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The paper is dedicated to Hans Wondratschek on the occasion of his 85th birthday

The application of *eigensymmetries* of face forms to anomalous scattering and twinning by merohedry in X-ray diffraction

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The face form (crystal form) {*hkl*} which corresponds to an X-ray reflection *hkl* is considered. The *eigensymmetry* (inherent symmetry) of such a face form can be used to derive general results on the intensities of the corresponding X-ray reflections. Two cases are treated. (i) Non-centrosymmetric crystals exhibiting anomalous scattering: determination of reflections *hkl* for which Friedel's rule is strictly valid, *i.e.* $I(hkl) = I(\bar{h}\bar{k}\bar{l})$ (Friedel pair, centric reflection), or violated, *i.e.* $I(hkl) \neq I(\bar{h}\bar{k}\bar{l})$ (Bijvoet pair, acentric reflection). It is shown that those reflections *hkl* strictly obey Friedel's rule, for which the corresponding face form {*hkl*} is centrosymmetric. If the face form {*hkl*} is non-centrosymmetric, Friedel's rule is violated due to anomalous scattering. (ii) Crystals twinned by merohedry: determination of reflections *hkl*, the intensities of which are affected (or not affected) by the twinning. It is shown that the intensity is affected if the twin element is not a symmetry element of the *eigensymmetry* of the corresponding face form {*hkl*}. The intensity is *not* affected if the twin element belongs to the *eigensymmetry* of {*hkl*} ('affected' means that the intensities of the twin-related reflections are different for different twin domain states owing to differences either in geometric structure factors or in anomalous scattering or in both). A simple procedure is presented for the determination of these types of reflections from Tables 10.1.2.2 and 10.1.2.3 of *International Tables for Crystallography*, Vol. A [Hahn & Klapper (2002). *International Tables for Crystallography*, Vol. A, Part 10, edited by Th. Hahn, 5th ed. Dordrecht: Kluwer]. The application to crystal-structure determination of crystals twinned by merohedry (reciprocal space) and to X-ray diffraction topographic mapping of twin domains (direct space) is discussed. Relevant data and twinning relations for the 63 possible twin laws by merohedry in the 26 merohedral point groups are presented in Appendices *A* to *D*.

1. Introduction

1.1. *Eigensymmetries* of face forms

A *face form* (crystal form) {*hkl*} is a closed or open polyhedron which consists of all faces equivalent to a given face (*hkl*) with respect to a (generating) crystallographic point group. Depending on the (general and special) values of h, k, l, general, special and limiting face forms are distinguished (*cf.* Hahn & Klapper, 2002, §10.1.2.2). Each form has a well defined *eigensymmetry*[1] which is the full symmetry (inherent symmetry) of a form, considered as a polyhedron by itself. The *eigensymmetry* of a form is a proper or improper supergroup of the generating point group. Among the 47 different crystallographic face forms, 22 have centrosymmetric and 25 non-centrosymmetric *eigensymmetries*. These face forms and their *eigensymmetries* are listed in Table 1 [*cf.* also Table 10.1.2.3 in *International Tables for Crystallography*, Vol. A (Hahn & Klapper, 2002) (hereafter *IT* A)].[2] Illustrations of all 47 face forms are contained in Chapter 3.2 (pp. 184–188) of the book by Vainshtein (1994) and Chapter 10 of Buerger (1956).

In *centrosymmetric* point groups *all* face forms are centrosymmetric. In most *non-centrosymmetric* point groups *some* face forms are centrosymmetric. They are built up by pairs of faces parallel to evenfold (twofold) rotation axes or parallel to mirror planes of the generating point group, resulting in pinacoids (parallelohedra) of centrosymetric *eigensymmetry* ∞/mm. These pinacoids are further 'multiplied' by the other

[1] It is pointed out that the terms '*eigensymmetry*' and 'oriented *eigensymmetry*' are also used in another sense in twinning and domain structures. They denote the point-group symmetry of a twin component (single domain) of a twinned crystal, in contrast to the 'composite symmetry' of the twin (*cf.* §3.3.4 of Hahn & Klapper, 2003 and §2.2 of the present paper).

[2] The duals to the 47 face forms are the 47 point forms which are also listed in Table 10.1.2.3 of IT *A*; corresponding face and point forms have the same *eigensymmetry*.

Table 1
The 47 crystallographic face forms, their *eigensymmetries* and number of faces.

The centrosymmetric face forms are marked with an asterisk, all other forms are non-centrosymmetric. For the Miller indices of these forms in various point groups, see Table 2.

No.	Face form	*Eigensymmetry*	No. of faces
1	Pedion, monohedron	∞m	1
2	Pinacoid, parallelohedron	∞/mm*	2
3	Dihedron†	$mm2$	2
4	Rhombic disphenoid	222	4
5	Rhombic pyramid	$mm2$	4
6	Rhombic prism	mmm*	4
7	Rhombic dipyramid	mmm*	8
8	Tetragonal pyramid	$4mm$	4
9	Tetragonal disphenoid	$\bar{4}2m$	4
10	Tetragonal prism	$4/mmm$*	4
11	Tetragonal trapezohedron	422	8
12	Ditetragonal pyramid	$4mm$	8
13	Tetragonal scalenohedron	$\bar{4}2m$	8
14	Tetragonal dipyramid	$4/mmm$*	8
15	Ditetragonal prism	$4/mmm$*	8
16	Ditetragonal dipyramid	$4/mmm$*	16
17	Trigonal pyramid	$3m$	3
18	Trigonal prism	$\bar{6}2m$	3
19	Trigonal trapezohedron	32	6
20	Ditrigonal pyramid	$3m$	6
21	Rhombohedron	$\bar{3}m$*	6
22	Ditrigonal prism	$\bar{6}2m$	6
23	Hexagonal pyramid	$6mm$	6
24	Trigonal dipyramid	$\bar{6}2m$	6
25	Hexagonal prism	$6/mmm$*	6
26	Ditrigonal scalenohedron	$\bar{3}m$*	12
27	Hexagonal trapezohedron	622	12
28	Dihexagonal pyramid	$6mm$	12
29	Ditrigonal dipyramid	$\bar{6}2m$	12
30	Dihexagonal prism	$6/mmm$*	12
31	Hexagonal dipyramid	$6/mmm$*	12
32	Dihexagonal dipyramid	$6/mmm$*	24
33	Tetrahedron	$\bar{4}3m$	4
34	Cube, hexahedron	$m\bar{3}m$*	6
35	Octahedron	$m\bar{3}m$*	8
36	Pentagon-tritetrahedron	23	12
37	Pentagon-dodecahedron	$m\bar{3}$*	12
38	Tetragon-tritetrahedron	$\bar{4}3m$	12
39	Trigon-tritetrahedron	$\bar{4}3m$	12
40	Rhomb-dodecahedron	$m\bar{3}m$*	12
41	Didodecahedron	$m\bar{3}$*	24
42	Trigon-trioctahedron	$m\bar{3}m$*	24
43	Tetragon-trioctahedron	$m\bar{3}m$*	24
44	Pentagon-trioctahedron	432	24
45	Hexatetrahedron	$\bar{4}3m$	24
46	Tetrahexahedron	$m\bar{3}m$*	24
47	Hexaoctahedron	$m\bar{3}m$*	48

† Alternative names for the dihedron are *dome* and *sphenoid*.

symmetry elements of the generating point group and thus create a face form of centrosymmetric *eigensymmetry*. In point groups 1 and 3 without evenfold axes and mirror planes, centrosymmetric face forms do not occur.

Three examples are given:

(i) The rhombohedron has the centrosymmetric *eigensymmetry* $\bar{3}2/m$. Possible generating symmetries are the centrosymmetric point groups $\bar{3}$ and $\bar{3}2/m$ and the non-centrosymmetric point group 32.

(ii) The cube has the centrosymmetric *eigensymmetry* $4/m\bar{3}2/m$. Possible generating symmetries are all five cubic point groups 23, $2/m\bar{3}$, 432, $\bar{4}3m$ and $4/m\bar{3}2/m$.

(iii) The pinacoid (parallelohedron) has the centrosymmetric *eigensymmetry* ∞/mm. Possible generating symmetries are the centrosymmetric point groups $\bar{1}$, $2/m$, mmm, $4/m$, $4/mmm$, $\bar{3}$, $\bar{3}m$, $6/m$, $6/mmm$ and the non-centrosymmetric groups 2, m, 222, $mm2$, $\bar{4}$, 422, $\bar{4}2m$ ($\bar{4}m2$), 32, $\bar{6}$, 622, $\bar{6}2m$ ($\bar{6}m2$). The possible generating symmetries of all 47 face forms are listed in Table 10.1.2.3 of *IT* A.

In 'multi-axial' point groups (*e.g.* 222, $4mm$, $3m1$, $31m$, 622, 23) the orientations of the *eigensymmetry* elements of a face form always coincide with the orientations of the symmetry elements of the lattice point group (holohedry). In the trigonal, tetragonal and hexagonal 'mono-axial' groups (3, $\bar{3}$; 4, $\bar{4}$, $4/m$; 6, $\bar{6}$, $6/m$), however, they coincide only for *special values* of h, k, l with the orientations of the lattice symmetry elements. Such special values of h, k, l, leading to 'fixed' *eigensymmetries*, do not exist for the monoclinic point groups because the holohedry $2/m$ and the two merohedries 2 and m are all mono-axial. (This assumes that in all cases the coordinate axes a, b, c of the point group are parallel to the conventional axes a, b, c of the lattice.) In the triclinic point groups 1 and $\bar{1}$ no symmetry directions exist.

This is illustrated by the following examples:

(i) In the multi-axial point group $4mm$ all face forms $\{hk0\}$ are ditetragonal prisms with fixed *eigensymmetry* $4/m2/m2/m$. The *eigensymmetry* elements of all these prisms coincide with the elements of the lattice point group (holohedry) $4/m2/m2/m$ and, of course, with the symmetry elements of the generating symmetry $4mm$, which is a subgroup of the *eigensymmetry*.

(ii) In the mono-axial point groups 4, $\bar{4}$ and $4/m$, however, all face forms $\{hk0\}$ are tetragonal prisms. They all have the same (type of) *eigensymmetry* $4/mmm$ but different ('floating') orientations of their symmetry elements, *i.e.* different 'oriented *eigensymmetries*'. Only the orientation of their tetragonal axes (*i.e.* the common generating symmetry element) is the same. Merely for the special prisms $\{100\}$ and $\{110\}$ are the orientations of all *eigensymmetry* elements and of the lattice symmetry elements (holohedry) the same. This is illustrated in Fig. 1.

Face forms are commonly used to describe and illustrate the morphology of crystals and their symmetries geometrically. Beyond morphology, face forms are considered here as sets of symmetrically equivalent (reflecting) net planes $\{hkl\}$ and their parallel 'opposites' $\{\bar{h}\bar{k}\bar{l}\}$. We present the application of face forms and their *eigensymmetries* to two experimentally important problems concerning the intensities (structure-factor moduli) of X-ray reflections hkl. They are (i) the strict validity of Friedel's rule, $I(hkl) = I(\bar{h}\bar{k}\bar{l})$, for certain pairs of reflections of *non-centrosymmetric* crystals containing anomalous scatterers, and (ii) for crystals twinned by merohedry, the determination of twin-related reflections hkl and $h'k'l'$ the intensities of which are affected (or are not affected) by the twinning (compared with the untwinned single crystal). For a first mention of this topic see Klapper & Hahn (1987).

For these purposes the face form $\{hkl\}$ corresponding to the reflection (reflecting netplane) hkl of interest is considered. It should be noted that all higher-order reflections $nh\ nk\ nl$ (with integer n) have the same face form $\{hkl\}$. In order to bring out

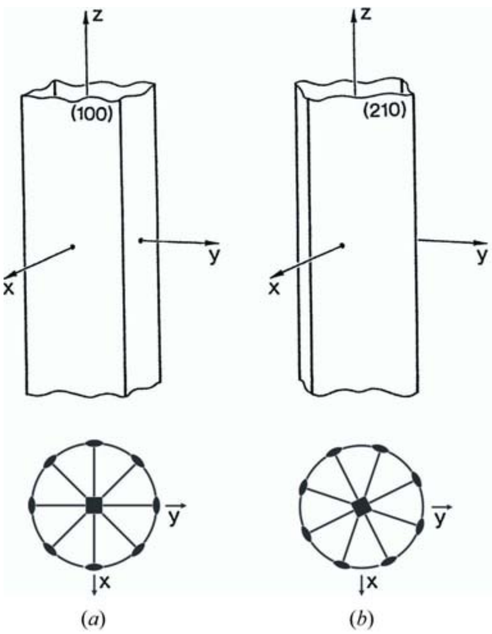


Figure 1
Face forms *tetragonal prism* {100} and {210} (generating point group 4), with two differently oriented *eigensymmetries* of type $4/m2/m2/m$ and their stereographic projections, which differ by a counter-clockwise angle of arctan (1/2) = 26.56° around the z axis. The axes x and y are the lattice directions [100] and [010]. In (*a*) the *eigenymmetry* orientation corresponds to the lattice orientation.

the correspondence between face form and reflection, we use in this treatment the higher-order notation also for face forms, *e.g.* $\{hhh\}$, $\{h0\bar{h}0\}$ or $\{00l\}$ instead of {111}, $\{10\bar{1}0\}$ and {001}, respectively.[3]

1.2. Anomalous scattering and Friedel/Bijvoet pairs

Anomalous scattering (anomalous dispersion) of atoms is always present in X-ray diffraction by crystals. The effect is small (and usually neglected) for X-ray wavelengths far from the absorption edges of all atoms in the crystal, but is substantial for wavelengths close to the absorption edge of at least one atom. Hence, the structure-factor moduli $|F(hkl)|$ do not only depend on the nature of the atoms and the crystal structure, but also on the X-ray wavelength employed.

In this paper we consider sets of reflections which are *symmetrically equivalent* under the *point group of the crystal*. The structure-factor moduli (intensities) of these reflections are always *exactly equal*, no matter whether the crystal is centrosymmetric or non-centrosymmetric or whether anomalous scattering is appreciable or not. [An algebraic proof of this equality is given by Waser (1955).] In crystal structure determination, especially of the *absolute structure*, pairs of 'opposite reflections' hkl and $\bar{h}\bar{k}\bar{l}$ are of particular importance. In *all centrosymmetric* point groups the F-moduli of these reflections are equivalent and hence *exactly equal*, whereas in *non-centrosymmetric* crystals *two cases* occur in the presence of anomalous scattering: $|F(hkl)|$ and $|F(\bar{h}\bar{k}\bar{l})|$ may be either exactly equal or different (with respect to anomalous scattering), depending on the values of h, k, l.

The following terms are used:

(i) *Friedel pair*, if $|F(hkl)| = |F(\bar{h}\bar{k}\bar{l})|$ is exactly fulfilled owing to the centrosymmetric *eigensymmetry* of the relevant face form $\{hkl\}$, even in the presence of anomalous scattering (*Friedel's rule*).

(ii) *Bijvoet pair*, if $|F(hkl)| \neq F(\bar{h}\bar{k}\bar{l})|$ owing to different anomalous scattering contributions.

These equalities or inequalities apply to the entire sets of reflections which are symmetrically equivalent to hkl and $\bar{h}\bar{k}\bar{l}$ each; hence, we extend the terms Friedel/Bijvoet pairs to Friedel/Bijvoet sets and to Friedel/Bijvoet face forms and point forms.

Note that Friedel's rule is *exactly valid* also for opposite acentric reflections hkl and $\bar{h}\bar{k}\bar{l}$ (corresponding to non-centrosymmetric opposite face forms) under special structural conditions, even in the presence of anomalous scattering. It always holds for crystals containing only *one* kind of scatterers, *i.e.* for all non-centrosymmetric element crystals. An example is provided by crystals of stable selenium and tellurium (hexagonal lattice, space groups $P3_121$ and $P3_221$). Examples of hypothetical structures with several *different* atoms are given by Iwasaki (1975).

A detailed treatment of anomalous scattering and its influence upon the diffraction intensities is provided by Rogers (1975). Brief accounts, including structure-factor diagrams, can be found in textbooks of X-ray diffraction, *e.g.* by Ladd & Palmer (1993), Woolfson (1997) and Massa (2004). Intensity statistics of 'Friedel opposites' are treated by Shmueli *et al.* (2008), Flack & Bernardinelli (2008) and Shmueli & Flack (2009).

