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A Tensor Classification of Twinning in Crystals

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Abstract

A classification scheme for twinning in crystals is proposed. It is based on a tensor distinction of properties across the twin interface. The classification employs concepts from the theory of transformation twinning. The adequacy of such a scheme for other main types of twins, namely growth twins and nonferroelastic mechanical twins, is examined, and found to be satisfactory. All twins can be divided into four fundamentally distinct categories: ferroelastic or *S*-twins, nonferroelastic–ferroic or *N*-twins, Bollmann or *B*-twins and translation or *T*-twins. A comparison is made of the attributes of these four types of twins. A compact and informative twin symbol is introduced, which carries information about the tensor properties in which the twins differ. Information about any underlying total or partial coincidence sublattice is incorporated by attaching a subscript *t* or *p* to the main twin symbol. Several examples are discussed. This paper also introduces a rigorous definition of prototype symmetry in terms of the 'nondisruption condition'.

1. Introduction

In terms of the origin of twinning, twinned crystals can be of three main types: transformation twins, mechanical twins and growth twins. The two principal schemes for classification of twinning are those due to Friedel (1926) and Donnay & Donnay (1974).

In Friedel's classification (see Cahn, 1954), one first looks for a sublattice (if any) that is common to the two crystals comprising the twin. Such a lattice is called the coincidence lattice. The inverse of the fraction of lattice sites common to the two components is called the twin index, Σ. There are two primary categories of twins in this scheme: Those with $\Sigma = 1$ and those with $\Sigma > 1$. Each of these categories is further divided in terms of twin obliquity ω, which is a measure of disorientation of one component with respect to the other; either $\omega = 0$ or $\omega \neq 0$. Thus, the four types of twinning in this scheme are: twinning by merohedry ($\Sigma = 1$, $\omega = 0$); twinning by pseudomerohedry ($\Sigma = 1$, $\omega \neq 0$); twinning by reticular merohedry ($\Sigma > 1$, $\omega = 0$); and twinning by reticular pseudomerohedry ($\Sigma > 1$, $\omega \neq 0$).

Donnay & Donnay's (1974) classification scheme lays emphasis on the characteristics of the diffraction pattern of the twin. The most obvious feature to look for in the diffraction pattern is whether or not there is a 'splitting' of diffraction spots. If $\omega \neq 0$, such a splitting would be present, and one then speaks of twinning by twin-lattice quasi-symmetry (TLQS). If $\omega = 0$, the twinning is by twin-lattice symmetry (TLS). For each of these primary divisions, further subdivision is made in terms of the twin index Σ. Thus, compared to the Friedel scheme, the relative importance of Σ and ω is interchanged in this scheme.

These classification schemes, though significant and useful, do not pack a large amount of crystallophysical information, and are therefore not very discriminative. For example, both Dauphiné and Brazil twins of quartz come under the TLS category of Donnay & Donnay (1974), in spite of the fact that the response of these two types of twins to mechanical stress is very different. Dauphiné twins are transformation twins and they can be readily detwinned (see, for example, Wadhawan, 1982). Brazil twins, on the other hand, are growth twins and it is practically impossible to detwin a Brazil twin because of the nature of bonding frozen into the structure at the nucleation and/or growth stage. It would be desirable to place the Dauphiné and Brazil twins of quartz into separate categories.

Twinning in NH_4Cl is another example of the fact that the existing classification schemes are not sufficiently discriminative. The twin individuals of this crystal differ in the sign of the piezoelectric coefficient d_{123} (Newnham & Cross 1974). However, this information is ignored in the descriptions 'twinning by merohedry' (Friedel) and 'twinning by TLS' (Donnay & Donnay) for this case.

To absorb such additional information, one must invoke the space-group symmetry of the crystal. The details of twinning are determined by the crystal structure and the crystal structure is properly described by the space group to which a crystal belongs. A classification scheme for twinning based on space-group considerations was formulated earlier by the present author (Wadhawan, 1987) [referred to hereafter as VKW1]. The primary subdivision of twinning in this scheme was done in terms of Aizu's (1970) concept of prototype symmetry (see §2 for some definitions). All twins were first divided into two categories, namely Aizu twins and Bollmann twins, depending on whether or not the twin

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mapping operator is an operator of the prototype space group. Further subcategories of Aizu twins were defined in terms of the nature of the reduction of the symmetry of the prototype space group. Similarly, Bollmann twins were further categorized in terms of the coincidence sublattice. This classification scheme, being based on space-group considerations, is adequately informative and discriminative. However, it is still not a perfect scheme because it does not always ensure uniqueness of categorization: it is sometimes possible to assign the same twin to two different categories in this scheme. VKW1 tried to overcome this problem by stipulating that the Aizu category be assigned a higher priority over the Bollmann category. This is not an entirely satisfactory arrangement. To overcome this deficiency, we formulate a different classification scheme in this paper, using a different primary-subdivision criterion.

The tensor-classification scheme, as the new scheme formulated here is called, draws on a number of concepts from the theory of ferroic and nonferroic phase transformations in crystals. Therefore, we first introduce some definitions in §2 before describing the modified classification in §3. In §4, an informative and compact twin symbol is introduced, which is similar in concept to the symbol defined in VKW1. Finally, in §5, we discuss additional details of the present formalism and advance further arguments in support of its generality and completeness.

