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A Relativistic *n*-Beam Dynamical Theory for Fast Electron Diffraction

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Abstract

From the Dirac equation with a periodic scalar potential, an *n*-beam dynamical formula for the matrix representation of high-energy electron diffraction by a crystal is developed. By combining this with the layer-doubling method, the diffraction from an assembly of crystal slabs having different structures and thicknesses can be evaluated. Dynamical calculations of aluminium, copper and gold at several accelerating voltages have been carried out in a completely parallel manner by the present method and the Bethe method derived from the relativistic Schrödinger equation by replacing corrected mass and wavelength. The relativistic Schrödinger equation is found to be applicable for *n*-beam dynamical calculations for the Laue case.

1. Introduction

Because of the strong scattering of electrons by all but the lightest atoms, scattered beams from even a thin crystal are sufficiently large to ensure there is considerable multiple scattering, because of which the dynamical theory is indispensable for an accurate interpretation of electron diffraction. Since Bethe (1928) developed a dynamical theory for electron diffraction, the problems of the dynamical scattering by a crystal have been tackled by many workers (Cowley & Moodie, 1957; Fujimoto, 1959; Van Dyck, 1980; Watanabe, Kikuchi, Hiratsuka & Yamaguchi, 1990). Until now, all theoretical approaches may be divided into two groups: those based on the layer-by-layer scheme and those on the eigenvalue problem for three-dimensional Bloch waves. Correlations between these approaches have also been discussed in detail (Jap & Glaeser, 1978; Gratias & Portier, 1983). In addition, there is a new theory which consists of the layer-by-layer and the eigenvalue methods combined with the layer-doubling methods (Peng & Whelan, 1990; Mitsuishi, Watanabe & Hashimoto, 1994).

Electrons in a conventional electron microscope travel at 55% of the velocity of light at 100 kV and at 89% at 600 kV so that the relativistic effect is not far from negligible. For such electrons, Fujiwara (1961) pointed out, by solving the Dirac equation with the perturbation method, that the relativistic Schrödinger equation might be obtained by the replacements of corrected mass and wavelength. The same result was obtained from the second-order equation deduced from the Dirac equation (Howie; see Fujiwara, 1962). The reliability of the relativistic Schrödinger equation had been confirmed by comparison with the extinction distances of aluminium (Hashimoto, 1964). In fact, almost *n*-beam dynamical calculations are set up under this relativistic correction. However, it is not certain whether any limitation of the correction exists or not.

Gevers & David (1982) derived the first-order differential equations for the coefficients in a spinor plane-wave expansion of the total wave function from the Dirac equation without making any prior approximation. However, it is not easy to handle nor does it have computer efficiency, and no published studies can be found dealing further with the relevant relativistic dynamical theory.

In this paper, a new scheme for the relativistic *n*-beam dynamical theory based on the Dirac equation is established. Basically, the idea is to generalize the scattering matrix for a spinor plane wave and to derive reflection and transmission matrices under an appropriate boundary condition. The *n*-beam dynamical calculations for aluminium, copper and gold are carried out by taking account of the accelerating voltage.

2. Theory

2.1. *The dynamical equation*

The Dirac equation for an electron interacting with a scalar crystal potential V is given by

$$E\psi = H\psi = (\hat{\boldsymbol{\alpha}}\hat{\mathbf{p}}c + \hat{\boldsymbol{\beta}}m_0c^2 - eV)\psi, \qquad (1)$$

where c is the light velocity, m_0 the rest mass, $\hat{\mathbf{p}}$ the momentum operator and $-e$ the charge of an electron. $\hat{\boldsymbol{\alpha}}$ and $\hat{\boldsymbol{\beta}}$ are given by

$$\hat{\boldsymbol{\alpha}}_i = \begin{pmatrix} 0 & \hat{\boldsymbol{\sigma}}_i \\ \hat{\boldsymbol{\sigma}}_i & 0 \end{pmatrix}, \quad i = 1, 2, 3, \quad \hat{\boldsymbol{\beta}}_i = \begin{pmatrix} \hat{\mathbf{1}} & 0 \\ 0 & -\hat{\mathbf{1}} \end{pmatrix},$$

where

$$\hat{\sigma}_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \hat{\sigma}_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \hat{\sigma}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},$$
$$\hat{1} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}.$$