2. Results and discussion

2.1. Intensities of Friedel- and Bijvoet-pair reflections in the presence of anomalous scattering

By consideration of 'opposite face forms' $\{hkl\}$ and $\{\bar{h}\bar{k}\bar{l}\}$, corresponding to opposite reflections hkl and $\bar{h}\bar{k}\bar{l}$, Friedel and Bijvoet pairs (sets) can be distinguished by the following rule: Friedel pairs, *i.e.* $|F(hkl)| = |F(\bar{h}\bar{k}\bar{l})|$, occur for all reflections if the *eigensymmetry* of the corresponding face form is centrosymmetric. If the *eigensymmetry* of $\{hkl\}$ is non-centrosymmetric, they form Bijvoet pairs: $|F(hkl)| \neq |F(\bar{h}\bar{k}\bar{l})|$.

This can be understood as follows. All faces of a face form $\{hkl\}$ and their corresponding reflections hkl are symmetrically equivalent with respect to the generating point group (*i.e.* the point group of the crystal under investigation), no matter whether the *eigensymmetry* of $\{hkl\}$ is higher or not. This means that *all* reflections belonging to the set $\{hkl\}$ have the same structure-factor moduli and, thus, the same intensity. (The phases of the structure factors, however, may be different owing to glide or screw components or the location of the origin.) Since a centrosymmetric face form consists of pairs of parallel and symmetrically equivalent faces $(hkl)/(\bar{h}\bar{k}\bar{l})$, the intensities of reflections hkl and $\bar{h}\bar{k}\bar{l}$ are equal (Friedel set). If

[3] As mentioned above, instead of the face forms used in this paper, the point forms, representing reciprocal-lattice points or face poles, could also be used.

the face form is non-centrosymmetric, the (opposite) sets $\{hkl\}$ and $\{\bar{h}\bar{k}\bar{l}\}$ are two non-equivalent 'inverted' face forms (with pairs of mutually parallel faces), *i.e.* their reflections differ in anomalous scattering (Bijvoet sets, Bijvoet forms).

The face form corresponding to a reflection hkl may be easily determined as follows. Find the face form $\{hkl\}$ for the relevant point group in Table 10.1.2.2 of *IT* A. The *eigensymmetries* of all 47 face forms are listed in Table 1 of the present paper (*cf.* also Table 10.1.2.3 of *IT* A).

Examples: (i) All face forms of centrosymmetric crystals are themselves centrosymmetric; thus all pairs $F(hkl)$ and $F(\bar{h}\bar{k}\bar{l})$ are Friedel pairs.

(ii) Reflection $h0\bar{h}l$ of a crystal with point group 321 (*e.g.* quartz, $GaPO_4$): from the listing of face forms of point group 321 in Table 10.1.2.2 it is found that this reflection corresponds to the face form 'rhombohedron', which has the centrosymmetric *eigensymmetry* $\bar{3}m1$. Thus, the pair of reflections $h0\bar{h}l$/$\bar{h}0h\bar{l}$ is a Friedel pair, *i.e.* also their anomalous-scattering contributions are equal.

(iii) Reflection $hh2\bar{h}l$ of the same crystal corresponds to the face form 'trigonal dipyramid' with non-centrosymmetric *eigensymmetry* $\bar{6}2m$; hence, reflections $hh2\bar{h}l$ and $\bar{h}\bar{h}2h\bar{l}$ are a Bijvoet pair, *i.e.* have different structure-factor moduli owing to different anomalous-scattering contributions; the corresponding face forms are a pair of 'morphologically inverse' trigonal bipyramids (*cf.* Fig. 3).

It is noted that the reflections with *centrosymmetric* and *non-centrosymmetric* face forms correspond to Rogers' '*centric*' and '*acentric*' (or '*sensitive*') reflections, as used in his tests to determine the absolute structure of enantiomorphic or polar crystals from diffraction intensities (Rogers, 1981; *cf.* Shmueli & Flack, 2009, Table 1). Flack, who suggests another approach ('*Flack parameter*'), uses the terms '*E reflections*' and '*non-E reflections*' for the centric and acentric reflections (Flack, 1983). Non-E reflections form Bijvoet pairs and are suitable for distinguishing between two enantiomorphic or polar forms of a crystal (absolute structure), whereas E reflections (forming Friedel pairs) are not (centric reflections). The first absolute structure was determined by Bijvoet (1949) and Bijvoet *et al.* (1951); it was recently confirmed and refined (Lutz & Schreurs, 2008).

In Table 2 the non-centrosymmetric point groups, their face forms and the corresponding X-ray reflections strictly obeying (centric reflections) or violating (acentric reflections) Friedel's rule are collected. Note that in point groups 1 and 3 no 'symmetry-enforced' Friedel pairs exist.

2.2. X-ray intensities of crystals twinned by merohedry

2.2.1. General considerations[4]. Twinning by merohedry can occur if the point-group symmetry of the crystal is a proper subgroup of its lattice symmetry (holohedry). The twin element[5] is a symmetry element of the lattice but not of the merohedral point group of the crystal (Friedel, 1926; Catti & Ferraris, 1976; Hahn & Klapper, 2003, §3.3.9). As a consequence, the lattice is mapped upon itself by the twin operation, and the lattices of the different twin components are exactly parallel to each other ('parallel-lattice twins'). Thus, all domains of the different (twin) orientation states are simultaneously in exact reflection position for all reflections hkl, *i.e.* the two reciprocal lattices are superimposed exactly.

In general, a twin operation 'interchanges' a reflection hkl of one twin domain with a symmetrically non-equivalent reflection $h'k'l'$ of the other domain (these reflections, however, would be equivalent in the holohedral point group). Since the structure-factor moduli of these reflections are, as a rule, different from each other, different domain states reflect with different intensities. Thus, the intensity diffracted from a crystal twinned by merohedry is affected by the twinning in two ways, first by the type of reflections $hkl/h'k'l'$ involved in the superposition and second by the volume fractions of the twin components. There are, however, always certain superimposed reflections, which are *not* affected by the twinning, *i.e.* have the same intensity for any volume ratio of the twin components.

Three cases of twin-related reflections $hkl/h'k'l'$, with intensities affected or not affected by twinning, are distinguished; henceforth these are called '*twin diffraction cases*' in order to avoid confusion with the term '*twin reflection*' (*twin reflection* plane) as a twin operation (twin element).

Case A. The twin element belongs to the 'oriented *eigensymmetry*' (*cf.* §1.1) of the face form $\{hkl\}$ corresponding to reflection hkl, *i.e.* the two (twin-related) face forms $\{hkl\}$ and $\{h'k'l'\}$ are symmetrically equivalent and transformed by the twin element into each other. Hence, the F-moduli of their reflections hkl and $h'k'l'$ are equal. Case A face forms may be centrosymmetric or not. An example is given in Fig. 2(*a*).

Case B. The twin element does *not* belong to the 'oriented *eigensymmetry*' of the face form $\{hkl\}$, *i.e.* the two twin-related (geometrically equal) face forms, which are transformed by the twin element into each other, are *not* symmetrically equivalent and thus their corresponding reflections hkl are not equal (Fig. 2*b*). This twin type, however, is further subdivided with respect to the contributions of the geometric (trigonometric) structure factor and of the anomalous scattering.

Case B1. Both the geometric structure factors *and* the anomalous scattering contributions of the twin-related reflections are different. In this case the F-moduli of the two reflections are different even for negligible anomalous scattering. This case corresponds to twins of type II by Catti & Ferraris (1976), *i.e.* the twin element does not belong to the Laue symmetry of the crystal but to its holohedry.

Case B2. The geometric structure factors are equal, but the anomalous-scattering contributions are different. This case occurs always for non-centrosymmetric face forms which are 'morphologically inverted' by the twin operation into their 'opposites' (Fig. 3). Case B2 is present in *inversion* twins (type I by Catti & Ferraris, 1976) for *all* non-centrosymmetric face

[4] For a review of the group-theoretical background, especially with respect to coset decomposition of a group and group–subgroup relations, the reader is referred to Wondratschek (2002), especially §§8.1.6 and 8.3.3.

[5] The singular term 'twin element' or 'twin operation' stands for any member of the 'coset of alternative twin operations' (which represents the twin law) as defined in §3.3.4 of Hahn & Klapper (2003).

Table 2
Intensity relations of Friedel opposites in the 21 non-centrosymmetric point groups in the presence of anomalous scattering.

$I(hkl) = I(\bar{h}\bar{k}\bar{l})$ is strictly obeyed ('Yes', centric reflection, Friedel pair) if the corresponding face form is centrosymmetric (Friedel's rule). The number of faces is given in parentheses. Asterisks (*) refer to the footnote at the end of the table.

Point group	Reflection	Face form (number of faces)	Centrosymmetry $I(hkl) = I(\bar{h}\bar{k}\bar{l})$
1	*hkl*	Pedion (monohedron) (1)	No
2*	*hkl*	Dihedron (sphenoid) (2)	No
(2 ∥ *b*)	*h*0*l*	Pinacoid (parallelohedron) (2)	Yes
	0*k*0	Pedion (monohedron) (1)	No
*m**	*hkl*	Dihedron (dome) (2)	No
(*m* ⊥ *b*)	*h*0*l*	Pedion (monohedron) (1)	No
	0*k*0	Pinacoid (parallelohedron) (2)	Yes
222	*hkl*	Rhombic disphenoid (4)	No
	*hk*0, *h*0*l*, 0*kl*	Rhombic prisms (4)	Yes
	*h*00, 0*k*0, 00*l*	Pinacoids (parallohedra) (2)	Yes
*mm*2	*hkl*	Rhombic pyramid (4)	No
	*hk*0	Rhombic prism (4)	Yes
	*h*0*l*, 0*kl*	Dihedra (domes) (2)	No
	*h*00, 0*k*0	Pinacoids (parallelohedra) (2)	Yes
	00*l*	Pedion (monohedron) (1)	No
4	*hkl*, *h*0*l*, *hhl*	Tetragonal pyramids (4)	No
	*hk*0, *h*00, *hh*0	Tetragonal prisms (4)	Yes
	00*l*	Pedion (monohedron) (1)	No
$\bar{4}$	*hkl*, *h*0*l*, *hhl*	Tetragonal disphenoids (4)	No
	*hk*0, *h*00, *hh*0	Tetragonal prisms (4)	Yes
	00*l*	Pinacoid (parallelohedron) (2)	Yes
422	*hkl*	Tetragonal trapezohedron (8)	No
	*h*0*l*, *hhl*	Tetragonal dipyramids (8)	Yes
	*hk*0	Ditetragonal prism (8)	Yes
	*h*00, *hh*0	Tetragonal prisms (4)	Yes
	00*l*	Pinacoid (parallelohedron) (2)	Yes
4*mm*	*hkl*	Ditetragonal pyramid (8)	No
	*h*0*l*, *hhl*	Tetragonal pyramids (4)	No
	*hk*0	Ditetragonal prism (8)	Yes
	*h*00, *hh*0	Tetragonal prisms (4)	Yes
	00*l*	Pedion (monohedron) (1)	No
$\bar{4}2m$*	*hkl*	Tetragonal scalenohedron (8)	No
	*h*0*l*	Tetragonal dipyramid (8)	Yes
	hhl	Tetragonal disphenoid (4)	No
	*hk*0	Ditetragonal prism (8)	Yes
	*h*00, *hh*0	Tetragonal prisms (4)	Yes
	00*l*	Pinacoid (parallelohedron) (2)	Yes
3*	*hkil*, $h0\bar{h}l$, $hh2\bar{h}l$	Trigonal pyramids (3)	No
(hexagonal axes)	*hki*0, $h0\bar{h}0$, $hh2\bar{h}0$	Trigonal prisms (3)	No
	000*l*	Pedion (monohedron) (1)	No
321*	*hkil*	Trigonal trapezohedron (6)	No
(hexagonal axes)	$h0\bar{h}l$	Rhombohedron (6)	Yes
	$hh2\bar{h}l$	Trigonal dipyramid (6)	No
	*hki*0	Ditrigonal prism (6)	No
	$h0\bar{h}0$	Hexagonal prism (6)	Yes
	$hh2\bar{h}0$	Trigonal prism (3)	No
	000*l*	Pinacoid (parallelohedron) (2)	Yes
3*m*1*	*hkil*	Ditrigonal pyramid (6)	No
(hexagonal axes)	$h0\bar{h}l$	Trigonal pyramid (3)	No
	$hh2\bar{h}l$	Hexagonal pyramid (6)	No
	*hki*0	Ditrigonal prism (6)	No
	$h0\bar{h}0$	Trigonal prism (3)	No
	$hh2\bar{h}0$	Hexagonal prism (3)	Yes
	000*l*	Pedion (monohedron) (1)	No
6	*hkil*, $h0\bar{h}l$, $hh2\bar{h}l$	Hexagonal pyramids (6)	No
	*hki*0, $h0\bar{h}0$, $hh2\bar{h}0$	Hexagonal prisms (6)	Yes
	000*l*	Pedion (monohedron) (1)	No
$\bar{6}$	*hkil*, $h0\bar{h}l$, $hh2\bar{h}l$	Trigonal dipyramids (6)	No
	*hki*0, $h0\bar{h}0$, $hh2\bar{h}0$	Trigonal prisms (3)	No
	000*l*	Pinacoid (parallelohedron) (2)	Yes
622	*hkil*	Hexagonal trapezohedron (12)	No
	$h0\bar{h}l$, $hh2\bar{h}l$	Hexagonal dipyramids (12)	Yes
	*hki*0	Dihexagonal prism (12)	Yes
	$h0\bar{h}0$, $hh2\bar{h}0$	Hexagonal prisms (6)	Yes
	000*l*	Pinacoid (parallelohedron) (2)	Yes

forms, as well as in *rotation* and *reflection* twins for *certain* non-centrosymmetric face forms (*cf*. Appendix *B*).

These three cases and their effects on structure determination (reciprocal space) and X-ray diffraction topography (direct space) are summarized in Table 3. In the following two sections they are illustrated by two crystals exhibiting merohedral twins with three different twin laws: by quartz SiO_2 and its homeotypes $AlPO_4$ (berlinite), $GaPO_4$ *etc.* (point group 321), and by $KLiSO_4$ III (point group 6). Cubic merohedral twins are treated in §2.2.4.

2.2.2. Twinning of quartz SiO_2 and its homeotypes. Quartz [point group 321, lattice symmetry (holohedry) 6/*m*2/*m*2/*m*] provides a particularly illustrative example because it exhibits three different kinds of merohedral twinning: *Brazil twins* (inversion twins, type I by Catti & Ferraris, 1976), Dauphiné twins and combined (Leydolt) twins (both type II by Catti & Ferraris, 1976).

(*a*) Brazil twins [inversion twins, composite (twin) symmetry $\bar{3}2/m1$]. A twin is an 'inversion twin' if there is an inversion $\bar{1}$ among the alternative twin operations each of which represents the twin law (*cf*. the footnotes to §2.2.1). Thus, the six alternative twin operations of the Brazil twin of quartz are the three mirror planes {11$\bar{2}$0} normal to the twofold symmetry axes of 321, and the three roto-inversions $\bar{3}$ around the threefold axis: $\bar{3}^1$, $\bar{3}^3 = \bar{1}$, $\bar{3}^5 = \bar{3}^{-1}$, classifying this twin as an inversion twin.

For the influence of this twinning on the X-ray intensities of reflections *hkil* two cases are distinguished:

(i) The face form is centrosymmetric: the superimposed intensities are not affected, even if anomalous scattering is taken into account (twin diffraction case A). For Brazil twins these reflections are {$h0\bar{h}l$} (rhombohedron), {$h0\bar{h}0$} (hexagonal prism) and {000l} (pinacoid).

(ii) The face form is non-centrosymmetric: the superimposed intensities of reflections *hkil* *versus* $\bar{h}\bar{k}\bar{i}\bar{l}$ are different, but only due to anomalous scattering (Bijvoet sets, case B2). For the Brazil twin these reflections are {*hkil*} (trigonal trapezohedron), {$hh2\bar{h}l$} (trigonal dipyramid), {*hki*0} (ditrigonal prism) and {$hh2\bar{h}0$} (trigonal prism). As for all inversion twins, case B1 reflections do not occur.

