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The Diffraction of X-rays by Close-packed Crystals Containing both 'Growth Stacking Faults' and 'Deformation or Transformation Stacking Faults'

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The general theory is developed of the X-ray diffraction effects exhibited by (*a*) different close-packed structures (*h, k, hk, hkk, hhk, hhkk* types) containing 'growth stacking faults', assuming a four-layer influence; (*b*) hexagonal and close-packed cubic crystals in which 'growth stacking faults' and 'deformation or transformation stacking faults' coexist; and (*c*) *hk*- and *hkk*-crystals (SiC–4*H* and SiC–6*H* types) containing 'deformation stacking faults'.

The manner in which a 'fault' of the first or second type disturbs the regular sequences of the layers is stated in a general way. Expressions are derived for the reciprocal-space intensity distribution as a function of the parameters (four, two, one, one in the different cases) which describe the densities of the faults. It is shown how these parameters can be determined from measurements of the X-ray intensities or the displacements of the peaks of the diffuse reflexions.

1. Introduction

If a crystal lattice can be described as the superposition of close-packed layers in a well defined manner, stacking 'disorder' arises whenever errors are introduced into the regular sequence of the layers.

2. Influence of the 'faults' upon the sequence of the layers

The notation *h* or *k* means that the layer is arranged with respect to the two preceding ones in a hexagonal or cubic manner. The regular sequences of the layers in a close-packed crystal can then generally be represented by symbols, e.g. *hkk* (Jagodzinski, 1949*a*), and this means that a regular sequence can be constructed by periodic repetition of that prescription, starting with two layers.

The irregular sequence of the layers in a close-packed crystal can however only be described with the aid of the transition probabilities, which define the one-dimensional disorder.

Two types of 'faults' will be considered here.

(*A*) '*Growth stacking faults*'

A 'stacking fault' is a 'growth fault' if it arises during growth.

We shall say that an '*n*-layer' influence is present whenever the way in which a new layer is added depends on the arrangement of the *n* preceding ones. A 'growth fault' occurs when the growth prescription is not followed in the new layer.

Since *n* layers may be arranged in 2^{n-2} ways $(kh)_i$, we have to introduce in this case 2^{n-2} parameters, α_i, which are the transition probabilities for continuing the sequence in the cubic way. The irregular sequence is then described statistically by the expressions ω_i, which are the probabilities that *n* successive layers are in a $(kh)_i$ arrangement.

Wilson (1942) and Hendricks & Teller (1942) dealt with $n = 2$, Jagodzinski (1949*b*) with $n = 3$ and Gevers (1952) treated two special cases $n = 4$ and $n = 6$.

In § 3 of this article we shall give the general calculations for $n = 4$, using the method of Wilson (1942).

In this case the continuation of the irregular stacking of the close-packed layers is described by the rule:

$$(kh)_i \begin{cases} \rightarrow h \text{ probability } 1-\alpha_i & (1a) \\ \rightarrow k \text{ probability } \alpha_i & (1b) \end{cases}$$

$$\text{if } (kh)_1 = hh,\ (kh)_2 = kh,\ (kh)_3 = hk,\ (kh)_4 = kk\,. \quad (1c)$$

(*B*) '*Deformation or transformation stacking faults*'

Imagine an extended block of a crystal based on any pattern, regular or irregular. If one half moves relative to the other, so that the discontinuity is confined to one layer only, we have a 'deformation fault' occurring at the discontinuity and characterized by a continuance of the displacement throughout subsequent layers.

If the displacement is so that one part of the crystal shifts out of the hollows of the layer beneath it into a possible neighbouring set of hollows of the other type, the 'fault' is a 'stacking fault' and the result is a change of type in the layers, beginning with the one in which the discontinuity occurs.

Symbolically we have

$$\text{either} \qquad A \rightarrow B,\ B \rightarrow C,\ C \rightarrow A\,, \quad (2a)$$

$$\text{or} \qquad A \rightarrow C,\ B \rightarrow A,\ C \rightarrow B\,. \quad (2b)$$

Both displacements can be characterized by a vector, respectively

$$\mathbf{d}_1 = \tfrac{1}{3}\mathbf{a}_1 + \tfrac{2}{3}\mathbf{a}_2$$

and

$$\mathbf{d}_2 = \tfrac{2}{3}\mathbf{a}_1 + \tfrac{1}{3}\mathbf{a}_2$$

($\mathbf{a}_1$, $\mathbf{a}_2$: translation vectors of the close-packed layer).

It is clear that we have:

$$\begin{cases} \mathbf{d}_1 + \mathbf{d}_1 = \mathbf{d}_2\,, & (3a) \\ \mathbf{d}_1 + \mathbf{d}_2 = 0 \quad \text{(mod. a lattice vector)}\,, & (3b) \\ \mathbf{d}_2 + \mathbf{d}_2 = \mathbf{d}_1\,. & (3c) \end{cases}$$

This type of 'fault' can be produced either during plastic deformation or during certain phase transformations or even spontaneously at an appropriate temperature.

A 'deformation or transformation fault' not only changes the arrangement (h or k) of the layer in which the discontinuity occurs, but also that of the next one.

