

THE ELASTIC SCATTERING OF FAST ELECTRONS

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1. Elastic scattering in High Energy Electron Diffraction (HEED)

Since electrons interact strongly with matter in order to interpret the diffraction patterns and images observed in an electron microscope we nearly always have to solve the Schrödinger equation (Sivardiere: this volume)

$$\left[\nabla^2 + k^2 - U(\underline{r}) \right] \psi(\underline{r}) = 0 \quad (1)$$

without invoking the Born approximation or other weak scattering models. In fact, by writing down a single particle equation such as (1) we have already considerably restricted the scope of our theory because this simple Schrödinger equation with a potential $U(\underline{r})$ having no internal degrees of freedom cannot describe the inelastic scattering of the electron beam by a real solid.

At best, (1), can only describe the elastic scattering if an optical potential is used, i.e.

(i) $U(\underline{r})$ is complex to allow for absorption of the elastically scattered electrons due to the inelastic processes (Dederichs 1972). It is called an optical potential by analogy with the use of a complex dielectric constant in optics or X-ray diffraction.

(ii) The effect of the thermal vibration of the atoms is included not only in $\text{Im}(U(\underline{r}))$ where the possibility of inelastic phonon scattering is catered for, but also by using for the real part of the potential an average $\langle U(\underline{r}) \rangle$ in which each atomic potential is smeared or averaged by convolution with the probability density function for thermal vibrations. In a crystal this is equivalent to multiplying each Fourier coefficient of U by a Debye-Waller factor.

(iii) Some relativistic effects are included because the kinetic energy E of the electron beams usually used in HEED ($E \gtrsim 100$ keV) is comparable to the rest mass energy $m_0 c^2$ of an electron (511 keV). Thus, the wave number k ($= 2\pi/\lambda$) and the mass m of the incident electrons are calculated relativistically:

$$k = (c/\hbar) \sqrt{m^2 - m_0^2}$$

and $m = m_0 (1 + E/m_0 c^2)$. (2)

Since the effective potential in the Schrödinger equation (1) is given by

$$U(\underline{r}) = 2m V(\underline{r})/\hbar^2 \quad (3)$$

in terms of the potential energy $V(\underline{r})$, the strength of U increases as the accelerating voltage of the incident electron beam is raised.

2. Forward scattering approximation

In HEED we want solutions of the Schrödinger equation describing the scattering of a beam of fast electrons which is fired through a thin sample (fig. 1).

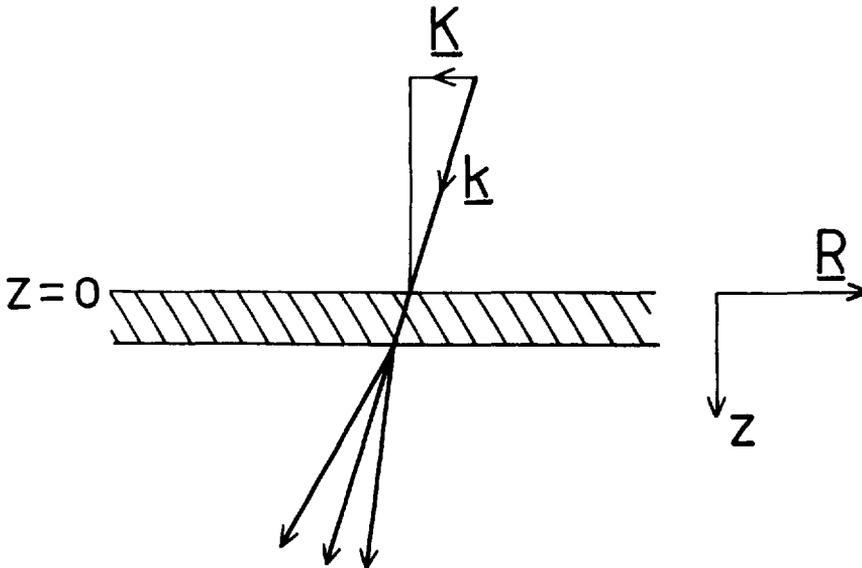


Fig. 1.

