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Multislice Formula for Inclined Illumination

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Abstract

A multislice formula for many-beam calculation of dynamical electron diffraction amplitudes which may be applied for inclined illumination has been derived based on the Schrödinger equation. This formula clearly shows the following points: a specimen should be considered as sliced parallel to the entrance surface; a spherical propagation function should be used, giving the exact excitation error measured along the surface normal; the interaction constant in the phase-grating function should be changed; the phase grating should be projected along the beam direction. A new parameter for the strength of the upper-layer interaction in the case of the inclined illumination is proposed in terms of the excitation error of the reciprocal-lattice points used to define the projected potential. It is pointed out that the potential projected along the beam direction may better represent the upper-layer interactions for the inclined illumination.

I. Introduction

The multislice formula derived by Cowley & Moodie (1957) included small inclination of illumination assuming a specimen of the orthogonal system. This formula was developed to include small tilts such as those occurring in the case of convergent-beam experiments (Lynch, 1971; Goodman & Moodie, 1974). There the effect of the tilt appeared only in the propagation function through an excitation error. Self (1979) made extensive investigations on the effect of specimen tilt intuitively and gave expressions different from the present results. In the matrix method it is easy to derive an equation taking into account the tilt theoretically (*e.g.* van Dyck, 1976), but this has problems for practical calculation in many-beam cases compared with the multislice method (van Dyck, 1979).

Normal convergent-beam patterns have been successfully interpreted using the usual multislice formula for small-unit-cell crystals composed of light atoms (Goodman & Lehmpfuhl, 1968) and of heavy atoms (Lynch, 1971). However, asymmetries of diffraction intensity have been observed in tilted convergent-beam patterns (Goodman, 1974; Buxton, Eades, Steeds & Rackham, 1976), and this effect has not been thoroughly investigated (Steeds, 1979). Some experiments with intentional large tilt angles have been done in order to determine the polarity of noncentrosymmetric crystals (Taftø, 1981; Taftø & Spence, 1982). Although these reports presented a simple method of understanding the diffraction intensities, detailed analysis should be carried out based on n-beam dynamical theory.

Upper-layer interactions (out-of-zone effects) are considered by Lynch (1971) and Goodman & Moodie (1974). They showed that the upper-layer interactions will introduce serious errors in the multislice calculation when the slice thickness multiplied by the excitation error becomes large.

In this paper, a multislice formula which may be applied to the inclined illumination is derived based on the Schrödinger equation, extending the previous paper of Ishizuka & Uyeda (1977). Some necessary changes from the usual formula for normal incidence are discussed. A parameter for the strength of upper-layer interactions, different from that of Goodman & Moodie (1974), is proposed. Some calculations in progress in order to test the present theory will be published separately.

II. Derivation of the formula

The solution of the Schrödinger equation written in Cartesian coordinates,

$$(\nabla^2 + k^2)\,\psi(\mathbf{r}) = (2m/\hbar^2)\,V(\mathbf{r})\,\psi(\mathbf{r}), \qquad (1)$$

is given in the following integral form

$$\varphi(\mathbf{r}) = 1 - \frac{2m}{4\pi\hbar^2}\int \frac{\exp i\{k|\mathbf{r}-\mathbf{r}'| - \mathbf{k}(\mathbf{r}-\mathbf{r}')\}}{|\mathbf{r}-\mathbf{r}'|} \times V(\mathbf{r}')\,\varphi(\mathbf{r}')\,\mathrm{d}\mathbf{r}', \qquad (2)$$

after the substitution: $\psi(\mathbf{r}) = \exp(i\mathbf{kr})\,\varphi(\mathbf{r})$. Here $k = |\mathbf{k}|$, $\mathbf{k}(k_x,k_y,k_z)$ is the wave vector of the incident electrons, $V(\mathbf{r})$ is the potential energy (Ishizuka &

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Uyeda, 1977). If the propagation function $p(\mathbf{r})$ is defined as

$$p(\mathbf{r}) = \frac{1}{\lambda i}\frac{1}{r}\exp i\{kr - \mathbf{kr}\}, \tag{3}$$

where $r = |\mathbf{r}|$, then (2) is rewritten

$$\varphi(\mathbf{r}) = 1 - \frac{i}{\hbar v}\int p(\mathbf{r}-\mathbf{r}')V(\mathbf{r}')\varphi(\mathbf{r}')\,\mathrm{d}\mathbf{r}'. \tag{4}$$

Here the relationship $2m/4\pi\hbar^2 = (i/\hbar v)(1/\lambda i)$ is used, where v and λ are the velocity and wavelength of the incident electrons respectively. This propagation function has the following properties:

$$\int p(\mathbf{b},z)\,\mathrm{d}\mathbf{b} = k/k_z, \tag{5a}$$

where $\mathbf{r} = (\mathbf{b},z)$, and

$$p(\mathbf{b}_2-\mathbf{b}_1, z_2-z_1) \simeq (k_z/k)\int_{\Sigma} p(\mathbf{b}-\mathbf{b}_1, z-z_1) \times p(\mathbf{b}_2-\mathbf{b}, z_2-z)\,\mathrm{d}\mathbf{b}. \tag{5b}$$

Here the plane of integration Σ is arbitrarily located between the points z_1 and z_2. These relationships can easily be proved using the Fourier transform of the propagation function (Appendix).

