**Bloch waves and many beam theory**

**1. General**

So much of the theory of electron diffraction depends upon Bloch waves that it is useful to explore some of their properties in a little explicit detail. The basic concept of a Bloch wave stems from a theorem called Bloch's theorem which states that any standing wave in a periodic solid must have the form

ψ(r) = exp(2πik.r) b(r) B1.1

where b(r) is a function which has the symmetry of the crystal lattice, i.e. must look the same at all points separated by translations vectors of the lattice as sketched in >>> Figure 1. Just as a plane wave looks the same (except for a phase term) in vacuum since all points in vacuum are the same, in a crystal a wave must look the same at identical points in the periodic crystal lattice (again, except for a phase term.) Thus

b(r) = b(r+la+mb+nc) B1.2

where a, b and c are the lattice translation vectors. The form

b(r) = Σ Cg exp(2πig.r) B1.3

g

satisfies this condition, e.g.

b(r+la+mb+nc) = Σ Cgexp(2πig.(r+la+mb+nc)) B1.4

g

= Σ Cgexp(2πig.r) B1.5

g

since g.(la+mb+nc) = an integer from the definition of the reciprocal lattice vectors.

In general there is more than one possible value for the wavevector k for a given energy of the electron, with a different set of coefficients for each particular value. We write each particular solution (general crystal wave) in the form

φ(r,kj) = exp(2πikj.r)b(r,kj) B1.6

with

b(r,kj) = Σ Cjg(kj)exp(2πig.r) B1.7

g

(The specific dependence on kj is often omitted.) Here kj is the wavevector of our crystal or Bloch wave, analogous to the wavevector of a plane wave in vacuum. Conventionally we order the Bloch waves in terms of decreasing values of kj, i.e. k1 > k2 > kn. The family of different values of kj (as we vary the momentum normal to the crystal zone axis) for a specific value of j is called a branch, and we refer to the plot of the values kj as the dispersion surface, each family therefore corresponding to one branch of the dispersion surface. An alternative way of thinking of the different solutions is as different bands for the high energy electron in the solid, degenerate bands since they all have the same energy. Some of these bands have more potential energy and are more concentrated in the deep attractive potential around the atoms, while others have more kinetic energy and are concentrated more between the atoms. The most general solution for the electron wave is as some combination of these Bloch waves, the excitation of each Bloch wave depending upon the boundary value at the entrance surface where we must match the wave amplitude and its derivative across the surface to that of the incoming wave so that there are no jumps or discontinuities in the wave and so that the momentum across the crystal boundary is conserved. We cannot have any sudden jumps in the amplitude as this would correspond to losing or gaining electrons, and the flux across the boundary inwards from the vacuum must be equal to the flux away from the boundary inside the crystal.

To determine the values of the (currently unknown) Bloch wavevectors and the coefficients Cjg(kj) we insist that the Bloch waves satisfy the standard Schroedingers equation,

∇2φ(r,kj) + (8π2me/h2)[ E + V(r) ] φ(r,kj) = 0 B1.8

Substituting in the form for the Bloch wave, we have what is called the secular equation for the coefficients Cjg (after collecting terms with common exponential terms and expanding the crystal potential as a Fourier series V(r)= ΣVgexp(2πig.r))

{ -4π2(kj+g)2 + (8π2me/h2)E}Cjg(kj) + ΣVg-hCjh(kj) = 0 B1.9

h

This is a matrix equation which can be solved analytically in a few simple cases, more generally is solved numerically. Using the notation

k2 = 2me(E+Vo)/h2 B1.10

where Vo is the mean inner potential of the crystal and k a free electron wavevector corrected for the mean inner potential (a change of order 10-5 from the wavevector in vacuum), it is convenient to write B1.9 as a matrix equation, i.e.

**D** C = 0 B1.11

where C is the vector with coefficients Cjg i.e.

C = (Cjo, Cjg, Cj2g, ... ) B1.12

and **D** is a matrix with diagonal elements

Dgg = k2 - (kj + g)2 B1.13

and off diagonal elements

Dgh = 2meVg-h/h2 B1.14

For there to be non trivial solutions, the determinant of **A** must be zero. (Trivial solutions are when C = 0.) When multiplied out this gives a polynomial of order 2n in kj where n is the number of different Fourier coefficients of the potential included in the calculation (also the number of diffraction beams). Solving this equation will give us 2n different possible values of kj, possible Bloch wavevectors. Half of these solutions will represent waves going backwards through the crystal, which can be discarded (at high energes). This will leave us with n possible Bloch waves. We can then go back to equation B1.11 and solve for the particular values of Cjg(kj). The final solution will be some sum of these different possible Bloch waves, i.e.

