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On the Diffraction of X-rays by Face-Centred Cubic Crystals containing Extrinsic Stacking Faults

By C. J. Howard*

Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, England

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A solution is given to that problem of extrinsic faulting in face-centred cubic crystals in which additional layers may be inserted with equal probability after layers of original crystal or after previously inserted layers. The method employed involves the derivation and solution of an appropriate difference equation. The distribution of diffracted intensity differs from that given previously [Sabine, *Acta Cryst.* (1966), **21**, 882–884], but it does show expected behaviour both for small values of the fault probability and for values near unity.

1. Introduction

Two distinct models for faulting which have been considered in the literature might be classified under the heading of extrinsic faults in face-centred cubic crystals, and corresponding to these two models for faulting there are two problems about the diffraction of X-rays. In the first model it is supposed that a layer may be inserted after any layer of the original crystal, but that an inserted layer cannot follow another inserted layer. The probability that any original layer is followed by an inserted layer is assumed to be constant throughout the crystal, and is denoted here by p. The corresponding diffraction problem has been solved by Johnson (1963), who used random-walk techniques to evaluate first the phase changes and then the diffracted intensity. Holloway & Klamkin (1969) (hereafter HK) have given another solution after using probability trees to set up a difference equation in the probabilities. Though the methods used in the two solutions are quite different, the expressions given for the diffracted intensity are identical. For $p \rightarrow 0$ the crystal tends to the original f.c.c. crystal, while for $p \rightarrow 1$ the crystal becomes the f.c.c. crystal which is the twin of the original (Johnson, 1963) – the diffracted intensity shows the expected peaks at these two limits. Evidently, these solutions to the first problem are quite satisfactory.

The situation has been less satisfactory for the second problem. In this it is supposed that a layer may be inserted after any layer of the original crystal, or after another inserted layer. The probability that any layer is followed by an inserted layer is denoted by p. For p small it is expected that the distribution of diffracted intensity should match that calculated in the first problem, for the probability of having two or more inserted layers after a given original layer is p^2 which is much less than the probability $p(1-p)$ of having a single inserted layer. For $p \rightarrow 1$ the structure becomes hexagonal close-packed (Fig. 1) so the diffracted intensity should show characteristic h.c.p. features in this limit. This second problem (which has been termed the problem of condensation faults) has been considered by Sabine (1966), but the result he gives fails to show the expected behaviour at small values of the fault probability. Sabine's work has been criticised by Johnson (1968) and again by HK. The latter gave an indication of how the problem might be approached, but until now no satisfactory solution has been published.

The purpose of this paper is to present a detailed solution to the second of the problems mentioned above. First, a difference equation in the probabilities is set up *via* the methods of HK, and a direct confirmation is made of its validity. It is shown that the difference equation for the first problem generates probabilities which are correct for the second problem to first order in p, while Sabine's difference equation generates probabilities which are in error by terms of first order in p. The solution of our difference equation is straightforward, if tedious. From this solution the expression for diffracted intensity is written down, and a brief description of its behaviour is given. This expression is shown to give the expected behaviour both at small values of p, and for $p \rightarrow 1$.

2. The difference equation

The difference equation is derived here following the methods of HK, although our notation differs slightly from theirs. We suppose that the original (or regular) stacking sequence is $ABCABC\ldots$, and that the probability that any layer is followed by an inserted layer (not in the regular stacking sequence) is p. At any layer which is described by the stacking symbol A we may distinguish layers A^+ and A^*: A^+ denotes a layer which, if the regular stacking sequence is followed or resumed, is followed by B, while A^* denotes a layer which, if regular stacking sequence is resumed, is followed by C. At layers described by the stacking symbols B or C analogous distinctions can be made.

* Science Research Council Visiting Fellow. On leave from Australian Atomic Energy Commission Research Establishment, Lucas Heights, New South Wales, Australia.

Fig. 1 shows probability trees drawn from A^+ and A^* – analogous trees can be drawn from B^+, B^*, C^+ and C^*.

The expected number of inserted layers after any layer of the original crystal is

$$p(1-p)+2p^2(1-p)+3p^3(1-p)+4p^4(1-p)+\ldots = p/(1-p)$$

so the fraction of condensed layers in the crystal is

$$\frac{p/(1-p)}{[1+p/(1-p)]}=p\,.$$

A layer will be of type A^+, B^+ or C^+ if it is a layer of the original crystal or if it is 2nd, 4th, 6th... in a sequence of inserted layers. It is not difficult to establish that the fraction of layers of these types is

$$(1+p^2+p^4+\ldots)/(1+p+p^2+p^3+\ldots)=1/(1+p)\,.$$

A layer will be of type A^*, B^*, C^* if it is 1st, 3rd, 5th, ... in a sequence of inserted layers. The fraction of layers of these types is $p/(1+p)$.

