3.3. TWINNING OF CRYSTALS

provided by a Brazil twin of quartz, with each Brazil domain containing Dauphiné twins. This results in four domain states and three twin laws; *cf.* Example 3.3.6.4.

The distinction of simple and multiple twins is important for the following morphological classification. Further examples are given in Section 3.3.6.

3.3.3.1. *Morphological classification*

The morphology of twinned crystals, even for the same species and the same orientation relation, can be quite variable. For a given orientation relation the morphology depends on the geometry of the twin boundary as well as on the number of twin partners. A typical morphological feature of growth-twinned crystals is the occurrence of re-entrant angles. These angles are responsible for an increased growth velocity parallel to the twin boundary. This is the reason why twinned crystals often grow as platelets parallel to the composition plane (*cf.* Section 3.3.7.1). Detailed studies of the morphology of twins *versus* untwinned crystals were carried out as early as 1911 by Becke (1911). As a general observation, twinned crystals grow larger than untwinned crystals in the same batch.

The following classification of twins is in use:

(i) *Contact twins*. Two twin partners are in contact across a single composition plane $\pm(hkl)$, the Miller indices h, k, l of which have the same values for both partners ('common' plane). The contact plane usually has low indices. For reflection twins, the composition plane is frequently parallel to the twin mirror plane [see, however, Note (4) in Section 3.3.2.4]. Examples are shown in Fig. 3.3.6.3 for gypsum, in Fig. 3.3.6.7 for calcite and in Fig. 3.3.6.8(*a*) for a spinel (111) twin. In most cases, contact twins are growth twins.

(ii) *Polysynthetic (lamellar) twins* are formed by repetition of contact twins and consist of a linear sequence of domains with two alternating orientation states (Fig. 3.3.3.1*b*). The contact planes are parallel (lamellar twinning). This is typical for reflection twins if the twin mirror plane and the composition plane coincide. An illustrative example is the albite growth twin shown in Fig. 3.3.6.13. Polysynthetic twins may occur in growth, transformation and deformation twinning.

(iii) *Penetration twins*. The name 'penetration twin' results from the apparent penetration of two or more (idiomorphic) single crystals. The most prominent examples are twins of the spinel law in cubic crystals (*e.g.* spinels, fluorite, diamond). The spinel law is a reflection across (111) or a twofold rotation around [111]. Ideally these twins appear as two interpenetrating cubes with a common threefold axis, each cube representing one domain state, Fig. 3.3.6.8(*b*). In reality, these twins usually consist of 12 pyramid-shaped domains, six of each domain state, all originating from a common point in the centre of the twinned crystal (as shown in Figs. 3.3.6.6 and 3.3.6.8). Another famous example is the Carlsbad twin of orthoclase feldspar with [001] as a twofold twin axis (Fig. 3.3.7.1). Penetration twins are always growth twins.

(iv) *Cyclic twins and sector twins*. In contrast to the linear sequence of domains in polysynthetic twins, *cyclic twins* form a circular arrangement of domains of suitable shape. They are always multiple twins (three or more orientations states) which are (formally) generated by successive application of equivalent twin laws. The twin aggregate may form a full circle or a fraction of a circle (see Figs. 3.3.3.1*c*, 3.3.6.9 and 3.3.6.12). Impressive examples are the 'sixlings' and 'eightlings' of rutile (Fig. 3.3.6.11, *cf.* Example 3.3.6.10).

A special case of cyclic twins is provided by *sector twins*. Three or more domains of nearly triangular shape (angular sectors) extend from a common centre to form a twinned crystal with a more or less regular polygonal outline. The boundaries between two sector domains are usually planar and low-indexed. Such twins can be interpreted in two ways:

(*a*) they can be described by repeated action of (equivalent) reflection planes or twofold twin axes with suitable angular spacings;

(*b*) they can be described by approximate twin rotation axes of order three or more (including noncrystallographic axes such as fivefold); *cf.* Section 3.3.2.3.2 and Note (6) in Section 3.3.2.4.

Prominent examples are the growth twins of NH_4LiSO_4 (Fig. 3.3.7.2), aragonite (Fig. 3.3.2.4), K_2SO_4 (Fig. 3.3.6.9) and certain alloys with pseudo-fivefold twin axes (Fig. 3.3.6.10). Cyclic and sector twins are always growth twins.

(v) *Mimetic twins*. The term 'mimetic' is often applied to growth twins which, by their morphology, simulate a higher crystal symmetry. Regular penetration twins and sector twins are frequently also 'mimetic' twins. Particularly impressive examples are the harmotome and phillipsite twins, where monoclinic crystals, by multiple twinning, simulate higher symmetries up to a cubic rhomb-dodecahedron, described in Section 3.3.6.16(iii).

(vi) Common names of twins. In addition to the morphological description of twins mentioned above, some further shape-related names are in use:

(*a*) dovetail twins (prominent example: gypsum, *cf.* Figs. 3.3.4.1 and 3.3.6.3);

(*b*) elbow twins (rutile, cassiterite, *cf.* Fig. 3.3.6.11*a*);

(*c*) arrowhead twins (diamond);

(*d*) iron-cross twins (pyrite);

(*e*) butterfly twins (perovskite).

It is obvious from the morphological features of twins, described in this section, that crystals – by means of twinning – strive to simulate higher symmetries than they actually have. This will be even more apparent in the following section, which deals with the composite symmetry of twins and the twin law.

