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Plesiotwins versus diperiodic twins

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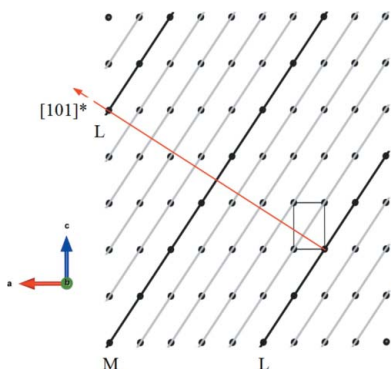
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Plesiotwins and diperiodic twins have in common the fact of being characterized by a low degree of lattice restoration. Plesiotwins differ from twins by the fact that the relative orientation of the individuals is obtained by a non-crystallographic rotation about the normal to the composition plane, whereas for twins this rotation is crystallographic, apart from possible small deviations coming from metric pseudosymmetries. In the case of plesiotwins, the low degree of lattice restoration comes from a large coincidence site lattice (CSL) in the composition plane. Diperiodic twins, instead, have a small CSL in the composition plane but the second plane of the same family contributing to the overall lattice restoration is too far away from the composition plane to be considered significant. It is shown that plesiotwins can occur as reflection twins if the composition plane is not parallel to the twin plane, and as rotation twins in the case of parallel hemitropy. Diperiodic twins can in principle occur in any category, but either the metric conditions to obtain a diperiodic twin are actually in contrast with the metric pseudosymmetry required for twinning or the result is actually a hybrid twin. This justifies why no confirmed examples of diperiodic twins are known to date.

1. Introduction

A high degree of lattice restoration is considered a necessary condition for the formation of twins. Some kinds of oriented associations of the same crystalline phase may however violate this requirement. One example is diperiodic twins, a potential category of twins predicted long ago, which however has no confirmed examples to date. Another example is that of plesiotwins, a special category of oriented crystal association based on a coincidence site lattice (CSL; Grimmer, 1976) in the composition plane, which typically occurs in crystals of flat tabular morphology (Nespolo, Ferraris *et al.*, 1999). The aim of this article is to provide precise criteria to differentiate between twins and plesiotwins and the likely reasons why we do not have examples of diperiodic twins. For that, we need to remind the reader of a few basic facts about twinning. The definitions used below have been reviewed several times and are easily accessible in the literature: see, for example, our recent review (Nespolo, 2015*a*) and references therein. Below is a minimal set of definitions indispensable to follow our derivation.

(a) When a lattice node of an individual crystal is mapped, exactly or approximately, on a lattice node of another individual, that node is said to be *restored* by the twin operation. The whole set of restored nodes defines the *twin lattice*, which may coincide with the lattice of the individual (twinning by merohedry, or pseudo-merohedry if the restoration is approximate) or be a sublattice of it (twinning by reticular merohedry, or reticular pseudo-merohedry).



(b) Depending on whether the lattice (or a sublattice) is restored by the twin operation exactly or approximately, one speaks of twin lattice symmetry (TLS) or twin lattice quasi-symmetry (TLQS) (Donnay & Donnay, 1974).

(c) The inverse of the fraction of lattice nodes (quasi-) restored by the twin operation is called the *twin index*.

(d) The angular deviation between the twin plane (axis) and the lattice direction (plane) quasi-perpendicular to it is called the *obliquity*.

(e) In the case of zero-obliquity TLQS, which occurs when the twin operation is of order higher than two and the twin lattice has a metric pseudosymmetry in the plane perpendicular to the twin axis, the linear divergence from exact overlap of lattice nodes is measured by the distance between lattice nodes quasi-restored by the twin operation that are closest to the origin; this linear divergence is called the *twin misfit* (Nespolo & Ferraris, 2007).

Twins can be classified according to different criteria. For our purpose, two classifications are particularly useful.

(i) *Geometric classification*. On the basis of the nature of the twin operation, twins are classified as inversion twins, reflection twins and rotation twins. For manifold twins [in which at least one of the twin operations has order $n > 2$ (Nespolo, 2004)], the possibility of roto-inversion twins has to be considered as well.

(ii) *Genetic classification*. From the genetic viewpoint, twins are classified in three categories: transformation twins (occurring as the result of a phase transition with a group-subgroup relation), mechanical (deformation, secondary) twins (due to the action of an external constraint) and growth twins. The latter may form at an early stage, when the normal crystal growth is hampered by some obstacles (impurities, dislocations *etc.*); to overcome this a change in orientation occurs. Although the probability of twinning is higher at the nucleation stage, growth twins may also form by oriented attachment (Sunagawa, 2005), a phenomenon known as *synneusis* in the petrological community (Vance, 1969), which may occur at an early or at a late stage (Nespolo & Ferraris, 2004).

Twinned crystals are physically separated by an interface called the *composition surface*. When this surface is planar, one speaks of *composition plane* (Friedel, 1926; Hartman, 1956). In the case of reflection twins, the composition plane can be parallel to the twin plane, as in most of the twins known to date, or differently oriented. In the former case the resulting twin is a contact twin; otherwise, the result is a penetration twin in the case of TLQS, but can also be a contact twin in the case of TLS (Curien & Kern, 1957; Sunagawa, 2005).

The old, morphological classification of twins used the concept of *hemitropy* (from the Greek $\eta\mu\iota$ 'half' and $\tau\rho\omicron\pi\iota\alpha$ 'to turn') to tentatively explain the formation of twins as the result of a twofold rotation about a direction perpendicular to the composition plane (normal hemitropy) or contained in this plane (parallel hemitropy). This categorization has been criticized by Friedel (1926) for two reasons: (i) it cannot explain twins in which the twin operation is of order higher

than two (manifold twins); (ii) reflection twins were treated as equivalent to twofold rotation twins, which is not correct for TLQS.

As will be discussed, from the reticular viewpoint some manifold twins can actually be interpreted (exactly in TLS; approximately in TLQS) as parallel or normal hemitropy. Moreover, for rotation twins the orientation of the twin axis with respect to the composition plane is an important feature. For these reasons, in the following we do make reference to the concept of hemitropy. As a useful bridge for readers with a background in physics or metallurgy and dealing with mechanical twinning, we emphasize the following correspondences (Crocker, 1962):

(a) Reflection twins in which the twin plane is parallel to the composition plane, or twofold rotation twins by normal hemitropy, are known as type I twins.

(b) Reflection twins in which the twin plane is perpendicular to the composition plane, or twofold rotation twins by parallel hemitropy, are known as type II twins.

Inversion twins are always twins by merohedry, *i.e.* the whole lattice is restored by the twin operation, because a three-dimensional lattice is always centrosymmetric. In the case of reflection or rotation twins, instead, the lattice restoration can be partial and the (quasi-)restored nodes define a sublattice which can in principle extend to three, two or just one direction; accordingly, twins are classified into triperiodic, diperiodic and monoperoiodic (Friedel, 1933). Almost all known twins are triperiodic; monoperoiodic twins were introduced to account for the special case of the Zinnwald twin in quartz, where a common periodicity could be found only in one direction. The possible existence of diperiodic twins was then considered as an intermediate case between the triperiodic and the monoperoiodic case, in analogy with the case of diperiodic epitaxy (Royer, 1928, 1954). However, in the case of epitaxy, the lack of common periodicity outside the composition plane is a direct consequence of the chemical, and thus structural, heterogeneity of the individuals; whereas a common two-dimensional supercell in the composition plane is a necessary condition for the formation of the epitaxy, a common periodicity outside this plane is not indispensable and is not realized in general, because the two crystals forming an epitaxy do not have, in general, a commensurate periodicity outside the composition plane. On the other hand, because the twinned individuals have by definition the same structure,¹ the presence of a common two-dimensional supercell at the composition plane without a (reasonably small) common periodicity outside the plane represents an unlikely situation.

In the case of late-stage synneusis, crystals undergoing oriented attachment have already developed their characteristic morphology; the existence of a common three-dimensional sublattice with a high degree of restoration is then a less stringent requirement. Two crystals coming into

¹ This statement has to be softened in the case of allotwins, *i.e.* the oriented associations of different polytypes of the same compound (Nespolo, Kogure *et al.*, 1999). However, because the individuals differ by the stacking of identical layers, the out-of-plane periodicity is again commensurate.

contact on a large, flat face with a random mutual orientation are likely to form an incoherent interface; in that case, they tend to re-orient so that the interface is separated into coherent and defect regions, with overall reduction of the interfacial energy (Leo & Schwartz, 2000). The result is the semi-coherent interface of a twin, whose limiting case is the coherent interface of a parallel growth. The relative orientation corresponding to a plesiotwin may represent a compromise between the more stable situation of the semi-coherent interface of a twin, and the incoherent interface of a completely random orientation realized at the moment of the attachment, if the former is significantly away from the latter. If now we express the relative orientation of the two individuals through a rotation φ about a direction $[hkl]_c^*$ perpendicular to the composition plane $(hkl)_c$, depending on whether $[hkl]_c^*$ does or does not approach a lattice direction $[uvw]$ in direct space, the oriented crystal association is triperiodic or diperiodic, respectively. Furthermore, the following categories are easily obtained on the basis of the value of the rotation φ :

(a) φ is a crystallographic operation which does not belong to the point symmetry of the individuals: the pair of crystals forms a twin;

(b) φ is a non-crystallographic operation: the pair of crystals forms a plesiotwin.

List of symbols used below

$[uvw]$: a lattice direction in direct space; uvw are the direction indices.

(hkl) : a lattice plane in direct space; hkl are the Miller indices of the plane.

$(hkl)_c$: the composition plane separating two twinned or plesiotwinned individual crystals.

$(hkl)_\perp$: the direct-lattice plane mapping the orientation of two twinned or plesiotwinned individuals in the case of reflection (plesio)twins.

