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References

Booth, A. D. (1946). *Proc. Roy. Soc.* A, **188**, 86.
Booth, A. D. (1947*a*). *J. Appl. Phys.* **18**, 664.
Booth, A. D. (1947*b*). *Nature, Lond.*, **160**, 196.
Cox, E. G. & Shaw, W. F. B. (1930). *Proc. Roy. Soc.* A, **127**, 71.
Frevel, L. K. (1937). *Rev. Sci. Instrum.* 8, 475.
Ingold, C. K. (1921). *J. Chem. Soc.* p. 305.
de Lange, J. J., Robertson, J. M. & Woodward, I. (1939). *Proc. Roy. Soc.* A, **171**, 398.
Piper, S. H. (1929). *J. Chem. Soc.* p. 234.
Riley, D. P. (1944). *Nature, Lond.*, **153**, 587.
Robertson, J. M. (1934). *Phil. Mag.* **18**, 729.
Vand, V. (1947). *J. Sci. Instrum.* **24**, 326.
Vand, V., Lomer, T. R. & Lang, A. (1947). *Nature, Lond.*, **159**, 507.
Vand, V. (1948*a*). *Nature, Lond.*, **161**, 600.
Vand, V. (1948*b*). *J. Appl. Phys.* **19**, 852.
Vand, V. (1948*c*). *J. Sci. Instrum.* **25**, 352.

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The Diffraction of X-rays by Distorted-Crystal Aggregates. II. Diffraction by Bent Lamellae

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The apparent particle size of distorted crystallites varies from independence of θ and λ (particle-size broadening) to proportionality with λ cosec θ (distortion broadening) as the parameter $E\eta \sin\theta/\lambda$ increases, where E is a particle size and η is an average strain. For a bent 'orthorhombic' lamella of thickness T with axes chosen so that z is parallel to the axis of bending, x perpendicular to the lamella, and y perpendicular to x and z, the apparent particle size ϵ is given by

$$\epsilon = \frac{2T}{p}\int_0^1 \frac{\sin\left[2\pi A\xi(1-\xi)\right]}{2\pi A\xi}\,d\xi,$$

where $A = \frac{T}{p}\frac{|q^2 - c_{21}p^2/c_{11}|}{R}\frac{T\sin\theta}{\lambda}$, R is the radius of curvature, p, q, r are the direction cosines of the reflecting planes, and c_{21} and c_{11} are elastic constants.

Introduction

Under the main title above, Dr A. R. Stokes and the writer published a crude but general treatment of the Debye-Scherrer pattern given by an aggregate of distorted crystals (Stokes & Wilson, 1944*a*). The treatment was general, in that no particular type of strain was postulated, but crude in that strain gradients were neglected. This note contains some more refined results, without neglect of strain gradients, for the particular case of diffraction by bent lamellae, of thickness small in comparison with their other dimensions. Such bent lamellae would be produced in the 'kinking' described by Orowan (1942). Publication of this incomplete investigation at the present time is prompted by a paper given by Dr H. Ekstein at the Harvard meeting of the International Union of Crystallography, in which he concluded that moderate bending would lead to no line broadening. When certain effects neglected by him are taken into account an expression is obtained which changes continuously from 'small-particle' to 'distortion' broadening as the ratio $T^2/\lambda R$ increases, where T is the thickness of the lamellae and R is their radius of curvature. This behaviour is not, however, confined to the particular model of bent lamellae, and may be of interest in connexion with some recent experiments (Wood & Rachinger, 1948) (see, however, Wood (1943) and Lipson & Stokes (1943)) in which the apparent strain in cold-worked metals was found to vary with the wave-length.

