# THE "OPTICAL POTENTIAL" AND MULTIPLE DIFFUSE SCATTERING IN DYNAMICAL ELECTRON DIFFRACTION AND IMAGING

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## Abstract

The theory for the absorption potential (or optical potential) in electron diffraction was established many years ago by Yoshioka. However, few studies have been devoted to examining the approximations originally introduced when the potential was derived. In this paper, the absorption potential first proposed by Yoshioka is revised for dynamic electron diffraction with consideration of the effects arising from thermal diffuse scattering and point defect scattering. A rigorous theoretical proof is given to show that the inclusion of this "potential" in the dynamical calculation automatically recovers the contributions made by the high order diffuse scatter-ing, although the calculation is done using the equation derived for single diffuse scattering. If Yoshioka's approximation is made, i.e., the Green's function is replaced by its form in free-space, then the inclusion of the optical potential in dynamical calculations still recovers the multiple diffuse scattering terms except the dynamic Bragg reflection after each diffuse scattering event. This conclusion establishes the basis for expanding the conventional diffraction theories developed under the first order diffuse scattering approximation to cases where the specimen thickness is large and the degree of disorder is high. It has been shown that the "optical potential" depends also on the structure of the crystal. The Fourier coefficients of this function are given in the Bloch wave representation for transmission electron diffraction.

**Key Words**: Multiple diffuse scattering, distorted wave Born approximation, Green's function optical potential, thermal diffuse scattering, point defect, short-range ordering.

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## Introduction

Refining crystal structures by quantitative electron diffraction (Spence and Zuo, 1992; Midgley *et al.*, 1995) is experiencing a rapid development owing to the advances in electron energy-filtering and digital data recording systems (see many articles in the book edited by Reimer, 1995). Quantitative analysis of structure information provided by transmission electron diffraction and imaging strongly relies on computer simulations. An important quantity in dynamical calculation is the "absorption" potential (or the optical potential, Dederichs, 1972), given based on model calculations. The absorption here actually means that the electron is not absorbed by the specimen rather it is scattered out of the elastic state (or Bragg peaks) due to energy-loss and momentum transfer, resulting in a decrease in the intensity of the elastic wave. This is the effect of inelastic scattering (or diffuse scattering) on the Bragg reflected waves (Yoshioka, 1957), which is equivalent to introducing an absorption potential in dynamical calculation (Heidenreich, 1962). This work has been the basis of almost all of the later theoretical calculations on electron diffraction and imaging. The theoretical modeling of the absorption potential has been a focus of research for many years and substantial progress has been made in including the contributions made by single electron excitation (Gjønnes, 1962; Howie and Stern, 1972; Whelan, 1965a; Radi, 1970; Humphreys and Whelan, 1969; Allen and Rossouw, 1990; Wang, 1990), thermal diffuse scattering (Whelan, 1965b; Radi, 1970; Humphreys and Hirsch, 1968; Rossouw *et al.*, 1990) and disorder point defect scattering (Howie and Stern, 1972; Dudarev *et al.*, 1992). The absorption potential has been published in forms of parametric fitting and FORTRAN programs for the access of public users (Coene and Van Dyck, 1990; Dudarev *et al.*, 1995; Bird and King, 1990).

Almost all of the model calculations for the absorption potential have been based on the approximation originally introduced by Yoshioka (1957), in which the Green's function is approximated by its form in free-space. Thus, the absorption potential is a non-local function that depends only on the nature of the inelastic scattering and the crystallographic structure but has no relation with the diffracting condition of the incident beam. All of the calculations later were performed

## List of symbols

| | |
|---|---|
| $V(\mathbf{r},t)$ | Instantaneous crystal potential |
| $V_0(\mathbf{r})$ | Time and spatially averaged crystal poten-tial |
| $\Delta V(\mathbf{r},t)$ | Deviation potential from the average struc-ture |
| $<>_{ts}$ | Statistical time and structure average |
| $\Psi$ | Electron wave function |
| $U_0$ | Accelerating voltage of the electron micro-scope |
| $m_0$ | Electron mass |
| $\gamma$ | Relativistic factor |
| $v$ | Electron velocity |
| $\Psi_0$ | Elastic wave initiated by an inci-dent plane wave of wave vector $\mathbf{K}_0$ |
| $G$ | Green's function |
| $\hat{G}$ | Fourier transform of G |
| $\mathbf{K}$ | Electron wave vector |
| $V'$ | Optical potential |
| $\Psi_0^{(0)}$ | Bragg scattered wave due to the aver-age periodic lattice $V_0$ ex-cluding $V'$ |
| $B_i(\mathbf{K}_0,\mathbf{r})$ | Bloch wave |
| $C$ | Bloch wave coefficients |
| $S(\mathbf{Q},\mathbf{Q}')$ | Dynamic form factor |
| $G_0$ | Green's function in free-space |
| $\mathbf{u},\mathbf{v},\boldsymbol{\tau}$ | Reciprocal space vectors |
| $\mathbf{g},\mathbf{h}$ | Reciprocal space lattice vectors |

based on this approximation, but no study has been initiated to examine the consequence caused by Yoshioka's approxi-mation.

On the other hand, calculations of the thermal diffuse scattering are usually carried out based on the distorted potential method first proposed by Takagi (1958), in which the thermal diffuse scattering is considered as a perturbation and only the first order diffuse scattering is considered. This approximation may be referred to as the distorted wave Born approximation (DWBA), which has been applied to calculate the diffuse scattering produced by rough surfaces in reflection high-energy electron diffraction (RHEED) (Dudarev et al., 1993) and short-range order of point defects in transmission electron diffraction (Wang, 1996a). It appears that, at first sight, the optical potential is primarily responsible for taking into account the reduction of intensity due to diffuse scattering, and it may have no relation with the multiple scattering effects. In fact, they are correlated with each other, as to be shown in this paper. Since the contribution made by inelastic scattering processes with energy-loss larger than 2 eV can be removed from the images and diffraction patterns using an energy filter but the diffuse scattering remains, our theory here is about thermal diffuse scattering (TDS) and short-range order of point defects.

This paper aims to derive a more precise form of the

"optical potential" with the use of the Green's function for a real crystal system instead of its form in free-space. It will be shown that the inclusion of this potential in the dynamical calculation automatically recovers the contributions made by the high order diffuse scattering, although the calculation is done using the equation derived for single diffuse scattering. Therefore, the calculated wave function is the full solution of the Schrödinger equation including all orders of diffuse scattering. Details are given to show the calculation of the optical potential using an improved Green's function.
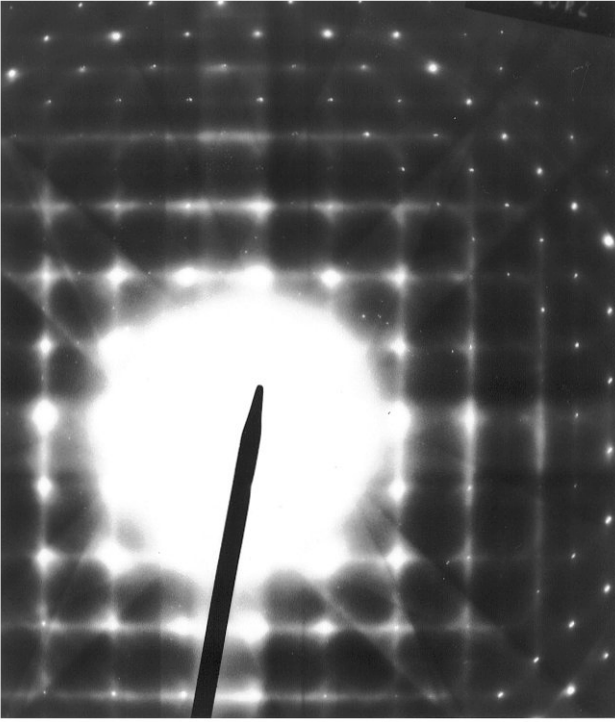
### A General Approach to Diffuse Scattering

Diffuse scattering is produced by structure modula-tion in a crystalline specimen, and the diffusely scattered electron intensity is distributed between Bragg reflected peaks. The Bragg reflections are generated by the periodically structured lattice of the crystal, while the diffuse scattering is produced by the non-periodical components including thermal vibrations of the crystal atoms and short-rage order (SRO) of point defects. Figure 1 shows an electron diffraction pattern recorded at 100 kV from Si foil. The TDS streaks observed in the pattern are determined by phonon dispersion relations of the acoustic branches (Honjo et al.,1964; Wang, 1992a). A general feature in the TDS diffraction pattern is that all of the streaks run along the lines interconnecting the Bragg peaks. For a monoatomic cubic structure, a simple rule has been proposed to predict the directions of the streaks in diffraction patterns from the unit cell structure of the crystal (Wang and Bentley, 1991). Dynamical theories for calculation of diffraction patterns and images of TDS electrons have been extensively developed based on the multislice approach (Fanidis et al., 1989, 1992, 1993; Coene and Van Dyck, 1990; Wang, 1991, 1992a and b, 1995a; Dinges et al., 1995), the Bloch wave approach (Howie, 1963; Rossouw, 1985; Rossouw and Bursill, 1985; Rez et al., 1977) and the Green's function approach (Dudarev et al., 1991; Wang and Li, 1995). All of these theories (for a review see Wang, 1995b) are based on the first order diffuse scattering approximation, as described in following.

To illustrate the focal point of this paper, we first review the classical approach of Takagi (1958), in which an *average crystal structure* is introduced. The crystal potential $V(\mathbf{r},t)$ is written into a form of

$$V(\mathbf{r},t) = V_0(\mathbf{r}) + \Delta V(\mathbf{r},t) \qquad (1)$$

where $V_0(\mathbf{r}) = <V(\mathbf{r},t)>_{ts}$ is the crystal potential for the average lattice, defined to be *time independent and periodic*, $<>_{ts}$ indicates the statistical time and struc-ture average, and $\Delta V(\mathbf{r})$ represents the deviation from the average lattice with $<\Delta V(\mathbf{r},t)>_{ts} = 0$, and it is *non-periodic and time-dependent* (for TDS). The statistical structure average $<>_s$ takes into account the

**Figure 1**. A [100] electron diffraction pattern recorded from a thin Si foil showing <110> streaks produced by thermal diffuse scattering.

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SRO of point defects. Figure 2 gives a schematic illustration of this theoretical approach for a case involving point defects.

