Coincidence sites for rational lattices

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Abstract. The problem of identifying the coincidence site lattices produced by the disorientations of any rational lattice is investigated. A very simple new procedure for computing the CSLs is presented, based on Grimmer's reciprocity theorem and application of the reduction algorithm introduced by Buerger for finding reduced cells. The point symmetries of a lattice imply that any orientation relationship that produces a CSL can be alternatively expressed by a variety of different rotations. Two simple numerical methods (one based on integral matrix operations and one based on quaternions) are demonstrated for finding all the equivalent rotations that produce the same CSL and for obtaining a single 'canonical' rotation as a representative of an equivalence class.

1. Introduction

The concept of a *coincidence-site lattice* arose from the need to understand the structure of grain boundaries in polycrystalline materials. Those boundaries are energetically preferred for which there is a 'good fit' between the structures to either side. A 'good fit' corresponds to a high density of atomic positions that belong to both stuctures. In reality, of course, some small adjustments of atomic positions at the boundary and close to it are to be expected and real grain boundaries are not strictly two-dimensional. This more realistic picture requires an extension of the coincidence-site concept to take *near*-coincidences into account; and is dealt with in the theory of 'secondary dislocations' and Bollmann's O-lattice theory (Bollmann, 1970, 1977; Iwasaki, 1976). The coincidence-site lattice approach ignores this complication, and represents a crystalline structure simply by its Bravais lattice. In this approach two superimposed lattices are considered (usually two congruent lattices in a specific relative orientation) and those relative orientations of the two lattices are sought that produce a high density of points common to both. When coincidence points exist, they constitute a lattice – the *coincidence-site lattice* or CSL. A grain boundary is then represented as a two-dimensional section of the CSL.

A major step in the mathematical theory of CSLs was provided by the formulae proposed by Ranganathan (1966) for a cubic lattice and a congruent lattice related to it by a rotation through an angle θ about a crystallographic axis ($h_1\ h_2\ h_3$). A CSL exists whenever

$$\tan\left(\frac{\theta}{2}\right) = \frac{Y\sigma}{X}\,,$$

where X and Y are mutually prime integers and

$$\sigma = \sqrt{(h_1{}^2 + h_2{}^2 + h_3{}^2)}\,.$$

The density of the CSL (relative to that of the given cubic lattice) is $1/\Sigma$, where

$$m\Sigma = X^2 + \sigma^2 Y^2\,,$$

in which m is 1 if $X^2 + \sigma^2 Y^2$ is odd and is equal to 2 or 4 according as 2 or 4 is the even factor of $X^2 + \sigma^2 Y^2$.

The Ranganathan formulae are valid for *fcc* and *bcc* as well as for a primitive cubic lattice.

The literature on the mathematical theory of CSLs has become extensive. We can mention just a few of the key developments. Grimmer (1973) developed a matrix approach to the particular case of CSLs for cubic lattices. Grimmer, Bollmann and Warrington (1974) computed and tabulated all CSLs obtained from a cubic lattice and a rational rotation, for values of Σ up to 49. Two rotations are equivalent in this context if they describe the same relative orientation of two lattices. Equivalence classes of rational rotations correspond to equivalence classes of integral quaternions (Grimmer, 1974a). The elegant quaternion approach is implicit in Grimmer's earlier work (Grimmer, 1973) and in Mykura's extension of the list of all possible CSLs for a cubic lattice to $\Sigma = 101$ (Mykura 1979). Zeiner (2005) has developed a quaternion approach to the problem of identifying the Bravais classes to which CSLs arising from cubic lattices belong and has presented the the results, up to $\Sigma = 59$, and in a recent paper has dealt with the CSLs of four-dimensional hypercubic lattices (Zeiner 2006). The quaternion approach can be applied also in the case of non-cubic lattices (Grimmer, 1980; Heinz, Neumann, 1991). The rhombohedral case exhibits some special features (Grimmer, 1989 a, b). A comprehensive unified treatment of the CSL theory for cubic lattices has been given recently (Reed *et al.*, 2005). The matrix

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formulation of the CSL problem in its greatest generality (in n-dimensions, for pairs of lattices that are not necessarily congruent) was presented by Santoro and Mighell (1973) and by Fortes (1983). Coincidence sites for quasilattices have also received attention – for planar quasilattices with N-fold symmetry (Pleasants *et al.*, 1996) and for the icosahedral case (Warrington, Lück, 1996; Baake 1997). The icosahedral case is related to the CSLs for a cubic lattice in six dimensions. A significant feature of these investigations was the derivation of the number of orientations that give rise to coincidences for each value of the CSL density, expressed by successive terms in the series expansion of a generating function. Grain boundaries between quasicrystals and periodic structures have also been considered (Proult *et al.*, 1996; Ranganathan *et al.,* 2000)

Janner's investigation (Janner, 2004a) of the statistical distribution of c/a ratios in hexagonal and tetragonal crystals has shown that far more lattices of real crystals are rational or near-rational, than would be expected by chance. The reason for this is unknown. This discovery suggests that theoretical investigations of properties of *rational* lattices may turn out to be relevant to understanding real materials, rather than being just an interesting academic exercise. The discovery was followed by Janner's development of a mathematical theory of sublattices of rational lattices and equivalence classes of rational lattices (Janner, 2004b).

In Section 2 a concise matrix formulation of the CSL problem is presented, that leads, in Section 3, to a very brief proof of Grimmer's reciprocity theorem – much simpler than previous proofs. The Ranganathan formulae are then shown to have a much greater generality than their origin in the context of cubic lattices. Subsequent sections demonstrate our method of finding CSLs. The method is completely general: it is applicable to any rational lattice in any number of dimensions. The idea is to exploit the fact that a displacement shift lattice (DSL) is given, by definition, in terms of a linearly-dependent set of lattice translations. We shall demonstrate how Buerger's algorithm for finding reduced cells can be applied to the problem of removing this redundancy. Having obtained a linearly-independent set of base vectors for a DSL, the corresponding CSL for the reciprocal lattices follows immediately from Grimmer's reciprocity theorem. Finally, simple matrix and quaternion methods for obtaining a unique representative rotation to characterise a disorientation is demonstrated by presenting a specific numerical example.

2. Notation and definitions

Let $\{\mathbf{e}_i,\ i=1,\dots m\}$ be a set of vectors in Euclidean n-dimensional space E^n. The set of all points with position vectors of the form

$$\mathbf{r}=\mathbf{e}_1u^1+\mathbf{e}_2u^2+\dots\mathbf{e}_mu^m\,, \tag{2.1}$$

where the u^i are integers, is a *point lattice* Λ in E^n provided there is a non-zero minimal distance between any two distinct points of the set. A *translation* lattice is also defined by (2.1), with the vectors $\mathbf{r}$ interpreted as translations rather than position vectors. The lattices we shall be dealing with may be regarded either as point lattices or translation lattices; it makes no difference to the algebraic expressions. For definiteness we employ the language of point lattices. The vectors $\mathbf{e}_i$ are the *base vectors*. The set of base vectors may be redundant, that is, the base vectors may be linearly dependent, in which case $m>n$. We shall encounter this kind of redundancy in dealing with displacement shift lattices. The set of integers $[u_1,u_2,\dots,u_m]$ is a generalisation of the *zone axis symbol* of crystallography.

Equation (2.1) can be written as

$$\mathbf{r}=Eu\,,\quad (u\in Z^m) \tag{2.2}$$

where E denotes the *row* of vectors $(\mathbf{e}_1,\ \mathbf{e}_2,\dots\mathbf{e}_m)$ and u denotes the m integers u^i, written as a *column*.

If the vectors $\mathbf{e}_i$ are specified by their components in a Cartesian reference system, then the Cartesian components of the position vectors $\mathbf{r}$ of the points of Λ are given by (2.2), with E now interpeted as an $n\times m$ matrix whose columns are the Cartesian components of the vectors $\mathbf{e}_i$. E is a *generating matrix* (or, simply, a *generator*) for the lattice Λ. If the rank of E is p then Λ lies in a p-dimensional subspace E^p of E^n. The generator of a given lattice is not unique: if E is a generator for Λ then so is EQ where Q is any unimodular matrix of integers. (In particular, the lattice is unaffected by the order of the columns of the matrix E that generates it and by arbitrary changes of overall sign of any column.)

The columns of a *non-singular* generating matrix E are the vectors giving the translations that define a primitive unit cell of the lattice.

Denoting the transpose of a matrix by a superscript T, the symmetric matrix

$$M=E^{\mathrm{T}}E \tag{2.3}$$

is the *metric matrix* (or, for brevity, simply the *metric*) associated with the generator E. Its elements are the scalar products of the translation vectors given by the columns of E. M is invariant under rotations $E\to RE$ $(RR^{\mathrm{T}}=I)$ of Λ but dependent on the choice of generating matrix; a change $E\to EQ$ induces

$$M\to Q^{\mathrm{T}}MQ\,. \tag{2.4}$$

A lattice is *rational* if its metric M is a rational matrix (a matrix of rational numbers), and is an *integral lattice* if M is an integral matrix (a matrix of integers). In what follows we shall deal only with integral lattices. This involves no loss of generality because, of course, integral lattices and rational lattices (or rational lattices scaled with an irrational factor) differ simply by a scale factor.

