

Symmetry Analysis of Interface Triple Junctions

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Abstract

The symmetry properties of triple junctions formed by crystal interfaces are presented using colour crystallography. Triple junctions are treated as composites of three crystals and are studied using the principle of superposition of symmetries. The conclusions are used for the understanding of the equivalences and invariances within one tricrystal composite and for studying such composites from the point of view of interfacial connectivity. A dissymmetrization procedure is established through which tricrystals equivalent by symmetry and their ways of coexistence can be found. Also, the relations between the symmetry properties of the tricrystal and those of the three bicrystals participating in the junction are given. Emphasis is placed on a particular class of composites that are termed variant constituted tricrystals. In these, two out of three bicrystals belong to orientation variants of the same dichromatic pattern. Such composites are usual in polycrystals and can exhibit interesting symmetry properties. The ordinary symmetry and antisymmetry in the third bicrystal can be predicted using a given algorithm. Appropriate examples are given to illustrate the application of the analysis.

1. Introduction

The problem of crystal interface triple junctions (TJs) has been a subject of interest for many years owing to their importance in affecting the behaviour and properties of polycrystalline materials. TJs can be classified as *homophase* or *heterophase*. Homophase TJs are the grain boundary TJs, *i.e.* TJs in which the three participating crystals have the same structure. In heterophase TJs, only two out of three crystal components have the same structure. Therefore, such TJs are composed of two heterophase interfaces and one homophase interface. Heterophase TJs are often observed (or constructed) in epitaxial deposition, precipitation and other phase-transformation processes. For example, epitaxy has been used for the fabrication of particular grain boundaries in the epitaxial deposit (Dahmen & Thangarai, 1993). In these cases, the composite of grain boundary plus epitaxial interface is a heterophase TJ. The grain boundary (GB) is the homophase boundary that

emanates from the junction of two variants of the epitaxial interface. Analogous is the case of internal twinning of precipitates that is observed with the twins emanating from the interphase boundary (Hugo & Muddle, 1989).

A lot of work has concentrated on grain-boundary TJs and a number of phenomena related to their properties have been studied. These include material mechanical behaviour (Hashimoto, Fujii & Miura, 1987; Sisanbaev & Valiev, 1992), wetting phenomena (Indekeu & Nikas, 1991), diffusion effects (Mikhailovskii, Rabukhin & Velikodnaya, 1991; Randle, 1993) and intergranular corrosion (Palumbo & Aust, 1988).

Grain-boundary TJs have also been classified according to their disclination character into *U* lines and *I* lines (Bollmann, 1984, 1988). *I* lines do not possess disclination character. The disclination character of *U* lines arises from the lack of nodal balancing of adjoining GB dislocation arrays. In addition, it is possible that TJ lines may exhibit partial dislocation character. Such a dislocation can be introduced, for example, if the crystal components are also related by rigid-body displacements that cannot be mutually accommodated at the TJ line.

It has been recognised that, in the coexistence of three crystals, symmetry may play an important role. However, previous work has concentrated on homophase TJs formed by crystals in coincidence-site-lattice (CSL) relative orientations. In such orientations, a lattice of common sites can always be found if the lattices of any two of the participating crystals are imagined to interpenetrate. The geometric properties of special CSL TJs have been analysed by Bleris & Karakostas (1989). Their results have been applied to experimental observations in polycrystalline Si (Komninou, Doni, Karakostas, Bleris & Delavignette, 1991).

The effort for the symmetry analysis of TJs cannot be regarded as complete. The role of symmetry in non-CSL TJs as well as heterophase TJs has not been elucidated. In addition, in CSL descriptions, only lattices and not actual crystal structures are considered. The question therefore remains as to what are the symmetry properties of any given TJ when the structures and relative orientations of the three crystals have been taken into account. It is expected that any symmetry that is shared by all three participating crystals will also characterize the TJ. Such common symmetry will express the invariances of all physical properties in a TJ. In addition, it may be

possible to obtain equivalence relations between the three crystals.

A second question is whether TJs that are equivalent by symmetry can be found. Such TJs will be variants obtained through systematic symmetry analysis. In a polycrystal, a number of such TJ variants can occur and their coexistence is possible. When equivalent-by-symmetry TJs coexist (for example at a node of TJs), what information can be obtained from symmetry as to how this coexistence is accommodated? If such questions can be answered, important conclusions will be obtained towards an *a priori* understanding of interfacial connectivity and the significance of TJs to polycrystal topology (Randle, 1993, 1994).

In this work, it will be shown that a unified approach can be adopted to study all types of TJs regardless of whether they are of the homophase or heterophase type. This can be accomplished by treating TJs as composite systems of three crystals mutually separated by three interfaces. Such composites will be termed *tricrystals*. General rules can be obtained to express equivalences and invariances in these systems. All equivalent-by-symmetry TJs can be predicted and their coexistence can be considered. A geometrically necessary defect content, other than that arising from nodal balancing of dislocation arrays, can also be predicted and assigned to particular TJs.

The study of composite systems can be facilitated through the use of the principle of superposition of symmetries (Shubnikov & Koptsik, 1974). According to this principle, the symmetry of a heterogeneous composite is the symmetry that is common in all its constituents depending on their relative arrangement. It follows that the composite will usually exhibit symmetry lower than that of its parts. This reduction of symmetry is known as dissymmetrization. Any symmetry that is suppressed by dissymmetrization will then relate equivalent variants of the same composite. In a homogeneous composite, it is possible to have additional symmetry, *i.e.*, apart from the common symmetry, there may also exist symmetry operations that relate the constituents of the composite and express equivalence between them. This is referred to as symmetrization.

Composite systems can be studied under the framework of generalized colour symmetry. A two-colour (dichromatic) approach has been employed for the case of bicrystals, *i.e.* composites of two crystals separated by one interface (Kalonji & Cahn, 1982; Pond & Vlachavas, 1983). Each crystal has been assigned a different colour, *i.e.* black or white. The bicrystal symmetry is expressed by a group in which two types of symmetry operation are represented. The first is ordinary symmetry operations, *i.e.* operations that leave invariant both the white and the black component and therefore the bicrystal as well. The second is antisymmetry (colour-reversing) operations that relate the black and white components. For such systems, a dissymmetrization procedure has been

established through which the symmetry of a given bicrystal as well as all equivalent bicrystal variants can be found. The symmetry analysis can then give information on the ways such variants can coexist (Pond, 1989).

For a tricrystal composite, it is natural to assume that the appropriate framework to use is that of three-colour crystallography. Such an approach will be developed in this work. A different colour will be assigned to each crystal component of the composite, say white, black and red. Then it is possible to distinguish three types of symmetry operations characterizing such a composite, *i.e.* ordinary, colour-exchange and mixed operations (Dahmen & Thangarai, 1993). Colour-exchange operations express equivalence between all three components, *i.e.* they involve a cyclic permutation of colour (white-to-black-to-red-to-white or *vice versa*). Mixed symmetry operations relate two of the three components (*e.g.* black-to-red) while leaving the third (*e.g.* white) invariant.

The symmetry group of a composite is obtained by dissymmetrizing a fundamental or embracing symmetry group. This is a group expressing the maximum possible symmetry associated with a given composite. For bicrystals, it has been established that the embracing group is the one characterizing the symmetry of the holosymmetric dichromatic pattern. This is the three-dimensional pattern that is constructed if we consider the black and white lattices to interpenetrate in space, having a common origin. The bicrystal is regarded as having been created from such a pattern by a sequence of imaginary steps. In each step, the symmetry is reduced and hence variants arise. It is easy to extend such an analysis for the case of tricrystals. The same sequence of dissymmetrization steps can be followed if we associate the embracing group for the composite with a holosymmetric tricrystal pattern, *i.e.* a three-dimensional composite constructed if we consider all three lattices to interpenetrate having a common origin. Establishing a dissymmetrization methodology similar to that used in bicrystallography is useful considering that, in a tricrystal, three bicrystals coexist. So, in order to understand interfacial connectivity, it is necessary to obtain a connection between the symmetry properties of a tricrystal and a bicrystal. It should be noticed that ordinary and colour symmetry operations in a composite can be found by a simple inspection of the relative orientation of its parts, making the matter trivial. However, this gives partial information since it is the embracing group that describes the highest symmetry associated with an imaginary medium from which the composite is developed. Thus, dissymmetrization of this group is the necessary analysis for a complete study of the symmetry properties and their influence in the defect content of such systems.

Special attention will be paid to a particular class of tricrystals that we term variant constituted tricrystals (VCTs). By this expression, we mean tricrystals in which

two out of three bicrystals originate from orientation variants of one dichromatic pattern. Our interest is based on their frequent occurrence in polycrystals and in a number of processes, such as phase transformations. For example, consider the well known case of $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ CSL TJs (the multiplicity Σ expresses the reciprocal density of coincident-lattice nodes in a CSL). The two $\Sigma 3$ bicrystals originate from dichromatic pattern variants. In materials with low stacking-fault energy, such bicrystals usually have $\{111\}$ interfacial orientations and are abundant. It is reasonable to assume that, in a polycrystalline material, all $\Sigma 3\{111\}$ variants have equal probability of existing since they all have low energy. Then, it is also highly probable that such variants will often form VCTs and so the number of $\Sigma 9$ bicrystals will also be large. Symmetry constraints are imposed by the $\Sigma 3\{111\}$ variants on the $\Sigma 9$ bicrystals. These can be established so that they are known *a priori*. In general, the effect of low-energy interfaces on polycrystal topology can be largely considered through their coexistence in VCTs, as well as through the coexistence of VCTs. In addition, the information obtained on VCTs can facilitate the study of tricrystals that are directly related to them, as will be demonstrated. In the CSL case, such tricrystals are those in which the three bicrystals have multiplicities related by a power law (*i.e.* $\Sigma_i = f^n$ bicrystals, for example $\Sigma 3$ - $\Sigma 9$ - $\Sigma 27$). The symmetry relations obtained can also aid the design and fabrication of VCTs, for example, by epitaxy, as in the method of Dahmen & Thangarai (1993).

The structure of the paper is divided into three main sections. The first section involves the characterization of the symmetry of tricrystal composite systems by examining the interrelationships of the participating crystals. For this purpose, the principle of superposition of symmetries is used. It is shown how suppressed symmetry can be used to predict equivalent tricrystal variants and their possible ways of coexistence. The second section is devoted to VCTs. It focuses on the symmetry of the third bicrystal introduced between bicrystals originating from dichromatic pattern orientation variants and examines how this symmetry is affected by the variants. The final section gives appropriate examples that are studied using the analysis of the previous two sections.

2. Symmetry of tricrystal composites

2.1. The dissymmetrization procedure in dichromatic bicrystallography

The analysis of the symmetry of a tricrystal can be formulated following a dissymmetrization procedure similar to that used in bicrystallography. For this reason, and because certain results concerning the symmetry of a single bicrystal are necessary for the study of a tricrystal,

this procedure is reviewed here in brief. Detailed analyses can be found elsewhere (Kalonji & Cahn, 1982; Gratias & Portier, 1982; Pond & Vlachavas, 1983).

