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The Scattering of X-rays by Face Centred Cubic Crystals Containing Condensed Sheets of Interstitial Atoms

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A calculation has been made of the distribution in reciprocal space of X-rays scattered by a face centred cubic crystal containing sheets of interstitial atoms condensed at random to form stacking faults. It is shown that these faults produce asymmetric peaks at non-integral values of *hkl* and the appearance of peaks at the position corresponding to reflexion from the crystal of opposite stacking sequence. In neutron-irradiated crystals where interstitial atoms condense on $\{111\}$ planes these effects should be detectable.

Introduction

There are many references in the literature to defect clusters produced in copper by neutron irradiation and the supposition is made that clusters greater than 50 Å in diameter result from interstitial condensation on $\{111\}$ planes (Barnes & Mazey, 1960; Makin, Whapham & Minter, 1960, 1961; Mazey, Barnes & Howie, 1962).

Makin & Manthorpe (1963) found that after irradiation to $2{\cdot}5\times10^{18}$ nvt (>1 MeV) at 27°C the concentration of point defects in these clusters is $2{\cdot}8\times10^{-4}$.

It is probable that irradiation in the 10^{19}–10^{20} nvt dose range will produce sufficient concentration of these defects for direct observation by X-ray diffraction.

Johnson (1962) calculated the intensity distribution of X-rays scattered by a face-centred cubic crystal containing extrinsic stacking faults, but he assumed that only one condensation can take place between any two close-packed layers of the original crystal; thus in the standard notation for cubic close-packing, if a fault produces a C layer after an A layer the following layer must be B.

In the case of irradiation damage the condensation will be a continuous process and successive condensations are possible.

In this paper the intensity distribution in reciprocal space is calculated assuming that interstitial atoms are continually condensing on one set of $\{111\}$ planes. It is assumed that the fault completely covers the plane and that the crystal is large enough for particle size effects to be ignored.

Theory

The usual transformation to hexagonal axes is made (Paterson, 1952) so that two axes are along $(\bar{1}10)$ and $(0\bar{1}1)$ in the close packed layers while the third axis is normal to these layers.

The relation between the Miller indices in the hexagonal lattice and in the cubic lattice is

$$h_{\rm hex}=\tfrac{1}{2}(-h_c+k_c)$$
$$k_{\rm hex}=\tfrac{1}{2}(-k_c+l_c)$$
$$l_{\rm hex}=(h_c+k_c+l_c)\,.$$

The method of calculation is based on that of Wilson (1942) for growth faults in cobalt in which it is shown that the diffracted intensity from a mono-dimensionally disordered layer structure can be written (when crystal size effects are ignored),

$$I(hkl)=\delta(H-h)\,\delta(K-k)\sum_{-\infty}^{+\infty}J_m\exp\frac{2\pi iml}{3}\,,$$

or, equivalently,

$$I(HKl)=\delta(H-h)\,\delta(K-k)\left\{J_0+2\sum_1^{\infty}\mathscr{R}(J_m)\cos\frac{2\pi iml}{3}-2\sum_1^{\infty}\mathscr{I}(J_m)\sin\frac{2\pi\,iml}{3}\right\},\quad(1)$$

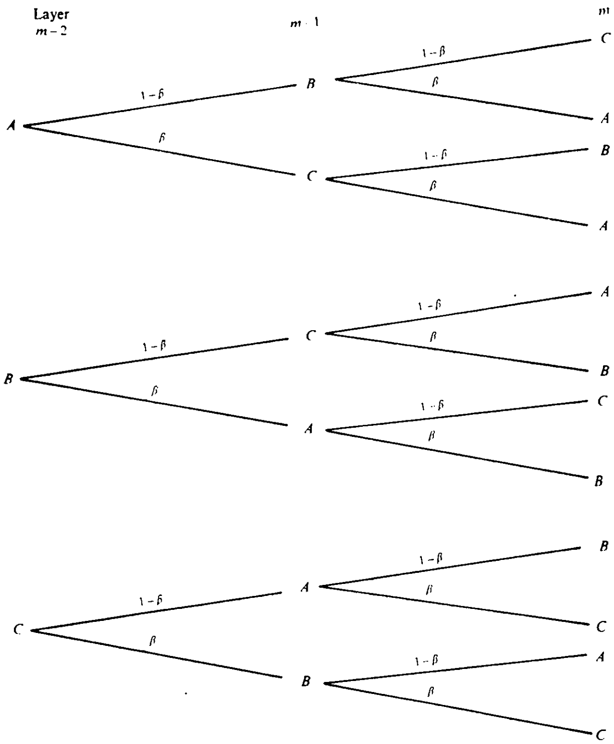
where hkl are continuous variables based on the reciprocal of the hexagonal lattice defined above, with integral values H, K, and L. The factor $\frac{1}{3}$ occurs in the exponent because l refers to a cell three layers high. J_m is the average value of the product $F_jF_{j+m}^*$ where j runs over all layers in the crystal separated by m interlayer spacings.

