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► **To cite this version:**

Massimo Nespolo. The chromatic symmetry of twins and allotwins. *Acta Crystallographica Section A: Foundations and Advances* [2014-..], 2019, 75 (3), pp.551-573. 10.1107/S2053273319000664. hal-02116283

**HAL Id: hal-02116283**

**<https://hal.univ-lorraine.fr/hal-02116283>**

Submitted on 30 Apr 2019

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*Acta Cryst.* (2019). **A75**, 551–573



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# The chromatic symmetry of twins and allotwins

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Received 3 June 2018

Accepted 14 January 2019

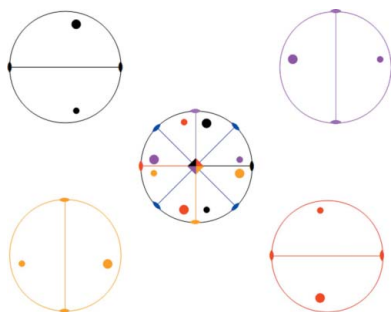
Edited by J.-G. Eon, Universidade Federal do Rio de Janeiro, Brazil

**Keywords:** allotwinning; groupoids; chromatic symmetry; twin point groups; twinning.

The symmetry of twins is described by chromatic point groups obtained from the intersection group  $\mathcal{H}^*$  of the oriented point groups of the individuals  $\mathcal{H}_i$  extended by the operations mapping different individuals. This article presents a revised list of twin point groups through the analysis of their groupoid structure, followed by the generalization to the case of allotwins. Allotwins of polytypes with the same type of point group can be described by a chromatic point group like twins. If the individuals are all differently oriented, the chromatic point group is obtained in the same way as in the case of twins; if they are mapped by symmetry operation of the individuals, the chromatic point group is neutral. If the same holds true for some but not all individuals, then the allotwin can be seen as composed of twinned regions described by a twin point group, that are then allotwinned and described by a colour identification group; the allotwin is then described by a chromatic group obtained as an extension of the former by the latter, and requires the use of extended symbols reminiscent of the extended Hermann–Mauguin symbols of space groups. In the case of allotwins of polytypes with different types of point groups, as well as incomplete (allo)twins, a chromatic point group does not reveal the full symmetry: the groupoid has to be specified instead.

## 1. Introduction

A twin is an oriented crystal association of two or more crystals (usually called ‘individuals’ or ‘components’)<sup>1</sup> of the same phase, oriented one with respect to the other according to precise geometric relations (Friedel, 1904). The operations mapping the orientation of the individuals building the twin are crystallographic operations<sup>2</sup> about direct lattice elements (Mallard’s law: Friedel, 1926, p. 436). Crystal associations with no specific relation between the individuals are called *bicrystals* (Hahn *et al.*, 1999) and are not considered here. The individuals building a twin are homogeneous crystals (not considering defects) and are therefore characterized by a crystallographic point (and space) group. A twin is instead heterogeneous: the interface separating two individuals, which is known as the *composition surface* (of which a special case is a *composition plane*), represents a structural discontinuity across which some of the crystallographic orbits building the structure are (exactly or approximately) continuous



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<sup>1</sup> In a domain structure, each domain occupies a physically distinct position in the crystal space; all the domains with the same orientation constitute a single ‘domain state’, which corresponds to an ‘individual’ in the case of a macroscopic twin (Nespolo, 2015a). Transformation twins, formed during a phase transition, are typically domain structures; growth twins, formed by a change of orientation due to perturbation of the normal crystal growth, or by synneusis (oriented attachment), are typically macroscopic twins (Nespolo & Ferraris, 2004).

<sup>2</sup> A special case of oriented crystal associations exists in which the relative orientation of the individuals can be expressed through a non-crystallographic rotation about the normal to the composition plane, although the operation mapping the individuals is a twofold rotation in the composition plane. These associations are known as plesiotwins (Nespolo, Ferraris *et al.*, 1999; Nespolo, 2018).

(Marzouki *et al.*, 2014). Because of this structural heterogeneity, a twin cannot be described by a space group; and because of the orientation heterogeneity, it cannot be described by a point group either. Nevertheless, because the operations mapping individuals in a twin are still crystallographic operations, a twin can be described by a generalized point group, which contains both symmetry operations (all or some of them, depending on the relative orientations) of the individual and operations mapping the orientation of the different individuals. If one attributes a colour to each individual, the point group of the twin is a chromatic group containing colour-preserving (achromatic) and colour-exchanging (chromatic) operations (Nespolo, 2004). Depending on the number of individuals, the point group of the twin is dichromatic (two individuals), trichromatic (three individuals) and so on; the term *polychromatic* is used for three or more colours (individuals).

A special case of oriented crystal association occurs when individuals are different polytypes of the same compound; these are known as *allotwins* (Nespolo, Kogure *et al.*, 1999). Because polytypes differ by the stacking of a common layer, they have a common two-dimensional lattice in the (*hkl*) plane of the layer, which explains the formation of oriented crystal associations much like in the case of twins. However, because the periodicity along the third direction is different, these associations differ from twins. The relative orientation of two twinned crystals is expressed by the mapping of the corresponding basis vectors; this can be obtained from the relations between the lattices of the individuals and the twin lattice. If  $(\mathbf{abc})_i$  are the basis vectors of the *i*th individual and  $(\mathbf{abc})_T$  those of the twin lattice, the mapping of the former to the latter is obtained by a matrix  $\mathbf{M}_i$  whose determinant corresponds to the ratio of the volumes of the unit cell of the twin lattice and of the individual lattice. The mapping of  $(\mathbf{abc})_i$  to  $(\mathbf{abc})_j$  is obtained by the following transformation:

$$(\mathbf{abc})_j = (\mathbf{abc})_T \mathbf{M}_j^{-1} = (\mathbf{abc})_i \mathbf{M}_i \mathbf{M}_j^{-1} \quad (1)$$

where the matrix  $\mathbf{M}_i \mathbf{M}_j^{-1}$  has determinant 1. In the case of allotwins, the transformation is obtained in the same way but the determinant of the matrix  $\mathbf{M}_i \mathbf{M}_j^{-1}$  is no longer restricted to 1: it coincides with the ratio of the volumes of the unit cell of the two individuals. Another fundamental difference is that the mapping  $\mathbf{M}_i \mathbf{M}_j^{-1}$  may correspond to a symmetry operation of one or more individuals, which is instead impossible in the case of twins. This calls for an extension of the treatment we have presented for twins.

## 2. The chromatic symmetry of a twinned crystal

We remind the reader that a twin operation  $\Phi(i \rightarrow j)$  is a three-dimensional isometry (distance-preserving transformation) mapping the orientation of the *i*th individual onto the orientation of the *j*th individual of the twin. The twin operations are associative and invertible [ $\Phi(i \rightarrow j)^{-1} = \Phi(j \rightarrow i)$ ] but they do not obey the closure property of a group: in particular, the identity is not a twin operation.

The symmetry operations of the oriented point group  $\mathcal{H}_i$  of the *i*th individual leave invariant the orientation in space of that individual. The isometries that leave invariant the orientation in space of all the individuals of the twin define the intersection group  $\mathcal{H}^* = \cap_i \mathcal{H}_i$ , *i.e.* the maximal common subgroup of the oriented point groups  $\mathcal{H}_i$ . This group can be extended by the twin operation(s) to form a group  $\mathcal{K}$  which leaves invariant the orientation in space of the twinned edifice, while exchanging the orientation of some or all of the individuals: it represents the symmetry of the twin.

When considering the result of the action of the operations in  $\mathcal{K}$  on each individual building the twin, these operations can be classified into two types: those which leave invariant each of the individuals, which belong to  $\mathcal{H}^*$  and are denoted  $\Phi(i \rightarrow i)$  in the following; and those which map different individuals, denoted  $\Phi(i \rightarrow j)$ . We can assign to each individual a colour; then, the operations  $\Phi(i \rightarrow i)$  fix the colours whereas the operations  $\Phi(i \rightarrow j)$  exchange them. The group  $\mathcal{K}$  is therefore a chromatic group, in the following indicated as  $\mathcal{K}^{(p)}$ , where *p* is the number of colours (*i.e.* individuals or domain states of the twin). Hereafter we define *chromaticity* as the pattern of colour exchange (two different operations that exchange the same colours are said to have the same chromaticity) and *chromatic index* as the number of colours exchanged, which coincides with the index of  $\mathcal{H}^*$  in  $\mathcal{K}^{(p)}$ , *i.e.* the ratio of the order of the two point groups:  $p = |\mathcal{K}^{(p)}|/|\mathcal{H}^*|$ . The combination of two colour-fixing operations is again a colour-fixing operation and the combination of a colour-fixing operation and a colour-exchanging operation is a colour-exchanging operation. However, the combination of two colour-exchanging operations  $\Phi(i \rightarrow j)\Phi(j \rightarrow k)$  can be either a colour-exchanging or a colour-fixing operation, depending on whether  $k \neq i$  or  $k = i$ . If all the combinations  $\Phi(i \rightarrow j)\Phi(j \rightarrow k)$  correspond to an existing colour, *i.e.* to the orientation of an individual developed and present in the twin, one speaks of a complete twin; in the opposite case, the twin is incomplete (Nespolo, 2004). An incomplete twin can be seen as a twin in which some of the individuals have zero volume: from the algebraic viewpoint this is simply a special case, but from the physical viewpoint the absence of some individuals has a significant influence on the properties of the twin, in particular on the diffraction pattern [see the analysis of merohedric twins in Nespolo *et al.* (2014)], and for this reason the case of incomplete twins is emphasized in the following.

By decomposing the twin point group  $\mathcal{K}^{(p)}$  with respect to  $\mathcal{H}^*$  one obtains *p* cosets, where *p* is the chromatic index defined above, the first of which is  $\mathcal{H}^*$  itself. A *twin law* is defined as any coset of  $\mathcal{H}^*$  in  $\mathcal{K}^{(p)}$  but  $\mathcal{H}^*$  itself; a twin whose twin point group is  $\mathcal{K}^{(p)}$  is composed of *p* individuals (domain states) in *p* different orientations. The operations mapping different individuals are called *twin operations*; all the operations with the same chromaticity (*i.e.* that map the same pair of individuals) belong to the same coset of  $\mathcal{H}^*$  in  $\mathcal{K}^{(p)}$  and constitute a twin law. In other terms, the relative orientation of two individuals is equivalently obtained by any of the twin operations belonging to the same twin law (coset of  $\mathcal{H}^*$  in  $\mathcal{K}^{(p)}$ ). A twin element is defined in the same way as a symmetry

element, *i.e.* as the combination of the geometric element (point, line or plane) left invariant by the twin operation, and the element set, *i.e.* the set of twin operations sharing the same geometric element. The chromatic index applies not only to the twin group  $\mathcal{K}$  but also to the twin elements. A mirror plane or an inversion centre is either a symmetry element (achromatic: no chromatic index) or a twin element (dichromatic: its chromatic index is 2). A (direct or inverse) rotation axis  $[uvw]$  can act both as twin element and as (non-trivial) symmetry element if the twin operation performed about it is of higher order: a 6 or  $\bar{6}$  twin operation about a twofold (chromatic index 3) or threefold (chromatic index 2) symmetry axis, a 4 or  $\bar{4}$  twin operation about a twofold symmetry operation (chromatic index 2), a  $\bar{3}$  twin operation about a threefold symmetry axis (chromatic index 2). We (Nespolo, 2004) have called *twin multiplicity* the number of individuals in a twin, and *twin degree* the number of independent twin elements: first-degree twins and higher-degree twins are twins in which only one or more than one independent twin elements exist, respectively. Two twin elements are here considered *independent* if the corresponding twin operations belong to different twin laws (cosets of  $\mathcal{K}^{(p)}$  with respect to  $\mathcal{H}^*$ ). Each twin element brings a chromatic index  $p_i$ , and the chromatic index of the group  $p$  may coincide with one of the  $p_i$  if the corresponding twin element exchanges all the individuals in the twin, or be a multiple of it otherwise. In first-degree twins, there is necessarily one twin element (possibly the only one) whose chromatic index  $p_i$  coincides with the chromatic index  $p$  of the group.

The operations in the chromatic point group  $\mathcal{K}^{(p)}$  can be classified as follows:

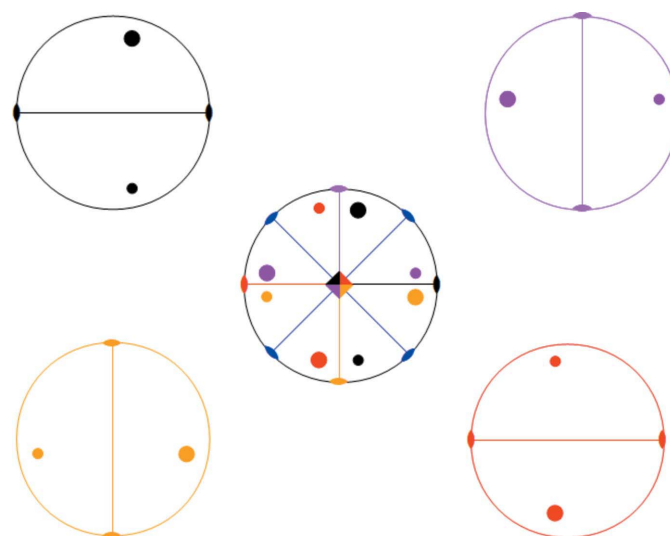
- (i) The achromatic operations  $\Phi(i \rightarrow i)$ , which belong to  $\mathcal{H}^*$ .
- (ii) The operations  $\Phi(i \rightarrow j)$  which exchange all the individuals of the twin are *totally chromatic operations*.
- (iii) Some operations  $\Phi(i \rightarrow j; k \rightarrow k)$  may fix a subset of the individuals ( $k \rightarrow k$ ) while exchanging another subset ( $i \rightarrow j$ ), and are *partially chromatic operations*.

Partially chromatic operations are operations of  $\mathcal{H}_i$  which do not appear in  $\mathcal{H}^*$  because they do not leave invariant the orientation of *all* of the individuals but only of some of them; they however leave invariant the orientation of the whole twinned edifice. For example, a twin composed of four individuals with point group of type 121, all oriented with their [001] directions parallel to each other, rotated by  $n \times 90^\circ$  about this direction ( $n$  integer) has intersection group  $\mathcal{H}^* = 1$  because the twofold rotation in each orientation does not leave invariant all the individuals but only two of them (Fig. 1). The four individuals are identified by the codes B, M, R, Y for black, magenta, red and yellow, respectively. The twin lattice is tetragonal and the fourfold rotation about the [001] direction exchanges all the four individuals and is therefore tetrachromatic:  $\Phi(i \rightarrow j) = 4(B \rightarrow M, M \rightarrow R, R \rightarrow Y, Y \rightarrow B)$ . The twofold rotations about the  $\langle 100 \rangle$  directions are partially chromatic operations of the twin, because they leave invariant two individuals while exchanging the other two:  $\Phi(i \rightarrow j; k \rightarrow k) = 2_{[010]} (B \rightarrow B, M \rightarrow Y, Y \rightarrow M, R \rightarrow R)$  and  $\Phi(i \rightarrow j; k \rightarrow k) = 2_{[100]} (B \rightarrow R, M \rightarrow M, R \rightarrow B, Y \rightarrow Y)$ . Finally, the combination of the

chromatic fourfold rotation with the partially chromatic twofold rotations generates chromatic twofold rotation about the  $\langle 110 \rangle$  directions  $\Phi(i \rightarrow j) = 2_{[110]} (B \rightarrow Y, Y \rightarrow B, R \rightarrow M, M \rightarrow BY)$  and  $\Phi(i \rightarrow j) = 2_{[\bar{1}\bar{1}0]} (B \rightarrow M, M \rightarrow B, R \rightarrow Y, Y \rightarrow R)$ , so that the twin point group is tetrachromatic, with symbol  $\mathcal{K}^{(4)} = (4^{(4)}2^{(2,2)}2^{(2)})^{(4)}$ .

In the example above, the operations of  $\mathcal{H}^*$  that are not present in the point group  $\mathcal{H}$  occur in  $\mathcal{K}^{(p)}$  as partially chromatic operations. This does not always happen. For example, the twins of quartz with inclined axes, *i.e.* twins in which the threefold axes of the individuals are not parallel to each other, occur with various relative orientations depending on the twin and composition plane: Japan  $\{11\bar{2}2\}$ , Esterel  $\{10\bar{1}1\}$ , Sella  $\{10\bar{1}2\}$ , Belowda Beacon  $\{30\bar{3}2\}$ , Breithaupt  $\{11\bar{2}1\}$ , Wheal Coates  $\{21\bar{3}1\}$ , Cornouailles  $\{20\bar{2}1\}$ , Pierre-Levée  $\{21\bar{3}3\}$  (Friedel, 1923). In all these twins,  $\mathcal{H}^* = 1$ ,  $\mathcal{K} = m'$ . None of the symmetry operations of the individuals remains in the intersection group, nor do they appear in the chromatic point group.

Depending on whether partially chromatic operations do or do not occur in the twin point group,  $\mathcal{K}^{(p)}$  is a non-invariant or invariant extension of the intersection point group  $\mathcal{H}^*$ . We (Nespolo, 2004) have listed the twin point groups on the basis of the detailed derivation of chromatic point groups given by Shubnikov & Koptsik (1974). Here we present a revised list of



**Figure 1**  
Stereographic projection of the symmetry elements and general form of four point groups of type 2 oriented at  $90^\circ$  from each other around the vertical to the plane containing the twofold axis. The stereographic poles of the four individuals are drawn with four different colours; poles on the different hemispheres are distinguished by their size (smaller one in the northern hemisphere). The projection in the centre of the figure is the superposition of the projections corresponding to the four orientations; it also shows the twin elements mapping the four individuals. The fourfold axis normal to the plane of the projection is tetrachromatic and represented by a square composed of four right triangles of different colours. The twofold axes along  $\langle 100 \rangle$  are partially chromatic: they are symmetry elements for two individuals and exchange the two other individuals; for this reason they are shown with two different colours. The twofold axes along  $\langle 110 \rangle$  are totally chromatic: they exchange all the four individuals' colours in pairs; for this reason they are shown with a single colour. The tetrachromatic point group is  $\mathcal{K}^{(4)} = (4^{(4)}2^{(2,2)}2^{(2)})^{(4)}$ .

Table 1

List of chromatic point groups expressing the possible symmetries of twinned crystals.

Separate sequential numbers are given for dichromatic  $\mathcal{K}^{(2)}$ , polychromatic invariant  $\mathcal{K}^{(p>2)}$  and polychromatic non-invariant  $\mathcal{K}_{\text{WB}}^{(p>2)}$  extensions of achromatic point groups. In the construction of cubic twin point groups, different twin elements along directions that become equivalent in the cubic axial setting are indicated by a Greek suffix. In particular,  $2_{\alpha}$  and  $2_{\beta}$  stand for any two of the three directions (100) that are equivalent in a cubic crystal. For cubic polychromatic groups, whenever necessary to avoid ambiguity, the symbol of the intersection group  $\mathcal{H}^*$  is given twice: once as oriented symbol with respect to the cubic basis vectors, and once as standard symbol with respect to the basis vectors of  $\mathcal{H}^*$ .

