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A 'three-beam' analysis of resonance scattering in reflection high-energy electron diffraction

G. R. Anstis

Department of Applied Physics, University of Technology, Sydney, PO Box 123, Broadway, NSW, Australia.
E-mail: geoff.anstis@uts.edu.au

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Dedicated to Professor A. F. Moodie on the occasion of his 75th birthday

Abstract

Enhanced reflection of fast electrons from a crystal surface and a decrease in the depth of penetration of the primary beam occurs when diffraction conditions are such as to set up a wave travelling just beneath the crystal surface. This is the surface resonance condition for reflection high-energy electron diffraction (RHEED). Quantitative prediction of these effects can be achieved by assuming that only the primary and two diffracted beams are significant. Expressions for the coefficient of reflection and the depth of penetration in terms of a few Fourier coefficients of an effective potential are derived. These coefficients depend sensitively on incident-beam direction and are significantly different from the values for the bulk crystal. In particular, the mean potential experienced by the electrons in the resonance state is increased. It can be estimated using Bethe's perturbation approach. Predictions of the position, height and width of the peak in reflectivity resulting from resonance scattering from the (111) surface of platinum are in reasonable agreement with the values obtained from many-beam computations. The three-beam approach gives insight into resonance scattering using the standard formalism of diffraction theory.

1. Introduction

Accurate analysis of electron diffraction patterns requires that the interaction between many diffracted beams be considered. Under some conditions, however, it is possible to assume that only the primary beam and two diffracted beams are significant. For instance, Moodie (1979) and Moodie *et al.* (1996) have shown that the three-beam approximation can be used to determine structure factors from transmission diffraction patterns. In reflection high-energy electron diffraction (RHEED), as shall be shown, a three-beam analysis can be applied to the phenomenon of surface resonance scattering. When the conditions for resonance scattering are set up, one observes an increase in the intensity of the specularly reflected beam along with a decrease in penetration of the fast electrons into the bulk of the crystal. A wavefield travelling parallel to the crystal surface is set up.

When diffraction from a surface occurs, one does not observe the diffracted beams that exist within the crystal directly, unlike the situation of diffraction associated with transmission through a thin foil. In the kinematic approximation to diffraction, the amplitudes of diffracted beams are given by the convolution of the Fourier components of the crystal potential with the shape function of the crystal (*e.g.* Cowley, 1995). The shape function of a semi-infinite crystal that is used in reflection experiments depends only on the coordinate perpendicular to the crystal surface and varies relatively slowly with distance in reciprocal space. The diffraction pattern resulting from reflection can thus be considered as being the intersection of the Ewald sphere with reciprocal-lattice rods which are perpendicular to the surface. The rods represent the scattering power as a function of angle of scattering for a system which is periodic in two dimensions. This scattering power can be described in terms of the Fourier components of the potential. If there is no surface reconstruction, the only potential components are those associated with reciprocal-lattice points lying along a line perpendicular to the surface.

The 'three-beam' analysis presented here involves two rods, labelled (0, 0) and (0, G). The (0, 0) rod has a scattering power described by a Fourier series with two terms [from the reciprocal-lattice points (0, 0, 0) and (0, 0, $-M_0 g$)] and the (0, G) rod has a scattering power described by a Fourier series with a single term related to the (0, G, $-M_g g$) reciprocal-lattice point. These three components of the scattering power lead to a diffracted wave with three Fourier components. These are the three beams that exist within the crystal. Fig. 1 shows two rows of reciprocal-lattice points and indicates three points that are close to the intersection of the Ewald sphere with the plane in reciprocal space containing these lattice points. Peng & Cowley (1987) provide a detailed description of the use of the Ewald construction in RHEED.

As for the case of transmission diffraction, it is possible to include, to some degree of approximation, through the use of perturbation theory as first set out by

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Bethe (*e.g.* Cowley, 1995), the effects of the Fourier components of the potential which have not been included in the three-beam approximation. Bethe's approach leads to an effective potential which depends on the direction of the incident beam.

In this paper, approximate expressions are obtained for the reflectivity and for the depth of penetration of the primary beam as functions of the angle of incidence of the beam and the Fourier components of the potential. These expressions are applied to resonance scattering from the (111) surface of platinum and the results are compared with those obtained by Dudarev & Whelan (1994*a*,*b*, 1995) through many-beam computations.