ψ(r) = Σ Aj φ(r,kj) B1.15

j

where the coefficients Aj as mentioned above have to be obtained by matching the wave amplitude and the first derivative across the entrance surface of the crystal.

**2. High Energy Solution**

The general matrix equation B1.9 as it stands has no particularly simple solution. However, in the case when the incident beam is normal to the entrance surface of the crystal it can be substantially simplified. Before we look at this simplified High Energy solution, we should not one feature about the boundary conditions. Taking the entrance surface to be normal to the z direction, if the incoming wave has the form

ψ(r) = exp(2πiχ.r) B2.1

then on the entrance surface

exp(2πiχ.[x + y ]) = Σ Aj b(z=0,x,y,kj) B2.2

j

= Σ Aj Σ Cjg(kj)exp(2πi(kj+g).(x+y))

j g B2.3

(We are neglecting reflection which for the case that we are considering here will be small. We will return to this later.) The only way that equation B2.3 can balance for all possible different values of x and y is if the wavevectors tangential to the entrance surface match, i.e.

χx = kjx ; χy = kjy B2.4

Returning to the matrix equation B1.9 we write

(8π2me/h2)(E+Vo) - 4π2(kj+g)2 = 4π2(k2 - (kj+g)2) B2.5

where k is the wavevector corrected for the mean inner potential, and then assuming that kj is along the z axis and perpendicular to g, we can approximate (following Metherell, in "Electron Microscopy in Materials Science", Part 2, Editors U.Valdre and E. Ruedl, Commission of the European Communities, 1973)

k2 - (kj+g)2 = k2 -kj2z - G B2.6

where G = (χx2 + χy2 + 2χxgx + 2χygy + gx2 + gy2 ) B2.7

and we have built in our boundary conditions at the entrance surface by using χx and χy in equation B2.7. The matrix equation can now be written as an eigenvalue problem. i.e

**D** C- kj2 C= 0 B2.8

where C is the vector that was defined in section 1 above and **D** is a matrix with diagonal elements

Dgg = k2 - G B2.9

and off diagonal elements

Dgh = (2me/h2)Vg-h B2.10

Some numerical method in general will give us both the eigenvalues (Cjg(kj) coefficients ) and eigenvectors kj2. We have in fact identical solutions for kj positive and negative. Physically the negative solutions correspond to waves propagating backwards through the lattice which we know physically cannot be of interest to us so we only need to consider the positive solutions.

**3. Orthonormality**

When evaluating the boundary conditions and for scattering approaches to the diffraction of electrons by defects we often make use of orthonormality conditions between the different Bloch waves to simplify the algebra. Strictly speaking this is a consequence of the high energy approximation that we have used above in the case when any absorption of the electrons by the crystal is neglected. In this case the matrix **D** is Hermitian, i.e.

Dgh = D\*hg B3.1

As shown in many mathematical textbooks, the eigenvalues of a Hermitian matrix are real and the eigenvectors are orthogonal, i.e.

Σ C\*jg(kj) Clg(kl) = δjl B3.2

g

For simplicity, we always choose the sum when j=l to equal one, what is called normalizing the eigenvectors. This relationship does not hold when an imaginary component is introduced into the crystal potential in which case the wavevectors of the Bloch waves will have imaginary components corresponding to damping by the absorption terms. All that we can use in these cases is the fact that different solutions of Schroedingers equation are orthogonal, which follows for the Bloch waves since the wavevectors are different, i.e.

∫b\*(r,kl) b(r,kj)dr =

Σ Σ ∫C\*lg(kl)Cjh(kj)exp(2πi[kj-kl+h-g).r)dr = δlj B3.3

g h

Note that we choose the integral to have the value of 1 conventionally in order to simplify the algebra.

**4. Periodicity of Cg and k values.**

A useful relationship is the periodicity of the solutions. We can write

ψ(r,kj) = exp(2πikj.r) Σ Cjg(kj)exp(2πig.r) B4.1

g

=exp(2πi[kj-h].r) Σ Cjg+h(kj)exp(2πi[g+h].r) B4.2

g

where h is any reciprocal lattice vector. In addition,

ψ(r,kj-h) = exp(2πi[kj-h].r) Σ Cjg(kj-h)exp(2πig.r) B4.3

g

One of the alternative ways of writing Bloch's theorem is to say that the solutions above for kj and kj-h are the same (except for a phase factor). Therefore

Cjg(kj) = Cjg+h(kj-h) B4.4

This is called the periodicity relationship since it represents the fact that the Bloch waves are periodic both in the image plane and in reciprocal space.