2. Some relevant concepts and definitions

Description of transformation twinning involves a comparison of a 'new' crystal structure and an 'old' crystal structure. The two crystal structures have different space-group symmetries and there is necessarily a loss or gain of symmetry operators in going from one structure to the other. In this context, statements regarding lost or gained symmetry operators have meaning only if the so-called nondisruption condition is satisfied (Guymont, 1981; Hahn & Wondratschek, 1994). Satisfaction of this condition means that the new structure arising from a phase transition can be described (*i.e.* its symmetry elements, Wyckoff positions of atoms, atomic parameters *etc.* can be located) in the frame of reference of the old structure after making (if necessary) continuous distortions (affine transformations) that do not themselves entail any additional changes of symmetry. Correlation of the symmetries of the two structures is a necessary, but not always a sufficient, condition for deriving one crystal structure from the other. The nondisruption condition provides a sufficiency criterion.

The notion of prototype symmetry was introduced by Aizu (1970, 1978). He defined the possible ferroic phases of a crystal as 'slight distortions' of one another and the prototype as the highest-symmetry structure from which the concerned ferroic phases can be derived by a slight distortion. The phrase 'slight distortion' was not quantified or defined rigorously. We introduce here a new definition of prototype symmetry that is intended to be more rigorous and precise:

> The prototype symmetry for one or more phase transitions in a crystal is the highest space-group symmetry attainable by, or conceivable for, that crystal by a distortion (affine mapping) of the crystal structure that does not violate the nondisruption condition.

Full justification and implications of such a definition will be discussed in a parallel publication.

Let G and H denote the space-group symmetries of the old and the new phases involved in a nonisomorphic phase transition. For determining the domain structure of the new phase, it is not necessary that H be a subgroup of G. It is sufficient that the nondisruption condition is satisfied (Guymont, 1981). However, for carrying out a group-theoretical analysis of the phase transition $G \rightarrow H$, it is necessary that either G corresponds to the prototype symmetry or we replace G by a suitably chosen prototype symmetry group. [Alternative approaches involve working with a group, I, which is the intersection group of G and H.]

Assuming that G denotes prototype symmetry, the possible number n of the types of single-domain states is given by (Aizu, 1970; Janovec, 1972):

$$n = (|G_p| : |H_p|)(Z_H : Z_G). \quad (1)$$

Here $|G_p|$ and $|H_p|$ are orders of the point groups G_p and H_p underlying the space groups G and H. The ratio $Z_H : Z_G$ is the number of times the primitive unit cell of the distorted phase is larger than that of the prototype.

The single-domain states can thus be numbered as $D_1, D_2, \ldots, D_n$, with n given by (1). If any pair in this set is a ferroelastic domain pair, (1) is valid only under the so-called parent-clamping approximation (PCA) (Zikmund, 1984; Janovec, Schranz, Warhanec & Zikmund, 1989).

All the domains have the same symmetry, described by the space group H. However, since they can differ in orientation, chirality and/or relative location, the positions and orientations of the crystallographic symmetry elements in each domain can be different. Let us choose a common origin (fixed in the prototype) and denote the symmetry groups of $D_1, D_2, \ldots, D_n$ by $H_1, H_2, \ldots, H_n$.

Symmetry operators of H_1 transform domain D_1 back onto itself. Operators g_j belonging to G, but not to H_1, will transform D_1 to other domain states (under the PCA, if needed). It is readily verified that all operators of G that transform D_1 to a particular domain state D_j are given by the left coset g_jH_1 (Aizu, 1970; Janovec, 1972):

$$D_j = (g_jH_1)D_1, \quad j = 1, 2, \ldots, n. \quad (2)$$

The choice of the domain-mapping operators g_j is not unique; however, there are exactly n such distinct (or representative) operators.

Since H_1 is a proper subgroup of G, the following coset decomposition can be written:

$$G = H_1 + g_2H_1 + \ldots + g_jH_1 + \ldots + g_nH_1. \qquad (3)$$

There is a one-to-one correspondence between the domain states $D_1, D_2, \ldots, D_n$ and the n left cosets in (3).

3. Tensor classification of twinning

Since we want to make a tensor distinction of properties across the twin boundary, we begin by dividing all twins into two main categories: those for which at least one tensor coefficient is different and those for which all macroscopic tensor coefficients are the same (Fig. 1). We call the latter category of twins translation twins or *T*-twins.

Next we note that twins that differ in at least one macroscopic tensor property can be of two types: those for which a prototype symmetry is definable (we call them Aizu twins) and those for which the prototype is not definable (we call them Bollmann twins, or *B*-twins) (Bollmann, 1970).

For Aizu twins, within the PCA, all the twin mapping operators are from among the n domain mapping operators g_j defined by (3). Since we have already recognized *T*-twins as a separate category, all Aizu twins are necessarily ferroic twins, *i.e.* for them the mapping operator necessarily has a rotational component; it may also sometimes have a fractional translation component arising out of a screw-axis or glide-plane operation (Guymont, Gratias, Portier & Fayard, 1976).