When the integral in (4) is divided into two regions, V_{I} and V_{II} (see Fig. 1), (4) will be modified using (5*a*) and (5*b*) within the forward scattering approximation (Ishizuka & Uyeda, 1977):

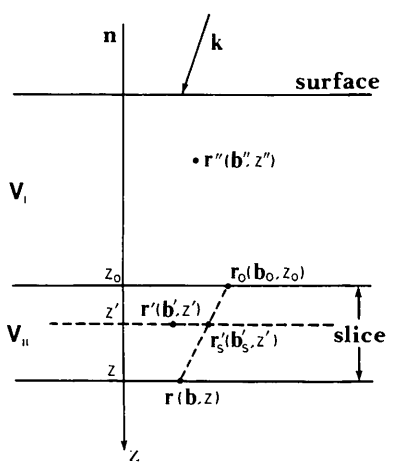

Fig. 1. A division of an integration in the integral form of the Schrödinger equation appropriate for the forward scattering approximation. The region V_{I} shows a part of specimen preceding the region V_{II} which forms a slice. Here a wave function at the bottom surface of the slice V_{II} is considered. The incident surface is normal to the z direction, and the system is described by Cartesian coordinates.

$$\varphi(\mathbf{b},z) = (k_z/k)\int \varphi(\mathbf{b}_0,z_0)p(\mathbf{b}-\mathbf{b}_0, z-z_0)\,\mathrm{d}\mathbf{b}_o + \left(-\frac{i}{\hbar v}\right)\iint_{V_{\mathrm{II}}} V(\mathbf{b}',z')\varphi(\mathbf{b}',z') \times p(\mathbf{b}-\mathbf{b}', z-z')\,\mathrm{d}\mathbf{b}'\,\mathrm{d}z'. \tag{6}$$

This integral equation is solved by expanding $\varphi(\mathbf{b},z)$ in an infinite series:

$$\varphi(\mathbf{b},z) = \sum_{n=0}^{\infty}\left(-\frac{i}{\hbar v}\right)^n f_n(\mathbf{b},z). \tag{7}$$

The substitution of (7) into (6) results in the following set of equations:

$$f_n(\mathbf{b},z) = \iint_{V_{\mathrm{II}}} V(\mathbf{b}',z')f_{n-1}(\mathbf{b}',z')p(\mathbf{b}-\mathbf{b}', z-z') \times \mathrm{d}\mathbf{b}'\,\mathrm{d}z', \tag{8a}$$

for $n \geq 1$, and

$$f_0(\mathbf{b},z) = (k_z/k)\int \varphi(\mathbf{b}_0,z_0)p(\mathbf{b}-\mathbf{b}_0, z-z_0)\,\mathrm{d}\mathbf{b}_0. \tag{8b}$$

If the potential varies slowly in the region of order $\{|\mathbf{r}-\mathbf{r}_0|/k\}^{1/2}$, then f_1 becomes

$$\begin{aligned} f_1(\mathbf{b},z) &= \int \mathrm{d}\mathbf{b}_0\,\varphi(\mathbf{b}_0,z_0)(k_z/k) \\ &\quad \times \iint_{V_{\mathrm{II}}} \mathrm{d}z'\,\mathrm{d}\mathbf{b}'\,V(\mathbf{b}',z')p(\mathbf{b}'-\mathbf{b}_0, z'-z_0) \\ &\quad \times p(\mathbf{b}-\mathbf{b}', z-z') \\ &\simeq \iint_{V_{\mathrm{II}}} V(\mathbf{b}'_s,z')\varphi(\mathbf{b}_0,z_0)p(\mathbf{b}-\mathbf{b}_o, z-z_0) \\ &\quad \times \mathrm{d}z'\mathrm{d}\mathbf{b}_0, \end{aligned} \tag{9}$$

where $\mathbf{b}'_s = [(z'-z_0)\mathbf{b} + (z-z')\mathbf{b}_0]/(z-z_0)$. Here the small-angle-scattering approximation is assumed and the method of stationary phase is applied (Appendix). The integral on the potential energy V over z' in (9) should be done on the stationary points $\mathbf{b}'_s$ which are located on the line $\mathbf{r}-\mathbf{r}_0$ approximately parallel to the incident-beam direction for the small-angle approximation. When the potential change with $\mathbf{b}$ is neglected in the region of $|\mathbf{r}-\mathbf{r}_0|\alpha$ where α is the maximum scattering angle, the integral over z' can be carried out along the direction of the incident electrons [see (15)], specified by the variable z_k, and (9) finally becomes

$$f_1(\mathbf{b},z) = (k_z/k)\int [(k/k_z)\int V(\mathbf{b}_0,z_k)\,\mathrm{d}z_k] \times \varphi(\mathbf{b}_0,z_0)p(\mathbf{b}-\mathbf{b}_0, z-z_0)\,\mathrm{d}\mathbf{b}_0. \tag{10}$$

