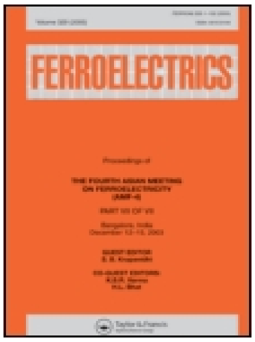

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PYROMAGNETIC DOMAIN WALLS CONNECTING ANTIFERROMAGNETIC NON-FERROELASTIC MAGNETOELECTRIC DOMAINS

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We describe a group-theoretical procedure that enables one to find necessary conditions for the appearance of spontaneous magnetization in domain walls. We illustrate the derivation of wall symmetries on example of Cr_2O_3 and present a brief summary of a systematic analysis of domain walls in antiferromagnetic non-ferroelastic and magnetoelectric phases which shows that in more than 50% of possible domain walls a spontaneous magnetization may appear.

Keywords: Non-ferroelastic domain structures; magnetoelectric domain structures; antiferromagnetic domain structures; structural domain walls; magnetic domain walls; symmetry analysis of domain structures

1. INTRODUCTION

It is well known that a non-homogeneity can induce effects that are forbidden in a homogeneous systems. Thus, e.g. a non-homogeneous temperature or non-homogeneous deformation give rise to an electric polarization in solid or liquid crystals. The very existence of such effects follows from a simple consideration: a non-homogeneity usually decreases the symmetry and thus allows the existence of some properties that are forbidden in the homogeneous system by its higher symmetry.

Domain walls represent a special kind of a non-homogeneity. The lowering of the translation symmetry to two dimensions confines the appearance of new effects to a certain layer the symmetry of which is described by so called *layer* (or *net*) *groups*. These groups further exclude some symmetry elements that may exist in domain bulks, e.g. rotation and inversion axes that are not perpendicular or parallel to the wall (Further information on layer groups can be found, e.g. in Ref. [1]. On the other hand, a planar domain wall (and corresponding three-dimensional twin as well) may be invariant under operations that interexchange domains on two sides of the wall. Since these operations cannot exist in the bulk of domains we encounter an enhancement of symmetry. Thus the symmetry difference between the domain bulks (domain states) and the wall is, in general, not a simple symmetry lowering and can thus induce not only an appearance of new effects in the domain wall that do not exist in the domain states of adhearing domains but also a disappearance of some properties existing in domain states.

A special interest attracts situations where domain walls acquire properties that do not exist in domain bulks. Thus Baryakhatar *et al.* [2] have studied theoretically the appearance of electric polarization in domain walls in magnetically ordered crystals. In this contribution we examine a complementary question of possible appearance of spontaneously magnetized (pyromagnetic) domain walls joining antiferromagnetic domains with zero average magnetization. Since in this problem the interaction between the spin system and the crystal lattice plays a key role we shall confine ourselves to magnetoelectric crystals where this interaction is expected to be particularly strong. We shall further restrict our attention to non-ferroelastic domain walls which bridge domains with the same spontaneous deformation.

For examining tensor properties, e.g. magnetization, polarization, etc., of domains and domain walls a continuum description of the medium is relevant. In this approach the symmetry properties are described by point groups. In what follows, continuum description and point groups are used. Similarly as the appearance of a non-zero magnetizaion $\mathbf{M} \neq 0$ in domain states is conditioned by certain symmetries described by magnetic point groups,[3] a spontaneous magnetization can appear only in those domain walls the layer group symmetries of which are compatible with $\mathbf{M} \neq 0$. In the next section we give an account of all possible layer symmetries of pyromagnetic (spontaneously magnetized) layers. Then we describe how the layer symmetry of a domain wall can be determined and demonstrate this procedure on an example of Cr_2O_3. Finally, we summarize results of a systematic search for pyromagnetic walls in all possible crystallographically different domain walls in antiferromagnetic non-ferroelastic magnetoelectric phases.

