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Multiple Scattering Theory for Fast Electrons in Single Crystals and Kikuchi Patterns

BY S. L. DUDAREV AND M. I. RYAZANOV

Moscow Engineering Physics Institute, Moscow 115409, *USSR*

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Abstract

By solving the kinetic equation for the density matrix of fast electrons the intensity distribution of the diffuse background and the Kikuchi lines and bands is found. It is shown that the Kikuchi pattern contrast depends on the angle of deflection of the scattered particles and the crystal thickness. For a thin crystal the expressions obtained coincide with the usual results found in the single inelastic scattering approximation. The theory takes into account both the dynamical diffraction and multiple inelastic scattering of electrons and gives a simple interpretation of a variety of contrast effects observed in thick single crystals.

1. Introduction

As is well known the diffraction of inelastically scattered electrons in crystals leads to Kikuchi line and band formation (Tomas & Goringe, 1979; Reimer, 1984). Kainuma (1955), Fujimoto & Kainuma (1963), Okamoto, Ichinokawa & Ohtsuki (1971) and Ohtsuki (1983) calculated the intensity distribution in Kikuchi patterns using the single inelastic scattering approximation. This approximation is only valid for very thin crystals (the thickness should be smaller than the mean free path with respect to inelastic collisions). In the case of a thick crystal one needs to take into account multiple inelastic scattering. As was pointed out by Hall (1970), Ishida (1970, 1971) and Chukhovskii, Alexanjan & Pinsker (1973), the contrast reversal of Kikuchi bands can be explained by means of absorption of inelastically scattered electrons. This procedure, however, does not conserve the total probability and leads to an exponential decrease of the scattered electron density (Thomas & Humphreys, 1970; Serneels, Van Roost & Knuyt, 1982). This result is at variance with experimental observation (Uyeda & Nonoyama, 1967, 1968) of the Kikuchi pattern in thick crystals (where the thickness is larger than the mean free path with respect to inelastic collisions). The application of an iterative method to the problem of electron multiple scattering has been reported by Høier (1973). This approach leads to a result in the form of an infinite series with many difficulties for quantitative evaluation.

The most consistent treatment of this problem is the use of the quantum kinetic equation for a single-particle density matrix (Blum, 1981). This formulation of multiple scattering theory was first used by Migdal (1955) for the case of random space-distribution scatterers. Kagan & Kononets (1970, 1973, 1974) in their theory of proton channelling generalized this method for the case of a crystalline medium. Bird & Buxton (1980) discussed the application of similar equations to electron diffraction.

Most of the known analytical solutions of the quantum kinetic equation in single crystals were obtained in the diffusive approximation (Kagan & Kononets, 1973). This approximation does not work for 10-1000 keV electrons, *i.e.* in the energy range used for observations by transmission electron microscopy.

For the two-beam case the quantum kinetic equation was solved by Dudarev & Ryazanov (1985) without using the diffusive approximation.

Nevertheless, the two-beam approximation fails in many cases for fast electrons, especially if they move along high-symmetry directions. For this reason, to

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calculate the electron angular distribution in thick crystals one should know some non-diffusive many-beam solutions of the quantum kinetic equation. A suitable mathematical technique is presented below.

The electron–atom differential cross section $d\sigma(\vartheta)/d\Omega$ has a rather sharp maximum in the forward direction. Consequently, the electron velocity direction in a thin crystal does not really change if the crystal thickness L is smaller than the transport length

$$l_{tr} = [N_1 \int d\sigma(\vartheta)(1-\cos\vartheta)]^{-1}$$

where N_1 is the number of atoms per unit volume. The total path length R_B for fast electrons is always larger than l_{tr} ($R_B > l_{tr}$) and the electron energy does not vary appreciably in a thin crystal. Under these conditions the penetration depth z is related to the time t by $z = v_0 t$, where v_0 is the initial electron velocity (Kagan & Kononets, 1973, 1974; Gratias & Portier, 1983). In this case one can obtain the electron angular distribution from the solution of the time-dependent kinetic equation taken at the moment $T = L/v_0$.

This equation should incorporate both diffraction by the crystal lattice mean potential and inelastic scattering accompanied by excitation of the electron and phonon subsystems. Such an equation is derived below [equation (23)]. In the limiting case of pure elastic scattering this equation (23) yields the well known elastically scattered electron distribution (37), (45) (*i.e.* intensity of Bragg spots). As is demonstrated below, the first iteration of (23) leads to the Kikuchi pattern intensity distribution discovered by Kainuma (1955), Fujimoto & Kainuma (1963), Okamoto *et al.* (1971) and Ohtsuki (1983). The present paper is concerned with a new approximate solution of the quantum kinetic equation for thick crystals. This method gives some analytical expressions for the Kikuchi line and band contrast, (74), (75), which are in good agreement with the observed angular distribution of electrons transmitted through a thick crystalline foil.

2. A quantum kinetic equation for fast-electron multiple scattering

As usual we describe a dissipative quantum system evolution in terms of a density matrix $\rho(x_1, x_2; t)$, where x_1 and x_2 are the system phase-space coordinates and t is time. The density matrix for the isolated stationary quantum system is a simple product

$$\rho(x_1, x_2; t) = \Psi(x_1; t)\Psi^*(x_2; t) \qquad (1)$$

where $\Psi(x; t)$ is the system wave function. The probability density is related to the matrix diagonal elements

$$\rho(x, x; t) = |\Psi(x; t)|^2. \qquad (2)$$

The density-matrix off-diagonal elements describe the mutual coherence properties of the system states at the phase-space points x_1 and x_2 (Blum, 1981).

In the case considered the phase-space coordinates are the fast-electron momentum $\mathbf{p}$ and the set of crystal state coordinates n.

The motion of an electron can be described by means of the electron single-particle density matrix

$$\rho(\mathbf{p}, \mathbf{p}'; t) = \sum_n \rho(\mathbf{p}, \mathbf{p}'; n, n; t). \qquad (3)$$

From (3) the electron momentum probability distribution has a form

$$w(\mathbf{p}, T) = \rho(\mathbf{p}, \mathbf{p}, T). \qquad (4)$$

By making use of (4), the electron angular distribution can be written down as

$$dw(\mathbf{\Omega})/d\Omega = \int_0^\infty dp\, p^2 \rho(p\mathbf{\Omega}, p\mathbf{\Omega}; T) \qquad (5)$$

and the electron energy spectrum is

$$dw(E)/dE = \int d^3p\, \rho(\mathbf{p}, \mathbf{p}; T)\delta(E - \mathbf{p}^2/2m) \qquad (6)$$

where m is the electron mass. The momentum probability distribution of fast electrons transmitted through a crystal slab can be obtained from (4)–(6) by means of the change $T \to L/v_0$, where L is the thickness of the slab.

A self-consistent equation for $\rho(\mathbf{p}, \mathbf{p}'; t)$ will be developed below in the case of low electron-beam intensity (the crystal properties do not change under the electron irradiation). An equation of motion for the density matrix has the form (Blum, 1981)

$$\begin{aligned} i\hbar\, \partial\rho(\mathbf{p}, \mathbf{p}'; n, l; t)/\partial t \\ = (\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'} + E_n - E_l)\rho(\mathbf{p}, \mathbf{p}'; n, l; t) \\ + \int [d^3q/(2\pi)^3] \sum_f \langle n|U(\mathbf{q})|f\rangle \rho(\mathbf{p} - \hbar\mathbf{q}; \mathbf{p}'; f, l; t) \\ - \int [d^3q/(2\pi)^3] \sum_f \rho(\mathbf{p}, \mathbf{p}' + \hbar\mathbf{q}; n, f; t)\langle f|U(\mathbf{q})|l\rangle \end{aligned} \qquad (7)$$

where $\varepsilon_{\mathbf{p}} = \mathbf{p}^2/2m$. E_n is the energy of a crystal excited state,

$$U(\mathbf{q}) = \int d^3r\, U(\mathbf{r}) \exp(-i\mathbf{q}\mathbf{r}),$$

and $U(\mathbf{r})$ is the electron–crystal interaction potential

$$\begin{aligned} U(\mathbf{r}) = -\sum_a Z_a e^2/|\mathbf{r} - \mathbf{R}_a - \mathbf{u}_a| \\ + \sum_{a,b} e^2/|\mathbf{r} - \mathbf{R}_a - \mathbf{u}_a - \mathbf{r}_{ba}| + \sum_v e^2/|\mathbf{r} - \mathbf{r}_v| \end{aligned} \qquad (8)$$

where Z_a is the atomic number of an atom of the medium, $\mathbf{R}_a$ denotes the equilibrium position of the atom, and $\mathbf{u}_a$ is its thermal displacement. The second and third terms in (8) describe the interaction of a fast incident electron with, respectively, inner-shell electrons, whose coordinates $\mathbf{r}_{ba}$ are reckoned from

the instantaneous position of the centre of the atom, and weakly bound conduction electrons.

