The corresponding symmetry-breaking mechanism gives
with two 180
simultaneous polarization along (0
the dimensional-order-parameter symmetry associated with
paraelectric
modynamic path occurring at the transition between the
served in the ferroelectric phase of YMnO
3
proper ferroelectric structure arises directly at the Curie
oretical investigations agree in that the cell-tripled im-
3
tric domain pattern below
of this direct path is the observation of a ferroelec-
N
emerging from one point (Fig. 1(b)). In contrast,
like pattern with six domains of alternating polarization
consistent with the two-
C
C
0
0
0
6 
1
3
0).
The corresponding symmetry-breaking mechanism gives
rise to a total of six domain states. These combine
three antiphase domain states resulting from the loss of
the (a, 0, 0), (a, a, 0) and (0, a, 0) paraelectric translations
with two 180
domain states representing opposite spontaneous polarization along (0, 0, c).
The most surprising feature of the domain pattern ob-
erved in the ferroelectric phase of YMnO
3 [Fig. (1a)] is that the six domain states form an exclusive vortex-like pattern with six domains of alternating polarization emerging from one point (Fig. (1b)). In contrast, N-
branched arrangements of domains with N ≠ 6 [Fig. (1c)]
or their combination into fragmented domain configur-
ations [Fig. (1d)] are absent. The absence of three-
branched domains is particularly puzzling since from a geometrical point of view these states are structurally stable: A shift of a domain wall may shift the point of in-
tersection but this would not break up the three-branch configuration as such. By contrast, a single-center six-branched interlocked domain state is geometrically less stable as it may split into fragmented domains under any small deformation [Fig. (1d)].

The vortex-like six-branch domain pattern in the
hexagonal manganites has been investigated in a variety of approaches so far. Chae et al. analyzed the network of ferroelectric domains by graph theory and Monte-
Carlo simulations. Their work is focused on the description of the experimentally observed domain pattern which is based on the distribution of a single type of vortex and antivortex. A derivation of the structure of the vortices and the resulting domain network was not intended. Artyukhin et al. apply Landau theory for deriving the energetically most beneficial arrangement of domain states around the vortex and find that this is a configuration with six states of alternating polarization. Kumagai et al. investigate the structure and energy of the walls between domain states by density functional theory and confirm the arrangement of states with alternating polarization. All these investigations, however, leave the question of the structure of the domain vortex point on the length scale of the unit cell open. This issue was investigated by transmission electron microscopy with controversial results. Yu et al. report that on the length scale of a unit cell the domains avoid one another so that there is no actual point in which all the domain states meet. In contrast, Zhang et al. observe a meeting of all six domain states within a diameter of four unit cells.

In this report we derive the structure of the domain vortex state in the hexagonal manganites from a combi-
natorial analysis based on symmetry and geometry. We find that three classes of six-branch domain (anti-) vortices are compatible with the site symmetry of the hexag-
onal manganites, even though only one of these classes is realized. Vortices in the basal (a, b) plane originate in the 6m2 high-symmetry lattice point of the paraelectric unit cell. In contrast, vortices in planes parallel to the c axis are expected to fragment which is supported by
recent electron microscopy results. Aspects of topology of the domain vortices and the relation to other types of vortex domain structures are discussed.

II. COMBINATORIAL MODEL FOR DOMAIN VORTICES

A. Symmetry

The exclusive stabilization of the six-branched domain-state vortex shown in Fig. 1(b) in favor of vortices involving a different number of domain states [Fig. 1(c)] or fragmented states around the vortex center. The 684 remaining configurations have a lower symmetry and are therefore unstable and prone to fragmentation. The 36 allowed configurations are divided into three classes of vortex-antivortex configurations denoted \(A^\pm, B^\pm, C^\pm\) [Fig. 2(c)]. They differ by the order of the domain sequences: \(A^+ (032541), B^+ (012345), C^+ (052143)\) and the reversed sequences for \(A^-, B^-, C^-\). Each vortex or antivortex class can exist in six different configurations depending on the position of the domains with respect to an arbitrary fixed direction. Figure 2(d) shows the six configurations of the \(A^+\) class which are denoted \(A_0^+, A_1^+, A_2^+, A_3^+, A_4^+, A_5^+\) depending on the location of the domains 0,1,2,3,4,5 with respect to a fixed in-plane axis, e.g. the two-fold rotation \(2_y\) of the 32 group. Each of the six sites (\(\alpha\) to \(\varphi\)) is compatible with only six vortex configurations as listed in Table 1 the (\(\alpha, \beta, \gamma, \delta, \epsilon, \varphi\)) sites are associated with the \((A^+, B^+, C^+)\) vortices whereas \((\beta, \delta, \phi)\) sites correspond to the \((A^-, B^-, C^-)\) vortices.

