

## Disorientations and Coincidence Rotations for Cubic Lattices

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As a result of the cubic symmetry operations there are up to 1152 rotations describing the same relative orientation of two cubic lattices. The relation between these equivalent rotations is made transparent and it is shown how the usual definition of the disorientation has to be modified so that, in each case, the definition picks out a unique rotation among all the equivalent ones. A convenient method is described for determining all the classes of rotation that lead to coincidence-site lattices of a given density and for finding the number of rotations in each class.

### Introduction

This paper considers questions that arise in studying the statistical distribution of the relative orientation of neighbouring crystals. In particular, it deals with the description of the relative orientation of two cubic lattices, 1 and 2. The paper consists of two parts. In the first part, the two lattices may have different lattice constants and may belong to different cubic Bravais classes. We describe the various rotations by which lattice 2 may have reached the position under consideration starting from a position where its four-fold axes were parallel to those of lattice 1. Our description gives a transparent view of all the rotations with this property. We show that the disorientation, as usually defined, is not always determined uniquely by the relative orientation; a minor change in the definition makes the disorientation unique.

In the second part, we specialize to two equal lattices related by a coincidence rotation, *i.e.* a rotation giving rise to a three-dimensional pattern of coincidence sites. These sites form the coincidence-site lattice or CSL. [For a discussion of the importance of the CSL to interpret experimental results, we refer the reader to Grimmer, Bollmann & Warrington (1974) and references quoted therein.] We give a convenient method for determining all the relative orientations and, hence, all the disorientations that give rise to a large density of coincidence sites. For each disorientation we obtain the number of rotations that give rise to the same relative orientation of two lattices.

The reader should not be discouraged by the fact that we state some of our results as theorems and lemmas. These terms do not mean that we shall use highbrow mathematics; they are introduced only to emphasize the results that become important further on.

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### 1. Two cubic lattices with arbitrary relative orientation

#### 1.1 *The description of the relative orientation of two cubic lattices*

A right-handed orthogonal coordinate system with axes in  $\langle 100 \rangle$  type directions of a cubic lattice can be chosen in 24 different ways. Consider two cubic lattices. Their relative orientation can be described by any of the rotations that map a coordinate system  $C_1$  of one of the lattices onto a coordinate system  $C_2$  of the other. In this way, we obtain up to  $24 \cdot 24 \cdot 2 = 1152$  different descriptions of the same relative orientation of two cubic lattices. Rotations describing the same relative orientation of two cubic lattices will be called (cubically) equivalent.

The lattice connected with  $C_1$  will be called lattice 1, the other lattice 2. The rotation that maps  $C_1$  onto  $C_2$  is described by a matrix  $\mathbf{R}$  in the coordinate system  $C_1$ . Consider new coordinate systems  $C'_1$  and  $C'_2$ . Expressed in  $C_1$ , the mapping  $C'_1 \rightarrow C_1$  has the form  $\mathbf{S}$  and the mapping  $C_2 \rightarrow C'_2$  the form  $\mathbf{RTR}^{-1}$ , where  $\mathbf{S}$  and  $\mathbf{T}$  are symmetry rotations of lattice 1. The mapping  $C'_1 \rightarrow C'_2$  becomes then  $\mathbf{RTR}^{-1}$ .  $\mathbf{R} \cdot \mathbf{S} = \mathbf{RTS}$  in system  $C_1$  and  $\mathbf{S} \cdot \mathbf{RTS} \cdot \mathbf{S}^{-1} = \mathbf{SRT}$  in system  $C'_1$ . Taking into account that either of the two lattices may take over the role of lattice 1, we conclude that cubic equivalence can be defined as follows:

*Definition.* Two rotation matrices  $\mathbf{R}, \mathbf{R}'$  are called (cubically) equivalent if and only if

$$\mathbf{R}' = \mathbf{SRT} \quad \text{or} \quad \mathbf{R}' = \mathbf{SR}^{-1}\mathbf{T}, \quad (1)$$

where  $\mathbf{S}$  and  $\mathbf{T}$  describe cubic symmetry rotations.