(*b*) Dauphiné twins (composite symmetry 622). The twin law is usually described as 'twofold twin axis parallel to the threefold symmetry axis'. Alternative twin operations,

Table 2 (continued)

Point group	Reflection	Face form (number of faces)	Centrosymmetry $I(hkl) = I(\bar{h}\bar{k}\bar{l})$
$6mm$	$hkil$	Dihexagonal pyramid (12)	No
	$h0\bar{h}l$, $hh2\bar{h}l$	Hexagonal pyramids (6)	No
	$hki0$	Dihexagonal prisms (12)	Yes
	$h0\bar{h}0$, $hh2\bar{h}0$	Hexagonal prisms (6)	Yes
	$000l$	Pedion (monohedron) (1)	No
$\bar{6}2m$*	$hkil$	Ditrigonal dipyramid (12)	No
	$h0\bar{h}l$	Hexagonal dipyramid (12)	Yes
	$hh2\bar{h}l$	Trigonal dipyramid (6)	No
	$hki0$	Ditrigonal prism (6)	No
	$h0\bar{h}0$	Hexagonal prism (6)	Yes
	$hh2\bar{h}0$	Trigonal prism (3)	No
	$000l$	Pinacoid (parallelohedron) (2)	Yes
23	hkl	Pentagon-tritetrahedron (12)	No
	hhl, $\lvert h\rvert < \lvert l\rvert$	Trigon-tritetrahedron (12)	No
	hhl, $\lvert h\rvert > \lvert l\rvert$	Tetragon-tritetrahedon (12)	No
	$hk0$	Pentagon-dodecahedron (12)	Yes
	$hh0$	Rhomb-dodecahedron (12)	Yes
	hhh	Tetrahedron) (4)	No
	$h00$	Cube (hexahedron) (6)	Yes
432	hkl	Pentagon-trioctahedron (24)	No
	hhl, $\lvert h\rvert < \lvert l\rvert$	Tetragon-trioctahedron (24)	Yes
	hhl, $\lvert h\rvert > \lvert l\rvert$	Trigon-trioctahedron (24)	Yes
	$hk0$	Tetrahexahedron (24)	Yes
	$hh0$	Rhomb-dodecahedron (12)	Yes
	hhh	Octahedron (8)	Yes
	$h00$	Cube (hexahedron) (6)	Yes
$\bar{4}3m$	hkl	Hexatetrahedron (24)	No
	$hk0$	Tetrahexahedron (24)	Yes
	hhl, $\lvert h\rvert < \lvert l\rvert$	Trigon-tritetrahedron (12)	No
	hhl, $\lvert h\rvert > \lvert l\rvert$	Tetragon-tritetrahedron (12)	No
	$hh0$	Rhomb-dodecahedron (12)	Yes
	hhh	Tetrahedron) (4)	No
	$h00$	Cube (hexahedron) (6)	Yes

* Each of these point groups is represented here by only one structural setting. For the monoclinic groups 2 and m the 'unique axis c' setting is not included. The settings $\bar{4}m2$, 312, $31m$, $\bar{6}m2$, as well as the 'rhombohedral-axes' settings of 3, 32 and $3m$, are not given for reasons of compactness. In all these (not included) settings, face forms, multiplicities and Yes/No entries agree with the settings contained in this table because these data do *not* change if another reference system is chosen. Only the *hkl* or *hkil* indices depend on the setting and can easily be obtained as follows. In the settings $\bar{4}2m$ and $\bar{4}m2$ the reflection indices of the pairs $h0l/hhl$ and $h00/hh0$ are interchanged, in the settings 321/312, $3m1/31m$, $\bar{6}2m/\bar{6}m2$ the reflection indices of the pairs $h0\bar{h}l/hh2\bar{h}l$ and $h0\bar{h}0/hh2\bar{h}0$ are interchanged (*cf.* Table 10.1.2.2 of IT *A*). Statistical and symmetry data for the different types of 'Friedel opposites' for all non-centrosymmetric point groups were recently provided by Shmueli & Flack (2009).

representing the same twin law, are rotations by 60°, 180°, 300° (*i.e.* rotations 6^1, $6^3 = 2$, 6^5) around [001] and the three twofold rotations 2[120], 2[210], $2[1\bar{1}0]$ between the twofold symmetry axes of 321 (*cf.* Hahn & Klapper, 2003, p. 404). The following three types of face forms, corresponding to the three twin diffraction cases A, B1 and B2, are distinguished:

(i) Face forms $\{h0\bar{h}0\}$ (hexagonal prism, *eigensymmetry* $6/mmm$) and $\{000l\}$ (pinacoid, *eigensymmetry* ∞/mm) are centrosymmetric *and* contain the twin elements as *eigensymmetry* elements. The corresponding sets of twin-related reflections are equivalent (case A) and, hence, not influenced by the twinning, not even in the presence of anomalous scattering.

(ii) Face forms $\{hkil\}$ (trigonal trapezohedron, *eigensymmetry* 321) and $\{h0\bar{h}l\}$ (rhombohedron, *eigensymmetry* $\bar{3}2/m1$): the (alternative) twin operations do *not* belong to the *eigensymmetries* of these forms. This twinning transforms the 'large rhombohedron' $\{10\bar{1}1\}$ of quartz into the non-equivalent 'small rhombohedron' $\{\bar{1}011\}$ and *vice versa*. The intensities of the corresponding reflections are affected by the twinning (case B1) due both to different geometrical structure factors *and* different anomalous scattering contributions.

(iii) Face forms $\{hh2\bar{h}l\}$ (trigonal dipyramid), $\{hki0\}$ (ditrigonal prism) and $\{hh2\bar{h}0\}$ (trigonal prism) possess the *eigensymmetry* $\bar{6}2m = 3/m2m$. The twofold twin axis does *not* belong to this symmetry, and the corresponding reflections are modified by this twinning. This case, however, is special because the combination of the *eigen* mirror plane in $\bar{6}$ (= $3/m$) of the face forms with the twofold twin axis normal to it generates the twin inversion. Thus, the twin-related face forms are pairs of 'inverted' polyhedra, and the Dauphiné twinning has for these special forms the same effect as an inversion twin. This is illustrated in Figs. 3(*a*) and explained in more detail in Appendix *B*. Consequently, the intensities of the corresponding reflections are affected only by the different anomalous-scattering contributions (case B2).

(*c*) Combined (Leydolt, Liebisch) twins (composite symmetry $\bar{6}2m = 3/m2m$). The twin law is represented by the coset of the following six alternative twin operations: three twin reflections across the planes $\{10\bar{1}0\}$ and three rotoinversions $\bar{6}$ around [001]: $\bar{6}^1$, $\bar{6}^3 = m(0001)$, $\bar{6}^5 = \bar{6}^{-1}$. The twin reflection plane $m(0001)$ normal to the threefold axis is the most illustrative twin element.

(i) The twin element belongs to the *eigensymmetry* $\bar{6}2m$ of the forms $\{hh2\bar{h}l\}$ (trigonal bipyramid), $\{hki0\}$ (ditrigonal prism), $\{h0\bar{h}0\}$ (hexagonal prism), $\{hh2\bar{h}0\}$ (trigonal prism) and $\{000l\}$ (pinacoid, *eigensymmetry* ∞/mm); thus, the corresponding twin-related reflections are equivalent and not affected by the twinning (case A).

(ii) The *eigensymmetries* of the face forms $\{hkil\}$ (trigonal trapezohedron, 321) and $\{h0\bar{h}l\}$ (rhombohedron, $\bar{3}2/m1$) do not contain the twin element: the corresponding reflections differ both in geometric structure factors and anomalous-scattering contributions (case B1). Case B2 does not occur in this twinning.

The intensity characteristics of the seven types of reflections for the three merohedral twin laws of quartz are summarized in Table 4 and derived in Appendix *C*.

2.2.3. Twinning of $KLiSO_4$ III. The room-temperature phase III of $KLiSO_4$ has the mono-axial point group 6 (lattice symmetry $6/mmm$) and forms three kinds of twins according to the laws 'twin reflection plane parallel to [001]', 'twofold twin axis normal to [001]' and 'twin reflection plane normal to [001]' (inversion twin). The two former are type II twins, the latter is a type I twin by Catti & Ferraris (1976). This example was chosen because rich experimental data are available (Klapper *et al.*, 1987) and because of the complexity of the twinning owing to its mono-axial point group.

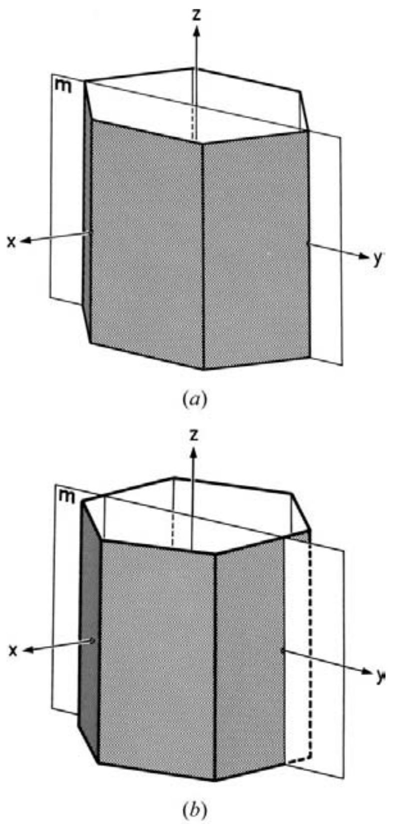


Figure 2
Face forms *hexagonal prism* $\{h0\bar{h}0\}$ (special) (*a*) and $\{hki0\}$ (general) (*b*), generating point group 6, with two differently oriented *eigensymmetries* of type $6/m2/m2/m$. In (*a*) the twin mirror plane *m* belongs to the oriented *eigensymmetry*, in (*b*) not. Hence, reflections of type $\{hki0\}$ are affected by the twinning, reflections $\{h0\bar{h}0\}$ are not (*cf.* Fig. 5). The other special hexagonal prism $\{hh2\bar{h}0\}$ contains the twin mirror plane in its *eigensymmetry* too: hence, reflections $\{hh2\bar{h}0\}$ are also not affected by the twinning.

(*a*) Twin law 'twin reflection plane parallel [001]' (composite symmetry $6mm$). The six (*i.e.* 3 + 3) twin mirror planes $\{10\bar{1}0\}$ and $\{11\bar{2}0\}$ parallel to the sixfold axis are 'alternative' twin elements. Since 6 is a mono-axial point group, the *oriented eigensymmetries* of the face forms have to be considered.

(i) Face forms $\{h0\bar{h}l\}$ and $\{hh2\bar{h}l\}$ (both hexagonal pyramids with the same oriented *eigensymmetry* $6mm$), $\{h0\bar{h}0\}$ and $\{hh2\bar{h}0\}$ (both hexagonal prisms with the same oriented *eigensymmetry* $6/mmm$), and $\{000l\}$ (pedion or monohedron, *eigensymmetry* ∞m) contain the twin elements within their *eigensymmetries* [*cf.* Fig. 2(*a*) for face form $\{h0\bar{h}0\}$], *i.e.* the corresponding superimposed sets of reflections are equivalent and thus not affected by this twinning; their intensities are the same for any volume ratio of the twins and do not generate twin domain contrast in X-ray topography of the twins (case A reflections).

(ii) Face forms $\{hkil\}$ (hexagonal pyramids, all with differently oriented *eigensymmetries* of type $6mm$) and $\{hki0\}$ (hexagonal prisms, all with differently oriented *eigensymmetries* of type $6/mmm$): the oriented *eigensymmetries* do *not* contain the twin mirror planes [*cf.* Fig. 2(*b*) for a prism $\{hki0\}$], *i.e.* the intensities of the corresponding superimposed non-equivalent reflections change with the volume ratio of the twins. In X-ray topography they generate domain contrast (case B1 reflections; case B2 reflections do not occur).

(*b*) Twin law 'twofold twin axis normal to [001]' (composite symmetry 622). The six (*i.e.* 3 + 3) alternative twin elements are the twofold twin axes along ⟨100⟩ and ⟨120⟩.

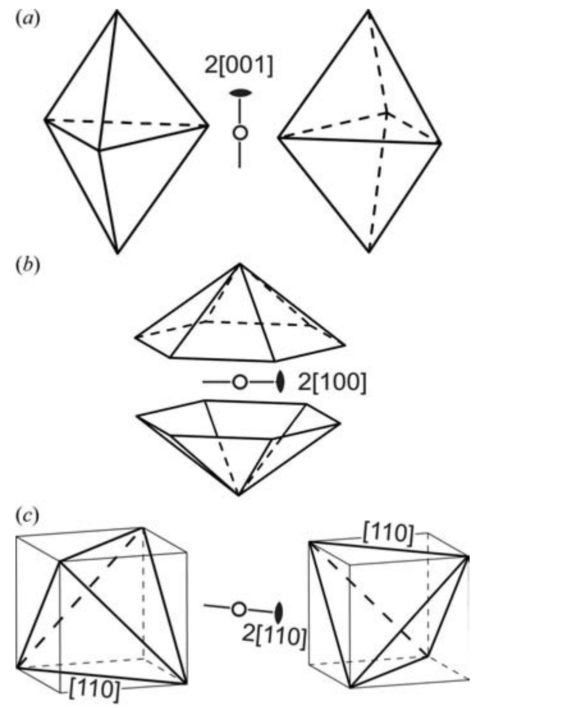


Figure 3
'Morphological (geometrical) inversion' of non-centrosymmetric face forms, illustrated for three selected examples. (*a*) Face forms *trigonal dipyramid* $\{hh\overline{2h}l\}$ and $\{\bar{h}\bar{h}2hl\}$ (*e.g.* of quartz, point group 321, *cf.* §2.2.2), *morphologically* related by a twofold axis parallel [001] (rotation twins), but also by a *morphological* inversion centre $\bar{1}$ indicated by a small circle (inversion twins). The combination of these two forms results in the centrosymmetric form *hexagonal dipyramid*. (*b*) Face forms *hexagonal pyramid* $\{h0\bar{h}l\}$ and $\{h0\bar{h}\bar{l}\}$ (*e.g.* of $KLiSO_4$, point group 6, *cf.* §2.2.3), *morphologically* related by a twofold axis parallel [100] (rotation twin), as well as by a mirror plane (0001) (reflection twin), but also by a *morphological* inversion centre $\bar{1}$ (inversion twin). Their combination results in the centrosymmetric form *hexagonal dipyramid*. (*c*) The same for *tetrahedra* $\{hhh\}$ and $\{\bar{h}\bar{h}\bar{h}\}$ with the centrosymmetric combination *octahedron*. The X-ray diffraction sets corresponding to these forms are affected by both rotation (or reflection) twinning as well as by inversion twinning (twin diffraction cases B2, defined in §2.2.1). These pairs of non-centrosymmetric face forms are called here '*morphologically (geometrically) inverted*' forms, independent of whether the two related crystal structures are inverted (opposite handedness) or not (equal handedness) (*cf.* Appendices *B* and *C*). These inverted forms correspond to sets of 'opposite reflections' as used by Flack & Bernardinelli (1999, 2008). Note that a morphological inversion of the three given face forms can also be achieved by twin reflection planes: in (*a*) by the twin planes $\{11\bar{2}0\}$, in (*b*) by the plane (0001) and in (*c*) by the planes {100}. In these cases the twin reflection planes are normal to a twofold symmetry axis of the generating point groups 321 (*a*), 6 (*b*) and 23 or $\bar{4}3m$ (*c*) (and thus also of the face-form *eigensymmetries*). Hence, these twins are inversion twins with the mirror plane as an alternative twin element of the twin coset and, thus, also exhibit inverted domain states.

Table 3
Sensitivity of twin-related reflections $hkl/h'k'l'$ of face forms $\{hkl\}$ to twinning by merohedry and its effect on intensities of superimposed reflections and X-ray topographic domain contrast.

Twin element part of *eigensymmetry* of $\{hkl\}$	Geometrical (trigonometric) structure factors	Contributions of anomalous scattering to $\|F(hkl)\|$ and $\|F(h'k'l')\|$	Sensitive to twinning	Intensities of superimposed reflections different†	Domain contrast in X-ray topography
Yes (case A)	Equal	Equal	No	No	No
No (case B1)	Different	Different	Yes	Yes	Yes
No (case B2)	Equal	Different	Yes	Yes	Yes

† 'Yes' also means that the intensity of the superimposed reflection depends on the volume ratio of the twin components.

Table 4
The seven face forms of point group 321 and of the composite symmetries of Brazil, Dauphiné and combined (Leydolt) twins of quartz.

The names of the face forms of the twin composite symmetries, which differ from the forms of untwinned point group 321, are printed in bold. They have twice the number of faces and correspond to X-ray reflections modified by the twinning (*i.e.* their intensities are different in different twin domain states, type B reflections). The reflections corresponding to the forms marked with an asterisk (*) differ only by anomalous scattering (Bijvoet forms, type B2). Unchanged form names imply unchanged intensities (Friedel forms, type A). The labels I and II refer to different orientations of the same (type of) face form, here hexagonal dipyramids and prisms rotated by 30° (or 90°) around the hexagonal axis.