$[hkl]_\perp$: the lattice direction in direct space quasi-perpendicular to a direct-lattice plane (hkl) ; it coincides with $[hkl]^*$ when the obliquity ω is zero.

$(uvw)_\perp$: the direct-lattice plane quasi-perpendicular to lattice direction $[uvw]$ in direct space; it coincides with $(uvw)^*$ when the obliquity ω is zero.

\mathcal{H}_i : the point group of the i th individual crystal.

\mathcal{H}^* : the intersection group of the point groups of the individual crystals in their respective orientations.

\mathcal{H}_L^* : the holohedral supergroup of \mathcal{H}^* (possibly coincident with \mathcal{H}^* if the latter is already holohedral).

\mathcal{K} : the chromatic supergroup of \mathcal{H}_L^* obtained as an extension of the latter by the twin operation(s).

ω , obliquity: the angle between the lattice direction (plane) quasi-perpendicular to the twin plane (axis) and the direction (plane), in general irrational, exactly perpendicular to it; $\omega = 0$ in TLS and in some manifold TLQS twins having a quasi-specialized metric perpendicular to the twin axis.

δ , twin misfit: the linear divergence between lattice nodes quasi-restored by the twin operation in zero-obliquity TLQS.

Ξ : the two-dimensional coincidence index for a plane of the family (hkl) .

ξ : the number of planes of the family (hkl) which (i) have a finite value of Ξ and (ii) are found between two successive planes having a node on the direction $[hkl]^*$.

2. Twin operations, twin elements, twin laws, twin lattice

A twin operation is a crystallographic isometry that maps, exactly or approximately, the orientation of the individuals of a twin. A twin element is a geometric element (point, line, plane) about which the twin operation is performed. If \mathbf{L}_1 is the lattice of the first individual of the twin taken as reference and t is a twin operation, the lattice of a second individual is $\mathbf{L}_2 = t\mathbf{L}_1$ and the twin lattice \mathbf{L}_T is the sublattice obtained as the intersection of the two lattices \mathbf{L}_1 and \mathbf{L}_2 ; the twin operation is a (pseudo)-symmetry operation for the twin lattice:

$$\mathbf{L}_T = \mathbf{L}_1 \cap \mathbf{L}_2 = \mathbf{L}_1 \cap t\mathbf{L}_1; t\mathbf{L}_T \simeq \mathbf{L}_T. \quad (1)$$

Equation (1) is easily generalized to a twin composed of $n > 2$ individuals.

Let \mathcal{H}_i be the point group of the i th individual, where the running index i points to each of the n individuals. All the \mathcal{H}_i groups are of the same type (same Hermann–Mauguin symbol) but they differ in their orientation. If \mathcal{H}^* is the intersection group of the n groups \mathcal{H}_i , the twin point group \mathcal{K} is generated from \mathcal{H}^* by $n - 1$ independent twin operations and can be decomposed in cosets with respect to \mathcal{H}^* :

$$\mathcal{K} = \mathcal{H}^* \cup t_1\mathcal{H}^* \cup t_2\mathcal{H}^* \dots t_{n-1}\mathcal{H}^* \quad (2)$$

where n is the number of cosets, $n - 1$ is the number of twin laws and t_i is the representative (twin operation) of the i th coset. If, about the same geometric element, both symmetry and twin operations coexist (a sixfold rotation as twin operation collinear with a threefold or twofold rotation as symmetry operation; a sixfold roto-inversion as twin operation collinear with a threefold rotation or perpendicular to a mirror reflection as symmetry operation; a fourfold rotation or roto-inversion as twin operation collinear with a twofold rotation as symmetry operation), then only the corresponding subset of operations occurs in equation (2).

\mathcal{K} is a chromatic point group in which each colour represents an isometry mapping a different individual to the one taken as reference (Nespolo, 2004). Each coset in equation (2) apart from \mathcal{H}^* is a twin law, and each element of the coset is a possible twin operation which maps the corresponding individuals. Depending on whether twinning is by TLS or TLQS, each operation in the same coset is a symmetry or pseudo-symmetry operation for the twin lattice, which means that from a reticular viewpoint these operations are exactly or only approximately equivalent. From the structural viewpoint, \mathcal{H}^* can be any type of crystallographic point group with the exception of the cubic and hexagonal holohedries, because these cannot be extended to a supergroup. We are interested in the lattice restoration; we therefore consider the holohedral supergroup \mathcal{H}_L^* of \mathcal{H}^* , which can only be of type $\bar{1}$, $2/m$, mmm , $4/mmm$ or $\bar{3}m$; the extension by $\{t\}$ is therefore one of the six non-triclinic holohedries. Let us indicate by $|\mathcal{H}_L^*|$ the order of

\mathcal{H}_L^* , which is also the length of each coset. Because \mathcal{H}_L^* is centrosymmetric, each coset contains as many operations of the second kind (handedness reversing) as of the first kind (handedness preserving). If the number of rotations of order $n > 2$ is p ($p = 2$ when $\mathcal{H}_L^* = \bar{3}m$ or $4/mmm$, 0 otherwise), then \mathcal{H}_L^* also contains p roto-inversions of order $n > 2$, $|\mathcal{H}_L^*|/2 - p - 1$ rotations of order $n = 2$ and $|\mathcal{H}_L^*|/2 - p - 1$ reflections (the two operations left are the identity and the inversion). In the case of TLS, any of the operations in a twin law [coset in equation (2)] defines exactly the same twin lattice; in some cases, the latter can also be obtained by other isometries which do not correspond to a crystallographic operation and, as such, do not appear in a twin law. This is the case of non-crystallographic rotations φ about $[hkl]_c^*$ which characterize plesiotwins.

In the case of TLQS the operations in a twin law are no longer rigorously equivalent. In particular:

(a) A rotation of order $n = 2$ about a direction $[uvw]$ is equivalent to a mirror reflection about a plane $(uvw)^*$ perpendicular to $[uvw]$. In TLS, the plane $(uvw)^*$ is rational in the direct space and its Miller indices will be indicated as $(uvw)_\perp$. In TLQS, the plane $(uvw)^*$ is not rational in the direct space; the angle ω between this plane and the rational plane $(uvw)_\perp$ quasi-perpendicular to $[uvw]$ is the obliquity. Accordingly, in TLS the twofold rotation about $[uvw]$ or the mirror reflection about $(uvw)_\perp$ lead to the same lattice restoration so that from the reticular viewpoint they are equivalent; the twin can be equivalently described as a rotation twin or reflection twin. In TLQS, instead, the two operations are not equivalent even from the reticular viewpoint; they lead to two different twins, known as *reciprocal twins* (Mügge, 1898) or *corresponding twins* (Friedel, 1926).

(b) A twin rotation of order $n > 2$ about a direction $[uvw]$ is always equivalent, from a reticular viewpoint, to a roto-inversion \bar{n} about the same direction (the inversion point being at the origin).

The systematic study of twins initiated in the late 18th century has shown that obliquities larger than 6° are very rare and twin indices larger than 6 are rather exceptional (Friedel, 1926). These empirical limits were adopted to distinguish between *Friedelian* and *non-Friedelian* twins (Nespolo & Ferraris, 2005), the latter being exceptional counter-examples. Most non-Friedelian twins are actually *hybrid twins*, i.e. twins in which two or more concurrent sublattices can be found within the empirical limit of 6° for the obliquity; the overall degree of lattice restoration is then a function of the fraction of lattice nodes belonging to any of these sublattices and defines the *effective twin index* (Nespolo & Ferraris, 2006), which, in most cases, is again within the empirical limit of 6. A screening of a large number of known twins (Nespolo & Ferraris, 2009; Pignatelli *et al.*, 2011) has indeed shown that non-Friedelian twins are in most cases hybrid twins with a low effective twin index. A small minority of cases with a larger (effective) twin index concern rare twins in minerals reported long ago, for which the identification of the twin element may be considered doubtful.

Operations that are equivalent for the lattice are in general no longer equivalent from the structural viewpoint. The

structural analysis of twins requires consideration of the pseudo-eigensymmetry of the crystallographic orbits, in particular close to the composition surface, and is beyond the scope of this article. Interested readers will find the detailed procedure e.g. in Marzouki *et al.* (2014) and references therein.

3. Calculation of the twin index and of the obliquity

The Miller indices of a family of lattice planes, (hkl) , identify the orientation of these planes. The position of the m th plane of the family (hkl) obeys the equation

$$hx + ky + lz = m \quad (3)$$

where m is an integer and x, y, z are the coordinates of a point in direct space on the plane. In the following, we assume that the unit cell used to describe the crystal undergoing twinning is primitive, in which case Miller indices are relatively prime [this is no longer true if the unit cell is centred (Nespolo, 2015b)]; if the conventional cell is not primitive, a change of basis to a primitive unit cell can always be adopted.