2. POINT-GROUP SYMMETRY OF PYROMAGNETIC LAYERS

Point group symmetry of domain walls in materials with magnetic structure is described by crystallographic magnetic layer groups. These groups are expressing possible symmetries of magnetic layers. Magnetic layer groups comprise the 31 classical (without time inversion) point groups of two-sided plane [4] and the 63 non-trivial magnetic layer groups that can be derived from the halving subgroups of the 31 groups of two-sided plane. From these 94 magnetic layer groups only 42 are pyromagnetic allowing a non-zero magnetization which can be in 20 cases accompanied by a simultaneous appearence of a non-zero spontaneous polarization.

With a symmetry operation of a magnetic layer group one can associate two attributes:

(i) The first one, common to all layer groups, indicates whether the operation keeps unchanged the normal **n** to the plane or inverts it into-**n**. We shall *underline* those operations that invert **n** and, therefore, also the sides of the layer. If the symbol of the layer group contains underlined operation(s) then the layer has a non-trivial symmetry of a two-sided plane whereas in the opposite case the symmetry properties of the layer are described by an one-sided plane group. The underlining further specifies the orientation of the operataions 2 and m with respect to the layer (e.g., 2 and m are perpendicular to the layer, whereas $\underline{2}$ and $\underline{m}$ are parallel to the layer).

(ii) The second attribute, specific to magnetic layer groups, indicates whether the operation includes time-inversion or not. We shall use a *prime* to indicate the presence of the time-inversion. If the symbol of a magnetic layer group contains primed operation(s) then the layer has a non-trivial magnetic symmetry.

With exception of trivial triclinic symmetry 1 or $\bar{1}$, the magnetic layer group L predetermines (in most cases completely, in a few layer groups only partially) the orientation of the magnetization **M** and polarization **P** with respect to the normal **n** of the layer. We arrange, therefore, the list of pyromagnetic layer groups L according to these configurations:

1. Pyromagnetic ($\mathbf{M} \neq 0$) non-pyroelectric ($\mathbf{P} = 0$) layers

1.1. **Magnetization M is perpendicular to the layer, i.e. M || n. for the layer groups**

$$L = 2/\underline{m},\ \underline{2}'\underline{2}'2\ (\mathrm{me}), m'm'\underline{m}, \underline{\bar{4}}(\mathrm{me}), 4\underline{2}'\underline{2}'(\mathrm{me}), \underline{\bar{4}2}'m'(\mathrm{me}), 4/\underline{m},$$
$$4/\underline{m}m'm', 3\underline{2}'(\mathrm{me}), \underline{\bar{3}}, \underline{\bar{3}}m', \underline{\bar{6}}, 6/\underline{m},\ 6\underline{2}'\underline{2}'(\mathrm{me}), \underline{\bar{6}}m'\underline{2}', 6/\underline{m}m'm'$$

1.2. **Magnetization M is parallel to the layer, i.e. M $\perp$ n, and its direction is specified by symmetry** for the layer groups

$$L = \underline{2}/m(\underline{2}\|M \perp m), \underline{22}'2'(\mathrm{M}\|\underline{2})(\mathrm{me}), m'm\underline{m}'(2\|\mathrm{M} \perp m).$$

1.3. **Magnetization M is parallel to the layer i.e. M $\perp$ n and its direction is** *not* **specified by symmetry** for the layer group

$$L = 2'/\underline{m}'(2' \perp \mathrm{M}\|\underline{m}').$$

1.4. **Magnetization M is out of the layer and its direction is confined to a plane perpendicular to the layer** for the layer group

$$L = \underline{2}'/m'(\underline{2}' \perp \mathrm{M}\|m').$$

1.5. **Direction of M is not restricted by symmetry** for the non-trivial triclinic layer group

$$L = \underline{\bar{1}}.$$

The symbol (me) indicates that the group L is magnetoelectric, nevertheless, the form of the magnetoelectric tensor is such that the polarization **P** equals zero.

