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An Exact Analytic Solution of Darwin's Difference Equations

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Abstract

Darwin derived a system of difference equations which relate the incident and reflected amplitudes of X-rays in a crystal in Bragg diffraction. These equations are recast in matrix form. Using a well known result from optical multilayer theory, an exact analytic solution of these difference equations is obtained for a crystal of an arbitrary number of atomic planes. In the limiting case of a semi-infinite crystal, and with the appropriate approximations, Darwin's expression for the reflectivity of a crystal in Bragg diffraction is obtained.

Introduction

To treat the diffraction of X-rays from perfect crystals, there have historically been two dynamical diffraction theories. One is the elegant mathematical theory introduced by Ewald (1916, 1917) and extended by von Laue (1931, 1940). The other is the simpler phenomenological theory introduced by Darwin (1914) and modified by Prins (1930). Because it is simpler, the Darwin–Prins formulation was at first favored over the Ewald–von Laue theory. However, with further work, the Ewald–von Laue theory was able to account for many phenomena, such as the Borrmann effect (Borrmann, 1950; von Laue, 1949), which could not be explained with the Darwin–Prins theory. Part of this had to do with the methods used to solve Darwin's difference equations. We present here an exact solution to Darwin's difference equations for a perfect crystal of any thickness using a matrix formulation. Berreman (1976) introduced the concept of solving the diffraction problem for the symmetric Bragg case using a matrix formulation. However, he did not apply his formulation to Darwin's difference equations. The method may also be extended to Borie's difference equations for Laue diffraction (Borie, 1966), and to the difference equations for asymmetrical reflection (Warren, 1969).

A future paper will use the results of this paper to show the theoretical relationship between the Darwin–Prins and Ewald–von Laue formulations of dynamical diffraction.

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Darwin's difference equations

We consider here the formulation of Darwin's difference equations as given by James (1965). Fig. 1 is similar to Fig. 24 of James. The horizontal lines represent planes of atoms parallel to the surface of a perfect crystal. The planes of atoms are numbered starting with the surface plane. The atomic planes are separated by a thickness of vacuum a. It is assumed that there are N atomic planes in total. T_0 represents the amplitude of a beam of plane X-rays incident on the crystal. S_0 represents the amplitude of the reflected X-rays. T_r and S_r represent the amplitudes of the forward and reverse propagating waves, respectively, of a beam of X-rays at the rth plane. The glancing angle of incidence is θ. The atoms in each plane are assumed to scatter in phase so that S_r is a plane wave whose angle is also θ relative to the atomic planes.

We define the reflection coefficient for a single plane of atoms to be $-iq$, the transmission coefficient through the plane of atoms to be $1-iq_0$, and the phase change for a wave travelling the distance a between two successive planes to be $\varphi=(2\pi a/\lambda)\sin\theta$. The wavelength of the X-rays is λ. The reflection coefficient for a plane of atoms is given by

$$q=-\frac{n\lambda}{\sin\theta}\frac{e^2}{mc^2}f(2\theta)P(2\theta), \qquad (1)$$

where n is the number of atoms per unit area in the plane, $f(2\theta)$ is the atomic scattering factor, and $P(2\theta)$ is a polarization factor. $P(2\theta)$ is equal to unity for σ-polarized waves and $\cos(2\theta)$ is equal to unity for

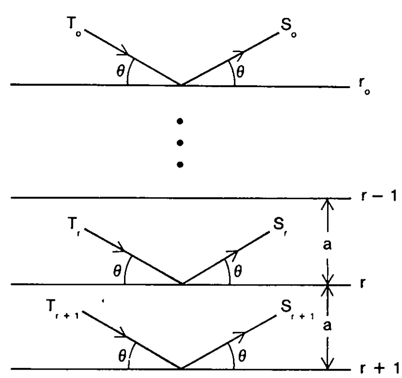

Fig. 1. Diffraction from a set of atomic planes.

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π-polarized waves. The other symbols have their usual significance. Although a lattice of one kind of atom is considered here, the results are easily generalized in the usual manner of replacing the atomic scattering factor by an appropriate structure factor.