#### 1.2 *The homomorphism between unit quaternions and rotations*

In order to gain a synopsis of a class of cubically equivalent rotations, it is convenient to make use of the two-to-one homomorphism between the group  $Q$  of unit quaternions and the rotation group  $SO(3)$  (see,

*e.g.*, Du Val, 1964). A unit quaternion  $\mathbf{a}$  is an ordered set of four real numbers,

$$\mathbf{a} := \{a_0, a_1, a_2, a_3\}$$

satisfying

$$a_0^2 + a_1^2 + a_2^2 + a_3^2 = 1. \quad (2)$$

The unit quaternions form a group  $Q$  with respect to the multiplication law

$$\begin{aligned} \mathbf{a} \cdot \mathbf{a}' = \{ & a_0 a_0' - a_1 a_1' - a_2 a_2' - a_3 a_3', \\ & a_0 a_1' + a_1 a_0' + a_2 a_3' - a_3 a_2', \\ & a_0 a_2' - a_1 a_3' + a_2 a_0' + a_3 a_1', \\ & a_0 a_3' + a_1 a_2' - a_2 a_1' + a_3 a_0' \}. \end{aligned} \quad (3)$$

The homomorphism associates with  $\pm \mathbf{a}$  the rotation

$$\begin{pmatrix} a_0^2 + a_1^2 - a_2^2 - a_3^2 & 2(a_1 a_2 - a_0 a_3) & 2(a_1 a_3 + a_0 a_2) \\ 2(a_1 a_2 + a_0 a_3) & a_0^2 - a_1^2 + a_2^2 - a_3^2 & 2(a_2 a_3 - a_0 a_1) \\ 2(a_1 a_3 - a_0 a_2) & 2(a_2 a_3 + a_0 a_1) & a_0^2 - a_1^2 - a_2^2 + a_3^2 \end{pmatrix}. \quad (4)$$

The quaternions associated with a right-handed rotation by an angle  $\theta$ ,  $0 \leq \theta \leq \pi$ , about an axis with direction cosines  $n_1, n_2, n_3$  can be found from (4) if we recall the matrix describing this rotation:

$$\begin{pmatrix} (1 - \cos \theta)n_1^2 + \cos \theta & (1 - \cos \theta)n_1 n_2 - n_3 \sin \theta \\ (1 - \cos \theta)n_1 n_2 + n_3 \sin \theta & (1 - \cos \theta)n_2^2 + \cos \theta \\ (1 - \cos \theta)n_1 n_3 - n_2 \sin \theta & (1 - \cos \theta)n_2 n_3 + n_1 \sin \theta \\ & (1 - \cos \theta)n_1 n_3 + n_2 \sin \theta \\ & (1 - \cos \theta)n_2 n_3 - n_1 \sin \theta \\ & (1 - \cos \theta)n_3^2 + \cos \theta \end{pmatrix} \quad (5a)$$

$$\Leftrightarrow \pm \{ \cos \frac{1}{2}\theta, n_1 \sin \frac{1}{2}\theta, n_2 \sin \frac{1}{2}\theta, n_3 \sin \frac{1}{2}\theta \}. \quad (5b)$$

(5b) shows that  $\pm \mathbf{a}$  determines a right-handed rotation by an angle

$$\theta = 2 \arccos |a_0|, \quad 0 \leq \theta \leq \pi, \quad (6a)$$

about the axis

$$[n_1, n_2, n_3] = \text{sign}(a_0) (1 - a_0^2)^{-1/2} [a_1, a_2, a_3], \quad (6b)$$

where  $\text{sign}(a_0)$  is  $+1$  for  $a_0 \geq 0$  and  $-1$  for  $a_0 \leq 0$ .

It is a straightforward if somewhat tedious matter to derive from (1) and (5b) the connexion between cubically equivalent unit quaternions, *i.e.* between the quaternions corresponding to cubically equivalent rotations. The result is:

