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LETTER TO THE EDITOR

Domain configurations and their symmetry in domain average engineered structures

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Abstract
An algorithmic method is presented for determining all *domain configurations* and their symmetries in domain average engineered structures. This method is applied to PZN–PT single crystals by determining the domain configurations of domain states which arise in a phase transition between $\boldsymbol{G} = m\bar{3}m$ and $\boldsymbol{F} = 3m$.

In domain average engineered samples of ferroic crystals [1], the sample is subdivided into a very large number of domains representing m domain states S_i, $i = 1, \ldots, m$, where m is equal to or smaller than the theoretically allowed maximum number n. The tensorial properties of such crystals are taken to be averages over all domains of the sample:

$$T_{\mathrm{ave}} = \sum_{i=1}^{m} V_i T_i \tag{1}$$

where T_i is a tensor property of the ith domain state S_i and V_i is the fraction of the total volume of the sample made up of domain states S_i. A sample, defined by its constituent domain states S_i, $i = 1, \ldots, m$, and their corresponding fractional volumes V_i, $i = 1, \ldots, m$, will be referred to as a *domain configuration*. A specific domain configuration consisting of the domain states S_i, $i = 1, \ldots, m$, will be denoted by $[S_1, \ldots, S_a][S_b, \ldots, S_c] \cdots [S_k, \ldots, S_m]$ where all domain states in each subtuplet of domain states, within the same square brackets, have the same fractional volume. In this letter we give an algorithmic method for determining *all* domain configurations in a domain average engineered sample of domains that have arisen in a phase transition between a phase of higher point group symmetry $\boldsymbol{G}$ and one of lower point group symmetry $\boldsymbol{F}$.

A method was given in [2] for determining all domain configurations in the special case where the domain states were of equal fractional volume. This method was applied to PZN–PT by considering the phase transition between $\boldsymbol{G} = m\bar{3}m$ and $\boldsymbol{F} = 3m$. A highly mathematical analysis was given in [3] for determining the domain configurations in the general case of non-equal fractional volumes. This was also applied to the case of domain configurations of PZN–PT, but an exhaustive listing of all domain configurations was not explicitly given.

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Table 1. Representative subsets of domain states for $\boldsymbol{G} = m\bar{3}m$ and $\boldsymbol{F} = \bar{3}_{xyz}m_{\bar{x}y}$, and the subgroups $\boldsymbol{H}$ of $m\bar{3}m$ that leave them invariant.

Representative subset	Symmetry $\boldsymbol{H}$
{1}; {2345678}	$3_{xyz}m_{\bar{x}y}$
{13}; {245678}	$m_{xy}m_{\bar{x}y}2_z$
{15}; {234678}	$\bar{3}_{xyz}m_{\bar{x}y}$
{16}; {234578}	$m_x m_{\bar{y}z}2_{yz}$
{123}; {45678}	$3_{x\bar{y}z}m_{\bar{x}z}$
{135}; {24678}	$m_{\bar{x}y}$
{136}; {24578}	m_{xy}
{1234}	$\bar{4}_z 3_{xyz}m_{xy}$
{1235}	$m_{\bar{x}z}$
{1238}	$3_{x\bar{y}z}m_{\bar{x}z}$
{1356}	$2_{\bar{x}z}$
{1357}	$m_{xy}m_{\bar{x}y}m_z$
{1368}	$4_z m_x m_{xy}$

Table 2. Domain configurations and their symmetries derived from the subset of domain states {1368}.

Subgroup $K \subseteq 4_z m_x m_{xy}$	Domain configuration	Symmetry
(1) $4_z m_x m_{xy}$	[1368]	$4_z m_x m_{xy}$
(2) 4_z	[1368]	$4_z m_x m_{xy}$
(3) $m_{xy}m_{\bar{x}y}2_z$	[13][68]	$m_{xy}m_{\bar{x}y}2_z$
(4) $m_x m_y 2_z$	[1368]	$4_z m_x m_{xy}$
(5) m_{xy}	[6][8][13]	m_{xy}
(6) $m_{\bar{x}y}$	[1][3][68]	$m_{\bar{x}y}$
(7) m_x	[16][38]	m_x
(8) m_y	[18][26]	m_y
(9) 2_z	[13][68]	$m_{xy}m_{\bar{x}y}2_z$
(10) 1	[1][3][6][8]	1

Here we give an alternative method for determining all domain configuration in the general case based not on the highly mathematical concepts of [3] but on a generalization of the symmetry analysis of [2]. As we present this algorithmic method, we shall in parallel derive all domain configurations in the case of PZN–PT.

