[Note added in proof. It has now been shown (Gaydon, 1944b) that the predissociation in the First Positive bands of N2, on which the value of 7.38 ev. for $D(N_2)$ is based, is capable of a different interpretation which leads to $D(N_2) = 9.76$ ev., giving D(NO) = 6.49 ev. (= 52500 cm⁻¹). This is within the range of values (51 000 to 61000 cm^{-1}) required by the results reported above.

Dr. W. Jevons has kindly called my attention to a recent note by Gerö, Schmid and Szily (1943) in which a rotational analysis of the (0, 2) and $(0, 3) \epsilon$ bands of NO, as obtained in *absorption*, is briefly reported. They agree in regarding the ϵ and γ systems as separate and giving B' = 1.99, compared with the above value of 1.99₉. No details of the analysis or discussion of the nature of the upper electronic state are given.]

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THE DIFFRACTION OF X RAYS BY DISTORTED CRYSTAL AGGREGATES-I

By A. R. STOKES,

Senior Rouse Ball Student, Trinity College, Cambridge,

AND A. J. C. WILSON,

Cavendish Laboratory, Cambridge

MS. received 18 December 1943

ABSTRACT Broadening of the Debye-Scherrer lines in x-ray photographs of coldworked metals has been attributed (1) to breaking up of the crystals into " crystallites " whose linear dimensions are $\leq 10^{-5}$ cm., (ii) to the presence of crystal grains of different lattice parameters, and (iii) to distortion of comparatively large crystal grains. The broadening to be expected on the last hypothesis is worked out approximately. It is "found that the "apparent strain" is given by

 $\eta \equiv \beta \cot \theta = 2/\phi_{hin}(0),$

where β is the (corrected) integral breadth of the *hkl* reflection, θ is the Bragg angle, and $\phi_{hkl}(e)de$ is the fraction of the crystal for which the tensile strain in the *hkl* direction is between *e* and e+de.

To relate η with the internal stresses requires some approximations. In cubic crystals fairly plausible assumptions lead to the equation

$$\eta^2 = A + BH,$$

where A and B are constants involving the elastic moduli and the mean square values of the direct and shear stresses, and $H \equiv (k^2 l^2 + l^2 h^2 + h^2 k^2)/(h^2 + k^2 + l^2)^2$. This equation is verified within the rather large experimental error for metal filings and wire. Details of the experimental work will be published elsewhere.

§1. INTRODUCTION

LINE-BROADENING in x-ray photographs of cold-worked metals has been the subject of many investigations. Three suggestions have been made as to its origin: 10^{-5} to 10^{-5} cm.

(i) that the metal is broken up into "crystallites" so small $(10^{-5} \text{ to } 10^{-6} \text{ cm.})$ in linear dimensions) that diffraction broadening occurs;

(ii) that the metal is broken up into crystals ($\sim 10^{-4}$ cm. in linear dimensions) with different mean lattice parameters; and

(iii) that the broadening is due to distortion of fairly large crystals ($\sim 10^{-4}$ cm. in linear dimensions).

The distinction between the second and third hypotheses is chiefly in the distortion assumed to be present. In the second hypothesis, the only "distortion" is a uniform expansion or contraction of the whole crystal, whereas in the third, non-uniform expansions and contractions, as well as twists and bends, are allowed.

The first hypothesis is found chiefly in the later papers of Wood (1941, 1943), the second is associated with Brindley (1940), and special cases of the third have been treated by Dehlinger (1927), Boas (1937), and others. On the first hypothesis, $\beta \cos \theta / \lambda$ (where β is the corrected integral breadth of the Debye-Scherrer line, θ the Bragg angle and λ the x-ray wave-length) should be independent of θ and λ , and on the second and third hypotheses $\beta \cot \theta$ should be independent of θ and λ . The problem is complicated by the fact that, in addition to the general variation of β with θ , β may vary in a complicated way with the indices of reflection. On the first ("fragmentation") hypothesis, the variation will depend on the shape of the fragments (Patterson, 1939; Waller, 1939; Stokes and Wilson, 1942), but can hardly be very great. On the second hypothesis, at any rate in the simple form stated above, no variation would be expected. On the third ("distortion") hypothesis, the variation would be zero only if the tensile strain were, on the average, independent of crystallographic direction. As, however, even cubic crystals are not elastically isotropic, this would involve a variation of stress with crystallographic direction. On general considerations of static equilibrium it seems more likely that the stress will be, on the average, independent of crystallographic direction. The resulting variation of tensile strain will then produce a variation of $\beta \cot \theta$.