No. $\mathcal{K}^{(2)}$	No. $\mathcal{K}^{(p>2)}$	No. $\mathcal{K}_{\text{WB}}^{(p>2)}$	$\mathcal{K}$	$\mathcal{H}^*$	1st twin element	2nd twin element	3rd twin element	4th twin element
1			$\bar{1}'$	1	$\bar{1}$			
2			$m'$	1	$m$			
3			$2'$	1	$2$			
4			$2'/m$	$m$	$\bar{1}$			
5			$2/m'$	$2$	$\bar{1}$			
6			$2'/m'$	$\bar{1}$	$2 (m)$			
7	1		$(2^{(2)}/m^{(2)})^{(4)}$	1	$\bar{1}$		2	
8			$m'm2'/mm'2'$	$1m1 / m11$	112			
9	2		$m'm'2$	112	$1m1$			
10			$(m^{(2)}m^{(2)}2^{(2)})^{(4)}$	1	$1m1$		$m11$	
11	3		$2'22'/22'2'$	$121 / 211$	112			
12			$(2^{(2)}2^{(2)}2^{(2)})^{(4)}$	1	121		112	
13	4		$2'/m'2/m2'/m'$	$12/m1$	112			
14			$2'/m2'/m2/m'$	$mm2$	121			
15			$2/m2'/m2'/m'$	222	$\bar{1}$			
16			$(2^{(2)}/m^{(2)}2/m^{(2)}2^{(2)}/m^{(2)})^{(4)}$	121	$\bar{1}$		112	
17			$(2^{(2)}/m^{(2)}2^{(2)}/m2^{(2)}/m^{(2)})^{(4)}$	$1m1$	$\bar{1}$		112	
18			$(2^{(2)}/m^{(2)}2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(4)}$	$\bar{1}$	121		112	
19			$(2^{(2)}/m^{(2)}2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(8)}$	1	$\bar{1}$		121	112
20	8		$4'$	2	4			
21			$4^{(4)}$	1	4			
22	9		$4'$	2	$\bar{4}$			
23			$\bar{4}^{(4)}$	1	$\bar{4}$			
24			$4'/m$	$2/m$	$\bar{4}$			
25			$4/m'$	4	$\bar{1}$			
26			$4'/m'$	$\bar{4}$	$\bar{1}$			
27	10		$(4^{(4)}/m^{(2)})^{(4)}$	$\bar{1}$	4			
28			$(4^{(2)}/m^{(2)})^{(4)}$	2	$\bar{1}$		4	
29			$(4^{(4)}/m)^{(4)}$	$m$	4			
30			$(4^{(4)}/m^{(2)})^{(8)}$	1	$\bar{1}$		411	
31	18		$4m'm'$	4	$1m1$			
32			$4'mm'/4'm'm$	$2mm. / 2.mm$	411			
33	14		$(4^{(2)}/m^{(2)}/m^{(2)})^{(4)}$	211	411		$1m1$	
34			$(4^{(4)}/m^{(2,2)}/m^{(2)})^{(4)}$	$1m1 / 11m$	411			
35	15	1	$(4^{(4)}/m^{(2)}/m^{(2)})^{(8)}$	1	$1m1$		411	
36			$42'2'$	4	121			
37			$4'22' / 4'2'2$	$222. / 2.22$	411			
38	16		$(4^{(2)}/2^{(2)}/2^{(2)})^{(4)}$	211	411		121	
39			$(4^{(4)}/2^{(2,2)}/2^{(2)})^{(4)}$	$121 / 112$	411			
40	17	2	$(4^{(4)}/2^{(2)}/2^{(2)})^{(8)}$	1	121		411	
41			$4m'2' / 42'm'$	$4$	$1m1 / 121$			
42			$4m2' / 4'2'm$	$2mm. / 2.mm$	411			
43	18		$(\bar{4}^{(2)}/m^{(2)}/2^{(2)})^{(4)}$	211	411		$1m1 / 121$	
44			$(\bar{4}^{(4)}/m^{(2,2)}/2^{(2)})^{(4)}$	$1m1 / 11m$	411			
45	3		$(\bar{4}^{(4)}/m^{(2)}/2^{(2,2)})^{(4)}$	112 / 121	411			
46			$(\bar{4}^{(4)}/m^{(2)}/2^{(2,2)})^{(8)}$	1	$1m1 / 121$		411	
47	19	4	$(\bar{4}^{(4)}/m^{(2)}/2^{(2)})^{(8)}$	222. / 2.22	411			
48			$4'2m' / \bar{4}'m'2$	4/m	121			
49			$4/m2'/m'2'/m'$	$4mm$	$\bar{1}$			
50			$4/m'2'/m2'/m$	422	$\bar{1}$			
51			$4/m'2'/m2'/m'$	$\bar{4}m2 / \bar{4}2m$	$\bar{1}$			
52			$4'/m'2'/m2'/m' / 4'/m'2'/m'2'/m$	$2/m2/m2/m. / 2/m.2/m2/m$	411			
53	20		$(4/m^{(2)}/2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(4)}$	4	$\bar{1}$		121	
54			$(4^{(2)}/m^{(2)}/2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(4)}$	4	$\bar{1}$		121	
55			$(4^{(2)}/m^{(2)}/2/m^{(2)}/2^{(2)}/m^{(2)})^{(4)}$	$222. / 2.22$	$\bar{1}$		411	
56			$(4^{(2)}/m^{(2)}/2^{(2)}/m2^{(2)}/m^{(2)})^{(4)}$	$2mm. / 2.mm$	$\bar{1}$		411	
57	24	5	$(4^{(4)}/m2^{(2,2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(4)}$	$m2m / mm2$	411			
58			$(4^{(2)}/m2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(4)}$	$2/m11$	411		121	
59			$(4^{(4)}/m^{(2)}/2^{(2,2)}/m^{(2,2)}/m^{(2)})^{(4)}$	$12/m1$	411			
60			$(4^{(4)}/m^{(2)}/2^{(2,4)}/m^{(2)}/2^{(2)}/m^{(2)})^{(8)}$	121	$\bar{1}$		411	
61			$(4^{(4)}/m^{(2)}/2^{(2)}/m^{(2,4)}/2^{(2)}/m^{(2)})^{(8)}$	$1m1$	$\bar{1}$		121	
62	25		$(4^{(4)}/m2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(8)}$	$m11$	411		121	
63			$(4^{(2)}/m^{(2)}/2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(8)}$	211	$\bar{1}$		411	121
64			$(4^{(4)}/m^{(2)}/2^{(2)}/m^{(2)}/2^{(2)}/m^{(2)})^{(8)}$	1	121		411	

Table 1 (continued)

No. $\mathcal{K}^{(2)}$	No. $\mathcal{K}^{(p>2)}$	No. $\mathcal{K}_{WB}^{(p>2)}$	$\mathcal{K}$	$\mathcal{H}^*$	1st twin element	2nd twin element	3rd twin element	4th twin element
28			$(4^{(4)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2)}/m^{(2)})^{(16)}$	1	$\bar{1}$		411	
29			$3^{(3)}$	1	$\bar{3}$			
30			$\bar{3}'$	3	$\bar{1}$			
30			$\bar{3}^{(3)}$	1	$\bar{3}$			
31			$\bar{3}^{(6)}$	1	$\bar{3}$			
31			$3m' / 3m'1 / 31m'$	3	$1m / 1m1 / 11m$			
32	9		$(3^{(3)}m^{(2,1)}_1)^{(3)} / (3^{(3)}m^{(2,1)}_1)^{(3)} / (3^{(3)}1m^{(2,1)})^{(3)}$	$1m / 1m1 / 11m$	3			
32			$(3^{(3)}m^{(2)}_1)^{(6)} / (3^{(3)}m^{(2)}_1)^{(6)} / (3^{(3)}1m^{(2)})^{(6)}$	1	$1m / 1m1 / 11m$	3		
32			$32' / 32'1 / 312'$	3	$12 / 121 / 112$			
33	10		$(3^{(3)}2^{(2,1)}_1)^{(3)} / (3^{(3)}2^{(2,1)}_1)^{(3)} / (3^{(3)}12^{(2,1)})^{(3)}$	$12 / 121 / 112$	3			
33			$(3^{(3)}2^{(2)}_1)^{(6)} / (3^{(3)}2^{(2)}_1)^{(6)} / (3^{(3)}12^{(2)})^{(6)}$	1	$12 / 121 / 112$	3		
33			$32'/m' / 32'/m'1 / 312'/m'$	$\bar{3}$	$12 / 121 / 112$			
34			$\bar{3}2/m' / \bar{3}2/m'1 / \bar{3}12/m'$	32 / 321 / 312	$\bar{1}$			
35			$\bar{3}2'/m / \bar{3}12'/m / \bar{3}2'/m1$	$3m / 3m1 / 31m$	$\bar{1}$			
35	11		$(\bar{3}^{(3)}2^{(2,1)}_1/m^{(2,1)}_1)^{(3)} / (\bar{3}^{(3)}2^{(2,1)}_1/m^{(2,1)}_1)^{(3)} / (\bar{3}^{(3)}12^{(2,1)}/m^{(2,1)})^{(3)}$	$12m / 12/m1 / 112/m$	3			
34			$(\bar{3}2^{(2)}_1/m^{(2)}_1)^{(4)} / (\bar{3}2^{(2)}_1/m^{(2)}_1)^{(4)} / (\bar{3}12^{(2)}/m^{(2)})^{(4)}$	$\bar{3}$	$12 / 121 / 112$			
34	12		$(\bar{3}^{(6)}2^{(2,2)}_1/m^{(2)}_1)^{(6)} / (\bar{3}^{(6)}2^{(2,2)}_1/m^{(2)}_1)^{(6)} / (\bar{3}^{(6)}12^{(2,2)}/m^{(2)}_1)^{(6)}$	$12 / 121 / 112$	$\bar{3}$			
34	13		$(\bar{3}^{(6)}2^{(2)}/m^{(2,2)})^{(6)} / (\bar{3}^{(6)}12^{(2)}/m^{(2,2)})^{(6)} / (\bar{3}^{(6)}2^{(2)}/m^{(2,2)})^{(6)}$	$1m / 1m1 / 11m$	$\bar{3}$			
35			$(\bar{3}^{(3)}2^{(2)}/m^{(2)})^{(6)} / (\bar{3}^{(3)}12^{(2)}/m^{(2)})^{(6)} / (\bar{3}^{(3)}2^{(2)}/m^{(2)})^{(6)}$	$\bar{1}$	$12 / 121 / 112$	3		
36			$(\bar{3}^{(6)}2^{(2)}/m^{(2)})^{(12)} / (\bar{3}^{(6)}12^{(2)}/m^{(2)})^{(12)} / (\bar{3}^{(6)}2^{(2)}/m^{(2)})^{(12)}$	1	$12 / 121 / 112$	$\bar{3}$		
36			$6'$	3	2			
37			$6^{(3)}$	2	3			
38			$6^{(6)}$	1	6			
37			$\bar{6}'$	3	$m$			
39			$\bar{6}^{(3)}$	$m$	3			
40			$\bar{6}^{(6)}$	1	$\bar{6}$			
38			$6/m'$	6	$\bar{1}$			
39			$6'/m$	$\bar{6}$	$\bar{1}$			
40			$6'/m'$	$\bar{3}$	2			
41			$(6^{(3)}/m)^{(3)}$	$2/m$	$\bar{3}$			
42			$(6^{(2)}/m^{(2)})^{(4)}$	3	$\bar{1}$	2		
43			$(6^{(6)}/m^{(2)})^{(6)}$	1	$m$	3		
44			$(6^{(6)}/m)^{(6)}$	$m$	6			
45			$(6^{(3)}/m^{(2)})^{(6)}$	2	$\bar{3}$			
46			$(6^{(6)}/m^{(2)})^{(12)}$	1	$m$	6		
41			$6'mm' / 6'm'm$	$3m1 / 31m$	211			
42			$6m'm'$	6	$1m1$			
42	14		$(6^{(3)}m^{(2,1)}_1/m^{(2,1)}_1)^{(3)}$	$2mm (mm2)$	311			
47			$(6^{(2)}/m^{(2)}_1)^{(4)}$	311	211	$1m1$		
48			$(6^{(3)}/m^{(2)}_1)^{(6)}$	211	311	$1m1$		
48	15		$(6^{(6)}/m^{(2,2)}_1)^{(6)} / (6^{(6)}/m^{(2,2)}_1)^{(6)}$	$1m1 / 11m$	611			
49			$(6^{(6)}/m^{(2)}_1)^{(12)}$	1	611	$1m1$		
43			$6'22' / 6'2'2$	$321 / 312$	211			
44			$62'2'$	6	121			
44	16		$(6^{(3)}2^{(2,1)}_1)^{(3)}$	222	311			
50			$(6^{(2)}/2^{(2)}_1)^{(4)}$	311	211	121		
51	17		$(6^{(6)}/2^{(2,2)}_1)^{(6)} / (6^{(6)}/2^{(2,2)}_1)^{(6)}$	$121 / 112$	611			
51			$(6^{(3)}/2^{(2)}_1)^{(6)}$	211	311	121		
52			$(6^{(6)}/2^{(2)}_1)^{(12)}$	1	611	121		
45			$\bar{6}2'/m' / \bar{6}m'2'$	$\bar{6}$	$121 / 112$			
46			$\bar{6}2m' / \bar{6}m'2$	$321 / 312$	$m..$			
47			$\bar{6}2'm / \bar{6}m2'$	$31m / 3m1$	$m..$			
47	18		$(\bar{6}^{(3)}2^{(2,1)}_1/m^{(2,1)}_1)^{(3)} / (\bar{6}^{(3)}2^{(2,1)}_1/m^{(2,1)}_1)^{(3)}$	$m2m / mm2$	311			
53			$(\bar{6}^{(2)}/2^{(2)}_1)^{(4)} / (\bar{6}^{(2)}/2^{(2)}_1)^{(4)}$	311	$m11$	$121 / 112$		
53	19		$(\bar{6}^{(6)}/2^{(2,2)}_1)^{(6)} / (\bar{6}^{(6)}/2^{(2,2)}_1)^{(6)}$	$121 / 112$	$\bar{6}11$			
53	20		$(\bar{6}^{(6)}/2^{(2)}_1)^{(6)} / (\bar{6}^{(6)}/2^{(2)}_1)^{(6)}$	$11m / 1m1$	$\bar{6}11$			
54			$(\bar{6}^{(3)}/2^{(2)}_1)^{(6)} / (\bar{6}^{(3)}/2^{(2)}_1)^{(6)}$	$m11$	311	$121 / 112$		
55			$(\bar{6}^{(6)}/2^{(2)}_1)^{(12)} / (\bar{6}^{(6)}/2^{(2)}_1)^{(12)}$	1	$\bar{6}11$	$121 / 112$		
48			$6'/m'2/m2'/m' / 6'/m'2'/m'2/m'$	$32/m1 / \bar{3}12/m$	$m11$	$112 / 121$		
59			$6/m2'/m'2'/m'$	$6/m$	121			
50			$6/m'2'/m'2'/m'$	622	$\bar{1}$			
51			$6/m'2'/m2'/m$	$6mm$	$\bar{1}$			
52			$6'/m2'/m'2'/m' / 6'/m2'/m2'/m'$	$\bar{6}2m / \bar{6}m2$	$\bar{1}$			
56			$(6^{(6)}/2^{(2)}/m^{(2)}_1)^{(4)}$	611	$\bar{1}$	121		
57			$(6^{(2)}/m2^{(2)}/m^{(2)}_1)^{(4)}$	$\bar{6}11$	$\bar{1}$	121		
58			$(6^{(2)}/m^{(2)}_1)^{(4)}$	311	211	121		
59			$(6^{(2)}/m^{(2)}_1)^{(4)} / (6^{(2)}/m^{(2)}_1)^{(4)}$	$321 / 312$	$\bar{1}$	$m11$	$112 / 121$	
60			$(6^{(2)}/m^{(2)}_1)^{(4)} / (6^{(2)}/m^{(2)}_1)^{(4)}$	$3m1 / 31m$	$\bar{1}$	$m11$	$112 / 121$	
60	21		$(6^{(3)}/m2^{(2,1)}_1)^{(3)} / (6^{(3)}/m2^{(2,1)}_1)^{(3)}$	$2/m2/m2/m$	3			
61			$(6^{(3)}/m2^{(2)}/m^{(2)}_1)^{(6)}$	$2/m11$	3	121		

Table 1 (continued)

No. $\mathcal{K}^{(2)}$	No. $\mathcal{K}^{(p>2)}$	No. $\mathcal{K}_{WB}^{(p>2)}$	$\mathcal{K}$	$\mathcal{H}^*$	1st twin element	2nd twin element	3rd twin element	4th twin element
		22	$(6^{(3)}/m^{(2)}2^{(2,2)}/m^{(2)}2^{(2,2)}/m^{(2)})^{(6)}$	222	$\bar{3}$			
		23	$(6^{(3)}/m^{(2)}2^{(2)}/m^{(2,2)}2^{(2)}/m^{(2,2)})^{(6)}$	2mm	$\bar{3}$			
		24	$(6^{(6)}/m2^{(2)}/m^{(2,2)}2^{(2,2)}/m^{(2)})^{(6)}$ / $(6^{(6)}/m2^{(2,2)}/m^{(2)}2^{(2)}/m^{(2,2)})^{(6)}$	mm2 / m2m	$\bar{3}$			
		25	$(6^{(6)}/m^{(2)}2^{(2,2)}/m^{(2,2)}2^{(2)}/m^{(2)})^{(6)}$ / $(6^{(6)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2,2)}/m^{(2,2)})^{(6)}$	12/m1 / 112/m	611			
	62		$(6^{(2)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2)}/m^{(2)})^{(8)}$	311	$\bar{1}$		211	121
	63		$(6^{(6)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2)}/m^{(2)})^{(12)}$	$\bar{1}$	611		121	
	64		$(6^{(6)}/m2^{(2)}/m^{(2)}2^{(2)}/m^{(2)})^{(12)}$	m11	611		121	
	65		$(6^{(3)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2)}/m^{(2)})^{(12)}$	211	$\bar{3}11$		121	
	26		$(6^{(6)}/m^{(2)}2^{(2,4)}/m^{(2)}2^{(2)}/m^{(2)})^{(12)}$ / $(6^{(6)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2,4)}/m^{(2)})^{(12)}$	121 / 112	611		$\bar{1}$	
	27		$(6^{(6)}/m^{(2)}2^{(2)}/m^{(2,4)}2^{(2)}/m^{(2)})^{(12)}$ / $(6^{(6)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2)}/m^{(2,4)})^{(12)}$	1m1 / 11m	611		$\bar{1}$	
	66		$(6^{(6)}/m^{(2)}2^{(2)}/m^{(2)}2^{(2)}/m^{(2)})^{(24)}$	1	611		$\bar{1}$	121
	67		$(2^3)^{(3)}$	2.. (222)	13			
	28		$(2^2)^3^{(3,1)}$	.3.	21			
	29		$(2^{(2,2)}3^3)^{(6)}$	2..	13			
	68		$(2^2)^3^{(3)}$	1	$2_\alpha^1$	$2_\beta^1$		13
53			$2/m' \bar{3}$	23	$\bar{1}$			
	69		$(2/m \bar{3}^3)^{(3)}$	$2/m.. (2/m2/m2/m)$	3			
	30		$(2^2)^2/m^{(2)} \bar{3}^{(3,1)}$	3	$2/m$			
	31		$(2^{(2,2)}/m^{(2,4)} \bar{3}^6)^{(6)}$	mm2.. (mm2)	2		131	
	32		$(2^{(2,2)}/m^{(2,2)} \bar{3}^3)^{(6)}$	$2/m.. (2/m)$	2		131	
	70		$(2/m^{(2)} \bar{3}^6)^{(6)}$	2.. (222)	$\bar{1}31$			
	33		$(2^2)^2/m^{(2)} \bar{3}^{(6,0)}$	13	$\bar{1}$		211	
	71		$(2^2)^2/m^{(2)} \bar{3}^{(3)}$	$\bar{1}$	$2_\alpha$		$2_\beta$	131
	34		$(2^{(2,4)}/m^{(2)} \bar{3}^6)^{(12)}$	2.. (2)	2		131	
	35		$(2^2)^2/m^{(2,4)} \bar{3}^6)^{(12)}$	m.. (m)	2		$\bar{1}31$	
	72		$(2^2)^2/m^{(2)} \bar{3}^6)^{(24)}$	1	$2_\alpha$	$2_\beta$		131
54			$4'32'$	23	411			
	36		$(4^{(2,1)}/3^3)^2^{(2,1)}$	422	131			
	37		$(4^4)^3^{(3,1)}2^{(2,2)}$	312/321	411			
	73		$(4^2)^3^3^2^{(2)}$	2.. (222)	131		112	
	38		$(4^{(4,2)}/3^3)^2^{(2)}$	4..	131			
	39		$(4^{(4,0)}/3^3)^2^{(2,2)}$	2.22 (222)	131			
	40		$(4^4)^3^{(3,2)}2^{(2)}$	131	411			
	41		$(4^{(4,0)}/3^3)^2^{(2)}$	2.. (2)	131		411	
	42		$(4^4)^3^{(3)}2^{(2,2)}$	..2 (2)	131			
	74		$(4^4)^3^{(3)}2^{(2)}$	1	$2_\alpha$	$2_\beta$	131	112
55			$4'3m'$	23	411			
	43		$(4^{(2,1)}/3^3)^m^{(2,1)}$	4.2m	131			
	44		$(4^4)^3^{(3,1)}m^{(2,2)}$	.3m	411			
	45		$(4^{(4,2)}/3^3)^m^{(2)}$	4..	131			
	75		$(4^2)^3^3^m^{(2)}$	2.. (222)	131		11m	
	46		$(4^{(4,0)}/3^3)^m^{(2,2)}$	2.mm (2mm)	411		131	
	47		$(4^4)^3^{(3,2)}m^{(2)}$	131	411			
	48		$(4^{(4,0)}/3^3)^m^{(2)}$	2.. (2)	131		$\bar{4}11$	
	49		$(4^4)^3^3^m^{(2,2)}$	..m (m)	131		411	
	76		$(4^4)^3^3^m^{(2)}$	1	$2_\alpha$	$2_\beta$	131	11m
56			$4'/m \bar{3}2'/m'$	$2/m\bar{3}$	411			
57			$4/m' \bar{3} 2/m'$	432	$\bar{1}$			
58			$4'/m' \bar{3} 2'/m$	$\bar{4}3m$	411			
	50		$(4^{(2,1)}/m \bar{3}^3)^2^{(2,1)}/m^{(2,1)}$	4/m2/m2/m	131			
	77		$(4^2)^2/m^{(2)} \bar{3}^2^2/m^{(2)}$	23	112		$\bar{1}$	
	51		$(4^4)^m^{(2)} \bar{3}^{(3,1)}2^{(2,2)}/m^{(2,2)}$	$\bar{3}2/m1 / \bar{3}12/m$	411			
	78		$(4^2)^2/m \bar{3}^3^2^2/m^{(2)}$	m.. (mmm)	$\bar{1}31$			
	52		$(4^{(4,2)}/m^{(2,2)} \bar{3}^3)^2^2/m^{(2)}$	4/m	131			
	53		$(4^{(4,2)}/m^{(2,4)} \bar{3}^6)^2^2/m^{(2)}$	4.mm (4mm)	$\bar{1}31$			
	54		$(4^{(2,2)}/m^{(2)} \bar{3}^6)^2^2/m^{(2)}$	4.22 (422)	$\bar{1}31$			
	55		$(4^{(4,0)}/m^{(2,2)} \bar{3}^3)^2^2/m^{(2,2)}$	m.mm (mmm)	$\bar{1}31$			
	56		$(4^{(4,0)}/m^{(2,4)} \bar{3}^6)^2^2/m^{(2)}$	4m.2 (4m2)	$\bar{1}31$			
	57		$(4^{(2,2)}/m^{(2)} \bar{3}^6)^2^2/m^{(2)}$	42.m (42m)	131			
	58		$(4^4)^m^{(2)} \bar{3}^{(3,2)}2^2/m^{(2)}$	.3.	411		11m	
	59		$(4^4)^m^{(2)} \bar{3}^{(6,0)}2^{(2,4)}/m^{(2,4)}$	.3m	$\bar{1}$		411	
	60		$(4^4)^m^{(2)} \bar{3}^{(6,0)}2^{(2,4)}/m^{(2)}$	.32	$\bar{1}$		411	
	79		$(4^2)^2/m^{(2)} \bar{3}^6^2^2/m^{(2)}$	2.. (222)	$\bar{1}31$		112	
	61		$(4^{(4,4)}/m^{(2)} \bar{3}^6)^2^2/m^{(2)}$	4..	$\bar{1}31$			
	62		$(4^{(4,0)}/m^{(2)} \bar{3}^6)^2^2/m^{(2)}$	4..	$\bar{1}31$			
	63		$(4^{(4,0)}/m^{(2)} \bar{3}^6)^2^2/m^{(2)}$	2.22 (222)	$\bar{1}31$		411	
	64		$(4^{(4,0)}/m^{(2,8)} \bar{3}^6)^2^2/m^{(2)}$	2mm.. (mm2)	131		411	
	65		$(4^{(4,0)}/m^{(2)} \bar{3}^6)^2^2/m^{(2,4)}$	2.mm (mm2)	$\bar{1}31$		411	
	66		$(4^4)^m^{(2,4)} \bar{3}^6^2^2/m^{(2,2)}$	m.m2 (mm2)	$\bar{1}31$		411	