Consider the phase transition from $\boldsymbol{G}$ to $\boldsymbol{F}$. The symmetry analysis of the domain configurations is based on the coset decomposition of the point group $\boldsymbol{G}$ with respect to its subgroup:

$$\boldsymbol{G} = \boldsymbol{F} + g_2\boldsymbol{F} + g_3\boldsymbol{F} + \cdots + g_n\boldsymbol{F} \tag{2}$$

where the elements g_i are coset representatives of the decomposition and $g_1 = 1$. The relative orientations of the domain states are given by $S_i = g_i S_1$. The closure of the group $\boldsymbol{G}$ under multiplication implies a permutation of the cosets of the coset decomposition (2) and in turn a permutation of the domain states S_i under elements g of $\boldsymbol{G}$. The action of an element g of $\boldsymbol{G}$ on S_i is defined by $gS_i = gg_iS_1 = g_k f S_1 = g_k S_1 = S_k$, where f is an element of $\boldsymbol{F}$, and the domain state S_i is transformed by the element g into the domain state S_k. The action of an element g of $\boldsymbol{G}$ on a domain configuration is

$$\begin{aligned} g[S_1, \ldots, S_a][S_b, \ldots, S_c]\cdots[S_k, \ldots, S_m] \\ = [gS_1, \ldots, gS_a][gS_b, \ldots, gS_c]\cdots[gS_k, \ldots, gS_m]. \end{aligned} \tag{3}$$

Table 3. Representative domain configurations of domain states in the phase transition between $m\bar{3}m$ and $\bar{3}_{xyz}m_{\bar{x}y}$.