The *reciprocal lattice* Λ^* of a lattice Λ can be defined as the set of all points in the subspace to which Λ belongs, whose position vectors $\mathbf{r}^*$ satisfy

$$\mathbf{r}^*\cdot\mathbf{r}\in Z\quad\text{for all}\quad\mathbf{r}\in\Lambda\,. \tag{2.5}$$

Proof: Let $\{\mathbf{e}_i,\ i=1,\dots,\ n\}$ be a linearly independent set of base vectors for Λ. Then Λ is given by

$$\mathbf{r}=Eu\,,\quad (u\in Z^n)$$

with a non-singular $n \times n$ generator E. Equation (2.5) then states that $\mathbf{r}^* Eu \in Z$ for all $u \in Z^n$ and, in particular, $h = \mathbf{r}^* E \in Z^n$. Therefore, $\mathbf{r}^*$ has the form

$$\mathbf{r}^* = hE^{-1}, \quad h \in Z^n, \tag{2.6}$$

which can be rewritten as

$$\mathbf{r}^* = h_1 \mathbf{e}^{*1} + h_2 \mathbf{e}^{*2} \dots h_n \mathbf{e}^{*n}$$

with the Cartesian components of the vectors $\mathbf{e}^{*i}$ given by the rows of E^{-1}. Since $E^{-1}E = I$, it follows that the base vectors $\mathbf{e}_i$ for Λ and the base vectors $\mathbf{e}^{*i}$ for Λ^* satisfy

$$\mathbf{e}^{i*} \cdot \mathbf{e}_i = \delta^i{}_j .$$

Therefore Λ^*, defined by (2.5) is the lattice reciprocal to Λ, as usually defined.

Since any matrix equation and its transpose convey the same information Eq. (2.6) can be written in the alternative form

$$\mathbf{r}^* = E^{-\mathrm{T}} h^{\mathrm{T}}, \quad h \in Z^n, \tag{2.7}$$

in which $\mathbf{r}^*$ and h^{T} are now interpreted as columns. (We use superscript T to denote matrix transpose, and $-\mathrm{T}$ to denote a transposed inverse of a matrix). Comparing this with (2.2) allows us to state that, if a lattice Λ is generated by a non-singular generator E, then its reciprocal lattice Λ^* is generated by $E^{-\mathrm{T}}$. This change of viewpoint allows us to treat vectors and reciprocal vectors on the same footing. This is expedient in later sections, where we are dealing with the relationship between a CSL and a DSL and *all* lattice vectors are are treated as *columns* of Cartesian components

If E generates Λ, the *sublattices* of Λ are generated by matrices of the form EQ, where Q is an integral $m \times m$ matrix. The *density* of the sublattice (ratio of the volume of the unit cell of Λ to that of the sublattice) is the absolute value of $1/|Q|$. If two lattices Λ_1 and Λ_2 occupy the same space, the set of all points contained in both (if any exist) constitute a lattice, the *coincidence-site lattice* (CSL) of Λ_1 and Λ_2. It is the lattice of greatest density contained in Λ_1 and Λ_2. The dual of this concept is the *displacement-shift lattice* (DSL), the lattice of least density that contains Λ_1 and Λ_2.

Note that, for the CSL and DSL of Λ_1 and Λ_2 to exist as 'true' lattices in E^n (generated by n linearly independent vectors), a necessary and sufficient condition is that Λ_1 and Λ_2, generated respectively by the non-singular matrices E and F, be commensurate in the sense that a set of base vectors for each should be a *rational* linear combination of the base vectors of the other. That is, $E^{-1}F$ should be rational.

3. Grimmer's theorem

A result of fundamental importance in the theory of coincidence sites, given by Grimmer (1974b), is:

The CSL of two lattices is the reciprocal of the DSL of their reciprocals.

Various proofs have appeared in the literature. The following proof is presumably the simplest. Let E and F be non-singular generating matrices for two lattices in E^n. Denote the CSL by Γ and the DSL of the reciprocals by Δ. Then, by definition,

$$\Gamma = \{\mathbf{r} : \mathbf{r} = Eu = Fv,\ u \in Z^n,\ v \in Z^n\},$$

$$\Delta = \{\mathbf{r}^* : \mathbf{r}^* = hE^{-1} + kF^{-1},\ h \in Z^n,\ k \in Z^n\}.$$

Therefore, for any $\mathbf{r} \in \Gamma$ and any $\mathbf{r}^* \in \Delta$,

$$\begin{aligned}\mathbf{r}^* \cdot \mathbf{r} &= (hE^{-1} + kF^{-1})\,\mathbf{r} = hE^{-1}\mathbf{r} + kF^{-1}\mathbf{r} \\ &= hE^{-1}Eu + kF^{-1}Fv = hu + kv \in Z,\end{aligned}$$

i.e., $\Gamma = \Delta^*$.

4. Reorientation of an integral lattice

A rotation in E^3 about the origin can be expressed as $\mathbf{r} \to R\mathbf{r}$, where R is an orthogonal matrix ($RR^{\mathrm{T}} = I$). For a rotation through an angle θ about an axis in the direction specified by a unit vector $\mathbf{n}$ this matrix takes the form

$$R = \mathrm{e}^{\theta N} = I + \theta N + \theta^2 N^2/(2!) \ + \theta^3 N^3/(3!) + \dots, \tag{4.1}$$

$$N = \begin{pmatrix} 0 & -n_3 & n_2 \\ n_3 & 0 & -n_1 \\ -n_2 & n_1 & 0 \end{pmatrix}$$

where n_1, n_2 and n_3 are the Cartesian components of $\mathbf{n}$. Because $N^3 = -N$,

$$R = I + N \sin\theta + N^2(1 - \cos\theta). \tag{4.2}$$

We shall be concerned with the CSL of an *integral* lattice Λ in E^3 generated by a nonsingular matrix E, and a reorientation of it, generated by RE. Recalling (2.7) and the remarks following it, we see that the DSL of the reciprocal lattices is generated by the 3×6 matrix

$$[E^{-\mathrm{T}}, RE^{-\mathrm{T}}] = E^{-\mathrm{T}}[I, E^{\mathrm{T}}RE^{-\mathrm{T}}]. \tag{4.3}$$

If R is taken to be a rotation through angle θ about an axis indexed by $(h_1\ h_2\ h_3)$ (indices referred to the primitive unit cell given by E) then, denoting the index set $(h_1\ h_2\ h_3)$ (a row of integers) by the symbol h, the unit vector $\mathbf{n}$ along this axis is given by

$$\mathbf{n} = \frac{hE^{-1}}{\sqrt{hM^{-1}h^T}} = \frac{|E|\ hE^{-1}}{\sigma}, \qquad \sigma = \sqrt{hM^{\mathrm{A}}h^T}. \tag{4.4}$$

M^{A} denotes the matrix *adjoint* to M,

$$M^{\mathrm{A}} = |M|\ M^{-1}. \tag{4.5}$$

Note that if M is an integral matrix, then so is M^{A}; σ^2 is therefore an integer. In order to obtain the form of (4.3) explicitly in terms of $(h_1\ h_2\ h_3)$ and θ, we need to investigate the form of $E^{\mathrm{T}}NE^{-\mathrm{T}}$. In terms of matrix components,

$$N_{ij} = -\varepsilon_{ijk} n_k = -\varepsilon_{ijk} h_\alpha (E^{-1})^\alpha{}_k\ |E|/\sigma.$$

ε_{ijk} is the *alternating symbol*, equal to $+1$ or -1 when ijk is an even or an odd permutation, respectively, of 123. Otherwise, it is zero. We have also made use here of the *summation convention*: if a subscript or superscript appears twice in an expression, a summation over all its values is implied.

