MAGNETIC COMPLETELY TRANSPOSABLE TWIN LAWS AND TENSOR DISTINCTION

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ABSTRACT

Macroscopic tensorial physical properties which are different in two domains of a ferroic crystal provide a *tensor distinction* of the two domains. This tensor distinction is determined from a symmetry relationship, called a *twin law,* between the bulk structures, the *domain states*, of the two domains. The simplest type of twin law is the so called *completely transposable* twin law. We extend here the concept of completely transposable twin laws from non-magnetic to magnetic completely transposable twin laws. We establish the structure of and tabulate the 380 classes of magnetic completely transposable twin laws. The relationship between magnetic completely transposable twin laws and double antisymmetry groups is then given. Examples of the application of magnetic completely transposable twin laws are given in the tensor distinction of non-ferroelastic magnetoelectric domain pairs.

1. INTRODUCTION

We consider crystalline domains which arise in a phase transition from a high symmetry phase of symmetry **G** to a low symmetry phase of symmetry **F**. We shall refer to the bulk structures of these domains in polydomain samples as *domain states*. Several disconnected domains of possibly different shape can have the same domain state. Consequently, domain states of a polydomain sample represent structures that appear in the sample, irrespective of in which domain and irrespective of the domain's shape. We shall be interested in the tensor distinction of domains, distinguishing the domains by the values of components of macroscopic tensorial physical properties. This tensor distinction of domains is identical with the tensor distinction of the domains' corresponding domain states. Because the domains, due to their shape, are not necessarily symmetry related, while their corresponding domain state are, we consider, in what follows, the tensor distinction of domain states. As we shall consider macroscopic tensorial physical properties, we shall use the continuum description of the domain states and our symmetry analysis will be based on point group considerations only.

Two domain state S_i and S_j form a *domain pair* $\{S_i,S_j\}$ (Janovec, 1972). We shall call the *twin law* of the domain pair $\{S_i,S_j\}$ that symmetry information which specifies the point groups of the two domain states S_i and S_j and their relationship. This symmetry information is provided for by the point group $\mathbf{F}_i$, the point group of the domain state S_i, and the element g_{ij}, called the twinning operation, which transforms the domain state S_i into the domain state S_j, i.e. $g_{ij}S_i=S_j$. The point group of the domain state S_j is given by $\mathbf{F}_j = g_{ij}\mathbf{F}_i g_{ij}^{-1}$, and the relationship between the domain states is given by the twinning operation g_{ij}.

The twin law of a domain pair can alternatively be given by a group $\mathbf{J} = <\mathbf{F}_i,g_{ij}>$, the group generated by the group $\mathbf{F}_i$ and the twinning operation g_{ij}. In its simplest form the twin law is of the form

$$\mathbf{J} \equiv <\mathbf{F}_i, g_{ij}> = \mathbf{F}_i + g_{ij}\mathbf{F}_i \qquad\qquad (1)$$

where **J** consists of only two cosets of the point group $\mathbf{F}_i$. In this case the twin law is refered to as a *completely transposable* twin law (Janovec, Litvin, and Richterova, 1994). This is refered to as a *transposable* twin law (This was previously refered to as an *ambivalent* twin law (Janovec, 1981)) as the twinning operation g_{ij} not only transforms the domain state S_i into S_j but in addition transforms the

domain state S_j into S_i, i.e.

$$S_j = g_{ij}S_i \text{ and } S_i = g_{ij}S_j.$$

This is refered to as a *completely* transposable twin law because in addition to being transposable, the point groups of the two domain states S_i and S_j are identical, that is:

$$F_j \equiv g_{ij}F_ig_{ij}^{-1} = F_i$$

A ferroic phase is non-ferroelastic if all the domains have the same (zero) spontaneous deformation (Aizu,1973). In a non-ferroelastic phase there are $n = |G|/|F|$ domain states where $|G|$ and $|F|$ denote the order of the groups G and F, respectfully, which are all related by the coset representatives of the coset decomposition of G with respect to F. The twin laws of non-ferroelastic domain pairs are of the form of Equation (1), i.e. are completely transposable twin laws (Janovec, Richterova, and Litvin, 1993).