For a free electron with spin up, the wave function is given by a plane-wave form:

$$\psi(z) = N \begin{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \\ \dfrac{c\hat{\sigma}_3 \hbar \mathbf{k}}{m_0 c^2 + E} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \end{pmatrix} \exp(ik_z z), \tag{2}$$

where N is the normalization factor, and the wave-number vector $\mathbf{k}$ and the energy E satisfy the following relation:

$$E^2 = c^2 \hbar^2 \mathbf{k}^2 + m_0^2 c^4. \tag{3}$$

Since a scalar crystal potential V is periodic, it can be expanded in a Fourier series:

$$V(\mathbf{r}) = \sum_{\mathbf{h}} V_{\mathbf{h}} \exp(i\mathbf{h}\mathbf{r}). \tag{4}$$

The solution of (1) can be expressed as a linear combination of a four-component basis function:

$$\{\psi_j(\mathbf{r})\} = \sum_{\mathbf{g}} \{C_j\}_{\mathbf{g}} \exp[i(\mathbf{k}+\mathbf{g})\mathbf{r}], \tag{5}$$

where $\{C_j\}_{\mathbf{g}}$ is the four-component spinor.

Substituting (4) and (5) into (1) and following the same procedure as in Bethe's eigenvalue method, we can easily show that the coefficients (spinors) are obtained by

$$\left[\hbar c(\mathbf{k}+\mathbf{g})\hat{\boldsymbol{\alpha}} + m_0 c^2 \hat{\boldsymbol{\beta}} - E\right]\{C_j\}_{\mathbf{g}} - \sum_{\mathbf{h}} V_{\mathbf{h}} \{C_j\}_{\mathbf{g}-\mathbf{h}} = 0. \tag{6}$$

In the diffraction problem, an eigenvalue is $\mathbf{k}_z$, unlike in the band calculation, so this equation cannot be solved directly through the eigenvalue problem. In order to change this equation into the eigenvalue problem, it must be transformed by the following two steps. First, (6) is transformed by the unitary matrix

$$U = \begin{pmatrix} \tilde{u} & & 0 \\ & \ddots & \\ 0 & & \tilde{u} \end{pmatrix}$$

with

$$\tilde{u} = \tfrac{1}{2} \begin{pmatrix} 1 & 1 & 1 & -1 \\ 1 & 1 & -1 & 1 \\ 1 & -1 & 1 & 1 \\ -1 & 1 & 1 & 1 \end{pmatrix}. \tag{7}$$

Next, by multiplication from the left by the matrix A',

$$A' = \begin{pmatrix} a' & & 0 \\ & \ddots & \\ 0 & & a' \end{pmatrix} \quad \text{with } a' = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix},$$

(6) can be recast into the form

$$\hbar c \begin{pmatrix} k_z & & 0 \\ & \ddots & \\ 0 & & k_z \end{pmatrix} \{C'_j\}_{\mathbf{g}} = \begin{pmatrix} D_{\mathbf{g}1} & V'_{\mathbf{g}1-\mathbf{g}2} & & \\ V'_{\mathbf{g}2-\mathbf{g}1} & \ddots & & \\ & & & D_{\mathbf{g}n} \end{pmatrix} \{C'_j\}_{\mathbf{g}}, \tag{8}$$

where

$$D_{\mathbf{g}} = \begin{pmatrix} E - V_0 & -m_0 c^2 + ic\hbar(k_y + g_{ij}) & 0 & -c\hbar(k_x - g_x) \\ m_0 c^2 + ic\hbar(k_y + g_y) & -E + V_0 & c\hbar(k_x + g_x) & 0 \\ 0 & -c\hbar(k_x + g_x) & E - V_0 & m_0 c^2 + ic\hbar(k_y + g_y) \\ c\hbar(k_x + g_x) & 0 & -m_0 c^2 + ic\hbar(k_y + g_y) & -E + v_0 \end{pmatrix}$$

with

$$V'_{\mathbf{g}} = \begin{pmatrix} -V_{\mathbf{g}} & 0 & 0 & 0 \\ 0 & -V_{\mathbf{g}} & 0 & 0 \\ 0 & 0 & -V_{\mathbf{g}} & 0 \\ 0 & 0 & 0 & -V_{\mathbf{g}} \end{pmatrix}.$$