Using the notation of *International Tables for Crystallography* (Hahn, 1983), let us denote by $\mathcal{G}(\lambda)$ and $\mathcal{G}(\mu)$ the space groups of symmetry operations of the white (λ) and black (μ) lattice, respectively. $\mathcal{G}(\lambda)$ and $\mathcal{G}(\mu)$ are each isomorphic to one of the 14 crystallographic space groups of lattices, although not necessarily the same. Let us now assume that, when referred to the coordinate system of crystal (λ), $\mathcal{G}(\lambda)$ is represented by the isomorphic group of 4×4 matrices $\{\mathcal{W}(\lambda)_i\}$. For lattices, the matrix representations of symmetry operations $\mathcal{W}(\lambda)_i = (\mathbf{W}(\lambda)_i, \mathbf{w}(\lambda)_i)$ are either of the form $(\mathbf{W}(\lambda)_i, \mathbf{o})$ for point symmetry operations or $(\mathbf{I}, \mathbf{t}(\lambda)_i)$ for translation operations. Similarly, let the space group $\mathcal{G}(\mu)$ be represented by the isomorphic group $\{\mathcal{W}(\mu)_i\}$ when referred to the coordinate system of crystal (μ).

An affine transformation of the coordinate system can be expressed as $\mathcal{P} = (\mathbf{P}, \mathbf{p})$, where \mathbf{P} is the linear part of the transformation expressing a change in length and/or orientation of the basis vectors and \mathbf{p} a shift of origin corresponding to a possible rigid-body translation between the two components. The linear part \mathbf{P} can always be expressed in the form of a product $\mathbf{P} = \mathbf{R}\mathbf{D}$, where \mathbf{R} corresponds to a rotation and \mathbf{D} to a pure strain. For the purpose of our analysis, it will always be assumed that the relative rotation is not the identity ($\mathbf{R} \neq \mathbf{I}$). Let us assume that \mathcal{P} is defined so that the vector bases are related by $(\mathbf{a}(\mu), \mathbf{b}(\mu), \mathbf{c}(\mu)) = (\mathbf{a}(\lambda), \mathbf{b}(\lambda), \mathbf{c}(\lambda))\mathbf{P}$. For a given bicrystal, the set of all equivalent-by-symmetry descriptions of this transformation is given by $\mathcal{PW}(\mu)_i$. Any such transformation can be used for obtaining the symmetry of a bicrystal. The imaginary steps for doing so are given below.

We consider initially the construction of the holosymmetric dichromatic pattern. We imagine the lattices of the two crystals to interpenetrate, assuming a common origin (*i.e.* $\mathbf{p} = \mathbf{o}$). Let the symmetry of this composite be expressed by the space group $\mathcal{G}'(p)$ (p denotes pattern). This is the embracing group of the bicrystal (Pond & Vlachavas, 1983) and, in the general case, it is a two-colour group. The actual bicrystal symmetry is always expressed by a subgroup of this group.

We next denote by $\mathcal{G}(p)$ the ordinary symmetry subgroup of the embracing group ($\mathcal{G}(p) \subseteq \mathcal{G}'(p)$), *i.e.* the group of symmetry operations that leave invariant both crystal components. This is a subgroup of the space groups of the two lattices ($\mathcal{G}(p) \subseteq \mathcal{G}(\lambda), \mathcal{G}(\mu)$) isomorphic to the group of matrices satisfying the expression $\mathcal{W}(p)_i = \mathcal{W}(\lambda)_i = \mathcal{P}_o \mathcal{W}(\mu)_i \mathcal{P}_o^{-1}$, where $\mathcal{P}_o = (\mathbf{P}, \mathbf{o})$. Symmetry operations belonging to $\mathcal{G}(p)$ are rotations and mirrors for which the corresponding geometric elements are parallel in the two crystals, as well as parallel translations of the same length. Also, the inversion is always an element of $\mathcal{G}(p)$ since its matrix is always invariant under the above transformation by \mathcal{P}_o .

If antisymmetry is not present, then $\mathcal{G}(p) \equiv \mathcal{G}'(p)$, whereas, if it is (*i.e.* in homophase bicrystals), $\mathcal{G}'(p)$ can be found as the extension of $\mathcal{G}(p)$ by a symmetrizing point group containing the identity and one second-order antisymmetry operation. The rules for the introduction of antisymmetry have been given by Vlachavas (1984).

For the analysis of TJs, the point symmetry is particularly important. Therefore, a few comments on it are necessary at this point. Let us denote by $\mathcal{Q}(\lambda)$ and $\mathcal{Q}(\mu)$ the point groups corresponding to the two lattices [*i.e.* $\mathcal{Q}(\lambda) \subset \mathcal{G}(\lambda)$, $\mathcal{Q}(\mu) \subset \mathcal{G}(\mu)$]. As known, each is isomorphic to one of the crystallographic point groups $\bar{1}$, $2/m$, mmm , $4/mmm$, $\bar{3}m$, $6/mmm$, $m\bar{3}m$. Then, if $\mathcal{Q}(p)$ is the point group corresponding to the dichromatic pattern [*i.e.* $\mathcal{Q}(p) \subseteq \mathcal{G}(p)$], it can only be isomorphic to one of the centrosymmetric groups $\bar{1}$, $2/m$, mmm , $4/m$, $4/mmm$, $\bar{3}$, $3m$, $6/m$ and $6/mmm$. The rest of the centrosymmetric groups are overruled for relative rotations. The above groups can only be extended to the antisymmetric groups $2'/m'$, $m'mm'$, $4'/mm'm'$, $4'/mmm'$, $8'/mmm'$, $\bar{3}m'$, $6'/m'mm'$, $6'/mm'm'$ and $12'/mmm'$, respectively.

As we have seen, $\mathcal{Q}(p)$ will always contain at least the identity and the operation of inversion. Let us now examine for which relative rotations, corresponding to \mathbf{R} , $\mathcal{Q}(p)$ can contain additional symmetry operations. As known, any symmetry operation corresponds to a geometric symmetry element such as a rotation axis or mirror plane. For operations belonging to $\mathcal{Q}(p)$, the corresponding geometric elements must be invariant under the relative rotation of the two crystals. Such geometric elements are symmetry axes parallel to the axis of relative rotation as well as mirror planes normal to this axis. Also, under a relative rotation, axes or mirrors for which the corresponding symmetry operations belong to different equivalence classes may be brought into coincidence. For crystal symmetries, this is possible only for second-order symmetry operations [*i.e.* when $\mathcal{Q}(p)$ is isomorphic to one of the groups mmm , $4/mmm$, $6/mmm$]. Finally, by a 180° relative rotation, symmetry axes perpendicular to the axis of relative rotation are left invariant, as well as mirrors to the planes of which the axis of relative rotation belongs. For example, in cubic crystals such 180° rotations must be around axes $\langle hk0 \rangle$ (for $\langle 001 \rangle$ coincidence), $\langle hhk \rangle$ (for $\langle 110 \rangle$ coincidence) or $\langle 211 \rangle$ (for $\langle 111 \rangle$ coincidence). The above arguments have been written for proper rotations, but can easily be extended for improper ones.

For homophase composites, the $2/m$ ordinary symmetry corresponds to a $\langle 110 \rangle$ coincidence in cubic and tetragonal crystals and to a $\langle 11\bar{2}0 \rangle$ or $\langle 1\bar{1}00 \rangle$ coincidence in hexagonal crystals. The mmm symmetry corresponds to second-order axes $\langle 100 \rangle$, $\langle 011 \rangle$, $\langle 011 \rangle$ (90° $\langle 110 \rangle$ rotation). A coincidence with axes of type $\langle 100 \rangle$ only corresponds to a 45° $\langle 100 \rangle$ rotation (symmetry $4/mmm$). The groups $\bar{3}m$ and $6/mmm$ also correspond to special rotations, *i.e.* 60° $\langle 111 \rangle$ and 30° $[0001]$, respectively.

A coset analysis of $\mathcal{Q}(\lambda)$ with respect to $\mathcal{Q}(p)$ determines the variants of $\mathcal{Q}(p)$ in $\mathcal{Q}(\lambda)$ and therefore the orientation variants of the holosymmetric dichromatic pattern inside the white lattice. If we denote by $W(\lambda)_i$ a symmetry operation of $\mathcal{Q}(\lambda)$ corresponding to $\mathcal{W}(\lambda)_i = (W(\lambda)_i, \mathbf{o})$, such an analysis can be written as

$$\mathcal{Q}(\lambda) \equiv W(\lambda)_{v_1} \mathcal{Q}(p) \cup W(\lambda)_{v_2} \mathcal{Q}(p) \cup \dots \cup W(\lambda)_{v_n} \mathcal{Q}(p), \quad (1)$$

where $W(\lambda)_{v_1} = I$ (the identity) and $W(\lambda)_{v_\kappa} \in \mathcal{Q}(\lambda)$, $W(\lambda)_{v_\kappa} \notin \mathcal{Q}(p)$, $\kappa = 2, \dots, n$, are the variant generating group elements, which in the following will be abbreviated as VGEs. The latter are symmetry operations corresponding to the matrix representations $\mathcal{W}(\lambda)_{v_\kappa} = (\mathbf{W}(\lambda)_{v_\kappa}, \mathbf{o})$. A similar analysis can be written with respect to $\mathcal{Q}(\mu)$. If the coset analysis (1) was written with respect to space-group symmetries, translational variants of the dichromatic pattern would also have been obtained.

The VGEs express the dissymmetry (or suppressed symmetry) and interrelate the orientation variants. This suppression is due to the relative arrangement of the two lattices in space or to there being no corresponding operation in $\mathcal{Q}(\mu)$.

There are also cases where one or both crystals can be considered as transformation products of phases of higher symmetry. The parent phase lattice may not be the same for both components. The interpenetration of such phases should be considered in obtaining the embracing group as they can lead to additional orientation variants due to different possible orientations of the black or white lattice within the parent phases. Other authors have analysed the issue (Van Tendeloo & Amelinckx, 1974; Guymont, Gratiias, Portier & Fayard, 1976).

After the embracing group of the bicrystal has been obtained, the second step in the dissymmetrization process is to obtain the space group characterizing the composite that is formed if the actual crystal structures (*i.e.* the lattice complexes) interpenetrate. This composite is known as the dichromatic complex. The space groups $\mathcal{G}(\lambda)^s$ and $\mathcal{G}(\mu)^s$ of the crystal structures are considered (each being isomorphic to one of the 230 crystallographic space groups). The ordinary symmetry group $\mathcal{G}(c)$ (c denotes complex) is obtained through the expression $\mathcal{W}(c)_i = \mathcal{W}(\lambda)_i = \mathcal{P}\mathcal{W}(\mu)_i\mathcal{P}^{-1}$ for the matrix representations of symmetry operations, where now the representations of point symmetry operations are of the form $\mathcal{W}_i = (\mathbf{W}_i, \mathbf{w}_i)$ with $\mathbf{w}_i \neq \mathbf{o}$ for non-symmorphic operations. Any relative displacement of the two components is also introduced, *i.e.* the transformation $\mathcal{P} = (\mathbf{P}, \mathbf{p})$ is used.

This step enables the treatment of non-holosymmetric and/or non-symmorphic crystals. In the non-holosymmetric case, there will exist certain symmetry operations that leave the lattice invariant but not the lattice complex.

Therefore, such operations will not leave invariant the dichromatic complex although they may do so for the dichromatic pattern. Also, in the non-symmorphic case, there may be cases where the lattice symmetry operation corresponding to $(\mathbf{W}_i, \mathbf{o})$ was a symmetry operation of the dichromatic pattern but its non-symmorphic counterpart in the lattice complex corresponding to $(\mathbf{W}_i, \mathbf{w}_i)$ does not leave invariant the dichromatic complex. This will be due to the fact that the vectors \mathbf{w}_i are not parallel and/or not identical in the two crystals. Additional dissymmetrization may arise due to the shift in origin \mathbf{p} .