Miller–Bravais indices	Untwinned Point group 321	Brazil twin Composite symmetry $\bar{3}2/m1$		Dauphiné twin 622		Combined (Leydolt) twin $\bar{6}2m$	
$\{hkil\}$	Trigonal trapezohedron	**Ditrigonal scalenohedron**	B1	**Hexagonal trapezohedron**	B1	**Ditrigonal dipyramid**	B1
$\{h0\bar{h}l\}$	Rhombohedron	Rhombohedron	A	**Hexagonal dipyramid I**	B1	**Hexagonal dipyramid I**	B1
$\{hh2\bar{h}l\}$	Trigonal dipyramid	**Hexagonal dipyramid II (*)**	B2	**Hexagonal dipyramid II (*)**	B2	Trigonal dipyramid	A
$\{hki0\}$	Ditrigonal prism	**Dihexagonal prism (*)**	B2	**Dihexagonal prism (*)**	B2	Ditrigonal prism	A
$\{h0\bar{h}0\}$	Hexagonal prism I	Hexagonal prism I	A	Hexagonal prism I	A	Hexagonal prism I	A
$\{hh2\bar{h}0\}$	Trigonal prism	**Hexagonal prism II (*)**	B2	**Hexagonal prism II (*)**	B2	Trigonal prism	A
$\{000l\}$	Pinacoid	Pinacoid	A	Pinacoid	A	Pinacoid	A

(i) Face forms $\{h0\bar{h}0\}$ and $\{hh2\bar{h}0\}$ (both hexagonal prisms with the same oriented *eigensymmetry* $6/mmm$) contain the twin elements within their *eigensymmetries*, *i.e.* the corresponding superimposed reflections are equivalent and not affected by this twinning (case A).

(ii) Face forms $\{hkil\}$ (hexagonal pyramids, all with differently oriented *eigensymmetries* $6mm$) and $\{hki0\}$ (hexagonal prisms, all with differently oriented *eigensymmetries* $6/mmm$): the oriented *eigensymmetries* do *not* contain the twofold twin axes; the corresponding superimposed non-equivalent reflections are affected by the twinning (case B1), *i.e.* they differ both in their geometric structure factors and their anomalous-scattering contribution.

(iii) Face forms $\{h0\bar{h}l\}$ and $\{hh2\bar{h}l\}$ [both 'special' hexagonal pyramids ('apices up') with the same oriented *eigensymmetry* $6mm$] do *not* contain the twin elements in their *eigensymmetry*. In these special cases, however, in contrast to (ii) above, the twin axis generates the 'inverted' pyramids $\{\bar{h}0h\bar{l}\}$ and $\{\bar{h}\bar{h}2h\bar{l}\}$ with 'apices down' (*cf.* Fig. 3*b*). The intensities of these superimposed reflections are modified only by different anomalous-scattering contributions (case B2). The same applies to the pedion $(000l)$ (*eigensymmetry* ∞m). For these special sets of reflections the twin law (*b*) appears as an inversion twin.

(*c*) Twin law 'twin reflection plane normal to [001]' (composite symmetry $6/m$, inversion twin). The coset of twin operations consists of $\bar{6}^1$, $\bar{6}^3 = m(0001)$, $\bar{6}^5$, $\bar{3}^1$, $\bar{3}^3 = \bar{1}$, $\bar{3}^5$.

(i) The general hexagonal prisms $\{hki0\}$ and the two special hexagonal prisms $\{h0\bar{h}0\}$ and $\{hh2\bar{h}0\}$ (oriented *eigensymmetry* $6/mmm$) all *include* the twin law in their *eigensymmetry* and, hence, represent twin diffraction case A, *i.e.* they are not affected by the twinning.

(ii) The general hexagonal pyramids $\{hkil\}$ and the special hexagonal pyramids $\{h0\bar{h}l\}$ and $\{hh2\bar{h}l\}$ (differently oriented *eigensymmetries* $6mm$) do *not* contain the twin element in their *eigensymmetries*, but every twin element generates the 'inverted' pyramids $\{\bar{h}\bar{k}\bar{i}\bar{l}\}$ *etc.*, thus forming case B2 reflections. The same applies to the pedion $(000l)$ with *eigensymmetry* ∞m. As in all inversion twins, case B1 reflections do not occur.

The intensity characteristics of the three merohedral twin laws above are summarized in Table 5.

2.2.4. Cubic merohedral twins of point group 23. To complement the previous trigonal and hexagonal examples, the cubic cases are treated in this section. Point group 23 (order 12), the lowest-symmetry group of the cubic system, can form three kinds of merohedral twins, each of index 2.

(i) Inversion twin, composite symmetry $2/m\bar{3}$ (order 24); coset of 12 alternative twin operations: three reflections $m\{100\}$, four $\bar{3}^1$ and four $\bar{3}^5$ rotoinversions around the four directions $\langle 111\rangle$ and $\bar{3}^3 = \bar{1}$ (inversion).

(ii) Rotation twin with 90° rotation around [100], composite symmetry 432 (order 24); coset: three 4^1 (90°) and three 4^3 (270°) rotations around $\langle 100\rangle$, six twofold rotations around $\langle 110\rangle$.

(iii) Reflection twin with twin mirror plane (110), composite symmetry $\bar{4}3m$ (order 24); coset: six reflections $m\{110\}$, three $\bar{4}^1$ and three $\bar{4}^3$ rotoinversions around $\langle 100\rangle$.

Table 5
Point group 6: the same data as in Table 4 for the three twin cases 6 → 6/*m* (inversion twin), 6 → 622 (rotation twin) and 6 → 6*mm* (reflection twin) of $KLiSO_4$.

Miller–Bravais indices	Untwinned Point group 6	Inversion twin Composite symmetry 6/*m*		Rotation twin 622		Reflection twin 6*mm*	
{*hkil*}	Hexagonal pyramid	**Hexagonal dipyramid (*)**	B2	**Hexagonal trapezohedron**	B1	**Dihexagonal pyramid**	B1
{$h0\bar{h}l$}	Hexagonal pyramid I	**Hexagonal dipyramid I (*)**	B2	**Hexagonal dipyramid I (*)**	B2	Hexagonal pyramid I	A
{$hh\overline{2h}l$}	Hexagonal pyramid II	**Hexagonal dipyramid II (*)**	B2	**Hexagonal dipyramid II (*)**	B2	Hexagonal pyramid II	A
{*hki*0}	Hexagonal prism	Hexagonal prism	A	**Dihexagonal prism**	B1	**Dihexagonal prism**	B1
{$h0\bar{h}0$}	Hexagonal prism I	Hexagonal prism I	A	Hexagonal prism I	A	Hexagonal prism I	A
{$hh\overline{2h}0$}	Hexagonal prism II	Hexagonal prism II	A	Hexagonal prism II	A	Hexagonal prism II	A
{000*l*}	Pedion	**Pinacoid (*)**	B2	**Pinacoid (*)**	B2	Pedion	A

Table 6
The seven reflection types of cubic point groups, multiplicities and *eigensymmetries* of the corresponding face forms in the untwinned group 23 and in the twin composite groups $2/m\bar{3}$, 432 and $\bar{4}3m$†.

For the three composite groups the twin reflection cases A, B1 and B2 (*cf.* §2.2.1) are given.

	Untwinned group 23		Composite group $2/m\bar{3}$			Composite group 432			Composite group $\bar{4}3m$		
Reflection types	Multipl.	*Eigensym.*	Multipl.	*Eigensym.*	Twin refl.	Multipl.	*Eigensym.*	Twin refl.	Multipl.	*Eigensym.*	Twin refl.
{*hkl*}	12	23	24	$m\bar{3}$	B2	24	432	B1	24	$\bar{4}3m$	B1
{*hhl*} $h < l$	12	$\bar{4}3m$	24	$m\bar{3}m$	B2	24	$m\bar{3}m$	B2	12	$\bar{4}3m$	A
{*hhl*} $h > l$	12	$\bar{4}3m$	24	$m\bar{3}m$	B2	24	$m\bar{3}m$	B2	12	$\bar{4}3m$	A
{*hk*0}	12	$m\bar{3}$	12	$m\bar{3}$	A	24	$m\bar{3}m$	B1	24	$m\bar{3}m$	B1
{*hh*0}	12	$m\bar{3}m$	12	$m\bar{3}m$	A	12	$m\bar{3}m$	A	12	$m\bar{3}m$	A
{*h*00}	6	$m\bar{3}m$	6	$m\bar{3}m$	A	6	$m\bar{3}m$	A	6	$m\bar{3}m$	A
{*hhh*}	4	$\bar{4}3m$	8	$m\bar{3}m$	B2	8	$m\bar{3}m$	B2	4	$\bar{4}3m$	A

† For the names of the face forms and their *eigensymmetries*, see the cubic point groups in Tables 10.1.2.2 and 10.1.2.3 of *IT* A.

These three cases are characterized in Table 6.[6]

Concerning (i): the twinning $23 \rightarrow 2/m\bar{3}$ is the simple case of an inversion twin with two domain states of opposite handedness, involving only twin diffraction cases A and B2, *i.e.* reflections not twin-affected (A) and twin-affected by anomalous scattering alone (B2).

Concerning (ii): the second case, $23 \rightarrow 432$, is a more complicated pure rotation twin preserving the handedness of the twin domains. All three twin diffraction cases A, B1 and B2 are present. Case B1 involves both different geometrical structure factors and different anomalous-scattering contributions.

Concerning (iii): the third case, $23 \rightarrow \bar{4}3m$, is a reflection twin with opposite handedness of the domain states. Here only the twin diffraction cases A and B1 occur.

Cases B2 of the two merohedral twins (i) and (ii) above involve 'morphologically inverted' face forms. This is illustrated for the tetrahedron {*hhh*} in Fig. 3(*c*). Morphologically inverted forms are discussed in detail under the aspect of equal and opposite handedness of the structure in Appendix *B*.

2.2.5. Summary of twinning by merohedry (Σ1 twins). The occurrence of the three diffraction cases, A, B1 and B2, in inversion and non-inversion twinning of the 26 types of merohedral point groups[7] is summarized as follows (*cf.* Appendix *A* for the 35 'structural settings' of these groups).

(i) *Inversion* twins, possible only in the 21 *non-centrosymmetric* merohedral point groups:

Possible twin diffraction cases:

A: twin-related sets of reflections have exactly equal *F*-moduli;

B2: twin-related sets of reflections have equal geometric (trigonometric) structure factors but differ in their anomalous-scattering contributions (Bijvoet sets).

Point groups 3 and 1: only case B2 is possible.

(ii) *Non-inversion* twins in the five *centrosymmetric* merohedral groups $4/m$, $\bar{3}$, $\bar{3}2/m$ (hexagonal lattice), $6/m$ and $2/m\bar{3}$:

Possible twin diffraction cases:

A: as above;

B1: twin-related sets of reflections differ in both their geometric structure factors and their anomalous-dispersion contributions.

(iii) *Non-inversion* twins in the 16 *non-centrosymmetric* merohedral groups of the tetragonal, trigonal, hexagonal and cubic systems:[8]

Possible twin diffraction cases:

A, B1, B2: all as above.

It is emphasized that the above rules, which are based on symmetry considerations alone, do *not* give any information on the degree by which the structure-factor moduli of the (superimposed) reflections may differ. The difference is dependent both on the individual structure-factor moduli of the twin-related reflections *hkl* and $h'k'l'$ and on the volume

[6] It should be mentioned that the three cubic twin composite groups $2/m\bar{3}$, 432 and $\bar{4}3m$ (all order 24) can further twin to the cubic holohedry $m\bar{3}m$ (order 48).

[7] Here the 'rhombohedral holohedry' $\bar{3}2/m$ is included as a merohedry of the hexagonal holohedry $6/m2/m2/m$.

[8] In the triclinic, monoclinic and orthorhombic point groups only merohedral inversion twins are possible, case (i).

ratio of the twin domain states. Note that for a twin volume ratio near 50:50, twin-related reflections exhibit equal intensities and, hence, twins of types (ii) and (iii) above may simulate a higher-symmetric Laue class, whereas twins of type (i) may simulate a centrosymmetric (disordered) structure. In X-ray diffraction topography, however, the volume ratio does not play a role, because the different mapped twin domains are spatially resolved (direct space). Here the 'domain contrast' (*i.e.* the difference between the *F*-moduli of twin-related reflections) may be strong or weak or not detectable (the latter for inversion twins, case B2, if anomalous scattering is negligible). Examples are given in §3.3.

2.3. Finding reflections affected by merohedral twinning

For several purposes it is desirable to know which reflections are affected by a twin element (twin operation) of a given merohedral point group. In the following we suggest a very simple procedure to find these reflections by using Table 10.1.2.2 of *IT* A. First, the composite symmetry of the twin can be determined by combining the point group of the untwinned crystal with the twin operation and establish the supergroup, which is, for merohedral twins, always minimal of index 2.[9] This is, for example, easily done by adding a twin element to the stereographic projection of the point group given in Table 10.1.2.2 and thus generating the stereographic projection of the 'twin-composite' point group. Similarly, the group–subgroup Table 10.1.3.2 of *IT* A may be used. Now the lists of the face forms of the 'untwinned' point group and the 'twin-composite' point group have to be compared. If, for the same type of Miller indices $\{hkl\}$, the face forms are the same (same name, same number of faces), the twin element belongs to the *eigensymmetry* of this form (diffraction case A), and the corresponding set of reflections is *not* modified by the twinning. If, however, the two face forms of the untwinned and the composite symmetry are different, the corresponding *F*-moduli are affected by the twinning (case B) in two different ways. If the two twin-related forms (of the untwinned point group) are non-centrosymmetric and 'morphologically inverted' polyhedra (*cf.* Fig. 3), case B2 exists. If the two twin-related forms are not 'inverted', case B1 applies, *i.e.* both their geometric (trigonometric) structure factors and their anomalous-scattering contributions are different. Note that these procedures are only correct if both point groups are referred to the same coordinate system.

This procedure is illustrated in condensed form in Table 4, where the face forms are listed for point group 321 and the 'twin-composite' groups of Brazil, Dauphiné and combined (Leydolt) twins of quartz, with the representative twin elements 'twin inversion centre $\bar{1}$' (composite symmetry $\bar{3}2/m1$), 'twofold twin axis along [001]' (composite symmetry 622) and 'twin reflection plane parallel (0001)' (composite symmetry $\bar{6}2m$), respectively. All reflections for which the corresponding face forms of the composite group differ from those of the 'untwinned' group are modified by the twinning, and their face forms are printed in bold face.

In Appendix *D* the types of reflections *hkl* affected and not affected by merohedral twinning are listed for all 63 possible twin laws in all 26 merohedral crystallographic point groups.

[9] Note, however, that superposition of two or three independent merohedral twin laws, *i.e.* twins of twins, can occur, leading to (composite) supergroups of index 4 and even 8, *e.g.* $4 \rightarrow 4/m \rightarrow 4/mmm$ or $3 \rightarrow \bar{3} \rightarrow 6/m \rightarrow 6/mmm$.

3. Applications

3.1. Determination of the absolute structure

The determination of the 'absolute structure' of non-centrosymmetric enantiomorphic crystals containing chiral molecules or atomic groups, or of polar crystals ('absolute orientation') requires measurement and interpretation of the (often small) differences due to anomalous scattering in many $|F(hkl)| \neq |F(\bar{h}\bar{k}\bar{l})|$ Bijvoet pairs. §2.1 provides complete and simple geometric methods for the selection of those sets of reflections which form Bijvoet pairs, *i.e.* are suitable for the determination of the absolute structure ('acentric' or 'sensitive' or E-reflections; Rogers, 1975, 1981; Flack, 1983; Flack & Bernardinelli, 1999, 2008; Shmueli *et al.*, 2008; Shmueli & Flack, 2009).

3.2. Structure determination of crystals twinned by merohedry

In single-crystal structure determination twinning is a frequently encountered serious problem for non-merohedral twins ('*ferroelastic*' twins), but even more for merohedral ('*non-ferroelastic*') twins (*cf.* Janovec & Přívratská, 2003, §3.4.2.2.1). In the 'ferroelastic' case certain diffraction spots split, and the split intensities can be measured separately if the spots are sufficiently resolved, and the volume ratio of the twin states can be measured experimentally. The distribution of non-split and split spots and the direction of their splitting allows one to identify the twin law involved. In crystals twinned by merohedry the reflections (diffraction spots) of differently oriented twin domains coincide completely and exactly, and their intensities cannot be measured separately. If the twin law is known, the reflections modified by the twinning are easily determined as shown in §2.3. In turn, if the affected reflections are known from the diffraction data, the twin law can be identified. An intermediate situation occurs for twins by pseudo-merohedry. In this border case of 'ferroelastic' twins the splitting of diffraction spots is too small to permit a separate measurement of their intensities, at least for low-index reflections, *i.e.* small diffraction angles.