**Theorem 1.** The quaternions that are cubically equivalent to  $\pm \mathbf{a}$  are obtained by arbitrary permutations and sign changes from one of the following six expressions

$$\{a_0, a_1, a_2, a_3\} (= \mathbf{a}) \quad (7a)$$

$$2^{-1/2} \{a_0 + a_1, a_0 - a_1, a_2 + a_3, a_2 - a_3\} \quad (7b)$$

$$2^{-1/2} \{a_0 + a_2, a_0 - a_2, a_1 + a_3, a_1 - a_3\} \quad (7c)$$

$$2^{-1/2} \{a_0 + a_3, a_0 - a_3, a_1 + a_2, a_1 - a_2\} \quad (7d)$$

$$\frac{1}{2} \{a_0 + a_1 + a_2 + a_3, a_0 + a_1 - a_2 - a_3, a_0 - a_1 + a_2 - a_3, a_0 - a_1 - a_2 + a_3\} \quad (7e)$$

$$\frac{1}{2} \{a_0 + a_1 + a_2 - a_3, a_0 + a_1 - a_2 + a_3, a_0 - a_1 + a_2 + a_3, a_0 - a_1 - a_2 - a_3\}. \quad (7f)$$

Let us discuss this result. The rotation angles appearing in a cubic equivalence class are given by:  $\theta = 2 \cos^{-1} |b_0|$ , where  $b_0$  is one of the 24 numbers that appear in (7), *i.e.*  $a_0, a_1, a_2, a_3, 2^{-1/2}(a_0 + a_1), \dots$ . Consider a fixed choice of  $\theta$ : the remaining three numbers on the same line of (7) determine the possible rotation axes connected with  $\theta$ . As an example, choose  $b_0 = 2^{-1/2}(a_0 - a_1)$ . Each of the corresponding rotation axes is parallel to one of the directions obtained from  $[a_0 + a_1, a_2 + a_3, a_2 - a_3]$  by arbitrary permutations and sign changes of the three components.

### 1.3 Disorientations

Since cubically equivalent rotations describe the same superposition of two cubic lattices, it will often be convenient to make a particular choice of the rotation describing the superposition. In the cubic equivalence class under consideration, we choose a rotation with the smallest rotation angle  $\theta$ . Theorem 1 tells us that there is always such a rotation with an axis  $[n_1, n_2, n_3]$  in the standard stereographic triangle (SST), *i.e.* with  $n_1 \geq n_2 \geq n_3 \geq 0$ . Rotations with the smallest angle and an axis in the SST are called disorientations (see, *e.g.* Handscomb, 1958). The axis of  $\pm \mathbf{a}$  with  $a_0 > 0$  lies in the SST if  $a_1 \geq a_2 \geq a_3 \geq 0$ . According to Theorem 1, the condition that  $\theta$  be smallest is  $a_0 \geq 2^{-1/2}(a_0 + a_1)$  and  $a_0 \geq \frac{1}{2}(a_0 + a_1 + a_2 + a_3)$ .

**Definition.** A rotation  $\pm \mathbf{a}$  is called a disorientation if

$$a_1 \geq a_2 \geq a_3 \geq 0, \quad (8a)$$

$$a_1 \leq (\sqrt{2} - 1)a_0, \quad (8b)$$

$$a_1 + a_2 + a_3 \leq a_0. \quad (8c)$$

If the equality sign does not hold in (8b, 8c), the disorientation is determined uniquely. If the equality sign holds in (8c) only, the expression (7e) will coincide with  $\mathbf{a}$  and we still have a unique disorientation. However, if the equality sign holds in (8b), then (7b) will give

$$\{a_0, a_1, 2^{-1/2}(a_2 + a_3), 2^{-1/2}(a_2 - a_3)\},$$

which coincides with (7a) only if  $a_3 = (\sqrt{2} - 1)a_2$ . We conclude: if  $\mathbf{a}$  is a disorientation then it will be the unique disorientation in its class unless  $a_1 = (\sqrt{2} - 1)a_0$  and  $a_3 \neq (\sqrt{2} - 1)a_2$ .

Consider for a moment all rotations, not just a cubic equivalence class. Equations (2) and  $a_1 = (\sqrt{2} - 1)a_0$  give for the maximal disorientation angle  $\theta_m$  about the axis  $[l m n]$  in the SST:

$$\text{tg}(\frac{1}{2}\theta_m) = [(3 - 2\sqrt{2})(1 + m^2 + n^2)]^{1/2}, \quad (9a)$$

whereas equations (2) and  $a_1 + a_2 + a_3 = a_0$  give

$$\text{tg}(\frac{1}{2}\theta_m) = (1 + m^2 + n^2)^{1/2} (1 + m + n)^{-1}. \quad (9b)$$

These results are illustrated in Fig. 1.  $\theta_m$  is determined by (9a) below the dotted line and by (9b) above. The disorientation becomes unique if we require its axis not to lie below the dashed line if  $\theta = \theta_m$ .