Now,

$$-\varepsilon_{ijk}(E^{-1})^{\beta}{}_i\,(E^{-1})^{\gamma}{}_j\,(E^{-1})^{\alpha}{}_k = -\varepsilon^{\alpha\beta\gamma}|E^{-1}|$$

(an identity expressing the definition of a determinant), from which it follows that

$$-\varepsilon_{ijk}(E^{-1})^{\alpha}{}_k\,|E| = -\varepsilon^{\alpha\beta\gamma}E_{i\beta}E_{j\gamma}\,.$$

Therefore,

$$N_{ij} = -h_\alpha\varepsilon^{\alpha\beta\gamma}E_{i\beta}E_{j\gamma}/\sigma\,.$$

If we define a matrix H,

$$H^{\beta\gamma} = -h_\alpha\varepsilon^{\alpha\beta\gamma}\,,$$

$$\text{i.e.,}\quad H = \begin{pmatrix} 0 & -h_3 & h_2 \\ h_3 & 0 & -h_1 \\ -h_2 & h_1 & 0 \end{pmatrix}, \tag{4.6}$$

then

$$N_{ij} = H^{\beta\gamma}E_{i\beta}E_{j\gamma}/\sigma\,,$$

$$\text{i.e.,}\quad N = EHE^{\mathrm{T}}/\sigma$$

and, finally,

$$E^{\mathrm{T}}NE^{-\mathrm{T}} = \frac{MH}{\sigma}\,. \tag{4.7}$$

Substituting this into the expression (4.2) for a rotation,

$$E^{\mathrm{T}}RE^{-\mathrm{T}} = I + \frac{\sin\theta}{\sigma}\,MH + (1-\cos\theta)\,\frac{(MH)^2}{\sigma^2}\,. \tag{4.8}$$

Recalling the note on commensurability at the end of Section 2, we can conclude that a 'true' DSL and CSL will arise if and only if this matrix is rational. Noting that σ^2 is an integer and that H is a matrix of integers, we see that $E^{\mathrm{T}}RE^{-\mathrm{T}}$ will be rational if and only if $(\sin\theta)/\sigma$ and $\cos\theta$ are rational, or, in other words, if and only if $\tan(\theta/2)/\sigma$ is rational. We therefore can write

$$\tan(\theta/2) = \frac{Y\sigma}{X} \tag{4.9}$$

where X and Y are mutually prime integers. Then

$$\frac{\sin\theta}{\sigma} = \frac{2XY}{m\Sigma} \quad\text{and}\quad \frac{(1-\cos\theta)}{\sigma^2} = \frac{2Y^2}{m\Sigma}\,,$$

$$m\Sigma = X^2 + \sigma^2Y^2. \tag{4.10}$$

(m is an integer, still to be chosen). Observe that (4.9) and (4.10) are homologous to the Ranganathan formulae for cubic lattices. The difference lies in the form of the quadratic expression σ^2.

We now have

$$E^{\mathrm{T}}RE^{-\mathrm{T}} = I + \frac{2Y}{m\Sigma}\,(XMH + Y(MH)^2)\,. \tag{4.11}$$

If m is now chosen to be the greatest common factor of $X^2+\sigma^2Y^2$ and all the elements of the integral matrix $2Y(XMH+Y(MH)^2$, the 3×6 matrix (4.3) that generates the DSL can be written in the form

$$\frac{E^{-\mathrm{T}}}{\Sigma}\,[\Sigma I, W]\,, \tag{4.12}$$

in which

$$W = \Sigma(E^{\mathrm{T}}RE^{-\mathrm{T}} - I) = 2Y(XMH + Y(MH)^2)/m \tag{4.13}$$

is an *integral* matrix.

To find the CSL, it is necessary to deduce a non-singular 3×3 generating matrix L for the DSL by eliminating the redundancy of the set of translations given by the columns of (4.12); the CSL is then generated by $L^{-\mathrm{T}}$.

5. The reduction algorithm

Let M be the metric for a lattice in E^n and let $M_{\alpha\beta}$ be the off-diagonal element with greatest absolute value such that

$$2|M_{\alpha\beta}| > M_{\alpha\alpha}\,,\qquad M_{\alpha\alpha} \le M_{\beta\beta}\,. \tag{5.1}$$

Changing the α-th column ($\mathbf{e}_\alpha$) of E according to

$$\mathbf{e}_\alpha \to \mathbf{e}_\alpha - \mu\mathbf{e}_\beta\,, \tag{5.2}$$

where μ is the sign of $M_{\alpha\beta}$, reduces the length of the vector $\mathbf{e}_\alpha$. If, after this step, the column $\mathbf{e}_\alpha$ is null it is to be eliminated from E. Iteration of these instructions will, *in general*, terminate when E has become an $n\times n$ nonsingular matrix whose columns are n of the shortest translations of Λ, and all elements of the corresponding metric matrix M satisfy

$$2|M_{\alpha\beta}| \le M_{\alpha\alpha}\,,\qquad 2|M_{\alpha\beta}| \le M_{\beta\beta}\,. \tag{5.3}$$

The phrase 'in general' refers to the fact that there exist exceptional cases where the iterative procedure has produced a metric satisfying (5.3) — so that the iterative process has terminated — but the matrix E has not been converted to a square matrix. A very simple example of this situation is

$$E = \begin{pmatrix} 1 & -1 & -1 & 1 \\ -1 & 1 & -1 & 1 \\ -1 & -1 & 1 & 1 \end{pmatrix},$$

$$M = \begin{bmatrix} 3 & -1 & -1 & -1 \\ -1 & 3 & -1 & -1 \\ -1 & -1 & 3 & -1 \\ -1 & -1 & -1 & 3 \end{bmatrix}$$

(body-centered cubic in E^3). In these circumstances the process of finding a non-singular generating matrix E can be readily completed by finding the relationship of linear dependence of its columns and eliminating a redundant column. A different kind of anomaly occurs when the algorithm terminates, but the columns of E are not the three shortest translations. A simple example is a rhombohedral lattice,

$$E = \begin{pmatrix} 2 & -1 & -1 \\ 0 & \sqrt{3} & -\sqrt{3} \\ \lambda & \lambda & \lambda \end{pmatrix},$$

$$M = \begin{bmatrix} 4+\lambda^2 & \lambda^2-2 & \lambda^2-2 \\ \lambda^2-2 & 4+\lambda^2 & \lambda^2-2 \\ \lambda^2-2 & \lambda^2-2 & 4+\lambda^2 \end{bmatrix},$$

for which $\lambda = 2(c/a)/\sqrt{3} < 1/\sqrt{2}$. Then M is already in reduced form (it satisfies (5.3)) but the columns of E are not the three shortest translations; $\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3$ is shorter. Observe, however, that in this case the unit cell corresponding to E is the primitive rhombohedral cell, which possesses the threefold symmetry of the lattice, and is thus 'more appropriate' than one based on the three shortest translations.

The idea of introducing a standard primitive unit cell for every lattice in E^3, based on the three shortest lattice translations, is due to Niggli (1928), who employed the theory of the classification of quadratic forms to obtain a unique form for the metric matrix for any three-dimensional lattice. There are 44 types, so this is a more refined classification than the Bravais classification. Niggli's classification is more readily accessible in Mighell and Rodgers (1980). The algorithm given above is the essence of the method given by Buerger (1957) for finding a 'reduced' cell (a primitive unit cell based on the three shortest lattice translations) for any lattice in E^3 from a given arbitrary unit cell. The end result, a *Buerger cell* for the given lattice, is not unique – some lattices can have as many as five inequivalent Buerger cells (Gruber, 1973). The problem posed by this lack of uniqueness is that of finding a standard unique metric for any lattice in E^3 (Santoro, Mighell, 1970; Krívý, Gruber, 1976; Gruber, 1992; Zuo *et al.*, 1995). Simply stated, it is the problem of finding the transformation (2.4) that will convert any nonsingular symmetric 3×3 matrix M to one of Niggli's canonical metrics. However, in the present context this need not concern us. *We can simply employ the iterative procedure to obtain a* nonsingular *matrix L with a metric satisfying* (5.3), *starting from a rectangular matrix of the form* (4.3). The matrix $L^{-\mathrm{T}}$ then generates the required CSL. The problem of the stability of the algorithm (Grosse-Kunstleve *at al.*, 2004) does not arise in the present context since only integer arithmetic is involved.

The reduction algorithm can be applied directly to the matrix M, without involving E. The step (5.2) can be carried out as two-stage process:

$$M_{\alpha\gamma} \to M_{\alpha\gamma} - \mu M_{\beta\gamma}$$

for the α-th row, followed by

$$M_{\gamma\alpha} \to M_{\gamma\alpha} - \mu M_{\gamma\beta}$$

for the α-th column. The α-th row and column are then to be removed if they are null. Having arrived at the final M satisfying (5.3) the corresponding reduced E can be obtained in one step

$$E \to EQ\,,$$

where Q is the integral matrix obtained as the product of all the matrices q that express the changes $M \to q^{\mathrm{T}}Mq$ undergone by M at each step.

6. An example

We illustrate the technique by applying the reduction algorithm to a simple rational lattice. We choose Frank's 'cubic hexagonal' lattice (Frank, 1965), which is a curious case of some intrinsic interest (Ranganathan *et al.*, 2002). The operation of the algorithm is in fact so simple and straightforward that it can be carried out quite quickly by pencil and paper calculation. The 'cubic hexagonal' lattice is a hexagonal lattice with a c/a ratio of $\sqrt{(3/2)}$. The generator and metric can be taken to be

$$E = \begin{pmatrix} \sqrt{2} & -1/\sqrt{2} & 0 \\ 0 & \sqrt{3/2} & 0 \\ 0 & 0 & \sqrt{3} \end{pmatrix},$$

$$M = \begin{bmatrix} 2 & -1 & 0 \\ -1 & 2 & 0 \\ 0 & 0 & 3 \end{bmatrix}, \qquad M^{\mathrm{A}} = 3 \begin{bmatrix} 2 & 1 & 0 \\ 1 & 2 & 0 \\ 0 & 0 & 1 \end{bmatrix}.$$

(We have employed a scaling factor so that M is an *integral* matrix rather than simply rational). As an (arbitrarily chosen) example, consider $h = (0\ 1\ 1)$, $X = 3$, $Y = 1$. This gives $\sigma^2 = hM^{\mathrm{A}}h = 9$, $\theta = 2\tan^{-1}(1) = 90°$, $m\Sigma = 18$.