As usual we assume the initial crystal state to be the thermodynamical equilibrium state with the density matrix (Feynman, 1972)

$$(\rho_{\text{cryst}})_{nn'} = \delta_{nn'} \exp(-E_n/\theta)/\sum_l \exp(-E_l/\theta). \quad (9)$$

The initial condition for (7) has the form

$$\rho(\mathbf{p}, \mathbf{p}'; n, n'; 0) = \rho_0(\mathbf{p}, \mathbf{p}')\delta_{nn'} \times \exp(-E_n/\theta)/\sum_l \exp(-E_l/\theta). \quad (10)$$

If the wave function of an incident electron coincides with a planar wave

$$\Psi_0(\mathbf{r}) = (2\pi\hbar)^{-3/2} \exp(i\mathbf{\Pi r}/\hbar),$$

the initial electron density matrix can be written as

$$\rho_0(\mathbf{p}, \mathbf{p}') = \int dr\, dr' \exp[i(\mathbf{p}'\mathbf{r}' - \mathbf{p}\mathbf{r})/\hbar]\Psi_0(\mathbf{r})\Psi_0^*(\mathbf{r}') = (2\pi\hbar)^3\delta(\mathbf{p}-\mathbf{\Pi})\delta(\mathbf{p}'-\mathbf{\Pi}). \quad (11)$$

For a further treatment it is suitable to split the potential (8) into two parts, namely the elastic scattering potential $U_0(\mathbf{r})$ and the inelastic scattering potential $\delta U(\mathbf{r})$,

$$U(\mathbf{r}) \equiv U_0(\mathbf{r}) + \delta U(\mathbf{r}). \quad (12)$$

The 'elastic' part of the scattering potential $U_0(\mathbf{r})$ describes all the processes without change of the crystal state. Electron diffraction is the principal of such processes. $U_0(\mathbf{r})$ has only diagonal matrix elements:

$$\langle n|U_0(\mathbf{q})|n'\rangle = \delta_{nn'}\sum_l \langle l|U_0(\mathbf{q})|l\rangle Z^{-1}\exp(-E_l/\theta) \quad (13)$$

$$Z = \sum_l \exp(-E_l/\theta).$$

All other processes like inelastic electron scattering are described by means of a 'fluctuating part of the potential'

$$\langle n|\delta U(\mathbf{q})|n'\rangle = \langle n|U(\mathbf{q}) - U_0(\mathbf{q})|n'\rangle.$$

Direct methods of summation over the excited states of the crystal have been developed by Afanas'ev & Kagan (1967). By making use of their results one can obtain

$$\langle n|U_0(\mathbf{q})|n'\rangle = \delta_{nn'}(2\pi)^3\sum_{\mathbf{K}} \Lambda(\mathbf{K})\delta(\mathbf{q}-\mathbf{K}), \quad (14)$$

where $\mathbf{K}$ is the reciprocal-lattice vector and

$$\Lambda(\mathbf{K}) = (4\pi e^2 N/K^2)\sum_\alpha [-Z_\alpha + f_\alpha(\mathbf{K})] \times \exp(-i\mathbf{K}\mathbf{r}_\alpha)\exp[-\tfrac{1}{2}M_\alpha(\mathbf{K})] \quad (15)$$

with $M_\alpha(\mathbf{q}) = \langle(\mathbf{q}\mathbf{u}_\alpha)^2\rangle$; $\exp[-\frac{1}{2}M_\alpha(\mathbf{q})]$ is the Debye–Waller factor, $f_\alpha(\mathbf{K})$ is the X-ray scattering amplitude for a single atom, and N is the number of unit cells per unit volume. After the summation over the crystal states in (7), using (15) we can write down the formula

$$\begin{aligned} i\hbar\,\partial\rho(\mathbf{p}, \mathbf{p}'; t)/\partial t &= (\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'})\rho(\mathbf{p}, \mathbf{p}'; t) \\ &+ \sum_{\mathbf{K}} \Lambda(\mathbf{K})[\rho(\mathbf{p}-\hbar\mathbf{K}, \mathbf{p}'; t) - \rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{K}; t)] \\ &+ \sum_{f,n}\int [d^3q/(2\pi)^3]\langle n|\delta U(\mathbf{q})|f\rangle \\ &\times \rho(\mathbf{p}-\hbar\mathbf{q}; \mathbf{p}'; f, n; t) \\ &- \sum_{f,n}\int [d^3q/(2\pi)^3]\rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{q}; n, f; t) \\ &\times \langle f|\delta U(\mathbf{q})|n\rangle. \end{aligned} \quad (16)$$

Let us consider the crystal state near thermodynamical equilibrium. In this case

$$\rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'; f, n; t) \simeq \rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'; t) \times \delta_{nf} Z^{-1}\exp(-E_n/\theta) \quad (17)$$

and the matrix elements containing δU in (16) tend to zero. To prove this one should use the equality

$$\sum_n \langle n|\delta U(\mathbf{q})|n\rangle Z^{-1}\exp(-E_n/\theta) = 0.$$

To calculate the right-hand side of (16) one needs to take into account the electron–crystal interaction and excitation of the crystal. In the first non-vanishing approximation with respect to δU we can write

$$\begin{aligned} i\hbar\,\partial\rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'; f, n; t)/\partial t &= (\varepsilon_{\mathbf{p}-\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}'} + E_f - E_n)\rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'; f, n; t) \\ &+ \sum_{\mathbf{K}} \Lambda(\mathbf{K})[\rho(\mathbf{p}-\hbar\mathbf{q}-\hbar\mathbf{K}; p'; f, n; t) \\ &- \rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'+\hbar\mathbf{K}; f, n; t)] \\ &+ \int [d^3k/(2\pi)^3]\langle f|\delta U(\mathbf{k})|n\rangle \\ &\times \rho(\mathbf{p}-\hbar\mathbf{q}-\hbar\mathbf{k}, \mathbf{p}'; t)Z^{-1}\exp(-E_n/\theta) \\ &- \int [d^3k/(2\pi)^3]\rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'+\hbar\mathbf{k}; t) \\ &\times \langle f|\delta U(\mathbf{k})|n\rangle Z^{-1}\exp(-E_f/\theta); \end{aligned} \quad (18)$$

$$\begin{aligned} i\hbar\,\partial\rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{q}; n, f; t)/\partial t &= (\varepsilon_p - \varepsilon_{\mathbf{p}'+\hbar\mathbf{q}} + E_n - E_f)\rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{q}; n, f; t) \\ &+ \sum_{\mathbf{K}} \Lambda(\mathbf{K})[\rho(\mathbf{p}-\hbar\mathbf{K}, \mathbf{p}'+\hbar\mathbf{q}; n, f; t) \\ &- \rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{K}+\hbar\mathbf{q}; n, f; t)] \\ &+ \int [d^3k/(2\pi)^3]\langle n|\delta U(\mathbf{k})|f\rangle \\ &\times \rho(\mathbf{p}-\hbar\mathbf{k}, \mathbf{p}'+\hbar\mathbf{q}; t)Z^{-1}\exp(-E_f/\theta) \\ &- \int [d^3k/(2\pi)^3]\rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{q}+\hbar\mathbf{k}; t) \\ &\times \langle n|\delta U(\mathbf{k})|f\rangle Z^{-1}\exp(-E_n/\theta). \end{aligned} \quad (19)$$

The effective values of q in (18) and (19) are comparable in order of magnitude with the reciprocal atomic

radius, $q \sim me^2 Z_a^{1/3}/\hbar^2$. This fact leads to the inequality $|\varepsilon_{\mathbf{p}+\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}}| \gg |\Lambda(\mathbf{K})|$. For this reason the contribution of the Fourier transforms of the regular potential $\Lambda(\mathbf{K})$ will be negligible. In other words, the effective linear dimension of the single-inelastic-scattering region is much smaller than the extinction distance $\hbar v_0/|\Lambda(\mathbf{K})|$. As was pointed out by Howie (1963), the region of small q, $q \lesssim |\Lambda(\mathbf{K})|/\hbar v_0$, contributes a negligible value to the total inelastic cross section. Taking into account this circumstance we can derive from (10), (18) and (19)

$$\begin{aligned}
&\rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'; f, n; t) \\
&\quad = -(i/\hbar)\int_0^t \mathrm{d}t \exp\left[-(i/\hbar)\right. \\
&\qquad \times (\varepsilon_{\mathbf{p}-\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}'} + E_f - E_n)(t-\tau)] \\
&\qquad \times \{\int [\mathrm{d}^3k/(2\pi)^3] \langle f|\delta U(\mathbf{k})|n\rangle \\
&\qquad \times \rho(\mathbf{p}-\hbar\mathbf{q}-\hbar\mathbf{k}, \mathbf{p}'; \tau) Z^{-1} \exp(-E_n/\theta) \\
&\qquad -\int [\mathrm{d}^3k/(2\pi)^3] \rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'+\hbar\mathbf{k}; \tau) \\
&\qquad \times \langle f|\delta U(\mathbf{k})|n\rangle Z^{-1} \exp(-E_f/\theta)\}. \qquad (20)
\end{aligned}$$