Domain configuration	Symmetry	Conditions on fractional volumes
[1]	$3_{xyz}m_{\bar{x}y}$	
[13]	$m_{xy}m_{\bar{x}y}2_z$	
[1][3]	$m_{\bar{x}y}$	$V_{[1]} \neq V_{[3]}$
[15]	$\bar{3}_{xyz}m_{\bar{x}y}$	
[1][5]	$3_{xyz}m_{\bar{x}y}$	$V_{[1]} \neq V_{[5]}$
[16]	$m_x m_{\bar{y}z}2_{yz}$	
[1][6]	$m_{\bar{y}z}$	$V_{[1]} \neq V_{[6]}$
[123]	$3_{x\bar{y}z}m_{\bar{x}z}$	
[1][23]	$m_{\bar{x}z}$	$V_{[1]} \neq V_{[23]}$
[1][2][3]	1	$V_{[2]} \neq V_{[3]}$
[1][3][5]	$m_{\bar{x}y}$	
[6][13]	m_{xy}	
[1][3][6]	1	$V_{[1]} \neq V_{[3]}$
[1234]	$\bar{4}_z3_{xyz}m_{xy}$	
[1][234]	$3_{xyz}m_{\bar{x}y}$	$V_{[1]} \neq V_{[234]}$
[12][34]	$m_{yz}m_{\bar{y}z}2_x$	$V_{[12]} \neq V_{[34]}$
[2][4][13]	m_{xy}	$V_{[2]} \neq V_{[4]}$ or $V_{[2]} \neq V_{[13]}$ or $V_{[4]} \neq V_{[13]}$
[1][2][3][4]	1	$\left\langle \begin{array}{c} V_{[1]} \neq V_{[3]} \\ V_{[1]} \neq V_{[2]} \text{ or } V_{[3]} \neq V_{[4]} \\ V_{[3]} \neq V_{[4]} \text{ or } V_{[2]} \neq V_{[4]} \text{ or } V_{[2]} \neq V_{[3]} \end{array} \right\rangle$
[1][5][23]	$m_{\bar{x}z}$	
[1][2][3][5]	1	$V_{[2]} \neq V_{[3]}$
[8][123]	$3_{x\bar{y}z}m_{\bar{x}z}$	
[2][8][13]	m_{xy}	$V_{[2]} \neq V_{[13]}$
[1][2][3][8]	1	$V_{[1]} \neq V_{[3]}$
[15][36]	$2_{\bar{x}z}$	
[1][3][5][6]	1	$V_{[1]} \neq V_{[5]}$ or $V_{[3]} \neq V_{[6]}$
[1357]	$m_{xy}m_{\bar{x}y}m_z$	
[17][35]	$m_{\bar{x}y}m_z2_{xy}$	$V_{[17]} \neq V_{[35]}$
[13][57]	$m_{xy}m_{\bar{x}y}2_z$	$V_{[13]} \neq V_{[57]}$
[15][37]	$2_{\bar{x}y}/m_{\bar{x}y}$	$V_{[15]} \neq V_{[37]}$
[1][3][5][7]	$m_{\bar{x}y}$	$\left\langle \begin{array}{c} V_{[1]} \neq V_{[5]} \text{ or } V_{[3]} \neq V_{[7]} \\ V_{[1]} \neq V_{[3]} \text{ or } V_{[5]} \neq V_{[7]} \\ V_{[1]} \neq V_{[7]} \text{ or } V_{[3]} \neq V_{[5]} \end{array} \right\rangle$
[1368]	$4_zm_xm_{xy}$	
[13][68]	$m_{xy}m_{\bar{x}y}2_z$	$V_{[13]} \neq V_{[68]}$
[6][8][13]	m_{xy}	$V_{[6]} \neq V_{[8]}$
[16][38]	m_x	$V_{[16]} \neq V_{[38]}$
[1][3][6][8]	1	$\left\langle \begin{array}{c} V_{[1]} \neq V_{[3]} \\ V_{[1]} \neq V_{[6]} \text{ or } V_{[3]} \neq V_{[8]} \end{array} \right\rangle$
[2][4][8][57]	m_{xy}	
[2][4][5][7][8]	1	$V_{[5]} \neq V_{[7]}$
[7][24][68]	$m_{\bar{x}y}$	
[2][4][6][7][8]	1	$V_{[2]} \neq V_{[4]}$ or $V_{[6]} \neq V_{[8]}$
[4][8][567]	$3_{x\bar{y}z}m_{\bar{x}z}$	
[4][6][8][57]	m_{xy}	$V_{[6]} \neq V_{[57]}$
[4][5][6][7][8]	1	$V_{[5]} \neq V_{[7]}$

Table 3. (Continued.)

