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The 'Frozen-Lattice' Approach for Incoherent Phonon Excitation in Electron Scattering. How Accurate Is It?

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Abstract

The 'frozen-lattice' model is a semi-classical approach for calculating electron diffuse scattering in crystals that has arisen from thermal vibration of crystal atoms. This quasi-elastic scattering approach is, however, unproven since its equivalence with the incoherent phonon-excitation model is not yet established. As quantitative electron microscopy is becoming a realistic method, it is necessary to examine the accuracy of the model. In this paper, based on a rigorous quantum-mechanical phonon excitation theory, it is proved that an identical result would be obtained using the frozen-lattice model and the formal phonon-excitation model if (i) the incoherence between *different orders* of thermal diffuse scattering is considered in the frozen-lattice-model calculation and (ii) the specimen thickness and the mean-free-path length for phonon excitation are both smaller than the distance travelled by the electron within the lifetime of the phonon ($\tau_0 v$, which is 5 μm for 100 kV electrons). Condition (ii) is usually absolutely satisfied and condition (i) can be precisely accounted for in the calculation with the introduction of the mixed dynamic form factor $S(\mathbf{Q}, \mathbf{Q}')$. The conclusion holds for each and all orders of diffuse scattering, thus, the quantum-mechanical basis of the frozen-lattice model is established, confirming the validity, reliability and accuracy of using this model in quantitative dynamical electron diffraction and imaging calculations. It has also been shown that the frozen-lattice model is suitable for low-energy electrons.

1. Introduction

In high-energy electron scattering, the frozen-lattice model is assumed in describing diffuse scattering arising from the thermal vibration of crystal atoms (Hall & Hirsch, 1965), which means that, although atom vibration is a time-dependent process, the crystal lattice appears as if in a stationary instantaneous configuration for an incident electron since the interaction time of the electron with the crystal is much shorter than the vibration period of the crystal atom, but the crystal lattice can be in another configuration for the next incoming electron. Thus, for each lattice configuration, the scattering of the electron can be considered as a time-independent quasi-elastic scattering process and the finally observed diffraction pattern contributed to by millions of electrons is equivalent to a time average on the intensities calculated for the different lattice configurations.

This model is the basis of many theoretical approaches for treating phonon scattering (or thermal diffuse scattering, TDS) in electron diffraction/imaging (Rossouw & Bursill, 1985; Fanidis *et al.*, 1992, 1993; Dinges *et al.*, 1995; Wang & Cowley, 1990; Wang, 1991; Amali & Rez, 1997; Anstis *et al.*, 1998; for a review, see Wang, 1995*a*, chs. 6–15), and in some cases quantitative matching with experimental observations have been obtained (Loane *et al.*, 1991; Xu *et al.*, 1991; Muller *et al.*, 1997). There are, however, two major concerns about this model. First, this is a quasi-elastic scattering model in which the electrons diffusely scattered by one lattice configuration are considered coherent although the scattering from different lattice configurations are treated incoherently. This deviates from the fundamental result of quantum mechanics that phonon scattering is an incoherent process (Van Dyck, 1997). Secondly, this model treats a time-dependent atom vibration process as an integration of many mini time-independent processes, thus, a semi-classical concept is introduced in this quantum excitation process. Hence, the accuracy and adequacy of this model might need to be examined because quantitative analysis of electron diffraction and imaging data is desired in current research (Spence & Zuo, 1992).

To answer these questions, the objective of this paper is to rigorously prove the equivalence of the frozen-lattice-model approach with the quantum-mechanical phonon excitation theory and to investigate conditions under which the equivalence holds, aiming at establishing the theoretical basis of the model in quantitative electron microscopy. For convenience of the proof, we first outline the result of the TDS theory in the frozen-lattice model (§2). Following a rigorous approach using Yoshioka (1957) equations, a quantum-mechanical theory is given in §3. A comparison of the two theories gives the conditions under which the two theories are

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equivalent. Finally, these conditions are examined in detail to establish the completeness of the proof.

2. The frozen-lattice model for thermal diffuse scattering in electron scattering

To properly treat TDS using the frozen-lattice model, the instantaneous crystal potential $V(\mathbf{r})$ is written as a sum of a time-averaged potential V_0 and a fluctuation term that characterizes the distortion of a crystal lattice owing to the displacement of an atom from its equilibrium position:

$$V(\mathbf{r}) = V_0(\mathbf{r}) + \Delta V(\mathbf{r}), \tag{1a}$$

where the instantaneous crystal potential is

$$V(\mathbf{r}) = \sum_\kappa V_\kappa(\mathbf{r} - \mathbf{r}_\kappa - \mathbf{u}_\kappa); \tag{1b}$$

$\mathbf{r}_\kappa$ is the equilibrium position of the κth atom in the crystal and $\mathbf{u}_\kappa$ is its displacement; the time average of the crystal potential is

$$\begin{aligned} V_0(\mathbf{r}) &= \Big\langle \sum_\kappa V_\kappa(\mathbf{r} - \mathbf{r}_\kappa - \mathbf{u}_\kappa) \Big\rangle \\ &= \sum_\kappa \int \mathrm{d}\boldsymbol{\tau} f^e_\kappa(\boldsymbol{\tau}) \exp[-W_\kappa(\boldsymbol{\tau})] \\ &\quad \times \exp[2\pi i \boldsymbol{\tau} \cdot (\mathbf{r} - \mathbf{r}_\kappa)], \end{aligned} \tag{1c}$$