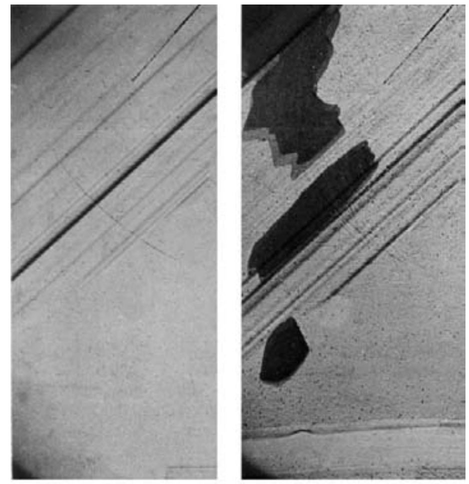
Several modern programs for the determination of crystal structures from measured single-crystal diffraction data (intensities) contain a routine to refine the structure of crystals twinned by merohedry, especially the program suite *SHELX97* (Sheldrick, 1998), the program *TWIN3.0* by Kahlenberg & Messner (2001) and the program *CRYSTALS 12* by Betteridge *et al.* (2003). Tests of different methods for the detection of merohedral twinning have been devised by Kahlenberg (1999). A description of the use of the program *SHELXL97* for merohedrally twinned crystals is provided by Herbst-Irmer (2006) and Herbst-Irmer & Sheldrick (1998).

For this procedure the twin law should be known or at least suspected. These programs consider the intensity of a reflection as a sum of the intensities of the different superimposed twin components (domains), weighted by their volume portions (whereby the volume ratios are also subject to refinement). This is done for all reflections, regardless of whether their intensities are affected by the twinning or not. Thus, the distinction between affected and non-affected reflections is usually not performed in routine structure refinements of twinned crystals. In problematic cases, *e.g.* if there are doubts about the twin law involved (or whether the crystal is twinned at all, or if the volume fraction of one domain state is very small), a special examination of the diffraction intensities that should be affected by a suspected twin law must be undertaken.

3.3. X-ray topographic imaging of merohedral twin domains

By X-ray diffraction topography [for an introduction see Tanner (1976) and Authier (2005), ch. 17] twin domains are visualized either by 'domain contrast', owing to different structure-factor moduli[10] of different domains states, or by contrast of their domain boundaries (Lang, 1965*a*, 1967; Lang & Miuskov, 1969; Phakey, 1969; Klapper *et al.*, 1983, 1987; Klapper, 1987). The visualization of the domain boundaries is important in those cases where domain contrast does not occur, *e.g.* for inversion twins in the case of negligible anomalous scattering. Here we consider only the domain contrast which arises when, owing to the twin law, the intensity of the reflection used for imaging is different in the various domain states. An early study of Dauphiné and Brazil twins of quartz has been reported by Lang (1965*a*). For mapping of Dauphiné domains he used the twin-related reflections $10\bar{1}1/\bar{1}011$ and $30\bar{3}1/\bar{3}031$, which belong to the two non-equivalent rhombohedra $\{h0\bar{h}l\}$ and $\{\bar{h}0hl\}$ (diffraction cases B1). Since the structure-factor moduli $|F(\bar{1}011)|$ and $|F(10\bar{1}1)|$ differ by a factor of about 1.5, the domain contrast is only moderate for this pair of reflections (*cf.* Fig. 4), whereas it is extremely strong for the second pair: $|F(\bar{3}031)|$ is about ten times larger than $|F(30\bar{3}1)|$. This study was made in transmission (Laue case) with moderately absorbing silver $K\alpha_1$ radiation (λ = 0.56 Å). For the mapping of Brazil (inversion) domains, Lang (1965*a*) used reflections $11\bar{2}1$ and $\bar{1}\bar{1}21$ [non-centrosymmetric face forms 'trigonal dipyramid' $\{hh2\bar{h}l\}$ and $\{\bar{h}\bar{h}2hl\}$, *cf.* Fig. 3(*a*), diffraction case B2] and cobalt $K\alpha_1$ radiation (λ = 1.789 Å), which is subject to a sufficiently strong anomalous scattering by silicon. His topographs, recorded by reflection (Bragg case) from both surfaces of a 1 mm-thick quartz plate, clearly revealed Brazil twin domains by domain contrast. Similarly, antiparallel 180° domains in ferroelectric $BaTiO_3$ (point group 4*mm*) have been imaged in reflections $003/00\bar{3}$ (non-centrosymmetric face form pedion $\{00l\}$) by domain contrast owing to anomalous scattering of chromium $K\alpha_1$ radiation (λ = 2.290 Å) by Ba and Ti (Niizeki & Hasegawa, 1964). Another example is given by Wallace (1970), who mapped 180° domains of rhombohedral lithium niobate (low-temperature point group 3*m*) by faint domain contrast of reflections $0006/000\bar{6}$ by surface reflection (Bragg case) using Cu $K\alpha$ radiation (diffraction case B2). Of particular benefit in this respect is the use of synchrotron white-beam topography, because the continuous synchrotron spectrum allows one to select an imaging wavelength close to the absorption edge of a significant atom, providing relatively strong contrast of inversion domains. This has been shown, *e.g.* by Huang *et al.* (1996) and Liu *et al.* (1996), for ferroelectric $KTiOAsO_4$ (point group *mm*2) with reflections $004/00\bar{4}$ and wavelengths (0.928 and 0.964 Å) close to the *K*-absorption edge of As (1.044 Å).

[10] Note that in the X-ray topography of large and perfect crystals the diffracted intensity is proportional to $|F|$ (*dynamical* reflection, *cf.* Zachariasen, 1945; Authier, 2005, §§4.8.8, 4.9.2 and 4.9.3), in contrast to the *kinematical* intensity proportional to $|F|^2$, which is applied in the structure determination of small ('extinction-free') crystals.



(*a*) (*b*)

Figure 4
X-ray topograph of an originally untwinned $(11\bar{2}0)$ plate of quartz before (*a*) and after (*b*) heating to above 846 K (transition to high-quartz) and cooling back to room temperature, recorded in the reflection $\bar{1}011$ ($|F|$ = 24.7). The Dauphiné-twin domains generated by this temperature cycle are imaged in the non-equivalent twin-related reflection $10\bar{1}1$ with higher structure-factor modulus ($|F|$ = 38.6) and, hence, appear in (*b*) with higher intensity as dark 'areas' (twin diffraction case B1). The line contrasts in (*a*) and (*b*) are growth striations. Mo $K\alpha_1$ radiation, imaged section ~8 mm × 18 mm (*cf.* Klapper *et al.*, 1983).

Fig. 4 shows the Dauphiné transformation twinning of quartz. A detailed study of the growth twinning (Dauphiné, Brazil and Leydolt twins) of quartz-homeotype gallium orthophosphate, $GaPO_4$, by etching, optical activity and X-ray topography is presented by Engel *et al.* (1989). Dauphiné and Leydolt domains have been mapped with Ag $K\alpha$ radiation, using twin-related 'rhombohedral' reflections $10\bar{1}1/\bar{1}011$ and $30\bar{3}1/\bar{3}031$, the former providing moderate, the latter very strong B1 domain contrast, similar to quartz (see above).

Fig. 5 presents four topographs of hexagonal $KLiSO_4$ (space group $P6_3$) which very frequently exhibits growth twins with twin law 'reflection plane *m* parallel to the hexagonal axis 6' (composite symmetry 6*mm*) with sharply defined (0001) twin boundaries (Klapper *et al.*, 1987). The topographs recorded in reflections 0003 and $30\bar{3}0$ do not show these twins (diffraction cases A), but are useful for the characterization of other growth defects (*e.g.* dislocations, growth striations, faulted growth-sector boundaries), whereas reflections $12\bar{3}0$ and $21\bar{3}0$ (diffraction case B1) reveal the twinning by very strong domain contrast ($|F(12\bar{3}0)| = 1.6$, $|F(21\bar{3}0)| = 18.2$; Chung, 1972). In this case the extreme domain contrast is useful for a clear distinction of twin domains from other growth defects, since the latter generate local intensity variations (film blackening) by a factor of five or even more. In another case of growth twins of $NaLiSO_4$ (point group $3m$, merohedal twin law 'm normal to threefold axis', composite symmetry $\bar{6}m2$) the twin domains can hardly be recognized because of the low domain contrast, compared with the strong local intensity changes owing to other growth defects (Klapper, 1987).

Another instructive example is provided by the X-ray topographic study of the very sluggish low-temperature phase transition III ($P6_3$) $\leftrightarrow$ IV ($P31m$) of $KLiSO_4$ at about 225 K (hysteresis $\simeq 50°$) (Klapper *et al.*, 2008). During this transition the growth twinning (*cf.* Fig. 5) vanishes owing to the evolution of the twin mirror plane $m \parallel [001]$ of phase III ($P6_3$) into the structural symmetry plane of phase IV ($P31m$) and a new transition twinning appears owing to the reduction of the hexagonal structural symmetry axis in $P6_3$ to the trigonal symmetry axis 3 in $P31m$. The twin law '$2 \parallel [001]$', and thus the space group $P31m$, is ascertained by the topographic B1 domain contrast characteristic of this twin law.

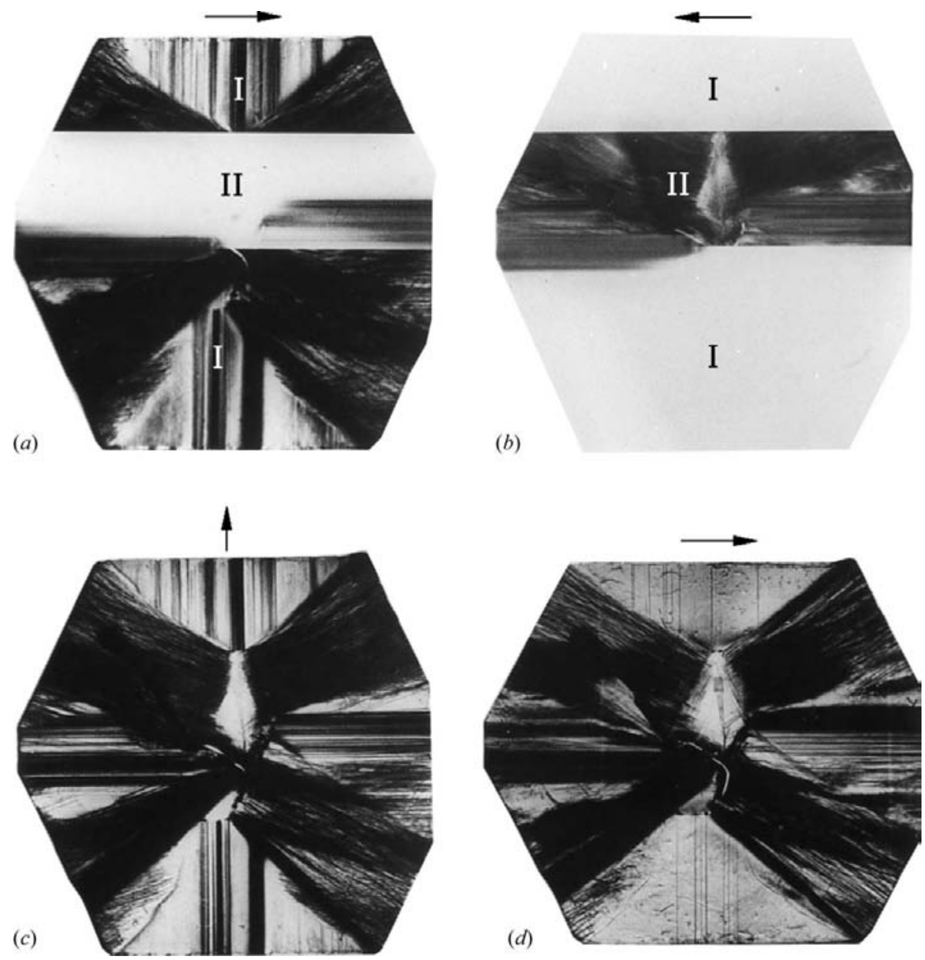


Figure 5
X-ray topographs (Mo $K\alpha_1$ radiation) of a $(11\bar{2}0)$ plate (12 mm $\times$ 12 mm, about 1 mm thick) cut from a hexagonal $KLiSO_4$ (modification III) crystal (point group 6; *cf.* §2.2.3), grown from aqueous solution, exhibiting growth reflection twinning of law $m \parallel [001]$ with two domain states I and II. The twin-related reflections $21\bar{3}0_{(I)}/12\bar{3}0_{(II)}$ (*a*) and $12\bar{3}0_{(I)}/21\bar{3}0_{(II)}$ (*b*) (diffraction case B1; *cf.* Fig. 2*b*), with very different moduli $F(12\bar{3}0) = 1.6$ and $F(21\bar{3}0) = 18.2$ (Chung, 1972), map the domain states I and II with very strong domain contrast, which is reversed in (*b*) when the two twin-related reflections are interchanged. The imaging reflections 0003 (*c*) and $30\bar{3}0$ (*d*), both twin diffraction case A (*cf.* Fig. 2*a*), do not reveal the domain states I and II (no domain contrast), but exhibit other growth defects, such as dislocations, faulted growth-sector boundaries *etc.* Note that these defects are not visible in the faintly diffracting domains of (*a*) and (*b*) owing to their small *F*-modulus which leads to kinematical (*i.e.* extinction-free) diffraction (*cf.* Klapper *et al.*, 1987).

Note: the present paper treats only twins by merohedry with *complete* lattice coincidence, *i.e.* $\Sigma 1$ twins. Twins by reticular merohedry with *partial* lattice coincidence, *i.e.* twins with $\Sigma > 1$, will be presented in a subsequent paper.

APPENDIX *A*
Classification of the laws of twinning by merohedry

In the previous sections of this paper we have shown with many examples that the face forms are an illustrative and useful tool for the treatment of intensity relations in the diffraction of chiral, polar and merohedrally twinned crystals. In this Appendix we present a systematic listing of the twin laws in all 26 merohedral point groups[11] under the aspect of the index $[n]$ between the merohedral group and its holohedral supergroup as an 'order parameter', whereas the index $[t]$ of a merohedral twin ('*untwinned* $\rightarrow$ *composite group*') is always 2. All merohedral twins can be considered as either inversion or twofold rotation or reflection twins.

Note the particular complexity in the hexagonal crystal family with the five 'trigonal' groups 3, $\bar{3}$, 32, $3m$, $\bar{3}2/m$, which are based on both a rhombohedral and a hexagonal lattice with holohedries $\bar{3}2/m$ and $6/m2/m2/m$; for the hexagonal holohedry the two point-group settings $\bar{3}2/m1$ and $\bar{3}12/m$ are two different merohedral subgroups of index $[n] = 2$. Note furthermore that symbols such as 32, 321 and 312 or $\bar{4}2m$ and $\bar{4}m2$ do not only refer to different descriptions of the same point group but rather imply different structures with different

[11] Inclusion of $\bar{3}2/m$ as merohedry of the hexagonal lattice holohedry brings the number of merohedral point group types from 25 (= 32 − 7 holohedries) to 26.

Table 7
Merohedral twins with index $[n] = 2$ between merohedral and holohedral group; only one twin law exists for each setting of a merohedral group.

Subtable (i): the twin laws can be considered as both *rotation* and *reflection* twins, because both types of operations belong to the twin coset. All face forms in these groups are centrosymmetric. Only twin diffraction cases A and B1 (but not B2) occur. The merohedral twins of point group $\bar{3}$ with hexagonal lattice (index $[n] = 4$) are given in Table 8(i).

Subtable (ii): all these twins are *inversion* twins. Centrosymmetric face forms are mapped onto themselves (twin diffraction case A), non-centrosymmetric forms onto their 'inverted' face forms (twin diffraction case B2). Diffraction case B1 does not occur. Note that for the twin $1 \rightarrow \bar{1}$ case A does not occur. Further inversion twins are given in Tables 8(ii) and 8(iii).