Equation (1) clearly indicates that TDS is a time-dependent process and time-dependent Schrödinger equation should be used. However, as to be shown below, this time dependent process can be converted into a series of time-independent processes. In high energy electron scattering, the "frozen" lattice model is assumed in describing TDS (Hall and 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 final observed diffraction pattern contributed by mil-lions of electrons is equivalent to a time average on the intensities calculated for the different lattice configurations, and the *time t simply serves as a parameter signifying the spontane-ous lattice configura-tion in the calculation*. In fact, it has been proved by Wang (1998a) that the result obtained using the frozen lattice model is identical to the result of quantum phonon
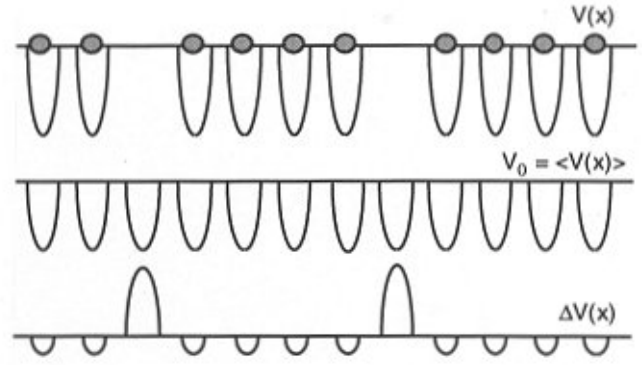


**Figure 2**. One-dimensional representation of a crystal potential V, the structurally averaged potential $V_0 = <V>$ and the deviation potential $\Delta V = V - V_0$ for a crystal containing point vacancies. $V_0$ is a periodic function but $\Delta V$ is not.

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excitation theory if (1) the incoherence between *different orders* of thermal diffuse scattering is considered in the frozen lattice model calculation, and (2) the specimen thickness and the mean-free-path length for phonon excitation both are smaller than the distance traveled by the electron within the life-time of the phonon. For the thin crystal case, the latter is absolutely satisfied.

The first objective in our theory is to find the scattered electron wave for a given frozen lattice config-uration, then a statistical time average is made on the electron diffraction *intensities* for a vast number of different thermal vibration configurations. For simplicity, we start from the time-independent Schrödinger equation with relativistic correc-tion (Humphreys, 1979; Spence, 1988),

$$\left( -\frac{\hbar}{2m_0}\nabla^2 - e\gamma V_0 - e\gamma\Delta V - E \right)\Psi = 0 \qquad (2)$$

where

$$E = eU_0\left[1 + \frac{eU_0}{2M_0c^2}\right]$$

$U_0$ is the accelerating voltage of the electron micro-scope, the relativistic factor $\gamma = (1 - v^2/c^2)^{-1/2}$, and v the electron velocity. For electron scattering, Equation (2) is converted into an integral equation with the use of the Green's function $G(\mathbf{r},\mathbf{r}_1)$ (Kainuma *et al.*, 1976):

$$\Psi(\mathbf{r},t)=\Psi_0(\mathbf{K}_0, \mathbf{r})+\int d\mathbf{r}_1\, G(\mathbf{r},\mathbf{r}_1)[e\gamma\Delta V(\mathbf{r}_1,t)\Psi(\mathbf{r}_1,t)] \qquad (3)$$

where G is the solution of

$$\left(-\frac{\hbar^2}{2m_0}\nabla^2 - e\gamma V_0 - E\right)G(r,r_1) = \delta(r - r_1) \qquad (4)$$

and $\Psi_0(\mathbf{K}_0,\mathbf{r})$ represents the elastic wave initiated by an incident plane wave of wave vector $\mathbf{K}_0$ after being scattered by the periodic, time-independent average potential $V_0$, and it satisfies

$$(-\frac{\hbar^2}{2m_0}\nabla^2 - e\gamma V_0 - E)\Psi_0 = 0 \qquad (5)$$

Equation (5) can be solved using the Bloch wave or multislice theory, while the solution of Equation (4) is not straightforward because a point source is located at $\mathbf{r} = \mathbf{r}_1$. It must be pointed out that the time variable in Equation (3) represents the instantaneous lattice configuration of the crystal due to thermal vibration. Equation (3) is usually solved iteratively so that each expansion term represents an order of the diffuse scattering. To illustrate our approach, the diffraction pattern is calculat-ed under the *first order diffuse scattering approximation*: $\Psi(\mathbf{r}_1,t)$ is replaced by $\Psi_0(\mathbf{K}_0,\mathbf{r}_1)$ in Equation (3). We first make this assumption, then we will come back to modify our approach to pick up the high order terms dropped by this approximation. Thus

$$\Psi(\mathbf{r},t)\approx$$
$$\Psi_0(\mathbf{K}_0,\mathbf{r})+\int d\mathbf{r}_1\, G(\mathbf{r},\mathbf{r}_1)[e\gamma\Delta V(\mathbf{r}_1,t)\Psi_0(\mathbf{K}_0,\mathbf{r}_1)] \qquad (6a)$$

where the first term stands for the elastic Bragg reflected waves and the second term is the first order diffuse scattering produced by TDS and/or SRO. This approximation holds if the specimen is thin. A consequence of this approximation is that the total number of incident electrons is not conserved because of the drop of higher order diffuse scattering terms. The diffraction pattern is calculated by

$$I(\mathbf{u}_b)=|\Phi_0(\mathbf{K}_0,\mathbf{u}_0,z=\infty|^2$$
$$+(e\gamma)^2\!\int d\mathbf{r}_1\!\int d\mathbf{r}_2 \hat{G}(\mathbf{u}_b,z=\infty,\mathbf{r}_1)\hat{G}*(\mathbf{u}_b,z=\infty,\mathbf{r}_1)$$
$$\times[<\Delta V(\mathbf{r}_1,t)\Delta V(\mathbf{r}_2,t)>_{ts}]\Psi_0(\mathbf{K}_0\mathbf{r}_1)\Psi_0*(\mathbf{K}_0\mathbf{r}_2) \qquad (6b)$$

where $\Phi_0$ and are the 2D Fourier transforms (FTs) of $\Psi_0$ and $G(\mathbf{r},\mathbf{r}_1)$ at $z = \infty$, respectively. The time and structure average of $<\Delta V(\mathbf{r}_1,t)\Delta V(\mathbf{r}_2,t)>_{ts}$ can be performed analytically before numerical calculation (Dudarev *et al.*, 1991; Wang, 1995a, 1996a). From the *reciprocity theorem*, $G(x, y, z=\infty, \mathbf{r}_1) = G(\mathbf{r}_1, x, y, z=\infty)$, provided there is no absorption. This relation means that the wave observed at $z = \infty$ when a point source is placed at $\mathbf{r}_1$ within the specimen is the same as the wave observed at $\mathbf{r}_1$ (in the specimen) when a point source is placed at $z = \infty$ (the image plane). In practice, when a point source is placed at $z = \infty$, the spherical wave emitted from the source is a plane wave when falls on the crystal surface, thus, $G(\mathbf{r}_1,x,y,z=\infty)$ is equivalent to the solution of the Schrödinger equation for an incident plane wave. This relation can be proven mathematically (Dudarev *et al.*, 1993) as

$$\hat{G}(\mathbf{u}_b,\mathbf{r}_i)=A_z\Psi_0(-\mathbf{K}-\mathbf{u}_b,\mathbf{r}_i) \qquad (6c)$$

where $\Psi_0(-\mathbf{K}, \mathbf{r}_1)$ is the solution of the Schrödinger equation (Eqn. 5) for an incident plane wave of wave vector $(-\mathbf{K})$ ($\mathbf{K} = \mathbf{K}_0 + \mathbf{u}_b$) and

$$A_z = -[im_0 \exp(2\pi iK_z z)](\pi\hbar^2 K_{0z})^{-1} \qquad (6d)$$

The negative sign of the wave vector means that the electron strikes the crystal along the negative z-axis direction. The elastic scattering wave $\Psi_0(-\mathbf{K}, \mathbf{r}_1)$ can be obtained using the conventional dynamical approaches, such as Bethe's theory. Thus, Equation (6b) is the basis of dynamical calculations under the first order diffuse scattering approximation, and it has been the fundamental equation for diffuse scattering.

To compensate for the loss of high order diffuse scattering terms in Equation (6a), an optical potential $V'$ is added in the Schrödinger equation (Eqn. 5) to modify the solution of $\Psi_0$ so that Equation (6a) may approach the exact solution of Equation (2)

$$(-\frac{\hbar^2}{2m_0}\nabla^2 - e\gamma V_0 - e\gamma V'-E)\Psi_0 = 0 \qquad (7)$$

This technique works if a unique solution of $V'$ can be found. The optical potential $V'$ is chosen in such a form that both Equations (6a) and (7) exactly satisfy Equation (2), the substitution requires

$$[V'\Psi_0]=e\gamma\int d\mathbf{r}_1\,[G(\mathbf{r},\mathbf{r}_1)\Delta V(\mathbf{r},t)\Delta V(\mathbf{r}_1,t)\Psi_0(\mathbf{K}_0,\mathbf{r}_1)] \qquad (8)$$
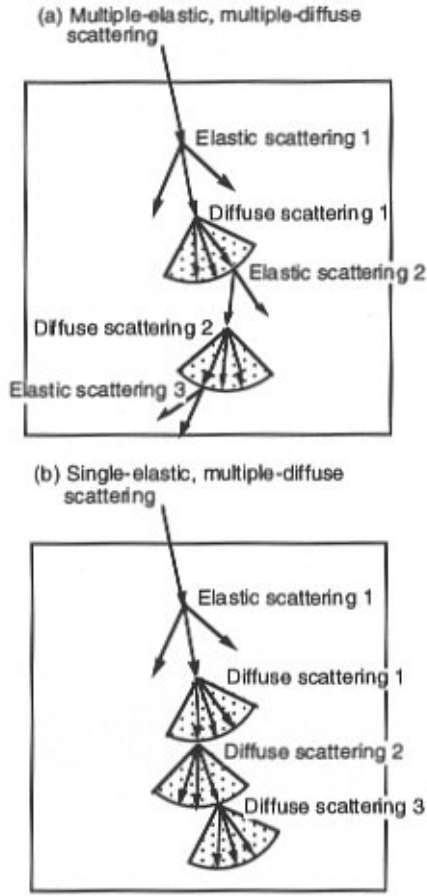
The function $V'$ defined in Equation (8) has two impor-tant characteristics. $V'$ is a non-local function since $V'$ cannot be separated from wave function $\Psi_0$. Strictly speaking, it is inadequate to call $V'$ a potential because of its dependence on the Green's function $G$, the solu-tion of which is determined by the crystal structure. To match the terminology that has been used in the literature, $V'$ is still referred as a "potential" function in the following discussion.

