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Dynamical Effects of Thermal Diffuse Scattering in RHEED

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Abstract

A general form for the intensity distribution of thermal diffuse scattering (TDS) is derived within the framework of quantum mechanics. The formulation takes into account the dynamical diffraction effects for both the initial and the final states of high-energy electrons and uses a single incoherent scattering approximation. When applied to estimate the intensity distribution of TDS in reflection high-energy electron diffraction (RHEED), using a two-wave approximation for the elastically scattered wave field, an analytical expression is obtained. It is shown that the dynamical diffraction effects may lead to an enhancement in the TDS intensities near the diffraction spots. The influence of correlated thermal atomic displacements on the contrast of the Kikuchi line parallel to the surface shadow edge has been discussed.

1. Introduction

It is now well established that inelastically scattered electrons contribute considerably to the image formation of reflection electron microscopy (Yagi, 1987; Peng & Cowley, 1989). The analysis of the angular and energy spectra of electrons scattered at grazing incidence from a crystal surface shows that the observed distribution results from inelastic collisions accompanied by phonon, plasmon and single-electron excitations of the target (Howie, 1983; Wang, 1988). In particular, Holloway & Beeby (1978), Agrawal (1984) and Peng & Cowley (1988, 1989) have shown that it is the thermal diffuse scattering that gives the appearance of the intensity peaks in reflection high-energy electron diffraction (RHEED) patterns in directions which prove to be forbidden by the energy and momentum conservation laws for pure elastic scattering. To analyze the intensity distribution in the thermal diffuse scattering (TDS) peaks, Holloway & Beeby (1978), Agrawal (1984), Albrecht & Meyer-Ehmsen (1988) and Korte & Meyer-Ehmsen (1990) used a kinematical theory and assumed that the states of electrons before and after the inelastic scattering are planar waves.

In the case of transmission high-energy electron diffraction (THEED), Rez, Humphreys & Whelan (1977) have shown that the kinematical approach to TDS can be used only when the effects of dynamical diffraction by the time-averaged crystal potential is negligible. The calculations of Maksym & Beeby (1981), Ichimiya (1983) and Peng & Cowley (1986) indicate that the criteria for the validity of the kinematical approximation in RHEED are not satisfied, and the diffraction by a periodical potential changes greatly the wave function of a high-energy electron inside the crystal in comparison with the incident planar wave.

The mutual influence of dynamical diffraction and inelastic scattering of high-energy electrons in the crystal is also of interest in the problem of Kikuchi-pattern intensity distribution analysis (Miyake, Hayakawa, Kawamura & Ohtsuki, 1975; Kawamura, Ichikawa & Goldstaub, 1973). It is worth mentioning that the theory of Kikuchi patterns (Kainuma, 1955; Chukhovskii, Alexanjan & Pinsker, 1973; Rossouw & Bursill, 1986; Bird & Wright, 1989) is based usually on the account of the diffraction of inelastically scattered electrons. In the RHEED case, however, the TDS intensity peaks are located in the vicinity of diffraction spots and the effects of elastic collisions are to be taken into account both before and after the inelastic scattering.

In this paper we give a general formulation for the TDS intensity distribution, treating the inelastic collision kinematically and the diffraction of electrons before and after the collision dynamically. An analytical expression for the angular distribution of TDS intensity is obtained using a two-wave approximation which permits a simple analytical representation of the dynamical diffraction processes. It is shown that the dynamical effects may lead to a considerable increase of TDS intensity in the vicinity of diffraction

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spots compared with the predictions of a kinematical TDS theory. The influence of the correlation of the thermal atomic displacements is shown to decrease with the increase of the scattering angle, and consequently the contrast of a horizontal Kikuchi line reverses from the center to the edge as observed experimentally by Miyake *et al.* (1975).

2. Single incoherent scattering approximation

As known, at the collisions with phonons a high-energy electron loses only a small portion of its energy $\Delta E \simeq k_B T \lesssim 0{\cdot}025$ eV, and the thermal scattering may be considered as quasielastic. This means that, in calculating the TDS intensity, a time averaging over the thermal motion gives the same results as a simple configuration averaging used in the statistical theory of X-ray diffraction (see, for example, Holy & Gabrielyan, 1987). To determine the TDS intensity distribution we consider the averaged direct product of the wave functions of the scattering problem (*i.e.* the density matrix)

$$\rho(\mathbf{r}, \mathbf{r}') = \langle \psi(\mathbf{r}) \psi^*(\mathbf{r}') \rangle. \tag{1}$$

Let us separate from the crystal potential $U(\mathbf{r})$ the perturbation potential describing the atomic thermal oscillations

$$\delta U(\mathbf{r}) = U(\mathbf{r}) - \langle U(\mathbf{r}) \rangle \tag{2}$$

and write the solution of the scattering problem in an integral form:

$$\psi(\mathbf{r}) = \psi(\mathbf{r}, \mathbf{p}_0) + \int \mathrm{d}^3 R \, G(\mathbf{r}, \mathbf{R}) \delta U(\mathbf{R}) \psi(\mathbf{R}), \tag{3}$$

where the function $\psi(\mathbf{r}, \mathbf{p}_0)$ represents the solution to the elastic scattering problem in the effective non-Hermitian potential (Dudarev, 1988; Gorodnichev, Dudarev, Rogozkin & Ryazanov, 1989) ($\hbar = 1$)

$$-\frac{1}{2m} \frac{\partial^2}{\partial \mathbf{r}^2} \psi(\mathbf{r}, \mathbf{p}_0) + V(\mathbf{r}) \psi(\mathbf{r}, \mathbf{p}_0) = (\mathbf{p}_0^2 / 2m) \psi(\mathbf{r}, \mathbf{p}_0). \tag{4}$$