3.3.4. Composite symmetry and the twin law

In this section we turn our attention to the symmetry relations in twinning. The starting point of all symmetry considerations is the *eigensymmetry* $\mathcal{H}$ of the untwinned crystal, *i.e.* the point group or space group of the single crystal, irrespective of its orientation and location in space. All domain states of a twinned crystal have the same (or the enantiomorphic) *eigensymmetry* but may exhibit different orientations. The orientation states of each two twin components are related by a twin operation k which cannot be part of the *eigensymmetry* $\mathcal{H}$. The term *eigensymmetry* is introduced here in order to provide a short and crisp distinction between the symmetry of the untwinned crystal (single-domain state) and the composite symmetry $\mathcal{K}$ of a twinned crystal, as defined below. It should be noted that in crystal morphology the term *eigensymmetry* is also used, but with another meaning, in connection with the symmetry of face forms of crystals (Hahn & Klapper, 2005, pp. 764 and 791).

3.3.4.1. *Composite symmetry*

For a comprehensive characterization of the symmetry of a twinned crystal, we introduce the important concept of *composite symmetry* $\mathcal{K}$. This symmetry is defined as the extension of the *eigensymmetry* group $\mathcal{H}$ by a twin operation k. This extension involves, by means of left (or right) *coset composition* $k \times \mathcal{H}$, the generation of further twin operations until a supergroup is obtained. This supergroup is the composite symmetry group $\mathcal{K}$.

In the language of group theory, the relation between the composite symmetry group $\mathcal{K}$ and the *eigensymmetry* group $\mathcal{H}$ can be expressed by a (left) coset decomposition of the supergroup $\mathcal{K}$ with respect to the subgroup $\mathcal{H}$:

$$\mathcal{K} = k_1 \times \mathcal{H} \cup k_2 \times \mathcal{H} \cup k_3 \times \mathcal{H} \cup \ldots \cup k_i \times \mathcal{H},$$

where k_1 is the identity operation; $k_1 \times \mathcal{H} = \mathcal{H} \times k_1 = \mathcal{H}$.

The number i of cosets, including the subgroup $\mathcal{H}$, is the *index* [i] of $\mathcal{H}$ in $\mathcal{K}$; this index corresponds to the number of different
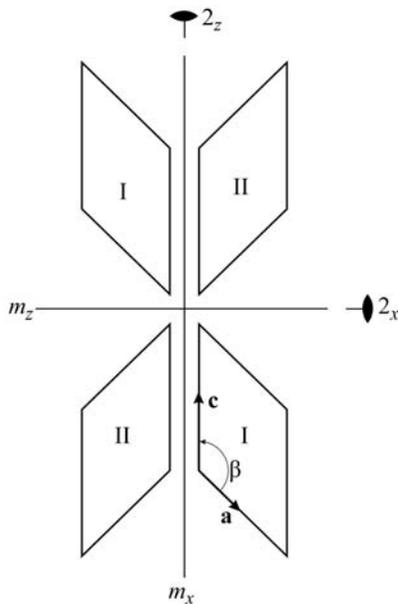
Fig. 3.3.4.1. Gypsum dovetail twin: schematic illustration of the coset of alternative twin operations. The two domain states I and II are represented by oriented parallelograms of *eigensymmetry* $2_y/m_y$. The subscripts x and z of the twin operations refer to the coordinate system of the orthorhombic composite symmetry $\mathcal{K}_D$ of this twin; **a** and **c** are the monoclinic coordinate axes.

orientation states in the twinned crystal. If $\mathcal{H}$ is a normal subgroup of $\mathcal{K}$, which is always the case if $i = 2$, then $k \times \mathcal{H} = \mathcal{H} \times k$, *i.e.* left and right coset decomposition leads to the same coset. The relation that the number of different orientation states n equals the index $[i]$ of $\mathcal{H}$ in $\mathcal{K}$, *i.e.* $n = [i] = |\mathcal{K}| : |\mathcal{H}|$, was first expressed by Zheludev & Shuvalov (1956, p. 540) for ferroelectric phase transitions.

These group-theoretical considerations can be translated into the language of twinning as follows: although the *eigensymmetry* $\mathcal{H}$ and the composite symmetry $\mathcal{K}$ can be treated either as point groups (finite order) or space groups (infinite order), in this and the subsequent sections twinning is considered only in terms of point groups [see, however, Note (8) in Section 3.3.2.4, as well as Section 3.3.10.4]. With this restriction, the number of twin operations in each coset equals the order $|\mathcal{H}|$ of the *eigensymmetry* point group $\mathcal{H}$. All twin operations in a coset represent the same orientation relation, *i.e.* each one of them transforms orientation state 1 into orientation state 2. Thus, the complete coset characterizes the orientation relation comprehensively and is, therefore, defined here as the *twin law*. The different operations in a coset are called *alternative twin operations*. A further formulation of the twin law in terms of black–white symmetry will be presented in Section 3.3.5. Many examples are given in Section 3.3.6.

This extension of the 'classical' definition of a twin law from a single twin operation to a complete coset of alternative twin operations does not conflict with the traditional description of a twin by the one morphologically most prominent twin operation. In many cases, the morphology of the twin, *e.g.* re-entrant angles or the preferred orientation of a composition plane, suggests a particular choice for the 'representative' among the alternative twin operations. If possible, twin mirror planes are preferred over twin rotation axes or twin inversion centres.