The unit cell of the twin lattice is built on a two-dimensional unit cell in a lattice plane (hkl) and a lattice direction $[uvw]$ (quasi-)perpendicular to (hkl) ; to emphasize its relation with the (hkl) plane, in the following it is indicated as $[hkl]_\perp$ but for calculation purposes its direction indices u, v, w are used. Let $\Xi \geq 1$ be the two-dimensional coincidence index for a plane of the family (hkl) (usually indicated as Σ in the CSL literature) under the twin operation: the two extreme cases are $\Xi = 1$ for a complete restoration and $\Xi = \infty$ for no restoration at all. Between two successive planes of the family (hkl) having a lattice node on the direction $[hkl]^*$ there are $N - 1$ lattice planes of the same family ($N \geq 1$); of these, ξ have finite Ξ and $N - \xi$ have infinite Ξ (no lattice node restored). The overall lattice restoration depends on the three parameters and the twin index is (Nespolo & Ferraris, 2006)

$$n = N\Xi/\xi \quad (4)$$

where only finite Ξ are considered. The number N of lattice planes with a node on the $[uvw]$ direction is obtained by computing the scalar product of the corresponding vectors in the dual spaces, \mathbf{r}_{hkl}^* , and of \mathbf{r}_{uvw} :

$$\begin{aligned} N &= \mathbf{r}_{hkl}^* \cdot \mathbf{r}_{uvw} = (|\mathbf{r}_{hkl}^*| |\mathbf{r}_{uvw}|) \mathbf{e}_{hkl}^* \cdot \mathbf{e}_{uvw} \\ &= |\mathbf{r}_{hkl}^*| |\mathbf{r}_{uvw}| \cos \omega \cong |\mathbf{r}_{uvw}| / d_{hkl} \\ &= (hkl | \mathbf{a}^* \mathbf{b}^* \mathbf{c}^*) (\mathbf{abc} | uvw) = |hu + kv + lw| \end{aligned} \quad (5)$$

where \cdot indicates a scalar product, \mathbf{e} stands for a unitary vector, $\mathbf{e}_{hkl}^* \cdot \mathbf{e}_{uvw} = \cos \omega \cong 1$ because (hkl) and $[uvw]$ are almost perpendicular, $|\mathbf{r}|$ is the norm of the vector \mathbf{r} , d_{hkl} is the interplanar distance of the family (hkl) , the absolute value of $hu + kv + lw$ is taken because the number of planes is a positive number, and the last equality comes from the fact that the scalar product of basis vectors in direct and reciprocal space is either 1 or 0. The obliquity is easily computed from the scalar product in equation (5):

$$\begin{aligned}\omega &= \cos^{-1}(\mathbf{r}_{hkl}^* \cdot \mathbf{r}_{uvw}) / (|\mathbf{r}_{hkl}^*| |\mathbf{r}_{uvw}|) \\ &= \cos^{-1} |hu + kv + lw| / (|\mathbf{r}_{hkl}^*| \cdot |\mathbf{r}_{uvw}|).\end{aligned}\quad (6)$$

The twin index does not depend on the orientation of the twin element (axis or plane) with respect to the composition plane, but the construction of the unit cell of the twin lattice does. In fact, because a pair of crystals is in contact on $(hkl)_c$, the unit cell of the twin lattice has to be built on a two-dimensional unit cell in $(hkl)_c$, for which Ξ can take finite values larger than 1 if $(hkl)_c$ is not parallel to the $(hkl)_t$. Reflection twins having the twin plane parallel to the composition plane or rotation twins by normal hemitropy, for which $(hkl)_c$ is parallel to $(uvw)_\perp$, have $\Xi = 1$ or $\Xi = \infty$. In fact, the minimal symmetry of a two-dimensional lattice is 2 and the two-dimensional lattice is therefore fully restored ($\Xi = 1$) by a twofold rotation about a lattice node or the mid-point between two lattice nodes, and not restored at all ($\Xi = \infty$) by a twofold rotation about any other point. Similarly, a mirror reflection about the twin plane $(hkl)_c$ fully restores the two-dimensional lattice of a plane $(h'k'l')$ if the projection of the lattice nodes of $(h'k'l')$ onto $(hkl)_c$ either coincides with the lattice nodes of $(hkl)_c$ or is located on the mid-points between two lattice nodes of $(hkl)_c$. On the other hand, ξ can only take the values 1 or 2; in fact, between two successive lattice planes which have a lattice node on the direction $[hkl]_c^*$, there may be one which has the mid-point between two lattice nodes again on $[hkl]_c^*$. Such a plane exists when N in equation (5) is even, and thus $\xi = 2$; otherwise, $\xi = 1$. The calculation of the twin index is then reduced to $N/p(N)$, where $p(N)$ is a simple parity function valuing 1 or 2 when N is odd or even, respectively (the reader is reminded that we assume the chosen unit cell is primitive).

On the other hand, reflection twins having the twin plane differently oriented with respect to the composition plane, or rotation twins by parallel hemitropy, may have finite values of Ξ larger than 1. Equation (4) can be generalized as follows:

$$n = N_t \Xi_t / \xi_t = N_\perp \Xi_\perp / \xi_\perp = N_c \Xi_c / \xi_c \quad (4a)$$

where the subscripts t, \perp and c indicate the twin plane $(hkl)_t$, the plane (quasi-)perpendicular to the twin axis $(uvw)_\perp$ and the composition plane $(hkl)_c$, respectively. Whereas $\Xi_t = \Xi_\perp = 1$ or ∞ and $\xi_t = \xi_\perp = 1$ or 2, the same restrictions do not apply to Ξ_c and ξ_c . The same happens when the twin operation is a rotation or roto-inversion of order $n > 2$.

4. Can diperiodic twins and plesiotwins occur as reflection twins?

As discussed above, whether the twin plane is parallel to the composition plane or not, the twin index does not change but the values of N , Ξ and ξ do. When $[hkl]_c^*$ coincides with a $[uvw]$ direction in direct space, then the restoration of lattice nodes outside the composition is exact; otherwise, it is only approximate. The obliquity measures this approximation. If no direction $[uvw]$ coincides, exactly or approximately, with $[hkl]_c^*$, then only the lattice nodes on the

composition plane are restored by the twin operation: the twin is diperiodic.

To find the $[uvw]$ direction close to $[hkl]_c^*$, we need to compute the expression of $[hkl]_c^*$ in direct space, and this is easily obtained by imposing that the expressions of this direction in the two dual spaces coincide (Nespolo, 2015a). The indices of the $[hkl]_c^*$ direction in direct space are

$$[uvw] = \mathbf{G}^* |hkl|. \quad (7)$$

If the direction is rational, the u , v , w indices can then be reduced so that they correspond to the h , k , l indices of the first lattice node along that direction. If the period of this direction is large, then n and N_t in equation (4a) would be so large as to have no physical meaning and the twin would be diperiodic. This requirement is however loosened by allowing a tolerance which coincides with the empirical limit on the obliquity ω ; in other words, a divergence up to $\sim 6^\circ$ between $[uvw]$ and $[hkl]_c^*$ is considered acceptable.

Let us suppose that two crystals of the same phase form a diperiodic twin with the composition plane parallel to the twin plane. This means that the direction $[hkl]_t^*$ is not rational in direct space within the empirical limits on the obliquity, so that N_t in equation (4a) is so large that only the lattice nodes on a single plane of the family (hkl) – namely, the composition plane – are restored by the twin operation. Equation (4a) reduces to $n = \Xi_t$. In our survey of hybrid twins, we have found very few examples of potential candidates for diperiodic twinning. The most likely example was the (104) twin in chalcostibite, with two directions quasi-perpendicular to the twin and composition plane, both having however a very large value of N_t . The contribution of the second restored plane, far away from the composition plane, could be considered insignificant. However, we later realized that the indexing of the twin and composition plane and the cell parameters were reported in the literature with respect to two different settings of the space group ($Pnma$ and $Pnam$). Once re-indexed consistently with respect to the standard setting, the twin becomes a normal, low-index hybrid twin (Nespolo, 2018). It turns out that, although it is possible to find a combination of cell parameters and Miller indices leading to a diperiodic twin, examples of this category are, to the best of our knowledge, unknown.

If instead $(hkl)_c$ is not parallel to $(hkl)_t$, $\Xi_c \geq 1$ but n is constant, so that $N_c \leq N_t$ [equation (4a)]; the larger is Ξ_c the smaller is N_c and the oriented crystal association quickly becomes triperiodic. Whether it is a twin or a plesiotwin depends on the rotation φ about $[hkl]_c^*$. Now, twins of this type are very scarce in the literature: they may be really rare or may have gone unnoticed if the composition plane has been incorrectly described as a twin plane.

The conclusion is that a reflection twin in which the composition plane is parallel to the twin plane cannot produce a plesiotwin ($\Xi = 1$); it can in principle lead to a diperiodic twin, if $[hkl]_t^*$ is irrational within the usual limits of the obliquity, but our survey of a large number of twins taken from the literature has shown that when N_t is large, there normally exist two or more concurrent sublattices based on the same twin

plane which contribute to the overall lattice restoration, so that the twin is still triperiodic, although hybrid. If instead the composition plane is not parallel to the twin plane, the result can be a plesiotwin.

5. Can diperiodic twins and plesiotwins occur as twofold rotation twins?

In the case of normal hemitropy, the same conclusions drawn for reflections twins with $(hkl)_t$ parallel to $(hkl)_c$ are obtained. This is a direct consequence of the centrosymmetric nature of the lattice: the twin lattice can be alternatively described (exactly, in the case of TLS; approximately in the case of TLQS) as a reflection twin about $(uvw)_\perp$, which is (almost) parallel to the composition plane.

In the case of parallel hemitropy, $\Xi = 1$ or ∞ and $\xi = 1$ or 2 for $(uvw)_\perp$ but not for $(hkl)_c$. The twin axis $[uvw]$ may correspond to a lattice direction of relatively large periodicity; this is typically the case for late-stage synnesis, when the contact on large, flat surfaces becomes the dominant factor. The twin index n can be unusually large, which means that the value of N in equations (4) and (5) is unusually large as well. In fact, the point group of a two-dimensional lattice can be 2, $2mm$, $4mm$ or $6mm$ and the corresponding possible holohedral plane groups are $p2$, $p2mm$, $c2mm$, $p4mm$ or $p6mm$. When out-of-plane operations are added, the result is a subset of holohedral layer groups, namely those layer groups obtained by extending the holohedral plane groups through an inversion. These groups are $p112/m$, $pmmm$, $cmmm$, $p4/mmm$ and $p6/mmm$ [symbols after Kopský & Litvin (2010)] or $P11(2/m)$, $P2/m2/m(2/m)$, $C2/m2/m(2/m)$, $P(4/m)2/m2/m$ and $P(6/m)2/m2/m$ [symbols after Dornberger-Schiff (1959)]. The value of the two-dimensional coincidence index Ξ depends on the orientation of the twin axis with respect to the symmetry directions of these layer groups.