Table 1 (continued)

No. $\mathcal{K}^{(2)}$	No. $\mathcal{K}^{(p>2)}$	No. $\mathcal{K}_{WB}^{(p>2)}$	$\mathcal{K}$	$\mathcal{H}^*$	1st twin element	2nd twin element	3rd twin element	4th twin element
		67	$(4^{(4,0)}/m^{(2,4)}\bar{3}^{(3)}2^{(2)}/m^{(2)})^{(12)}$	$2/m.. (2/m)$	131	411		
		68	$(4^{(4)}/m^{(2)}\bar{3}^{(3)}2^{(2,2)}/m^{(2,2)})^{(12)}$	$..2/m (2/m)$	131	411		
		69	$(4^{(4)}/m^{(2)}\bar{3}^{(6,0)}2^{(2)}/m^{(2)})^{(16)}$	$..3.$	$\bar{1}$	411	411	
80			$(4^{(4)}/m^{(2)}\bar{3}^{(3)}2^{(2)}/m^{(2)})^{(24)}$	$\bar{1}$	411	131		
		70	$(4^{(4,0)}/m^{(2)}\bar{3}^{(6)}2^{(2)}/m^{(2)})^{(24)}$	$2.. (2)$	$\bar{1}\bar{3}\bar{1}$	411		
		71	$(4^{(4)}/m^{(2)}\bar{3}^{(6)}2^{(2,4)}/m^{(2)})^{(24)}$	$..2 (2)$	$\bar{1}\bar{3}\bar{1}$	411		
		72	$(4^{(4)}/m^{(2,8)}\bar{3}^{(6)}2^{(2)}/m^{(2)})^{(24)}$	$m.. (m)$	$\bar{1}\bar{3}\bar{1}$	411		
		73	$(4^{(4)}/m^{(2)}\bar{3}^{(6)}2^{(2)}/m^{(2,4)})^{(24)}$	$..m (m)$	$\bar{1}\bar{3}\bar{1}$	411		
81			$(4^{(4)}/m^{(2)}\bar{3}^{(6)}2^{(2)}/m^{(2)})^{(48)}$	1	$2_\alpha$	$2_\beta$	$\bar{1}\bar{3}\bar{1}$	112

twin point groups (Table 1). With respect to the previous list we have introduced the following differences:

(i) In the previous list, the dichromatic groups were ordered according to the achromatic group, in contrast to the case of polychromatic groups; in the new list, all the entries are ordered according to the chromatic group, which makes the analysis easier and more straightforward.

(ii) A typo has been corrected: the dichromatic extension of  $\bar{4}$  is  $4'/m'$  and not  $4/m'$ , which corresponds instead to the dichromatic extension of  $4$ .

(iii) The classification is more consistently done in terms of the minimal set of twin *elements*, which in some cases leads to a reduction in the twin degree; for example, the twin point group  $(4^{(4)}/m)^{(4)}$ , obtained as an extension of  $\mathcal{H}^* = m11$ , was listed as a second-degree twin obtained by twinning according to 211 followed by twinning according to 411. The two twin operations share the same geometric element and the intermediate case corresponds to  $2'/m$ , which is a dichromatic extension corresponding to a different twin point group; we do not need to consider it explicitly when it occurs as part of a tetrachromatic twin.

We proceed then to extend the chromatic analysis of the case of allotwins.

Allotwins are oriented crystal associations of two or more different polytypes of the same compound. Polytypes are built up by stacking layers of (nearly) identical structure and composition, in which the two-dimensional translations in the  $(hkl)$  plane of the layer are (essentially) preserved whereas the period along the stacking direction depends on the stacking sequence (Guinier *et al.*, 1984). The allotwin lattice, *i.e.* the sublattice common to the allotwinned crystals, is based on the common two-dimensional lattice in the plane of the layer and an out-of-plane translation which is a simple multiple of the out-of-plane translation of the allotwinned polytypes. In the case of individuals with the same crystal structure, operations belonging to  $\mathcal{H}^*$  would lead to a parallel growth (for which  $\mathcal{H}^* = \mathcal{H}$ ), not to a twin; in the case of individuals with different crystal structure, operations belonging to  $\mathcal{H}^*$  are among those that can produce epitaxies, if a common two-dimensional sublattice with a small supercell exists in the composition plane (Royer, 1928, 1954). Allotwins are an intermediate case between twins and epitaxies: because they are built by stacking the same layer, the individuals have a common two-dimensional lattice in the plane of the layer; but

because the stacking sequence is different, they have a different crystal structure. As a consequence, operations belonging to  $\mathcal{H}^*$  are always among the possible allotwin operations; as we are going to show by analysing some concrete examples taken from the literature, allotwins built by operations belonging to  $\mathcal{H}^*$  are actually the most frequent cases. In the analysis of these examples, we have retained the Miller indices used in the original publications, although some of them are incorrectly reduced to relative prime integers despite the use of centred unit cells (Nespolo, 2015b).

The operations mapping individuals in incomplete twins and in some allotwins do not obey the closure property of groups; to give a full account of them we need to use a generalization of the concept of group, namely a groupoid,<sup>3</sup> which in our case will be a chromatic point groupoid. The usefulness of a groupoid analysis however goes well beyond the case of incomplete twins, as we are going to show. Before developing our treatment, we need to remind the reader of the symbols of chromatic point groups.

### 3. Symbols for chromatic point groups

The direct product  $(\mathcal{H}, 1^{(p)})$  of an achromatic point group  $\mathcal{H}$  with the colour identification group  $1^{(p)}$ , *i.e.* the group that permutes all  $p$  colours, is a neutral chromatic group. The colour identification group is a cyclic group  $\{1^{(p)}\} = \{1^{(p)1}, 1^{(p)2}, \dots, 1^{(p)p} = 1\}$  where the number outside the parentheses  $^{(p)}$  indicates the number of times the operation  $1^{(p)}$  is applied; the generator of the group is obviously  $1^{(p)1}$ . The number of possible orders of the three-dimensional crystallographic point groups is 10 (1, 2, 3, 4, 6, 8, 12, 16, 24, 48), which is also the number of possible values for the chromatic index  $p$ . In neutral crystallographic point groups, every operation is associated with the exchange of all the  $p$  colours (Shubnikov & Koptsik, 1974). In particular:

(i)  $(\mathcal{H}, \{1^{(1)}\})$  are the 32 monochromatic groups of order  $|\mathcal{H}|$  in which each operation attributes one and the same colour to each individual on which it acts; alternatively, the operations can attribute no colour at all so that the 32 groups  $(\mathcal{H}, \{1^{(1)}\})$  can also be called achromatic.

<sup>3</sup> We use the term 'groupoid' in the sense of Brandt (1927), *i.e.* as a small connected category in which every morphism is invertible; the reader should be aware that some authors use the term 'groupoid' in a different sense, to indicate a magma, *i.e.* a set equipped with a single binary operation.

(ii)  $(\mathcal{H}, \{1^{(2)}\})$  are the 32 grey groups of order  $2|\mathcal{H}|$ , in which each operation attributes two colours (usually ‘black’ and ‘white’, and thus ‘grey’ for their coexistence) to each individual on which it acts.

(iii)  $(\mathcal{H}, \{1^{(p>2)}\})$  are the  $32 \times 8 = 256$  neutral polychromatic groups of order  $p|\mathcal{H}|$ , in which each operation attributes  $p$  colours to each individual on which it acts; 8 is the number of chromaticities  $p > 2$ :  $p = 3, 4, 6, 8, 12, 16, 24, 48$ .

True (non-neutral) polychromatic point groups  $\mathcal{K}^{(p)}$  isomorphic to the crystallographic monochromatic point groups  $\mathcal{H}$  are obtained as extensions of the subgroups of  $\mathcal{H}$ . Invariant extension gives 51 dichromatic point groups (‘black and white’ groups, Shubnikov groups,  $\mathcal{K}^{(2)}$ ) and 81 polychromatic point groups (Koptisk groups,  $\mathcal{K}^{(p>2)}$ ). The other 73 true polychromatic point groups are obtained as non-invariant extensions and are termed Van der Waerden & Burckhardt (1961) groups,  $\mathcal{K}_{\text{WB}}^{(p>2)}$ ; the Wittke & Garrido (1959) groups,  $\mathcal{K}_{\text{WG}}^{(p>2)}$ , are isomorphic to them.  $\mathcal{K}^{(p)}$  and  $\mathcal{K}_{\text{WB}}^{(p)}$  differ in the fact that all the elements in  $\mathcal{K}^{(p)}$  are either achromatic or totally chromatic, whereas in  $\mathcal{K}_{\text{WB}}^{(p>2)}$  some of the elements are partially chromatic.

Shubnikov groups  $\mathcal{K}^{(2)}$  are minimal supergroups whose order is twice the order of the monochromatic groups  $\mathcal{H}$ . Only binary (first-degree twofold) twins can be described by  $\mathcal{K}^{(2)}$  groups; the twin operations are marked with a prime (′) instead of a superscript  $^{(2)}$ .

Koptisk groups  $\mathcal{K}^{(p>2)}$  are indicated by a full Hermann–Mauguin symbol in which each chromatic element (twin element) brings a chromatic index  $p_i$  ( $p_i = 2$  in Shubnikov groups is replaced by the prime); if more than one chromatic element occurs, the whole symbol is included in parentheses and a chromatic index  $p$  for the whole group is given, which corresponds to the index of  $\mathcal{H}$  in  $\mathcal{K}^{(p)}$ .

Van der Waerden–Burckhardt groups  $\mathcal{K}_{\text{WB}}^{(p)}$  are non-invariant extensions of  $\mathcal{H}$  which contain partially chromatic operations  $n^{(p_1, p_2)}$ ;  $p_1$  is the number of colours exchanged and  $p_2$  the number of colours fixed by the operation. Symbols of partially chromatic operations are of two types:

(i)  $p_2 \neq 0, p_1 + p_2 \leq p$ : the operation fixes  $p_2$  individuals and exchanges the other  $p - p_2$  individuals in sets of  $p_1$  individuals.

(ii)  $p_2 = 0, 3 < p_1 < p$ : they occur in cubic twin point groups, where symmetry operations and twin operations share the same geometric element.  $p_1$  coincides with the order of the twin operation and is twice the order of the symmetry operation.

As an example of the first case, we can consider  $\mathcal{K}_{\text{WB}}^{(4)} = (4^{(4)}2^{(2,2)}2^{(2)})^{(4)}$ , which corresponds to Fig. 1 already discussed. The second case corresponds to a fourfold twin (proper or improper) rotation about a twofold symmetry axis, symbol  $4^{(4,0)}$ , or a threefold twin rotoinversion about a threefold symmetry axis, symbol  $\bar{3}^{(6,0)}$ . Odd powers of the twin operation exchange all the  $p$  individuals, leaving fixed  $p_2 = 0$  individuals; even powers of the twin operation correspond to symmetry operations for the individual and thus fix it: they however exchange  $m \cdot p_1$  individuals but leave fixed the other  $p - m \cdot p_1$  individuals, where  $m = \lfloor p/p_1 \rfloor$  is the floor of  $p/p_1$  (the ‘floor’ is the ‘rounding down to integer’ function).

For example, the group  $(4^{(4,0)}3^{(3)}2^{(2,2)})^{(6)}$  is obtained by extending the orthorhombic group 2.22 (oriented symbol indicating symmetry elements along  $[100]$ ,  $[110]$  and  $[\bar{1}\bar{1}0]$  directions of a cubic reference) through threefold chromatic rotations about the cubic  $\langle 111 \rangle$  directions. The twofold rotations are symmetry operations of the individual and are mapped onto the corresponding elements of the other individuals by the threefold twin rotation. As a consequence, the fourfold rotation about the cubic basis vectors, which is not a symmetry operations for any of the individuals, exchanges four individuals and fixes none; a second application of the same operation gives a twofold rotation, which is a symmetry operation for two of the individuals, those with basis vectors parallel to each other, but exchanges the other four individuals. Accordingly, the fourfold rotation exchanges  $m \cdot p_1 = 1 \cdot 4 = 4$  individuals and fixes  $p - m \cdot p_1 = 6 - 4 = 2$  individuals. Similarly, the twofold rotations about the cubic  $\langle 110 \rangle$  directions are symmetry operations for two of the six individuals; they fix two individuals and exchange the other four in pairs.

In the same way, the group  $(2^{(2)}/m^{(2)}\bar{3}^{(6,0)})^{(8)}$  is obtained as an extension of 3 by inversion and twofold rotation about the cubic  $\langle 100 \rangle$  directions. The twin is composed of eight individuals. Odd powers of the  $\bar{3}$  operation exchange all the eight crystals; even powers correspond to the threefold rotation, which are symmetry operations for the individual: they fix  $8 - 6 = 2$  individuals, and exchange the other 6 individuals. The twofold rotations and the mirror reflections are totally chromatic.

A revised list of chromatic point groups is presented in Table 1.

#### 4. Groupoid structure of the mappings of twinned individuals

Let us consider a twin built by  $p$  individuals and let  $\mathcal{H}_i$  be the point group of the  $i$ th individual, with order  $|\mathcal{H}_i|$ ,  $h(m_i)$  the  $m_i$ th operation in  $\mathcal{H}_i$ ,  $m_i = [1, |\mathcal{H}_i|]$ . Let us indicate by  $\Phi(i \rightarrow 1)$  a twin operation mapping the orientation of the  $i$ th individual to the orientation of the first individual, taken as reference. The point groups  $\mathcal{H}_i$  and  $\mathcal{H}_1$  are related as follows ( $X_i = i$ th individual):

$$\begin{aligned} \Phi(i \rightarrow 1)X_i &= X_1; \Phi(1 \rightarrow i)X_1 = X_i; \Phi(1 \rightarrow i) = \Phi(i \rightarrow 1)^{-1} \\ \downarrow h(m_1)X_1 &= X_1; h(m_i)X_i = X_i \\ \Phi(i \rightarrow 1)X_i &= \Phi(i \rightarrow 1)[h(m_i)X_i] = X_1 = h(m_1)X_1 \\ &= h(m_1)\Phi(i \rightarrow 1)X_i \\ \Phi(i \rightarrow 1)h(m_i) &= h_1(m_1)\Phi(i \rightarrow 1) \\ h(m_i) &= \Phi(i \rightarrow 1)^{-1}h(m_1)\Phi(i \rightarrow 1). \end{aligned}$$

This relation is a similarity transformation known as *conjugation*. It holds for any  $h(m_i)$ , and thus also for the whole sets of operations, i.e. the point groups  $\mathcal{H}_1$  and  $\mathcal{H}_i$ :

$$\mathcal{H}_i = \Phi(i \rightarrow 1)^{-1}\mathcal{H}_1\Phi(i \rightarrow 1) = \Phi(1 \rightarrow i)\mathcal{H}_1\Phi(i \rightarrow 1). \quad (2)$$

The point groups of the individuals are therefore conjugate under the twin operation: they are of the same type (same

**Table 2**

The point groupoid built by the whole set of mappings relating any pair of individuals  $i \rightarrow j$ .

$\mathcal{H}_i$  ( $= \mathcal{C}_{i \rightarrow i}$ ) is the point group of the  $i$ th individual and the nucleus of the  $i$ th mixed group;  $\mathcal{C}_{i \rightarrow j}$  is the complex containing the operations mapping the orientation of the  $i$ th individual onto the orientation of the  $j$ th individual.