It has been proven (Pond & Vlachavas, 1983) that $\mathcal{G}(c) \subseteq \mathcal{G}(p)$. As the ordinary symmetry is reduced, antisymmetry must be reduced accordingly, *i.e.* $\mathcal{G}'(c) \subseteq \mathcal{G}'(p)$. Any suppression of symmetry when going from the dichromatic pattern to the complex leads to a number of variants of the latter in the former. Such variants are termed complex variants.

In the final dissymmetrization step, the bicrystal two-dimensional symmetry is obtained by considering the interface as a mathematical plane. Only the symmetry and antisymmetry operations of $\mathcal{G}'(c)$ that leave this plane invariant are conserved at this stage. Operations that do not do so relate variants of the bicrystal within the complex. Such variants have been termed morphological variants (Pond & Vlachavas, 1983).

The symmetry operations relating any two bicrystal variants, whether orientation, complex or morphological, have been used in order to characterize *a priori* the geometrically necessary interfacial defect accommodating their coexistence (Pond, 1989). Since a similar dissymmetrization methodology will be adopted below for tricrystals, it will be useful to examine whether geometrically necessary defects that accommodate the coexistence of tricrystal variants can be predicted.

2.2. Relations between the rotations of the three crystals and construction of a variant constituted tricrystal

After giving the dissymmetrization procedure for bicrystals, and before proceeding with the appropriate analysis for tricrystals, we recall that, in TJs, the linear transformations between the three crystal components (as shown in Fig. 1) can always be related by the equation

$$\mathbf{P}_3 = \mathbf{P}_2 \mathbf{P}_1^{-1} \quad (2)$$

when expressed in the (λ) coordinate frame (providing, of course, that there is no rotational closure failure between the three crystals). Among the set of equivalent transformations relating any two crystals, three transformations for which (2) is valid can always be found. However, this expression may not be valid for the rigid-body translations \mathbf{p} [*i.e.* for operations of the form $\mathcal{P} = (\mathbf{P}, \mathbf{p})$].

In this subsection, we also consider the construction of the particular class of VCTs. According to the definition given in the *Introduction*, such a tricrystal is obtained

when there is no crystal continuity at the junction of two bicrystals originating from orientation variants of the same dichromatic pattern (crystal continuity necessitates the existence of a disclination at their junction). Then a third bicrystal is always geometrically necessary at this junction. This approach is in variance to the treatment of Pond (1989), where continuity is always assumed.

The definition of a VCT does not impose any conditions on the participating complex variants and does not limit the interfacial orientations in the orientation variants to be crystallographically equivalent. So this is a quite general category of tricrystals. For example, consider the well known case of $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ CSL TJs. The $\Sigma 3$ bicrystals are obtained from dichromatic pattern orientation variants. Usually, these are both $\Sigma 3\{111\}$ bicrystals, but it has also been observed that one $\Sigma 3\{111\}$ can coexist with one $\Sigma 3\{211\}$ bicrystal in the same TJ (Garg, Clark & Hirth, 1989). In both cases, we have a VCT.

Let $\mathcal{G}(\lambda)$, $\mathcal{G}(\mu)$, $\mathcal{G}(\varepsilon)$ be the space groups of symmetry operations of the three lattices (ε , taken from the Greek word *erithros*, denotes the red crystal). In accordance with the classification of TJs as either homophase or heterophase, we impose the condition $\mathcal{G}(\mu) \equiv \mathcal{G}(\varepsilon)$ when referred to their respective coordinate frames. Let also $\mathcal{G}'(p)_1$, $\mathcal{G}'(p)_2$, $\mathcal{G}'(p)_3$ be the embracing groups of the three bicrystals (as shown in Fig. 1) and $\mathcal{G}(p)_1$, $\mathcal{G}(p)_2$, $\mathcal{G}(p)_3$ the corresponding ordinary symmetry subgroups.

Then, if bicrystal 2 originates from an orientation variant of the dichromatic pattern of bicrystal 1 corresponding to a VGE $W(\lambda)_{v_k} \notin \mathcal{G}(p)_1$, we have $\mathcal{G}(p)_2 \equiv W(\lambda)_{v_k} \mathcal{G}(p)_1 W(\lambda)_{v_k}^{-1}$ [note that the coset analysis (1) can easily be extended for space groups if translation symmetries are added, without changing the number of orientation variants].

For the second bicrystal, we obtain the transformation $\mathcal{P}_{o2} = \mathcal{W}(\lambda)_{v_k} \mathcal{P}_{o1} \mathcal{W}(\varepsilon)_m^{-1}$, having linear part $\mathbf{P}_2 = \mathbf{W}(\lambda)_{v_k} \mathbf{P}_1 \mathbf{W}(\varepsilon)_m^{-1}$, where $\mathcal{W}(\lambda)_{v_k} = (\mathbf{W}(\lambda)_{v_k}, \mathbf{o})$ is the matrix representation corresponding to the symmetry operation $W(\lambda)_{v_k}$, and $\mathcal{W}(\varepsilon)_m = (W(\varepsilon)_m, \mathbf{o})$ corresponds to a symmetry operation of $\mathcal{G}(\varepsilon)$. [Multiplying \mathcal{P}_{o1} from the left by $\mathcal{W}(\lambda)_{v_k}$ gives a transformation of bicrystal 2. We are then at liberty to follow by multiplying from the right by any matrix $\mathcal{W}(\varepsilon)_m$ corresponding to a symmetry

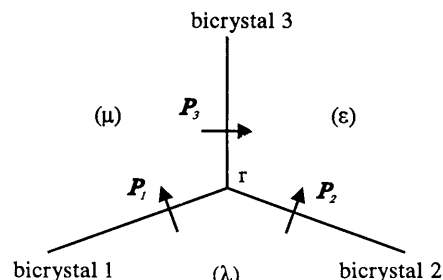


Fig. 1. Relative rotations and notations used in the analysis.

operation of $\mathcal{G}(\varepsilon)$.] So (2) is re-written as $\mathbf{P}_3 = \mathbf{W}(\lambda)_{\nu_k} \mathbf{P}_1 \mathbf{W}(\varepsilon)_m^{-1} \mathbf{P}_1^{-1}$ [or $\mathcal{P}_{o3} = \mathcal{W}(\lambda)_{\nu_k} \mathcal{P}_{o1} \mathcal{W}(\varepsilon)_m^{-1} \mathcal{P}_{o1}^{-1}$], when expressed in the (λ) coordinate frame. This relation becomes the Q_{ij} expression of Pond (1989) if continuity between the bicrystals obtained from dichromatic pattern orientation variants is assumed. Transformed in the (μ) coordinate system, \mathbf{P}_3 becomes $\mathbf{C} = \mathbf{P}_1^{-1} \mathbf{P}_3 \mathbf{P}_1 = \mathbf{P}_1^{-1} \mathbf{W}(\lambda)_{\nu_k} \mathbf{P}_1 \mathbf{W}(\varepsilon)_m^{-1}$. Then, since there is always an operation of $\mathcal{G}(\mu)$ such that $\mathbf{W}(\varepsilon)_m \equiv \mathbf{W}(\mu)_l$ (when referred to their respective coordinate frames), one of the transformations equivalent to \mathbf{C} is

$$\mathbf{C}_k = \mathbf{C} \mathbf{W}(\mu)_l = \mathbf{P}_1^{-1} \mathbf{W}(\lambda)_{\nu_k} \mathbf{P}_1 \quad (3)$$

corresponding to the transformation $\mathcal{C}_{ok} = (\mathbf{C}_k, \mathbf{o})$. This is an expression at the (μ) coordinate system of the matrix representation corresponding to the VGE in the coordinate system of (λ) .

2.3. Ordinary symmetry of the holosymmetric tricrystal pattern

After giving the relations between the three transformations, we can proceed with the symmetry analysis of tricrystals using the steps that have been described in §2.1 and the principle of superposition of symmetries for composite systems. Under such a methodology, the characteristic symmetry of any tricrystal can be found. Since in any tricrystal three bicrystals coexist, using the same sequence of dissymmetrization steps for both bi- and tricrystals will enable us to obtain a connection between their symmetry properties.

The methodology involves the dissymmetrization of an embracing group. Symmetry operations that are suppressed at the various steps of dissymmetrization relate tricrystal variants. So the steps enable us to distinguish between the ways such variants can coexist.

The first step is the characterization of the symmetry of the holosymmetric tricrystal pattern. The space group of this composite is given by the general expression $\mathcal{G}(p)_{(\text{tricrystal})} \equiv \mathcal{G}(p)_{(\text{tricrystal})} \odot \mathcal{S}$, where subgroup $\mathcal{G}(p)_{(\text{tricrystal})}$ is the space group of ordinary symmetry operations leaving the composite invariant and \mathcal{S} is a symmetrizing point group of colour-associated operations (\odot here indicates the operation of symmetrization). The space group $\mathcal{G}(p)_{(\text{tricrystal})}$ is the embracing group of the composite.

In this subsection, we are interested in the ordinary symmetry of the tricrystal pattern, *i.e.* in the group $\mathcal{G}(p)_{(\text{tricrystal})}$. For it, the following theorem is self-evident.

Theorem 1: In order to find $\mathcal{G}(p)_{(\text{tricrystal})}$, it is sufficient to consider the intersection $\mathcal{G}(p)_{(\text{tricrystal})} \equiv \mathcal{G}(p)_1 \cap \mathcal{G}(p)_2$, where $\mathcal{G}(p)_{(\text{tricrystal})} \subseteq \mathcal{G}(p)_1, \mathcal{G}(p)_2$.

This intersection expresses the addition of the dissymmetries in the composite (Shubnikov & Koptsik,

Table 1. *Common intersections for all permissible point symmetries of $\mathcal{Q}(p)_1$ for all crystal systems (with the exception of triclinic) in the case of homophase VCTs*

$\mathcal{Q}(p)_{(\text{tricrystal})} = \mathcal{Q}(p)_1 \cap \mathcal{Q}(p)_2$						
$\mathcal{Q}(p)_1$	Mono- clinic (2/m)	Ortho- rhombic (mmm)	Tetragonal (4/mmm)	Trigonal (3m)	Hexagonal (6/mmm)	Cubic (m3m)
$\bar{1}$	$\bar{1}$	$\bar{1}$	$\bar{1}$	$\bar{1}$	$\bar{1}$	$\bar{1}$
2/m		2/m	$\bar{1}, 2/m$		$\bar{1}, 2/m$	$\bar{1}, 2/m$
mmm			mmm			$\bar{1}, mmm$
4/m			4/m			$\bar{1}, 4/m$
4/mmm						mmm
$\bar{3}$				$\bar{3}$		$\bar{1}, \bar{3}$
3m						2/m
6/m					6/m	

1974). Any operation that belongs to both $\mathcal{G}(p)_1$ and $\mathcal{G}(p)_2$ will also belong to $\mathcal{G}(p)_3$ and hence to $\mathcal{G}(p)_{(\text{tricrystal})}$ as well.

Then, for VCTs, a second theorem is easily obtained by considering the expressions for $\mathbf{P}_1, \mathbf{P}_2$ and \mathbf{P}_3 given in §2.2.

Theorem 2: In VCTs, $\mathcal{G}(p)_{(\text{tricrystal})}$ is the subgroup of $\mathcal{G}(p)_1$ that is invariant under the VGE $\mathcal{W}(\lambda)_{\nu_k}$.

