Reciprocity relations for Bollmann's o-lattice

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A reciprocity relation for Bollmann's O-lattice is introduced. This result completes the existing Grimmer's reciprocity results between coincidence sites and displacement shift complete lattices. We show that the lattice generated by $a_i^* - b_i^*$ $(i = 1, 2, 3)$ is reciprocal to the O-lattice. This result, supported by Multislice calculations, indicates that it is possible to observe the O-lattice under an electron microscope using annular apertures, thus allowing the study of strain fields existing in interfaces or between a thin film growing onto a crystalline substrate.

Keywords: O-lattices; grain boundaries.

Se presenta una relación de reciprocidad para las redes O de Bollmann. Este resultado complementa el obtenido por Grimmer quien estableció relaciones de reciprocidad entre las redes de coincidencia y las redes DSC. Demostramos que la red generada por $a_i^* - b_i^*$ $(i = 1, 2, 3)$ es recíproca a la red O. Este resultado, apoyado en simulaciones por el método multicapas, indica que podría ser factible observar la red O en el microscopio electrónico usando aperturas anulares, permitiendo con ello el estudio de los campos de esfuerzos en interfaces o entre una película delgada y el sustrato sobre el que crece.

Descriptores: Redes O; fronteras de grano.

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1. Introduction

For the geometric analysis of the interfaces between crystals, several geometrical constructs have been introduced, among which the most useful are, probably, the Coincidence Site lattice (CSL), the DSC lattice, and Bollmann's O-lattice, which can be quickly defined as follows:

- Given two lattices $\mathcal{L}_1$ and $\mathcal{L}_2$, the CSL is the intersection lattice, $\mathcal{L}_1 \cap \mathcal{L}_2$ [1]. In other words, the CSL is the lattice formed by all points in both lattices. In this communication, it will be assumed that the two lattices have a rational orientation relationship, so the CSL is a discrete lattice.
- The DSC lattice is defined as the sum of the lattices ($\mathcal{L}_1 + \mathcal{L}_2 = \{a + b \mid a \in \mathcal{L}_1 \, , \, b \in \mathcal{L}_2\}$). In other words, the DSC lattice is the lattice formed by vectors that are sums (or differences) of points in the two lattices.
- The Bollmann's O-lattice [2], which is formally defined below, represents points of a good geometrical fit between the two lattices.

The CSL has been studied, among others, for lattices and modules by Baake and Pleasants [3], by Baake [4,5] and also by Warrington, Radulescu and Lück [6] and Radulescu [7]. A useful review of O-lattices has been presented by Smith and Pond [8].

Many works on grain boundaries use these concepts in various ways, but the main objective has been to provide a geometrical background to aid the solution of a still unresolved fundamental problem: to find a relation between internal structure (grain boundary crystallography) and physical properties.

Along these lines, Grimmer [9] showed that the DSC and the CSL satisfy certain reciprocity relations. Roughly speaking, the CSL and the DSC are reciprocal to each other (a more precise formulation is given below). This is a fundamental result since it relates observable (diffraction) information to structural properties of interfaces.

In the present work (restricting the attention basically to two dimensions since our main interest lies in boundaries and epitaxy), we show that reciprocity relations equivalent to those found by Grimmer exist for O-lattices. These relationships show that O-lattices, or rather, regions of good and bad fit between two crystals can be directly observed under the electron microscope. This is important not only in the field of interfaces, but also in problems of epitaxy and in the study of the strain fields existing in thin films growing on crystalline substrates. Multislice simulations are shown to illustrate this point. Also, the reported relationships open up the possibility of experimentally confirming or discarding models of interface structure.

2. O-lattices

Let $\mathcal{L}_1$ and $\mathcal{L}_2$ be two three-dimensional lattices with a common origin, spanned by $\alpha = \{\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3\}$ and $\beta = \{\mathbf{a}_4, \mathbf{a}_5, \mathbf{a}_6\}$, respectively.

Given a vector $\mathbf{P} \in R^3$, there exists a unique $\mathbf{l}_1$ in $\mathcal{L}_1$ and numbers $0 \leq \alpha_i < 1$, $(i = 1, 2, 3)$ such that

$$\mathbf{P} = \mathbf{l}_1 + \sum_{i=1}^{3} \alpha_i \, \mathbf{a}_i .$$

The (integer) coordinates of $\mathbf{l}_1$ are called "external coordinates of $\mathbf{P}$" (with respect to $\mathcal{L}_1$) and the numbers α_i, $(i = 1, 2, 3)$ are called "internal coordinates of $\mathbf{P}$" (with respect to $\mathcal{L}_1$). The external and internal coordinates of $\mathbf{P}$ with respect to $\mathcal{L}_2$ are defined in a similar way [2, 8]. A point (or vector) $\mathbf{O}$ is called an O-point if it has the same internal coordinates with respect to both $\mathcal{L}_1$ and $\mathcal{L}_2$.