$\mathcal{C}_{1 \rightarrow 1} = \mathcal{H}_1$	$\mathcal{H}_1 \Phi(2 \rightarrow 1) = \mathcal{C}_{2 \rightarrow 1}$	...	$\mathcal{H}_1 \Phi(n \rightarrow 1) = \mathcal{C}_{n \rightarrow 1}$
$\Phi(1 \rightarrow 2) \mathcal{H}_1 = \mathcal{C}_{1 \rightarrow 2}$	$\Phi(1 \rightarrow 2) \mathcal{H}_1 \Phi(2 \rightarrow 1) = \mathcal{C}_{2 \rightarrow 2} = \mathcal{H}_2$	...	$\Phi(1 \rightarrow 2) \mathcal{H}_1 \Phi(n \rightarrow 1) = \mathcal{C}_{n \rightarrow 2}$
...	...	...	...
$\Phi(1 \rightarrow n) \mathcal{H}_1 = \mathcal{C}_{1 \rightarrow n}$	$\Phi(1 \rightarrow n) \mathcal{H}_1 \Phi(2 \rightarrow 1) = \mathcal{C}_{2 \rightarrow n}$	...	$\Phi(1 \rightarrow n) \mathcal{H}_1 \Phi(n \rightarrow 1) = \mathcal{C}_{n \rightarrow n} = \mathcal{H}_n$

Hermann–Mauguin symbol), but differently oriented in space; in particular, they have the same order:  $|\mathcal{H}|$ . The intersection group  $\mathcal{H}^*$  is a proper or trivial subgroup of  $\mathcal{H}_i$ :  $\mathcal{H}^* = \cap_i \mathcal{H}_i \subseteq \mathcal{H}_i$ . If  $\mathcal{H}^* = \mathcal{H}_i$ , then  $\mathcal{H}_i = \mathcal{H}$  for all individuals, which means that  $\mathcal{H}$  is self-conjugate by the twin operations and is therefore a normal (invariant) subgroup<sup>4</sup> of the chromatic group:  $\mathcal{H} \triangleleft \mathcal{K}^{(p)}$ .

The operations mapping the  $i$ th and the  $j$ th individuals are obtained as a combination of the operations mapping each of them to the reference individual (No. 1):

$$\Phi(i \rightarrow j) X_i = X_j;$$

$$\Phi(i \rightarrow j) = \Phi(1 \rightarrow j) \Phi(i \rightarrow 1) = \Phi(j \rightarrow 1)^{-1} \Phi(i \rightarrow 1) \quad (3)$$

which represents the mapping from  $i$  to 1 followed by the mapping from 1 to  $j$ , i.e. a combined mapping  $i$  to  $j$  via 1. For any pair of individuals  $i$  and  $j$  a set of equivalent mappings is obtained by combining  $\Phi(i \rightarrow j)$  with the symmetry operations of the target individual  $j$ , i.e. the operations of the point group  $\mathcal{H}_j$ ; the result is a set of  $|\mathcal{H}|$  operations  $\mathcal{C}_{i \rightarrow j} = \mathcal{H}_j \Phi(i \rightarrow j) = \cup_k \Phi_k(j \rightarrow j) \Phi(i \rightarrow j)$ ,  $k = [1, |\mathcal{H}|]$ . Indeed, the combination of a mapping  $i \rightarrow j$  with all the mappings  $j \rightarrow j$  gives the set of all the mappings  $i \rightarrow j$ ; in that case, it would not form a group but a complex (Ledermann, 1964),<sup>5</sup> unless  $i = j$ , which corresponds to the group  $\mathcal{H}_i$ . In the same way, the combination  $\Phi(i \rightarrow j)^{-1} \mathcal{H}_i = \Phi(j \rightarrow i) \mathcal{H}_i = \Phi(j \rightarrow i) \cup_k \Phi_k(j \rightarrow j)$ ,  $k = [1, |\mathcal{H}|]$ , represents the combination of all the mappings  $j \rightarrow j$ , i.e. the symmetry operations of the source individual, with a mapping  $j \rightarrow i$  and gives the set of all the mappings  $j \rightarrow i$ ; the result is the complex  $\mathcal{C}_{j \rightarrow i}$ . Finally, the set of all the complexes,  $\mathcal{D} = \{\mathcal{C}_{i \rightarrow j}\}$ ,  $\forall i, j = \{\Phi(k \rightarrow j) \Phi(k \rightarrow k) \Phi(i \rightarrow k)\}$ ,  $\forall i, j, k$  includes all the mappings relating the orientation of any pair of individuals, including the pairs  $k \rightarrow k$ . The structure of  $\mathcal{D}$  can be described in a matrix form with respect to the point groups  $\mathcal{H}_i$ ,  $i = [1, p]$ , of the  $p$  individuals (Table 2). Each row in Table 2 contains the whole set of mappings from any individual to a given, fixed individual. For example, the first row can be rewritten as  $\mathcal{M}_1 = \cup_i \mathcal{C}_{i \rightarrow 1} = \mathcal{C}_{1 \rightarrow 1} \cup_{i \neq 1} \mathcal{C}_{i \rightarrow 1} = \mathcal{H}_1 \cup_{i \neq 1} \mathcal{C}_{i \rightarrow 1}$ , i.e. the union of the point group of individual No. 1,  $\mathcal{H}_1$ , and of the whole set of operations mapping all the other individuals to No. 1.  $\mathcal{M}_1$  is known as *Mischgruppe* (Loewy, 1927), translated

<sup>4</sup> Rigorously speaking, because  $\mathcal{H}$  is achromatic whereas  $\mathcal{K}$  is chromatic, one can say that the former is a normal subgroup of the latter (or not) when considering the achromatic group  $\mathcal{K}_A$  obtained by removing every chromaticity from  $\mathcal{K}$ .

<sup>5</sup> When decomposing a group in terms of one of its subgroups, each coset but the subgroup is a complex.

in English as *mixed group* (Dixon, 1963), *hybrid group* (Sadanaga, unpublished manuscript) or *compound group* (Brown, 1987); hereafter we follow Dixon (1963) and use *mixed group* which is closer to the original German.  $\mathcal{C}_{1 \rightarrow 1}$  has been called *kern* (kernel or core) (Loewy, 1927), *kernel* (Sadanaga et al., 1980) and *nucleus* (Dixon, 1963). The term *kernel* may lead to confusion because it is also used in group theory to indicate the subset of elements of a

group that are homomorphically mapped to the unit element of a subgroup. Similarly, *core* may lead to confusion because it is also used in group theory to indicate a special normal subgroup. Hereafter we follow Dixon (1963) and use the term *nucleus*, which is the least ambiguous.<sup>6</sup> Finally,  $\cup_{i \neq 1} \mathcal{C}_{i \rightarrow 1}$  has been called *shale* (skin or shell) (Loewy, 1927), *hull* (Sadanaga et al., 1980) and *shell* (Sadanaga, unpublished manuscript). Hereafter we use *shell*, which is closer to the original German and, with its natural counterpart *nucleus*, is related to physics, whereas the pair kernel–hull is more related to botany. [In Japanese,  $\mathcal{C}_{1 \rightarrow 1}$  and  $\cup_{i \neq 1} \mathcal{C}_{i \rightarrow 1}$  are indicated as

核 and 莢,

respectively, with a clear analogy to the kernel and hull of a drupe (Sadanaga, 1980). In his unpublished manuscript, Sadanaga used shell instead of hull, which would correspond to

殻;

the pair kernel–shell therefore shifts the botanical analogy from drupe to nut.]

The structure of  $\mathcal{D}$  is precisely the structure of a Brandt groupoid (Brandt, 1927). Groupoids have been used in crystallography, especially in the framework of the OD (order–disorder) theory (Dornberger-Schiff, 1979), to describe the whole set of operations mapping modules (bricks, rods, layers) in a modular structure and in that context they are known as space groupoids; the diagonal elements in the matrix  $\mathcal{D}$  (Table 2) are local operations of the modules (nuclei of the mixed groups) and the extra-diagonal elements are partial operations mapping different modules (shells of the mixed groups). In our case, the symmetry operations do not contain any translational component, so that  $\mathcal{D}$  is a point groupoid. The application of groupoids to domain structures has been presented by Cayron (2006) in a more abstract approach. A very similar treatment was adopted by Roth (1982) in the study of colouring of geometric drawings: his ‘bisets’ are mixed groups and his ‘array of bisets’ is nothing other than a groupoid (p. 280), although Roth’s presentation is the transpose of ours because he applies the operations from the right instead of from the left. The stabilizer of each colour corresponds to the nucleus of each mixed group.

<sup>6</sup> The term *nucleus* is also used in ring theory to indicate the set of elements that associate with all others; the risk of confusion with its use in the present context is however minimal.

Table 3

Groupoid for example 1 case 1.

Here and in the following the horizontal line separates two expressions of the groupoid, the first with the elements indicated as  $\Phi(k \rightarrow i)\mathcal{H}^*\Phi(i \rightarrow j)$ , the second with the operations listed explicitly.

$\{1, 2_{[001]}, 2_{[100]}, 2_{[010]}\}$	$\{1, 2_{[001]}, 2_{[100]}, 2_{[010]}\}4^+_{[001]}$
$4^-_{[001]}\{1, 2_{[001]}, 2_{[100]}, 2_{[010]}\}$	$4^-_{[001]}\{1, 2_{[001]}, 2_{[100]}, 2_{[010]}\}4^+_{[001]}$
$\{1, 2_{[001]}, 2_{[100]}, 2_{[010]}\}$	$\{4^+_{[001]}, 4^-_{[001]}, 2_{[1\bar{1}0]}, 2_{[110]}\}$
$\{4^-_{[001]}, 4^+_{[001]}, 2_{[1\bar{1}0]}, 2_{[110]}\}$	$\{1, 2_{[001]}, 2_{[100]}, 2_{[010]}\}$

Our treatment addresses the derivation of the chromatic symmetry of both twins and allotwins and for this purpose we need to find the subset of operations of  $\mathcal{D}$  which build the chromatic group  $\mathcal{K}^{(p)}$ . This will be done in analogy with the extraction of space-group operations from a space groupoid (Nespolo & Aroyo, 2016a).

The operations  $\Phi(i \rightarrow j)$  in the groupoid  $\mathcal{D}$  are specific to the given pair of individuals  $i$  and  $j$  and do not act, in general, on other pairs. They are known as local ( $i = j$ ) or partial ( $i \neq j$ ) operations. A subset of these actually do act on the whole crystal space and are therefore total (or global) operations, building a group. The total operations are identified by checking which of the operations of the groupoid occur in each and every mixed group. In the case of a modular structure, the operations identified in this way build the space group of the structure. In the case of a twin, the analysis of the groupoid structure allows a straightforward derivation of the chromatic point group. In fact:

- (i) The operations occurring in each mixed group are total operations, either chromatic or achromatic.
- (ii) Among these, the operations occurring only in the nuclei are achromatic, those occurring only in the shells are totally chromatic, those occurring in the nucleus of some mixed groups and in a shell of the other mixed groups are partially chromatic.
- (iv) The operations occurring in some, but not all of the mixed group, are partial operations; their presence is indicative of an incomplete twin.

To illustrate the procedure we present a few examples.

#### 4.1. Example 1: complete versus incomplete twins

A point groupoid contains as many identity operations as the number of individuals (because they are contained in the nucleus of each mixed group) and may contain partial operations as well, in the case of incomplete twins. A twin point group, on the other hand, contains only one identity and does not contain partial operations; the incomplete nature of a twin is not reflected in the twin point group, so that two different twins, one complete and the other incomplete, may be described by the same type of twin point group. The groupoid instead shows the difference. We illustrate the procedure through three simple cases.

*Case 1.* A crystal with a point group of type 222 twinned by fourfold rotation about [001] gives a twin whose point group is  $\mathcal{K}^{(2)} = 4'22'$ . The point groups of the two individuals have their basis vectors parallel to each other, so that  $\mathcal{H}_1 = \mathcal{H}_2 = \mathcal{H}^* = 222$ . The groupoid is trivial (see Table 3).

The operations in the nuclei of the two mixed groups define the intersection group  $\mathcal{H}^* = 222$  whereas the operations in the two shells are totally chromatic.

*Case 2.* Let us then consider a crystal with point group of type 422 possessing a (pseudo)-cubic (sub)lattice, which forms a three-individual twin with twin operations a 90° rotation about the [100] direction and another 90° rotation about the [010] direction. The groupoid is obtained as shown in Table 4.

All the operations occur in all the three mixed groups and are therefore total operations. They are however of three different types:

- (i) The operations that occur in the three nuclei are achromatic; these are 1,  $2_{[100]}$ ,  $2_{[010]}$  and  $2_{[001]}$ , so that  $\mathcal{H}^* = 222$ .
- (ii) The operations that occur in a shell but not in a nucleus are totally chromatic; these are the threefold rotations about the four cubic directions  $\langle 111 \rangle$ .
- (iii) The operations that occur in the nucleus of one mixed group and in a shell of the two other mixed groups are partially chromatic; these are the three powers of the fourfold rotations about the directions of the basis vectors, as well as the twofold rotations about the six cubic directions  $\langle 110 \rangle$ ; they exchange two individuals and fix one.

Table 4

Groupoid for example 1 case 2.

$\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}$	$\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}4^+_{[100]}$	$\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}4^+_{[010]}$
$4^-_{[100]}\{1, 4^+_{[001]}, 2_{[001]}, 4^+_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}$	$4^-_{[100]}\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}4^+_{[100]}$	$4^-_{[100]}\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}4^+_{[010]}$
$4^-_{[010]}\{1, 4^+_{[001]}, 2_{[001]}, 4^+_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}$	$4^-_{[010]}\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}4^+_{[100]}$	$4^-_{[010]}\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}4^+_{[010]}$
$\{1, 4^+_{[001]}, 2_{[001]}, 4^-_{[001]}, 2_{[100]}, 2_{[010]}, 2_{[110]}, 2_{[1\bar{1}0]}\}$	$\{4^+_{[100]}, 3^+_{[111]}, 2_{[011]}, 3^+_{[1\bar{1}1]}, 4^-_{[100]}, 2_{[01\bar{1}]}, 3^+_{[1\bar{1}1]}, 3^+_{[1\bar{1}\bar{1}]} \}$	$\{4^+_{[010]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[0\bar{1}0]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[101]}, 4^-_{[010]}, 3^+_{[111]}, 3^+_{[1\bar{1}\bar{1}]} \}$
$\{4^-_{[100]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[011]}, 3^+_{[111]}, 4^+_{[100]}, 2_{[01\bar{1}]}, 3^+_{[1\bar{1}1]}, 3^+_{[1\bar{1}\bar{1}]} \}$	$\{1, 4^+_{[010]}, 2_{[010]}, 4^-_{[010]}, 2_{[100]}, 2_{[001]}, 2_{[10\bar{1}]}, 2_{[101]} \}$	$\{3^+_{[1\bar{1}\bar{1}]}, 2_{[1\bar{1}0]}, 3^+_{[1\bar{1}\bar{1}]}, 4^-_{[001]}, 3^+_{[111]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[110]}, 4^+_{[001]} \}$
$\{4^-_{[010]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[0\bar{1}0]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[101]}, 4^+_{[010]}, 3^+_{[111]}, 3^+_{[1\bar{1}\bar{1}]} \}$	$\{3^+_{[1\bar{1}\bar{1}]}, 4^+_{[001]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[1\bar{1}0]}, 3^+_{[111]}, 3^+_{[1\bar{1}\bar{1}]}, 2_{[110]}, 4^+_{[001]} \}$	$\{1, 4^-_{[100]}, 2_{[100]}, 4^+_{[100]}, 2_{[001]}, 2_{[010]}, 2_{[011]}, 2_{[01\bar{1}]} \}$

**Table 5**  
Groupoid for example 2.

$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}3^+_{[001]}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}3^-_{[001]}$
$3^-_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$3^-_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}3^+_{[001]}$	$3^-_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}3^-_{[001]}$
$3^+_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$3^+_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}3^+_{[001]}$	$3^+_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}3^-_{[001]}$
$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{3^+_{[001]}, 2_{[110]}, \bar{3}^+_{[001]}, m_{[110]}\}$	$\{3^-_{[001]}, 2_{[100]}, \bar{3}^-_{[001]}, m_{[100]}\}$
$\{3^-_{[001]}, 2_{[110]}, \bar{3}^-_{[001]}, m_{[110]}\}$	$\{1, 2_{[100]}, \bar{1}, m_{[100]}\}$	$\{3^+_{[001]}, 2_{[010]}, \bar{3}^+_{[001]}, m_{[010]}\}$
$\{3^+_{[001]}, 2_{[100]}, \bar{3}^+_{[001]}, m_{[100]}\}$	$\{3^-_{[001]}, 2_{[010]}, \bar{3}^-_{[001]}, m_{[010]}\}$	$\{1, 2_{[110]}, \bar{1}, m_{[110]}\}$

From this analysis, the trichromatic point group is obtained straightforwardly as  $\mathcal{K}^{(3)} = (4^{(2,1)}3^{(3)}2^{(2,1)})^{(3)}$ .

*Case 3.* Let us now suppose that the same crystal, with point group of type 422, forms a twin with only two individuals, for example by fourfold rotation about [100]. The groupoid consists of the top-left  $2 \times 2$  block of the last example. Quite obviously,  $\mathcal{H}^* = 222$  as before. The totally chromatic operations, occurring in the two shells, are  $4^+_{[100]}$ ,  $4^-_{[100]}$ ,  $2_{[011]}$  and  $2_{[01\bar{1}]}$ . All the other operations, namely the threefold rotations about the body diagonals, occur in the counterclockwise sense only in one shell and in the clockwise sense only in the other shell, and are therefore partial operations. The twin point group  $\mathcal{K}^{(p)}$  is obtained as an extension of  $\mathcal{H}^* = 222$  by a fourfold rotation about [100]. By re-labelling the basis vectors so that the fourfold twin rotation is now about [001],  $\mathcal{K}^{(2)} = 4'22'$ .

Both  $\mathcal{H}^*$  and  $\mathcal{K}^{(p)}$  are the same in case 1 and in case 3, but the former is a complete twin whereas the latter is an incomplete twin, characterized by the presence of partial operations. The groupoids of the two twins (which for case 1 degenerates into a group) clearly show the difference.

#### 4.2. Example 2: a non-invariant extension of a monoclinic holohedral point group

Let us suppose that a monoclinic crystal with point group of type  $12/m1$  possesses a (pseudo)-hexagonal (sub)lattice based on the (001) plane and a direction  $[u0v]$  (quasi)-perpendicular to (001); this (sub)lattice is the twin lattice. The crystal can therefore undergo twinning by a threefold or sixfold rotation about  $[u0v]$ ; the deviation from exact perpendicularity is measured by the obliquity, *i.e.* the angle  $\omega$  between  $[u0v]$  and  $[001]^*$ . For the derivation of the twin point group  $\mathcal{K}^{(p)}$ , we assume  $\omega = 0^\circ$ ; if that is not the case, then  $\mathcal{K}^{(p)}$  represents the *pseudo*-symmetry of the twin. For the sake of brevity, we assume that the twin operation is a threefold rotation; the case of a sixfold rotation can be worked out exactly in the same way, although the derivation is longer. The directions  $[uvw]$  are expressed with respect to the hexagonal basis of the twin lattice, where the directions [010] of the first individual and of the twin lattice are taken parallel (see Table 5).

The only operations  $\Phi(i \rightarrow i)$  that are common to  $i = 1, 2, 3$  are the identity and the inversion; accordingly,  $\mathcal{H}^* =$

$\mathcal{H}_1 \cap \mathcal{H}_2 \cap \mathcal{H}_3 = \bar{1}$ . The operations  $3^+_{[001]}$ ,  $3^-_{[001]}$ ,  $\bar{3}^+_{[001]}$ ,  $\bar{3}^-_{[001]}$  occur in an extra-diagonal block of each mixed group, namely  $\mathcal{C}_{2 \rightarrow 1}$  and  $\mathcal{C}_{1 \rightarrow 3}$  for the counterclockwise (proper or improper) rotation, and in  $\mathcal{C}_{3 \rightarrow 1}$  and  $\mathcal{C}_{1 \rightarrow 2}$  for the clockwise (proper or improper) rotation: these are therefore totally chromatic operations and the corresponding element is represented as  $\bar{3}^{(3)}$ . This symbol indicates that the threefold rotoinversion (operation of order 6) is trichromatic (exchanges three colours, *i.e.* three individuals); the inversion component (of order 2) is therefore

achromatic, as one should expect, knowing that  $\mathcal{H}^* = \bar{1}$ . All of the twofold rotations about an axis in the (001) plane and the reflections about the plane normal to them occur also in each mixed group, but once in the nucleus and twice in the shells:

- $2_{[010]}$  and  $m_{[010]}$  occur in  $\mathcal{C}_{1 \rightarrow 1} = \mathcal{H}_1$ , in  $\mathcal{C}_{2 \rightarrow 3}$  and in  $\mathcal{C}_{3 \rightarrow 2}$ ;
- $2_{[100]}$  and  $m_{[100]}$  occur in  $\mathcal{C}_{2 \rightarrow 2} = \mathcal{H}_2$ , in  $\mathcal{C}_{1 \rightarrow 3}$  and in  $\mathcal{C}_{3 \rightarrow 1}$ ;
- $2_{[110]}$  and  $m_{[110]}$  occur in  $\mathcal{C}_{3 \rightarrow 3} = \mathcal{H}_3$ , in  $\mathcal{C}_{1 \rightarrow 2}$  and in  $\mathcal{C}_{2 \rightarrow 1}$ .

