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New *n*-Beam Dynamical Calculations Combining Two- and Three-Dimensional Bloch Waves

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Abstract

A new scheme for the matrix representation of high-energy electron diffraction by a crystal is developed. The theory consists of the matrix formula of two-dimensional Bloch waves and that of three-dimensional ones combined with the layer-doubling method. The new method reduces computing time to about one-tenth of that required for two-dimensional Bloch waves alone and makes it possible to include the surface effect accurately.

1. Introduction

Since Bethe (1928) developed a dynamical theory of electron diffraction, the problems of dynamical scattering have been studied by many researchers (Fujimoto, 1959; Cowley & Moodie, 1957; Van Dyck, 1980; Watanabe, Kikuchi, Hiratsuka & Yamaguchi, 1990). Up to the present, all theoretical approaches may be divided into two groups: those based on the layer-by-layer scheme and those based on the eigenvalue problem.

The first approach has its origin in Darwin's (1914) theory of X-ray diffraction and has found powerful extensions to the multislice method for high-energy electrons in the transmission case (Cowley & Moodie, 1957; Ishizuka & Uyeda, 1977; Wang, 1990), in low-energy electron diffraction calculations using both plane waves and spherical waves (Pendry, 1974) and in reflection high-energy electron diffraction (RHEED) calculations using two-dimensional Bloch waves. In particular, the layer-by-layer method of two-dimensional Bloch waves can be seen properly in high-resolution transmission-electron-microscopy (HRTEM) simulations as well as in RHEED ones because the nonperiodic variations of the potential in the surface-normal direction can be taken into account. However, the layer-by-layer method using two-dimensional Bloch waves requires much computing time compared with Bethe's eigenvalue and the multislice methods.

Peng & Whelan (1990) proposed a matrix formula based on three-dimensional Bloch waves for both transmission and reflection cases. However, in a qualitative sense, it is not completely satisfactory because it is assumed that the triply periodic potential field ceases suddenly at crystal–vacuum or crystal–crystal interface planes.

In this paper, a new scheme for the matrix formula for the transmission case that consists of two- and three-dimensional Bloch-wave theories is proposed so as to accurately incorporate the surface effect and defects.

2. Two-dimensional Bloch wave

On the basis of two-dimensional periodicity, scattering-matrix techniques for solving coupled second-order differential equations have been proposed by many authors (Maksym & Beeby, 1981; Ichimiya, 1983; Zhao, Poon & Tong, 1988; Nagano, 1990). With the introduction of the column vector $\overline{\Phi}$ combining Fourier coefficients of the wave function and its derivatives, the Schrödinger equation can be written in matrix form as

$$\frac{\mathrm{d}\overline{\Phi}}{\mathrm{d}z} = \begin{pmatrix} \bar{0} & \bar{I} \\ \overline{W} & \bar{0} \end{pmatrix} \overline{\Phi}, \tag{1}$$

where $\bar{I}$ is a unit matrix, $\bar{0}$ is a zero matrix and $\overline{W}$ is defined as

$$\{\overline{W}\}_{g_\parallel g'_\parallel} = (2m/\hbar^2) V_{g_\parallel - g'_\parallel} - k_{g_z}^2 \delta_{g_\parallel g'_\parallel}. \tag{2}$$

$V_{g_\parallel}$ is a two-dimensional Fourier coefficient of the crystal potential, k_{g_z} is the z component of the diffracted wave vector and $\delta_{g_\parallel g'_\parallel}$ is the Kronecker δ.