If A is a linear mapping such that $A(\mathbf{a}_i) = \mathbf{a}_{i+3}$, $(i = 1, 2, \ldots, 3)$, then it can be shown [2,8] that $\mathbf{O}$ is an O-point if and only if it satisfies

$$(I - A^{-1})\mathbf{O} = \mathbf{l}_1 \tag{1}$$

for some $\mathbf{l}_1 \in \mathcal{L}_1$ or, equivalently,

$$(I - A)\mathbf{O} = \mathbf{l}_2 \tag{2}$$

for some $\mathbf{l}_2 \in \mathcal{L}_2$; I represents the identity mapping. If the inverse of $(I - A)$ [or, equivalently, the inverse of $(I - A^{-1})$] exists, then the O-points form a point lattice.

It is important to notice that the O-lattice depends on $\mathcal{L}_1$,$\mathcal{L}_2$ and a choice of bases α and β or, alternatively, a choice of a linear transformation A mapping $\mathcal{L}_1$ into $\mathcal{L}_2$.

3. Reciprocity relations for $\mathcal{L}_1+\mathcal{L}_2$ and $\mathcal{L}_1\cap\mathcal{L}_2$

In what follows, let $\mathcal{L}_1$ and $\mathcal{L}_2$ be the two lattices spanned by $\alpha = \{a_1, a_2, a_3\}$ and $\beta = \{b_1, b_2, b_3\}$, respectively, and let $\mathcal{L}_1^*$ and $\mathcal{L}_2^*$ be the corresponding reciprocal lattices spanned by $\alpha^* = \{a_1^*, a_2^*, a_3^*\}$ and $\beta^* = \{b_1^*, b_2^*, b_3^*\}$.

Grimmer [9] has shown that:

$$\begin{aligned} \mathcal{L}_1 + \mathcal{L}_2 &= (\mathcal{L}_1^* \cap \mathcal{L}_2^*)^* \\ \mathcal{L}_1 \cap \mathcal{L}_2 &= (\mathcal{L}_1^* + \mathcal{L}_2^*)^* \end{aligned} \tag{3}$$

This result is absolutely general and it does not depend at all on the choice of lattice bases or structure matrices.

4. A reciprocity relation for the O-lattice

Theorem: Let $\mathcal{L}_1$ and $\mathcal{L}_2$ be two lattices spanned by $\alpha = \{a_1, a_2, a_3\}$ and $\beta = \{b_1, b_2, b_3\}$, respectively, and let the corresponding reciprocal lattices $\mathcal{L}_1^*$ and $\mathcal{L}_2^*$ be spanned by $\alpha^* = \{a_1^*, a_2^*, a_3^*\}$ and $\beta^* = \{b_1^*, b_2^*, b_3^*\}$ respectively. Let $\mathcal{O}(\mathcal{L}_1, \mathcal{L}_2)$ be the O-lattice generated by $\mathcal{L}_1$ and $\mathcal{L}_2$ (and with respect to the given bases). If $\mathcal{O}^*(\mathcal{L}_1, \mathcal{L}_2)$ is the lattice reciprocal to $\mathcal{O}(\mathcal{L}_1, \mathcal{L}_2)$, we have that

$$\mathcal{O}^*(\mathcal{L}_1, \mathcal{L}_2) = \operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)$$

(here $\operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)$ is the lattice generated by $\{a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*\}$).

Proof: Let $P \in \mathcal{O}(\mathcal{L}_1, \mathcal{L}_2)$, so

$$\begin{aligned} P &= h_1 a_1 + h_2 a_2 + h_3 a_3 + x_1 a_1 + x_2 a_2 + x_3 a_3 \\ &= h_1' b_1 + h_2' b_2 + h_3' b_3 + x_1 b_1 + x_2 b a_2 + x_3 b a_3, \end{aligned}$$

where $h_1, h_1', h_2, h_2', h_3$ and h_3' are integers (external coordinates) and $x_1, x_2, x_3 \in [0, 1)$ (internal coordinates). Then, for $i = 1, 2, 3$:

