Computer simulation of convergent-beam electron diffraction patterns from bicrystals. By N. S. BLOM and F. W. SCHAPINK, Laboratory of Metallurgy, Delft University of Technology, Rotterdamseweg 137, 2628 AL Delft, The Netherlands

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Abstract

A computer program for simulating convergent-beam electron diffraction patterns from single crystals and bicrystals is described. The program is based on the dynamical theory of electron diffraction, and the intensity of a convergent-beam disc is constructed from the individual intensities of a number of plane waves incident on the specimen. For the bicrystal case, this program allows the influence of parameters such as the location of the boundary plane and a rigid translation at the boundary to be investigated. These effects are discussed for a horizontal (111) twin boundary in silicon.

1. Introduction

Convergent-beam electron diffraction (CBED) is a well-known technique for determining the point- and space-group symmetry of single crystals (Goodman, 1975; Tinnapel, 1975; Buxton, Eades, Steeds & Rackham, 1976). Recently an extension to the determination of bicrystal symmetry has been proposed (Schapink, Forghany & Buxton, 1983; Buxton, Forghany & Schapink, 1984). It has been shown that for this purpose bicrystal symmetry is best classified by one of the 58 possible dichromatic point groups obtained from the interpenetrating pattern (or complex) of the lattices (or structures) of both crystals. The symmetry of a bicrystal is obtained from the dichromatic pattern by an appropriate sectioning procedure (Pond & Vlachavas, 1983). The bicrystal is assumed to consist of a thin parallel-sided specimen with the boundary coincident with the mid-plane of the specimen (Fig. 1). For this idealized geometry of a bicrystal specimen the interrelation between dichromatic point groups and diffraction groups, characterizing the CBED pattern symmetry, has been established.

However, in practice deviations from the idealized boundary position assumed above will often occur, and so far the effects of such deviations on CBED patterns have not been investigated. Accordingly we have set up a computer program for simulating CBED patterns of bicrystals, based upon the N-beam dynamical theory of electron diffraction including higher-order Laue-zone effects. Such a simulation enables not only the effects of boundary location to be investigated in detail, but also, for example, changes in CBED patterns due to a rigid translation at the boundary. It is the purpose of this paper to describe this program and discuss some of the results obtained.

2. Theory

The program for simulating CBED patterns to be described here is based on the dynamical theory of electron diffraction originally developed by Bethe (1928). For a detailed discussion of the theory the reader is referred to Humphreys (1979). Using Humphreys' notation the theory results in a set of equations of the type

\[ \{K^2-(k^{(j)}+g)^2\}C_{g}^{(j)}+\sum_{h\neq n}U_{n-h}C_{h}^{(j)}=0, \]

(1)

one for each reflection \( g \) considered. Here \( K \) is the magnitude of the mean incident electron wave vector corrected for refraction, \( k^{(j)} \) is the wave vector for the \( j \)th Bloch wave, \( C_{g}^{(j)} \) is the \( g \)th Fourier coefficient of the \( j \)th Bloch wave and \( U_{n} \) is the \( n \)th Fourier coefficient of the reduced potential \( U(r)=(2mlel/h^2)V(r) \). Since we wish to include higher-order Laue zone (HOLZ) reflections, we write

\[ K^2-(k^{(j)}+g)^2=K^2-k_{z}^{(j)}-2k_{z}^{(j)}g_{z}, \]

(2)

for those reflections, neglecting a term containing \( g_{z}^2 (g_{z}<<k_{z}) \). Here \( k_{z} \), \( g_{z} \) and \( k_{z} \) are the \( z \) components (parallel to the surface normal) and the tangential components of \( k \) and \( g \), respectively. The component \( k_{z} \) is taken along the direction of the vector \( g-g_{s} \) where \( s \) is the surface normal. It should be noted that omitting the higher-order term \( 2k_{z}^{(j)}g_{z} \) yields the projection approximation. Substitution of (2) in (1) represents an eigenvalue problem of a hermitian square matrix resulting in a set of eigenvectors \( C^{(j)} \) = \( k^{(j)} + K \) and eigenvectors \( C^{(j)} \). The amplitude of the \( g \)th diffracted beam is then calculated from

\[ \psi_{g}(t)=\sum_{j}C_{j}^{(j)*}C_{g}^{(j)}\exp 2\pi i(k_{z}^{(j)}+g_{z}t), \]

