A NEW DERIVATION OF THE DARWIN-PRINS FORMULA OF X-RAY REFLECTION

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1. *Introduction*

DARWIN (1914) and PRINS (1930) have investigated the intensity of reflection of X-rays by crystal planes, and have derived formulæ for the variation of intensity with the angle of incidence. The special case of a non-absorbing crystal was studied by Darwin, who found that the reflection is total over a small range of angles of incidence, outside which it falls away rapidly. Prins has extended Darwin's formulæ to an absorbing crystal. In essence the method is identical in both cases; it depends on the formation of a set of difference equations, and the solution of these for the case when the number of planes in the crystal is infinite. An excellent resume of the Darwin-Prins analysis is given in the book, " X-rays in Theory and Experiment," pp. 365 to 391, by Compton and Allison (hereafter referred to as C and A), whose notation we shall follow in this paper, on account of their compact nature.

An alternative method would be to solve the equations for the case when the number of planes is finite and then to extend the results for the infinite crystal. This procedure is more instructive in that it shows the manner in which the principal maximum is built up. In fact, it is found, as will be shown later, that, outside the region of perfect reflection indicated by Darwin's theory, the curve is not smooth as shown by his, but that there are an infinite number of maxima and minima, which, however, are so close together that it will not be possible to detect them. This important feature is altogether missed in Darwin's theory.

The author has actually considered such a finite stratification in an earlier paper (1942). There, a most general case of a periodic stratification was considered, in which the variation in the properties of the medium was supposed to occur periodically, the nature of this variation being unspecified. The theory can easily be applied to crystal planes, and then it can be extended to the case of a crystal of infinite depth. This is done in this paper, and the results obtained are substantially the same as those of Darwin and Prins.

It is however found that Darwin's theory leads to a different formula from that in the present paper for the region outside that of perfect reflection, which obviously arises from the failure of his theory to take cognisance of the fluctuations of intensity which occur in this region. Incidentally, it is found that a certain assumption made by Darwin and Prins is not altogether justified. The present method is free from any such, and is consequently more rigorous.

2. *General Formulæ*

Suppose that we have a medium consisting of n stratifications, and that a beam of electromagnetic radiation is incident on it. Let T_1 be the amplitude of the incident beam at the beginning of the first stratification. It is evident that there will be a large number of multiple reflections, and consequently there will be two streams of energy in the medium, one building up the transmitted beam, and the other the reflected beam. Let T_1, R_1; T_2, R_2; ....; T_{n+1} represent the electric vectors in the two streams at the beginning of the first, second, etc., stratification. If now we denote by r and t the complex reflection and transmission coefficients of a single stratification, on which the radiation is incident at the angle concerned, then the following difference equations are obtained:

$$R_s = rT_s + tR_{s+1}; \quad T_s = tT_{s-1} + rR_s \tag{1}$$

Eliminating the R's and the T's successively from these,

$$T_{s-1} = yT_s - T_{s+1}; \quad R_{s-1} = yR_s - R_{s+1} \tag{2}$$

where $y = (1 + t^2 - r^2)/t$. These can be solved by making use of the condition that $R_{n+1} = 0$. One obtains in this way

$$R_1/T_{n+1} = (r/t)\cdot f_n(y); \quad T_1/T_{n+1} = f_n(y)/t - f_{n-1}(y) \tag{3}$$

where
$$f_n(y) = y^{n-1} - \frac{(n-2)}{(1)!} y^{n-3} + \frac{(n-4)(n-3)}{(2)!} y^{n-5} - \ldots\ldots \tag{4}$$

The series appearing as $f_n(y)$ can be summed up by putting $y = 2\cosh\beta$, and comes out as $= \sinh n\beta/\sinh\beta$ (Jolley, 1925). Also, making the substitution

$$\frac{r}{\sinh\beta} = \frac{t}{\sinh\alpha} = \frac{1}{K}$$

where K can be evaluated to be equal to $\sinh(\alpha + \beta)$ from the relation $\cosh\beta = (1 + t^2 - r^2)/2t$, one finally obtains,

$$\frac{R_1}{\sinh n\beta} = \frac{T_{n+1}}{\sinh\alpha} = \frac{T_1}{\sinh(\alpha + n\beta)}, \tag{5}$$

where $\cosh\beta = (1 + t^2 - r^2)/2t$ and $\cosh\alpha = (1 - t^2 + r^2)/2r$ (6)

