Group-Theoretical Consideration of the CSL Symmetry

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Abstract

A theoretical analysis for the computation of the coincidence site lattice (CSL) symmetry is presented. It is shown that three types of symmetry elements can exist and each one can be found by properly using the CSL's rotation matrix of the smallest-angle description. Thus, from the existence of the subgroup \( H_1 \), the order of which is directly connected with the number of the different orientations that the sublattice \( A_{\Sigma} \) can have, a low-symmetry \( H_1 \) group implies more possibilities for the formation of the corresponding CSL. From the existence of the symmetry elements of the second type, the smallest-angle rotation matrix can be a symmetry element but only of the fourth or sixth order. From the third type of elements a connection between CSLs of different \( \Sigma \) values can exist. Since the analytical form of this smallest-angle rotation matrix can be deduced for every crystallographic system, the procedure described here is of general use. Thus a new classification of the different CSLs is possible according to their symmetry group. This allows the study of the CSL model from the symmetry point of view.

1. Introduction

In this paper we present results of a study of the CSL model, which is based on the theory established by Bleris & Delavignette (1981). According to this theory a rotation matrix, which produces the CSL describing a bicrystal, can be expressed as a function of the integral numbers \( m, n, \alpha \) and the rotation axis \([uvw]\). The algorithms for the calculation of these values have been recently given (Bleris, Karakostas & Delavignette, 1983) for any CSL of the cubic and hexagonal systems. With a complete classification of all CSLs according to the rotation matrix of their smallest-angle description, the question to be answered was 'how can we use this rotation matrix in order to define the crystallographic elements of a given CSL, i.e. the crystal system of the CSL and its base?'.

In this paper we shall give the group-theoretical information for the computation of the symmetry of a given CSL. The use of group theory for the study of the CSL was first introduced by Pond & Bollmann (1979), where the stability of a CSL boundary was examined by means of some selection rules based on the black and white symmetries. Later, Gratias & Portier (1982) presented an extension of Pond's idea. Neither of these works deals with the CSL symmetry. Thus, the only existing information for comparison with our results is the Bravais classes of the cubic CSLs, which have been determined by a method described by Mighell, Santoro & Donnay in International Tables for X-ray Crystallography (1969), and have been given for the CSLs up to \( \Sigma = 49 \) by Grimmer, Bollmann & Warrington (1974).

In the following paper (Bleris, Doni, Karakostas, Antonopoulos & Delavignette, 1985) we shall present the necessary analytical expressions for the CSL's base computation, using some of the results of the present work.

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2. Basic definitions

Two lattices $A_1$ and $A_2$, related by the equation

$$A_2 = RA_1,$$  \hspace{1cm} (1)

where $R$ is a rotation matrix, form a CSL $A_2$ if

$$A_{12} = A_1 \cap RA_1$$  \hspace{1cm} (2)

is not the empty set.

In the following the CSL, which is the common sublattice of the two lattices, will be considered as part of one of the two lattices only, which will be called 'the parent lattice'. Let us now suppose that $g$ is a symmetry operator of this sublattice. If we apply this operator to the parent lattice, the following possibilities only can exist:

(i) the sublattice and the parent lattice remain invariant;
(ii) the sublattice remains invariant but not the parent lattice;
(iii) a sublattice with a smaller $\Sigma$ value, which contains all the points of the first (higher $\Sigma$ value) sublattice, remains invariant, but not the parent lattice.

The above cases lead to the following obvious properties. Firstly, if we take into account the fact that every symmetry element may be a pure rotation or a combination of a rotation and the inversion operator $I$, we can say that, in the case of simple lattices, it is sufficient to deal with pure rotations only.