For $p112/m$, $\Xi > 1$ for any orientation of the twin axis, because the layer group does not contain a twofold rotation axis in the plane of the layer.

For $pmmm$ and $cmmm$, $\Xi = 1$ if the twin axis is parallel to the $[100]$ or $[010]$ direction of the layer; $\Xi > 1$ otherwise.

For $p4/mmm$, $\Xi = 1$ if the twin axis is parallel to the $[100]$, $[010]$, $[110]$ or $[1\bar{1}0]$ direction of the layer; $\Xi > 1$ otherwise.

For $p6/mmm$, $\Xi = 1$ if the twin axis is parallel to the $[100]$, $[010]$, $[110]$, $[210]$, $[120]$ or $[1\bar{1}0]$ direction of the layer; $\Xi > 1$ otherwise.

When $\Xi > 1$, the actual value of Ξ depends on the orientation of the twin axis within the composition plane. The same lattice restoration can be expressed through a rotation φ about the direction $[hkl]_c^*$ contained in $(uvw)_\perp$. The result is therefore: a triperiodic twin if N_c is small and φ is crystallographic; a triperiodic plesiotwin if N_c is small and φ is not crystallographic; a diperiodic twin if N_c is large and φ is crystallographic; a diperiodic plesiotwin if N_c is large and φ is not crystallographic.

6. Can diperiodic twins and plesiotwins occur as manifold rotation twins?

When the twin operation is a rotation of higher order, the twin can be composed of more than two individuals. This is usually the case, unless the twin axis of order p is coaxial with a symmetry axis of order $p/2$. In fact, a 4 or $\bar{4}$ twin axis parallel to a twofold symmetry axis, as well as a 6 or $\bar{6}$ twin axis parallel to a threefold symmetry axis result in two twinned individuals, although the operation mapping the two individuals is of order $n > 2$. Another case of a twin with two individuals despite a twin operation of order $n > 2$ is when one or more individuals are physically missing, in which case one speaks of an incomplete twin (Nespolo, 2004).

The twin axis along $[uvw]$ is perpendicular to the reciprocal-lattice plane $(uvw)^*$. Because the twin axis is a (pseudo)-symmetry axis for the lattice or for a sublattice, there exists a direct-lattice plane $(uvw)_\perp$ which (almost) coincides with $(uvw)^*$; the Miller indices (hkl) corresponding to $(uvw)_\perp$ can be computed from equation (7) by multiplying both members by the metric tensor \mathbf{G} :

$$|hkl\rangle = \mathbf{G}|uvw\rangle_\perp.$$

The composition plane $(hkl)_c$ can be perpendicular to the twin axis and thus coincide with $(uvw)_\perp$, or be parallel to it. When $(hkl)_c$ coincides with $(uvw)_\perp$, the individuals of the twin are stacked along $[uvw]$ and different planes of the same $(hkl)_c$ family physically separate two successive individuals; this is typical of polysynthetic twins, although polysynthetic twins are not necessarily manifold twins (a classical counterexample is that of albite twinning in feldspars). Ξ_c [equation (4a)] can take finite values larger than 1, but the φ rotation about the normal to the composition plane, *i.e.* about $[uvw]$, is the twin rotation, or the rotational component of it in the case of roto-inversion twins, and is therefore certainly crystallographic, apart from possible small deviations due to metric pseudo-symmetries. The result is therefore a twin, not a plesiotwin. Further, in the case of TLS all planes of the family $(uvw)_\perp$ have the same Ξ_c value, which means that $N_c = \xi_c = 1$. The result is therefore a triperiodic twin with twin index $n = \Xi_c$. In the twin law [equation (2)] we can always find a direction in the $(uvw)_\perp$ plane about which a twofold rotation maps the orientation of two successive individuals exactly like the n -fold rotation about $[uvw]$. Contrary to Friedel's conclusions, the manifold twin can be described as due to parallel hemitropy, if the composition plane is perpendicular to the twin axis and if the focus is on each pair of individuals physically in contact through the composition plane. This interpretation is also in line with Sunagawa's view of 'twinning by higher than three-fold rotation as a repeated symmetry operation of a twofold axis' (Sunagawa, 2005).

In the case of TLQS, the twin could become diperiodic. It is however necessary that N_c [equation (4a)] gets a value sufficiently large so that the second plane of the family with finite Ξ_c is too far away from the composition plane for its contribution to the overall lattice restoration to be considered significant. This result can in principle be obtained when

$\omega > 0^\circ$, provided that two conditions are fulfilled at the same time: (i) the ratio of the cell parameters along the $[uvw]$ direction and in the $(uvw)_\perp$ plane – typically, the c/a ratio, if the standard setting is adopted – is such that the shortest-period lattice direction within the empirical limit on the obliquity from $[uvw]$ leads to an unreasonably large value of N_c ; (ii) the twin does not become hybrid. Although such a special case cannot be ruled out, the requirement of a pseudosymmetry higher than orthorhombic makes it an unlikely event. On the other hand, a metric pseudosymmetry in the $(uvw)_\perp$ plane, due to an interaxial angle slightly departing from the metrical value (90° or 120°) or to a slight difference in the in-plane cell parameters – typically, a and b if the standard setting is adopted – leads to a nonzero twin misfit δ and an angle φ relating the orientation of the two individuals about $[uvw]$ which slightly deviates from a crystallographic value. This deviation does not transform the twin, which remains triperiodic, into a plesiotwin but simply reflects the departure of the metric from exact symmetry.

The situation is different when the composition plane is parallel to the twin axis, because the twin operation repeats it and the result is a set of composition planes, one for each pair of individuals, typical of cyclic twins, unless the twin is incomplete with only two individuals left. The relative orientation of two individuals of a pair can be described as a twofold rotation about a direction contained in the composition plane $(hkl)_c$ and perpendicular to the twin axis $[uvw]$. If the twin rotation or roto-inversion is even, there exists also a twofold rotation about the direction $[hkl]_c^*$ perpendicular to the composition plane $(hkl)_c$, but not if the twin rotation is odd, namely three. In both cases the result is a twin, not a plesiotwin, with however a fundamental difference: in the former case, there exists a direction perpendicular to the composition plane about which a twofold rotation relates two individuals of a pair, whereas in the latter such a direction does not exist and the concept itself of plesiotwinning does not even apply. In the case of TLS, the twin is triperiodic. In the case of zero-obliquity TLQS ($\omega = 0^\circ$, $\delta \neq 0 \text{ \AA}$), the twin is still triperiodic but the twin operation about $[uvw]$ is a pseudo-symmetry operation for the lattice. Finally, in the case of nonzero obliquity the same conclusions drawn for the case above apply again: although the possibility of a diperiodic twin cannot be ruled out, the metric pseudosymmetry makes it an unlikely event.

A couple of simple examples for even and odd rotations can be useful to concretely illustrate the situation of manifold twins.

6.1. $\mathcal{K} = [4^{(4)}/m^{(2)}2^{(2,2)}/m^{(2,2)}2^{(2)}/m^{(2)}]^{(4)}$, $\mathcal{H}_L^* = 2/m$

The fourfold twin axis is perpendicular to a twofold symmetry axis and the twin is composed of four individuals. Each of the twofold symmetry axes along the secondary direction of the twin lattice acts also as a twin axis for two other individuals, and the same holds for the mirror plane normal to it: the elements are called *partially chromatic* and denoted by the superscript (2,2) because they fix two colours

(i.e. each of them is a symmetry element for one crystal and maps the top and the bottom of the other crystal) but exchange the two other colours (i.e. they map the other two crystals). The coset decomposition is the following:

$$\mathcal{K} = \{1, 2_{[010]}, \bar{1}, m_{(010)}\} \cup \{4_{[100]}^+, 2'_{[110]}, \bar{4}_{[100]}^+, m'_{(1\bar{1}0)}\} \\ \cup \{2'_{[001]}, 2'_{[100]}, m'_{(001)}, m'_{(100)}\} \cup \{4_{[100]}^-, 2'_{[1\bar{1}0]}, \bar{4}_{[100]}^-, m'_{(110)}\}.$$

Let us first consider the composition plane being (001): the twin is polysynthetic. The four individuals are 90° apart, with their unique symmetry direction alternatively along $[010]$ and $[100]$ with respect to the basis vectors of the twin lattice. Two individuals physically in contact on the composition plane are mapped by a twofold rotation about $[110]$ or $[1\bar{1}0]$, i.e. the chromatic rotations in the twin point group. The partially chromatic rotations, about $[100]$ and about $[010]$, map individuals that are not physically in contact but separated by a third individual between them, for which these rotations are symmetry operations.