Dudarev & Whelan (1994*a*,*b*, 1995) have also established relatively simple expressions for the reflectivity and penetration depth. The relationship between the two methods is discussed and it is shown that they are of similar accuracy, as judged by comparison with the many-beam results.

The paper is set out as follows. In §2, the equations for the three-beam approximation are given. Their solution requires determination of the eigenvalues and eigenvectors of a 6×6 matrix. Through the use of Hill's determinant (Lamla, 1938; Moon, 1972), it is shown in Appendix *A* that the eigenvalues can be expressed as power series in the Fourier components of the potential. The first few terms of these series are determined by using a computer algebra package, *Mathematica* (Wolfram, 1991). The inverse of the imaginary part of one of these eigenvalues is directly proportional to the depth of penetration of the primary beam. Expressions for the eigenvectors (Bloch waves) associated with these eigenvalues are given. The reflection coefficient is then obtained by relating the wavefield inside the crystal to that outside.

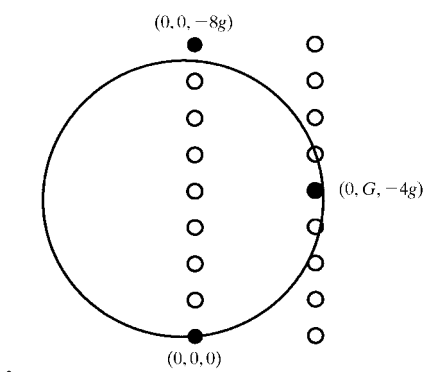

Fig. 1. The Ewald construction for analysing resonance scattering. The intersection of the Ewald sphere with a plane in reciprocal space and two rows of reciprocal-lattice points in that plane are shown. Three points closest to the sphere are shown as filled circles. These correspond to the beams used in the three-beam analysis.

In §3, the effective potentials which result from including the effects of other Fourier components on the amplitudes of the three beams are discussed. The analysis differs somewhat from that of the transmission case in that the eigenvalues are required to determine the correction terms for the original potential.

After the expressions for the depth of penetration and the reflection coefficient in terms of three components of an effective potential have been determined, in §4 they are applied to the surface resonance scattering of 100 keV electrons by the (111) surface of platinum, and curves showing how these two quantities vary with the azimuthal angle are compared with those obtained by Dudarev & Whelan (1994*a*,*b*).

2. Expressions for the reflection coefficient and penetration depth

Expressing the potential as a Fourier series with components V_g labelled by reciprocal-lattice vectors $\mathbf{g}$ results in Schroedinger's equation being transformed into an infinite set of equations for the Fourier components $C_g(\mathbf{k})$ of the wave function (Hirsch *et al.*, 1977). The countably infinite set of solutions that arise are labelled by $\mathbf{k}$.

$$[\chi^2 + U_0 - (\mathbf{k}+\mathbf{g})^2]C_g(\mathbf{k}) + \sum_{h\neq g} U_{g-h}C_h(\mathbf{k}) = 0, \quad (1)$$

where χ is the modulus of the wavevector of the incident plane wave and

$$\chi^2 = 2mE/\hbar^2(1 + eE/2m_0c^2) \quad (2)$$

and

$$U_g = 2meV_g/\hbar^2. \quad (3)$$

Let $K^2 = \chi^2 + U_0$ with $\mathbf{K} = \boldsymbol{\chi} + \gamma_0\mathbf{n}$ and where $\mathbf{n}$ is a unit vector normal to the surface and pointing towards the crystal. Instead of labelling solutions by $\mathbf{k}$ we use γ defined by

$$\mathbf{k} + \mathbf{g} = \mathbf{K} + \gamma\mathbf{n} + \mathbf{g}. \quad (4)$$

Then the scattering equations can be written as

$$\{(\gamma + \mathbf{K}\cdot\mathbf{n} + \mathbf{g}\cdot\mathbf{n})^2 - [K^2 - (\mathbf{K}+\mathbf{g})_t^2]\}C_g(\mathbf{k}) - \sum_{h\neq g} U_{g-h}C_h(\mathbf{k}) = 0, \quad (5)$$

where $(\mathbf{K}+\mathbf{g})_t$ is the tangential component of the vector.

Assume there are two rods which account for the scattering. Label these as $(0, 0)$ and $(0, G)$. The beams which contribute to the scattering can be labelled as $(0, 0, mg)$ and $(0, G, mg)$ where m is any integer.