The solution of (1) can be obtained by an exponential series for small slice thickness h (Magnus, 1954):

$$\begin{aligned}\bar{\Phi}(n) &= M_n\bar{\Phi}(n-1)\\ &= \exp\begin{pmatrix}\bar{0} & h\bar{I}\\ h\bar{W} & \bar{0}\end{pmatrix}\bar{\Phi}(n-1)\\ &= M_nM_{n-1}\ldots M_2\bar{\Phi}(1)\\ &= B\bar{\Phi}(1).\end{aligned} \tag{3}$$

From (3) and upper and lower boundary conditions, transmission- and reflection-coefficient matrices are expressed by Nagano (1990) as

$$T^{s-} = 2[(B_{11} - iB_{12}K) + iK^{-1}(B_{21} - iB_{22}K)]^{-1}, \tag{4}$$

$$\begin{aligned}R^{s-} = &[(B_{11} - iB_{12}K) - iK^{-1}(B_{21} - iB_{22}K)]\\ &\times [(B_{11} - iB_{12}K) + iK^{-1}(B_{21} - iB_{22}K)]^{-1},\end{aligned} \tag{5}$$

$$T^{s+} = 2[(b_{11} + ib_{12}K) - iK^{-1}(b_{21} + ib_{22}K)]^{-1}, \tag{6}$$

$$\begin{aligned}R^{s+} = &[(b_{11} + ib_{12}K) + iK^{-1}(b_{21} + ib_{22}K)]\\ &\times [(b_{11} + ib_{12}K) - iK^{-1}(b_{21} + ib_{22}K)]^{-1}\end{aligned} \tag{7}$$

with $K = k_{g_z}\delta_{g_\parallel g'_\parallel}$ and B_{ij} and b_{ij} being submatrices of B and B^{-1}, respectively. The superscript s refers to surface. These transmission and reflection matrices can also be evaluated by other scattering-matix methods (Maksym & Beeby, 1981; Ichimiya, 1983; Zhao *et al.*, 1988).

3. Three-dimensional Bloch wave

In three-dimensional periodicity, Bethe's (1928) fundamental equation is given by

$$(K^2 - k_g^2)C_g + \sum_h V_{g-h}C_h = 0 \tag{8}$$

with

$$k_g = (k_\parallel + g_\parallel, k_z + g_z + \gamma). \tag{9}$$

γ is the conventional *Ampassung* and K is the wave vector of the incident electron waves in the crystal. In the Laue case, (8) can be reduced to a generalized eigenvalue problem (Kim & Sheinin, 1982)

$$\begin{aligned}&\{2k_z\gamma - [K^2 - (k_\parallel + g_\parallel)^2 - k_z^2]\}C^i_{g_\parallel} - \sum_{h_\parallel \neq g_\parallel} V_{g_\parallel - h_\parallel}C^i_{h_\parallel}\\ &= 0\end{aligned} \tag{10}$$

where $C^i_{g_\parallel}$ is the $g_\parallel$ element of the eigenvector of the ith eigenvalue. The electron wave function with $g_\parallel$ and its derivative can be written as

$$\varphi_{g_\parallel}(z) = \sum_j^n C^j_{g_\parallel}\exp[i(k_z + \gamma^j)z]\alpha^j \tag{11}$$

and

$$\mathrm{d}\varphi_{g_\parallel}(z)/\mathrm{d}z = \sum_j^n C^j_{g_\parallel} i(k_z + \gamma^j + g_z)\exp[i(k_2 + \gamma^j)z]\alpha^j. \tag{12}$$

In matrix notation, (11) and (12) can be written as

$$\varphi_{g_\parallel}(z) = \bar{C}(z)\bar{\Gamma}(z)\bar{A} \tag{13}$$

and

$$\mathrm{d}\varphi_{g_\parallel}/\mathrm{d}z = \bar{C}(z)\bar{Z}\,\bar{\Gamma}(z)\bar{A}. \tag{14}$$

$\bar{C}$ is the matrix of $C^j_{g_\parallel}$, $\bar{Z}$ and $\bar{\Gamma}$ are the diagonal matrices consisting of $i(k_2 + \gamma^j)_z$ and $\exp[i(k_z + \gamma^j)z]$ and $\bar{A}$ is the column matrix of α^j. With the same column vector $\bar{\Phi}$ combining Fourier coefficients $\varphi_{g_\parallel}$ and its derivatives, and with the same boundary condition at $z = z_A$ and $z = z_B$ as above, the Bloch-wave amplitude vector $\bar{A}$ is eliminated from (13) and (14) as follows:

$$\bar{\Phi}(z_B) = \bar{M}\,\bar{\Phi}(z_A). \tag{15}$$

Therefore, the reflection and transmission matrices T^{B-}, T^{B-}, T^{B+} and R^{B+} can be obtained in the same forms as in the two-dimensional Bloch-wave case. The reflection matrix R^{B-} is identical to that of Peng & Whelan (1990).