In the case of TLS ($\omega = 0^\circ$, $\delta = 0 \text{ \AA}$), each pair of twinned individuals forms a parallel hemitropy, the twin axis in the composition plane being $[110]$ or $[1\bar{1}0]$ for the three pairs of individuals. The rotation φ relating the orientation of the two individuals is the $\pm 90^\circ$ rotation about $[001]^*$ (here coinciding with $[001]$), which is a crystallographic rotation. Further, all planes of the family (001) have the same Ξ_c value, which means that $N_c = \xi_c = 1$. This is therefore a triperiodic twin with twin index $n = \Xi_c$. In order for the twin to become diperiodic, we need N_c to get a value large enough so that the second plane of the family with finite Ξ_c is sufficiently far away from the composition plane so that its contribution to the overall lattice restoration can be considered insignificant. This result can in principle be for $\omega > 0^\circ$ and a specialized value of the c/a ratio for which there is no direction close to $[001]^*$ leading to a small value of N_c or making the twin hybrid. Although such a special case cannot be ruled out, the requirement of a tetragonal pseudosymmetry makes it an unlikely event. On the other hand, a pseudo-tetragonal metric in the (001) plane, due to a γ angle slightly departing from 90° or to a slight difference in the a and b cell parameters, leads to a nonzero twin misfit δ , and an angle φ relating the orientation of the two individuals about $[001]^*$ which slightly deviates from 90° . This deviation does not obviously make the oriented association a plesiotwin but simply reflects the departure of the metric from exact tetragonal symmetry.

If instead the composition plane is parallel to the twin axis, the four individuals are in contact with alternatives on (110) and on $(1\bar{1}0)$, resulting in a cyclic twin. The directions perpendicular to these two planes, and thus mutually perpendicular to each other, are contained in the composition plane and normal to it; in fact, $2'_{[110]}$ and $2'_{[1\bar{1}0]}$ occur in a different coset (twin law). The twin would be at the same time normal and parallel hemitropy, a contradiction which fully justifies Friedel's original criticism of the concept of hemitropy in this case. The same conclusion holds for the other type of even-fold twins, namely sixfold, but not in the case of odd-fold (namely, threefold) twins, as we are going to show in the next

example. The rotation φ relating the two individuals is the 180° rotation about $[110]$ or $[\bar{1}\bar{1}0]$, which is a crystallographic rotation. In the case of TLQS, we have to distinguish the following cases:

(a) $\omega > 0^\circ$, $\delta = 0$: the directions $[001]$ and $[001]^*$ are not parallel, but the two-dimensional lattice in the (001) plane is tetragonal. The rotation φ relating the two individuals is the 180° rotation about $[110]^*$ or $[\bar{1}\bar{1}0]^*$, which are no longer parallel to $[110]$ and $[\bar{1}\bar{1}0]$, respectively. $[110]$ and $[110]^*$ are in the $[\bar{1}\bar{1}0]$ zone, whereas $[\bar{1}\bar{1}0]$ and $[\bar{1}\bar{1}0]^*$ are in the $[110]$ zone.

(b) $\omega = 0^\circ$, $\delta > 0$: the directions $[001]$ and $[001]^*$ are parallel, but the two-dimensional lattice in the (001) plane is only pseudo-tetragonal. As in the previous case, the rotation φ relating the two individuals is the 180° rotation about $[110]^*$ or $[\bar{1}\bar{1}0]^*$, which are no longer parallel to $[110]$ and $[\bar{1}\bar{1}0]$, respectively, but are all in the $[001]$ zone.

(c) $\omega > 0^\circ$, $\delta > 0$: none of the five directions above are parallel in direct and reciprocal space. The rotation φ relating the two individuals is once again the 180° rotation about $[110]^*$ or $[\bar{1}\bar{1}0]^*$, which are not in zone with any pair of the three directions $[001]$, $[110]$ and $[\bar{1}\bar{1}0]$.

Whatever the case above, the result is a twin, not a plesiotwin. Only for special values of the cell parameters would the result be dipericodic; this is an unlikely event under the hypothesis of a tetragonal pseudosymmetry.

$$6.2. \mathcal{K} = [\bar{3}^{(3)}2^{(2,1)}/m^{(2,1)}]^{(3)}, \mathcal{H}_L^* = 2/m$$

The threefold twin axis is perpendicular to the twofold symmetry axis and the twin is composed of three individuals, *i.e.* three pairs of twinned crystals. Each of the twofold symmetry axes of an individual crystal acts also as a twin axis for the other two individuals, and the same holds for the mirror plane normal to it: these are therefore partially chromatic elements and denoted by the superscript (2,1) because they fix one colour (*i.e.* they are symmetry elements for one crystal) but exchange the two other colours (*i.e.* they map the two other crystals). The coset decomposition is the following:

$$\mathcal{K} = \{1, 2_{[010]}, \bar{1}, m_{[\bar{1}\bar{2}\bar{1}0]}\} \cup \{3_{[001]}^{+'}, 2'_{[100]}, \bar{3}_{[001]}^{+'}, m'_{[\bar{2}\bar{1}\bar{0}]}\} \\ \cup \{3_{[0010]}^{-'}, 2'_{[110]}, \bar{3}_{[001]}^{-'}, m'_{[1\bar{1}\bar{2}0]}\}.$$

The operations in each twin law (second and third coset) are exactly equivalent in the case of TLS, but only approximately equivalent in the case of TLQS.

If the composition plane is (001) , then the twin is polysynthetic. The same conclusions obtained in the previous example hold, the only difference being the number of individuals and the rotation φ , 120° here instead of 90° . If instead the composition plane is parallel to $[001]$, the twin is cyclic and the three composition planes coincide with the partially chromatic planes. Pairs of twinned crystals can also be mapped by twofold rotations about the partially chromatic axes perpendicular to the composition planes, so that each pair can also be described as a normal hemitropy, again against Friedel's conclusions, differently from the case of an even-fold twin discussed above. The analysis of TLQS does not differ

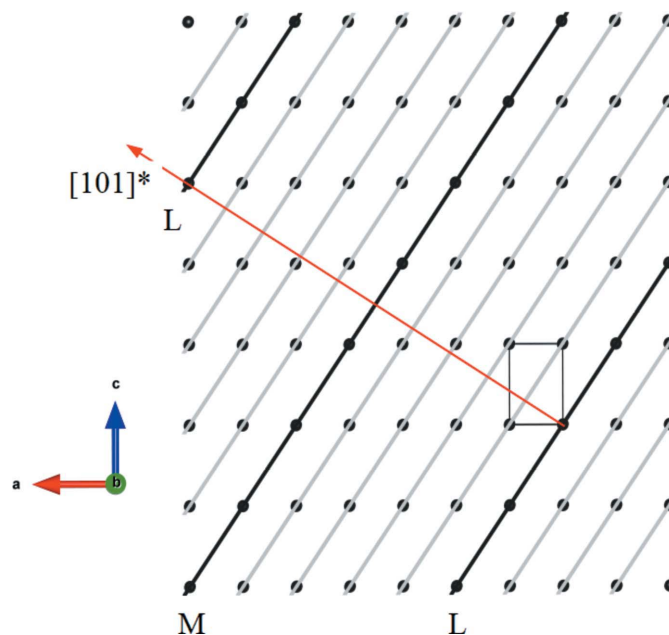


Figure 1
The lattice of atacamite observed in projection along the $[010]$ direction. Lattice planes of the family (101) are shown in black (when a lattice node or a mid-point between two lattice nodes is located on the direction $[101]^*$) or in grey (otherwise). The two-dimensional coincidence index Ξ is 1 for black planes and ∞ for the grey planes. Of the $N = 10$ planes between two black planes, $\xi = 2$ have $\Xi = 1$ so that the twin index is $n = 10 \times 1/2 = 5$. Cell parameters used in the calculation are taken from Parise & Hyde (1986). The figure was drawn with *VESTA* (Momma & Izumi, 2011).

from the previous case and the conclusion is the same: the result is a triperiodic twin, unless special values of the cell parameters make it dipericodic, an unlikely event under the hypothesis of a rhombohedral pseudosymmetry.

7. Case study

In the following, a few examples of real or hypothetical twins are analysed to illustrate the approach described in the previous sections. The twin index, obliquity and indices of directions quasi-perpendicular to planes have been obtained with the software *geminography* (Nespolo & Ferraris, 2006); the scan for direction in the composition plane was performed with a new software that will be presented soon elsewhere.

7.1. (101) twin in atacamite

Atacamite, $\text{Cu}_2(\text{OH})_3\text{Cl}$, is a secondary mineral formed through the oxidation of other copper minerals, especially in arid, saline conditions. It crystallizes in a space group of type *Pnma* (No. 62) and gives a twin by reflection on (101) (both twin and composition plane).² Fig. 1 shows the projection of the lattice of atacamite on the (010) plane. Lattice nodes are shown as black circles; planes of the family (101) from $m = -3$ to $m = 10$ [equation (3)] are shown: in black those for which Ξ

² In the mineralogical literature, the twin plane is often reported as (110) because the axial ratio obtained from the old morphological studies corresponds to the *Pnam* setting of the space group (*e.g.* Smith, 1898).

Table 1
Angular relations between lattice elements in staurolite.

Cell parameters are $a = 7.860$, $b = 16.645$, $c = 5.606$ Å, $\beta = 90.00^\circ$ (Caucia *et al.*, 1994), which lead to cell parameters for the twin lattice $a = 16.645$, $b = 16.818$, $c = 7.860$ Å, $\alpha = \beta = \gamma = 90.00^\circ$. The two sets of indices in each box separated by the oblique bar are expressed with respect to the basis vectors of the individual lattice (first) and of the twin lattice (second).