The potential $V(\mathbf{r})$ is a sum of two terms: a Hermitian one which describes the usual elastic electron scattering and a non-Hermitian one which corresponds to the effective particle absorption due to the inelastic transitions (Whelan, 1965; Radi, 1970; Dudarev & Ryazanov, 1988).

For a planar wave $\exp(i\mathbf{p}_0\mathbf{r})$ incident on a crystal from infinity, the boundary condition for (4) is of the form

$$\psi_{\mathrm{inc}}(\mathbf{r}) = \exp(i\mathbf{p}_0\mathbf{r}). \tag{5}$$

The Green function $G(\mathbf{r}, \mathbf{R})$ appearing in (3) describes the field due to a point source

$$\left[\frac{p_0^2}{2m} + \frac{1}{2m} \frac{\partial^2}{\partial \mathbf{r}^2} - V(\mathbf{r}) \right] G(\mathbf{r}, \mathbf{R}) = \delta(\mathbf{r} - \mathbf{R}). \tag{6}$$

It should be noted that the formulation suggested above differs from the previous ones (Gjønnes, 1972; Kambe, 1967; Serneels, Haentjens & Gevers, 1980) in the form of the Green function (6). In our case the function is so chosen to include the dynamical diffraction effects by the effective periodical potential of the crystal $V(\mathbf{r})$ [for detailed treatment see Gomoyunova, Dudarev & Pronin (1990)].

The averaging of a bilinear combination of the wave functions (1) is easier to perform by expansion into a series in powers of the potential $\delta U(\mathbf{r})$. By virtue of the relation

$$\langle \delta U(\mathbf{r}) \rangle = 0, \tag{7}$$

we have

$$\begin{aligned} \rho(\mathbf{r}, \mathbf{r}') = {} & \psi(\mathbf{r}, \mathbf{p}_0) \psi^*(\mathbf{r}', \mathbf{p}_0) \\ & + \int \mathrm{d}^3 R \, \mathrm{d}^3 R' \, G(\mathbf{r}, \mathbf{R}) G^*(\mathbf{r}', \mathbf{R}') \\ & \times \langle \delta U(\mathbf{R}) \delta U(\mathbf{R}') \rangle \\ & \times \psi(\mathbf{R}, \mathbf{p}_0) \psi^*(\mathbf{R}', \mathbf{p}_0) + \ldots . \end{aligned} \tag{8}$$

The series (8) has a simple physical meaning. The first term of this series describes elastic scattering, the second term corresponds to a single incoherent scattering, and $\rho(\mathbf{r}, \mathbf{r}')$ is the density matrix of a fast electron with the energy close to its initial energy $|E - p_0^2/2m| \leq k_B T$. It is worth mentioning that (8) can also be obtained by expanding the stationary kinetic-equation solution into a series of integral of inelastic collisions (Dudarev, 1988; Gomoyunova *et al.* 1990).

The angular and energy distributions of particles are described in quantum mechanics by the diagonal elements of the density matrix in its momentum representation (Blum, 1981)

$$\rho(\mathbf{p}, \mathbf{p}') = \int \mathrm{d}^3 r \, \mathrm{d}^3 r' \exp(-i\mathbf{p}\mathbf{r} + i\mathbf{p}'\mathbf{r}') \rho(\mathbf{r}, \mathbf{r}'). \tag{9}$$

Calculation of these elements generally requires the integration of (1) over all three spatial coordinates. However, in cases where all the electrons emerging from the crystal have nearly the same energy $E \cong p_0^2/2m$, the angular distribution can be determined if we know the distribution of particles between the components of the momentum parallel to the surface $\mathbf{p}_\parallel = (p_x, p_y)$ in the limit $z \to -\infty$, where the direction of axis z coincides with the inner normal to a crystal surface.

$$\rho(\mathbf{p}_\parallel, z; \mathbf{p}_\parallel, z) = \int \mathrm{d}^2 r_\parallel \, \mathrm{d}^2 r'_\parallel \exp[i\mathbf{p}_\parallel(\mathbf{r}'_\parallel - \mathbf{r}_\parallel)] \rho(\mathbf{r}_\parallel, z; \mathbf{r}'_\parallel, z). \tag{10}$$

It is common practice in RHEED to characterize the momentum direction of a particle emerging from the crystal by two angles θ_1 and φ_1 using a spherical system of coordinates. In this case the diagonal elements (10) are related to the angular distribution of electrons scattered in a half-space $z < 0$