As the electron wavelength is very small compared to atomic dimensions ($\lambda < 0.05\text{\AA}$ if $E \geq 100$ keV), the scattering is strongly peaked in the forward direction in HEED and we may simplify (1) if the electrons are fired into the sample at near normal incidence. We let

$$\psi(\underline{r}) = e^{ikz} \tau(\underline{R}, z) \quad (4)$$

and neglect the second derivative $\frac{\partial^2 \tau}{\partial z^2}$ on the assumption that $\tau(\underline{R}, z)$ is slowly varying.

Thus:

$$2ik \frac{\partial \tau}{\partial z} = \left[-\nabla_{\underline{k}}^2 + U(\underline{R}, z) \right] \tau(\underline{R}, z), \quad (5)$$

which is like a 'time' dependent Schrödinger equation. Back scattering is neglected in this approximation so τ satisfies the initial condition at the entrance surface $z = 0$ that it matches onto the incident plane wave, i.e.

$$\tau(\underline{R}, 0) = e^{i \cdot \underline{K} \cdot \underline{R}} \quad (6)$$

where \underline{K} , the transverse component of the incident wave vector \underline{k} , defines the orientation of the incident beam (fig. 1).

3. The phase object approximation

A wide variety of techniques may now be used to solve (5). In the simplest approximation we neglect the $-\nabla_{\underline{R}}^2$ term which represents the transverse kinetic energy of the waves. (5) can then be integrated i.e.

$$\tau(\underline{R}, z) = \exp\left[\frac{-i}{2k} \int_0^z dz' U(\underline{R}, z')\right] \tau(\underline{R}, 0) \quad (7)$$

which is just the eikonal approximation (Glauber 1959). Although it is only valid for moderately thin samples, (7) is a kind of dynamical approximation as the potential U enters non-linearly in contrast to the kinematic (or Born) approximation which is linear in U and may be obtained by expanding the exponential to first order when

$$\frac{1}{2k} \int_0^z dz' U(\underline{R}, z') \ll 1 \quad (8)$$

(7) is then equivalent to the first Born approximation provided the forward scattering assumption is incorporated in the latter and it is further assumed that $\underline{k}^2 z / 2k \ll 1$.

Note that since both U and k increase as m increases, the Born approximation is not a high energy approximation in HEED, but is valid for very thin samples when $\int_0^z dz' U(\underline{R}, z')$ is small. The phase object approximation is therefore often used to indicate the qualitative effects of dynamic diffraction e.g. Cowley (1978) uses it to discuss high resolution imaging in electron microscopy. It should also be noted that (7) can be applied just as easily to disordered or amorphous materials as to crystals and that it is invertable, i.e. given the wave function $\tau(\underline{R}, z)$ we can infer

the projected potential $\int_0^z dz' U(\underline{R}, z')$. Attempts have therefore been made to find explicit formulae similar to (8) but of wider validity. Jap and Glaeser (1978) adopt an approach based on the Feynman path integral which has been discussed by Van Dyck (1975).

4. The multi-slice method

However, the phase object approximation neglects the transverse kinetic energy $-\nabla_{\underline{R}}^2$ in (5) which imparts the wave nature to the propagation of the electrons through the solid and must be included in more sophisticated approximations. One way is to convolute (7) with the Fresnel approximation to the wave propagator as we know that the important secondary wavelets travel in the forward direction almost parallel to z :

$$\tau(\underline{R}, z) = \int d^2 \underline{R}' \left(\frac{k}{2\pi i z} \right) \exp \left[i \frac{k(\underline{R}-\underline{R}')^2}{2z} - \frac{i}{2k} \int_0^z dz' U(\underline{R}', z') \right] \tau(\underline{R}', 0) . \quad (9)$$

This formula, which is easily shown to be a solution of the Schrödinger equation (5) to first order in z (which is assumed to be small) is the basis of the multi-slice method when it is used to describe the evolution of the wavefunction from a plane at depth z_n to $z_{n+1} = z_n + \Delta z$ rather than from 0 to z as above. Propagation through the whole sample is accomplished by dividing it into many such thin slices and repeatedly applying (9).

Although (9) is the form of the multi-slice employed by Ishizuka and Uyeda (1977), a slightly different equation is usually used in which the phase-grating $\exp \left[-i/2k \int_0^z dz' U(\underline{R}, z') \right]$ is evaluated at \underline{R} after the initial wave has propagated to the plane z . This is an equally good solution of the Schrödinger equation (5) to first order in z . Van Dyck (1978) has recently exploited this ambiguity in the order of propagation to obtain a more accurate solution.