2. Pyromagnetic (M $\neq$ 0) and pyroelectric (P $\neq$ 0) layers

2.1. **Magnetization M is perpendicular to the layer, M||n,** and

2.1.1. polarization **P** is also perpendicular to the layer, **P**||**n** for the layer symmetries

$$L = 2, m'm'2, 4, 4m'm', 3, 3m', 6, 6m'm'$$

(in all these layer groups both **M** and **P** are parallel to the unique polar axis),

2.1.2. polarization **P** is parallel to the layer, **P**$\perp$**n**, and its direction is determined by symmetry for the layer group

$$L = \underline{2}'\underline{m}m'(\mathrm{P}\|\underline{2}'),$$

2.1.3. polarization **P** is parallel to the layer, **P**$\perp n$, but its direction is *not* determined by symmetry for the layer group

$$L = \underline{m}(\mathbf{P}\|\underline{m}).$$

2.2. Magnetization M is parallel to the layer , M⊥n, its direction is specified by symmetry, and

2.2.1. polarization **P** is perpendicular to the layer, **P**||**n**, for the layer group

$$L = m'm2'(\mathbf{M} \perp m, \mathbf{P}\|2'),$$

2.2.2. polarization **P** is parallel to the layer and parallel to **M**, **P**||**M**⊥**n**, and the direction of **P** is specified by symmetry for the layer groups

$$L = \underline{2}, \underline{2}m'\underline{m}'$$

(in both layer groups $\mathbf{M}\|\mathbf{P}\|\ \underline{2}$),

2.2.3. polarization **P** is parallel to the layer, **P**⊥**n**, perpendicular to **M**, **P**⊥**M** and its direction is determined by symmetry for the layer group

$$L = \underline{2}'m\underline{m}'(\mathbf{M} \perp m, \mathbf{P}\|\underline{2}'),$$

2.2.4. polarization **P** is confined to a plane perpendicular to the layer for the layer group

$$L = m(\mathbf{M} \perp m, \mathbf{P}\|m).$$

2.3. Magnetization M is parallel to the layer, M ⊥ n its direction is ***not*** **specified by symmetry,** and

2.3.1. polarization is perpendicular to the layer, **P**||**n**, for the layer group

$$L = 2'(\mathbf{P}\|2'),$$

2.3.2. polarization **P** is parallel to the layer, **P** ⊥ **n** but its direction is *not* determined by symmetry for the layer group

$$L = \underline{m}'.$$

2.4. Magnetization M is out of the layer but its direction is confined to a plane perpendicular to the layer, and

2.4.1. polarization **P** is parallel to the layer, **P**⊥**n**, and perpendicular to **M** for the layer group

$$L = \underline{2}'(\mathbf{M} \perp \mathbf{P}\|\underline{2}'),$$

2.4.2. polarization **P** is in the same plane as **M** but there is no specific relation between directions of **P** and **M** for the layer group

$$L = m'(\mathbf{M}\|m', \mathbf{P}\|m').$$

2.5. Direction of neither M nor P is determined by symmetry for the layer group of trivial triclinic symmetry

$$L = 1.$$

3. WALL SYMMETRY AND SECTIONAL LAYER GROUPS

Let us consider a planar domain wall with orientation (hkl) between two domains with domain states S_1 and S_2 (*domain states* are bulk structures of domains with no specification of the domain boundaries). We shall use for such a wall the symbol $|S_1(hkl)S_2|$. The orientation can be also expressed by the normal **n** to the plane, $|S_1(hkl)S_2| \equiv |S_1(\mathbf{n})S_2|$. If the orientation of the wall is not essential, or if it is known from the context, we shall use a short symbol $|S_1|S_2|$.

Symmetry of the wall $|S_1(\mathbf{n})S_2|$ is described by a layer group $T_{12}(\mathbf{n})$. An operation u of this group must fulfill two *necessary* conditons:

(i) Being an operation of a layer group, u must either keep **n** unchanged or invert it into $-\mathbf{n}$. Operations of the latter type are underline operations (according to the convention introduced in the preceding Part 2).

(ii) An operation $u \in T_{12}(\mathbf{n})$ either keeps both S_1 and S_2 unchanged or exchanges them, i.e. $uS_1 = S_2$. We shall denote such *state-exchanging operations* by an asterisk *.

