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Electron Virtual Inelastic Scattering in the Multislice Scheme

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Abstract

A multislice theory has been developed for including the virtual inelastic scattering in dynamical calculations of high-energy electron diffraction. The effects on elastic waves of all inelastic processes, such as single-electron excitation, plasmon excitation and phonon scattering, can be characterized by a complex correction potential. Its real part describes the virtual inelastic process and its imaginary part represents the inelastic absorption effect. This potential is directly related to the generalized dielectric response function of the crystal.

1. Introduction

The early theory of simulating electron inelastic scattering in a crystal was described by Yoshioka (1957). Based on quantum mechanical theory, he derived a set of coupled Schrödinger equations by considering all the possible inelastic transitions between the ground state and the excited states of the crystal. These equations, in principle, govern the wave behavior of electrons, but it is extremely difficult to solve them even numerically.

The first application of Yoshioka's theory was to consider the effects of inelastic scattering on the elastic scattered wave. In electron diffraction, an electron can lose a certain amount of energy (inelastic scattering) and then regain the same amount of energy to become an 'elastic' electron again. This process is the so-called virtual inelastic scattering initiated by Yoshioka (1957). Besides the crystal potential due to atomic arrangement, there will be a real addition to the crystal potential from virtual inelastic scattering as well as the imaginary absorption correction. Yoshioka (1957) gave an expression for this correction potential in the Bloch wave scheme. The corrections due to single-electron inelastic scattering were calculated using the Thomas-Fermi atomic model (Yoshioka, 1957) and atomic wave functions (Whelan, 1965). This correction has also been calculated by Humphreys & Hirsch (1968) for different inelastic excitation processes. The contribution due to plasmon excitation was investigated by Radi (1970) and Yoshioka & Kainuma (1962).

Several methods have been developed to solve Yoshioka's equations in order to investigate the inelastic scattering behavior of the electrons in a crystal. Based on the Bloch wave approach under small-angle approximation, Howie (1963) gave some analytical solution for phonon excitations under some simplified conditions. This Bloch wave approach is usually limited by the assumption of periodic structures, which gives the difficulty of approaching general crystal defects. Serneels, Haentjens & Gevers (1980) have proposed an iteration method for solving these coupled equations based on the Bloch wave theory. However, this method may be limited by the convergence of the iteration.

Recently, Wang (1989) has proposed a generalized multislice method for solving Yoshioka's coupling equations. This approach can easily introduce a non-periodic structure in the calculation and considers all the possible transitions among the excited states. This theory has been applied to describe the thermal diffuse scattering in simulating high-angle annular-dark-field scanning transmission electron microscopy (STEM) lattice images (Wang & Cowley, 1990*a*, *b*). In practice, however, it is difficult to calculate all the inelastic waves individually because a large number of excited states are always involved. This makes it almost impossible to estimate the contribution of virtual inelastic scattering to the elastic wave. Therefore, it is desirable to seek an approximate method which can take into account the effects of virtual inelastic processes without knowing each individual inelastic component during the scattering. This is useful in the quantitative simulation of elastic electron images in high-resolution electron microscopy (HREM), especially when an energy filter is used. This is the purpose of the present paper.

2. Virtual inelastic process in the multislice scheme

For the convenience of theoretical analysis, it would be helpful to review the coupling equations derived by Yoshioka (1957) and the multislice solution given by Wang (1989). If one considers the interaction of an incident electron with a crystal, the Schrödinger equation of the system (the electron and the crystal) is

$$[-(\hbar^2/2m_0)\nabla^2 + H_c + H']\Phi = E\Phi, \qquad (1)$$

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where $-(\hbar^2/2m_0)\nabla^2$ is the kinetic energy of the electron, H_c is the crystal Hamiltonian and H' describes the interaction between the electron and the solid. $\Phi(\mathbf{r}, \mathbf{r}_1, \ldots, \mathbf{r}_M)$ is the wave function of the system, depending on $\mathbf{r}$, the coordinates of the incident electron, and on $\mathbf{r}_1, \ldots, \mathbf{r}_M$ the coordinates of the electrons and ions of the crystal. Neglecting exchange effects one can write

$$\Phi(\mathbf{r}, \mathbf{r}_1, \ldots, \mathbf{r}_M) = \sum_n a_n(\mathbf{r}_1, \ldots, \mathbf{r}_M)\Psi_n(\mathbf{r}), \quad (2)$$

where a_n is the wave function of the crystal in its nth excited state of energy ε_n so that