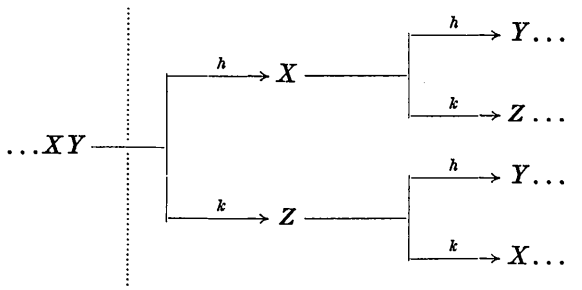


Fig. 1.

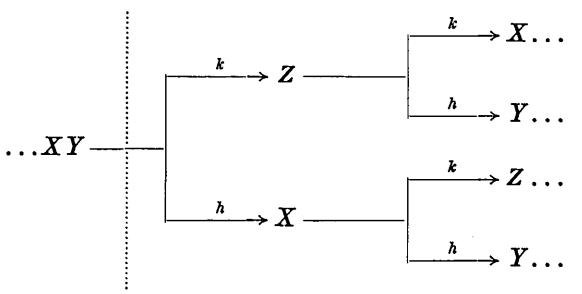


Fig. 2.

Indeed, when X, Y, Z stand respectively for A (B, C), B or C (C or A, A or B), C or B (A or C, B or A), Fig. 1 gives the four possible sequences of four layers in any regular or irregular pattern, with the indication of their arrangement hh, kh, hk or kk.

Suppose now that the two parts on each side of the dotted line be displaced with respect to each other so that a 'deformation or transformation stacking fault' is created.

The symbols of Fig. 1 for the close-packed layers of the upper part have then to be changed in a manner corresponding with (2a) or (2b). The result is Fig. 2, where the arrangements of the four layers are again indicated.

This scheme proves clearly that the arrangement of the layer in which the discontinuity occurs and that of the next one are both changed ($h \to k$, $k \to h$). The arrangement h or k of the other layers is not altered by the process, since the three layers, which form the arrangement h or k, are all either displaced or not.

From now on we shall call the layer itself in which the discontinuity occurs 'a deformation or transformation stacking fault', and it will be marked with a dash (A', ..., h', k').

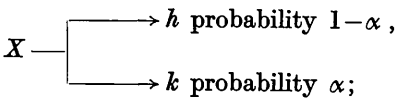
Paterson (1952) has considered the case of a close-packed cubic crystal with randomly distributed 'deformation stacking faults'.

In §§ 5 and 6 we shall deal with the pattern of the hk- and hkk-type ($ABAC$ and $ABCACB$) (SiC-4H and SiC-6H type), where the regular sequence is disturbed by the occurrence of randomly distributed 'deformation faults' with density β.

(C) Faults of both types

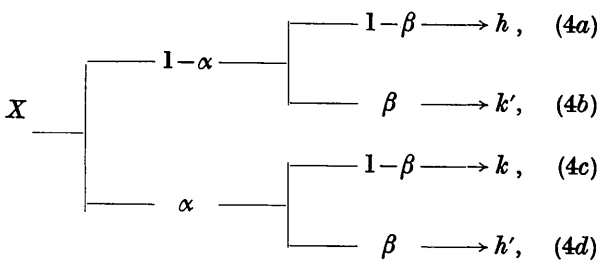
Imagine that, during one-dimensional disordered 'growth' an irregular pattern of close-packed layers is built up, and that this pattern is afterwards altered by the appearance of 'deformation or transformation stacking faults'.

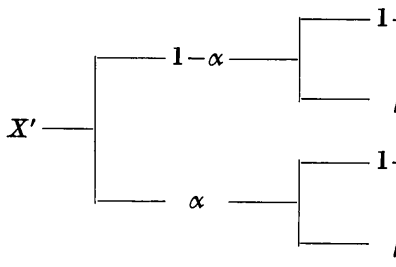
In § 4 we shall deal with the case in which (1) the irregular sequence is the one obtained in the case of a 'two-layer' influence (Wilson, 1942; Hendricks & Teller, 1942); this means that the pattern is one of a close-packed hexagonal or close-packed cubic crystal with 'growth faults', where the continuation of the irregular stacking of the close-packed layers is given by the rule



(2) the density of the 'deformation faults', introduced into the irregular sequence, is β.

Remembering that the introduction of a 'fault' of the second type has the effect of interchanging the arrangement h or k of two layers, we obtain the following rule:





(4e) (4f) (4g) (4h)

in which a layer which is a 'deformation fault' is marked with a dash (X', h', k').

3. Calculation of P_m for a sequence with 'growth faults' for four-layer influence

We shall first calculate the probabilities $\omega_i (i=1,\ldots,4)$ of finding, when running through the crystal, a layer arranged with respect to its predecessors in a $(kh)_i$ manner.