Having solved this eigenvalue problem to obtain k_z and $\{c_j\}_{\mathbf{g}}$, we may write the wave function as

$$\psi(\mathbf{r}) = \sum_{\mathbf{g},i} \chi^i \{C_j^i\}_{\mathbf{g}} \exp[i(\mathbf{k}^i + \mathbf{g})\mathbf{r}], \tag{9}$$

where χ^i are the Bloch-wave excitation amplitudes. In matrix notation, (9) can be rewritten as

$$\Psi = U^{-1} C' \Gamma(z) \chi, \tag{10}$$

where

$$\Gamma(z) = \begin{pmatrix} \exp(ik^1 z) & 0 & \cdots & 0 \\ 0 & & & \vdots \\ \vdots & & \ddots & 0 \\ 0 & \cdots & \cdots & \exp(ik^n z) \end{pmatrix}. \tag{11}$$

With the same boundary condition at $z = z_A$ and $z = z_B$, the wave amplitude vector χ is eliminated as follows:

$$\begin{aligned} \Psi(z_B) &= U^{-1} C' \Gamma(z_B) \Gamma^{-1}(z_A) C'^{-1} U \Psi(z_A) \\ &= M \Psi(z_a) \\ &= \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} \Psi(z_A). \end{aligned} \tag{12}$$

Using a proper boundary condition, we can determine the unique electron scattering wave function. In other words, reflection and transmission coefficients can be evaluated.

2.2. *Boundary conditions*

Now, let us study the scattering case shown in Fig. 1, where the wave functions are written as

$$\Psi(z_A) = \left\{ z \begin{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \\ \dfrac{c\hat{\sigma}_3 \hbar K_g}{m_0c^2 + E} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \end{pmatrix} \exp(iK_{g,z} z_A) \right\}_g \delta_{g,0} + \left\{ \begin{pmatrix} \begin{pmatrix} r_{g,u} \\ r_{g,d} \end{pmatrix} \\ \dfrac{-c\hat{\sigma}_3 \hbar K_g}{m_0c^2 + E} \begin{pmatrix} r_{g,u} \\ r_{g,d} \end{pmatrix} \end{pmatrix} \exp(-iK_{g,z} z_A) \right\}_g \tag{13}$$

and

$$\Psi(z_B) = \left\{ \begin{pmatrix} \begin{pmatrix} t_{g,u} \\ t_{g,d} \end{pmatrix} \\ \dfrac{c\hat{\sigma}_3 \hbar K_g}{m_0c^2 + E} \begin{pmatrix} t_{g,u} \\ t_{g,d} \end{pmatrix} \end{pmatrix} \exp(iK_{g,z} z_B) \right\}_g, \tag{14}$$

where $r_{g,u}$, $r_{g,d}$, $t_{g,u}$ and $t_{g,d}$ are reflection coefficients and transmission coefficients for spin up and spin down, respectively, and s is the coefficient of an incident electron. Here, we restrict our discussion to the solution for incident beams with spin up. At a boundary, the wave function must be continuous because of conservation and hence reflection and transmission coefficients can be combined by using (12)–(14):

$$\left\{ \begin{pmatrix} \begin{pmatrix} t_{g,u} \\ t_{g,d} \end{pmatrix} \\ \dfrac{c\hat{\sigma}_3 \hbar K_g}{m_0c^2 + E} \begin{pmatrix} t_{g,u} \\ t_{g,d} \end{pmatrix} \end{pmatrix} \exp(iK_{g,z} z_B) \right\}_g = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} \left(\left\{ s \begin{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \\ \dfrac{c\hat{\sigma}_3 \hbar K_g}{m_0c^2 + E} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \end{pmatrix} \exp(iK_{g,z} z_A) \right\}_g \delta_{g,0} + \left\{ \begin{pmatrix} \begin{pmatrix} r_{g,u} \\ r_{g,d} \end{pmatrix} \\ \dfrac{-c\hat{\sigma}_3 \hbar K_g}{m_0c^2 + E} \begin{pmatrix} r_{g,u} \\ r_{g,d} \end{pmatrix} \end{pmatrix} \exp(-iK_{g,z} z_A) \right\}_g \right). \tag{15}$$