(Gorodnichev, Dudarev, Rogozkin & Ryazanov, 1987)

$$|\cos\theta_1|\rho(\mathbf{p}_\parallel; -\infty; \mathbf{p}_\parallel; -\infty)[\mathrm{d}^2p_\parallel/(2\pi)^2\Sigma] = I(\theta_1; \varphi_1)\,\mathrm{d}(\cos\theta_1)\,\mathrm{d}\varphi_1,$$

where

$$\mathbf{p}_\parallel = (p_0 \sin\theta_1 \cos\varphi_1; p_0 \sin\theta_1 \sin\varphi_1); \cos\theta_1 < 0$$

and Σ is a crystal surface area. Taking into account the relation

$$\mathrm{d}^2p_\parallel = p_0^2|\cos\theta_1|\,\mathrm{d}\varphi_1\,\mathrm{d}(|\cos\theta_1|), \tag{11}$$

we obtain

$$I(\theta_1; \varphi_1) = (p_0 \cos\theta_1/2\pi)^2 \times \Sigma^{-1}\rho(\mathbf{p}_\parallel; -\infty; \mathbf{p}_\parallel; -\infty). \tag{12}$$

To calculate the angular distribution of the scattered electrons, using (8) and (12) one needs to evaluate the Fourier component of the Green function (6) between the coordinates parallel to the surface

$$g_{\mathbf{p}_\parallel}(-\infty; \mathbf{R}) = \int \mathrm{d}^2r_\parallel\, G(\mathbf{r}_\parallel, -\infty; \mathbf{R}) \exp(-i\mathbf{p}_\parallel\mathbf{r}_\parallel). \tag{13}$$

Taking into account the reciprocity theorem (Pogany & Turner, 1968),

$$G(\mathbf{r}, \mathbf{R}) = G(\mathbf{R}, \mathbf{r}), \tag{14}$$

and using (13), we obtain to an accuracy of a phase factor (Gorodnichev *et al.*, 1989)

$$g_{\mathbf{p}_\parallel}(-\infty; \mathbf{R}) = -(im/p_0|\cos\theta_1|)\psi(\mathbf{R}, -\mathbf{p}_1). \tag{15}$$

It is interesting to note that the function $\psi(\mathbf{R}, -\mathbf{p}_1)$ corresponds to the known 'reciprocal wave' used by Kainuma (1955) in his theory of Kikuchi patterns. A substitution of (15) into (12) and application of the expression (8) then allows us to obtain the following expression for the angular distribution of TDS

$$I(\theta_1; \varphi_1) = \Sigma^{-1}(m/2\pi)^2 \int \mathrm{d}^3R\,\mathrm{d}^3R' \times \psi(\mathbf{R}, -\mathbf{p}_1)\psi^*(\mathbf{R}', -\mathbf{p}_1) \times \langle\delta U(\mathbf{R})\delta U(\mathbf{R}')\rangle\psi(\mathbf{R}, \mathbf{p}_0)\psi^*(\mathbf{R}', \mathbf{p}_0). \tag{16}$$

Equation (16) shows that, in order to find the intensity of TDS in a direction $\mathbf{p}_1$, it is necessary to calculate the double integral of the product of the potential correlator (2) times the four solutions of the dynamical diffraction problem. It should be noted that a formula which is similar to (16) was obtained by Dmitrienko & Kaganer (1987) in the statistical dynamical theory of X-rays.

If we neglect the dynamical diffraction effects, (16) then reduces to the known kinematical formula (Holloway & Beeby, 1978; Agrawal, 1984). In what follows, we will give a solution for the elastic scattering problem (4) using a two-wave approximation. The two-wave solution then allows us to calculate the integral (16) in an explicit analytical form. A generalization of the corresponding formulae to the many-beam case can be obtained by using one of the numerical methods as developed, for example, by Collela (1972), Moon (1972) and Ma & Marks (1989).

3. The wave function of the elastic scattering problem

To solve the problem (4), (5), let us consider the case of the two-wave Bragg diffraction when the potential $V(\mathbf{r})$ in the region $z > 0$ may be presented in the form

$$V(\mathbf{r}) = V_0 + V_1 \exp(iGz) + V_{-1} \exp(-iGz), \tag{17}$$

where in the general case $V_1^* \neq V_{-1}$. Outside the crystal $z < 0$ the potential $V(\mathbf{r})$ is equal to zero. It is known that the two-wave approximation can be used in cases when

$$G^2/2m|V_1| \gg 1. \tag{18}$$

If the condition (18) is satisfied the wave function $\psi(\mathbf{r}, \mathbf{p}_0)$ may then be represented in the form $\psi(\mathbf{r}, \mathbf{p}_0) = \exp[i(\mathbf{p}_0)_\parallel\mathbf{r}_\parallel]\varphi(z)$, where

$$\varphi(z) = \begin{cases} \alpha \exp(i\kappa z) + \beta \exp[i(\kappa - G)z] & \text{at } z > 0 \\ \exp(ik_0 z) + R \exp(-ik_0 z) & \text{at } z < 0, \end{cases} \tag{19}$$

where $k_0 = (\mathbf{p}_0)_z = p_0 \cos\theta_0$, and the quasimomentum κ is a solution of the equation

$$\left(\frac{\kappa^2}{2m} - \frac{k_0^2}{2m} + V_0\right)\left[\frac{(\kappa - G)^2}{2m} - \frac{k_0^2}{2m} + V_0\right] - V_1V_{-1} = 0. \tag{20}$$