Evaluation of J_m

Let α be the (random) probability that a condensed layer forms, after any given layer. If this layer is A the succeeding layer will be B unless there is an odd number of successive condensations after A. Thus the probability that the layer sequence is AB is

$$1-\alpha+\alpha^2-\alpha^3+\alpha^4-\ldots=\frac{1}{1+\alpha}\,.$$

The packing sequence in the crystal leading to the mth layer being of a particular type can be written:



where $\beta = \dfrac{\alpha}{1+\alpha}$.

The probability of the mth layer being A can be written from these sequences as:

$$P_m^A = \beta P_{m-2}^A + (1-\beta)^2 P_{m-2}^B + \beta(1-\beta) P_{m-2}^C .$$

From the two additional relations:

$$P_{m-1}^A = \beta P_{m-2}^B + (1-\beta) P_{m-3}^C$$
$$P_{m-2}^A = 1 - P_{m-2}^B - P_{m-2}^C ,$$

the difference equation for P_m^A can be written:

$$P_m^A + (1-\beta) P_{m-1}^A + (1-2\beta) P_{m-2}^A = 1 - \beta (\beta \neq \tfrac{1}{2}). \quad (2)$$

This equation can be solved for positive values of m in the usual way by writing $P_m^A = a + bx^m$ which leads to

$$a = \tfrac{1}{3} \text{ and } x^2 + (1-\beta)x + 1 - 2\beta = 0 . \quad (3)$$

The roots of the quadratic equation are:

$$\tfrac{1}{2}\{-(1-\beta) \pm \sqrt{-3+6\beta+\beta^2}\} .$$

For values of β less than $2\sqrt{3}-3$ ($a < \sqrt{3}/2$) these roots are complex and can be written:

$$x = Re^{\pm i\gamma} ,$$

where

$$R = (1-2\beta)^{\frac{1}{2}}$$
$$R\cos\gamma = -(1-\beta)/2$$
$$R\sin\gamma = \sqrt{3-6\beta-\beta^2}/2 .$$

The solution of (2) is then:

$$P_m^A = \tfrac{1}{3} + b_1 Re^{i\gamma} + b_2 Re^{-i\gamma} .$$

The coefficients b_1 and b_2 can be found by using as boundary conditions the state of the zero and first layers for the three cases P_A^A, P_B^A, P_C^A. When this is done we have specific values of the probabilities to insert into the expression for J_m which is, for positive m,

$$J_m = \tfrac{1}{3} \left\{ \begin{array}{l} P_A^A F_A F_A^\star + P_B^A F_A F_B^\star + P_C^A F_A F_C^\star \\ + P_A^B F_B F_A^\star + P_B^B F_B F_B^\star + P_C^B F_B F_C^\star \\ + P_A^C F_C F_A^\star + P_B^C F_C F_B^\star + P_C^C F_C F_C^\star \end{array} \right\} \quad (4)$$

The F's are the layer structure factors, and, taking the X-ray scattering factor as unity, can be written:

$$F_A = 1$$
$$F_B = e^{i\varphi}$$
$$F_C = e^{-i\varphi}$$

where

$$\varphi = \frac{2\pi}{3}(H-K) .$$

Substitution in (4) gives:

$$J_m = P^{AA} + P^{CA} e^{-i\varphi} + P^{BA} e^{i\varphi} .$$

Substitution of the explicit values for the probabilities gives:

$$J_m = 1 \text{ for } H-K = 3N$$

and for $H-K = 3N \pm 1$

$$\mathscr{R}(J_m) = \frac{-R^m}{2R\sin\gamma}(\sin m\gamma + 2R\sin(m-1)\gamma)$$

$$\mathscr{I}(J_m) = \frac{\pm\sqrt{3}(2\beta-1)R^m \sin m\gamma}{2R\sin\gamma} .$$

For $\beta \geq 2\sqrt{3}-3$ the roots of (3) are real and the solution of (2) is written:

$$P_m^A = \tfrac{1}{3} + b_1 p^m + b_2 q^m ,$$

where

$$p = \tfrac{1}{2}\{-(1-\beta) + \sqrt{-3+6\beta+\beta^2}\} ,$$
$$q = \tfrac{1}{2}\{-(1-\beta) - \sqrt{-3+6\beta+\beta^2}\} .$$

The boundary conditions are identical with those for complex roots of equation (3) and the b's are found in the same way.