$$2Y(XMH + Y(MH)^2) = 6 \begin{pmatrix} -4 & -2 & 2 \\ 3 & 0 & 0 \\ -2 & 2 & -2 \end{pmatrix}$$

so $m = 6$, $\Sigma = 3$ and the generator (4.12) for the DSL can be replaced by $(^1/_3)\, E^{-\mathrm{T}}[3I, W]$, which is

$$\frac{E^{-\mathrm{T}}}{3} \begin{pmatrix} 3 & 0 & 0 & -4 & -2 & 2 \\ 0 & 3 & 0 & 3 & 0 & 0 \\ 0 & 0 & 3 & -2 & 2 & -2 \end{pmatrix}.$$

The components in the last three columns can be immediately reduced modulo 3 by additions and subtractions of the first three columns. Also note that the fifth and sixth columns differ only by a sign, and so one of them can be eliminated. The algorithm terminates at

$$L = \frac{E^{-\mathrm{T}}}{3} \begin{pmatrix} 2 & 0 & 1 \\ 0 & 3 & 0 \\ 1 & 0 & -1 \end{pmatrix}.$$

The CSL is therefore generated by $L^{-\mathrm{T}} = EQ$,

$$Q = \begin{pmatrix} 1 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & -2 \end{pmatrix}.$$

The determinant of Q is -3, so the density of the CSL is $^1/_3$.

(Note that there are many integral matrices Q that describe in this way the same CSL; they are related to each other by multiplication on the right or the left by any integral matrix with determinant ± 1.)

The metric of the CSL is

$$Q^{\mathrm{T}}MQ = \begin{bmatrix} 5 & -1 & -4 \\ -1 & 2 & -1 \\ -4 & -1 & 14 \end{bmatrix}.$$

This does not satisfy (5.3) so the algorithm can be applied to reduce it to a more recognisable form. The result is

$$\begin{bmatrix} 5 & -1 & 0 \\ -1 & 2 & 0 \\ 0 & 0 & 9 \end{bmatrix},$$

showing that the CSL is primitive monoclinic.

7. Centred cells

In the method suggested above for finding CSLs the indices $h = (h_1\ h_2\ h_{3'})$ of the rotation axis $\mathbf{n}$ are defined with reference to a *primitive* unit cell. For lattices of types I, F or S this is not the accepted way of indexing an axis. Thus, if the axis is given by its index triple k referred to a centred cell, a conversion must be made to obtain the triple h needed as input for the algorithm. Let E be the given nonsingular generator for a lattice Λ and let G be the matrix whose columns are the translations along the edges of a centred cell. Then

$$\mathbf{n} \sim hE^{-1} \sim kG^{-1}\,,$$

so that

$$h \sim kG^{-1}E\,, \qquad k = hE^{-1}G\,.$$

We employ the symbol $\sim$ to denotes equality to within a scalar factor. The vector $\mathbf{n}$ is to be normalised to a unit vector and h or k to a triplet of mutually prime integers). Thus, conversion of the usual indexing in terms of centred cell to the indices h required in the present context is achieved with the aid of the matrix $G^{-1}E$. The following forms may be adopted (the matrix E has been chosen in each case in a convenient form. It should be noted that the corresponding choice of primitive cell here is *not* in general a Buerger cell). These particular forms for the generating matrices E and G of the Bravais lattices will be adopted as canonical in subsequent sections of this paper.

$$E = \begin{pmatrix} a & -a & -a \\ -b & b & -b \\ -c & -c & c \end{pmatrix}, \qquad G = \begin{pmatrix} 2a & & \\ & 2b & \\ & & 2c \end{pmatrix},$$

$$G^{-1}E = {}^1\!/_2 \begin{pmatrix} 1 & -1 & -1 \\ -1 & 1 & -1 \\ -1 & -1 & 1 \end{pmatrix},$$

$$E^{-1}G = -\begin{pmatrix} 0 & 1 & 1 \\ 1 & 0 & 1 \\ 1 & 1 & 0 \end{pmatrix}$$

for body-centred (cubic, tetragonal or orthorhombic);

$$E = \begin{pmatrix} 0 & a & a \\ b & 0 & b \\ c & c & 0 \end{pmatrix}, \qquad G = \begin{pmatrix} 2a & & \\ & 2b & \\ & & 2c \end{pmatrix},$$

$$G^{-1}E = {}^1\!/_2 \begin{pmatrix} 0 & 1 & 1 \\ 1 & 0 & 1 \\ 1 & 1 & 0 \end{pmatrix},$$

$$E^{-1}G = \begin{pmatrix} -1 & 1 & 1 \\ 1 & -1 & 1 \\ 1 & 1 & -1 \end{pmatrix}$$

for face-centred (cubic or orthorhombic);

$$E = \begin{pmatrix} a & -a & 0 \\ b & b & d \\ 0 & 0 & 2c \end{pmatrix}, \qquad G = \begin{pmatrix} 2a & & \\ & 2b & d \\ & & 2c \end{pmatrix},$$

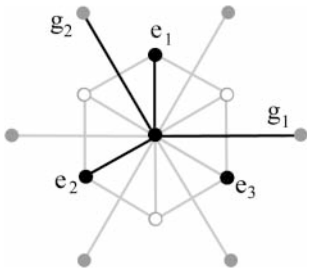

Fig. 1. Relationship between the base vectors of the rhombohedral reference system (E) and those of the hexagonal system (G), for a rhombohedral lattice. (Viewed along g_3)

$$G^{-1}E = {}^1\!/_2 \begin{pmatrix} 1 & -1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix}, \qquad E^{-1}G = \begin{pmatrix} 1 & 1 & 0 \\ -1 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

for base-centred (orthorhombic or monoclinic).

A similar consideration applies a to a *rhombohedral* lattice, where the need is to convert an index set k referred to a hexagonal system to an index set h referred to a primitive rhombohedral cell. Taking the base vectors of the two reference systems in the relationship indicated in Fig. 1,

$$E = \begin{pmatrix} 0 & -a/2 & a/2 \\ a/\sqrt{3} & -a/2\sqrt{3} & -a/2\sqrt{3} \\ c/3 & c/3 & c/3 \end{pmatrix},$$

$$G = \begin{pmatrix} a & -a/2 & 0 \\ 0 & a\sqrt{3}/2 & 0 \\ 0 & 0 & c \end{pmatrix},$$

$$G^{-1}E = {}^1\!/_3 \begin{pmatrix} 1 & -2 & 1 \\ 2 & -1 & -1 \\ 1 & 1 & 1 \end{pmatrix},$$

$$E^{-1}G = \begin{pmatrix} 0 & 1 & 1 \\ -1 & 0 & 1 \\ 1 & -1 & 1 \end{pmatrix}.$$

8. Planar integral lattices

The simpler cases of CSLs for rational lattices in the Euclidean plane E^2 is not without interest. They have the advantage for illustrative purposes that the geometrical situation underlying the algebra is more readily visualised. The planar cases can of course be approached as special cases of the three dimensional case. Consider for example a hexagonal lattice in E^3 and rotate it about its sixfold axis (chosen along the z-axis). Then we can conveniently omit the third row and column from the matrices and write

$$E = \frac{1}{\sqrt{2}} \begin{pmatrix} 2 & -1 \\ 0 & \sqrt{3} \end{pmatrix}, \qquad H = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix},$$

$$M = \begin{pmatrix} 2 & -1 \\ -1 & 2 \end{pmatrix}$$

The adjoint M^{A} is to be replaced by the single integer $|M| = 3 = \sigma^2$. Take, for example, $X = 9$, $Y = 1$, so that $\tan(\theta/2) = {}^1\!/_3\sqrt{3}$ and $m\Sigma = 84$. The generator (4.3) for the DSL is in this case

$$\frac{E^{-\mathrm{T}}}{7} \begin{pmatrix} 7 & 0 & -2 & -3 \\ 0 & 7 & 3 & 1 \end{pmatrix} \to \frac{E^{-\mathrm{T}}}{7} \begin{pmatrix} 3 & 1 \\ -1 & 2 \end{pmatrix} = L$$

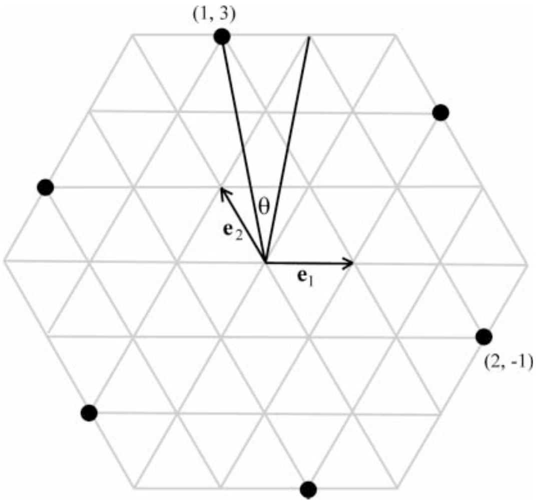

Fig. 2. A CSL with $\Sigma = 7$ for a plane hexagonal lattice. The network of triangles is the original hexagonal lattice. The black circles are the coincidence sites that arise from rotation through angle $\theta = 2\tan^{-1}(1/3\sqrt{3})$.