Following HK once again, we suppose that the probabilities of occurrence of $A^+, B^+, C^+, A^*, B,^* C^*$ in the mth layer are respectively $G_1, G_2, G_3, G_4, G_5, G_6$. Then we have

$$G_1+G_2+G_3+G_4+G_5+G_6=1 \tag{1}$$

$$P_m^A=G_1+G_4 \tag{2}$$

$$P_{m+1}^A=pG_2+(1-p)G_3+(1-p)G_5+pG_6 \tag{3}$$

$$P_{m+2}^A=pG_1+(1-p)^2G_2+p(1-p)G_3 +(1-2p+2p^2)G_4+p(1-p)G_5+p(1-p)G_6 \tag{4}$$

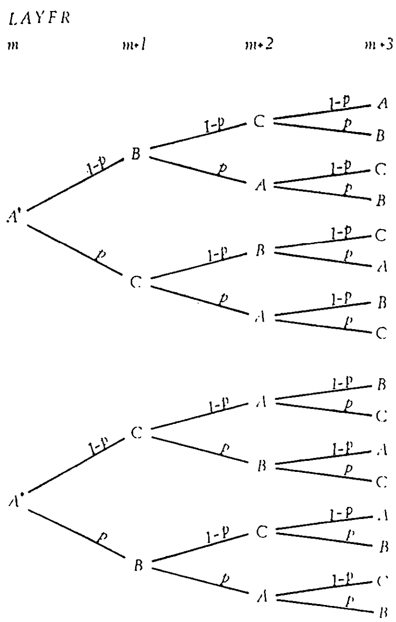

Fig. 1. Probability trees used in the derivation of the difference equation.

$$P_{m+3}^A=(1-3p+4p^2-2p^3)G_1+p(2-4p+3p^2)G_2 +p(1-p^2)G_3+2p(1-2p+p^2)G_4+p(1-p^2)G_5 +(1-3p+4p^2-p^3)G_6\,. \tag{5}$$

Equations (2) to (5) are written from inspection of the probability trees. In these equations the symbol P_m^A denotes the probability that the mth layer is in stacking position A (whether A^+ or A^*). Equations (1) to (5) involve the unknowns in just four combinations: (G_1+G_6), (G_2+G_4), (G_3+G_5) (G_4-G_6). Equations (1) to (4) can be solved for these four quantities and the results substituted in (5) to give

$$P_{m+3}^A+(1-2p)P_{m+2}^A+(1-2p)P_{m+1}^A +p(1-3p+3p^2)P_m^A=1-p-p^2+p^3 \tag{6}$$

which is the required difference equation. The probabilities P_m^B and P_m^C satisfy precisely analogous difference equations – it is necessary only to replace A by B or C in (6).

The validity of (6) can be examined in a straightforward manner. For instance it can be supposed that we have A^+ on the 0th layer. Then a probability tree can be constructed as in the upper part of Fig. 1 (but extended beyond the 3rd layer), and the probabilities $P_0^A, P_1^A, \ldots, P_m^A$, can be written from inspection. Given P_0^A, P_1^A and P_2^A, successive applications of (6) should generate the probabilities correctly. In Table 1 are shown some probabilities obtained by inspection of the probability tree, together with probabilities generated from (6). It is of interest to compare these probabilities with those generated from HK's difference equation for the first problem:

$$P_{m+4}^A+(1-p)P_{m+3}^A+(1-p+p^2)P_{m+2}^A +2p(1-p)P_{m+1}^A+p^2P_m^A=1 \tag{7}$$

and from Sabine's difference equation

$$P_{m+2}^A+(1-p)P_{m+1}^A+(1-2p)P_m^A=1-p\,. \tag{8}$$

The results indicate that (6) is the correct equation for the problem under consideration, that (7) generates probabilities which are correct to first order in p, while (8) generates probabilities which are in error by terms of first order in p.