A physically allowed root of (20) is selected by the conditions that $\mathrm{Im}\,\kappa > 0$ and $\lim\kappa = k_0$ at V_0, V_1, $V_{-1} = 0$. Applying these conditions, we find

$$\kappa = (G/2) + \{k_0^2 + (G^2/4) - 2mV_0[G^2(k_0^2 - 2mV_0) + 4m^2V_1V_{-1}]^{1/2}\}^{1/2}, \tag{21}$$

where a complex square root is determined by the rule $z^{1/2} = |z|^{1/2}\exp(i\delta/2)$ where $z = |z|\exp(i\delta)$; $0 \leq \delta < 2\pi$. Using the conditions of continuity for $\varphi(z)$ and $\varphi'(z)$ at $z = 0$, one can find the numerical coefficients in (19):

$$R = \frac{k_0 - \kappa + D(k_0 - \kappa + G)}{k_0 + \kappa + D(k_0 + \kappa + G)};$$
$$\alpha = \frac{2k_0}{k_0 + \kappa + D(k_0 + \kappa + G)}; \quad \beta = D\alpha; \tag{22}$$
$$D = \frac{2mV_{-1}}{k_0^2 - (\kappa - G)^2 - 2mV_0}.$$

At $k_0^2/2m \gg |V_0|$, (22) give $\alpha \simeq 1$ and $\beta \simeq D \simeq R$ and this property is very important for our further analysis. In the case of zero absorption and in the region of values

$$[(G^2/4) + 2mV_0 - 2m|V_1|]^{1/2} \leq k_0 \leq [(G^2/4) + 2mV_0 + 2m|V_1|]^{1/2}, \tag{23}$$

the real part of the quasimomentum Re κ is constant and equals Re $\kappa = G/2$, giving $|R| = 1$. An imaginary part of the quasimomentum reaches its maximum of Im $\kappa = 2m|V_1|/G$ at the point

$$k_0 = [(G^2/4) + 2mV_0 - (2m|V_1|/G)^2]^{1/2}. \quad (24)$$

At this point the wave function penetrates into the crystal by the minimal depth

$$l \sim (2 \text{ Im } \kappa)^{-1} = G/4m|V_1|. \quad (25)$$

The penetration depth (25) is many times the lattice constant along the z direction. This circumstance allows us to limit ourselves to the case of bulk phonon scattering only.

4. Structure factor of phonon scattering

To calculate the correlator of the potential fluctuations in formula (8) we represent $\delta U(\mathbf{R})$ in the form of a sum of the potentials of separate atoms

$$\begin{aligned}\delta U(\mathbf{R}) &= \sum_a \delta U_a(\mathbf{R}) \\ &= \sum_a U_a(\mathbf{R} - \mathbf{R}_a - \mathbf{u}_a) - \langle U_a(\mathbf{R} - \mathbf{R}_a - \mathbf{u}_a)\rangle \\ &= \int d^3q/(2\pi)^3 \sum_a U_a(\mathbf{q}) \exp[i\mathbf{q}(\mathbf{R} - \mathbf{R}_a)] \\ &\quad \times [\exp(-i\mathbf{q}\mathbf{u}_a) - \langle \exp(-i\mathbf{q}\mathbf{u}_a)\rangle]. \quad (26)\end{aligned}$$

Neglecting the anharmonicity of the thermal motion, one can perform averaging of the product of (26) over atomic displacements, following the method as developed by Afanas'ev & Kagan (1968).

$$\begin{aligned}\langle \delta U(\mathbf{R}) \delta U(\mathbf{R}')\rangle &= \int [d^3q\, d^3k/(2\pi)^6] \\ &\quad \times \sum_{q,b} U_a(\mathbf{q}) U_b(-\mathbf{k}) \\ &\quad \times \exp[i\mathbf{q}(\mathbf{R} - \mathbf{R}_a) - i\mathbf{k}(\mathbf{R}' - \mathbf{R}_b)] \\ &\quad \times \exp[-\tfrac{1}{2}\langle(\mathbf{q}\mathbf{u}_a)^2\rangle - \tfrac{1}{2}\langle(\mathbf{k}\mathbf{u}_b)^2\rangle] \\ &\quad \times \{\exp[\langle(\mathbf{q}\mathbf{u}_a)(\mathbf{k}\mathbf{u}_b)\rangle] - 1\}, \quad (27)\end{aligned}$$

where $U_a(\mathbf{q}) = -(4\pi e^2/q^2)[Z_a - f_a(\mathbf{q})]$ is a Fourier component of the atomic potential. Following Afanas'ev & Kagan, we divide a summation over the coordinates of atoms $\mathbf{R}_a$ in (27) into a summation over centers of unit cells $\mathbf{R}_\mathfrak{n}$ and a summation over atoms within a cell j, $\mathbf{R}_a = \mathbf{R}_\mathfrak{n} + \mathbf{r}_j$. Then, for values included in (27) we may write

