symmetry as Q_1.

APPENDIX C: PHASE FACTORS

In this section we discuss how the definition of order parameters is subject to inclusion of arbitrary phase factors. As mentioned, this ambiguity is similar to that encountered in a two-sublattice antiferromagnet where one defines the staggered magnetization order parameter $\mathbf{N}$ in terms of the sublattice magnetizations, either as $\mathbf{N} = \mathbf{M}_1 - \mathbf{M}_2$ or as $\mathbf{N} = \mathbf{M}_2 - \mathbf{M}_1$. A macroscopic observable will not depend on the sign of $\mathbf{N}$. We now see how such a phase factor affects our analysis. Equations (41) and (42) define the wave functions $\Psi_{\alpha n}^{(B,\sigma)}$. In principle we can introduce arbitrary phases $\exp(i\phi_\sigma)$ as prefactors in these definitions. It is not useful to go to that level of generality. So we will modify these definitions

by writing

$$\Psi^{(B,1)}_{\alpha n} = \xi_1 \lambda'_\alpha \Psi^{(A,1)}_{\alpha \bar{n}} = \xi_1 \lambda'_\alpha [\mu_\alpha b_\alpha, b_\alpha, \mu_\alpha a_\alpha, a_\alpha; z_\alpha, \mu_\alpha z_\alpha, z^*_\alpha, \mu_\alpha z^*_\alpha]_n, \tag{C1}$$

$$\Psi^{(B,2)}_{\alpha n} = -\xi_2 \lambda'_\alpha \Psi^{(A,2)}_{\alpha \bar{n}} = -\xi_2 \lambda'_\alpha [\mu_\alpha d_\alpha, -d_\alpha, \mu_\alpha c_\alpha, -c_\alpha; -w_\alpha, \mu_\alpha w_\alpha, -w^*_\alpha, \mu_\alpha w^*_\alpha]_n. \tag{C2}$$

with each $\xi_\sigma = \pm 1$. Then, in this version of the theory we have

$$\mathcal{M}_\sigma(m_y) = (-1)^{\sigma+1} \begin{bmatrix} 0 & \xi_1 e^{ik_y/2} & 0 & 0 \\ \xi_2 e^{-ik_y/2} & 0 & 0 & 0 \\ 0 & 0 & 0 & \xi_1 e^{-ik_y/2} \\ 0 & 0 & \xi_2 e^{ik_y/2} & 0 \end{bmatrix}. \tag{C3}$$

These modifications do not affect $\mathcal{M}_\sigma(m_z)$ or $\mathcal{M}_\sigma(2_z)$ because m_z and 2_z transform wave functions into themselves. Similarly, we now have the modified result

$$\mathcal{M}_\sigma(m_x) = (-1)^{\sigma+1} \begin{bmatrix} 0 & 0 & 0 & \xi_2 e^{i(k_x+k_y)/2} \\ 0 & 0 & \xi_1 e^{i(k_x-k_y/2)} & 0 \\ 0 & \xi_2 e^{-i(k_x+k_y)/2} & 0 & 0 \\ \xi_1 e^{-i(k_x-k_y)/2} & 0 & 0 & 0 \end{bmatrix}. \tag{C4}$$

One can verify that these choices of phase do not affect Eq. (73) and thus do not affect the results for the invariant potentials U, V_n, and W.

[1] T. Kimura, T. Goto, H. Shintani, K. Ishizaka, T. Arima, and Y. Tokura, Nature (London) **426**, 55 (2003).

[2] N. Hur, S. Park, P. A. Sharma, J. S. Ahn, S. Guha, and S.-W. Cheong, Nature (London) **429**, 392 (2004).

[3] T. Goto, T. Kimura, G. Lawes, A. P. Ramirez, and Y. Tokura, Phys. Rev. Lett. **92**, 257201 (2004).

[4] H. Katsura, N. Nagaosa, and A. V. Balatsky, Phys. Rev. Lett. **95**, 057205 (2005).