With the successive application of the stationary-phase method using the mathematical induction (Ishizuka & Uyeda, 1977) f_n finally reduces to

$$f_n(\mathbf{b},z) = (k_z/k)\int \frac{1}{n!}[(k/k_z)\int V(\mathbf{b}_0,z_k)\,\mathrm{d}z_k]^n \times \varphi(\mathbf{b}_o,z_o)p(\mathbf{b}-\mathbf{b}_0, z-z_0)\,\mathrm{d}\mathbf{b}_0. \tag{11}$$

The substitution of (8*b*) and (11) into (7) and the summation over n give φ as

$$\varphi(\mathbf{b},z) = (k_z/k) \int \exp\left\{-\frac{i}{\hbar v}\frac{k}{k_z} \int V(\mathbf{b}_0,z_k)\,\mathrm{d}z_k\right\} \times \varphi(\mathbf{b}_0,z_0)\,p(\mathbf{b}-\mathbf{b}_0,\, z-z_0)\,\mathrm{d}\mathbf{b}_0. \quad (12)$$

which is simply rewritten with $z_n = z_0$ and $z_{n+1} = z$, using the convolution integral

$$\varphi_{n+1}(\mathbf{b}) = [\varphi_n(\mathbf{b})\, q_n(\mathbf{b})]*[(k_z/k)\, p_n(\mathbf{b})]. \quad (12a)$$

Here $\varphi_n(\mathbf{b}) = \varphi(\mathbf{b},z_n)$ and $q_n(\mathbf{b})$ defines the phase grating:

$$q_n(\mathbf{b}) = \exp\left\{-\frac{i}{\hbar v}\frac{k}{k_z}\int_{z_n}^{z_{n+1}} V(\mathbf{b},z_k)\,\mathrm{d}z_k\right\}. \quad (13)$$

So far an orthogonal system with the z axis perpendicular to the crystal surface has been assumed. When the c axis of the crystal coordinates is not perependicular to the surface as in the case of non-orthogonal systems (Fig. 2), the **b** coordinates of origins of the phase gratings lying on the c axis will shift parallel to the surface. The wave function at the bottom of the nth slice will satisfy (12). The origin of φ_{n+1} must coincide with that of the next phase grating q_{n+1} during the next iteration. If this shift of origin is denoted by $\mathbf{b}_0$ (Fig. 2), then (12*a*) becomes

$$\varphi_{n+1}(\mathbf{b}-\mathbf{b}_0) = [\varphi_n(\mathbf{b})\, q_n(\mathbf{b})]*[(k_z/k)\, p_n(\mathbf{b})]. \quad (14)$$

The Fourier transform of this equation becomes

$$\Phi_{n+1}(\mathbf{u}) = [\Phi_n(\mathbf{u})*Q_n(\mathbf{u})][(k_z/k)P_n(\mathbf{u})\exp(2\pi i\mathbf{u}\mathbf{b}_0)], \quad (14a)$$

where capital letters represent the corresponding Fourier pairs, and **u** defines the reciprocal-lattice plane conjugate to the **b** plane (Fig. 3).

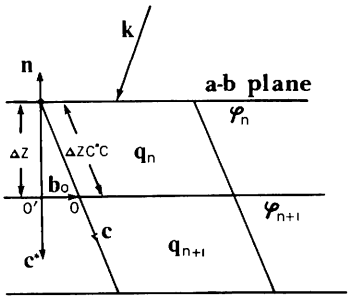

Fig. 2. The relationship of successive slices showing a shift of origins in the Cartesian coordinates. After an appropriate transformation, the x–y plane and z direction of the Cartesian coordinates are regarded as the a–b (real) plane and c^* (reciprocal) direction of crystal coordinates respectively. Then the shift of the origins denoted by $\mathbf{b}_0$ can be simply related in the crystal coordinates.