### The "Optical Potential" and Multiple Diffuse Scat-tering

The first objective is to prove that the optical potential $V'$ given by Equation (8) can be applied to recover the high order diffuse scattering terms droped when $\Psi(\mathbf{r}_1,t)$ is replaced by $\Psi_0(\mathbf{K}_0,\mathbf{r}_1)$ in deriving Equation (6a) under the first order diffuse scattering approximation. Starting from the integral form of Equation (7) with the use of Green's function and iterative calculation, the elastic wave is expanded as

$$\Psi_0(\mathbf{K}_0,\mathbf{r})=$$
$$\Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r})+e\gamma\int d\mathbf{r}_1 G(\mathbf{r},\mathbf{r}_1)[V'(\mathbf{r}_1)\Psi_0(\mathbf{K}_0,\mathbf{r}_1)]$$
$$= \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r})+(e\gamma)^2\int d\mathbf{r}_1 G(\mathbf{r},\mathbf{r}_1)\int d\mathbf{r}_2\,[G(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times\Delta V(\mathbf{r}_1,t)\Delta V(\mathbf{r}_2,t)\Psi_0(\mathbf{K}_0,\mathbf{r}_2)]$$

(a) Multiple-elastic, multiple-diffuse scattering

Elastic scattering 1

Diffuse scattering 1

Elastic scattering 2

Diffuse scattering 2

Elastic scattering 3

(b) Single-elastic, multiple-diffuse scattering

Elastic scattering 1

Diffuse scattering 1

Diffuse scattering 2

Diffuse scattering 3

**Figure 3**. Schematic diagrams showing multiple diffuse scattering processes in a crystal (**a**) with (G) and (**b**) without ($G_0$) consideration of dynamical diffraction between consecutive diffuse scattering events. The fan-shape intensity distribution indicates a diffuse scattering process.

$$= \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}) + (e\gamma)^2 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \, [G(\mathbf{r},\mathbf{r}_1)\, G(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times \Delta V(\mathbf{r}_1,t)\, \Delta V(\mathbf{r}_2,t)\, \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_2)]$$
$$+ (e\gamma)^4 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 \int d\mathbf{r}_4 \, [G(\mathbf{r},\mathbf{r}_1)\, G(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times G(\mathbf{r}_2,\mathbf{r}_3)\, G(\mathbf{r}_3,\mathbf{r}_4)\, \Delta V(\mathbf{r}_1,t)\, \Delta V(\mathbf{r}_2,t)$$
$$\times \Delta V(\mathbf{r}_3,t)\, \Delta V(\mathbf{r}_4,t)\, \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_4)] + \cdots \quad (9)$$

where $\Psi_0^{(0)}$ is the Bragg scattered wave due to the average periodic lattice at the absence of V′ (e.g., no absorption)

$$\left(-\frac{h^2}{2\,m_0}\nabla^2 - e\gamma V_0 - E\right)\Psi_0^{(0)} = 0 \quad (10)$$

This equation can be solved using conventional dynamic electron diffraction theories. Substituting Equation (9) into Equation (6a), the total scattered wave is

$$\Psi(\mathbf{r},t) = \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r})$$
$$+ (e\gamma) \int d\mathbf{r}_1 \, G(\mathbf{r},\mathbf{r}_1)\, \Delta V(\mathbf{r}_1,t)\, \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_1)$$
$$+ (e\gamma)^2 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \, [G(\mathbf{r},\mathbf{r}_1)\, G(\mathbf{r}_1,\mathbf{r}_2)\, \Delta V(\mathbf{r}_1,t)$$
$$\times \Delta V(\mathbf{r}_2,t)\, \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_2)]$$
$$+ (e\gamma)^3 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 \, [G(\mathbf{r},\mathbf{r}_1)\, G(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times G(\mathbf{r}_2,\mathbf{r}_3)\Delta V(\mathbf{r}_1,t)\Delta V(\mathbf{r}_2,t)\Delta V(\mathbf{r}_3,t)\Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_3)]$$
$$+ (e\gamma)^4 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 \int d\mathbf{r}_4 \, [G(\mathbf{r},\mathbf{r}_1)\, G(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times G(\mathbf{r}_2,\mathbf{r}_3)\, G(\mathbf{r}_3,\mathbf{r}_4)\, \Delta V(\mathbf{r}_1,t)\, \Delta V(\mathbf{r}_2,t)$$
$$\times \Delta V(\mathbf{r}_3,t)\, \Delta V(\mathbf{r}_4,t)\, \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_4)] \cdots \quad (11)$$

where all of the higher order terms have been recovered. The third term in Equation (11) is taken as an example to show its physical meaning, as schematically shown in Figure 3a. The Bragg scattered wave is diffusely scattered at $\mathbf{r}_2$ by $\Delta V(\mathbf{r}_2,t)$. The diffusely scattered wave is elastically scattered by the crystal lattice while propagating from $\mathbf{r}_2$ to $\mathbf{r}_1$ [$G(\mathbf{r}_1,\mathbf{r}_2)$], then, the second order diffuse scattering occurs at $\mathbf{r}_1$ [$\Delta V(\mathbf{r}_1,t)$]. Finally, the double diffusely scattered wave exits the crystal at $\mathbf{r}$ after elastic scattering when propagating from $\mathbf{r}_1$ to $\mathbf{r}$ [$G(\mathbf{r}, \mathbf{r}_1)$]. The integrals over $\mathbf{r}_1$ and $\mathbf{r}_2$ are to sum over the contributions made by all of the possible scattering sources in the crystal.

It can be proven directly from Equation (11) with the use of Equations (10) and (4)

$$\left(-\frac{h^2}{2\,m_0}\nabla^2 - e\gamma V_0 - E\right)\Psi(\mathbf{r},t) = e\gamma\Delta V(\mathbf{r},t)\,\Psi(\mathbf{r},t) \quad (12)$$

which is exactly the form of Equation (2), the equation we started with. Therefore, the multiple diffusely scattered waves are comprehensively included in the calculation of Equation (6a) if the optical potential V′ given by Equation (8) is introduced in the calculation of $\Psi_0$ (Eqn. 6a). This is a key conclusion which means that, by introducing a proper form of the optical poten-tial, the multiple diffuse scattering terms are automatical-ly included in the calculation using Equation (6a), although it was derived for the first order diffuse scattering. Thus, introduction of the optical potential V′ in the calculation of the elastic wave makes the existing theories available for calculating high order time-depen-dent systems (TDS). This conclusion is universal for a time-independent system because no assumption and approximation was made in the proof. There are two major steps in this calculation: the solution of Equation (6a) and the optical potential V′. These quantities are calculated separately in following sections.

### The Bloch Wave Solution

For transmission electron diffraction of a thin slab crystal, as shown in Figure 4, the Bloch wave theory is the best suited approach for solving the Schrödinger equation if the average crystal lattice is periodic. The wave function $\Psi_0$ is a linear superposition of the Bloch waves $B_i$

$$\Psi_0(K_0, r) = \sum_i \alpha_i(K_0) B_i(K_0, r) \tag{13a}$$

where

$$B_i(K_0, r) = \sum_g C_g^{(i)}(K_0) \exp[2\pi i(K_0 + g)\cdot r + 2\pi i v_i z] \tag{13b}$$

which is the eigen-solution of Equation (7); $\alpha_i$ are the superposition coefficients determined by the boundary conditions, and the result is $\alpha_i = C_{I0}^{(i)*}$, where $C_{I0}^{(i)}$ are the elements of the first column of the inverse of the matrix whose elements are $C_g^{(i)*}$ (row i and column g) (Spence and Zuo, 1992). Since the average crystal potential is a periodic function, it can be written into a Fourier series,

$$V(r) = \sum_g V_g \exp[2\pi i g \cdot r] \tag{14}$$

Thus, the substitution of Equations (13b) and (14) into Equation (7) gives

$$[K_0^2 - (k_i + g)^2]C_g^{(i)} + \frac{2\gamma m_0 e}{h^2}\sum_h V_{g-h} C_h^{(i)}$$
$$= -\frac{2\gamma m_0 e}{h^2 V_c}\int dr \exp[-2\pi i(k_i + g)\cdot r][V' B_i(K_0, r)] \tag{15}$$

where $\mathbf{k}_i = \mathbf{K}_0 + v_i\hat{z}$, and $V_c$ is the volume of the crystal. We now use Equation (8) to perform the calcu-lation on the right-hand side of Equation (15). Since $[V^{(i)}\Psi_0]$ given in Equation (8) is a time-dependent (or lattice configura-tion-dependent) function, it is approxi-mately represented by its time/structure average in the following calculation
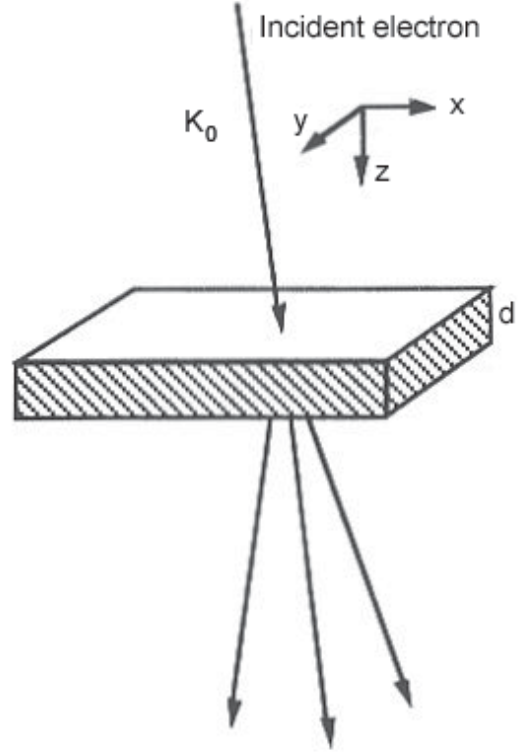
$$\int d\mathbf{r} \exp[-2\pi i(\mathbf{k}_i + \mathbf{g})\cdot\mathbf{r}] < [V' B_i(\mathbf{K}_0, \mathbf{r})]>_{ts}$$
$$= e\gamma\int d\mathbf{r}\int d\mathbf{r}_1 \exp[-2\pi i(\mathbf{k}_i + \mathbf{g})\cdot\mathbf{r}][G(\mathbf{r}, \mathbf{r}_1)$$
$$\times <\Delta V(\mathbf{r}, t)\Delta V(\mathbf{r}_1, t)>_{ts} B_i(\mathbf{K}_0, \mathbf{r}_1)] \tag{16}$$

To proceed with this calculation, a dynamic form factor $S(\mathbf{Q}, \mathbf{Q}')$ (for a review see Kohl and Rose, 1985) is introduced

$$<\Delta V(\mathbf{r}, t)\Delta V(\mathbf{r}_1, t)>_{ts}$$
$$= \int d\mathbf{Q}\int d\mathbf{Q}' \exp[2\pi i(\mathbf{r}\cdot\mathbf{Q} - \mathbf{r}_1\cdot\mathbf{Q}')] S(\mathbf{Q}, \mathbf{Q}') \tag{17}$$