Secondly, the axis of the $g$ operator should be a vector of the sublattice and its angle can only have one of the allowable values $2\pi/2$, $2\pi/3$, $2\pi/4$ or $2\pi/6$. Furthermore, in case (i), $g$ is a symmetry element of the parent lattice as well. In case (ii), the new orientation of the parent lattice and the old one have a common sublattice and also $g$ is a CSL rotation, having a rotation angle equal to one of the previously mentioned values. In case (iii), $g$ should be a CSL rotation of the higher $\Sigma$ value sublattice and also a symmetry element of that with the smaller $\Sigma$ value. Moreover, in this case the unit volume of the former sublattice is a multiple of the unit volume of the latter sublattice. Since the smaller $\Sigma$ value sublattice has a unit volume that is a multiple of the unit volume of the parent lattice, we can conclude that the one with the higher $\Sigma$ value has a unit volume that is a composite multiple of the unit volume of the parent lattice.

Let us now examine (2). This relation implies that the existence of a CSL is equivalent to the existence of two sublattices $A_1^1$ and $A_1^2$ of $A_1$ such that

$$RA_1^1 = A_1^2.$$  \hspace{1cm} (3)

3. The elements of the first type (the subgroup $H_1$)

Let $G$ be the symmetry group of the parent lattice $A_1$ and $H_1$ the set of the elements of $G$ defined by the relation:

$$H_1 = \{g_i = R^{-1} g g: g_i A_1^1 = A_1^2, g_i, g \in G\}.$$  \hspace{1cm} (5)

Relation (4) expresses the one-to-one correspondence between the elements of these two groups; therefore, it suffices for our purpose to find the group $G_{A_1^1}$ only. Then the symmetry group $G_{A_1^2}$, i.e. the CSL's symmetry, is immediately obtained by the similarity transformation (4).

In the following sections we shall determine the symmetry elements of the three types classified above. Moreover, since we are dealing with matrices and their numerical properties, we choose a constant base, that of $A_1^1$, for their representation.
In fact, \( H_1 \) is a subgroup of \( G_{A_1} \) because

\[
\forall g_i \in H_1 \rightarrow g_i A_1 = R^{-1} g_i R A_1 = R^{-1} A_1^2 = A_1
\]

(6)

and also \( H_1 \) is a subgroup of \( G \).

It is well known that every group can be analysed as a coset sum of one of its subgroups (see for instance Bradley & Cracknell, 1972; Van Tendeloo & Amelinckx, 1974). By using this property, \( G \) can be written as a coset sum of \( H_1 \):

\[
G = g_1 H_1 + g_2 H_1 + \cdots + g_v H_1,
\]

(7)

where \( g_i \) (\( i = 1, 2, 3, \ldots, v \)) does not belong to \( H_1 \) and \( g_1 = E \) is the identity operator. The integer \( v \), named the index of \( H_1 \) in \( G \), is the ratio of the orders \([G]\) and \([H_1]\) of the groups \( G \) and \( H_1 \), respectively.

All of the cosets in (7) correspond to a variant, i.e. to a different orientation of the sublattice \( A_1 \) within lattice \( A_1 \), produced by the application on \( A_1 \) of one of the \( g_i \) elements that do not belong to \( H_1 \). There are \( v \) possible variants of \( A_1 \), the integer \( v \) being dependent on the order of \( H_1 \). The smaller the order of \( H_1 \) the larger the number of variants. As an example of the relation between the symmetry elements of the parent lattice \( A_1 \) and the variants of \( A_1 \), let us again consider the previously examined case of \( 2 = 5 \), in Fig. 1. If we consider a 90° rotation around the [001] axis, which is a symmetry element of \( A_1 \), it can be easily seen that this is also a symmetry element of \( A_1 \), that is this rotation is a symmetry element of \( H_1 \). On the contrary, if we consider a 180° rotation around the [110] axis, which is also a symmetry element of \( A_1 \), when this rotation is applied on \( A_1 \) it produces \( A_2 \), so that this rotation is one of the \( h_i \in H_1 \) elements.

Similarly, \( G_{A_1} \) can be analysed as

\[
G_{A_1} = h_1 H_1 + h_2 H_1 + \cdots + h_t H_1,
\]

(8)

where the \( h_i \notin H_1 \) elements are not symmetry elements of \( G \). Since \( h_i \) are symmetry elements of \( G_{A_1} \), they are of the second or third kind, according to our classification in § 2. An example of the second kind of operations is a 180° rotation around the [210] axis in the previously examined case of \( \Sigma = 5 \) (Fig. 1), which leaves \( A_1 \) invariant while it changes the orientation of \( A_1 \).