Aizu twins neatly divide themselves into two inherently distinct subclasses: ferroelastic or Seignette-elastic twins (or *S*-twins), and nonferroelastic–ferroic twins (or *N*-twins). An *S*-twin can be considered as arising from a ferroelastic phase transition, *i.e.* a real or hypothetical phase transition involving a spontaneous distortion of the prototype lattice. [In *S*-twins, *S* stands for 'Seignette-elastic' or for 'spontaneous distortion'. The words 'ferroic' and 'ferroelastic' both begin with the letter *F*, so we use the letter *S* for ferroelastic twins to avoid confusion with the letter *F* used for 'ferroic' by Aizu (1970) for defining his symbol for ferroic species].

Proper ferroelastic transitions are well described by mean-field theories like the Landau theory (Salje, 1990). The same cannot be said of nonferroelastic–ferroic tran-

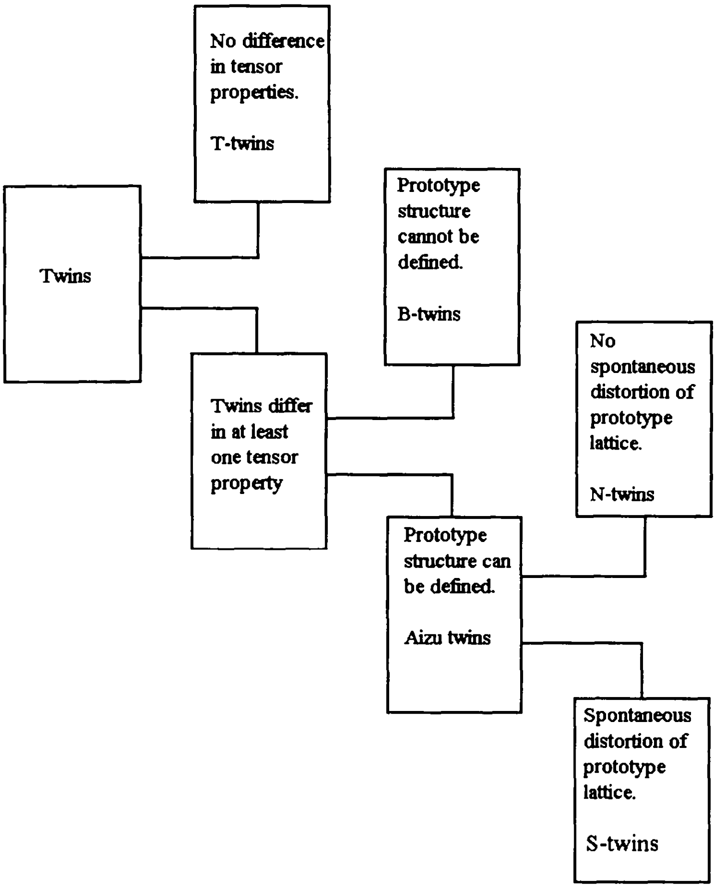

Fig. 1. The proposed tensor classification of twinning in crystals.

Table 1. *Comparison of attributes of the four types of twinning in crystals*

Attribute Twin type	Is a prototype structure conceivable?	Twins differ in which macroscopic tensor property ?	Generalized susceptibilities near the Curie temperature	Is detwinning possible ?	Origin of the twin
S	Yes	Second-rank polar. May differ in other properties also	At least one of them becomes arbitrarily large	Yes. Theory of ferroic transitions provides a systematic approach	Phase transition from the prototypic phase
N	Yes	Other than second-rank polar	At least one of them becomes arbitrarily large	Yes. Theory of ferroic transitions provides a systematic approach	Phase transition from the prototypic phase
B	No	No restriction on tensor rank, but such twins are not derivative structures	?	Very difficult. No general theoretical approach exists	Growth twin; non-ferroelastic mechanical twin; disruptive phase transition
T	'Yes' in some cases, 'no' in others	None	There is no relevant generalized susceptibility	No	(*a*) Transformation twin (*b*) Some types of stacking fault

sitions, from which *N*-twins arise. *N*-twinning occurs when there is a ferroic transition that does not involve a distortion of the lattice of the prototype, with *N* standing for 'no distortion' or for 'nonferroelastic–ferroic'.

Our approach to twinning thus leads to the result that all twins can be divided into four fundamentally different categories: *S*-twins, *N*-twins, *B*-twins and *T*-twins (Fig. 1). Since we have arrived at this conclusion through the transformation-twinning route, it remains to be seen whether this classification scheme is also adequate for growth twins and nonferroelastic mechanical twins. We examine this question in §5.

3.1. *S-twins*

The distinguishing feature of *S*-twins is a change of orientation of the optical indicatrix across the twin wall. In fact, they differ in at least one component of all second-rank polar tensor properties (the spontaneous strain tensor, the dielectric tensor, the magnetic permeability tensor *etc.*); see Table 1. Their presence is readily revealed under the polarizing microscope (because of the relative disorientation of the optical indicatrices) or in a diffraction experiment (because of a difference in the orientations of the respective crystallographic axes). Examples of purely ferroelastic *S*-twins are: $BiVO_4$, $BaCl_2 \cdot 2H_2O$ and $Pb_3(PO_4)_2$ (see Wadhawan, 1982).