The dynamic form factor $S(\mathbf{Q}, \mathbf{Q}')$ is an important quantity in dynamical inelastic electron diffraction. For TDS and SRO, the calculation of $S(\mathbf{Q}, \mathbf{Q}')$ is given elsewhere (Wang, 1995a, 1996a), in which the statistical time average over the instantaneous thermal vibration configurations of the crystal atoms and the statistical spatial distribution of the point defects with spatial short-ranged ordering have been evaluated analytically. Thus,

$$\int d\mathbf{r} \exp[-2\pi i(\mathbf{k}_i + \mathbf{g})\cdot\mathbf{r}] < [V' B_i(\mathbf{K}_0, \mathbf{r})]>_{ts}$$
$$= e\gamma\int d\mathbf{Q}\int d\mathbf{Q}' S(\mathbf{Q}, \mathbf{Q}')\int d\mathbf{r}\int d\mathbf{r}_1 \{\exp[2\pi i(\mathbf{k}_i + \mathbf{g})\cdot\mathbf{r}]$$
$$\times \exp[2\pi i(\mathbf{r}\cdot\mathbf{Q} - \mathbf{r}_1\cdot\mathbf{Q}')][G(\mathbf{r}, \mathbf{r}_1) B_i(\mathbf{K}_0, \mathbf{r}_1)]\}$$



**Figure 4**. A coordinate system used to describe trans-mission electron diffraction by a thin crystal slab of thickness d. The incident beam is nearly parallel to the z-axis.

$$= e\gamma\sum_h C_h^{(i)}(K)\int dQ\int dQ' S(Q, Q')$$
$$\times\int d\mathbf{r}\int d\mathbf{r}_1 \exp[-2\pi i(\mathbf{k}_i + \mathbf{g} - \mathbf{Q})\cdot\mathbf{r} - 2\pi i(\mathbf{Q}' - \mathbf{k}_i - \mathbf{h})\cdot\mathbf{r}_1]$$
$$\times G(\mathbf{r}, \mathbf{r}_1)$$
$$= \sum_h V_{gh}^{'(i)} C_h^{(i)}(K_0) \tag{18}$$

where

$$V_{gh}^{'(i)} = \frac{e\gamma}{V_c}\int dQ\int dQ' S(Q, Q') \hat{G}(k + g - Q, Q' - k_i - h) \tag{19}$$

and $\hat{G}(\mathbf{u}, \mathbf{u}_1)$ denotes the 3-D double Fourier transform of $G(\mathbf{r}, \mathbf{r}_1)$. Substituting Equation (18) into Equation (15), a matrix equation is obtained (Humphreys, 1979; Spence and Zuo, 1992)

$$[K^2 - (k_i + g)^2]C_g^{(i)} + \frac{2\gamma m_0 e}{h^2}\sum_h (V_{g-h} + V_{gh}^{'(i)}) C_h^{(i)} = 0 \tag{20}$$

This is just the eigen-equation of the Bloch wave theory except $V_{g-h}$ is replaced by $[V_{g-h} + V_{gh}^{'(i)}]$ (Note $V'$ depends also on the branch i of the Bloch wave). This is an elegant approach which

has automatically included the non-local effect of the optical potential. Equations (19) and (20) are equivalent to that derived by Radi (1968) for a general inelastic scattering case. The numerical calculation can be performed using the FORTRAN program developed by Spence and Zuo (1992).

The second objective here is to prove that Equation (19) is a generalized form of the optical potential first introduced by Yoshioka (1957). If the diffraction effect of the crystal is ignored so that the Green's function is replaced by its form in free-space,

$$G(\mathbf{r},\mathbf{r}_1) = G_0(\mathbf{r},\mathbf{r}_1) = \frac{2 m_0}{\hbar^2} \frac{\exp(2\pi i K_0 | \mathbf{r} - \mathbf{r}_1 |)}{4\pi |\mathbf{r} - \mathbf{r}_1 |} \quad (21)$$

after some calculation, Equation (19) gives

$$V_{gh}^{\prime,(i)} \approx -\frac{e m_0 \gamma}{2\pi^2 \hbar^2 V_c} \lim_{\varepsilon \to 0} \int d\mathbf{u} \frac{S(g+k_i-u,h+k_i-u)}{K_0^2 - u^2 + i\varepsilon}$$

$$= \frac{e m_0 \gamma}{2\pi^2 \hbar^2 V_c} \{ \int d\tau(u) \frac{S(g+k_i-u,h+k_i-u)}{u^2 - K_0^2}$$

$$+ i \frac{\pi}{2 K_0} \int d\sigma(u) S(g+k_i-u,h+k_i-u) \} \quad (22)$$

where the integral $\tau(\mathbf{u})$ is over all reciprocal space $\mathbf{u}$ except a spherical shell defined by $|\mathbf{u}| = K_0$, and the integral $\sigma(\mathbf{u})$ is over the Ewald sphere surface defined by $|\mathbf{u}| = K_0$. This is the form of the optical potential first obtained by Yoshioka (1957) and Yoshioka and Kainuma (1962). This function is complex, and its real part is usually ignored because it is much smaller than the crystal potential. But in recovering the high order terms, the real component needs to be included. The imaginary component is just the absorption potential that has been frequently used in dynamical calculations. The V′ given by Equation (22) is independent of the dynamical diffracting condition because of the substitution of G by $G_0$. In this case, V′ can be called an optical potential, but, in a general case, the function V′ is not simply a potential function because of the involvement of Bloch wave coefficients and crystal thickness.

From the discussion above, one might wonder: what is missing in dynamical calculations if the optical potential takes the form given by Yoshioka (1957)? If the Green's function is replaced by its form in free-space, Equation (11) is approximated as

$$\Psi(\mathbf{r},t) \approx \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r})$$
$$+ (e\gamma) \int d\mathbf{r}_1 G_0(\mathbf{r},\mathbf{r}_1) \Delta V(\mathbf{r}_1,t) \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_1)$$
$$+ (e\gamma)^2 \int d\mathbf{r}_1 \int d\mathbf{r}_2 [G_0(\mathbf{r},\mathbf{r}_1) G_0(\mathbf{r}_1,\mathbf{r}_2) \Delta V(\mathbf{r}_1,t)$$
$$\times \Delta V(\mathbf{r}_2,t) \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_2)]$$
$$+ (e\gamma)^3 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 [G_0(\mathbf{r},\mathbf{r}_1) G_0(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times G_0(\mathbf{r}_2,\mathbf{r}_3) \Delta V(\mathbf{r}_1,t) \Delta V(\mathbf{r}_2,t) \Delta V(\mathbf{r}_3,t)$$
$$\times \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_4)]$$
$$+ (e\gamma)^4 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 \int d\mathbf{r}_4 [G_0(\mathbf{r},\mathbf{r}_1) G_0(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times G_0(\mathbf{r}_2,\mathbf{r}_3) G_0(\mathbf{r}_3,\mathbf{r}_4) \Delta V(\mathbf{r}_1,t) \Delta V(\mathbf{r}_2,t)$$

$$\times \Delta V(\mathbf{r}_3,t) \Delta V(\mathbf{r}_4,t) \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_4)] + \cdots \quad (23)$$

We take the second term as an example to illustrate the physical meaning of this equation: the elastic wave at $\Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_1)$ is diffusely scattered at $\mathbf{r}_1$ by $\Delta V(\mathbf{r}_1,t)$, followed by a propagation in "free-space" from $\mathbf{r}_1$ to $\mathbf{r}$, where the wave exits the crystal. This approximation simply ignores the dynamical diffraction of the electron after the diffuse scattering, and it can be schematically shown by Figure 3b, where dynamical diffraction occurs only prior to the diffuse scattering (Høier, 1973). Equation (23) includes all of the orders of diffuse scattering but not the dynamical diffraction of the diffusely scattered waves. This is the consequence arisen from the Yoshioka's approximation. On the other hand, the multiple diffuse scattering is included in the optical potential. Therefore, the high order diffuse scattering has already been included in the dynamical calculation although the calculation is performed using the equation derived under the first order diffuse scattering approxi-mation. The optical potential has a rich meaning beyond the conventional interpretation of an absorption potential.

## The Green's Function Solution and The Optical Potential

The Green's function represents the electron wave distributed in the space due to a point source located at $\mathbf{r} = \mathbf{r}'$ in the crystal. The Green's function can be expressed into an integral form of the eigen Bloch states (Radi, 1968). In this section, we use the Green's function of Dudarev *et al.* (1994) to calculate the optical potential for a general case. The Green's function for electron scattering has been proven to be in the form of

$$G(\mathbf{r},\mathbf{r}') = \frac{m_0}{2\pi^2 \hbar^2} \lim_{\varepsilon \to 0} \int d\kappa \frac{\exp(-2\pi i \kappa \bullet \mathbf{r}')}{(\kappa^2 - K_0^2 - i\varepsilon)} \Psi_0^{(0)}(\kappa,\mathbf{r})$$

$$= \frac{m_0}{2\pi \hbar^2} \int d\mathbf{r}'' \frac{\exp(2\pi i K_0 |\mathbf{r}'-\mathbf{r}''|)}{|\mathbf{r}'-\mathbf{r}''|} \int d\kappa [\exp(2\pi i \kappa \mathbf{r}'')$$

$$\times \Psi_0^{(0)}(-\kappa,\mathbf{r})]$$

$$= \frac{m_0}{2\pi \hbar^2} \left[ \frac{\exp(2\pi i K_0 r')}{r'} \right] \otimes \int d\kappa [\exp(2\pi i \kappa \bullet \mathbf{r}') \times \Psi_0^{(0)}(-\kappa,\mathbf{r})]$$
$$(24)$$

where $\Psi_0^{(0)}(\hat{\mathbf{e}},\mathbf{r})$ is the solution of Equation (10) for an incident plane wave with wave vector $\kappa$, which can be calculated using conventional dynamical diffraction theories. This is a key relation which correlates the Green's function with the solution of the Schrödinger equation. We now take a double Fourier transform of $G(\mathbf{r},\mathbf{r}')$

$$\hat{G}(\mathbf{u},\mathbf{v}) = \int d\mathbf{r} \int d\mathbf{r}' \exp(-2\pi i \mathbf{u}\cdot\mathbf{r}) \exp(-2\pi i \mathbf{v}\cdot\mathbf{r}') G(\mathbf{r},\mathbf{r}')$$

$$= \frac{m_0}{2\pi^2\hbar^2} \lim_{\varepsilon \to 0} \frac{\Phi_0^{(0)}(-v,u)}{(v^2 - K_0^2 - i\varepsilon)} \qquad (25)$$

where $\Phi_0^{(0)}(\kappa,\mathbf{u})$ is the Fourier transform of $\Phi_0^{(0)}(\kappa,\mathbf{r})$, and the negative sign of the wave vector indicates that the incident plane wave strikes the crystal from the bottom surface. Substituting this equation into Equation (19), the optical potential is

$$V'^{(i)}_{gh} = \frac{e\gamma m_0}{2\pi^2\hbar^2 V_c} \lim_{\varepsilon \to 0} \int d\mathbf{u} \, [\int d\kappa \, S\,(k_i + g - u, k_i + h - \kappa)$$
$$\times \frac{\Phi_0^{(0)}(\kappa,u)}{\kappa^2 - K_0^2 - i\varepsilon} \, ]$$

$$= \frac{e\gamma m_0}{2\pi^2\hbar^2 V_c} \int d\mathbf{u}[\int d\tau(\kappa) \, S\,(k_i + g - u, k_i + h - \kappa) \frac{\Phi_0^{(0)}(\kappa,u)}{\kappa^2 - K_0^2}$$

$$+ i\frac{\pi}{2K_0}\int d\sigma(\kappa) \, S\,(k_i + g - u, k_i + h - \kappa)\Phi_0^{(0)}(\kappa,u)] \qquad (26)$$

where the integral $\tau(\kappa)$ is over all reciprocal space except a spherical shell defined by $|\kappa| = K_0$; the integral $\sigma(\kappa)$ is over the Ewald sphere surface defined by $K = K_0$; and $\mathbf{k}_i = \mathbf{K} + v_i\hat{z}$. The integral of $\mathbf{u}$ is to sum over the components scattered to the entire recipro-cal space. Since $1/(\kappa^2 - K_0^2)$ is an antisymmetric function when $\kappa \to (K_0-\varepsilon)$ and $\kappa \to (K_0+\varepsilon)$, thus the integral around the singular point $\kappa = K_0$ is approximately zero, thus, no abnormal numerical singularity is expected in numerical calculation (Rez, 1976). Equation (26) is a general solution, and its meaning can be illustrated by the following two cases, and a detailed analytical illustration of which will be given later.