4. Layer-doubling method

The scattering matrix for the combined system of two slabs can be considered as shown in Fig. 1. Following the layer-doubling scheme, transmission and reflection matrices are given by

$$R^{AB-} = R^{A-} + T^{A+}R^{B-}(I - R^{A+}R^{B-})^{-1}T^{A-}, \tag{16}$$

$$T^{AB-} = T^{B-}(I - R^{A+}R^{B-})^{-1}T^{A-}, \tag{17}$$

$$R^{AB+} = R^{B+} + T^{B-}R^{A-}(I - R^{B-}R^{A+})^{-1}T^{B+}, \tag{18}$$

$$T^{AB+} = T^{A+}(I - R^{B-}R^{A+})^{-1}T^{B+}. \tag{19}$$

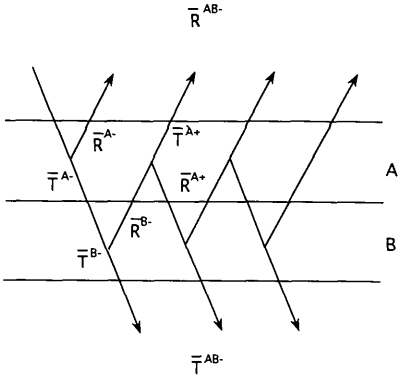

Fig. 1. Schematic view of the layer-doubling method.

Since this calculation scheme is naturally taken into the layer-doubling method, computing time is reduced considerably as follows. As shown in Fig. 2, a specimen for the Laue case is divided into three regions. In regions I and II, the crystal potential is nonperiodic along the z direction but in region II the crystal potential is periodic. Then, treating I and III as a set, many scattering matrices need to be calculated. However, in region II, only one matrix is required. Similar treatment using the layer-doubling method was suggested by Maksym (1985).

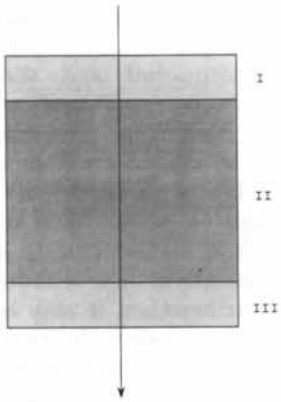

Fig. 2. Schematic view of the crystal system.

5. Results

Compared to the Bethe method, the validity of the present method is discussed for n-beam dynamical calculations for copper at 100 and 300 kV excluding and including a surface effect. The orientation was fixed at [001]. The crystal potentials were constructed from superposing free atoms (Doyle & Turner, 1967).

5.1. *The validity of the present method*

In order to illustrate its validity, the present method has been tested by n-beam dynamical calculations with the same crystal potential as the Beth method, which stops suddenly at the crystal–vacuum interface. For the Bethe method, an 11×11 beam number is adopted, which makes the calculations for this material converge. The present method also uses an 11×11 beam number and an $a/128$ slice thickness. Fig. 3 shows plots of 000 and 200 beam intensities against thickness. The present method is in good agreement with Bethe method whether the accelerating voltage is 100 or 300 kV. The computing time at $z = 20a$ is about one-tenth of that for a two-dimensional Bloch wave only.

5.2. *The effect of surface*

As for the first step, a surface effect is discussed using a crystal potential that is simply reconstructed