4. The elements of the second type (\( h_i \notin H_1 \))

As was pointed out before, the elements of the second type are CSL rotations. This means that \( h_i \) corresponds to one of the symmetrically equivalent descriptions of the same CSL discussed by Karakostas, Bleris & Antonopoulos (1979). By its definition every symmetrically equivalent description is a rotation connecting a variant of \( A_1 \) with \( A_1^2 \). Thus the general expression of the rotation matrix describing a CSL and operating on \( A_1 \) is

\[
R_i = g_i^{-1} R
\]

(9)

Let us consider the set of the elements \( h_i \) for which it holds that

\[
h_i = g_i^{-1} R \in G_{A_1}
\]

(10)

and \( g_i^{-1} \neq E \), the identity operator. Since \( h_i \) is an element of \( G_{A_1} \) and a CSL rotation as well, for a convenient \( t \), it follows that

\[
G_{A_1} \ni h_i^{-1} = g_i^{-1} R.
\]

(11)

Let us now consider an element \( h \in G_{A_1} \) to be constant. Then, for every other \( h_i \in G_{A_1} \), we have either

\[
h_i^{-1} h = R^{-1} g_i^{-1} R = g_k^{-1} R \in G_{A_1},
\]

(12a)

for a convenient \( k \), or

\[
h_i^{-1} h = R^{-1} g_i^{-1} R = g_i^{-1} \in G_{A_1},
\]

(12b)

for a convenient \( t \). Relation (12a) leads to \( R \in G \), which is impossible. On the other hand, from (12b) one can see that

\[
h_i^{-1} h \in H_1.
\]

(13)

Moreover, from (12b) it is obvious that

\[
\forall h_i \in G_{A_1} \rightarrow \exists g_i \in H_1 : h g_i = h_i
\]

(14)

and this means that all the symmetry elements \( h_i \) of (14) belong to the left coset \( h H_1 \). Since (12b) is unique for the existence of \( h_i \), we can conclude that

\[
H_1 + h H_1 = G_{A_1}
\]

(15)

and if there are no elements of the third type in \( G_{A_1} \) we have

\[
H_1 + h H_1 = G_{A_1}.
\]

(16)

From (16) it is obvious that \([G_{A_1}] / [H_1] = 2\), which means that \( H_1 \) is an invariant subgroup of \( G_{A_1} \) (see Bradley & Cracknell, 1972). The previous statement immediately implies that \( h H_1 = H_1 \) and therefore

\[
(h h_1)(h h_1) = h (h_1 h) h_1 = h^2 h_1 = H_1,
\]

(17)

which means that \( h^2 = E \) and so \( h \) is a 180° rotation. Let us examine the order \( p \) that \( h \) may have in (15). Supposing that \( p > 2 \), we have

\[
E = h . h . h \ldots h = g_1^{-1} R g_1^{-1} R \ldots g_1^{-1} R,
\]

(18)

where \( E \) is the identity operator. From (11) we have

\[
R g_1^{-1} R = g_1
\]

(19)

By substituting (19) into (18) \( p - 1 \) times, we have

\[
E = g_1^{-1} R g_1^{-1} g \ldots g_1^{-1} g,
\]

(20)

This relation cannot be true, since \( R \notin G \). Thus, if \( R \) is the smallest-angle rotation matrix of a given CSL,
then
\[ G_{A_1} \ni h = g^{-1} R, \text{if and only if } h^2 = E. \]

Finally, there remains the case
\[ h_t = g_t^{-1} R \quad \text{and} \quad g_t^{-1} = E. \]

In this case (11) is still valid and therefore
\[ h_t^{-1} = R^{-1} = g_t^{-1} R \]
for an appropriate \( g_t \in G \). From this relation we have
\[ R^2 = g_t \]
and, since \( R \in G_{A_1} \), the element \( R^2 \) belongs to \( G_{A_1} \) and \( g_t \in H_1 \). Thus, the smallest-angle rotation matrix \( R \) can be a symmetry element if its square is an element of \( H_1 \).