$$\begin{aligned}
&\rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{q}; n, f; t) \\
&\quad = -(i/\hbar)\int_0^t \mathrm{d}\tau \exp\left[-(i/\hbar)\right. \\
&\qquad \times (\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'+\hbar\mathbf{q}} + E_n - E_f)(t-\tau)] \\
&\qquad \times \{\int [\mathrm{d}^3k/(2\pi)^3] \langle n|\delta U(\mathbf{k})|f\rangle \\
&\qquad \times \rho(\mathbf{p}-\hbar\mathbf{k}, \mathbf{p}'+\hbar\mathbf{q}; \tau) Z^{-1} \exp(-E_f/\theta) \\
&\qquad -\int [\mathrm{d}^3k/(2\pi)^3] \rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{q}+\hbar\mathbf{k}; \tau) \\
&\qquad \times \langle n|\delta U(\mathbf{k})|f\rangle Z^{-1} \exp(-E_n/\theta)\}. \qquad (21)
\end{aligned}$$

Both the exponential factors in (21) and (22),

$$\exp\left[-(i/\hbar)(\varepsilon_{\mathbf{p}-\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}} + E_f - E_n)(t-\tau)\right]$$

and

$$\exp\left[-(i/\hbar)(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'+\hbar\mathbf{q}} + E_n - E_f)(t-\tau)\right]$$

are rapidly oscillating functions of τ. Hence we can change the variable τ into t in the argument of the density matrix ρ and let t tend to infinity in the upper limits of the integrals. Using the well known relation

$$\int_0^\infty \mathrm{d}\tau \exp(i\omega\tau) = \pi\delta(\omega) + \text{v.p.}\,(i/\omega), \qquad (22)$$

where v.p. denotes the principal value, we can easily transform (16) into the closed self-consistent kinetic equation describing the evolution of the single-particle density matrix,

$$\begin{aligned}
&\hbar\,\partial\rho(\mathbf{p}, \mathbf{p}'; t)/\partial t + i(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'})\rho(\mathbf{p}, \mathbf{p}'; t) \\
&\quad = -i\sum_{\mathbf{K}} \Lambda(\mathbf{K})\{\rho(\mathbf{p}-\hbar\mathbf{K}, \mathbf{p}'; t) - \rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{K}; t)\} \\
&\qquad + \pi\int [\mathrm{d}^3q/(2\pi)^3] \int \mathrm{d}\omega \sum_{\mathbf{K}} V(\mathbf{K}-\mathbf{q}) V(\mathbf{q}) \\
&\qquad \times [S(\mathbf{K}-\mathbf{q}; -\mathbf{q}; \omega)\delta(\varepsilon_{\mathbf{p}-\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}'} - \hbar\omega) \\
&\qquad \times \rho(\mathbf{p}-\hbar\mathbf{q}, \mathbf{p}'+\hbar\mathbf{K}-\hbar\mathbf{q}; t) \\
&\qquad - S(\mathbf{q}; \mathbf{q}-\mathbf{K}; \omega)\delta(\varepsilon_{\mathbf{p}-\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}'} + \hbar\omega) \\
&\qquad \times \rho(\mathbf{p}-\hbar\mathbf{K}; \mathbf{p}'; t)] \\
&\qquad + \pi\int [\mathrm{d}^3q/(2\pi)^3] \int \mathrm{d}\omega \sum_{\mathbf{K}} V(\mathbf{K}-\mathbf{q}) V(\mathbf{q}) \\
&\qquad \times [S(\mathbf{q}; \mathbf{q}-\mathbf{K}; \omega)\delta(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'+\hbar\mathbf{q}} + \hbar\omega) \\
&\qquad \times \rho(\mathbf{p}-\hbar\mathbf{K}+\hbar\mathbf{q}; \mathbf{p}'+\hbar\mathbf{q}; t) \\
&\qquad - S(\mathbf{K}-\mathbf{q}; -\mathbf{q}; \omega)\delta(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'+\hbar\mathbf{q}} - \hbar\omega) \\
&\qquad \times \rho(\mathbf{p}, \mathbf{p}'+\hbar\mathbf{K}; \omega)], \qquad (23)
\end{aligned}$$

where $S(\mathbf{q}; \mathbf{q}-\mathbf{K}; \omega)$ is the Fourier transform of the inelastic excitation structure factor:

$$s(\mathbf{q}; \mathbf{k}; \omega) = (2\pi)^3 \sum_{\mathbf{K}} S(\mathbf{q}; \mathbf{q}-\mathbf{K}, \omega)\delta(\mathbf{q}-\mathbf{k}-\mathbf{K}) \qquad (24)$$

and

$$\begin{aligned}
s(\mathbf{q}, \mathbf{k}, \omega) = [V(\mathbf{q})V(\mathbf{k})]^{-1} \sum_{l,n} Z^{-1} \exp(-E_l/\theta) \\
\times \langle l|\delta U(\mathbf{q})|n\rangle\langle n|\delta U(-\mathbf{k})|l\rangle \\
\times \delta[\omega + (E_l - E_n)/\hbar] \qquad (25)
\end{aligned}$$

where $V(\mathbf{k}) = 4\pi e^2/k^2$. In (23) we neglect all terms with principal values of integrals (Humphreys, 1979). These terms are smaller by an order of magnitude and go beyond the accuracy of experiments.

3. Inelastic excitation structure factor

During the practical calculation of the angular distribution of electrons transmitted through a single crystal one needs an analytical expression for the structure factor (25). This factor describes the set of inelastic excitations of a crystal. In the case of metals or semiconductors the structure factor (25) can be written as a sum of several terms describing various kinds of excitations, such as lattice vibrations, electron-hole excitations and plasmons (Okamoto, Ichinokawa & Ohtsuki, 1971):

$$s(\mathbf{q}; \mathbf{k}; \omega) = s_{\mathrm{ph}}(\mathbf{q}; \mathbf{k}; \omega) + s_{\mathrm{el}}(\mathbf{q}; \mathbf{k}; \omega) + s_{\mathrm{pl}}(\mathbf{q}; \mathbf{k}; \omega). \qquad (26)$$

Both the experimental results of Cundy, Howie & Valdre (1969) and the theoretical considerations of Platzman & Wolff (1973), Radi (1970) and Kawamura

& Shimamoto (1975) lead to the conclusion of a homogeneous space distribution of valence electrons in some crystals like Al and Si. In this case the plasmon excitation structure factor can be written in the form (Platzman & Wolff, 1973)

$$s_{\rm pl}(\mathbf{q};\mathbf{k};\omega) = (2\pi)^3\delta(\mathbf{q}-\mathbf{k})(N_0q^2/2m\omega_p)\delta(\omega-\omega_p)\eta(\mathbf{q}_c-\mathbf{q}) \quad (27)$$

where N_0 is the number of valence electrons per unit volume, $\hbar\omega_p$ is the plasmon excitation energy, $\mathbf{q}_c$ is the plasmon cut-off vector and $\eta(x)$ is the step function

$$\eta(x)=\begin{cases}1, & x\geq 0\\ 0, & x<0.\end{cases} \quad (28)$$

The electron-hole excitation factor was considered by Yoshioka (1957) and Afanas'ev & Kagan (1967). Assuming all the crystal electrons are in their ground states we can easily show that

$$s_{\rm el}(\mathbf{q};\mathbf{k};\omega) = N\sum_{n\neq 0}\sum_{\alpha}\exp[-i(\mathbf{q}-\mathbf{k})R_\alpha]\exp[-\tfrac{1}{2}M_\alpha(\mathbf{q}-\mathbf{k})] \times f_{0n}^{(\alpha)}(\mathbf{q})f_{n0}^{(\alpha)}(-\mathbf{k})\delta[\omega+(E_0-E_n)/\hbar] \quad (29)$$

where

$$f_{0n}^{(\alpha)}(\mathbf{q})=\langle 0|\sum_{b\alpha}\exp(-i\mathbf{q}\mathbf{r}_{b\alpha})|n\rangle$$

is the atomic electron excitation amplitude of the transition from the ground state $|0\rangle$ into the excited state $|n\rangle\neq|0\rangle$. The lattice-thermal-vibrations structure factor was considered by Hall & Hirsch (1965) and Afanas'ev & Kagan (1967). In the Einstein model for thermal motion (the thermal displacements of different atoms are assumed to be non-correlated) it is easy to derive the expression (Hall & Hirsch, 1965)

$$s_{\rm ph}(\mathbf{q};\mathbf{k};\omega) = N\delta(\omega)\sum_{\alpha}[-Z_\alpha+f_\alpha(\mathbf{q})][-Z_\alpha+f_\alpha(-\mathbf{k})] \times\exp[-i(\mathbf{q}-\mathbf{k})\mathbf{R}_\alpha]\{\exp[-\tfrac{1}{2}M_\alpha(\mathbf{q}-\mathbf{k})] -\exp[-\tfrac{1}{2}M_\alpha(\mathbf{q})-\tfrac{1}{2}M_\alpha(\mathbf{k})]\}. \quad (30)$$