These two conditions can be fulfilled in four different ways each of which specifies how the operation transforms the normal **n** and the domain states S_1,S_2:

a) An operation f_{12} that neither changes the normal **n** nor the states S_1 and S_2 obviously keeps the wall $|S_1|S_2|$ unchanged. Such operations are called *trivial symmetry operations of a domain wall.*

b) An operation $\underline{s}_{12}$ that inverts the normal n exchanges half-spaces on the left and right sides of the wall. Since these half-spaces are occupied by domain states S_1 and S_2 (say e.g., that S_1 is on the left side and S_2 on the right side of the wall) this exchange of half-spaces is accompanied by an exchange of domain states on both sides of the wall. If the opeartion $\underline{s}_{12}$

changes directly (i.e. not via half-spaces exchange) neither S_1 nor S_2 then this operation results in a domain wall which has domain state S_2 on the left side and the domain state S_1 on the right side of the wall. We call such a wall $|S_2|S_1|$ the *reversed domain wall* with respect to the initial wall $|S_1|S_2|$.

c) An operation r^*_{12} that exchanges domain states S_1 and S_2 but does not invert the normal **n** (i.e. keeps the half-spaces on both sides of the wall in their positions) transforms the initial domain wall also into a reversed wall $|S_2|S_1|$.

d) An operation $\underline{t}^*_{12}$ that exchanges half spaces (inverts **n**) *and independently* exchanges domain states S_1, S_2, transforms the wall into itself. This operation $\underline{t}^*_{12}$ is, therefore, a symmetry operation of the wall. Such operations are called *non-trivial symmetry operations of a domain wall.*

Table I summarizes the action of these four types of operations on the normal **n**, on both domain states S_1, S_2 and on the domain wall $|S_1|S_2|$. To make the effect of the operation more comprehensible, we add in the wall symbol to the lower end of the central vertical line a small horizontal "foot" which can be associated with the minus side of the normal **n**. The change in the direction of this small horizontal line indicates the exchange of the half-spaces.

The layer group $T_{12}(hkl)$ that describes the symmetry of the wall $|S_1(hkl)S_2|$ consists of all trivial and non-trivial symmetry operations of that wall. We shortly recapitulate the procedure that enables one to find, for a given domain states S_1, S_2 and the orientation (hkl), the group $T_{12}(hkl)$.[5–7] We restrict out considerations to non-ferroelastic domain walls that join domain states with equal deformation. Then the symmetry groups of both domain states S_1 and S_2 are equal, $F_1 = F_2$.[8]

First, let us consider a trivial domain wall $|S_1|S_1|$ with the same domain state S_1 on both sides of the wall. Its symmetry consists of all operations of the group F_1 that leave invariant a plane (hkl) transecting the domain state S_1 with symmetry F_1. Such a layer group is called the *sectional layer group of the plane (hkl)*[4, 9] and will be denoted $\bar{F}_1(hkl)$, or, shortly, $\bar{F}_1$. This sectional layer group contains all trivial symmetry operations f_{12} of the domain wall and the set of all these operations forms a one-sided layer group $\hat{F}_1$ which is a subgroup of the (generally two-sided) sectional layer group $\bar{F}_1, \hat{F}_1 \leq \bar{F}_1$. If $\underline{s}_{12}$ is a side-reversing operation then the left coset $\underline{s}_{12}\hat{F}_1$ comprises all such side-reversing operations of $\bar{F}_1$ and

$$\bar{F}_1 = \hat{F}_1 + \underline{s}_{12}\hat{F}_1. \qquad (1)$$

Thus the trivial part $\hat{F}_1$ of the wall symmetry can be deduced from the sectional layer group $\bar{F}_1$ as its halving one-sided subgroup.