The concept of the twin law as a coset of alternative twin operations, defined above, has been used in more or less complete form before. The following authors may be quoted: Mügge (1911, pp. 23–25); Tschermak & Becke (1915, p. 97); Hurst *et al.* (1956, p. 150); Raaz & Tertsch (1958, p. 119); Takano & Sakurai (1971); Takano (1972); Van Tendeloo & Amelinckx (1974); Donnay & Donnay (1983); Zikmund (1984); Wadhawan (1997, 2000); Nespolo *et al.* (2000). A systematic application of

Table 3.3.4.1. *Gypsum, dovetail twins: coset of alternative twin operations (twin law), given in orthorhombic axes of the composite symmetry $\mathcal{K}_D$*

$\mathcal{H}$	$k \times \mathcal{H}$
1	$m_x \times 1 = m_x$
2_y	$m_x \times 2_y = m_z$
m_y	$m_x \times m_y = 2_z$
$\bar{1}$	$m_x \times \bar{1} = 2_x$

left and double coset decomposition to twinning and domain structures has been presented by Janovec (1972, 1976) in a key theoretical paper. An extensive group-theoretical treatment with practical examples is provided by Flack (1987).

Example: dovetail twin of gypsum (Fig. 3.3.4.1). Eigensymmetry:

$$\mathcal{H} = 1\frac{2_y}{m_y}1.$$

Twin reflection plane (100):

$$k_2 = k = m_x.$$

Composite symmetry group $\mathcal{K}_D$ (orthorhombic):

$$\mathcal{K} = \mathcal{H} \cup k \times \mathcal{H},$$

given in orthorhombic axes, x, y, z. The coset $k \times \mathcal{H}$ contains all four alternative twin operations (Table 3.3.4.1) and, hence, represents the twin law. This is clearly visible in Fig. 3.3.6.3(a). In the symbol of the orthorhombic composite group,

$$\mathcal{K} = \frac{2'_x}{m'_x}\frac{2_y}{m_y}\frac{2'_z}{m'_z},$$

the primed operations indicate the coset of alternative twin operations. The above black-and-white symmetry symbol of the (orthorhombic) composite group $\mathcal{K}$ is another expression of the twin law. Its notation is explained in Section 3.3.5. The twinning of gypsum is treated in more detail in Example 3.3.6.3.

It should be noted that among the four twin operations of the coset $k \times \mathcal{H}$ two are rational, m_x and 2_z, and two are irrational, m_z and 2_x (Fig. 3.3.4.1). All four are equally correct descriptions of the same orientation relation. From morphology, however, preference is given to the most conspicuous one, the twin mirror plane $m_x = (100)$, as the representative twin element.

The concept of composite symmetry $\mathcal{K}$ is not only a theoretical tool for the extension of the twin law but has also practical aspects:

(i) *Morphology of growth twins*. In general, the volume fractions of the various twin domains are different and their distribution is irregular. Hence, most twins do not exhibit regular morphological symmetry. If, however, the twin aggregate consists of p components of equal volumes and shapes (p = 'length' of the coset of alternative twin operations) and if these components show a regular symmetrical distribution, the morphology of the twinned crystal displays the composite symmetry. In minerals, this is frequently very well approximated, as can be inferred from Fig. 3.3.6.3 for gypsum. It is emphasized that, apart from this rare and idealized case, the composite symmetry does *not* give the symmetry of a twin specimen (twin symmetry), but rather a group-theoretical representation of the twin law including the coset of *all* alternative twin elements.

(ii) *Diffraction pattern*. The 'single-crystal diffraction pattern' of a twinned crystal exhibits its composite symmetry $\mathcal{K}$ if the volume fractions of all domain states are (approximately) equal.

(iii) *Permissible twin boundaries*. The composite symmetry $\mathcal{K}$ in its black–white notation permits immediate recognition of the 'permissible twin boundaries' (W-type composition planes), as explained in Section 3.3.10.2.1.

3.3.4.2. *Equivalent twin laws*

In the example of the dovetail twin of gypsum above, the twin operation $k = m_x = m(100)$ is of a special nature in that it maps the entire *eigensymmetry* $\mathcal{H} = 12/m1$ onto itself and, hence, generates a *single* coset, a single twin law and a finite composite group $\mathcal{K}$ of index [2] (simple twin). There are other twin operations, however, which do not leave the entire *eigensymmetry* invariant, but only a part (subgroup) of it, as shown for the hypothetical (111) twin reflection plane of gypsum in Example 3.3.6.3. In this case, extension of the complete group $\mathcal{H}$ by such a twin operation k does not lead to a single twin law and a finite composite group, but rather generates in the same coset two or more twin operations $k_2, k_3, \ldots$, which are *independent* (non-alternative) but symmetry *equivalent* with respect to the *eigensymmetry* $\mathcal{H}$, each representing a different but equivalent twin law. If applied to the 'starting' orientation state 1, they generate two or more new orientation states 2, 3, 4, ... In the general case, continuation of this procedure would lead to an *infinite* set of domain states and to a composite group of infinite order (*e.g.* cylinder or sphere group). Specialized metrics of a crystal can, of course, lead to a 'multiple twin' of small finite order.