Coordinates (x, y, z) are used, with z normal to the surface and x along the zone axis. Then χ_z depends on the tilt angle and χ_y depends on the azimuthal and tilt angles of the incident beam. It is convenient to define

$$x = (\gamma + \mathbf{K}\cdot\mathbf{n})/g \tag{6}$$

$$c_0^2 = K_z^2/g^2 \tag{7}$$

and

$$\begin{aligned} c_G^2 &= [K^2 - (\mathbf{K}+\mathbf{g})_t^2]/g^2 \\ &= (\chi_z^2 + U_0 - 2K_y G - G^2)/g^2 \end{aligned} \tag{8}$$

so that (5) becomes

$$[(x+m)^2 - c_{G'}^2]C_{(0,G',mg)}(\mathbf{k}) - \sum_{h\neq g}(U_{g-h}/g^2)C_h(\mathbf{k}) = 0 \tag{9}$$

with $G' = 0$ or G.

Resonance scattering corresponds to the situation in which the wavefield has significant components $(0, 0, mg)$ and $(0, G, ng)$ for some values of m and n. The conditions for resonance to occur can be expressed in terms of c_0 and c_G, as shown below. Bragg conditions correspond to c_0 being an integer.

The eigenvalues of the above system can be determined by the special methods developed by Hill, which leads to the eigenvalues being expressed in terms of the roots of a biquadratic equation (Lamla, 1938; Moon, 1972). Two independent sets of eigenvalues are obtained. Each set consists of complex conjugate pairs which differ from each other by an integer. When resonance is established, the reflection coefficient equals 1 if the Fourier coefficients of the potential are real, and the two sets of eigenvalues are generated from numbers which have equal non-zero imaginary parts and real parts of opposite sign.

In the following it is assumed that the only significant components of the crystal potential are $(0, 0, 0)$, $(0, 0, \pm 2g)$ and $(0, \pm G, \pm g)$. Equation (31) of Appendix A shows that equal imaginary parts occur when $(y_0^2 - y_G^2)^2 + 8A_0 y_0 y_G^2 + 8A_G y_G y_0^2 < 0$. This inequality can be satisfied if $c_0 \simeq 1 - c_G$ when $y_0 \simeq -y_G$ and $A_0 \simeq A_G$. Peng (1994) has discussed the conditions for resonance in terms of the eigenvalues of the scattering matrix. He concludes that the two eigenvalues are equal and therefore there is a singularity in the reflection coefficient. In the present work, the eigenvalues are closely related and there is no anomaly in the reflection coefficient.

Now consider the orientation of the crystal at which $c_0 \simeq 1$. This is the situation considered by Dudarev & Whelan (1994*a*,*b*, 1995, 1997); the results developed here will be used in §4. In this orientation, $y_0 \simeq \pi(c_0 - 1)$ and $y_G \simeq \pi c_G$. To lowest order in the potential coefficient U_{0Gg} and assuming $U_{002g} \ll U_{0Gg}$,

$$\begin{aligned} &2A_0 y_0 y_G^2 + 2A_G y_G y_0^2 \\ &\quad = -\pi^4 (U_{0Gg}/g^2)^2 (1 - c_G^2/4)(1 - c_0). \end{aligned} \tag{10}$$

When $c_0 + c_G \simeq 1$, it is found from (31) that the eigenvalues are approximately

$$\{(c_0 - 1)^2 \pm i(U_{0Gg}/g^2)[(1 - c_G^2/4)(1 - c_0)]^{1/2}\}^{1/2}. \tag{11}$$

The depth of penetration of the electron wave into the crystal is proportional to the inverse of the imaginary part of the eigenvalues.

If absorption is significant, the condition $c_0 + c_G = 1$ cannot be satisfied exactly and it becomes possible to use the first two terms in the binomial expansion for the square root in (31). One of the roots of (31) is approximately y_G^2 and the other root is approximately $y_0^2 + 4A_0 y_0 y_G^2 + 4A_G y_G y_0^2$ and very much less than 1. Further use of the binomial expansion leads to the following expressions for the eigenvalues:

$$x^{(1)} = c_G \tag{12}$$

$$\begin{aligned} x^{(2)} &= (c_0 - 1) - (U_{0Gg}/g^2)^2 \\ &\quad \times (1 - c_G^2/4)(c_0 - 1)/[(c_0 - 1)^2 - c_G^2]. \end{aligned} \tag{13}$$