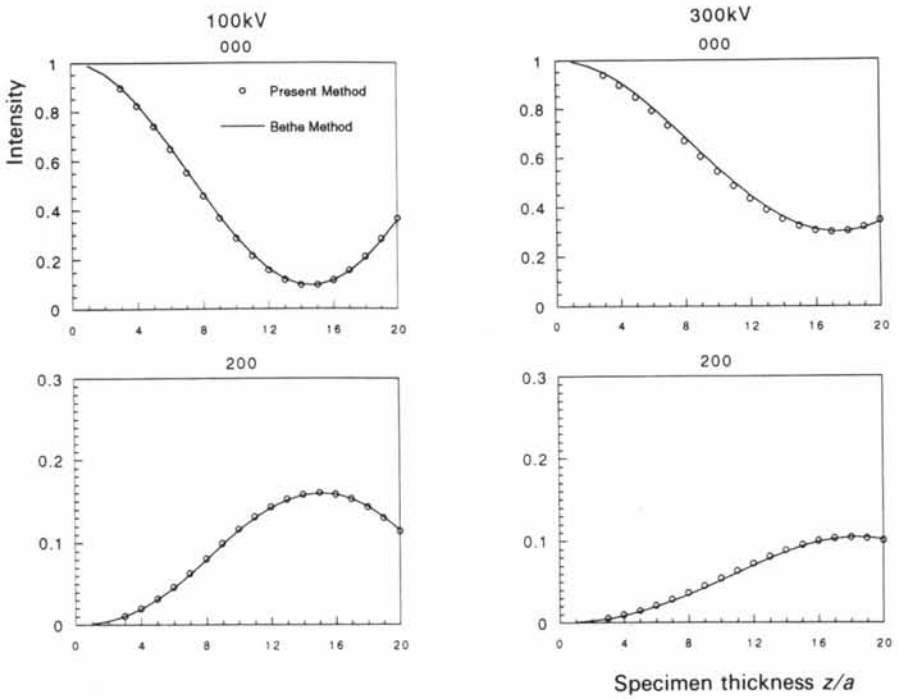

Fig. 3. Beam intensities of the 000 and 200 reflections for [001] n-beam dynamical calculations excluding a surface effect for copper at 100 and 300 kV.

from an ideal superslab of neutral atoms, the surface relaxation and reconstruction being thereby ignored. The planar average potentials for the Bethe method and for an 11-layer film with the present method are plotted as a function of z coordinate perpendicular to the surface, as shown in Fig. 4. The potential of the last atomic layer is very similar to that of the bulk and the surface potential leaks out into the vacuum over the dotted line.

The parameters used in the present method are the same as those above. Fig. 5 shows the thickness variation of the 000 and 200 beam intensities. The effect of the surface is a relatively small difference in the main-beam intensities almost independent of the accelerating voltage. Although the effect of this ideal surface seems to be relatively small, the surface effect is one of the important factors for accurate HRTEM image simulations. The present method appears to be useful not only for accurate HRTEM simulations of a wide variety of materials but also for the study of properties such as surface polarity and reconstruction. A more detailed understanding of the surface effect is possible by making use of real surface structure without any serious problems.

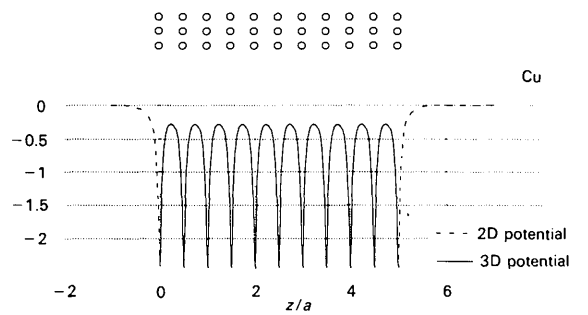

Fig. 4. The planar average potentials for the Bethe method and an 11-layer film by the present method.

6. Concluding remarks

A general scheme has been proposed for the dynamical theory of fast electron diffraction on the basis of two-dimensional and three-dimensional Bloch waves that are combined with the layer-doubling method. Reduction of computing time and required memory is achieved in comparison with the two-dimensional Bloch-wave method. This method is applicable not only to perfect bulk crystals but also to the ideal surface. It can readily include surface relaxation or reconstruction without any serious complications. Furthermore, it may be applicable to simulations of defects using the periodic continuation approximation (Fields & Cowley, 1978).