(3)

where \( t \) is the specimen thickness. Equation (3) does not contain a summation over \( g_{z} \), since in our case the set of \( N \) reflections is chosen in such a way that none of them are
related by a reciprocal translation along the zone axis (Jones, Rackham & Steeds, 1977; Buxton, 1976). Absorption, including the contribution from HOLZ reflections, can be introduced by perturbation methods as described by Humphreys (1979), resulting in
\[ v_\phi(t) = \sum_j C_0^j* C_\phi^j \exp[2\pi i (h_x^j + g_x^j) t] \exp(-2\pi q^j(t)) \]
with
\[ q^j = \sum_k \left[ U_{g-h/2} C_g^j C_h^j \right] \]

In order to apply the above theory to the calculation of a CBED pattern from a bicrystal, the following extensions have to be made. Firstly, the simulation should be carried out for a large number of plane incident waves with tangential components \( k_x \). The different components \( k_x \) can be said to form a grid, the spacing of which determines the amount of detail obtainable in the convergent-beam discs. In the actual computations it is assumed that there is no phase relationship between the plane waves with different \( k_x \), i.e. the electron source is assumed to be perfectly incoherent. This implies that the intensity in each point of a convergent-beam disc is uniquely determined by the scattering of a single incident wave. For computation purposes both a rectangular grid as well as a radial grid have been implemented, of which the latter most clearly reveals the symmetry in the CBED patterns.

Secondly, electron diffraction in a bicrystal specimen can be described in two ways. If the individual lattices of both crystals are chosen, the diffracted beams leaving the first crystal should be treated as incoming waves in the second crystal (Sheinin & Corbett, 1976; Sutton & Pond, 1978). For a single incident wave this procedure involves the solution of one eigenvalue problem of dimension \( N^1 \), for the first crystal and \( N^2 \) eigenvalue problems of dimension \( N^2 \) for the second crystal, if \( N^1 \) and \( N^2 \) reflections are considered in the first and second crystals, respectively. Since the computation time for a single \( N \)-beam eigenvalue equation is proportional to \( N^3 \), the total computation time in this case is proportional to \( N^3 + N^1 N^2 \).

Alternatively, electron diffraction in a bicrystal can be treated on the basis of the coincidence site lattice (CSL) common to both crystals. In this case, of course, the lattice is continuous across the interface, whereas the location of atoms in the unit cell is different in both crystals. The advantage is that the reciprocal lattice of the CSL contains all possible diffraction vectors relevant to diffraction from the bicrystal, i.e. the Bloch waves in both crystals have the same number of components. Consequently, matching at the interface is a simple procedure. The number of eigenvalue problems for a single incident wave to be solved is 2, one for each part of the bicrystal, with dimension \( N_1 \) if \( N_1 \) reflections are taken into account. Thus, the computation time for a single incident wave in this case is proportional to \( 2N_1^3 \). The total computation time in both methods depends therefore on the relative magnitude of \( N^1 \), \( N^2 \) and \( N_1 \), respectively.

With regard to the equivalence of both methods for calculating intensities the following remarks should be made. For coincidence-type boundaries the Sutton & Pond method gives scattering matrices in both crystals, which may be of different dimensions. Hence, the number of solutions for the eigenvalue \( \gamma^1 \), i.e. the dispersion surface, may be different in both crystals. As shown by Sutton & Pond (1978), the amplitude of a particular reflection \( I \) in their method is obtained by summing the contributions from a large number of (double) diffracted beams. Specifically, one has to take into account all diffracted beams from the sets \( \{ g \} \) and \( \{ h \} \) for which \( g + h = l \), if \( \{ g \} \) and \( \{ h \} \) represent the sets of diffraction vectors associated with the first and second crystals, respectively. On the other hand, the CSL method gives the same number of solutions of \( \gamma^1 \) in both parts of the bicrystal, the dimension of the eigenvalue problem (or scattering matrix) being equal to the number of diffraction vectors in the set \( \{ g \} \cup \{ h \} \). This is easily seen since the reciprocal lattice of the CSL is the displacement-shift-complete (DSC) lattice in reciprocal space (Grimmer, 1974). The latter is constructed from the difference vectors of the reciprocal lattices of both crystals, i.e. the set \( \{ g \} \cup \{ h \} \). It appears that the CSL method is conceptually more satisfactory, although the two methods should give identical results, provided sufficiently large sets of reflections \( \{ g \} \) and \( \{ h \} \) are taken into account in the actual many-beam computations.