In order to overcome this problem of the 'infinite sets' and to ensure a finite composite group (of index [2]) for a pair of adjacent domains, we consider only that subgroup of the *eigensymmetry* $\mathcal{H}$ which is left invariant by the twin operation k. This subgroup is the '*intersection symmetry*' $\mathcal{H}^*$ of the two '*oriented eigensymmetries*' $\mathcal{H}_1$ and $\mathcal{H}_2$ of the domains 1 and 2 (shown in Fig. 3.3.4.2): $\mathcal{H}^* = \mathcal{H}_1 \cap \mathcal{H}_2$. This group $\mathcal{H}^*$ is now extended by k and leads to the '*reduced composite symmetry*' $\mathcal{K}^*$ of the domain pair (1, 2): $\mathcal{K}^*(1, 2) = \mathcal{H}^* \cup k_2 \times \mathcal{H}^*$, which is a finite supergroup of $\mathcal{H}^*$ of index [2]. In this way, the complete coset $k \times \mathcal{H}$ of the *eigensymmetry* $\mathcal{H}$ is split into two (or more) smaller cosets $k_2 \times \mathcal{H}^*$, $k_3 \times \mathcal{H}^*$ *etc.*, where $k_2, k_3, \ldots$, are symmetry-equivalent twin operations in $\mathcal{H}$. Correspondingly, the differently oriented '*reduced composite symmetries*' $\mathcal{K}^*(1, 2) = \mathcal{H}^* \cup k_2 \times \mathcal{H}^*$, $\mathcal{K}^*(1, 3) = \mathcal{H}^* \cup k_3 \times \mathcal{H}^*$ *etc.* of the domain pairs (1, 2), (1, 3) *etc.* are generated by the representative twin operations k_2, k_3 *etc.* These cosets $k_i \times \mathcal{H}^*$ are considered as the twin laws for the corresponding domain pairs.

As an *example*, an orthorhombic crystal of *eigensymmetry* $\mathcal{H} = 2/m\,2/m\,2/m$ with equivalent twin reflection planes $k_2 = m(110)$ and $k_3 = m(\bar{1}10)$ is shown in Fig. 3.3.4.2. From the 'starting' domain 1, the two domains 2 and 3 are generated by the two twin mirror planes (110) and ($\bar{1}$10), symmetry equivalent with respect to the oriented *eigensymmetry* $\mathcal{H}_1$ of domain 1. The intersection symmetries of the two pairs of oriented *eigensymmetries* $\mathcal{H}_1$ & $\mathcal{H}_2$ and $\mathcal{H}_1$ & $\mathcal{H}_3$ are identical: $\mathcal{H}^* = 112/m$. The three oriented *eigensymmetries* $\mathcal{H}_1, \mathcal{H}_2, \mathcal{H}_3$, as well as the two differently oriented reduced composite symmetries $\mathcal{K}^*(1, 2) = \mathcal{K}^*(110)$ and $\mathcal{K}^*(1, 3) = \mathcal{K}^*(\bar{1}10)$ of the domain pairs (1, 2) and (1, 3), are all isomorphic of type $2/m\,2/m\,2/m$, but exhibit different orientations.

3.3.4.3. *Classification of composite symmetries*

The discussions and examples briefly presented in the previous section are now extended in a more general way. For the classification of composite symmetries $\mathcal{K}$ we introduce the notion of *oriented eigensymmetry* $\mathcal{H}_j$ of an orientation state j and attach to it its geometric representation, the *framework of oriented eigensymmetry elements*, for short *framework of oriented eigensymmetry*. Twin partners of different orientation states have the same *eigensymmetry* $\mathcal{H}$ but exhibit different *oriented eigensymmetries* $\mathcal{H}_j$, which are geometrically represented by their frameworks of oriented *eigensymmetry*. The well known crystallographic term 'framework of symmetry' designates the spatial arrangement of the symmetry elements (planes, axes, points) of a point group or a space group, as represented by a stereographic projection or by a space-group diagram (*cf.* Hahn, 2005, Parts 6, 7 and 10).
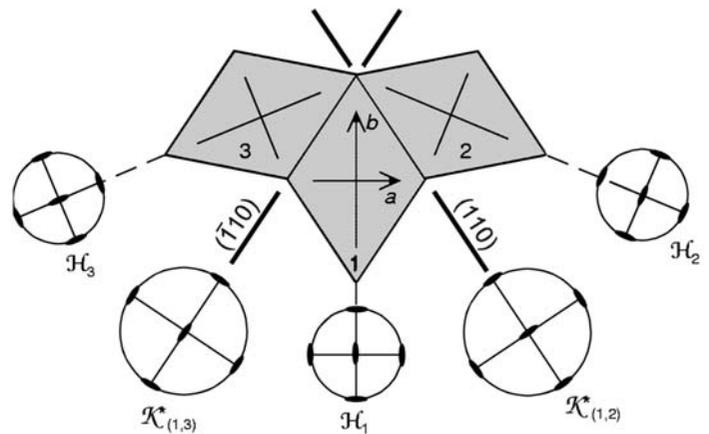

Fig. 3.3.4.2. Twinning of an orthorhombic crystal (*eigensymmetry* $2/m\,2/m\,2/m$) with equivalent twin mirror planes (110) and ($\bar{1}$10). Three twin domains 1, 2 and 3, bound by {110} contact planes, are shown. The oriented *eigensymmetries* $\mathcal{H}_1, \mathcal{H}_2, \mathcal{H}_3$ and the reduced composite symmetries $\mathcal{K}^*(1, 2) = \mathcal{K}^*(110)$ and $\mathcal{K}^*(1, 3) = \mathcal{K}^*(\bar{1}10)$ of each domain pair are given in stereographic projection. The intersection symmetry of all domains is $\mathcal{H}^* = 112/m$.