$$H_c a_n = \varepsilon_n a_n. \quad (3)$$

Ψ_0 in (2) describes the elastic scattered wave of energy $E_0 = E$, and Ψ_n describes the inelastically scattered wave of energy $E_n = E - \varepsilon_n$, with $n = 1, 2, \ldots m$. Substituting (2) and (3) into (1), multiplying by a_n^* integrating over the coordinates $\mathbf{r}_1, \ldots, \mathbf{r}_M$, we have

$$(\nabla^2 + k_0^2)\Psi_0 = \sum_m (2m_0/\hbar^2)H'_{0m}(\mathbf{r})\Psi_m, \quad (4a)$$

$$(\nabla^2 + k_n^2)\Psi_n = \sum_m (2m_0/\hbar^2)H'_{nm}(\mathbf{r})\Psi_m, \quad (4b)$$

where

$$k_n^2 = (2m_0/\hbar^2)E_n, \quad (4c)$$

and

$$H'_{nm} = \int a_n^* H' a_m \, d\mathbf{r}_1, \ldots, d\mathbf{r}_M. \quad (4d)$$

These are Yoshioka's coupling equations for inelastic scattering. By assuming $\Psi_n = \exp(i\mathbf{k}_n \cdot \mathbf{r})\varphi_n$, Wang (1989) has shown that the waves φ_n going into a crystal slice at $z = z_0$ and those coming out of the slice at $z = z$ are related by

$$\begin{pmatrix} \varphi_0(\mathbf{b}, z) \\ \varphi_1(\mathbf{b}, z) \\ \vdots \\ \varphi_m(\mathbf{b}, z) \end{pmatrix} = \int d\mathbf{b}_0 \begin{pmatrix} P_0(\mathbf{b}-\mathbf{b}_0, \Delta z) & 0 & \cdots & 0 \\ 0 & P_1(\mathbf{b}-\mathbf{b}_0, \Delta z) & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \cdots & P_m(\mathbf{b}-\mathbf{b}_0, \Delta z) \end{pmatrix} \times \exp\left\{ -\frac{i}{\hbar v} \begin{pmatrix} h'_{00}(\mathbf{b}_0, \Delta z) & h'_{01}(\mathbf{b}_0, \Delta z) & \cdots & h'_{0m}(\mathbf{b}_0, \Delta z) \\ h'_{10}(\mathbf{b}_0, \Delta z) & h'_{11}(\mathbf{b}_0, \Delta z) & \cdots & h'_{1m}(\mathbf{b}_0, \Delta z) \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0}(\mathbf{b}_0, \Delta z) & h'_{m1}(\mathbf{b}_0, \Delta z) & \cdots & h'_{mm}(\mathbf{b}_0, \Delta z) \end{pmatrix} \right\} \times \begin{pmatrix} \varphi_0(\mathbf{b}_0, z_0) \\ \varphi_1(\mathbf{b}_0, z_0) \\ \vdots \\ \varphi_m(\mathbf{b}_0, z_0) \end{pmatrix} \quad (5a)$$

where

$$h'_{nm} = \int_{z_0}^{z} H'_{nm}(\mathbf{b}, z')\, dz', \quad (5b)$$

$\mathbf{b} = (x, y)$ and $\mathbf{b}_0 = (x_0, y_0)$. P_n is defined as a propagation function,

$$P_n(\mathbf{b}, \Delta z) = (1/i\lambda_n \Delta z)\exp(i\pi b^2/\lambda_n \Delta z). \quad (5c)$$

λ_n is the wavelength of the electron with energy E_n. Equation (5) is the multislice solution of Yoshioka's coupling equations. This solution can be conveniently applied to different inelastic processes, such as plasmon diffuse scattering and thermal diffuse scattering (Wang, 1989). It can be proved that the total intensity $\sum_{n=0}^{m} \int d\mathbf{b}|\varphi_n|^2$ before and after penetrating through a crystal slice is conserved (see Appendix *A*). For $|H_{nm}| \ll |H_{nn}|$, $H'_{00} \simeq H'_{11} \simeq \ldots \simeq H'_{mm}$ and $\sigma|H_{nm}| \ll 1$ for $n \neq m$, with $\sigma = (1/\hbar v)$ then (5) can be written as (6) under the first-order approximation.

$$\begin{pmatrix} \varphi_0(\mathbf{b}, z) \\ \varphi_1(\mathbf{b}, z) \\ \vdots \\ \varphi_m(\mathbf{b}, z) \end{pmatrix} = \begin{pmatrix} \{\exp(-i\sigma h'_{00})[\varphi_0(\mathbf{b}, z_0) - i\sigma \sum_{n\neq 0} h'_{0n}\varphi_n(\mathbf{b}, z_0)]\} * P_0 \\ \{\exp(-i\sigma h'_{11})[\varphi_1(\mathbf{b}, z_0) - i\sigma \sum_{n\neq 1} h'_{1n}\varphi_n(\mathbf{b}, z_0)]\} * P_1 \\ \vdots \\ \{\exp(-i\sigma h'_{mm})[\varphi_m(\mathbf{b}, z_0) - i\sigma \sum_{n\neq m} h'_{mn}\varphi_n(\mathbf{b}, z_0)]\} * P_m \end{pmatrix} \quad (6)$$

where * indicates convolution.