In a ferroelastic phase, the orientations of the domain states are controlled by disorientations, i.e. rotations of single domain states (domain states invariant under F or any conjugate subgroup of F in G) needed to achieve a coherent interface of two ferroelastic domain states along a planar wall. Consequently, the number of domain states in a ferroelastic phase is, in general, greater than $n = |G|/|F|$ and the twin law of a ferroelastic domain pair is, in general, not a completely transposable twin law (Janovec, Litvin, and Richterova, 1994).

The tensor distinction of two domain states of a domain pair can be determined from the domain pair's twin law. The form T_i of a tensor T in the domain state S_i is determined by the point group F_i and the form T_j of the tensor T in the domain state S_j can be determined by transforming T_i by g_{ij}:

$$T_j = g_{ij}T_i \tag{2}$$

By comparing the forms T_i and T_j related by Equation (2), one determines the components of the tensor T which are distinct in the two domain states, i.e. one determines the tensor distinction of the domain pair $\{S_i,S_j\}$.

Non-magnetic completely transposable twin laws, Equation (1), can be written as

$$J = F + g^*F$$

where J is a non-magnetic point group (i.e. a point group belonging to one of the thirty-two classes of

crystallographic point groups), and the element "g" has been stared to denote and emphasize that this is an element of **J** which transposes the two domain states. The non-magnetic completely transposable twin law is uniquely characterized by the point group **J** and a subgroup **F** of index two of **J**. Consequently, the mathematical structure of non-magnetic completely transposable twin laws is the same as that of dichromatic (black and white, antisymmetry) point groups (Heesch, 1930; Shubnikov, 1951). Completely transposable non-magnetic twin laws have been used in determining the macroscopic tensorial physical properties which distinguish domains of a domain pair in the cases of non-ferroelastic and ferroelectric non-ferroelastic domains (Janovec, Richterova, and Litvin, 1992,1993).

In Section 2 we extend the concept of completely transposable twin laws from that of non-magnetic completely transposable twin laws to magnetic completely transposable twin laws, i.e. to the case where in Equation (1) **J** is a magnetic point group. The structure of magnetic completely transposable twin laws is then considered, two types of notation are introduced and all classes of magnetic completley transposable twin laws are then derived. In Section 3 we discuss the application of magnetic completely transposable twin laws in determining the tensor distinction of non-ferroelastic magnetoelectric domain pairs. It is shown in Appendix 1 that the mathematical structure of magnetic completely transposable twin laws is the same as that of the so called *double antisymmetry groups* introduced by Zamorzaev and Sokolov (1957), see also Zamorzaev (1976) and Zamorzaev and Palistrant (1980). A third type of notation is introduced there for magnetic transposable twin laws based on the notation used for double antisymmetry groups.

In the remainder of this paper, for typographical and linguistic simplicity, we shall refer to "completely transposable twin laws" simply as "twin laws."

2. MAGNETIC TWIN LAWS

Let **J** denote a magnetic point group, i.e. a point group belonging to one of the 122 classes of crystallographic magnetic point groups (Opechowski, 1986). Let **F** denote a subgroup of index two of **J**. A magnetic twin law, **J** = **F** + g***F**, with a magnetic point group **J**, is uniquely characterized and can be denoted, in a double group notation, by **J**[**F**]. A second single group notation for a magnetic twin law

can be had by using the Hermann-Mauguin (International) notation for the magnetic group **J**. Individual symbols in the group symbol of **J** representing elements of **J** not contained in **F** are starred. For example, the magnetic twin laws $2_z/m_z' = 2_z + m_z'* 2_z$ and $4_z'm_x'm_{xy} = m_x'm_y'2_z + 4_z'* m_x'm_y'2_z$ are denoted in the double group notation **J[F]** respectively as $2_z/m_z'[2_z]$ and $4_z'm_x'm_{xy}[m_x'm_y'2_z]$ and in the single group notation, respectfully as $2_z/m_z'*$ and $4_z'*m_x'm_{xy}*$.

The equivalence of two magnetic twin laws is defined as follows: Two magnetic twin laws $J_1[F_1]$ and $J_2[F_2]$ are said to be equivalent and belong to the same class of magnetic twin laws if there exists a Euclidian transformation that simultaneously transforms J_1 into J_2 and F_1 into F_2.