B. Connections between two vortices

From the preceding considerations one can deduce the rules which determine the connectivity of a domain network with A, B, and C-type vortices: Two vortices can

TABLE I: Distribution of vortex configurations among the six lattice sites \(\alpha\) to \(\varphi\) shown in Fig. 2(b).

\[
\begin{array}{c|cccccc}
\alpha & A_0^+ & B_0^+ & C_1^+ & B_2^+ & A_3^+ & C_4^+ \\
\beta & B_0^+ & A_2^+ & C_1^+ & A_2^+ & B_3^+ & C_5^+ \\
\gamma & B_0^+ & C_2^+ & A_1^+ & C_2^+ & B_4^+ & A_5^+ \\
\delta & A_0^+ & C_3^+ & B_1^+ & C_3^+ & A_4^+ & B_5^+ \\
\epsilon & C_0^+ & A_1^+ & B_1^+ & A_2^+ & C_4^+ & A_5^+ \\
\varphi & C_0^+ & B_2^+ & A_2^+ & B_3^+ & C_5^+ & A_5^+ \\
\end{array}
\]
be connected by one to six curves separating two domains as shown in Fig. 3. Rule 1: A connection involving more than one curve occurs between a vortex and its antivortex; Rule 2: A connection by a single curve can occur either between a vortex and its antivortex or between two vortices or two antivortices of a univocally determined couple of different classes. Rule 1 is deduced from the property that a vortex is fully determined by an ordered sequence of three numbers. As shown in Fig. 2(a) the sequences (032), (123) and (052) designate the vortices $A^+$, $B^+$ and $C^+$, respectively. Turning in the opposite sense around a vortex, the same sequence of numbers are exclusive to the corresponding antivortices $A^-$, $B^-$ and $C^-$, respectively. Fig. 3(a) illustrates the impossibility of connecting two vortices belonging to different classes by more than one curve. Rule 2 stems from the property that an ordered sequence of two numbers always typifies two vortex classes: For example, the (30) sequence characterizes the $C^+$ and $A^-$ vortices whereas the reversed sequence (03) is associated with $C^-$ and $A^+$, so that $C^+$ and $A^+$ can be connected by a single curve, while an $A^+$ vortex cannot be paired with a $B^+$ or $A^+$ vortex. Table II lists the couples of vortices which can be connected by a single curve.

Figure 3(b) summarizes the different types of connections observed between domain vortices in the network shown in Fig. 3(c). To begin with, a single vortex-antivortex pair connected by six curves is found, forming an “island” (label “6”) surrounded by a single domain. The corresponding pair is topologically trivial since a loop surrounding it runs within a single domain, justifying the terms vortex/antivortex. Two pairs forming “bubbles” (label “5”) surrounded by domains of opposed polarities are observed in Fig. 3(c), each pair being interconnected by five curves. The number of vortex-antivortex pairs increases with the decreasing number of curves connecting them, i.e. seven, eleven, and seventeen pairs are found in Fig. 3(c) for two vortices connected by, respectively, four, three, and one (labels accordingly) curves.