$W_\kappa = 2\pi^2 \langle |\boldsymbol{\tau} \cdot \mathbf{u}_\kappa|^2 \rangle$ is the Debye–Waller factor, where $\langle\ \rangle$ stands for time average, $f^e_\kappa(\boldsymbol{\tau})$ is the electron scattering factor and $\boldsymbol{\tau}$ is a reciprocal-space vector; and the deviation potential depending on the lattice configuration is

$$\Delta V(\mathbf{r}) = V(\mathbf{r}) - V_0(\mathbf{r}). \tag{1d}$$

The electron wave function scattered by a given crystal lattice is determined by the time-independent Schrödinger equation

$$[(\hbar^2/2m_0)\nabla^2 - eV_0 - e\Delta V - E]\Psi = 0, \tag{2}$$

where E is the electron energy. For electron scattering, (2) is converted into an integral equation with the use of Green's function $G(\mathbf{r}, \mathbf{r}_1)$:

$$\Psi(\mathbf{r}) = \Psi_0(\mathbf{r}) + e \int \mathrm{d}\mathbf{r}_1 G(\mathbf{r}, \mathbf{r}_1) \Delta V(\mathbf{r}_1) \Psi(\mathbf{r}_1), \tag{3a}$$

where G is the solution of

$$[-(\hbar^2/2m_0)\nabla^2 - eV_0 - E] G(\mathbf{r}, \mathbf{r}_1) = \delta(\mathbf{r} - \mathbf{r}_1), \tag{3b}$$

which corresponds to the electron wave coming from a point source at $\mathbf{r}'$ after being scattered by the crystal, and it can be solved for a general case (Wang, 1998*a*); and $\Psi_0(\mathbf{r})$ represents the elastic wave initiated by an incident plane wave of wave vector $\mathbf{K}_0$ after being scattered by the time-independent potential V_0, and it satisfies

$$[-(\hbar^2/2m_0)\nabla^2 - eV_0 - E]\Psi_0 = 0. \tag{3c}$$

For the purpose of this paper, we define a density matrix

$$\rho_f(\mathbf{r}, \mathbf{r}') = \langle \Psi(\mathbf{r}) \Psi^*(\mathbf{r}') \rangle, \tag{4}$$

where $\langle\ \rangle$ represents an average over lattice configurations. The electron diffraction pattern including TDS can be calculated by taking a double Fourier transform of the density matrix

$$I(\boldsymbol{\tau}) = \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' \exp[-2\pi i \boldsymbol{\tau} \cdot (\mathbf{r} - \mathbf{r}')] \rho_f(\mathbf{r}, \mathbf{r}'). \tag{5}$$

The intensity distribution in the electron image can be calculated by

$$I(\mathbf{r}) = \int \mathrm{d}\mathbf{r} \int \mathrm{d}\mathbf{r}' \rho_f(\mathbf{r}_1, \mathbf{r}_2) t(\mathbf{r} - \mathbf{r}_1) t^*(\mathbf{r} - \mathbf{r}_2), \tag{6}$$

where $t(\mathbf{r})$ characterizes the information transfer property of the optical system in an electron microscope. Therefore, the entire scattering process can be described by the density-matrix equation and the key here is to find its solution. We now derive the equation that governs the evolution of the density matrix.

Starting from (3*a*) and using $\langle \Delta V(\mathbf{r}) \rangle = 0$, we have

$$\begin{aligned} \rho_f(\mathbf{r}, \mathbf{r}') = \rho_0(\mathbf{r}, \mathbf{r}') + e^2 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2\, [G(\mathbf{r}, \mathbf{r}_1) G^*(\mathbf{r}', \mathbf{r}_2) \\ \times \langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \Psi(\mathbf{r}_1) \Psi^*(\mathbf{r}_2) \rangle], \end{aligned} \tag{7}$$

where the density matrix of the Bragg scattered electrons is $\rho_0(\mathbf{r}, \mathbf{r}') = \Psi_0(\mathbf{r}) \Psi_0^*(\mathbf{r}')$. Using the iterative solution of (3*a*) and treating the different orders of diffuse scattering incoherently, which means

$$\begin{aligned} &\langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \Delta V(\mathbf{r}_3) \Delta V(\mathbf{r}_4) \rangle \\ &\quad = \langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \rangle \langle \Delta V(\mathbf{r}_3) \Delta V(\mathbf{r}_4) \rangle, \\ &\langle \Delta V \Delta V \Delta V \Delta V \Delta V \Delta V \rangle \\ &\quad = \langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \rangle \langle \Delta V(\mathbf{r}_3) \Delta V(\mathbf{r}_4) \rangle \\ &\qquad \times \langle \Delta V(\mathbf{r}_5) \Delta V(\mathbf{r}_6) \rangle \end{aligned} \tag{8a}$$

and similar relations hold for higher-order terms, it has been proved (Wang, 1996*b*) that

$$\begin{aligned} &\langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \Psi(\mathbf{r}_1) \Psi^*(\mathbf{r}_2) \rangle \\ &\quad = \langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \rangle \langle \Psi(\mathbf{r}_1) \Psi^*(\mathbf{r}_2) \rangle. \end{aligned} \tag{8b}$$

Thus, (7) becomes

$$\begin{aligned} \rho_f(\mathbf{r}, \mathbf{r}') = \rho_0(\mathbf{r}, \mathbf{r}') + e^2 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2\, [G(\mathbf{r}, \mathbf{r}_1) G^*(\mathbf{r}', \mathbf{r}_2) \\ \times \langle \Delta V(\mathbf{r}_1) \Delta V(\mathbf{r}_2) \rangle \rho_f(\mathbf{r}_1, \mathbf{r}_2)]. \end{aligned} \tag{9}$$