S-twins may differ not only in second-rank polar tensor properties but also in other macroscopic properties like spontaneous polarization, spontaneous magnetization, spontaneous optical activity, compliance coefficients *etc.* Several examples are mentioned in VKW1, where the name *F*-twins is used for them.

For a contiguous ferroelastic domain pair, the domains on the two sides of the domain wall have different values of coefficients of the spontaneous-strain tensor. Such domains must therefore undergo small rotations (disorientations) for making contact at the domain wall; otherwise cracks can appear [see *e.g.* Wadhawan (1988) for a detailed analysis of this problem for the specific case of the ferroelastic superconductor Y–Ba–Cu–O]. Because of this, the operators that map one such domain to another (the twin operators) are not exactly equal to the g_j in (3). Since the disorientations involved do not generally entail additional changes of symmetry, one can usually work under the PCA, if needed.

Since the disorientations can occur in two or more equivalent ways, the actual possible number of disorientation states is more (often much more) than the number expected under the PCA (Boulesteix, 1984; Shuvalov, Dudnik & Wagin, 1985; Wadhawan, 1988).

The distinction between *S*-twins and *N*-twins is of a very basic nature. The spontaneous distortion of the prototype lattice, leading to the formation of *S*-twins, has several important consequences (Salje, 1990). One is the occurrence of disorientations and a variety of 'tweed structures' (Salje, 1994). Another is the effect of the spontaneous distortion on the kinetics of the ferroic phase transition (Salje, 1995) and on the critical fluctuations near the Curie temperature; the emerging phase tends to suppress the critical fluctuations because of the mismatch between the lattices of the old phase and the new phase (Friedel, 1981; Wadhawan, 1985). The nature of the critical fluctuations has a serious influence on response functions in the vicinity of the Curie temperature.

3.2. *N-twins*

The distinctive feature of *N*-twins is that they differ with respect to at least one macroscopic tensor property other than a second-rank polar tensor property and, in addition, their twinning pattern is describable in terms of a prototype space group (Table 1). Several examples of nonferroelastic ferroic properties, or combinations of such properties, are described in VKW1.

It is pertinent to point out here that polar second-rank tensors are invariant under an inversion operation. Therefore, if a contiguous domain pair is related only by an inversion operation, it cannot be a ferroelastic domain pair and is thus a (ferrogyrotropic) *N*-twin (see VKW1). It may alternatively be just a *B*-twin.

Apart from inversion, a mirror operation can also change the optical gyration tensor. A second-rank polar tensor is not necessarily invariant under a mirror operation. Thus it is possible to have ferrogyrotropic *S*-twins. Dicalcium strontium propionate (DSP) is an example of this (Wadhawan, 1979; Glazer, Stadnicka & Singh, 1981).

3.3. *B-twins*

Bollmann twins or *B*-twins differ from *S*-twins and *N*-twins in that a physically meaningful prototype structure is not conceivable for them. The following are their important subclasses:

(i) Twins resulting from phase transitions for which the nondisruption condition is violated. This includes reconstructive phase transitions.

(ii) All twinned crystals in their prototypic phase, rather than in a ferroic phase. This includes all mechanical twinning in crystals with a cubic or hexagonal point-group symmetry. An example is that of twinning induced in Mg crystals across $(10\bar{1}2)$ planes by a shear force along $[10\bar{1}\bar{1}]$.

(iii) Growth twins like the Brazil twins of α-quartz and β-quartz. Similarly, the 60°-rotation growth twins in crystals of GaAs, GaP and InAs are examples of *B*-twins (Chen *et al.*, 1992).

(iv) Twins resulting from special coincidence-lattice configurations (see §5.3).

Although a group-theoretical analysis of a *B*-twin configuration cannot make use of the powerful and highly successful concept of prototype symmetry, an alternative approach making use of the notion of the intersection group (Guymont, 1981; Toledano & Pascoli, 1981; Gratias & Portier, 1982; Kalonji, 1985) is still possible, which can even help in understanding the mechanism of formation of such twins. The basic idea of this approach rests on the often observed fact that, for such twins, a fraction of the structural edifice is common for the two components of the twin and remains undisrupted right across the twin boundary. Let H and H_2 denote the space-group symmetries of component 1 and component 2 of the *B*-twin. The two components have the same crystal structure but H and H_2 are distinct (with reference to a common system of coordinates) because the Seitz operator $\{R|\tau\}$ that maps component 1 to component 2 is not an identity operator; in fact, it necessarily has a rotational part. The two groups are related as follows:

$$H_2 = \{R|\tau\}H\{R|\tau\}^{-1}. \qquad (4)$$

Let I denote the group (the intersection group) comprising elements common to H and H_2:

$$I = H \cap H_2 = H \cap [\{R|\tau\}H\{R|\tau\}^{-1}]. \qquad (5)$$

Operators of I map component 1 back onto itself and, simultaneously, component 2 back onto itself. This group is a subgroup of H: $I \subset H$. Seitz operators h_j present in H but not present in I define the possible variants of the *B*-twin and can be identified with the cosets in the following coset decomposition:

$$H = I + h_2 I + \ldots + h_j I + \ldots + H_m I. \qquad (6)$$

Here m is the ratio of the orders of the groups H and I, in the spirit of (1).