These are therefore partially chromatic operations, fixing one colour (individual) and exchanging the two others. The corresponding elements are represented as  $2^{(2,1)}$  and  $m^{(2,1)}$ . The chromatic point group is therefore  $\mathcal{K}^{(3)} = (\bar{3}^{(3)}2^{(2,1)}/m^{(2,1)})^{(3)}$ .

#### 5. Extension to the treatment of allotwins

In the analysis of twins we have seen that the mappings  $\Phi(i \rightarrow i)$  are symmetry operations for each individual and are achromatic (colour-preserving), whereas the mappings  $\Phi(i \rightarrow j)$  relate different individuals and are chromatic (colour-exchanging). In the matrix representing the groupoid  $\mathcal{D}$ , the diagonal blocks contain achromatic operations, whereas the extra-diagonal blocks contain chromatic operations. For a  $p$ -individual twin, the matrix contains  $p^2$  blocks and, if we consider only its chromaticity (colour-exchanging) properties, it is symmetric: in fact, by assigning two different colours to the individuals  $i$  and  $j$ , the chromaticity of the mapping  $\Phi(j \rightarrow i)$  is the same as the chromaticity of the mapping  $\Phi(i \rightarrow j)$ . Therefore, the number of columns, or the number of rows, coincides with the number of individuals (colours).  $\Phi(i \rightarrow j)$  is always different from any  $\Phi(i \rightarrow i)$ , because the orientation of each individual in a twin is different from that of any other individual. This is no longer true in the case of allotwins. In particular, whereas the relative orientation of individuals in twins is uniquely defined by equation (1), in the case of allotwins only two of the three basis vectors – those in the  $(hkl)$  plane of the layer – can be uniquely related in different polytypes: the ‘relative orientation’ of different polytypes in an allotwin needs to be defined in an unambiguous way.

Different polytypes of the same compound are built by stacking the same layer in various orientations/positions. The resulting structure is different but the two basis vectors in the  $(hkl)$  plane of the layer have identical length and interaxial

angle. If the allotwin A is composed of a number of polytypes  $P_1, P_2, \dots, P_n$ , then the allotwin lattice is the sublattice common to all the  $P_j$  and one can find the basis vector transformation from each of the  $P_j$  to A; these transformations also provide the relative orientation of the different  $P_j$ . The matrix  $\mathbf{M}_i \mathbf{M}_j^{-1}$  in equation (1) gives the transformation of the basis vectors and, for allotwins, does not, in general, represent an isometry, because the unit cell of the two polytypes does not have, in general, the same volume. The difference in volume comes from the different number of layers, reflected in the different unit-cell parameter along the stacking direction. If we consider a block-diagonal matrix built on the  $2 \times 2$  block of the  $\mathbf{M}_i \mathbf{M}_j^{-1}$  matrix that represents the transformation of the

basis vectors in the  $(hkl)$  plane of the layer, we obtain an isometry relating the orientation of the two polytypes in the allotwin. The classification of allotwins is based on whether or not this mapping belongs to  $\mathcal{H}_{ij}^* = \mathcal{H}_i \cap \mathcal{H}_j$ . In the positive case, we speak of ‘parallel orientation of polytypes’; otherwise, of ‘different orientation’. If the conventional cell of a polytype has basis vectors in the  $(hkl)$  plane of the layer that are no longer equivalent to those of the layer itself, a non-conventional unit cell may be useful to find the relative orientation. For example, mica polytypes are obtained by stacking a layer parallel to the (001) plane along an out-of-plane direction with a rotation of  $n \times 60^\circ$  ( $n$  integer). In the 3T polytype each layer is rotated  $+120^\circ$  or  $-120^\circ$  with respect to

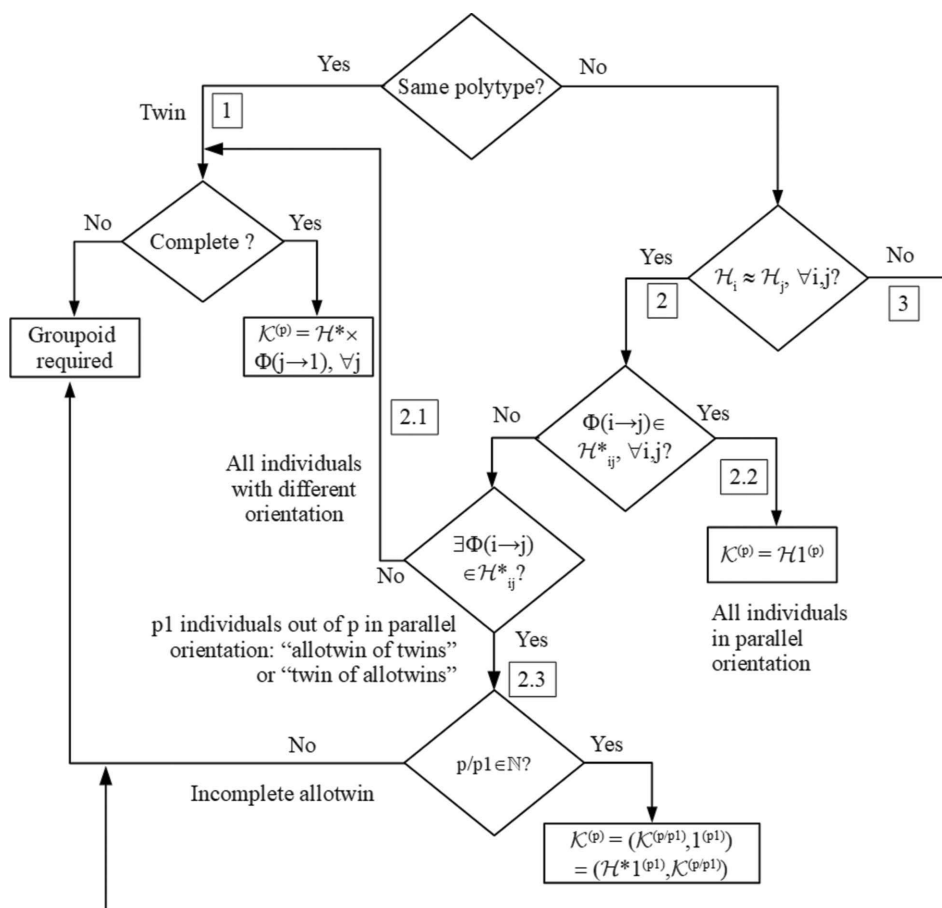


Figure 2

Flowchart of the derivation of the chromatic point group for twins and allotwins.  $\forall$  means ‘any’,  $\exists$  means ‘exists’,  $\in$  means ‘belongs to’,  $\cong$  means ‘of the same type (conjugate)’. Numbers in boxes correspond to the hierarchy of cases described in Section 5. (1) is the branch corresponding to a twin of identical polytypes: the chromatic point group is the extension of the intersection point groups of the individuals by the operation(s) mapping the individuals. If the twin is incomplete, for its faithful description the full groupoid is required. (2.1) is the branch corresponding to an allotwin of different polytypes with the same type of point group but all with different orientation. The resulting point group is obtained as in the case of a twin. If the allotwin is incomplete, for its faithful description the full groupoid is required. (2.2) is the branch corresponding to an allotwin of different polytypes with the same oriented point group  $\mathcal{H}$  (all individuals occur in parallel orientation): the operation(s) mapping the individuals belong(s) to the point group and the allotwin is described by a neutral point group obtained by extending  $\mathcal{H}$  by the colour identification group  $1^{(p)}$  (the result is a grey group in the case of two individuals). (2.3) is the branch corresponding to an allotwin of individuals in which the operation mapping the individuals does not belong to the point group of one or more individuals, *i.e.* in which some but not all the individuals occur in parallel orientation. This case can be described as an ‘allotwin of twins’ or a ‘twin of allotwins’ and requires extended chromatic symbols constructed in a similar way as the extended Hermann–Mauguin symbols of the space group. The chromatic group  $\mathcal{K}^{(p)}$  is obtained as an extension of the chromatic group describing the twinned regions by the colour identification group describing regions in which polytypes are in parallel orientation. If the allotwin is incomplete, for its faithful description the full groupoid is required. (3) is the branch corresponding to an allotwin of individuals with different types of point group: this case cannot be described by a chromatic point group but requires explicitly the (Ehresmann) groupoid, although a chromatic symbol can, at least in some cases, be used to represent the groupoid, as shown in Section 5.4.

the previous one, in a spiral sequence; the space-group type is  $P3_112$  or  $P3_212$ , depending on the sign of the rotation angle. The conventional basis is hexagonal (Nespolo & Kogure, 1998). One can, however, always use a orthohexagonal basis in which the **a** and **b** vectors are the same in length as those of the single layer, although this would lead to a non-standard setting of the space group, with symbol  $C123_1$  or  $C123_2$  (Nespolo & Aroyo, 2016b).

An oriented association of polytypes can include the following cases (see the flowchart in Fig. 2):

(1) Individuals corresponding to the same polytype building up a twin; the individuals are all in different orientations [ $\Phi(i \rightarrow j) \notin \mathcal{H}_{ij}^*$  for all the pairs  $i, j$ ], otherwise the oriented crystal association would be a parallel growth.

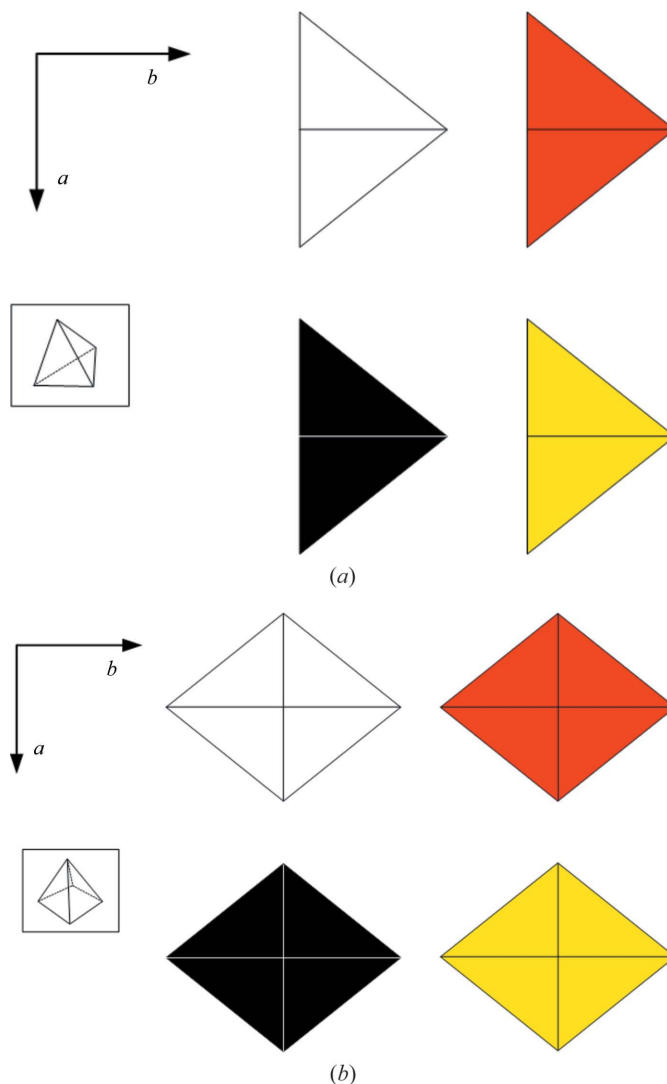
(2) Individuals corresponding to different polytypes but all with the same type of point group  $\mathcal{H}$ :

(2.1) All in different orientations [ $\Phi(i \rightarrow j) \notin \mathcal{H}_{ij}^*$  for all the pairs  $i, j$ ]. This case can be dealt with in a similar way as done for twinning: the allotwin can be described by a chromatic point group  $\mathcal{K}^{(p)}$  obtained as an extension of  $\mathcal{H}^*$  through the  $p - 1$  operations  $\Phi(1 \rightarrow j)$ ,  $j = [2, p]$ .

(2.2) All in parallel orientations [ $\Phi(i \rightarrow j) \in \mathcal{H}_{ij}^*$  for all the pairs  $i, j$ ]. In this case  $\mathcal{H}^* = \mathcal{H} = \mathcal{H}_i, \forall i$ . In fact,  $\Phi(i \rightarrow j) \in \mathcal{H}_{ij}^*$  implies  $\Phi(i \rightarrow j) \in \mathcal{H}_i$  and  $\Phi(i \rightarrow j) \in \mathcal{H}_j$  and the same is true for the inverse operations. Then [equation (2)]  $\Phi(j \rightarrow i)\mathcal{H}_i\Phi(i \rightarrow j) = \Phi(i \rightarrow j)^{-1}[\mathcal{H}_i\Phi(i \rightarrow j)] = \Phi(i \rightarrow j)^{-1}\mathcal{H}_i = \mathcal{H}_i$ . The chromatic point group obtained as an extension of  $\mathcal{H}^* = \mathcal{H}$  is neutral,  $\mathcal{H}1^{(p)}$ , in particular a grey group  $\mathcal{H}1'$  in the case of two individuals (Figs. 3a, 3b).

(2.3)  $\Phi(i \rightarrow j) \in \mathcal{H}_{ij}^*$  for  $p1$  individuals out of  $p$ . This case can be described as an ‘allotwin of twins’ or as a ‘twin of allotwins’ because the allotwinning of the individuals for which  $\Phi(i \rightarrow j) \notin \mathcal{H}_{ij}^*$  can be treated as the case of twinning [point (2.1) above]. If the ratio  $p/p1$  is not integer, the allotwin is incomplete and the groupoid has to be given explicitly for a faithful description of the allotwin. If instead the allotwin is complete, the chromatic group  $\mathcal{K}^{(p)}$  is the supergroup obtained by extending the chromatic group  $\mathcal{K}^{(p/p1)}$ , which describes the regions where polytypes have different orientations, by the colour identification group  $1^{(p1)}$ , which describes the regions in which polytypes have the same orientation. The same result is obtained by extending the neutral group  $\mathcal{H}^*1^{(p1)}$ , which describes the regions in which polytypes have the same orientation, by the chromatic group  $\mathcal{K}^{(p/p1)}$ , which describes the regions where polytypes have different orientations. If all the polytypes have their symmetry elements parallel to each other, then  $\mathcal{H}^* = \mathcal{H}$ :  $\mathcal{H}$  is a normal subgroup of  $\mathcal{K}^{(p)}$  ( $\mathcal{H} \triangleleft \mathcal{K}^{(p)}$ ) and the neutral group is  $\mathcal{H}1^{(p)}$  [Figs. 4(a), 4(b) and 5; see the detailed analysis in Section 5.3].

(3) Individuals corresponding to different polytypes and with different types of point group. This case is more complex and has to be treated in terms of groupoids according to Ehresmann (Taylor, 1977), which are more general than Brandt groupoids. The latter are *connected*, which means that the nuclei of the mixed groups are all of the same type, whereas this restriction does not apply to an Ehresmann groupoid. Mixed groups with nuclei of different types build



**Figure 3**

(a) Schematic representation of a four-individual allotwin in which all the individuals have the same type of point group,  $\mathcal{H}_i = m11$ , represented by pyramids with isosceles triangles as basis (in the projection, the vertex of a pyramid is above the middle point of the vertical line representing the unequal edge of the triangular basis; the inset shows one pyramid in elevation). All the individuals are in parallel orientation (path 2.2 of the flowchart in Fig. 2). The allotwin is described by the neutral group  $m11,1^{(4)}$ . (b) The same scheme, this time with individuals having point group  $\mathcal{H}_i = mm2$ , represented by rhombic pyramids. The allotwin is described by the neutral group  $mm2,1^{(4)}$ .

therefore an Ehresmann groupoid. Accordingly, the matrix of the groupoid presents a block-diagonal structure, where each block includes mixed groups whose nuclei correspond to a type of point group. The nuclei in mixed groups of different blocks are no longer conjugate groups but different groups, and the intersection group  $\mathcal{H}^*$  contains only the operations that are common to these groups. The remaining operations may occur only in the corresponding nucleus, in which case they are local operations of that individual. If they also occur in the shells, they are partially chromatic operations; and if they occur in some but not all of the shells, they are *partially chromatic partial operations*, meaning that the allotwin is incomplete.

A chromatic point group does not bring much information if the (allo)twin is incomplete or if the individuals have point groups of a different type. An informative description requires the point groupoid (Brandt or Ehresmann, depending on

whether the polytypes have the same type of point group or not). In some cases this point groupoid can be synthetically represented by a chromatic symbol built in the same way as the chromatic symbol of a twin point group, as we are going to show with some examples of allotwins reported in the literature.

5.1. Allotwins with point groups of the same type and individuals all in different orientation:  $\Phi(i \rightarrow j) \notin \mathcal{H}_{ij}^* \forall i, j$

This case corresponds to path 2.1 of the flowchart in Fig. 2. The only examples we are aware of are the two  $1M-2M_1$  allotwins in micas we have reported (Nespolo *et al.*, 2000a), in which the two individuals were rotated by  $120^\circ$  and by  $60^\circ$ , respectively, about the normal to the (001) plane. The two-dimensional lattice in the (001) plane of the layer is pseudo-hexagonal, whereas the layer itself is only monoclinic. This explains not only the large number of polytypes found in micas, but also the occurrence of allotwins in which the operations mapping the individuals do not belong to the point groups of the individuals.

