Spatial anisotropy of topological domain structure in hexagonal manganites

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The domain structure of hexagonal manganites is simulated based on the phenomenological Ginzburg-Landau theory, and special attention is paid to the evolution of a topological vortex-antivortex pattern with the varying out-of-plane anisotropies of two stiffness parameters for the in-plane (x,y)-plane trimerization amplitude P and out-of-plane (z-axis) polarization Q. It is revealed that the topological domain structure can be remarkably modulated by the stiffness anisotropies. A larger stiffness for Q along the z axis causes the trajectory lines of the vortex nodes and antivortex nodes to be seriously stretched along the z axis, eventually leading to the topological striplike domain pattern. The larger stiffness for either Q or P along the z axis makes the domain walls perpendicular to the z axis wider, while the domain walls parallel to the z axis remain less affected. The present work suggests that the topological domain structure may be controlled by some approaches (e.g., lattice strain) which can change the trimerization stiffness and polarization stiffness in hexagonal manganites.

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I. INTRODUCTION

Improper ferroelectricity has been attracting substantial attention recently not only for a set of challenging issues from the viewpoint of fundamental research but also due to promising application potentials. It is known that ferroelectric (FE) polarization (P) in conventional ferroelectrics appears as the primary order parameter arising from the spatial inversion symmetry breaking [1–4]. However, for those improper ferroelectrics, the polarization plays as the secondary order parameter, and the ferroelectricity may be determined by specific spin ordering (e.g., multiferroicity), charge ordering, and/or structural distortion [5–10]. Along this line, the FE domain structure and domain walls in these improper ferroelectrics may exhibit much more fascinating properties than those in conventional FE systems.

One representative case is the topological domain structures (the so-called cloverleaf domain patterns) observed in hexagonal manganites RMnO 3 [11], where R denotes species Y or a rare-earth ion of small radius (R = Dy, Ho, Er, Tm, Yb, Lu, etc.). Hexagonal YMnO 3 is a FE system with high Curie temperature (T C) [12,13], and the FE transition from the centrosymmetric P6 3/mmc phase to the polar P6 3cm ferroelectric phase has been under debate for a long time [14–18]. Although a direct P6 3/mmc ↔ P6 3cm transition without intermediate phase was reported recently [14–17,19,20], details of the transitions remain yet unclear. For other RMnO 3 compounds, similar problems exist and details of the transitions are under investigation [21,22]. Furthermore, the FE polarization in hexagonal RMnO 3 is generated by the nonlinear coupling between the zone-center polar Γ 1 2 mode and zone-boundary K 3 mode, leading to the improper ferroelectricity. Here, the improper ferroelectricity is believed to be irrelevant to the magnetism although the Mn ion is magnetic [23,24], different from the magnetism-induced multiferroicity.

Nevertheless, the extraordinary topological domain structures in this hexagonal RMnO 3 is highly surprising. The topological features, microscopic mechanism, thermodynamics, and dynamic evolution of such topological domain structures are under intensive investigation [25–29]. As a guide for the eyes, a schematic drawing of the topological domain structure is presented in Fig. 1(a) where the domains are self-organized into a cloverleaf vortex-antivortex pattern in the a(bxy)-planar view. Such a cloverleaf pattern is believed to be driven by an a(bxy)-plane trimerization-type structural transition at temperature T m, which results from the difference in the ionic radii of R and Mn as well as the resultant periodic modulation of the atomic lattice [18,30]. The trimerization corresponds to the periodic tilting of the MnO 3 trigonal bipyramids from the high-symmetry P6 3/mmc structure and the resulting lattice distortion makes the unit cell size three times larger and thus lowers the symmetry to the polar P6 3cm space group. In order to understand such fascinating topology, a phenomenological theory was proposed [19,31], and two order parameters are defined. One is the trimerization amplitude Q which describes the amplitude of MnO 3 trigonal bipyramidal tilting from the c(z) axis and the other is the trimerization phase Φ which scales the tilting orientation. This theory has been successful in explaining the major characteristic observed experimentally, and the main ingredients can be highlighted as follows.

First, given the hexagonal symmetry, the trimerization leads to three types of antiphase domains (α, β, γ), described by the Z 3 symmetry. Each antiphase can supply two different polarizations (upward and downward along the c(z) axis), based on the downward distortion of two-thirds of the R ions and the upward distortion of one-third of R ions or vice versa set by the in or out orientations of the K 3 tilting, leading to the Z 2 symmetry. Second, the consequent six interlocked antiphase domains and FE domains (α +, α −, β +, β −, γ +, γ −) emerge from the same point (node) and form one Z 2 × Z 3 topological defect. The core ingredient of the physics is the interlocking between the structurally antiphase domain walls and FE domain walls. Around a topological defect, there are only two winding orders which are (α +, β −, γ +, α −, β +, γ −) and (α +, γ −, β +, α −, γ +, β −) in a clockwise manner, corresponding to a vortex (clockwise) and an antivortex (counterclockwise) constituting a sixfold topological structure, as shown in Fig. 1(a). This definition may be questionable in the strict sense. To identify a vortex
or an antivortex topological defect in a two-dimensional plane is not difficult. However, we discuss here the three-dimensional topological vortex-antivortex domain networks. It is not straightforward to identify whether a defect is a vortex or an antivortex, and it depends on the orientation of the viewing plane on which the vortex or antivortex emerges [28]. Third, such topological structure depends on the thermal annealing details [32,33], and two types of patterns can be observed. The type-I domain patterns refer to those with roughly equal fractions of upward and downward polarization domains along the c axis and the type-II patterns to those with one dominant polarization either upward or downward. It was indeed found that the surface defects, impurities, or processing details may result in strong self-poling which distorts the topological structure. The so-called “isotropic domain pattern” means that the shapes of domains viewed on two arbitrary planes are statistically similar to each other and do not show serious elongation along any specific direction. If the shape of the domains is seriously elongated along some direction, e.g., the c(z) axis, the domain pattern is said to be anisotropic.