In the present paper certain consequences of the third hypothesis are investigated. In his discussion of line-broadening by small crystals, Jones (1938) found it convenient to introduce a quantity, the "apparent particle size", defined by

which is independent of wave-length and angle of reflection, and depends only on the thickness of the crystal measured perpendicular to the reflecting planes (Waller, 1939; Stokes and Wilson, 1942). Similarly, in discussing linebroadening by internal strains, it is convenient to introduce the "apparent tensile strain" defined by

$$\eta = \beta \cot \theta, \qquad \dots \dots (2)$$

which is independent of the wave-length and angle of reflection, and, to the approximation to which it has been possible to carry the calculations, depends only on the distribution of tensile strains perpendicular to the reflecting planes. If the fraction of the crystal for which the tensile strain in the *hkl* direction lies between e and e + de is $\phi_{hkl}(e)de$, then (§ 2 below)

$$\eta = 2/\phi_{hkl}(0); \qquad \dots \dots (3)$$

i.e., the apparent strain is twice the integral breadth of the curve giving the distribution of tensile strain in the hkl direction. It is therefore possible to obtain $\phi_{hkl}(0)$ from measurements of integral breadth. In principle it is possible to obtain the whole curve $\phi_{hkl}(e)$ from photometry of both sharp and broadened lines, but it requires a rather complicated analysis of the photometer curves, involving the numerical evaluation of Fourier transforms.

Thus, for any particular reflection, the consequences of hypotheses (ii) and (iii) are indistinguishable—to the approximation reached it does not matter whether the variation of lattice parameter takes place in a single crystal grain or from grain to grain, each grain having a constant lattice parameter. Hypothesis (iii), however, offers some hope of correlating the variation of η with the mean stresses and the elastic constants of the material. In the present paper the consequences of the assumption that the stresses are statistically isotropic are worked out for two approximations to the stress distribution. Both lead to the result that η is a function of $H \equiv (k^2l^2 + l^2h^2 + h^2k^2)/(h^2 + k^2 + l^2)^2$ only. The second approximation seems preferable, and leads to the relation

$$\eta^2 = A + BH, \qquad \dots \dots (4)$$

where A and B are constants depending on the elastic constants of the crystal and the mean square values of the direct and shear stresses. This equation is verified within the rather large experimental error for Cu and Cu₃Ni filings and cold-worked Cu wire. The experimental results will be given in detail elsewhere.

§2. GENERAL CALCULATION OF THE INTEGRAL BREADTHS

It has been shown (Wilson, 1943) that the apparent particle size of imperfect crystals is given by

$$\epsilon = J_0^{-1} \int_{-\infty}^{+\infty} J_t dt, \qquad \dots \dots (5)$$

where J_t is the mean value of the product FF^* of the structure amplitudes of two cells separated a distance t in the *hkl* direction. In a distorted crystal the structure amplitude of the unit cell originally at r will differ from that at the

origin for two reasons: the cell is displaced bodily by an amount $\mathbf{u}(\mathbf{r})$, and suffers a rotation of $\frac{1}{2}\nabla \times \mathbf{u}$. The effect of the rotation on the phase of the x rays scattered from the cell is small compared with that of the displacement, and may be neglected in a first approximation. The structure amplitude of the cell at **r** for the *hkl* reflection becomes, therefore,