A layer can only be hh-arranged if it continues h-wise a sequence of four layers which is arranged either in an hh- or a kh-manner. Thus,

$$\omega_1 = \omega_1(1-\alpha_1)+\omega_2(1-\alpha_2) .$$

In the same way,

$$\omega_2 = \omega_3(1-\alpha_3)+\omega_4(1-\alpha_4) ,$$
$$\omega_4 = \omega_3\alpha_3+\omega_4\alpha_4 ,$$

and of course

$$\omega_1+\omega_2+\omega_3+\omega_4 = 1 .$$

From these relations we can calculate easily

$$\omega_1 = (1-\alpha_2)(1-\alpha_4)t,\ \omega_2 = \omega_3 = \alpha_1(1-\alpha_4)t ,$$
$$\omega_4 = \alpha_1\alpha_3 t , \quad (5a)$$

where

$$1/t = (1-\alpha_2)(1-\alpha_4)+2\alpha_1(1-\alpha_4)+\alpha_1\alpha_3 . \quad (5b)$$

P_m will be the probability that two layers, m layers apart, are in the relationship $A\ (B, C)\ldots A\ (B, C)$ and $p_m^{(i)}$ the partial probability that the last $A\ (B, C)$ is arranged with the three preceding layers in a $(kh)_i$ manner.

Thus,

$$P_m = p_m^{(1)}+p_m^{(2)}+p_m^{(3)}+p_m^{(4)} . \quad (6)$$

If the mth layer is A (zero layer A) and is arranged in an hh- (or kh-) manner, the $(m-2)$th layer must also be A and can be related to its three predecessors in any of the four ways.

If we take (1) into account we obtain the two formulae

$$p_m^{(1)} = p_{m-2}^{(1)}(1-\alpha_1)(1-\alpha_1)+p_{m-2}^{(2)}(1-\alpha_2)(1-\alpha_1)$$
$$+p_{m-2}^{(3)}(1-\alpha_3)(1-\alpha_2)+p_{m-2}^{(4)}(1-\alpha_4)(1-\alpha_2) , \quad (7)$$

and

$$p_m^{(2)} = p_{m-2}^{(1)}\alpha_1(1-\alpha_3)+p_{m-2}^{(2)}\alpha_2(1-\alpha_3)$$
$$+p_{m-2}^{(3)}\alpha_3(1-\alpha_4)+p_{m-2}^{(4)}\alpha_4(1-\alpha_4) . \quad (8)$$

When the mth layer is not A, say B, and is arranged in a k-manner, only the sequences ACB and CAB are possible. Thus either the $(m-2)$th or the $(m-1)$th layer has to be A.

We then have immediately two further relations:

$$p_m^{(3)}+p_{m-2}^{(1)}(1-\alpha_1)\alpha_1+p_{m-2}^{(2)}(1-\alpha_2)\alpha_1+p_{m-2}^{(3)}(1-\alpha_3)\alpha_2$$
$$+p_{m-2}^{(4)}(1-\alpha_4)\alpha_2+p_{m-1}^{(1)}\alpha_1+p_{m-1}^{(2)}\alpha_2 = \omega_3 , \quad (9)$$

$$p_m^{(4)}+p_{m-2}^{(1)}\alpha_1\alpha_3+p_{m-2}^{(2)}\alpha_2\alpha_3+p_{m-2}^{(3)}\alpha_3\alpha_4+p_{m-2}^{(4)}\alpha_4\alpha_4$$
$$+p_{m-1}^{(3)}\alpha_3+p_{m-1}^{(4)}\alpha_4 = \omega_4 . \quad (10)$$

In these, the first term gives the probability that the mth layer is A, hk (or kk) arranged; the sum of the four following terms that the mth layer is B or C, not preceded by A, hk (or kk) arranged; the sum of the last two that the mth layer is B or C, preceded by A, hk (or kk) arranged.

Equations (6), (7), (8), (9) and (10) form a system which can be easily solved by the method used in our previous article (Gevers, 1952). We get

$$P_m = \tfrac{1}{3}+\sum_{n=1}^{8} C_n x_n^m , \quad (11)$$

where x_n are the roots of the characteristic equation*

$$\begin{vmatrix} 1 & 1 & 1 & 1 & -1 \\ x^2-(1-\alpha_1)^2 & -(1-\alpha_1)(1-\alpha_2) & -(1-\alpha_2)(1-\alpha_3) & -(1-\alpha_2)(1-\alpha_4) & 0 \\ -\alpha_1(1-\alpha_3) & x^2-\alpha_2(1-\alpha_3) & -\alpha_3(1-\alpha_4) & -\alpha_4(1-\alpha_4) & 0 \\ \alpha_1 x+\alpha_1(1-\alpha_1) & \alpha_2 x+\alpha_1(1-\alpha_2) & x^2+\alpha_2(1-\alpha_3) & \alpha_2(1-\alpha_4) & 0 \\ \alpha_1\alpha_3 & \alpha_2\alpha_3 & \alpha_3 x+\alpha_3\alpha_4 & x^2+\alpha_4 x+\alpha_4^2 & 0 \end{vmatrix} = 0$$

or

$$x^8+\alpha_4 x^7-(1-\alpha_1-\alpha_4)(1-\alpha_1+\alpha_4)x^6$$
$$-[\alpha_4(1-\alpha_1)^2-\alpha_1(1-\alpha_2)(1-\alpha_3)]x^5$$
$$-[\alpha_2^2(1-\alpha_3)^2+\alpha_4^2(1-\alpha_1)^2-\alpha_1\alpha_4(1-\alpha_2)(1-\alpha_3)$$
$$-\alpha_1\alpha_3(1-\alpha_2)(1-\alpha_4)]x^4$$
$$+a_5x^3+a_6x^2+a_7x+(\alpha_3-\alpha_4)^2(\alpha_1-\alpha_2)^2 = 0 . \quad (12)$$

* After this article had been written in its original form, a paper by Kakinoki & Komura (1952) came to our notice, in which the general form of the equation for an 'n-layer influence' was derived by the generalized method of Hendricks & Teller. It can be proved that the form given here can also be derived by their method.