(i) 6 centrosymmetric merohedral groups

Untwinned group	Composite group
$4/m$	$4/m2/m2/m$
$\bar{3}$ (rhombohedral lattice)	$\bar{3}2/m$
$\bar{3}2/m1$ (hexagonal lattice)	$6/m2/m2/m$
$\bar{3}12/m$ (hexagonal lattice)	$6/m2/m2/m$
$6/m$	$6/m2/m2/m$
$2/m\bar{3}$	$4/m\bar{3}2/m$

(ii) 17 non-centrosymmetric merohedral groups

Untwinned group	Composite group
1	$\bar{1}$ (only B2)
2, m	$2/m$
222, $mm2$	$2/m2/m2/m$
422, $4mm$, $\bar{4}2m$, $\bar{4}m2$	$4/m2/m2/m$
32, $3m$ (rhombohedral lattice)	$\bar{3}2/m$
622, $6mm$, $\bar{6}2m$, $\bar{6}m2$	$6/m2/m2/m$
432, $\bar{4}3m$	$4/m\bar{3}2/m$

space groups (*e.g.* $R32$, $P321$ and $P312$ or $P\bar{4}2m$ and $P\bar{4}m2$); these 'structural settings' were first introduced and listed by Donnay (1977). In the present paper, 35 such structural settings of the 26 merohedral point-group types are treated, leading to a total of 63 possible merohedral twin laws.

In conclusion, it is apparent from Tables 7 and 8 that the 'complexity' of the twinning by merohedry depends upon the 'distance' of the crystal point group from its holohedral point group, *i.e.* upon the index $[n]$ of the merohedral group with respect to its holohedry. For the possible indices $[n]$ = 2, 4, 8 the number of (binary) merohedral twin laws is $n - 1$ = 1, 3, 7. They consist always of one *inversion*, $(n - 2)/2$ *rotation* and $(n - 2)/2$ *reflection* twins, *i.e.* for $[n] = 4$ of one *inversion*, one *rotation* and one *reflection twin*, for $[n] = 8$ of one *inversion*, three *rotation* and three *reflection* twins.

The value of the index n has a further meaning which is important for the refinement of twinned crystal structures: n is the maximal number of domain states (orientation states) which can coexist in a merohedral twin. For $n = 2$ this is trivial, for $n = 4$ illustrative examples are quartz and $KLiSO_4$ described in §2.2.2, §2.2.3 and Appendix *C*. For $n = 8$ (point group 3) no twin with eight domain states is known yet, but refinement of an organometallic Os compound (space group $P3_1$) resulted in three possible twin laws and four of the eight possible domain states with, however, quite different volume fractions, dominated by a pair of reflection twin domains (Herbst-Irmer & Sheldrick, 1998, example 1; Herbst-Irmer, 2006, pp. 122–127; Flack & Bernardinelli, 1999, §8.2). A twin with n merohedral domain states would be the 'complete twin' of Curien & Donnay (1959), with the holohedral point group as twin-composite symmetry, in the three cases above $6/m2/m2/m$ (*cf.* Hahn & Klapper, 2003, §3.3.6.14)

A different aspect of the values n = 2, 4 and 8 has been pointed out by Le Page *et al.* (1984, especially Table 1): n is the number of non-equivalent 'structural settings' of a merohedral point group in its holohedry, *i.e.* the number of different

Table 8
Merohedral twins with indices $[n] = 4$ and $[n] = 8$ between merohedral and holohedral group; for $[n] = 4$ there are three twin laws for each setting of a merohedral group, for $[n] = 8$ seven twin laws [last line in subtable (ii)].

Subtable (i): only twin diffraction cases A and B1 occur. All three twins can be considered as either *rotation* or *reflection* twins, because their cosets contain both types of operations.

Subtable (ii): for *inversion* twins only twin diffraction cases A and B2 occur (except for the twin $3 \rightarrow \bar{3}$, where case A does not occur, because there is no centrosymmetric face form in point group 3). Note that in the *reflection* twins (column 4) with $[n] = 4$ morphologically inverted forms (case B2) do not occur, whereas for $[n] = 8$ all three cases are realised. In the last line the unique twin case of index $[n] = 8$ is presented, point group 3 with respect to the hexagonal holohedry $6/m2/m2/m$. Again, diffraction case A does not occur for the *inversion* twin $3 \rightarrow \bar{3}$. For the *rotation* and *reflection* twins all three cases A, B1 and B2 occur.

Subtable (iii). Here *inversion* twins (column 2) and 'mixed' *rotation* and *reflection* twins occur; in the latter, the twin coset contains a twofold rotation as well as a reflection, but both are not perpendicular to each other (columns 3 and 4; see also Appendix *B* below).

(i) 1 centrosymmetric merohedral group

Untwinned group	Rotation and reflection twins, twin diffraction cases A, B1		
$\bar{3}$ (hexagonal lattice)	$\bar{3}2/m1$	$\bar{3}12/m$	$6/m$

(ii) 6 + 1 enantiomorphic (chiral) merohedral rotation groups, $[n] = 4$ and 8

Untwinned group	Inversion twins A, B2	Rotation twins A, B1, B2	Reflection twins A, B1
4	$4/m$	422	$4mm$
3 (rhombohedral lattice)	$\bar{3}$ (only B2)	32	$3m$
321 (hexagonal lattice)	$\bar{3}2/m1$	622	$\bar{6}2m$
312 (hexagonal lattice)	$\bar{3}12/m$	622	$\bar{6}m2$
6	$6/m$	622	$6mm$
23	$2/m\bar{3}$	432	$\bar{4}3m$
3 (hexagonal lattice) ($[n] = 8$)	$\bar{3}$ (only B2)	6, 321, 312	$\bar{6} = 3/m$ (A, B1), $3m1$, $31m$ (+ B2)

(iii) 4 non-enantiomorphic non-centrosymmetric merohedral groups

Untwinned group	Inversion twins A, B2	Rotation and reflection twins A, B1, B2	
$\bar{4}$	$4/m$	$\bar{4}2m$	$\bar{4}m2$
$3m1$ (hexagonal lattice)	$\bar{3}2/m1$	$6mm$	$\bar{6}m2$
$31m$ (hexagonal lattice)	$\bar{3}12/m$	$6mm$	$\bar{6}2m$
$\bar{6}$	$6/m$	$\bar{6}2m$	$\bar{6}m2$

transformations (including the identity) of the base vectors **a**, **b**, **c**, the atomic coordinates x, y, z and the Miller indices (structure factors) h, k, l within a crystal family. These settings are in a one-to-one correspondence with the possible merohedral twin laws of a given point group. Thus, for point group 3 the seven (non-identity) transformations within the hexagonal holohedry $6/m2/m2/m$ correspond to the seven possible merohedral twin laws, listed in the last line of Table 8(ii).

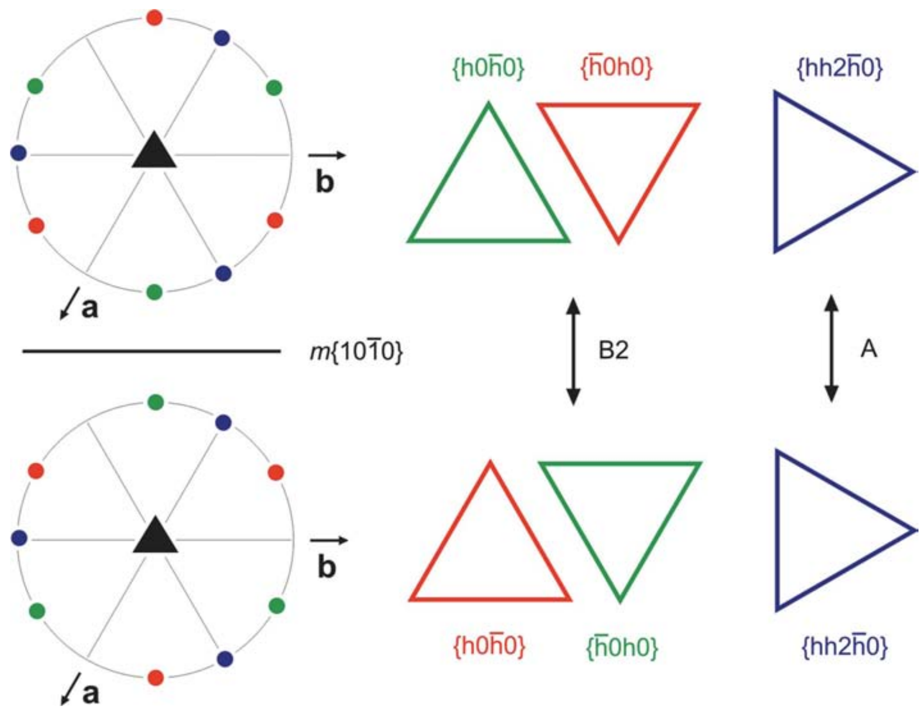


Figure 6
Reflection twinning $3 \rightarrow 3m1$ of index 2 with twin element 'reflection plane $\{10\bar{1}0\}$' (index $[n] = 8$ with respect to the hexagonal holohedry $6/m2/m2/m$). Two pairs of twin-related non-centrosymmetric face forms ('acentric' reflection sets) are considered: (i) the *trigonal prism* I $\{h0\bar{h}0\}$ (green) and its inverted ('opposite') form $\{\bar{h}0h0\}$ (red); (ii) the *trigonal prism* II $\{hh2\bar{h}0\}$ (blue), superimposed upon itself, because the twin element is part of its *eigensymmetry*. The stereographic projections of these forms in the two twin orientation states are shown. Forms with different F-moduli (owing to different anomalous-scattering contributions) are distinguished by different colours (green and red). The superposition of the coloured face poles (reciprocal-lattice points, reciprocal diffraction vectors) of the two orientation states reveals the intensity characteristics of the twin-related reflections: reflection set $\{h0\bar{h}0\}$ (green) is superimposed upon its non-equivalent inverted (opposite) set $\{\bar{h}0h0\}$ (red, different colours, twin diffraction case B2), whereas the set $\{hh2\bar{h}0\}$ (blue) is superimposed upon itself (equal colour, case A). This is further illustrated by the corresponding coloured face forms *trigonal prism* viewed along the threefold axis. For the other twin reflection plane $\{11\bar{2}0\}$ in $3 \rightarrow 31m$ (not shown here) the prism $\{h0\bar{h}0\}$ is superimposed upon itself (blue, case A), whereas the prism $\{hh2\bar{h}0\}$ is superimposed upon its inverted form (green/red, case B2).

APPENDIX *B*
Morphologically (geometrically) inverted face forms

In this Appendix *general* and *special* face forms $\{hkl\}$, which are of particular significance for the B2 diffraction case, are discussed. These face forms themselves are non-centrosymmetric but are mapped by a twin operation onto their 'inverted' form $\{\bar{h}\bar{k}\bar{l}\}$. Superposition (combination) of these two forms results in a centrosysmmetric set $\{hkl\}$, which is the face form $\{hkl\}$ of the twin composite group, no matter whether the latter is centrosymmetric or not. Hence, these pairs of inverted forms have '*Bijvoet character*' and correspond to twin diffraction case B2. Examples are shown in Fig. 3.

These '*morphological inversions* $\bar{1}_{\rm morph}$' can be the result of three twin cases:

(1) a twin inversion centre $\bar{1}' \rightarrow \bar{1}_{\rm morph}$;

(2) a twofold twin axis $2'$ perpendicular to an *eigensymmetry* mirror plane m of the special face form, $2'/m \rightarrow \bar{1}_{\rm morph}$;

(3) a twin reflection plane m' normal to a twofold *eigensymmetry* axis 2 of the special face form, $2/m' \rightarrow \bar{1}_{\rm morph}$.

The prime indicates the twin operation in order to distinguish it from the *eigensymmetry* operation of the face form.[12] Note that the unprimed symmetry elements 2 and m belong to the *eigensymmetry* of the face form but may not belong to the symmetry group of the untwinned crystal; otherwise the twin would be an inversion twin of case (1).

Case (1) occurs in all 21 non-centrosymmetric merohedral point groups listed in Appendix *A*, Table 7(ii) (index $[n] = 2$) and Tables 8(ii) and 8(iii), column 2 (index $[n] = 4, 8$). Case (2) occurs only in rotation twins of chiral and achiral non-centrosymmetric groups of $[n] = 4$ and 8, Tables 8(ii) and 8(iii). Case (3) occurs in a 'pure way' only in the $[n] = 8$ point group 3 (hexagonal lattice), Table 8(ii), last line, column 4, which is discussed below.

Simultaneous occurrence of cases (2) and (3) is found in all non-centrosymmetric non-enantiomorphic (achiral) groups, the twins of which can be considered as rotation as well as reflection twins ('mixed way'). These are listed in Table 8(iii), columns 3 and 4.

The three cases (1)–(3) above are now illustrated by examples and the following section 'Concerning (3): reflection twin' with Figs. 6 and 7.

Concerning (1): *inversion twins*. All centrosymmetric face forms are mapped upon themselves (twin diffraction case A), all non-centrosymmetric forms $\{hkl\}$ (*general* and *special*) are mapped upon their inverted 'opposite' forms $\{\bar{h}\bar{k}\bar{l}\}$ (case B2).

Examples: (i) Twin $321 \rightarrow \bar{3}2/m1$ (Brazil twin of quartz, *cf.* Table 4): the following non-centrosymmetric face forms are mapped upon their inverted opposites: trigonal bipyramid $\{hh2\bar{h}l\}$, ditrigonal prism $\{hki0\}$ and trigonal prism $\{hh2\bar{h}0\}$. Their twin combinations result in the centrosymmetric forms

[12] The prime is commonly used in the treatment of black–white symmetries (*cf.* Hahn & Klapper, 2003, §3.3.4).

hexagonal bipyramid, dihexagonal prism, hexagonal prism, respectively, of composite group $\bar{3}2/m1$ (*cf.* Fig. 3*a*).

(ii) Cubic twin 23 → $2/m\bar{3}$ (*cf.* Table 6): the face form tetrahedron $\{hhh\}$ is mapped upon its inverted tetrahedron $\{\bar{h}\bar{h}\bar{h}\}$, resulting in the centrosymmetric face form octahedron of composite group $2/m\bar{3}$ (Fig. 3*c*).

Concerning (2): *rotation twins*. Certain face forms are mapped upon themselves (diffraction case A), others upon their inverted opposites (case B2), and still others upon a 'non-inverted' form (case B1).

Examples: (i) Twin 321 → 622 (Dauphiné twin of quartz, *cf.* Table 4): the face forms mapped upon their inverted antipodes are the same as for the inversion (Brazil) twin above (Fig. 3*a*).

(ii) Cubic twin 23 → 432 (*cf.* Table 6): as above, the tetrahedron $\{hhh\}$ is morphologically inverted, leading to the combined form octahedron (Fig. 3*c*).

Concerning (3): *reflection twins*. This case occurs as a '*pure*' reflection twin 3 → $3m1$ only in point group 3 (hexagonal lattice, $[n]$ = 8): the 'fixed' (specially oriented) trigonal prism $\{h0\bar{h}0\}$ (*eigensymmetry* $\bar{6}2m$) is mapped upon its inverted form by m' = $(10\bar{1}0)$ [normal to the twofold *eigensymmetry* axis of the prism; twin diffraction case B2], whereas the other fixed prism $\{hh2\bar{h}0\}$ is mapped upon itself (diffraction case A), *cf.* Fig. 6. For the other pure reflection twin 3 → $31m$ [m' = $(11\bar{2}0)$] the diffraction cases are A for prism $\{h0\bar{h}0\}$ and B2 for prism $\{hh2\bar{h}0\}$. Similarly, reflection twin 3 → $3/m$ [m' = (0001)]: case B2 for the pedion $(000l)$ (*eigensymmetry* ∞m).

A twin is simultaneously a *rotation* and a *reflection* twin ('mixed way'), when non-perpendicular twin elements $2'$ and m' are contained in the twin coset [Table 8(iii), columns 3 and 4].

Example: Twin $\bar{4}$ → $\bar{4}2m$ (mixed rotation and reflection twin): the 'fixed' face form 'tetragonal tetrahedron (disphenoid)' $\{h0l\}$ is mapped upon its inverted form by $2'\langle 100\rangle$ as well as by $m'\{110\}$ (case B2), resulting in the centrosymmetric combination 'tetragonal dipyramid'. The other fixed tetragonal tetrahedron $\{hhl\}$, however, is mapped upon itself (diffraction case A) by these two twin operations (Fig. 7).

Note that for the different twin $\bar{4}$ → $\bar{4}m2$ the twin operations are $2'\langle 110\rangle$ and $m'\{100\}$; both invert the face form $\{hhl\}$ (case B2), but leave the form $\{h0l\}$ unchanged (case A).