[5] G. Lawes, A. B. Harris, T. Kimura, N. Rogado, R. J. Cava, A. Aharony, O. Entin-Wohlman, T. Yildrim, M. Kenzelmann, C. Broholm, and A. P. Ramirez, Phys. Rev. Lett. **95**, 087205 (2005).

[6] M. Kenzelmann, A. B. Harris, S. Jonas, C. Broholm, J. Schefer, S. B. Kim, C. L. Zhang, S.-W. Cheong, O. P. Vajk, and J. W. Lynn, Phys. Rev. Lett. **95**, 087206 (2005).

[7] A. B. Harris, T. Yildirim, A. Aharony, and O. Entin-Wohlman, Phys. Rev. B **73**, 184433 (2006).

[8] M. Mostovoy, Phys. Rev. Lett. **96**, 067601 (2006).

[9] A. B. Harris, Phys. Rev. B **76**, 054447 (2007); **77**, 019901(E) (2008).

[10] G. Lawes, M. Kenzelmann, and C. Broholm, J. Phys.: Condens. Matter **20**, 434205 (2008).

[11] M. Kenzelmann, G. Lawes, A. B. Harris, G. Gasparovic, C. Broholm, A. P. Ramirez, G. A. Jorge, M. Jaime, S. Park, Q. Huang, A. Ya. Shapiro, and L. A. Demianets, Phys. Rev. Lett. **98**, 267205 (2007).

[12] T. A. Kaplan and S. D. Mahanti, Phys. Rev. B **83**, 174432 (2011).

[13] T. Honda, J. S. White, A. B. Harris, L. C. Chapon, A. Fennell, B. Roessli, O. Zaharko, Y. Murakami, M. Kenzelmann, and T. Kimura, Nat. Commun. **8**, 15457 (2017).

[14] Go to the website "ISODISTORT."

[15] J. S. White, T. Honda, K. Kimura, T. Kimura, Ch. Niedermayer, O. Zaharko, A. Poole, B. Roessli, and M. Kenzelmann, Phys. Rev. Lett. **108**, 077204 (2012).

[16] In Ref. [15] it is shown that below the phase transition at 5.5 K the incommensurate order parameters appear discontinuously and the commensurate order parameters have discontinuities.

[17] T. Honda, Y. Ishiguro, H. Nakamura, Y. Wakabayashi, and T. Kimura, J. Phys. Soc. Jpn. **81**, 103703 (2012).

[18] J. S. White, T. Honda, R. Sibille, N. Gauthier, V. Dmitriev, Th. Strässle, Ch. Niedermayer, T. Kimura, and M. Kenzelmann, Phys. Rev. B **94**, 024439 (2016).

[19] For the complete result see Eq. (35).

[20] I. F. Silvera, Rev. Mod. Phys. **52**, 393 (1980).

[21] R. Sachidanandam and A. B. Harris, Phys. Rev. Lett. **67**, 1467 (1991); Phys. Rev. B **78**, 014407 (2008).

[22] *Space-Group Symmetry*, International Tables for Crystallography Vol. A, edited by T. Hahn, 5th ed. (Springer, New York, 2005).

[23] P. Pfeuty and G. Toulouse, *Introduction to the Renormalization Group and to Critical Phenomena* (Wiley, New York, 1976); A. Aharony, *Dependence of Universal Critical Behavior on Symmetry and Range of Interaction*, in Phase Transitions and Critical Phenomena Vol. 6, edited by C. Domb and M. S. Green (Academic, New York, 1976), pp. 357–424.

[24] E. P. Wigner, *Group Theory and its Applications to the Quantum Mechanics of Atomic Spectra* (Academic, New York, 1959), pp. 105–106.

[25] A. B. Harris, A. Aharony, and O. Entin-Wohlman, J. Phys.: Condens. Matter **20**, 434202 (2008), Sec. 3.2.

[26] Umklapp terms would have to be of very high order and should be completely negligible here.

[27] Taken from ISODISTORT in Feb. 2015.

[28] The fact that this table does not use the exactly correct value for $\mathbf{k}_A$ does not affect the results.

[29] A. B. Harris, arXiv:1701.04976.