In order to separate the electron and positron parts, we transform the basis by the unitary matrix. Then (15) can be recast into the form

$$\begin{pmatrix} \tilde{K}(z_B) & 0 \\ 0 & \tilde{K}(z_B) \end{pmatrix} \begin{pmatrix} t_B \\ \tilde{p}t \end{pmatrix} = \begin{pmatrix} m'_{11} & m'_{12} \\ m'_{21} & m'_{22} \end{pmatrix} \left\{ \begin{pmatrix} \tilde{K}(z_A) & 0 \\ 0 & \tilde{K}(z_A) \end{pmatrix} \begin{pmatrix} s \\ \tilde{p}s \end{pmatrix} + \begin{pmatrix} \tilde{K}(-z_a) & 0 \\ 0 & \tilde{K}(-z_A) \end{pmatrix} \begin{pmatrix} r \\ -\tilde{p}r \end{pmatrix} \right\}, \tag{16}$$

where

$$t = \begin{pmatrix} t_{0,u} \\ t_{0,d} \\ \vdots \\ t_{m,u} \\ t_{m,d} \\ \vdots \end{pmatrix}, \quad r = \begin{pmatrix} r_{0,u} \\ r_{0,d} \\ \vdots \\ r_{m,u} \\ r_{m,d} \\ \vdots \end{pmatrix}, \quad s = \begin{pmatrix} s_{0,u} \\ 0 \\ \vdots \\ \vdots \\ \vdots \\ 0 \end{pmatrix}, \tag{17}$$

$$\tilde{p} = \begin{pmatrix} \dfrac{c\hbar K_{g_0}}{E + m_0c^2} & 0 & \cdots & 0 \\ 0 & \dfrac{-c\hbar K_{g_0}}{E + m_0c^2} & \cdots & \vdots \\ \vdots & & \ddots & \\ & & \dfrac{c\hbar K_{g_{n-1}}}{E + m_0c^2} & 0 \\ 0 & & 0 & \dfrac{-c\hbar K_{g_{n-1}}}{E + m_0c^2} \end{pmatrix} \tag{18}$$

and

$$\tilde{K}(z) = \begin{pmatrix} \exp(iK_z z) & 0 & \cdots & 0 \\ 0 & \exp(iK_z z) & \cdots & \vdots \\ \vdots & \vdots & \ddots & \\ & & \exp(iK_{z,n-1} z) & 0 \\ 0 & \cdots & 0 & \exp(iK_{z,n-1} z) \end{pmatrix}. \tag{19}$$

Therefore, the columns for transmission and reflection coefficients, that is t and r, can be expanded with the so-called transmission and reflection matrices:

$$\tilde{K}(z_B)t = \tilde{T}\tilde{K}(z_A)s \tag{20}$$

and

$$\tilde{K}(-z_A)r = \tilde{R}\tilde{K}(z_A)s, \tag{21}$$

where

$$\begin{aligned} \tilde{T} = {} & \{(m'_{11} - m'_{12}\tilde{p})^{-1} - (m'_{21} - m'_{22}\tilde{p})^{-1}\tilde{p}\} \\ & \times \{(m'_{11} - m'_{12}\tilde{p})^{-1}(m'_{11} + m'_{12}\tilde{p}) \\ & - (m'_{21} - m'_{22}\tilde{p})^{-1}(m'_{21} + m'_{22}\tilde{p})\} \end{aligned} \tag{22}$$

and

$$\tilde{R} = \{(m'_{11} - m'_{12}\tilde{p}) - \tilde{p}^{-1}(m'_{21} - m'_{22}\tilde{p})\}^{-1} \times \{\tilde{p}^{-1}(m'_{21} + m'_{22}p) - (m'_{11} - m'_{12}\tilde{p})\}. \quad (23)$$

For the case where the incident electrons come from the reverse side, reflection and transmission matrices can also be evaluated in the same manner. Thus, reflection and transmission matrices for a combined system of two slabs can be calculated with the layer-doubling method as shown in Fig. 2 (Nagano, 1990). Therefore, the diffraction from an assembly of crystal slabs having different structures and thicknesses is easily calculated by this scheme without any serious problems.

3. Results

The n-beam dynamical calculations are evaluated by taking account of the atomic number of the materials and the accelerating voltages in this calculation. The crystal potentials are constructed from superposing free atoms (Doyle & Turner, 1968). In order to investigate how the beam number affects the diffraction intensities, (100) n-beam dynamical calculations for the present method are compared with the relativistically corrected Bethe method. Figs. 3–5 show the thickness series for aluminium, copper and gold at 100, 400 and 600 kV. The present method and the corrected Bethe method are superimposed everywhere at 100, 400 and 600 kV irrespective of atomic number and 121 beams are enough for convergent (100) n-beam dynamical calculations. The results for 121 beams are also in good agreement with the other methods (Van Dyck, 1980; Watanabe *et al.*, 1990; Mitsuishi *et al.*, 1994).