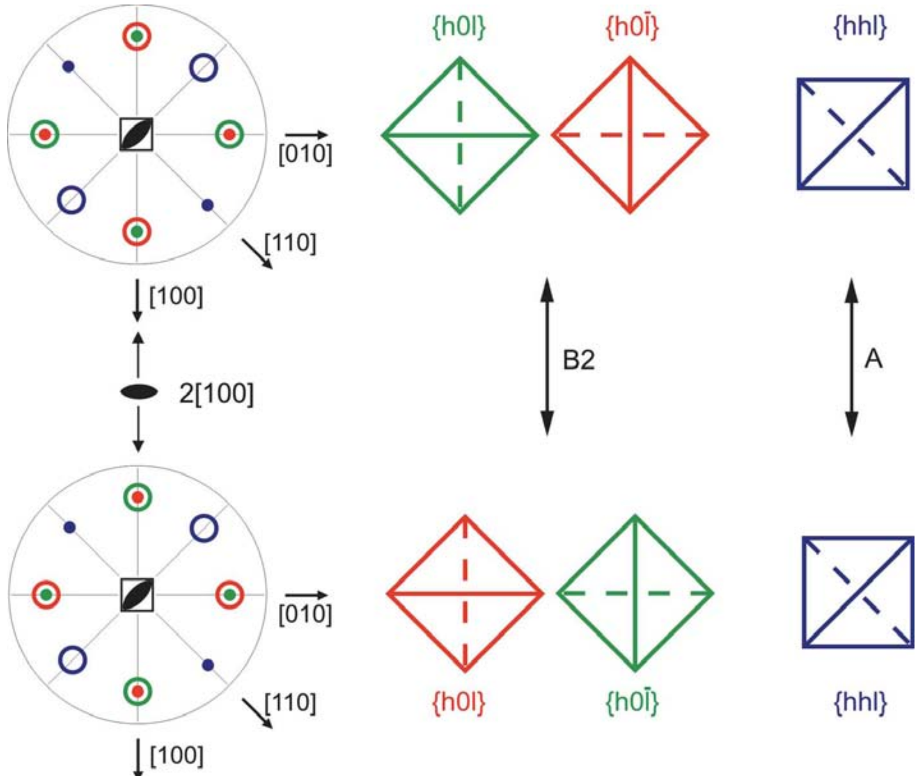


Figure 7
'Mixed' rotation and reflection twinning $\bar{4} \rightarrow \bar{4}2m$ (coset of twin elements: $2_{[100]}$, $2_{[010]}$, $m_{(110)}$, $m_{(1\bar{1}0)}$, here represented by $2_{[100]}$), applied to two twin-related non-centrosymmetric face forms ('acentric' reflection sets): (i) the *tetragonal tetrahedron* (*disphenoid*) $\{h0l\}$ (green) and its inverted (opposite) form $\{h0\bar{l}\}$ (red); (ii) the *tetragonal tetrahedron* $\{hhl\}$ (blue), superimposed upon itself. [Note: $\{h0\bar{l}\} \equiv \{\bar{h}0\bar{l}\}$ (red) and $\{hhl\} \equiv \{\bar{h}\bar{h}l\}$ (blue) (different representatives).] The stereographic projections of these forms in the two twin orientation states are shown. Dots represent the face poles (reciprocal-lattice points, reciprocal diffraction vectors) on the upper, circles on the lower half of the projection sphere. Forms with different F-moduli (owing to different anomalous-scattering contributions) are distinguished by different colours (green and red). The superposition of the face poles reveals the intensity characteristics of twin-related reflections: reflection set $\{h0l\}$ (green) is superimposed upon its non-equivalent inverted (opposite) set $\{h0\bar{l}\}$ (red, different colours, twin diffraction case B2), whereas the set $\{hhl\}$ (blue) is superimposed upon itself (equal colour, case A). This is further illustrated by the corresponding coloured tetragonal tetrahedra viewed along the axis $\bar{4}$.

APPENDIX *C*

Diffraction intensity relations of twinned enantiomorphic crystals

In this Appendix only merohedral twins of *enantiomorphic* (*chiral*) crystals are treated. They deserve special consideration with respect to the structure-factor moduli of their twin partners, because partners either of equal or of opposite handedness are combined. Three cases are distinguished:

(i) Inversion twins: partners of opposite handedness and inverted ('opposite') orientation are combined.

(ii) Rotation twins: partners of equal handedness but different orientation are combined.

(iii) Reflection twins: partners of opposite handedness but with mirror-related orientation are combined, *i.e.* the orientation relation differs from that of an inversion twin.

Following international usage, the same right-handed coordinate system is chosen here for both enantiomorphs. The following relations between the structure-factor moduli of right- and left-handed structures, indicated by subscripts 'right' and 'left', are generally valid for *all non-centrosymmetric* sets of reflections, independent of their face forms:

(a) $|F(hkl)_{\rm right}| \neq |F(\bar{h}\bar{k}\bar{l})_{\rm right}|$, $|F(hkl)_{\rm left}| \neq |F(\bar{h}\bar{k}\bar{l})_{\rm left}|$, owing to different anomalous-scattering contributions (Bijvoet pairs).

(b) $|F(hkl)_{\rm right}| \neq |F(hkl)_{\rm left}|$, again owing to different anomalous-scattering contributions.

(c) $|F(hkl)_{\rm right}| = |F(\bar{h}\bar{k}\bar{l})_{\rm left}|$, $|F(hkl)_{\rm left}| = |F(\bar{h}\bar{k}\bar{l})_{\rm right}|$, because of equal anomalous-scattering contributions.

For the special case of *centrosymmetric* face forms, all four F-moduli are equal:

$$|F(hkl)_{\rm right}| = |F(\bar{h}\bar{k}\bar{l})_{\rm right}| = |F(hkl)_{\rm left}| = |F(\bar{h}\bar{k}\bar{l})_{\rm left}|.$$

In the following, only those special *non-centrosymmetric* face forms $\{hkl\}$ which are 'morphologically inverted' into their opposite forms (antipodes) $\{\bar{h}\bar{k}\bar{l}\}$ by the twinning (*cf.* Appendix B) are considered. For these face forms the above relations (a)–(c) have the following consequences for *enantiomorphic* crystals:

Inversion twins [Appendix A, Table 8(ii), column 2]: all twin-related non-centrosymmetric sets of reflections $\{hkl\}_{\rm (right)}$ and $\{hkl\}_{\rm (left)}$, with the *same* signs of their indices but different F-moduli, are superimposed [diffraction case B2, relation (b) above]. Note that $\{hkl\}_{\rm (right)}$ would be superimposed with $\{\bar{h}\bar{k}\bar{l}\}_{\rm (left)}$ (different signs of indices) if the left-handed structure would be referred to a left-handed coordinate system.

For inversion twins of non-centrosymmetric but non-enantiomorphic (achiral) crystals, $\{hkl\}$ pairs with *opposite* signs of their indices are always superimposed, independent of the handedness of the coordinate system.

Rotation twins [Table 8(ii), column 3]: both twin partners have the same handedness. If the twofold twin rotation axis is perpendicular to a mirror plane of the *eigensymmetry* of an acentric face form $\{hkl\}$, twin-related sets of reflections $\{hkl\}_{\rm (right)}$ and $\{\bar{h}\bar{k}\bar{l}\}_{\rm (right)}$, or $\{hkl\}_{\rm (left)}$ and $\{\bar{h}\bar{k}\bar{l}\}_{\rm (left)}$, with *opposite* signs of their indices, are superimposed and form case B2 [different F-moduli

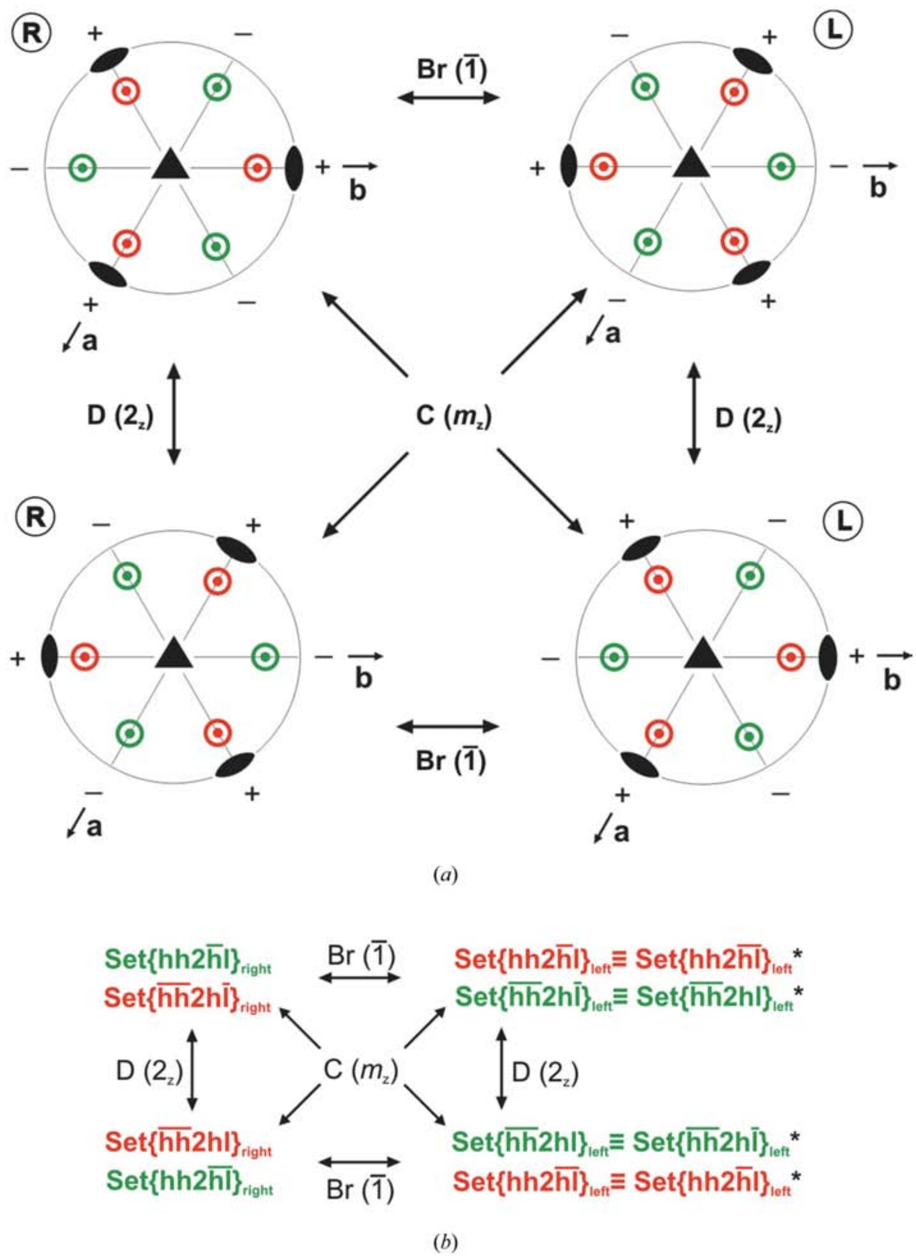


Figure 8

(a) Stereographic projections of the *non-centrosymmetric* face form *trigonal dipyramid* $\{hh2\bar{h}l\}$ and its inverse opposite form $\{\bar{h}\bar{h}2hl\}$ of a quartz crystal for the three twin types $321 \rightarrow \bar{3}2/m1$ (Brazil, *inversion*), $321 \rightarrow 622$ (Dauphiné, *rotation*) and $321 \rightarrow \bar{6}2m$ (Leydolt, *reflection*). Dots represent the face poles (reciprocal-lattice points, reciprocal diffraction vectors) on the upper, circles on the lower half of the projection sphere. R and L indicate right- and left-handed crystals. Forms with different F-moduli (owing to different anomalous-scattering contributions) are distinguished by different colours (green and red). The following pairs of stereographic projections represent the same twin law: horizontal pairs, inversion twins 'Br'; vertical pairs, Dauphiné twins 'D'; diagonal pairs, Leydolt ('combined') twins 'C'. The superposition of the diagrams of a pair shows the intensity characteristics of the twinned crystal: if face poles of different colour are superimposed (Brazil and Dauphiné twins), diffraction case B2 with different F-moduli results; if the superimposed face poles have the same colour (Leydolt twins), case A with equal moduli results. Note that in Brazil and Dauphiné twins the polar twofold axes (indicated by + and −) are reversed ('electrical twinning'), whereas in Leydolt twins they remain unchanged. (b) The same twin relations as in (a), but in terms of Miller–Bravais indices. Note: $F[{\rm set}\{hh2\bar{h}l\}_{\rm right}] = F[{\rm set}\{\bar{h}\bar{h}2hl\}_{\rm left}]$ (both green) and $F[{\rm set}\{\bar{h}\bar{h}2hl\}_{\rm right}] = F[{\rm set}\{hh2\bar{h}l\}_{\rm left}]$ (both red). The two equations correspond to the two general equations (c) of the present Appendix C. They explain why reflections $\{hh2\bar{h}l\}$ related by the Leydolt twin law exhibit the same F-moduli. The asterisks (*) indicate different representatives of the set related by the *eigensymmetry plane* m_z of the trigonal dipyramids.

owing to anomalous scattering, relation (*a*) above]. This leads to a morphologically inverted form (*cf.* Appendix *B*).

Reflection twins [Table 8(ii), column 4]: both twin partners have opposite handedness. In contrast to the inversion and rotation twins, here the non-centrosymmetric twin-related face forms are mapped *upon themselves* and *not upon their inverted ones*. Reflection sets $\{hkl\}_{\rm (right)}$ and $\{\bar{h}\bar{k}\bar{l}\}_{\rm (left)}$ or $\{hkl\}_{\rm (left)}$ and $\{\bar{h}\bar{k}\bar{l}\}_{\rm (right)}$, both with equal moduli, are superimposed [diffraction case A, relation (*c*) above]. The superimposed diffraction intensities are independent of the twin volume ratio and there is no domain contrast in X-ray topography. Face forms which are not mapped upon themselves or upon their 'opposites' provide B1 diffraction cases (different geometrical structure factors).

Example: reflection twin $321 \rightarrow \bar{6}2m$ (*cf.* Table 4, column 5): the acentric face forms $\{hh2\bar{h}l\}$, $\{hki0\}$ and $\{hh2\bar{h}0\}$ are mapped upon themselves, as well as the centric forms $\{h0\bar{h}0\}$ and $\{000l\}$ (all diffraction case A). The forms $\{hkil\}$ (acentric) and $\{h0\bar{h}l\}$ (centric) are not mapped upon themselves or upon their opposites: diffraction cases B1 (different geometrical structure factors). Case B2 does not occur, as in all other reflection twins, except for those with $[n] = 8$ [Table 8(ii)].

A special case, however, exists for the $[n] = 8$ reflection twins $3 \rightarrow 3m1$, $31m$ [Table 8(ii), last line, column 4]. Of the two fixed trigonal prisms $\{h0\bar{h}0\}$ and $\{hh2\bar{h}0\}$ (differently oriented *eigensymmetries* $\bar{6}2m$), always one is transformed into itself (case A), the other into its inverted (opposite) form [case B2, *cf.* Fig. 6 and Table 9(*c*)]. For the reflection twin $3 \rightarrow \bar{6} = 3/m$ all trigonal pyramids constitute case B1 and all trigonal prisms case A. Only the pedion $(000l)$ is a B2 case.