Domain configuration	Symmetry	Conditions on fractional volumes
[25][3478]	$m_x m_{\bar{y}z} 2_{yz}$	
[25][38][47]	m_x	$V_{[38]} \neq V_{[47]}$
[25][37][48]	2_{yz}	$V_{[37]} \neq V_{[48]}$
[2][5][34][78]	$m_{\bar{y}z}$	$V_{[2]} \neq V_{[5]}$ or $V_{[34]} \neq V_{[78]}$
[2][3][4][5][7][8]	1	$\left\langle \begin{array}{c} V_{[3]} \neq V_{[4]} \text{ or } V_{[7]} \neq V_{[8]} \\ V_{[2]} \neq V_{[5]} \text{ or } V_{[3]} \neq V_{[7]} \text{ or } V_{[4]} \neq V_{[8]} \\ V_{[2]} \neq V_{[5]} \text{ or } V_{[3]} \neq V_{[8]} \text{ or } V_{[4]} \neq V_{[7]} \end{array} \right\rangle$
[234678]	$\bar{3}_{xyz} m_{\bar{x}y}$	
[234][678]	$3_{xyz} m_{\bar{x}y}$	$V_{[234]} \neq V_{[678]}$
[37][2468]	$2_{\bar{x}y}/m_{\bar{x}y}$	$V_{[37]} \neq V_{[2468]}$
[28][37][46]	$2_{\bar{x}y}$	$V_{[28]} \neq V_{[46]}$
[26][37][48]	$\bar{1}$	$V_{[26]} \neq V_{[48]}$
[3][7][24][68]	$m_{\bar{x}y}$	$\left\langle \begin{array}{c} V_{[3]} \neq V_{[7]} \text{ or } V_{[24]} \neq V_{[68]} \\ V_{[3]} \neq V_{[24]} \text{ or } V_{[7]} \neq V_{[68]} \end{array} \right\rangle$
[2][3][4][6][7][8]	1	$\left\langle \begin{array}{c} V_{[6]} \neq V_{[8]} \\ V_{[2]} \neq V_{[6]} \text{ or } V_{[3]} \neq V_{[7]} \text{ or } V_{[4]} \neq V_{[8]} \\ V_{[2]} \neq V_{[8]} \text{ or } V_{[3]} \neq V_{[7]} \text{ or } V_{[4]} \neq V_{[6]} \end{array} \right\rangle$
[24][57][68]	$m_{xy} m_{\bar{x}y} 2_z$	
[5][7][24][68]	$m_{\bar{x}y}$	$V_{[5]} \neq V_{[7]}$
[2][4][6][8][57]	m_{xy}	$V_{[2]} \neq V_{[4]}$ or $V_{[6]} \neq V_{[8]}$
[2][4][5][6][7][8]	1	$\left\langle \begin{array}{c} V_{[5]} \neq V_{[7]} \\ V_{[2]} \neq V_{[4]} \text{ or } V_{[6]} \neq V_{[8]} \end{array} \right\rangle$
[5][234][678]	$3_{xyz} m_{\bar{x}y}$	
[3][5][7][24][68]	$m_{\bar{x}y}$	$V_{[3]} \neq V_{[24]}$ or $V_{[7]} \neq V_{[68]}$
[2][3][4][5][6][7][8]	1	$V_{[2]} \neq V_{[4]}$ or $V_{[6]} \neq V_{[8]}$
[12345678]	$m\bar{3}m$	
[1234][5678]	$\bar{4}_z 3_{xyz} m_{xy}$	$V_{[1234]} \neq V_{[5678]}$
[15][234678]	$\bar{3}_{xyz} m_{\bar{x}y}$	$V_{[15]} \neq V_{[234678]}$
[1][5][234][678]	$3_{xyz} m_{\bar{x}y}$	$\left\langle \begin{array}{c} V_{[1]} \neq V_{[5]} \text{ or } V_{[234]} \neq V_{[678]} \\ V_{[1]} \neq V_{[234]} \text{ or } V_{[5]} \neq V_{[678]} \end{array} \right\rangle$
[1368][2457]	$4_z m_x m_{xy}$	$V_{[1368]} \neq V_{[2457]}$
[1357][2468]	$m_{xy} m_{\bar{x}y} m_z$	$V_{[1357]} \neq V_{[2468]}$
[28][46][1357]	$m_{xy} m_z 2_{\bar{x}y}$	$V_{[28]} \neq V_{[46]}$
[13][24][57][68]	$m_{xy} m_{\bar{x}y} 2_z$	$\left\langle \begin{array}{c} V_{[13]} \neq V_{[57]} \text{ or } V_{[24]} \neq V_{[68]} \\ V_{[13]} \neq V_{[68]} \text{ or } V_{[24]} \neq V_{[57]} \\ V_{[13]} \neq V_{[24]} \text{ or } V_{[57]} \neq V_{[68]} \end{array} \right\rangle$
[26][48][1357]	$2_{xy}/m_{xy}$	$V_{[26]} \neq V_{[48]}$
[17][28][35][46]	m_z	$V_{[17]} \neq V_{[35]}$ or $V_{[28]} \neq V_{[46]}$
[17][26][35][48]	2_{xy}	$V_{[17]} \neq V_{[35]}$
[15][26][37][48]	$\bar{1}$	$\left\langle \begin{array}{c} V_{[15]} \neq V_{[37]} \\ V_{[26]} \neq V_{[37]} \text{ or } V_{[26]} \neq V_{[48]} \text{ or } V_{[37]} \neq V_{[48]} \end{array} \right\rangle$
[2][4][6][8][13][57]	m_{xy}	$\left\langle \begin{array}{c} V_{[2]} \neq V_{[6]} \text{ or } V_{[4]} \neq V_{[8]} \text{ or } V_{[13]} \neq V_{[57]} \\ V_{[2]} \neq V_{[4]} \text{ or } V_{[6]} \neq V_{[8]} \\ V_{[2]} \neq V_{[8]} \text{ or } V_{[4]} \neq V_{[6]} \text{ or } V_{[13]} \neq V_{[57]} \end{array} \right\rangle$
[1][2][3][4][5][6][7][8]	1	$\left\langle \begin{array}{c} V_{[5]} \neq V_{[7]} \\ V_{[1]} \neq V_{[5]} \text{ or } V_{[2]} \neq V_{[6]} \text{ or } V_{[3]} \neq V_{[7]} \text{ or } V_{[4]} \neq V_{[8]} \\ V_{[1]} \neq V_{[7]} \text{ or } V_{[2]} \neq V_{[6]} \text{ or } V_{[3]} \neq V_{[5]} \text{ or } V_{[4]} \neq V_{[8]} \\ V_{[1]} \neq V_{[7]} \text{ or } V_{[2]} \neq V_{[8]} \text{ or } V_{[3]} \neq V_{[5]} \text{ or } V_{[4]} \neq V_{[6]} \\ \left(\begin{array}{c} V_{[2]} \neq V_{[3]} \text{ or } V_{[2]} \neq V_{[4]} \text{ or } V_{[3]} \neq V_{[4]} \text{ or } \\ V_{[6]} \neq V_{[7]} \text{ or } V_{[6]} \neq V_{[8]} \text{ or } V_{[7]} \neq V_{[8]} \end{array} \right) \end{array} \right\rangle$