(The phonon energy is equal in order of magnitude to the temperature $\theta\sim 0{\cdot}025$ eV and the thermal scattering can be considered as pure elastic.) Integrating the diagonal element of (23) over all momenta we find a total probability conservation law

$$\partial[\int d^3p\,\rho(\mathbf{p},\mathbf{p}';t)]/\partial t=0. \quad (31)$$

4. Elastic electron scattering

Let us consider an incident electron beam with a small angular width. In such a case the initial wave function coincides with a planar wave (11) and the scattered electron angular distribution consists of Bragg spots and a wide fan of inelastically scattered particles (Uyeda & Nonoyama, 1967, 1968). The solution of (23) with the initial condition (11) has the form

$$\rho(\mathbf{p},\mathbf{p}';t) = (2\pi\hbar)^3\sum_{\mathbf{G}_h;\mathbf{G}_l}\rho_{hl}(t)\delta(\mathbf{p}-\mathbf{\Pi}-\hbar\mathbf{G}_h) \times\delta(\mathbf{p}'-\mathbf{\Pi}-\hbar\mathbf{G}_l)+\rho_{\rm inel}(\mathbf{p},\mathbf{p}';t). \quad (32)$$

The summation over $\mathbf{G}_h$ and $\mathbf{G}_l$ in (32) is carried out over all the reciprocal-lattice vectors of the crystal. The density matrix $\rho_{\rm inel}(\mathbf{p},\mathbf{p}';t)$ describes the inelastically scattered electrons. From (11)

$$\rho_{hl}(0)=\delta_{h0}\delta_{l0}, \qquad \rho_{\rm inel}(\mathbf{p},\mathbf{p}';0)=0. \quad (33)$$

The diagonal elements of the density matrix $\rho_{hl}(t)$ are related to the Bragg spot intensity

$$I_h(T)=\rho_{hh}(T). \quad (34)$$

The equations (23) and (32) constitute a closed system for matrix elements $\rho_{hl}(t)$ which is equivalent to the equations of the usual many-beam dynamical theory in an absorbing crystal:

$$\hbar\,\partial\rho_{hl}/\partial t+i(\varepsilon_h-\varepsilon_l)\rho_{hl} = -i\sum_s[\Lambda_{hs}-(i/2)\gamma_{hs}]\rho_{sl}+i\sum_s\rho_{hs}[\Lambda_{sl}+(i/2)\gamma_{sl}] \quad (35)$$

where

$$\Lambda_{hs}=\Lambda(\mathbf{G}_h-\mathbf{G}_s),$$
$$\varepsilon_h=(\mathbf{\Pi}+\hbar\mathbf{G}_l)^2/2m,$$
$$\gamma_{hs}=\int[d^3k/(2\pi)^2]\int d\omega\; V(\mathbf{\Pi}/\hbar+\mathbf{G}_h-\mathbf{k}) \times V(\mathbf{\Pi}/\hbar+\mathbf{G}_s-\mathbf{k})S(\mathbf{\Pi}/\hbar+\mathbf{G}_h-\mathbf{k};\ \mathbf{\Pi}/\hbar+\mathbf{G}_s-\mathbf{k};\omega)\delta(\varepsilon_{\hbar\mathbf{k}}-\varepsilon_{\mathbf{\Pi}}+\hbar\omega).$$

As is easily seen from (35), the quantities Λ_{hs} and $-\frac{1}{2}\gamma_{hs}$ are the Hermitian and non-Hermitian parts of the effective potential for electron elastic scattering in a crystal (Whelan, 1965; Radi, 1970).

In the case of the two-beam approximation the system (35) turns into four equations:

$$\begin{aligned}
\hbar\,\partial\rho_{00}/\partial t &= -\gamma_{00}\rho_{00}-i\Lambda_{01}\rho_{10}+i\rho_{01}\Lambda_{10} -\gamma_{01}\rho_{10}/2-\rho_{01}\gamma_{10}/2\\
\hbar\,\partial\rho_{10}/\partial t+i(\varepsilon_1-\varepsilon_0)\rho_{10} &= -\gamma_{00}\rho_{10}+i\Lambda_{10}(\rho_{11}-\rho_{00})-\gamma_{10}(\rho_{11}+\rho_{00})/2\\
\hbar\,\partial\rho_{01}+i(\varepsilon_0-\varepsilon_1)\rho_{01} &= -\gamma_{00}\rho_{01}+i\Lambda_{01}(\rho_{00}-\rho_{11})-\gamma_{01}(\rho_{11}+\rho_{00})/2\\
\hbar\,\partial\rho_{11} &= -\gamma_{00}\rho_{11}+i\rho_{10}\Lambda_{01}-i\Lambda_{10}\rho_{01} -\gamma_{10}\rho_{01}/2-\rho_{10}\gamma_{01}/2.
\end{aligned} \quad (36)$$

The quantities ρ_{00} and ρ_{11} are the intensities of the transmitted and diffracted waves.

In the case of fast electrons $|\gamma_{10}| \ll |\Lambda_{10}|$ (Reimer, 1984) and we can derive a solution of (36) in the form

$$\begin{aligned}
\rho_{00}(t) &= [2(1+y^2)]^{-1} \exp(-\gamma_{00}t/\hbar) \\
&\times \{\cos[2|\Lambda_{10}|(1+y^2)^{1/2}t/\hbar] \\
&+ (1+2y^2)\cosh[|\gamma_{10}|\cos(\theta_0)t/(1+y^2)^{1/2}\hbar] \\
&- 2y(1+y^2)^{1/2} \\
&\times \sinh[|\gamma_{10}|\cos(\theta_0)t/(1+y^2)^{1/2}\hbar]\} \\
\rho_{11}(t) &= [2(1+y^2)]^{-1} \exp(-\gamma_{00}t/\hbar) \\
&\times \{\cosh[|\gamma_{10}|\cos(\theta_0)t/(1+y^2)^{1/2}\hbar] \\
&- \cos[2|\Lambda_{10}|(1+y^2)^{1/2}t/\hbar]\} \\
\rho_{10}(t) &= \rho_{01}^*(t) \\
&= -\tfrac{1}{2}\exp(i\theta_\Lambda)\exp(-\gamma_{00}t/\hbar) \\
&\times ((1+y^2)^{-1/2} \\
&\times \sinh[|\gamma_{10}|\cos(\theta_0)t/(1+y^2)^{1/2}\hbar] \\
&- [y/(1+y^2)] \\
&\times \{\cosh[|\gamma_{10}|\cos(\theta_0)t/(1+y^2)^{1/2}\hbar] \\
&- \cos[2|\Lambda_{10}|(1+y^2)^{1/2}t/\hbar]\}); \\
y &= (\varepsilon_0 - \varepsilon_1)/2|\Lambda_{10}| \\
\Lambda_{10} &= |\Lambda_{10}|\exp(i\theta_\Lambda) \\
\gamma_{10} &= |\gamma_{10}|\exp(i\theta_\gamma) \\
\theta_0 &= \theta_\Lambda - \theta_\gamma. \qquad (37)
\end{aligned}$$

These results coincide with the expressions found by Hashimoto, Howie & Whelan (1962) in the dynamical diffraction theory. If the initial electron momentum direction is far from any crystallographic axis, we find from (37) that

$$\rho_{hl}(t) = \delta_{h0}\delta_{l0}\exp(-\gamma_{00}t/\hbar). \qquad (38)$$

In the case of pure elastic scattering $\gamma_{lh} = 0$ and we can introduce a linear transformation

$$\rho_{jj'}(t) = \sum_{h,l}[c^+(\mathbf{\Pi})]_{jh}\rho_{hl}(t)c_{lj'}(\mathbf{\Pi}) \qquad (39)$$

where

$$\sum_{h,l}[c^+(\mathbf{\Pi})]_{jh}(\varepsilon_h\delta_{hl} + \Lambda_{hl})c_{lj'}(\mathbf{\Pi}) = \delta_{jj'}E_j \qquad (40)$$

and

$$(c^+)_{jh} = (c_{hj})^*.$$

The probability density $\sum_l \rho_{ll}$ does not depend on the wave-function basis, so (39) is the unitary transformation

$$\begin{aligned}
\sum_h [c^+(\mathbf{\Pi})]_{jh}c_{hj'}(\mathbf{\Pi}) &= \delta_{jj'} \\
\sum_j c_{hj}(\mathbf{\Pi})[c^+(\mathbf{\Pi})]_{jl} &= \delta_{hl}.
\end{aligned} \qquad (41)$$