To derive the number of classes of magnetic twin laws **J[F]** we first introduce a notation for non-magnetic point groups and the more detailed notation for the magnetic point groups: We denote a non-magnetic point group by **Q**. There are three types of magnetic point groups:

1) **J = Q** . There are 32 such classes. These are the 32 classes of crystallographic non-magnetic point groups.

2) **J = Q1'** . There are 32 such classes. These are direct products of a non-magnetic point group **Q** and the group **1'** consisting of the identity 1 and time inversion 1'.

3) **J = H + a'H** . There are 58 such classes. Magnetic groups of this type are also denoted by **Q(H)** where

$$Q = H + aH .$$

All magnetic groups **J = Q, J = Q1'**, and **J = Q(H)** are said to belong to the same *family* of the class of the non-magnetic point group **Q**. Consequently, all 122 classes of magnetic point groups can be categorized according to their family into 32 families.

In deriving the magnetic twin laws **J[F]** it is advantageous to subdivide the derivation according

to the type of magnetic point group $\mathbf{J}$:

1) $\mathbf{J} = \mathbf{Q}$. $\mathbf{F}$ must then be a subgroup $\mathbf{H}$ of index 2 of the non-magnetic point group $\mathbf{Q}$. The twin law is then of the type $\mathbf{J[F]} = \mathbf{Q[H]}$, i.e. a non-magnetic twin law which in the format of Equation (1) is written as $\mathbf{Q} = \mathbf{H} + a*\mathbf{H}$. The number of classes of magnetic twin law $\mathbf{Q[H]}$ is the same as the number of classes of magnetic point groups $\mathbf{Q(H)}$, that is, 58. For example, for $\mathbf{Q} = 2_x2_y2_z$ and $\mathbf{H} = 2_z$ we have the twin law $2_x2_y2_z = 2_z + 2_x*2_z$ which is denoted by $2_x2_y2_z [2_z]$ or $2_x*2_y*2_z$.

2) $\mathbf{J} = \mathbf{Q1'}$. There are three possibilities for $\mathbf{F}$:

i) If $\mathbf{F} = \mathbf{Q}$ then the twin law is $\mathbf{J[F]} = \mathbf{Q1'[Q]}$ and $\mathbf{J} = \mathbf{Q} + 1'*\mathbf{Q} = \mathbf{Q1'*}$. The number of classes of magnetic twin law $\mathbf{Q1'[Q]}$ is the same as the number of classes of magnetic point groups $\mathbf{Q1'}$, that is, 32. For example, for $\mathbf{Q} = 2_x2_y2_z$ we have the twin law $2_x2_y2_z1' = 2_x2_y2_z + 1'* 2_x2_y2_z$ which is denoted by $2_x2_y2_z1'[2_x2_y2_z]$ or $2_x2_y2_z1'*$.

ii) If $\mathbf{F} = \mathbf{H1'}$, where $\mathbf{H}$ is a subgroup of index 2 of $\mathbf{Q}$, then the twin law is $\mathbf{J[F]} = \mathbf{Q1'[H1']}$. Since

$$\mathbf{J} = \mathbf{H1'} + a*\mathbf{H1'} = (\mathbf{H} + a*\mathbf{H})1'$$

and $\mathbf{H} + a*\mathbf{H}$ is a non-magnetic twin law $\mathbf{Q[H]}$, the above type of magnetic twin law can be denoted by $\mathbf{Q[H]1'}$. The number of classes of such magnetic twin laws is the same as the number of classes of twin laws $\mathbf{Q[H]}$, that is, 58. For example, for $\mathbf{Q} = 2_x2_y2_z$ and $\mathbf{H} = 2_z$ we have the twin law $2_x2_y2_z1' = 2_z1' + 2_x*2_z1'$ which is denoted by $2_x2_y2_z1'[2_z1']$ or $2_x*2_y*2_z1'$.

iii) If $\mathbf{F} = \mathbf{Q(H)}$ then the magnetic twin law is $\mathbf{J[F]} = \mathbf{Q1'[Q(H)]}$ and :

$$\mathbf{J} = \mathbf{Q(H)} + 1'*\mathbf{Q(H)} = \mathbf{Q(H)1'*}$$

The number of classes of such magnetic twin laws is the same as the number of classes of magnetic point groups $Q(H)$, that is, 58. For example, for $Q = 2_x2_y2_z$ and $H = 2_z$ we have the twin law $2_x2_y2_z1' = 2_x'2_y'2_z + 1'^* 2_x'2_y'2_z$ which is denoted by $2_x2_y2_z1'[2_x'2_y'2_z]$ or $2_x'2_y'2_z1'^*$.