This can be illustrated by the following contraction mechanism which is required for increasing the number of connections between a vortex and its antivortex. The mechanism shown in Fig. 3(d) consists of the removal of an interstitial domain between two identical domains in the loop surrounding a pair of domain vortices. By the removal the two identical domains are merged into a single one, and the connectivity of the pair of domain vortices is increased by one. Thus, step by step, the connectivity is increased by decreasing the number of domains crossed by the loop surrounding the domain-vortex pair until, in the ultimate case, an island-like pair of a vortex and an antivortex with a connectivity of six is obtained. The surrounding domain is determined by the order obeyed during the sequence of successive contractions. If the contraction does not proceed as shown in Fig. 3(d), because of interaction with other nearby vortices or if the contraction involves vortices from different classes, the contraction mechanism leads to a smaller number of connecting curves between the vortices forming a pair.

TABLE II: Pair of neighboring domain states (bold numbers) and the associated couple of vortex classes where they occur. For example, the sequence 43 of domain states occurs in the classes $B^-$ and $C^+$.

|   | 0 | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|---|
| 0 | $A^+ B^+$ | $A^+ C^-$ | $B^- C^+$ |
| 1 | $A^- B^-$ | $B^- C^+$ | $A^- C^-$ |
| 2 | $A^- C^+$ | $A^+ B^-$ | $B^- C^+$ |
| 3 | $A^+ C^-$ | $B^- C^+$ | $B^+ A^-$ |
| 4 | $B^+ C^+$ | $A^- C^+$ | $A^+ B^-$ |
| 5 | $A^- B^-$ | $B^- C^+$ | $A^- C^-$ |

FIG. 2: (a) High-symmetry sites in the basal $(a, b)$ plane of the paraelectric $P6_3/mmc$ unit-cell of the hexagonal manganites. Red ions: O$^{2-}$, blue ions: Y$^{3+}$. (b) Location of the six sites $\alpha$ to $\varphi$ with $6m2$ symmetry in relation to the tripled ferroelectric unit-cell. This permits the formation of a domain vortex pattern which has the chiral point symmetry $32m$, a subgroup of $6m2$. (c) The three classes of vortex-antivortex domain configurations $A^+, B^+, C^-$. (d) The six domain configurations forming the $A^+$ class.
FIG. 3: (a) Illustration of the impossibility of joining two vortices belonging to different classes by more than one curve. (b) Possible connections by 1 to 6 domain walls (labels) between a vortex and its antivortex, and (c) their occurrence in the YMnO$_3$ domain network. (d) Contraction mechanism reducing the number of connecting curves for the $B^-_{1} - B^+_{2}$ pair.

C. Type of vortex networks

The preceding discussion was restricted to the case of two vortices being connected in forming a domain, i.e., a 2-gon configuration. Figure 3(a) shows that $n$-gon vortex configurations with $n$ even and ranging from 0 to at least 16 are observed in YMnO$_3$. The restriction to even-gons, which laid the basis for the graph-theoretical description by Chae et al., points to a pattern consisting of a single class of vortex and antivortex. These can only occur in pairs since identical vortices (like $A^+ - A^+$), as they would be present in the associated odd-gons, cannot be connected, see Rules 1 and 2. In principle, even-gons and odd-gons composed of different types of (anti-)vortices like the one in Fig. 3(c) are allowed by rule 2 but they are never observed. Thus, applying the connectivity rules to the observed domain pattern leads to the conclusion that only one type of vortex-antivortex pair is present in YMnO$_3$. Furthermore, applying the paraelectric symmetry operations to the three classes of vortices shows that subsets of vortices within the $A^\pm$ and $C^\pm$ classes transform into one another under these symmetry operations. Hence, they are energetically equivalent and should therefore both be present or absent in the domain pattern. By contrast, $B^\pm$ vortices always transform into $B^\pm$ vortices. Therefore, one can unambiguously identify the type of the exclusively occurring class of vortices as $B$. Table 1 provides the type of $B^\pm$ vortex associated with each 6$m$2 site in Fig. 2(d).

Two aspects remain to be understood, namely (i) the process leading to the formation of vortices and vortex pairs; (ii) the restriction to a solitary type of vortex and antivortex.