Calculation

In the simplest case, when the glide lamellae are assumed to possess orthorhombic symmetry and to be bent into portions of circular cylinders, the displacement u, v, w of the point x, y, z from its position in the undistorted state is given by

$$\left.\begin{aligned} u &= -\alpha x^2/2R - y^2/2R, \\ v &= xy/R, \\ w &= 0, \end{aligned}\right\} \qquad (1)$$

where x is taken perpendicular to the glide plane, y in the glide direction, z parallel to the axis of the cylinder, and $\alpha = c_{21}/c_{11}$ is a sort of Poisson's ratio for these axes.

Cube terms are omitted, and the origin is taken at the centre of the lamella. For the intensity distribution in reciprocal space these displacements lead to complicated functions involving Fresnel integrals, but a fairly simple expression for the apparent particle size from Debye-Scherrer lines is obtainable. If the direction cosines of the reflecting plane are p, q, r relative to the above axes, the displacement normal to the plane is $pu+qv$, so that the effective structure amplitude of cells in the neighbourhood of x, y, z is changed from F to

$$F' = F\exp\{-2\pi i(2\sin\theta/\lambda)(pu+qv)\}. \quad (2)$$

The product of this by the complex conjugate of F' for a cell distant t in the direction p, q, r is

$$\begin{aligned} F'F'^* &= FF^*\exp\{2\pi i(\sin\theta/\lambda R)\\ &\quad\times[p\{\alpha x^2+y^2-\alpha(x+pt)^2-(y+qt)^2\}\\ &\quad+2q\{(x+pt)(y+qt)-xy\}]\}\\ &= FF^*\exp\{2\pi i(\sin\theta/\lambda R)(q^2-\alpha p^2)(pt^2+2xt)\}. \end{aligned} \quad (3)$$

The mean value of this with respect to x is J_t, the function required for calculating integral breadths (Wilson, 1943; Stokes & Wilson, 1944*b*). The limits for x are $\pm\frac{1}{2}T$, but for x positive F'^* is zero for $x>\frac{1}{2}T-pt$, and for t negative F' is zero for $x<-\frac{1}{2}T-pt$. For t positive therefore

$$\begin{aligned} J_t &= \frac{FF^*}{T-pt}\int_{-\frac{1}{2}T}^{\frac{1}{2}T-pt}\exp\{2\pi i(\sin\theta/\lambda R)(q^2-\alpha p^2)\\ &\qquad\times(pt^2+2xt)\}\,dx\\ &= \frac{FF^*}{T-pt}\frac{\lambda R}{2\pi(q^2-\alpha p^2)t\sin\theta}\\ &\quad\times\sin\{2\pi(q^2-\alpha p^2)(\sin\theta/\lambda R)(tT-pt^2)\}. \end{aligned} \quad (4)$$

For t negative the same expression is found, except that t is replaced by $|t|$. Then the apparent particle size is

$$\epsilon = (V_0J_0)^{-1}\int V_tJ_t\,dt,$$

where V_t is the volume common to the crystal and its 'ghost' shifted a distance t in the pqr direction, and V_0, J_0 are the values of V_t, J_t for $t=0$. Since

$$V_t = (\text{area of lamella})(T-p|t|),$$

$$\begin{aligned} \epsilon &= \frac{1}{T}\int_{-T/p}^{T/p}\frac{\lambda R}{2\pi(q^2-\alpha p^2)|t|\sin\theta}\\ &\quad\times\sin\{2\pi(q^2-\alpha p^2)(\sin\theta/\lambda R)(|t|T-pt^2)\}\,dt\\ &= \frac{2T}{p}\int_0^1\frac{\sin\{2\pi A\xi(1-\xi)\}}{2\pi A\xi}\,d\xi, \end{aligned} \quad (5)$$

where

$$\xi = p|t|/T, \quad (6)$$

$$A = \frac{T}{p}\frac{|q^2-\alpha p^2|}{R}\frac{T\sin\theta}{\lambda}. \quad (7)$$

The integral in (5) is a function of the single parameter A. For A small it reduces to $\frac{1}{2}$, and for A large to $1/(4A)$.