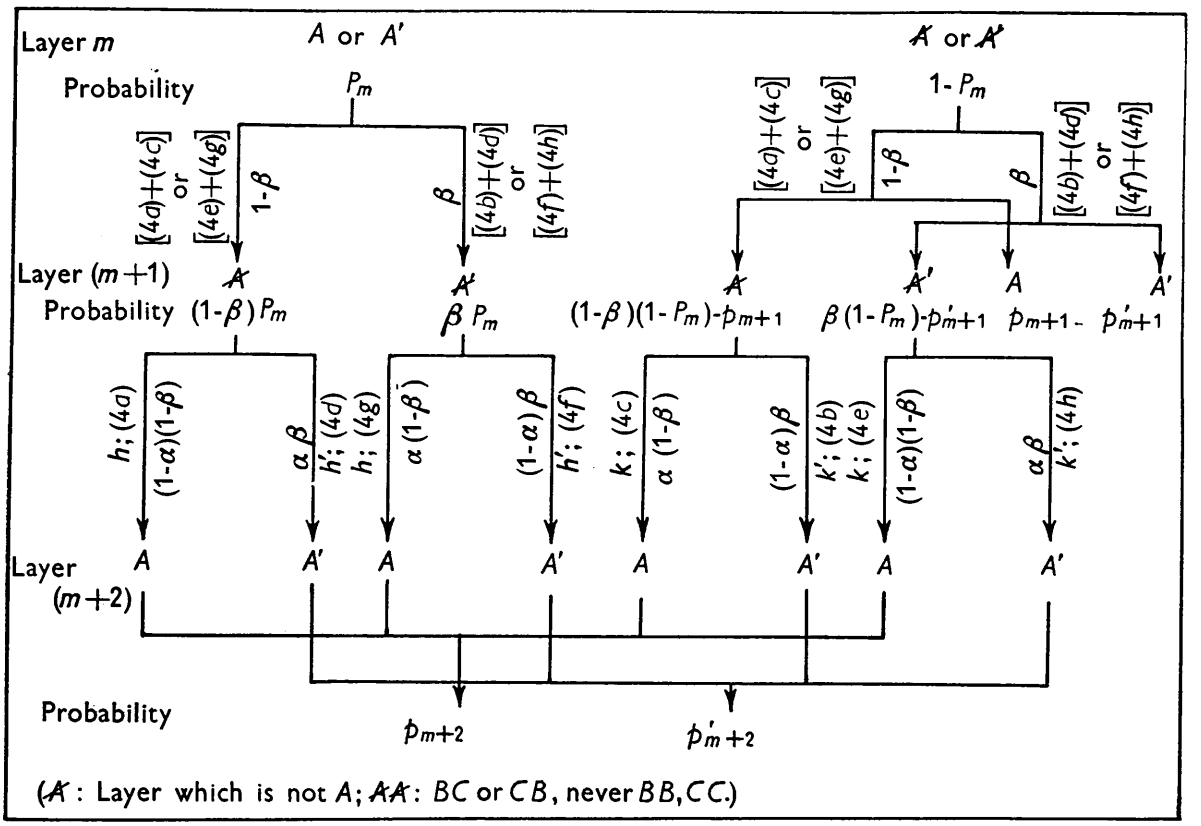


Fig. 3.

The results of Wilson (1942), Jagodzinski (1949*b*) and Gevers (1952) can be obtained as special cases ($\alpha_1=\alpha_2=\alpha_3=\alpha_4$; $\alpha_1=\alpha_2$, $\alpha_3=\alpha_4$; $\alpha_1=\alpha_2=1$, $\alpha_4=0$).

The C_n's are the solution of the system

$$\sum_{n=1}^{8} C_n x_n^m = P_m - \tfrac{1}{3} \quad (m = 0, 1, \ldots, 7)\,. \tag{13}$$

The values of P_m can be calculated with the aid of (6), (7), (8), (9), (10), $p_0^{(i)} = \omega_i$, $p_1^{(i)} = 0$ $(i = 1, \ldots, 4)$ and (5).

4. Calculation of P_m for hexagonal or cubic close-packed crystals with both types of 'stacking faults'

P_m will again be the probability that two layers, m layers apart, are in the relationship A $(B, C)\ldots$ A (B, C). Two cases can occur: (1) the last layer is not a 'deformation fault': probability p_m; (2) the last layer is a 'deformation fault': probability p'_m. Thus

$$P_m = p_m + p'_m\,. \tag{14}$$

Consider now the scheme of Fig. 3. In this figure all possible transitions from layer m to layer $(m+1)$ are shown, and only those transitions from layer $(m+1)$ to layer $(m+2)$ leading to an A for the last layer.