The waves at the rth plane are related by

$$S_r = -iqT_r + (1 - iq_0)\, e^{-i\varphi} S_{r+1}, \qquad (2)$$

$$T_{r+1} = (1 - iq_0)\, e^{-i\varphi} T_r - i\bar{q}\, e^{-2i\varphi} S_{r+1}, \qquad (3)$$

where $\bar{q}$ is the reflection coefficient from the lower side of an atomic plane and may be, but is not necessarily, equal to q. Equation (2) says that S_r is composed of two parts: the part of T_r that is reflected from the rth layer and the part of S_{r+1} that is transmitted through the rth layer. Equation (3) is interpreted similarly. Equations (2) and (3) are known as Darwin's difference equations.

Solution

The primary quantity of interest is the reflectivity of X-rays from a crystal,

$$R = \frac{I}{I_0} = \left(\frac{S_0}{T_0}\right)^2, \qquad (4)$$

where R is the reflectivity and I_0 and I are the incident and reflected intensity, respectively. Darwin (1914) found an approximate expression for the ratio (S_0/T_0) by assuming a form of the solution for an infinite crystal. Later he (Darwin, 1922) found an approximate solution for a finite number of layers. Henke (1981) has also found a solution for a finite number of layers. There may be others reported in the literature.

We present here an exact solution to (2) and (3). The method of solution used the fact that (2) and (3) may be rewritten in matrix form as

$$\begin{bmatrix} T_r \\ S_r \end{bmatrix} = \frac{1}{(1 - iq_0)} \times \begin{bmatrix} e^{i\varphi} & i\bar{q}\, e^{-i\varphi} \\ -q\, e^{i\varphi} & [(1 - iq_0)^2 + \bar{q}q]\, e^{-i\varphi} \end{bmatrix} \begin{bmatrix} T_{r+1} \\ S_{r+1} \end{bmatrix}, \qquad (5)$$

which, for convenience, we write as

$$\begin{bmatrix} T_r \\ S_r \end{bmatrix} = \mathbf{A} \begin{bmatrix} T_{r+1} \\ S_{r+1} \end{bmatrix}. \qquad (6)$$

It is apparent from this that the amplitudes of the waves at any two lattice planes are related by a power of the 2×2 matrix in (5) and (6). If there are N total planes of atoms in the crystal then the relation between the incident, reflected and transmitted wave is

$$\begin{bmatrix} T_0 \\ S_0 \end{bmatrix} = \mathbf{A}^N \begin{bmatrix} T \\ 0 \end{bmatrix}. \qquad (7)$$

Here T denotes the amplitude of the transmitted wave. The 2×2 matrix in question, $\mathbf{A}$, is unimodular (*i.e.* its determinant equals unity).

A unimodular matrix raised to a power N is related to Chebyschev polynomials of the second kind (Abelés, 1950; Born & Wolf, 1959; Knittl, 1976). If we define a matrix $\mathbf{B}$,

$$\mathbf{B} = \begin{bmatrix} a & b \\ c & d \end{bmatrix}, \qquad (8)$$

such that

$$\det \mathbf{B} = 1, \qquad (9)$$

then $\mathbf{B}^N$ may be written as follows;

$$\mathbf{B}^N = \begin{bmatrix} aU_{N-1}(x) - U_{N-2}(x) & bU_{N-1}(x) \\ cU_{N-1}(x) & dU_{N-1}(x) - U_{N-2}(x) \end{bmatrix}. \qquad (10)$$

Here $U_N(x)$ are the Chebyschev polynomials of the second kind,

$$U_N(x) = \frac{\sin[(N-1)\cos^{-1} x]}{(1 - x^2)^{1/2}}, \qquad (11)$$

$$x = (a + d)/2.$$

If $x > 1$, then hyperbolic functions must be used (Knittl, 1976). This result is well known in optical multilayer work (Abelés, 1950; Born & Wolf, 1959; Knittl, 1976).