For transmission electron diffraction, the wave function takes the form of the Bloch waves given by Equations (13a) and (13b), thus

$$\Phi_0^{(0)}(\kappa,u) = \sum_{j}\sum_{g'} C'^{(j)*}_0(\kappa) \, C'^{(j)}_{g'}(\kappa)\delta(u - \kappa - v_j\hat{z} - g') \quad (27)$$

where $C_{g'}^{(j)}$ are the solutions of Equation (20) without including the correction potential $V'^{(i)}_{gh}$, the corresponding optical potential is:

$$V'^{(i)}_{gh} = \frac{e\gamma m_0}{2\pi^2\hbar^2 V_c} \sum_{j}\sum_{g'} [\int d\tau(\kappa) \, C'^{(j)*}_0(\kappa) \, C'^{(j)}_{g'}(\kappa)$$

$$\times \frac{S(k_i + g - \kappa - v_j\hat{z} - g', k_i + h - \kappa)}{\kappa^2 - K_0^2} \, ]$$

$$+ i\frac{\pi}{2K_0}\int d\sigma(\kappa) \, C'^{(j)*}_0(\kappa) \, C'^{(j)}_{g'}(\kappa)$$

$$\times S(k_i + g - \kappa - v_j\hat{z} - g', k_i + h - \kappa)] \qquad (28)$$

As pointed out in Equation (8), $V'$ is a non-local func-tion depending on the Green's function sparked by a point source in the specimen. The finite thickness of the specimen affects the solution of the Green's function, resulting in the thickness dependence of $V'$. By the same token, $V'$ also depends on the Bloch wave coefficients that are responsible for the dynamical diffraction effect, which, however, was dropped in the theory of Yoshioka (1957). This is the key difference that distinguishes our theory from the conventional approach. This is also the critical point that the potential given by Equation (26) can automatically include multiple diffuse scattering in dynamical calculation with the use of the first order diffuse scattering equation (Eqn. 6a).

Under the zero order approximation, in which the wave function is the incident plane wave, $\Phi_0^{(0)}(\kappa,\mathbf{u}) = \delta(\kappa - \mathbf{u})$, Equation (26) gives the exact result of Equation (22).

**The Multiple Diffusely Scattered Electron Intensity**

The theory for HRTEM image calculation using a combined Bloch wave-multislice theory has been proposed (Wang, 1998b). We now use the Bloch wave theory to calculate the diffractiion pattern. The objective in this theory is to calculate the angular distribution of the multiple diffusely scattered electrons. We now consider the main steps in this calculation. Since $V'$ is approximated by its average value $<V'>_{ts}$ in Equation (16), which means that $\Psi_0(\mathbf{r},t)$ is replaced by $<\Psi_0(\mathbf{r},t)>_{ts}$, thus,

$$<\Phi_0(\mathbf{K}_0,\mathbf{u}_b,z=\infty)\,\Delta V(\mathbf{r},t)>_{ts}$$
$$= <\{\mathbf{FT}[<\Psi_0(x,y,z=\infty,t)>_{ts}]\,\Delta V(\mathbf{r},t)\}>_{ts} = 0 \qquad (29)$$

Therefore, with the use of Equations (6c) and (17), the diffraction pattern formed by the Bragg reflected and all diffusely scattered electrons is calculated by

$$I(\mathbf{u}_b) = |<\Phi_0(\mathbf{K}_0,\mathbf{u},t)]>|^2$$
$$+ D\int d\mathbf{Q}\int d\mathbf{Q}'\, S(\mathbf{Q},\mathbf{Q}') \, \{\int d\mathbf{r}_1 \, [\exp(2\pi i(\mathbf{r}_1\cdot\mathbf{Q})$$
$$\times \Psi_0^{(0)}(-\mathbf{K}_0-\mathbf{u}_b,\mathbf{r}_1) < \Psi_0(\mathbf{K}_0,\mathbf{r}_1)>]$$
$$\times \int d\mathbf{r}_2 \, [\exp(-2\pi i\mathbf{r}_2\cdot\mathbf{Q}')\,\Psi_0^{(0)*}(-\mathbf{K}_0-\mathbf{u}_b,\mathbf{r}_2)$$
$$\times <\Psi_0^*(\mathbf{K}_0,\mathbf{r}_2)>] \} \qquad (30)$$

where $D = e^2\gamma^2 m_0[2\pi^2\hbar^2 E\,\cos^2\phi_0]^{-1}$, and $\phi_0$ is the angle between $\mathbf{K}$ and z-axis, and $<\Psi_0(\mathbf{K}_0,\mathbf{r}_1)>$ is the solution of

$$(-\hbar^2/2m_0 \nabla^2 - e\gamma V_0 - E) <\Psi_0>$$
$$= e\gamma\int d\mathbf{r}_1[G(\mathbf{r},\mathbf{r}_1)<\Delta V(\mathbf{r},t)\Delta V(\mathbf{r}_1,t)>_{ts}<\Psi_0(\mathbf{K}_0,\mathbf{r}_1)>] \quad (31)$$

which is converted into Equation (20) in the Bloch wave theory; and $\Psi_0^{(0)}$ is the solution of Equation (10).

An iterative calculation of Equation (30) shows that all of

the even order terms of $\Delta V$ (e.g., $\Delta V^2$, $\Delta V^4$ etc.) are included, but the odd order terms (e.g., $\Delta V^3$, $\Delta V^5$ etc.) are dropped as a consequence of replacing $V'$ by its average. This result is equivalent to treating the electrons diffusely scattered by different orders of diffuse scattering as incoherent. In fact, it has been proven that the calculation according to Equations (30) and (31) recovers the entire multiple diffusely scattered electrons (Wang, 1996b,c). Therefore, Equation (30) holds even for highly distorted crystal structures. This is the point that we have proven in this paper. This result could have important applications in RHEED of a growing surface, where the surface roughness could be so high that the single diffuse scattering theory is insufficient.

### What is Missing in Conventional Calculations?

The theory for HRTEM image calculation using a combined Bloch wave-multislice theory has been pro-posed by Wang (1998b). Listed below is our calculation for the electron diffraction, which is more convenient with the use of Bloch's wave theory. The purpose of this paper is to illustrate the rela-tionship between the optical potential and multiple diffuse scattering. It is possible to cover all the high order diffuse scattering terms if the optical potential is introduced in an elegant way as showed earlier. The current dynamical calculations reported in the literature are usually performed with including the Debye-Waller factor and an imaginary absorption potential following an equation

$$[(-\hbar^2/2m_0)\,\nabla^2 - e\gamma V_0 - e\gamma V' - E]\,\Psi_0 = 0 \qquad (32)$$

We would like to know what is missing in this type of traditional calculation. To illustrate this point one starts from the Born series solution of Equation (32), which is

$$
\begin{aligned}
\Psi_0(\mathbf{K}_0,\mathbf{r}) &= \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}) \\
&+ e\gamma \int d\mathbf{r}_1\, G(\mathbf{r},\mathbf{r}_1)\,[V'(\mathbf{r}_1)\Psi_0(\mathbf{K}_0,\mathbf{r}_1)] \\
&= \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}) + (e\gamma)^2 \int d\mathbf{r}_1\,\{G(\mathbf{r},\mathbf{r}_1) \\
&\times \int d\mathbf{r}_2\,[G(\mathbf{r}_1\mathbf{r}_2)\,\Delta V(\mathbf{r}_1,t)\,\Delta V(\mathbf{r}_2,t)\,\Psi_0(\mathbf{K}_0,\mathbf{r}_2)]\} \\
&= \Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}) + (e\gamma)^2 \int d\mathbf{r}_1 \int d\mathbf{r}_2\,[G(\mathbf{r},\mathbf{r}_1)\,G(\mathbf{r}_1,\mathbf{r}_2) \\
&\times \Delta V(\mathbf{r}_1,t)\,\Delta V(\mathbf{r}_2,t)\,\Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_2)] \\
&+ (e\gamma)^4 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3 \int d\mathbf{r}_3\,[G(\mathbf{r},\mathbf{r}_1)\,G(\mathbf{r}_1,\mathbf{r}_2) \\
&\times G(\mathbf{r}_2,\mathbf{r}_3)\,G(\mathbf{r}_3,\mathbf{r}_4)\,\Delta V(\mathbf{r}_1,t)\,\Delta V(\mathbf{r}_2,t) \\
&\times \Delta V(\mathbf{r}_3,t)\,\Delta V(\mathbf{r}_4,t)\,\Psi_0^{(0)}(\mathbf{K}_0,\mathbf{r}_4)] + \cdots
\end{aligned}
\qquad (33)
$$

In comparison to Equation (11), the odd power terms of $\Delta V$ are missing. Therefore, in the classical dynamical calculation using either the Bloch wave or multislice theory, the contribution made by the second, fourth and all the even power order diffuse scattering terms are included but the first, third and all the even order diffuse scattering terms are ignored. Thus, the calculation includes only a small portion of the diffuse scattering, and the calculated results should be considered as pure-elastic Bragg scattering only.