Table 1 gives all possible point groups isomorphic to $\mathcal{Q}(p)_{(\text{tricrystal})} \subseteq \mathcal{G}(p)_{(\text{tricrystal})}$ for all point groups $\mathcal{Q}(p)_1$, and for all crystal systems (with the exception of the trivial triclinic), in the case of homophase VCTs. This table is constructed using the analysis on $\mathcal{Q}(p)$ permissible symmetries that has been given in §2.1. For heterophase VCTs, we may need to dissymmetrize the given groups by any operations of $\mathcal{Q}(\lambda)$ that are not also operations of $\mathcal{Q}(\mu)$. From this table, we observe that, in general, $\mathcal{Q}(p)_{(\text{tricrystal})}$ includes operations other than just the identity and the inversion only when $\mathcal{Q}(p)_1$ is invariant under the VGE. Then, $\mathcal{Q}(p)_{(\text{tricrystal})} \equiv \mathcal{Q}(p)_1$. There are, however, two cases that do not conform to this rule. These correspond to special rotations in the cubic system. The first is the 60° $\langle 111 \rangle$ rotation (intersection symmetry 2/m) and the second is the 45° $\langle 001 \rangle$ rotation (intersection symmetry mmm).

In addition, from Table 1, the following theorem can be obtained

Theorem 3: In VCTs, the necessary condition for the existence of intersection symmetry [$\mathcal{Q}(p)_{(\text{tricrystal})}$] greater than $\bar{1}$ is the existence of a second-order VGE.

This theorem can be proven by considering the arguments for the coincidence of geometric symmetry elements given in §2.1. Assume that there are some symmetry operations of $\mathcal{Q}(p)_1$ (other than the identity and the inversion) that belong to $\mathcal{Q}(p)_{(\text{tricrystal})}$. Since the VGE does not belong to $\mathcal{Q}(p)_{(\text{tricrystal})}$, it cannot correspond to a 60, 90 or 120° rotation axis parallel to the axis or mirror normal corresponding to a symmetry operation of $\mathcal{Q}(p)_{(\text{tricrystal})}$. However, the geometric elements of the operations of $\mathcal{Q}(p)_{(\text{tricrystal})}$ must be left invariant under

the operation of the VGE. The only way to satisfy both requirements is for the VGE to be a second-order symmetry operation. For example, consider a rotation symmetry axis corresponding to an operation of $\mathcal{Q}(p)_1$. This axis will also be invariant under the operation of the VGE if the VGE corresponds to a 180° rotation around an axis perpendicular to it or to a mirror plane to which this axis belongs. Then the corresponding symmetry operation will belong to $\mathcal{Q}(p)_{(\text{tricrystal})}$. So the conditions that have been outlined in §2.1 for relative rotations must be combined with the conditions imposed on the VGE if the intersection symmetry is to exceed $\bar{1}$.

The above theorems make straightforward the identification of the ordinary symmetry of the tricrystal pattern and make possible the *a priori* construction of high-symmetry patterns. Also, by following the rules of dissymmetrization, the equivalent patterns (*i.e.* the tricrystal pattern orientation variants) can be determined by performing the coset analysis of $\mathcal{Q}(\lambda)$ [or $\mathcal{Q}(\mu) \equiv \mathcal{Q}(\varepsilon)$] with respect to $\mathcal{Q}(p)_{(\text{tricrystal})}$. Tricrystal pattern orientation variants are the analogues of dichromatic pattern orientation variants in the bicrystal case. In addition, by performing a coset analysis of $\mathcal{Q}(p)_1$ with respect to $\mathcal{Q}(p)_{(\text{tricrystal})}$, we can find all equivalent tricrystal patterns that can be constructed by keeping one dichromatic pattern [*i.e.* that corresponding to $\mathcal{Q}(p)_1$] invariant. The coexistence of tricrystals belonging to equivalent tricrystal patterns is possible along nodes of TJ lines. This will be illustrated in §4.1.1.

In the case of VCTs, we see that the total number of distinct tricrystal patterns that can be created by the pairwise combination of dichromatic pattern orientation variants [given by (1)] is $\varphi = \sum_{i=1}^{n-1} i$, where n is the number of such variants. However, not all of them are equivalent by symmetry. The actual number of the latter is given by $[i]/2$, where $[i]$ is the index of $\mathcal{Q}(p)_{(\text{tricrystal})}$ in $\mathcal{Q}(\lambda)$ (the denominator is introduced because each dichromatic pattern orientation variant is considered twice). For example, consider the case of $\mathcal{Q}(p)_1 \equiv 4/m$ when $\mathcal{Q}(\lambda) \equiv \mathcal{Q}(\mu) \equiv \mathcal{Q}(\varepsilon) \equiv m\bar{3}m$. The number of dichromatic pattern orientation variants is $n = 6$. So, $\varphi = 15$ tricrystal patterns can be created. However, only $[i]/2 = 3$ of them have symmetry $\mathcal{Q}(p)_{(\text{tricrystal})} \equiv 4/m$ (*i.e.* equivalent tricrystal patterns constructed around each of the three $\langle 001 \rangle$ directions). The other 12 have symmetry $\bar{1}$.

2.4. Colour symmetry of the holosymmetric tricrystal pattern

Once the ordinary symmetry of the tricrystal's embracing group has been obtained, we need to consider the conditions for the existence of colour symmetry in the composite. This will reveal any equivalences between the participating crystals (although at this stage the equivalences are only between the lattices). For this purpose, we need to find the symmetrizing group S . As

mentioned earlier, there are two kinds of symmetry operations that can belong to S , *i.e.* mixed or colour exchange operations. The former are operations relating two of the three crystals while leaving the third invariant. The latter are operations causing a cyclic permutation of the crystals.

It is obvious that the mixed operations can only be selected among the antisymmetry operations of $\mathcal{G}'(p)_1$, $\mathcal{G}'(p)_2$, $\mathcal{G}'(p)_3$. However, all the antisymmetry operations of the bicrystal embracing groups may not belong to the tricrystal's embracing group. This has interesting consequences from the point of view of predicting equivalent tricrystals as will be seen later. At this point, it is simply recalled that any suppression of symmetry (ordinary or colour) when going from one dissymmetrization stage to the next has to lead to a number of equivalent composites that are related by this symmetry.

Three cases can be distinguished in which colour symmetry may be present in the tricrystal pattern.

(a) *The mixed symmetry case.* The symmetrizing group S contains two operations, *i.e.* the identity and one second-order mixed operation involving a permutation of the form $\begin{pmatrix} \lambda & \mu & \varepsilon \\ \lambda & \varepsilon & \mu \end{pmatrix}$. Obviously, the given permutation can only be obtained by a second-order antisymmetry operation of $\mathcal{G}'(p)_3$ that, when referred to the coordinate system of the white crystal, becomes a second-order ordinary operation of (λ) . So, S is isomorphic to the matrix group $\{I, \mathcal{W}'_3\}$ where $I = (I, \mathbf{o})$ and \mathcal{W}'_3 is the matrix representative of the mixed symmetry operation. For this case, the following theorem can be proven:

Theorem 4: Mixed symmetry in a tricrystal pattern is possible only for VCTs and occurs if, and only if, there exists a second-order VGE.

The above theorem is proven as follows: When a symmetry operation of the white crystal is referred to the coordinate system of the black crystal, we obtain the matrix C_{ok} of (3). If C_{ok} expresses a transformation between the black and the red crystals, a VCT is obtained according to (2). It is also obvious that the symmetry operation of the white crystal cannot belong to $\mathcal{Q}(p)_1$, otherwise it is impossible to obtain a tricrystal. Hence, it must be a VGE. If $\mathcal{W}(\lambda)_{v_x}$ is the representative of this operation of (λ) , then C_{ok} always represents an antisymmetry operation of $\mathcal{G}'(p)_3$ if the VGE is a second-order operation. This will be proven in §3. On the other hand, if there is no VGE that is of second order, an appropriate symmetrizing group S can never be found.

Mixed symmetry operations can occur in either homophase or heterophase VCTs but not in any other type of tricrystal. In the isomorphic to S matrix group, $\mathcal{W}'_3 = C_{ok}$ or, if referred to the white coordinate frame, $\mathcal{W}'_3 = \mathcal{W}(\lambda)_{v_x}$. Mixed symmetry cases are discussed in §§4.1 and 4.2.

(b) *The colour-exchange symmetry case.* The symmetrizing group S is a three-colour cyclic group isomorphic

to $3^{(3)}$, involving the crystal permutations

$$\left\{ \begin{pmatrix} \lambda & \mu & \varepsilon \\ \lambda & \mu & \varepsilon \end{pmatrix}, \begin{pmatrix} \lambda & \mu & \varepsilon \\ \varepsilon & \lambda & \mu \end{pmatrix}, \begin{pmatrix} \lambda & \mu & \varepsilon \\ \mu & \varepsilon & \lambda \end{pmatrix} \right\}.$$

In this case, S is isomorphic to the matrix group $\{\mathcal{I}, \mathcal{W}'', \mathcal{W}''^{-1}\}$, where double-primed symbols correspond to colour-exchange operations. All three components can be interrelated only if the composite is homogeneous, *i.e.* in homophase tricrystals. In a homophase composite, the transformations \mathcal{P}_i do not express strain but only rotation. Also, it is obvious that, when colour-exchange symmetry exists, all three dichromatic patterns have the same symmetry. It is evident that colour-exchange symmetry exists in a tricrystal pattern if among the set of equivalent descriptions of the transformation \mathcal{P}_{o1} there exists one corresponding to a 120° rotation and, at the same time, among the set of equivalent descriptions of the transformation \mathcal{P}_{o2} there exists one corresponding to a 120° rotation around the same axis but in the opposite sense. If the equivalent to \mathcal{P}_{o1} transformation is represented say by $\mathcal{P}_{o1}\mathcal{W}(\mu)_q$ and the equivalent to \mathcal{P}_{o2} by $\mathcal{P}_{o2}\mathcal{W}(\varepsilon)_p$, we have $\mathcal{W}'' = \mathcal{P}_{o1}\mathcal{W}(\mu)_q$ and $\mathcal{W}''^{-1} = \mathcal{P}_{o2}\mathcal{W}(\varepsilon)_p$. Then, according to (2), among the equivalent descriptions of \mathcal{P}_{o3} there is one corresponding to \mathcal{W}'' .