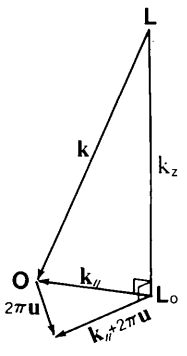

Fig. 3. A relationship of reciprocal-space vectors. The **u** plane is conjugate to the incident surface, *i.e.* the x–y plane. $\mathbf{k}_{\parallel}$ and k_z are components of **k**, which are parallel and perpendicular to the incident surface respectively. L and L_0 are centers of Ewald sphere and Laue circle respectively.

III. Discussion

The new derived formula points out three changes from the usual multislice formula for the normal incidence of electrons as follows.

(1) The interaction constant $\sigma = 1/\hbar v$ should be changed to $\sigma' = (k/k_z)\sigma$. The same modified interaction constant has been used by Anstis & Cockayne (1979), and can be expressed in terms of the wave number as $m/\hbar^2 k_z$, which shows that the specimen *feels* not the whole wave number k but only its component k_z. This result is in accord with the boundary conditions on the entrance surface, which leave the wave component $\mathbf{k}_{\parallel}$ parallel to the surface unchanged.

(2) The direction of integration for calculation of the phase grating should be along the incident direction of the electrons. Since it may not be necessary to divide the atomic potential into two slices (Goodman & Moodie, 1974) the projected potential of one slice will be calculated from the atoms whose centers belong to that slice. Hence a modified Fourier theorem of projection can be applied as follows:

$$\begin{aligned} V_n(\mathbf{b}) &= \int_{z_n}^{z_{n+1}} V(\mathbf{b},z_k)\,\mathrm{d}z_k \\ &\simeq \int_{-\infty}^{\infty} V'(x + zk_x/k_z,\, y + zk_y/k_z,\, z)\,\mathrm{d}z \\ &= \int\int_{-\infty}^{\infty} F'(\mathbf{g})\exp\{2\pi i[g_x(x + zk_x/k_z) \\ &\qquad + g_y(y + zk_y/k_z) + g_z z]\}\,\mathrm{d}\mathbf{g}\,\mathrm{d}z \\ &= \int F'(\mathbf{u}, -\mathbf{u}\mathbf{k}_{\parallel}/k_z)\exp(2\pi i\mathbf{u}\mathbf{b})\,\mathrm{d}\mathbf{u}. \end{aligned} \quad (15)$$

Here $\mathbf{k}_{\parallel} = (k_x,k_y)$ and $\mathbf{u} = \mathbf{g}_{\parallel} = (g_x,g_y)$. $V'(\mathbf{r})$ represents the potential distribution due to the atomic potentials, and $F'(\mathbf{g})$ its Fourier transform. The function V' may

extend out of the slice and the integration should be carried out over $(-\infty,\infty)$. Equation (15) shows that the projected potential should be calculated from the Fourier transform $F'(\mathbf{g})$ for the value of $\mathbf{g}$ located on the plane perpendicular to the incident direction of the electrons: $(\mathbf{u},-\mathbf{u}\mathbf{k}_{\parallel}/k_z)$. $(\mathbf{k}_{\parallel},k_z) = 0$. In the case of periodic objects the integral in (15) becomes a summation over discrete points on the $\mathbf{u}$ plane. It will be noted that the reciprocal points $(\mathbf{u},-\mathbf{u}\mathbf{k}_{\parallel}/k_z)$ defining the projected potential do not coincide with the reciprocal-*lattice* points of the crystal except in the case of zone-axis orientation.

(3) The propagation function based on the spherical wave should be modified as follows:

$$p'_n(\mathbf{b}) = (k_z/k)\, p_n(\mathbf{b} + \mathbf{b}_0).$$

and

$$P'_n(\mathbf{u}) = (k_z/k)\, P_n(\mathbf{u}) \exp(2\pi i \mathbf{u}\mathbf{b}_0),$$

in the real and reciprocal spaces respectively. The factor k_z/k guarantees the normalization as

$$\int p'_n(\mathbf{b})\, \mathrm{d}\mathbf{b} = P'_n(0) = 1. \qquad (5a')$$

The entrance surface (x–y plane) may be regarded as a crystal plane (001), and the z direction may coincide with the reciprocal-lattice vector $\mathbf{c}^*$, since the crystal axes can be transformed if necessary (Fig. 2). Hence $\mathbf{u}\mathbf{b}_0$ will be expressed as

$$\mathbf{u}\mathbf{b}_0 = \mathbf{g}_{\zeta\parallel}\, \mathbf{b}_0 = -\Delta z(g_{\zeta z} - c^*\zeta).$$

Here we used the following relationships:

$$\mathbf{u} = \mathbf{g}_{\zeta\parallel} = (g_{\zeta x}, g_{\zeta y}),$$
$$\mathbf{b}_0 = \Delta z\mathbf{n} + \Delta z c^* \mathbf{c}$$

