I hate diffraction... always have, always will

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Abstract

A reading of Zachariesen's theory of dynamical diffraction applied to neutrons with a (hopefully) more transparent notation and treatment. I have tried to present a complete derivation showing all the steps and explaining as well as I can the physical reasoning behind the assumptions. The section 6. presents a summary description of the calculation and it might be a good idea to try reading it first.

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1 Basic Equations

Schroedinger Equation:

$$\nabla^2 \psi + \left(k_o^2 - \frac{2m}{\hbar^2} V(r) \right) \psi = 0$$

k_o is the incident k vector in vacuum

$$\begin{aligned} V(r) &= \frac{2\pi\hbar^2}{m} a \sum_j \delta(r - r_j) \\ \frac{2m}{\hbar^2} V(r) &= 4\pi \sum_L F_L e^{i \vec{G}_L \cdot \vec{r}} \end{aligned}$$

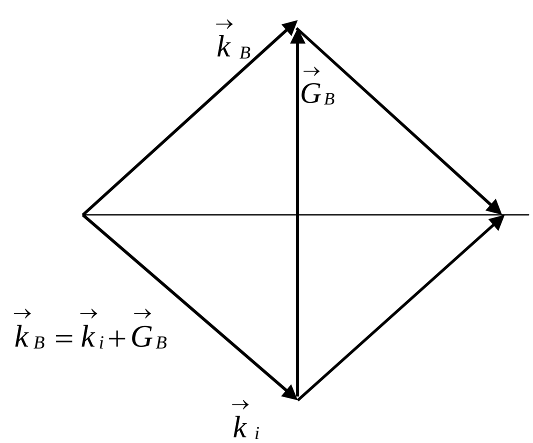
The last relation holds because V is a periodic function with periods given by the inverse lattice, $\vec{G}_L$. Similarly we can write

$$\psi = \sum_H D_H e^{i \vec{k}_H \cdot \vec{r}}$$

with

$$\vec{k}_H = \vec{k}_i + \vec{G}_H \qquad (1)$$

i.e. as a Bloch wave...a plane wave times a periodic function.



Note that $H = 0$ has a definite meaning, $\overrightarrow{G}_{H=0} = 0$, corresponding to $F_{H=0}$ being proportional to the average value of the potential

$$F_{L=0} \sim \int V(\overrightarrow{r})\, d^3r$$

and

$$\overrightarrow{k}_{H=0} = \overrightarrow{k}_i, \qquad \left(\overrightarrow{G}_{H=0} = 0\right)$$

Substituting in the S.E.

$$\begin{aligned}
\sum_H D_H\left(-k_H^2 + k_o^2\right) e^{i\overrightarrow{k}_H\cdot\overrightarrow{r}} - 4\pi \sum_{H',L} F_{H'} e^{i\overrightarrow{G}_{H'}\cdot\overrightarrow{r}} D_L e^{i\overrightarrow{k}_L\cdot\overrightarrow{r}} &= 0 \\
\sum_H D_H\left(-k_H^2 + k_o^2\right) e^{i\overrightarrow{k}_H\cdot\overrightarrow{r}} - 4\pi \sum_{H,L} F_{H-L} e^{i\overrightarrow{G}_{H-L}\cdot\overrightarrow{r}} D_L e^{i\overrightarrow{k}_L\cdot\overrightarrow{r}} &= 0 \\
\left(\sum_H D_H\left(-k_H^2 + k_o^2\right) - 4\pi \sum_{H,L} F_{H-L} D_L\right) e^{i\overrightarrow{k}_H\cdot\overrightarrow{r}} &= 0
\end{aligned}$$

since $\overrightarrow{k}_L + \overrightarrow{G}_{H-L} = \overrightarrow{k}_H$ by (1). Since this last equation must hold for every value of r, each term of the sum must be equal to zero:

$$D_H\left(-k_H^2 + k_o^2\right) - 4\pi \sum_L F_{H-L} D_L = 0$$

2 Case of one excited wave: refraction

Now let $H = i$ refer to the incident wave (inside the crystal) and consider as a first case $D_H \approx 0$ for $H \neq i$. This will be the case where the Ewald sphere corresponding to the incident $\overrightarrow{k}_i$ doesn't overlap any points of the reciprocal lattice. In this case

$$\begin{aligned}
D_i\left(-k_i^2 + k_o^2\right) - 4\pi \sum_L F_{H-L} D_L &= 0 \\
D_i\left(-k_i^2 + k_o^2 - 4\pi F_o\right) &= 0
\end{aligned}$$

or

$$k_o^2 = k_i^2 + 4\pi F_o \tag{2}$$

yielding the normal index of refraction for the case where the Bragg condition is not met for any reflection.

3 2 wave case..diffraction

We now consider the case where the Ewald sphere intersects one reciprocal lattice point so there will be 2 waves the incident $H = i$ and the diffracted wave, $H = B$.

Writing the equation for the two waves we find

$$\begin{array}{ll} H = i & D_i \left(k_o^2 - k_i^2 - 4\pi F_o\right) - 4\pi F_B D_B = 0 \\ H = B & -4\pi F_B D_i + \left(k_o^2 - k_B^2 - 4\pi F_o\right) D_B = 0 \end{array}$$

We have assumed for simplicity that $F_H = F_{-H}$. Writing $k'^2 = k_o^2 - 4\pi F_o$ we have

$$\begin{aligned} D_i \left(k'^2 - k_i^2\right) - 4\pi F_B D_B &= 0 \\ -4\pi F_B D_i + \left(k'^2 - k_B^2\right) D_B &= 0 \end{aligned}$$

These equations have a non-trivial solution only if the determinant of the coefficients is zero:

$$\left(k'^2 - k_i^2\right)\left(k'^2 - k_B^2\right) - (4\pi)^2 F_B^2 = 0$$

This is the dispersion relation for the waves in the crystal $\left(\vec{k}_i, \vec{k}_B\right)$.