Similarly, we also consider the *intersection group* $\mathcal{H}^* = \mathcal{H}_1 \cap \mathcal{H}_2$ of the oriented *eigensymmetries* $\mathcal{H}_1$ and $\mathcal{H}_2$ and its geometric representation, the *framework of intersection symmetry*. Two cases of intersection symmetries have to be distinguished:

Case (I): $\mathcal{H}^* = \mathcal{H}_1 \cap \mathcal{H}_2 = \mathcal{H}$. Here, all twin operations map the *complete* oriented frameworks of the two domain states 1 and 2 onto each other, *i.e.* the oriented *eigensymmetries* $\mathcal{H}_1$ and $\mathcal{H}_2$ and their intersection group $\mathcal{H}^*$ coincide. Hence, for binary twin operations there is only one coset $k \times \mathcal{H} = k \times \mathcal{H}^*$ and one twin law. The composite symmetry $\mathcal{K} = \mathcal{H}^* \cup k \times \mathcal{H}^*$ is *crystallographic*. An example is provided by the dovetail twins of gypsum, described above: $\mathcal{H}^* = \mathcal{H} = 12/m1$ (*cf.* Table 3.3.4.1).

Case (II): $\mathcal{H}^* = \mathcal{H}_1 \cap \mathcal{H}_2 < \mathcal{H}_1$ and $< \mathcal{H}_2$ (index [i] $\geq$ 2). Here, the twin operations map only a *fraction* $1/i$ of the oriented symmetry elements of domain states 1 and 2 onto each other. Hence, the intersection group $\mathcal{H}^*$ of the two oriented *eigensymmetries* $\mathcal{H}_1$ and $\mathcal{H}_2$ is a proper subgroup of index [i] $\geq$ 2 of both $\mathcal{H}_1$ and $\mathcal{H}_2$. The coset $k \times \mathcal{H}^*$ leads to the crystallographic *reduced composite symmetry*

$$\mathcal{K}^* = \mathcal{H}^* \cup k \times \mathcal{H}^*,$$

which is crystallographic as for case (I) above. The number of twin laws, which are different but equivalent with respect to the 'starting' *eigensymmetry* $\mathcal{H}_1$ of the first domain state 1, equals the index [i]. This implies i differently oriented domain pairs $(1, j)$ ($j = 1, 2, \ldots, i$). The composite symmetry of such a domain pair is now defined by $\mathcal{K}^*_{1,j} = \mathcal{H}^* \cup k_j \times \mathcal{H}^*$ and is called the *reduced composite symmetry* $\mathcal{K}^*$. All twin laws can also be expressed by the black–white symbol of the reduced composite symmetry $\mathcal{K}^*_{1,j}$, as described in Section 3.3.5.

The orthorhombic example given in Section 3.3.4.1 (Fig. 3.3.4.2) is now extended as follows:

Eigensymmetry $\mathcal{H} = 2/m\,2/m\,2/m$, intersection symmetry $\mathcal{H}^* = 112/m$, $k_1 =$ identity, $k_2 = m(110)$, $k_3 = m(\bar{1}10)$, [i] = 2. The two cosets $k_2 \times \mathcal{H}^*$ and $k_3 \times \mathcal{H}^*$ are listed in Table 3.3.4.2. From these cosets the two different reduced composite symmetries $\mathcal{K}^*(1, 2)$ and $\mathcal{K}^*(1, 3)$ of type $2/m\,2/m\,2/m$ are derived as follows:

$$\mathcal{K}^*(1, 2) = \mathcal{H}^* \cup k_2 \times \mathcal{H}^* \quad \text{and} \quad \mathcal{K}^*(1, 3) = \mathcal{H}^* \cup k_3 \times \mathcal{H}^*.$$

These groups of reduced composite symmetry are always *crystallographic* and *finite*.

421

Table 3.3.4.2. *Reduced composite symmetries* $\mathcal{K}^*(1,2) = \mathcal{H}^* \cup k_2 \times \mathcal{H}^*$ *and* $\mathcal{K}^*(1,3) = \mathcal{H}^* \cup k_3 \times \mathcal{H}^*$ *for the orthorhombic example in Fig. 3.3.4.2*

$\mathcal{H}^*$	$k_2 \times \mathcal{H}^*$	$k_3 \times \mathcal{H}^*$
1	$m(110)$	$m(\bar{1}10)$
2_z	$m \perp [\bar{1}10]$	$m \perp [110]$
m_z	$2 \parallel [\bar{1}10]$	$2 \parallel [110]$
$\bar{1}$	$2 \perp (110)$	$2 \perp (\bar{1}10)$

Note that the twin operations in these two reduced cosets would form *one* coset if one of the operations (k_2 or k_3) were applied to the *full eigensymmetry* $\mathcal{H}$ (twice as long as $\mathcal{H}^*$): $k_2 \times \mathcal{H} = k_3 \times \mathcal{H} = k_2 \times \mathcal{H}^* \cup k_3 \times \mathcal{H}^*$. This process, however, would not result in a finite group, whereas the *two reduced* cosets lead to groups of finite order.