We shall investigate carefully the consequence of the out-of-plane stiffness anisotropies for both Q and P in terms of the domain structure evolution and topology. It is found that the topological characteristic remain unaffected in the parameter space under consideration, while the domain pattern can be seriously modulated by such anisotropies. The remaining part of this paper is organized as follows. In Sec. II we have a brief description of the Ginzburg-Landau theory on the domain structure and also the simulation details. The domain patterns as viewed from an arbitrary orientation upon a spatial coordination transformation will be illustrated in Sec. III. The dependence of the domain structure on the two stiffness parameters will be discussed in Sec. IV. A brief conclusion will be given in Sec. V.

II. PHENOMENOLOGICAL MODEL AND SIMULATIONS

A. Free-energy formulation

We start from the Ginzburg-Landau theory in terms of the variations of trimerization and polarization for hexagonal RMnO₃ [31]. The free-energy density is expressed as

\[ \mathbf{f} = f_L + f_G, \]

where the Landau energy \( f_L \) and gradient energy \( f_G \) can be written as

\[ f_L = \frac{d_0}{2} Q^2 + \frac{b_0}{4} Q^4 + \frac{c_0}{6} Q^6 + \frac{c_0'}{6} Q^6 \cos 6\Phi \]

\[ -g Q^3 P \cos 3\Phi + \frac{g'}{2} Q^2 P^2 + \frac{a_0}{2} P^2, \]

\[ f_G = \frac{1}{2} \sum_{l=x,y,z} \left[ \mathbf{s}_l^\mathbf{Q} (\mathbf{h}_l \mathbf{Q} \mathbf{Q} + Q^2 \mathbf{h}_l \Phi \mathbf{h}_l \Phi) + \mathbf{s}_l^P \mathbf{h}_l \mathbf{P} \mathbf{h}_l \mathbf{P} \right], \]

\[ \mathbf{h}_l = \frac{\partial}{\partial l}. \]

needs additional ingredients besides the intrinsic dependence of the domain pattern on the structural trimerization.

In this work, we intend to discuss this issue from the phenomenological framework [31]. One defines the center of a vortex or antivortex as a node. If the spatial trajectory of a node is a straight line along the c(z) axis, the domain pattern observed on any plane parallel to the c(z) axis must be stripelike [Fig. 1(b)]. However, if the spatial trajectory is curved or spatially entangled, the sixfold vortex-antivortex pattern may be observable on any plane. We shall demonstrate that a coordinate transformation of the topological structure presents a qualitative explanation of the vortex-antivortex pattern on an arbitrary plane as long as some conditions are satisfied. In fact, the stiffness parameters for two order parameters \( Q \) and \( P \) are the controlling factors, making the topological domain pattern to be either isotropic or anisotropic. Here, we may define what is the “isotropic domain pattern” or “anisotropic domain pattern,” which is not strict but a qualitative description of the shape characteristic of the domain structure. The so-called “isotropic domain pattern” means that the shapes of domains viewed on two arbitrary planes are statistically similar to each other and do not show serious elongation along any specific direction. If the shape of the domains is seriously elongated along some direction, e.g., the c(z) axis, the domain pattern is said to be anisotropic.
where $Q$ and $\Phi$, as mentioned, are the amplitude and phase of the trimerization mode ($K_3$), $P$ is the local polarization (or the local amplitude of polar mode $\Gamma_2^3$), $a$ and $b$ are the stiffness parameters for $Q$ and $\Phi$, and $c$ are the constants for the free-energy polynomial on $Q$ extended up to the sixth order, $c_0$ is the amplitude and phase of the trimerization mode ($K_3$) and mode $\Gamma_2^3$, $g'$ is the coupling factor between $Q$ and $P$, and $\alpha_P$ is the self-energy factor of $P$. Coefficients $s_Q$ and $s_P$ scale the energy costs for the spatial variations of $Q$ and $P$ and they are defined as the stiffness parameters for $Q$ and $P$, respectively.

The ground state can be obtained by minimizing the Landau term ($f_L$). The six degenerate ground states can be predicted [36], as determined from the six extrema of $f_L$, by solving the following set of equations:

$$\frac{\partial f_L}{\partial Q} = [a + bQ^2 + (c + c'\cos 6\Phi)]Q^4 - 3gQP \cos 3\Phi + g'P^2]Q = 0,$$

$$\frac{\partial f_L}{\partial \Phi} = (-2c'Q^6 \cos 3\Phi + 3gQP^3) \sin 3\Phi = 0,$$

$$\frac{\partial f_L}{\partial P} = -gQ^3 \cos 3\Phi + (g'Q^2 + \alpha_P)P = 0. \tag{3}$$

It is noted that parameters $Q$, $\Phi$, and $P$ should be scalar fields and the energy terms in Eq. (2) and the derivatives in Eq. (3) should be functionals. However, here we only discuss the ground-state values of these parameters and no spatial variations of them are considered; i.e., only the Landau energy needs to be considered. For simplification consideration, Eqs. (2) and (3) are written in the form of normal derivatives instead of functionals.