Regarding (i), we assume, supported by theory, that interfaces between odd-numbered or even-numbered domains are so energetically unfavorable that they are
strictly avoided. Therefore, when two domains of same parity, e.g. 0 and 2, get closer they form an intermediate region of opposed parity (012) with the creation of an additional domain-wall. This explains the scarcity of vortex-antivortex pairs connected by a single curve. The interstitial domain created between even or odd domains within the $B^\pm$ pairs is pre-determined as shown in Fig. 2(c): the domain state 1 is formed between 0 and 2, domain state 4 separates 3 and 5 etc. Figures 5(a) and 5(b) illustrate the according defragmentation process and the creation of interstitial domains leading to the formation of a $B^+$ vortex. Figure 5(c) shows how these mechanisms can combine to form a vortex-antivortex pair.

Regarding (ii), we assume that interfaces between domain states as they would occur in A- or C-type vortices are energetically unfavorable in comparison to the interfaces in B-type vortices. Therefore, when two such domain states (such as 0 and 3) get closer, they form intermediate domain states (like 0123) with additional walls of reduced energy. This is supported by Landau and density-functional theory which show that the B-type vortices indeed represent the arrangement with the lowest energy between neighboring domains. Walls between different antiphase domains of different polarization possess the lowest energy. Walls between different antiphase domains of the same polarization are less preferred, and walls between the same type of antiphase domain but opposite polarization are least favorable.

These results lead to the B-type vortices if we associate the sequence 0, 1, 2, 3, 4, 5 of domains to the physical domain states $\alpha^+$, $\beta^-$, $\gamma^+$, $\alpha^-$, $\beta^+$, $\gamma^-$. Note that according to our combinatorial derivation other domain patterns than in the hexagonal manganites may occur in isostructural materials with different energies of the domain states and the walls separating them. An investigation of the hexagonal ferrites might
prove interesting in this respect.\textsuperscript{20} In addition, the preference of a symmetric non-fragmented vortex over a lower-symmetry fragmented one is not necessarily universal. A modification of the thermodynamic parameters might interchange the stability of the two types of configurations. Thus, varying the temperature can, in principle, lead to a \textit{fragmentation transition}. However, this has not yet been observed. Furthermore, because of the perturbation exerted by the polarization field near the vortex core a slight defragmentation on the length scale below the resolution limit of force microscopy experiments may occur after all. This will be investigated further in the following section.

\section{Experimental Investigation of the Vortex Center}

Figure \ref{fig:6} shows low-energy electron microscopy data adapted from Ref.\textsuperscript{21}. The images show two faces of an \textit{ErMnO}_3 sample oriented, respectively, perpendicular (basal \((a,b)\) plane) and parallel to the direction of the spontaneous polarization. While the domain vortex on the \((a,b)\) plane does not show any fragmentation within the resolution limit of about 5 nm, fragmentation on a length scale of 100 nm perpendicular to this plane is obvious. This experimental observation is a striking confirmation of our original conclusion that the high-symmetry points within the \((a,b)\) plane are essential for stabilizing the domain vortex structure with a central meeting point of six domains and 32 symmetry. In the \((a,c)\) or \((b,c)\) plane a point with 32 symmetry or higher does not exist which promotes the observed fragmentation of the vortex. This result is partially supported by transmission electron microscopy. According to Yu et al. domains in the plane parallel to the polarization avoid one another so that there is no actual point in which all the domain states meet.\textsuperscript{19} Zhang et al. observe that the domains approach one another down to a distance of a few unit cells but below that the distinction between domain states is no longer possible. Investigations of domain vortices by transmission electron microscopy in the plane perpendicular to the polarization have not been reported so far.

In summary, starting from the high-symmetry lattice points allowing the formation of a domain-vortex pattern in \textit{YMnO}_3 we showed that three classes of vortex-antivortex configurations need to be considered. The connectivity rules within the domain network reveal that only one of these classes is stabilized. In the formation of the network a number of properties regarding the energy of the walls between different types of domain states have been concluded and found to be in agreement with theory (with a possibility of remaining high-energy islands as shown in Fig.\ref{fig:7}). Electron microscopy confirmed our prediction regarding the formation and fragmentation of domain vortices of different crystallographic orientation.

