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A numerical study of the dynamical theory of scattering from a distorted crystal. By W. J. FITZGERALD, *Institut Laue–Langevin, B.P. n° 156, 38042-Grenoble Cedex, France* and C. N. W. DARLINGTON, *Department of Physics, The University of Birmingham, P.O. Box 363, Birmingham B15 2TT, England*

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The Darwin difference equations are solved numerically for the case of a crystal having a depth-dependent *d* spacing.

Introduction

The application of dynamical theory, based on Ewald's ideas, to distorted crystals (Kato, 1963) is generally complicated and leads to equations that cannot be easily applied in solving a particular problem. The earlier treatment due to Darwin (see Warren, 1969) is readily extended to include certain forms of distortion, and meaningful results can be obtained with the aid of a computer.

An example is given for a crystal with a depth-dependent *d* spacing. Such a situation can be realized by applying an electric field to $BaTiO_3$ for example. These crystals are *n*-type semiconductors, so that close to the electrodes Schottky barrier layers are formed. On the application of a d.c. bias a large field gradient is set up across the surface region near the negative electrode, and a depth-dependent *d* spacing results from electrostrictive coupling between field and strain.

In the Darwin (1914) approach to the dynamical theory of the diffraction of X-rays from crystals, two difference equations, the so-called Darwin difference equations, are formulated which relate the amplitude and phase of the total transmitted wave T_r just above the $(r+1)$th plane to the total reflected wave S_r in the same position. Hence T_0 and S_0 are the incident and reflected waves respectively. The two difference equations are

$$S_r = iqT_r + (1-h+iq)S_{r+1} \exp(-i\varphi) \quad (1)$$

$$T_{r+1} = (1-h+iq_0)T_r \exp(-i\varphi) + iqS_{r+1} \exp(-2i\varphi) \quad (2)$$

where

$$q = \left(\frac{e^2}{mc^2}\right)\frac{M\lambda f(2\theta)}{\sin\theta}, \quad q_0 = \left(\frac{e^2}{mc^2}\right)\frac{M\lambda f(0)}{\sin\theta}$$

and M is the number of atoms per unit area. h is a small number which partially allows for absorption of the beam in passing through a layer (Prins, 1930).

The phase factor φ is given by

$$\varphi = \frac{2\pi}{\lambda} d \sin\theta$$

where d is the interplanar spacing.

The solution of the Darwin difference equations for a crystal containing p layers may be found by using a trial solution of the form (Warren, 1969)

$$S_r = S_0\left(\frac{x^{r-p} - x^{p-r}}{x^{-p} - x^p}\right).$$

By making suitable approximations and by substituting this trial solution for S_r into the difference equation found from (1) and (2) by eliminating the T's, one finds that

$$\frac{S_0}{T_0} = \frac{iq}{(h+iv) + \eta\coth(p\eta)}$$

where

$$v = \frac{2\pi}{\lambda} d(\theta - \theta_B)\cos\theta,$$

θ_B = Bragg angle corrected for refraction, and

$$\eta = \pm[q^2 + (h+iv)^2]^{1/2}.$$

As p tends to infinity, coth $(p\eta)$ tends to unity and for an infinitely thick crystal

$$\frac{S_0}{T_0} = \frac{iq}{(h+iv) \pm [q^2 + (h+iv)^2]^{1/2}}.$$

The absolute square of this quantity is related to the intensity of the scattered radiation.

Consider a crystal which has a depth-dependent d spacing such that

$$d_r = d_0 + r\Delta$$

where Δ is a small constant. The two Darwin difference equations may then be written as

$$S_r = iqT_r + aS_{r+1} \exp(-irA) \quad (3)$$

$$T_{r+1} = aT_r \exp(-irA) + bS_{r+1} \exp(-2iAr) \quad (4)$$

where

$$a = (1-h+iq_0)\exp(-i\varphi)$$

$$b = iq \exp(-2i\varphi)$$

and

$$A = \frac{2\pi}{\lambda}\Delta \sin\theta.$$

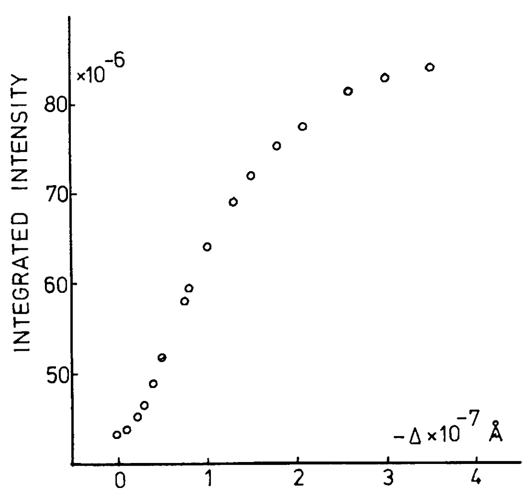

Fig. 1. Integrated intensity for the 002 reflexion from $BaTiO_3$ as a function of Δ derived from the computer calculation.