$$F' = F \exp\{-2\pi i \mathbf{h} \cdot \mathbf{u}\}; \qquad \dots \dots (6)$$

F is the structure amplitude of a unit cell of the undistorted crystal and $\mathbf{h} = h\mathbf{b}_1 + k\mathbf{b}_2 + l\mathbf{b}_3$, where the **b**s are the edges of the unit cell of the reciprocal lattice. The product of the structure amplitudes of two cells separated a distance t in the direction of \mathbf{h} is therefore

$$F'F'^* = FF^* \exp\{-2\pi i \mathbf{h} \cdot \mathbf{u}(\mathbf{r})\} \exp\{2\pi i \mathbf{h} \cdot \mathbf{u}(\mathbf{r} + t\mathbf{h}/|\mathbf{h}|)\}$$
$$= FF^* \exp\{2\pi i \mathbf{h} \cdot [\mathbf{u}(\mathbf{r} + t\mathbf{h}/|\mathbf{h}|) - \mathbf{u}(\mathbf{r})]\}. \qquad (....(7)$$

The mean value of $F'F'^*$ cannot be evaluated exactly without assuming a particular variety of distortion. It is, however, highly improbable that a cold-worked metal would contain *periodic* distortions, and for non-periodic distortions the following plausible arguments lead to an approximate general evaluation. For large values of t the relative displacements of the cells will be large and random, so that the mean value of $F'F'^*$ will vanish. For small values of t, $\mathbf{u}(\mathbf{r}+t\mathbf{h}/|\mathbf{h}|)$ may be expanded in a power series in t, and terms in t^2 and beyond neglected. Then

$$J_{t} = \overline{F'F'^{*}} = FF^{*} \exp \left\{ 2\pi i \mathbf{h} \cdot \left[\mathbf{u}(\mathbf{r}) + (t/|\mathbf{h}|) \mathbf{h} \cdot \nabla \mathbf{u} + \dots - \mathbf{u}(\mathbf{r}) \right] \right\}$$

$$= FF^{*} \exp \left\{ 2\pi i t \mathbf{h} \cdot \nabla \mathbf{u} \cdot \mathbf{h}/|\mathbf{h}| \right\}. \qquad (8)$$

In this equation $\nabla \mathbf{u}$ is the tensor of which the strain tensor e_{ij} is the symmetrical part and the rotation tensor ω_{ij} is the anti-symmetrical part. It is readily verified that the tensile strain in the *hkl* direction, e_{hk} , is equal to $|\mathbf{h}|^{-2}\mathbf{h}$. $\nabla \mathbf{u} \cdot \mathbf{h}$, so

Equation (9) should therefore be a good approximation to $\overline{F'F'^*}$ for any distribution of strains that makes J, approach zero rapidly for large t. If the fraction of the crystal for which e_{hh} lies between e and e + de is $\phi_{hkl}(e)de$, then

$$J_t = FF^* \int_{-\infty}^{+\infty} \phi_{hkl}(e) \exp\left\{2\pi i \left| \mathbf{h} \right| te\right\} de \qquad \dots \dots (10)$$

and

$$\epsilon = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \phi_{hkl}(e) \exp\{2\pi i |\mathbf{h}| te\} de dt,$$

= $|\mathbf{h}|^{-1} \phi_{hkl}(0),$
= $(\lambda/2 \sin \theta) \phi_{hkl}(0),$ (11)
 $\beta = \lambda/\epsilon \cos \theta = 2 \tan \theta/\phi_{hkl}(0),$ (12)