In our case, we combine (7), (10) and (11) and obtain the coefficient of reflection,

$$\frac{S_0}{T_0} = -iq/\{1 - (1 - iq_0)\, e^{-i\varphi} \times \sin[(N-1)\cos^{-1} x]/\sin[N \cos^{-1} x]\} \qquad (12)$$

and the coefficient transmission,

$$\frac{T}{T_0} = (1 - x^2)^{1/2}(1 - iq_0)/\{e^{i\varphi} \sin(N \cos^{-1} x) - (1 - iq_0)\sin[(N-1)\cos^{-1} x]\}, \qquad (13)$$

where now

$$x \equiv \frac{e^{i\varphi} + [(1 - iq_0)^2 + \bar{q}q]\, e^{-i\varphi}}{2(1 - iq_0)}. \qquad (14)$$

Equations (12) and (13) are exact solutions of Darwin's difference equations [(2) and (3)] for any number of atomic layers.

Derived next is the expression for the coefficient of reflection for a thick crystal. It will be shown to reduce to Darwin's result for an infinite crystal using the appropriate approximations (James, 1965). To do this one needs to consider the following term from (12) in the limit as the number of layers in the crystal goes to infinity:

$$\lim_{N\to\infty} \frac{\sin[(N-1)\cos^{-1}x]}{\sin[N\cos^{-1}x]}. \quad (15)$$

This may be rewritten as

$$\lim_{N\to\infty} \{x-(1-x^2)^{1/2}\cot[N\cos^{-1}x]\}. \quad (16)$$

It is convenient for the analysis to write $\cos^{-1}x$ with real and imaginary parts

$$\cos^{-1}x = \alpha + i\beta. \quad (17)$$

Then we need to examine

$$\begin{aligned}\lim_{N\to\infty} &\cot[N\cos^{-1}x] \\ &= \lim_{N\to\infty}\left[i\frac{e^{iN(\alpha+i\beta)}+e^{-iN(\alpha+i\beta)}}{e^{iN(\alpha+i\beta)}-e^{-iN(\alpha+i\beta)}}\right], \\ &= \begin{cases}+i, & \beta<0\\ -i, & \beta>0\end{cases}. \end{aligned} \quad (18)$$

Therefore, for an infinite crystal,

$$R = I/I_0 = |-iq/\{1-(1-iq_0)[x \pm i(1-x^2)^{1/2}]\,e^{i\varphi}\}|^2. \quad (19)$$

This is an exact solution of Darwin's difference equations for an infinite crystal. To investigate the form of this about a Bragg peak, write

$$\varphi = (2\pi a/\lambda)\sin\theta = m\pi + v, \quad (20)$$

where v is small. It may then be shown, for small v, that (19) reduces to

$$R = I/I_0 \simeq |-q/[q_0 + v \pm (q_0 + v^2)^{1/2} - \bar{q}q]|^2. \quad (21)$$

This is identical to equation (2.75) of James (1965) and is Darwin's result for the reflectivity of an infinite crystal near a Bragg peak.

Discussion

We have presented here an exact solution to Darwin's difference equations for any number of atomic planes in a crystal. An exact expression for the reflectivity of X-rays from an infinite crystal has been derived. This solution has been shown to simplify to Darwin's result. The key for doing this is to rewrite Darwin's difference equations in matrix form and then apply results from optical multilayer theory.

The method of solution presented here is extendable to other difference equations. We have used it to obtain exact solutions to Borie's difference equations for Laue diffraction and the Borrmann effect (Borie, 1966). We have used it, as well, to obtain exact solutions to the difference equations for asymmetrical Bragg reflection (Warren, 1969).

The matrix form [(5)] of Darwin's difference equations has been used to demonstrate the relationship between the Ewald–von Laue dynamical diffraction theory and the Darwin–Prins theory. This will be the subject of a future paper.

Many people are familiar with the treatment of the Ewald–von Laue dynamical diffraction theory given by Zachariasen (1945). Upon the recommendation of one of the referees, the work presented here will also be related to the results of the Ewald–von Laue theory as given by Zachariasen.

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