In (6), for the elastic scattered wave φ_0, the first term is the phase grating result of the crystal slice, which is the elastic penetration of the incident elastic wave. The terms containing h'_{0n} are the 'transitions' of the electrons from the excited states to the ground state, which is the result of virtual inelastic scattering. It is obvious that the generated virtual inelastic scattering from each slice depends on the incident inelastic waves. In other words, the contribution of virtual inelastic scattering to the elastic scattered wave depends strongly on the incident inelastic waves, which is related to the 'history' of electron scattering. The multislice solution (6) cannot be easily performed in practical calculation, because a real inelastic scattering can involve many excited states of different energy and different momentum, such as valence excitations. It is necessary to look for an approximate solution in the multislice scheme for φ_0 if one is interested only in the elastic scattered wave. This is the goal of the following work.

We start firstly from the original Schrödinger equations (4). By neglecting the transitions from the other excited states to the nth state and keeping only

the transition from the ground state, an integral solution of (4b) can be written as (Yoshioka, 1957)

$$\Psi_n(\mathbf{r}) \simeq -(2m_0/4\pi\hbar^2)\int (\exp ik_n|\mathbf{r}-\mathbf{r}'|/|\mathbf{r}-\mathbf{r}'|) \times H'_{n0}(\mathbf{r}')\Psi_0(\mathbf{r}')\,d\mathbf{r}'. \quad (7)$$

Insert (7) into (4a), then the wave equation for the elastic wave is

$$[\nabla^2 + k_0^2 - (2m_0/\hbar^2)H'_{00}(\mathbf{r})]\Psi_0(\mathbf{r}) + (2m_0/\hbar^2)\int A(\mathbf{r},\mathbf{r}')\Psi_0(\mathbf{r}')\,d\mathbf{r}' = 0, \quad (8a)$$

where

$$A(\mathbf{r},\mathbf{r}') = \sum_{n\neq 0} (m_0/2\pi\hbar^2)H'_{0n}(\mathbf{r})H'_{n0}(\mathbf{r}') \times [\exp(ik_n|\mathbf{r}-\mathbf{r}'|)/|\mathbf{r}-\mathbf{r}'|]. \quad (8b)$$

It is interesting to note that the virtual inelastic processes introduce an extra term containing a core function $A(\mathbf{r},\mathbf{r}')$ in (8a). By taking $\Psi_0 = \exp(i\mathbf{k}_n\cdot\mathbf{r})\varphi_0$, the integral solution of (8a) is

$$\varphi_0(\mathbf{r}) = 1 - (2m_0/4\pi\hbar^2)\int F(\mathbf{r}-\mathbf{r}',\mathbf{k}_0)\{H'_{00}(\mathbf{r}')\varphi_0(\mathbf{r}') - \int A(\mathbf{r}',\mathbf{r}'')\varphi_0(\mathbf{r}'')\,d\mathbf{r}''\}\,d\mathbf{r}', \quad (9a)$$

with

$$F(\mathbf{r}-\mathbf{r}',\mathbf{k}_n) = (\exp\{i[k_n|\mathbf{r}-\mathbf{r}'| - \mathbf{k}_n\cdot(\mathbf{r}-\mathbf{r}')]\})/|\mathbf{r}-\mathbf{r}'|). \quad (9b)$$