4. Concluding remarks

A general scheme has been proposed for the dynamical theory of fast electron diffraction on the basis of the Dirac equation, in which the scattering matrix is combined with the layer-doubling method. Thus, it can be applied not only to multilayer materials but also to

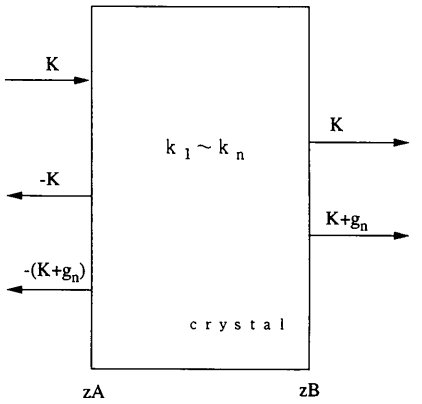

Fig. 1. Schematic view of the electron scattering.

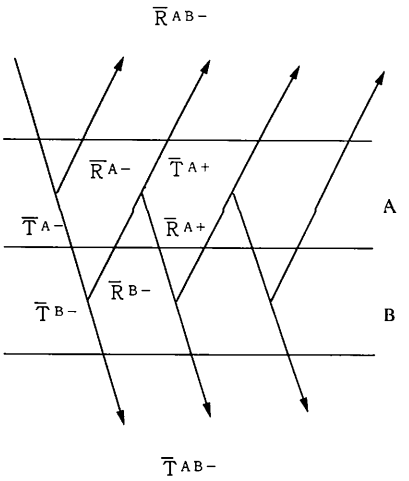

Fig. 2. Schematic view of the layer-doubling method.

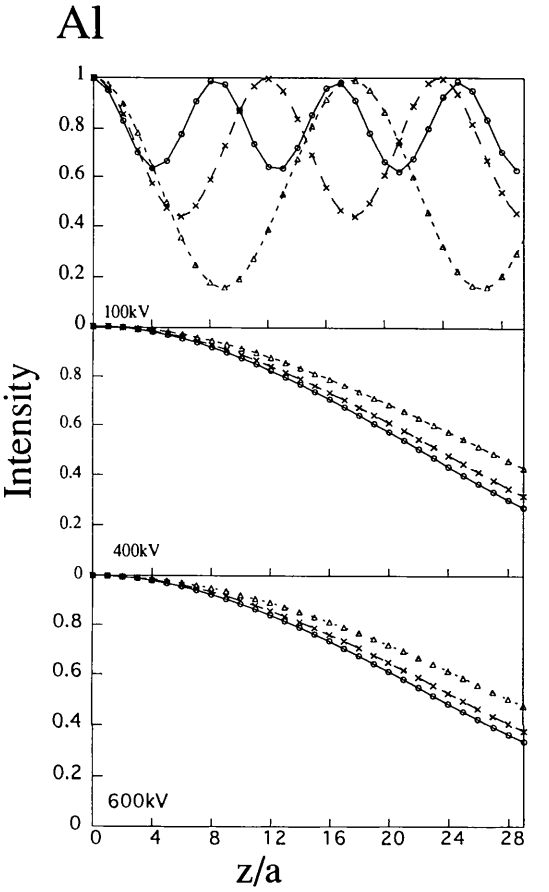

Fig. 3. Beam intensities of 000 for aluminium at 100, 400 and 600 kV. Solid curves: present method with 121 beams. Dashed curve: present method with 25 beams. Dotted curve: present method with 9 beams. Circles: Bethe method with 121 beams. Crosses: Bethe method with 25 beams. Triangles: Bethe method with 9 beams.