The multi-slice has most frequently been used to calculate the amplitudes of the waves diffracted by a perfect crystalline sample when (9) is Fourier transformed and applied in reciprocal space. However, in recent work it has been used to calculate high resolution images of defects in crystals (e.g. : Fields and Cowley 1978, Cowley and Fields 1979).

5. The Howie-Whelan equations

We do not however have to use the multi-slice to calculate the diffracted wave amplitudes: we can follow the spirit of Darwin's original treatment of the dynamical

theory of X-ray diffraction and represent the wavefunction inside a crystalline sample as a sum of diffracted waves:

$$\tau(\underline{R}, z) = e^{-ikz} \sum_{\underline{g}} a_{\underline{g}}(z) e^{i(\underline{k} + \underline{g}) \cdot \underline{r}} \quad (10)$$

If we similarly expand the periodic potential within a crystalline sample in a Fourier series,

$$U(\underline{r}) = \sum_{\underline{g}} U_{\underline{g}} e^{i\underline{g} \cdot \underline{r}} \quad (11)$$

the Schrödinger equation (5) will be satisfied if the amplitudes $a_{\underline{g}}$ change with depth z according to:

$$\frac{da_{\underline{g}}}{dz} = -is_{\underline{g}} a_{\underline{g}} - i/2k \sum_{\underline{g}'} U_{\underline{g}-\underline{g}'} a_{\underline{g}'}, \quad (12)$$

where, the excitation error

$$s_{\underline{g}} = [(\underline{K} + \underline{G})^2 - K^2] / 2k + g_z \quad (13)$$

has been introduced. (\underline{G} is the component of \underline{g} \perp to z , g_z its component \parallel to z). $s_{\underline{g}}$ is the distance in the z direction from the Ewald sphere to the reciprocal lattice point \underline{g} (fig. 2).

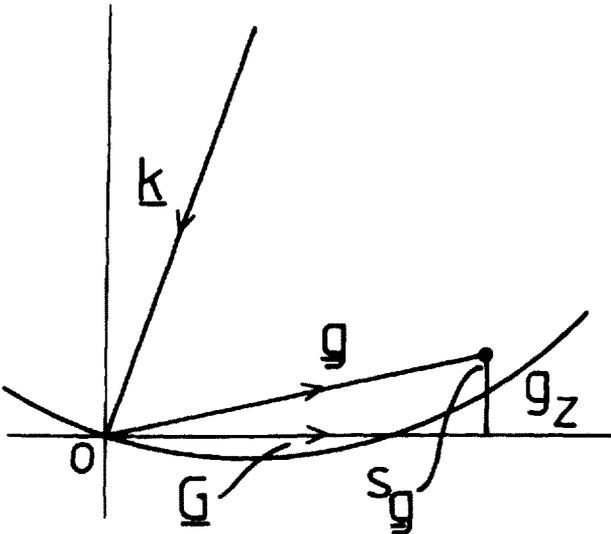


Fig. 2.

The excitation error $s_{\underline{g}}$. For clarity the curvature of the Ewald sphere has been much exaggerated.

It should be noted that (12) cannot be derived from the Schrödinger equation (5) because (11) is only valid for z within the sample (U vanishes outside the crystal). In general, there are too many diffracted wave amplitudes $a_{\underline{g}}$ to be uniquely determined by (5), but (12) is a convenient choice which ensures that $\tau(\underline{R}, z)$ satisfies the Schrödinger equation. Similarly, we choose

$$a_{\underline{g}}(0) = \delta_{\underline{g}, 0} \quad (14)$$

which automatically ensures that $\tau(\underline{R}, 0)$ is equal to the incident plane wave at $z = 0$.

