K8 Two beam theory

We will start from the Schroedinger equation for the electron travelling through the solid,

$$\nabla^2\psi(\underline{\mathbf{r}}) + (8\pi^2 me/h^2)[\, E + V(\underline{\mathbf{r}})\,]\psi(\underline{\mathbf{r}}) = 0 \qquad \text{K8.1}$$

We know that in electron diffraction the scattering angles of the electron are in general small. It is therefore reasonable to factorize out the wavevector of the incident wave (taken along the z-axis as before) and write

$$\psi(\underline{\mathbf{r}}) = \phi(\underline{\mathbf{r}})\exp(2\pi ikz) \qquad \text{K8.2}$$

We now have a wavefunction $\phi(\underline{\mathbf{r}})$ which will be slowly varying as it goes through the crystal. Substituting this form into equation K4.1 we obtain (using e for the electron charge and dropping the negative sign)

$$\{-4\pi k^2\phi(\underline{\mathbf{r}}) + 4\pi i k d\phi(\underline{\mathbf{r}})/dz + d^2\phi(\underline{\mathbf{r}})/dz^2 + \nabla_r{}^2\phi(\underline{\mathbf{r}})$$

$$+ (8\pi^2 me/h^2)[\, E + V(\underline{\mathbf{r}})\,]\phi(\underline{\mathbf{r}})\}\exp(2\pi i k z) = 0 \qquad \text{K8.3}$$

where

$$\nabla_r{}^2\phi(\underline{\mathbf{r}}) = d^2\phi(\underline{\mathbf{r}})/dx^2 + d^2\phi(\underline{\mathbf{r}})/dy^2 \qquad \text{K8.4}$$

Remembering that

$$(8\pi^2 me/h^2)E = 4\pi k^2 \qquad \text{K8.5}$$

and neglecting the term $d^2\phi(\underline{\mathbf{r}})/dz^2$ on the basis that k is fairly large, to obtain the equation

$$d\phi(\underline{\mathbf{r}})/dz = - \{(i/4\pi k)\nabla_r^2 + (2\pi i m e/h^2 k)V(\underline{\mathbf{r}})\}\phi(\underline{\mathbf{r}}) \qquad \text{K8.6}$$

Equation K8.6 is mathematically the same as the equations that are solved in the Kinematical theory, and as yet we have made only one small justifiable approximation (neglecting the second derivative term in z). Before we proceed any further, it is informative to consider the physical sense of equation K4.6. The wavefunction $\phi(\underline{\mathbf{r}})$, really the wave with the swiftly varying z dependence stripped away, changes as it moves with z through the specimen; in effect the electron travels down through the specimen. How the electron changes depends upon two different terms. The first one, $(i/4\pi k)\nabla_r^2$ is rather like a diffusion term. The spirit of this term is therefore to spread the wavefunction in the x,y plane as it travels. The second term contains all the scattering of the wave by the specimen potential. Comparing the magnitude of the two, with a typical estimate of $eV(\underline{\mathbf{r}})$ of 20 eV,

$$(2\pi i m e/h^2 k)V(\underline{\mathbf{r}})/(1/4\pi k) = 8\pi^2 m e V(\underline{\mathbf{r}})/h^2 \sim 10 \text{ Å}^{-2} \qquad \text{K8.7}$$

Therefore unless the wave is changing very fast in the x,y plane, which only occurs when we have to consider large scattering vectors, the second term is substantially larger than the first, and this effect will become more important at higher voltages as the mass increases. ***This is a very important point.*** Because of relativistic effects at relatively high energies, the scattering by the potential becomes ***stronger relative to the transverse "diffusion"*** of the electrons. A simple mistake that is often made (by the uninitiated) is that at high energies the interaction of the electron is weak, so simple models can be used – due to relativistic effects this is not in fact the case.

If we ignore the first term, we are in effect ignoring sideways spreading of the information in the electron, in effect the column approximation. Let us now write

$$\phi(\underline{\mathbf{r}}) = \Sigma\ \phi_g(z)\exp(2\pi i[\underline{\mathbf{g}}.\underline{\mathbf{r}} - s_z(\underline{\mathbf{g}})z]) \qquad \text{K8.8}$$

Then

$$d\phi(\underline{\mathbf{r}})/dz = \sum_{g} \{d\phi_g(z)/dz+i[4\pi^2 g^2/4\pi k-2\pi s_z+2\pi g_z]\, \phi_g(z)\}\exp(2\pi i[\underline{\mathbf{g}}.\underline{\mathbf{r}} -s_z(\underline{\mathbf{g}})z]) \quad \text{K8.9}$$

$$= (2\pi i m e/h^2 k)V(\underline{\mathbf{r}}) \sum_{g} \phi_g(z)\exp(2\pi i[\underline{\mathbf{g}}.\underline{\mathbf{r}} -s_z(\underline{\mathbf{g}})z]) \quad \text{K8.10}$$

The term inside the square brackets "**[ ]**" is small to zero; neglecting it is equivalent to invoking a column approximation. Using:

$$V(\underline{\mathbf{r}}) = \sum_{\mathbf{q}} \exp(2\pi i\underline{\mathbf{q}}.\underline{\mathbf{r}})\, V(\underline{\mathbf{q}}) \quad \text{K8.11}$$