(see Fig. 2), and

$$\mathbf{g}_\zeta = \xi\mathbf{a}^* + \eta\mathbf{b}^* + \zeta\mathbf{c}^* = (g_{\zeta x}, g_{\zeta y}, g_{\zeta z})$$

(see Fig. 4). The modified propagation function in the reciprocal space is written for the reflection $\mathbf{g}_\zeta$ explicitly as (Appendix § 1):

$$P'(\mathbf{g}_\zeta) = k_z/[k^2 - (\mathbf{k}_{\parallel} + 2\pi\mathbf{g}_{\zeta\parallel})^2]^{1/2} \exp\{-2\pi i \Delta z S(\mathbf{g}_\zeta)\}. \qquad (16)$$

Here

$$S(\mathbf{g}_\zeta) = \{k_z + 2\pi(g_{\zeta z} - c^*\zeta) - [k^2 - (\mathbf{k}_{\parallel} + 2\pi\mathbf{g}_{\zeta\parallel})^2]^{1/2}\}/2\pi, \qquad (17)$$

which expresses the excitation error for each reflection. It should be noted that $S(\mathbf{g}_\zeta)$ is determined only by $\mathbf{g}_{\zeta\parallel}$ equal to $\mathbf{u}$. The excitation error evaluated at $\mathbf{g}_0$ on the $a^* - b^*$ plane is exactly equal to the distance from the reciprocal-lattice point to the Ewald sphere measured along the surface normal. In the multislice formulation all possible reflections $\mathbf{g}$ will be represented at $\mathbf{g}_\parallel$ in the $\mathbf{u}$ plane, which defines the reciprocal-lattice plane conjugate to the surface. The exact expression for the excitation error (17) can be approximated by

$$S(\mathbf{g}) \simeq \frac{1}{2\pi}\frac{k^2 - (\mathbf{k} + 2\pi\mathbf{g}_0)^2}{2(\mathbf{k} + 2\pi\mathbf{g}_0)_z} \qquad (18a)$$

$$\simeq \frac{1}{2\pi}\frac{k^2 - (\mathbf{k} + 2\pi\mathbf{g}_0)^2}{2k_z}. \qquad (18b)$$

Here $\mathbf{g}_0$ is used to represent the reflection at $\mathbf{u}$, so that $\zeta = 0$. It is noted that the slice thickness in (16) is measured along the z direction. If the slice thickness Δz_k measured along the beam direction is used in (16) instead of Δz, k_z in (18b) will be replaced by k.

The upper limit of the slice thickness will be estimated from the above derivation of the multislice formula as follows. The potential distribution should change slowly over the region $(\Delta z_k/k)^{1/2}$ ($A9$) or $\Delta z_k\, \alpha$ (10). If d defines the distance over which the potential does not change appreciably, these two conditions give the same order of the upper limit for the slice thickness Δz_k or Δz as

$$\Delta z_k \lesssim kd^2/\pi \text{ or } \Delta z \lesssim k_z\, d^2/\pi, \qquad (19)$$

where a well-known relationship, $\alpha \simeq 2\pi/kd$, is used.

The maximum inclination angle of the incident electrons will be limited as follows. It is obvious that this formula cannot be applied to the glancing incidence of electrons. Here the forward scattered electrons will go back into the former slices, but that possibility is neglected in (6). Another limitation on the inclination angle is imposed by the approximation ($A5$) which has been used in (6), (9) and (11) through the relation (5b). The approximation is reduced to

$$[k_z^2 - (4\pi\mathbf{k}_\parallel \mathbf{g}_\parallel + 4\pi^2 \mathbf{g}_\parallel^2)]^{1/2} \simeq k_z,$$

so that

$$2\pi g/k \ll \cos^2\beta/\sin\beta$$

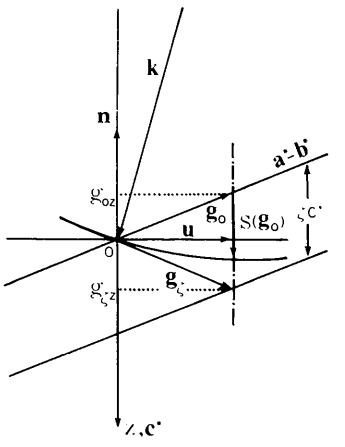

Fig. 4. A diagram showing an excitation error defined by the reciprocal-space propagation function. The excitation error should be measured along the surface normal. If the crystal transformation is assumed as in Fig. 2, a simple expression for the excitation error is obtained. Here $\mathbf{g}_0 = \mathbf{g}_\zeta$, and these points give the same excitation error.

where β is the inclination angle. This inequality will hold more satisfactorily for electrons accelerated by higher voltages. However, the limitation of the inclination angle may be relaxed appreciably for all voltages, because (5*b*) has been obtained with the stationary-phase method in the Fourier transform of (*A*5) (Appendix).