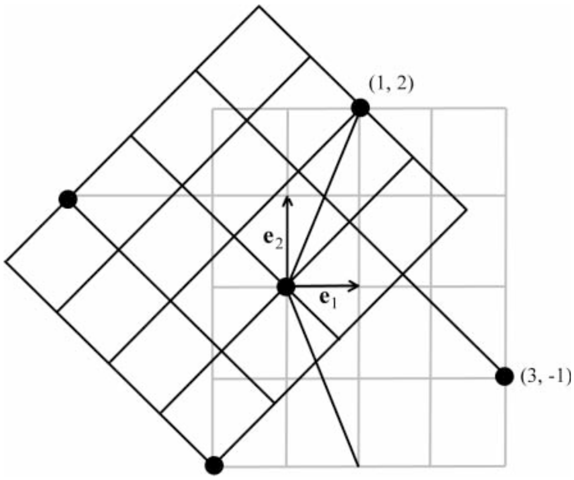

Fig. 3. A plane rectangular lattice generated by translations of length ratio $\sqrt{2}$: $\sqrt{3}$, and the CSL with $\Sigma = 7$ obtained by rotation through $\theta = 2\tan^{-1}(\sqrt{6})$.

so the CSL is generated by $L^{-\mathrm{T}} = EQ$ with

$$Q = \begin{pmatrix} 2 & 1 \\ -1 & 3 \end{pmatrix}.$$

The determinant of Q is 7 so the density of the CSL is $^1/_7$. The metric of the CSL is $Q^{\mathrm{T}}MQ = 7M$; the CSL is an enlarged and rotated version of the original lattice – as are all CSLs of a planar hexagonal lattice. The geometrical situation is illustrated in Fig. 2.

As a second simple example, consider the plane rectangular lattice with

$$E = \begin{pmatrix} \sqrt{2} & \\ & \sqrt{3} \end{pmatrix}, \quad M = \begin{bmatrix} 2 & \\ & 3 \end{bmatrix}, \quad \sigma^2 = |M| = 6.$$

Then $H = \begin{pmatrix} & -1 \\ 1 & \end{pmatrix}$ (as for all planar cases) gives

$$MH = \begin{pmatrix} & -2 \\ 3 & \end{pmatrix}, \qquad (MH)^2 = -6\begin{pmatrix} 1 & \\ & 1 \end{pmatrix}.$$

For the case $X = Y = 1$ we then get $\tan(\theta/2) = \sqrt{6}$, $m\Sigma = 7$, and the DSL is generated by

$$\frac{E^{-\mathrm{T}}}{7}\begin{pmatrix} 7 & 0 & -12 & -4 \\ 0 & 7 & 6 & -12 \end{pmatrix}$$
$$\to \frac{E^{-\mathrm{T}}}{7}\begin{pmatrix} 7 & 0 & 2 & -4 \\ 0 & 7 & -1 & 2 \end{pmatrix} \to \frac{E^{-\mathrm{T}}}{7}\begin{pmatrix} 2 & 1 \\ -1 & 3 \end{pmatrix},$$

from which it follows that the CSL is generated by EQ,

$$Q = \begin{pmatrix} 3 & 1 \\ -1 & 2 \end{pmatrix}.$$

The metric for the CSL is

$$Q^{\mathrm{T}}MQ = 7\begin{bmatrix} 3 & \\ & 2 \end{bmatrix} = \begin{bmatrix} 21 & \\ & 14 \end{bmatrix}.$$

The geometrical situation is illustrated in Fig. 3.

9. Grain boundary lattices

According to coincidence-site lattice theory the structure of a planar portion of a grain boundary is represented by a section of a CSL. Hence a prescription is needed for finding the two-dimensional section of a given CSL in a plane normal to a given direction.

Lemma: The two-dimensional sublattice of a lattice Λ in an $(h_1\ h_2\ h_3)$ plane is generated by EH. (Proof: An $(h_1\ h_2\ h_3)$ plane is normal to the vector $\mathbf{r}^* = hE^{-1}$. All the points $\mathbf{r} = Eu$ of Λ that lie on the $(h_1\ h_2\ h_3)$ plane through the origin satisfy $\mathbf{r}^* \cdot \mathbf{r} = hu = 0$. The integral solutions u of this equation are given by Hv, for any integral v. The sublattice of Λ in the plane h consists of all points $\mathbf{r}$ of the form $\mathbf{r} = EHv$.)

Consider a lattice Λ and a sublattice of Λ (*e.g.*, CSL) generated, respectively, by nonsingular E and $L^{-\mathrm{T}} = EQ$, where Q is an integral matrix. Let h' be an index set for a lattice plane of Λ. The vector perpendicular to the plane is $\mathbf{r}^* = h'E^{-1}$. With reference to the sublattice the index set for the same plane is $k = \mathbf{r}^* L^{-\mathrm{T}} = h'Q$. From the above lemma it then follows that the planar lattice of the sublattice generated by $L^{-\mathrm{T}}$ that lies in a plane h' is generated by

$$EQK, \qquad K = \begin{pmatrix} 0 & -k_3 & k_2 \\ k_3 & 0 & -k_1 \\ -k_2 & k_1 & 0 \end{pmatrix}, \qquad k = h'Q. \tag{9.1}$$

As an illustrative example, take again the CSL of the cubic hexagonal lattice given by $h = (0\,1\,1)$, $X = 3$, $Y = 1$, that we obtained in section 6:

$$Q = \begin{pmatrix} 1 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & -2 \end{pmatrix}.$$

Suppose we wish to find the two-dimensional lattice of coincidence sites in a (2 1 0) plane (the grain boundary). Thus, $h' = (2\ 1\ 0)$, $k = (2\ 1\ 2)$,

$$K = \begin{pmatrix} 0 & -2 & 1 \\ 2 & 0 & -2 \\ -1 & 2 & 0 \end{pmatrix}, \quad QK = \begin{pmatrix} -1 & 0 & 1 \\ 2 & 0 & -2 \\ 2 & -6 & 1 \end{pmatrix},$$

$$EQK = \begin{pmatrix} -2\sqrt{2} & 0 & 2\sqrt{2} \\ \sqrt{6} & 0 & -\sqrt{6} \\ 2\sqrt{3} & -6\sqrt{3} & \sqrt{3} \end{pmatrix}.$$

The metric of this singular (rank 2) generating matrix is $\begin{bmatrix} 26 & -36 & -8 \\ -36 & 108 & -18 \\ -8 & -18 & 17 \end{bmatrix}$. The reduction algorithm converts this to the more appropriate 2×2 metric of the *grain boundary lattice*:

$$\begin{bmatrix} 26 & -8 \\ -8 & 17 \end{bmatrix}.$$

10. Equivalence classes of rotations

In terms of a generating matrix E of a lattice Λ, a *point symmetry* of Λ corresponds to an orthogonal matrix S for which

$$SE = EQ \tag{10.1}$$

for some unimodular matrix Q of integers. The integral matrices corresponding in this way to symmetries of Λ satisfy $Q^{\mathrm{T}}MQ = M$. The formula (10.1) can be interpreted as a definition of a representation of the symmetry group of the lattice in terms of integral matrices Q. It will be convenient to refer to these representations as '*Q-representations*'.

The orientation relationship between two congruent lattices is represented by an *equivalence class* of rotations, a rotation R being *equivalent* to $S'RS$, where S and S' are orthogonal matrices associated with lattice symmetries. Any rotation in the equivalence class of a given rotation R can be expressed as $S(S'R)\,S^{-1}$ where S and S' are rotations belonging to the symmetry group of Λ. Since trace $(R) = 1 + 2\cos\theta$ it follows that any two rotations R and SRS^{-1} are rotations by the *same angle* θ, about different axes, and therefore that all the angles of the rotations in the equivalence class of R are given by the subset of rotations of the form $S'R$.