It is well known that the Howie-Whelan equations (12) can be integrated analytically if only two diffracted wave amplitudes are included (the incident beam a_0 and one other strong reflexion) to obtain the 'pendulum' solution.

$$a_0 = \exp\left[\frac{-iz}{2} (s_{\underline{g}} + U_0/k)\right] \left\{ \cos\left(\frac{\pi z}{\xi_{\underline{g}}}\right) + \frac{is_{\underline{g}}\xi_{\underline{g}}}{2\pi} \sin\left(\frac{\pi z}{\xi_{\underline{g}}}\right) \right\} \quad (15)$$

$$a_{\underline{g}} = \frac{-iU_{\underline{g}}\xi_{\underline{g}}}{2\pi k} \exp\left[\frac{-iz}{2} (s_{\underline{g}} + U_0/k)\right] \sin\left(\frac{\pi z}{\xi_{\underline{g}}}\right), \quad (15)$$

in which intensity is 'periodically' transferred from the incident beam to the diffracted beam and back again in the extinction distance,

$$\xi_{\underline{g}} = 2\pi / \sqrt{s_{\underline{g}}^2 + U_{\underline{g}}U_{-\underline{g}}/k^2} \quad (16)$$

Of more interest here is the fact that (12) can be formally integrated

$$\underline{a}(z) = \exp\left[\frac{-izM}{2k}\right] \underline{a}(0) \quad (17)$$

(see for example Hirsch et al 1965), by writing the amplitudes $a_{\underline{g}}$ as a column vector \underline{a} and utilizing the scattering matrix

$$S = \exp\left[\frac{-izM}{2k}\right] \quad (18)$$

where M is the matrix of the coefficients in the Howie-Whelan equations (12). The matrix exponential in (18) can be evaluated from its series expansion, or recursively, or by using the Bloch wave method described in the next section. However, we should first note that the scattering matrix itself has been used in the calculation of electron microscope images of defects (Hirsch et al 1965).

6. Bloch waves and the dispersion surface

As mentioned above, we could introduce the Bloch waves as a formal device for evaluating the scattering matrix, but let us instead return to the physics. It is known from solid state physics that electrons in a crystal do not propagate as plane waves (which are scattered by the periodic potential as (12) shows) but as Bloch

waves $\psi_j(\underline{r})$ each of which can be written as a sum of plane waves

$$\psi_j(\underline{r}) = \sum_{\underline{g}} c_{\underline{g}}^{(j)} e^{i(\underline{k}^{(j)} + \underline{g}) \cdot \underline{r}} \quad (19)$$

Since we are considering elastic scattering the Bloch waves excited in the crystal all have the same energy k^2 and, to match onto the incident plane wave at $z = 0$ they all have the same transverse component of the Bloch wavevector $\underline{k}_{\perp}^{(j)} = \underline{K}$, but the z components of the wavevectors $k_z^{(j)}$ differ in general. The loci of the wavevectors $k_z^{(j)}$ as functions of \underline{K} or the incident beam orientation form the dispersion surface. We could calculate the dispersion surface and the Bloch waves by substituting (19) in the original Schrödinger equation (1), but we really only want those solutions which correspond to waves propagating almost parallel to the z axis so it is easier to use (5) which neglects back-scattering even though from (5) the curvature of the dispersion surface is only approximately correct. To calculate the Bloch waves we assume that (11) is valid both inside and outside the sample. Thus, if we let

$$\gamma_j = k_z^{(j)} - k_z \approx k_z^{(j)} - k + \underline{K}^2/2k \quad , \quad (20)$$

we find that

$$\sum_{\underline{g}} \left[(\gamma_j + s_{\underline{g}}) \delta_{\underline{g} \underline{g}'} + \frac{1}{2k} U_{\underline{g} - \underline{g}'} \right] c_{\underline{g}}^{(j)} = 0 \quad (21)$$

These are the many-beam equations which, being of standard eigenvalue-eigenvector form can be solved easily on a computer. The only problem is that it may be necessary to use a rather large matrix ($\sim 100 \times 100$) to obtain accurate results, especially for heavy materials at high voltages or when the incident beam is nearly parallel to a low index zone axis of the crystal. In the latter case there are many strong reflexions in the zero Laue zone with small excitation errors $s_{\underline{g}}$ which must be included in the many-beam matrix. However, the major asset of the Bloch wave method is not that it is a particularly efficient computational scheme (for large calculations the multi-slice is usually faster and can handle many more reflexions) but, the insight it provides via the intermediate concept of the Bloch waves which propagate independently through the crystal. The electron wavefunction is thus given by a sum of Bloch waves

$$\psi(\underline{r}) = \sum_j \epsilon_j \psi_j(\underline{r}) \quad (22)$$

with constant excitation amplitudes ϵ_j determined by matching (22) to the incident wave at $z = 0$. The diffracted wave amplitudes may be obtained by substituting (19) into (22) and comparing with (10) from which we see that the diffracted wave

intensities are largely determined by interference of the Bloch waves excited in the crystal.