As an example, the above general relations of the *F*-moduli and their superposition are illustrated by the three merohedral twins of quartz (treated in detail in §2.2.2) for two types of face forms:[13]

(i) The *non-centrosymmetric* form $\{hh2\bar{h}l\}$ and its inverted opposite form $\{\bar{h}\bar{h}2h\bar{l}\}$ (two symmetrically non-equivalent *trigonal dipyramids*). From the stereographic projections in Fig. 8 it is immediately clear that in the Brazil and the

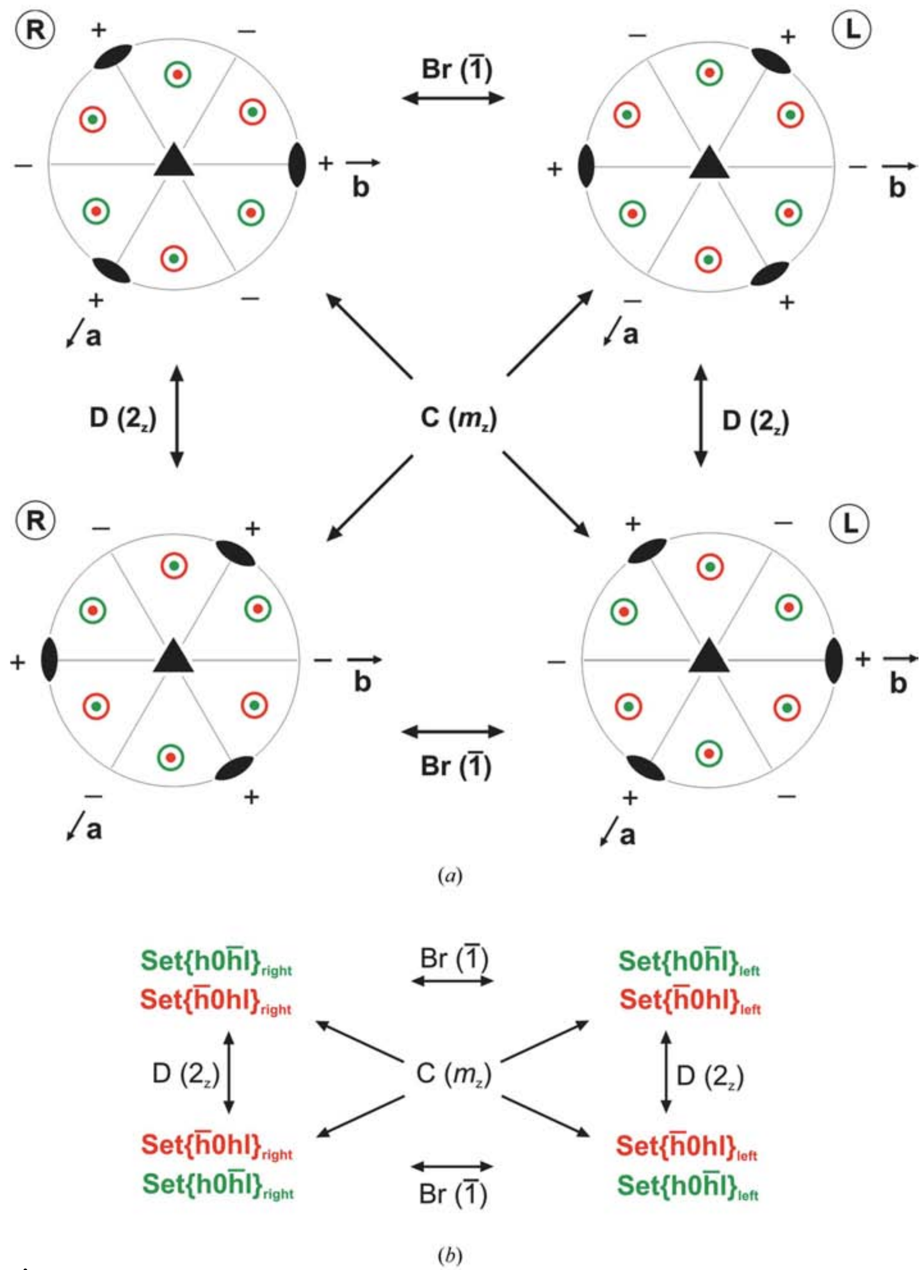


Figure 9
(*a*) The same as in Fig. 8 but for the two *centrosymmetric* non-equivalent face forms *rhombohedron* $\{h0\bar{h}l\}$ (green) and *rhombohedron* $\{\bar{h}0hl\}$ (red). Each of these two rhombohedra has the same *F*-moduli for right- and left-handed crystals, *i.e.* $|F\{h0\bar{h}l\}_{\rm left}| = |F\{h0\bar{h}l\}_{\rm right}|$, whereas the *F*-moduli of the two (green and red) rhombohedra differ (different geometric *and* anomalous-scattering contributions, diffraction case B1, in contrast to diffraction case B2 of Fig. 8). Again, the superposition of the diagrams of twin-related rhombohedra reveals whether the reflections sets are affected by the twinning (different colours for the Dauphiné and Leydolt twins, diffraction case B1) or not affected (equal green and red colours for the Brazil twin, case A). (*b*) The same twin relations as in (*a*), but in terms of Miller–Bravais indices. Note that a rhombohedron $\{h0\bar{h}l\}$ (green) is, owing to its centrosymmetric *eigensymmetry*, transformed into the rhombohedron $\{\bar{h}0hl\} \equiv \{h0\bar{h}\bar{l}\}$ (red) by both, the twofold axis $D(2_z)$ and the mirror plane $C(m_z)$, whereas it is mapped upon itself by the Brazil twin Br($\bar{1}$).

[13] To achieve a correct 'setting' of low-quartz can be a rather complicated affair, full of pitfalls, owing to the various choices involved: right-quartz *versus* left-quartz, right-handed *versus* left-handed coordinate system, enantiomorphic space group $P3_121$ *versus* $P3_221$, sense of optical rotation, choice of origin along the *c* axis and possible positive directions of all the axes. These problems are extensively discussed by Lang (1965*b*) and Donnay & Le Page (1978).

Table 9
(*a*) Triclinic, monoclinic and orthorhombic point groups (only meroheral inversion twins possible).

Point group	Composite group	Composite group (black–white notation)	Twin diffraction cases for different types of reflections (face forms)						
			hkl	*h*0*l*	0*kl*	*hk*0	*h*00	0*k*0	00*l*
1	1	$\bar{1}'$	All reflections B2						
2 ($\parallel b$)	$2/m$	$2/m'$ ($\bar{1}'$)	B2	A	B2	B2	A	B2	A
m ($\perp b$)	$2/m$	$2'/m$ ($\bar{1}'$)	B2	B2	B2	B2	B2	A	B2
222	$2/m2/m2/m$	$2/m'2/m'2/m'$ ($\bar{1}'$)	B2	A	A	A	A	A	A
$mm2$	$2/m2/m2/m$	$2'/m2'/m2/m'$ ($\bar{1}'$)	B2	B2	B2	A	A	A	B2

(*b*) Tetragonal point groups

Point group	Composite group	Composite group (black–white notation)	Twin diffraction cases for different types of reflections (face forms)						
			hkl	*h*0*l*	*hhl*	*hk*0	*h*00	*hh*0	00*l*
4	$4/m$	$4/m'$ ($\bar{1}'$)	B2	B2	B2	A	A	A	B2
	422	$42'2'$	B1	B2	B2	B1	A	A	B2
	$4mm$	$4m'm'$	B1	A	A	B1	A	A	A
$\bar{4}$	$4/m$	$4'(2)/m'$ ($\bar{1}'$)	B2	B2	B2	A	A	A	A
	$\bar{4}2m$	$\bar{4}2'm'$	B1	B2	A	B1	A	A	A
	$\bar{4}m2$	$\bar{4}m'2'$	B1	A	B2	B1	A	A	A
$4/m$	$4/m2/m2/m$	$4/m2'/m'2'/m'$	B1	A	A	B1	A	A	A
422	$4/m2/m2/m$	$4/m'2/m'2/m'$ ($\bar{1}'$)	B2	A	A	A	A	A	A
$4mm$	$4/m2/m2/m$	$4/m'2'/m2'/m$ ($\bar{1}'$)	B2	B2	B2	A	A	A	B2
$\bar{4}2m$	$4/m2/m2/m$	$4'(2)/m'2/m'2'/m$ ($\bar{1}'$)	B2	A	B2	A	A	A	A
$\bar{4}m2$	$4/m2/m2/m$	$4'(2)/m'2'/m2/m'$ ($\bar{1}'$)	B2	B2	A	A	A	A	A

(*c*) Trigonal point groups (hexagonal lattice)

Point group	Composite group	Composite group (black–white notation)	Twin diffraction cases for different types of reflections (face forms)						
			hkil	$h0\bar{h}l$	$hh2\bar{h}l$	*hki*0	$h0\bar{h}0$	$hh2\bar{h}0$	000*l*
3	$\bar{3}$	$\bar{3}'(3)$ ($\bar{1}'$)	All reflections B2						
	321	$32'1$	B1	B2	B1	B1	B2	A	B2
	312	$312'$	B1	B1	B2	B1	A	B2	B2
	$3m1$	$3m'1$	B1	A	B1	B1	A	B2	A
	$31m$	$31m'$	B1	B1	A	B1	B2	A	A
	6	$6'(3)$	B1	B1	B1	B2	B2	B2	A
	$\bar{6}$	$\bar{6}'(3) = 3/m'$	B1	B1	B1	A	A	A	B2
$\bar{3}$	$\bar{3}2/m1$	$\bar{3}2'/m'1$	B1	A	B1	B1	A	A	A
	$\bar{3}12/m$	$\bar{3}12'/m'$	B1	B1	A	B1	A	A	A
	$6/m$	$6'(3)/m'$	B1	B1	B1	A	A	A	A
321	$\bar{3}2/m1$	$\bar{3}'(3)2/m'1$ ($\bar{1}'$)	B2	A	B2	B2	A	B2	A
	$\bar{6}2m$	$\bar{6}'(3)2m'$	B1	B1	A	A	A	A	A
	622	$6'(3)22'$	B1	B1	B2	B2	A	B2	A
312	$\bar{3}12/m$	$\bar{3}'(3)12/m'$ ($\bar{1}'$)	B2	B2	A	B2	B2	A	A
	$\bar{6}m2$	$\bar{6}'(3)m'2$	B1	A	B1	A	A	A	A
	622	$6'(3)2'2$	B1	B1	B2	B2	B2	A	A
$3m1$	$\bar{3}2/m1$	$\bar{3}'2'/m1$ ($\bar{1}'$)	B2	B2	B2	B2	B2	A	B2
	$\bar{6}m2$	$\bar{6}'(3)m2'$	B1	B1	B2	A	A	A	B2
	$6mm$	$6'(3)mm'$	B1	B1	A	B2	B2	A	A
$31m$	$\bar{3}12/m$	$\bar{3}'12'/m$ ($\bar{1}'$)	B2	B2	B2	B2	A	B2	B2
	$\bar{6}2m$	$\bar{6}'(3)2'm$	B1	B2	B1	A	A	A	B2
	$6mm$	$6'(3)m'm$	B1	A	B1	B2	A	B2	A
$\bar{3}2/m1$	$6/m2/m2/m$	$6'(3)/m'2/m2'/m'$	B1	B1	A	A	A	A	A
$\bar{3}12/m$	$6/m2/m2/m$	$6'(3)/m'2'/m'2/m$	B1	A	B1	A	A	A	A

(*d*) Trigonal point groups (rhombohedral lattice)†

Point group	Composite group	Composite group (black–white notation)	Twin diffraction cases for different types of reflections (face forms)						
			hkl	*hhl*	$hk(2k-h)$	$hk(\bar{h}+\bar{k})$	$hh2\bar{h}$	$0h\bar{h}$	*hhh*
3	$\bar{3}$	$\bar{3}'(3)$	All reflections B2						
	32	$32'$	B1	B2	B1	B1	B2	A	B2
	$3m$	$3m'$	B1	A	B1	B1	A	B2	A
$\bar{3}$	$\bar{3}2/m$	$\bar{3}2'/m'$	B1	A	B1	B1	A	A	A
32	$\bar{3}2/m$	$\bar{3}'(3)2/m'$ ($\bar{1}'$)	B2	A	B2	B2	A	B2	A
$3m$	$\bar{3}2/m$	$\bar{3}'(3)2'/m$ ($\bar{1}'$)	B2	B2	B2	B2	B2	A	B2

(*e*) Hexagonal point groups

Point group	Composite group	Composite group (black–white notation)	Twin diffraction cases for different types of reflections (face forms)						
			hkil	$h0\bar{h}l$	$hh2\bar{h}l$	*hki*0	$h0\bar{h}0$	$hh2\bar{h}0$	000*l*
6	6/*m*	$6/m'$ $(\bar{1})$	B2	B2	B2	A	A	A	B2
	622	$62'2'$	B1	B2	B2	B1	A	A	B2
	6*mm*	$6m'm'$	B1	A	A	B1	A	A	A
$\bar{6} = 3/m$	6/*m*	$6'(3)/m$ $(\bar{1})$	B2	B2	B2	B2	B2	B2	A
	$\bar{6}2m$	$\bar{6}2'm'$	B1	B2	A	B1	B2	A	A
	$\bar{6}m2$	$\bar{6}m'2'$	B1	A	B2	B1	A	B2	A
6/*m*	6/*m*2/*m*2/*m*	$6/m2'/m'2'/m'$	B1	A	A	B1	A	A	A
622	6/*m*2/*m*2/*m*	$6/m'2/m'2/m'$ $(\bar{1})$	B2	A	A	A	A	A	A
6*mm*	6/*m*2/*m*2/*m*	$6/m'2'/m2'/m$ $(\bar{1})$	B2	B2	B2	A	A	A	B2
$\bar{6}2m$	6/*m*2/*m*2/*m*	$6'(3)/m2/m'2'/m$ $(\bar{1})$	B2	A	B2	B2	A	B2	A
$\bar{6}m2$	6/*m*2/*m*2/*m*	$6'(3)/m2'/m2/m'$ $(\bar{1})$	B2	B2	A	B2	B2	A	A

(*f*) Cubic point groups

Point group	Composite group	Composite group (black–white notation)	Twin diffraction cases for different types of reflections (face forms)						
			hkl	*hhl*, $h < l$	*hhl*, $h > l$	*hk*0	*hh*0	*hhh*	*h*00
23	$2/m\bar{3}$	$2/m'\bar{3}'(3)$ $(\bar{1})$	B2	B2	B2	A	A	B2	A
	432	$4'(2)32'$	B1	B2	B2	B1	A	B2	A
	$\bar{4}3m$	$\bar{4}'(2)3m'$	B1	A	A	B1	A	A	A
$2/m\bar{3}$	$4/m\bar{3}2/m$	$4'(2)/m\bar{3}2'/m'$	B1	A	A	B1	A	A	A
432	$4/m\bar{3}2/m$	$4/m'\bar{3}'(3)2/m'$ $(\bar{1})$	B2	A	A	A	A	A	A
$\bar{4}3m$	$4/m\bar{3}2/m$	$4'(2)/m'\bar{3}'(3)2'/m$ $(\bar{1})$	B2	B2	B2	A	A	B2	A

† For the rhombohedral Miller indices of the different types of reflections (face forms) see Hahn & Klapper (2002), Table 10.1.2.2, especially pp. 776–782.

Dauphiné twins the above sets with *F*-moduli differing only in their anomalous scattering contributions are superimposed (diffraction cases B2), whereas for the Leydolt twin the *F*-moduli are equal (case A). At first glance the latter result seems to be strange, but it is easily understood from Fig. 8 by considering the Leydolt twin [in the literature often called 'combined quartz twin', *e.g.* Frondel (1962)] as the combination of a Dauphiné (twin rotation $2'$) followed by a Brazil (inversion $\bar{1}'$) twin, resulting in the Leydolt twin (twin reflection plane $m'(0001)$ which is an *eigensymmetry* plane of the trigonal dipyramid).

(ii) As a second kind of face forms, the two twin-related non-equivalent centrosymmetric rhombohedra $\{h0\bar{h}l\}$ and $\{\bar{h}0hl\}$ are shown in Fig. 9. Owing to their *eigen-centrosymmetry* each rhombohedron displays the same *F*-moduli for left- and right-handed crystals. The moduli of the two rhombohedra, however, differ in both anomalous scattering and geometric structure-factor contributions. Thus, they provide diffraction case A for Brazil and B1 for Dauphiné and Leydolt twins. The special forms $\{10\bar{1}1\}$ and $\{\bar{1}011\}$ are the major and minor rhombohedron of the quartz morphology which develop differently during growth in nature. Their *F*-moduli are $|F\{10\bar{1}1\}| = 38.6$, $|F\{\bar{1}011\}| = 24.7$.

Similar sets of four diagrams can be drawn also for the other face forms (reflection sets) of the point group 321 of quartz, as well as for the other merohedral point groups in which *inversion*, *rotation* and *reflection* twins are possible [Tables 8(ii) and 8(iii) of Appendix *A*].

APPENDIX *D*
Survey of the diffraction cases of all 63 merohedral twin laws

Table 9 in this Appendix provides a complete summary of the 63 possible merohedral twin laws in the 35 structural settings of the 26 merohedral crystal classes. Each entry contains the symbols of the untwinned point group and the twin composite group, the latter both in standard and 'black–white' notation (columns 2 and 3). Columns 4 to 10 contain the seven types of reflections (face forms) and their twin diffraction cases A, B1 or B2 (*cf.* §2.2.1).

In the black–white symmetry symbols of a twin (column 3) the primed symbols indicate the twin operation. If the primed operation contains (as sub-element) an operation of the untwinned group, the latter is added in parentheses, *e.g.* 321 → $\bar{3}'(3)2/m'1$ $(\bar{1}')$. Inversion twins are always indicated by $(\bar{1}')$. For names and *eigensymmetries* of the face forms, see Table 1 and Tables 10.1.2.2 and 10.1.2.3 of Hahn & Klapper (2002).

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