Table 1. *Parameters used for the* 002 *reflexion from* $BaTiO_3$

θ (Bragg) corrected for refraction	d spacing	p	q	M	$h=\mu d/2 \sin\theta$
12·4643°	1·996 Å	10^4	$2·54\times10^{-4}$	$3·125\times10^{14}$	$1·411\times10^{-5}$

Eliminating T_r from equations (3) and (4) one obtains

$$cS_{r-1}+aS_{r+1}-m(r)S_r=0 \quad (5)$$

where $c=a \exp iA$ and

$$m(r)=\exp(irA)+(a^2-ibq)\exp[-i(r-2)A].$$

A numerical method was used to solve this difference equation for S_r. Defining an 'operator' G_r such that

$$G_rS_r=S_{r+1}$$

then from equation (5)

$$[aG_rG_{r-1}-m(r)G_{r-1}+c]S_{r-1}=0.$$

Therefore

$$G_{r-1}=\frac{c}{m(r)-aG_r}. \quad (6)$$

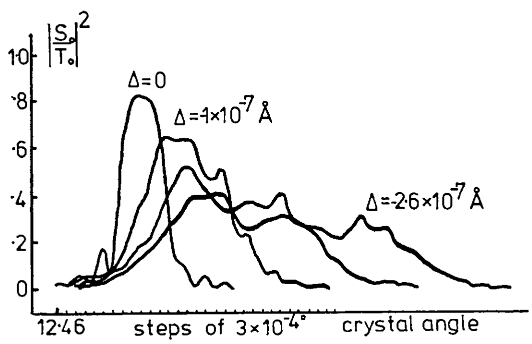

Fig. 2. Intensity of the scattered radiation from the 002 reflexion from $BaTiO_3$ as a function of crystal angle derived from the computer calculations.

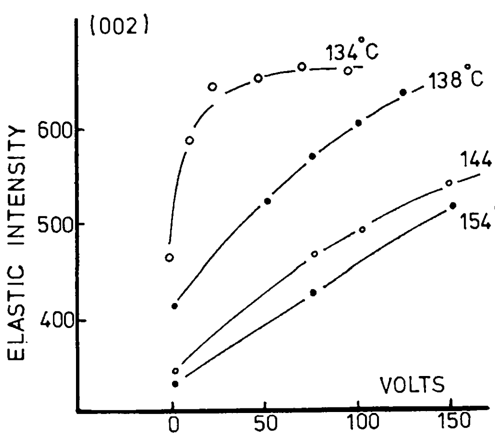

Fig. 3. Measured integrated intensity of the 002 reflexion at the negative electrode as a function of applied voltage. The variation with temperature is caused by the temperature dependence of the permittivity which results in an increase in field-induced strain as the temperature decreases for the same value of the applied d.c. bias.

If a thin crystal of p layers is considered, where r runs from 0 to $(p-1)$, then $S_p=0$ and

$$cS_{p-2}-m(p-1)S_{p-1}=0$$

which is obtained from equation (5). Therefore,

$$G_{p-2}=\frac{S_{p-1}}{S_{p-2}}=\frac{c}{m(p-1)}. \quad (7)$$

With equations (6) and (7) it is now possible to generate all the values of G_r $[r=(p-2)\rightarrow 0]$. Now, from equation (3)

$$iqT_r=S_r-a\exp(-irA)G_rS_r.$$

Therefore,

$$\frac{S_0}{T_0}=\frac{iq}{1-aG_0}$$

where G_0 may be found from the continued fraction

$$G_0=\cfrac{c}{m(1)-\cfrac{ac}{m(2)-\cfrac{ac}{\ddots\; G_{p-2}}}}.$$

A computer was used to calculate the real and imaginary parts of G_0 and hence the scattered intensity.

Figs. 1 and 2 show the computer results obtained for the 002 reflexion from $BaTiO_3$ as a function of Δ and crystal angle. The parameters used in the calculation are given in Table 1. The number of layers chosen was limited to 10^4 since the iterative calculation required large amounts of computer time. The range of Δ was chosen such that the integrated intensity did not vary greatly with any further increase in Δ. These results compare with experimentally measured changes in the integrated 'elastic' intensity of 002 shown in Fig. 3. The experiments were performed with highly monochromatic radiation obtained from a Mössbauer γ-ray source ($\lambda\sim0·86$ Å). The results were found to be reproducible, and the temperature stability of the crystal was $\pm0·5$ K. The technique (O'Connor, 1972) has the further benefit of distinguishing between elastically and inelastically scattered γ-rays and so phonon-assisted scattering can be subtracted experimentally from the measured integrated intensity of a Bragg reflexion (Fitzgerald, Darlington & O'Connor, to be published). Although there is no direct experimental evidence for a model having $d_r=d_0+r\Delta$, it has been shown by other computations that any depth-dependent distortion produces much the same result for the integrated intensity as a function of distortion.

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