The computations reported in this paper were carried out for a twinned Si crystal, and in this case many reflections common to both matrix and twin crystals occurred. The total computation time in this situation is much lower for the CSL method, and hence this method has been used. In the general case where both common and non-common reflections are taken into account, the total computation time depends on the magnitude of \( N^1 \), \( N^2 \) and \( N_1 \). It can be shown that in the limiting case of no common reflections, when \( N_1 = N^1 + N^2 - 1 \), the CSL method requires less computation time for \( N_1 = N_2 > 13 \).

The influence of a rigid-body translation at the boundary plane can be introduced in the same way in both methods by

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**Fig. 2. Block diagram of the computer program for the simulation of CBED patterns.** Main part of the program is an eigenvalue and eigenvector calculation. Detailed plots up to a grid of 64 x 64 are possible. A line-scan and a radial-scan option are included in order to reduce computation time.
a displacement of the atomic positions in one lattice relative to the other. Such a translation introduces additional phase factors in the potentials $U_g$ and in the Bloch-wave components.

3. Program description

Fig. 2 shows a block diagram of the program construction. The program consists of a number of subprograms that handle input and output. Input is handled by the use of two input files: one with relatively rigid parameters like structure parameters and EM parameters, the second with relatively fluctuating parameters such as the thickness of the sample, depth of the grain boundary if present, means of output etc. The central part of the program calculates the eigenvalues and eigenvectors with the use of standard NAG Library routines involving eigenvalue evaluation of complex hermitian matrices.

Output can be obtained in several ways. One possibility is a plot facility enabling each CBED disc to be plotted in any desired grid, depending on the amount of detail that is required. The digitized plot is visualized on a 256 grey-level video screen, connected to a 16 grey-level plotter. Other means of output are a line scan or radial scan method in which only one line or circle within the convergent-beam discs is calculated, respectively. Both methods make it possible to deduce the symmetry present from a minimum amount of information, thereby reducing calculation time. Another option makes it possible to calculate and plot dispersion-surface sections.

The programs are written in Fortran 77 (V) and run on an Amdahl 470. At present it uses 2100 Kbyte of memory, which can easily be reduced to 1000 Kbyte. The main program can both run in a batch execution mode as well as interactively, whereas the input and output handling programs are mainly interactive programs. Copies of the programs can be obtained from the authors.

4. Results

The possible symmetries observed in CBED patterns can be classified according to 31 diffraction groups (Buxton et al., 1976). A diffraction-group symmetry results from the crystal point-group symmetry by considering the effect of the symmetry elements seen along the incident-beam axis. For instance, a silicon single crystal seen along $\langle 111 \rangle$ has point-group symmetry $3m$. This results in diffraction group $6\overline{mm}g$ having a $3m$ symmetry in both the bright field and whole pattern. Fig. 3 gives the result of the simulated (000) disc intensity for Si. In the calculations presented here the effect of absorption has not been taken into account. Calculations are based on a 22-beam case $(000), 6\langle 2\overline{2}0 \rangle, 6\langle 2\overline{4}2 \rangle, 6\langle 1\overline{9}9 \rangle$ and $3\langle 5115 \rangle$ with the incident beam exactly along the $[111]$ zone axis.

The fact that the three $\langle 5115 \rangle$ first-order lines intersect at the centre of the disc is due to the accelerating voltage of 102.7 kV, in agreement with the experimental results of Jones et al. (1977, Fig. 1). Fig. 4 gives the same (000) disc plotted as $R$ against $\varphi$, in which $R \leq R_{\text{disc}}$, the latter clearly revealing the expected $3m$ symmetry.

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Fig. 3. Simulated density map of a (000) disc for silicon $\langle 111 \rangle$ single crystal. The accelerating voltage is 102.7 kV resulting in the intersection of the three $\langle 5115 \rangle$ first-order Laue zone lines at the centre of the disc. The total specimen thickness is 200 nm. The angle of convergence is 0.2° (3.5 mrad).