For a given $\boldsymbol{G}$ and $\boldsymbol{F}$, all domain configurations are classified into classes of equivalent domain configurations: two domain configurations $[S_1, \ldots, S_a][S_b, \ldots, S_c] \cdots [S_k, \ldots, S_m]$ and $[S'_1, \ldots, S'_a][S'_b, \ldots, S'_c] \cdots [S'_k, \ldots, S'_m]$ are said to belong to the same class of equivalent domain configurations if there exists an element g of $\boldsymbol{G}$ such that

$$g[S_1, \ldots, S_a][S_b, \ldots, S_c] \cdots [S_k, \ldots, S_m] = [S'_1, \ldots, S'_a][S'_b, \ldots, S'_c] \cdots [S'_k, \ldots, S'_m]. \quad (4)$$

That is, the unprimed domain configuration is transformed by g into the primed domain configuration. In determining all domain configurations for a given $\boldsymbol{G}$ and $\boldsymbol{F}$ we shall explicitly list only one domain configuration from each class. The remaining domain configurations in each class are derived by applying all elements g of $\boldsymbol{G}$ to these representative domain configurations.

The subsets $\{S_1, \ldots, S_m\}$ of all domain states in the domain configurations can also be classified into classes of equivalent subsets of domain states: two subsets $\{S_1, \ldots, S_m\}$ and $\{S'_1, \ldots, S'_m\}$ are said to belong to the same class of equivalent subsets of domain states if there exists an element g of $\boldsymbol{G}$ such that

$$g\{S_1, \ldots, S_m\} = \{gS_1, \ldots, gS_m\} = \{S'_1, \ldots, S'_m\}. \quad (5)$$

The symmetry group $\boldsymbol{H}$ of a subset of domain states is defined as the subgroup of all elements g of $\boldsymbol{G}$ which leaves the subset invariant:

$$g\{S_1, \ldots, S_m\} = \{gS_1, \ldots, gS_m\} = \{S_1, \ldots, S_m\}. \quad (6)$$

This classification was used in [2] and is equivalent to the classification of all domain configurations of domain states with equal fractional volumes. A computer program is available [4] which calculates the classes of equivalent subsets of domains and their symmetries for any pair of point groups $\boldsymbol{G}$ and $\boldsymbol{F}$.