Near the vertical lines we indicate the transition probabilities for the special ways and (in brackets) the rules which are applied, while under each layer symbol (A, A', $\mathcal{A}$, $\mathcal{A}'$) the corresponding occurrence probability is written.

With the aid of the scheme of Fig. 3, we find immediately the two following relations:

$$\begin{aligned} p_{m+2} = {} & (1-\beta)P_m(1-\alpha)(1-\beta) + \beta P_m \alpha(1-\beta) \\ & + [(1-\beta)(1-P_m) - p_{m+1}]\alpha(1-\beta) \\ & + [\beta(1-P_m) - p'_{m+1}](1-\alpha)(1-\beta) \end{aligned} \tag{15}$$

and

$$\begin{aligned} p'_{m+2} = {} & (1-\beta)P_m\alpha\beta + \beta P_m(1-\alpha)\beta + [(1-\beta)(1-P_m) \\ & - p_{m+1}](1-\alpha)\beta + [\beta(1-P_m) - p'_{m+1}]\alpha\beta\,. \end{aligned} \tag{16}$$

Equations (14), (15) and (16) form a linear system which can be solved in the usual way (Gevers, 1952). We obtain

$$P_m = \tfrac{1}{3} + C_1 x_1^m + C_2 x_2^m\,, \tag{17}$$

where x_1 and x_2 are the roots of the equation

$$\begin{vmatrix} x^2+\alpha(1-\beta)x & (1-\alpha)(1-\beta)x & -(1-\beta)(1-2\alpha)(1-2\beta) \\ \beta(1-\alpha)x & x^2+\alpha\beta x & \beta(1-2\alpha)(1-2\beta) \\ 1 & 1 & -1 \end{vmatrix} = 0$$

or

$$x^2+\alpha x+(2\alpha-1)(1-3\gamma) = 0\,, \qquad (18a)$$

where

$$\gamma = \beta(1-\beta)\,. \qquad (18b)$$

If $\beta = 0$, we verify the formula given by Wilson (1942) and if $\alpha = 1$, that given by Paterson (1952). $\alpha = 0$ would give a formula equivalent to that of Paterson, but for the hexagonal close-packed structure.

C_1 and C_2 are obtained by solving the system

$$\left.\begin{aligned} C_1 + C_2 &= \tfrac{2}{3}\,, \\ x_1C_1+x_2C_2 &= -\tfrac{1}{3}\,. \end{aligned}\right\} \qquad (19)$$

5. Calculation of P_m for *hk*-structure (SiC-4*H* type) with 'deformation stacking faults'

In an undisturbed *hk*-crystal we can meet with equal probability:

Layer m $\quad m+1 \quad m+2 \quad m+3 \quad m+4$

$$X \longrightarrow Y \begin{cases} \xrightarrow[(2)]{} X \xrightarrow[(2)]{} Z \xrightarrow[(1)]{} X \ldots, & (20a) \\ \xrightarrow[(1)]{} Z \xrightarrow[(2)]{} Y \xrightarrow[(2)]{} X \ldots. & (20b) \end{cases}$$

The numbers in brackets indicate the manner (1 or 2) in which that particular layer may shift:

Manner (1): $X \longrightarrow Y$, $Y \longrightarrow Z$, $Z \longrightarrow X$;

Manner (2): $X \longrightarrow Z$, $Y \longrightarrow X$, $Z \longrightarrow Y$.

We have that the $(m+4)$th layer will be an A (zero layer: A) (1) when the mth layer is an A and the displacement resulting from all the faults present is 0 (mod. a lattice vector), or (2) when the mth layer is a B or C (or C or B) and the resulting displacement is $\mathbf{d}_2$ (or $\mathbf{d}_1$) (mod. a lattice vector).

We have, if we take into account (3), (20) and all the possible arrangements of correct and faulted layers,

$$P_{m+4} = P_m[(1-\beta)^4+4\beta^2(1-\beta)^2+\beta^4] + (1-P_m)[2\beta(1-\beta)^3+\beta^2(1-\beta)^2+2\beta^3(1-\beta)]$$

or

$$P_{m+4}-(1-3\gamma)^2P_m = \gamma(2-3\gamma)\,, \qquad (21a)$$

where

$$\gamma = \beta(1-\beta)\,. \qquad (21b)$$

Thus

$$P_m = \tfrac{1}{3}+\sum_{n=1}^{4} C_n x_n^m\,, \qquad (22)$$

where x_n are the roots of

$$x^4-(1-3\gamma)^2 = 0\,. \qquad (23)$$

$P_0, P_1, \ldots, P_8$ can be calculated by considering (20) (with $m = 0$), and by taking (21a) into account for the last four.