        3) $J = Q(H)$. There are two possibilities for F:

        i) $F = H$. The magnetic twin law is $Q(H)[H]$ and:

$$J = H + a'^*H$$

The number of classes of such magnetic twin laws is the same as the number of classes of magnetic point groups $Q(H)$, that is, 58. For example, for $Q = 2_x2_y2_z$ and $H = 2_z$ we have the twin law $2_x'2_y'2_z = 2_z + 2_x'^*2_z$ which is denoted by $2_x'2_y'2_z[2_z]$ or $2_x'^*2_y'^*2_z$.

        ii) $F \neq H$. The magnetic twin law is $Q(H)[K(R)]$ where $K(R)$ is a magnetic subgroup of index 2 of $Q(H)$ and:

$$J = R + a_1'R + a_2^*R + a_3'^*R$$

$K = R + a_1R$ and $H = R + a_2R$ are subgroups of index 2 of Q. R is a subgroup of index 2 of both H and K and a subgroup of index 4 of Q. There are 116 classes of magnetic twin laws $Q(H)[K(R)]$. For example, for $Q = 2_x2_y2_z$, $H = 2_z$, $K = 2_x$, and $Q = 1$, we have the twin law $2_x'2_y'2_z = 2_x' + 2_z'^*2_x'$ ($= 1 + 2_x'1 + 2_z^*1 + 2_y'^*1$) which is denoted by $2_x'2_y'2_z[2_x']$ or $2_x'2_y'^*2_z^*$.

    There are then six types of magnetic twin laws $J[F]$

1) $Q[H]$	2) $Q1'[Q] = Q1'^*$
3) $Q1'[H1'] = Q[H]1'$	4) $Q1'[Q(H)] = Q(H)1'^*$ (4)
5) $Q(H)[H]$	6) $Q(H)[K(R)]$

and a total of 380 classes of magnetic twin laws[*]. The magnetic twin laws can be classified into 32 families according to the family of the magnetic group J. A representative magnetic twin law of each class of magnetic twin laws belonging to the family of $Q = 222$ is given in Table 1. The numbers assigned to each magnetic twin law consists first of the serial number given to the magnetic twin law's family followed by a decimal point and the serial number of the class of the magnetic twin laws in that family. This is followed, in parentheses, by a number denoting the type of the magnetic twin law, see Equation (4). In the first column is the symbol for the magnetic twin law in double group notation followed in the second column with the corresponding single group notation. In the third column is a third notation based on the notation for the six types of twin laws given in Equation (4) and the symbols which denote double antisymmetry groups. This third notation and the relationship between the magnetic twin laws derived in this section and the double antisymmetry groups is given in Appendix I.

3. TENSOR DISTINCTION

We consider the tensor distinction of non-ferroelastic magnetoelectric domain pair (Litvin, Janovec, & Litvin, 1994). In such domain pairs, the domains states S_i and S_j have the same (zero) spontaneous deformation and their magnetic twin laws $J = F + g*F$ are completely transposable. We denote a physical property tensor by T, and the form of this tensor in the two domain states S_i and S_j, respectively, by T_i and T_j. The components of the tensor T_i will be denoted by $T_i^{\alpha\beta\gamma\cdots}$.

The types of physical property tensors considered are denoted by symbols for their transformational properties. Each symbol consists of the symbol V^n, which denotes the n-th product of a polar vector tensor, possibly preceded by "a" and/or "e", symbols which denote rank zero tensors that change sign, respectively, under time inversion $1'$ and spatial inversion $\overline{1}$. The physical meaning of many types of these tensors are given by Sirotin and Shaskolskaya (1975). We list eight tensor types in the first column of Table 2.