$$\begin{aligned} &P\cdot(a_i^* - b_i^*) \\ &= P \cdot a_i^* - P \cdot b_i^* \\ &= (h_1 a_1 + h_2 a_2 + h_3 a_3 + x_1 a_1 + x_2 a_2 + x_3 a_3) \cdot a_i^* \\ &\quad - (h_1' b_1 + h_2' b_2 + h_3' b_3 + x_1 b_1 + x_2 b_2 + x_3 b_3) \cdot b_i^* \\ &= h_i + x_i - (h_i' + x_i) = h_i - h_i'; \end{aligned}$$

so

$$P \cdot (a_i^* - b_i^*) = h_i - h_i' \in \mathbb{Z},$$

meaning that

$$P \in \operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)^*,$$

where $\operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)^*$ is the lattice reciprocal to $\operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)$.

Conversely, if $P \in \operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)^*$, then for $i = 1, 2, 3$

$$P \cdot (a_i^* - b_i^*) \in \mathbb{Z};$$

and if

$$\begin{aligned} P &= h_1 a_1 + h_2 a_2 + h_3 a_3 + x_1 a_1 + x_2 a_2 + x_3 a_3 \\ &= h_1' b_1 + h_2' b_2 + h_3' b_3 + x_1' b_1 + x_2' b a_2 + x_3' b a_3 \end{aligned}$$

with $h_1, h_1', h_2, h_2', h_3$ and h_3' integers (external coordinates), and

$$x_1, x_2, x_3, x_1', x_2', x_3' \in [0, 1)$$

(internal coordinates) we have that

$$\begin{aligned} P \cdot a_i^* &= h_i + x_i \\ P \cdot b_i^* &= h_i' + x_i' \end{aligned}$$

and

$$P \cdot (a_i^* - b_i^*) = (h_i - h_i') + (x_i - x_i') \in \mathbb{Z},$$

implying that

$$(x_i - x_i') \in \mathbb{Z}$$

and

$$-1 < (x_i - x_i') < 1$$

and, finally, that

$$x_i = x_i';$$

so

$$P \in \mathcal{O}(\mathcal{L}_1, \mathcal{L}_2),$$

thus establishing that $\mathcal{O}(\mathcal{L}_1, \mathcal{L}_2)$ and $\operatorname{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*)$ are reciprocal to each other.

5. A matrix approach

Here we provide a simple, alternative, matrix proof of the fact that, if $\mathcal{O}(\mathcal{L}_1, \mathcal{L}_2)$ is the O-lattice generated by $\mathcal{L}_1$ and $\mathcal{L}_2$ (with respect to the given bases), and if $\mathcal{O}^*(\mathcal{L}_1, \mathcal{L}_2)$ is the lattice reciprocal to $\mathcal{O}(\mathcal{L}_1, \mathcal{L}_2)$, then we have, as before, that

$$\mathcal{O}^*(\mathcal{L}_1, \mathcal{L}_2) = \mathrm{gen}(a_1^* - b_1^*, a_2^* - b_2^*, a_3^* - b_3^*).$$

The structure matrix S for lattice $\mathcal{L}_1$ is defined as that matrix having as columns the coordinates of the generators $\{a_1, a_2, a_3\}$ with respect to a given, fixed, orthonormal frame of reference. The structure matrix S' for $\mathcal{L}_2$ is defined analogously.

It is well known that the structure matrices for the reciprocal lattices are given by $S^* = (S^T)^{-1}$ and $S'^* = (S'^T)^{-1}$, respectively; here S^T denotes the transpose matrix.

If the matrix A maps lattice one into lattice two (so $S' = AS$), Bollmann's basic equation states that the structure matrix O for the O-lattice satisfies (provided the inverse exists, when it doesn't pseudoinverses can be used as shown in [11])

$$O = (I - A^{-1})^{-1} S$$

so that

$$\begin{aligned} O^{T(-1)} &= (I - A^{-1})^T S^{T(-1)} \\ &= (I - A^{T(-1)}) S^{T(-1)} \end{aligned} \tag{4}$$

and, calling $S^* = S^{T(-1)}$ and $O^* = O^{T(-1)}$, we have that

$$\begin{aligned} O^* &= (I - A^{T(-1)}) S^* \\ &= S^* - A^{T(-1)} S^*. \end{aligned} \tag{5}$$

However, if $S' = AS$ then

$$S'^{T(-1)} = S'^* = A^{T(-1)} S^{T(-1)} = A^* S^*$$

and

$$O^* = S^* - S'^*$$

thus proving our theorem.