The extreme values of ϵ are therefore

$$\epsilon = T/p \quad (A\ll 1), \quad (8)$$

$$\epsilon = \lambda\,\mathrm{cosec}\,\theta(R/2T|q^2-\alpha p^2|) \quad (A\gg 1). \quad (9)$$

The first of these is small-particle broadening, the second distortion broadening. It may be expressed in the form of 'apparent strain' (Stokes & Wilson, 1944*a*)

$$\eta \equiv \beta\cot\theta = \lambda/\epsilon\sin\theta = 2T|q^2-\alpha p^2|/R. \quad (10)$$

For small A (5) can be expanded in a power series in A, giving

$$\begin{aligned} \frac{p\epsilon}{2T} &= \sum_n\frac{(-)^n(2\pi A)^{2n}}{(2n+1)!}\int_0^1\xi^{2n}(1-\xi)^{2n+1}\,d\xi\\ &= \sum_n\frac{(-)^n(2n)!(2\pi A)^{2n}}{(4n+2)!}. \end{aligned} \quad (11)$$

This converges rapidly for $A<2$, but becomes inconvenient for larger values.

In deriving these expressions no account has been taken of possible coherence between the lamellae for reflexions with $p=1$. For other reflexions coherent scattering from different lamellae is very unlikely, but for these, in the region approximating to 'small-particle' broadening, the apparent particle size may be anomalously large.

Equation (5) gives a continuous variation from small-particle broadening to distortion broadening. It is, of course, unnecessary to postulate the particular model of bent lamellae in order to obtain a continuous transition with variation of a parameter involving the wavelength. To a first approximation (Stokes & Wilson, 1944*a*)

$$J_t = FF^*\int_{-\infty}^{\infty}\phi(e)\exp\{2\pi i(2\sin\theta/\lambda)te\}\,de, \quad (12)$$

where $\phi(e)\,de$ is the fraction of the crystal for which the strain perpendicular to the reflecting planes lies between e and $e+de$. The apparent particle size is then

$$\epsilon = V_0^{-1}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}V_t\phi(e)\exp\{2\pi i(2\sin\theta/\lambda)te\}\,de\,dt. \quad (13)$$

In terms of the variable s, equal to $2(\sin\theta-\sin\theta_0)/\lambda$, where θ_0 is the Bragg angle for a large crystal, the line profile $I(s)$ for an undistorted crystal is

$$I(s) = U^{-1}\int_{-\infty}^{\infty}V_t\exp\{2\pi ist\}\,dt, \quad (14)$$

where U is the volume of a unit cell, so that

$$\epsilon = (U/V_0)\int_{-\infty}^{\infty}\phi(e)\,I(2e\sin\theta/\lambda)\,de, \quad (15)$$

or

$$\epsilon = (U/V_0)(\lambda/2\sin\theta)\int_{-\infty}^{\infty}\phi(\lambda s/2\sin\theta)\,I(s)\,ds. \quad (16)$$

(These relations, at first sight surprisingly simple, are no more than the Parseval theorem for Fourier transforms in a particular case. Analogous relations would apply in the simultaneous consideration of particle size and 'mistakes'.) If $\phi(e)$ is a wide curve in

comparison with $I(s)$ it may be set equal to $\phi(0)$ in the integration, so that, since $\int I(s)\,ds = V_0/U \equiv I$,

$$\epsilon = \lambda\phi(0)/2\sin\theta. \tag{17}$$

On the other hand, if $I(s)$ is the wide curve it may be set equal to $I(0)$ for the integration over $\phi(e)$, so that

$$\epsilon = (U/V_0)\,I(0) = I(0)/I. \tag{18}$$

Equations (17) and (18) express distortion and small-particle broadening respectively. The functional form of the transitions from one to the other depends on the exact shape of $\phi(e)$ and $I(s)$. With the common, but unreliable, Gaussian approximations