In the case where \( R \) is an element of \( G_{A_1} \), we also form the product
\[ h_t^{-1} R = R^{-1} g_t R, \quad \forall h_t \in G_{A_1} \]
and
\[ h_t \neq R, h_t \neq R^{-1}. \]

From (24), following the procedure used for (12), it can be easily proved that
\[ h_t^{-1} R = R^{-1} g_t R = g_m^{-1} \in H_1. \]

Relation (25) can be rewritten as
\[ h_t^{-1} R = h_t R = g_m^{-1} = g_t^{-1} R R = g_t^{-1} R^2 = g_t^{-1} g_t. \]

But \( g_t \in H_1 \), so it is obvious that \( g_t^{-1} \in H_1 \). Moreover, (25) implies that
\[ \forall h_t \in G_{A_1} \rightarrow \exists g_m \in H_1: h_t = R g_m \in R H_1, \]
which means that all the symmetry elements \( h_t \) belong to the left coset \( RH_1 \) of \( H_1 \) in \( G_{A_1} \). From (25) it is also obvious that
\[ \forall h_t \in G_{A_1} \rightarrow \exists g_k \in H_1: R = h_t g_k \in h_t H_1, \]
which means that \( R \) is among the elements of \( h_t H_1 \). The same is true for \( R^{-1} \). This can be possible only if \( H_1 \) contains a number of elements equal to the number of 180° rotations that it has plus two. So, when \( R \in G_{A_1} \), \( H_1 \) contains two more elements, besides the \( g_t \) element, which are given by (23).

As we have mentioned before, \( R \), being a symmetry element, can be a rotation of the third, fourth or sixth order. If \( R \) is of the third order, then necessarily \( R^3 = E \) and because of (23) we have \( R = g_t^{-1} \). This is impossible and we may conclude that \( R \) can only be of the fourth or sixth order.

5. The elements of the third type

As has been pointed out above (§ 2), this case can only exist if \( \Sigma \) is a composite integer. Let us now suppose that \( \Sigma = pq \), where \( p, q \) are prime integers. Then, a symmetry element of the third type for the CSL with volume \( \Sigma \) can be found from the symmetry elements of the CSLs with volume either \( \Sigma = p \) or \( \Sigma = q \).

For the symmetry element \( R_p \) (or \( R_q \)) to be a symmetry element of the CSL with volume \( \Sigma \), in both cases the following conditions are necessary:

(i) \( R_p \) is not a symmetry element of \( G \). Then \( R_p \) is either a 180° rotation or one of the exceptional cases of 90° or 60° rotations.

(ii) The axis \( r \) of \( R_p \) is a vector of the exceptional cases. This condition can be easily checked by taking the transformation \( R_p^2 \), where \( R_p \) is the smallest-angle rotation matrix of the CSL with volume \( \Sigma \).

(iii) The product \( R_p^{-1} R_p \) gives a rotation matrix that describes a CSL with volume \( p \).

If such an element exists, then the symmetry group \( G_{A_1} \) of this composite CSL can be obtained by joining this element to the group of the elements that have been found by the steps of §§ 3 and 4.

6. Practical procedure for the construction of \( G_{A_1} \)

In this section we shall develop the practical procedure for the construction of \( G_{A_1} \), taking into account the previously established properties of the symmetry group \( G_{A_1} \).

Let \( G \) be the symmetry group of the parent lattice. Since we are dealing with simple lattices, \( G \) has the form
\[ G = G_0 + i G_0, \]
where \( I \) is the inversion operator and \( G_0 \) is a subgroup of \( G \) containing only pure rotations. As we have mentioned before, we shall use the subgroup \( G_0 \). For the cubic system \( G_0 \) is the \( O (432) \) group containing 24 elements (Karakostas et al., 1979), and for the hexagonal system \( G_0 \) is the \( D_6 (622) \) group containing 12 elements (Hagège, Nouet & Delavignette, 1980).