Element 1	(001) / (010)	(010) / (100)	(001) / (010)	(010) / (100)	(031) / (110)	$[\bar{0}\bar{1}3]$ / $[\bar{1}10]$	$[\bar{0}\bar{1}3]$ / $[\bar{1}\bar{1}0]$
Element 2	(031) / (110)	(031) / (110)	(0 $\bar{3}$ 1) / ($\bar{1}$ 10)	(03 $\bar{1}$) / (110)	(0 $\bar{3}$ 1) / ($\bar{1}$ 10)	$[013]$ / $[110]$	(0 $\bar{3}$ 1) / ($\bar{1}$ 10)
Angle	45.3°	44.7°	45.3°	44.7°	90.6°	89.4°	89.4°

is finite ($\Xi = 1$ in this case: all lattice nodes are restored), in grey those for which Ξ is infinite (no lattice node is restored). $N = 10$ planes of the family (101) occur between two planes having a lattice node on $[101]^*$; of these, $\xi = 2$ planes of the (hkl) family, namely those for which $m = 0$ and $m = 5$ [equation (3)], are restored by the twin operation: one ($m = 0$) has a lattice node on the direction $[101]^*$, the other ($m = 5$) has the mid-point between two lattice nodes on it. Indeed, $p(10) = 2$, so that the twin index is $10/2 = 5$. The point group of the lattice is of type mmm for both individuals and the intersection group is of type $2/m$: the only symmetry direction in common is $[010]$. The lattice direction closest to $[101]^*$ is $[703]$; the plane containing $[703]$ and $[010]$ is $(\bar{3}07)$ and lattice directions in the (101) plane defining the two-dimensional conventional unit cell in this plane are $[010]$ and $[\bar{1}01]$. The basis transformation is (I and T stand for individual and twin, respectively): $\mathbf{a}_T = 7\mathbf{a}_I + 3\mathbf{c}_I$, $\mathbf{b}_T = \mathbf{b}_I$, $\mathbf{c}_T = -\mathbf{a}_I + \mathbf{c}_I$; accordingly, the symmetry of the twin lattice obtained by applying equation (2) is

$$\begin{aligned} \mathcal{K} &= \{1, m'_{(101)}\}_I \times \{1, 2'_{[010]}, \bar{1}, m_{(010)}\}_I \\ &= \{1, 2'_{[703]}, 2'_{[010]}, 2'_{[\bar{1}01]}, \bar{1}, m'_{(101)}, m_{(010)}, m'_{(\bar{3}07)}\}_I \\ &= \{1, 2'_{[100]}, 2'_{[010]}, 2'_{[010]}, \bar{1}, m_{(100)}, m_{(010)}, m_{(001)}\}_T \\ &= 2'/m'2/m2'/m' \\ &= \{1, 2'_{[010]}, \bar{1}, m_{(010)}\}_I \cup \{2'_{[703]}, 2'_{[\bar{1}01]}, m'_{(101)}, m'_{(\bar{3}07)}\}_I. \end{aligned}$$

This is actually an approximation because the obliquity is $\omega = 0.52^\circ$, based on the cell parameters measured by Parise & Hyde (1986). Let us ignore this small deviation from the exact perpendicularity; in this case, we can consider the four operations in the second coset (the twin law) as exactly equivalent. $2'_{[703]}$ is (quasi-)perpendicular to the twin and composition plane and obviously provides the same lattice restoration as the reflection in the (101) plane. $2'_{[\bar{1}01]}$ is instead contained in the twin and composition plane. The relative orientation about the normal to the composition plane of two individuals related by a twofold rotation about $[\bar{1}01]$ is a 180° rotation about $[101]^*$, which almost coincides with the $[703]$ direction. This result corresponds to a crystallographic rotation φ and the oriented crystal association is therefore a twin, not a plesiotwin.

7.2. Greek cross twin in staurolite

Nespolo & Ferraris (2007) have presented a detailed reticular analysis of the twins in staurolite, which has a space group of type $C2/m$ but $\beta = 90^\circ$; this is therefore a case of metric merohedry (Nespolo & Ferraris, 2000) and the symmetry of the lattice is of type $Cmmm$. Fig. 2 shows a

drawing of the Greek cross twin, which is a penetration twin where the two individuals are rotated by 180° about the $\langle 013 \rangle$ direction, so that $\mathcal{H}^* = \bar{1}$ but $\mathcal{H}_L^* = mmm$. The basis vectors of the twin lattice are obtained by the basis transformation $\mathbf{a}_T = \mathbf{b}_I$, $\mathbf{b}_T = 3\mathbf{c}_I$, $\mathbf{c}_T = \mathbf{a}_I$; the inverse transformation is $\mathbf{a}_I = \mathbf{c}_T$, $\mathbf{b}_I = \mathbf{a}_T$, $\mathbf{c}_I = \mathbf{b}_T/3$. The twin lattice is pseudo-tetragonal, with $a_T/b_T = 0.9897$ [cell parameters from Caucia *et al.* (1994)]; the twin axis becomes $[110]$ (or $[\bar{1}\bar{1}0]$) when indexed in the basis of the twin lattice. The result of extending \mathcal{H}_L^* by $\{t\}$ is the following:

$$\mathcal{K} = \{1, 2'_{[110]}\} \times mmm = 4'/m2/m2'/m'.$$

If we now decompose \mathcal{K} in terms \mathcal{H}_L^* , we obtain two cosets including the subgroup, and thus only one twin law:

$$\begin{aligned} \mathcal{K} &= \mathcal{H}_L^* \cup \{2'_{[110]}, 2'_{[\bar{1}\bar{1}0]}, 4'_{[001]}, 4'_{[001]}, m'_{[110]}, m'_{[\bar{1}\bar{1}0]}, \bar{4}'_{[001]}, \bar{4}'_{[001]}\}_T \\ &= \mathcal{H}_L^* \cup \{2'_{[013]}, 2'_{[\bar{0}\bar{1}3]}, 4'_{[100]}, 4'_{[\bar{1}00]}, m'_{[013]}, m'_{[\bar{0}\bar{1}3]}, \bar{4}'_{[100]}, \bar{4}'_{[\bar{1}00]}\}_I. \end{aligned}$$

From the reticular viewpoint, any of the operations in the twin law (second coset) would be equivalent if the twin lattice were

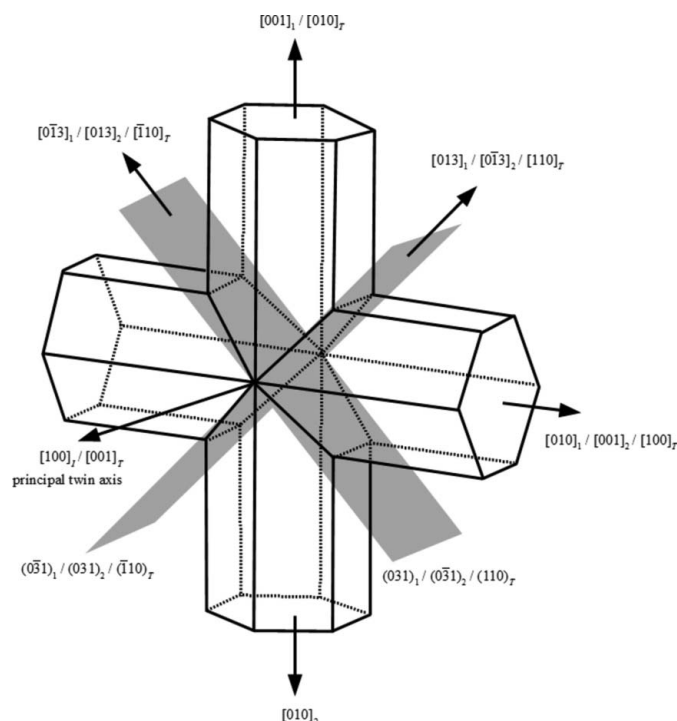


Figure 2
The Greek cross in staurolite, drawn after Fig. 4, plate 51, volume 8 of Goldschmidt (1923). The grey planes bisect the re-entrant angles and have the appearance of composition planes separating the two twinned crystals, although this is actually a penetration twin. Indexing of planes and directions is expressed with respect to the basis vectors of both the individuals (subscripts 1 and 2) and the twin (subscript T) lattices.

Table 2

Parallel hemitropy twins and plesiotwins (index up to 100) in a tetragonal lattice with composition plane (001) and (plesio)twin axis $[uv0]$ (cf. Fig. 3).

The relative rotation φ between the two crystals takes a crystallographic value for $n = 1$ (directions $[100]$ and $[110]$), which corresponds to twinning by merohedry for crystals with point groups of type $4, \bar{4}, 4mm, \bar{4}m2$ and $4/m$, and to parallel growth for crystals with point groups of type $422, 4\bar{2}m$ and $4/mmm$. All the other cases correspond to plesiotwinning.

φ (°)	n	$[uv0]$	φ (°)	n	$[uv0]$	φ (°)	n	$[uv0]$	φ (°)	n	$[uv0]$
0	1	$[100]$	25.99	89	$[13,3,0]$	47.92	97	$[940]$	71.08	37	$[750]$
8.80	85	$[13,1,0]$	28.07	17	$[410]$	48.89	73	$[11,5,0]$	73.74	25	$[430]$
10.39	61	$[11,1,0]$	30.51	65	$[11,3,0]$	53.13	5	$[210]$	75.75	65	$[970]$
12.68	41	$[910]$	31.89	53	$[720]$	58.11	53	$[950]$	77.32	41	$[540]$
14.25	65	$[810]$	36.87	5	$[310]$	59.49	65	$[740]$	79.61	61	$[650]$
16.26	25	$[710]$	41.11	73	$[830]$	61.93	17	$[530]$	81.20	85	$[760]$
18.92	37	$[610]$	42.08	97	$[13,5,0]$	64.01	89	$[850]$	90	1	$[110]$
22.62	13	$[510]$	43.60	29	$[520]$	64.94	85	$[11,7,0]$			
25.06	85	$[920]$	46.40	29	$[730]$	67.38	13	$[320]$			

exactly tetragonal, *i.e.* in the case of TLS. The Greek cross twin would then be a two-individual fourfold twin with the fourfold twin axis coaxial with a twofold symmetry axis. However, because the metric symmetry is only pseudo-tetragonal, the eight operations in the twin law all lead to a slightly different result. The obliquity computed for the $[110]_T/(\bar{1}\bar{1}0)_T$ pair {or $[\bar{1}\bar{1}0]_T/(110)_T$ } is 0.6° (Table 1), which means that the sublattices defined by the rotation and by the reflection slightly differ in orientation. On the other hand, the $[001]_T$ direction is exactly perpendicular to the $(001)_T$ plane but, precisely because the two directions $[110]_T$ and $[\bar{1}\bar{1}0]_T$ diverge from perpendicularity by 0.6° , they are not exactly overlapped by a fourfold rotation or roto-inversion about $[001]_T$. Although the obliquity computed for the $[001]_T/(001)_T$ pair is zero, the twin operation still corresponds to TLQS twinning and the deviation from exact overlap of lattice nodes is measured by the twin misfit $\delta = |a_T - b_T| = 0.173 \text{ \AA}$. The composition surface is not planar in a penetration twin like this. Nevertheless, the re-entrant corners (Fig. 2) are bisected precisely by the $(011)_T$ and $(0\bar{1}1)_T$ planes, which have the appearance of composition

planes. The twin axes $[110]_T$ and $[\bar{1}\bar{1}0]_T$ are contained and (almost) perpendicular to these planes, so that the φ angle mapping the two individuals is a 180° rotation, confirming the interpretation of this twin as a triperiodic twin.