The solution is then given by

$$x = \frac{D_B}{D_i} = \frac{\left(k'^2 - k_i^2\right)}{4\pi F_B}$$

3.1 Physical preliminaries

Before discussing the solution of this dispersion relation we need some physical preliminaries.

boundary condition: At the input boundary plane of the crystal defined by $\vec{n} \cdot \vec{r} = 0$, the incident wave in vacuum, $e^{i\vec{k}_o \cdot \vec{r}}$ must be equal to the incident wave in the crystal $e^{i\vec{k}_i \cdot \vec{r}}$ so that $\vec{k}_o$ and $\vec{k}_i$ can differ only by a term proportional to $\vec{n}$

$$\vec{k}_i = \vec{k}_o + \Delta\vec{n}$$

Defining

$$\begin{aligned} k_i^2 &= k_o^2 \left(1 + 2\delta_o\right) \\ k_B^2 &= k_o^2 \left(1 + 2\delta_B\right) \end{aligned} \tag{3a}$$

we find

$$\Delta = \frac{k_o \delta_o}{\gamma_o} \tag{4}$$

where $\gamma_o = \overrightarrow{n} \cdot \overrightarrow{k}_o / k_o$ is the direction cosine of the incident wave in vacuum with respect to the surface normal. The fact that the quantities (3a) differ from k_o^2 represents the key physical insight to the whole theory. The reasons for this seem to be that the waves are travelling at slightly different directions with respect to the crystal planes and that the effective indices of refraction are different for the different waves because they are determined by different Fourier components of the potential, F_B. Now since

$$\begin{aligned}
\overrightarrow{k}_B &= \overrightarrow{k}_i + \overrightarrow{G}_B = \overrightarrow{k}_i = \overrightarrow{k}_o + \Delta\overrightarrow{n} + \overrightarrow{G}_B \\
k_B^2 &= k_o^2 (1 + 2\delta_B) \\
&= k_o^2 + G_B^2 + \Delta^2 + 2\overrightarrow{k}_o \cdot \overrightarrow{G}_B + 2\Delta\overrightarrow{n} \cdot \overrightarrow{G}_B + 2\Delta\overrightarrow{k}_o \cdot \overrightarrow{n}
\end{aligned}$$

Thus

$$\begin{aligned}
\delta_B &= \alpha + \frac{\left(\overrightarrow{n} \cdot \overrightarrow{G}_B + \overrightarrow{k}_o \cdot \overrightarrow{n}\right)}{k_o^2} \frac{k_o \delta_o}{\gamma_o} = \\
&= \delta_o \left(1 + \frac{\overrightarrow{n} \cdot \overrightarrow{G}_B}{\gamma_o k_o}\right) + \alpha \equiv \frac{\delta_o}{b} + \alpha
\end{aligned} \tag{5}$$

This relation between δ_o and δ_B or k_B^2, k_o^2 and k_i^2 is crucial to what follows.

$$\alpha = \left(G_B^2 + 2\overrightarrow{k}_o \cdot \overrightarrow{G}_B\right) / 2k_o^2$$

is the variable which changes when the incident wavelength, direction or crystal orientation is varied. We can call it the scan variable.

3.2 Dispersion relation

We now rewrite the dispersion relation

$$\left(k'^2 - k_i^2\right)\left(k'^2 - k_B^2\right) - (4\pi)^2 F_B^2 = 0$$

in terms of these quantities.

$$\left[k'^2 - k_o^2 (1 + 2\delta_o)\right]\left[k'^2 - k_0^2 (1 + 2\delta_B)\right] - (4\pi)^2 F_B^2 = 0$$

$$\left(-4\pi F_o - 2k_o^2 \delta_o\right)\left[-4\pi F_o - 2k_o^2 \left(\frac{\delta_o}{b} + \alpha\right)\right] - (4\pi)^2 F_B^2 = 0$$

or introducing

$$\psi_{o,B} = \frac{4\pi F_{o,B}}{k_o^2} \tag{6}$$

$$(\psi_o + 2\delta_o)\left(\psi_o + 2\left(\frac{\delta_o}{b} + \alpha\right)\right) - \psi_B^2 = 0 \tag{7}$$

This equation is the dispersion relation for our problem.

In terms of these quantities the solution x becomes

$$\begin{aligned} x &= \frac{D_B}{D_i} = \frac{\left(k'^2 - k_i^2\right)}{4\pi F_B} = \frac{k_o^2 - 4\pi F_o - k_o^2 - k_o^2 2\delta_o}{4\pi F_B} \\ &= -\frac{\psi_o + 2\delta_o}{\psi_B} \end{aligned} \tag{8}$$