A consequent algebraic formulation with Eq. (3) allows the order parameters of the six states:

$$a + bQ^2 + (c + c')Q^4 - 3gQP \frac{gQ^3}{gQ^2 + \alpha_P} + g'\left(\frac{gQ^3}{gQ^2 + \alpha_P}\right)^2 = 0$$

$$\Phi = 0, \pm \pi / 3, \pm 2\pi / 3, \pi$$

$$P = \frac{gQ^3 \cos 3\Phi}{gQ^2 + \alpha_P}. \tag{4}$$

It is clear that polarization $P$ with two degenerate states $\pm P$ is parallel to the $z$ axis, given the six possible values of parameter $\Phi$.

B. Ginzburg-Landau theory on domain structure

Now we consider the domain structure in three-dimensional space. The order parameter $Q$ only has two components ($Q_x, Q_y$), and it scales the amplitude of lattice structure trimerization on the $ab(xy)$ plane [37]:

$$Q_x = Q \cos \Phi, \quad Q_y = Q \sin \Phi. \tag{5}$$

Equation (2) can be rewritten as

$$f_L = \frac{a}{2}(Q_x^2 + Q_y^2) + \frac{b}{4}(Q_x^2 + Q_y^2)^2 + \frac{c}{6}(Q_x^2 + Q_y^2)^3 + \frac{c'}{6}(Q_x^3 - 15Q_xQ_y^2 + 15Q_yQ_x^2 - Q_y^3)$$

$$- g(Q_x^3 - 3Q_yQ_x^2)P + g'\left(Q_x^2 + Q_y^2\right)^2P^2 + \frac{\alpha_P}{2}P^2.$$  

$$f_G = \frac{1}{2} \sum_{l=x,y,z} \left\{s_Q^2 \left[\left(\frac{\partial Q_x}{\partial l}\right)^2 + \left(\frac{\partial Q_y}{\partial l}\right)^2\right] + s_P^2 \left[\left(\frac{\partial P}{\partial l}\right)^2\right]\right\}. \tag{6}$$

The total free energy is

$$F = \iiint_V f \, dV = \iiint_V (f_L + f_G) \, dV, \tag{7}$$

where $V$ is the lattice volume under consideration. The equilibrium state is obtained by locally minimizing the free energy with respect to an arbitrary variable among ($\delta Q_x, \delta Q_y, \delta P$), yielding

$$\delta F = \delta \iiint_V f \, dV$$

$$= \iiint_V \delta Q_x \left(\frac{\partial f_L}{\partial Q_x} - s_Q^2 \frac{\partial^2 Q_x}{\partial x^2} - s_Q^2 \frac{\partial^2 Q_x}{\partial y^2} - s_Q^2 \frac{\partial^2 Q_x}{\partial z^2}\right) \, dV + \iiint_V \delta Q_y \left(\frac{\partial f_L}{\partial Q_y} - s_Q^2 \frac{\partial^2 Q_y}{\partial x^2} - s_Q^2 \frac{\partial^2 Q_y}{\partial y^2} - s_Q^2 \frac{\partial^2 Q_y}{\partial z^2}\right) \, dV$$

$$+ \iiint_V \delta P \left(\frac{\partial f_L}{\partial P} - s_P^2 \frac{\partial^2 P}{\partial x^2} - s_P^2 \frac{\partial^2 P}{\partial y^2} - s_P^2 \frac{\partial^2 P}{\partial z^2}\right) \, dV + \iiint_S \delta Q_x \left(s_Q^2 \frac{\partial Q_x}{\partial x} + s_Q^2 \frac{\partial Q_x}{\partial y} + s_Q^2 \frac{\partial Q_x}{\partial z}\right) \cdot dS$$

$$+ \iiint_S \delta Q_y \left(s_Q^2 \frac{\partial Q_y}{\partial x} + s_Q^2 \frac{\partial Q_y}{\partial y} + s_Q^2 \frac{\partial Q_y}{\partial z}\right) \cdot dS + \iiint_S \delta P \left(s_P^2 \frac{\partial P}{\partial x} + s_P^2 \frac{\partial P}{\partial y} + s_P^2 \frac{\partial P}{\partial z}\right) \cdot dS = 0. \tag{8}$$
where \((i, j, k)\) stand for the unit vectors along the coordinates \((x, y, z)\), and \(S\) is the surface area of the lattice under consideration. Here, the derivatives of \(Q\) and \(P\) are written in the functional form instead of the normal form since the gradient energy is considered. Following the standard procedure, the surface integral terms of Eq. (8) should be zero, allowing the time-independent Ginzburg-Landau equations:

\[
\begin{align*}
\frac{s^2}{2} Q_x^{\prime\prime} + s_y Q_x^{\prime\prime} + s^2 Q_y^{\prime\prime} + s_y Q_y^{\prime\prime} &= -\partial f_L/Q_x = 0, \\
\frac{s^2}{2} P^{\prime\prime} + s_y P^{\prime\prime} + s^2 P^{\prime\prime} + s_y P^{\prime\prime} &= -\partial f_L/P = 0, \\
\end{align*}
\]

noting that parameter \(P\) aligns along the \(z\) axis. The solutions of Eq. (9) accurately describe the topological vortex-antivortex domain structure in the ground state. However, a rigorous solution to Eq. (9) as a general case is not available to us. To track the spatial and temporal evolution of the order parameters, one instead tracks the time-dependent Ginzburg-Landau equation, as shown below.