(c) *The high-colour-symmetry case.* The symmetrizing group S is isomorphic to $(3^{(3)}2^{(2)})^{(6)}$, or $(3^{(3)}m^{(2)})^{(6)}$, involving the crystal permutations

$$\left\{ \begin{pmatrix} \lambda & \mu & \varepsilon \\ \lambda & \mu & \varepsilon \end{pmatrix}, \begin{pmatrix} \lambda & \mu & \varepsilon \\ \mu & \lambda & \varepsilon \end{pmatrix}, \begin{pmatrix} \lambda & \mu & \varepsilon \\ \varepsilon & \mu & \lambda \end{pmatrix}, \right. \\ \left. \begin{pmatrix} \lambda & \mu & \varepsilon \\ \lambda & \varepsilon & \mu \end{pmatrix}, \begin{pmatrix} \lambda & \mu & \varepsilon \\ \varepsilon & \lambda & \mu \end{pmatrix}, \begin{pmatrix} \lambda & \mu & \varepsilon \\ \mu & \varepsilon & \lambda \end{pmatrix} \right\}.$$

Such symmetrization is a combination of the previous two cases where there are both mixed operations and 120° colour-exchange operations. Obviously, this case can occur only in homophase VCTs. The symmetrizing group S is isomorphic to the matrix group $\{\mathcal{I}, \mathcal{W}'_1, \mathcal{W}'_2, \mathcal{W}'_3, \mathcal{W}''', \mathcal{W}'''^{-1}\}$. The second-order mixed operations of S are chosen from the set of antisymmetry operations of the three embracing groups $\mathcal{G}'(p)_1, \mathcal{G}'(p)_2, \mathcal{G}'(p)_3$. The appropriate theorem for this case is

Theorem 5: High-colour symmetry will exist in the tricrystal pattern of a homophase VCT if, and only if, among the set of equivalent descriptions of the transformation \mathcal{P}_{o1} there exists one corresponding to a 120° rotation, represented say by $\mathcal{P}_{o1}\mathcal{W}(\mu)_q$ and, at the same time, there exists a second-order VGE [represented by $\mathcal{W}(\lambda)_{v_x}$] such that

$$\mathcal{W}(\lambda)_{v_x}(\mathcal{P}_{o1}\mathcal{W}(\mu)_q)\mathcal{W}(\lambda)_{v_x}^{-1} = (\mathcal{P}_{o1}\mathcal{W}(\mu)_q)^{-1}.$$

The above expression gives the representative of the 120° rotation among the set of equivalent descriptions of the transformation \mathcal{P}_{o2} [note that since we have a

homophase TJ, there always exists a symmetry operation of the red crystal such that $\mathcal{W}(\varepsilon)_w = \mathcal{W}(\lambda)_{v_x}$]. Then, when expressed to the white coordinate frame, $\mathcal{W}'_3 = \mathcal{W}(\lambda)_{v_x}$ and $\mathcal{W}'' = \mathcal{P}_{o1}\mathcal{W}(\mu)_q$. The matrices $\mathcal{W}'_1, \mathcal{W}'_2$ can be found from the multiplication table of the isomorphic to S matrix group. Also, using this table, theorem 5 can be proved. It can be seen that, if the above expression is valid, the group S always exists for the given operations. On the other hand, if this expression is not valid, the group S can only exist if $\mathcal{W}(\lambda)_{v_x}$ does not represent a VGE, which is against our initial assumption for the occurrence of mixed symmetry.

The geometric element of the VGE must be a 180° axis perpendicular to the 120° axis or a mirror containing the 120° axis. Also, high colour symmetry can easily be identified if a second-order VGE can be found for which the corresponding geometric element has an angle of 60° to the element corresponding to one of the second-order antisymmetry operations of $\mathcal{G}'(p)_1$ [or $\mathcal{G}'(p)_2$]. A high-colour-symmetry case is given in §4.3.

Using the above rules, the colour group $\mathcal{G}'(p)_{(\text{tricrystal})}$, of the tricrystal pattern can be found. As mentioned earlier, the suppression of colour symmetry can give rise to a second class of equivalent tricrystal patterns, *i.e.* patterns related by antisymmetry operations of $\mathcal{G}'(p)_1, \mathcal{G}'(p)_2$ or $\mathcal{G}'(p)_3$, which do not belong to $\mathcal{G}'(p)_{(\text{tricrystal})}$. This situation is analogous to that of antisymmetry-related bicrystals (Pond, 1989) and can occur only in homophase tricrystals. For example, consider a VCT in which an antisymmetry operation of $\mathcal{G}'(p)_1$ is not present in $\mathcal{G}'(p)_{(\text{tricrystal})}$. Then this operation relates two tricrystal patterns obtained using the same VGE, except that, in the first, the VGE is an operation of the white crystal while, in the second, the VGE is an operation of the black crystal. Similarly, an antisymmetry operation of $\mathcal{G}'(p)_2$ relates two tricrystal patterns where, in the first, the VGE is in the white and, in the second, in the red crystal. An analysis using such variants is given in §4.1.2.

2.5. Symmetry of the tricrystal complex

After the embracing group of the tricrystal has been found, the second dissymmetrization step must follow in the procedure of obtaining the symmetry of a tricrystal. This involves the introduction of the space groups $\mathcal{G}(\lambda)^s$ and $\mathcal{G}(\mu)^s \equiv \mathcal{G}(\varepsilon)^s$ of the respective crystal structures, as well as the introduction of the rigid-body translations of the three bicrystals [*i.e.* we consider the transformations $\mathcal{P}_1 = (\mathbf{P}_1, \mathbf{p}_1), \mathcal{P}_2 = (\mathbf{P}_2, \mathbf{p}_2), \mathcal{P}_3 = (\mathbf{P}_3, \mathbf{p}_3)$]. The three-dimensional composite formed by the interpenetration of the three crystals will be termed the tricrystal complex. Any ordinary operation of $\mathcal{G}(p)_{(\text{tricrystal})}$ that is not common in the groups of all three dichromatic complexes is suppressed so that $\mathcal{G}(c)_{(\text{tricrystal})} \subseteq \mathcal{G}(p)_{(\text{tricrystal})}$. As the ordinary symmetry is reduced, the colour symmetry must be reduced accordingly. The

dissymmetrization at the complex level gives us information on equivalent tricrystals and TJ defects introduced due to the suppression of symmetry because of the material's non-symmorphic and/or non-holosym-metric structure.

Let us assume initially a tricrystal complex with no rigid-body translations between the three crystals. Such a complex will be termed holosymmetric since it exhibits the highest possible symmetry for a complex. Then, we have $\mathcal{G}(c)_{(\text{tricrystal})} \equiv \mathcal{G}(c)_1 \cap \mathcal{G}(c)_2$. For VCTs, this relation is written $\mathcal{G}(c)_{(\text{tricrystal})} \equiv \mathcal{G}(c)_1 \cap W(\lambda)_v \mathcal{G}(c)_1 W(\lambda)_v^{-1}$, where now the VGE $W(\lambda)_v$ may be a non-symmorphic operation, *i.e.* the matrix representative can be in general $\mathcal{W}(\lambda)_v = (\mathbf{W}(\lambda)_v, \mathbf{w}(\lambda)_v)$.

The rigid-body translations $\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3$ among the three crystals have to be introduced independently from one another (based on experimental observation), since they are due to interfacial energy minimization and may not be related. In other words, it cannot be known before-

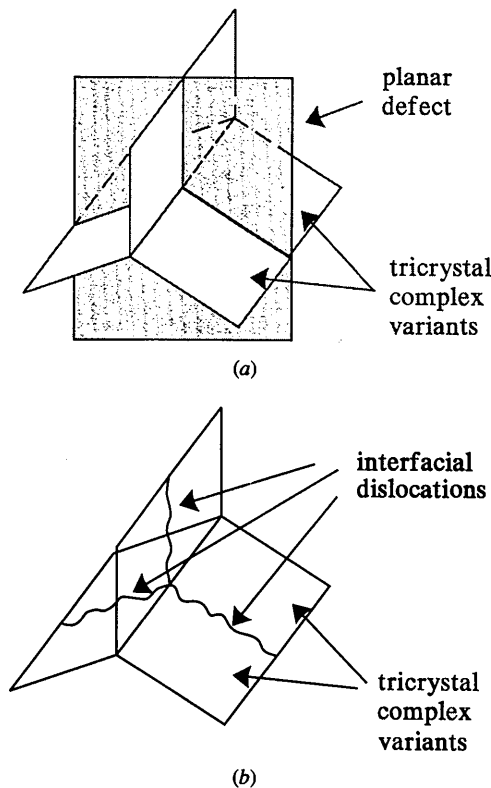


Fig. 2. Schematic drawing showing two possible ways of coexistence of tricrystals originating from variants of one tricrystal complex within the same tricrystal pattern. In (a), the complex variants are separated by a planar defect (shaded) that is introduced in order to compensate for the suppression of symmetry due to a material's non-holosymmetric crystal structure. In (b), the variants coexist through the introduction of frustrated symmetry dislocations placed in order to compensate for the suppression of symmetry due to the material's non-symmorphic structure. The dislocations must form a node on the TJ line.

hand whether mutual accommodation of the rigid-body translations exists. It may be possible to have a partial dislocation along the TJ line which accommodates incompatible rigid-body translations. By using transformations of the form $\mathcal{P}_i = (\mathbf{P}_i, \mathbf{p}_i)$, further dissymmetrization may be obtained. Then, an operation that is suppressed in any of the three dichromatic complexes is also suppressed in the tricrystal complex.

Through dissymmetrization, one can identify all tricrystal complex variants within one tricrystal pattern by performing the coset analysis of $\mathcal{G}'(p)_{(\text{tricrystal})}$ with respect to $\mathcal{G}(c)_{(\text{tricrystal})}$. In a polycrystal, all or some of these variants may be found and their coexistence is possible. For example, in a non-holosymmetric crystal, if two tricrystal complex variants coexist, a planar defect (such as an antiphase boundary, inversion domain boundary, twin or translation-twin) must be introduced between them (Fig. 2a). The character of the defect is defined by the suppressed operation relating the variants. Also, in the case of suppression of non-symmorphic symmetry, two tricrystal complex variants will coexist *via* frustrated symmetry interfacial dislocations (Pond, 1989) forming a node (Fig. 2b). A combination of the cases of Figs. 2(a) and (b) is also possible. Note that a suppressed operation relates tricrystal variants only if it is acted upon all three crystal components. By combining tricrystal pattern variants with tricrystal complex variants, one can enumerate the ways for constructing a given TJ and characterize the geometrically necessary defects between equivalent tricrystals.

For the case of VCTs, the complex level is additionally important for the *a priori* characterization of the geometrically necessary defect content along the junction line. This is defined by the VGE. If the VGE is a non-symmorphic operation, then a frustrated symmetry partial dislocation will exist between two interfaces obtained

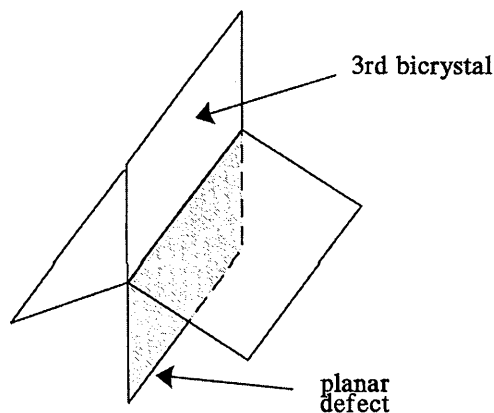


Fig. 3. Schematic drawing of a VCT where the two orientation variant bicrystals are related by a VGE that is present in the lattice, but not in the actual crystal structure because the material is non-holosymmetric. Then, in order to conserve the interfacial structure when going from bicrystal 1 to bicrystal 2, a planar defect is geometrically necessary (shaded).

from orientation variants of one dichromatic pattern, providing the translation part of the operation is not invariant with respect to the transformation \mathbf{P}_1 (Pond, 1989). The existence of the partial dislocation is imperative unless the distortion can be absorbed by the third bicrystal [in which case, the third bicrystal is a rotation-translation boundary (Guymont, Gratias, Portier & Fayard, 1976)]. In addition, if the white crystal is non-holosymmetric, the VGE may be an operation leaving invariant the lattice but not the structure (e.g. as in ordering). Then a planar defect must emanate from the TJ line (Fig. 3). If analogous suppression of symmetry occurs in $\mathfrak{G}(\mu)^s \equiv \mathfrak{G}(\varepsilon)^s$, the planar defect character may continue along the third boundary. This will be illustrated in §4.1.1.