3.3 Two solutions for the incident and diffracted waves in the crystal

The dispersion relation, (7) is now an equation for δ_o with 2 solutions, $\delta_o^{(1,2)}$ the parameter α will be seen to be a function of the scan variables, *e.g.* wavelength λ or angle. Thus there will be two incident waves inside the crystal with two values of $\overrightarrow{k}_i$ or, alternately, two values of the parameter Δ, and two solutions for the diffracted wave characterized by the two values of x. Thus the complete solution for the incident wave inside the crystal will be

$$e^{i\overrightarrow{k}_o \cdot \overrightarrow{r}} \left[D_{i,1} e^{i\Delta_1 t} + D_{i,2} e^{i\Delta_2 t}\right] \tag{9}$$

where t is the normal distance from the crystal surface

$$t = \overrightarrow{n} \cdot \overrightarrow{r}$$

Similarly the solution for the diffracted wave is made up of the superposition of the two solutions

$$e^{i\left(\overrightarrow{k}_o + \overrightarrow{G}_B\right) \cdot \overrightarrow{r}} \left[x_1 D_{i,1} e^{i\Delta_1 t} + x_2 D_{i,2} e^{i\Delta_2 t}\right] \tag{10}$$

Before proceeding further we will look at the scan parameter α in some more detail

$$\alpha = \frac{\left(G_B^2 + 2\overrightarrow{k}_o \cdot \overrightarrow{G}_B\right)}{2k_o^2} \tag{11}$$

As a function of θ we write (note: $2k_B \sin\theta_B = G_B$)

$$\alpha_\theta = \frac{\left(G_B^2 - 2k_B G_B \sin\left(\theta_B + \varepsilon\right)\right)}{2k_B^2} \approx -\frac{1}{k_B} G_B \cos\theta_B \varepsilon \tag{12}$$

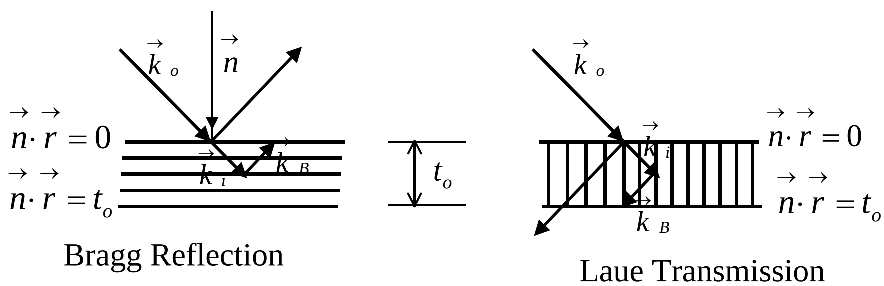
where $\theta = \theta_B + \varepsilon$. As a function of λ we have

$$\alpha_\lambda = 2\sin^2\theta_B \frac{(\lambda - \lambda_B)}{\lambda_B} \tag{13}$$

We now return to the general form of the solution, (9) and (10). To proceed further we need to impose an additional boundary condition which will be different for the Bragg and Laue cases.

Bragg: $D_B = 0$ at $\overrightarrow{r} \cdot \overrightarrow{n} = t_o$ where t_o is the thickness of the crystal

Laue: $D_B = 0$ at $\overrightarrow{r} \cdot \overrightarrow{n} = 0$



In the following we discuss the Bragg case. At $\overrightarrow{r} \cdot \overrightarrow{n} = t = 0$ we have to match the incident intensity coming from the vacuum. This is just a normalization constant for the problem which we set to unity.

$$D_{i,1} + D_{i,2} = 1 \tag{14}$$

while the Bragg boundary condition at $\overrightarrow{r} \cdot \overrightarrow{n} = t_o$ is

$$x_1 D_{i,1} e^{i\Delta_1 t_o} + x_2 D_{i,2} e^{i\Delta_2 t_o} = 0 \tag{15}$$

or

$$D_{i,1}\left[x_1 e^{i\Delta_1 t_o} - x_2 e^{i\Delta_2 t_o}\right] + x_2 e^{i\Delta_2 t_o} = 0$$

$$D_{i,1} = \frac{x_2 e^{i\Delta_2 t_o}}{x_2 e^{i\Delta_2 t_o} - x_1 e^{i\Delta_1 t_o}}$$

Similarly

$$D_{i,2} = 1 - D_{i,1} = \frac{-x_1 e^{i\Delta_1 t_o}}{x_2 e^{i\Delta_2 t_o} - x_1 e^{i\Delta_1 t_o}}$$

Now the reflected intensity at the entrance surface ($t = 0$) is given by

$$\begin{aligned} \frac{I_B}{I_o} &= \left|D_{B,1} + D_{B,2}\right|^2 = \left|x_1 D_{i,1} + x_2 D_{i,2}\right|^2 \qquad (16) \\ &= \left|(x_1 - x_2) D_{i,1} + x_2\right|^2 = \left|\frac{x_1 x_2 (c_1 - c_2)}{(x_2 c_2 - x_1 c_1)}\right|^2 \end{aligned}$$

where

$$c_{1,2} \equiv e^{i\Delta_{1,2}t_o}$$

so that

$$|c_2 - c_1|^2 = 2\left[1 - \cos\left(\Delta_1 - \Delta_2\right)t_o\right] = 4\sin^2\frac{\left(\Delta_1 - \Delta_2\right)t_o}{2}$$

By our previous definition

$$\left(\Delta_1 - \Delta_2\right) = \frac{k_o}{\gamma_o}\left(\delta_{0,2} - \delta_{0,1}\right)$$

In order to find δ_o we have to solve

$$\begin{aligned}
\left(\psi_o + 2\delta_o\right)\left(\psi_o + 2\frac{\delta_o}{b} + 2\alpha\right) - \psi_B^2 &= 0 \\
\psi_o^2 + \frac{2\delta_o\psi_o}{b} + 2\alpha\psi_o + 2\delta_o\psi_o + \frac{4\delta_o^2}{b} + 4\delta_o\alpha - \psi_B^2 &= 0 \\
\delta_o^2 + \frac{\delta_o}{2}\left[\psi_o + b\psi_o + 2b\alpha\right] + \frac{b}{4}\left(2\alpha\psi_o + \psi_o^2 - \psi_B^2\right) &= 0
\end{aligned}$$