The two allotwins reported in the literature are represented by the symbols  $Z_T = {}^3_5$  and  $Z_T = {}^3_4$ , respectively, where the upper and lower digits give the orientation of the first and second individuals with respect to a fixed reference, in which the six possible orientations are numbered from 1 to 5 and the first one is taken with orientation 3 (Nespolo *et al.*, 2000b). The two polytypes  $1M$  and  $2M_1$  crystallize in space-group types  $C2/m$  and  $C2/c$ , respectively, so that they have the same type of point group,  $2/m$ . The axial ratio  $b/a$  closely approaches the orthohexagonal ratio  $3^{1/2}$  and the projection of the  $c$  axis on the (001) plane, *i.e.*  $c \cos \beta$ , approaches  $-a/3$ . For each polytype, a three-dimensional pseudo-hexagonal sublattice can therefore be obtained by the basis vector transformation  $\mathbf{a}_T = \mathbf{a}_1$ ,  $\mathbf{b}_T = (-\mathbf{a}_1 + \mathbf{b}_1)/2$ ,  $\mathbf{c}_T = \mathbf{a}_1 + 3\mathbf{c}_1$  ( $T = \text{twin}$ ,  $I = \text{individual}$ ). Moreover, the axial setting of the  $1M$  polytype can be transformed to the axial setting of the  $2M_1$  polytype by the transformation  $\mathbf{a}_{2M1} = -\mathbf{a}_{1M}$ ,  $\mathbf{b}_{2M1} = \mathbf{b}_{1M}$ ,  $\mathbf{c}_{2M1} = \mathbf{a}_{1M} + 2\mathbf{c}_{1M}$  (Nespolo *et al.*, 1998), so that a common hexagonal sublattice (the allotwin sublattice) is obtained by the

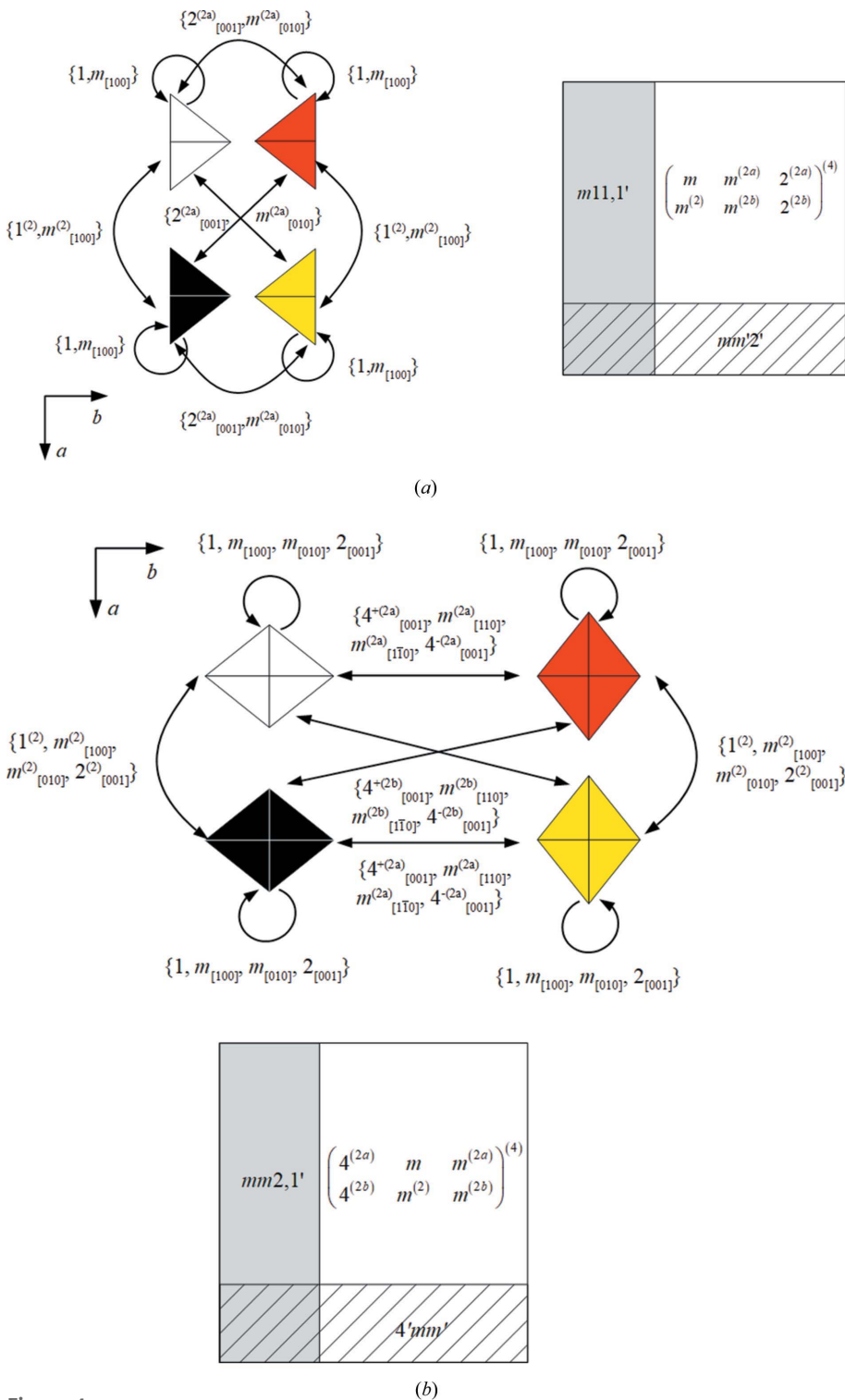


Figure 4 (a) Schematic representation of a four-individual allotwin in which all the individuals have the same type of point group,  $\mathcal{H}_i = m11$ , in two different orientations (path 2.3 of the flowchart in Fig. 2). Left: orientation of the four individuals; right: construction of the tetrahromatic point group. (b) The same scheme, this time with individuals having point group  $\mathcal{H}_i = mm2$ .

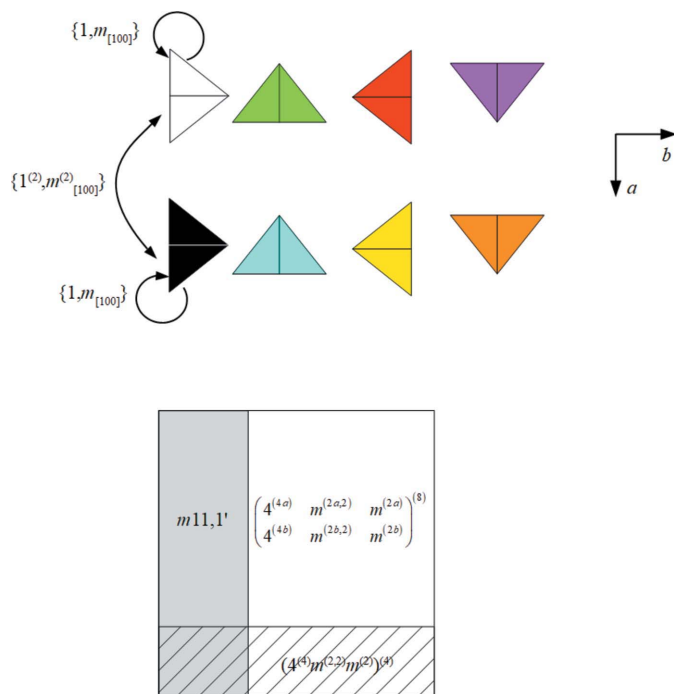


**Table 6**  
Groupoid for Section 5.1.

$Z_T = {}^3_5$		$Z_T = {}^3_4$	
$\{1, 2_{[120]}, \bar{1}, m_{[120]}\}$	$\{1, 2_{[120]}, \bar{1}, m_{[120]}\}3^+_{[001]}$	$\{1, 2_{[120]}, \bar{1}, m_{[120]}\}$	$\{1, 2_{[120]}, \bar{1}, m_{[120]}\}6^+_{[001]}$
$3^-_{[001]}\{1, 2_{[120]}, \bar{1}, m_{[120]}\}$	$3^-_{[001]}\{1, 2_{[120]}, \bar{1}, m_{[120]}\}3^+_{[001]}$	$6^-_{[001]}\{1, 2_{[120]}, \bar{1}, m_{[120]}\}$	$6^-_{[001]}\{1, 2_{[120]}, \bar{1}, m_{[120]}\}6^+_{[001]}$
$Z_T = {}^3_5$		$Z_T = {}^3_4$	
$\{1, 2_{[120]}, \bar{1}, m_{[120]}\}$	$\{3^+_{[001]}, 2_{[210]}, \bar{3}^+_{[001]}, m_{[210]}\}$	$\{1, 2_{[120]}, \bar{1}, m_{[120]}\}$	$\{6^+_{[001]}, 2_{[110]}, \bar{6}^+_{[001]}, m_{[110]}\}$
$\{3^-_{[001]}, 2_{[210]}, \bar{3}^-_{[001]}, m_{[210]}\}$	$\{1, 2_{[1\bar{1}0]}, \bar{1}, m_{[1\bar{1}0]}\}$	$\{6^-_{[001]}, 2_{[110]}, \bar{6}^-_{[001]}, m_{[110]}\}$	$\{1, 2_{[210]}, \bar{1}, m_{[210]}\}$

following transformation:  $\mathbf{a}_T = \mathbf{a}_{2M1} = -\mathbf{a}_{1M}$   $\mathbf{b}_T = (-\mathbf{a}_{2M1} + \mathbf{b}_{2M1})/2 = (\mathbf{a}_{1M} + \mathbf{b}_{1M})/2$ ,  $\mathbf{c}_T = \mathbf{a}_{2M1} + 3\mathbf{c}_{2M1} = 2\mathbf{a}_{1M} + 6\mathbf{c}_{1M}$ . In the setting of the allotwin lattice, the symmetry direction of  $\mathcal{H} = 2/m$  is oriented parallel to the  $[120]$  direction so that the groupoid has the structure given in Table 6.

The operations common to the two nuclei are the identity and the inversion, so that  $\mathcal{H}^* = \bar{1}$ . The operations  $2_{[210]}$  and  $m_{[210]}$  (for  $Z_T = {}^3_5$ ) and  $2_{[110]}$  and  $m_{[110]}$  (for  $Z_T = {}^3_4$ ) occur in both shells and are therefore totally chromatic operations. The other operations (threefold rotations or  $Z_T = {}^3_5$  and sixfold rotations for  $Z_T = {}^3_4$ ) are partial and show that this is actually an incomplete allotwin. The chromatic point group, which leaves the whole edifice invariant, is in both cases  $\mathcal{K}^{(2)} = 2'/m'$ : because of the incomplete nature of the allotwin, the chromatic point group does not convey much information, as is instead the case of the groupoid.



**Figure 5**  
Extension of the case in Fig. 3(a) to an eight-individual allotwin with four orientations. In this case,  $\mathcal{H}^* = 1 \subset \mathcal{H}_i$  and the allotwin can be divided into two regions containing only individuals with different orientation, forming each a four-individual twin.

**5.2. Polytypes with the same type of point group all in parallel orientation:**  $\Phi(i \rightarrow j) \in \mathcal{H}_{ij}^* \forall i, j$

The case of an allotwin in which all the individuals have the same type of point group and all the mappings belong to the intersection group of the point groups of the individuals corresponds to path 2.2 of the flowchart in Fig. 2 and can be schematically represented by the example in Fig. 3(a), where four pyramids with isosceles triangles as basis, decorated with four colours, white, black, red and yellow (W, B, R, Y in the following), represent four polytypes all with point group  $m11$ . The different colours indicate a different structure, *i.e.* polytypes with a different number of layers defining the period along the stacking direction, or a different stacking sequence of the same number of layers. The mapping of any pair of individuals is realized through operations that correspond to the same isometry but differ in their chromaticity. These operations can be symbolized by adding a sequence letter to the chromatic index.  $\{1^{(4a)}, m^{(4a)}\}$  are mappings of neighbour colours, *i.e.* the cyclic permutation (2341):  $W \rightarrow B \rightarrow Y \rightarrow R \rightarrow W$ .  $\{1^{(4b)}, m^{(4b)}\}$  are mappings of second neighbours, *i.e.* the permutation (13)(24):  $W \leftrightarrow Y$  and  $B \leftrightarrow R$ .  $\{1^{(4c)}, m^{(4c)}\}$  indicate the reverse cyclic permutation (4321):  $W \rightarrow R \rightarrow Y \rightarrow B \rightarrow W$ . Finally, the achromatic operations retain their usual symbols,  $\{1, m\}$  and correspond to mappings  $W \leftrightarrow W$ ,  $B \leftrightarrow B$ ,  $R \leftrightarrow R$  and  $Y \leftrightarrow Y$ . The chromatic group of this allotwin is the grey group  $m11,1^{(4)}$ . Fig. 3(b) shows the same analysis for the case of individuals with point group  $mm2$ , represented by rhombic pyramids. The chromatic group of this allotwin is the neutral group  $mm2,1^{(4)}$ . The analysis is easily extended to higher chromatic indices.

In the literature we have found two examples, both dichromatic. The first one is the rhenium pincer complex  $cis\text{-Re}[(\text{PNP}^{\text{CH}_2}\text{-iPr})(\text{CO})_2\text{Cl}]$  studied by Glatz *et al.* (2017), who reported samples made up of two polytypes with respective symmetries  $P2_1/c$  ( $a = 9.6475$ ,  $b = 10.7392$ ,  $c = 25.629$  Å,  $\beta = 68.684^\circ$ ) and  $I2/a$  ( $a = 18.6854$ ,  $b = 10.7708$ ,  $c = 25.599$  Å,  $\beta = 107.480^\circ$ ). The layers in both polytypes are parallel to the (100) plane, differently stacked along the  $[100]$  direction. The allotwin lattice is the common sublattice obtained from the unit cell of the  $P2_1/c$  polytype with transformation  $4\mathbf{a} - \mathbf{c}$ ,  $\mathbf{b}$ ,  $\mathbf{c}$  [inverse transformation  $(\mathbf{a} + \mathbf{c})/4$ ,  $\mathbf{b}$ ,  $\mathbf{c}$ ] and from the unit cell of the  $I2/a$  polytype with transformation  $2\mathbf{a}$ ,  $\mathbf{b}$ ,  $\mathbf{c}$  (inverse transformation  $\mathbf{a}/2$ ,  $\mathbf{b}$ ,  $\mathbf{c}$ ) neglecting the small metric differences between the two polytypes. The transformation from  $P2_1/c$  to

$12/a$  is obtained by combining the former with the inverse of the latter, the result being  $2\mathbf{a} - \mathbf{c}/2$ ,  $\mathbf{b}$ ,  $\mathbf{c}$ , which is the inverse of the transformation in equation (1) of Glatz *et al.* (2017) for the corresponding transformation in reciprocal space. This shows that the basis vectors in the plane of the layer, *i.e.*  $\mathbf{b}$  and  $\mathbf{c}$ , are parallel in the allottwinned polytypes. Therefore [equation (1)]:

$$\mathbf{M}_1 = \begin{pmatrix} 4 & 0 & 0 \\ 0 & 1 & 0 \\ \bar{1} & 0 & 1 \end{pmatrix}; \mathbf{M}_2 = \begin{pmatrix} 2 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix};$$

$$\mathbf{M}_2^{-1} = \begin{pmatrix} \frac{1}{2} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \mathbf{M}_1\mathbf{M}_2^{-1} = \begin{pmatrix} 2 & 0 & 0 \\ 0 & 1 & 0 \\ \bar{1} & 0 & 1 \end{pmatrix}$$

and the  $2 \times 2$  block of the  $\mathbf{M}_1^{-1}\mathbf{M}_2$  matrix corresponding to the  $\mathbf{b}$  and  $\mathbf{c}$  basis vectors, which represents the mapping of the two polytypes in the allottwin, is the identity matrix. The point group of both individuals is therefore the same (both in type and orientation) so that the allottwin point group is the grey group  $12/m1,1'$ .

The second example is an allottwin of two polytypes of  $\text{KAgCO}_3$  reported by Hans *et al.* (2015), with space groups of type *Ibca* and *Pccb*. The layer is again parallel to the (100) plane, differently stacked along the [100] direction. The *a* parameter of the *Ibca* polytype is twice the *a* parameter of the *Pccb* polytype, and the unit cell of the allottwin lattice coincides with the unit cell of the *Ibca* polytype. As in the previous example, the basis vectors in the plane of the layer, *i.e.*  $\mathbf{b}$  and  $\mathbf{c}$ , are parallel in the allottwinned polytypes. Therefore [equation (1)]:

$$\mathbf{M}_1 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \mathbf{M}_2 = \begin{pmatrix} 2 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix};$$

$$\mathbf{M}_2^{-1} = \begin{pmatrix} \frac{1}{2} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \mathbf{M}_1\mathbf{M}_2^{-1} = \begin{pmatrix} \frac{1}{2} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

and the  $2 \times 2$  block of the  $\mathbf{M}_1\mathbf{M}_2^{-1}$  matrix corresponding to the  $\mathbf{b}$  and  $\mathbf{c}$  basis vectors is again the identity matrix. The allottwin point group is the grey group  $mmm,1'$ .

### 5.3. Allotwins with point groups of the same type and individuals both in parallel and different orientation:

$\Phi(i \rightarrow j) \in \mathcal{H}_{ij}^*$  for  $p1$  individuals out of  $p$

This case corresponds to path 2.3 of the flowchart in Fig. 2 and requires the use of extended chromatic symbols not yet described.<sup>7</sup> Let us first consider the case  $\mathcal{H}^* = \mathcal{H}$ , *i.e.* the case when  $\mathcal{H}$  is a normal subgroup of  $\mathcal{K}^{(p)}$ , starting from the four-individual allottwin with point group  $mm2$  in Fig. 3(b). We reduce the symmetry of the individuals to  $m11$ , as in the case of Fig. 3(a), by cutting the pyramids in half, retaining however the left half for two of them, and the right half for the other

two (Fig. 4a). The resulting crystal association can be described as an 'allotwin of twins' or as a 'twin of allottwins' and is composed of four individuals in two different orientations, with the same type of point group, related by the two operations  $m_{[010]}$  and  $2_{[001]}$  which have been removed in moving from the group  $mm2$  to the subgroup  $m11$ . In other words, the four operations that were neutral in Fig. 3(b) are now divided into two types: the identity and the reflection through the plane (100) act once as achromatic (symmetry operations of the individuals) and once as chromatic (mapping  $W \leftrightarrow B$  and  $R \leftrightarrow Y$ ); the reflection through the plane (010) and the rotation about the axis [001] act twice as chromatic operations, with different chromaticity. Concretely, the result of their action is once  $W \leftrightarrow R$ ,  $B \leftrightarrow Y$  and once  $W \leftrightarrow Y$ ,  $B \leftrightarrow R$ . Individuals in parallel orientation ( $B$  and  $W$ ;  $Y$  and  $R$ ) form two regions, each described by the grey group  $m11,1'$  (grey rectangle on the left); those with different orientation ( $B$  and  $Y$ ;  $W$  and  $R$ ) form two twinned regions described by the dichromatic group  $mm'2'$  (hatched rectangle on the bottom). The intersection group coincides with the group of the individuals:  $\mathcal{H}^* = \cap_i \mathcal{H}_i = m11$ .

Each geometric operation acts twice, with different chromatic components.  $1$  and  $m_{[100]}$  act once as achromatic operations (symmetry operations of each polytype) and once as chromatic operations, mapping different polytypes in parallel orientations ( $W \leftrightarrow B$  and  $R \leftrightarrow Y$ ).  $m_{[010]}$  and  $2_{[001]}$  act twice as chromatic operations but with different chromatic components: in one case they result in the exchanges  $W \leftrightarrow R$  and  $B \leftrightarrow Y$  (operation 'a'), in the second case  $W \leftrightarrow Y$  and  $B \leftrightarrow R$  (operation 'b'). The point group is tetrachromatic; it is a supergroup of both the grey group  $m11,1'$  and of the dichromatic group  $m2'm'$ ; its symbol, constructed on the model of the extended symbols of space groups, is

$$\mathcal{K}^{(4)} = \begin{pmatrix} m & m^{(2a)} & 2^{(2a)} \\ m^{(2)} & m^{(2b)} & 2^{(2b)} \end{pmatrix}^{(4)}$$

(white square on the right), which can be shortened to  $(mm^{(2)}2^{(2)})^{(4)}$ . The difference with respect to the dichromatic symbol  $mm'2'$  is evident from the external superscript (4), which shows that the group is tetrachromatic and not dichromatic, and from the internal superscript (2), which is used instead of ' when a dichromatic operation occurs in a polychromatic group.

The situation is similar to the case of two glide reflections that share the same geometric element in a space group, which occurs in space groups whose conventional cell is centred. For example, in a space group of type  $Amm2$ , the (100) plane (geometric element) acts both as mirror plane and as  $n$ -glide plane: the two operations are related by the  $A$  centring vector. In the case of the four polytypes in Fig. 4(a), the identity and the (100) plane act once as achromatic and once as chromatic: to differentiate them the symbols  $1, 1^{(2)}$ , as well as  $m_{[100]}$  and  $m^{(2)}_{[100]}$  are used below. On the other hand, the (010) plane acts twice, each time with a different chromaticity; to differentiate these two operations, we adopt the symbols  $m^{(2a)}_{[010]}$  and  $m^{(2b)}_{[010]}$ . In the same way, the twofold rotation about

<sup>7</sup> This example came to my attention following a discussion with one of the reviewers, to whom I express my gratitude.