$$\langle(\mathbf{q}\mathbf{u}_a)(\mathbf{k}\mathbf{u}_b)\rangle = Y_{jj'}(\mathbf{q}, \mathbf{k}, \mathbf{R}_\mathfrak{n} - \mathbf{R}_{\mathfrak{n}'}) \quad (28)$$

where

$$\begin{aligned}&Y_{jj'}(\mathbf{q}, \mathbf{k}, \mathbf{R}_\mathfrak{n} - \mathbf{R}_{\mathfrak{n}'}) \\ &= \Omega_0/(2\pi)^3 \int d^3f \sum_\alpha \frac{\bar{n}(\mathbf{f}, \alpha) + \frac{1}{2}}{\omega_\alpha(\mathbf{f})(M_j M_{j'})^{1/2}} \\ &\quad \times [\mathbf{q}\mathbf{e}_j(\mathbf{f}, \alpha)][\mathbf{k}\mathbf{e}^*_{j'}(\mathbf{f}, \alpha)] \exp[i\mathbf{f}(\mathbf{R}_\mathfrak{n} - \mathbf{R}_{\mathfrak{n}'})]. \quad (29)\end{aligned}$$

In (29), Ω_0 is the volume of a unit cell, M_j is the mass of the jth atom, the summation over $\mathbf{f}$ and α is carried out over the wave vectors and the branches of the phonon spectrum of the crystal with the frequencies $\omega_\alpha(\mathbf{f})$ and polarization vectors $\mathbf{e}_j(\mathbf{f}, \alpha)$, and for equilibrium

$$\bar{n}(\mathbf{f}, \alpha) = \{\exp[\omega_\alpha(\mathbf{f})/k_B T] - 1\}^{-1}.$$

At $j = j'$ and $\mathbf{R}_\mathfrak{n} = \mathbf{R}_{\mathfrak{n}'}$, (29) coincides with the usual Debye-Waller factor

$$Y_{jj}(\mathbf{q}, \mathbf{q}, 0) = W_j(\mathbf{q}) = \langle(\mathbf{q}\mathbf{u}_j)^2\rangle. \quad (30)$$

In the limiting case $|\mathbf{R}_\mathfrak{n} - \mathbf{R}_{\mathfrak{n}'}| \gg \Omega_0^{-1/3}$ the acoustic branches of the phonon spectrum give the main contribution to the sum (29) and in the cubic (b.c.c. or f.c.c.) lattice one can obtain

$$Y_{jj'}(\mathbf{q}, \mathbf{k}, \mathbf{R}_\mathfrak{n} - \mathbf{R}_{\mathfrak{n}'}) = \frac{k_B T}{Mc^2} \frac{\Omega_0}{4\pi} \frac{(\mathbf{q}\mathbf{k})}{|\mathbf{R}_\mathfrak{n} - \mathbf{R}_{\mathfrak{n}'}|}, \quad (31)$$

where $M = \sum_j M_j$ and c is the velocity of sound. We have already mentioned that the calculation of diffuse intensity (16) reduces to the integration of the potential fluctuations correlator (27) over all values of $\mathbf{R}$ and $\mathbf{R}'$ inside the crystal. It is seen that the result of the integration may be represented in the form of a linear combination of integrals of $\langle \delta U(\mathbf{R}) \delta U(\mathbf{R}')\rangle$ times the four exponents decreasing at infinity

$$\psi(\mathbf{R}) \simeq \exp(i\mathbf{Q}\mathbf{R} - \boldsymbol{\mu}\mathbf{R}), \quad (32)$$

where the vector $\boldsymbol{\mu}$ lies along the z axis, $|\boldsymbol{\mu}| = \mu \ll G$.

Let us introduce the notation

$$\begin{aligned}&(m/2\pi)^2 \Sigma^{-1} \int d^3R\, d^3R' \exp(i\mathbf{K}_1\mathbf{R} - i\mathbf{K}_2\mathbf{R}') \\ &\quad \times \exp[-(\boldsymbol{\mu}_0 + \boldsymbol{\mu}_1)(\mathbf{R} + \mathbf{R}')] \langle \delta U(\mathbf{R}) \delta U(\mathbf{R}')\rangle \\ &\quad = [2(\mu_0 + \mu_1)]^{-1} S(\mathbf{K}_1; \mathbf{K}_2). \quad (33)\end{aligned}$$

According to (19) and (16), to estimate the diffuse background we must calculate the integral (33) at $\mathbf{K}_1 - \mathbf{K}_2 = \mathbf{G}'$ where $\mathbf{G}'$ is a reciprocal-lattice vector. Since the general expression for $S(\mathbf{K} + \mathbf{G}_1; \mathbf{K} + \mathbf{G}_2)$ corresponding to an arbitrary lattice symmetry proves to be very cumbersome, we limit ourselves to the calculation of this factor in the case of a monoatomic cubic lattice, one atom of which is located at the origin. To realize this we separate all terms corresponding to $\mathfrak{n} = \mathfrak{n}'$ and $j = j'$ from the sum over $a = (\mathfrak{n}, j)$ and $b = (\mathfrak{n}', j')$. Summing over $\mathbf{R}_\mathfrak{n}$, we obtain