We now define a parameter, z, so that the coefficient of δ_o in this equation will be

$$\begin{aligned}
z + \psi_o &= \frac{1}{2}\left(\psi_o + \psi_o b + 2\alpha b\right) \\
z &= \frac{1}{2}\left(-\psi_o + \psi_o b + 2\alpha b\right) \qquad (17)
\end{aligned}$$

We will see that this represents an extremely helpful trick. We now calculate "$b^2 - 4ac$" for the quadratic equation for δ_o.

$$\begin{aligned}
\text{"}b^2 - 4ac\text{"} &= \left(z + \psi_o\right)^2 - b\left(2\alpha\psi_o + \psi_o^2 - \psi_B^2\right) \\
&= z^2 + 2z\psi_o + \psi_o^2 - b\left(2\alpha\psi_o + \psi_o^2 - \psi_B^2\right) \\
&= z^2 - \psi_o^2 + b\psi_o^2 + 2\alpha b\psi_o + \psi_o^2 - b\left(2\alpha\psi_o + \psi_o^2 - \psi_B^2\right) \\
&= z^2 + b\psi_B^2 = z^2 + q
\end{aligned}$$

with $q = b\psi_B^2$. Therefore the solutions to the quadratic equation (dispersion relation) are

$$\delta_{o,(1,2)} = \frac{1}{2}\left[-\left(z + \psi_o\right) \pm \sqrt{z^2 + q}\right] \qquad (18)$$

and therefore

$$\begin{aligned}
|c_2 - c_1|^2 &= 4\sin^2\frac{\left(\Delta_1 - \Delta_2\right)t_o}{2} \\
&= 4\sin^2\frac{k_o\left(\sqrt{z^2 + q}\right)t_o}{2\gamma_o} \\
&= 4\sin^2\frac{k_o\sqrt{b}\psi_B\left(\sqrt{z^2/b\psi_B^2 + 1}\right)t_o}{2\gamma_o}
\end{aligned}$$

Now returning to

$$\frac{I_B}{I_o} = \left|\frac{x_1 x_2 (c_1 - c_2)}{(x_2 c_2 - x_1 c_1)}\right|^2 = \left|\frac{(c_1 - c_2)}{\left(\frac{c_2}{x_1} - \frac{c_1}{x_2}\right)}\right|^2$$

we calculate the denominator as:

$$\left(\frac{e^{i\Delta_2 t_o}}{x_1} - \frac{e^{i\Delta_1 t_o}}{x_2}\right)\left(\frac{e^{-i\Delta_2 t_o}}{x_1} - \frac{e-^{i\Delta_1 t_o}}{x_2}\right) =$$
$$\frac{1}{x_1^2} + \frac{1}{x_1^2} - \frac{2\cos(\Delta_2 - \Delta_1)\, t_o}{x_1 x_2} \tag{19}$$

and then using

$$x_{1,2} = -\frac{\psi_o + 2\delta_{o,(1,2)}}{\psi_B} = \frac{-1}{\psi_B}\left(-z \pm \sqrt{z^2 + q}\right)$$

again showing the usefulness of the trick. Then

$$\begin{aligned} x_{1,2}^2 &= \frac{1}{\psi_B^2}\left(2z^2 + q \mp 2z\sqrt{z^2 + q}\right) \\ &\equiv \eta \mp \omega \end{aligned}$$

and

$$x_1 x_2 = \frac{1}{\psi_B^2}\left(z^2 - \left[z^2 + q\right]\right) = \frac{-q}{\psi_B^2}$$

so that in order to evaluate expression (19), we calculate

$$\begin{aligned} \frac{1}{x_1^2} + \frac{1}{x_1^2} &= \frac{1}{\eta + \omega} + \frac{1}{\eta - \omega} = \frac{2\eta}{\eta^2 - \omega^2} \\ &= \frac{2\left(2z^2 + q\right)\psi_B^2}{\left((2z^2 + q)^2 - \left(2z\sqrt{z^2 + q}\right)^2\right)} \\ &= \frac{2\left(2z^2 + q\right)\psi_B^2}{(4z^4 + 4z^2 q + q^2 - 4z^4 - 4z^2 q)} \\ &= \frac{2\left(2z^2 + q\right)\psi_B^2}{q^2} \end{aligned}$$

and then (19) becomes

$$\begin{aligned} \frac{2\left(2z^2 + q\right)\psi_B^2}{q^2} - \frac{2\cos(\Delta_2 - \Delta_1)\, t_o}{x_1 x_2} &= \\ \frac{4z^2\psi_B^2}{q^2} + \frac{\psi_B^2 2}{q}\left(1 + \cos(\Delta_2 - \Delta_1)\, t_o\right) &= \\ \frac{4z^2\psi_B^2}{q^2} + \frac{\psi_B^2 4}{q}\cos^2\left[\frac{k_o\sqrt{b}\psi_B\left(\sqrt{z^2/b\psi_B^2 + 1}\right) t_o}{2\gamma_o}\right] &= \end{aligned}$$

$$\frac{4z^2\psi_B^2}{q^2} + \frac{\psi_B^2 4}{q}\cos^2 A\sqrt{1-y^2} \tag{20}$$

where we have introduced the famous (and, to newcomers to the subject, most obscure parameter) y, defined by $y^2 = z^2/\,|b|\,\psi_B^2$. Note that