If $\underline{t}^*_{12}$ is a side and state reversing operation then all operations of the set (left coset) $\underline{t}^*_{12}\hat{F}_1$ are side and state reversing operations as well. It can be shown that these are all non-trivial symmetry operations of the wall. Therefore, the symmetry group T_{12} of the wall equals

$$T_{12} = \hat{F}_1 + \underline{t}^*_{12}\hat{F}_1. \tag{2}$$

To derive the side and state exchanging operations that appear in Eq. (2) it is useful to introduce the concept of a *non-ordered domain pair* $\{S_1, S_2\}$ which is just a set consisting of two domain states S_1, S_2. The symmetry group J_{12} of this non-ordered non-ferroelastic domain pair equals[8]

$$J_{12} = F_1 + j^*_{12}F_1, \tag{3}$$

where F_1 is the symmetry of S_1 and S_2, and j^*_{12} exchanges S_1 and S_2, $j^*_{12}S_1 = S_2, j^*_{12}S_2 = S_1$. Since for a non-ordered domain pair $\{S_1, S_2\} = \{S_2, S_1\}$, the symmetry operations comprised in the left coset $j^*_{12}F_1$ are also symmetry operations of the domain pair $\{S_1, S_2\}$.

It can be shown that the sectional layer group $\bar{J}_{12}$ of J_{12} along the plane (hkl) has the form[5-7]

$$\bar{J}_{12} = \hat{F}_1 + \underline{t}^*_{12}\hat{F}_1 + r^*_{1j}\hat{F}_1 + \underline{s}_{12}\hat{F}_1, \tag{4}$$

where the operations $\underline{t}^*_{12}, \underline{s}_{12}$ and r^*_{12} are defined in Table I. Comparing Eq. (4) with Eq. (2) we see that the symmetry group T_{12} of the wall is a halving subgroup of the sectional layer group $\bar{J}_{12}$. Thus the task of finding the symmetry group $T_{12}(hkl)$ of the wall $|S_1(hkl)S_2|$ consists in the determination of two sectional layer groups $\bar{F}_1(hkl)$ and $\bar{J}_{12}(hkl)$, and their halving subgroups $\hat{F}_1(hkl)$ and $T_{12}(hkl)$, resp.

The last two left cosets in Eq. (4) assemble operations that transform the wall $|S_1(hkl)S_2|$ into a *reversed wall* $|S_2(hkl)S_1|$ with opposite order of

TABLE I Action of four types of operations u on a domain wall

u	un	uS_1	uS_2	$u\lvert S_1\rfloor S_2\rvert$	wall
f_{12}	$\mathbf{n}$	S_1	S_2	$\lvert S_1\rfloor S_2\rvert$	initial wall
$\underline{s}_{12}$	$-\mathbf{n}$	S_1	S_2	$\lvert S_2\lfloor S_1\rvert$	side-reversed wall
r^*_{12}	$\mathbf{n}$	S_2	S_1	$\lvert S_1\rceil S_2\rvert$	state-reversed wall
$\underline{t}^*_{12}$	$-\mathbf{n}$	S_2	S_1	$\lvert S_1\lfloor S_2\rvert$	initial wall

domain states. Accordingly, one can distinguish *reversible walls* for which $\bar{J}_{12} > T_{12}$, and *irreversible walls* with $\bar{J}_{12} = T_{12}$. In the former case the wall $|S_1|S_2|$ is symmetrically equivalent with the reversed wall $|S_2|S_1|$, whereas in the latter case it is not.

4. ANALYSIS OF DOMAIN WALLS IN ANTIFERROMAGNETIC MAGNETOELECTRIC PHASES

As an illustrative example we shall first analyse the walls in Cr_2O_3 crystal with symmetries $G = \bar{3}m.1' = \bar{3}'m'.1'$ and $F = \bar{3}'m'$ of the high temperature paramagnetic and the low temperature antiferromagnetic phase, resp. Two domain states S_1 and S_2 have the same symmetry $F_1 = F_2 = F$ and the operation $g^*_{12} = 1'$ exchanges these domain states. The domain pair $\{S_1, S_2\}$ has the symmetry

$$J_{12} = F_1 + g^*_{12} F_1 = \bar{3}'m' + 1'^*\{\bar{3}'m'\}. \qquad (5)$$

In Table II we present layer groups $\hat{F}_1$, the wall symmetry T_{12}, $\bar{J}_{12}$, the spontaneous magnetization $\mathrm{M}(W_{12})$ and the spontaneous polarization $\mathbf{P}(W_{12})$ in the wall $|S_1(hkl)S_2|$, and the spontaneous magnetization $\mathbf{M}(W_{21})$ and the spontaneous polarization $\mathbf{P}(W_{21})$ in the reversed wall $|S_2(hkl)S_1|$.