We have then

$$\left.\begin{aligned} &P_0 = 1, P_1 = 0, P_2 = \tfrac{1}{2}, P_3 = \gamma, \\ &P_4 = 1-4\gamma+6\gamma^2, P_5 = \gamma(2-3\gamma)\,, \\ &P_6 = \tfrac{1}{2}(1-2\gamma+3\gamma^2), P_7 = 3\gamma(1-3\gamma+3\gamma^2), \\ &P_8 = 1-8\gamma+36\gamma^2-72\gamma^3+54\gamma^4\,. \end{aligned}\right\} \qquad (24)$$

6. Calculation of P_m for *hkk* structure (SiC-6*H* type) with 'deformation stacking faults'

The same method gives us

Layer

$m \quad m+1 \quad m+2 \quad m+3 \quad m+4 \quad m+5 \quad m+6$

$$X \longrightarrow Y \begin{cases} \xrightarrow[(2)]{} X \xrightarrow[(2)]{} Z \xrightarrow[(2)]{} Y \xrightarrow[(1)]{} Z \xrightarrow[(1)]{} X \ldots, & (25a) \\ \xrightarrow[(1)]{} Z \xrightarrow[(1)]{} X \xrightarrow[(1)]{} Z \xrightarrow[(2)]{} Y \xrightarrow[(2)]{} X \ldots, & (25b) \\ \xrightarrow[(1)]{} Z \xrightarrow[(2)]{} Y \xrightarrow[(2)]{} X \xrightarrow[(2)]{} Z \xrightarrow[(1)]{} X \ldots, & (25c) \end{cases}$$

$$P_{m+6} = P_m[(1-\beta)^6+9\beta^2(1-\beta)^4+\beta^3(1-\beta)^3+\beta^3(1-\beta)^3 +9\beta^4(1-\beta)^2+\beta^6]+(1-P_m)[3\beta(1-\beta)^5+3\beta^2(1-\beta)^4 +9\beta^3(1-\beta)^3+3\beta^4(1-\beta)^2+3\beta^5(1-\beta)]\,,$$

or

$$P_{m+6}-(1-3\gamma)^3P_m = 3\gamma(1-3\gamma+3\gamma^2)\,, \qquad (26a)$$

where

$$\gamma = \beta(1-\beta)\,. \qquad (26b)$$

Thus,

$$P_m = \tfrac{1}{3}+\sum_{n=1}^{6} C_n x_n^m\,, \qquad (27)$$

where x_n are the roots of

$$x^6-(1-3\gamma)^3 = 0\,. \qquad (28)$$

We have then

$$\left.\begin{aligned} &P_0 = 1,\ P_1 = 0,\ P_2 = \tfrac{1}{3}(1+2\gamma),\ P_3 = \tfrac{1}{3}(1-\gamma), \\ &P_4 = \tfrac{1}{3}(1+2\gamma-6\gamma^2),\ P_5 = \gamma(2-3\gamma)\,, \\ &P_6 = 1-6\gamma+18\gamma^2-18\gamma^3,\ P_7 = 3\gamma(1-3\gamma+3\gamma^2), \\ &P_8 = \tfrac{1}{3}(1+2\gamma-18\gamma^2+54\gamma^3-54\gamma^4), \\ &P_9 = \tfrac{1}{3}(1-\gamma+9\gamma^2-27\gamma^3+27\gamma^4), \\ &P_{10} = \tfrac{1}{3}(1+2\gamma-24\gamma^2+108\gamma^3-216\gamma^4+162\gamma^5), \\ &P_{11} = \gamma(5-30\gamma+90\gamma^2-135\gamma^3+81\gamma^4), \\ &P_{12} = 1-12\gamma+90\gamma^2-360\gamma^3+810\gamma^4-972\gamma^5 \\ &\qquad +486\gamma^6\,. \end{aligned}\right\} \qquad (29)$$

7. Calculation of the diffraction intensity

It has been proved by Jagodzinski (1949a, b) that the diffracted intensity is proportional to

$$I = \frac{\sin^2\frac{1}{2}N_1A_1}{\sin^2\frac{1}{2}A_1}\cdot\frac{\sin^2\frac{1}{2}N_2A_2}{\sin^2\frac{1}{2}A_2}\left\{\frac{1+2Q}{3}\cdot\frac{\sin^2\frac{1}{2}N_3A_3}{\sin^2\frac{1}{2}A_3} +(1-Q)\sum_n N_3C_n\frac{1-x_n^2}{1-2x_n\cos A_3+x_n^2}\right\}, \qquad (30)$$

with $\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$: translation vectors,
N_1, N_2, N_3: number of translations,
$\mathbf{s}_0, \mathbf{s}$: unit vectors in the direction of incidence and diffraction,
H, K, L: hexagonal indices,

$$A_\nu = (2\pi/\lambda)\mathbf{a}_\nu \cdot (\mathbf{s}-\mathbf{s}_0)\,,$$
$$A_1 = 2\pi H,\ A_2 = 2\pi K,\ A_3 = 2\pi L/n\,, \qquad (31)$$

n: number of close-packed layers in the hexagonal cell,

$$Q = \begin{cases} -\frac{1}{2} & \text{for } H-K \neq 3m\,, \\ 1 & \text{for } H-K = 3m\,. \end{cases}$$

(*A*) $H-K = 3m$

There are sharp intensity peaks for $A_3 = 0$ (mod. 2π) and zero intensity elsewhere.