General speaking, it is difficult to solve (9a), and certain approximations have to be made for different cases. When the high-energy electron scattering satisfies the following conditions: (1) $\alpha^2 \ll 1$, α is the scattering angle, and (2) $|\mathbf{r}-\mathbf{r}'| \simeq z-z'$, thus

$$k_n|\mathbf{r}-\mathbf{r}'| - \mathbf{k}_n\cdot(\mathbf{r}-\mathbf{r}') \simeq k_n[|\mathbf{b}-\mathbf{b}'|^2/2(z-z')]$$

then (9a) becomes

$$\varphi_0(\mathbf{r}) = 1 - (i/\hbar v)\int P_0(\mathbf{b}-\mathbf{b}', z-z')\{H'_{00}(\mathbf{r}')\varphi_0(\mathbf{r}') - \int A(\mathbf{r}',\mathbf{r}'')\varphi_0(\mathbf{r}'')\,d\mathbf{r}''\}\,d\mathbf{r}', \quad (10)$$

where v is the velocity of the electron and P_0 is the propagation function defined in (5c). By defining a crystal potential

$$U = H'_{00}/e \quad (11a)$$

and

$$U'(\mathbf{r},\mathbf{r}') = -\int P_0(\mathbf{b}-\mathbf{b}'', z-z'') \times A(\mathbf{r}'',\mathbf{r}')\,d\mathbf{r}''/eP_0(\mathbf{b}-\mathbf{b}', z-z'), \quad (11b)$$

switching the variables $\mathbf{r}''$ and $\mathbf{r}'$ in the last term of (10), we have

$$\varphi_0(\mathbf{r}) = 1 - (ie/\hbar v)\int P_0(\mathbf{b}-\mathbf{b}', z-z') \times\{U(\mathbf{r}') + U'(\mathbf{r},\mathbf{r}')\}\varphi_0(\mathbf{r}')\,d\mathbf{r}'. \quad (12)$$

In order to find the multislice solution of (12), we need to derive a relationship between the wave, $\varphi_0(\mathbf{b}, z_0)$, going into a crystal slice of thickness $(z-z_0)$ and that, $\varphi_0(\mathbf{b}, z)$, after being scattered by the crystal slice. From (12), if we look particularly at the wave at $z=z$, then

$$\begin{aligned}\varphi_0(\mathbf{b},z) &= 1 + (-ie/\hbar v)\int_{z'=-\infty}^{z'=z}\int P_0(\mathbf{b}-\mathbf{b}', z-z') \times\{U(\mathbf{b}',z') + U'(\mathbf{b},z,\mathbf{b}',z')\} \times\varphi_0(\mathbf{b}',z')\,d\mathbf{b}'\,dz' \\ &= 1 + (-ie/\hbar v)\int_{z''=-\infty}^{z''=z_0}\int P_0(\mathbf{b}-\mathbf{b}'', z-z'') \times\{U(\mathbf{b}'',z'') + U'(\mathbf{b},z,\mathbf{b}'',z'')\} \times\varphi_0(\mathbf{b}'',z'')\,d\mathbf{b}''\,dz'' \\ &\quad + (-ie/\hbar v)\int_{z'=z_0}^{z'=z}\int P_0(\mathbf{b}-\mathbf{b}', z-z') \times\{U(\mathbf{b}',z') + U'(\mathbf{b},z,\mathbf{b}',z')\} \times\varphi_0(\mathbf{b}',z')\,d\mathbf{b}'\,dz'. \end{aligned} \quad (13)$$

From the following properties (Ishizuka & Uyeda, 1977):

$$\int P_0(\mathbf{b}-\mathbf{b}_0, z-z_0)\,d\mathbf{b}_0 = 1 \quad (14a)$$

and

$$P_0(\mathbf{b}-\mathbf{b}'', z-z'') = \int P_0(\mathbf{b}-\mathbf{b}_0, z-z_0)P_0(\mathbf{b}_0-\mathbf{b}'', z_0-z'')\,d\mathbf{b}_0, \quad (14b)$$

(13) can be rewritten as

$$\begin{aligned}\varphi_0(\mathbf{b},z) &= \int P_0(\mathbf{b}-\mathbf{b}_0, z-z_0)\varphi_0(\mathbf{b}_0,z_0)\,d\mathbf{b}_0 \\ &\quad + (-ie/\hbar v)\int_{z'=z_0}^{z'=z}\int P_0(\mathbf{b}-\mathbf{b}', z-z') \times\{U(\mathbf{b}',z') + U'(\mathbf{b},z,\mathbf{b}',z')\} \times\varphi_0(\mathbf{b}',z')\,d\mathbf{b}'\,dz'. \end{aligned} \quad (15)$$

The first term in (15) indicates the expanding propagation of the incident wave through the 'vacuum' for a distance $z-z_0$; the second term is the scattering results of the atoms within the slice. In general, (15) cannot be solved accurately, because the interaction term U' is involved. To find the first-order solution, we assume that (15) can be expanded in the powers of $(-ie/\hbar v)$,

$$\varphi_0(\mathbf{b},z) = \int P_0(\mathbf{b}-\mathbf{b}_0, z-z_0)\varphi_0(\mathbf{b}_0,z_0) \times\sum_{L=0}^{\infty}[(-ie/\hbar v)^L f_L(\mathbf{b},z,\mathbf{b}_0,z_0)]\,d\mathbf{b}_0. \quad (16)$$