### C. Dynamic evolution of order parameters

The time-dependent Ginzburg-Landau equation reads

\[
\partial \xi(r, t)/\partial t = -L \delta F/\delta \xi(r, t) + \xi(r, t), \quad \xi = Q_x, Q_y, P, \\
\langle \xi(r, t) \rangle = 0, \\
\langle \xi(r, t) \xi(r', t') \rangle = 2L(k_B T)K_{\epsilon \epsilon} \delta(r - r') \delta(t - t'), \\
\xi', \xi'' = Q_x, Q_y, P,
\]

where \(L\) is the kinetic coefficient, \(r\) is the spatial coordinate, \(t\) is time, \(\delta F/\delta \xi\) represents the thermodynamic driving force for the spatial and temporal evolution of variant \(\xi\), \(\xi\) is the Langevin noise term, \(k_B\) is the Boltzmann constant, \(T\) is temperature, \(K_{\epsilon \epsilon}\) is the Kronecker symbol, and the delta function \(\delta\) applies to both spatial coordinate \(r'(r')\) and time \(t'(t')\). The noise term satisfies the Gaussian distribution and its correlation properties meet the requirements of the fluctuation-dissipation theorem [38,39].

It should be mentioned that we do not consider the kinetic aspect of the domain structure evolution and the temperature is assumed to be far below the ferroelectric transition temperature (close to zero). Therefore, the noise term is simply ignored in the present work. This simplification treatment is based on the consideration that only the quasi-ground-state behaviors are discussed, as often done in the literature (Refs. [40,41]). By the way, the \(T\)-dependent behaviors across the phase transition point were discussed in an earlier work (Ref. [36]) and they will not be addressed in this work.

In proceeding, the spatial and temporal evolutions of parameters \(Q_x, Q_y,\) and \(P\) can be written, respectively, as

\[
\begin{align*}
\frac{\partial Q_x(r, t)}{\partial t} &= L \left[ s^2 \partial^2 Q_x(r, t)/\partial x^2 + s_y \partial^2 Q_x(r, t)/\partial y^2 \right] \\
+ s^2 \partial^2 Q_y(r, t)/\partial z^2 - \partial f_L/Q_x(r, t), \\
\frac{\partial P(r, t)}{\partial t} &= L \left[ s^2 \partial^2 P(r, t)/\partial x^2 + s_y \partial^2 P(r, t)/\partial y^2 \right] \\
+ s^2 \partial^2 P(r, t)/\partial z^2 - \partial f_L/P(r, t).
\end{align*}
\]

We set factor \(L = 1\) and the left-hand side of Eq. (9) is exactly the same as the right-hand side of Eq. (11). It means that the order parameter fields \((Q_x, Q_y, P)\) are the solutions of Eq. (9), given the conditions \(\partial Q_x/\partial t = 0, \partial Q_y/\partial t = 0,\) and \(\partial P/\partial t = 0\) in Eq. (11). Surely, for realistic calculations, these conditions cannot be strictly met unless \(t \to \infty\). We make a truncation of the results as the solutions of Eq. (9), upon sufficient iteration cycling when these parameters remain nearly unchanged with time.

In practical calculations, we take the parameter values given by Artyukhin et al. unless stated elsewhere, as shown in Table I (including \(L = 1\)) which were obtained from the first-principles calculations on YMnO\(_3\) [31]. The standard finite difference scheme (the forward difference equation for parabolic equations) for the first-order and second-order derivatives is employed. The cubic lattice of 320 (x) × 320 (y) × 320 (z) is taken with \(\Delta x = \Delta y = \Delta z = 0.3\) nm and periodic boundary conditions, unless stated elsewhere. The time step for the time integration is \(\Delta t = 0.01\). For a test, we once chose several different time steps from 0.001 to 0.05, and no identifiable difference among them was found in terms of the final domain structure, indicating good convergence of the calculations. The final domain structure is obtained by a sufficient iteration of cycling steps (>1 × 10\(^6\) cycling steps) when parameters \(Q = (Q_x, Q_y)\) and \(P\) do change very slowly with time. The initial values of \(Q_x, Q_y,\) and \(P\) (at \(t = 0\)) over the whole lattice are chosen randomly within the ranges.

**Table I.** The values of physical parameters used in the present calculations (e.g., YMnO\(_3\)) [31]. Note: These values are used in all the calculations unless stated elsewhere.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a_0) (eV Å(^-2))</td>
<td>(-2.626)</td>
</tr>
<tr>
<td>(b_0) (eV Å(^-4))</td>
<td>(3.375)</td>
</tr>
<tr>
<td>(c_0) (eV Å(^-6))</td>
<td>(0.117)</td>
</tr>
<tr>
<td>(c'_0) (eV Å(^-6))</td>
<td>(0.108)</td>
</tr>
<tr>
<td>(a_p) (eV Å(^-2))</td>
<td>(0.866)</td>
</tr>
<tr>
<td>(g) (eV Å(^-4))</td>
<td>(1.945)</td>
</tr>
<tr>
<td>(g') (eV Å(^-4))</td>
<td>(9.931)</td>
</tr>
<tr>
<td>(s_x^2) (eV)</td>
<td>(5.14)</td>
</tr>
<tr>
<td>(s_y^2) (eV)</td>
<td>(5.14)</td>
</tr>
<tr>
<td>(s_z) (eV)</td>
<td></td>
</tr>
<tr>
<td>(\xi) (eV Å(^2))</td>
<td>(15.4)</td>
</tr>
<tr>
<td>(\eta) (eV Å(^2))</td>
<td>(8.88)</td>
</tr>
<tr>
<td>(\zeta) (eV Å(^2))</td>
<td>(8.88)</td>
</tr>
<tr>
<td>(\eta') (eV Å(^2))</td>
<td>(52.7)</td>
</tr>
<tr>
<td>(\eta'') (eV Å(^2))</td>
<td>(1)</td>
</tr>
</tbody>
</table>
Given the values of constants \((a_0, b_0, c_0, g'_0, g''_0, \alpha P)\) and stiffness coefficients \((s'_Q, s'_P)\), a sixfold vortex-antivortex domain structure on the \(xy\) plane can be obtained from Eq. (11). One example is shown in Fig. 2(a) where the contrast scales the variation of \(P\). A closeup image showing a marked antivortex pattern is given on the side. The as-simulated domain pattern is consistent with observed results.