(*B*) $H-K \neq 3m$

(1) The diffuse intensity varies along the lines H, K = constant ($H-K \neq 3m$); L = variable, of the reciprocal lattice according to the sum in (30). Furthermore, we have then

$$\int_0^{2\pi} I dA_3 = 2\pi N_3 \frac{\sin^2 \frac{1}{2}N_1A_1}{\sin^2 \frac{1}{2}A_1} \cdot \frac{\sin^2 \frac{1}{2}N_2A_2}{\sin^2 \frac{1}{2}A_2} \qquad (32)$$

since we have

$$\sum_n C_n = P^4 - \tfrac{1}{3} = 1 - \tfrac{1}{3} = \tfrac{2}{3}$$

if we take (13) into account.

Thus,

$$\varepsilon = \frac{I}{\int_0^{2\pi} I dA_3} = \frac{1}{2\pi} \cdot \frac{3}{2} \sum_n C_n \frac{1-x_n^2}{1-2x_n \cos A_3 + x_n^2}\,. \qquad (33)$$

We can prove that

$$\varepsilon = \frac{1}{2\pi}\frac{3}{2}\frac{C+2\sum_{p=1}^{n-1} D_p \cos pA_3}{A+2\sum_{p=1}^{n} B_p \cos pA_3} \qquad (34)$$

(n = degree of the characteristic equation), where

$$\left.\begin{aligned} &A = \sum_{m=0}^{n} a_m^2,\ B_p = \sum_{m=0}^{n-p} a_m \cdot a_{m+p}\,, \\ &C = \sum_{m=0}^{2n} c_m P_m,\ D_p = \sum_{m=0}^{2n-p} d_m^{(p)} P_m\,, \\ &\sum_{m=0}^{2n} c_m x^m = (1-x^2)\sum_{s=0}^{n-1}\left(\sum_{r=0}^{s} a_r x^{s-r}\right)^2, \\ &\sum_{m=0}^{2n-p} d_m^{(p)} x^m = (1-x^2)\sum_{s=0}^{n-p-1}\left(\sum_{r=0}^{s} a_r x^{s-r}\right)\left(\sum_{r=0}^{s+p} a_r x^{s+p-r}\right), \end{aligned}\right\} \qquad (35)$$

when $a_m (m = 0, 1, \ldots, n)$ are the coefficients of the characteristic equation $\sum_{m=0}^{n} a_m x^{n-m} = 0$ and $P_m (m = 0, \ldots, 2n)$ can be calculated.

(2) In the case of hexagonal close-packed or cubic face centred crystals with both types of 'stacking faults' we obtain, by substituting the result of § 4 in equation (34),

$$\varepsilon = \frac{1}{2\pi} \times \{[3\alpha(1-\alpha)+3(2\alpha-1)^2\gamma(2-3\gamma)]+3(2\alpha-1)\gamma\cos A_3\}/ \{[(2-4\alpha+5\alpha^2)-3(2\alpha-1)^2\gamma(2-3\gamma)] +2\alpha[2\alpha-3(2\alpha-1)\gamma]\cos A_3 +2(2\alpha-1)(1-3\gamma)\cos 2A_3\}\,. \qquad [36]$$

The maxima and minima for ε are given by

$$d\varepsilon/dA_3 = 0\,. \qquad (37)$$

With the aid of (23) and (24), (28) and (29), we obtain by substitution in equation (34):

(3) For an *hk*-crystal:

$$\varepsilon = \frac{1}{2\pi} \times \{3\gamma(4-18\gamma+36\gamma^2-27\gamma^3)-9\gamma(1-3\gamma+3\gamma^2)\cos A_3 +\tfrac{3}{2}\gamma(2-3\gamma)\cos 2A_3 - 3\gamma(1-3\gamma)\cos 3A_3\}/ \{1+(1-3\gamma)^4-2(1-3\gamma)^2\cos 4A_3\}\,. \qquad (38)$$

(4) For an *hkk*-crystal:

$$\varepsilon = \frac{1}{2\pi} \times \{9\gamma(2-15\gamma+60\gamma^2-135\gamma^3+162\gamma^4-81\gamma^5)-3\gamma(5-30\gamma +90\gamma^2-135\gamma^3+81\gamma^4)\cos A_3+6\gamma^2(4-18\gamma +36\gamma^2-27\gamma^3)\cos 2A_3-9\gamma^2(1-3\gamma+3\gamma^2)\cos 3A_3 +6\gamma^2(2-9\gamma+9\gamma^2)\cos 4A_3-3\gamma(1-3\gamma)^2\cos 5A_3\}/ \{1+(1-3\gamma)^6-2(1-3\gamma)^3\cos 6A_3\}\,. \qquad (39)$$

8. Calculation of the probability parameters for the various problems of the preceding sections

(1) *'Growth faults' with* $n = 4$

The formula (34) expresses ε as a function of A_3, which can be measured. With the aid of ε for 16 different values of A_3, a homogeneous system of 16 equations can be constructed. This will enable us to calculate the 17 unknown's A, B_p, C, D_p apart from an indefinite factor.