The $(0, G, -g)$ component of the wavefield associated with the set $x^{(1)}$ is the largest near resonance. The other components are given by

$$\begin{aligned} C^{(1)}_{(002\bar{g})}/C^{(1)}_{(0G\bar{g})} &= \frac{U_{0Gg}^2/(U_{0Gg}g^2) + x^{(1)2} - c_G^2}{U_{002g}/g^2 + [x^{(1)} - 1]^2 - c_0^2} \\ &\quad \times \left(\frac{U_{002g}}{U_{0Gg}}\right) \end{aligned} \tag{14}$$

$$\begin{aligned} C^{(1)}_{(000)}/C^{(1)}_{(0G\bar{g})} &= \frac{U_{0Gg}^2/(U_{0Gg}g^2) + x^{(1)2} - c_G^2}{U_{002g}/g^2 + [x^{(1)} + 1]^2 - c_0^2} \\ &\quad \times \left(\frac{U_{002g}}{U_{0Gg}}\right). \end{aligned} \tag{15}$$

The wavefield associated with $x^{(2)}$ has components given by

$$C^{(2)}_{(002\bar{g})}/C^{(2)}_{(000)} = \frac{U_{002g}/g^2 + [x^{(2)} + 1]^2 - c_0^2}{U_{002g}/g^2 + [x^{(2)} - 1]^2 - c_0^2} \tag{16}$$

$$\begin{aligned} C^{(2)}_{(0G\bar{g})}/C^{(2)}_{(000)} &= \frac{U_{002g}/g^2 + [x^{(2)} + 1]^2 - c_0^2}{U_{0Gg}^2/(U_{0Gg}g^2) + x^{(1)2} - c_G^2} \\ &\quad \times \left(\frac{U_{0Gg}}{U_{002g}}\right). \end{aligned} \tag{17}$$

The amplitude of the specularly reflected wave can be found by matching the solution within the crystal to solutions corresponding to plane waves travelling away from the surface. The reflection coefficient is given by solving a set of four simultaneous equations for the two components of the reflected wave and the amplitudes of the two Bloch waves within the crystal. Away from a resonance, to a good approximation, only the wave corresponding to eigenvalue $x^{(2)}$ is significant for calculating the coefficient of specular reflection. Then the reflection coefficient is given by

$$R_{\rm pot} = [a^{(2)} - b^{(2)}]/[a^{(2)} + b^{(2)}], \quad (18)$$

where

$$a^{(2)} = 1 + C^{(2)}_{(002\bar{g})}/C^{(2)}_{(000)} \quad (19)$$

and

$$b^{(2)} = [1 - x^{(2)} - (1 + x^{(2)})C^{(2)}_{(002\bar{g})}/C^{(2)}_{(000)}]g/\chi_z. \quad (20)$$

Note that $R_{\rm pot}$ depends on the azimuthal angle since the eigenvalue $x^{(2)}$ depends on the parameter c_G. $R_{\rm pot}$ has a maximum near the angle for resonance scattering, but near a resonance, an additional term becomes important. To a good approximation, the total reflection coefficient is

$$R = R_{\rm pot} + \frac{[b^{(1)} - a^{(1)}][1 - x^{(2)}g/\chi_z]}{[a^{(2)} + b^{(2)}][1 - x^{(1)}g/\chi_G]} C^{(2)}_{(0G\bar{g})}/C^{(2)}_{(000)}, \quad (21)$$

where

$$a^{(1)} = [C^{(1)}_{(000)} + C^{(1)}_{(002\bar{g})}]/C^{(1)}_{(00G\bar{g})} \quad (22)$$

and

$$b^{(1)} = \left\{\frac{[x^{(1)} - 1]g}{\chi_z} C^{(1)}_{(000)} + \frac{[x^{(1)} + 1]g}{\chi_z} C^{(1)}_{(002\bar{g})}\right\} \Big/ C^{(1)}_{(00G\bar{g})} \quad (23)$$

and

$$\chi_G^2 = \chi_z^2 - 2\chi_y G - G^2. \quad (24)$$

3. Estimation of many-beam effects by perturbation theory

In order to allow for the effects of the Fourier coefficients of the potential which have not been explicitly considered in the three-beam approximation, we may employ perturbation theory in the way first used by Bethe (*e.g.* Cowley, 1995). An iterative process can be used to calculate the components $C_h(\mathbf{k})$ which have not been explicitly included from the three components that have. Label the three largest components as $(0, 0, 0)$, $(0, 0, -M_0 g)$ and $(0, G, -M_G g)$, where M_0 and M_G are integers corresponding to the reciprocal-lattice points closest to the Ewald sphere (Fig. 1). The first iteration results in the estimates for the amplitudes of the other components, labelled $(0, G', mg)$,