It is worthwhile to consider upper-layer interactions for inclined illumination. Here the upper-layer interactions are defined in real space as a multiple scattering of electrons by atoms in the unit cell. In other words, the upper-layer interactions mean the breakdown of the projection approximation for the phase grating. Therefore, in the multislice calculations, the upper-layer interactions will be introduced when the unit cell is divided into more than one slice to make better phase gratings. In order to get the appropriate phase gratings two points should be considered: the slice thickness and the projection direction. The upper-layer reflections are usually included in the multislice calculation, provided a sufficient number of beams is taken into account. However the calculated reflections will not show correct scattering amplitudes if the slice thickness is not sufficiently small. Goodman & Moodie (1974) showed that 'false' upper-layer reflections will be generated when the slice thickness multiplied by the excitation error, corresponding to (18*b*), becomes more than one. If the slice thickness is made sufficiently small, the phase gratings made from the potential projected along any direction, *e.g.* the zone axis, will introduce the correct upper-layer interactions.

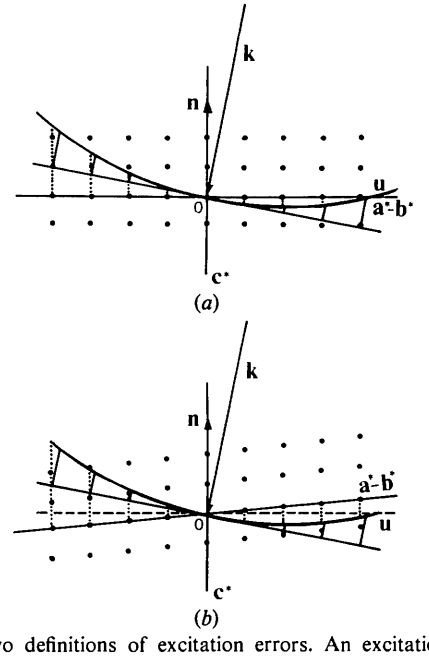

Fig. 5. Two definitions of excitation errors. An excitation error $S(\mathbf{g})$ measured along the surface normal controls the excitation of reflection. The other excitation error $S_k(\mathbf{g})$ measured along the beam direction defines the strength of an 'upper-layer' interaction. (*a*) and (*b*) show these two excitation errors in the case of an orthogonal and a non-orthogonal system respectively. The lengths of the dotted lines show $S(\mathbf{g})$, whereas the lengths of the solid lines show $S_k(\mathbf{g})$. The maximum value of $S_k(\mathbf{g})$ within any resolution limit is smaller than that of $S(\mathbf{g})$.

However, the upper-layer interactions can be better taken into account in the small-angle scattering approximation, if the potential is projected along the incident beam direction, *i.e.* the principal scattering direction. Here the projected potential is calculated from the structure factors on the plane perpendicular to the incident beam. A parameter for the strength of the upper-layer interaction will be defined in terms of the excitation error of the reciprocal points used to define the projected potential. This excitation error may be measured along the projection direction, *i.e.* the beam direction, and expressed as

$$S_k(\mathbf{g}) = \{k - [k^2 - (2\pi g)^2]^{1/2}\}/2\pi,$$

where $\mathbf{g}$ is the reciprocal point defining the projected potential. The parameter of the 'pseudo' upper-layer interaction due to a finite slice thickness is defined by this excitation error multiplied by the slice thickness Δz_k as

$$\Delta z_k\, S_k(\mathbf{g}) \simeq \pi \Delta z_k\, g^2/k.$$

The upper limit of the slice thickness given by (19) limits this parameter to one. This means the slice thickness given by (19) may guarantee the 'correct' upper-layer interactions. It can be seen from Fig. 5 that the maximum value of $S_k(\mathbf{g})$ within any resolution limit is smaller than that of $S(\mathbf{g})$, so that the pseudo upper-layer interactions due to a finite slice thickness are small when the potential is projected along the beam direction. Some reflections of small excitation error $S(\mathbf{g})$ may have large upper-layer interactions $S_k(\mathbf{g})$ in the case of tilted illumination. However, in the normal illuminated zone-axis orientation, two excitations, $S(\mathbf{g})$ and $S_k(\mathbf{g})$, become identical.

The modified multislice formula may be used for wedge crystals (Goodman & Moodie, 1974) except at the edge of the wedge. Here the wedge will be sliced parallel to the entrance surface for the electrons, and the outgoing wave, *i.e.* the wave function at the exit surface, will be expressed by the summation of the wave functions for each specimen thickness.

Some calculations are in progress to test the present theory for tilted-convergent-beam patterns and electron diffraction from non-orthogonal systems, and will be published elsewhere. They will include examinations of some approximations for the excitation error and of the upper-layer interactions.