Insertion of explicit values for the probabilities into (4) leads to

$$J_m = 1, \text{ for } H-K = 3N,$$

and, for $H-K = 3N \pm 1$,

$$\mathscr{R}(J_m) = -\frac{1}{2(p-q)}\left\{(2q+1)p^m - (2p+1)q^m\right\}$$

$$\mathscr{I}(J_m) = \mp \frac{\sqrt{3}(1-2\beta)}{2(p-q)}\left\{p^m - q^m\right\} .$$

Evaluation of intensity distribution

The values of the real and imaginary parts of J_m are inserted into (1) and by using the general summations:

$$\sum_{m=1}^{\infty} x^m \cos m\varphi = \frac{x\cos\varphi - x^2}{1 - 2x\cos\varphi + x^2}$$

$$\sum_{m=1}^{\infty} x^m \sin m\varphi = \frac{x\sin\varphi}{1 - 2x\cos\varphi + x^2} ,$$

and expressing the products of trigonometrical expressions as sums, the following expressions are obtained for the roots of (2), complex and real respectively:

$$I(HKl)=1+\frac{1}{1-2R\cos A+R^2}\left\{-R\sin A\left(\frac{1}{2R\sin\gamma}+\cot\gamma\right)+(R\cos A-R^2)\left(1\pm\frac{\sqrt{3}(2\beta-1)}{2R\sin\gamma}\right)\right\}$$

$$+\frac{1}{1-2R\cos B+R^2}\left\{-R\sin B\left(\frac{1}{2R\sin\gamma}+\cot\gamma\right)+(R\cos B-R^2)\left(1\mp\frac{\sqrt{3}(2\beta-1)}{2R\sin\gamma}\right)\right\},$$

where

$$A=\gamma+\theta,\ B=\gamma-\theta,\ \theta=\frac{2\pi l}{3},$$

and

$$I(HKl)=1+\frac{1}{p-q}\times\left\{\frac{(2p+1)(q\cos\theta-q^2)\mp\sqrt{3}(1-2\beta)q\sin\theta}{1-2q\cos\theta+q^2}-\frac{(2q+1)(q\cos\theta-p^2)\mp\sqrt{3}(1-2\beta)p\sin\theta}{1-2p\cos\theta+p^2}\right\}.$$

Machine computations have been carried out to determine the line profiles as the number of condensed sheets increase. The following effects emerge.

(1) Assymetric line profiles are produced with peak shifts away from integral values of l towards $l=3N+3/2$. Simultaneously reflexions, whose profiles are also asymmetric, appear at the positions corresponding to reflexions from a crystal which has the packing sequence *CBA*.

(2) For $\alpha>\frac{1}{2}$ the twin and normal peaks coalesce to an asymmetric peak displaced from $1=3N+3/2$ toward the position of the normal peak.

(3) At the limit ($\alpha=1$) the intensity distribution is symmetric about $1=3N+3/2$.

These effects are illustrated in Fig. 1, where the calculated profiles for various value of α are shown.

At low fault densities the present analysis should agree with that of Johnson, but the predicted behaviour from the two calculations is quite different. Johnson finds a shift of the peak away from the twin reflexion position for low fault densities whereas the present analysis predicts a continuous movement toward the twin position as the fault probability increases, and the appearance of a peak at the twin position immediately a fault is introduced.

Conclusion

In an irradiated crystal these condensation faults, if they occur, will be distributed with equal probability on all four $\{111\}$ planes so that the observed effects will be a superposition of those due to faults on each

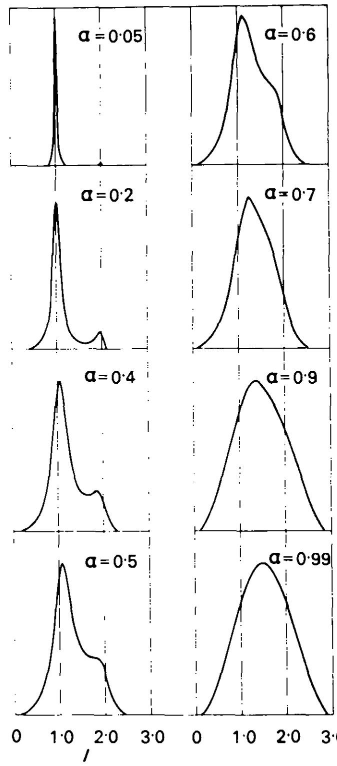


Fig. 1. Variation of reflexion profiles, with condensation probability.

plane. However the appearance of reflexions at the twin position should be characteristic of this cluster formation. For defect densities of the order of those produced by irradiation the ratio of the peak intensities of the normal and twin spots are given below.

α	I_N/I_T
0·001	2000
0·002	1000
0·005	400
0·01	200

They should therefore be detectable.

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