$$C_{(0,G',mg)}(\mathbf{k}) = \sum_h {}^{(3)}(U_{g-h}/g^2)C_h(\mathbf{k})/[(x + m)^2 - c^2_{G'}], \quad (25)$$

where the summation is over the three significant components. The most significant additional components are those which correspond to reciprocal-lattice points close to the Ewald sphere, *i.e.* the $[0, G, -(M_G + 1)g]$, $[0, G, -(M_G - 1)g]$ and $(0, 0, \pm g)$ components. Note that unlike the situation when transmission through a thin foil occurs and the eigenvalues are small enough to be insignificant, in the case of reflection an estimate of the eigenvalues x must be made. The estimate can be made using equations (12) and (13) with the potential coefficients for the bulk. The additional components calculated by (25) are then included in the summation in the scattering equations (9) and result in a set of equations of the same form as the original set but with modified potential coefficients $\bar{U}(g, h)$. Solution of these three-beam equations provides an improved estimate of the amplitudes of the three most important components of the wavefield. Close to the resonance condition, the most significant differences between the modified and original potential coefficients are for $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$, $\bar{U}[(0, G, -M_G g), (0, -M_0 g, -M_G g)]$ and $\bar{U}[(0, 0, 0), (0, G, -M_G g)]$.

The modified potential coefficients lead to the modified parameter $\bar{c}_G$ given by

$$\bar{c}_G^2 = \{\chi_z^2 + \bar{U}[(0, G, -M_G g), (0, G, -M_G g)] - 2K_y G - G^2\}/g^2. \quad (26)$$

4. Application of the theory

An example analysed in some detail by Dudarev & Whelan (1994*a*,*b*) and by Derlet & Smith (1997) is addressed here. These authors considered the resonance scattering produced when 100 keV electrons are incident on a (111) surface of platinum at an angle just off a (100) zone. At a glancing angle of 58.6 mrad, which is slightly less than the angle for satisfying the (888) reflection, a many-beam calculation of the reflection coefficient as a function of azimuthal angle shows a peak at 12.0 mrad of height 0.039 and width at half-maximum of 3.3 mrad. The approximation of Dudarev & Whelan (1994*a*,*b*) when applied to this situation reproduces the many-beam results very closely. [Note, however, that Derlet & Smith (1997) show their results as having a

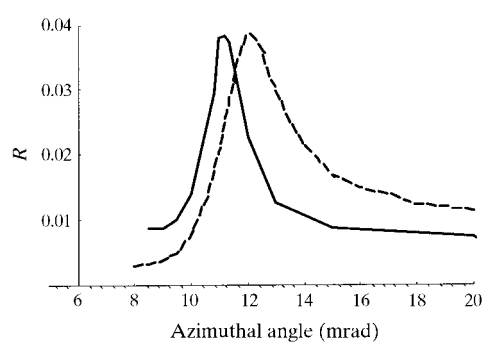

Fig. 2. The coefficient of reflection as a function of azimuthal angle for 100 keV electrons incident on the (111) surface of platinum, calculated using the three-beam approximation, showing a peak when the surface resonance state is established. The dashed line shows the many-beam results of Dudarev & Whelan (1994*a*,*b*).

peak at 10.8 mrad and a height of 0.049 and a width of 2.7 mrad. The present author, on using the formulae of Dudarev & Whelan, has found a peak at 12.1 mrad of height 0.048 and width 3.3 mrad.] The approximation of Dudarev & Whelan (1994*a*,*b*) requires three parameters, which they estimate from many-beam calculations. The relationship of these parameters to the modified potential coefficients introduced in the previous section is discussed below.