$$\begin{aligned}S(\mathbf{K} + \mathbf{G}_1, \mathbf{K} + \mathbf{G}_2) &= S_E(\mathbf{K} + \mathbf{G}_1, \mathbf{K} + \mathbf{G}_2) \\ &\quad + S_D(\mathbf{K} + \mathbf{G}_1, \mathbf{K} + \mathbf{G}_2) \quad (34)\end{aligned}$$

$$\begin{aligned}&S_E(\mathbf{K} + \mathbf{G}_1; \mathbf{K} + \mathbf{G}_2) \\ &\quad = n(m/2\pi)^2 U(-\mathbf{K} - \mathbf{G}_1) U(\mathbf{K} + \mathbf{G}_2) \\ &\quad \times \{\exp[-\tfrac{1}{2} W(\mathbf{G}_2 - \mathbf{G}_1)] \\ &\quad - \exp[-\tfrac{1}{2} W(\mathbf{K} + \mathbf{G}_1) - \tfrac{1}{2} W(\mathbf{K} + \mathbf{G}_2)]\}, \quad (35)\end{aligned}$$

where n is the number of atoms per unit volume. It

should be noted that (35) coincides with the phonon structure factor calculated within the Einstein model of thermal motion by Hall & Hirsch (1965) and Whelan (1965). The second term in (34) results from the scattering by correlated atomic displacements. Afanas'ev & Kagan (1968) have shown that the first term of the expansion of (27) in powers of the $Y_{jj'}(\mathbf{q}, \mathbf{k}, \mathbf{R}_n - \mathbf{R}_{n'})$ function gives the main contribution to the second summand. Using the Debye model (31) we obtain a simple expression for the second term in (34)

$$\begin{aligned} S_D(\mathbf{K}+\mathbf{G}_1; \mathbf{K}+\mathbf{G}_2) &= n\nu U(-\mathbf{K}-\mathbf{G}_1)\,U(\mathbf{K}+\mathbf{G}_2) \\ &\times (k_B T/Mc^2)\exp[-\tfrac{1}{2}W(\mathbf{K}+\mathbf{G}_1)] \\ &\times \exp[-\tfrac{1}{2}W(\mathbf{K}+\mathbf{G}_2)](\mathbf{K}+\mathbf{G}_1, \mathbf{K}+\mathbf{G}_2) \\ &\times \sum_G [(|\mathbf{K}_\parallel+\mathbf{G}_\parallel|+\mu_0+\mu_1) \\ &\times \{|\mathbf{K}_\parallel+\mathbf{G}_\parallel|[(K_z+G_z)^2 \\ &+ (|\mathbf{K}_\parallel+\mathbf{G}_\parallel|+\mu_0+\mu_1)^2]\}^{-1}], \end{aligned} \tag{36}$$

where ν is the number of atoms in a unit cell. In this case only one summand closest to the pole $|\mathbf{K}+\mathbf{G}|^{-1}$ in the sum over $\mathbf{G}$ in (36) should always be preserved.

It is interesting to note that a similar expression to (36) has been obtained by Afanas'ev, Kagan & Chukhovskii (1968) in the dynamical theory of X-ray diffraction.

Finally, inserting (19) in (16) and taking into account the definitions (33) and (22) one obtains

$$\begin{aligned} I(\theta_1; \varphi_1) = 2(\mu_0+\mu_1)]^{-1}\{&S(\mathbf{K}, \mathbf{K}) \\ &+ |R_0+R_1|^2 S(\mathbf{K}+\mathbf{G}, \mathbf{K}+\mathbf{G}) \\ &+ |R_0 R_1|^2 S(\mathbf{K}+2\mathbf{G}, \mathbf{K}+2\mathbf{G}) \\ &+ 2\,\mathrm{Re}\,(R_0+R_1) S(\mathbf{K}, \mathbf{K}+\mathbf{G}) \\ &+ 2\,\mathrm{Re}\,(R_0 R_1) S(\mathbf{K}, \mathbf{K}+2\mathbf{G}) \\ &+ 2\,\mathrm{Re}\,\{|R_0|^2 R_1 + |R_1|^2 R_0\} \\ &\times S(\mathbf{K}+\mathbf{G}, \mathbf{K}+2\mathbf{G})\}, \end{aligned} \tag{37}$$

where the vector $\mathbf{G}$ lies opposite the z axis, $(\mathbf{G})_z = -G$, $\mathbf{K}_\parallel = (\mathbf{p}_0 - \mathbf{p}_1)_\parallel$, $K_z = \mathrm{Re}\,\kappa_0 + \mathrm{Re}\,\kappa_1$, $\mu_{1,0} = \mathrm{Im}\,\kappa_{1,0}$ and κ_i denotes the quasimomentum (21) corresponding to the wave vector $k_i = p_0|\cos\theta_i|$; $i = 0,1$.

Expression (37) gives the complete solution of the thermal diffuse background evaluation problem in RHEED under the conditions of two-wave Bragg diffraction for both the incident wave on a crystal and the inelastically scattered electrons.