### III. ISOTROPIC DOMAIN STRUCTURE

We perform extensive calculations on the domain structures and evaluate these structures from various aspects. In addition to Fig. 2(a), we also image the domain patterns on arbitrary cross sections and two examples on the \(xz\) and \(yz\) planes are plotted in Figs. 2(b) and 2(c) where a closeup image showing a vortex pattern is given on the side for each case. We do observe statistically similar domain structures on these planes in spite of a slight elongation of the domain shape along a specific direction (e.g., along the \(z\) axis here). It means that the domain structure is roughly isotropic. This feature raises two issues. First, how different are the domain patterns on an arbitrary plane and, second, what is the controlling factor of the domain pattern? For the first issue, we look at the Ginzburg-Landau equation in an arbitrary coordinate frame \((X, Y, Z)\) with the coordinate transformation from \((x, y, z)\) as

\[
\begin{align*}
X &= x \cos \phi + y \cos \theta \sin \phi - z \sin \theta, \\
Y &= -x \sin \phi + y \cos \phi, \\
Z &= x \sin \theta \cos \phi + y \sin \phi + z \cos \theta,
\end{align*}
\]

where \(\phi\) and \(\theta\) are the azimuthal angle and polar angle, respectively. The time-independent Ginzburg-Landau equation [Eq. (9)] is rewritten as

\[
\begin{align*}
(s'_Q \cos^2 \theta + s'_Q \sin^2 \theta) \frac{\partial^2 Q_x}{\partial X^2} + s'_Q \frac{\partial^2 Q_x}{\partial Y^2} + (s'_Q \sin^2 \theta + s'_Q \cos^2 \theta) \frac{\partial^2 Q_x}{\partial Z^2} + (s'_Q - s'_Q) \sin \theta \frac{\partial^2 Q_x}{\partial X Z} + (s'_Q - s'_Q) \cos \theta \frac{\partial^2 Q_x}{\partial Y Z} & = 0, \\
(s'_P \cos^2 \theta + s'_P \sin^2 \theta) \frac{\partial^2 P}{\partial X^2} + s'_P \frac{\partial^2 P}{\partial Y^2} + (s'_P \sin^2 \theta + s'_P \cos^2 \theta) \frac{\partial^2 P}{\partial Z^2} + (s'_P - s'_P) \sin \theta \frac{\partial^2 P}{\partial X Z} + (s'_P - s'_P) \cos \theta \frac{\partial^2 P}{\partial Y Z} & = 0,
\end{align*}
\]

where conditions \(s'_Q = s'_Q\) and \(s'_P = s'_P\) are assumed, implying the \(xy\)-plane isotropy of the stiffness properties. The lattice has the hexagonal symmetry and no in-plane polarization, making this assumption reasonable. In this case, the azimuthal angle \(\phi\) disappears.

It is noted that Eq. (14) recovers back to Eq. (9) as long as the following conditions are satisfied:

\[
\begin{align*}
s'_Q &= s'_Q (s'_Q = s'_Q), \\
s'_P &= s'_P (s'_P = s'_P).
\end{align*}
\]

The simulated domain structures as seen from arbitrary cross sections remain highly similar if Eq. (15) is satisfied. In other words, the domain structure would be spatially isotropic on the scale of domain size, irrelevant of the specific hexagonal symmetry. Nevertheless, it is known that Eq. (15) does not work for \(RMMO_3\), as shown in Table I, but the difference between \(s'_Q\) and \(s'_Q\) is not huge. The fact that \(s'_P > s'_P\) explains why the experimentally observed domain patterns on the \(xz\) and \(yz\) planes are slightly deformed along the \(z\) axis [35]. The relatively big gap between \(s'_P\) and \(s'_P\) does not affect the domain pattern much. Furthermore, Eq. (15) implies that the domain structure can be controlled by the stiffness anisotropy. For example, given the parameters in Table I, the evaluated domain patterns on the \((X, Y), (X, Z),\) and \((Y, Z)\) planes with \(\theta = 145^\circ\) are plotted in Figs. 3(a)–3(c).
for each case a closeup image showing one node is given on the side for reference. It is indicated that the domain structures exhibit the cloverleaf patterns with coherent sixfold topological characteristic on all these planes without much difference. In this sense, one may argue that the domain structure is not so sensitive to the stiffness difference along various orientations. Here it is also noted that a relationship between the stiffness parameters and the hexagonal lattice details is not yet available. In the following, one discusses the influence of the stiffness anisotropies for $Q$ and $P$, i.e., $s_Q^l$ and $s_P^l$ with $l = (x, y, z)$ on the domain structure.