R and R^{-1} (rotation by θ or by $-\theta$ about the same axis) are also equivalent, because to describe the orientation relation between two congruent lattices either of them can be taken as the reference lattice. To find all rotations of the equivalence class to which a given rotation R belongs, those rotations $S'RS$ and $S'R^{-1}S$ with S and S' belonging to the *rotational* subgroup of the symmetry group of Λ need be computed. This 'reduced' equivalence class may contain as many as $2g^2$ rotations, where g is the order of the group of rotational symmetries of Λ. An important problem is the selection of a unique rotation as a definitive representative of its equivalence class. The convention usually adopted is to select the rotation with the smallest rotation angle θ and with axis $\mathbf{n}$ corresponding to a point in a chosen fundamental region of the stereogram associated with the point symmetry group of Λ (Grimmer, 1980; Heinz, Neumann, 1991). Clarification of this latter concept is provided by Fig. 4, which illustrates the spherical space of unit vectors $\mathbf{n}$. The points marked $\mathbf{g}_1$, $\mathbf{g}_2$, $\mathbf{g}_3$ correspond to the directions of the basis vectors $\mathbf{g}_1$, $\mathbf{g}_2$, $\mathbf{g}_3$, which define the edges of the unit cell and whose Cartesian components are given by the columns of the matrices G of Section 7 (for primitive lattices, $G = E$). The criteria for a vector $\mathbf{n}$ to lie in the chosen fundamental region are given in terms of the components ($\varkappa_1\ \varkappa_2\ \varkappa_3$) of $\mathbf{n}$ referred to this basis: $\mathbf{n} \sim \mathbf{g}_1\varkappa_1 + \mathbf{g}_2\varkappa_2 + \mathbf{g}_3\varkappa_3 = G\varkappa$, so that

$$\varkappa \sim G^{-1}\mathbf{n} = \mathbf{n}G^{-\mathrm{T}}.$$

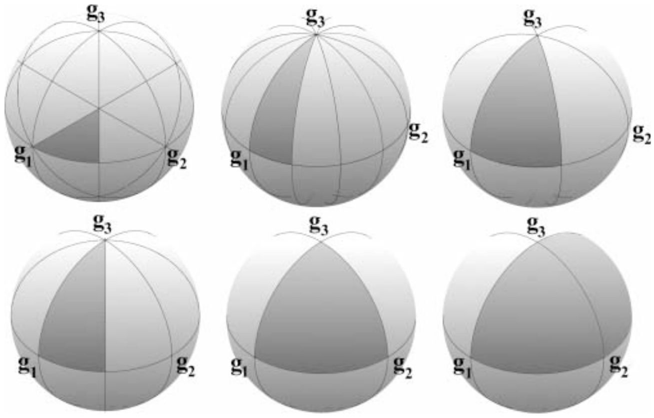

Fig. 4. Spherical representations of the fundamental regions (darker grey) for the symmetry groups of Bravais lattices. First row: cubic, hexagonal, rhombohedral. Second row: tetragonal, orthorhombic, monoclinic.

The criteria for $\mathbf{n}$ to lie in the chosen fundamental region are

Cubic: $0 \le \varkappa_2 \le \varkappa_1, \quad 0 \le \varkappa_3 \le \varkappa_1$
Hexagonal: $0 \le \varkappa_2 \le \varkappa_1, \quad 0 \le \varkappa_3$
Rhombohedral: $0 \le \varkappa_1, \quad 0 \le \varkappa_2, 0 \le \varkappa_3$
Tetragonal: $0 \le \varkappa_1, \quad 0 \le \varkappa_2, 0 \le \varkappa_3$
Orthorhombic: $0 \le \varkappa_1, \quad 0 \le \varkappa_3$
Monoclinic: $0 \le \varkappa_3$

10.1 Matrix method

Q-representations for the rotational symmetry groups of the various Bravais lattices can be obtained as products of the sets of integral matrices listed below. These particular representations correspond to the choices of E employed in Section 7. As a space-saving device, each 3×3 matrix is presented as a row of three number triples, each triple represents a *column* of a matrix Q):

Cubic 432 (order 24):
P (010, $\bar{1}$00, 001), (001, 100, 010), (00$\bar{1}$, 0$\bar{1}$0, $\bar{1}$00)
I (00$\bar{1}$, 111, 0$\bar{1}$0), (001, 100, 010), (00$\bar{1}$, 0$\bar{1}$0, $\bar{1}$00)
F (10$\bar{1}$, 100, 1$\bar{1}$0), (001, 100, 010), (00$\bar{1}$, 0$\bar{1}$0, $\bar{1}$00)
Hexagonal 622 (order 12)
(110, $\bar{1}$00, 001), (100, $\bar{1}\bar{1}$0, 00$\bar{1}$)
Rhombohedral 322 (order 6)
(001, 100, 010), (0$\bar{1}$0, $\bar{1}$00, 00$\bar{1}$)
Tetragonal 422 (order 8)
P (010, $\bar{1}$00, 001), (010, 100, 00$\bar{1}$)
I (00$\bar{1}$, 111, 0$\bar{1}$0), ($\bar{1}$00, 0$\bar{1}$0, 111)
Orthorhombic 222 (order 4)
P ($\bar{1}$00, 0$\bar{1}$0, 001), (100, 0$\bar{1}$0, 00$\bar{1}$)

I (010, 100, $\bar{1}\bar{1}\bar{1}$), ($\bar{1}\bar{1}\bar{1}$, 001, 010)
F (01$\bar{1}$, 10$\bar{1}$, 00$\bar{1}$), ($\bar{1}$00, $\bar{1}$01, $\bar{1}$10)
S ($\bar{1}$00, 0$\bar{1}$0, 001), (0$\bar{1}$0, $\bar{1}$00, 00$\bar{1}$)
Monoclinic 2 (order 2)
P ($\bar{1}$00, 010, 00$\bar{1}$)
S (010, 100, 00$\bar{1}$)

For completeness, we list the relations satisfied by the generators (in the sense of Coxeter and Moser (1972) – a different usage, here, of the term 'generator'), that serve to define the group:

Cubic $Q_1^4 = Q_2^3 = Q_3^2 = (Q_2Q_3)^2 = (Q_3Q_1)^3 = (Q_1Q_2)^4 = I$, $Q_3 = Q_1Q_2Q_1^2$
Hexagonal $Q_1^6 = Q_2^2 = (Q_1Q_2)^2 = I$
Rhombohedral $Q_1^3 = Q_2^2 = (Q_1Q_2)^2 = I$
Tetragonal $Q_1^4 = Q_2^2 = (Q_1Q_2)^2 = I$
Orthorhombic $Q_1^2 = Q_2^2 = (Q_1Q_2)^2 = I$
Monoclinic $Q_1^2 = I$

In each case, the Q-representation of the whole group consists of all possible products of the matrices we have listed; for the cubic group 432 for example it follows from the relations that these products can all be written in the form $Q_3^{\gamma}Q_2^{\beta}Q_1^{\alpha}$ ($\alpha = 0, 1, 2, 3$; $\beta = 0, 1, 2$; $\gamma = 0, 1$) and analogous expressions for the other Bravais types: all elements are given by $Q_2^{\mu}Q_1^{\nu}$ ($\mu = 0, 1, \nu = 0, \ldots, n-1$, $n = 6, 3, 4, 2$ for hexagonal, rhombohedral, tetragonal, or orthorhombic, respectively).

Equivalence of CSL-producing rotations is now expressible in terms of integral matrices W (4.13): $Q'WQ^{\mathrm{T}}$ and $Q'W^{\mathrm{T}}Q^{\mathrm{T}}$ will lead to the same CSL, where Q and Q' are any matrices belonging to the Q-representation of the lattice's rotational symmetry group. Thus we have a means of obtaining, from a given angle θ and axis h for a CSL of a rational lattice, all other $\{\theta, h\}$ pairs that describe the same orientation relation and, consequently, the same CSL: for each case, employing (4.2) and (4.13).

$$R' = E^{-\mathrm{T}}(Q'WQ^{\mathrm{T}})\,E^{\mathrm{T}} - I, \qquad N \sin\theta = (R - R^{\mathrm{T}})/2$$

(and similarly with W^{T} replacing W) gives the corresponding angle θ and axis $\mathbf{n}$. Then $h \sim \mathbf{n}E$.

If the aim is to find the canonical rotation that is equivalent to a given rotation R, then only g rotations, rather than $2g^2$, need be computed to arrive at the desired result. This we shall demonstrate in the following section after introducing the quaternion representation of rotations.

10.2 Quaternion method

Rotations in three dimensions are very elegantly and conveniently represented by quaternions. (Hamilton, 1844; Tait, 1890; du Val, 1964; Altmann, 1966, Conway, Smith, 1992; etc.). The application of quaternions to the description of reorientations of a cubic lattice and the associated equivalence classes of rotations was introduced by Grimmer (1974a) and extended to other lattice types by Grimmer (1980) and Heinz and Neumann (1991).