Finally, let us note that the $\text{Im}(U(\underline{r}))$ is small so that absorption effects can usually be included by perturbation theory. The eigenvectors obtained from the many-beam equations (21) are then orthogonal and we can evaluate the excitation amplitudes:

$$\epsilon_j = C_0^{(j)*} \quad (23)$$

However, the eigenvalues γ_j and the Bloch wavevectors $k_z^{(j)}$ become complex when $\text{Im}(U(\underline{r}))$ is included showing that, due to the inelastic scattering, the elastically scattered intensity decreases as the waves propagate through the crystal.

7. Conclusion

We can connect the approaches described here by noting that in the Bloch wave method we are effectively calculating the scattering matrix S by diagonalizing the matrix M . It is not surprising therefore that the many-beam equations (21) can be solved when only two plane waves (C_0 and $C_{\underline{g}}$) are included in the expansion of the Bloch wave (19):

$$\gamma_{\pm} = -1/2 \left[s_{\underline{g}} + U_0/k \mp \sqrt{s_{\underline{g}}^2 + U_{\underline{g}} U_{-\underline{g}}/k^2} \right] \quad (24)$$

and, if we ignore $\text{Im}(U(\underline{r}))$, the eigenvectors are given by:

$$\left. \begin{aligned} C_0^{(\pm)} &= \frac{1}{\sqrt{2}} \sqrt{1 \mp s_{\underline{g}} \xi_{\underline{g}}/2\pi} \\ C_{\underline{g}}^{(\pm)} &= \pm \frac{e^{i \arg(U_{\underline{g}})}}{\sqrt{2}} \sqrt{1 \pm s_{\underline{g}} \xi_{\underline{g}}/2\pi} \end{aligned} \right\} \quad (25)$$

The dispersion surface is thus obtained in the familiar two beam approximation as sketched in fig. 3 near the Brillouin zone boundary perpendicular to \underline{g} . Like the phase object approximation this simple approximation has also been extensively used to discuss qualitatively the effects produced by dynamical diffraction in electron microscopy. However, it should be emphasized that the usefulness of the Bloch wave concept goes far beyond the two beam theory, for example in the explanation of channelling effects, the critical voltage effect, higher order Laue zone diffraction effects or the multiple refraction effects observed when steep wedge shaped crystal specimens are used.

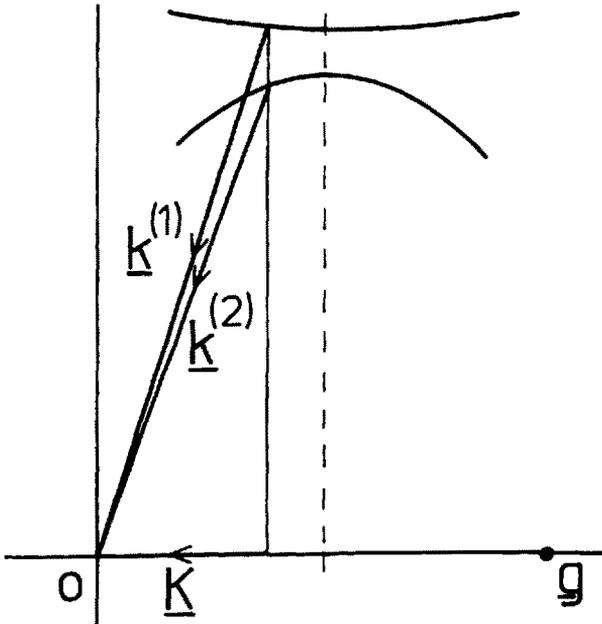


Fig. 3. The two beam dispersion surface (the curvature is exaggerated for clarity).

Finally, it should be noted that low energy electron diffraction has not been discussed at all here. A different kind of dynamical theory is usually used in LEED in which the scattering of the electron waves by the atoms in a solid is described by using the phase shifts as discussed by Sivardiere and these scattered waves are in turn themselves scattered by other atoms in the sample. The details of this multiple scattering cannot be discussed here (Pendry 1974), but let us note that such an approach would not be fruitful in HEED because of the large number of phase shifts that would be required to describe the scattering of the 100 keV electrons by an atom.

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