7.3. $[uv0]$ twins and plesiotwins in a tetragonal crystal

Let us consider the case of a tetragonal crystal with twin axis contained in the (001) plane, *i.e.* with direction indices of type $[uv0]$. Because in a tetragonal lattice there is always a plane exactly perpendicular to any $[uv0]$ direction, with Miller indices $(uv0)$, the obliquity is systematically zero. This means that, from the reticular viewpoint, the twin is equivalently described as a twofold rotation twin about $[uv0]$ as twin axis, or as a reflection twin about $(uv0)$ as twin plane.

The point group of the lattice is of type $4/mmm$ for both individuals and the intersection group is of type $4/m$. The shortest directions in the $(uv0)$ plane are $[\bar{v}u0]$ and $[001]$, so that the basis transformation is $\mathbf{a}_T = u\mathbf{a}_I + v\mathbf{b}_I$, $\mathbf{b}_T = -v\mathbf{a}_I + u\mathbf{b}_I$, $\mathbf{c}_T = \mathbf{c}_I$; with a volume increase by $u^2 + v^2$. The symmetry of the twin lattice obtained by applying equation (2) is

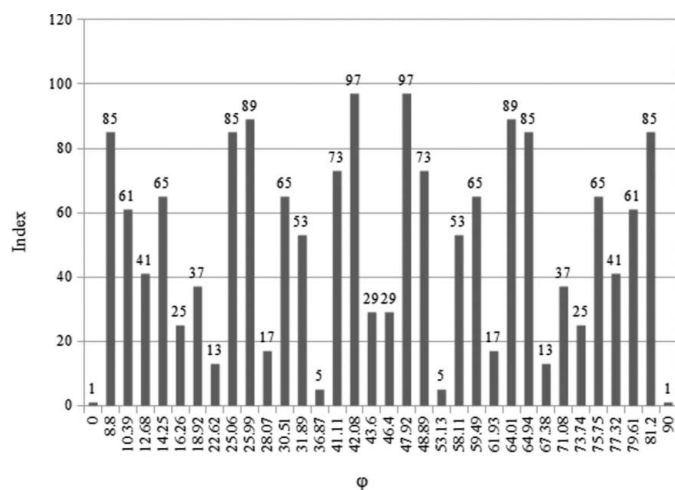


Figure 3

Histogram showing the Ξ_c value as a function of φ for the oriented association of two tetragonal crystals with composition plane (001). The $[uv0]$ indices of the directions corresponding to each bar in the histogram are in Table 2. Only φ values corresponding to $\Xi_c < 100$ are shown in the symmetrically independent octant between $[100]$ and $[110]$; these directions correspond to twins, all the others to plesiotwins.

$$\begin{aligned} \mathcal{K} &= \{1, 2'_{[uv0]}\}_I \times \{1, 4^+_{[001]}, 4^-_{[001]}, 2_{[001]}, \bar{1}, \bar{4}^+_{[001]}, \bar{4}^-_{[001]}, m'_{(001)}\}_I \\ &= \{1, 4^+_{[001]}, 4^-_{[001]}, 2_{[001]}, \bar{1}, \bar{4}^+_{[001]}, \bar{4}^-_{[001]}, m'_{(001)}, 2'_{[uv0]}, 2'_{[\bar{v}u0]}, \\ &\quad 2'_{[\bar{v}u0]}, 2'_{[uv0]}, m'_{(vu0)}, m'_{(uv0)}, m'_{(u\bar{v}0)}, m'_{(v\bar{u}0)}\}_I \\ &= \{1, 4^+_{[001]}, 4^-_{[001]}, 2_{[001]}, \bar{1}, \bar{4}^+_{[001]}, \bar{4}^-_{[001]}, m'_{(001)}, \\ &\quad 2'_{[100]}, 2'_{[010]}, 2'_{[110]}, 2'_{[\bar{1}\bar{1}0]}, m'_{(100)}, m'_{(010)}, m'_{(110)}, m'_{(\bar{1}\bar{1}0)}\}_T \\ &= 4/m 2' / m' 2' / m' \\ &= \{1, 4^+_{[001]}, 4^-_{[001]}, 2_{[001]}, \bar{1}, \bar{4}^+_{[001]}, \bar{4}^-_{[001]}, m'_{(001)}\}_I \\ &\quad \cup \{2'_{[uv0]}, 2'_{[\bar{v}u0]}, 2'_{[\bar{v}u0]}, 2'_{[uv0]}, m'_{(vu0)}, m'_{(uv0)}, m'_{(u\bar{v}0)}, m'_{(v\bar{u}0)}\}_I. \end{aligned}$$

The (chromatic) symmetry of the twin lattice is of the same type as the (achromatic) symmetry of the lattice of the individual, a special case of twinning known as reticular polyholohedry (Nespolo & Ferraris, 2004).

If we perform a scan of the $[uv0]$ directions in the symmetrically independent octant between $[100]$ and $[110]$ we obtain the results shown in Fig. 3 and Table 2 (only indices below 100 are shown). The rotation angle about the normal to (001) , *i.e.* the $[001]$ direction, varies between 0° and 90° .

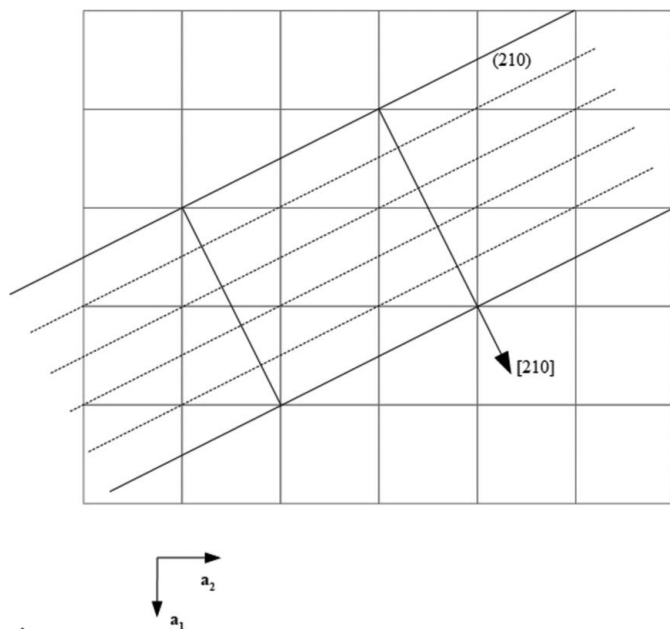


Figure 4
 (001) projection of a tetragonal lattice showing the unit cell of the (plesio)twin lattice based on the pair (210)/[210]. Of the five lattice nodes contained in the unit cell, only the one at the corners of the unit cell is restored by the twofold rotation about [210] or the mirror reflection about (210). Solid lines identify planes of the family (210) having $\Xi = 1$, dotted lines those for which $\Xi = \infty$.

Suppose that the composition plane is (001), so that twinning corresponds to parallel hemitropy. As a rotation twin about $[uv0]$, the twin index is $n = \Xi_c$ because $N_c = \xi_c = 1$ [equation (4a)]; in fact, the same two-dimensional CSL is obtained in every plane of the (001) family. As a reflection twin about $(hk0)$, the twin index is $n = N_{\perp} \Xi_{\perp} / \xi_{\perp}$, where $\xi_{\perp} = 1$ or 2, Ξ_{\perp} is either 1 or ∞ and N_{\perp} is the number of planes of the family $(hk0)$ between two successive planes for which $\Xi_{\perp} = 1$. Whether the twin is considered a rotation or a reflection twin, the restoration in the composition plane is partial and the relative orientation of two twinned individuals is the rotation φ in Fig. 3 and Table 2, which is non-crystallographic for all the directions but [100] and [010]. Accordingly, all the directions but these two lead to plesiotwins. Fig. 4 shows the case of [210]/(210), which corresponds to $n = 5$. As a rotation twin about [210], one lattice node out of five in the (001) plane is restored (the node at the corner of the unit cell of the twin lattice), so that $\Xi_c = 5$; the same result is valid for any plane of the family (001) so that $N_c = \xi_c = 1$. As a reflection twin about (210), instead, $\Xi_{\perp} = 1$ for the lattice planes corresponding to $m = 0$ and $m = 5$ in equation (1) (a trace of the planes is drawn as a solid line in Fig. 4), while $\Xi_{\perp} = \infty$ for all the other planes of the family, which means that $N_{\perp} = 5$ and $\xi_{\perp} = 1$. The result is obviously again $n = 5$. The relative orientation of two twinned individuals is the rotation $\varphi = 53.13^\circ$ about [001], which is non-crystallographic and makes this a plesiotwin.