The approximations made in conventional calcula-tions are summarized in following. First, the Green function G is replaced by its form in free-space $G_0$, which means that the dynamical elastic diffraction of the electrons is ignored once they are diffusely scattered. Second, the optical potential $V'$ is usually approximated as an imaginary function and the real part is ignored. This might be a good approximation for TDS, but it may not hold for SRO of point defects. Third, the first, third and all odd power terms of the diffuse scattering terms are dropped. Since the diffuse scattering is mainly distributed at high scattering angles, in the low scattering angular range the calculation accounts only the purely Bragg reflections although the Debye-Waller factor is included; in the high angle range, the calculation accounts only a small portion of the diffuse scattering. Finally, it must be pointed out that the Debye-Waller factor characterizes the weakening of atomic scattering factor due to the blurring effect of the atom thermal vibration, but the inclusion of this factor does not mean that the diffuse scattering is included in the calculation. This has been misunderstood by many readers. To account for the contribution of entire diffuse scattering, Equation (6a) (or Eqn. 30) must be used.

### Calculation of V' by the Born Series Method

The calculation of the Green's function, in principle, can be carried out following Equation (24), the computa-tion of this equation, however, could be very large because of the requirement of the elastic waves with a wide range of wave vectors and incident beam direc-tions. In the literature, the Green's function has been approximated by its form in free-space (Yoshioka, 1957)

$$
\begin{aligned}
G_0(\mathbf{r},\mathbf{r'}) &= \frac{m_0}{2\pi\hbar^2} \int du\, \frac{\exp[2\pi i \mathbf{u}\bullet(\mathbf{r}-\mathbf{r'})]}{\pi\,(u^2 - K_0^2 - i0)} \\
&= \frac{m_0}{2\pi\hbar^2}\,\frac{\exp[2\pi i K_0\,|\mathbf{r}-\mathbf{r'}|]}{|\mathbf{r}-\mathbf{r'}|}
\end{aligned}
\qquad (34)
$$

The consequence of this approximation is that the dynamical Bragg diffraction between consecutive diffuse scattering is ignored. We now introduce an approximated method for this calculation, and the purpose is to improve the original approximation proposed by Yoshioka (1957). We now use the Born series technique to give an approximated solution of the Green's func-tion (Wang, 1998c). Rewrite Equation (4) into the following form:

$$[(-\hbar^2/2m_0)\nabla^2 - E]\,G(\mathbf{r},\mathbf{r}_1) = \delta(\mathbf{r}-\mathbf{r}_1) + e\gamma V_0\,G(\mathbf{r},\mathbf{r}_1) \quad (35)$$

the Born series solution of this equation is

$$G(\mathbf{r},\mathbf{r'}) = G_0(\mathbf{r},\mathbf{r'}) + (e\gamma)\int d\mathbf{r}_1\, G_0(\mathbf{r},\mathbf{r}_1)\,V_0(\mathbf{r}_1)\,G(\mathbf{r}_1,\mathbf{r'})$$

$$= G_0(\mathbf{r},\mathbf{r}') + (e\gamma)\int d\mathbf{r}_1\, G_0(\mathbf{r},\mathbf{r}_1)\, V_0(\mathbf{r}_1)\, G_0(\mathbf{r}_1,\mathbf{r}')$$
$$+ (e\gamma)^2 \int d\mathbf{r}_1 \int d\mathbf{r}_2\, [G_0(\mathbf{r},\mathbf{r}_1)\, G_0(\mathbf{r}_1,\mathbf{r}_2)\, V_0(\mathbf{r}_1)$$
$$\times V_0(\mathbf{r}_2)\, G_0(\mathbf{r}_2,\mathbf{r}')]$$
$$+ (e\gamma)^3 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \int d\mathbf{r}_3\, [G_0(\mathbf{r},\mathbf{r}_1)\, G_0(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times G_0(\mathbf{r}_2,\mathbf{r}_3)\, V_0(\mathbf{r}_1)\, V_0(\mathbf{r}_2)\, V_0(\mathbf{r}_3)\, G_0(\mathbf{r}_3,\mathbf{r}')] + \cdots \qquad (36)$$

where the first term is the Green's function in free-space and the second term is the kinematical scattering compo-nent of the Green's function. The nth term $\mathbf{g}_n(\mathbf{r},\mathbf{r}')$ in Equation (36) is related to the (n-1)th term $\mathbf{g}_{n-1}(\mathbf{r},\mathbf{r}')$ by

$$\mathbf{g}_n(\mathbf{r},\mathbf{r}') = (e\gamma)\int d\mathbf{r}_1\, G_0(\mathbf{r},\mathbf{r}_1)\, V_0(\mathbf{r}_1)\, \mathbf{g}_{n-1}(\mathbf{r}_1,\mathbf{r}') \qquad (37)$$

We now take a double Fourier transform of $\mathbf{g}_n$, with the use of Equation (34)

$$\hat{g}_n(\mathbf{u},\mathbf{v}) =$$
$$\int d\mathbf{r} \int d\mathbf{r}'\, \exp(-2\pi i\mathbf{u}\cdot\mathbf{r})\, \exp(2\pi i\mathbf{v}\cdot\mathbf{r}')\, g_n(\mathbf{r},\mathbf{r}')$$

$$= \left[\,\frac{e\gamma m_0}{2\pi^2\hbar^2}\,\right]\frac{1}{\left(u^2 - K_0^2 - i0\right)}\int dr_1\{\exp[2\pi i u\cdot r_1]V_0(r_1)$$
$$\times \hat{g}_{n-1}(r_1,v)\}$$
$$= \left[\,\frac{e\gamma m_0}{2\pi^2\hbar^2}\,\right]\frac{1}{\left(u^2 - K_0^2 - i0\right)}\left[V_0(u)\otimes \hat{g}_{n-1}(u,v)\right]$$
$$(38)$$

where $V_0(\mathbf{u})$ is the Fourier transform of $V_0(\mathbf{r})$. The calculation of $\mathbf{g}_n(\mathbf{u},\mathbf{v})$ is given in the Appendix.

We now use the first order approximation to illustrate the consequence of including the crystal potential in the calculation of the optical potential $V'$. If only the first two terms are kept, with the use of Equation (34), then for a periodically structured crystal (see Appendix) the double Fourier transform of G under the first order approximation is

$$\hat{G}(u,v) = \left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\frac{\delta(u+v)}{\left(v^2 - K_0^2 - i0\right)}$$
$$+ (e\gamma)\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]^2 \frac{\sum_{g1} V_{g1}\delta(u+v-g_1)}{\left(u^2 - K_0^2 - i0\right)\left(v^2 - K_0^2 - i0\right)} \qquad (39)$$

A mathematical identity is used for the following calculation

$$\frac{1}{x - x' - i0} = P\left[\,\frac{1}{x - x'}\,\right] + i\pi\delta(x - x') \qquad (40)$$

where P signifies the principal value (i.e., the function $1/(x-x')$ is given as $1/(x-x')$ for all values of $x'$ except at the point $x = x'$, for which $1/(x-x')$ is taken to be identically zero). From Equations (19) and (39)

$$V'^{(i)}_{gh} = \frac{e\gamma}{V_c}\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\int\int dQ \int dQ' \frac{S(Q,Q')}{\left(|k_i + g - Q|^2 - K_0^2 - i0\right)}$$
$$\times\{\delta(Q'-Q+g-h) + (e\gamma)\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\frac{\sum_{g1} V_{g1}\delta(Q'-Q+g-h-g_1)}{\left(|Q'-k_i-h|^2 - K_0^2 - i0\right)}\}$$
$$= \frac{e\gamma}{V_c}\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\int du \int dv \frac{S(k_i+g-u,v+k_i+h)}{U_2 - K_0^2 - i0}\{\delta(u+v)$$
$$+ (e\gamma)\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\frac{\sum_{g1} V_{g1}\delta(u+v-g_1)}{v_2 - K_0^2 - i0}\}$$
$$= \frac{e\gamma}{V_c}\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\int\int du\{\frac{S(k_i+g-u,k_i+h-u)}{u^2 - K_0^2 - i0}$$
$$+ (e\gamma)\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\sum_{g1} V_{g1}$$
$$\times\frac{S(k_i+g-u,k_i+h-u+g_1)}{\left(u^2 - K_0^2 - i0\right)\left(|u-g_1|^2 - K_0^2 - i0\right)}\}$$
$$= \frac{e\gamma}{V_c}\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]\{(\int d\tau(u)\frac{S(k_i+g-u,k_i+h-u)}{u^2 - K_0^2}$$
$$+ i\frac{\pi}{2K_0}\int d\sigma(u)S(k_i+g-u,k_i+h-u)\}$$
$$+ \frac{e^2\gamma^2}{V_c}\left[\,\frac{m_0}{2\pi^2\hbar^2}\,\right]^2 \sum_{g1} V_{g1}$$
$$\times\{\int d\tau'(u)\frac{S(k_i+g-u,k_i+h-u+g_1)}{\left(u^2 - K_0^2\right)\left(|u-g_1|^2 - K_0^2\right)}$$
$$+ i\frac{\pi}{2K_0}\int d\sigma'(u)\frac{S(k_i+g-u,k_i+h-u+g_1)}{\left(|u-g_1|^2 - K_0^2\right)}$$
$$+ i\frac{\pi}{2K_0}\int d\sigma'(u-g_1)\frac{S(k_i+g-u,k_i+h-u+g_1)}{\left(u^2 - K_0^2\right)}$$
$$- \pi^2 \int d\sigma''(u)S(k_i+g-u,k_i+h-u+g_1)\times\delta(|u-g_1|-K_0)\}$$

$$(41)$$

where the terms in the first {} are those first derived by Yoshioka (1957) when the Green's function is replaced by its form in free-space. The terms contained in the second {} are those from the first order kinematical electron diffraction as included in the Green's function calculation: the integral $\tau'(\mathbf{u})$ is over all reciprocal space except the spherical shells defined by $|\mathbf{u}| = K_0$ and $|\mathbf{u}-\mathbf{g}_1| = K_0$, the two Ewald spheres (Fig. 5a); the integral $\sigma'(\mathbf{u})$ is over the Ewald sphere surface defined by $|\mathbf{u}| = K_0$ except the points falling on the other Ewald sphere defined by $|\mathbf{u}-\mathbf{g}_1| = K_0$; the integral $\sigma'(\mathbf{u}-\mathbf{g}_1)$ is over the Ewald sphere surface defined by $|\mathbf{u}-\mathbf{g}_1| = K_0$ except the points intersecting the other Ewald sphere defined by $|\mathbf{u}| = K_0$; the last integral $\sigma''(\mathbf{u})$ covers the intersection lines of the two Ewald spheres

(i.e., a line integral). The volume and line integrals give a real component correction to the potential, the surface integrals give an imaginary component which is usually referred as the absorption potential. Since the calculation includes the first order diffrac-tion effect in the Green's function, the Ewald sphere $|\mathbf{u}\text{-}\mathbf{g}_1| = K_0$ represents the newly generated scattering center at $\mathbf{g}_1$ due to Bragg reflection. The sum over $\mathbf{g}_1$ is to consider the contributions from all of the possible Bragg reflections scaled kinematically by the structure factor $V_{g1}$.