After this transformation (35) can be written as

$$\begin{aligned}
&\hbar\,\partial\rho_{jj'}/\partial t + i(E_j - E_{j'})\rho_{jj'} \\
&\quad = -\tfrac{1}{2}\sum_{j''}\mu_{jj''}\rho_{j''j'} - \tfrac{1}{2}\sum_{j''}\rho_{jj''}\mu_{j''j'}
\end{aligned} \qquad (42)$$

where

$$\mu_{jj'} = \sum_{l,h}[c^+(\mathbf{\Pi})]_{jh}\gamma_{hl}c_{lj'}(\mathbf{\Pi}).$$

The initial condition for (42) has the form

$$\rho_{jj'}(0) = c_{0j}^*(\mathbf{\Pi})c_{0j'}(\mathbf{\Pi}). \qquad (43)$$

In the case of pure elastic scattering

$$\rho_{jj'}(t) = c_{0j}^*(\mathbf{\Pi})c_{0j'}(\mathbf{\Pi})\exp[-i(E_j - E_{j'})t/\hbar] \qquad (44)$$

and for Bragg-spot intensities one finds directly

$$\begin{aligned}
\rho_{hh}(T) &= \sum_{j,j'} c_{hj}(\mathbf{\Pi})c_{0j}^*(\mathbf{\Pi})c_{0j'}(\mathbf{\Pi})c_{hj'}^*(\mathbf{\Pi}) \\
&\times \exp[-i(E_j - E_{j'})T/\hbar].
\end{aligned} \qquad (45)$$

The existence of inelastic scattering leads to a decrease in the total intensity of the Bragg spots

$$\begin{aligned}
\hbar\,\partial\sum_h \rho_{hh}/\partial t = \hbar\,\partial\sum_j \rho_{jj}/\partial t &= -\sum_{h,l}\gamma_{lh}\rho_{hl} \\
&= -\sum_{j,j'}\mu_{jj'}\rho_{j'j} < 0.
\end{aligned} \qquad (46)$$

In this case the diagonalization of (35) by a unitary transformation is impossible and we need to use some approximate methods to find a solution of (42) (Reimer, 1984).

5. Single inelastic scattering approximation

Let us consider the case when the initial beam direction does not coincide with any of the crystallographic axes and the crystal slab is thin enough, $T \ll \hbar/\gamma_{00}$. In this case (32) can be solved by means of perturbation theory:

$$\rho(\mathbf{p}, \mathbf{p}'; t) = (2\pi\hbar)^3\delta(\mathbf{p} - \mathbf{\Pi})\delta(\mathbf{p} - \mathbf{p}') + \rho^{(1)}(\mathbf{p}, \mathbf{p}'; t) \qquad (47)$$

where

$$\int \mathrm{d}^3p\,\rho^{(1)}(\mathbf{p}, \mathbf{p}; t) \ll 1.$$

Substitution of (47) into (23) gives a general form of the solution of first order

$$\rho^{(1)}(\mathbf{p}, \mathbf{p}'; t) = (2\pi\hbar)^3\sum_h \varphi_{0h}(\mathbf{p}, t)\delta(\mathbf{p}' - \mathbf{p} - \hbar\mathbf{G}_h). \qquad (48)$$

The functions $\varphi_{0h}(\mathbf{p}, t)$ obey an infinite set of inhomogeneous differential equations

$$\begin{aligned}
&\hbar\,\partial\varphi_{hl}/\partial t + i[\varepsilon_h(\mathbf{p}) - \varepsilon_l(\mathbf{p})]\varphi_{hl} \\
&\quad = -i\sum_s(\Lambda_{hs}\varphi_{sl} - \varphi_{hs}\Lambda_{sl}) + \mathscr{S}_{hl}(\mathbf{p})
\end{aligned} \qquad (49)$$

with the initial condition

$$\varphi_{hl}(\mathbf{p}, 0) = 0, \tag{50}$$

where we have introduced the definitions $\varphi_{lh}(\mathbf{p}, t) \equiv \varphi_{0s}(\mathbf{p}+\hbar\mathbf{G}_l; t)$ if $\mathbf{G}_l + \mathbf{G}_s = \mathbf{G}_h$ and

$$\begin{aligned}\mathcal{S}_{hl}(\mathbf{p}) = [1/2(2\pi)^2] \int d\omega \\ \times V[(\mathbf{\Pi}-\mathbf{p})/\hbar - \mathbf{G}_l] V[(\mathbf{p}-\mathbf{\Pi})/\hbar + \mathbf{G}_h] \\ \times S[(\mathbf{\Pi}-\mathbf{p})/\hbar - \mathbf{G}_l; (\mathbf{p}-\mathbf{\Pi})/\hbar + \mathbf{G}_h; \omega] \\ \times [\delta(\varepsilon_{\mathbf{\Pi}} - \varepsilon_{\mathbf{p}+\hbar\mathbf{G}_l} - \hbar\omega) \\ + \delta(\varepsilon_{\mathbf{\Pi}} - \varepsilon_{\mathbf{p}+\hbar\mathbf{G}_h} - \hbar\omega)].\end{aligned} \tag{51}$$

The function $\mathcal{S}_{hl}(\mathbf{p})$ arises from the substitution of the zero-order approximation (47) into (23). In accordance with (4) the diagonal element $\varphi_{00}(\mathbf{p}, T)$ gives the inelastically scattered electron momentum distribution. To solve (49) we can transform a set of equations for elements $\varphi_{hl}(\mathbf{p}, t)$ as in (39),

$$\varphi_{jj'}(t) = \sum_{h,l} [c^{+}(\mathbf{p})]_{jh} \varphi_{hl}(\mathbf{p}, t) c_{lj'}(\mathbf{p}). \tag{52}$$

From (49) and (52) we obtain

$$\hbar\, \partial\varphi_{jj'}/\partial t + i[E_j(\mathbf{p}) - E_{j'}(\mathbf{p})]\varphi_{jj'} = \mathcal{S}_{jj'}(\mathbf{p}). \tag{53}$$

The solution of (53) has the form

$$\begin{aligned}\varphi_{jj'}(T) = (1/\hbar)\mathcal{S}_{jj'}(\mathbf{p}) \int_0^T d\tau \\ \times \exp\{-i[E_j(\mathbf{p}) - E_{j'}(\mathbf{p})](T-\tau)/\hbar\}.\end{aligned} \tag{54}$$

In the limiting case $T \gg \hbar/|E_j - E_{j'}|$ we can neglect the rapidly oscillating terms in (54) and find

$$\varphi_{jj'}(\mathbf{p}, T) = (T/\hbar)\delta_{jj'}\mathcal{S}_{jj'}(\mathbf{p}). \tag{55}$$

This quantity is connected with the inelastically scattered electron distribution $\varphi_{00}(\mathbf{p}, T)$ by means of unitary transformation (52). Using the inverse transformation we obtain

$$\varphi_{00}(\mathbf{p}, T) = (T/\hbar) \sum_j \sum_{h,l} |c_{0j}|^2 c^*_{hj} c_{lj} \mathcal{S}_{hl}(\mathbf{p}). \tag{56}$$

The distribution (56) is valid in the general case of many-beam dynamical diffraction. In the particular two-beam case we can use the well known expression for c_{hj} (Reimer, 1984, p. 288) and write down analytically the intensity distribution in the Kikuchi patterns:

$$\begin{aligned}\varphi_{00}(\mathbf{p}, T) = (T/\hbar)\{\mathcal{S}_{00}(\mathbf{p}) \\ + [1/2(1+y^2)][\mathcal{S}_{11}(\mathbf{p}) - \mathcal{S}_{00}(\mathbf{p})] \\ - [y/2(1+y^2)][\mathcal{S}_{01}(\mathbf{p}) + \mathcal{S}_{10}(\mathbf{p})]\}\end{aligned} \tag{57}$$

$$\begin{aligned}\varphi_{00}(\mathbf{p}+\hbar\mathbf{G}, T) = (T/\hbar)\{\mathcal{S}_{11}(\mathbf{p}) \\ + [1/2(1+y^2)][\mathcal{S}_{00}(\mathbf{p}) - \mathcal{S}_{11}(\mathbf{p})] \\ + [y/2(1+y^2)][\mathcal{S}_{01}(\mathbf{p}) + \mathcal{S}_{10}(\mathbf{p})]\}.\end{aligned} \tag{58}$$

These expressions coincide with the results of Kainuma (1955), Fujimoto & Kainuma (1963), Okamoto, Ichinokawa & Ohtsuki (1971) and Ohtsuki (1983). One should note that the observed small oscillations of the inelastically scattered electron fan are related to some small terms neglected during the transformation from (54) to (55). The solution (54) is valid only in the case of a thin crystal with $T \ll \hbar/\gamma_{00}$. If the thickness is larger than the mean free path with respect to inelastic collisions ($L > \hbar v_0/\gamma_{00}$) we need to take multiple inelastic scattering of electrons into account.