The effect of anomalous absorption resulting from the depletion of the elastic wave by inelastic scat-

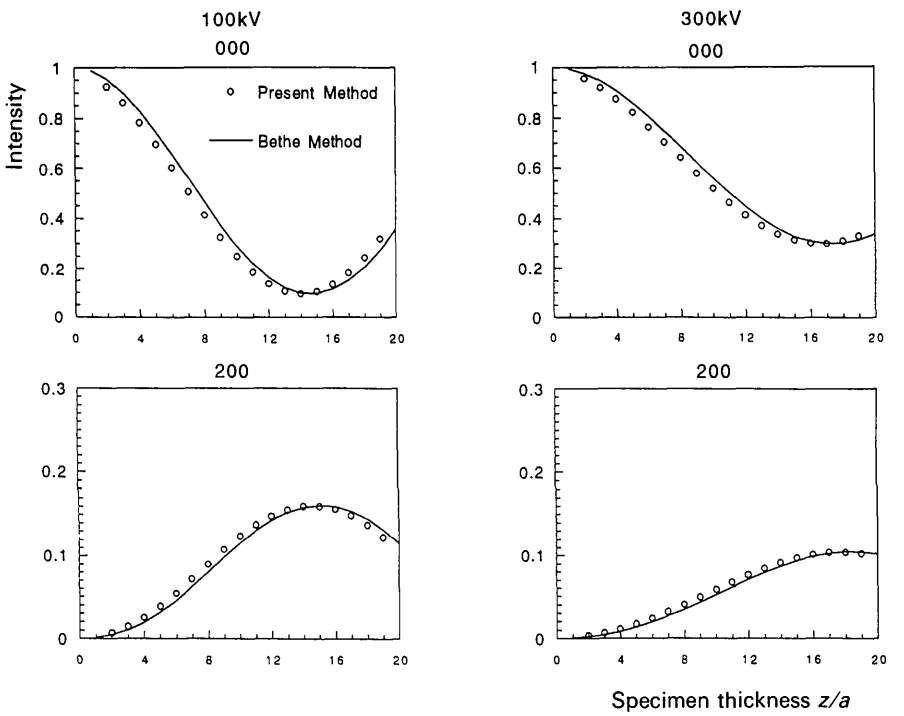

Fig. 5. Beam intensities of the 000 and 200 reflections for [001] n-beam dynamical calculations including a surface effect for copper at 100 and 300 kV.

tering can be easily taken into account by adding additional complex contributions $V'_{g\parallel}(z)$ and V'_g to (2) and (8) (Yoshioka, 1957; Nagano, 1990) in two- and three-dimensional Bloch-wave theories, respectively.

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On the Statistical Analysis of Orientation Data

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Abstract

The statistical analysis of data in the form of orientations is a relatively new discipline and results from the literature on this subject are not yet widely known outside the statistics community. This paper provides an introduction to and the key references for statistical methods for analysing orientation data. More specifically, the problem of estimating an unknown orientation is considered and results on the precision of such an estimated orientation are described. The calculation of average orientation and dispersion parameters for a sample of orientations is also considered. Finally, procedures for generating and testing for random orientations are described. The methodology is illustrated with crystal orientation data obtained from the analysis of electron back-scattering patterns.

1. Introduction

Orientation data arise naturally in many scientific areas, notably in the earth sciences, astronomy and biology. Within the field of materials science, for example, the development of techniques for measuring local lattice orientations in polycrystalline materials has opened up the way for wholly new types of investigations. These techniques, especially the electron back-scattering pattern (EBSP) technique in scanning electron microscopy and the Kikuchi diffraction technique in transmission electron microscopy, have recently been a major subject at several conferences and workshops (Bunge, 1993, 1994). These modern techniques for measuring local lattice orientations are convenient and rapid in use and the EBSP technique has recently even been fully automated (Wright & Adams, 1992), thus allowing large numbers of orientation data to be collected. Statistical methods for analysing such data, however, have only very rarely been applied. As a result of this, for example, little is known about the precision by which crystal orientations can be determined. Another example of the application of orientation statistics is when the average and dispersion of a sample of orientations is to be determined. For example, it is demonstrated in the final section of this paper that using the arithmetic mean of Euler angles as a measure of an average orientation cannot generally be recommended.