Measurements of integral breadths will therefore give $\phi_{hkl}(0)$. It is, however, possible to obtain $\phi_{hkl}(e)$ by a complete analysis of photometer curves of broadened and unbroadened lines. Suppose that the radiation reflected with an angular

deviation between $2(\theta + \gamma)$ and $2(\theta + \gamma + d\gamma)$ is $I(\gamma)d\gamma$. Then, in the notation of this paper, equation (4) of Wilson (1943) may be written

$$I(\gamma) = N \int_{-\infty}^{+\infty} J_t \exp\left\{2\pi i M \gamma t\right\} dt, \qquad \dots \dots (14)$$

where N is the number of unit cells in the crystal and M is $2\cos\theta/\lambda$. On substituting the value of J_t from equation (10), this becomes

$$I(\gamma) = NFF* \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \phi_{hkl}(e) \exp\{2\pi i |\mathbf{h}| te\} \exp\{2\pi i M\gamma t\} dt$$
$$= NFF*\phi_{hkl}(-M\gamma/|\mathbf{h}|) \qquad \dots \dots (15)$$

or

$$\frac{\phi_{hkl}(e)}{\phi_{hkl}(0)} = \frac{I(-|\mathbf{h}| e/M)}{I(0)} = \frac{I(-e \tan \theta)}{I(0)}.$$
 (16)

Thus ϕ_{hkl} is the same function of e as I is of $-e \tan \theta$. The elimination of incidental broadenings from the observed photometer curves to obtain $I(\gamma)$ involves Fourier analyses of both sharp and broadened lines. It is being treated elsewhere by one of us (Stokes, 1944).

§3. CALCULATION OF THE INTEGRAL BREADTHS FOR PARTICULAR MODELS

The problem of calculating β is thus reduced to the problem of calculating the fraction of the crystal for which the tensile strain in the *hkl* direction lies between 0 and *de*, i.e. $\phi_{hkl}(0)de$. This can be done if some particular model for the distortion is assumed, or by the use of approximations. Various approximations which suggest themselves lead to the result that $\beta \cot \theta$ is a function of $H \equiv (k^2l^2 + l^2h^2 + h^2k^2)/(h^2 + k^2 + l^2)^2$, but the exact form of the function depends on the approximations. Two of the simplest are

(i) all values of $|e_{hh}|$ between zero and a maximum equally likely, and e_{hh} = Direct Stress/Young's Modulus; or

(ii) mean value of $\exp\{ix\}$ equal to $\exp\{-\overline{x^2}/2\}$, where $\sqrt{\overline{x^2}}$ is the root mean square value of x, the stress being isotropic on the average in each case.

The assumption (ii) is exact if x is distributed according to a Gaussian error curve, and is in any case true as far as terms in x^3 . One would expect, therefore, that (ii) would always be a fair approximation.

On approximation (i), $\phi(0) = 1/2(e_{hh})_{max}$, and

$$\eta = \beta \cot \theta = 4(e_{hh})_{\max}. \qquad \dots \dots (17)$$

If the assumption that the maximum stress is independent of direction is made, the maximum value of e_{hh} will be the maximum stress p_{max} divided by the appropriate value of Young's Modulus, i.e.

$$\beta \cot \theta = 4p_{\max}\{s_{12} + 2s_{44} + (s_{11} - s_{12} - 2s_{44})(h^4 + h^4 + h^4)/(h^2 + h^2 + h^2)^2\}.$$
(18)

Except that it contains the maximum stress instead of the yield stress, this is the expression used by Stokes, Pascoe and Lipson (1943) in interpreting their results on copper filings. Since $h^4 + k^4 + l^4 = (h^2 + k^2 + l^2)^2 - 2(k^2l^2 + l^2h^2 + h^2k^2)$, equation (18) may be written

$$\beta \cot \theta = 4p_{\max}\{s_{11} - 2(s_{11} - s_{12} - 2s_{44})(k^2l^2 + l^2h^2 + h^2k^2)/(h^2 + k^2 + l^2)^2\}$$

= A + BH,(19)

where A and B are constants for a given specimen, and

$$H \equiv \frac{(k^2 l^2 + l^2 h^2 + h^2 k^2)}{(h^2 + k^2 + l^2)^2}.$$

Values of H for the reflections commonly appearing on Debye-Scherrer photographs are given in the following table.