6. Kikuchi lines and bands in the thick crystal

As is well known the main contribution to fast-electron angular straggling in a crystal is due to thermal diffuse scattering. The scattering by plasmon or electron-hole excitations leads to energy losses but practically does not change the velocity direction. The diffraction-contrast conservation by plasmon and electron-hole inelastic scattering proves this fact in an experimental way (Cundy, Howie & Valdre, 1969). In the remaining discussion we shall not deal with the energy distribution of scattered electrons. Keeping in mind the following integration over the absolute value of p at a fixed observation direction we neglect below plasmon and electron-hole scattering. We thus use for the structure factor $S(\mathbf{q}, \mathbf{k}; \omega)$ and for the non-Hermitian part of the regular potential the phonon parts given by Hall & Hirsch (1965). To find a general form of the solution (23) let us consider the solution of (23) in a particular case without diffraction, *i.e.* without taking into account all terms with $\mathbf{K} \neq 0$. In this case the solution of (23) with the initial condition (11) has the form

$$\rho(\mathbf{p}, \mathbf{p}'; t) = (2\pi\hbar)^3 \delta(\mathbf{p}-\mathbf{p}')\, W_0(\mathbf{p}, t) \tag{59}$$

where $W_0(\mathbf{p}, t)$ satisfies the transport equation

$$\begin{aligned}\partial W_0(\mathbf{p}, t)/\partial t = \int [d^3q/(2\pi)^2]\sigma(\mathbf{q})\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}}) \\ \times W_0(\mathbf{p}+\hbar\mathbf{q}) - (\gamma_{00}/\hbar)\, W_0(\mathbf{p}, t)\end{aligned} \tag{60}$$

and

$$\begin{aligned}\sigma(\mathbf{q}) = (1/\hbar)(4\pi e^2/q^2)^2 N \\ \times \sum_{\alpha} |-Z_\alpha + f_\alpha(\mathbf{q})|^2 \{1 - \exp[-M_\alpha(\mathbf{q})]\}.\end{aligned}$$

The initial condition for (60) is

$$W_0(\mathbf{p}, 0) = \delta(\mathbf{p}-\mathbf{\Pi}). \tag{61}$$

The solution of (60) with the condition (61) is called the Moliere distribution (Moliere, 1948). In a thick slab $T \gg \hbar/\gamma_{00}$ this distribution has the analytical form (Mott & Massey, 1965)

$$\begin{aligned}W_0(\mathbf{p}, t) = (1/mp)\delta(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{\Pi}}) \\ \times [\pi\vartheta_0^2 x \ln(4x)]^{-1} \exp[-\vartheta^2/\vartheta_0^2 x \ln(4x)]\end{aligned} \tag{62}$$

where

$$x = \gamma_{00} t/\hbar \gg 1;\ \mathbf{p}\boldsymbol{\Pi} = p\Pi \cos\vartheta;$$

$\vartheta_0^2 = m^2 e^2 Z^{2/3}/\hbar^2 p^2$ is the mean square angle of electron scattering by the thermal fluctuation of a single-atom potential.

Taking into account the diffraction, *i.e.* the terms with $\mathbf{K} \neq 0$, one can find a quite general form for the solution of (23):

$$\rho(\mathbf{p}, \mathbf{p}'; t) = (2\pi\hbar)^3 \delta(\mathbf{p}-\mathbf{p}')\, W_0(\mathbf{p}, t) + (2\pi\hbar)^3 \sum_h \phi_{0h}(\mathbf{p}, t)\, \delta(\mathbf{p}'-\mathbf{p}-\hbar\mathbf{G}_h). \tag{63}$$

Substitution of (23) into (63) yields

$$\begin{aligned}
&\hbar\, \partial\phi_{hl}(\mathbf{p}, t)/\partial t + i[\varepsilon_h(\mathbf{p}) - \varepsilon_l(\mathbf{p})]\phi_{hl}(\mathbf{p}; t) \\
&\quad = -i \sum_s [\Lambda_{hs} - (i/2)\gamma_{hs}]\phi_{sl}(\mathbf{p}; t) \\
&\qquad + i \sum_s \phi_{hs}(\mathbf{p}, t)[\Lambda_{sl} + (i/2)\gamma_{sl}] \\
&\qquad + \pi \int [\mathrm{d}^3 q/(2\pi)^3] \sum_s V(\mathbf{G}_s - \mathbf{G}_l + \mathbf{q})\, V(\mathbf{q}) \\
&\qquad \times S(\mathbf{G}_s - \mathbf{G}_l + \mathbf{q}; \mathbf{q}) \\
&\qquad \times \delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{G}_h+\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}+\hbar\mathbf{G}_l})\phi_{hs}(\mathbf{p}+\hbar\mathbf{q}; t) \\
&\qquad + \pi \int [\mathrm{d}^3 q/(2\pi)^3] \sum_s V(\mathbf{q})\, V(\mathbf{G}_h - \mathbf{G}_s - \mathbf{q}) \\
&\qquad \times S(\mathbf{q}; \mathbf{q} - \mathbf{G}_h - \mathbf{G}_s) \\
&\qquad \times \delta(\varepsilon_{p+\hbar\mathbf{G}_h} - \varepsilon_{\mathbf{p}+\hbar\mathbf{q}+\hbar\mathbf{G}_l})\phi_{sl}(\mathbf{p}+\hbar\mathbf{q}; t) \\
&\qquad + i(1-\delta_{hl})\{W_0(\mathbf{p}+\hbar\mathbf{G}_h; t) \\
&\qquad \times [\Lambda_{hl} + (i/2)\gamma_{hl}] - W_0(\mathbf{p}+\hbar\mathbf{G}_l; t) \\
&\qquad \times [\Lambda_{hl} - (i/2)\gamma_{hl}]\} \\
&\qquad + \pi(1-\delta_{hl}) \int [\mathrm{d}^3 q/(2\pi)^3] V(\mathbf{G}_h - \mathbf{G}_l + \mathbf{q})\, V(\mathbf{q}) \\
&\qquad \times S(\mathbf{G}_h - \mathbf{G}_l + \mathbf{q}; \mathbf{q}) \\
&\qquad \times \delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{G}_h+\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}+\hbar\mathbf{G}_l})\, W_p(\mathbf{p}+\hbar\mathbf{G}_h+\hbar\mathbf{q}; t) \\
&\qquad + \pi(1-\delta_{hl}) \int [\mathrm{d}^3 q/(2\pi)^3] V(\mathbf{q})\, V(\mathbf{G}_h - \mathbf{G}_l - \mathbf{q}) \\
&\qquad \times S(\mathbf{q}; \mathbf{q} + \mathbf{G}_l - \mathbf{G}_h) \\
&\qquad \times \delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{G}_l+\hbar\mathbf{q}} - \varepsilon_{\mathbf{p}+\hbar\mathbf{G}_h})\, W_0(\mathbf{p}+\hbar\mathbf{G}_l+\hbar\mathbf{q}; t).
\end{aligned} \tag{64}$$

The initial condition for (64), as for (49), has the form

$$\phi_{hl}(\mathbf{p}; 0) = 0. \tag{65}$$

For $h \neq l$ the function $\phi_{hl}(\mathbf{p}, t)$ describes the mutual coherence properties of electron states with momenta $\mathbf{p}+\hbar\mathbf{G}_h$ and $\mathbf{p}+\hbar\mathbf{G}_l$ (Blum, 1981).