Fig. 4. (a) The same density map as in Fig. 3, now in the $R-\varphi$ representation. This plot does reveal the $3m$ symmetry more clearly than Fig. 3. (b) $R-\varphi$ representation: a CBED disc is scanned in a radial way and the intensities are plotted in a rectangular $R-\varphi$ coordinate system.
As mentioned in the Introduction, bicrystal symmetry can be described with dichromatic point groups (Schapink et al., 1983). In the case of a bicrystal specimen with a (111) twin boundary, the associated coloured point group is expected to be $6/m'm'm$, the primes denoting symmetry relations describing a transformation from the 'black' to the 'white' lattice, and vice versa. According to Table 1 of Schapink et al. (1983) the resulting diffraction group is $3m1_r$. Therefore, the bright field will have $6mm$ symmetry provided that the boundary plane of the bicrystal specimen coincides with its mid-plane (c.f. Fig. 1). Figs. 5 and 6 show the simulated $x$-$y$ plot and $R$-$\phi$ plot of the bright-field intensity respectively, clearly revealing a $6mm$ symmetry. Calculations were based on a 31-beam case, using the CSL description as discussed in § 2. This is equivalent to the 22-beam single-crystal situation extended to 31 beams to account for the non-coincidence of nine first-order reflections in the first and second parts of the bicrystal.

Fig. 5. Simulated density map of a (000) disc of a silicon (111) bicrystal specimen. The (111) twin boundary is taken at 0.5 of the specimen thickness of 200 nm. The accelerating voltage is 102.7 kV. The white spots are artefacts caused by calculation errors.

Fig. 6. The same density map as in Fig. 5 in the $R$-$\phi$ representation. Again the observed symmetry ($6mm$) is more clearly revealed than in Fig. 5.

Fig. 7. The effect of the grain-boundary location on CBED zone-axis patterns for silicon (111) bicrystals. The total specimen thickness is 200 nm; the accelerating voltage is 102.7 kV. (a) Radial scan of a simulated (000) disc at $R = 0.5R_{\text{disc}}$. The (111) twin boundary is located at 0.5 of the specimen thickness, resulting in $6mm$ symmetry. (b) Radial scan similar to (a), but now the twin boundary is at 0.4 of the specimen thickness. The symmetry is reduced from $6mm$ to $3m$.

Fig. 8. Radial scan of a simulated (000) disc at $R = 0.5R_{\text{disc}}$ for a silicon (111) bicrystal specimen. The accelerating voltage and specimen thickness are 102.7 kV and 200 nm, respectively. The (111) twin boundary is located at 0.5 of the specimen thickness. A rigid translation along the boundary of 1/12 [121] is applied. The original $6mm$ symmetry is thereby reduced to $2mm$. 

In this book the author aims to present 'a complete and readily understood treatment of all the known phenomena occurring in liquid crystals under the influence of electrical and magnetic fields. Major emphasis is given to explaining the qualitative aspects of the phenomena and to portraying their physical basis'. The result of setting out to perform such an ambitious task in under 350 pages is remarkably successful. Inevitably, much has been culled directly from the literature, sometimes less critically and with less depth of understanding than one might wish, but the sources of information are well referenced, although it is unfortunate that the bibliography is arranged by chapter and order of citation without any attempt at an alphabetical listing or an author index. Blinov himself has made original contributions in this field and naturally writes with particular authority on areas in which he has worked.

The book is divided into two sections. The first (chs. 1–3) describes the basic physics of liquid crystals, while the second (chs. 4–8) looks in detail at the orientational and electrohydrodynamic effects in nematic phases and texture changes and instabilities in cholesteric and smectic liquid crystals. The final chapter (ch. 8) attempts to give a perspective of the practical applications of electro-optical effects in liquid crystals; a particularly hazardous task in so fast moving a field, which nevertheless manages to be laudably comprehensive bearing in mind that the book is a translation of a Russian volume, which appears from the references to have been published about 1980. The publishers, incidentally, appear somewhat coy about the pedigree of this book as a translation from the Russian.

In summary, this is a nicely produced and readable book, which will be valuable to anyone interested in thermotropic liquid crystals for its broad overview of the interesting and important physical phenomena that may occur in such phases.

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