2.6. Symmetry of the tricrystal

We have reached the final step in the dissymmetrization procedure, where the symmetry of the tricrystal itself is obtained. The tricrystal is obtained from the tricrystal complex in a manner analogous to the bicrystal case. This means that, in an imaginary process, the three interfaces are introduced in the complex. Then, one crystal is left between each pair of interfaces and the other two are discarded. So we now have, instead of the one invariant plane condition (for the case of one bicrystal), the conditions for (i) an invariant line (i.e. the TJ line), and (ii) the invariance of three planes (for the three bicrystals). If the TJ line is a rational direction and the translation operation along it is coincident, then the tricrystal symmetry is described by a rod group. If the direction is irrational, then the tricrystal group is a rosette group.

Each participating bicrystal is characterized by two-dimensional symmetry. The only permissible operations in a bicrystal group are ordinary operations corresponding to rotation axes and mirrors perpendicular to the interface, as well as antisymmetry operations corresponding to $2'$ axes and anti-mirrors parallel to the interface, and $4'$ and $6'$ axes perpendicular to the interface (Pond & Vlachavas, 1983). The anti-inversion $\bar{1}'$ and the antirotoinversion $\bar{3}'$ are not permitted for relative rotations ($\mathbf{R} \neq \mathbf{I}$) between two crystals, as can be seen from the point groups that have been given in §2.1.

Let the interfaces intersect along the common direction \mathbf{r} . Their introduction breaks any ordinary rotation symmetry of the tricrystal complex along \mathbf{r} . So the only ordinary operation that can be common to all three bicrystal groups corresponds to a possible coincident mirror perpendicular to \mathbf{r} . In other words, if such an operation exists in two bicrystal groups, it must also exist in the third. The interfaces themselves can have any orientation, although it is common that antisymmetry mirrors are the energetically preferred orientations.

In addition to an ordinary mirror, a tricrystal may possess colour symmetry. In the mixed symmetry case,

the tricrystal has colour symmetry if the geometric element corresponding to a mixed-symmetry operation is parallel to the interface of the bicrystal for which this operation is an antisymmetry operation and, at the same time, this operation relates the orientations of the other two interfaces. In the colour-exchange symmetry case, the tricrystal has colour symmetry if \mathbf{r} coincides with the 120° colour-exchange axis and at the same time the orientations of the three interfaces are related by the colour-exchange operation. Finally, in the high-colour-symmetry case, the tricrystal can exhibit colour-exchange symmetry or mixed symmetry or both. A tricrystal will have both types of symmetry if \mathbf{r} coincides with the 120° colour axis and the second-order mixed symmetry elements at 120° angle with each other are parallel to the interface planes.

Any operation of $\mathfrak{G}'(c)_{(\text{tricrystal})}$ that does not belong to the tricrystal's group relates tricrystal morphological variants within the same complex. In particular, if such an operation corresponds to a geometric element along the TJ line, it leaves this line invariant and so relates tricrystal variants around it. This will be illustrated in §4.1.

3. The embracing group of the third bicrystal in variant constituted tricrystals

As mentioned in the *Introduction*, the task at hand in this section is to determine the symmetry constraints imposed on the embracing group of bicrystal 3 in VCTs. According to the principle of symmetry superposition, we have $\mathfrak{G}(p)_3 \supseteq \mathfrak{G}(p)_{(\text{tricrystal})}$, i.e. the intersection group may be further symmetrized so that the third embracing group can contain additional symmetry operations. This can only be done by an appropriate symmetrizing group. However, the operations of the symmetrizing group cannot belong to either $\mathfrak{G}(p)_1$ or $\mathfrak{G}(p)_2$ because, according to theorem 1 (or theorem 2), they would then belong to $\mathfrak{G}(p)_{(\text{tricrystal})}$. So they must be chosen from the set $\{\mathfrak{G}(\mu) - \mathfrak{G}(p)_1 - \mathfrak{G}(p)_2\}$ [or, equivalently, $\{\mathfrak{G}(\varepsilon) - \mathfrak{G}(p)_1 - \mathfrak{G}(p)_2\}$]. The matrix representatives of operations in the symmetrizing group must satisfy the condition $\mathcal{W}(\mu)_i = C_{ok} \mathcal{W}(\varepsilon)_j C_{ok}^{-1}$, where $C_{ok} = \mathcal{P}_{o1}^{-1} \mathcal{W}(\lambda)_v \mathcal{P}_{o1}$. For point symmetries in particular, if $\mathbf{r}_i, \mathbf{r}_j$ are the orientation vectors of the corresponding geometric elements, then $\mathbf{W}(\lambda)_v \mathbf{P}_1 \mathbf{r}_j = \mathbf{P}_1 \mathbf{r}_i$.

The dichromatic group $\mathfrak{G}'(p)_3$ is a factor-two supergroup of $\mathfrak{G}(p)_3$. In order to find the antisymmetry operations of $\mathfrak{G}'(p)_3$, we employ the rules of Vlachavas (1984). Then C_{ok} [given in (3)] represents an antisymmetry operation of $\mathfrak{G}'(p)_3$ (i.e. $C_{ok} = \mathcal{W}_3$) if $C_{ok}^2 = \mathcal{P}_{o1}^{-1} \mathcal{W}(\lambda)_v^2 \mathcal{P}_{o1}$ represents an operation of $\mathfrak{G}(p)_3 \subseteq \mathfrak{G}(p)_3$. However, as we have mentioned (theorem 3), whenever the $\mathfrak{G}(p)_{(\text{tricrystal})}$ symmetry is greater than $\bar{1}$, the VGE is of second order. For such a VGE, $C_{ok}^2 = (\mathbf{I}, \mathbf{o})$, i.e. C_{ok} represents an antisymmetry operation of second order. Then, $\mathfrak{G}'(p)_3$ is the extension

of $\mathcal{G}(p)_3$ by a symmetrizing group containing the identity and this second-order antisymmetry operation.

It remains to find the conditions for antisymmetry to exist when the VGE is not of second order [in which case $\mathcal{R}(p)_{(\text{tricrystal})} \equiv \bar{1}$]. Then, C_{ok} corresponds to a 60, 90 or 120° rotation. So, among the equivalent transformations $C_{om} = C_{ok} \mathcal{W}(\varepsilon)_m$, we need to find a second-order one. It is easy to see that C_{om} will be of second order if, and only if, $\mathcal{W}(\varepsilon)_m$ is also of second order (Landau & Lifshitz, 1952). Moreover, the axis or mirror normal corresponding to $\mathcal{W}(\varepsilon)_m$ must be perpendicular to the axis of C_{ok} . If say $\mathcal{W}(\varepsilon)_m$ expresses a 180° rotation, then so does C_{om} . The axis of C_{om} is also perpendicular to the axis of C_{ok} and has an angle to the axis of $\mathcal{W}(\varepsilon)_m$ equal to half the rotation angle of C_{ok} . Similar arguments can be written for the case when $\mathcal{W}(\varepsilon)_m$ corresponds to a mirror if instead of axes the mirror-plane normals are used. We note that, in general, antisymmetry in the third embracing group is possible even if it does not exist in the dichromatic pattern orientation variants (e.g. as in heterophase TJs).

Before closing this section, a case of particular importance must be distinguished. When $\mathcal{R}(p)_1$ is invariant under the VGE (i.e. $\mathcal{R}(p)_1 \equiv \mathcal{W}(\lambda)_{v_x} \mathcal{R}(p)_1 \mathcal{W}(\lambda)_{v_x}^{-1}$), we have $\mathcal{R}(p)_3 \supseteq \mathcal{R}(p)_1 \cap \mathcal{R}(p)_2 \equiv \mathcal{R}(p)_1$. As mentioned in §2.1, this is the most usual case when $\mathcal{R}(p)_{(\text{tricrystal})}$ contains operations other than the identity and the inversion. Then, for one of the transformations \mathcal{P}_{o1} , we can have

$$\mathcal{P}_{o2} = \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1} \mathcal{W}(\varepsilon)_j^{-1} = \mathcal{P}_{o1}^{-1}. \quad (4)$$

Hence, in the two dichromatic pattern orientation variants, the transformations \mathcal{P}_o have opposite senses. This is interesting since now the coordinate transformations of the third dichromatic pattern do not intrinsically depend on the VGE. Indeed, one of the equivalent to C_{ok} transformations is \mathcal{P}_{o1}^{-2} . Then, it is possible that $\mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1}^{-2} = \mathcal{W}(\lambda)_{v_x}$. So, when $\mathcal{R}(\lambda) \equiv \mathcal{R}(\mu) \equiv \mathcal{R}(\varepsilon)$, the VGE can be one of the extending elements in the symmetrization of $\mathcal{R}(p)_1$ to $\mathcal{R}(p)_3$. It can be proven (see Appendix A) that this is possible if, and only if, \mathcal{P}_{o1}^{-2} represents an antisymmetry operation of $\mathcal{G}(p)_3$. So the procedure for obtaining $\mathcal{G}(p)_3$ is simplified.

In order to find the symmetry group $\mathcal{G}(c)_3$ of the dichromatic complex, we need to dissymmetrize $\mathcal{G}(p)_3$ by considering the space group $\mathcal{G}(\mu)^s \equiv \mathcal{G}(\varepsilon)^s$ and the rigid-body translation \mathbf{p}_3 , as described in §2.1. Finally, the two-dimensional bicrystal symmetry is found by introducing the interface orientation.

In concluding this section, we remark that, as has been shown, the symmetry of the third bicrystal in VCTs is significantly constrained by the VGEs relating the orientation variants. This information can be employed to design VCTs in which the third bicrystal will be a desired bicrystal characterized by particular symmetry and antisymmetry operations. This will be demonstrated

in §4.2. The method of Dahmen & Thangarai (1993) can be used to fabricate such a bicrystal. In this method, a VCT is obtained by the coexistence of two variants of an epitaxial interface.