Fig. 2 shows the variation of reflection coefficient with respect to azimuthal angle, calculated using equations (12), (13) and (21) with $g = 2\pi(4/d_{111})$ and $G = 2\pi/d_{440}$. The mean inner potential for the bulk was taken to be $(34.34 - i1.89)$ eV and the other Fourier coefficients of the potential are calculated using the formula for atomic scattering factors given by Dudarev & Whelan (1995). Fig. 2 was calculated by first computing the total reflection coefficient R as given by equation (21), then subtracting the coefficient of reflection assuming only scattering of the (888) reflection, and finally adding a constant background of $(-1.3032 - i7.60212) \times 10^{-3}$, as given by Derlet & Smith (1997). The modified potential calculations were performed for each value of the azimuthal angle since they depend on the parameter c_G, which depends on the y component of the incident wavevector. Equation (25) was used with $g = 2\pi/d_{111}$, $G = 2\pi/d_{440}$, $M_0 = 8$ and $M_G = 4$. When the azimuthal angle is about 7.5 mrad, the $(0, G, -5g)$ beam is nearly satisfied and the set of three strong beams has to be redefined. The same situation arises at an azimuthal angle near 5.2 mrad when the $(0, G, -3g)$ beam is approximately satisfied. Fig. 2 shows a peak of height 0.038 and a width of 2.0 mrad centred at 11.1 mrad. Thus, compared with the results of the many-beam calculations, the width is too small and the peak position is in error by 0.9 mrad. The approximate method of Dudarev & Whelan (1994*a*,*b*), using values of the parameters given by these authors, is found to give a peak at the same position and of the same width as given by many-beam calculations, but with a height of 0.048.

The depth of penetration D as a function of azimuthal angle is shown in Fig. 3. It is the inverse of the imaginary part of $2\pi g x^{(2)}$. Fig. 3 also shows the curve obtained using the approximate method of Dudarev & Whelan (1994*a*,*b*). The minima are separated by about 1 mrad. If the value of the parameter controlling peak height in their method is reduced to a value to give a peak height of 0.038, then the curve for the depth of penetration generated with this value is close to the curve shown in Fig. 3 produced by the methods described herein, apart from a shift of 1 mrad.

The positions of the peak in the reflection-coefficient curve and the minimum in the penetration-depth curve depend critically on the real part of $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$. This coefficient can be considered as the average potential experienced by electrons in the resonance state, in which state they are travelling parallel to the surface. Peng *et al.* (1988) and Yao & Cowley (1990) provide a detailed discussion of this point. These authors have estimated the mean potential to be about 70 V by analysing the differences in the positions of the enhanced reflectivity at resonance and the positions of Kikuchi lines, which depend on the mean potential of the bulk. In the approach of Dudarev & Whelan (1994*a*,*b*) it is the parameters ε_0 that determine the position of maximum reflectivity. ε_0 is the energy of a bound state in the potential of (111) planes. Fig. 4 shows how the real part of this coefficient of potential varies with azimuthal angle and also indicates the value of $-\varepsilon_0/e$ which was determined from many-beam calculations. The peaks in the curve are an indication that the assumption that only one Fourier component is sufficient to describe the scattering power along the $(0, G)$ rod breaks down. The curve indicates that the three-beam approximation should be adequate for azimuthal angles greater than about 9 mrad. The corrected potential coefficients differ by up to 200% from the original coefficients. Thus, use of first-order perturbation theory to estimate the coefficients of the modified potential requires that further iterations of the

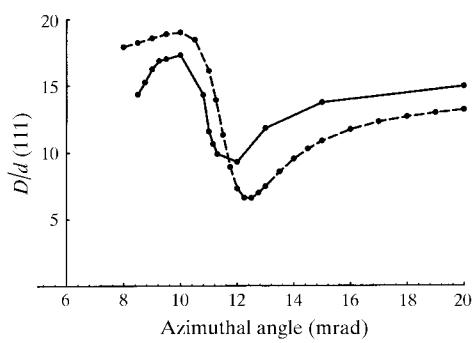

Fig. 3. Variation of depth of penetration (D) of the primary beam, divided by the spacing of the (111) planes, with azimuthal angle. The dashed line shows the approximation of Dudarev & Whelan (1994*a*,*b*).

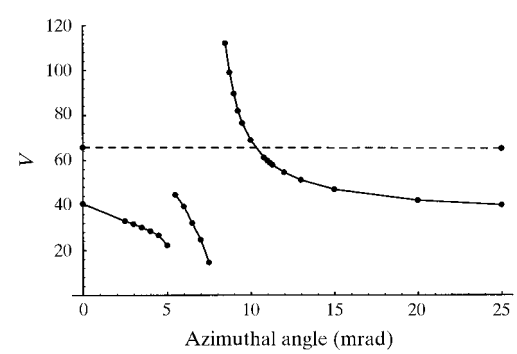

Fig. 4. The real part of the coefficient $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$ as a function of azimuthal angle. Also shown, by the dashed line, is the parameter $-\varepsilon_0/e$ from the approximate method of Dudarev & Whelan (1994*a*,*b*).

scheme for including many-beam effects be carried out. This is not attempted in this paper. However, the results from many-beam computations indicate that first-order perturbation theory has a useful degree of accuracy.