The solutions of (6) and its analogues for P_m^B and P_m^C are (see HK)

$$P_m^A=\tfrac{1}{3}+K_1X_1^m+K_2X_2^m+K_3X_3^m \tag{9}$$

$$P_m^B=\tfrac{1}{3}+L_1X_1^m+L_2X_2^m+L_3X_3^m \tag{10}$$

$$P_m^C=\tfrac{1}{3}+M_1X_1^m+M_2X_2^m+M_3X_3^m \tag{11}$$

where the X's are the roots of

$$X^3+(1-2p)X^2+(1-2p)X+p(1-3p+3p^2)=0\,. \tag{12}$$

These roots are shown in Table 2. Following HK we choose the 0th layer at random, and arbitrarily assign to it the stacking symbol A. It may be A^+ [with probability $1/(1+p)$] or A^* [with probability $p/(1+p)$]. Then the probabilities for $m=0,1,2$, which are the boundary conditions, can be written from inspection of the trees in Fig. 1, after assigning appropriate weights to the two trees. This allows determination of the constants K, L, M in (9), (10), (11) and the solution of the difference equation is complete. The assumed probabilities and the constants are included in Table 2.

3. The intensity distribution

To discuss the intensity distribution we use the same hexagonal coordinates $[hk.l]$ as HK, so that we can take over their results immediately. For $(h-k)\equiv 0$ (mod 3) there are sharp intensity maxima when l is an even integer. For $(h-k)\not\equiv 0$(mod 3) we obtain spots which show streaking in a direction normal to the close-packed planes. Since for $0<p<1$

$$|X_v|<1 \quad v=1,2,3$$

which is condition (21) in HK, we can use the HK result (23) for the intensity, *viz*

$$I=\sum_{v=1}^{3}\frac{3K_v(1-X_v^2)/2\pm\sqrt{3}(L_v-M_v)X_v\sin\pi l}{1-2X_v\cos\pi l+X_v^2} \qquad (13)$$

where the upper and lower signs correspond to the cases $(h-k)\equiv 1$(mod 3) and $(h-k)\equiv 2$(mod 3) respectively. Using first the 'remarks' from Table 2 to eliminate (L_2-M_2), (L_3-M_3) from (13), then substituting for the X's, K's, etc., we obtain

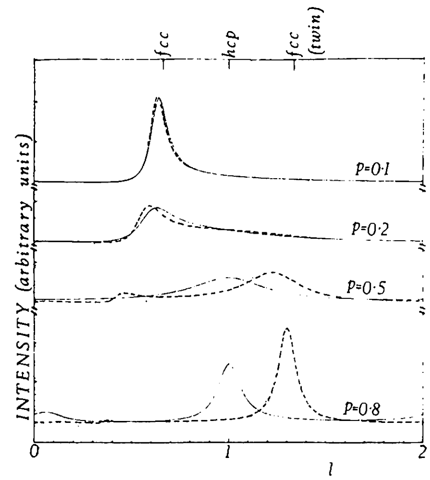

Fig. 2. Intensity profiles resulting from solutions of the first problem (broken line) and of the second problem (full line). The profiles are drawn for various values of the fault probability.

Table 1. *Some probabilities P_m^A, given that the 0th layer is A^+*

m	P_m^A by inspection	P_m^A generated by equation (6)	P_m^A generated by equation (7)	P_m^A generated by equation (8)
0	1	—	—	—
1	0	—	—	—
2	p	—	—	—
3	$(1-p)(1-2p+2p^2)$	$(1-p)(1-2p+2p^2)$	—	p
4	$p(3-9p+11p^2-4p^3)$	$p(3-9p+11p^2-4p^3)$	$p(3-7p+5p^2-2p^3)$	$(1-p)^2$
5	$p(1-p)(1+4p-11p^2+8p^3)$	$p(1-p)(1+4p-11p^2+8p^3)$	$p(1-p^2+p^3)$	$p(1-p+p^2)$

Table 2. *Solutions of the difference equations* (6)

See equations (9) to (12) in text.

Roots of (12)

$X_1=-p$ $\quad X_2=[(3p-1)+i\sqrt{3}(1-p)]/2$ $\quad X_3=[(3p-1)-i\sqrt{3}(1-p)]/2$

Boundary conditions, given that 0th layer is A

$P_0^A=1$	$P_1^A=0$	$P_2^A=2p(1-p+p^2)/(1+p)$
$P_0^B=0$	$P_1^B=(1-p+p^2)/(1+p)$	$P_2^B=p(1-p)$
$P_0^C=0$	$P_1^C=p(2-p)/(1+p)$	$P_2^C=(1-2p+2p^2-p^3)/(1+p)$

Constants in equations (9) to (11)