5. Analysis of intensity distribution

As known, the diffuse background (37) possesses the highest intensity in the vicinity of the diffraction spots (Peng & Cowley, 1988, 1989), which, within the two-wave approximation, correspond to the directions $\mathbf{K} \simeq -\mathbf{G}$. In the vicinity of such points the main contribution is due to the second term of the structure factor (34) and

$$\begin{aligned} I(\theta_1; \varphi_1) \cong \frac{n\nu}{2(\mu_0+\mu_1)} &\left|\frac{m}{2\pi} U(\mathbf{G})\right|^2 \\ &\times \frac{k_B T}{Mc^2} \exp[-W(\mathbf{G})]\mathbf{G}^2 |1 - R_0 R_1|^2 \\ &\times \frac{|\mathbf{K}_\parallel| + \mu_0 + \mu_1}{|\mathbf{K}_\parallel|[(K_z - G)^2 + (|\mathbf{K}_\parallel| + \mu_0 + \mu_1\})^2]}. \end{aligned} \tag{38}$$

From (38) we can see that a 'kinematical' divergence of the intensity in the vicinity of a mirror reflection $I \sim |\mathbf{K}_\parallel|$ found by Holloway & Beeby (1978) and Agrawal (1984) is preserved in the dynamical theory. However, the total intensity integrated over θ_1 and φ_1 turns out to be essentially greater in dynamical theory than that in the kinematical case. This effect arises from the constant difference $K_z - G$ within the region of 'total' Bragg reflection (23). Indeed, if the inequalities (23) are satisfied and the absorption is weak, *i.e.* $\mathrm{Re}\,\kappa_0 \simeq G/2$ and $\mathrm{Re}\,\kappa_1 \simeq G/2$, then one can obtain $|K_z - G| \to 0$. This means the diffuse intensity around the Bragg reflection reduces insignificantly even at relatively high angular deviations $|\mathbf{K}_\parallel| \simeq \mu_0, \mu_1$ of the momentum $\mathbf{p}_1$ from the mirror direction (see Fig. 1). It is interesting to note that in the Laue case discussed by Takagi (1958) and Afanas'ev & Kagan (1968) the diffuse intensity remains finite even in the vicinity of the diffraction spots. However, the divergence of the above-obtained type exists in the Bragg geometry as was shown by Afanas'ev, Kagan & Chukhovskii (1968) for X-rays.

The effect of background increase on the opposite (with respect to the position of elastic reflection) side of the total reflection region is an interesting peculiarity of diffuse scattering in Bragg geometry (Figs. 1 and 2).

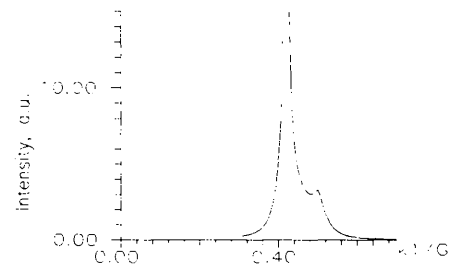

Fig. 1. Thermal diffuse scattering intensity *versus* the normal momentum component of emerging electrons k_1/G. Momentum p_1 is located in the plane of incidence. Values of Fourier components of potential (17) $2mV_0/G^2 = -0{\cdot}045 - i0{\cdot}009$; $2mV_1/G^2 = -0{\cdot}045 - i0{\cdot}003$. Normal component of the incident electron momentum k_0/G is equal to 0·42; $p_0/G = 20$.

In (38), the factor $|1-R_0R_1|^2$ promotes this effect. Actually, the real part of the reflection coefficient R has different signs on different sides of the Bragg reflection region (23). It results in the fact that a maximum of the coefficient $|1-R_0R_1|$ at $R_0=1$ corresponds to the value $R_1\simeq -1$ and *vice versa*. It may lead to the appearance of an additional peak in the distribution of diffuse intensity similar to that shown in Fig. 2. The physical nature of this additional peak is connected with the predominance of the interbranch phonon scattering over the intrabranch one in the vicinity of the diffraction spots (Takagi, 1958; Rez, Humphreys & Whelan, 1977). In fact, within the approximation (36) a correlator of potential fluctuations from (8), (16) may be written in the form

$$\langle \delta U(\mathbf{R})\delta U(\mathbf{R}')\rangle = \sum_{a,b} \langle (\mathbf{u}_a\,\partial/\partial\mathbf{R})\langle U_a(\mathbf{R}-\mathbf{R}_a-\mathbf{u}_a)\rangle \times (\mathbf{u}_b\,\partial/\partial\mathbf{R}')\langle U_b(\mathbf{R}'-\mathbf{R}_b-\mathbf{u}_b)\rangle\rangle. \quad (39)$$

In the vicinity of atomic equilibrium positions, (39) is an odd function of its arguments $\mathbf{R}-\mathbf{R}_a$ and $\mathbf{R}'-\mathbf{R}_b$. As a result, the phonon transitions among Bloch states of opposite parity contribute most of all to the diffuse intensity in the vicinity of the diffraction maxima. These states correspond to vacuum wave vectors k_0 and k_1 lying on different sides of the Bragg reflection region (23) [for a detailed discussion of symmetry properties of Bloch-wave functions in the Bragg case see the review by Koval'chuk & Kohn (1986)]. It is important to stress that the occurrence of the second diffuse maximum in the diffraction pattern had been mentioned by Takagi (1958) but he discussed the effect only qualitatively.