Put (16) into (15) and consider that the crystal slice is so thin that the variation of U' in the range $(z-z_0)$ is small. By comparing the coefficients of $(-ie/\hbar v)^L$, one has

$$f_0 = 1 \quad (17a)$$

and for $L \geq 1$

$$\int P_0(\mathbf{b}-\mathbf{b}_0, z-z_0)\varphi_0(\mathbf{b}_0, z_0) f_L(\mathbf{b}, z, \mathbf{b}_0, z_0)\, d\mathbf{b}_0$$
$$= \int \int_{z'=z_0}^{z'=z} P_0(\mathbf{b}-\mathbf{b}', z-z')[U(\mathbf{b}', z') + U'(\mathbf{b}, z, \mathbf{b}', z')$$
$$\times \int P_0(\mathbf{b}'-\mathbf{b}_0, z'-z_0)\varphi_0(\mathbf{b}_0, z_0)$$
$$\times f_{L-1}(\mathbf{b}', z', \mathbf{b}_0, z_0)]\, d\mathbf{b}_0\, d\mathbf{b}'\, dz'. \quad (17b)$$

Integrating over $\mathbf{b}'$ using the method of stationary phase (Ishizuka & Uyeda, 1977) and assuming that U, U' and f_L vary slowly in the region of $z-z_0$, we have

$$f_L(\mathbf{b}, z, \mathbf{b}_0, \Delta z) \simeq \int_{z'=z_0}^{z'=z} [U(\mathbf{b}', z') + U'(\mathbf{b}, z, \mathbf{b}', z')] \times f_{L-1}(\mathbf{b}', z', \mathbf{b}_0, z_0)\, dz', \quad (18a)$$

where $\mathbf{b}' = [(z'-z_0)\mathbf{b} + (z-z')\mathbf{b}_0/z-z_0]$. Using the non-deflection approximation, $\mathbf{b}' \simeq \mathbf{b}_0$, (18$a$) can be written approximately as

$$f_L(\mathbf{b}, z, \mathbf{b}_0, \Delta z) \simeq \int_{z'=z_0}^{z'=z} [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')] \times f_{L-1}(\mathbf{b}_0, z', \mathbf{b}_0, z_0)\, dz'. \quad (18b)$$

It can be proved that the solution of (18b) is (see Appendix B)

$$f_L = \left(\frac{1}{L!}\right)\left\{\int_{z'=z_0}^{z'=z} [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')]\, dz'\right\}^L. \quad (19)$$

Then the solution of (16) is

$$\varphi_0(\mathbf{b}, z) \simeq \int P_0(\mathbf{b}-\mathbf{b}_0, z-z_0) \times \exp\left[-i\sigma e \int_{z'=z_0}^{z'=z} [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')]\, dz'\right] \varphi_0(\mathbf{b}_0, z_0)\, d\mathbf{b}_0$$
$$= \left\{\exp\left[-i\sigma e \int_{z'=z_0}^{z'=z} [U(\mathbf{b}, z') + U'(\mathbf{b}, z_0, \mathbf{b}, z')]\, dz'\right] \varphi_0(\mathbf{b}, z_0)\right\} * P_0, \quad (20)$$

where $*$ indicates a convolution operator. Equation (20) is basically the same as the original multislice theory (Cowley & Moodie, 1957) except that an extra potential U' is added. As expected, the effect of virtual inelastic processes on the elastic wave is equivalent to adding a complex correction potential U' to the crystal potential U, whose real part describes the perturbation of virtual inelastic processes and whose imaginary part represents the inelastic absorption effect.

3. Deriving the correction potential of virtual inelastic scattering

It has been shown that the effect of virtual inelastic processes on elastic waves can be characterized approximately by a correction potential, which is defined by (11b). Putting (8b) into (11b), we obtain

$$U'(\mathbf{r}, \mathbf{r}') = -\int P_0(\mathbf{b}-\mathbf{b}'', z-z'') \sum_{n\neq 0} (m_0/2\pi\hbar^2) H'_{0n}(\mathbf{r}'') \times H'_{n0}(\mathbf{r}')[\exp(ik_n|\mathbf{r}'-\mathbf{r}''|)/|\mathbf{r}'-\mathbf{r}''|] \times d\mathbf{r}''/eP_0(\mathbf{b}-\mathbf{b}', z-z'). \quad (21)$$