(it is a sum, so it does not matter if we use **g** or **q**) and

$$\xi_g = 1/\{ (2me/h^2 k)\, V(\underline{\mathbf{g}}) \} \quad \text{K8.12}$$

Then

$$\sum_{g} d\phi_g(z)/dz \exp(2\pi i[\underline{\mathbf{g}}.\underline{\mathbf{r}} - s_z(\underline{\mathbf{g}})z] = \sum_{g}\sum_{q} (\pi i/\xi_q)\, \phi_g(z)\exp(2\pi i[\{\underline{\mathbf{g}}+\underline{\mathbf{q}}\}.\underline{\mathbf{r}} - s_z(\underline{\mathbf{g}})z]) \qquad \text{K8.13}$$

We next note that the left-hand side contains an exponential with "$\underline{\mathbf{g}}.\underline{\mathbf{r}}$" while the right contains "$\{\underline{\mathbf{g}}-\underline{\mathbf{q}}\}.\underline{\mathbf{r}}$". This equation must be true for all (x,y), which means that these two must be the same for each individual term. We can do this by replacing "$\underline{\mathbf{g}}$" by "$\underline{\mathbf{g}}-\underline{\mathbf{q}}$" on the right, i.e.:

$$\sum_{g} d\phi_g(z)/dz \;\exp(2\pi i[\underline{\mathbf{g}}.\underline{\mathbf{r}} - s_z(\underline{\mathbf{g}})z]) = \sum_{g}\sum_{q} (\pi i/\xi_q)\, \phi_{g-q}(z)\exp(2\pi i[\underline{\mathbf{g}}.\underline{\mathbf{r}} - s_z(\underline{\mathbf{g}}-\underline{\mathbf{q}})z] \qquad \text{K8.14}$$

and now eliminating $\exp(2\pi i\underline{\mathbf{g}}.\underline{\mathbf{r}})$ from both sides

$$d\phi_g(z)/dz = \sum_{q} (\pi i/\xi_q)\, \phi_{g-q}(z) \exp(2\pi i \{ s_z(\underline{\mathbf{g}}-\underline{\mathbf{q}}) - s_z(\underline{\mathbf{g}})\} z) \qquad \text{K8.15}$$

These are what Williams and Carter call the "Howie-Whelan" equations. To understand them, note that the left-hand side is the change in the (complex) amplitude of a given diffracted beam as a function of depth, the second term being a phase change. It is easy to check that this second phase term ensures that the Ewald sphere curvature effect is taken into account. On the right of this equation we have scattering from $\phi_{g-q}(z)$ into $\phi_g(z)$ as a function of depth, with a Ewald sphere curvature term. Taking the simple case where we assume that $\phi_{g-q}(z)$ is very small unless **g**=**q** (Kinematical model) we have:

$$d\phi_g(z)/dz \quad = \quad (\pi i/\xi_g) \;\; \phi_0(z) \exp(-2\pi i \; s_z(\mathbf{g}) \; z) \qquad \text{K8.16}$$

This will reduce down to Kinematical theory, albeit in a slightly different form since equation K8.8 used a slightly different definition than that which was used in section K3. If instead we assume that there are only two beams (reciprocal lattice values) that are strong and of interest to us, we can add a second equation to K8.16, namely

$$d\phi_0(z)/dz \quad = \quad (\pi i/\xi_{-g}) \;\; \phi_g(z) \exp(-2\pi i s_z(-\mathbf{g})\} \; z) \qquad \text{K8.17}$$

K9 Two Beam Solutions

Our task is to solve the two equations

$$d\phi_g(z)/dz \quad = \quad (\pi i/\xi_g) \;\; \phi_0(z) \exp(-2\pi i \; s_z(\mathbf{g}) \; z) \qquad \text{K9.1}$$

$$d\phi_0(z)/dz \quad = \quad (\pi i/\xi_{-g}) \;\; \phi_g(z) \exp(-2\pi i s_z(-\mathbf{g})\} \; z) \qquad \text{K9.2}$$

Writing K9.2 as

$$\exp(-2\pi i s_z z) d\phi_o(z)/dz = \quad (\pi i/\xi_g)\phi_g(z) \qquad \text{K9.3}$$

and then differentiating with respect to z (and dropping the "**g**" for the excitation error) we obtain

$$\exp(-2\pi i s_z z)\{ \; d^2\phi_o(z)/dz^2 - 2\pi i s_z d\phi_o(z)/dz \; \}$$

$$= \;\; (\pi i/\xi_g) d\phi_g(z)/dz \qquad \text{K9.4}$$

Substituting for ϕ_g from equation K9.1 we obtain

$$d^2\phi_o(z)/dz^2 - 2\pi i s_z d\phi_o(z)/dz + (\pi/\xi_g)^2\phi_o(z) = 0 \qquad \text{K9.5}$$

This is a fairly simple differential equation which has a general solution of the form

$$\phi_o(z) = \exp(2\pi i\alpha z) \qquad \text{K9.6}$$

where

$$4\alpha^2 - 4\alpha s_z + (1/\xi_g)^2 = 0 \qquad \text{K9.7}$$

which has roots for α of

$$\alpha = (s_z \pm \sqrt{[s_z^2+1/\xi_g^2]})/2 \qquad \text{K9.8}$$
$$= (s_z \pm s^{eff})/2 \qquad \text{K9.9}$$

where we are introducing the ***effective excitation error*** $\boldsymbol{s^{eff}}$ here which plays the same role in two-beam theory as the excitation error in Kinematical theory.