From (5), the reflecting power, R, of the stratified medium is given by

$$\frac{1}{R} = \left|\frac{T_1}{R_1}\right|^2 = |\cosh \alpha + \sinh \alpha \coth n\beta|^2 \qquad (7)$$

This is our fundamental equation, from which the intensity of the reflected beam can be calculated by substituting the values of α and β. It may be noted that, in general, both α and β are complex.

3. *Derivation of the Formulæ for an Absorbing Crystal*

In order to determine the values of α and β, we must know those of r and t. For these, we make use of the relations, first derived by Darwin and reproduced in C and A, pp. 365 to 376. If we take the boundary between adjacent stratifications as the planes midway between the atomic planes, it is easy to show from the above formulæ in C and A that

$$r = -ise^{-ik\,\delta/2} \text{ and } t = (1 - i\sigma)e^{-ik\,\delta/2} \qquad (8)$$

where s and σ are two constants depending on the nature of the planes, $k = 2\pi/\lambda$, and $\delta = 2d \sin \psi$, d being the distance between the planes, ψ the angle of incidence, and λ the wavelength of the X-rays. The quantities σ and s are given by

$$(\lambda \sin \psi/2\pi d)\,\sigma = \mu_c - 1 = -(\delta + i\beta) \qquad (9)$$

where μ_c is the complex refractive index of the crystal, whose real and imaginary parts are, respectively, $(1 - \delta)$ and β, and

$$s = f(2\psi, k)\,\sigma/f(0, k),$$

so that $(\lambda \sin \psi/2\pi d)\, s = -(\delta + i\beta).\ f(2\psi, k)/f(0, k) = -(\Delta + ib)$ (10)

where $f(2\psi, k)$ and $f(0, k)$ are quantities proportional to the atomic structure factors for angles 2ψ and zero. (C and A, p. 376). In the above expressions, σ and s are to be taken as small.

Now, we are interested in the phenomena that occur near about the Bragg angle, for which $k\,\delta/2 \simeq m\pi$. Putting $k\,\delta/2 = m\pi + \xi$, where ξ is small, $\exp(-ik\,\delta/2) = (-1)^m \exp(-i\xi)$, and

$$r = (-1)^{m+1}\, is\, e^{-i\xi}, \text{ and } t = (-1)^m (1 - i\sigma)e^{-i\xi} \qquad (11)$$

From these, treating σ, s and ξ as small quantities, one obtains

$$\cosh \alpha = (-1)^{m+1} (\sigma + \xi)/s;\ \cosh \beta = (-1)^m \{2 + s^2 - (\sigma + \xi)^2\}/2 \qquad (12)$$

In the general case of an absorbing crystal, both σ and s are complex quantities, so that both $\cosh \alpha$ and $\cosh \beta$, and hence α and β, are complex. Now, the reflecting power of a finite stratification is given by (7). In order to determine the same for an infinite one, we must put $n \to \infty$ in the right hand side of (7). If β is complex, it is easily shown that $\underset{n\to\infty}{\mathrm{L}t} \coth n\beta = 1$, so that for an infinite stratification,

$$\underset{n\to\infty}{\mathrm{L}t} \frac{\mathrm{T}_1}{\mathrm{R}_1} = \cosh\alpha + \sinh\alpha \tag{13}$$

Now, from (12), $\cosh\alpha = (-1)^{m+1} (\sigma + \xi)/s$, so that $\sinh\alpha = \pm\sqrt{(\sigma+\xi)^2 - s^2}/s$, and

$$\underset{n\to\infty}{\mathrm{L}t} \frac{\mathrm{R}_1}{\mathrm{T}_1} = -s/\{(-1)^m (\sigma + \xi) \pm \sqrt{(\sigma+\xi)^2 - s^2}\} \tag{14}$$