This is the equation that determines the solution of the density matrix and it can be solved iteratively:

$$\begin{aligned}\rho_f(\mathbf{r},\mathbf{r}') = {} & \rho_0(\mathbf{r},\mathbf{r}') + e^2 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2\, [G(\mathbf{r},\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2) \\ & \times \langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle \rho_0(\mathbf{r}_1,\mathbf{r}_2)] \\ & + e^4 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 \int \mathrm{d}\mathbf{r}_3 \int \mathrm{d}\mathbf{r}_4 \\ & \times [G(\mathbf{r},\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2)G(\mathbf{r}_1,\mathbf{r}_3)G^*(\mathbf{r}_2,\mathbf{r}_4) \\ & \times \langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle \\ & \times \langle \Delta V(\mathbf{r}_3)\Delta V(\mathbf{r}_4)\rangle \rho_0(\mathbf{r}_3,\mathbf{r}_4)] + \ldots, \end{aligned} \tag{10}$$

where the higher orders of TDS are included. For the general purpose of the proof, all the higher-order terms are kept. On the other hand, the time (or configurational) average of $\Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)$ is directly related to the mixed dynamic form factor $S(\mathbf{Q},\mathbf{Q}')$ by (Wang, 1996*a*)

$$\begin{aligned}&\langle \Delta V(\mathbf{r},t)\Delta V(\mathbf{r}_1,t)\rangle \\ &\quad = \int \mathrm{d}\mathbf{Q} \int \mathrm{d}\mathbf{Q}' \exp[2\pi i(\mathbf{r}\cdot\mathbf{Q} - \mathbf{r}_1\cdot\mathbf{Q}')] S(\mathbf{Q},\mathbf{Q}'), \end{aligned} \tag{11a}$$

and for TDS with consideration of the phase correlation among atom vibrations, the mixed dynamic form factor is given by

$$\begin{aligned} S(\mathbf{Q},\mathbf{Q}') = {} & \sum_{\kappa}\sum_{\kappa'} \exp[2\pi i(\mathbf{r}_{\kappa'}\cdot\mathbf{Q}' - \mathbf{r}_{\kappa}\cdot\mathbf{Q})] \\ & \times f^e_{\kappa}(\mathbf{Q}) f^e_{\kappa'}(\mathbf{Q}') \exp[-W_{\kappa}(\mathbf{Q}) - W_{\kappa'}(\mathbf{Q}')] \\ & \times \{\exp[2F_{\kappa\kappa'}(\mathbf{Q},\mathbf{Q}')] - 1\}, \end{aligned} \tag{11b}$$

where

$$F_{\kappa\kappa'}(\mathbf{Q},\mathbf{Q}') = 2\pi^2 \langle (\mathbf{u}_{\kappa}\cdot\mathbf{Q})(\mathbf{u}_{\kappa'}\cdot\mathbf{Q}')\rangle \tag{11c}$$

is defined as a correlation function that is determined by the coupling between vibrations of the κ and κ' atoms. The calculation of $W_{\kappa}(\mathbf{Q})$ and $F_{\kappa\kappa'}(\mathbf{Q},\mathbf{Q}')$ using the harmonic oscillators model has been given elsewhere (Sears & Shelley, 1991; Wang, 1995*a*,*b*). Equation (9) is the main result of the frozen-lattice model and the introduction of the mixed dynamic form factor $S(\mathbf{Q},\mathbf{Q}')$ [see (3*a*)] automatically considers the incoherence between the different orders of diffuse scattering. We now derive a similar equation using the rigorous quantum-mechanical approach.

3. Quantum-mechanical description of phonon excitation in electron scattering

Phonon excitation is an inelastic scattering process that is most adequately described using Yoshioka's (1957) approach, in which the eigenstate of the incident electron–crystal system is expressed as a product of the eigenstate of the crystal with that of the incident electron,

$$\Phi_s(\mathbf{r},\mathbf{R}) = \sum_{n=0}^{\infty} a_n(\mathbf{R})\Psi_n(\mathbf{r}), \tag{12}$$

where a_n is the nth eigenstate of the crystal of energy ε_n; Ψ_0 is the elastic scattered wave of energy E and Ψ_n is the inelastically scattered wave of energy $E_n = E - \varepsilon_n$ after exciting the nth crystal state; $\mathbf{R}$ represents the coordinates of crystal electrons $(\mathbf{r}_1 \ldots, \mathbf{r}_M)$. The fundamental equations that govern the generation, scattering and transition between/among the elastic wave and the inelastic waves are:

$$[-(\hbar^2/2m_0)\nabla^2 + H_{00} - E]\Psi_0 = -\sum_{m=1} H_{0m}\Psi_m \tag{13a}$$

and

$$[-(\hbar^2/2m_0)\nabla^2 + H_{nn} - E_n]\Psi_n = -\sum_{m\neq n} H_{nm}\Psi_m, \tag{13b}$$

where the transition matrix from state a_m to a_n is

$$H_{nm} = \int a_n^* H a_m \,\mathrm{d}\mathbf{R} = \langle a_n|H|a_m\rangle. \tag{13c}$$

H is the interaction Hamiltonian between the incident electron with the electrons and nuclei in the crystal.

Phonon excitation is a quantum transition process, in which the creation and annihilation of phonons are both possible. The excitation process can be adequately described using the transition matrix, similar to other inelastic electron transitions. Phonon excitation has the following characteristics. Firstly, the phonon energy is on the order of 0.1 eV or less, much smaller than the incident energy of the electron (typically 100 keV), thus, the electron energy can be considered to be unaffected by the energy transfer due to phonon excitation, which means $E_n \simeq E$ but this energy loss has destroyed the coherence of the electron. Hence, *the incoherence among all of the crystal states (e.g. different phonon states) must be preserved.* Secondly, a phonon is characterized by a group of quantities of $(\mathbf{q}, \omega_j, \mathbf{e})$, where $\mathbf{q}$ is the phonon wave vector, ω_j the phonon frequency belonging to the jth branch and $\mathbf{e}$ the polarization vector, which means that an excited state a_n is also characterized by $(\mathbf{q}, \omega_j, \mathbf{e})$, which is usually denoted as $|n_s\rangle$ in phonon excitation. Finally, the interaction Hamiltonian of the electron with the crystal is determined by the instataneous crystal potential V as given by (1*b*), thus, the diagonal matrix element H_{nn} is

$$H_{nn} \simeq H_{00}. \tag{14a}$$

The approximation of $H_{nn} \simeq H_{00}$ is considered to be almost exact since the excitation of phonons does not affect the time-averaged potential of the crystal. For the statistical distribution of phonons in the ground state at a finite temperature T, a weight factor must be added, which is related to the energy of the harmonic oscillator by

$$p(n_s) = \frac{\exp[-(n_s + 1/2)\hbar\omega_j/k_BT]}{\sum_{n_s=0}\exp[-(n_s + 1/2)\hbar\omega_j/k_BT]}$$
$$= [1 - \exp(-\hbar\omega_j/k_BT)]\exp(-n_s\hbar\omega_j/k_BT). \quad (14b)$$