For high-energy electrons, $k_0 \simeq k_n$ (*i.e.* $P_0 \simeq P_n$) is always satisfied, using the same procedure as in (9) and (10) and *assuming* that the conditions (1) small-angle scattering and (2) $|\mathbf{r}'-\mathbf{r}''| \simeq |z'-z''|$ are satisfied, then by use of the propagation function defined in (5c), (21) can be changed to

$$U'(\mathbf{r}, \mathbf{r}') \simeq -(i/\hbar v) \int\int P_0(\mathbf{b}-\mathbf{b}'', z-z'') \times P_0(\mathbf{b}''-\mathbf{b}', z''-z') \sum_{n\neq 0} H'_{0n}(\mathbf{r}'') H'_{n0}(\mathbf{r}') \times \exp[i\mathbf{k}_n \cdot (\mathbf{r}'-\mathbf{r}'')]\, d\mathbf{r}''/eP_0(\mathbf{b}-\mathbf{b}', z-z'). \quad (22)$$

Integrating over $\mathbf{b}''$ in (22) using the method of stationary phase, we have

$$U'(\mathbf{r}, \mathbf{r}') \simeq -(i/\hbar v) \int \sum_{n\neq 0} H'_{0n}(\mathbf{r}'') H'_{n0}(\mathbf{r}') \times \exp[i\mathbf{k}_n \cdot (\mathbf{r}'-\mathbf{r}'')]\, dz'', \quad (23a)$$

where

$$\mathbf{b}'' = [(z''-z')\mathbf{b} + (z-z'')\mathbf{b}'/(z-z')]. \quad (23b)$$

In general, H' can be written as

$$H'_{nm}(\mathbf{r})\left\langle m \left| \frac{1}{4\pi\varepsilon_0}\left\{\sum_j \frac{e^2}{|\mathbf{r}-\mathbf{r}_j|} - \sum_k \frac{Z_k e^2}{|\mathbf{r}-\mathbf{R}_k|}\right\} \right| n \right\rangle, \quad (24)$$

and

$$(1/|\mathbf{r}-\mathbf{r}_j|) = (1/2\pi^2) \int (d\mathbf{q}/q^2) \exp i\mathbf{q} \cdot (\mathbf{r}-\mathbf{r}_j), \quad (25)$$

where $\mathbf{r}_j$ and $\mathbf{R}_k$ are the positions of the jth electron and the kth nucleus respectively. Using (24) and (25), integrating over z'' in (23a) and through some algebra, one obtains

$$U'(\mathbf{r}, \mathbf{r}') = -\frac{i}{\hbar v}\frac{e^3}{32\pi^5\varepsilon_0^2} \int (d\mathbf{q}/q^2) \exp(i\mathbf{q} \cdot \mathbf{r}') \times \int (d\mathbf{q}'/q'^2) \exp(-i\mathbf{q}' \cdot \mathbf{r}') S(\mathbf{q}, \mathbf{q}')(z-z') \times \delta[(\mathbf{q}-\mathbf{k}_0) \cdot (\mathbf{r}-\mathbf{r}')], \quad (26)$$

where S is the so-called mixed dynamic form factor and is defined as

$$S(\mathbf{q}, \mathbf{q}') = \sum_{n\neq 0} F_{0n}(\mathbf{q}) F_{n0}(-\mathbf{q}'), \quad (27a)$$

where

$$F_{0n}(\mathbf{q}) = \langle n | \sum_j \exp(-i\mathbf{q} \cdot \mathbf{r}_j) | 0 \rangle. \quad (27b)$$

The dynamic form factor S is connected with the generalized dielectric function of the solid by (Kohl & Rose, 1985)

$$S(\mathbf{q}, \mathbf{q}') = \int_{-\infty}^{\infty} S(\mathbf{q}, \mathbf{q}', \omega)\, d\omega, \quad (28a)$$

$$S(\mathbf{q}, \mathbf{q}', \omega) = i\hbar\varepsilon_0 V / \{2\pi e^2 [1 - \exp(-\beta\hbar\omega)]\} \times [q^2/\varepsilon_{\mathbf{qq}'}(\omega) - q'^2/\varepsilon^*_{\mathbf{q}'\mathbf{q}}(\omega)], \quad (28b)$$

where $\beta = (1/k_B T)$, T is the temperature for the system and V is its volume. The dielectric function ε describes the response of the system as a function of total electric potential $\varphi^{\text{tot}}(\mathbf{r}, t) = \varphi^{\text{ext}}(\mathbf{r}, t) + \varphi^{\text{ind}}(\mathbf{r}, t)$, where φ^{ext} and φ^{ind} are the external applied and the induced potential respectively. ε is defined (Adler, 1962) by

$$\varphi^{\text{ext}}(\omega) = \sum_{\mathbf{q}'} \varepsilon_{\mathbf{qq}'}(\omega) \varphi^{\text{tot}}(\omega). \quad (29)$$