We consider a domain pair related by the magnetic twin law $J[F] = 4_z 2_x' 2_{xy}'[4_z]$, i.e. by the twin law $4_z 2_x' 2_{xy}' = 4_z + 2_x'*4_z$. In this case the point group of the domain S_i is $F = 4_z$ and the domain S_j is

related to the domain S_i by the element $2_x'$, i.e. $S_j = 2_x'S_i$. Consequently, the form of the tensor T_i is invariant under the point group $\mathbf{4_z}$ and the form of the tensor T_j is related to the form of the tensor T_i by the element $2_x'$, i.e. $T_j = 2_x'T_i$. The form of non-magnetic physical property tensors invariant under non-magnetic point groups, for a wide variety of tensor types can be found in Sirotin and Shaskolskaya (1975). These same tables can be also be used to determine the form of magnetic physical property tensor invariant under magnetic point groups (Litvin, 1994). For the magnetoelectric effect tensor, a tensor of the type aeV^2, the form of the tensor T_i, invariant under $\mathbf{4_z}$ is:

$$T_i^{\alpha\beta} = \begin{array}{ccc} A & C & 0 \\ -C & A & 0 \\ 0 & 0 & B \end{array}$$

The form of the tensor $T_j = 2_x'T_i$ is calculated via the standard transformation of a second rank tensor

$$T_j^{\alpha\beta} = - D^{1}(2_x)_{\alpha\alpha'}D^{1}(2_x)_{\beta\beta'}T_i^{\alpha'\beta'}$$

where the additional minus sign is present since the tensor type is aeV^2, which changes sign under time inversion, and $g^* = 2_x'$ is a primed element. The form of the tensor T_j is then:

$$T_j^{\alpha\beta} = \begin{array}{ccc} -A & C & 0 \\ -C & -A & 0 \\ 0 & 0 & -B \end{array}$$

The two domains can be distinguished by the T^{xx}, T^{yy}, and T^{zz} components of the magnetoelectric effect tensor.

In the second and third column of Table 2, we give the component forms of the tensors T_i and T_j for the tensor types listed in the first column. By comparing the forms of the tensors in the two domains of the domain pair related by this magnetic twin law we have that five tensor types distinguish between the two domains and three do not.

This domain pair is magnetoelectric as the form of the magnetoelectric effect tensor (a tensor of type aeV^2) is different in the two domains, and non-ferroelastic as the ferroelasticity effect tensor (of the type $[V^2]$) is identical in the two domains. From Table 2 we have that the domains of this domain pair can also be distinguished by spontaneous polarization (V), piezoelectric and second non-linear magnetoelectric tensors ($V[V^2]$), piezomagnetic and first non-linear magnetoelectric tensors ($aeV[V^2]$), and non linear electric tensors ($[V^3]$) (See Schmid (1975) for a complete discussion of these magnetoelectric phenomena). The domains can not be distinguished by spontaneous magnetization

(aeV), their electric and magnetic susceptibility ($[V^2]$), nor by their non-linear magnetic susceptibility ($ae[V^3]$).

[*] The complete tables have been deposited with the British Library Lending Division as Supplementary Publication No. SUP-----(--pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

APPENDIX 1. DOUBLE ANTISYMMETRY GROUPS

The magnetic twin laws, derived in Section 2, have the same mathematical structure as the double antisymmetry point groups introduced by Zamorzaev and Sokolov (1957). Double antisymmetry point groups can be defined as follows: All points of a finite object are assigned two signs, each of which can be a positive or negative sign. In addition to the point group transformations of the unsigned object, one defines transformations of the signs, a transformation 1' which reverses the value of the first sign, and 1* which reserves the value of the second sign. (In Zamorzaev and Sokolov (1957), the star is placed to the left of the symbol, i.e. *1.) A double antisymmetry group is an invariance group of such a signed finite object, the group of those point group transformations and point group transformations coupled with 1', 1*, or 1'* which leave the signed finite object invariant. Of the twelve types of double antisymmetry point groups, six correspond to the six types of magnetic twin laws. Listed in an order corresponding to the types of magnetic twin laws listed in Equation (4), these are:

1) **Q{H}**   2) **Q1'***

3) **Q{H}1'**   4) **Q(H)1'***   (A-1)

5) **Q(H){H}**   6) **Q(H){K} = Q(H){K(R)}**

Q denotes a point group and **H** and **K** subgroups of index 2 of **Q**. **R** is a subgroup of index 2 of both **H** and **K** and a subgroup of index 4 of **Q**.