$$b = \frac{\overrightarrow{n}\cdot\overrightarrow{k}_o}{\left(\overrightarrow{n}\cdot\overrightarrow{k}_o + \overrightarrow{n}\cdot\overrightarrow{G}_B\right)}$$

is negative for the Bragg case so that

$$y = \frac{z}{\psi_B\sqrt{|b|}} \tag{21}$$

and we have accordingly changed the sign of y^2 in (20). Further

$$A = \frac{k_o\sqrt{b}\psi_B t_o}{2\gamma_o}$$

is imaginary

$$A = i\frac{k_o\sqrt{|b|}\psi_B t_o}{2\gamma_o} = i\overline{A} \tag{22}$$

Thus finally we have

$$\begin{aligned}
\frac{I_B}{I_o} &= \left|\frac{(c_1-c_2)}{\left(\frac{c_2}{x_1}-\frac{c_1}{x_2}\right)}\right|^2 = \frac{4\sin^2 A\sqrt{1-y^2}}{\frac{4z^2\psi_B^2}{q^2}+\frac{\psi_B^2 4}{q}\cos^2 A\sqrt{1-y^2}} \\
&= \frac{b\sin^2 A\sqrt{1-y^2}}{\left(\frac{z^2}{b\psi_B^2}+\cos^2 A\sqrt{1-y^2}\right)} = \frac{b\sin^2 A\sqrt{1-y^2}}{\left(-y^2+\cos^2 A\sqrt{1-y^2}\right)} \\
&= \frac{b\sin^2 A\sqrt{1-y^2}}{\left(-y^2+1-\sin^2 A\sqrt{1-y^2}\right)}
\end{aligned} \tag{23}$$

using $q = b\psi_B^2$. Now equation (23) represents the *intensity* ratio of the reflected to the incident beam (Bragg case), while what we really want is the *power* ratio

$$\begin{aligned}
\frac{P_B}{P_o} &= \frac{A_B I_B}{A_o I_o} = -\frac{\left(\overrightarrow{n}\cdot\overrightarrow{k}_2\right)}{\left(\overrightarrow{n}\cdot\overrightarrow{k}_o\right)}\frac{I_B}{I_o} = -\frac{\left(\overrightarrow{n}\cdot\overrightarrow{k}_o+\overrightarrow{n}\cdot\overrightarrow{G}_B\right)}{\overrightarrow{n}\cdot\overrightarrow{k}_o}\frac{I_B}{I_o} \\
&= \frac{1}{-b}\frac{I_B}{I_o} = \frac{\sin^2 A\sqrt{1-y^2}}{\left(y^2-1+\sin^2 A\sqrt{1-y^2}\right)} \\
&= \frac{\sinh^2 \overline{A}\sqrt{1-y^2}}{\left(1-y^2+\sinh^2\overline{A}\sqrt{1-y^2}\right)} = \frac{1}{y^2+(1-y^2)\coth^2\overline{A}\sqrt{1-y^2}}
\end{aligned} \tag{24}$$

using $\sinh^2 x = -\sin^2 ix$.

4 Interpretation

The famous and obscure parameter, y is defined as

$$y = \frac{z}{\psi_B \sqrt{|b|}}$$

with

$$z = \frac{1}{2}\left(-\psi_o + \psi_o b + 2\alpha b\right)$$

so that

$$y = \frac{1}{2\psi_B \sqrt{|b|}}\left(-\psi_o + \psi_o b + 2\alpha b\right) \tag{25}$$

and α in turn is given by

$$\alpha = \frac{\left(G_B^2 + 2\overrightarrow{k}_o \cdot \overrightarrow{G}_B\right)}{2k_o^2}$$

and we had as a function of θ

$$\alpha_\theta \approx -\frac{1}{k_B} G_B \cos\theta_B \varepsilon \tag{26}$$

where $\theta = \theta_B + \varepsilon$ so that

$$\frac{dy}{d\theta} = \frac{\sqrt{b}}{\psi_B}\frac{1}{k_B} G_B \cos\theta_B = \frac{\sqrt{b}}{\psi_B}\sin 2\theta_B$$

Now equation (24) gives the reflected power as a function of incident angle θ when we use equations (25) and (26). The pattern is seen to be symmetrical with respect to $y = 0$ and we see from (25) that this does not correspond to $\alpha = 0$, *i.e.* the pattern is not centered on the Bragg angle but is slightly shifted from it by an amount that can be seen to be

$$\delta\theta = \varepsilon = \frac{\psi_o}{2\sin 2\theta_B}\left(\frac{1}{b} - 1\right)$$

The angular width of the diffraction pattern is given by $y = \pm 1$.

$$\begin{aligned} \alpha &= \frac{\psi_B \sqrt{|b|}}{b} = \sin 2\theta_B \varepsilon \\ \varepsilon &= \theta - \theta_B = \frac{\psi_B}{\sqrt{|b|}\sin 2\theta_B} \end{aligned}$$

5 Reflection from a single thin crystal - the mysterious Q

We had

$$\frac{P_B}{P_o} = \frac{1}{y^2 + (1-y^2)\coth^2 \overline{A\sqrt{1-y^2}}}$$

To find the total reflected power we need to calculate

$$R_B = \int \frac{P_B}{P_o} d\theta = \int \frac{P_B}{P_o} dy \frac{d\theta}{dy} = \frac{\psi_B}{\sqrt{b}\sin 2\theta_B} \int \frac{P_B}{P_o} dy$$