**IV. ANISOTROPIC DOMAIN STRUCTURE**

Given the stiffness anisotropy for $Q$ or $P$, one may define two stiffness anisotropic factors:

$$
\eta_Q = \frac{s_Q^l}{s_Q^Z}, \quad s_Q^l = s_Q^Y, \\
\eta_P = \frac{s_P^l}{s_P^Z}, \quad s_P^l = s_P^Y,
$$

(16)

which scale the anisotropies of $s_Q^l$ and $s_P^l$ with $l = (x, y, z)$. We perform extensive computations given a set of $\eta_Q$ and $\eta_P$. All the values of parameters in Table I except those of $s_Q^Z$ and $s_P^Z$ are taken in the calculations. In the other words, we only deal with the out-of-plane stiffness anisotropies for $Q$ and $P$, while the in-plane stiffness is isotropic for all the cases.

![Diagram](image-url)

**Fig. 3.** The simulated domain patterns on (a) $XY$-plane, (b) $XZ$-plane, and (c) $YZ$-plane, with the coordinate transform angle $\theta \sim 145^\circ$. The domain contrast is scaled by the out-of-plane polarization $P$ as labelled. For each case, a zoom-in image showing one vortex/antivortex node is given on the side as a guide for the eyes.

**Fig. 4.** The simulated domain patterns at three different values of $\eta_Q$ for $Q$. For each case, the domain patterns on the $xy$ plane and $xz$ plane are plotted, respectively. (a,b) $\eta_Q = 1$, (c,d) $\eta_Q = 20$, (e,f) $\eta_Q = 82$. The domain contrast is scaled by the out-of-plane polarization $P$ as labeled. For each case of (a), (b), (c), and (e), a closeup image showing one vortex or antivortex is given in (a1), (b1), (c1), and (e1), respectively. A closeup image from (d) showing one vortex-antivortex pair is given in (d1), and a closeup image from (f) showing the domain alignment along the $x$ axis is given in (f1).

**A. Power-law dependence on $\eta_Q$**

Given $\eta_p = 1.0$, the domain structures on the $xy$ and $xz$ planes corresponding to a series of $\eta_Q$ are calculated and three examples are plotted in Fig. 4 for $\eta_Q = 1.0$ (a,b), 20.0 (c,d), and 82.0 (e,f), respectively. A variation of $\eta_Q > 1$ does not have essential impact on the $xy$-plane domain pattern. However, the $xz$-plane pattern, which is also isotropic at $\eta_Q = 1.0$, experiences gradual stretching along the $z$ axis with increasing $\eta_Q$. This stretching becomes so serious that a striplike pattern is obtained at $\eta_Q = 82.0$. In consequence, the number of vortex-antivortex nodes as counted from the $xz$ plane decreases rapidly with increasing $\eta_Q$.

The invariance of the topology characteristic can be checked in spite of huge differences in domain geometry. Indeed, the topological properties remain robust against the variation of $\eta_Q$. First, the sixfold vortex-antivortex pairs on the $xz$ plane are still identifiable unless the domains become fully striplike, as shown in Fig. 4(d). A closeup image is given in Fig. 4(d1) where a vortex-antivortex pair is displayed. It is guessed that this pair is on the verge of annihilation. Even for an extremely large $\eta_Q (= 82.0)$ where the $xz$-plane pattern is fully striplike, the phase alignment along the $x$ axis still follows the topological series: $(\gamma^+, \alpha^-, \beta^+, \gamma^-, \beta^-, \alpha^-), (\gamma^+, \beta^-, \alpha^+)$, and
tivortex nodes (blue), and vortex-antivortex loops (olive). (a) antivortex nodes, plotted in Figs. 5(a)–5(c) for \( \eta_Q = 1 \), (b) \( \eta_Q = 20 \), and (c) \( \eta_Q = 82 \).

FIG. 5. The spatial trajectory lines of the vortex nodes (red), antivortex nodes (blue), and vortex-antivortex loops (olive). (a) \( \eta_Q = 1 \), (b) \( \eta_Q = 20 \), (c) \( \eta_Q = 82 \).

\( \langle \gamma^- \alpha^+, \beta^- \gamma^+, \alpha^- \rangle \), as demonstrated in Fig. 4(f1). In the other words, the topology is invariant although the domain geometry changes substantially [31,42]. It should be mentioned that the topology of the domain structure seems to be damaged as \( \eta_Q \ll 1 \) and this issue will be discussed elsewhere.

One can also track the spatial trajectories of the vortex and antivortex nodes, plotted in Figs. 5(a)–5(c) for \( \eta_Q = 1 \) (a), 20.0 (b), and 82.0 (c), respectively. Here, the node property (i.e., it is a vortex node or an antivortex node) is defined from the \( xy \)-plane pattern. For clarification, the vortex nodes and antivortex nodes are represented by red and blue lines, while the node loops are marked with olive lines. For \( \eta_Q = 1 \) (\( \eta_P = 1 \)), the trajectories of these nodes have no preferred orientation, suggesting the complexity of the topological domain structure. The trajectory lines and node loops at \( \eta_Q = 20 \) are drawn in Fig. 5(b), showing that the trajectory lines and loops are gradually stretched along the \( z \) axis. Upon \( \eta_Q \) up to 82.0, these trajectory lines become straight, as shown in Fig. 5(c), corresponding to the stripelike pattern shown in Fig. 4(f). The above-discussed results remain qualitatively the same for other \( \eta_P (>1) \).