**2. Two equal cubic lattices in coincidence orientation**

**2.1 The cubic equivalence classes of coincidence rotations**

In the following, we shall consider the special case that the unit quaternion has commensurable components, *i.e.* that all the components are integral multiples of one and the same number. Such a unit quaternion can be written in the form  $\mathbf{a} = c\{A_0, A_1, A_2, A_3\}$ , where the  $A_i$  are coprime integers (*i.e.* integers that have no common integral factor) and  $c$  is a constant multiplying each  $A_i$ . Since at least one of the numbers  $A_i$  is odd,  $A_0^2 + A_1^2 + A_2^2 + A_3^2$  cannot be a multiple of 8. We conclude that a commensurable unit quaternion has the form

$$\mathbf{a} = (N\sigma)^{-1/2}\{A_0, A_1, A_2, A_3\}, \tag{10}$$

where  $\sigma$  is odd,  $N = 1, 2, \text{ or } 4$ , and the  $A_i$  are coprime integers with  $A_0^2 + A_1^2 + A_2^2 + A_3^2 = N\sigma$ .

In this case, equation (4) gives a rotation matrix with rational matrix elements. The smallest common denominator (SCD) of the matrix elements is a factor of  $N\sigma$  that we call  $\sigma'$ . Denote the elements of the rotation matrix by  $r_{ij}$  then  $q_{ij} := \sigma' \cdot r_{ij}$  is an integer. The equations  $q_{i1}^2 + q_{i2}^2 + q_{i3}^2 = \sigma'^2$  for  $i = 1, 2, 3$  show that  $\sigma'$  is odd. (If  $\sigma'$  was even,  $\sigma'^2$  would be a multiple of 4, which is possible only if all the  $q_{ij}$  are even. But then  $\sigma'$  is not the SCD.) It follows from equations (3,4) that

$$\begin{aligned} 4a_0^2 &= 1 + r_{11} + r_{22} + r_{33} \\ 4a_1^2 &= 1 + r_{11} - r_{22} - r_{33} \\ 4a_2^2 &= 1 - r_{11} + r_{22} - r_{33} \\ 4a_3^2 &= 1 - r_{11} - r_{22} + r_{33} \end{aligned}$$

These equations tell us that  $N\sigma$  is a factor of  $4\sigma'$ , whence  $\sigma' = \sigma$ . It is not difficult to show that the matrix (4) does not have rational matrix elements if the corresponding quaternion does not have commensurable components. We conclude:

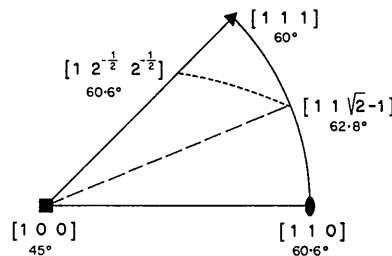


Fig. 1. The maximum disorientation angles depending on the direction of the rotation axis within the standard stereographic triangle. -----  $a_0 = (\sqrt{2}-1)^{-1}a_1 = a_1 + a_2 + a_3 \Rightarrow a_2 + a_3 = \sqrt{2}a_1$ . — — — — —  $a_3 = (\sqrt{2}-1)a_2$ .

*Lemma 1.* Each rotation of which the matrix elements have  $\sigma$  as SCD corresponds (one-to-one) to a pair of quaternions of the form

$$\pm (N\sigma)^{-1/2}\{A_0, A_1, A_2, A_3\},$$

with  $N = 1, 2, \text{ or } 4$  and the coprime integers  $A_i$  satisfying  $A_0^2 + A_1^2 + A_2^2 + A_3^2 = N\sigma$ .

A rotation with matrix elements having  $\sigma$  as SCD will be called a  $\sigma$  rotation. It has been shown by Grimmer, Bollmann & Warrington (1974) that a  $\sigma$  rotation applied to a cubic lattice produces a coincidence-site lattice with primitive unit cell  $\sigma$  times larger in volume than the primitive unit cell of the original lattice. Therefore,  $\sigma$  rotations will also be called *coincidence rotations*.