hkl	$h^2 + k^2 + l^2$	H	hkl	$h^2 + k^2 + l^2$	H
100	1	0.000	410*	17	0.055
110	2	0.250	322*	17	0.304
111	3	0.333	330*	18	0.250
200	4	0.000	411*	18	0.102
210	5	0.160	331	19	0.274
211	6	0.220	420	20	0.160
	7		421	21	0.190
220	8	0.220	332	22	0.316
300*	9	0.000		23	
221*	9	0.296	422	24	0.250
310	10	0.090	500*	25	0.000
311	11	0.157	430*	25	0.230
222	12	0.333	510*	26	0.037
320	13	0.213	431*	26	0.250
321	14	0.250	333*	27	0.333
—	15		511*	27	0.070
400	16	0.000			

Values of $H = (k^2 l^2 + l^2 h^2 + h^2 k^2)/(h^2 + k^2 + l^2)^2$

* Overlapping reflections.

On approximation (ii), equation (9) gives

$$J_{t} = FF^{*} \exp\left\{-2\pi^{2}|\mathbf{h}|^{2}\overline{e_{hh}^{2}}t^{2}\right\}, \qquad \dots \dots (20)$$

$$\epsilon = J_{0}^{-1} \int_{-\infty}^{+\infty} J_{t} dt = (2\pi |\mathbf{h}|^{2}\overline{e_{hh}^{2}})^{-\frac{1}{2}},$$

$$= (\lambda/2\sin\theta)(2\pi\overline{e_{hh}^{2}})^{-\frac{1}{2}},$$

$$\beta = \lambda/\epsilon\cos\theta = 2(2\pi\overline{e_{hh}^{2}})^{\frac{1}{2}}\tan\theta,$$

$$\eta = \beta\cot\theta = 2(2\pi\overline{e_{hh}^{2}})^{\frac{1}{2}}. \qquad \dots \dots (21)$$

The same result is of course obtained by assuming that e_{hh} is distributed according to a Gaussian error curve. Evaluating the constants in

$$\phi_{hkl}(e) = C \exp\{-\alpha e^2\}$$

in terms of $\overline{e_{hh}^2}$ gives

and hence

$$\phi_{hkl}(e) = (2\pi \overline{e_{hh}^2})^{-\frac{1}{2}} \exp\{-e^2/2\overline{e_{hh}^2}\},\\eta = 2/\phi_{hkl}(0) = 2(2\pi \overline{e_{hh}^2})^{\frac{1}{2}}.$$
 (21)

<u>so</u>

The calculation of $\overline{e_{hh}^2}$ calls for some consideration. In terms of the strains referred to the crystal axes,

$$e_{hh} = (h^2 e_{11} + k^2 e_{22} + l^2 e_{33} + 2k l e_{23} + 2l h e_{31} + 2h k e_{12})/(h^2 + k^2 + l^2),$$

or, in terms of the stresses referred to the crystal axes,

$$e_{hh} = [h^{2}(s_{11}p_{11} + s_{12}p_{22} + s_{12}p_{33}) + k^{2}(s_{12}p_{11} + s_{11}p_{22} + s_{12}p_{33}) + l^{2}(s_{12}p_{11} + s_{12}p_{22} + s_{11}p_{33}) + 4s_{44}klp_{23} + 4s_{44}lhp_{31} + 4s_{44}hkp_{12}]/(h^{2} + k^{2} + l^{2}). \qquad (22)$$

In finding the mean value $\overline{e_{hh}^2}$ of e_{hh}^2 , it is necessary to evaluate an expression containing mean values of products of pairs of the *p*s. It can be shown that, when the statistical distribution of stress is spherically symmetrical,

$$\overline{p_{11}^2} = \overline{p_{22}^2} = \overline{p_{33}^2}, \qquad \dots \dots (23)$$

$$p_{22}p_{33} = p_{33}p_{11} = p_{11}p_{22},$$
(24)

$$\overline{p_{11}^2} - \overline{p_{11}p_{22}} = 2\overline{p_{12}^2}, \qquad \dots \dots (26)$$