The product of two quaternions $p = (p_0, \mathbf{p})$ and $q = (q_0, \mathbf{q})$ is

$$pq = (p_0q_0 - \mathbf{p}\cdot\mathbf{q},\ \mathbf{p}\times\mathbf{q} + p_0\mathbf{q} + q_0\mathbf{p}). \tag{10.2}$$

The *conjugate* of a quaternion $q = (q_0, \mathbf{q})$ is $\bar{q} = (q_0, -\mathbf{q})$. A quaternion q is *unimodular* if $q\bar{q} = e$ where e denotes the unit quaternion (1 0 0 0). Equivalently, a unit quaternion is one for which $q_0^2 + q_1^2 + q_2^2 + q_3^2 = 1$. A rotation R through an angle θ about an axis in the direction of a unit vector $\mathbf{n}$ can be represented by a unimodular quaternion:

$$q = (\cos(\theta/2),\ \mathbf{n}\sin(\theta/2)). \tag{10.3}$$

The angle and axis of the rotation are given by the Rodrigues vector $\mathbf{q}/q_0 = \mathbf{n}\tan(\theta/2)$ (Rodriques, 1840; Altmann, 1989; Heinz, Neumann, 1991). Products of rotations correspond to products of their associated quaternions. For a rotation R that gives rise to a CSL of a rational lattice (4.9) allows us to rewrite this as

$$q \sim (X, \sigma Y\mathbf{n}). \tag{10.4}$$

The symbol $\sim$ denotes the omission of a scalar factor; for the purpose of evaluation of products of rotations scalar factors are irrelevant and can be chosen arbitrarily. This observation considerably simplifies computations and also permits us to impose the restriction that the component q_0 of the quaternions be non-negative, corresponding to $0 \geq \theta \geq \pi$. A method of finding rotations equivalent to a given rotation is now apparent: We simply have to find all the quaternions $s'qs$ corresponding to the rotations $S'RS$. Apart from irrelevant scaling factors the quaternions s that represent the generators of the rotational symmetry groups of the various Bravais lattices can be chosen as follows:

Cubic (1 0 0 1), (1 1 1 1), (0 1 1 0)
Hexagonal ($\sqrt{3}$ 0 0 1), (0 1 0 0)
Rhombohedral (1 0 0 $\sqrt{3}$), (0 1 0 0)
Tetragonal (1 0 0 1), (0 1 1 0)
Orthorhombic (0 0 0 1), (0 1 0 0)
Monoclinic (0 0 1 0)

If the aim is to find the canonical rotation representing the equivalence class to which a given rotation R belongs, it is not necessary to find all rotations of the equivalence class. Moreover, the quaternion methods reveal further simplifications; starting from a given quaternion q representing a rotation θ about an axis $\mathbf{n}$ (10.3), and any quaternion s,

$$q' = sq\bar{s} = (q_0,\ S\mathbf{q}) \tag{10.5}$$

where S is the orthogonal matrix associated with the quaternion s. Thus the rotations associated with q and q' have *the same angle of rotation* – around different axes.

All rotations equivalent to the rotation given by a quaternion q can be expressed as $s(s'q)\,\bar{s}$ where s and s' are quaternions associated with symmetries of Λ. It follows from this and the previous statement that the canonical rotation of the equivalence class can be found by computing the g quaternions $s'q$, picking out the quaternion q' of this set that has the least angle θ, then computing the g quaternions $sq'\bar{s}$ and selecting the one whose vector part lies in the principal fundamental region of the stereogram for the rotation group of Λ. The number of quaternion multiplications required in this search algorithm is abridged by making use of the following properties of the quaternions s that represent 180° rotations (for which $s_0 = 0$). For any quaternions $a = (a_0\ a_1\ a_2\ a_3)$,

$$s \sim (0\ 1\ 0\ 0) \to sa\bar{s} \sim (a_0\ a_1\ -a_2\ -a_3)\,, \quad sa \sim (a_1\ -a_0\ a_3\ -a_2)$$

$$s \sim (0\ 0\ 1\ 0) \to sa\bar{s} \sim (a_0\ -a_1\ a_2\ -a_3)\,, \quad sa \sim (a_2\ -a_3\ -a_0\ a_1)$$

$$s \sim (0\ 0\ 0\ 1) \to sa\bar{s} \sim (a_0\ -a_1\ -a_2\ a_3)\,, \quad sa \sim (a_3\ a_2\ -a_1\ -a_0)\,.$$

We demonstrate the method by means of a simple example — the same example we used in Section 6: the ‘cubic hexagonal lattice’ with $h = (0\ 1\ 1)$ and $X = 3$, $Y = 1$. $\mathbf{n} \sim hE^{-1} \sim (0\ \sqrt{6}\ \sqrt{3})$ so that $\mathbf{n} = (0\ \sqrt{2}\ 1)/\sqrt{3}$ and therefore

$$q \sim (3\ 0\ \sqrt{6}\ \sqrt{3})\,.$$

Finding all equivalent rotations is now simply a matter of multiplying this q, on the right and on the left, by all possible products of the quaternions $s_1 = (\sqrt{3}\ 0\ 0\ 1)$ and $s_2 = (0\ 1\ 0\ 0)$. There are 144 cases. The quaternions s' and s are of the form $s_2{}^{\alpha} s_1{}^{\beta}$ ($\alpha = 0,\ 1;\ \beta = 0,\ 1,\ \dots,\ 5$) and in each case, from $q' = s'qs$ we can extract

$$\tan(\theta/2) = \frac{|\mathbf{q}'|}{q'_0}\,, \qquad h \sim \mathbf{q}'E\,.$$

To simply obtain the canonical rotation rather than the whole equivalence class, the computation goes as follows:

$$\begin{array}{lll}
q & \sim (3\ 0\ \sqrt{6}\ \sqrt{3}) & \to \tan(\theta/2) = 1 \\
s_1q & \sim (2\ \sqrt{3}\ -\sqrt{6}\ 3\ \sqrt{2}\ 6) & \to \tan(\theta/2) = \sqrt{5} \\
s_1{}^2q & \sim (0\ -3\ \sqrt{2}\ \sqrt{6}\ 4\ \sqrt{3}) & \to \tan(\theta/2) = \infty \\
s_1{}^3q & \sim (\sqrt{3}\ \sqrt{6}\ 0\ -3) & \to \tan(\theta/2) = \sqrt{5} \\
s_1{}^4q & \sim (6\ 3\ \sqrt{2}\ \sqrt{6}\ -2\ \sqrt{3}) & \to \tan(\theta/2) = 1 \\
s_1{}^5q & \sim (4\ \sqrt{3}\ \sqrt{6}\ 3\ \sqrt{2}\ 0) & \to \tan(\theta/2) = 1/\sqrt{2} \\
 & & \\
s_2q & \sim (0\ -3\ \sqrt{3}\ -\sqrt{6}) & \to \tan(\theta/2) = \infty \\
s_2s_1q & \sim (\sqrt{6}\ 2\ \sqrt{3}\ -6\ 3\ \sqrt{2}) & \to \tan(\theta/2) = \sqrt{11} \\
s_2s_1{}^2q & \sim (3\ \sqrt{2}\ 0\ -4\ \sqrt{3}\ \sqrt{6}) & \to \tan(\theta/2) = \sqrt{3} \\
s_2s_1{}^3q & \sim (\sqrt{6}\ -\sqrt{3}\ -3\ 0) & \to \tan(\theta/2) = \sqrt{2} \\
s_2s_1{}^4q & \sim (3\ \sqrt{2}\ -6\ -2\ \sqrt{3}\ -\sqrt{6}) & \to \tan(\theta/2) = \sqrt{3} \\
s_2s_1{}^5q & \sim (\sqrt{6}\ -4\ \sqrt{3}\ 0\ -3\ \sqrt{2}) & \to \tan(\theta/2) = \sqrt{11}
\end{array}$$

The computation of the last six quaternions on the list is facilitated by noting that for any quaternion $a = (a_0\ a_1\ a_2\ a_3)$, $s_2a \sim (a_1\ -a_0\ a_3\ -a_2)$. The quaternion in this list that gives the smallest (positive) angle θ is $q' = s_1{}^5q$. We then compute all quaternions $sq'\bar{s}$ (for all of which, $\tan(\theta/2) = 1/\sqrt{2}$):

$$\begin{array}{lll}
q' & \sim (4\ \sqrt{3}\ \sqrt{6}\ 3\ \sqrt{2}\ 0) & \to \varkappa = (1\ 1\ 0) \\
s_1q's_1{}^5 & \sim (4\ \sqrt{3}\ -\sqrt{6}\ 3\ \sqrt{2}\ 0) & \to \varkappa = (0\ 1\ 0) \\
s_1{}^2q's_1{}^4 & \sim (\sqrt{6}\ -\sqrt{3}\ 0\ 0) & \to \varkappa = (\bar{1}\ 0\ 0) \\
s_1{}^3q's_1{}^3 & \sim (4\ \sqrt{3}\ -\sqrt{6}\ -3\ \sqrt{2}\ 0) & \to \varkappa = (\bar{1}\ \bar{1}\ 0) \\
s_1{}^4q's_1{}^2 & \sim (4\ \sqrt{3}\ \sqrt{6}\ -3\ \sqrt{2}\ 0) & \to \varkappa = (0\ \bar{1}\ 0) \\
s_1{}^5q's_1 & \sim (\sqrt{6}\ \sqrt{3}\ 0\ 0) & \to \varkappa = (1\ 0\ 0) \\
s_2q's_2 & \sim (4\ \sqrt{3}\ \sqrt{6}\ -3\ \sqrt{2}\ 0) & \to \varkappa = (0\ \bar{1}\ 0) \\
s_2s_1q's_1{}^5s_2 & \sim (4\ \sqrt{3}\ -\sqrt{6}\ -3\ \sqrt{2}\ 0) & \to \varkappa = (\bar{1}\ \bar{1}\ 0) \\
s_2s_1{}^2q's_1{}^4s_2 & \sim (\sqrt{6}\ -\sqrt{3}\ 0\ 0) & \to \varkappa = (\bar{1}\ 0\ 0) \\
s_2s_1{}^3q's_1{}^3s_2 & \sim (4\ \sqrt{3}\ -\sqrt{6}\ 3\ \sqrt{2}\ 0) & \to \varkappa = (0\ 1\ 0) \\
s_2s_1{}^4q's_1{}^2s_2 & \sim (4\ \sqrt{3}\ \sqrt{6}\ 3\sqrt{2}\ 0) & \to \varkappa = (1\ 1\ 0) \\
s_2s_1{}^5q's_1s_2 & \sim (\sqrt{6}\ \sqrt{3}\ 0\ 0) & \to \varkappa = (1\ 0\ 0)
\end{array}$$

The computation of the last six quaternions on the list is facilitated by noting that for any quaternion $a = (a_0\ a_1\ a_2\ a_3)$, $s_2as_2 \sim (a_0\ a_1\ -a_2\ -a_3)$. The quaternion of this list that satisfies the criterion $0 \le 2\varkappa_2 \le \varkappa_1$, $0 \le \varkappa_3$ for $\mathbf{n}$ to lie in the principal region of the spherical representation for the hexagonal lattice is $(\sqrt{6}\ \sqrt{3}\ 0\ 0) \sim (\sqrt{2}\ 1\ 0\ 0)$, which gives $\mathbf{n} = (1\ 0\ 0)$, $h = (2\ \bar{1}\ 0)$.