$K_1=p(1-p+4p^2)/D$

$K_2=(2-9p+9p^2+2p^3)/6D+i\sqrt{3}p(1-p)(1-2p)/2D$

K_3 is complex conjugate of K_2

$L_1=-p(2-5p+5p^2)/D$

$L_2=-(1-9p+18p^2-8p^3)/6D-i\sqrt{3}(1-3p+4p^3)/6D$

L_3 is complex conjugate of L_2

$M_1=p(1-4p+p^2)/D$

$M_2=-(1-9p^2+10p^3)/6D+i\sqrt{3}(1-6p+9p^2-2p^3)/6D$

M_3 is complex conjugate of M_2

with $D=(1+p)(1-4p+7p^2)$

Remarks

$K_1+L_1+M_1=0$

$K_2+L_2+M_2=0$

$K_3+L_3+M_3=0$

$3K_2-i\sqrt{3}(L_2-M_2)=0$

$3K_3+i\sqrt{3}(L_3-M_3)=0$

$$
\begin{aligned}
I= & \frac{3K_1(1-X_1^2)/2\pm\sqrt{3}(L_1-M_1)X_1\sin\pi l}{1-2X_1\cos\pi l+X_1^2} \\
& +\tfrac{3}{2}K_2\frac{[\exp(\pm i\pi l)+X_2]}{[\exp(\pm i\pi l)-X_2]} \\
& +\tfrac{3}{2}K_3\frac{[\exp(\mp i\pi l)+X_3]}{[\exp(\mp i\pi l)-X_3]} \\
= & \frac{3p(1-p)}{2(1+p)(1-4p+7p^2)} \\
& \times\left[\frac{(1+p)(1-p+4p^2)\pm 2p(1-2p)\sqrt{3}\sin\pi l}{1+2p\cos\pi l+p^2}\right. \\
& +\left.\frac{(2-9p+9p^2+2p^3)-3(1-p)(1-2p)\cos\pi l\mp(1-2p)(1-3p)\sqrt{3}\sin\pi l}{(2-3p+3p^2)+(1-3p)\cos\pi l\mp(1-p)\sqrt{3}\sin\pi l}\right]. \qquad (14)
\end{aligned}
$$

Fig. 2 shows intensity profiles for $h-k\equiv 1(\text{mod } 3)$ and for various values of the fault probability p. The figure also shows profiles for the case $h-k\equiv 1(\text{mod } 3)$ given by the solution to the first problem

$$
\begin{aligned}
I= & \frac{3p(1-p)}{(1+p)}\{(2-\cos\pi l\mp\sqrt{3}\sin\pi l)/[2(1-p+p^2) \\
& +(1+p-2p^2)\cos\pi l\mp(1-p)\sqrt{3}\sin\pi l \\
& +p\cos 2\pi l\mp p\sqrt{3}\sin 2\pi l]\}\,. \qquad (15)
\end{aligned}
$$

As expected, the profiles given by (14) and (15) are almost indistinguishable for p small (say $p\lesssim 0{\cdot}2$). On the other hand for p nearly unity (say $p\gtrsim 0{\cdot}8$), the intensity as given by (14) peaks at the h.c.p. position as expected.

4. Discussion

Sabine (1966) presented a solution to the (second) problem in which the diffraction profiles showed unusual features, but that solution has been shown to be in error. From the solution presented here it is evident that the two models for faulting result in diffraction profiles which are practically indistinguishable for small or moderate values of the fault probability.

A more general model for faulting can be conceived in which a layer of original crystal is followed by an inserted layer with probability p, while an inserted layer is followed by another inserted layer with probability q (not necessarily equal to p). This model reduces to the 'first model' above for $q=0$ and to the 'second model' for $q=p$. This model might also provide a more realistic representation of the faulting which occurs, for instance, in a crystal which has been subjected to heavy neutron damage. It is clear that the HK method would have the power to deal with this problem. At any layer described by stacking symbol A we would have to distinguish three types: A of the original crystal, inserted A followed on resumption of regular sequence by B, and inserted A followed on resumption of regular sequence by C. Unfortunately there would be nine unknowns G_1 to G_9 and although the HK procedure is clear the difference equation which results could have order as high as eight. At present, and particularly in view of the similarity of the results for $q=0$ and $q=p$, it does not seem worthwhile to carry out the longer calculation for the more general model.

It would be of some interest to derive the intensity distribution (14) from some general theory which avoids the use of difference equations. Kakinoki (1967) has given one general theory, but that theory does not seem to be applicable because the *Reichweite* for the (second) problem is not well defined. It is not clear to the author whether the intensity distribution can be obtained by a straightforward application of Cowley's (1976) theory.

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