From the Table II it follows that domain walls between two antiferromagnetic magnetoelectric domains in Cr_2O_3 can carry a non-zero spontaneous magnetization for any orientation of the wall. For any orientation the walls exhibit no magnetoelectric effect and carry no spontaneous polarization. All walls are reversible and the spontaneous magnetization in the initial and the reversed domain walls in antiparallel.

Now, we present a short summary of a systematic study of domain walls in a broader class of materials. We have restricted our analysis to domain

TABLE II Layer group symmetries, spontaneous magnetization **M** and spontaneous polarization **P** of domain walls in Cr_2O_3

(hkil)	$\hat{F}_{12}$	T_{12}	$\bar{J}_{12}$	$\mathbf{M}(W_{12})$	$\mathbf{M}(W_{21})$	$\mathbf{P}(W_{12})$	$\mathbf{P}(W_{21})$
(0001)	$3m'$	$\underline{\bar{3}}^*m'$	$\bar{3}m\cdot 1'$	$\mathbf{M}\|\mathbf{n}$	$-\mathbf{M}\|\mathbf{n}$	$\mathbf{P}=0$	$\mathbf{P}=0$
$(2\bar{1}\bar{1}0)$	2	$2/\underline{m}^*$	$2/m\cdot 1'$	$\mathrm{M}\|\mathrm{n}$	$-\mathbf{M}\|\mathbf{n}$	$\mathrm{P}=0$	$\mathrm{P}=0$
$(0h\bar{h}0)$	m'	$\underline{\underline{2}}'^*/m'$	$\underline{2}/m\cdot 1'$	M[1]	−M[1]	$\mathrm{P}=0$	$\mathrm{P}=0$
(hkil)	1	$\underline{\bar{1}}$	$\bar{1}\cdot 1'$	M[2]	−M[2]	$\mathrm{P}=0$	$\mathrm{P}=0$

[1]symmetry confines **M** to the plane m' perpendicular to the wall.
[2]direction of **M** is not restricted by symmetry.

walls that join two domain states with the same deformation (non-ferroelastic domain pairs) but with different magnetoelectric tensor components (in non-ferroelastic domain pairs components of the magneto-electric tensor differ only in sign or are equal). There exist 141 non-equivalent classes on non-ferroelastic magnetoelectric domain pairs.[10] From them we have chosen pairs with antiferromagnetic domain states, i.e. with zero average magnetization ($\mathbf{M}=0$). This condition requires that the group F_1 of both domain states must belong to one of $58-19=39$ magnetetoelectric point groups. Six of these groups are pyroelectric, i.e. they allow the existence of a non-zero spontaneous polarization, $\mathbf{P}\neq 0$.

From each class of crystallographically equivalent domain pairs we have examined just one representative domain pair. For each such domain pair we have found the layer groups $T_{12}(hkl)$ for all crystallographically non-equivalent orientations (hkl). It has turned out that from the total of 141 pairs only 83 domain pairs can generate walls that can carry, at least for some orientations (hkl), a non-zero magnetic moment ($\mathbf{M}\neq 0$). For 41 domain pairs the domain walls are for all possible orientations pyromagnetic, for 42 pairs are non-pyromagnetic only walls that are perpendicular to the axis of the order 3, 4 and 6. One exception of this rule appears in orhorhombic system where the non-pyromagnetic wall is perpendicular to one of the 2-fold axes. More detailed results and discussion will be published elsewhere.

We can conclude with a brief summary: Our analysis has shown that more than 50% of domain walls in antiferromagnetic non-ferroelastic magneto-electric phases can carry spontaneous magnetization. We should add, however, that the symmetry analysis provides only necessary conditions for the existence of spontaneous magnetization and does not say anything about the magnitude of this effect. Deduced layer groups of domain walls represent the highest possible symmetries. A concrete topological structure of the wall may only decrease this symmetry and cannot, therefore, change our general conclusions.

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