A statistical average needs to be added accordingly, thus

$$H_{00} = -e\sum_{n_s}\langle n_s|V(\mathbf{r})|n_s\rangle p(n_s), \quad (14c)$$

where the sum of n_s is over all of the phonon states and the number of phonons. Using the relationship between the electron scattering factor and the atomic potential, one has

$$\begin{aligned} H_{00} &= -e\sum_{n_s}\Big\langle n_s\Big|\sum_{\kappa}V_{\kappa}(\mathbf{r} - \mathbf{r}_{\kappa} - \mathbf{u}_{\kappa})\Big|n_s\Big\rangle p(n_s) \\ &= e\sum_{\kappa}\int \mathrm{d}\boldsymbol{\tau}\, f^e_{\kappa}(\boldsymbol{\tau})\exp[2\pi i\boldsymbol{\tau}\cdot(\mathbf{r} - \mathbf{r}_{\kappa})] \\ &\quad \times \sum_{n_s}\langle n_s|\exp(-2\pi i\boldsymbol{\tau}\cdot\mathbf{u}_{\kappa})|n_s\rangle p(n_s) \\ &= -e\sum_{\kappa}\int \mathrm{d}\boldsymbol{\tau}\, f^e_{\kappa}(\boldsymbol{\tau})\exp[2\pi i\boldsymbol{\tau}\cdot(\mathbf{r} - \mathbf{r}_{\kappa})] \\ &\quad \times \exp[-W_{\kappa}(\boldsymbol{\tau})], \end{aligned} \quad (14d)$$

where a general relationship used in the above derivation is $\langle\exp(x)\rangle = \exp[\langle x^2\rangle/2]$ if $\langle x\rangle = 0$, which has been proved to be exact (Glauber, 1955), and the Debye–Waller factor is given by

$$W_{\kappa} = 2\pi^2\sum_{n_s}\langle n_s|(\boldsymbol{\tau}\cdot\mathbf{u}_{\kappa})^2|n_s\rangle p(n_s). \quad (14e)$$

In comparison to the average crystal potential V_0 given in (1*c*), one has

$$H_{nn} = -eV_0, \quad (14f)$$

in which the Debye–Waller factor is included. Therefore, (13*a*) and (13*b*) are transformed into

$$[-(\hbar^2/2m_0)\nabla^2 - eV_0 - E]\Psi_0 = -\sum_{m=1}H_{0m}\Psi_m \quad (15a)$$

and

$$[-(\hbar^2/2m_0)\nabla^2 - eV_0 - E]\Psi_n = -\sum_{m\neq n}H_{nm}\Psi_m. \quad (15b)$$

The density matrix in this case is defined to be

$$\rho_p(\mathbf{r}, \mathbf{r}') = \sum_{n=0}\Psi_n(\mathbf{r})\Psi^*_n(\mathbf{r}'), \quad (16)$$

where the sum over n is over all the crystal states including the elastic scattering state (*i.e.* $n = 0$, ground state). We now derive the equation that governs the motion of the density matrix. By using Green's function $G(\mathbf{r}, \mathbf{r}')$ introduced in (3*b*), (15*a*) and (15*b*) are transformed into a unified integral equation

$$\Psi_n(\mathbf{r}) = \Psi_0(\mathbf{r})\delta_{n0} - \sum_{m\neq n}\int \mathrm{d}\mathbf{r}_1 G(\mathbf{r}, \mathbf{r}_1)H_{nm}(\mathbf{r}_1)\Psi(\mathbf{r}_1). \quad (17)$$

From the physics point of view, electrons scattered by different crystal states have no phase relationship. This is the random phase approximation and it is introduced from the practical consideration of electron diffraction, which means the interference terms of the elastic wave $\Psi_0(\mathbf{r})$ with the inelastic wave $\Psi_m(\mathbf{r})$ vanish. Using this approximation, a subtitution of (17) into (16) gives

$$\begin{aligned} \rho_p(\mathbf{r},\mathbf{r}') &= \rho_0(\mathbf{r}, \mathbf{r}') + \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 G(\mathbf{r}, \mathbf{r}_1)G^*(\mathbf{r}', \mathbf{r}_2) \\ &\quad \times \sum_{n\neq 0}\sum_{m}\sum_{m'}[H_{nm}(\mathbf{r}_1)H_{m'n}(\mathbf{r}_2)\Psi_m(\mathbf{r}_1)\Psi^*_{m'}(\mathbf{r}_2)]. \end{aligned} \quad (18)$$

The summation in (18) contains the interference terms of different crystal states. Using the random phase approximation again, the interference between the m and m' states vanishes from the sums unless $m = m'$. Therefore, (18) is approximated by

$$\begin{aligned} \rho_p(\mathbf{r}, \mathbf{r}') &= \rho_0(\mathbf{r}, \mathbf{r}') + \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 G(\mathbf{r}, \mathbf{r}_1)G^*(\mathbf{r}', \mathbf{r}_2) \\ &\quad \times \left\{\sum_{m}\left[\sum_{n\neq 0}H_{nm}(\mathbf{r}_1)H_{mn}\right]\Psi_m(\mathbf{r}_1)\Psi^*_m(\mathbf{r}_2)\right\}. \end{aligned} \quad (19)$$