This is identical with the Eq. (6·42) in C and A, p. 380, except for the factor $(-1)^m$ before $(\sigma + \xi)$ in our above expression. The difference is easily explained, for the amplitudes are measured at points on the planes in the derivation in C and A, while in ours, the reference point is at a distance $d/2$ above the first plane. It is obvious that this would produce a phase change of $\exp(-ik\delta/2)$, which, for small values of ξ, reduces to $(-1)^m$ exactly the factor by which the two expressions differ. It is thus seen that Prins's formula can be derived easily from our general formulæ by substituting the appropriate values of r and t.

4. *The Case of a Non-Absorbing Crystal*

We now consider the case of a non-absorbing crystal. Here, β and b in Eqns. (13) and (14) are equal to zero, so that both σ and s are real. Hence, both $\cosh\alpha$ and $\cosh\beta$ are real, and two separate cases have to be considered, *viz.*,

case (i) $|\cosh\alpha| < 1$, for which $|\cosh\beta| > 1$, and

case (ii) $|\cosh\alpha| > 1$, and $|\cosh\beta| < 1$.

From (12), $|\cosh\alpha| = 1$, if $(\sigma + \xi) = \pm s$, or $\xi = -\sigma \pm s$.

Hence, if $-\sigma - s < \xi < -\sigma + s$, case (i) is operative, so that $\sinh^2\alpha$ is $-ve$ and $\sinh^2\beta$ is $+ve$. If one writes $\alpha = \alpha_1 + i\alpha_2$, and $\beta = \beta_1 + i\beta_2$, then

$$\frac{1}{\mathrm{R}} = |\cos\alpha_2 + i\sin\alpha_2 \coth n\beta_1|^2 = 1 + \frac{\sin^2\alpha_2}{\sinh^2 n\beta_1} \tag{15}$$

Therefore, as n is increased, the intensity corresponding to any value of ξ within this range steadily increases. When $n\to\infty$, $\sinh^2 n\beta_1 \to \infty$, and $\mathrm{R}\to 1$, *i.e., the reflection is total throughout the whole range of values of* ξ *from* $-\sigma - s$ *to* $-\sigma + s$ *for an infinite stratification.* This region may be called the principal maximum, and its width is given by $\Delta\xi = 2s$.

Outside this range, case (ii) is operative, so that $\sinh^2\alpha$ is $+ve$ and $\sinh^2\beta$ $-ve$. Hence, in this case,

$$1/R = 1 + \sinh^2\alpha_1/\sin^2 n\beta_2 \tag{16}$$

In this region, a number of maxima and minima occur corresponding to $n\beta_2 = s\pi + 1/2$ and $s\pi$ respectively. These may be referred to as subsidiary maxima, and they all lie evidently on the curve $R = \operatorname{sech}^2 \alpha_1$. When the number n increases, the intensity corresponding to any value of ξ in this region does not tend to a limit, but only undergoes fluctuations. Also, the subsidiary maxima approach closer and closer together. When n actually tends to infinity, the subsidiary maxima would be so close together, and the fluctuations in the intensity would be so rapid, that they would not be detected. The observed intensity would only be an average over a cycle of changes, and the observed value of R would be given by

$$R_{obs} = \frac{1}{\pi}\int_{\theta}^{\theta+\pi} R\, d(n\beta_2) = \frac{1}{\pi}\int_{\theta}^{\theta+\pi} \frac{\sin^2 n\beta_2}{\sin^2 n\beta_2 + \sinh^2 \alpha_1}\, d(n\beta_2) \tag{17}$$