IV. Conclusion

A multislice formula which may be used for illumination inclined to the entrance-surface normal of a

crystal has been derived, based on the integral form of the Schrödinger equation. This formula shows the changes which are necessary to the propagation and phase-grating functions relative to the usual formula for normal incidence. The generalized propagation function for the spherical wave gives the exact excitation error measured along the surface normal. The phase grating parallel to the incidence surface should be calculated with a modified interaction constant and the potential projected along the direction of the incident electrons.

For the inclined incidence of electrons, the upper-layer interactions will give serious errors when the excitation errors measured along the beam direction multiplied by the slice thickness are large. The maximum slice thickness imposed by the approximations during the derivation of the multislice formula guarantees the correct upper-layer interactions.

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APPENDIX

1. Fourier transform of the propagation function $p(\mathbf{r})$

We consider the 2D Fourier transform of $p(\mathbf{b},z)$ on the $\mathbf{b}$ plane,

$$P(\mathbf{u}) = \int \frac{1}{\lambda i}\frac{1}{r}\exp i\{kr - \mathbf{kr}\}\exp(-2\pi i\mathbf{ub})\,\mathrm{d}\mathbf{b} \quad (A1)$$

$$= \frac{1}{\lambda i}\exp(-ik_z z)\int \frac{\exp ik(\mathbf{b}^2 + z^2)^{1/2}}{(\mathbf{b}^2 + z^2)^{1/2}} \times \exp\left\{-2\pi i\left(\mathbf{u} + \frac{\mathbf{k}_\parallel}{2\pi}\right)\mathbf{b}\right\}\mathrm{d}\mathbf{b},$$

where $\mathbf{k}_\parallel = (k_x,k_y)$ and $\mathbf{u}$ is a vector on 2D reciprocal space. This Fourier transform can be carried out successively over x and y, and results in

$$P(\mathbf{u}) = \{k/[k^2 - (\mathbf{k}_\parallel + 2\pi\mathbf{u})^2]^{1/2}\} \times \exp(-iz\{k_z - [k^2 - (\mathbf{k}_\parallel + 2\pi\mathbf{u})^2]^{1/2}\}), \quad (A2)$$

which propagates as an evanescent wave when $|\mathbf{k}_\parallel + 2\pi\mathbf{u}| > k$. The Fourier pair used in the first transform (say for x) is as follows:

$$\exp[ib(x^2 + a^2)^{1/2}]/(x^2 + a^2) \langle=\rangle \begin{cases} i\pi H_0^{(1)}\{a[b^2 - (2\pi\xi)^2]^{1/2}\} & \text{for } b > |2\pi\xi|, \\ 2K_0\{a[(2\pi\xi)^2 - b^2]^{1/2}\} & \text{for } b < |2\pi\xi|, \end{cases}$$

and in the second transform (say for y):

$$\exp[ib(a^2 - x^2)^{1/2}]/(a^2 - x^2)^{1/2} \langle=\rangle \pi H_0^{(1)}\{a[(2\pi\xi)^2 + b^2]^{1/2}\}$$

and

$$\exp[-b(x^2 + a^2)^{1/2}]/(x^2 + a^2) \langle=\rangle 2K_0\{a[(2\pi\xi)^2 + b^2]^{1/2}\}.$$

Here x and ξ are conjugate variables and $\langle=\rangle$ shows a Fourier pair. $H_0^{(1)} = J_0 + iN_0$. $H_0^{(1)}$, K_0, J_0 and N_0 are the zeroth-order first-kind Hankel function, second-kind modified Bessel function, Bessel function and Neumann function respectively.

2. Some properties of the spherical propagation function (3)

The normalization is given by

$$\int p(\mathbf{b},z)\,\mathrm{d}\mathbf{b} = P(0) = k/k_z. \quad (A3)$$

The next equation is readily shown using $(A2)$ as follows:

$$P(\mathbf{u}, z - z_1)P(\mathbf{u}, z_2 - z) = \frac{k}{[k^2 - (\mathbf{k}_\parallel + 2\pi\mathbf{u})^2]^{1/2}} \times P(\mathbf{u}, z_2 - z_1), \quad (A4)$$

which can be approximated for small-angle scattering as

$$P(\mathbf{u}, z - z_1)P(\mathbf{u}, z_2 - z) \simeq (k/k_z)P(\mathbf{u}, z_2 - z_1). \quad (A5)$$

The Fourier transform of this equation provides $(5b)$ using the convolution theorem:

$$p(\mathbf{b}, z - z_1) * p(\mathbf{b}, z_2 - z) \simeq (k/k_z)p(\mathbf{b}, z_2 - z_1). \quad (A6)$$

This approximation can be justified with the stationary-phase method applied to the Fourier transform of the r.h.s. of $(A4)$. Here a stationary point appears at

$$\mathbf{k}_\parallel + 2\pi\mathbf{u}_s = [k/(b^2 + z^2)^{1/2}]\mathbf{b},$$

so that the factor on the r.h.s. of $(A4)$ becomes

$$k/[k^2 - (\mathbf{k}_\parallel + 2\pi\mathbf{u}_s)^2]^{1/2} = (b^2 + z^2)^{1/2}/z \simeq k/k_z,$$

and the Fourier transform of $(A4)$ results in $(A6)$.