Put (28) into (26), we have

$$U'(\mathbf{r}, \mathbf{r}') = -(eV/64\pi^6\varepsilon_0 v) \int (d\mathbf{q}/q^2) \int (d\mathbf{q}'/q'^2) \times \exp[i(\mathbf{q}-\mathbf{q}') \cdot \mathbf{r}'] |z - z'| \times \delta[(\mathbf{q} - \mathbf{k}_0) \cdot (\mathbf{r} - \mathbf{r}')] \times \int_{-\infty}^{\infty} d\omega \{1/[1 - \exp(-\beta\hbar\omega)]\} \times [q^2/\varepsilon_{\mathbf{qq}'}(\omega) - q'^2/\varepsilon^*_{\mathbf{q}'\mathbf{q}}(\omega)]. \quad (30)$$

This is the final expression for the correction potential. It is apparent that U' is a complex function with its real part associated with $\text{Re}(-1/\varepsilon)$ and its imaginary part related to $\text{Im}(-1/\varepsilon)$. The latter is the energy-loss function of the solid and is responsible for the absorption effect. Now apply (30) to the U' which appears in (20):

$$U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z') = -(eV/64\pi^6\varepsilon_0 v) \int (d\mathbf{q}/q^2) \times \int (d\mathbf{q}'/q'^2) \exp[i(\mathbf{q}-\mathbf{q}') \cdot \mathbf{b}_0 + i(q_z - q'_z)z'] \delta(q_z - k'_{0z}) \times \int_{-\infty}^{\infty} d\omega \{1/[1 - \exp(-\beta\hbar\omega)]\} \times [q^2/\varepsilon_{\mathbf{qq}'}(\omega) - q'^2/\varepsilon_{\mathbf{q}'\mathbf{q}}(\omega)]. \quad (31)$$

The δ function characterizes the conservation of momentum in the z direction.

Several assumptions have been made in the derivation of all the above equations. Firstly, the crystal potential U and correction potential U' are assumed to vary slowly in the range of $\Delta z = z - z_0$. This is a good approximation if Δz approaches zero. In practical image simulation for high-energy electrons, the slice thickness is usually taken as 1-3 Å, which gives reasonably good accuracy (Ishizuka & Uyeda, 1977). Secondly, the energy loss of an electron is taken so small that $P_0 \simeq P_n$. This is usually satisfied because most of the electrons will loose an energy of less than a few hundred electron volts, which is much smaller than the kinetic energy of the electrons (100 keV). Finally, small-angle scattering is assumed. This is a good treatment especially for high-energy electrons.

4. Concluding remarks

A modified multislice approach for including virtual inelastic scattering in the dynamic calculations of high-energy electrons has been introduced. The effects of virtual inelastic processes on the elastic wave can be characterized by a correction potential, its real part indicates the virtual inelastic process and its imaginary part represents the inelastic absorption effect. This potential is derived in real space and is connected to the generalized dielectric function of the system. It is feasible, in principle, to simulate the energy-filtered elastic image and diffraction pattern quantitatively using this multislice theory. The total effects of inelastic processes, plasmon excitation, single electron excitation and phonon excitation, should be comprehensively included in the generalized dielectric function of the system. In other words, these effects should be contained in the newly introduced correction potential.

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APPENDIX A

Conservation of total scattering intensity governed by (5a)

From (5a), we define a transition matrix as

$$T(\mathbf{b}_0, \Delta z) = \begin{pmatrix} h'_{00}(\mathbf{b}_0, \Delta z) & h'_{01}(\mathbf{b}_0, \Delta z) & \cdots & h'_{0m}(\mathbf{b}_0, \Delta z) \\ h'_{10}(\mathbf{b}_0, \Delta z) & h'_{11}(\mathbf{b}_0, \Delta z) & \cdots & h'_{1m}(\mathbf{b}_0, \Delta z) \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0}(\mathbf{b}_0, \Delta z) & h'_{m1}(\mathbf{b}_0, \Delta z) & \cdots & h'_{mm}(\mathbf{b}_0, \Delta z) \end{pmatrix} \quad (A1)$$