The integration can be carried out, and leads to the remarkably simple result that

$$R_{obs} = 1 - |\tanh \alpha_1| \tag{18}$$

Now, the boundary between the two regions is obviously given by $\sinh \alpha_1 = 0$, for which $\tanh \alpha_1 = 0$ and $R_{obs} = 1$. Thus, in the second region, R_{obs} starts from unity and quickly drops down as the value of ξ is taken farther and farther away from a principal maximum. These features are all very similar to those predicted by Darwin's theory. In fact, the reflection curve is not symmetric about $\xi = 0$, but $\xi = -\sigma$, or, $k\,\delta/2 = m\pi - \sigma$. The value of this shift caused by refraction, as also of the width of the region of perfect reflection can be obtained in angular dimensions. If θ_0 be the Bragg angle defined by $k\,\delta/2 = kd \sin\theta_0 = m\pi$, and if θ be the angle of incidence corresponding to the centre of the diffraction pattern, then

$$\theta - \theta_0 = -\sigma/kd \cos\theta_0 = \delta \sec\theta_0 \operatorname{cosec}\theta_0 \tag{19}$$

Also, if the angular width of perfect reflection be $\Delta\psi$, then

$$\Delta\psi = s/kd \cos\theta_0 = \Delta \sec\theta_0 \operatorname{cosec}\theta_0 \tag{20}$$

Both these are identical with the relations derived in C and A, pp. 388 and 389.

5. *A Note on Darwin's Method of Solving the Difference Equations*

In the preceding section, it was shown that the reflection coefficient in the region outside the principal maximum is $R = 1 - |\tanh \alpha_1|$. Darwin's formula, put in terms of our symbols, gives $R = e^{-|2\alpha_1|}$ in the same region. The disagreement needs an explanation. It arises from a certain assumption made by Darwin in solving the difference equations. In terms of our symbols Darwin puts $T_2/T_1 = x$, and obtains R_1/T_1 in the form

$$R_1/T_1 = (r^2 - t^2 + tx)/r \tag{21}$$

The problem thus reduces to the evaluation of x, and for this, *Darwin makes the assumption that* $T_{s+1} = T_1 x^s$. By substituting this in (2), he gets

$$x = e^{\pm\beta} = \cosh\beta \pm \sinh\beta \text{ and } R_1/T_1 = \cosh\alpha \pm \sinh\alpha. \tag{22}$$

For the absorbing crystal, the same procedure is followed (C and A, p. 378).

It is to be noted that such an *a priori* assumption cannot be justified, since we are actually trying to find the values of the quantities T_s and R_s. A more serious objection to this assumption lies in the fact that it leads to an internal contradiction in the theory. Taking the case where it disagrees with the present theory, the value of x here is $\exp(\pm i\beta_2)$, so that $|x| = 1$. Hence, $|T_{s+1}| = |T_1|$, so that $\underset{s\to\infty}{Lt} |T_s| = |T_1|$. Thus the transmitted intensity is equal to the incident intensity and *the reflection must be zero*. However, by substituting the value of x in (21), Darwin actually gets $R = |\cosh\alpha \pm \sinh\alpha|^2 = \exp(\pm 2\alpha_1)$. Thus proceeding in two different ways from the same assumption, one gets different results, so that the assumption itself cannot be correct.

In fact, by making use of the method of the present paper, it can be proved that the assumption holds asymptotically for the first few planes (*i.e.*, for small values of s) in Prins's case, and in the region of perfect reflection in Darwin's case. In other words, the value of x as defined by the equation $x = T_2/T_1$ is actually equal to $\exp(\pm\beta)$ in these cases. However, the assumption is not generally true, and is invalid in the region outside the principal maximum for a non-absorbing crystal. Details are not given for want of space.

In conclusion, I wish to express my thanks to Prof. Sir C. V. Raman for the many helpful discussions I had with him during the investigation.

Summary

In this paper is described a new derivation of the formulæ for the reflection of X-rays by perfect crystals, which forms an alternative approach to the problem to that adopted by Darwin and Prins. It consists in obtaining a solution of the difference equations which occur in the problem for a crystal containing a finite number (n) of laminations. The case of a crystal

of infinite depth is obtained from this by proceeding to the limit when $n \to \infty$. The formulæ thus obtained are identical with those of Prins for an absorbing crystal, while for a non-absorbing crystal, the width of the region of perfect reflection is in agreement with Darwin's value. However, there is a difference in the formulæ for the variation of intensity with angle outside this region. This has been shown to be due to an assumption made by Darwin which is not altogether justifiable.

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