$$\overline{p_{11}p_{12}} = \overline{p_{22}p_{23}} = \overline{p_{33}p_{31}} = \overline{p_{11}p_{13}} = \overline{p_{22}p_{21}} = \overline{p_{33}p_{32}} = 0, \quad \dots \dots (27)$$

$$p_{31}p_{12} = p_{12}p_{23} = p_{23}p_{31} = 0. \qquad \dots \dots (29)$$

Using these relations and equation (22) gives

$$\overline{g_{hh}^2} = \{ [(h^2 s_{11} + k^2 s_{12} + l^2 s_{12})^2 + \text{cyclic permutations}] \overline{p_{11}^2} \\ + 2[(h^2 s_{12} + k^2 s_{11} + l^2 s_{12})(h^2 s_{12} + k^2 s_{12} + l^2 s_{11}) + \text{cyclic permutations}] \overline{p_{11}p_{22}} \\ + 16s_{44}^2 (k^2 l^2 + l^2 h^2 + h^2 k^2) \overline{p_{12}^2} / (h^2 + k^2 + l^2)^2,$$

which becomes on reduction

$$\overline{e_{hh}^{2}} = (s_{11} + 2s_{12})^{2} \overline{p_{11}^{2}} - 4s_{12}(s_{12} + 2s_{11}) \overline{p_{12}^{2}} - 4[(s_{11} - s_{12})^{2} - 4s_{44}^{2}](k^{2}l^{2} + l^{2}h^{2} + h^{2}k^{2})(h^{2} + k^{2} + l^{2})^{-2} \overline{p_{12}^{2}}, \qquad \dots \dots (30) = A' + B'H, \qquad \dots \dots (31)$$

where A' and B' are independent of the order of reflection and depend only on the elastic constants of the metal and the mean square stresses. Then

$$\eta = \beta \cot \theta = 2[2\pi(A' + B'H)]^{\frac{1}{2}} \qquad \dots \dots (32)$$

$$= (A+BH)^{\frac{1}{2}}, \qquad \dots \dots (33)$$

$$\eta^2 = A + BH, \qquad \dots \dots (34)$$

where A and B are constants for a given specimen and

$$H \equiv \frac{k^2 l^2 + l^2 h^2 + h^2 k^2}{(h^2 + k^2 + l^2)^2}$$

Experimental results obtained at this laboratory on metal filings and wire are in fair agreement with equations (19) and (34). The results are not sufficiently accurate to distinguish between them, but on the whole (34) seems preferable,

§4. CRITICISMS

Since the calculation given above depends on a number of approximations it will be well to reiterate them. They are as follows:

(a) The effect of the rotation of the cells has been neglected in finding $F'F'^*$.

(b) The distortion is assumed to be such that the mean value of $F'F'^*$ tends to zero as t increases, and that u(r+th/|h|) - u(r) can be expanded without going beyond the term in t for values of t for which $F'F'^*$ is not negligible.

(c) (1) In the approximation (i), leading to equation (19), only the direct stresses are taken into account. This renders equation (19) somewhat less plausible than equation (34).

(2) In the approximation (ii), leading to equation (34), the distribution of stresses is assumed nearly normal. A different distribution of stresses would. however, only have the effect of changing the constant $2(2\pi)^{\frac{1}{2}}$ in equation (21). and would not, therefore, alter the form of equation (34).

(d) It is assumed that the stress distribution is statistically isotropic. This is open to two objections. First, it gives rise to geometrically incompatible strains in differently oriented crystals. The true distribution is more likely to be something between isotropic stress and isotropic strain, but the variation of η with H should still be of the same general kind. Secondly, it neglects the fact that the shear stress in some directions is probably relieved by the action of slip planes in the crystal.

It seems certain, therefore, that with sufficiently precise measurements of integral breadths, equations (19) and (34) will be found inexact. The manner in which they break down will perhaps indicate where the treatment is in greatest need of refinement.

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