A possible improvement on perturbation theory for estimating this mean potential is to obtain an expression for the scattering by a one-dimensional sinusoidal potential with Fourier coefficient $U_{(00g)}$, an approach similar to that of Dudarev & Whelan (1994*a*,*b*).

The width of the peak is determined by the parameter Γ. Fig. 5 shows how the imaginary part of $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$ varies with azimuthal angle and also shows $\Gamma/2$ estimated from many-beam calculations. The reason that the peak in Fig. 2 is narrower than that obtained by many-beam computations is that the potential coefficient $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$ varies rapidly with azimuthal angle around 9 to 12 mrad. If it is assumed that this is because of the inapplicability of perturbation theory and that it should be a constant value equal to its value at 11.1 mrad, it may be calculated that the reflection coefficient shows a peak of width 3.2 mrad, which is close to the many-beam value.

The height of the peak depends on the modified potential coefficient $\bar{U}[(0, 0, 0), (0, G, -M_G g)]$. In Dudarev & Whelan (1994*a*,*b*), the peak height depends on the parameter Λ, which is related to scattering from a plane wave into the wave propagating parallel to the surface. In Fig. 6, the real part of this potential coefficient is plotted and the value of Λ given by Dudarev & Whelan, as well as that which gives a peak height of 0.038, is indicated.

The comparisons shown in Figs. 4, 5 and 6 suggest that the parameters used by Dudarev & Whelan (1994*a*,*b*) can be interpreted using the standard terminology of diffraction theory. Further justification for interpreting these parameters in this way is obtained by taking the values of ε_0 and Γ given by Dudarev & Whelan and a value of Λ which is 0.78 of their value (to obtain the peak height of 0.038) and using them in the approach described in the present paper. The resulting reflection coefficient has a peak at 12.1 mrad, is of height 0.041 and of width 3.2 mrad, and the plot of the penetration depth against azimuthal angle is very close to that generated by the theory of Dudarev & Whelan (1994*a*,*b*).

Dudarev & Whelan (1997) have proposed that the angles at which resonance occurs can be determined from the energies of the bound states of a one-dimensional potential perpendicular to the crystal surface. Assuming that bound-state energies are related to the real part of $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$, the condition for resonance according to these authors is $\bar{c}_G = 0$. The present analysis shows that while this condition is approximately satisfied when $c_0 \simeq 1$, as in the orientations chosen by Dudarev & Whelan (1997), it is not a necessary condition for resonance.

5. Conclusions

It has been shown that a three-beam approach predicts how the reflection coefficient for fast electrons diffracting under resonance conditions varies with the angle of incidence. A simple expression has been derived for the depth of penetration, as have expressions for the reflection coefficient. These expressions require only simple encoding for evaluation by computer. Provided Bethe's perturbation theory is sufficiently accurate, a full many-beam calculation is not required to estimate the potential coefficients that occur in these expressions. This study has shown that additional insights into electron diffraction by crystal surfaces can be achieved through a three-beam analysis, as Moodie *et al.* (1996) have shown for the transmission case.

APPENDIX *A*

Here we discuss in more detail how the expressions for the eigenvalues were obtained.

The series expansion, in powers of the Fourier coefficients of the potential, of the Hill's determinant

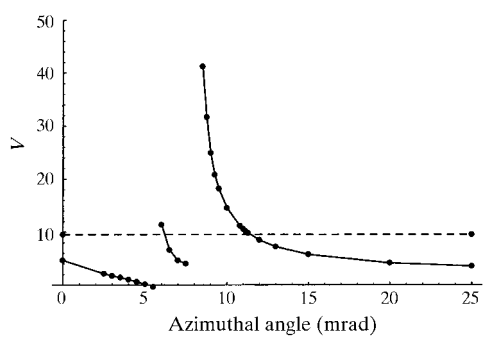

Fig. 5. The imaginary part of the coefficient $\bar{U}[(0, G, -M_G g), (0, G, -M_G g)]$ as a function of azimuthal angle. Also shown, by the dashed line, is the parameter $(\Gamma/2)/e$ from the approach of Dudarev & Whelan (1994*a*,*b*).