The following approximation is made in order to simplify this equation. In one-particle multiple-scattering theory, each time the electron inelastically interacts with the crystal atoms the collision is assumed to take place as if the crystal is in its ground state and the effect of previous collisions is very small. This condition is satisfied if $\tau_0 v > \Lambda$, where τ_0 is the lifetime of the crystal excitation state, v is the electron velocity and Λ is the mean-free-path length of phonon excitation. This approximation is mathematically expressed as

$$\sum_{n\neq 0}[H_{nm}(\mathbf{r}_1)H^*_{nm}(\mathbf{r}_2)] \simeq \sum_{n\neq 0}[H_{n0}(\mathbf{r}_1)H^*_{n0}(\mathbf{r}_2)]. \quad (20)$$

It is simply called the 'ground-state' approximation and its meaning will be discussed later. Thus, equation (19) is approximated as (Wang, 1995*a*, ch. 14)

$$\begin{aligned} \rho_p(\mathbf{r}, \mathbf{r}') &\simeq \rho_0(\mathbf{r}, \mathbf{r}') + \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 G(\mathbf{r}, \mathbf{r}_1)G^*(\mathbf{r}', \mathbf{r}_2) \\ &\quad \times \left[\sum_{n\neq 0}H_{n0}(\mathbf{r}_1)H_{0n}(\mathbf{r}_2)\right]\left[\sum_{m}\Psi_m(\mathbf{r}_1)\Psi^*_m(\mathbf{r}_2)\right] \\ &= \rho_0(\mathbf{r}, \mathbf{r}') + \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 G(\mathbf{r}, \mathbf{r}_1)G^*(\mathbf{r}', \mathbf{r}_2) \\ &\quad \times \left[\sum_{n\neq 0}H_{n0}(\mathbf{r}_1)H_{0n}(\mathbf{r}_2)\right]\rho_p(\mathbf{r}_1, \mathbf{r}_2), \end{aligned} \quad (21a)$$

which can be solved iteratively:

$$\begin{aligned}\rho_p(\mathbf{r},\mathbf{r}') = {} & \rho_0(\mathbf{r},\mathbf{r}') + \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 \Big\{ G(\mathbf{r}',\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2) \\ & \times \Big[\sum_{n\neq 0} H_{n0}(\mathbf{r}_1)H_{0n}(\mathbf{r}_2)\Big]\rho_0(\mathbf{r}_1,\mathbf{r}_2)\Big\} \\ & + \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 \int \mathrm{d}\mathbf{r}_3 \int \mathrm{d}\mathbf{r}_4 \\ & \times \Big\{ G(\mathbf{r},\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2)G(\mathbf{r}_1,\mathbf{r}_3)G^*(\mathbf{r}_2,\mathbf{r}_4) \\ & \times \Big[\sum_{n\neq 0} H_{n0}(\mathbf{r}_1)H_{0n}(\mathbf{r}_2)\Big] \\ & \times \Big[\sum_{n'\neq 0} H_{n'0}(\mathbf{r}_3)H_{0n'}(\mathbf{r}_4)\Big] \\ & \times \rho_0(\mathbf{r}_3,\mathbf{r}_4)\Big\} + \ldots . \qquad (21b)\end{aligned}$$

As illustrated in Appendix *A*, with a proper understanding of the ground state a_0 and an introduction of statistical average, it can be proved that

$$\sum_{n\neq 0} [H_{n0}(\mathbf{r}_1)H^*_{n0}(\mathbf{r}_2) = e^2\langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle, \qquad (22)$$

which means the sum over crossed transition-matrix elements is the time-averaged cross perturbation crystal potentials. This is a key equation that correlates the quantum model with the semi-classical model. Therefore, (21*a*) becomes

$$\begin{aligned}\rho_p(\mathbf{r},\mathbf{r}') \simeq {} & \rho_0(\mathbf{r},\mathbf{r}') + e^2 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2\, G(\mathbf{r},\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2) \\ & \times \langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle \rho_p(\mathbf{r}_1,\mathbf{r}_2), \qquad (23a)\end{aligned}$$

which can be solved iteratively:

$$\begin{aligned}\rho_p(\mathbf{r},\mathbf{r}') \simeq {} & \rho_0(\mathbf{r},\mathbf{r}') + e^2 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2\, [G(\mathbf{r},\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2) \\ & \times \langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle \rho_0(\mathbf{r}_1,\mathbf{r}_2)] \\ & + e^4 \int \mathrm{d}\mathbf{r}_1 \int \mathrm{d}\mathbf{r}_2 \int \mathrm{d}\mathbf{r}_3 \int \mathrm{d}\mathbf{r}_4 \\ & \times [G(\mathbf{r},\mathbf{r}_1)G^*(\mathbf{r}',\mathbf{r}_2)G(\mathbf{r}_1,\mathbf{r}_3)G^*(\mathbf{r}_2,\mathbf{r}_4) \\ & \times \langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle \\ & \times \langle \Delta V(\mathbf{r}_3)\Delta V(\mathbf{r}_4)\rangle \rho_0(\mathbf{r}_3,\mathbf{r}_4)] + \ldots . \qquad (23b)\end{aligned}$$

This is the identical form to (10), therefore, the density matrix calculated based on the frozen-lattice model is equal to the density matrix calculated following the phonon-excitation model in quantum mechanics:

$$\rho_p(\mathbf{r},\mathbf{r}') = \rho_f(\mathbf{r},\mathbf{r}'). \qquad (24)$$

This identity holds for all of the orders and each order of the diffuse scattering. Therefore, the result calculated using the frozen-lattice model is identical to the result calculated using the phonon-excitation model if the approximations made in §2 and §3 hold. We now evaluate these approximations.