This description applies when *B*-twins are produced by mechanical twinning or during crystal growth. We discuss next another aspect of *B*-twins.

Our definition of prototype symmetry is so strict that a substantial fraction of transformation twins resulting from structural phase transitions will qualify only as Bollmann twins, rather than Aizu twins. The most obvious examples of such transitions are, of course, the reconstructive transitions (*e.g.* graphite–diamond and calcite–aragonite). A symmetry analysis of *B*-twins can be carried out even in such cases by employing the notion of intersection symmetry. Let G_1 and G_2 denote the space groups of the two phases in any such case and let I be their intersection group:

$$I = G_1 \cap G_2. \qquad (7)$$

G_1 is not a subgroup of G_2 and G_2 is not a subgroup of G_1 but I is a subgroup of both G_1 and G_2. When we consider the phase transition $G_1 \rightarrow G_2$, the variants (*B*-twins) of the phase with symmetry G_2 can be listed by writing a coset decomposition of G_1 with respect to I:

$$G_1 = I + g_{11} I + g_{12} I + \ldots + g_{1j} I + \ldots + g_{1k} I. \qquad (8)$$

Similarly, when we consider the reverse phase transition $G_2 \rightarrow G_1$, the possible *B*-twin configurations in phase G_1 can be understood in terms of the coset decomposition of G_2 with respect to I:

$$G_2 = I + g_{21} I + g_{22} I + \ldots + g_{2j} I + \ldots + g_{2l} I. \qquad (9)$$

Physically what happens in such situations is that, although there is a change in the coordination numbers of a certain fraction of the atoms, some of the Wyckoff positions remain unchanged and define the group I. The phase transition in ZnS and CdS is an example of this type (Bol'shakov, Dmitrenko & Abalduev, 1979). For it, $G_1 = O_h^5$, $G_2 = D_{6h}^4$ and $I = C_{3v}^5$. One can not only carry out an enumeration of the possible *B*-twins, but can even define a 'composite order parameter' in the spirit of the

Landau theory by regarding the phase transition as a combination of (at least) two transitions: $G_1 \rightarrow I$ and $G_2 \rightarrow I$.

The above analysis suggests a method for a further subclassification of B-twins. Let us consider (6). I is a subgroup of H and m is the index of I in H ($m \geq 2$). A high value of m indicates, in general, a low commonality of Wyckoff positions across the twin boundary. Similarly, with reference to (8) and (9), high values of k and l are likely to point towards a strong violation of the nondisruption condition. We can very conveniently use m, k, l (as the case may be) as a subscript for the symbol B and describe the twin as a B_m-twin or a B_k-twin. Details of such an analysis, with illustrations, will be described elsewhere.

3.4. *T-twins*

Translation twins or T-twins do not differ in any macroscopic tensor property at all. Their presence can be detected with techniques such as HRTEM, etching and X-ray diffraction topography with a superlattice reflection. Twins across antiphase boundaries in the alloy Cu_3Au below 667 K are a familiar example of T-twins.

4. A symbol for twinning

Since our tensor classification of twinning is based primarily on notions drawn from the theory of transformation twinning, a compact symbol, which conveys a large amount of information about the physical nature of the twin, can be readily introduced, or rather reintroduced in the changed framework (see VKW1). The symbol consists of one of the letters S, N, B or T corresponding to the four basic types of twinning, followed by one or more lower-case letters in brackets, which denote the tensor properties of interest in which the twins differ. For T-twins, the second part of the symbol does not exist because such twins do not differ in any macroscopic tensor property.

We consider the case of twinning in quartz as an illustration of how the symbol is assigned. α-Quartz is ferrobielastic as well as ferroelastoelectric (see *e.g.* Wadhawan, 1982). The twin symbol for its Dauphiné twins is therefore $N(d, s)$, where d denotes the fact that the two components of the twin differ in at least one piezoelectric coefficient and s represents their difference with respect to the compliance tensor. On heating, when such a specimen makes a transition to the β phase, Dauphiné twinning disappears.

We consider next the Brazil twins of quartz. These are growth twins, with a mirror operation parallel to the optic axis as the twinning operation. This type of twinning does not disappear when α-quartz makes a phase transition to the higher-symmetry β-quartz. In any case, mirror symmetry is not present in the (prototypic) space-group symmetry $P6_222$ (or $P6_422$) of β-quartz, and cannot be a ferroic mapping operation g_j of (3). Brazil twins of quartz (both α and β) are thus B-twins (and not N-twins). The twin symbol for them is $B(g)$, where g represents the fact that the twin components have different optical gyration tensors (or opposite chiralities).

Table 2 gives several examples of the twin symbol. This table also serves to highlight the higher information content of the present tensor-classification scheme for twinning.

Information about the 'total' or 'partial' nature of a coincidence lattice (if any) underlying a twin can be included in our twin symbol by attaching a subscript t or p, as explained below in §5.3.