Our general solution for $\phi_o(z)$ is therefore

$$\phi_o(z) = C_o^{+}\exp(\pi iz(s_z+s^{eff})) + C_o^{-}\exp(\pi iz(s_z-s^{eff})) \qquad \text{K9.10}$$

where C_o^{+} and C_o^{-} are constants which we have to determine from our boundary conditions, i.e. the incoming electron wave. If we substitute back with our solutions we obtain a very similar equation for the diffracted beam, i.e.

$$\phi_g(z) =(\pi^2\xi_g)(s_z+s^{eff})C_o^{+}\exp(-\pi iz(s_z-s^{eff}))$$

$$(\pi^2\xi_g)(s_z-s^{eff})C_o^{-}\exp(-\pi iz(s_z+s^{eff})) \qquad \text{K9.11}$$

To complete our solution, we need to determine our C constants. ***To do this we use the fact that on the entrance surface of the crystal the wave within the crystal must match the incident wave on the entrance surface***, i.e.

$$\phi_o(0) = 1 = C_o^{+} + C_o^{-} \qquad \text{K9.12}$$

so that the incident wave has a value of 1 on the entrance surface z=0, and forcing the diffracted beam to have no amplitude on the incident surface we obtain:

$$\phi_g(0) = 0 = (\pi^2\xi_g)\{ (s_z+s^{eff})C_o^{+} + (s_z-s^{eff})C_o^{-} \} \qquad \text{K9.13}$$

substituting in for C_o^- we have

$$C_o^{+} \{(s_z+s^{eff}) - (s_z-s^{eff}) \} = - (s_z-s^{eff}) \qquad \text{K9.14}$$

which reduces to

$$C_o^{+} = (1 - s_z/s^{eff})/2 \qquad \text{K9.15}$$

$$C_o^{-} = (1 + s_z/s^{eff})/2 \qquad \text{K9.16}$$

Using these specific values, we obtain for the wave amplitudes

$$\phi_o(z) = (1 - s_z/s^{eff})/2\exp(\pi iz(s_z+s^{eff}))$$

$$+ (1 + s_z/s^{eff})/2\exp(\pi iz(s_z-s^{eff})) \qquad \text{K9.17}$$

which simplifies to:

$$\phi_o(z) = \exp(\pi izs_z)\{\cos(\pi s^{eff}z) - (is_z/s^{eff})\sin(\pi s^{eff}z)\} \qquad \text{K9.18}$$

and for the diffracted beam:

$$\phi_g(z) = (i\exp(-\pi is_z z)/\xi_g s^{eff})\sin(\pi s^{eff}z) \qquad \text{K9.19}$$

The intensity of the diffracted beam is therefore

$$|\phi_g(z)|^2 = (1/\xi_g)^2\ \{\sin(\pi s^{eff}z)/s^{eff}\}^2 \qquad \text{K9.20}$$

with $|\phi_o(z)|^2 = 1 - |\phi_g(z)|^2$ K9.21

The result that we have is qualitatively very similar to that of the Kinematical theory, with an effective excitation error replacing the true excitation error. As before, the intensity oscillates as a function of the crystal thickness, but whereas this did not occur for the exact Bragg condition in Kinematical Theory it now always occurs. In addition we see that the intensity of the incident beam is complimentary to that of the diffracted beam, what we invoked as an ad hoc correction in our earlier analysis.

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Bloch Waves

Going back to the original equation

$$\nabla^2\psi(\underline{\mathbf{r}}) + (8\pi^2 me/h^2)[\, E + V(\underline{\mathbf{r}})\,]\psi(\underline{\mathbf{r}}) = 0 \qquad \text{K8.1}$$

You can also solve this using a Bloch wave

$$b(\underline{\mathbf{k}},\underline{\mathbf{r}}) = Co \exp(2\pi i\gamma z)[\, \exp(2\pi i\underline{\mathbf{k}}.\underline{\mathbf{r}}) + 2\gamma\xi g \exp(2\pi i[\underline{\mathbf{k}}+\underline{\mathbf{g}}].\underline{\mathbf{r}})]$$

At the 2-beam condition

$$sz=0, \quad \gamma= \pm 1/2\xi g$$

$$b(k,r) = Co \exp(2\pi i\gamma z)[\, \exp(2\pi i\underline{\mathbf{k}}.\underline{\mathbf{r}}) \pm \exp(2\pi i[\underline{\mathbf{k}}+\underline{\mathbf{g}}].\underline{\mathbf{r}})]$$