**5 Current Flow.**

An important property of a Bloch wave is the direction that the energy flow through the solid of the wave. As shown in many texts on Quantum mechanics, the energy flow of an electron wave can be represented by the current flow vector S which is defined by

S = ∫(-ih/4πm) { ψ\*(r) ∇ψ(r) - ψ(r) ∇ψ\*(r) }dr B5.1

Of interest is the result where we take ψ(r) to be a Bloch wave, in which case S gives us the path of a given Bloch wave through the crystal, i.e. allows us to draw a ray diagram for these waves. We will show here that the direction of current flow is perpendicular to the dispersion surface. The Gradient of a Bloch wave is

∇φ(r,kj)= Σ 2πi[(kj+g]Cjg(kj)exp(2πi[kj+g].r) B5.2

g

Multiplying by ψ\*(r) and remembering that when we integrate over r that any remaining wave terms will vanish, we have

∫ φ\*(r,kj) ∇φ(r,kj)dr = Σ 2πi[kj+g] |Cjg(kj)|2 B5.3

g

Therefore,

S = ∫(-ih/4πm) { φ\*(r)∇φ(r) - φ(r)∇φ\*(r)}dr B5.4

= Σ (h/m) [kj+g] |Cjg(kj)|2 B5.5

g

(strictly we only include the real part of kj, so any imaginary component introduced to simulate absorption does not appear in the current flow).

Let us next consider Schroedingers equation in the most general case where we allow the electron energy to be a function of the wavevector kj, i.e.

∇2φ(r,kj) + (8π2me/h2)[ E(kj) + V(r) ] φ(r,kj) = 0 B5.6

Substituting in the form for the Bloch wave, we have the standard secular equation for the coefficients Cjg (after collecting terms with common exponential terms and expanding the crystal potential as a Fourier series)

{-4π2(kj+g)2+(8π2me/h2)E(kj)}Cjg(kj) + Σ Vg-hCjh(kj) = 0 B5.7

h

The Gradient of E(kj) with respect to the wavevector kj is a vector normal to the dispersion surface. Therefore we take the derivative with respect to kj which gives us

{ -8π2(kj+g) + (8π2me/h2) ∇k E(kj)}Cjg(kj)

+ (8π2me/h2)E(kj)} ∇k Cjg(kj)+ Σ ∇k Vg-h Cjh(kj) = 0 B5.8

h

Multiplying by C\*jg and summing over different values of g, remembering that (in the high energy approximation with no imaginary component to the potential)

Σ C\*jg Cjg = 1 B5.9

g

so that

Σ C\*jg ∇k Cjg = 0 B5.10

g

we have (since the last two terms in B5.8 vanish)

Σ { -8π2(kj+g) + (8π2me/h2) ∇k E(kj)}|Cjg(kj)|2 = 0 B5.11

g

or

Σ (h/m) [kj+g] |Cjg(kj)|2 = (e/h) ∇k E(kj) = S B5.12

g

Thus the direction of current flow, i.e. how the energy associated with each particular Bloch wave moves through the lattice is determined by the vector normal to the dispersion surface. Note that it is quite wrong to think of the diffracted beams as propagating through the lattice in a direction specified by their wavevectors; in fact the diffracted beams travel as part of the Bloch waves so each beam travels in many different directions. An analogue of this behavior is the polyrefringent behavior of light in some mediums.

>> Figure of current flow

>> Talk about different orientation effects - weak beam, chanelling

**6. Dispersion Surface**

One of the more important ways of looking at dynamical diffraction is via the dispersion surface, i.e. the surface of allowed kj vectors for a given energy. The reason for the importance is the inherent complication of dynamical diffraction theory. Whilst the basic theory is not particularly difficult, so that it is possible to write down analytical equations for the diffracted amplitudes, hard numbers are very difficult to obtain short of resorting to a full numerical calculation on a digital computer. Therefore any method which can give us an indication of the general sense of the results is particularly useful; we can use it for a back of the envelope calculation, which we cannot do always do with a computer program. Two definitions of the dispersion surface can be found in the literature; here we will use the definition which is consistent in spirit with band structure theory and the Ewald sphere construction. Given an origin, we draw downwards kj. As axes we choose the vertical direction (in general) to lie along a major zone axis of the crystal.