The two twin laws, based on $k_2(110)$ and $k_3(\bar{1}10)$, can be expressed by a black–white symmetry symbol of type $\mathcal{K}^* = 2'/m'\,2'/m'\,2/m$ with $\mathcal{H}^* = 112/m$. The frameworks of these two groups, however, are differently oriented (*cf.* Fig. 3.3.4.2).

In the limiting case, the intersection group $\mathcal{H}^*$ consists of the identity alone (index $[i]$ = order $|\mathcal{H}|$ of the *eigensymmetry* group), *i.e.* the two frameworks of oriented *eigensymmetry* have no symmetry element in common. The number of equivalent twin laws then equals the order $|\mathcal{H}|$ of the *eigensymmetry* group, and each coset consists of one twin operation only.

3.3.4.4. *Categories of composite symmetries*

After this preparatory introduction, the three categories of composite symmetry are treated.

(i) *Crystallographic composite symmetry.* According to case (I) above, only the following three types of twins have *crystallographic composite symmetry* $\mathcal{K}$ with two orientation states, one coset and, hence, one twin law:

(*a*) all merohedral twins, both with $\Sigma = 1$ and $Z > 1$ (reticular merohedry) (*cf.* Section 3.3.9);

(*b*) twins of 'monoaxial' *eigensymmetry* $\mathcal{H}$ that have either a twin reflection plane parallel or a twofold twin axis normal to the single *eigensymmetry* axis. Monoaxial *eigensymmetries* are $2, m = \bar{2}, 2/m, 3, \bar{3}, 4, \bar{4}, 4/m, 6, \bar{6} = 3/m, 6/m$;

(*c*) the triclinic *eigensymmetry* groups 1 and $\bar{1}$; here any binary twin element leads to a crystallographic composite symmetry $\mathcal{K}$.

Examples, including some special cases of trigonal crystals, are given in Section 3.3.6.

(ii) *Noncrystallographic composite symmetry.* As shown below, a noncrystallographic composite symmetry $\mathcal{K}$ results if the conditions of case (II) apply. Twins of this type are rather complicated because more than one twin law and more than two orientation states are involved. This case is illustrated in Figs. 3.3.3.1(*c*) and 3.3.4.2, where the twinning of an orthorhombic crystal with *eigensymmetry* $\mathcal{H} = 2/m\,2/m\,2/m$ and twin mirror plane (110) is considered. In case (II) above and in Fig. 3.3.4.2, domains 2 and 3 are generated from the starting domain 1 by the application of the equivalent twin elements $m_1(110)$ and $m_1(\bar{1}10)$. By applying the two twin elements $m_2(110)$ and $m_2(\bar{1}10)$ of domain 2, a new domain 4 is obtained and, at the same time, domain 1 is reproduced. Similarly, the twin elements $m_3(110)$ and $m_3(\bar{1}10)$ of domain 3 generate a further new domain 5, and domain 1 is reproduced again.

The continuation of this construction leads in the limit to a circular arrangement with an infinitely large number of domain states. The group-theoretical treatment of this process, based on the *full eigensymmetry*, results in the *infinite* composite symmetry group $\mathcal{K} = \infty/mm$, with the rotation axis parallel to the twofold axis of the intersection symmetry $112/m$, common to all these infinitely many domains. In an even more general case, for example an orthorhombic crystal with twin reflection plane (111), the infinite sphere group $\mathcal{K} = m\overline{\infty}$ would result as composite

symmetry. Neither of these cases is physically meaningful and thus they are not considered further here. It is emphasized, however, that the *reduced* composite symmetry $\mathcal{K}^*$ for any pair of domains in contact, as derived in case (II) above, is finite and crystallographic and, thus, of practical use.

(iii) *Pseudo-crystallographic composite symmetry.* Among twins with noncrystallographic composite symmetry, described above, those exhibiting structural or at least metrical pseudo-symmetries are of special significance. Again we consider an orthorhombic crystal with *eigensymmetry* $\mathcal{H} = 2/m\,2/m\,2/m$ and equivalent twin reflection planes (110) and (1$\bar{1}$0), but now with a special axial ratio $b/a \approx \left|\tan(180°/n)\right|$ ($n = 3, 4$ or 6).

The procedure described above in (ii) leads to three different orientation states for $n = 3$ and 6 and to two different orientation states for $n = 4$, forming a cyclic arrangement of sector domains (for cyclic and sector twins see Section 3.3.3). The intersection group $\mathcal{H}^*$ of all these domain states is $112/m$, with the twofold axis along the c axis. The reduced composite symmetry of any pair of domains in contact is orthorhombic of type $\mathcal{K}^* = 2'/m'\,2'/m'\,2/m$. If the only twin element is an n-fold rotation or roto-inversion axis ($m \geq 3$), *no restriction* on the b/a ratio is required. This case is discussed below under 'Remark'.

These multiple cyclic twins can be described in two ways (*cf.* Section 3.3.2.3.2):

(*a*) by repeated application of equivalent binary twin operations (reflections or twofold rotations) to a pseudosymmetrical crystal, as proposed by Hartman (1960) and Curien (1960). Note that each one of these binary twin operations is 'exact', whereas the closure of the cycle of sectors is only approximate; the deviation from $360°/n$ depends on the (metrical) pseudosymmetry of the lattice;

(*b*) by successive application of pseudo n-fold twin rotations around the zone axis of the equivalent twin reflection planes. Note that the individual rotation angles are not exactly $360°/n$, due to the pseudosymmetry of the lattice. This alternative description corresponds to the approach by Friedel (1926, pp. 435 and 464) and Buerger (1960*b*).