4. Examples

4.1. Variant constituted tricrystals of first-order twins in f.c.c. polycrystals

We first examine the commonly observed $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ VCTs. This system is useful for a first illustration of the dissymmetrization procedure introduced in this work, because its simplicity leads to a direct understanding of the results obtained. Consider such a tricrystal composed of $\Sigma 3$ {111} bicrystals in f.c.c. The f.c.c. structure is chosen in this first approach because it is both symmorphic and holosymmetric [$\mathcal{G}(j) \equiv \mathcal{G}(j)^s \equiv Fm\bar{3}m$, $j = \lambda, \mu, \varepsilon$], which means that there are no complex variants. Also let us make the hypothesis that there are no rigid-body translations involved in the TJ that could complicate the analysis. Then the symmetries for the $\Sigma 3$ are $\mathcal{G}(p)_1 \equiv \mathcal{G}(c)_1 \equiv P6'/m'mm'$ and the bicrystal symmetry is $\mathcal{G}(b)_1 \equiv p6'm2'$ as commonly known. The ordinary symmetries are $\mathcal{G}(p)_1 \equiv \mathcal{G}(c)_1 \equiv P\bar{3}m$ and $\mathcal{G}(b)_1 \equiv p3m$, respectively. For the tricrystal pattern and complex, the ordinary symmetry is $\mathcal{G}(p)_{(\text{tricrystal})} \equiv \mathcal{G}(c)_{(\text{tricrystal})} \equiv C2/m$. The existence of common intersection symmetry greater than 1 is possible because there are second-order VGEs. Also due to these elements, there exists a symmetrizing group $S \equiv 2'$, where the mixed symmetry operation introduces the permutation $\begin{pmatrix} \lambda & \mu & \varepsilon \\ \lambda & \varepsilon & \mu \end{pmatrix}$, i.e. we have a mixed-colour symmetry case. Therefore, $\mathcal{G}(p)_{(\text{tricrystal})} \equiv$

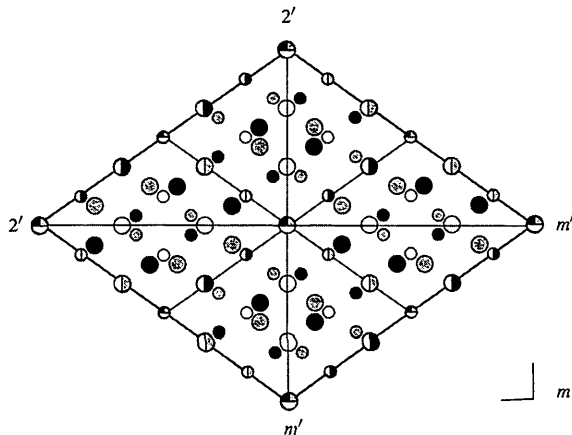


Fig. 4. Tricrystal complex for the $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ tricrystal. The three crystal components are coloured white, black and grey, respectively. The mixed symmetry elements are primed. In this example, these elements relate the black and grey atoms while leaving the white ones invariant. The orientations of the {111} planes where $\Sigma 3$ twins occur are also given. The projection direction is $\langle 011 \rangle$. Two layers of atoms are shown. Large circles denote atoms at zero height. Small circles denote atoms at height $a2^{1/2}/4$ (where a is the lattice parameter).

$\mathcal{G}'(c)_{(\text{tricrystal})} \equiv Cm'mm'$. This is also the symmetry of $\mathcal{G}'(p)_3 \equiv \mathcal{G}(c)_3$ since there is no further symmetrization. The tricrystal complex is given in Fig. 4.

From the above dissymmetrization, we conclude that six tricrystal patterns can be constructed among the four orientation variants of $\bar{3}m$ in $m\bar{3}m$, all of which are crystallographically equivalent. Also, if we keep one dichromatic pattern invariant, three equivalent tricrystal patterns can be constructed, since there are three variants of $\mathcal{Q}(p)_{(\text{tricrystal})}$ in $\mathcal{Q}(p)_1$. However, there are additionally three variants of $\mathcal{Q}(p)_{(\text{tricrystal})}$ related by the antisymmetry operations of $\mathcal{Q}(p)_1$, since these operations do not belong to the embracing group of the tricrystal. Such variants introduce antisymmetry-related tricrystal patterns.

Finally, for a $\{1\bar{2}2\}$ orientation of the $\Sigma 9$ interface, the tricrystal has one-dimensional symmetry described by the rod group $\rho 2'mm'$, where m corresponds to the perpendicular to the TJ-line mirror and the translation vector is $\frac{1}{2}(011)$. Hence, there are two morphological variants of the tricrystal inside the tricrystal complex that are both around the TJ line and are related by a 180° rotation (Fig. 5).

A number of interesting results can arise by considering the coexistence of tricrystal orientation variants. Two cases are given below.

4.1.1. *Coexistence of tricrystal variants in an assembly of octahedral morphology.* All tricrystal variants that are related by ordinary symmetry can be pictured in an octahedral assembly in $\Sigma 3$ orientation with the surrounding matrix, having faces of $\{111\}$ type. This configuration is shown in Fig. 6. $\Sigma 9$ twins emanate from the junctions of the $\Sigma 3$ bicrystals, thus forming 12 VCTs. The tricrystals coexist along the nodes of the octahedron. The $\Sigma 9$ twins may emanate either into the octahedron or into the matrix. Such an assembly would be difficult to observe experimentally and probably is energetically unstable. However, a half-octahedral arrangement of $\Sigma 3$ twins has been observed in epitaxial SiC on (001) Si (Stoemenos, 1994). This is a case of multiple microtwinning and the first-order twins must be related by $\Sigma 9$

microtwins. However, since SiC has the non-holosymmetric sphalerite structure, the octahedron will be polar (Holt, 1988). This means that the polarity of the first-order twins will be inverted in each face unless an inversion domain boundary emanates from each junction. The inversion character is then extended in the $\Sigma 9$ bicrystals. One such TJ is shown in Fig. 7. This figure is

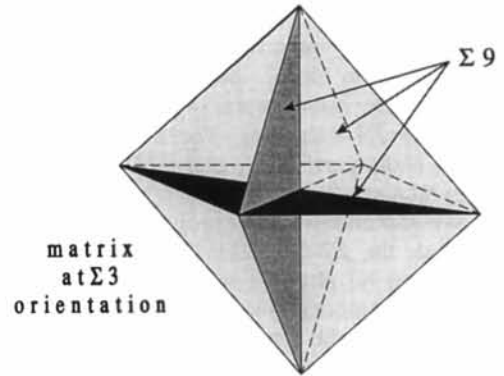


Fig. 6. Hypothetical octahedral assembly of tricrystal variants. The assembly is in $\Sigma 3$ orientation with the surrounding matrix crystal. The interfaces between matrix and inclusion are $\{111\}$ twins. $\Sigma 9$ twins are necessary for the coexistence of the $\Sigma 3$ twins and these have been taken in the drawing to emanate from their junctions into the octahedron (shown by shading). The figure illustrates the coexistence of all variants of the $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ tricrystal that are related by ordinary symmetry.

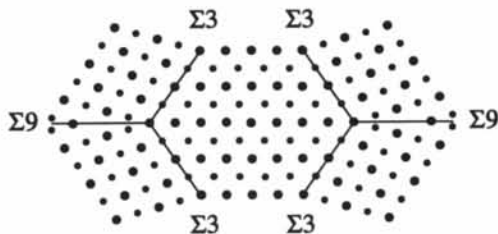


Fig. 5. Equivalent-by-symmetry tricrystals around the same TJ line originating from one tricrystal pattern in the $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ CSL case. The tricrystals are related by the 180° symmetry operation around the TJ line. The projection direction is (011) . Two layers of atoms are shown. Atom heights are as in Fig. 4. In this, as in all following figures, the interfacial structures are shown unrelaxed and no rigid-body translations between the crystals have been considered.

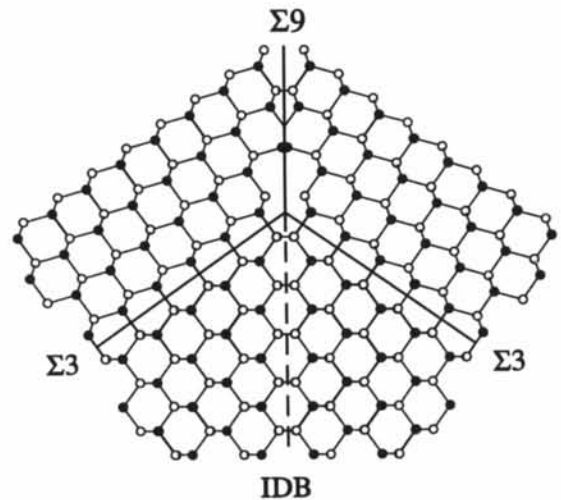


Fig. 7. $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ tricrystal in the sphalerite structure. Colouring of the atoms distinguishes the two atom types composing the sphalerite crystal basis. Bonding between the atoms is also shown for clarity. Since the $\Sigma 3$ bicrystals are related by a VGE that is present in the lattice but not in the crystal structure (non-holosymmetric crystal), an inversion domain boundary (IDB) must emanate from their junction in order to preserve the polarity in the twins (shown dashed). As this is a homophase VCT, atoms of the same type come opposite one another in the $\Sigma 9$ bicrystal, i.e. the inversion domain character is extended in the $\Sigma 9$. The projection direction is (011) . Two layers of atoms are shown.

in accordance with Fig. 3 and the analysis of §2.5. In general, polycrystals can be modelled using various assemblies of tricrystal variants if we choose the degree to which symmetry constrains the system.

4.1.2. *CSL TJs with multiplicities related by a power law.* Antisymmetry-related $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ tricrystal variants around the same line can be used to construct *a priori* the $\Sigma_i = 3^n$ CSL tricrystals. Such an analysis can be generalized for all $\Sigma_i = f^n$ CSL tricrystals (where f, n are integers).

We will consider only the simple case of $\Sigma 3$ - $\Sigma 9$ - $\Sigma 27a$ tricrystals in detail. Such a tricrystal can be considered as the combination of two antisymmetry-related $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ tricrystals. The model is shown in Fig. 8. The components ($\mu 1$) and ($\mu 2$) are in a $\Sigma 27a$ orientation relation. By superimposing the two tricrystal patterns, we can choose the appropriate crystal components to create the $\Sigma 3$ - $\Sigma 9$ - $\Sigma 27a$ tricrystal. It should be emphasized that no physical meaning can be currently ascribed to this model since it is only a geometric construction. Using the sequence of coordinate transformations, we can study the properties of this new composite. It is easy to conclude that the common ordinary symmetry is the same as in the VCTs. Also, this tricrystal has no colour symmetry. We can combine three VCTs to construct a $\Sigma 3$ - $\Sigma 27$ - $\Sigma 81$ tricrystal and so on. In general, it can be said that antisymmetry appears to play an important role for the modelling of interfacial connectivity through TJs.

4.2. Fabrication of a particular bicrystal by epitaxial deposition

This example serves as an illustration of the methodology developed in §3 for obtaining the symmetry of the third bicrystal in VCTs. It has been shown that epitaxy can be used for the fabrication of bicrystals of particular orientation in the epitaxial deposit (Dahmen & Thangarai, 1993). It is known that the relative rotation

Table 2. *Appropriate substrate orientations and relative rotations so that a $\Sigma 3$ twin can emanate in the epitaxial deposit for the case of cubic-on-cubic material epitaxy*

The relative rotations between substrate and epitaxial crystal are given in the form of sets of parallel axes (instead of axis/angle pairs) in accordance with common practice. The corresponding VGEs and tricrystal symmetries are also given.

Second-order VGEs	Suitable substrate orientations	Relative rotations (R_1) between substrate and epitaxial crystal	Tricrystal pattern symmetry
$\langle 110 \rangle$	$\langle 110 \rangle, \{h\bar{h}k\}$	$\langle 1\bar{1}0 \rangle_e // \langle 1\bar{1}0 \rangle_s, \langle 111 \rangle_e // \langle 110 \rangle_s, \langle 11\bar{2} \rangle_e // \langle 001 \rangle_s$	$2/m$
$\langle 001 \rangle$	$\langle 001 \rangle, \{hk0\}$	$\langle 1\bar{1}0 \rangle_e // \langle 1\bar{1}0 \rangle_s, \langle 112 \rangle_e // \langle 110 \rangle_s, \langle 111 \rangle_e // \langle 001 \rangle_s$	$(m'm'm')$
$\langle 110 \rangle$	$\langle 110 \rangle, \{h\bar{h}k\}$	$\langle 112 \rangle_e // \langle 112 \rangle_s, \langle 111 \rangle_e // \langle 1\bar{1}0 \rangle_s, \langle 110 \rangle_e // \langle 111 \rangle_s$	$\bar{1}(2'/m')$
$\langle 110 \rangle$	$\langle 110 \rangle, \{h\bar{h}k\}$	$\langle 111 \rangle_e // \langle 111 \rangle_s, \langle 112 \rangle_e // \langle 110 \rangle_s, \langle 110 \rangle_e // \langle 112 \rangle_s$	$\bar{3}(3m')$
$\langle 110 \rangle$	$\langle 110 \rangle, \{h\bar{h}k\}$	$\langle 1\bar{1}3 \rangle_e // \langle 1\bar{1}3 \rangle_s, \langle 211 \rangle_e // \langle 110 \rangle_s, \langle 471 \rangle_e // \langle 332 \rangle_s$	$\bar{1}(2'/m')$
$\langle 001 \rangle$	$\langle 001 \rangle, \{0hk\}$	$\langle 120 \rangle_e // \langle 120 \rangle_s, \langle 211 \rangle_e // \langle 001 \rangle_s, \langle 215 \rangle_e // \langle 210 \rangle_s$	$\bar{1}(2'/m')$

between the substrate and the epitaxial deposit depends on a number of factors such as the minimization of the lattice mismatch. So it is useful to know *a priori* which relative rotations and substrate orientations can be used to construct a given bicrystal. Then, appropriate substrate materials can be chosen. This is illustrated in the following problem.