One way to use the dispersion surface is to determine geometrically which Bloch waves are excited in the crystal for a given incident beam direction. Taking exp(2πi .r) as the incident wave, on the entrance surface of the crystal (assumed to be a plane normal to z) we must match the wave amplitude:

exp(2πiχ.[x + y ]) = T Σ Aj b(z=0,x,y,kj)

j

+ Σ Rgexp(2πi(χ'+g).[x + y ]) B6.1

g

where a fraction T is transmitted, and Rg reflected. Here χ is the wavevector of the incoming beam and χ' the wavevector of the specularly reflected beam where

χx = χ'x ; χy = χ'y ; χz = - χ'z B6.2

When we are dealing with diffraction we must include the possibility of waves which are not simply specularly reflected, but are reflected with an additional diffraction component, which is why we use a sum over reflected-diffracted waves with wavevectors χ' + g. Collecting terms with common components of the wavevector g,

(1-Ro) exp(2πiχ.[x + y ])

= T Σ Aj Cjo(kj)exp(2πikj.(x+y)) B6.3

j

Rgexp(2πiχ.[x + y ]) = T Σ Aj Cjg(kj)exp(2πikj.(x+y)) B6.4

j

As previously discussed for the High Energy case, the only way that this equation can hold for all different values of x and y is if

χx = kjx ; χy = kjy B6.5

We may represent this geometrically in the following fashion. On the dispersion surface construction we draw χ the incident wave, using the same origin as that of the dispersion surface. We now project along the z direction for the end point of χ (remember that z is not here along the beam direction) to the points where the normal direction cuts the branches of the dispersion surface. At all of these points the condition B6.5 above is satisfied so that we have a geometrical solution for the kj vectors. Exactly the same geometrical solution holds at the bottom surface of the crystal, the derivation of which is left to the reader. Here we match from each Bloch wave to any and all of the different possible plane waves, travelling in the transmitted and diffracted beam directions as shown in >>> Figure 2. We have therefore managed to obtain a geometrical understanding of what Bloch waves are excited in the crystal without truly solving the diffraction equations.

**Example: Wedge Shaped Crystal**

An informative example is a wedge shaped crystal for two branches of the dispersion surface since this is a problem which can be readily solved for Kinematical Theory and in the column approximation. The appropriate diagram is shown in >>> Figure 3. The net result after the crystal is four outgoing plane waves: a pair of satellites around both the transmitted and diffracted beam directions. This result differs from the Kinematical result which only shows satellites around the diffracted beam but is essentially the same as the column approximation result. Note that there is no need to solve the equations for the kj values or the Bloch wave coefficients in order to know the result, one of the advantages of using the dispersion surface.

>>> Figure showing spot splitting

**Evanescent waves**

It is possible to generate cases when the normal to the surface does not in fact cut the dispersion surface, particularly when the beam is nearly normal to the crystal surface as in RHEED and REM. In this case we still excite Bloch waves in the crystal, but rather than the waves extending infinitely as above, they are restricted to the surface. These waves are called evanescent waves. The idea is that instead of matching to real values kj, we consider using a complex wavevector. (There is no magic to real wavevectors, so why not so long as it works !) With the coordinate system that we used previously with z normal to the surface, we consider a complex wavevector h split into hrj and hij as real and imaginary components respectively, i.e. h = hrj + ihij.

Our first step is to utilize the matching condition for the wavevector in the plane of the surface (B6.5), from which we immediately see that

χx = hrjx ; χy = hrjy ; 0 = hijx ; 0 = hijy B6.6

All that remains is to work out the z component of h. For this we insist that our Bloch wave with a complex wavevector satisfies Schroedingers equation. The most obvious approach is to try a solution (Bloch wave) rather like those that we use inside the crystal with a form

φ(r,hj) = Σ Cjg(hj)exp(2πi[hrj+ihij+g].r) B6.7

g

With Schroedinger's equation

∇2φ(r,kj) + (8π2me/h2)[ E + V(r) ]φ(r,kj) = 0 B6.8

We obtain when we collect common exponential terms

{ -4π2(hj+g)2 + (8π2me/h2)E}Cjg(hj) + ΣVg-hCjh(hj) = 0 B6.9

h

To solve this directly, at first sight it appears that we have to solve numerically. Actually, we can solve using some of the known information about Bloch waves with a little care particularly if we assume that there is no imaginary coefficient to the potential (absorption) and assume that the potential is centered so that Vg = V-g. Writing B6.9 out with real and imaginary coefficients we have

{ -4π2[ (hrj+g)2 + 2i[hij.g +hijzhrjz] - hij2 ]

+ (8π2me/h2)E}Cjg(hj) + ΣVg-hCjh(hj) = 0 B6.10

h

The imaginary term 2i[hij.g +hijzhrjz] - hij2 ] will cause problems. We can avoid these problems is we choose a form where we cancel out this imaginary term, which we can do by choosing hrjz = 0 and both plus and minus terms in g, i.e. use instead a form