**Table 7**  
Groupoid for Section 5.3.

$\{1, m_{[100]}\}$	$\{1, m_{[100]}\} m^{(2a)}_{[010]}$	$\{1, m_{[100]}\} 1^{(2)}$	$\{1, m_{[100]}\} m^{(2b)}_{[010]}$
$m^{(2a)}_{[010]}\{1, m_{[100]}\}$	$m^{(2)}_{[010],1}\{1, m_{[100]}\} m^{(2a)}_{[010]}$	$m^{(2a)}_{[010]}\{1, m_{[100]}\} 1^{(2)}$	$m^{(2a)}_{[010]}\{1, m_{[100]}\} m^{(2b)}_{[010]}$
$1^{(2)}\{1, m_{[100]}\}$	$1^{(2)}\{1, m_{[100]}\} m^{(2a)}_{[010]}$	$1^{(2)}\{1, m_{[100]}\} 1^{(2)}$	$1^{(2)}\{1, m_{[100]}\} m^{(2b)}_{[010]}$
$m^{(2b)}_{[010]}\{1, m_{[100]}\}$	$m^{(2b)}_{[010]}\{1, m_{[100]}\} m^{(2a)}_{[010]}$	$m^{(2b)}_{[010]}\{1, m_{[100]}\} 1^{(2)}$	$m^{(2b)}_{[010]}\{1, m_{[100]}\} m^{(2b)}_{[010]}$
$\{1, m_{[100]}\}$	$\{m^{(2a)}_{[010]}, 2^{(2a)}_{[001]}\}$	$\{1^{(2)}, m^{(2)}_{[100]}\}$	$\{m^{(2b)}_{[010]}, 2^{(2b)}_{[001]}\}$
$\{m^{(2a)}_{[010]}, 2^{(2a)}_{[001]}\}$	$\{1, m_{[100]}\}$	$\{m^{(2b)}_{[010]}, 2^{(2b)}_{[001]}\}$	$\{1^{(2)}, m^{(2)}_{[100]}\}$
$\{1^{(2)}, m^{(2)}_{[100]}\}$	$\{m^{(2b)}_{[010]}, 2^{(2b)}_{[001]}\}$	$\{1, m_{[100]}\}$	$\{m^{(2a)}_{[010]}, 2^{(2a)}_{[001]}\}$
$\{m^{(2b)}_{[010]}, 2^{(2b)}_{[001]}\}$	$\{1^{(2)}, m^{(2)}_{[100]}\}$	$\{m^{(2a)}_{[010]}, 2^{(2a)}_{[001]}\}$	$\{1, m_{[100]}\}$

$[001]$  has two different chromatic actions that will be indicated as  $2^{(2a)}_{[001]}$  and  $2^{(2b)}_{[001]}$ . The groupoid is constructed in the usual way (see Table 7).

Each and every operation occurs once in every mixed group; they are therefore total operations and the groupoid degenerates into a group. A group like this, which contains different chromatic operations about the same geometric element, is fully represented by an extended symbol similar to those used for space groups. In the short Hermann–Mauguin symbol of the *Am*m2 example above, which in this case coincides with the full symbol, only the mirror planes and the rotation axis are shown, according to the priority rule for the construction of space-group symbols. The existence of the glide planes and screw axes becomes explicit when the extended symbol is used, namely

$$A \quad m \quad m \quad 2 \\ \quad \quad n \quad c \quad 2_1 :$$

the two planes perpendicular to  $[100]$  share the same geometric element, namely  $0yz$ , whereas the symmetry elements along the other direction correspond to different geometric elements:  $m$  for  $x0z$ ,  $c$  for  $x\frac{1}{2}z$ ,  $2$  for  $00z$  and  $2_1$  for  $0\frac{1}{2}z$ . The chromatic point group obtained by extending the dichromatic point group by the colour identification group,  $(mm^{(2)}2^{(2)}, 1^{(2)})^{(4)}$ , is represented by the extended symbol shown above. Fig. 4(b) shows the same analysis for the case of individuals with point group  $mm2$  in two orientations related by a fourfold rotation about the twofold symmetry axis. The dichromatic point group describing the region where polytypes have different orientation is  $4'mm'$ ; each region is then mapped by operations belonging to  $\mathcal{H}^*$  and is described by the grey group  $mm2, 1'$ ; the four-individual allotwin is eventually described by the extension of the former by the latter,  $(4^{(2)}mm^{(2)}, 1^{(2)})^{(4)}$ , with extended symbol

$$\left( \begin{matrix} 4^{(2a)} & m & m^{(2a)} \\ 4^{(2b)} & m^{(2)} & m^{(2b)} \end{matrix} \right)^{(4)}$$

and short symbol  $(4^{(2)}mm^{(2)})^{(4)}$ , which cannot be confused with the corresponding dichromatic symbol  $4'mm'$ . Two

examples of this type of allotwin were reported very recently and are discussed below.

The case of  $\mathcal{H}^* \subset \mathcal{H}$  is represented in Fig. 5, which shows an extension of the case in Fig. 4(a) with eight individuals in four orientations. In this case,  $\mathcal{H}^* = 1$ . Each of the two regions containing the four individuals with different orientation are described by the chromatic group  $\mathcal{K}^{(4)} = (4^{(4)}m^{(2,2)}m^{(2)})^{(4)}$ : the four individuals may correspond to the same or a different polytype. Because of the different orientation, each of the two regions is completely represented by the chromatic group  $\mathcal{K}^{(4)}$ . Each individual in a region has a corresponding individual in the other region which is oriented parallel to it: each region with two polytypes in parallel orientation is described by the grey group  $m\bar{1}1, 1'$ . Each of the operations in the group  $\mathcal{K}^{(4)}$  acts twice with two different chromatic components (colour codes: W = white, B = black, G = green, C = cyan, R = red, Y = yellow, M = magenta, O = orange):

(i) The identity acts once as achromatic operation and once mapping  $W \leftrightarrow B, G \leftrightarrow C, R \leftrightarrow Y$  and  $M \leftrightarrow O$ .

(ii)  $4^1_{[001]}$  exchanges once  $W \leftrightarrow G \leftrightarrow R \leftrightarrow M$  and  $B \leftrightarrow C \leftrightarrow Y \leftrightarrow O$  and once  $W \leftrightarrow C \leftrightarrow R \leftrightarrow O$  and  $B \leftrightarrow G \leftrightarrow Y \leftrightarrow M$ .

(iii)  $2_{[001]}$  exchanges once  $W \leftrightarrow R, G \leftrightarrow M, B \leftrightarrow Y, C \leftrightarrow O$  and once  $W \leftrightarrow Y, G \leftrightarrow O, B \leftrightarrow R, C \leftrightarrow M$ .

(iv)  $4^3_{[001]}$  exchanges once  $W \leftrightarrow M \leftrightarrow R \leftrightarrow G$  and  $B \leftrightarrow O \leftrightarrow Y \leftrightarrow C$  and once  $W \leftrightarrow O \leftrightarrow R \leftrightarrow C$  and  $B \leftrightarrow M \leftrightarrow Y \leftrightarrow G$ .

(v)  $m_{[100]}$  is partially chromatic and exchanges once  $G \leftrightarrow M, C \leftrightarrow O$  and once  $G \leftrightarrow O, C \leftrightarrow M$ , while fixing  $W, B, R$  and  $Y$ .

(vi)  $m_{[010]}$  is partially chromatic and exchanges once  $W \leftrightarrow R, B \leftrightarrow Y$  and once  $W \leftrightarrow Y, B \leftrightarrow R$ , while fixing  $G, M, C$  and  $O$ .

(vii)  $m_{[110]}$  exchanges once  $W \leftrightarrow G, R \leftrightarrow M, B \leftrightarrow C, Y \leftrightarrow O$  and once  $W \leftrightarrow C, R \leftrightarrow O, B \leftrightarrow G, Y \leftrightarrow M$ .

(viii)  $m_{[1\bar{1}0]}$  exchanges once  $W \leftrightarrow M, R \leftrightarrow G, B \leftrightarrow O, Y \leftrightarrow C$  and once  $W \leftrightarrow O, R \leftrightarrow C, B \leftrightarrow M, Y \leftrightarrow G$ .

The whole allotwin can be described by an octachromatic group obtained by extending the tetrachromatic twin point group  $\mathcal{K}^{(4)}$  by the colour identification group  $1^{(2)}$ : the extended symbol is obtained from the extension  $((4^{(4)}m^{(2,2)}m^{(2)})^{(4)}, 1^{(2)})^{(8)}$ , resulting in

**Table 8**  
Groupoid for vanadosilicate example.

$\{1, 2_{[010]}\}$	$\{1, 2_{[010]}\}m^{(2a)}_{[100]}$	$\{1, 2_{[010]}\}1^{(2)}$	$\{1, 2_{[010]}\}m^{(2b)}_{[100]}$
$m^{(2a)}_{[100]}\{1, 2_{[010]}\}$	$m^{(2a)}_{[100]}\{1, 2_{[010]}\}m^{(2a)}_{[100]}$	$m^{(2a)}_{[100]}\{1, 2_{[010]}\}1^{(2)}$	$m^{(2a)}_{[100]}\{1, 2_{[010]}\}m^{(2b)}_{[100]}$
$1^{(2)}\{1, 2_{[010]}, \bar{1}\}$	$1^{(2)}\{1, 2_{[010]}\}m^{(2a)}_{[100]}$	$1^{(2)}\{1, 2_{[010]}\}1^{(2)}$	$1^{(2)}\{1, 2_{[010]}\}m^{(2b)}_{[100]}$
$m^{(2b)}_{[100]}\{1, 2_{[010]}\}$	$m^{(2b)}_{[100]}\{1, 2_{[010]}\}m^{(2a)}_{[100]}$	$m^{(2b)}_{[100]}\{1, 2_{[010]}\}1^{(2)}$	$m^{(2b)}_{[100]}\{1, 2_{[010]}\}m^{(2b)}_{[100]}$
$\{1, 2_{[010]}\}$	$\{m^{(2a)}_{[100]}, m^{(2a)}_{[001]}\}$	$\{1^{(2)}, 2^{(2)}_{[010]}\}$	$\{m^{(2b)}_{[100]}, m^{(2b)}_{[001]}\}$
$\{m^{(2a)}_{[100]}, m^{(2a)}_{[001]}\}$	$\{1, 2_{[010]}\}$	$\{m^{(2b)}_{[100]}, m^{(2b)}_{[001]}\}$	$\{1^{(2)}, 2^{(2)}_{[010]}\}$
$\{1^{(2)}, 2^{(2)}_{[010]}\}$	$\{m^{(2b)}_{[100]}, m^{(2b)}_{[001]}\}$	$\{1, 2_{[010]}\}$	$\{m^{(2a)}_{[100]}, m^{(2a)}_{[001]}\}$
$\{m^{(2b)}_{[100]}, m^{(2b)}_{[001]}\}$	$\{1^{(2)}, 2^{(2)}_{[010]}\}$	$\{m^{(2a)}_{[100]}, m^{(2a)}_{[001]}\}$	$\{1, 2_{[010]}\}$

$$\begin{pmatrix} 4^{(4a)} & m^{(2a,2)} & m^{(2a)} \\ 4^{(4b)} & m^{(2b,2)} & m^{(2b)} \end{pmatrix}^{(8)},$$

or the short symbol  $(4^{(4)}m^{(2,2)}m^{(2)})^{(8)}$ .

If the number of individuals in each twinned region is different, the allotwin is incomplete and the groupoid should be explicitly given to provide a satisfactory description of the allotwinned edifice.

In the literature we have found two examples corresponding to this category of allotwins.

**5.3.1. Vanadosilicate VSH-13Na.** Danisi & Armbruster (2018) reported a complex allotwin in the microporous vanadosilicate VSH-13Na, with composition  $\text{Na}_2(\text{VO})\text{-}(\text{Si}_4\text{O}_{10})\cdot 3\text{H}_2\text{O}$ . This compound has been found to occur in two polytypes ( $P_1$  and  $P_2$ ), both crystallizing in  $P12_11$ , which differ in the orientation of the vanadyl groups (parallel and anti-parallel). Both polytypes are twinned by mirror reflection about (100) ( $P_1 \leftrightarrow P_1'$ ;  $P_2 \leftrightarrow P_2'$ ), resulting in the dichromatic point group  $m'2m'$  for each of them; twinning is by pseudo-merohedry, due to the metric of the lattice which is almost orthorhombic ( $\beta \simeq 90^\circ$ ). The two polytypes, whose cell parameters are almost identical, are then allotwinned in parallel orientation. The transformation matrices are trivial:

$$\begin{aligned} \mathbf{M}_1 &= \mathbf{M}_2 = \mathbf{M}_1^{-1} = \mathbf{M}_2^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \\ \mathbf{M}_1' &= \mathbf{M}_2' = \mathbf{M}_1'^{-1} = \mathbf{M}_2'^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \bar{1} \end{pmatrix}; \\ \mathbf{M}_i\mathbf{M}_j^{-1} &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \mathbf{M}_i\mathbf{M}_j'^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \bar{1} \end{pmatrix}. \end{aligned}$$

The groupoid is obtained as shown in Table 8.

The four nuclei identify the intersection group  $\mathcal{H}^* = \mathcal{H} = 121$ , which obviously coincides with the point group of the individuals. The identity as well as the operation  $2_{[010]}$  act as

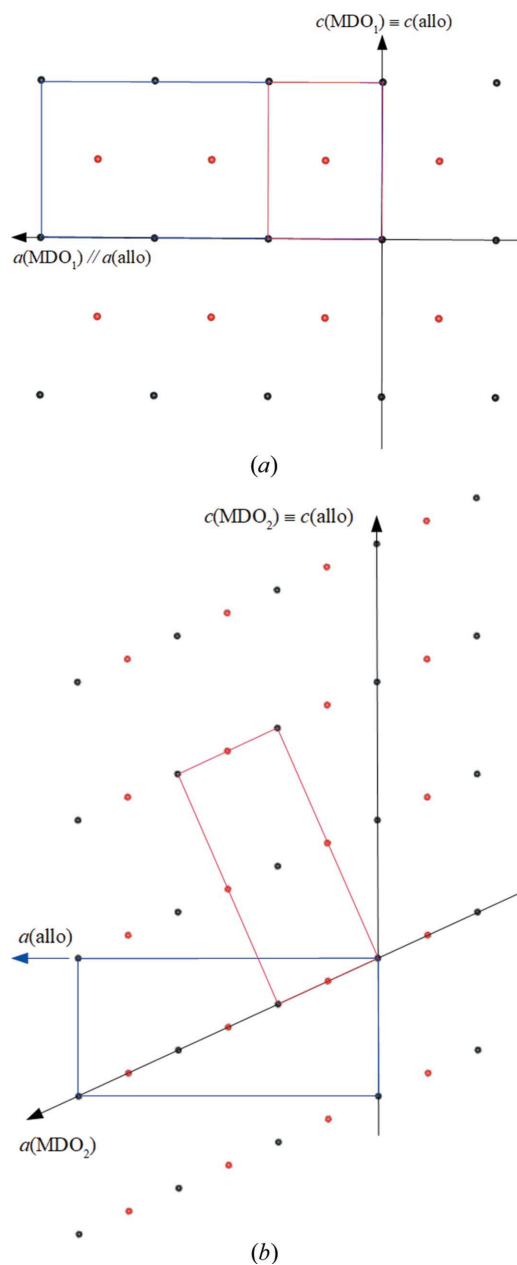
symmetry operation for each polytype,  $\{1, 2_{[010]}\}$ , but also as allotwin operations relating  $P_1$  and  $P_2$ , as well as  $P_1'$  and  $P_2'$ :  $\{1^{(2)}, 2^{(2)}_{[010]}\}$ . The two mirror reflections act twice as chromatic elements:  $\{m^{(2a)}_{[100]}, m^{(2a)}_{[001]}\}$  for  $P_1 \leftrightarrow P_1'$  and  $P_2 \leftrightarrow P_2'$ , and  $\{m^{(2b)}_{[100]}, m^{(2b)}_{[001]}\}$  for  $P_1 \leftrightarrow P_2'$  and  $P_1 \leftrightarrow P_2'$ . All the operations are global, so that the groupoid degenerated into a tetrachromatic group

$$\begin{pmatrix} m^{(2a)} & 2 & m^{(2a)} \\ m^{(2b)} & 2^{(2)} & m^{(2b)} \end{pmatrix}^{(4)},$$

with short symbol  $(m^{(2)}2m^{(2)})^{(4)}$ . The allotwin lattice coincides with the lattice of the individuals, if we neglect the small metric differences.

**5.3.2.  $\text{KCa}_3\text{Te}_5\text{O}_{12}\text{Cl}_3$ .** Larvor *et al.* (2018) reported the crystal structure of  $\text{KCa}_3\text{Te}_5\text{O}_{12}\text{Cl}_3$  from a four-domain sample composed of two polytypes built up of  $[\text{KTe}_5\text{O}_{12}]_3$  layers extending in the (100) plane, with calcium cations and chloride anions in the interlayer regions. The two polytypes have almost identical  $b$  and  $c$  cell parameters (these correspond to the basis vectors in the plane of the layer) and differ in the layer stacking, resulting in different  $a$  and  $\beta$  cell parameters. Both polytypes occurred as twins and the allotwin is built up by these two twins. The description of twinning was given without sufficient details but a comprehensive understanding of twinning in both samples is mandatory to understand the formation of the allotwin and to provide a satisfactory description of it.

The  $I2/a$  polytype ( $\text{MDO}_1$ : maximum degree of order) has cell parameters  $a = 17.847$ ,  $b = 7.5525$ ,  $c = 24.646$  Å,  $\beta = 89.82^\circ$ ; it undergoes twinning by pseudo-merohedry with obliquity  $0.18^\circ$ . The twin lattice is pseudo-orthorhombic and coincides with the lattice of the individual. The decomposition of the orthorhombic holohedry (symmetry of the twin lattice within the approximation of the obliquity) with respect to the monoclinic holohedry (symmetry of the individual) results in two cosets: the point group of the individual and the twin law; the twin point group is dichromatic:


**Figure 6**

(a) Lattice of the  $I2/a$  polytype ( $MDO_1$ ) of  $KCa_3Te_5O_{12}Cl_3$ : black and red nodes are at integer and semi-integer values of the  $y$  coordinate. Red unit cell: the unit cell of the individual, which is pseudo- $oI$ . The twin by pseudo-merohedry reported by Larvor *et al.* (2018) has this unit cell for the twin lattice. Blue unit cell: the pseudo- $oI$  unit cell obtained by the basis vector change  $3\mathbf{a}$ ,  $\mathbf{b}$ ,  $\mathbf{c}$ , which is metrically very close to the blue unit cell of the twin lattice of the  $C2/c$  ( $MDO_2$ ) polytype in (b). (b) Lattice of the  $C2/c$  ( $MDO_2$ ) polytype of  $KCa_3Te_5O_{12}Cl_3$ : same graphic conventions as in (a). Red unit cell: the pseudo- $oF$  unit cell obtained by the basis vector change  $\mathbf{a}$ ,  $\mathbf{b}$ ,  $\mathbf{a} + 2\mathbf{c}$ , which shows that the lattice is actually pseudo-orthorhombic. Blue unit cell: the pseudo- $oI$  unit cell obtained by the basis vector change  $3\mathbf{a} + \mathbf{c}$ ,  $\mathbf{b}$ ,  $\mathbf{c}$ , which shows the presence of a pseudo-orthorhombic sublattice. A twin by reflection through (001) of the individual would result in twinning by pseudo-merohedry with the red unit cell as cell of the twin lattice; a twin by reflection through (100) of the individuals would result in twinning by reticular pseudo-merohedry (twin index 3) with the blue unit cell as cell of the twin lattice. The latter is the twin reported by Larvor *et al.* (2018). The blue unit cell is also the unit cell of the allotwin lattice. The symbols  $\equiv$  and  $//$  stand for 'coincides with' and 'is parallel to', respectively.

$$2'/m'2/m2'/m'$$

$$= \{1, 2_{[010]}, \bar{1}, m_{[010]}\} \cup \{2_{[100]}, 2_{[001]}, m_{[100]}, m_{[001]}\}. \quad (4)$$

Any operation in the second coset (twin law) is a twin operation that relates the two individuals; they would be exactly equivalent if the obliquity were zero.