*Lemma 2.* To each  $\sigma$  rotation there is a cubically equivalent rotation given by  $\pm \sigma^{-1/2}\{B_0, B_1, B_2, B_3\}$ , where the  $B_i$  are coprime integers satisfying  $B_0 \geq B_1 \geq B_2 \geq B_3 \geq 0$ .

To prove this, let  $\pm \mathbf{a} = \pm (\bar{N}\sigma)^{-1/2}\{A_0, A_1, A_2, A_3\}$  be the quaternions of the  $\sigma$  rotation under consideration. If  $\bar{N} = 1$ , Lemma 2 follows immediately from Theorem 1. If  $\bar{N} = 2$ , two of the  $A_i$  are odd and two even. One of the quaternions (7b-7d) has the form (10) with  $N = 1$ , the other two with  $N = 4$ . If  $\bar{N} = 4$ , all the  $A_i$  are odd and (7e) has the form (10) with  $N = 1$ .

Take for (7a) one of the quaternions of Lemma 2. The expressions (7b-7d) then have the form (10) with  $N = 2$  and (7e, 7f) the form (10) with  $N = 4$ . It follows that the  $B_i$  of Lemma 2 are determined uniquely and we have:

*Theorem 2.* To find the cubic equivalence classes of  $\sigma$  rotations, we have to determine the different ways to express  $\sigma$  as

$$\sigma = B_0^2 + B_1^2 + B_2^2 + B_3^2,$$

where the  $B_i$  are coprime integers satisfying

$$B_0 \geq B_1 \geq B_2 \geq B_3 \geq 0.$$

We shall use the symbol  $[B_0, B_1, B_2, B_3]$  to denote a cubic equivalence class of coincidence rotations.

Example: there are two classes of rotation with  $\sigma = 21$ ; they correspond to  $21 = 4^2 + 2^2 + 1^2 + 0^2 = 3^2 + 2^2 + 2^2 + 2^2$ .

**2.2 The number of coincidence rotations**

To determine the total number of  $\sigma$  rotations, we make use of a number-theoretical result. Let  $S(\sigma)$  be the sum of the divisors of the odd number  $\sigma$ , *e.g.*  $S(21) = 1 + 3 + 7 + 21 = 32$ . The number of different quadruples  $\mathbf{A} = \{A_0, A_1, A_2, A_3\}$  of (not necessarily coprime) integers satisfying  $A_0^2 + A_1^2 + A_2^2 + A_3^2 = 2^n\sigma$  equals  $8 S(\sigma)$  if  $n = 0$  and  $24 S(\sigma)$  if  $n$  is a positive integer. This result is due to Jacobi (1828). If the different factors that appear more than once in the decomposition of  $\sigma$  as a product of primes are  $p_1, \dots, p_r$ , we shall write  $\sigma = p_1^2 \dots p_r^2 \cdot q$ . It follows from Jacobi's result that the number  $T(2^n\sigma)$  of different

quadruples  $\mathbf{A}$  of integers that are coprime and satisfy  $A_0^2 + A_1^2 + A_2^2 + A_3^2 = 2^n \sigma$  is given by

$$T(\sigma) = 8W(\sigma), \quad T(2\sigma) = 24W(\sigma), \quad T(4\sigma) = 16W(\sigma),$$

$$T(2^n \sigma) = 0 \quad \text{if } n > 2,$$

where

$$W(\sigma) = S(\sigma) - \sum_{1 \leq i \leq r} S(p_i^{-2} \cdot \sigma) + \dots$$

$$+ (-1)^n \sum_{1 \leq i_1 < \dots < i_n \leq r} S(p_{i_1}^{-2} \dots p_{i_n}^{-2} \sigma) + \dots$$

$$+ (-1)^r S(q).$$

Since each  $\sigma$  rotation corresponds to a pair  $\pm \mathbf{A}$ , the total number of  $\sigma$  rotations is  $\frac{1}{2}(8 + 24 + 16)W(\sigma) = 24W(\sigma)$ .