4. The approximations and conditions

4.1. *The incoherence between different orders of diffuse scattering*

The major approximation introduced in the calculation of the density matrix in the frozen-lattice model is

$$\begin{aligned}&\langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\Delta V(\mathbf{r}_3)\Delta V(\mathbf{r}_4)\rangle \\ &\quad = \langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle\langle \Delta V(\mathbf{r}_3)\Delta V(\mathbf{r}_4)\rangle.\end{aligned}$$

In electron diffraction, the first-order scattering intensity is determined by $\langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle$ and the second order is by $\langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle\langle \Delta V(\mathbf{r}_3)\Delta V(\mathbf{r}_4)\rangle$, which can be interpreted as the second order being a double scattering of two incoherent first-order events. The approximation simply means the scattering produced by the different orders of diffuse scattering (such as ΔV^2, ΔV^4 and ΔV^6 *etc.* or multiple diffuse scattering) must be treated as incoherent, and the higher-order scattering is the result of multiple incoherent first-order-scattering events. The second-order scattering, for example, is an incoherent double first-order scattering (*e.g.* $\langle \Delta V^4\rangle = \langle \Delta V^2\rangle\langle \Delta V^2\rangle$) and $\langle \Delta V^6\rangle = [\langle \Delta V^2\rangle \times \langle \Delta V^2\rangle\langle \Delta V^2\rangle]$ for the third-order scattering *etc.* (Wang, 1996*b*). This is in fact the practical situation and the approximation holds exactly. This approximation also reduces the coherence effect in the quasi-elastic scattering process. The consequence of this approach means that any theory that has been developed with use of the mixed dynamic form factor $S(\mathbf{Q},\mathbf{Q}')$ instead of using the real space $\Delta V(\mathbf{r}_1)$ has correctly considered this incoherence. Thus, the corresponding calculation is equivalent to the result of quantum phonon-excitation theory.

Calculations using the frozen-lattice model have frequently been performed in the multislice theory (Cowley & Moodie, 1957) following two different approaches. One approach (Wang, 1991; Loane *et al.*, 1991) assumes that the entire crystal in one instantaneous configuration is a frozen lattice and the diffuse scattering generated in the entire volume is coherent but the final intensity is an incoherent sum over the intensities contributed by different lattice configurations. The other approach assumes that the diffuse scattering generated within one thin crystal slice is coherent but incoherence is preserved for the scattering generated in different slices (Dinges *et al.*, 1995; Wang, 1995*b*). The former has properly considered the multiple-diffuse-scattering effect but ignored the incoherence between the different orders of diffuse scattering. This is likely to be an excellent approximation for thin specimens. The latter treats the incoherence properly but the multiple diffuse scattering among slices is neglected. The current study shows that the most significant error in the frozen-lattice-model calculation is the 'quasi-elastic' approximation, which can be compensated for with the introduction of the mixed dynamic form factor.

4.2. *The 'ground-state' approximation*

The approximation made in the phonon excitation model is the so-called 'ground-state' approximation. The approximation means that each time the electron inelastically interacts with the crystal atoms the collision is assumed to take place as if the crystal is in its ground state and the effect of any previous collision is very small. For a thin crystal of thickness $d = 100$ nm, the duration that the electron interacts with the crystal atoms for a 100 keV electron is on the order of $\Delta t \simeq 6 \times 10^{-15}$ s. Theorectical calculation has found that the lifetime of a phonon is on the order of $\tau_0 = 10^{-13}$ s (Björkman *et al.*, 1967; Woll & Kohn, 1962). The limited lifetime of phonons is due to anharmonic effects in atom vibrations. The lifetime of a phonon is much longer than the interaction time Δt of the electron with the crystal, thus, there is no phonon decay during the scattering of an incident electron. On the other hand, for a modern field-emission source, the electron flux is on the order of 10^{12} e s^{-1}, thus, the average time interval between two successive incident electrons is 10^{-12} s, much longer than the lifetime of the phonon. The phonons generated by the previous incident electron have annihilated before the next incoming electron interacts with the crystal. Therefore, the crystal is in its ground state for each incident electron.

The conditions for this approximation to be held are: the lifetime of the photon is larger than the time required to create a new phonon, *e.g.* $\tau_0 > \Lambda/v$ (or $\tau_0 v > \Lambda$); and the electron–specimen interaction time is shorter than the lifetime of the phonon, *e.g.* $d < \tau_0 v$. The first condition depends on the energy of the incident electron and the second condition is satisfied in almost all of our experiments. The conditions $\tau_0 v > \Lambda$ and $d < \tau_0 v$ mean that the specimen thickness and the mean-free-path length for phonon excitation are both required to be smaller than the distance travelled by the electron within the lifetime of the phonon. For a 100 kV electron, $\tau_0 v \simeq 5$ μm. Therefore, both conditions are absolutely satisfied in all of our current experiments.

On the other hand, the average number of phonons in a crystal is rather large at ambient temperature, thus, adding or subtracting a few phonons due to the electron–phonon interaction in the whole system have almost no effect on the potential distribution in the crystal. This is always assumed in electron scattering. Therefore, the ground-state approximation is excellent.

Finally, the random phase approximation introduced in (18) has been considered an excellent approach although it is an argument purely based on the physical picture (probably without mathematical proof). We believe this approximation also holds for phonon scattering.

4.3. *Frozen-lattice model for low-energy electrons*

The quantum phonon-excitation theory presented in §3 holds if the specimen thickness and the mean-free-path length for phonon excitation are both smaller than the distance travelled by the electron within the lifetime period of the phonon. We now examine these conditions for low-energy electrons. Taking an electron with 100 eV energy as an example, its moving speed v is $0.0198\,c$, where c is the speed of light, and the lifetime of a phonon is 10^{-13} s, independent of electron energy, the electron mean-free-path length and the specimen thickness are both required to be smaller than $\tau_0 v = 600$ nm, which is satisfied in almost all of the low-energy experiments. Thus, the density-matrix equation given in (23*a*) holds even for low-energy electrons. On the other hand, since (23*a*) is identical to (9), which means that the calculation using the frozen-lattice model is also valid for low-energy electrons. This is a rather surprising result because the frozen-lattice model was proposed for high-energy electrons with a simple assumption that the electron–specimen interaction time is much shorter than the vibration period of the crystal atoms. To understand this result, the penetration depth of a 100 V electron into the specimen is assumed to be less than 10 nm, the electron–specimen interaction time is $\sim 1.7 \times 10^{-15}$ s, much smaller than the vibration period of an atom (typically of 10^{-13} s). Therefore, the theoretical proof given in §2 and §3 has expanded the energy range in which the frozen-lattice model is applicable.