5. Discussion

A good classification scheme for twinning should have two important features: completeness and uniqueness. Completeness means that it should be possible to assign all observed types of twinning to one or other of the various categories defined by the classification. Uniqueness means that each observed example of twinning should be assignable to only one category. The earlier scheme (VKW1) did not meet the uniqueness criterion in a satisfactory manner, as exemplified by the case of Dauphiné twins of quartz. We have had to employ a different primary-classification criterion in the present paper to overcome this problem.

We have used concepts from the theory of phase transformations for formulating the present classification. Therefore, it is particularly suitable for classifying transformation twins. To satisfy the completeness requirement, it is necessary that the same scheme be applicable to nonferroelastic mechanical twins and to growth twins. We examine this question now. We also discuss here briefly the question of coincidence lattices in the context of twinning.

5.1. *Mechanical twins*

Two types of mechanical twins can be distinguished: those corresponding to ferroelastic domain pairs and the rest. The former are just S-twins and require no further discussion here. It should be possible to classify the rest of them as B-twins.

Type 1 mechanical twins (also called mirror twins) are defined as those for which the habit plane K_1, the second invariant direction η_2 and the shear plane have rational indices. The twinning operator for such twins can be written as follows in the Seitz notation (Stark, 1988):

$$\alpha_1 = \{m_{K_1} | t_1\}. \qquad (10)$$

Here m_{K_1} denotes the mirror operation across the habit plane and t_1 is an appropriate fractional translation, if any.

Table 2. *Comparison of the present classification scheme for twinning with two other schemes*

The application of the twin symbol introduced in this paper is also illustrated. When the twins differ in spontaneous polarization, the letter p appears in the twin symbol of the present scheme. Similarly, e denotes that the twins differ in spontaneous strain, m stands for spontaneous magnetization, g for a coefficient of the gyration tensor, d for a piezoelectric coefficient, s for compliance, q for magnetoelastic and α for magnetoelectric coefficients. The subscripts t and p denote the existence of total and partial coincidence sublattices, respectively, across the twin interface. Many of the entries are discussed at appropriate places in the text.

Twin	Present scheme (twin type)	Friedel's scheme (twinning by)	Donnay & Donnay's scheme (twinning by)	References
α-Quartz (Dauphiné)	$N(d, s)$	Merohedry	TLS	(1), (2)
$Pb_5Ge_3O_{11}$	$N(p, s, g)$	Merohedry	TLS	(3), (4)
$BaTiO_3$ (180° twin)	$N(p, g)$	Merohedry	TLS	(1), (4)
NH_4Cl	$N(d)$	Merohedry	TLS	(1), (4)
$CuCsCl_3$	$N(d, g)$	Merohedry	TLS	(1), (4), (5)
CoF_2, $FeCO_3$	$N(q)$	Merohedry	TLS	(1), (6)
Cr_2O_3	$N(\alpha)$	Merohedry	TLS	(1), (6)
α-Quartz (Brazil)	$B(g)$	Merohedry	TLS	(2)
Pyrite [(110) mirror]	B_t	Merohedry	TLS	(7), (8)
F.c.c. metals [(111) mirror twin]	B_p	Reticular merohedry	TLS	(7)
Mg	B	?	TLQS	(1)
Cu_3Au	T	?	TLS	(9)
$BaTiO_3$ (90° twin)	$S(p, e)$	Pseudomerohedry	TLQS	(1)
Fe_3O_4	$S(m, e)$	Pseudomerohedry	TLQS	(10)
Ni-I boracite	$S(p, m, e)$	Pseudomerohedry	TLQS	(10)
Aragonite	$S(e)$	Reticular pseudomerohedry	TLQS	(8)

References: (1) Wadhawan (1982); (2) Donnay & Donnay (1974); (3) Toledano & Toledano (1976); (4) Aizu (1972); (5) Wadhawan (1979); (6) Newnham & Cross (1974); (7) Gratias *et al.* (1979); (8) Cahn (1954); (9) Portier & Gratias (1982); (10) Aizu (1970).

Type 2 mechanical twins (or rotation twins) are characterized by a rational second-invariant plane K_2 and a rational shear direction η_1. A typical twin operator for them has the form

$$\alpha_2 = \{2_{\eta_1}|t_2\}. \qquad (11)$$

For a compound twin (Cahn, 1954), K_1, K_2, η_1 and η_2 all have rational indices.

It is clear from the above description that the twinning operator in all types of mechanical twins (type 1, type 2, compound) has the form of a Seitz operator indicated by (10) or (11). If this operator can be identified with one of the g_js in (3), the mechanical twin is an S-twin; if not, it can be identified with one of the h_js in (6) and we have a B-twin. But in either case our tensor classification of twinning is applicable to all mechanical twins (both ferroelastic and nonferroelastic).

5.2. *Growth twins*

We shall use the term 'growth twins' for twins formed during the nucleation and/or growth stages of crystal growth from a fluid phase. Twinning arising from the growth of a crystal from another solid phase is normally described as transformation twinning.