Now Zachariasen, quoting Darwin (Phil. Mag., **43,** 800, 1922) gives

$$\int_{-\infty}^{\infty} \frac{1}{y^2 + (1-y^2)\coth^2 \overline{A\sqrt{1-y^2}}} dy = \pi \tanh \overline{A} \sim \pi \overline{A}$$

for thin crystals ($A \ll 1$) so that

$$R_B = \pi \frac{\psi_B^2}{2\sin 2\theta_B} \frac{k_o t_o}{\gamma_o} = Q \frac{t_o}{\gamma_o}$$

since

$$\frac{t_o}{\gamma_o} = \frac{\delta V}{S_o}$$

with δV the exposed volume of the crystal and S_o the cross section area of the beam. Then since $\psi_B = \frac{4\pi F_B}{k_o^2}$

$$Q = \frac{\pi (4\pi)^2 F_B^2}{2k_o^3 \sin 2\theta_B} = \frac{\lambda^3 F_B^2}{\sin 2\theta_B}$$

The parameter Q is then the reflection of a single (weakly reflecting) mosaic block and serves as the input for calculations of the reflection and transmission of a mosaic crystal. The theory is relatively straightforward and is more or less understandable from the standard books.

6 Summary of the calculation: Main physical inputs

The calculation starts with the Schroedinger equation with a periodic potential. Since the potential is periodic it can be written as a Fourier series:

$$\frac{2m}{\hbar^2} V(r) = 4\pi \sum_L F_L e^{i\vec{G}_L \cdot \vec{r}}$$

and the wave function can be written as a Bloch wave: a plane wave times a periodic function

$$\psi = \sum_H D_H e^{i\vec{k}_H \cdot \vec{r}}$$

with

$$\overrightarrow{k}_H = \overrightarrow{k}_i + \overrightarrow{G}_H \tag{27}$$

Substituting this into the Schroedinger equation we obtain after redefining the indices in the sum

$$D_H \left(-k_H^2 + k_o^2\right) - 4\pi \sum_L F_{H-L} D_L = 0$$

This equation should hold for every H for which the associated D_H is significantly different from zero.

In the case where the input $\overrightarrow{k}$ vector is such that no diffracted wave is excited we get the standard result of a refracted wave with index of refraction related to $4\pi F_o$.

In the case where one diffracted wave is excited *i.e.* the Ewald sphere only intersects one point of the reciprocal lattice we get a pair of equations for $H = i$ and $H = B$, *i.e.* an incident and a Bragg wave.

$$\begin{aligned} D_i \left(k'^2 - k_i^2\right) - 4\pi F_B D_B &= 0 \\ -4\pi F_B D_i + \left(k'^2 - k_B^2\right) D_B &= 0 \end{aligned}$$

These equation only have a solution if the determinant is zero. This gives the dispersion relation

$$\left(k'^2 - k_i^2\right)\left(k'^2 - k_B^2\right) - (4\pi)^2 F_B^2 = 0$$

and the solution is then given by

$$x = \frac{D_B}{D_i} = \frac{\left(k'^2 - k_i^2\right)}{4\pi F_B}$$

A crucial point is that the waves $H = i$ and $H = B$ have different indices of refraction because they have different directions and interact with different Fourier components of the periodic potential. Thus defining

$$\begin{aligned} k_i^2 &= k_o^2 (1 + 2\delta_o) \\ k_B^2 &= k_o^2 (1 + 2\delta_B) \end{aligned} \tag{28a}$$

δ_o and δ_B are different and their relation follows from the boundary conditions and the Bragg condition

$$\overrightarrow{k}_B = \overrightarrow{k}_i + \overrightarrow{G}_B$$

so that we found

$$\delta_B = \frac{\delta_o}{b} + \alpha$$

where

$$\alpha = \left(G_B^2 + 2\overrightarrow{k}_o \cdot \overrightarrow{G}_B\right)/2k_o^2$$

is the 'scan parameter'. Substituting for δ_B in terms of δ_o in the dispersion relation yields two solution for δ_o so that there are two different incident and diffracted waves inside the crystal. Designating the two solutions by $1, 2$ we have for the incident wave

$$e^{i\overrightarrow{k}_o\cdot\overrightarrow{r}}\left[D_{i,1}e^{i\Delta_1 t}+D_{i,2}e^{i\Delta_2 t}\right] \tag{29}$$

and for the diffracted (B) wave

$$e^{i\left(\overrightarrow{k}_o+\overrightarrow{G}_B\right)\cdot\overrightarrow{r}}\left[x_1 D_{i,1}e^{i\Delta_1 t}+x_2 D_{i,2}e^{i\Delta_2 t}\right] \tag{30}$$

where

$$\Delta_{1,2}=\frac{k_o\delta_{o(1,2)}}{\gamma_o}$$

is determined by the solutions of the dispersion relation.