In cubic-on-cubic material epitaxy, we seek to determine the appropriate substrates and relative orientations so that a $\Sigma 3$ twin can appear in the epitaxial deposit emanating from the heterophase boundary. In order to do so, we need to determine the rotations R_1 for which the composite operation is one of the second-order antisymmetry operations of $\Sigma 3$, *i.e.* along $\{111\}$, $\{1\bar{2}1\}$, $\{1\bar{2}1\}$ or $\{112\}$. This is straightforward in this case, since we find that the composite operation can only be of the form of (3), where the VGE is one of the cubic structure's second-order operations. Using these elements, the relative rotations are determined. Then the substrate orientations are found by noting that the geometric element of the VGE must be perpendicular to the substrate so that it conserves its orientation. The results are summarized in Table 2. From this table, we observe that in all cases we have, in the third bicrystal, further symmetrization from the common symmetry of the tricrystal. Additionally, we observe that, for the 90° $\langle 112 \rangle$ and the 30° $\langle 111 \rangle$ rotations, the VGEs are also symmetrizing elements of $\mathcal{Q}(p)_{(\text{tricrystal})}$, *i.e.* we are in the special case of §3.

4.3. Variant constituted tricrystal exhibiting high colour symmetry

For the sake of completeness, we give here a theoretical example of occurrence of high colour symmetry for the $\Sigma 9$ orientation in f.c.c. ($Fm\bar{3}m$). For this CSL, we observe the existence of a second-order VGE corresponding to a geometric element of $\{10\bar{1}\}$ orientation that has an angle of 60° to the antisymmetry elements of $\{411\}$ orientation. So, according to theorem 5, we can construct a high-symmetry VCT where the 120° colour-exchange axis is along $\langle 15\bar{1} \rangle$. The

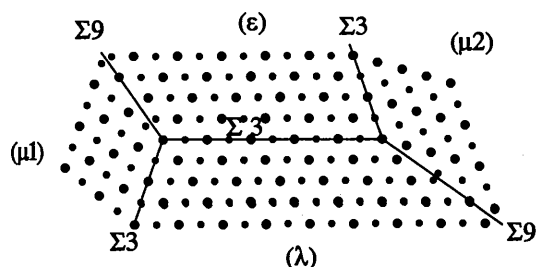


Fig. 8. Geometrical model showing the coexistence of two antisymmetry-related $\Sigma 3$ - $\Sigma 3$ - $\Sigma 9$ tricrystals in f.c.c. Antisymmetry related means, in this case, that the two holosymmetric tricrystal patterns are variants related by an antisymmetry operation of the (λ) - (ϵ) dichromatic pattern that does not belong to the tricrystal embracing group. They are related by the $\{111\}$ anti-mirror of the common $\Sigma 3$ bicrystal. The crystal components ($\mu 1$) and ($\mu 2$) are in a $\Sigma 27a$ orientation. The projection direction is $\langle 011 \rangle$. Two layers of atoms are shown. Atom heights are as in Fig. 4.

embracing group of this composite is $\mathcal{G}(p)_{(\text{tricrystal})} \equiv \mathcal{G}(c)_{(\text{tricrystal})} \equiv \rho(3^{(3)}2^{(2)})^{(6)} \equiv \rho I \otimes (3^{(3)}2^{(2)})^{(6)}$. The symmetry of the tricrystal is $\rho(3^{(3)}m^{(2)})^{(6)}$ [the superscripts denote the colour permutations (Shubnikov & Koptsik, 1974)]. Such a TJ is shown in Fig. 9. In this composite, the well known CSL multiplication rule is not valid (this rule would have predicted a $\Sigma 9$ - $\Sigma 9$ - $\Sigma 81$ combination). Out of the $\varphi = 66$ possible tricrystal patterns constructed using $\Sigma 9$ bicrystal orientation variants, only 12 can give $\Sigma 9$ - $\Sigma 9$ - $\Sigma 9$ tricrystals.

5. Conclusions

Whenever, in a high-symmetry medium such as a crystal, an effect is observed where the symmetry of the medium is reduced, an appropriate dissymmetrization should be sought. Such an effect can be a defect, an interface or a TJ. Dissymmetry leads to a number of equivalent effects that can occur in the medium and so it must be distinguished from asymmetry.

The above fundamental principle has prompted the investigation of the crystallography of TJs that has been presented in this work. The TJ has been treated as a three-component composite but the analysis has followed the same steps as those used in bicrystallography. The ordinary symmetry of a tricrystal has been defined by the addition of the dissymmetries of the participating bicrystals, in accordance with the basic principle for the superposition of symmetries. This symmetry expresses the invariances of any physical property in the composite. In addition, colour elements can be introduced that interrelate the components. Such elements express the equivalences in the system. Special attention has been paid to the information that can be obtained *a priori* and on the consequences of dissymmetrization on polycrystal topology and TJ defects.

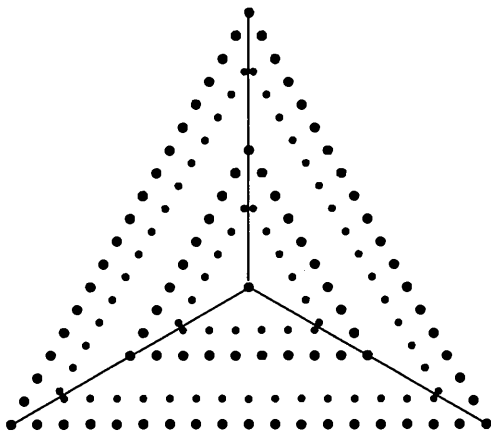


Fig. 9. $\Sigma 9$ - $\Sigma 9$ - $\Sigma 9$ VCT showing high colour symmetry. The projection direction is $\langle 1\bar{5}1 \rangle$. A 120° colour-exchange axis is parallel to the TJ line. Two layers of atoms are shown. Large circles denote atoms at zero height. Small circles denote atoms at height $a/3 \times 3^{1/2}$.

Particular emphasis has been placed on VCTs since these are the most symmetrical of all tricrystals and certainly the most important ones. Their significance is easily understood if one considers their frequent occurrence in processes such as crystal growth, precipitation processes and phase transformations in general *etc.* It has been shown that, in a VCT, the VGEs are crucial in determining the symmetry of the composite.

The analysis gives the experimentalist the opportunity to investigate the relationships of the three crystals. This could not be done easily in a graphic manner, since the sense of three-dimensional space would be lost. In addition, by using the given information, one could design the epitaxial deposition on special substrates so that particular bicrystals exist in the epitaxial deposit. So the analysis is useful for interface engineering. This is why the work has also focused on the symmetry of the third bicrystal in VCTs.

A number of appropriate examples have been given to illustrate the application and usefulness of the analysis. In §4.1, it was shown how the dissymmetrization procedure can provide a useful tool for predicting cases of interface connectivity. In this example, it was proposed that certain tricrystals, such as tricrystals composed of CSL bicrystals related by a power law, can be modelled using broken antisymmetry in a polycrystal. §4.2 gives a case of interface design by epitaxy. Finally, §4.3 illustrated a case of a VCT exhibiting three-colour symmetry.

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APPENDIX A

Let $\mathcal{Q}(\lambda) \equiv \mathcal{Q}(\mu) \equiv \mathcal{Q}(\varepsilon)$ (*i.e.* there is a black as well as a red operation such that, when referred to their respective coordinate frames, we have $\mathcal{W}(\mu)_i \equiv \mathcal{W}(\varepsilon)_j \equiv \mathcal{W}(\lambda)_{v_x}$ for the matrix representatives). Then, (4) can always be re-written as

$$\mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1} \mathcal{W}(\lambda)_{v_x}^{-1} = \mathcal{P}_{o1}^{-1}. \quad (5)$$

For (5) to be valid, the VGE can only be a second-order symmetry operation leaving invariant the common relative rotation axis corresponding to \mathcal{P}_{o1} . From (5), we have

$$\begin{aligned} & (\mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1} \mathcal{W}(\lambda)_{v_x}^{-1}) (\mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1} \mathcal{W}(\lambda)_{v_x}^{-1}) \\ & = \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x}^{-1} = \mathcal{P}_{o1}^{-2}. \end{aligned} \quad (6)$$

Let $\mathcal{W}(\lambda)_{v_x}$ represent a symmetry operation of $\mathcal{Q}(p)_3$. Then,

$$\mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1}^{-2} = \mathcal{W}_i, \quad (7)$$

where \mathcal{W}_i represents a symmetry operation of $\mathcal{Q}(p)_3$. From (6) and (7), we have

$$\mathcal{W}_i \mathcal{P}_{o1}^2 = \mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x} = \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1}^{-2} \Rightarrow (\mathcal{P}_{o1}^2)^2 = \mathcal{W}_i^{-1} \mathcal{W}(\lambda)_{v_x},$$

i.e. \mathcal{P}_{o1}^2 represents an antisymmetry operation of $\mathcal{Q}(p)_3$ according to the rules of Vlachavas (1984).

Now let \mathcal{P}_{o1}^2 represent an antisymmetry operation. Then, $(\mathcal{P}_{o1}^2)^2$ represents an operation of $\mathcal{Q}(p)_3 \subseteq \mathcal{Q}(\mu) \equiv \mathcal{Q}(\varepsilon)$. Then also $(\mathcal{P}_{o1}^2)^2 \mathcal{W}(\lambda)_{v_x} \mathcal{W}(\lambda)_{v_x}^{-1}$ represents an operation of $\mathcal{Q}(\mu) \equiv \mathcal{Q}(\varepsilon)$. But, using (6), we can write

$$\mathcal{P}_{o1}^2 \mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x} \mathcal{W}(\lambda)_{v_x}^{-1} = \mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1}^{-2} \mathcal{W}(\lambda)_{v_x}^{-1}.$$

However, since $\mathcal{W}(\lambda)_{v_x}$ represents an operation of $\mathcal{Q}(\mu) \equiv \mathcal{Q}(\varepsilon)$ so also does $\mathcal{P}_{o1}^2 \mathcal{W}(\lambda)_{v_x} \mathcal{P}_{o1}^{-2}$, i.e. $\mathcal{W}(\lambda)_{v_x}$ represents a coincident operation and hence an element of group $\mathcal{Q}(p)_3$.

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