Growth of a crystal from a fluid phase is necessarily a process of first-order phase transition. Because of the negative and large change of entropy in going from the fluid phase to the crystalline phase, the process occurs in two distinct steps, namely nucleation and growth. And because of the competition between the surface contribution and the bulk contribution to the free energy, nuclei ('embryos') of a size below a certain critical value tend to be redissolved and only those above the critical size are able to grow into larger crystals. Growth twins can be formed at both the nucleation and the growth stages.

5.2.1. *Nucleation.* At the beginning of the formation of the nucleus, the surface-energy term is very dominant (Mutaftschiev, 1993). The term clusters is used for aggregates of atoms that, unlike molecules, are not found in appreciable numbers in vapours in equilibrium (Martin, 1988). Clusters may consist of about 100–1000 growth units or less. Small clusters undergo a process of reconstruction: every time a unit of the crystallizing species attaches itself to the cluster, the units rearrange themselves completely. This goes on until a large enough size is reached, beyond which the addition of one more growth unit does not lead to drastic reconstruction and the microcrystal already has the symmetry of the final bulk crystal. This cluster-to-crystal transition necessarily involves a change of symmetry (Multani & Wadhawan, 1990; Haberland, 1994). For every symmetry operator lost at such a transition, equivalent configurations (twins) can appear in the microcrystal and the possible twin variants would have a one-to-one correspondence with the cosets in (8).

The symmetry of the clusters need not always be from among the 32 crystallographic point groups. Icosahedral symmetry is favoured quite frequently. Gold clusters, for example, have icosahedral symmetry in the 4–15 nm regime. Since icosahedral symmetry is not compatible with translational periodicity of a bulk single crystal in three-dimensional space, the cluster-to-crystal transition in such a case is not a nondisruptive transition, so that Aizu twins can be ruled out in such a case. It may also involve multiple twinning as an adjustment mechanism (Ajayan & Marks, 1990; Riley, 1990). Gold clusters change from icosahedral symmetry to face-centred cubic symmetry on reaching a size of 15 nm (Renou & Gillet, 1981).

In the ultimate analysis, peculiarities of the atomic structure determine the laws of twinning at the nucleation or growth stages. For example, the energy of formation of a faulted two-dimensional embryo having the configuration of a rotation twin is very low for Si or Ge crystals (Tiller, 1991*a*). Twinning at even the nucleation stage is therefore quite likely to occur in them. The space-group symmetries involved are either noncrystallographic or prototypic (cubic for the case of Si) with *B*-twins as the only possibility.

5.2.2. *Growth.* The growth rates of the various habit faces are different because of their specific atomic structures. Similarly, the likelihood of a particular habit face becoming a twin interface increases if it has a high reticular density of atoms and if a large fraction of the atomic sites is common to the individuals comprising the twin. For example, in crystals with f.c.c. symmetry, the twin plane is parallel to the octahedral face, which has the maximum density of atoms. This is typical of the spinel law of twinning. Similarly, in aragonite twins, a fraction of the structure of $CaCO_3$ has a common orientation in the two components.

In growth twins, a twin-operation configuration provides additional re-entrant corners or junctions where the growth units can bind more strongly, resulting in enhanced growth rates compared with surface or terrace sites. However, since dislocations are also normally present in real specimens, the generation rate of layers of the growing crystal is determined by three primary competing mechanisms: (i) two-dimensional or 'pill-box' nucleation; (ii) screw dislocations; and (iii) twin-plane re-entrant corners (Sunagawa, 1987). In both (i) and (iii), layers are initiated by nucleation of two-dimensional pill boxes but in (iii) only a partial pill box, with a lower formation energy, is needed (Tiller, 1991*a*).

Mechanical twinning can also occur during the growth of a crystal due to internal mechanical and thermal stresses (Tiller 1991*b*).

In the laboratory, large crystals are often grown by starting with a seed crystal, which grows in size on being surrounded by the nutrient fluid under appropriate conditions. Kotru & Raina (1982) and Kotru, Kachroo & Raina (1985) investigated the occurrence of microtwins in hydrothermally grown synthetic quartz. They concluded that the microtwins mainly originate at the surface of the seed, and usually exist beforehand, having been produced by the stress at high temperatures generated by the sawing procedures used for obtaining the seed from a larger crystal. The mechanical twins so produced on the surface of the seed crystal are not always dissolved away fully by the etching practices adopted.

5.2.3. *Summary.* Twinning can occur during both the nucleation stage and the growth stage of a crystal. The formation of variants at the cluster-to-crystal transition is not a thoroughly investigated subject yet. However, in view of the noncrystallographic symmetry often adopted by clusters, and also because of the reconstructive nature of this transition, such growth twins are not very likely to be *S*-twins or *N*-twins. After the nucleation has occurred and growth is progressing, twinning can also arise as a result of 'probability accidents' (Tiller 1991*b*), especially when such 'accidents' can lead to increased growth rates because of the peculiarities of the atomic structure at certain habit faces. The re-entrant sites appearing as a result of such twinning operations can serve as ledges where attachment of growth units can occur at a faster rate than on the surface or terrace sites of the growing crystal.