To complete the solution we need to impose boundary conditions which depend on the shape of the crystal and, for the rectangular slab considered here, are different for the Bragg (diffracted wave exits from the same surface as the incident wave enters) and Laue (diffracted wave exits from the side opposite to the entrance plane). At the entrance plane we have (normalizing the incident wave in the vacuum to unity)

$$D_{i,1}+D_{i,2}=1$$

while the Bragg boundary condition at $\overrightarrow{r}\cdot\overrightarrow{n}=t_o$ (the exit plane) is

$$x_1 D_{i,1}e^{i\Delta_1 t_o}+x_2 D_{i,2}e^{i\Delta_2 t_o}=0$$

We define

$$\psi_{o,B}=\frac{4\pi F_{o,B}}{k_o^2}$$

and solve the dispersion relation. After some algebra, putting it all together, we find for the power reflection coefficient

$$\frac{P_B}{P_o}=\frac{1}{y^2+(1-y^2)\coth^2\overline{A}\sqrt{1-y^2}}$$

where y is a basic parameter of the theory and is defined as

$$y=\frac{z}{\psi_B\sqrt{|b|}}$$

with

$$z=\frac{1}{2}\left(-\psi_o+\psi_o b+2\alpha b\right)$$

so that

$$y=\frac{1}{2\psi_B\sqrt{|b|}}\left(-\psi_o+\psi_o b+2\alpha b\right) \tag{31}$$

where α is the scan parameter. For $y^2>1$ the hyperbolic cotangent becomes a trigonometric cotangent and there are very rapid oscillations of the reflection. These are a result of beats between the two waves and are observable in special circumstances. They are often called *pendelösung* fringes. In other more usual cases one can average over these oscillations as Zachariasen does.

7 Larmor precession during penetration of Bragg reflected wave

We consider the case of Bragg reflection from a crystal located in a uniform magnetic field. In this case the Larmor precession will continue during the time the wave is inside the crystal. As the penetration depth is a function of the position in the Darwin peak (*i.e.* a function of the variable y) is is expected that the Larmor precession will vary in the same way and thus lead to a depolarization of the wave which may be measurable.

7.1 Larmor precession of a plane wave

If the plane wave has wave vector $\overrightarrow{k}_o$ in a vacuum it will have

$$\overrightarrow{k} = \overrightarrow{k}_o \pm \frac{\omega_L}{2v_o} \tag{32}$$

in a magnetic field B_o. $\omega_L = 2\mu B/\hbar$ is the Larmor frequency. The ($\pm$) signs apply to each of the 2 spin states. The reflected wave in the Bragg case is given by (10)

$$\psi_{ref} = e^{i\left(\overrightarrow{k}+\overrightarrow{G}_B\right)\cdot\overrightarrow{r}} \left|R(k)\right| e^{i\phi(k)}$$

Expanding $\phi(k)$ we find $\phi(k) = const + \phi'(k_o)(k - k_o)$ or

$$\psi_{ref} = e^{i\left(\overrightarrow{k}_o+\overrightarrow{G}_B\right)\cdot\overrightarrow{r}} \left|R(k_o)\right| e^{\pm i\phi'(k_o)\frac{\omega_L}{2v_o}}$$

using (32). The constant phase can be neglected as the Larmor precession angle is the difference in phase between the two spin states. Thus the phase difference between the two spin components of the wave function (Larmor precession angle) will be

$$\Delta\phi_L = \phi_+ - \phi_- = \phi'(k_o)\frac{\omega_L}{v_o} \tag{33}$$

so $\phi'(k_o)$ represents the "distance travelled" during the reflection.

7.2 Reformulation of Zachariesen variables in terms of experimental quantities

We consider the ideal case of Bragg reflection with the crystal boundary parallel to the Bragg Planes, $\left(\overrightarrow{G}_B \parallel \overrightarrow{n}\right)$ so that (5)

$$\frac{1}{b} = 1 + \frac{\overrightarrow{n}\cdot\overrightarrow{G}_B}{\gamma_o k_o}$$

with $\gamma_o = \overrightarrow{n}\cdot\overrightarrow{k_o}/k_o$. Thus $\gamma_o < 0$ and $\overrightarrow{n}\cdot\overrightarrow{G}_B = -2\gamma_o k_o$ so that

$$b = -1. \tag{34}$$

We consider a single-atom unit cell with no q dependence of the form factor. Neglecting Debye Waller factors we then have

$$\psi_o = \psi_B = \frac{4\pi F_{o,B}}{k_o^2},$$

(6) and

$$k_o^2 = k_i^2 + 4\pi F_o,$$

(2). Therefore we identify

$$F_{o,B} = Na,$$

N being the density of atoms/cc and a the coherent scattering length, (single atom/unit cell) and

$$\psi_o = \psi_B = \frac{4\pi Na}{k_o^2}. \tag{35}$$

Further

$$\begin{aligned} z &= \frac{1}{2}\left(-\psi_o + b\psi_o + 2\alpha b\right) \\ &= -\psi_o - \alpha \end{aligned}$$

(17) with

$$\alpha = \frac{\left(G_B^2 + 2\overrightarrow{k}_o \cdot \overrightarrow{G}_B\right)}{2k_o^2}$$

being the scan variable (11) given as a function of θ or λ by (12 or 13). Also (21)

$$\begin{aligned} y^2 &\equiv \frac{z^2}{|b|\,\psi_B^2} \\ y &= \frac{z}{\psi_B} = \frac{-\left(\psi_o + \alpha\right)}{\psi_B} \end{aligned} \tag{36}$$

7.3 Reflected wave function (Bragg case)

The wave function of the reflected wave is given by equation (10), ($t = 0$ at the entrance surface)

$$\begin{aligned} \psi_{ref}\left(t = 0\right) &= e^{i\left(\overrightarrow{k}_o + \overrightarrow{G}_B\right)\cdot\overrightarrow{r}}\left[x_1 D_{i,1} + x_2 D_{i,2}\right] \\ &\equiv \Gamma e^{i\left(\overrightarrow{k}_o + \overrightarrow{G}_B\right)\cdot\overrightarrow{r}} \end{aligned}$$