As is easy to see from (37), for fixed $\mathbf{p}$, the value $\phi_{hl}(\mathbf{p}, t)$ depends on the difference $\varepsilon_l - \varepsilon_h$ and decreases rapidly with increase in $|\varepsilon_l - \varepsilon_h|$. Actually, each diffracted wave has a rather high intensity only when the electron velocity direction lies within a fixed angular region. In the two-beam case this angular region is determined by the condition $|y| \lesssim 1$. In the general case the angular widths of these 'coherence regions' can be estimated as (Fujimoto, Takagi, Komaki, Koike & Uchida, 1972)

$$\Delta\vartheta \sim 4m|\Lambda_{0h}|/\hbar p G_h \quad \text{for } \hbar^2 G_h^2/2m|\Lambda_{0h}| \gg 1 \tag{66}$$

$$\Delta\vartheta \sim (8m|\Lambda_{10}|)^{1/2}/p \quad \text{for } \hbar^2 G_h^2/2m|\Lambda_{10}| \ll 1. \tag{67}$$

These inequalities correspond to the opposite cases of weak and strong coupling in the electron transverse motion. In other words, using the expressions (66) and (67) we can say that the off-diagonal matrix elements $\phi_{hl}(\mathbf{p}, t)$ [and also the diagonal element $\phi_{00}(\mathbf{p}, t)$] have non-vanishing values only inside some angular region defined by a relation

$$\Delta\vartheta \sim \min\,[4m|\Lambda_{0h}|/\hbar p G_h;\ (8m|\Lambda_{10}|)^{1/2}/p]. \tag{68}$$

In the particular case, discussed in the Appendix, where

$$\Delta\vartheta \ll me^2 Z^{1/3}/\hbar^2 p, \qquad \gamma_{00} t/\hbar \gg 1, \tag{69}$$

(64) yields

$$\begin{aligned}
&\hbar\, \partial\phi_{hl}(\mathbf{p}, t)/\partial t + i[\varepsilon_h(\mathbf{p}) - \varepsilon_l(\mathbf{p})]\phi_{hl}(\mathbf{p}, t) \\
&\quad = -i \sum_s [\Lambda_{hs} - (i/2)\gamma_{hs}]\phi_{sl} \\
&\qquad + i \sum_s \phi_{hs}[\Lambda_{sl} + (i/2)\gamma_{sl}] \\
&\qquad + i\Lambda_{hl}[W_0(\mathbf{p}+\hbar\mathbf{G}_h; t) - W_0(\mathbf{p}+\hbar\mathbf{G}_l; t)] \\
&\qquad + (1-\delta_{hl})(\gamma_{hl}/\gamma_{00})\hbar\, \partial W_0(\mathbf{p}, t)/\partial t.
\end{aligned} \tag{70}$$

As usual the fast-electron momentum distribution is related to the diagonal matrix element (63) $W_0(\mathbf{p}, T) + \phi_{00}(\mathbf{p}, T)$ where $W_0(\mathbf{p}, T)$ is the inelastically scattered electron fan. All the Bragg-scattering effects are described by $\phi_{00}(\mathbf{p}, T)$. For this reason we have to define a Kikuchi pattern contrast by the relation

$$\mathcal{K}(\mathbf{p}, T) = \phi_{00}(\mathbf{p}, T)/W_0(\mathbf{p}, T). \tag{71}$$

To solve (70) it is useful to transform this equation, like (39) and (52), into the Bloch-state representation

$$\begin{aligned}
&\hbar\, \partial\phi_{jj'}/\partial t + i(E_j - E_{j'})\phi_{jj'} \\
&\quad = -\tfrac{1}{2} \sum_{j''} (\mu_{jj''}\phi_{j''j'} + \phi_{jj''}\mu_{j''j'}) \\
&\qquad + iM_{jj'}(t) + (\mu_{jj'}/\gamma_{00} - \delta_{jj'})\hbar\, \partial W_0(\mathbf{p}, t)/\partial t,
\end{aligned} \tag{72}$$

where the quantities $\mu_{jj'}$ were defined in (42) and $M_{jj'}(t)$ has the form

$$M_{jj'} = \sum_{h,l} [c^+(\mathbf{p})]_{jh}\Lambda_{hl} \times [W_0(\mathbf{p}+\hbar\mathbf{G}_h; t) - W_0(\mathbf{p}+\hbar\mathbf{G}_l; t)]c_{lj'}(\mathbf{p}).$$

As is clear from (73), the matrix M is anti-Hermitian and its diagonal elements vanish. If we avoid the critical-voltage effect, when E_j coincides with $E_{j'}$ (Humphreys, 1979; Reimer, 1984), we can solve (72) by means of perturbation theory with the small parameters

$$\nu_{jj'} = |\mu_{jj'}/(E_j - E_{j'})| \ll 1, \quad j \neq j'. \tag{73}$$

Using (69) we can neglect all the time derivatives in (72) and obtain analytical expressions for the diagonal and off-diagonal elements,

$$\begin{aligned}\phi_{jj} = (1/\gamma_{00})(1 - \gamma_{00}/\mu_{jj})\hbar\,\partial W_0(\mathbf{p}, t)/\partial t \\ -\tfrac{1}{2}\sum_{j\neq j'}\{[\mu_{jj'}M_{j'j}/\mu_{jj}(E_{j'} - E_j)] \\ +[M_{jj'}\mu_{j'j}/\mu_{jj}(E_j - E_{j'})]\}\end{aligned} \tag{74}$$

and

$$\phi_{jj'} = M_{jj'}/(E_j - E_{j'}). \tag{75}$$

Consequently the Kikuchi pattern contrast (71) has the form

$$\mathcal{K}(\mathbf{p}, T) = \sum_{j,j'} c_{0j}(\mathbf{p})\phi_{jj'}(\mathbf{p}, T)c^*_{0j'}(\mathbf{p})/W_0(\mathbf{p}, T) \tag{76}$$

where we have taken into account (74) and (75). This expression gives the intensity distribution in the Kikuchi pattern for the case of a thick crystal (*i.e.* $\gamma_{00}T/\hbar \gg 1$). Using (76) we can give a very simple and clear interpretation for a number of experimental observations of the Kikuchi pattern contrast effects in thick crystalline foils (Uyeda & Nonoyama, 1967, 1968; Nakai, 1970; Komuro, Kojima & Ichinokawa, 1972). Actually, if $W_0(\mathbf{p}+\hbar\mathbf{G}_h, t) \simeq W_0(\mathbf{p}+\hbar\mathbf{G}_l; t)$, the diagonal elements of (74) define the differences between the Bloch-state occupation numbers with the same value of quasi-momentum $\mathbf{p}$. In the first-order approximation (63) (without the second term on the right-hand side), the Bloch-state occupation probability does not depend on j and is equal to $W_0(\mathbf{p}, t)$. In the solid angular region where $\partial W_0(\mathbf{p}, t)/\partial t > 0$ (the edges of the image) all Bloch states with $\mu_{jj} > \gamma_{00}$ (strongly absorbed states) are fully occupied. In another regular angular region in the central part of the picture where $\partial W_0(\mathbf{p}, t)/\partial t < 0$ other states with $\mu_{jj} < \gamma_{00}$ (weakly absorbed states) are occupied preferentially. Such a distribution over the occupation numbers, caused by multiple scattering of the average fan of electrons, can be described by the transport equation (60). This equation shows what happens in an element of phase space in the presence of scattering. The first term on the right-hand side describes the scattering from all other possible electron momentum states into the state being considered. The second term is an 'absorption' term which accounts for the scattering out of the momentum state considered into all other states. The initial condition (61) leads to an intensity distribution of Gaussian type with a sharp maximum in the central part of the image. To find a derivative in (72) one must use the analytical expression (62)

$$\partial W_0(\mathbf{p}, t)/\partial t \sim W_0(\mathbf{p}, t)(1/x)[\vartheta^2/\vartheta_0^2 x \ln(4x) - 1]. \tag{77}$$

In the small-deflection-angle region $\vartheta^2 < \vartheta_0^2 x \ln(4x)$, $\partial W_0/\partial t < 0$ and the average fan decreases. In outer parts of the image $\partial W_0(\mathbf{p}, t)/\partial t > 0$ and the scattered electron intensity increases.

A Bloch wave function of an electron in a single crystal has a nonhomogeneous space density distribution $|\psi(\mathbf{r})|^2$. Consequently the different Bloch states have different phonon scattering probabilities. Owing to this circumstance, in the central part of the picture the absorption rate of the states localized on atomic planes is higher than the absorption rate of the average fan. By contrast, in the outer part of the image the occupation numbers of strongly absorbed states turn out to be somewhat larger than the average level and so on. The Kikuchi band reversed-contrast effect confirms this quantitative picture. As follows from much experimental data the reversed-contrast region increases with the crystal thickness. From (77) it is easy to find the angular width of the reversed-contrast region:

$$\vartheta_r \sim \vartheta_0[(\gamma_{00}t/\hbar)\ln(4\gamma_{00}t/\hbar)]^{1/2}. \tag{78}$$

The absolute value of the Kikuchi-band contrast decreases with the crystal thickness

$$\mathcal{K}(\mathbf{p}, T) \sim \mathcal{K}_0(\mathbf{p})/(\gamma_{00}T/\hbar) \tag{79}$$

where

$$|\mathcal{K}_0(\mathbf{p})| \sim 1.$$

Assuming the level of contrast resolution is equal to 0·1 (Ichimiya, 1973) one can easily obtain the maximum crystal thickness which permits Kikuchi pattern detection:

$$L_{\max} = v_0 T_{\max} \sim 10 v_0 \hbar/\gamma_{00}. \tag{80}$$

The value of $L_{\max}$ and its velocity dependence are in good agreement with the experimental results reported by Uyeda & Nonoyama (1967, 1968).