The impact of \( \eta_Q \) on the domain stretching along the \( z \) axis is easily understood, noting that a large \( s_Q^z \) implies high antiphase wall energy given the same difference in \( Q \) across the antiphase wall. Therefore, the antiphase walls always prefer to align along the \( l \) axis in order to reduce this energy. To characterize the effect of \( \eta_Q \) quantitatively, sufficient sampling statistics on the domain patterns is performed. Without losing the generality, we take the \( 4352 \Delta x \times 4352 \Delta z \) \( xy \)-plane for the statistics and count the domain wall numbers along the \( x \) axis and \( z \) axis. Defining \( N_s \), as the average number of walls along the \( z \) axis and \( N_{xy} \), as the average number of walls along the \( x \) axis, we propose a normalized factor \( \xi = N_s/N_{xy} \) to scale the geometric anisotropy of the domain pattern. The sufficient statistics gives the good power-law dependence: \( \xi \propto 1/\eta_Q^m \) \((m = 0.496 \pm 0.006)\), as shown in Fig. 6 where the best fitting is given by the solid red line. This power law remains applicable for other finite values of \( \eta_P (>1) \).

It is interesting to note that the power-law exponent is \( m \sim 1/2 \) with small uncertainties. To explain this power-law dependence and why \( m = 1/2 \), one consults to a simplified and zero-order argument. Given the Langevin equation in a simplified form from Eq. (11),

\[
\frac{\partial Q_{x,y}(r,t)}{\partial t} = L_s Q_{x,y} \frac{\partial^2 Q_{x,y}(r,t)}{\partial x^2} + L_s \frac{\partial^2 Q_{x,y}(r,t)}{\partial y^2} + L_s \frac{\partial^2 Q_{x,y}(r,t)}{\partial z^2} - L_x \frac{\partial f_l}{\partial Q_{x,y}(r,t)}. \tag{17}
\]

One has the general solutions of two slightly different one-dimensional differential equations:

\[
\frac{d^2 f(x)}{dx^2} = f(x) \Rightarrow f(x) = C_1 e^x + C_2 e^{-x},
\]

\[
\frac{d^2 f(x)}{dx^2} = \frac{1}{\kappa} f(x) \Rightarrow f(x) = C_1 e^{\kappa x} + C_2 e^{-\kappa x}, \tag{18}
\]
where $C_1$, $C_2$, and $\kappa$ ($\neq 0$) are constants to be determined. Clearly, the function $f(x)$ will be $k^{1/2}$ times extended along the $x$ axis if $k^{1/2} > 1$ and $k^{1/2}$ times compressed if $k^{1/2} < 1$. Therefore, for the present cases, the domain scale in terms of $Q_{s}$, along the $z$ axis will be proportional to $S_0^{1/2}$ if $S_0^{1/2}$ stays unchanged. In the other words, exponent $m = 1/2$.

Surely, the power-law behavior may not be applicable if $\eta_Q$ is extremely large where the domains on any plane parallel to the $z$ axis (c axis) become stripelike. This issue deserves to have additional checking in a much larger lattice. It is also noted that the dependence of $\xi$ on $\eta_P$ is weak, and the power-law behavior remains the same upon a variation of $\eta_P$ in a finite range covered in the present simulations.

B. Domain wall width dependences on $\eta_P$ and $\eta_Q$

The influence of factors $\eta_Q$ on the domain structure is remarkable, while that of factor $\eta_P$ is much less. Nevertheless, it is seen that the domain walls can be modulated by both $\eta_Q$ and $\eta_P$, and one case is shown in Fig. 4 where those walls aligned in the $xy$ plane (in-plane walls or $x$ walls) are much wider than those walls along the $z$ axis (out-of-plane walls or $z$ walls), with increasing $\eta_Q$. A similar effect is identified if $\eta_P$ increases. To show this effect clearly, we present a closeup of the domain pattern on the $xz$ plane, focusing on one out-of-plane wall ($z$ wall) and one in-plane wall ($x$ wall). The $P$ profiles across the two types of walls at $\eta_P = 1.0$ and $\eta_P = 12.5$ are presented in Figs. 7(a) and 7(b), respectively, given $\eta_Q = 1.0$. The width of the $x$ walls does increase with increasing $\eta_P$, while the width of the $z$ walls remains less affected. Similar results are obtained if a larger $\eta_Q$ is taken, and the width of the $z$ walls does not change much with increasing $\eta_P$.

We also evaluate the profiles of the other two order parameters $\Phi$ and $Q$ together with the total Landau energy density $f$ across the domain walls. The data taken along the dashed white lines in Figs. 7(a) and 7(b) are plotted in Figs. 7(c)–7(f), respectively. The $Q$ profiles across the V2 line are much broader than those across the other three lines (H1, H2, V1). The $f$ profiles across all these lines remain less different. What should be mentioned is that the energy density at the domain center keeps the same for all the cases although the wall widths are different, giving rise to the same energy density difference $\Delta f_w \sim 0.2$ eV between the domain wall and inner domain. This difference is responsible for the fact that the domain walls are more conductive than the inner parts of the domain [43,44]. It implies that the Landau energy for the $x$ walls is higher than for the $z$ walls as $\eta_Q > 1$ and $\eta_P > 1$.

For a quantitative evaluation of the $\eta_Q$ and $\eta_P$ dependences of the wall widths, extensive simulation samplings on extremely large lattices give good statistics and the results are summarized in Figs. 8(a) and 8(b) where the wall widths $W_z$ for the $x$ walls and $W_z$ for the $z$ walls as functions of $\eta_Q$ and $\eta_P$, respectively, are plotted. Here $W_x$ and $W_z$ are measured by the full width at the half valley of the Landau energy profiles. For $\eta_P = 1$, the obtained $W_z$ as a function of $\eta_Q$ again exhibits a power-law dependence with the best fitted exponent $\beta_Q \sim 0.5$, while the obtained $W_x$ shows nearly no dependence on $\eta_Q$. Similarly, for the varying $\eta_P$, one obtains the power-law behavior for $W_x$ on $\eta_P$ but independence of $W_x$ on $\eta_Q$. The power-law exponent for $W_x(\eta_P)$ at $\eta_Q = 1$ is $\beta_P \sim 0.6$.