In Figs. 1 and 2 we illustrate this result graphically for two-dimensional lattices. In Fig. 1 one can appreciate two square lattices; the O-lattice is also shown. In Fig. 2 the corresponding reciprocal lattices are shown together with the basis vectors for all the lattices involved.

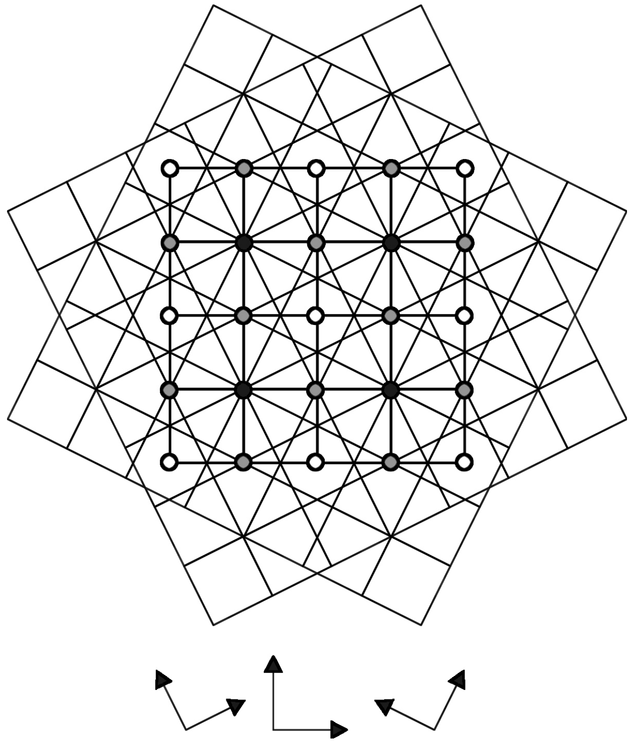

FIGURE 1. Two identical square lattices rotated 36.9 degrees with respect to each other ($\Sigma 5$). The corresponding O-lattice is also displayed with shaded circles. Each shade corresponds to a set of internal coordinates, black=$\{0, 0\}$ CSL), grey=$\{0, 1/2\}$, white=$\{1/2, 1/2\}$. The basis vectors for L_1, L_2 and the O-lattice are shown.

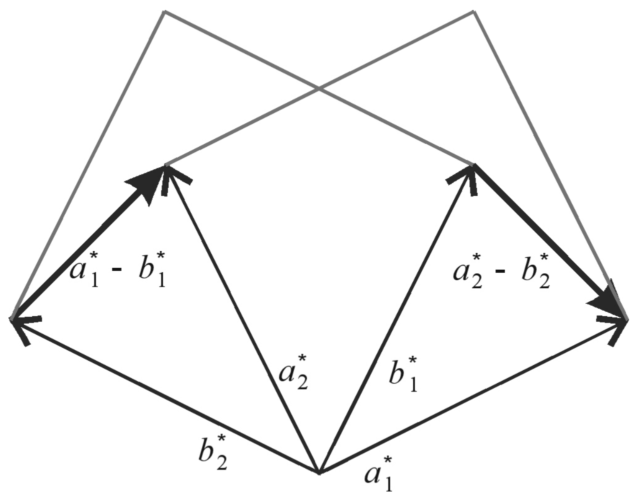

FIGURE 2.

6. Observing O-lattices under the electron microscope

Consider two crystals on top of each other with lattices $\mathcal{L}_1$ and $\mathcal{L}_2$. Under an electron microscope, the diffraction pattern will show spots corresponding to $\mathcal{L}_1^*$ and $\mathcal{L}_2^*$, and also some spots from $\mathcal{L}_1^* + \mathcal{L}_2^*$ (*i.e.* double diffraction spots). If an annular aperture is placed so that it includes spots of the form $\pm a_i^*, \pm b_i^*$ $(i = 1, 2)$, then the resulting image will be basically (*i.e.* in a kinematical approximation) a mapping of the O-lattice.