It is now reasonable to define an *extended composite symmetry* $\mathcal{K}(n)$ by adding the n-fold rotation as a further generator to the reduced composite symmetry $\mathcal{K}^*$ of a domain pair. This results in the composite symmetry $\mathcal{K}(n)$ of the complete twin aggregate, in the present case in a modification of the symmetry $\mathcal{K}^* = 2'/m'\,2'/m'\,2/m$ to:

$\mathcal{K}(6) = \mathcal{K}(3) = 6(2)/m\,2/m\,2/m$ (three orientation states, two twin laws) for $n = 3$ and $n = 6$;

$\mathcal{K}(4) = 4(2)/m\,2/m\,2/m$ (two orientation states, one twin law) for $n = 4$.

The *eigensymmetry* component of the main twin axis is given in parentheses.

This construction can also be applied to noncrystallographic twin rotations $n = 5, 7, 8$ *etc.* (*cf.* Section 3.3.6.9), *e.g.*:

$\mathcal{K}(10) = \mathcal{K}(5) = 10(2)/m\,2/m\,2/m$ (five orientation states, four twin laws) for $n = 5$ and $n = 10$ (Fig. 3.3.6.10).

The above examples are based on a twofold *eigensymmetry* component along the n-fold twin axis. An example of a pseudo-hexagonal twin, monoclinic gibbsite, Al(OH)$_3$, *without* a twofold *eigensymmetry* component along [001], is treated as Example 3.3.6.11 and Fig. 3.3.6.12.

It is emphasized that the considerations of this section apply not only to the particularly complicated cases of multiple growth twins but also to transformation twins resulting from the loss of higher-order rotation axes that is accompanied by a small metrical deformation of the lattice. As a result, the extended composite symmetries $\mathcal{K}(n)$ of the transformation twins resemble the symmetry $\mathcal{G}$ of their parent phase. The occurrence of both multiple growth and multiple transformation twins of ortho-rhombic pseudo-hexagonal K$_2$SO$_4$ is described in Example 3.3.6.8.

Remark. It is possible to construct multiple twins that cannot be treated as a cyclic sequence of binary twin elements. This case occurs if a pair of domain states 1 and 2 are related only by an *n*-fold rotation or roto-inversion ($n \geq 3$). The resulting coset again contains the alternative twin operations, but in this case *only* for the orientation relation $1 \Rightarrow 2$, and not for $2 \Rightarrow 1$ ('non-transposable' domain pair). This coset procedure thus does not result in a composite group for a domain pair. In order to obtain the composite group, further cosets have to be constructed by means of the higher powers of the twin rotation under consideration. Each new power corresponds to a further domain state and twin law.

This construction leads to a composite symmetry $\mathcal{K}(n)$ of supergroup index $[i] \geq 3$ with respect to the *eigensymmetry* $\mathcal{H}$. This case can occur only for the following $\mathcal{H} \Rightarrow \mathcal{K}$ pairs: $1 \Rightarrow 3$, $\bar{1} \Rightarrow \bar{3}$, $1 \Rightarrow 4$, $1 \Rightarrow \bar{4}$, $m \Rightarrow 4/m$, $1 \Rightarrow 6$, $2 \Rightarrow 6$, $m \Rightarrow \bar{6} = 3/m$, $m \Rightarrow 6/m$, $2/m \Rightarrow 6/m$ (monoaxial point groups), as well as for the two cubic pairs $222 \Rightarrow 23$, $mmm \Rightarrow 2/m\bar{3}$. For the pairs $1 \Rightarrow 3$, $\bar{1} \Rightarrow \bar{3}$, $m \Rightarrow \bar{6} = 3/m$, $2/m \Rightarrow 6/m$ and the two cubic pairs $222 \Rightarrow 23$, $mmm \Rightarrow 2/m\bar{3}$, the $\mathcal{K}$ relations are of index [3] and imply three non-transposable domain states. For the pairs $1 \Rightarrow 4$, $1 \Rightarrow \bar{4}$, $m \Rightarrow 4/m$, as well as $1 \Rightarrow 6$ and $m \Rightarrow 6/m$, four or six different domain states occur. Among them, however, domain pairs related by the second powers of 4 and $\bar{4}$ as well as by the third powers of 6 and $\bar{6}$ operations are transposable, because these twin operations correspond to twofold rotations or, for $\bar{6}$, to *m*.

No growth twins of this type are known so far. As a transformation twin, langbeinite ($23 \Longleftrightarrow 222$) is the only known example.

3.3.5. Description of the twin law by black–white symmetry

An alternative description of twinning employs the symbolism of colour symmetry. This method was introduced by Curien & Le Corre (1958) and by Curien & Donnay (1959). In this approach, a colour is attributed to each different domain state. Depending on the number of domain states, simple twins with two colours (*i.e.* 'black–white' or 'dichromatic' or 'anti-symmetry' groups) and multiple twins with more than two colours (*i.e.* 'polychromatic' symmetry groups) have to be considered. Two kinds of operations are distinguished:

(i) The symmetry operations of the *eigensymmetry* (point group) of the crystal. These operations are 'colour-preserving' and form the 'monochromatic' *eigensymmetry* group $\mathcal{H}$. The symbols of these operations are unprimed.