$$\phi(e) \doteqdot (2\pi\eta^2)^{-\frac{1}{2}} \exp\{-e^2/2\eta^2\}, \tag{19}$$

and

$$I(s) \doteqdot IE \exp\{-\pi E^2 s^2\}, \tag{20}$$

where η is the root-mean-square stress and E is an order-of-magnitude particle size, equation (15) gives

$$\epsilon \doteqdot E(2\pi\eta^2)^{-\frac{1}{2}} \int_{-\infty}^{\infty} \exp\left\{-e^2\left[\frac{1}{2\eta^2} + \frac{4\pi E^2 \sin^2\theta}{\lambda^2}\right]\right\} de$$
$$= 2E\{4 + 16A_1^2\}^{-\frac{1}{2}}, \tag{21}$$

where

$$A_1 = \sqrt{(2\pi)}\,E\eta\sin\theta/\lambda. \tag{22}$$

Equation (22) is quite analogous to (5), in which $T/p \sim E$, $A \sim A_1$, and the integral has the same limiting values as $\{4 + 16A_1^2\}^{-\frac{1}{2}}$. The decrease of ϵ with increase in A is, however, much more rapid for (21). Other approximations to $\phi(e)$ and $I(s)$ lead to the same general behaviour, but different functional dependence on A. Thus, for sufficiently small values of $\sin\theta/\lambda$, particle-size broadening will always mask distortion broadening; and for sufficiently large values distortion broadening will be predominant. The manner in which the transition takes place will depend on the nature of the distortion and the shape of the crystallites.

References

Lipson, H. & Stokes, A. R. (1943). *Nature, Lond.*, **152**, 20.

Orowan, E. (1942). *Nature, Lond.*, **149**, 643.

Stokes, A. R. & Wilson, A. J. C. (1944*a*). *Proc. Phys. Soc. Lond.* **56**, 174.

Stokes, A. R. & Wilson, A. J. C. (1944*b*). *Proc. Camb. Phil. Soc.* **40**, 197.

Wilson, A. J. C. (1943). *Proc. Roy. Soc.* A, **181**, 360.

Wood, W. A. (1943). *Nature, Lond.*, **151**, 585.

Wood, W. A. & Rachinger, W. A. (1948). *Nature, Lond.*, **162**, 891.

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Diffuse Scattering of X-rays by Ice

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The diffuse scattering of X-rays by single crystals of ice has been found to be of thermal origin. The existing theory is inadequate to explain the observations, and needs extending to include the effects of vibrations of shorter wave-lengths. The main features of the representation in reciprocal space of the diffuse scattering are as follows:

(1) There is a strong concentration of scattering power immediately round the reciprocal-lattice points, corresponding to rounded isodiffusion contours.

(2) Beyond these regions are weaker bridges, which connect reciprocal-lattice points; they are narrow, quite clearly defined, and their strength varies little along their length; they join together to form plane sheets of scattering power, which are parallel to the c^* and to one of the a^* axes, and intersect along lines joining rows of reciprocal-lattice points parallel to the c^* axis.

(3) There is no sheet of scattering power lying along any a^* axis, or passing through the origin of the reciprocal lattice, though there are a few bridges, which make only a small angle with the a^* axes, which do not fall into this general scheme.

(4) The sheets pass through many 'forbidden' points, but at some other 'real' reciprocal-lattice points they show sharp discontinuities; they are not always centrosymmetrical about the reciprocal-lattice points.

(5) The diffuse pattern does not become broader or less clearly defined with increasing angle of reflexion.

The amplitudes of vibration of the molecules are of the order of 0·4 A. at $-5°$ C. The strong local disturbances which cause the diffuse streaks could include movements of the hydrogen atoms between neighbouring oxygen atoms.

Introduction

The diffuse scattering of X-rays by any crystal depends on the directions and amplitudes of the vibrations executed by the atoms, and these, in turn, depend upon the forces within and between the molecules. Ice is known to be molecular, but although the arrangement