Given a point group $\boldsymbol{G}$ and subgroup $\boldsymbol{F}$, the first step in determining all domain configurations is to determine one representative subset of domain states from each class of equivalent subsets of domain states and its symmetry group $\boldsymbol{H}$. For $\boldsymbol{G} = m\bar{3}m$ and $\boldsymbol{F} = \bar{3}_{xyz}m_{\bar{x}y}$, the coset representatives $g_i, i = 1, \ldots, 8$, can be chosen as $1, 2_x, 2_z, 2_y, \bar{1}, m_x, m_z, m_y$, respectively. Representative subsets and their symmetries of the 20 classes of equivalent subsets of domain states are given in table 1. Each subset is denoted by listing the indices of the domain states contained in that subset.

Next, for each representative subset of domain states, we determine all domain configurations which contain these domain states. For each representative subset $\{S_1, \ldots, S_m\}$ of symmetry $\boldsymbol{H}$ we list all subgroups $\boldsymbol{K} \subseteq \boldsymbol{H}$. For each $\boldsymbol{K}$ we partition $\{S_1, \ldots, S_m\}$ into a set of subtuplets $\{S_1, \ldots, S_a\}\{S_b, \ldots, S_c\} \cdots \{S_k, \ldots, S_m\}$ where all domain states of each subtuplet can be obtained by acting on one domain state with all elements k of $\boldsymbol{K}$. (These subtuplets are known as $\boldsymbol{K}$-orbits of domain states in $\{S_1, \ldots, S_m\}$.) For each such partition we construct a domain configuration $[S_1, \ldots, S_a][S_b, \ldots, S_c] \cdots [S_k, \ldots, S_m]$ by taking the fractional volumes of all domain states in each subtuplet to be the same. We denote by $V_{[s \ldots v]}$ the fractional volume of *each* domain state in the subtuplet $[S_s, \ldots, S_v]$. We temporarily assume that the fractional volumes of the subtuplets are distinct, and determine the symmetry of the domain configuration. If the group $\boldsymbol{H}$ contains sets of conjugate subgroups $\boldsymbol{K}$, then one performs this construction only for one subgroup K of each conjugate set of subgroups. The use of additional subgroups in a set of conjugate subgroups will only give rise to equivalent domain configurations. One then lists all distinct domain configurations.

For example, for the representative subset $\{1368\}$ of symmetry $\boldsymbol{H} = 4_z m_x m_{xy}$ we list in table 2 the subgroups $\boldsymbol{K} \subseteq \boldsymbol{H}$, the domain configuration determined by the partitioning of the set of domain states with respect to $\boldsymbol{K}$, and the symmetry group of the domain configuration. The first, second, and fourth domain configurations so derived, as well as the third and ninth, are identical. Also, the fifth and sixth, and the seventh and eighth, use pairs of conjugate subgroups of $\boldsymbol{H} = 4_z m_x m_{xy}$. Consequently we have, derived from $\{1368\}$, five non-equivalent domain configurations where the fractional volumes of the subtuplets are distinct. All such non-equivalent domain configurations for $\boldsymbol{G} = m\bar{3}m$ and $\boldsymbol{F} = \bar{3}_{xyz} m_{\bar{x}y}$ and their symmetries are listed in the first two columns of table 3.

We now relax the condition that the fractional volumes of the domain states of each subtuplet in a domain configuration are distinct. All relationships among the fractional volumes are allowed under the criterion that the relationships do not lead to a domain configuration of a higher symmetry. In column three of table 3 we list conditions that the relationships must fulfil to satisfy this criterion. Some domain configurations have no conditions on the relationships among the fractional volumes. Others have optional conditions separated by the word *or*, and others multiple optional conditions given on separate lines. With these conditions table 3 is now a list of one representative domain configuration from each class of equivalent domain configurations. A complete listing of all domain configurations, in this example, is obtained by applying all elements of $\boldsymbol{G} = m\bar{3}m$ to each of the representative domain configurations listed in table 3.

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References

[1] Fousek J and Cross L E 2001 *Ferroelectrics* **252** 171–80
[2] Fousek J, Litvin D B and Cross L E 2001 *J. Phys.: Condens. Matter* **13** L1–6
[3] Fuksa J and Janovec V 2002 *J. Phys.: Condens. Matter* **14** 3795–812
[4] Shaparenko B, Schlessman J and Litvin D B 2002 *Ferroelectrics* **269** 9–14