Let \( R \) be the rotation matrix that gives a CSL with multiplicity \( \Sigma \) when it operates on \( \Lambda_1 \). We compute the products
\[ (a) \quad R^{-1} g R, \quad (b) \quad g^{-1} R \]
\[ \forall g \in G_0. \]

Relation (29a) gives the subgroup \( H_1 \) while (29b) gives the set of the different descriptions of the same CSL.

We can easily see that the order of \( H_1 \), i.e. the number of the symmetry elements \( R^{-1} g R \), can never be equal to the order of \( G_0 \). (Otherwise all the elements of \( G_0 \) would be symmetry elements of \( \Lambda_1 \) and there would be no variants of \( \Lambda_1 \), so that all the \( g^{-1} R \) rotations would describe one and the same geometrical operation.) On the other hand, the number of 180° rotations from the set of the \( g^{-1} R \) elements should be smaller or equal to one half of the order of \( G_0 \). This happens because, as we have seen before,
Table 1. Possible CSL symmetry according to the number of 180° rotations

<table>
<thead>
<tr>
<th>Number of 180° rotations</th>
<th>Σ prime</th>
<th>Σ composite</th>
</tr>
</thead>
<tbody>
<tr>
<td>None</td>
<td>Triclinic</td>
<td>Triclinic or higher</td>
</tr>
<tr>
<td>1</td>
<td>Monoclinic</td>
<td>Monoclinic or higher</td>
</tr>
<tr>
<td>2</td>
<td>Orthorhombic</td>
<td>Orthorhombic or higher</td>
</tr>
<tr>
<td>3</td>
<td>Rhombohedral</td>
<td>Rhombohedral or higher</td>
</tr>
<tr>
<td>4</td>
<td>Tetragonal</td>
<td>Tetragonal</td>
</tr>
<tr>
<td>5</td>
<td>Impossible, because there are no subgroups of order 5</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Hexagonal</td>
<td>Hexagonal or cubic</td>
</tr>
</tbody>
</table>

if there exists a set of 180° rotations, then the number of the elements of this set is equal to the number of the elements of \( hH_1 \). This number is equal to the number of the elements of \( hH_1 \) minus 2 in the special cases where \( R \) is an element of \( G_{AI} \). Taking into account that in all cases the symmetry elements of \( G_{AI} \) that do not belong to \( G \) are included in the products \( hH_1 \), we can write

\[
[H_1 + hH_1] = [G_0]
\]

and

\[
[H_1 + hH_1] = 2[H_1] = [G_0] \rightarrow [H_1] = [G_0]/2.
\]

The possibilities existing for the symmetry of the sublattice \( A_1 \) are shown in the following diagram, holding for both cubic and hexagonal systems.

Cubic O(432)       Hexagonal D₆(622)
Rhombohedral        \( D₃(32) \) : \( [D₃] = 6 \)
Tetragonal          \( D₂(422) \) : \( [D₂] = 8 \)
Orthorhombic        \( D₂(222) \) : \( [D₂] = 4 \)
Monoclinic          \( C₃(2) \) : \( [C₃] = 2 \)
Triclinic           \( C₁(1) \) : \( [C₁] = 1 \).

Taking this diagram into account, we have classified all the possibilities as a function of the existing 180° rotations in Table 1.

In all the above cases one must also take into account the possibility of \( R \) being a symmetry element of the CSL. In this case the symmetry of the CSL does not follow the rules of Table 1, but it is characterized by the order of \( R \).

In what follows, we shall treat two typical examples of constructing \( G_{AI} \) one of a composite \( \Sigma \)-value CSL and the case \( \Sigma = 2 \) CSL of the hexagonal system. Moreover, tables have been constructed for CSLs up to \( \Sigma = 49 \) for both cubic and hexagonal (\( \mu = \nu = 1; \mu = 5, \nu = 2; \mu = 8, \nu = 3 \)) systems. These tables present the step by step procedure for the computation of \( G_{AI} \) of every CSL.*

Cubic system: \( \Sigma = 15, [uvw] = [210], \theta = 48.18° \).