(ii) The twin operations, *i.e.* those operations which transform one orientation state into another, are 'colour-changing' operations. Their symbols are designated by a prime if of order 2: $2'$, m', $\bar{1}'$.

For *simple twins*, all colour-changing (twin) operations are binary, hence the two domain states are transposable. The composite symmetry $\mathcal{K}$ of these twins thus can be described by a 'black-and-white' symmetry group. The coset, which defines the twin law, contains only colour-changing (primed) operations. This notation has been used already in previous sections.

It should be noted that symbols such as $4'$ and $6'$, despite appearance to the contrary, represent *binary* black-and-white operations, because $4'$ contains 2, and $6'$ contains 3 and $2'$, with $2'$ being the twin operation. For this reason, these symbols are written here as $4'(2)$ and $6'(3)$, whereby the unprimed symbol in parentheses refers to the *eigensymmetry* part of the twin axis. In contrast, $6'(2)$ would designate a (polychromatic) twin axis which relates three domain states (three colours), each of *eigensymmetry* 2. Twin centres of symmetry $\bar{1}'$ are always added to the symbol in order to bring out an inversion twinning contained in the twin law. In the original version of Curien & Donnay (1959), the black–white symbols were only used for twinning by merohedry. In the present chapter, the symbols are also applied to

non-merohedral twins, as is customary for (ferroelastic) domain structures. This has the consequence, however, that the *eigensymmetries* $\mathcal{H}$ or $\mathcal{H}^*$ and the composite symmetries $\mathcal{K}$ or $\mathcal{K}^*$ may belong to different crystal systems and, thus, are referred to different coordinate systems, as shown for the composite symmetry of gypsum in Section 3.3.4.1.

For the treatment of *multiple twins*, 'polychromatic' composite groups $\mathcal{K}(n)$ are required. These contain colour-changing operations of order higher than 2, *i.e.* they relate three or more colours (domain states). Consequently, not all pairs of domain states are transposable. This treatment of multiple twins is rather complicated and only sensible if the composite symmetry group is finite and contains twin axes of low order ($n \leq 8$). For this reason, the symbols for the composite symmetry $\mathcal{K}$ of multiple twins are written without primes; see the examples in Section 3.3.4.4(iii). An extension of the dichromatic twin descriptions to polychromatic symbols for multiple twins was recently presented by Nespolo (2004).

3.3.6. Examples of twinned crystals

In order to illustrate the foregoing rather abstract deliberations, an extensive set of examples of twins occurring either in nature or in the laboratory is presented below. In each case, the twin law is described in two ways: by the coset of alternative twin operations and by the black–white symmetry symbol of the composite symmetry $\mathcal{K}$, as described in Sections 3.3.4 and 3.3.5.

For the description of a twin, the conventional crystallographic coordinate system of the crystal and its *eigensymmetry* group $\mathcal{H}$ are used in general; exceptions are specifically stated. To indicate the orientation of the twin elements (both rational and irrational) and the composition planes, no specific convention has been adopted; rather a variety of intuitively understandable simple symbols are chosen for each particular case, with the additional remark 'rational' or 'irrational' where necessary. Thus, for twin reflection planes and (planar) twin boundaries symbols such as m_x, $m(100)$, $m \parallel (100)$ or $m \perp [100]$ are used, whereas twin rotation axes are designated by 2_z, $2_{[001]}$, $2 \parallel [001]$, $2 \perp (001)$, 3_z, $3_{[111]}$, $4_{[001]}$ *etc.*

3.3.6.1. *Macroscopic identification of twins and of twin laws*

As an introduction to the subsequent examples, this section shows how to recognize and identify twinning in a crystal, either by morphological features and observations in polarized light, or by etching, decoration and X-ray diffraction topography. Diffraction effects of twins are treated in Section 3.3.11.

(i) *Re-entrant angles and twin striations*

The most prominent and easily recognizable morphological features are exhibited by penetration and contact twins with their *re-entrant angles* (*edges*). Re-entrant edges are typical for twins with non-parallel lattices (non-merohedral twins) and reticular merohedral twins with a $\Sigma > 1$ coincidence lattice (*e.g.* $\Sigma 3$ spinel twins). Merohedral $\Sigma 1$ twins with full lattice parallelism in general do not exhibit re-entrant edges but can often be identified by twin-related faces appearing in the morphology of a twinned crystal (*e.g.* $\Sigma 1$ twins of quartz). The re-entrant edges mark the outcrop of the twin boundaries, which are defined by the path of this edge during crystal growth. Note that re-entrant edges occur in any arbitrary intergrowth of crystals, which can sometimes be misinterpreted as twinning.

Illustrations of penetration twins are Figs. 3.3.6.6 ($FeBO_3$), 3.3.6.8 (spinel law, diamond), 3.3.6.15 (staurolite) and 3.3.7.1 (orthoclase feldspar). Contact growth twins with re-entrant angles are presented in Fig. 3.3.6.3 (gypsum) and 3.3.6.7 (calcite). In repeated growth twins (*e.g.* albite, Fig. 3.3.6.13) the parallel re-entrant edges form 'polysynthetic twins'. If the width of the twin lamellae gets smaller and smaller, *twin striations* develop as a characteristic feature of these twins. For further examples see the

references