It is important to note that matrix T has the property

$$T^\dagger = T, \quad (A2)$$

where † means an operation of Hermitian conjugate (*i.e.* complex-conjugate plus transpose). Also

$$\int d\mathbf{b}\, P_n(\mathbf{b} - \mathbf{b}_0, \Delta z) P_n^*(\mathbf{b} - \mathbf{b}_1, \Delta z) = \delta(\mathbf{b}_1 - \mathbf{b}_0). \quad (A3)$$

Then the total intensity after penetrating a crystal

slice can be calculated according to (5a),

$$\sum_n^m \int |\varphi_n(\mathbf{b}, z)|^2 \, d\mathbf{b}$$

$$= \int d\mathbf{b}[\varphi_0^*(\mathbf{b}, z)\varphi_1^*(\mathbf{b}, z) \ldots \varphi_m^*(\mathbf{b}, z)] \begin{pmatrix} \varphi_0(\mathbf{b}, z) \\ \varphi_1(\mathbf{b}, z) \\ \vdots \\ \varphi_m(\mathbf{b}, z) \end{pmatrix}$$

$$= \int d\mathbf{b}_1[\varphi_0^*(\mathbf{b}_1, z_0)\varphi_1^*(\mathbf{b}_1, z_0) \ldots \varphi_m^*(\mathbf{b}_1, z_0)]$$

$$\times \exp[(i/\hbar v)T^\dagger(\mathbf{b}_1, \Delta z)]$$

$$\times \int d\mathbf{b} \begin{pmatrix} P_0^*(\mathbf{b}-\mathbf{b}_1, \Delta z) & 0 & \cdots & 0 \\ 0 & P_1^*(\mathbf{b}-\mathbf{b}_1, \Delta z) & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \cdots & P_m^*(\mathbf{b}-\mathbf{b}_1, \Delta z) \end{pmatrix}$$

$$\times \int d\mathbf{b}_0 \begin{pmatrix} P_0(\mathbf{b}-\mathbf{b}_0, \Delta z) & 0 & \cdots & 0 \\ 0 & P_1(\mathbf{b}-\mathbf{b}_0, \Delta z) & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \cdots & P_m(\mathbf{b}-\mathbf{b}_0, \Delta z) \end{pmatrix}$$

$$\times \exp[-(i/\hbar v)T(\mathbf{b}_0, \Delta z)] \begin{pmatrix} \varphi_0(\mathbf{b}_0, z_0) \\ \varphi_1(\mathbf{b}_0, z_0) \\ \vdots \\ \varphi_m(\mathbf{b}_0, z_0) \end{pmatrix}. \quad (A4)$$

Using property ($A3$) and ($A1$), integrating over $\mathbf{b}$ first, then over $\mathbf{b}_1$, one obtains from ($A4$)

$$\sum_n^m \int |\varphi_n(\mathbf{b}, z)|^2 \, d\mathbf{b} = \sum_n^m \int |\varphi_n(\mathbf{b}_0, z_0)|^2 \, d\mathbf{b}_0. \quad (A5)$$

That is, the total intensity (elastic plus inelastic) is conserved before and after being scattering by a crystal slice.

APPENDIX *B*

Proof of (19)

Mathematical induction is used to prove (19). We first consider f_1. From (18b),

$$f_1(\mathbf{b}, z, \mathbf{b}_0, \Delta z) \simeq \int_{z'=z_0}^{z'=z} [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')] \, dz'. \quad (B1)$$

Now it is assumed that the solution of f_L is ($B3$),

$$f_L \simeq (1/L!)\left\{ \int_{z'=z_0}^{z'=z} [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')] \, dz' \right\}^L. \quad (B2)$$

According to (18b), one has

$$\begin{aligned} f_{L+1}(\mathbf{b}, z, \mathbf{b}_0, \Delta z) &\simeq \int_{z'=z_0}^{z'=z} [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')] \\ &\quad \times f_L(\mathbf{b}_0, z', \mathbf{b}_0, z_0) \, dz', \\ &\simeq (1/L!) \int_{z'=z_0}^{z'=z} dz' [U(\mathbf{b}_0, z') \\ &\quad + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')] \\ &\quad \times \left\{ \int_{z''=z_0}^{z''=z} dz'' [U(\mathbf{b}_0, z'') \right. \\ &\quad \left. + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z'')] \right\}^L. \end{aligned} \quad (B3)$$

Integrate over z'', we have

$$f_{L+1} = [1/(L+1)!]\left\{ \int_{z'=z_0}^{z'=z} dz' [U(\mathbf{b}_0, z') + U'(\mathbf{b}_0, z_0, \mathbf{b}_0, z')] \right\}^{L+1}. \quad (B4)$$

Thus, (19) is proved by mathematical induction.

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