The integrals are over the volumes excluding the shells of the spheres, the surfaces of the spheres and the intersection lines of the sphere shells. The number of spheres involved in each calculation depends on the order of scattering to be included in the Green's function calculation. For higher order scattering, the integral of points characterized by delta functions (such as the joint points of three spheres) is possible, as shown in Figure 5b for a second order scattering.

It appears that the calculation of Equation (41) may encounter some numerical singularities, the separation of the integrals into volume, surface and line integrals automatically resolved this problem. It must be pointed out that the function $1/(u^2\text{-}K_0{}^2)$ is an asymmetric function when $u \rightarrow K_0+0$ and $u \rightarrow K_0$-0, thus the integral is close to zero around $u = K_0$ (Fig. 6).

## Conclusion

In this paper, the absorption potential first proposed by Yoshioka (1957) is revised for dynamic electron diffraction with consideration of the effects arising from thermal diffuse scattering and point defect scattering. Using the Green's function for a crystal instead of its form in free-space, a rigorous theoretical proof is given to show that the inclusion of this potential in the dynami-cal calculation automatically recovers the contri-butions made by the high order diffuse scattering although the calculation is done using the equation derived for single diffuse scattering. This conclusion gives the basis for expanding the conventional diffraction theories devel-oped under the first order diffuse scattering to cases where the specimen thickness is large and/or the degree of disorder is high. Therefore, the calculated wave function is the full solution of the Schrödinger equation including all of the orders of diffuse scattering. Strictly speaking, the optical potential here is no longer a potential function, and it has a much more rich meaning than the conventional interpretation of an absorption effect.

## Appendix

If $V_0$ is a periodic function, then it can be written into a Fourier series

$$V_0(r) = \sum_g V_g \exp(2\pi i g \cdot r) \tag{A1}$$

or

$$V_0(u) = \sum_g V_g \delta(u - g)$$

where $V_g$ are the so called structure factors which are related to the atom types and atom distribution in the unit cell. From Equation (37)

$$\hat{g}_n(u,v) = \left[ \frac{e\gamma m_0}{2\pi^2\hbar^2} \right] \frac{1}{(u^2 - K_0^2 - i0)} \times \sum_g V_g \hat{g}_{n-1}(u - g, v) \tag{A2}$$

The zeroth order is

$$\hat{g}_0(u,v) = \left[ \frac{m_0}{2\pi^2\hbar^2} \right] \frac{\delta(u,v)}{v^2 - K_0^2 - i0} \tag{A3}$$

from which the first order is calculated

$$\hat{g}_1(u,v) = (e\gamma)\left[ \frac{m_0}{2\pi^2\hbar^2} \right]^2 \frac{1}{(u^2 - K_0^2 - i0)(v^2 - K_0^2 - i0)} \times \sum_{g1} V_{g1}\delta(u + v - g_1) \tag{A4}$$
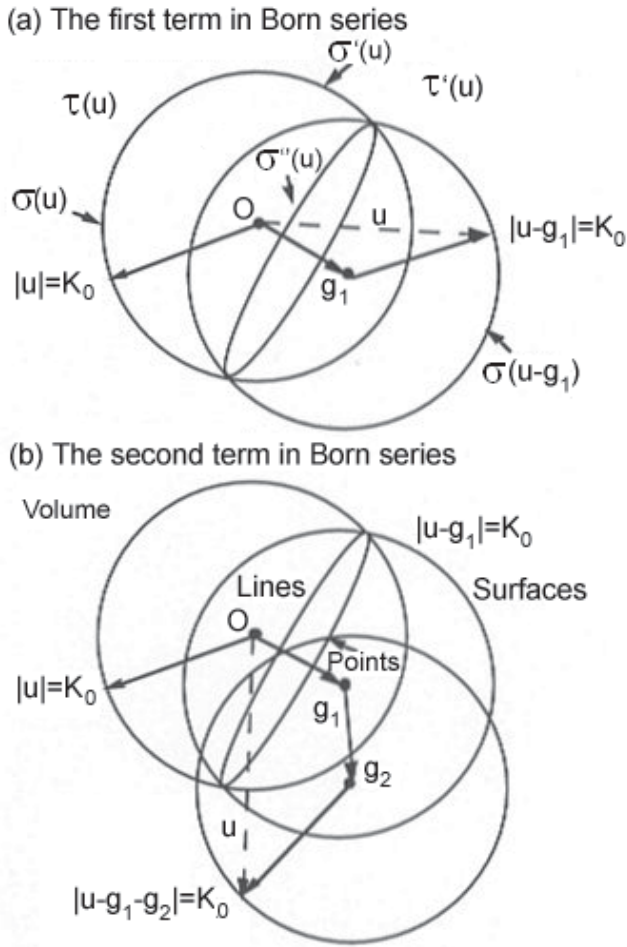
A substitution of Equation (A4) into (A2) gives the second order term

$$\hat{g}_2(u,v) = (e\gamma)^2\left[ \frac{m_0}{2\pi^2\hbar^2} \right]^3 \frac{1}{(u^2 - K_0^2 - i0)(v^2 - K_0^2 - i0)} \times \sum_{g1} \sum_{g2} \frac{V_{g1}V_{g2}\delta(u + v - g_1 - g_2)}{(|u - g_1|^2 - K_0^2 - i0)} \tag{A5}$$

The third order term is calculated similarly

$$\hat{g}_3(u,v) = (e\gamma)^3\left[ \frac{m_0}{2\pi^2\hbar^2} \right]^4 \frac{1}{(u^2 - K_0^2 - i0)(v^2 - K_0^2 - i0)} \times \sum_{g1} \sum_{g2} \sum_{g3} \frac{V_{g1}V_{g2}V_{g3}\delta(u + v - g_1 - g_2 - g_3)}{(|u - g_1|^2 - K_0^2 - i0)(|u - g_1 - g_2|^2 - K_0^2 - i0)} \tag{A6}$$

For the higher order terms with $n > 1$, and with the use of Equation (40)

(a) The first term in Born series



(b) The second term in Born series



**Figure 5**. (**a**) A model of two Ewald spheres showing the physical meaning of the integrals introduced in Equation (41). (**b**) A model for three Ewald spheres, where the point integral occurs.

$$\hat{g}_n(u,v) = (e\gamma)^n [\frac{m_0}{2\pi^2\hbar^2}]^{n+1} \frac{1}{(u^2 - K_0^2 - i0)(v^2 - K_0^2 - i0)}$$
$$\times \sum_{g1} \cdots \sum_{gn} V_{g1} \cdots V_{gn}$$
$$\times \frac{\delta(u + v - \sum_n g_n)}{(|u - g_1|^2 - K_0^2 - i0) \cdots (|u - g_1 \cdots - g_{n-1}|^2 - K_0^2 - i0)}$$
$$= (e\gamma)^n [\frac{m_0}{2\pi^2\hbar^2}]^{n+1} [P\{\frac{1}{u^2 - K_0^2}\} + i\frac{\pi}{2K_0}\delta(v - K_0)]$$
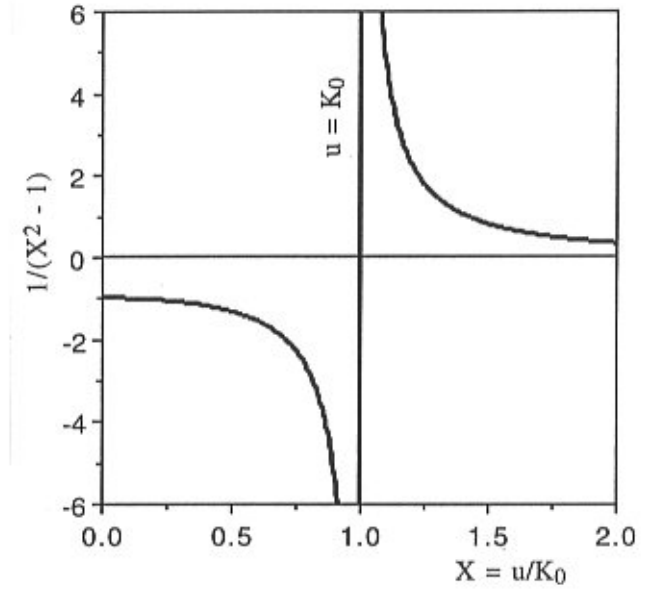


**Figure 6**. A plot of function $1/(u^2 - K_0^2)$ showing assym-metry around $u = K_0$.

$$\times [P\{\frac{1}{(v^2 - K_0^2)}\} + i\frac{\pi}{2K_0}\delta(V - K_0)]$$
$$\times \{\sum_{g1} \cdots \sum_{gn} V_{g1} \cdots V_{gn} \delta(u + v - \sum_n g_n)$$
$$\times [P\{\frac{1}{(|u - g_1|^2 - K_0^2)}\} + i\frac{\pi}{2K_0}\delta(|u - g_1| - K_0)]$$
$$\cdots [P\{\frac{1}{(|u - g_1 \cdots g_n - 1|^2 - K_0^2)}\}$$
$$+ i\frac{\pi}{2K_0}\delta(|u - g_1 \cdots g_{n-1}| - K_0)]\}$$

(A7)

These terms appear to have singularities while perform-ing the integrals, but as will be shown in Equation (41) that the use of Equation (40) separates the integrals into volume, surface, line and point (for higher order scatter-ing) integrals, thus the singularities are automatically resolved.

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*Erratum:

Equation (15) in Wang (1996c) and Equation (16b) in Wang (1996b) contain a typographic error. The correct form of these Equations should be as follows:

$$< |\Phi_0(\mathbf{K}_0, \mathbf{u}_b, t)|^2 >_{ts} \approx | <\Phi_0(\mathbf{K}_0, \mathbf{u}_b, t)] >_{ts}|^2$$

**Discussion with Reviewers**

**P. Rez**: Following Yoshioka (1957) the total wave function can be written as a product of the fast electron wave function and some generalized wave function representing the crystal in its ground or excited state. Wouldn't this affect the terms that are collected in Equations (9) and (11)?

**Author**: There are two approaches for calculating the intensity of diffusely scattered electrons. The first theory considers the phonon scattering to be an excitation of crystal state and the transition is described by a matrix element $H_{n0}'$, as described by Yoshioka's theory (1957). This approach is the quantum mechanical theory and it is suitable for both high and low energy electrons, but the theory can only be reasonably formulated under the single phonon scattering approximation. The theory becomes incredibly complex if multi- and multiple-phonon processes are included, such as the case de-scribed in this paper.

The other approach is specifically developed for high energy electrons under the frozen lattice approximation. Most of the theories in the literature are devel-oped following this scheme and the results have shown an excellent agreement with experimental observations (Loane *et al.*, 1991). This quasi-elastic scattering theory is the approach adopted in this paper, and I do not think the calculation following Yoshioka's theory, if possible, would produce any difference from the terms collected in Equations (9) and (11).