φ(r,hj) = ΣCjg(hj)exp(2πi[hrj+ihij].r)Cos(2πg.r) B6.11

g

which gives us in Schroedingers equation when we substitute and collect like terms

{ -4π2[hrj2+g2-hij2] + (8π2me/h2)E}Cjg(hj) + ΣVg-hCjh(hj) = 0 B6.12

h

The trick now is to compare equation B6.12 with the equation for a Bloch wave whose wavevector kj lies solely in the plane of the surface and perpendicular to the g vectors which is in the high energy approximation

{ -4π2(kj2+g2) + (8π2me/h2)E}Cjg(kj) + ΣVg-hCjh(kj) = 0 B6.13

h

By inspection the two are identical if we choose

kj2 = hrj2 - hij2 B6.14

Thus we have a solution with the same diffracted wave coefficients as our Bloch wave running along the surface with real and imaginary coefficients given in equation B6.14. It is should be noted that the current flow

S = Real Part of Σ (h/m) [hj+g] |Cjg(kj)|2 B6.15

g

lies in the plane of the surface. Therefore no current (energy) flows into the crystal; the wave amplitude decays into the crystal due to the imaginary component of the wavevector, but there is no actual movement of energy into the crystal. This corresponds to total reflection of the particular wavevector. If we employ a complex potential there is no so simple solution and as one might expect the current flow will now turn in towards the absorbing medium.

It is appropriate to mention here the question of what wavevectors to employ near to a reflection situation for the waves which do in fact get transmitted into the crystal. Considering the dispersion surface construction as shown in Figure 4, for any given branch of the surface there are two intersections with the surface normal. Considering the current flow vectors, of the two one is directed into the crystal, one away from it. Physically we can only employ waves where the energy flows into the crystal. Note that in this case we cannot use the directions of the wavevectors as a guide.

>> Example of REM calculation result

>> Figure illustrating evanescent waves

**7. Boundary Conditions**

The final step of interest is specifically to match the boundary conditions at the entrance surface. Using as the wave inside the crystal

ψ(r) = Σ Ajb(r,kj)exp(2πikj.r) B7.1

j

Taking into account the equality of the wavevectors in the plane of the surface, to match at the entrance surface we require for the amplitudes that

(1-Σ Rgexp(2πig.r)) = T Σ Aj b(r,kj) B7.2

g j

and for the derivative along the z direction (normal to the surface)

(χz - Σ Rgexp(2πig.r)(-χz + gz) ) =T Σ Aj kjz b(r,kj) B7.3

g j

remembering that the reflected beams have opposite signs of the wavevector along the z direction. Dividing equation B7.3 by z and subtracting B7.2 gives

Σ Rgexp(2πig.r)(-2+ gz/χz) =T Σ Aj (kjz/χz -1) b(r,kj) B7.4

g j

In the general transmission case, the difference between kjz and z is small and they are both far larger than gz, so that we can take the reflection coefficients Rg to be essentially zero. The boundary condition then simplifies to

1 = Σ Aj Σ Cg(kj) exp(2πig.r) B7.5

j g

In the most general case we define a matrix **C** which has the structure

**C =**  | Co(k1) Cg(k1) .... |

| Co(k2) Cg(k2) .... | B7.6

and writing the vector A as

A = ( A1 A2 . . An ) B7.7

and defining

U = ( 1 0 ... 0 )

we can write the boundary conditions as

U = **C** A B7.8

a matrix equation which can be solved formally as

A = **C**-1 U B7.9

If we use the high energy approximation and assume that the surface is normal to the incident beam direction, the orthonormality of the Bloch wave coefficients eases the solution which can be shown simply to be

A = ( Co(k1) Co(k2) ... ) B7.10

so that the wave inside is

ψ(r) = Σ Co(kj) b(r,kj)exp(2πikj.r) B7.11

j

Using the same approximation for the exit surface, we match each Bloch wave to a sum of plane waves. Below the crystal the wavefunction is

ψ(r) = Σ φgexp(2πi[ χ+g ] ) B7.12

g

= Σ Co(kj) b(r,kj)exp(2πikj.r) B7.13

g

where the two have to be equal when z=t, so that for a thickness of t

φg = Σ Co(kj)Cg(kj) exp(2πikjzt) B7.14

j

The oscillations in the intensities of the diffracted beams depend upon the phase terms exp(2πikjzt), the only factors which change with thickness. When we have two Bloch waves, we will obtain simple sin or cos fringes due to the interference the two exp(2πikjzt) terms. Similarly with more Bloch waves we obtain more complicated oscillations, as a rule superimposed.

>> Examples of solutions