To understand the dependences of the domain walls on $\eta_Q$ and $\eta_P$, it should be mentioned that the domain structure in such hexagonal manganites is basically the consequence of the interlocking between ferroelectric domain walls and structural antiphase domain walls. While the ferroelastic anisotropy can be measured by $\eta_Q$, the anisotropy in the electric dipole interaction may be scaled by $\eta_P$. It is reasonable that $W_x$ is a function of both $\eta_Q$ and $\eta_P$ due to the ferroelectric and ferroelastic domain wall interlocking. For the domain walls along the $z$ axis, these walls are the 180° ferroelectric and ferroelastic domain walls. The interlocking seems to be completely relaxed, leading to the independence of $W_x$ on $\eta_P$ and $\eta_Q$. The present simulations suggest that both the stiffness parameters impose comparable impact on locking the domain walls along the $x$ axis, with exponents $\beta_Q \sim 0.5$ and $\beta_P \sim 0.6$. 
$W_x = C_1 \eta_Q^{\beta_Q} + C_2 \eta_P^{\beta_P}$

(a) $W_x$ vs $\eta_Q$ (b) $W_x$ vs $\eta_P$

FIG. 8. The widths of the $x$ walls ($W_x$) and $z$ walls ($W_z$) as a function of $\eta_Q$ at $\eta_P = 1$ (a) and as a function of $\eta_P$ at $\eta_Q = 1$ (b), respectively. The $x$-wall width $W_x$ is fitted using a power-law scaling $W_x = C_1 \eta_Q^{\beta_Q} + C_2 \eta_P^{\beta_P}$ where $C_1$ and $C_2$ are the constants while $\beta_Q$ and $\beta_P$ are the exponents. The best-fitted values of them are inserted in the figures for reference.

C. Discussion

So far, the identified topological vortex-antivortex structures in a number of $R$MnO$_3$ systems are roughly isotropic [33,35]. If the Ginzburg-Landau theory is applicable, it would be expected that the in-plane trimerization and $z$-axis polarization stiffness parameters along various directions are similar. A preliminary understanding of this characteristic, to be checked in the future, can be described as the following.

The vortex-antivortex structure is originated from the in-plane trimerization of the MnO$_5$ bipyramids layers. For a consideration of elastic energy, such in-plane trimerization causes the trimerized MnO$_5$ bipyramids in neighboring in-plane layers to locate at different coordinates. In the other words, if the vortex and antivortex nodes are straight lines along the $z$ axis, the in-plane trimerized MnO$_5$ bipyramids in different layers stack vertically along the $z$ axis, generating very large interlayer elastic energy. This energy may be reduced if these bipyramids in different layers stagger from each other, resulting in the trajectories of the vortex and antivortex nodes wandering spatially. Therefore, one observes the close rings, curved lines, and intercrossed (entangled) trajectories of these nodes [28], and the topological domain pattern becomes roughly isotropic.

For tracking the stiffness anisotropy, it is proposed that a proper tension strain of the lattice along the $z$ axis should benefit the release of the interlayer elastic energy, and then the random wandering of the node trajectories will be gradually suppressed. This corresponds to the in-plane trimerization stiffness enhancement. In this sense, the roughly isotropic vortex-antivortex pattern will be stretched gradually along the $z$ axis with increasing stiffness along the $z$ axis, as observed in the present simulations. Similar discussion on the varying out-of-plane polarization stiffness can be made. The enhanced stiffness seems to be equivalent to the increased polarization gradient energy on the domain walls and a wide wall makes the gradient energy low. A tension strain along the $z$ axis would also benefit this release. In these senses, one expects that a tension strain along the $z$ axis for $R$MnO$_3$ would result in the evolution of the isotropic topological domain structure into stripelike topological domains, which certainly deserves experimental checking [27]. At this stage, we have no knowledge of the relationship between the lattice strain tensors and the stiffness tensor for either $Q$ or $P$, but an engineered strain does make sense in realizing the predictions of this work. Finally, one notices that we only discuss the consequence of the out-of-plane stiffness anisotropies for $Q$ and $P$, while more profound physics associated with the in-plane stiffness anisotropy for $Q$ or $P$ may be expected. For instance, a uniaxial or biaxial in-plane lattice strain can be an effective approach to unveil the physics, which is certainly deserving of future investigation.

V. CONCLUSION

In conclusion, we have investigated carefully the real-space topological domain structures of hexagonal $R$MnO$_3$ manganites from various aspects, based on the phenomenological Ginzburg-Landau theory. First, it has been revealed that the stiffness anisotropies of the domain structure into the out-of-plane stripelike topological geometry. Second, it has been proposed that the geometric anisotropy of the domain structure, which is dependent on the stiffness anisotropy for the in-plane trimerization, follows the power-law scaling while no such dependence on the stiffness anisotropy for the out-of-plane polarization is identified. Third, the stiffness anisotropies for both the in-plane trimerization and the out-of-plane polarization can widen the in-plane domain walls but have no influence on the out-of-plane walls along the $z$ axis. The interlocking between the ferroelectric domain walls and ferroelastic domain walls is suggested to be responsible for these behaviors. The present work provides a comprehensive understanding of the topological domain pattern evolution in hexagonal manganites, addressing the significance of the vortex-antivortex topology and proposing possible modulation of the domain structure by, e.g., strain engineering.

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