Q{H} denotes a group where the elements of **Q** not contained in the subgroup **H** are coupled with 1*. **Q(H)** denotes a group where the elements of **Q** not contained in the subgroup **H** are coupled with 1'. **Q(H){K}** denotes a group where the elements of **Q** not contained in **H** are coupled with 1' and the elements of **Q** not contained in **K** are coupled with 1*. In groups **Q(H){H}**, the elements of **Q** not contained in **H** are coupled with 1'*. In groups **Q(H){K}**, with **H** ≠ **K**, there are elements of **Q** coupled with 1', coupled with 1*, and coupled with 1'*. Those elements of **Q** not coupled with any of these constitute a subgroup **R** which is a subgroup of index 2 of both **H** and **K**, and a subgroup of index 4 of **Q**. Elements of **K** which are not in **R** are coupled with 1' and consequently double antisymmetry point groups of this type can be denoted by **Q(H){K(R)}**. The mathematical equivalence of the magnetic twin laws given in Equation (4) with the double antisymmetry groups listed in Equation (A-1) can now be easily seen: One can interexchange the corresponding types of magnetic twin laws and double antisymmetry groups by interexchanging the square brackets [] with the curly brackets {}. In the third column of symbols in Table 1 we give the double antisymmetry group symbol, Equation (A-1), of each of the listed magnetic twin laws.

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Table 1: Representative magnetic twin laws of classes belonging to the family of $\mathbf{Q} = \mathbf{222}$. The type of the magnetic twin law is given, in parentheses, following the magnetic twin law's serial number. Three symbols are given, the double group symbol $\mathbf{J}[\mathbf{F}]$, the single group symbol, and the corresponding double antisymmetry group symbol.

Table 2: For the magnetic twin law $\mathbf{J}[\mathbf{F}] = \mathbf{4}_z\mathbf{2}_x{}'\mathbf{2}_{xy}{}'[\mathbf{4}_z]$ and the eight types of tensors listed in the first column, the forms of the tensors T_i and $T_j = 2_x{}'T_i$ are given in the second and third column, respectively. The tensor notation used is that of Sirotin and Shaskolskaya (1975).

TABLE 1

6) **222**

6.1(1)	$2_x2_y2_z[2_z]$	$2_x*2_y*2_z$	$2_x2_y2_z\{2_z\}$
6.2(2)	$2_x2_y2_z1'[2_x2_y2_z]$	$2_x2_y2_z1'*$	$2_x2_y2_z1'*$
6.3(3)	$2_x2_y2_z1'[2_z1']$	$2_x*2_y*2_z1'$	$2_x2_y2_z\{2_z\}1'$
6.4(4)	$2_x2_y2_z1'[2_x'2_y'2_z]$	$2_x'2_y'2_z1'*$	$2_x2_y2_z(2_z)1'*$
6.5(5)	$2_x'2_y'2_z[2_z]$	$2_x'*2_y'*2_z$	$2_x2_y2_z(2_z)\{2_z\}$
6.6(6)	$2_x'2_y'2_z[2_x']$	$2_x'2_y'*2_z*$	$2_x2_y2_z(2_z)\{2_x(1)\}$

TABLE 2

Tensor Type	Domain State S_i T_i	Domain State S_j T_j
V	0 0 A	0 0 -A
aeV	0 0 A	0 0 A
$[V^2]$	A 0 0 0 A 0 0 0 B	A 0 0 0 A 0 0 0 B
aeV^2	A C 0 -C A 0 0 0 B	-A C 0 -C -A 0 0 0 -B
$V[V^2]$	0 0 0 B A 0 0 0 0 A -B 0 C C D 0 0 0	0 0 0 B -A 0 0 0 0 -A -B 0 -C -C -D 0 0 0
$aeV[V^2]$	0 0 0 B A 0 0 0 0 A -B 0 C C D 0 0 0	0 0 0 -B A 0 0 0 0 A B 0 C C D 0 0 0
$[V^3]$	0 0 0 0 0 0 0 A A B	0 0 0 0 0 0 0 -A -A -B
$ae[V^3]$	0 0 0 0 0 0 0 A A B	0 0 0 0 0 0 0 A A B