If A', B_p', C', D_p' is a solution of the system we have:

$$A+2\sum_{p=1}^{8} B_p \cos pA_3 \sim A'+2\sum_{p=1}^{8} B_p' \cos pA_3 \sim \prod_{p=1}^{8}(\cos A_3 - y_p)\,.$$

Since

$$A+2\sum_{p=1}^{8} B_p \cos pA_3 \equiv \prod_{p=1}^{8}(1-2x_p\cos A_3+x_p^2)\,,$$

the roots of the equation (12) are given by the systems

$$\begin{cases} (1+x_p^2)/2x_p = y_p \\ |x_p| \leqslant 1 \end{cases} \quad (p = 1, \ldots, 8)\,. \qquad (40)$$

The coefficients, a_1, a_2, a_3, a_4, of (12) enable us then to calculate the four transition probabilities α_i.

(2) *'Growth and deformation or transformation stacking faults' in hexagonal close-packed and cubic face centred crystals*

(*a*) *Hexagonal close-packing.*—Broadened maxima occur for $A_3 = 0$ and $A_3 = \pi$ (mod. 2π), and

$$\left.\begin{aligned}\varepsilon_0 &= \frac{1}{2\pi}\cdot\frac{(1-\alpha)-3(1-2\alpha)\gamma}{3[\alpha+(1-2\alpha)\gamma]},\\ \varepsilon_\pi &= \frac{1}{2\pi}\cdot\frac{3[(1-\alpha)-(1-2\alpha)\gamma]}{\alpha+3(1-2\alpha)\gamma}.\end{aligned}\right\} \quad (41)$$

Thus α and β can be calculated with the aid of (41).

(*b*) *Cubic close-packing.*—Minima occur for $A_3 = 0$ and $A_3 = \pi$ (mod. 2π); (41) is still valid and can eventually be used to calculate α and β. Broadened maxima occur for

$$A_3 = \pm\theta \text{ (mod. } 2\pi), \quad (42)$$

where θ is the root of (37), which will move towards $A_3 = \pm 2\pi/3$ (mod. 2π) as $\alpha \to 1$ and $\beta \to 0$ or 1.

With the aid of (42) and $\varepsilon_{\pm\theta}$, α and β can again be calculated. When $\alpha = 1$ (face centred cubic without 'growth faults'), we obtain the same equation (18), but the further calculations are different (Paterson, 1952).

(3)

For an *hk*-crystal we obtain from (38):

$$\varepsilon_0 = \frac{1}{2\pi}\cdot\frac{\frac{1}{2}-3\gamma(1-3\gamma)}{3\gamma(2-3\gamma)},\quad \varepsilon_{\pm\pi/2} = \frac{1}{2\pi}\cdot\frac{\frac{1}{2}-\gamma(2-3\gamma)}{\gamma(2-3\gamma)},$$
$$\varepsilon_\pi = \frac{1}{2\pi}\cdot\frac{3[\frac{1}{2}-\gamma(1-\gamma)]}{\gamma(2-3\gamma)}. \quad (43)$$

(4)

For an *hkk*-crystal we obtain from (39):

$$\varepsilon_0 = \frac{1}{2\pi}\cdot\frac{\gamma(4-9\gamma)}{3(1-3\gamma+3\gamma^2)},$$
$$\varepsilon_{\pm 2\pi/6} = \frac{1}{2\pi}\cdot\frac{2-14\gamma+51\gamma^2-54\gamma^3}{18\gamma(1-3\gamma+3\gamma^2)},$$
$$\varepsilon_\pi = \frac{1}{2\pi}\cdot\frac{4-10\gamma+30\gamma^2-27\gamma^3}{9\gamma(1-3\gamma+3\gamma^2)},$$
$$\varepsilon_{\pm 2\pi/3} = \frac{1}{2\pi}\cdot\frac{2-10\gamma+23\gamma^2+18\gamma^3}{6\gamma(1-3\gamma+3\gamma^2)}. \quad (44)$$

With one of the formulae (43) and one of the formulae (44) we can calculate β in either case.

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A Direct Approach to the Determination of Crystal Structures

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A direct method of solution of the X-ray crystallographic problem is described, which consists in tabulating the complete function connecting the structure factor with the atomic positions. The successive steps of the solution can be interpreted as intersections of $(N-1)$-dimensional surfaces in N-dimensional space, where N is the number of unknown co-ordinates. A card index, graphs and tables have been prepared and published for a one-dimensional unit cell with no centre of symmetry containing up to four equal point atoms and, with centre of symmetry, up to ten equal point atoms per cell. Centrosymmetric structures with up to twenty atoms per cell can be solved by a single convolution of the above tables.

Introduction

The determination of crystal structures and the corresponding phase problem of X-ray crystallography has been attacked in the past by a variety of methods. If the direct methods applicable to special cases, such as the heavy-atom and the isomorphous-replacement methods, are not considered, the usual methods are those of trial and error, Patterson synthesis and its variants, Harker–Kasper inequalities, solution of polynomials, and statistical methods.

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