5.2. Dynamical theory of electron diffraction

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5.2.1. Introduction

Since electrons are charged, they interact strongly with matter, so that the single scattering approximation has a validity restricted to thin crystals composed of atoms of low atomic number. Further, at energies of above a few tens of keV, the wavelength of the electron is so short that the geometry of two-beam diffraction can be approximated in only small unit cells.

It is therefore necessary to develop a scattering theory specific to electrons and, preferably, applicable to imaging as well as to diffraction. The development, started by Born (1926) and Bethe (1928), and continuing into the present time, is the subject of an extensive literature, which includes reviews [for instance: Howie (1978), Humphreys (1979)] and historical accounts (Goodman, 1981), and is incorporated in Chapter 5.1. Here, an attempt will be made to present only that outline of the main formulations which, it is hoped, will help the nonspecialist in the use of the tables. No attempt will be made to follow the historical development, which has been tortuous and not always logical, but rather to seek the simplest and most transparent approach that is consistent with brevity. Only key points in proofs will be sketched in an attempt to display the nature, rather than the rigorous foundations of the arguments.

5.2.2. The defining equations

No many-body effects have yet been detected in the diffraction of fast electrons, but the velocities lie well within the relativistic region. The one-body Dirac equation would therefore appear to be the appropriate starting point. Fujiwara (1962), using the scattering matrix, carried through the analysis for forward scattering, and found that, to a very good approximation, the effects of spin are negligible, and that the solution is the same as that obtained from the Schrödinger equation provided that the relativistic values for wavelength and mass are used. In effect a Klein–Gordon equation (Messiah, 1965) can be used in electron diffraction (Buxton, 1978) in the form

$$\nabla^2 \psi_b + \frac{8\pi^2 m |e| \varphi}{h^2} \psi_b + \frac{8\pi^2 m_0 |e| W}{h^2} \left(1 + \frac{|e| W}{2 m_0 c^2}\right) \psi_b = 0.$$

Here, W is the accelerating voltage and φ, the potential in the crystal, is defined as being positive. The relativistic values for mass and wavelength are given by $m = m_0 (1 - v^2/c^2)^{-1/2}$, and taking 'e' now to represent the modulus of the electronic charge, $|e|$,

$$\lambda = h[2 m_0 e W (1 + e W/2 m_0 c^2)]^{-1/2},$$

and the wavefunction is labelled with the subscript b in order to indicate that it still includes back scattering, of central importance to LEED (low-energy electron diffraction).

In more compact notation,

$$[\nabla^2 + k^2 (1 + \varphi/W)] \psi_b = (\nabla^2 + k^2 + 2 k \sigma \varphi) \psi_b = 0. \quad (5.2.2.1)$$

Here $k = |\mathbf{k}|$ is the scalar wavenumber of magnitude $2\pi/\lambda$, and the interaction constant $\sigma = 2\pi m e \lambda / h^2$. This constant is approximately 10^{-3} for 100 kV electrons.

For fast electrons, φ/W is a slowly varying function on a scale of wavelength, and is small compared with unity. The scattering will therefore be peaked about the direction defined by the incident beam, and further simplification is possible, leading to a forward-scattering solution appropriate to HEED (high-energy electron diffraction).

5.2.3. Forward scattering

A great deal of geometric detail can arise at this point and, further, there is no generally accepted method for approximation, the various procedures leading to numerically negligible differences and to expressions of precisely the same form. Detailed descriptions of the geometry are given in the references.

The entrance surface of the specimen, in the form of a plate, is chosen as the x, y plane, and the direction of the incident beam is taken to be close to the z axis. Components of the wavevector are labelled with suffixes in the conventional way; $\mathbf{K}_0 = \mathbf{k}_x + \mathbf{k}_y$ is the transverse wavevector, which will be very small compared to $\mathbf{k}_z$. In this notation, the excitation error for the reflection is given by

$$\zeta_{\mathbf{h}} = \frac{K_0^2 - |\mathbf{K}_0 + 2\pi \mathbf{h}|^2}{4\pi |\mathbf{k}_z|}.$$

An intuitive method argues that, since $\varphi/W \ll 1$, then the component of the motion along z is little changed by scattering. Hence, making the substitution $\psi_b = \psi \exp\{ik_z z\}$ and neglecting $\partial^2 \psi / \partial z^2$, equation (5.2.2.1) becomes

$$\frac{\partial \psi}{\partial z} = i \left[\frac{1}{2k_z} (\nabla^2_{x,y} + K_0^2) + \sigma \varphi\right] \psi, \quad (5.2.3.1)$$

where

$$\nabla^2_{x,y} \equiv \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2},$$

and $\psi(x, y, 0) = \exp\{i(k_x x + k_y y)\}$.

Equation (5.2.3.1) is of the form of a two-dimensional time-dependent Schrödinger equation, with the z coordinate replacing time. This form has been extensively discussed. For instance, Howie (1966) derived what is essentially this equation using an expansion in Bloch waves, Berry (1971) used a Green function in a detailed and rigorous derivation, and Goodman & Moodie (1974), using methods due to Feynman, derived the equation as the limit of the multislice recurrence relation. A method due to Corones *et al.* (1982) brings out the relationship between the HEED and LEED equations. Equation (5.2.2.1) is cast in the form of a first-order system,

$$\frac{\partial}{\partial z} \begin{pmatrix} \psi_b \\ \frac{\partial \psi_b}{\partial z} \end{pmatrix} = \begin{pmatrix} 0 & 1 \\ -(\nabla^2_{x,y} + k^2 + 2k\sigma\varphi) & 0 \end{pmatrix} \begin{pmatrix} \psi_b \\ \frac{\partial \psi_b}{\partial z} \end{pmatrix}.$$

A splitting matrix is introduced to separate the wavefunction into the forward and backward components, $\psi_b^{\pm}$, and the fast part of the phase is factored out, so that $\psi_b^{\pm} = \psi^{\pm} \exp\{\pm ik_z z\}$. In the resulting matrix differential equation, the off-diagonal terms are seen to be small for fast electrons, and equation (5.2.2.1) reduces to the pair of equations

$$\frac{\partial \psi^{\pm}}{\partial z} = \pm i \left[\frac{1}{2k_z} (\nabla^2_{x,y} + K_0^2) + \sigma \varphi\right] \psi^{\pm}. \quad (5.2.3.2)$$

The equation for $\psi^{\pm}$ is the Lontovich & Fock (1946) parabolic equation.

5.2.4. Evolution operator

Equation (5.2.3.1) is a standard and much studied form, so that many techniques are available for the construction of solutions. One of the most direct utilizes the causal evolution operator. A recent account is given by Gratias & Portier (1983).