The previous statement calls for further clarification. There will be interference between beams a_i and b_j $(i, j = 1, 2)$, and we have the same situation as when observing the beating between various waves. However, the modulations with greater wavelengths (analogous to the beating) correspond to the combinations $a_i - b_j$ of shortest length. These will dominate the image, so that we will "see" one of the possible O-lattices; in a way we have selected one of

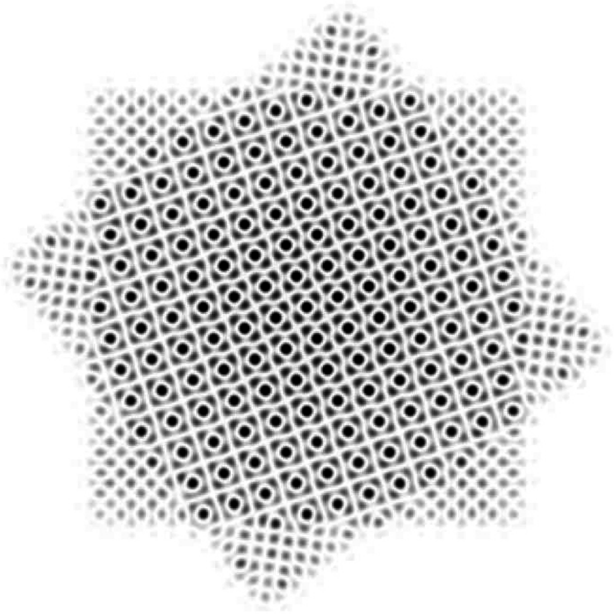

FIGURE 3. Multislice simulation (shown in reversed contrast) of two identical square lattices rotated 36.9° with respect to each other ($\Sigma5$). The dark wide spots mark the positions of O-lattice points.

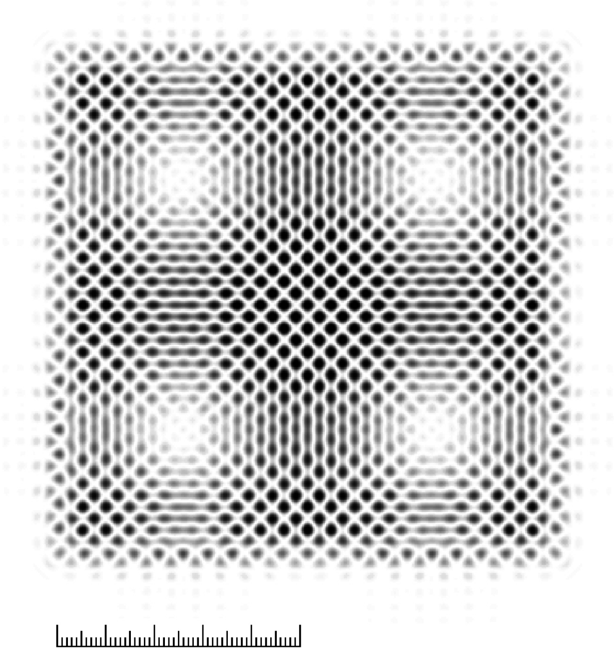

FIGURE 4. Multislice simulation of two parallel square lattices with lattice parameters differing by 10 per cent. An annular aperture was used (the contrast has been reversed for purposes of clarity).

them. This is completely analogous to what one observes in the so-called Moiré patterns; actually, Bollmann was well aware of the close relationship between O-lattices and Moiré patterns [2].

In order to illustrate this, we have used the Multislice program SimulaTem [10] to compute diffraction patterns and images from two superimposed, two-dimensional square crystals of gold misoriented 36.87 degrees in order to simulate the well known $\Sigma5$ CSL. We have used an annular aperture, admitting the strongest reflections from both crystals having wave-vectors between 0.2 Å^{-1} and 0.3 Å^{-1}.

The resulting image for Scherzer defocus is shown in Fig. 3 together with a purely geometrical representation of the O-lattice, making clear a complete agreement.

It is therefore possible to directly observe O-lattices using annular apertures.

Another case of importance is that of epitaxy; in Fig. 4 we again show two square lattices, but this time one of them has a lattice parameter 10 per cent larger than the other. Again the O-lattice can be clearly resolved with the annular aperture.

7. Conclusions

We have extended Grimmer's reciprocity results [9] between CSL and DSC lattices. In this paper, we have presented a new reciprocity relation for the O-lattice in which the reciprocal lattice to the O-lattice is the lattice generated by $a_i^* - b_i^*$, $(i = 1, 2, 3)$.

The similarities and differences between the CSL and the O-Lattice can be stated succinctly by saying that P is the in O-lattice iff $P \cdot (a_i^* - b_i^*) \in \mathbb{Z}$ for all $i = 1, 2, \ldots, n$, whereas P is in the CSL iff $P \cdot (a^* - b^*) \in \mathbb{Z}$ for all $a \in \mathcal{L}_1^*, b \in \mathcal{L}_2^*$

According to our results, supported by Multislice calculations, it seems possible to observe the O-lattice under an electron microscope using annular apertures, which offers the possibility of studying the strain fields generated during thin film growth on a crystalline substrate.

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