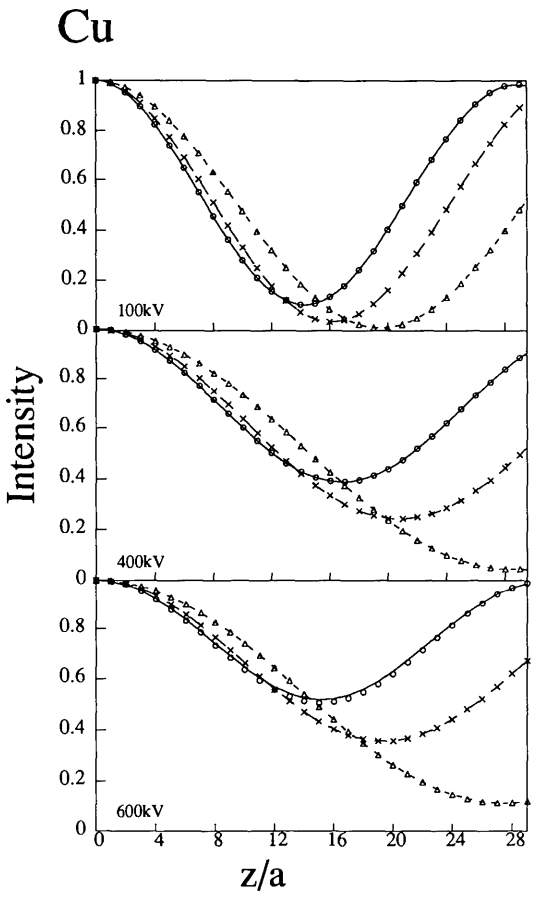

Fig. 4. Beam intensities of 000 for copper at 100, 400 and 600 kV. Solid curve: present method with 121 beams. Dashed curve: present method with 25 beams. Dotted curve: Present method with 9 beams. Circles: Bethe method with 121 beams. Crosses: Bethe method with 25 beams. Triangles: Bethe method with 9 beams.

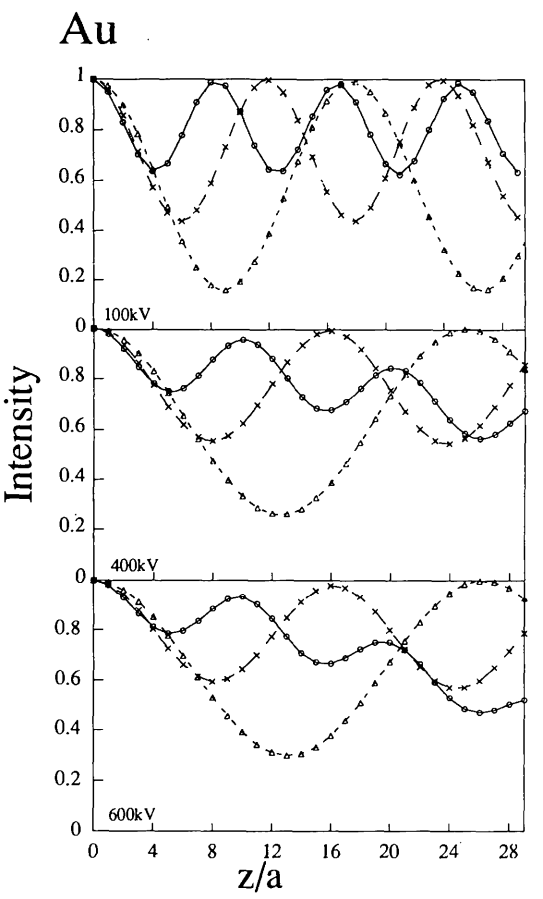

Fig. 5. Beam intensities for 000 for gold at 100, 400 and 600 kV. Solid curve: present method with 121 beams. Dashed curve: present method with 25 beams. Dotted curve: present method with 9 beams. Circles: Bethe method with 121 beams. Crosses: Bethe method with 25 beams. Triangles: Bethe method with 9 beams.

isolated defects by using the periodic continuation approximation (Fields & Cowley, 1978). This method also has the advantage of a linear system, thus being easily extended to RHEED calculations without any problems, unlike the Bethe method (Kambe, 1988; Ma & Marks, 1989). Furthermore, it turns out that the Schrödinger equation with relativistically corrected mass and wavelength is applicable for n-beam dynamical calculations under the small-angle approximation. Although reductions of computing time and memory are achieved in comparison with a system of first-order differential equations derived from the Dirac equation (Gevers & David, 1982) and the Schrödinger equations (Maksym & Beeby, 1981; Ichimiya, 1983; Zao, Poon & Tong, 1988), the matrix size is four times larger than for the Bethe method. However, the matrix size can usually be reduced by group theory and is not a serious problem with today's advanced computers. It promises to be a good starting point to discuss the relativistic effect of high-energy electron diffraction.

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