In this CSL the application of (29) gives the subgroup \( H_1 \), which contains only the identity operator and one element of the 180° type. This element corresponds to a 180° rotation around the [125] axis and it is in a matrix form:

\[
h = \frac{1}{15} \begin{bmatrix} 14 & 2 & 5 \\ 2 & 11 & 10 \\ 5 & 10 & 10 \end{bmatrix}.
\]

So the subgroup \( H_1 + hH_1 \) is

\[
H_1 + hH_1 = \{E\} + h\{E\} = \{E, h\}.
\]

Since the volume of this CSL is a composite number, we have to look for symmetry elements of the third type before the final classification. But the CSL rotation around the [210] axis of \( \Sigma = 15 \) corresponds to a symmetry element of 180°, because a symmetrical equivalent of this axis is among the equivalent descriptions of \( \Sigma = 5 \), corresponding to a 180° symmetry element. So, the 180° rotation around the [210] axis of \( \Sigma = 5 \) is a symmetry element of the third type of \( \Sigma = 15 \) and \( G_{AI} \) is constructed by joining this element, given in a matrix representation with the form

\[
R_5 = \frac{1}{5} \begin{bmatrix} 3 & 4 & 0 \\ 4 & 3 & 0 \\ 0 & 0 & 5 \end{bmatrix},
\]

to the subgroup \( H_1 + hH_1 \) according to the following relation:

\[
G_{AI} = \{E, h\} + R_5\{E, h\}.
\]

The element \( R_5h \) is in a matrix form:

\[
R_5h = hR_5 = \frac{1}{3} \begin{bmatrix} 2 & 2 & 1 \\ 2 & 1 & 2 \\ 1 & 2 & 2 \end{bmatrix} = R_3
\]

and corresponds to a 180° rotation around the [121] axis of \( \Sigma = 3 \), which is also a symmetry element of the third type of \( \Sigma = 15 \). It can be easily checked that the [121] axis is an axis of the CSL \( \Sigma = 15 \).

So, \( G_{AI} \), containing the identity operator \( E \) and three other elements corresponding to 180° rotations is isomorphic to the \( D₂ \) orthorhombic symmetry group.

Hexagonal system: \( \Sigma = 2, [uvw] = [210], \theta = 90° (\mu = \nu = 1) \).

By using (29) we obtain

\[
H_1 = \{E, C₂, C'₂, C''₂\}
\]

and two rotations of 180°. Taking into account that \( R \) is of the fourth order, we have a typical example of the case discussed in § 4. The different powers of

* Supplementary material giving the results of the application of the theory has been deposited with the British Library Lending Division as Supplementary Publication No. SUP 42161 (15pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.
$R$ have the following matrix form:

\[
R = \frac{1}{2} \begin{bmatrix} 2 & 0 & 0 \\ 1 & 0 & 2 \\ 1 & 2 & 0 \end{bmatrix}, \quad R^2 = \begin{bmatrix} 1 & 0 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix},
\]

\[
R^3 = \frac{1}{2} \begin{bmatrix} 2 & 0 & 0 \\ 1 & 0 & 2 \\ 2 & 1 & 0 \end{bmatrix}, \quad R^4 = E.
\]

$R^2$ is the $C_{22} \in H_1$ element and we may construct $G_{A_1}$ by making use of the $C_2 \in H_1$ element. Therefore,

\[
G_{A_1} = \{ R, R^2, R^3, R^4 \} + C_2 \{ R, R^2, R^3, R^4 \}
\]

\[
= \{ E, C_{22}, C_{22}', C_{2}, R, R^{-1}, C_2 R, C_2 R^3 \}
\]

and $G_{A_1}$ is isomorphic to the $D_4$ tetragonal symmetry group.

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**References**