The  $C2/c$  polytype ( $MDO_2$ ) has cell parameters  $a = 19.635$ ,  $b = 7.5496$ ,  $c = 24.670$  Å,  $\beta = 114.622^\circ$ . The value of  $-2c \cos \beta/a$  is 1.05, showing that the lattice is actually pseudo- $oF$ . Accordingly, this polytype can undergo twinning by pseudo-merohedry with obliquity  $1.18^\circ$ . The change of basis vector which best shows the orthorhombic pseudo-symmetry is ( $\mathbf{a}$ ,  $\mathbf{b}$ ,  $\mathbf{a} + 2\mathbf{c}$ ), resulting in cell parameters  $a = 19.635$ ,  $b = 7.5496$ ,  $c = 44.8633$  Å,  $\beta = 91.18^\circ$ , for the pseudo- $oF$  unit cell; the (100) and (001) lattice planes in the  $mC$  setting become (101) and (001), respectively, in the (pseudo)- $oF$  setting and the twin element would be the (001) plane. The dichromatic point group and the twin law would be the same as in equation (4). However, this is not the twin that was reported experimentally. A different twin was instead found, whose twin lattice is obtained by the transformation ( $3\mathbf{a} + \mathbf{c}$ ,  $\mathbf{b}$ ,  $\mathbf{c}$ ), corresponding to a pseudo- $oI$  unit cell with parameters  $a = 55.5493$ ,  $b = 7.5496$ ,  $c = 24.670$  Å,  $\beta = 89.86^\circ$ . Twinning is by reticular pseudo-merohedry, with twin index 3 and obliquity  $0.14^\circ$ . The (100) and (001) lattice planes in the  $mC$  setting become (100) and (101), respectively, in the (pseudo)- $oI$  setting and the twin element is the (100) plane. Again, with respect to this unit cell, the dichromatic point group and the twin law are expressed by equation (4).

Now, the cell parameters of the twin lattice obtained from the unit cell of the  $C2/c$  polytype can be made to almost coincide with those of a sublattice of the  $I2/a$  polytype having triple period along  $a$  ( $55.549/3 \times 17.847 = 1.04$ ) (Fig. 6). Therefore, apart from this small metric deviation, the twin lattice of the  $C2/c$  individual is also the allotwin lattice; the  $\mathbf{b}$  and  $\mathbf{c}$  vectors, *i.e.* the vectors in the (100) plane of the layer, are common to the polytypes and their twins:

$$\mathbf{M}_1 = \begin{pmatrix} 3 & 0 & 0 \\ 0 & 1 & 0 \\ 1 & 0 & 1 \end{pmatrix}; \mathbf{M}_2 = \begin{pmatrix} 3 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix};$$

$$\mathbf{M}_2^{-1} = \begin{pmatrix} \frac{1}{3} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}; \mathbf{M}_1\mathbf{M}_2^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ \frac{1}{3} & 0 & 1 \end{pmatrix}.$$

The  $\mathbf{M}_1\mathbf{M}_2^{-1}$  shows that the  $\mathbf{b}$  and  $\mathbf{c}$  basis vectors, which are the vectors in the plane of the layer, are common to the lattice of the two polytypes as well as to the allotwin lattice. The point groups of the four individuals, as well as the dichromatic point groups of the two twins, are all of type  $2/m$  and are all iso-oriented. The two twinned polytypes are therefore allotwinned in parallel orientation. The groupoid is obtained as shown in Table 9.

The tetrachromatic group expressing the symmetry of the allotwin is obtained as an extension of the dichromatic group (4) by the colour identification group  $1'$ , *i.e.*

**Table 9**  
Groupoid for KCaTeOCl example.

$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2a)}_{[001]}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}1^{(2)}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2b)}_{[001]}$
$m^{(2a)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$m^{(2a)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2a)}_{[001]}$	$m^{(2a)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}1^{(2)}$	$m^{(2a)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2b)}_{[001]}$
$1^{(2)}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$1^{(2)}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2a)}_{[001]}$	$1^{(2)}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}1^{(2)}$	$1^{(2)}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2b)}_{[001]}$
$m^{(2b)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$m^{(2b)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2a)}_{[001]}$	$m^{(2b)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}1^{(2)}$	$m^{(2b)}_{[001]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m^{(2b)}_{[001]}$
$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{m^{(2a)}_{[001]}, m^{(2a)}_{[100]}, 2^{(2a)}_{[001]}, 2^{(2a)}_{[100]}\}$	$\{1^{(2)}, 2^{(2)}_{[010]}, \bar{1}^{(2)}, m^{(2)}_{[010]}\}$	$\{m^{(2b)}_{[001]}, m^{(2b)}_{[100]}, 2^{(2b)}_{[001]}, 2^{(2b)}_{[100]}\}$
$\{m^{(2a)}_{[001]}, m^{(2a)}_{[100]}, 2^{(2a)}_{[001]}, 2^{(2a)}_{[100]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{m^{(2b)}_{[001]}, m^{(2b)}_{[100]}, 2^{(2b)}_{[001]}, 2^{(2b)}_{[100]}\}$	$\{1^{(2)}, 2^{(2)}_{[010]}, \bar{1}^{(2)}, m^{(2)}_{[010]}\}$
$\{1^{(2)}, 2^{(2)}_{[010]}, \bar{1}^{(2)}, m^{(2)}_{[010]}\}$	$\{m^{(2b)}_{[001]}, m^{(2b)}_{[100]}, 2^{(2b)}_{[001]}, 2^{(2b)}_{[100]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{m^{(2a)}_{[001]}, m^{(2a)}_{[100]}, 2^{(2a)}_{[001]}, 2^{(2a)}_{[100]}\}$
$\{m^{(2b)}_{[001]}, m^{(2b)}_{[100]}, 2^{(2b)}_{[001]}, 2^{(2b)}_{[100]}\}$	$\{1^{(2)}, 2^{(2)}_{[010]}, \bar{1}^{(2)}, m^{(2)}_{[010]}\}$	$\{m^{(2a)}_{[001]}, m^{(2a)}_{[100]}, 2^{(2a)}_{[001]}, 2^{(2a)}_{[100]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$

$$\left( \begin{array}{ccc} 2^{(2a)}/m^{(2a)} & 2/m & 2^{(2a)}/m^{(2a)} \\ 2^{(2b)}/m^{(2b)} & 2^{(2)}/m^{(2)} & 2^{(2b)}/m^{(2b)} \end{array} \right)^{(4)},$$

whose short symbol is  $(2^{(2)}/m^{(2)}2/m^{(2)}/m^{(2)})^{(4)}$ . One may wonder why the MDO<sub>2</sub> polytype was found twinned by reflection about (100), which leads to twin index 3, instead of about (001), for which the twin index is 1. The probability of occurrence of a twin is roughly inversely proportional to the twin index and the obliquity; although the obliquity indeed would favour the (100) twin, the difference of about 1° can hardly compensate that difference in the twin index. We suspect that the formation of a sublattice common to all the four individuals (the allotwin lattice) was perhaps responsible for the occurrence of the (100) twin instead of the (001) but that makes sense if the four individuals are juxtaposed in such a way as to produce a common composition plane. Unfortunately, no morphological observation was reported; morphological data are of paramount importance in the study of twins, but they are seldom reported in the modern literature.

#### 5.4. Allotwins of polytypes with different types of point group

This case corresponds to path 3 of the flowchart in Fig. 2. An allotwin of malonamide, C<sub>3</sub>H<sub>6</sub>N<sub>2</sub>O<sub>2</sub>, has been reported by Strand *et al.* (2016). The first polytype crystallizes in a space group of type  $P12_1/a1$ , which is however better described in the unconventional setting  $B12_1/e1$  (Nespolo & Aroyo, 2016*b*), where the presence of a metric orthorhombic lattice becomes evident ( $a = -2c \cos \beta$  in the primitive cell, corresponding to  $\beta = 90^\circ$  in the  $B$ -centred cell); the second polytype crystallizes in a space group of type  $Pca2_1$ .<sup>8</sup> The  $a$  and  $b$  cell parameters of the two polytypes are identical; the  $c$  parameter

<sup>8</sup> We remind the reader that point groups of the same type are isomorphic, whereas the opposite is not true in general. The two polytypes in this example have point groups of different type,  $2/m$  and  $2mm$ , but they are isomorphic (Nespolo & Souvignier, 2009).

of the  $oB$  cell of the monoclinic polytype (transformation  $\mathbf{a}, \mathbf{b}, \mathbf{a} + 2\mathbf{c}$  from the  $mP$  cell) is twice the  $c$  parameter of the  $oP$  cell of the orthorhombic polytype within one standard uncertainty, so that the unit cell of the allotwin lattice coincides with the  $oB$  unit cell of the monoclinic polytype and is twice the unit cell of the orthorhombic polytype. The respective basis vectors of the  $oB$  and  $oP$  unit cells are all parallel; the intersection group of the point groups of the two individuals is  $1m1$  and the chromatic point group is  $\mathcal{K}^{(2)} = 1m1, 1'$ . The symmetry operations of the two point groups,  $mmm$  and  $12/m1$ , not present in  $\mathcal{H}^*$ , are local operations of the individuals and do not appear in the neutral point group.

The structure of  $\alpha$ - and  $\beta$ -Sb<sub>3</sub>O<sub>4</sub>I has been reported by Hugonin *et al.* (2009) from a sample in which they formed an allotwin. Sb<sub>3</sub>O<sub>4</sub>I consists of rods of composition Sb<sub>3</sub>O<sub>4</sub> separated by isolated iodine ions; the two polymorphs  $\alpha$  and  $\beta$  are actually polytypes which differ only in the next nearest neighbour arrangement.  $\alpha$ -Sb<sub>3</sub>O<sub>4</sub>I crystallizes in a space group of type  $Pbn2_1$  (unconventional setting of  $Pna2_1$ ) whereas  $\beta$ -Sb<sub>3</sub>O<sub>4</sub>I crystallizes in a space group of type  $P12_1/c1$ . A common orthorhombic cell can be obtained with basis vector transformations  $\mathbf{a}, \mathbf{b}, 2\mathbf{c}$  from  $Pbn2_1$  and  $\mathbf{b}, \mathbf{c}, 2\mathbf{a} + \mathbf{c}$  from  $P12_1/c1$ ; in this common subcell the Sb<sub>3</sub>O<sub>4</sub> rods in both polytypes are oriented along  $\mathbf{a}$ . This subcell defines the allotwin lattice; in this setting, the point groups of the two polytypes are  $mmm$  and  $m11$ , respectively, and their intersection group is  $\mathcal{H}^* = m11$ . By extending this group through the colour identification group  $1'$  we can assign the grey group  $\mathcal{K}^{(2)} = m11, 1'$  to the allotwin. As in the case of malonamide, the symmetry operations of the two point groups,  $mmm$  and  $2/m11$ , not present in  $\mathcal{H}^*$ , are local operations of the individuals and do not appear in the neutral point group.

In both cases, a complete description of the allotwin requires the point groupoid.

We are not aware of cases of allotwins built by three or more different polytypes. However, some interesting examples

**Table 10**  
Groupoid for (1*R*,3*S*)-dimethyl 2-oxocyclohexane-1,3-dicarboxylate.

Twin of the first polytype	$\{1, m_{[010]}\}$	$\{1, m_{[010]}\}m_{[100]}$
	$m_{[100]}\{1, m_{[010]}\}$	$m_{[100]}\{1, m_{[010]}\}m_{[100]}$
Second polytype untwinned	$\{1, m_{[100]}, m_{[010]}, 2_{[001]}\}$	
Twin of the first polytype	$\{1, m_{[010]}\}$	$\{m_{[100]}, 2_{[001]}\}$
	$\{m_{[100]}, 2_{[001]}\}$	$\{1, m_{[010]}\}$
Second polytype untwinned	$\{1, m_{[100]}, m_{[010]}, 2_{[001]}\}$	

have been reported of allotwins of two polytypes with different types of point groups, in which either or both polytypes are twinned, leading to the presence of partially chromatic operations.

**5.4.1. Complex allotwinning in (1*R*,3*S*)-dimethyl 2-oxocyclohexane-1,3-dicarboxylate.** A complex allotwin of the title compound was reported by Jahangiri *et al.* (2013). Two polytypes, with space-group symmetry *Cc* and *Pmc*<sub>2</sub><sub>1</sub>, have a common orthorhombic sublattice obtained by the respective basis vector transformations **2a + c**, **b**, **c** and **2a**, **b**, **c**. The monoclinic polytype occurs as a two-individual twin with twin operation a mirror reflection through (100) of the orthorhombic sublattice, so that the twin point group is *m'**m**2'*, of the same type and iso-oriented with respect to the point group of the second polytype. The twin operation is a symmetry operation for the second polytype. The Ehresmann groupoid is obtained as shown in Table 10.

$\mathcal{H}^* = m$ , because the identity and the reflection  $m_{[010]}$  are common to the three nuclei, so that  $\mathcal{K}^{(3)} = m1^{(3)}$ , which does not convey much information about the allotwin. The operations  $m_{[100]}$  and  $2_{[001]}$  occur in the shell of the two mixed groups corresponding to the twin of the monoclinic polytype, but in the nucleus of the orthorhombic polytype and are therefore partially chromatic. In the case of twins, partially chromatic operations occur in higher-order twins, *i.e.* twins in which at least one of the twin operations is of order higher than two. The reason for this limitation is that a partially chromatic operation is at the same time a symmetry operation for a subset of the individuals and a twin operation for another subset; this implies the existence of a number of individuals larger than two, unless the twin is incomplete. In the case of allotwins, this limitation no longer exists if some of the symmetry operations of one polytype are twin operations for the other polytype, as is precisely the case under consideration. By collecting the operations according to their chromaticity we get the polychromatic symbol  $(m^{(2,1)}m2^{(2,1)})^{(3)}$ , which does not correspond to a point group but synthetically represents the Ehresmann point groupoid.

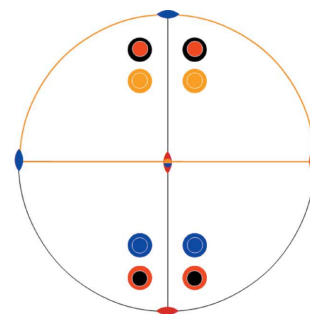
**5.4.2. Complex allotwinning in 9,9'-(2,5-dibromo-1,4-phenylene)bis[9*H*-carbazole].** The title compound was studied by Kautny *et al.* (2017), who reported two polytypes, both twinned, which eventually form a four-individual allotwin. The first polytype crystallizes in a space group of type *B*<sub>2</sub><sub>1</sub>/*d* [unconventional setting of *P*<sub>2</sub><sub>1</sub>/*c* (Nespolo & Aroyo, 2016*b*)], twinned by reflection through (100). The second polytype crystallizes in a space group of type *F*<sub>2</sub>*dd* twinned

by inversion. The relation between the cell parameters is (*m* = monoclinic, *o* = orthorhombic):  $a_o = a_m$ ,  $b_o = 2b_m$ ,  $c_o = c_m$ , with  $\beta_m = 90^\circ$ . The twin point group of the first polytype is *2'/m'2/m2'/m'*, that of the second polytype is *2/m'2'/m2'/m*; the two polytypes are allotwinned with their basis vectors parallel to each other, so that we can take the identity as the operation mapping their orientation (Table 11).

The intersection group of the four nuclei contains the identity and the reflection  $m_{[010]}$ , so that  $\mathcal{H}^* = m$  and  $\mathcal{K}^{(4)} = m1^{(4)}$ , again not very informative about the allotwin. The operations  $2_{[100]}$ ,  $2_{[010]}$ ,  $m_{[001]}$  and  $\bar{1}$  occur in the nucleus of one polytype and in the shell of the other polytype, so that they are partially chromatic. Finally, the operations  $2_{[001]}$  and  $m_{[100]}$  occur in the shell of both polytypes and are totally chromatic. By collecting the operations according to their chromaticity we get a polychromatic symbol for the Ehresmann point groupoid, which is  $(2^{(2,2)}/m^{(2,2)}2^{(2,2)}/m2^{(2,2)}/m^{(2,2)})^{(4)}$ . Fig. 7 shows that this is not the symbol of a group.

## 6. Discussion

In a previous publication (Nespolo *et al.*, 2004) we have emphasized the similarities between twins and modular structures. Twins are heterogeneous crystalline edifices whose symmetry can be described by a set of achromatic (individual-



**Figure 7**  
Stereographic projection of the symmetry, twin and allotwin elements and general forms of the complex allotwin of 9,9'-(2,5-dibromo-1,4-phenylene)bis[9*H*-carbazole] reported by Kautny *et al.* (2017). The general form of the first polytype, with space group of type *B*<sub>2</sub><sub>1</sub>/*d*, is shown closer to the circle; this polytype is twinned with dichromatic group *2'/m'2/m2'/m'* and the two twinned individuals are shown by black and red stereographic poles of their general form. The general form of the second polytype, with space group of type *F*<sub>2</sub>*mm*, is shown closer to the centre; this polytype is twinned with dichromatic group *2/m'2'/m2'/m* and the two twinned individuals are shown by blue and yellow stereographic poles of their general form. The intersection group of four nuclei is *m*, with achromatic mirror plane oriented perpendicular to the [010] direction. The twofold axis along [001] and the mirror plane perpendicular to [100] are totally chromatic: they exchange the four colours in pairs. The twofold axes along [100] and [010] and the mirror plane perpendicular to [001], as well as the inversion centre are partially chromatic: they are a symmetry element for one polytype and twin element for the other polytype. For this reason they are drawn with two different colours. The chromatic symmetry of the allotwin is represented by the symbol  $(2^{(2,2)}/m^{(2,2)}2^{(2,2)}/m2^{(2,2)}/m^{(2,2)})^{(4)}$  which shows the operations of the groupoid; because of the heterogeneity of the allotwin, it cannot be represented by a group.

**Table 11**  
Groupoid for 9,9'-(2,5-dibromo-1,4-phenylene)bis[9*H*-carbazole].

$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m_{[100]}$	$\{1, 2_{[100]}, m_{[010]}, m_{[001]}\}$	$\{1, 2_{[100]}, m_{[010]}, m_{[001]}\}\bar{1}$
$m_{[100]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$m_{[100]}\{1, 2_{[010]}, \bar{1}, m_{[010]}\}m_{[100]}$	$\bar{1}\{1, 2_{[100]}, m_{[010]}, m_{[001]}\}$	$\bar{1}\{1, 2_{[100]}, m_{[010]}, m_{[001]}\}\bar{1}$
$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{m_{[100]}, m_{[001]}, 2_{[100]}, 2_{[001]}\}$	$\{1, 2_{[100]}, m_{[010]}, m_{[001]}\}$	$\{\bar{1}, m_{[100]}, 2_{[010]}, 2_{[001]}\}$
$\{m_{[100]}, m_{[001]}, 2_{[100]}, 2_{[001]}\}$	$\{1, 2_{[010]}, \bar{1}, m_{[010]}\}$	$\{\bar{1}, m_{[100]}, 2_{[010]}, 2_{[001]}\}$	$\{1, 2_{[100]}, m_{[010]}, m_{[001]}\}$

fixing) and chromatic (individual-exchanging) operations, much like the symmetry of a modular structure can be described by a set of local (module-fixing) and partial (module-exchanging) operations. The whole set of operations constitutes a groupoid: a point groupoid in the case of twins, a space groupoid in the case of modular structures.

The existence of allotwins and their specificity were realized only about 20 years ago and since then a number of examples in both inorganic and organic crystals have been reported. A consistent symmetry description of allotwins was not available and the chromatic description already known for twins had not been applied to allotwins. We have shown that allotwins too can be described by chromatic operations. Allotwins in which the individuals have the same type of point group can be analysed in a similar way as twins: the whole set of operations constitutes a Brandt point groupoid, whereas the operations acting on all the individuals constitute a chromatic point group. With respect to twins, however, neutral chromatic groups, as well as extensions of twin point groups by neutral chromatic groups, have to be considered also: this comes from the fact that operations belonging to the point group of the individuals can also map different individuals, because of the difference in the structure, and thus in the space group. This calls for extended chromatic symbols built in the same way as the extended Hermann–Mauguin symbols of space groups. The corresponding short symbols are reminiscent of normal chromatic point groups, but the chromatic index clearly shows the difference. Nevertheless, the occasional user may experience some confusion by this similarity so that the extended symbols are safer and should be considered as preferable. When the individuals have instead a different type of point group, the whole symmetry of the allotwin is represented by a Ehresmann point groupoid. The chromatic point group which accounts for the operations acting on the whole allotwinned edifice may in some cases bring little information: the point groupoid has to be given, for which however a chromatic symbol can often be obtained much like the symbol of a point group. The use of groupoids represents a unifying approach to the symmetry of allotwins and provides important information complementary to the relation between the lattice of the individuals and the allotwin lattice.

## Acknowledgements

I wish to thank Professor Bernd Souvignier (Radboud University Nijmegen, The Netherlands) and Dr Bertold Stöger (Technical University Vienna, Austria) for profitable discussions. Extensive critical remarks by three anonymous reviewers were extremely useful for the revision of the original manuscript and are here gratefully acknowledged.

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