Then the boundary conditions give ($t_o \to \infty$ is crystal thickness), (equations 14, 15)

$$\begin{aligned} x_1 D_{i,1} e^{i\Delta_1 t_o} + x_2 D_{i,2} e^{i\Delta_2 t_o} &= 0 \\ D_{i,1} + D_{i,2} &= 1 \end{aligned}$$

Then

$$\Gamma = [(x_1 - x_2) D_{i,1} + x_2] \tag{37}$$

$$D_{i,1} = \frac{x_2 c_2}{x_2 c_2 - x_1 c_1}$$

$$\Gamma = \frac{(x_1 - x_2) x_2 c_2 + (x_2 c_2 - x_1 c_1) x_2}{x_2 c_2 - x_1 c_1}$$

$$= \frac{x_1 x_2 (c_2 - c_1)}{x_2 c_2 - x_1 c_1} \tag{38}$$

with $c_{1,2} = e^{i\Delta_{1,2} t_o}$. According to equation (4) $\Delta_{1,2} = \delta_{o(1,2)} k_o/\gamma_o$ and equation (18) gives the solutions to the dispersion relation as

$$\delta_{o(1,2)} = \frac{1}{2}\left[-(z + \psi_o) \pm \sqrt{z^2 + q}\right]$$

with $q = b\psi_B^2 = -\psi_B^2$. Thus $z^2 + q = \psi_B^2 \left(y^2 - 1\right)$ and $(\psi_B = \psi_o)$

$$\delta_{o(1,2)} = \frac{1}{2}\left[-(y+1)\psi_B \pm \psi_B \sqrt{y^2 - 1}\right]$$

$$\Delta_{1,2} = \frac{k_o}{2\gamma_o}\left[-(y+1)\psi_B \pm \psi_B \sqrt{y^2 - 1}\right]$$

According to equation (8)

$$x_{1,2} = \frac{-\left(\psi_o + 2\delta_{o(1,2)}\right)}{\psi_B} = \frac{-1}{\psi_B}\left[-y\psi_B \pm \psi_B \sqrt{y^2 - 1}\right]$$

$$= y \mp \sqrt{y^2 - 1} \tag{39}$$

$$c_{1,2} = e^{it_o \frac{k_o}{2\gamma_o}\left[-(y+1)\psi_B \pm \psi_B \sqrt{y^2 - 1}\right]}$$

We are interested in the case $y \le 1$ (inside the Darwin peak) so $\sqrt{y^2 - 1} \to i\sqrt{1 - y^2}$ and

$$c_{1,2} = e^{it_o \frac{k_o \psi_B}{2\gamma_o}\left[-(y+1) \pm i\sqrt{1 - y^2}\right]}$$

$$= e^{-i\overline{A}(y+1)} e^{\mp \overline{A}\sqrt{1 - y^2}}$$

with

$$\overline{A} = t_o \frac{k_o \psi_B}{2\gamma_o} \to \infty$$

(22). In this case (38, 39)

$$\Gamma = x_1 = y - \sqrt{y^2 - 1}$$

$$= y - i\sqrt{1 - y^2}$$

and the phase of the reflected wave (relative to the incident wave) is given by:

$$\tan\phi(k) = \sqrt{\frac{1}{y^2} - 1} \tag{40}$$

We need

$$\frac{d\phi}{dk} = \frac{d\phi}{dy}\frac{dy}{dk} = \frac{d\phi}{dy}\left(\frac{-1}{\psi_B}\right)\frac{d\alpha}{dk}$$

from equation (36). Equation (13) yields

$$\alpha_\lambda = 2\sin^2\theta_B \frac{\delta\lambda}{\lambda_B} = 2\sin^2\theta_B \frac{\delta k}{k_B}$$

so

$$\begin{aligned}
\frac{d\alpha}{dk} &= \frac{2\sin^2\theta_B}{k_B} \\
\frac{d\phi}{dk} &= \left(\frac{-1}{\psi_B}\right)\left(\frac{2\sin^2\theta_B}{k_B}\right)\frac{d\phi}{dy}
\end{aligned}$$

From (40)

$$\begin{aligned}
\phi(k) &= \tan^{-1}\sqrt{\frac{1}{y^2} - 1} = \tan^{-1}\eta(y) \\
\frac{d\phi}{dy} &= \frac{d\phi}{d\eta}\frac{d\eta}{dy}, \quad \frac{d\phi}{d\eta} = \frac{1}{1+\eta^2} \\
\frac{d\eta}{dy} &= -\frac{1}{y^2\sqrt{(1-y^2)}} \\
\frac{d\phi}{dy} &= -\frac{1}{\sqrt{(1-y^2)}} \\
\frac{d\phi}{dk} &= \left(\frac{1}{\psi_B}\right)\left(\frac{2\sin^2\theta_B}{k_B}\right)\frac{1}{\sqrt{(1-y^2)}} = \frac{\Delta_o}{\sqrt{(1-y^2)}}
\end{aligned}$$

with (remember $2k_B\sin\theta_B = 2\pi/d$, d= lattice spacing)

$$\Delta_o = \left(\frac{2k_B\sin^2\theta_B}{4\pi Na}\right) = \frac{\sin\theta_B}{2Nad}$$

Thus according to (33) the Larmor precession phase is given by:

$$\Delta\phi_L = \phi'(k_o)\frac{\omega_L}{v_o} = \frac{\Delta_o}{\sqrt{(1-y^2)}}\frac{\omega_L}{v_o}$$