**P. Rez**: Is there not a difference in treating diffuse scattering from disorder and diffuse scattering from crystal excitations (such as phonons, plasmons, etc.)? Would it not be better to use a density matrix formalism to explicitly bring out these differences?

**Author**: A condition for the quasi-elastic scattering theory in the frozen lattice model to be hold is that the electron energy-loss suffered in phonon excitation (< 0.1 eV) is much smaller than the energy spread of the filament in TEM (~ 0.5 eV). With consideration of the large difference in time scale, it is very reasonable to treat TDS as a quasi-elastic scattering process. For plasmon excitation, however, the electron energy-loss is usually more than a few eV, much larger than the energy spread of the filament, thus, the incoherence among the inelastically scattered electrons must be considered, and the Yoshioka's theory needs to be adopted. Therefore, the theoretical approaches for the phonon and plasmon excitations are quite different.

The density matrix theory could also give the higher order terms, but its numerical calculation is usually carried out iteratively and the accuracy depends on how fast the

calculation converge. The theory presented here, however, shows that the calculation under the first order approximation covers all of the higher order terms if the optical potential is introduced properly.

**P. Rez**: The real part in Equation (28) has already been shown to be vanishingly small.

**Author**: The numerical calculation of Rez (1978) showed that the real component of the optical potential for single electron excitation is very small, but the situation could be very different for TDS and point defects. A full calculation must be done before extrapo-lating the conclusion.

**P. Rez**: Can the author give an estimation on the magnitude of the contributions representing various orders of scattering to the optical potential $\Delta V$?

**Author**: From Figure 2, it can be seen that the shape of $\Delta V$ can be hardly approximated by its first order term which is proportional to $dV/dr$. The numerical calcula-tion shown by Wang (1995b, chapter 6) has demonstrat-ed this result, and the high order terms are undoubtedly important.

**G. Anstis**: Does your work have any implications for determining structure factors by convergent beam electron diffraction (CBED)?

**Author**: The dynamical calculations on CBED reported in the literature have been performed using Equations (20) (or Equations 7 and 22). Based on the Born series form of Equation (7), as given in Equation (9), the calculation covers only the contributions made by even order terms of $\Delta V$ (i.e., $\Delta V^2$, $\Delta V^4$ etc, see the discus-sion given below Equation 33), but those by all of the odd order terms (i.e., $\Delta V$, $\Delta V^3$ etc) are missing. This means that only the second, fourth and sixth orders TDS are included in the calculation, but the contributions made by the first, third and fifth orders TDS are ig-nored. Thus, the result of this paper will have an important impact on quantitative CBED since all of the TDS electrons remain although an energy filter is employed.

**G. Anstis**: Can you say what implications your results have for Cowley and Moodie's multislice method using complex potentials? In this method we cannot use thickness dependent scattering functions.

**Author**: In Cowley and Moodie's multislice theory, the phase grating function of each slice is
$Q = \exp[i\sigma(V_0 + \Delta V)]$. Following Cowley's (1995) approach, the absorption potential is introduced by taking a time average of Q,

$$<Q> = \exp[i\sigma V_0] <\exp[i\sigma \Delta V]>$$

$$= \exp[i\sigma V_0][1 - \frac{\sigma^2 <(\Delta V)^2 >}{2}]\}$$

$$= \exp\{i\sigma[V_0 + i\frac{\sigma <(\Delta V)^2 >}{2}]\}$$

This approach keeps the even order terms of $\Delta V$, but the odd terms are dropped. This means that the calcula-tion keeps the contributions made by the second, fourth and sixth orders of TDS, but those made by the first, third and fifth orders of TDS are not included. Thus the multislice theory needs to be extended to include all these odd terms following the similar scheme introduced in this paper (see Wang, 1995a for details).

To properly introduce the absorption effect in the multislice calculation, two approximations are made. First, the non-local property of the optical potential is ignored, which means $V'$ can be separated from $\Psi_0$. Thus, $\Psi_0(\mathbf{K}_0, \mathbf{r}')$ is replaced by $\Psi_0(\mathbf{r})$ and Equation (8) is approximated as

$$V'(\mathbf{r}) \approx e\gamma \int d\mathbf{r}_1 [G(\mathbf{r},\mathbf{r}_1) <\Delta V(\mathbf{r},t) \Delta V(\mathbf{r}_1,t) >_t]$$

Secondly, the Green's function is replaced by its form in free space, thus, the optical potential is approximated as

$$V'(\mathbf{r}) \approx e\gamma \int d\mathbf{r}_1 [G_0(\mathbf{r},\mathbf{r}_1) <\Delta V(\mathbf{r},t) \Delta V(\mathbf{r}_1,t) >_t]$$

$$= \frac{e\gamma m_0}{2\pi^2 \hbar^2} \int dQ \int dQ' \int dr_1 \int d\kappa \frac{\exp[2\pi i\kappa \cdot (r - r_1)]}{(\kappa^2 - K_0^2 - i0)}$$
$$\times \exp[2\pi i(r \cdot Q - r_1 \cdot Q')] S_{TDS}(Q, Q')$$

$$= \frac{e\gamma m_0}{2\pi^2 \hbar^2} \int dQ \int dQ' \frac{\exp[2\pi ir \cdot (Q - Q')]}{(Q'^2 - K_0^2 - i0)} S_{TDS}(Q, Q')$$

This complex potential should be used in image calcula-tions. The recent theory by Wang (1998b) has implicated this approach in the image calculation using the multi-slice theory.

**G. Anstis**: Does the non-local property of the optical potential can be incorporated into the multislice calculations? what errors are introduced by making a local approximation?

**Author**: It is possible to introduce the non-local property of the optical potential for the atoms distributed within one slice, but it may be difficult to incorporate the non-local property along the beam direction (or z-axis) since the fundamental assumption of the multislice theory is no backscattering. In other words, the scattering in the nth slice is assumed not being affected by the behavior of the slices located at deeper depths.

The error introduced by the approximation of local optical potential is equivalent to the error introduced by replacing

the Green's function with its form in vacuum, which means the multiple Bragg reflections between successive diffuse scattering events are ignored. Therefore, the accuracy of the calculated Kikuchi patterns using the multislice theory might be affected, particularly at high scattering angle region, but for high resolution TEM image calculations the error could vanish.

**G. Anstis**: Does your method apply only to uncorrelated atomic displacements?

**Author**: No. The theory is applicable for a general case with consideration of phonon dispersion relations. The calculation of dynamic form factors under the harmonic oscillators approximation for TDS and SRO of point defects have been given elsewhere (Wang, 1996a). The theory also works when TDS and SRO coexist.

**D. Van Dyck**: The paper claims to extend the Yoshioka theory for thermal diffuse scattering. However, the original Yoshioka theory was set up for inelastic scattering whereas the treatment in this paper only deals with elastic diffuse scattering. Indeed, in the Yoshioka paper the wave function of the whole system (electron + crystal) is considered and expanded in eigenstates of the crystal Hamiltonian so as to include inelastic electron-crystal interactions that change the state of the crystal (e.g., create a phonon). The present paper only describes the electron wave and leaves the crystal unaffect-ed by the electrons.

**Author**: I always claim that the aim of this paper is to expand the meaning and usefulness of the optical poten-tial, first introduced by Yoshioka, beyond the conven-tional interpretation. The paper was not intended to extend the Yoshioka's inelastic scattering theory (e.g., the coupled differential equations). This might be a misunderstanding.

The referee is correct that I did not consider the change of crystal states as a result of phonon excitations. If the electron and the crystal is considered as a system, then the heat (or energy) transfer between the crystal and the environment (i.e., the specimen holder in TEM) would also have a small affect on the form of the absorption potential (Fanidis *et al*., 1992, 1993), but this effect might be negligibly small in comparison to other factors.

**D. van Dyck**: If one uses a time-dependent potential one should in principle start from the time dependent Schrödinger equation unless it is proven that the approxi-mation of using t as a parameter is valid (for instance if the time scales are largely different).

**Author**: The frozen lattice model (or quasi-elastic scattering model) is usually adopted in the calculation of TDS for high energy electrons. It is this model which converts a time-dependent process into a sum over the time segments composed of many instantaneous time-independent processes because the interaction time between an incident electron with the crystal in TEM is much much shorter than the vibration period of the crystal atoms. Thus, the lattice configuration is seen as if stationary for one incident electron. For the next incident electron, the crystal atoms are "frozen" at different positions. Therefore, the observed diffraction pattern contributed by many electrons is a sum of the intensities contributed by these electrons, each of which has been scattered by a slightly distorted lattice, equivalent to taking a time average over the scattering intensity, while time t is taken as a parameter in the calculation. The validity of the frozen lattice model has been discussed by Wang (1995b, sections 2.1 and 7.1) and by Loan *et al*. (1991).

**D. van Dyck**: How are the boundary conditions as introduced at exit and entrance face of the crystal, or is the vacuum considered as a part of the object for which V = 0?

**Author**: The boundary condition is introduced at the entrance face of the crystal z = 0 (see the paragraph following Equation 13b).

**D. van Dyck**: The diffraction pattern is calculated as the 2D Fourier transform of $\Psi$ at $z = \infty$. I believe however that the diffraction pattern is the Fourier transform of the wave function at the exit face of the crystal, or in the Fraunhofer approximation at the wave function $\Psi$ itself at $z = \infty$.

**Author**: For diffraction pattern calculation, both ap-proaches give the same answer, as proven below. If the electron wave function at the exit face z = d of the crystal is $\Psi(\mathbf{b},d)$, the diffraction intensity is $|FT[\Psi(\mathbf{b},d)]|^2$, where $\mathbf{b} = (x, y)$ and FT stands for the 2D Fourier transform. We now shift the observation point to $z = L$ ($L \rightarrow \infty$), the propagation of the wave in free space for a distance of L-d needs to be considered. Thus, the wave function observed at z = L, under the small angle scattering approximation, is (Cowley, 1995)

$$\Psi(\mathbf{b},L) = \Psi(\mathbf{b},d) \otimes P(\mathbf{b},L\text{-}d)$$

where $\otimes$ stands for a convolution calculation of $\mathbf{b}$, and P is the Fresnell propagator

$$P(\,\mathbf{b},z) = \frac{\exp(\pi iK\,|\mathbf{b}|^2\,/z)}{i\lambda z}$$

Since $FT[P(\mathbf{b},z)] = \exp(\text{-}\pi iu^2\Delta z\lambda)$, one simply has

$$|FT[\Psi(\mathbf{b},L)|^2 = |FT[\Psi(\mathbf{b},d)]|^2\,|FT[P(\mathbf{b},L\text{-}d)]|^2$$
$$= |FT[\Psi(\mathbf{b},d)]|^2$$

Thus, the diffraction patterns are the same at the crystal exit face z = d and at $z = \infty$.

**D. van Dyck**: V′ is called a nonlocal function. In the strict sense it is not a function but rather a functional.
**Author**: I agree with the reviewer.

### Additional References

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