4.4. *The first-order diffuse scattering result*

It is worthwhile pointing out that the above two approximations are not needed if the specimen is much smaller than the mean-free-path length of thermal diffuse scattering so that the single diffuse scattering approximation holds. From (10) and (23*b*), the first-order diffuse scattering terms are identical regardless of the lifetime of the phonon. Therefore, the first-order diffuse scattering based on the frozen-lattice model gives exactly the same result as the quantum phonon-excitation theory. This simply proves the validity of the existing theories for thin specimens.

Based on current understanding, the density-matrix theory is likely to be the most precise approach for calculating the diffraction patterns or images of phonon-scattered electrons, but the calculation has to be carried out iteratively in order to include high-order diffuse scattering.

Before reaching a conclusion, we wish to comment on the theory developed by the author for recovering high-order diffuse scattering in dynamical calculations (Wang, 1996*b*). *By inclusion of a complex potential in* *dynamical calculation of the elastic wave, it has been proved analytically that the higher-order diffuse scattering is fully recovered in the calculations using the equation derived under the distorted-wave Born approximation.* The proof was based on the frozen-lattice model with consideration of the incoherence between different orders of diffuse scattering. We can claim now that the current study proves the quantum-mechanical equivalence of the proposed theory.

5. Conclusions

Based on rigorous Green-function and density-matrix theories, it has been proved that the calculation based on the frozen-lattice model for thermal diffuse scattering in electron diffraction gives an identical result to that obtained from the phonon excitation model if the following two conditions are satisfied: (i) the incoherence between different orders of thermal diffuse scattering is considered in the frozen-lattice-model calculation; and (ii) the specimen thickness and the mean-free-path length for phonon excitation are both smaller than the distance travelled by the electron within the lifetime of the phonon. Condition (ii) is absolutely satisfied by both low- and high-energy electrons, and satisfying condition (i) is the most critical requirement in the frozen-lattice-model calculation. More specifically, the diffuse scattering produced by the different orders of diffuse scattering (such as ΔV^2, ΔV^4 and ΔV^6 *etc.* or multiple diffuse scattering) must be treated as incoherent in the frozen-lattice model, and the higher-order scattering is the result of multiple incoherent first-order-scattering events. This incoherence can be precisely accounted for with the introduction of the mixed dynamic form factor $S(\mathbf{Q}, \mathbf{Q}')$, a key quantity for inelastic electron scattering. A new multislice approach has been proposed recently in which the multiple diffuse scattering and the incoherence effects are both accounted for using the $S(\mathbf{Q}, \mathbf{Q}')$ function (Wang, 1998*b*).

The conclusion, reached directly using the integral form of the Schrödinger equation without making the high-energy approximation or ignoring the back scattering, holds for each and all of the orders of diffuse scattering if they are treated *incoherently*, thus, the quantum-mechanical basis of the frozen-lattice model is established. *The frozen-lattice model can also be used to calculate the diffuse diffraction of low-energy electrons* because their effective penetration depth is relatively small. This conclusion confirms the validity, reliability and accuracy of using the frozen-lattice model in numerous dynamical theories of phonon excitation in electron diffraction and imaging of thin specimens.

APPENDIX A

In this section, our aim is to prove that

$$\sum_{n\neq 0} [H_{n0}(\mathbf{r}_1)H^*_{n0}(\mathbf{r}_2)] = e^2\langle \Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle.$$

Starting from the left-hand side and using a completeness equation of

$$\sum_n |a_n(\mathbf{R}_2)\rangle\langle a_n(\mathbf{R}_1)| = \delta(\mathbf{R}_2 - \mathbf{R}_1),$$

one has

$$\begin{aligned}
&\sum_{n\neq 0} H_{n0}(\mathbf{r}_1)H^*_{n0}(\mathbf{r}_2)\\
&\quad = \sum_{n\neq 0}\langle a_0(\mathbf{R}_2)|H(\mathbf{r}_2, \mathbf{R}_2)|a_n(\mathbf{R}_2)\rangle\\
&\qquad \times \langle a_n(\mathbf{R}_1)|H(\mathbf{r}_1, \mathbf{R}_1)|a_0(\mathbf{R}_1)\rangle\\
&\quad = \langle a_0(\mathbf{R}_2)|H(\mathbf{r}_2, \mathbf{R}_2)\Big[\sum_n |a_n(\mathbf{R}_2)\rangle\langle a_n(\mathbf{R}_1)|\\
&\qquad - |a_0(\mathbf{R}_2)\rangle\langle a_0(\mathbf{R}_1)|\Big]H(\mathbf{r}_1, \mathbf{R}_1)|a_0(\mathbf{R}_1)\rangle\\
&\quad = \langle a_0(\mathbf{R}_2)|H(\mathbf{r}_2, \mathbf{R}_2)[\delta(\mathbf{R}_2 - \mathbf{R}_1)\\
&\qquad - |a_0(\mathbf{R}_2)\rangle\langle a_0(\mathbf{R}_1)|]\, H(\mathbf{r}_1, \mathbf{R}_1)|a_0(\mathbf{R}_1)\rangle\\
&\quad = \langle a_0(\mathbf{R}_1)|H(\mathbf{r}_2, \mathbf{R}_2)H(\mathbf{r}_1, \mathbf{R}_1)|a_0(\mathbf{R}_1)\rangle\\
&\qquad - \langle a_0(\mathbf{R}_2)|H(\mathbf{r}_2, \mathbf{R}_2)|a_0(\mathbf{R}_2)\rangle\\
&\qquad \times \langle a_0(\mathbf{R}_1)|H(\mathbf{r}_1, \mathbf{R}_1)|a_0(\mathbf{R}_1)\rangle. \qquad (25)
\end{aligned}$$