3. Derivation of equation (9)

From the definition of the propagation function (3),

$$p(\mathbf{r}' - \mathbf{r}_0)p(\mathbf{r} - \mathbf{r}') = \frac{1}{(\lambda i)^2}\frac{1}{|\mathbf{r}' - \mathbf{r}_0|}\frac{1}{|\mathbf{r} - \mathbf{r}'|} \times \exp\{-i\mathbf{k}(\mathbf{r} - \mathbf{r}_0)\} \times \exp ik\{|\mathbf{r}' - \mathbf{r}_0| + |\mathbf{r} - \mathbf{r}'|\}. \quad (A7)$$

The last term is a function of $\mathbf{b}'$, which has a stationary point at

$$\mathbf{b}'_s = [(z' - z_0)\mathbf{b} + (z - z')\mathbf{b}_0]/(z - z_0).$$

In the small-angle-scattering approximation it is sufficient to consider the directions of $\mathbf{r}' - \mathbf{r}_0$ and $\mathbf{r} - \mathbf{r}'$ close to the incident direction of electrons, that is, the region of $\mathbf{b}'$ close to $\mathbf{b}'_s$. Then the distance $|\mathbf{r}' - \mathbf{r}_0| + |\mathbf{r} - \mathbf{r}'|$ will be approximated as

$$|\mathbf{r}' - \mathbf{r}_0| + |\mathbf{r} - \mathbf{r}'| \simeq |\mathbf{r}'_s - \mathbf{r}_0| + |\mathbf{r} - \mathbf{r}'_s| + \tfrac{1}{2}\left\{\frac{1}{|\mathbf{r}'_s - \mathbf{r}_0|} + \frac{1}{|\mathbf{r} - \mathbf{r}'_s|}\right\} \times (\mathbf{b}' - \mathbf{b}'_s)^2,$$

where $\mathbf{r}'_s = (\mathbf{b}'_s, z')$. Since $|\mathbf{r}'_s - \mathbf{r}_0| + |\mathbf{r} - \mathbf{r}'_s| = |\mathbf{r} - \mathbf{r}_0|$, $(A7)$ becomes

$$p(\mathbf{r}' - \mathbf{r}_0)p(\mathbf{r} - \mathbf{r}') \simeq \frac{1}{(\lambda i)^2}\frac{1}{|\mathbf{r}' - \mathbf{r}_0|}\frac{1}{|\mathbf{r} - \mathbf{r}'|} \times \exp i\{k|\mathbf{r} - \mathbf{r}_0| - \mathbf{k}(\mathbf{r} - \mathbf{r}_0)\} + \exp i\left\{\frac{k}{2}\frac{|\mathbf{r} - \mathbf{r}_0|}{|\mathbf{r}'_s - \mathbf{r}_0||\mathbf{r} - \mathbf{r}'_s|} \times (\mathbf{b}' - \mathbf{b}'_s)^2\right\}. \quad (A8)$$

When the potential distribution does not change appreciably over a distance of $\{|\mathbf{r} - \mathbf{r}_0|/2k\}^{1/2}$, which gives the upper limit of

$$\left\{\frac{2}{k}\frac{|\mathbf{r}'_s - \mathbf{r}_0||\mathbf{r} - \mathbf{r}'_s|}{|\mathbf{r} - \mathbf{r}_0|}\right\}^{1/2}$$

through a relationship between geometric and arithmetic means, the integration over $\mathbf{b}'$ in (9) can be carried out using the method of stationary phase (Eckart, 1948):

$$\begin{aligned}(k_z/k)\int V(\mathbf{b}',z')\,p(\mathbf{b}' - \mathbf{b}_0, z' - z_0) &\times p(\mathbf{b} - \mathbf{b}', z - z')\,\mathrm{d}\mathbf{b}' \\ &\simeq (k_z/k)V(\mathbf{b}'_s,z')\int p(\mathbf{b}' - \mathbf{b}_0, z' - z_0) \times p(\mathbf{b} - \mathbf{b}', z - z')\,\mathrm{d}\mathbf{b}' \\ &\simeq V(\mathbf{b}'_s,z')\,p(\mathbf{b} - \mathbf{b}_0, z - z_0), \quad (A9)\end{aligned}$$

where $(A6)$ has been used to derive the last equation.

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