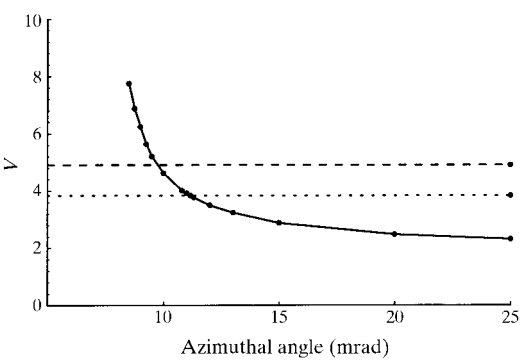

Fig. 6. The real part of the coefficient $\bar{U}[(0, 0, 0), (0, G, -M_G g)]$ as a function of azimuthal angle. Also shown is the real part of the parameter Λ/e from the approach of Dudarev & Whelan (1994*a*,*b*) as estimated by these authors (dashed line) and as estimated by fitting the peak height (dotted line).

associated with the scattering equations was found using the computer algebra package *Mathematica* (Wolfram, 1991).

The infinite determinant associated with the scattering equations (9) can be put into the form of a Hill's determinant after division by $(x+m)^2 - c_{G'}^2$ (Lamla, 1938; Moon, 1972). There are only two independent eigenvalues associated with the scattering matrix implied by (9). All other eigenvalues differ from these two by integers or they are the complex conjugates of these eigenvalues. They are obtained from the roots of the biquadratic equation

$$\begin{aligned} y^4(1 + 2A_0y_0 + 2A_Gy_G) + y^2[-y_0^2 - y_G^2 \\ + 2A_0y_0(1 - y_G^2) + 2A_Gy_G(1 - y_0^2)] \\ + (y_0^2y_G^2 - 2A_0y_0y_G^2 - 2A_Gy_Gy_0^2] = 0, \end{aligned} \quad (27)$$

where $y = \tan \pi x$, $y_0 = \tan \pi c_0$, $y_G = \tan \pi c_G$ and A_0 and A_G depend on the Hill's determinant $\Delta_1(x)$.

$$A_0 = \pi \lim_{x \to c_0} (x - c_0)\Delta_1(x)$$

$$A_G = \pi \lim_{x \to c_G} (x - c_G)\Delta_1(x).$$

Using computer algebra, we find to the lowest orders in U_{00g} and U_{0Gg} that

$$\begin{aligned} A_0 = -\pi \frac{(U_{0Gg}/g^2)^2}{2c_0[(1 - c_0)^2 - c_G^2]} \\ - \pi \frac{(U_{0Gg}/g^2)^2}{2c_0[(1 + c_0)^2 - c_G^2]} \\ - \pi \left\{ \frac{1}{2c_0[(-2 - c_0)^2 - c_0^2]} \right. \\ \left. + \frac{1}{2c_0[(2 - c_0)^2 - c_0^2]} \right\} (U_{00g}/g^2)^2 \end{aligned} \quad (28)$$

$$\begin{aligned} A_G = -\pi \frac{(U_{0Gg}/g^2)^2}{2c_G[(-c_0^2) + (1 + c_G)^2]} \\ - \pi \frac{(U_{0Gg}/g^2)^2}{2c_G[-c_0^2 + (1 - c_G)^2]} \\ - \pi \left\{ \frac{1}{2c_G[(2 + c_G)^2 - c_G^2]} \right. \\ \left. + \frac{1}{2c_G[(2 - c_G)^2 - c_G^2]} \right\} (U_{00g}/g^2)^2. \end{aligned} \quad (29)$$

Since the Fourier coefficients of the potential are small compared with 1, we find that equation (27) can be approximated by

$$\begin{aligned} y^4 + y^2(-y_0^2 - y_G^2) \\ + (y_0^2y_G^2 - 2A_0y_0y_G^2 - 2A_Gy_Gy_0^2) = 0. \end{aligned} \quad (30)$$

The roots of the quadratic equation (27) are

$$\begin{aligned} (1/2)(y_0^2 + y_G^2) \pm (1/2)[(y_0^2 - y_G^2)^2 \\ + 8A_0y_0y_G^2 + 8A_Gy_Gy_0^2]^{1/2}. \end{aligned} \quad (31)$$

I should like to express my gratitude to Professor Alec Moodie for showing me, at the start of my research career, the excitement associated with the field of electron crystallography and high-resolution electron microscopy. I should also like to thank members of the Electron Diffraction Group at Arizona State University, where this work was commenced, for providing a stimulating environment in which to work on quantitative aspects of electron diffraction and microscopy.

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