Thus, the Kikuchi bands in thick crystals arise from the anomalous transmission and anomalous absorption of inelastically scattered electrons. By contrast, the Kikuchi lines arise as a result of a simple diffraction transfer of a fan from the high-intensity regions to the regions of low intensity.

In the single inelastic scattering model (Kainuma, 1955; Ohtsuki, 1983) the Kikuchi line is described by the second term on the right-hand side of (57) and (58). In the thick-crystal case the Kikuchi lines are described by the matrix elements of $M_{jj'}$. Using the two-beam approximation one can obtain an analytical expression for the contrast distribution in the

pair of lines

$$\phi_{00}(\mathbf{p}, T) = \frac{-\frac{1}{2}\{[1-(\gamma_{l0}/\gamma_{00})^2]-y\gamma_{l0}/\gamma_{00}\}}{\{y^2+[1-(\gamma_{l0}/\gamma_{00})^2]\}} \times [W_0(\mathbf{p}, T) - W_0(\mathbf{p}+\hbar\mathbf{G}_l; T)] \quad (81)$$

$$\phi_{00}(\mathbf{p}+\hbar\mathbf{G}_l; T) = \frac{\frac{1}{2}\{[1-(\gamma_{l0}/\gamma_{00})^2]+y\gamma_{l0}/\gamma_{00}\}}{\{y^2+[1-(\gamma_{l0}/\gamma_{00})^2]\}} \times [W_0(\mathbf{p}; T) - W_0(\mathbf{p}+\hbar\mathbf{G}_l; T)] \quad (82)$$

where both Λ_{l0} and γ_{l0} are assumed to be real. In the limiting case $\gamma_{l0}=0$ (81) and (82) coincide with (57) and (58).

The above expressions, (82) and (81), show the asymmetry of the lines with regard to the exact Bragg reflection condition $y=0$. The angular width of the lines is

$$|y_{1/2}| \sim (1-\gamma_{l0}^2/\gamma_{00}^2)^{1/2} < 1.$$

The effect of the angular width decrease in thick crystals could reach about 10%. This effect takes place for the Kikuchi band also. In the two-beam approximation the Kikuchi-band intensity distribution has the form

$$\phi_{00}(\mathbf{p}; T) = -\frac{(\gamma_{l0}/\gamma_{00})^2+y\gamma_{l0}/\gamma_{00}}{y^2+[1-(\gamma_{l0}/\gamma_{00})^2]} \frac{\hbar}{\gamma_{00}} \frac{\partial}{\partial t} W_0(\mathbf{p}, t)\Big|_{t=T}$$

$$\phi_{00}(\mathbf{p}+\hbar\mathbf{G}_l; T) = -\frac{(\gamma_{l0}/\gamma_{00})^2-y\gamma_{l0}/\gamma_{00}}{y^2+[1-(\gamma_{l0}/\gamma_{00})^2]} \frac{\hbar}{\gamma_{00}} \frac{\partial}{\partial t} W_0(\mathbf{p}, t)\Big|_{t=T}.$$

We ought to note that the effect of the angular width decrease does not enable one to examine the validity of the two-beam approximation in thick crystals by a simple comparison of the line width with the Bragg angle.

APPENDIX

When the inequality $\Delta\vartheta \ll me^2Z^{1/3}/\hbar^2p$ is satisfied, the angular width of the Kikuchi lines and bands is much less than the deflection angle of the single scattering of an electron by an atom. Consequently, a single inelastic collision leads to scattering into momentum states satisfying the relation $|\varepsilon_h(\mathbf{p}) - \varepsilon_l(\mathbf{p})| \gg |\Lambda_{hl}|$. Owing to this circumstance in (64) we can neglect all integral terms including $\phi_{lh}(\mathbf{p}, t)$.

The integral terms including $W_0(\mathbf{p}, t)$ can be estimated from the closed expression for the mean square deflection angle (Mott & Massey, 1965)

$$\langle\vartheta_t^2\rangle = (\gamma_{00}t/\hbar)\ln(4\gamma_{00}t/\hbar)(me^2Z^{1/3}/\hbar^2p)^2.$$

Using the theorem of the mean one can obtain

$$\begin{aligned}&\int[d^3q/(2\pi)^2]V(\mathbf{G}_h-\mathbf{G}_l-\mathbf{q})V(\mathbf{q})\\ &\times S(\mathbf{G}_h-\mathbf{G}_l+\mathbf{q}, \mathbf{q})\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{G}_h+\hbar\mathbf{q}}-\varepsilon_{\mathbf{p}+\hbar\mathbf{G}_l})\\ &\times W_0(\mathbf{p}+\hbar\mathbf{G}_h+\hbar\mathbf{q}; t)\\ &-(\gamma_{lh}/\gamma_{00})\int[d^3q/(2\pi)^2]|V(\mathbf{q})|^2S(\mathbf{q}, \mathbf{q})\\ &\times\delta(\varepsilon_{\mathbf{p}+\hbar\mathbf{q}}-\varepsilon_{\mathbf{p}})W_0(\mathbf{p}+\hbar\mathbf{q}, t)\\ &=\gamma_{lh}[W_0(\mathbf{p}', t)-W_0(\mathbf{p}'', t)], \qquad (A1)\\ &|\mathbf{p}-'\mathbf{p}''| < \hbar^2\kappa(\kappa+|\mathbf{G}_h|)/\langle\vartheta_t^2\rangle p,\\ &\kappa = me^2Z^{1/3}/\hbar^2.\end{aligned}$$

Consequently, for strong first-order reflections the difference (A1) can be estimated as

$$|\gamma_{lh}|W_0(\mathbf{p}, t)(\hbar\kappa/p)^3(\langle\vartheta_t^2\rangle)^{-3/2} \ll \hbar|\partial W_0(\mathbf{p}, t)/\partial t|.$$

This expression enables us to derive equation (70).

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Image Processing in High-Resolution Electron Microscopy Using the Direct Method. III. Structure-Factor Extrapolation

BY LIU YI-WEI, FAN HAI-FU AND ZHENG CHAO-DE

Institute of Physics, Academia Sinica, Beijing, China

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Abstract

A resolution-enhancement method has been proposed which makes use of the Sayre equation [Sayre (1952). *Acta Cryst.* **5**, 60-65] to extrapolate both phases and magnitudes of structure factors. The starting point of the procedure is just a single deconvoluted electron microscopic image. No preliminary knowledge other than the chemical composition of the sample is necessary. A simulation on a theoretical image of copper perchlorophthalocyanine shows that the image resolution can be enhanced from 2 to 1 Å, resolving clearly individual atoms.

Introduction

Enhancement of the resolution of electron microscopic images by *a posteriori* processing techniques has long been attempted (Li Fang-hua, 1977; Ishizuka, Miyazaki & Uyeda, 1982; Fan Hai-fu, Zhong Zi-yang, Zheng Chao-de & Li Fang-hua, 1985). All the methods mentioned above rely on an additional electron diffraction pattern, which contains reflections corresponding to a higher resolution. Improvement in resolution can then be achieved simply by a phase extension procedure. However, without using the electron diffraction pattern, resolution enhancement is still possible. In X-ray crystallography, Fan Hai-fu & Zheng Qi-tai (1975) proposed a method using the Sayre equation (Sayre, 1952) to extrapolate both phases and magnitudes of structure factors. With this method a low-resolution image of a crystal structure can be enhanced to obtain a high-resolution picture without the necessity of collecting additional diffraction data in high-angle regions of reciprocal space. In this paper, the method has been improved and applied to high-resolution electron microscopy.

The philosophy of the method is as follows: For a crystalline sample, suppose that there are N atoms in the asymmetric unit; then, in order to reveal the structure with sufficiently high resolution, we only have to solve the $2N$ positional parameters (in the two-dimensional case). Now if we have in hand a low-resolution image of the crystal structure, which in reciprocal space can yield more than $2N$ symmetrically independent structure factors wihin its resolution limit, then in principle we can set up enough simultaneous equations to solve the $2N$ parameters. This implies that high-resolution structural information may be extracted from a low-resolution image. The Sayre equation (1) may be used for this purpose.

$$\mathbf{F}_{\mathbf{H}} = (\theta/V)\sum_{\mathbf{H}'} \mathbf{F}_{\mathbf{H}'}\mathbf{F}_{\mathbf{H}-\mathbf{H}'}. \qquad (1)$$

Structure factors beyond the resolution limit can be obtained from the left-hand side of (1) using a set of structure factors at low resolution on the right-hand side. A least-squares procedure based on (1) has accordingly been developed.

The least-squares procedure

Suppose that we have m known structure factors with reciprocal vectors all within a resolution limit H_L.

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