Using the Fourier transform of $H = -eV$, which is

$$\begin{aligned}
H(\mathbf{r}) &= -eV(\mathbf{r})\\
&= -e\sum_\kappa \int \mathrm{d}\mathbf{Q}\exp[2\pi i(\mathbf{r} - \mathbf{r}_\kappa)\cdot \mathbf{Q}]f^e_\kappa(\mathbf{Q}), \qquad (26)
\end{aligned}$$

equation (25) can be converted into the form

$$\begin{aligned}
&\sum_{n\neq 0} [H_{n0}(\mathbf{r}_1)H^*_{n0}(\mathbf{r}_2)]\\
&\qquad = e^2 \int \mathrm{d}\mathbf{Q} \int \mathrm{d}\mathbf{Q}' \exp[2\pi i(\mathbf{r}_1\cdot \mathbf{Q} - \mathbf{r}_2 \cdot \mathbf{Q}')]\\
&\qquad\quad \times S'(\mathbf{Q}, \mathbf{Q}'), \qquad (27)
\end{aligned}$$

where the S' function is given by

$$\begin{aligned}
&S'(\mathbf{Q}, \mathbf{Q}')\\
&\quad = \sum_\kappa\sum_{\kappa'} \exp[2\pi i(\mathbf{r}_{\kappa'}\cdot \mathbf{Q}' - \mathbf{r}_\kappa \cdot \mathbf{Q})]f^e_\kappa(\mathbf{Q})f^e_{\kappa'}(\mathbf{Q}')\\
&\qquad \times \big\{\langle a_0|\exp[2\pi i(\mathbf{Q}'\cdot \mathbf{u}_{\kappa'} - \mathbf{Q}\cdot \mathbf{u}_\kappa)]|a_0\rangle\\
&\qquad - \langle a_0|\exp(2\pi i\mathbf{Q}'\cdot \mathbf{u}_{\kappa'})|a_0\rangle\\
&\qquad \times \langle a_0|\exp(-2\pi i\mathbf{Q}\cdot \mathbf{u}_\kappa)|a_0\rangle\big\}\\
&\quad = \sum_\kappa\sum_{\kappa'} \exp[2\pi i(\mathbf{r}_{\kappa'}\cdot \mathbf{Q}' - \mathbf{r}_\kappa \cdot \mathbf{Q})]f^e_\kappa(\mathbf{Q})f^e_{\kappa'}(\mathbf{Q}')\\
&\qquad \times \big\{\exp[-2\pi^2\langle a_0|\mathbf{Q}'\cdot \mathbf{u}_{\kappa'} - \mathbf{Q}\cdot \mathbf{u}_\kappa)^2|a_0\rangle]\\
&\qquad - \exp[-2\pi^2\langle a_0|(\mathbf{Q}\cdot \mathbf{u}_{\kappa'})^2|a_0\rangle]\\
&\qquad \times \exp[-2\pi^2\langle a_0|(\mathbf{Q}\cdot \mathbf{u}_\kappa)^2|a_0\rangle]\big\}. \qquad (28)
\end{aligned}$$

From the notation, it may be misunderstood that there is no phonon in the ground state $|a_0\rangle$. This is true at absolute zero ($T = 0$ K). In practice, any system is not isolated from the environment, thus, energy exchange is involved in electron diffraction (Fanidis *et al.*, 1992, 1993). To incorporate the statistical distribution of phonons in the ground state at a finite temperature, the terms in (28) need to be modified to represent the expectation value, hence

$$\langle a_0|(\mathbf{Q}'\cdot\mathbf{u}_{\kappa'})^2|a_0\rangle = \sum_{n_s}\langle n_s|(\mathbf{Q}'\cdot\mathbf{u}_{\kappa'})^2|n_s\rangle p(n_s). \quad (29)$$

Therefore, (28) becomes

$$\begin{aligned} S'(\mathbf{Q},\mathbf{Q}') &\\ = \sum_{\kappa}\sum_{\kappa'} &\exp[2\pi i(\mathbf{r}_{\kappa'}\cdot\mathbf{Q}' - \mathbf{r}_{\kappa}\cdot\mathbf{Q})]f^e_{\kappa}(\mathbf{Q})f^e_{\kappa'}(\mathbf{Q}')\\ &\times \exp[-W_{\kappa}(\mathbf{Q}) - W_{\kappa'}(\mathbf{Q}')]\\ &\times \{\exp[2F_{\kappa\kappa'}(\mathbf{Q},\mathbf{Q}')] - 1\} \quad (30)\end{aligned}$$

where

$$F_{\kappa\kappa'}(\mathbf{Q},\mathbf{Q}') = 2\pi^2\sum_{n_s}\langle n_s|(\mathbf{u}_{\kappa}\cdot\mathbf{Q})(\mathbf{u}_{\kappa'}\cdot\mathbf{Q}')|n_s\rangle p(n_s). \quad (31)$$

Equation (31) gives the same result as defined in (11*c*) under the harmonic oscillators approximation. A comparison of (31) and (11*b*) gives

$$S(\mathbf{Q},\mathbf{Q}') = e^2 S'(\mathbf{Q},\mathbf{Q}'), \quad (32)$$

or

$$\sum_{n\neq 0}[H_{n0}(\mathbf{r}_1)H^*_{n0}(\mathbf{r}_2)] = e^2\langle\Delta V(\mathbf{r}_1)\Delta V(\mathbf{r}_2)\rangle. \quad (33)$$

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