Suppose now that the composition plane is $(hk0)$, so that twinning corresponds to normal hemitropy. As a rotation or as a reflection twin, the twin index is $n = N_t \Xi_t / \xi_t$ where $\xi_t = 1$ or 2, Ξ_t is either 1 or ∞ and N_t is the number of planes of the family

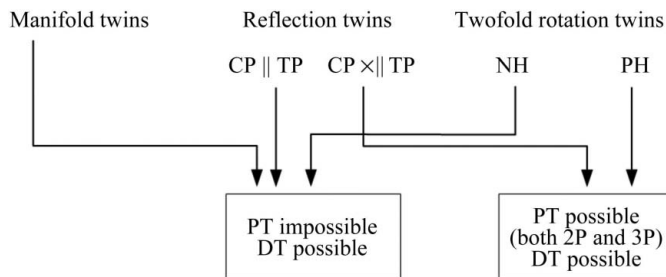


Figure 5
 Schematic summary of the possible occurrences of plesiotwins and diperiodic twins. PT: plesiotwins; DT: diperiodic twins; CP: composition plane; TP: twin plane; NH: normal hemitropy; PH: parallel hemitropy; 2P: diperiodic; 3P: triperiodic; ||: parallel to; x||: not parallel to.

$(hk0)$ between two successive planes for which $\Xi_t = 1$. Whether the twin is considered a rotation of a reflection twin, the restoration in the composition plane is complete ($\Xi_t = 1$) and the relative orientation of two twinned individuals is 180° about the lattice direction $[hk0]$; the rotation φ about [001] in Fig. 3 and Table 2 does not have a physical meaning in this case. Accordingly, the result is always a twin, but for large values of N_t the twin is actually diperiodic. By simple inspection of Fig. 3 and Table 2 we realize that all the twins besides those corresponding to $n = 5$ have a large value of N_t , the next one being already 13. A restoration every $N_t \geq 13$ lattice planes is hardly significant and these twins could therefore be considered diperiodic. However, almost all these twins are actually hybrid and in several cases the effective twin index is within or close to the empirical limit for a Friedelian twin, although hybrid (Table 3).

7.4. $[uv0]$ twins and plesiotwins in a pseudo-tetragonal crystal

The analysis in the previous section can be easily extended to look for possible diperiodic twins or plesiotwins by transforming the tetragonal lattice into a pseudo-tetragonal one. This can be done by making the interaxial angles depart from 90° . If the distortion is applied to the γ angle, the obliquity between $[uv0]$ and $(uv0)$ is no longer zero but depends on γ and, with the increase in the deformation, the twin may become hybrid and the plane $(uv0)$ may end outside the empirical limit on the obliquity. Table 4 shows the example of the lattice direction [310] for an angle γ variable from 90° to 99° . Until $\gamma = 92^\circ$, the only change is in the obliquity, which moves from 0 to 1.6° ; between 93° and 94° , the plane (410) becomes closer to $(310)^*$ and, together with (310), makes the twin hybrid; between 95° and 97° (410) is no longer within the empirical limits, so that the twin goes back to being a non-hybrid based on (310) but with increasing obliquity; finally, between 98° and 99° , the lattice plane closest to $(310)^*$ is (510), with the index now increasing to 8. Now, if the composition plane is (001), all these are triperiodic plesiotwins with the same two-dimensional value of Ξ_c in all the planes of the (001) family. If instead the composition plane is of type $(hk0)$, when the lattice plane closer to $(310)^*$ is (510) only one

Table 3

Normal hemitropy in a tetragonal lattice with composition plane (001) and twin axis $[uv0]$.

Most of these twins are actually hybrid twins with two or more concurrent sublattices (up to eight), leading to an effective twin index n_E much smaller than the twin index based on the $(uv0)/[uv0]$ sublattice (cf. Table 2). $(hk0)_1 = (uv0)$ so that $\omega_1 = 0^\circ$ by definition. Directions within 6° from $[100]$ or $[110]$ are not considered in this table, because they are quasi-perpendicular to (100) or (110), respectively.

$[uv0]$	n_E	$(hk0)_1$	$(hk0)_2$	ω_2	$(hk0)_3$	ω_3	$(hk0)_4$	ω_4	$(hk0)_5$	ω_5	$(hk0)_6$	ω_6	$(hk0)_7$	ω_7	$(hk0)_8$	ω_8
[910]	11.7	(910)	(710)	1.79	(510)	4.97										
[810]	8.1	(810)	(15,2,0)	0.47	(710)	1.01	(13,2,0)	1.62	(610)	2.34	(11,2,0)	3.18	(510)	4.18	(920)	5.40
[710]	12.5	(710)	(510)	3.18												
[610]	7.4	(610)	(11,2,0)	0.84	(510)	1.85	(920)	3.07	(410)	4.57						
[510]	13	(510)														
[920]	12.1	(920)	(14,3,0)	0.43	(510)	1.22	(410)	1.51								
[13,3,0]	8.1	(13,3,0)	(610)	3.53	(11,3,0)	2.26	(410)	1.04	(710)	4.86	(510)	1.68	(310)	5.44		
[410]	5.7	(410)	(720)	7.1	(310)	4.40										
[11,3,0]	14.4	(11,3,0)	(410)	1.22	(310)	3.18										
[720]	10.6	(720)	(10,3,0)	0.75	(410)	1.91										
[310]	5	(310)														
[830]	14.6	(830)	(11,4,0)	0.57	(820)	1.25										
[13,5,0]	14.9	(13,5,0)	(520)	0.76	(730)	2.16	(310)	2.60								
[520]	4.1	(520)	(830)	1.25	(310)	3.37	(210)	4.76								
[730]	6.4	(730)	(210)	3.37	(310)	4.76										
[940]	8.8	(940)	(16,7,0)	0.33	(730)	0.76	(12,5,0)	1.34	(520)	2.16	(210)	2.60				
[11,5,0]	14.6	(11,5,0)	(730)	1.25	(210)	2.12										
[210]	5	(210)														
[950]	11.8	(950)	(530)	1.91	(210)	2.49										
[740]	7.2	(740)	(12,7,0)	0.51	(530)	1.22	(850)	2.26	(210)	3.18						
[530]	8.5	(530)	(210)	4.40												
[850]	12.7	(850)	(13,8,0)	0.40	(530)	1.04	(320)	1.68								
[11,7,0]	7.4	(11,7,0)	(970)	5.40	(950)	3.42	(430)	4.40	(750)	3.07	(320)	1.22	(530)	1.51	(210)	5.91
[320]	6.5	(320)	(430)	3.18												
[750]	12.3	(750)	(320)	1.85	(530)	4.57										
[430]	6.2	(430)	(540)	1.79	(320)	3.18										
[970]	14.4	(970)	(430)	1.01	(750)	2.34	(320)	4.18								
[540]	10.2	(540)	(650)	1.15	(430)	1.79										

Table 4

Effect of a deviation from $\gamma = 90^\circ$ on the sublattice built on the $[310]$ direction of a pseudo-tetragonal crystal.

(hkl) is the lattice plane (quasi-)perpendicular to $[310]$. With the increase in the deviation, the (plesio)twins becomes hybrid ($\gamma = 93\text{--}94^\circ$), with a second concurrent sublattice based on the (410) plane contributing to the overall lattice restoration, before going back to non-hybrid based on (310) ($\gamma = 95\text{--}97^\circ$) or (510) ($\gamma = 98\text{--}99^\circ$). Beyond $5\text{--}6^\circ$, however, the lattice can hardly be considered pseudo-tetragonal.

$(hkl)/\gamma$	90	91	92	93	94	95	96	97	98	99
(310)	0	0.80	1.60	2.40	3.20	4.00	4.81	5.61		
(410)				1.87	1.2					
(510)									0.19	0.69
n or n_E	5	5	5	4.3	4.3	5	5	5	8	8

plane out of eight has $\Xi_\perp = 1$ and the twin could be considered diperiodic. Now, because $[310]$ is one of the directions that corresponds to the minimal ($n = 5$) index in a tetragonal crystal, the same conclusion also applies to the other directions. Note however that a deviation of 8° or more from $\gamma = 90^\circ$ is quite extreme and can hardly be considered compatible with a tetragonal pseudosymmetry.

If instead the deviation from the tetragonal metric occurs outside the (001) plane, the obliquity and the identification of the quasi-perpendicular plane also have to take into account the c/a ratio.

7.5. Conclusions

Fig. 5 summarizes the conclusion obtained through the analysis presented above. Plesiotwins can occur both in reflection twins, when the composition plane is not parallel to

the twin plane, and twofold rotation twins, in the case of parallel hemitropy. They cannot occur in manifold twins, because the rotation between pairs of twinned individuals is by definition crystallographic: either the higher-order rotation typical of manifold twins, or a twofold rotation about a direction perpendicular to the axis of the higher-order rotation. Since the distinction between a twin and a plesiotwin is based on the rotation about the normal to the composition plane, the latter obviously plays a primary role in the characterization of twins, contrary to the idea of Takano & Sakurai (1971) that it could be ignored in favour of a major role of the twin plane. Unfortunately, with the development of automatic diffraction equipment, the morphological analysis of crystals, which should be the first step in any crystallographic investigation unless the sample is anhedral, is commonly left out so that information of primary importance, like the orientation of the composition plane, is often not made available.

Diperiodic twins are in principle possible for any category. However, they are unlikely to occur in manifold twins because the high obliquity would be incompatible with the high degree of metric pseudosymmetry existing in these twins; and in twofold twins because a high-index twin is almost invariably a hybrid twin with a much lower effective twin index. These considerations might explain why no confirmed examples of diperiodic twins have been reported to date.

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