\section{Other Work on Vortex-Domains}

\subsection{Topology and scaling laws}

Griffin et al. interpreted the arrangement of the ferroelectric domains as the result of a topological phase transition and the domain vortex cores as manifestation of topological defects whose distribution is governed by universal scaling laws.\textsuperscript{23} On first glance this seems to be incompatible with the present work. Despite a number of common properties with topological defects the vortices in \textit{YMnO}_3 clearly appear from our description as geometric defects stabilized by symmetry and not for reasons of topology. Linear topological defects result from...
the breaking of a continuous symmetry: gauge symmetry for superconducting and superfluid vortices or rotational symmetry for disclinations in liquid crystals. These defects are stabilized for purely topological reasons related to the continuous character of the order-parameter space without any room for domains and domain walls. By contrast, the ferroelectric phase in YMnO\(_3\) results from a symmetry-breaking transition creating discrete domains in which domain walls play an essential role for the formation of the domain-vortex network. More generally, within the theory of defects the topological concept applies to the order-parameter space (in the sense of Goldstone variables) that is the topology of the high symmetry group, and not only to real space. It is the interconnection between the two topologies which makes the property of topological defects interesting, whereas in YMnO\(_3\) one simply refers to real space since the topology of the order-parameter space is discrete.

The apparent contradiction can be solved by taking into account the different temperature dependence of the primary \(K_3\) trimerization mode and the improper ferroelectric \(I_3\) mode. Right below \(T_C = 1260\) K the amplitude of the polarization with respect to the trimerization is orders of magnitude smaller than at room temperature. In this case the YMnO\(_3\) lattice is, up to the sixth power of the order parameter, energetically isotropic with respect to the azimuthal direction of the \(K_3\) tilt mode and, hence, isotropic with respect to the phase of the order parameter. Thus, right below \(T_C\) the direction of the \(K_3\)-related tilt, and with it the phase of the of order parameter, is expected to vary continuously as sketched in Fig. 8(a). Therefore, in spite of the discreteness of the YMnO\(_3\) lattice, the phase transition at \(T_C\) would share basic properties with transitions with continuous symmetry breaking being described by an order parameter of \(U(1)\) symmetry, which allows the formation of topological defects.

Upon cooling the ferroelectric polarization grows until it is no longer negligible. As a result, six energetically preferred directions for the \(K_3\)-related tilt emerge and the effective continuous symmetry of the order parameter breaks down into six discrete domain states. The domain pattern sketched in Fig. 8(b) is formed, and at room temperature we find the geometrically protected network of domains that is analyzed in the present work. Ideally, the verification of the model of continuous symmetry-breaking requires the observation of vortices without associated walls near \(T_C\) and the progressive formation of walls upon cooling. However, since the application of techniques like piezoresponse force microscopy in the range of \(10^3\) K is difficult, and because the temperature range in which the symmetry is effectively continuous can be very narrow, an indirect verification with annealing cycles in the vicinity of \(T_C\) with subsequent probing of the domain structure at ambient conditions may suffice.

## B. Other types of vortex domains

Similar domain patterns as the one discussed by us have been previously discussed in different systems. The skyrmion-vortex-like domain patterns predicted to occur in multi-axial ferroelectrics depend only partly on the order-parameter symmetry and require finite size effects for producing the energy-consuming depolarizing fields. The charge-density-wave domains in compounds like 2H-TaSe\(_2\) present six-channel domain configurations. They represent three spatial in-plane orientations and + or − out-of-plane ferroelastic strain, which, however, form an essentially different network with a variety of fragmented configurations related to the random array of dislocations and discommensurations required for their stabilization.

The hexagonal manganites represent, to our knowledge, the first system in which ferroelectric domains displaying an exclusive type of vortex-like configuration have been observed. This is because such configurations result not only from the symmetry of the order-parameter, but also from the high-symmetry of the lattice points on which they form.

### Acknowledgments

The authors thank Nicola A. Spaldin for constructive discussions and Dennis Meier for sharing the data in Fig. 6 with us prior to their publication. P. T. Thanks the ETH Zurich for supporting his stay as a guest professor. M. L. acknowledges support of his position by an ETH Research Grant.

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