11. Conclusions

The algorithmic method developed by Niggli and Buerger for finding a ‘reduced’ unit cell for a given lattice can be applied to any given set of generating vectors that serve to define the lattice; the generating vectors do not need to be linearly independent. The method can therefore be applied to the union of the sets of generators for two lattices, leading to a ‘reduced cell’ for the displacement shift lattice. Grimmer’s reciprocity theorem then gives immediately the corresponding CSL. This very simple approach to the coincidence site problem leads to an equally simple and illuminating approach to the concept of relative orientations and equivalence classes of rotations.

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References

Altmann, S. L.: Rotations, quaternions and double groups. Clarendon Press, Oxford (1966).

Altmann, S. L.: Hamilton, Rodrigues, and the quaternion scandal. Mathematics Magazine **62** (1989) 291–308.

Baake, M.; Pleasants, A. B.: Algebraic solution of the coincidence problem in two and three dimensions. Z. Naturforsch. **50a** (1995) 711–717.

Baake, M.: Solution of the coincidence problem in dimensions ≤4. Mathematics of Aperiodic Order (ed. Moody, R. V). Kluwer, Dordrecht (1996).

Baake, M.: Coincidence rotations for icosahedral patterns. In: *New Horizons in Quasicrystals: Research and Applications.* Ames, Iowa 19–23 August 1996. (Eds. Goldman, A. I.; Sordelet, D. J.; Thiel, P. A.) World Scientific, Singapore (1997).

Bollmann, W.: Crystal Defects and Crystalline Interfaces. Springer, Berlin (1970).

Bollmann, W.: Continuous distribution of dislocations in intercrystalline boundaries. Acta Cryst. A**33** (1977) 730–739.

Buerger, M. J.: Reduced cells. Z. Kristallogr. **109** (1957) 42–60.

Conway, J. H.; Smith, D. A.: On Quaternions and Octonions. AK Peters Ltd (1992).

Coxeter, H. S. M.; Moser, W. O. J.: Generators and Relations for Discrete Groups. Springer (1957).

de Wolff, P. M.; Gruber, B.: Niggli lattice characters: definition and graphical representation. Acta. Cryst. A**47** (1991) 29–36.

Du Val, P.: Homographies, Quaternions, and Rotations. Oxford University Press (1964).

Fortes, M. A.: N-dimensional coincidence-site-lattice theory. Acta. Cryst. A**39** (1983) 351–357.

Frank, F. C.: On Miller-Bravais indices and four-dimensional vectors. Acta Cryst. **18** (1965) 862–866.

Gómez, A.; Beltrán, J. L.; Rameu, D.: A generalized coincidence-site network model for grain boundaries. Scripta Materialia **38** (1998) 795–801.

Grimmer, H.: Coincidence rotations for cubic lattices. Scripta Metallurgica **7** (1973) 1295–1300.

Grimmer, H.: Disorientation and coincidence rotations for cubic lattices. Acta. Cryst. A**30** (1974a) 685–688.

Grimmer, H.: A reciprocity relation between the coincidence site lattice and the DSC lattice. Scripta Metallurgica **8** (1974b) 1221–1223.

Grimmer, H.; Bollmann, W.; Warrington, D. H.: Coincidence-site lattices and complete pattern-shift lattices in cubic crystals. Acta Cryst. A**30** (1974) 199–205.

Grimmer, H.: A unique description of the relative orientation of neighbouring grains. Acta Cryst. A**36** (1980) 382–389.

Grimmer, H.: Tables of all coincidence orientations with low multiplicity for arbitrary hexagonal and rhombohedral lattices. Scripta Metallurgica **23** (1989a) 1407–1412.

Grimmer, H.: Coincidence orientation of grains in rhombohedral materials. Acta Cryst. A**45** (1989b) 505–523.

Grosse-Kunstleve, R. W.; Sauter, N. K.; Adams, P. D.: Numerically stable algorithm for the computation of reduced unit cells. Acta Cryst. A**60** (2004) 1–6.

Gruber, B.: The relationship between reduced cells in a general Bravais lattice. Acta Cryst. A**29** (1973) 433–440.

Gruber, B.: Topological approach to the Niggli lattice characters. Acta Cryst. A**48** (1992) 461–470.

Hamilton, W. R.: On quaternions; or a new system of imaginaries in algebra. Phil. Mag. **25** (1844) 489–494.

Heinz, A.; Neumann, P.: Representation of orientation and disorientation data for cubic, hexagonal, tetragonal and orthorhombic crystals. Acta Cryst. A**47** (1991) 780–789.

Iwasaki, Y.: General theory of coincidence-site lattices, reduced 0-lattices and complete pattern-shift lattices in arbitrary crystals. Acta Cryst. A**32** (1976) 59–65.

Janner, A.: Integral lattices. Acta Cryst. A**60** (2004a) 198–200.

Janner, A.: Zones and sublattices of integral lattices. Acta Cryst. A**60** (2004b) 611–620.

Krívý, I.; Gruber, B.: A unified algorithm for determining the reduced (Niggli) cell. Acta. Cryst. A**32** (1976) 297–298.

Mighell, A. D.; Rodgers, J. R.: Lattice symmetry determination. Acta Cryst. A**36** (1980) 321–326.

Mykura, H.: A checklist of cubic coincidence site lattice relationships. Grain Boundary Structure and Kinetics: 1979 ASM Materials Science Seminar. American Society for Metals (1979).

Niggli, P.: Handbuch der Experimentalphysik (Vol. 7 part 1). Akademische Verlagsgesellschaft (1928).

Pleasants, A. B.; Baake, M.; Roth, J.: Planar coincidences for N-fold symmetry. J. Math. Phys. **37** (1996) 1029–1058.

Proult, A.; Redjaimia, A.; Donnadieu, P.: Interface between simple crystal and icosahedral-symmetry related crystal. In: *38ème Colloque de Métallurgie de l'INSTN*. (Journal de Physique IV **6**, Colloque C2) (1996).

Ranganathan, S.: On the geometry of coincidence-site lattices. Acta Cryst. **21** (1966) 197–199.

Ranganathan, S.; Ramakrishnan, K.; Kulkarni, U. D.; Mukhopadhyay, N. K.: On the relationship between cubic crystalline coincidence site lattices and quasiperiodic superlattices. Mater. Sci. Eng A**294–296** (2000) 429–433.

Ranganathan, S.; Singh, A.; Tsai, A. P.: Frank's 'cubic' hexagonal phase: an intermetallic cluster compound as an example. Phil. Mag. Lett. **82** (2002) 13–19.

Reed, B. W.; Minich, R. W.; Rudd, R. E.; Kumar, M.: The structure of the cubic coincidence-site rotation group. Acta Cryst. A**60** (2004) 263–277.

Rodrigues, O.: Des lois géométriques qui régissent les déplacements d'un systême solide dans l'espace, et la variation des coordonnées de ses deplacements considérés indépendamment des causes qui peuvent les produire. J. de Mathématiques Pures et Appliquées **5** (1840) 390–440.

Santoro, A.; Mighell, A. D.: Determination of reduced cells. Acta Cryst. A**26** (1970) 124–127.

Santoro, A.; Mighell, A. D.: Coincidence-site lattices. Acta Cryst. A**29** (1973) 169–175.

Tait, P. G.: An Elementary Treatise on Quaternions. (3rd edn) Cambridge University Press (1890).

Warrington, D. H.; Lück, R.: Rotational space and coincidence-site lattices for icosahedral symmetry. Mater. Sci. Forum **207–209** (1996) 825–828.

Zeiner, P.: Symmetries of coincidence site lattices of cubic lattices. Z. Kristallogr. **220** (2005) 915–925.

Zeiner. P.: Coincidences of hypercubic lattices in 4 dimensions. Z. Kristallogr. **221** (2006) 115–114.

Zhang, W.-Z.: Application of the DSCL in reciprocal space for the study of coincidence boundaries. Scripta Materialia **37** (1997) 187–92.

Zuo, L.; Muller, J.; Philippe, M.-J.; Esling, C.: A refined algorithm for the reduced cell determination. Acta. Cryst. A**51** (1995) 943–945.