To determine the number of rotations in the cubic equivalence class  $[A, B, C, D]$ , we first compute the number  $\nu$  of different quadruples obtained from  $\{A, B, C, D\}$  by permutations and sign changes. We write  $\nu = 8W$ , call  $W$  the 'weight' of the class, and list  $W$  in Table 1.

Table 1. *The weight  $W$  of the cubic equivalence class  $[A, B, C, D]$*

No other cases than those listed are possible because either one or three of the numbers  $A, B, C, D$  are odd, the others even.

$[A, B, C, D]$	$W$	
$A > B > C > D > 0$	48	(11a)
$D > 0$ , two numbers equal	24	(11b)
$A > B > C > (D=0)$	24	(11c)
$C > (D=0)$ , two numbers equal	12	(11d)
$D > 0$ , three numbers equal	8	(11e)
$A > B > (C=D=0)$	6	(11f)
$(A=B=C) > (D=0)$	4	(11g)
$A > (B=C=D=0)$	1	(11h)

It is easy to check whether we have found all classes of  $\sigma$  rotations: their weights must add up to  $W(\sigma)$ . Example:  $W(21) = 32$  whereas the two classes of rotation with  $\sigma = 21$  have weights 24 and 8.

Examining the expressions (7) for the various cases (11) and writing the quaternions in the form (10), one finds that a class containing  $\nu$  quaternions with  $N=1$  also contains  $3\nu$  quaternions with  $N=2$  and  $2\nu$  quaternions with  $N=4$ . We conclude that the number of rotations in a cubic equivalence class equals  $\frac{1}{2} \cdot 6\nu = 24W$ .

For a discussion of what  $W$  tells us about the symmetry of the coincidence-site lattice, we refer the reader to Grimmer (1973). In that paper it is also shown that, unless  $W=48$ , we can obtain twins by appropriately choosing the interface between the two crystals in coincidence orientation.

### 2.3 Coincidence disorientations

The following procedure to find the disorientation in the class  $[A, B, C, D]$  of coincidence rotations follows immediately from Theorem 1. We have to distinguish three cases according to which of the numbers  $2A, \sqrt{2}(A+B), A+B+C+D$  is the largest (Table 2).

Table 2. *The coincidence disorientation*

Largest number	Disorientation
$2A$	$\sigma^{-1/2}\{A, B, C, D\}$
$\sqrt{2}(A+B)$	$(2\sigma)^{-1/2}\{A+B, A-B, C+D, C-D\}$ reordered according to decreasing values
$A+B+C+D$	$(4\sigma)^{-1/2}\{A+B+C+D, A+B-C-D,$ $A-B+C-D,  A-B-C+D \}$

Each disorientation is given by a point in the disorientation region (DR) indicated in Fig. 1. Since  $a_1 = (\sqrt{2}-1)a_0$  requires  $a_1/a_0$  to be irrational, it follows for coincidence rotations that the disorientation is determined uniquely and that  $\theta = \theta_m$  is possible only if the axis lies above the dotted line of Fig. 1. An equivalence class of coincidence rotations determines a disorientation and, therefore a point in the DR. If from this point the DR appears under a solid angle  $4\pi f$ , the weight  $W$  of the class will be  $48f$ . For  $W=48$ , the disorientation lies in the interior of the DR, for  $W=24$  on a surface, for  $W=12, 8$ , or  $6$  on an edge, and for  $W=4$  or  $1$  on a vertex of the DR. We list the various possibilities in Table 3.

Table 3. *Axis  $[hkl]$  and angle  $\theta$  of the disorientation that corresponds to a class of coincidence rotations of weight  $W$*

$W$	Axis	Angle	$W$	Axis	Angle
48	$h > k > l > 0$	$\theta < \theta_m$	12	$h > (k=l) > 0$	$\theta = \theta_m$
24	$h > k > (l=0)$	$\theta < \theta_m$	8	[111]	$\theta < \theta_m$
	$(h=k) > l > 0$	$\theta < \theta_m$	6	[100]	$\theta < \theta_m$
	$h > (k=l) > 0$	$\theta < \theta_m$	4	[111]	$\theta = \theta_m$
12	$h > k > l > 0$	$\theta = \theta_m$	1		$\theta = 0$
	[110]	$\theta < \theta_m$			
	$(h=k) > l > 0$	$\theta = \theta_m$			

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