Growth twins are expected to be *B*-twins, in general. The twinning operations involved in them are normally not traceable to a prototype symmetry group in the spirit of (3). We have already seen the case of Brazil twins in quartz. Another example is that of twins in III–V compound semiconductors, which have been identified by Chen *et al.* (1992) to be 60°-rotation twins, a nonferroic operation in the present context.

5.3. *Coincidence sublattices and twinning*

The translational symmetry of a twin is described by the translation group underlying the intersection group *I* [*cf.* (5)]:

$$T_I = T \cap [\{R|\tau\}T\{R|\tau\}^{-1}], \qquad (12)$$

where *T* is the translation group of component 1 of the twin.

T_I can have a dimensionality of 3, 2, 1 or 0 (Pond & Bollmann, 1979). A three-dimensional T_I implies the existence of a coincidence lattice running right across the interface of the twin. Although the group T_I comprises only lattice translations, the presence of Seitz operators in (12) makes it possible to identify point-group operations that achieve the same invariance of the twin as that achieved by pure lattice translations of T_I. Coincidence lattices can occur in both Aizu and Bollmann twins.

A crystal structure can be considered as consisting of a number of sublattices corresponding to the various sets of Wyckoff positions. Often, even a subset of one type of Wyckoff position can constitute a sublattice (with an underlying translation group) (Gratias, Portier, Fayard & Guymont, 1979; Doni, Bleris, Karakostas, Antonopoulos & Delavignette, 1985). A sublattice L is called a total sublattice by Gratias *et al.* (1979) if it consists of one or more complete sets of Wyckoff positions. If at least one set of Wyckoff positions is included only partially in the sublattice, it is a partial sublattice. Any number of sublattices can be constructed from a total or partial sublattice.

Several important features of total sublattices have been discussed by Gratias *et al.* (1979). The space group of a crystal is always a subgroup of the space group of any of its total sublattices. Further, if a total sublattice consists of only one set of Wyckoff positions, the twin mapping operation (if present) is always 'translation reducible' (comprises only a point operation).

Available information about the total or partial nature of a sublattice in a given twinned crystal can be readily incorporated in our classification scheme by appending a subscript t (for total) or p (for partial) to the symbols S, N or B.

The (110) mirror twins of pyrite (FeS_2) are an example of twins with a total sublattice (Gratias *et al.*, 1979). They are also B-twins. Therefore, the twin symbol for them is B_t (Table 1).

The (111) mirror twins commonly found in f.c.c. metals have a partial underlying coincidence lattice and their twin symbol therefore is B_p (Table 2).

6. Conclusions

The limitations of the classification scheme for twinning formulated in VKW1 are overcome in the present scheme, which has been obtained by using an entirely different classification criterion. In particular, the present classification provides a unique place for every kind of twinning. Consequently, there is no need to assign a higher priority or seniority to Aizu twins over Bollmann twins. Another improvement is the following: In VKW1, coincidence lattices are taken note of only for Bollmann twins, although their occurrence for N-twins is also mentioned in passing. In the present paper, it is recognized explicitly that coincidence lattices can occur for both Aizu twins and Bollmann twins.

The principal result of the present work is that all twinning in crystals can be divided into four fundamentally different types: S, N, B and T. Table 1 provides a comparison of their attributes. S-twins (which are called F-twins in VKW1) and N-twins are derivative structures, derived from an appropriate prototype symmetry by the loss of one or more symmetry operators. They are ferroic twins and therefore always differ in at least one macroscopic tensor property. B-twins also differ in at least one macroscopic tensor property but are different from S-twins and N-twins in that they are not derived from a supergroup prototype symmetry in a nondisruptive manner. The rigorous definition of prototype symmetry introduced in this paper in terms of the nondisruption condition makes this distinction very sharp. Several types of mechanical twins and practically all growth twins fall into the category of B-twins. Reconstructive transformations also result in B-twins only. T-twins do not differ in any tensor property at all; they may or may not be derivative structures the way ferroic twins are.

The fundamental nature of the distinction between S-twins and N-twins on one hand and B-twins on the other becomes all the more apparent in situations where a phase transition to the prototypic structure can actually be realized at a suitable temperature T_c and pressure P_c. In the vicinity of (T_c, P_c), the response function of a ferroic crystal for a macroscopic tensor property (corresponding to the order parameter of the transition) becomes arbitrarily large, a fact that can be exploited, for example, for the detwinning of such crystals (Wadhawan, 1982). Generally speaking, no such blowing up of the response function may occur for B-twins, simply because they do not arise as derivative structures from a real or hypothetical prototypic phase. However, the question of response functions in the vicinity of phase transitions that do not respect the nondisruption condition needs to be investigated thoroughly.

A compact and informative twin symbol has been introduced. The presence of any total or partial coincidence sublattice across a twin interface is an additional piece of information, which is easily incorporated in the twin symbol by the use of a subscript (t or p) with the main symbol.

The present formulation of the classification of twinning in terms of tensor properties makes it possible to make practical use of the results of group-theoretical analyses of tensor distinction of domains resulting from ferroic phase transitions (Janovec, Richterova & Litvin, 1992, 1993; Litvin, Litvin & Janovec, 1995; Litvin & Janovec, 1996).

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