In the region of relatively high angles of deviation from the mirror direction, the first term of the structure factor (35) contributes most to the intensity. In this case, (37) describes the intensity distribution in the Kikuchi line parallel to the surface shadow edge (a horizontal Kikuchi line). It should then be noted that (37) contains an important factor $(\mu_0+\mu_1)^{-1}$ missed by Kawamura *et al.* (1973) and Miyake *et al.* (1975). A factor of this type is well known in all problems of radiative transfer in half-space targets (Chandrasekhar, 1960; Case & Zweifel, 1967; Dudarev, 1988; Gorodnichev *et al.*, 1989). In particular, in (37) this factor takes account of a change of the effective number of scattering atoms at varying diffraction conditions for incident or emerging electrons. The factor $(\mu_0+\mu_1)^{-1}$ reaches its minimal value at the center of a Bragg reflection region. It is evidently linked with a sharp reduction of the electron penetration depth into a crystal in this case.

Fig. 3 illustrates the intensity profile of a Kikuchi line calculated in accordance with (37) where the structure factor $S(\mathbf{K}_1;\mathbf{K}_2)$ is defined by (35). The angle between the plane of incidence and the plane of emergence is equal to $\arcsin(2G/p_0\sin\theta_0)\simeq 5{\cdot}5°$. The curve given is in good agreement with the experimental data of Miyake *et al.* (1975). Notice that the transition to large scattering angles is accompanied by the appearance of a typical minimum in the profile of the Kikuchi line [see, for example, Fig. 4 of Miyake *et al.* (1975)]. Miyake *et al.* (1975) explained the effect on the basis of the theoretical approach developed by Okamoto, Ichinokawa & Ohtsuki (1971). They showed that the use of the Einstein model allows one to calculate the intensity profiles similar to those observed experimentally. However, the contribution of the Debye term (36) is predominant in the area of small angles of scattering. This point should apparently always be taken into account in quantitative interpretation of the intensity distribution. In particular, the observed effect of line contrast variation with an increasing scattering angle (*i.e.* with a transfer from Fig. 1 to Fig. 3) may also be caused by a change of the form of the phonon structure factor from (36) to (35).

6. Concluding remarks

In this paper we have evaluated the thermal diffuse scattering background in RHEED within the framework of the two-wave approximation of dynamical diffraction theory. An enhancement in the diffuse intensity is found near the region of 'total' Bragg reflection, and the contrast of a horizontal Kikuchi line corresponding to the experimental conditions of Miyake *et al.* (1975) is calculated.

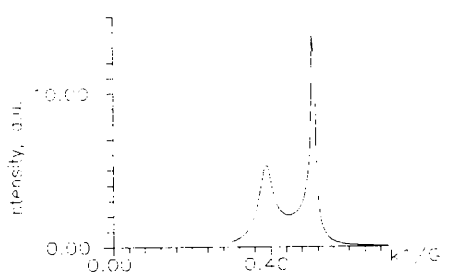

Fig. 2. Thermal diffuse scattering intensity in the plane of incidence for $k_0/G=0{\cdot}51$. Other parameters are the same as for Fig. 1.

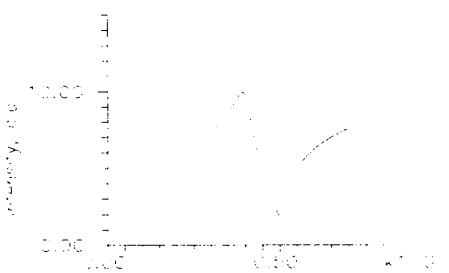

Fig. 3. Kikuchi line intensity profile, $k_0/G=0{\cdot}38$. The angle between the plane of incidence and the plane of emergence is equal to $\arcsin(2G/p_0\sin\theta_0)\simeq 5{\cdot}5°$.

To solve the problem of TDS in RHEED we have derived a general form for the intensity distribution of diffuse scattering, using a single incoherent scattering approximation. Dynamical elastic diffraction effects have been taken into account for both in the initial and final fast electron states. From the analysis of (8) one should note that the high-energy electron diffuse scattering pattern may be more complex than distribution (37). Firstly, an intensity pattern observed in the experiments of Peng & Cowley (1988, 1989) is formed under the many-beam diffraction conditions. The functions $\psi(\mathbf{r}, \mathbf{p}_0)$ and $\psi(\mathbf{r}, -\mathbf{p}_1)$ calculated using the many-beam theory methods may differ greatly in some cases from the corresponding two-wave solution (19). Secondly, statistics of the thermal motion near the surface (27) may not coincide with the statistics of thermal displacements in the bulk (Agrawal, Djafari-Rouhani & Dobrzynski, 1979). Moreover, for a rough surface the incoherent intensity resulting from the diffuse scattering of point defects, dislocations and atomic steps may be considerable. We believe that the approach given above may be applied effectively to analyze the diffuse intensity in all mentioned cases while investigating the surface structure by reflection high-energy electron diffraction.

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