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Simultaneous Reflections and the Mosaic Spread in a Crystal Plate

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The intensity of an X-ray (Bragg) reflection from a mosaic crystal plate under the conditions of multiple diffractions is discussed theoretically. The set of simultaneous differential equations is solved exactly for the two-beam and the three-beam cases. A second order approximation is given for the multiple beam case, and a third order approximation for the triple beam case. The fact that the ratio R of the peak intensity of a single diffracted beam to the intensity of the same beam under conditions of multiple diffraction depends on the mosaic spread η of the crystal provides a method to obtain this magnitude from the experiment. Preliminary measurements were performed on Si and Ge single-crystal plates under conditions of multiple diffraction. From them experimental values of R were obtained for different planes, which then were used to calculate the mosaic spread of the crystals. In this method only relative intensity measurements have to be used and most of the corrections found to be necessary in other procedures used to determine mosaic spread are not needed. The polarization factor for the case of a double reflection preceded by a monochromator is analyzed and the results are given in an Appendix.

Introduction

The experiments with which the present paper is concerned are similar to those performed by Renninger (1937) to show the effects of multiple diffraction in the 'forbidden' reflection 222 from diamond. The crystal is oriented so that a given reciprocal lattice point (RELP in what follows) is under reflecting conditions and in such a way that the crystal can be rotated around the diffraction vector **H** (Fig. 1). During the rotation the RELP H remains on the Ewald sphere, thus continuously diffracting in the direction CH. The detector D is positioned so as to measure the intensity of this diffracted beam which is then recorded as a function of the angle of rotation ω around the vector **H**. The intensity remains constant until one or more other RELP's contact the Ewald sphere, and then, depending on the particular pair or set of reflections involved, an increase or a decrease in intensity is observed.

We shall assume in what follows that:

1. The crystal is of mosaic type with an angular distribution of crystallites $W(\Delta)$ which is approximately Gaussian and isotropic:

$$W(\Delta)=\frac{1}{\eta\sqrt{2\pi}}\exp\left(-\Delta^2/2\eta^2\right).$$

2. The incident beam is nearly monochromatic and well collimated so that its angular width is much smaller than that associated to the mosaic distribution (η).

3. η is much bigger than the half-width of the diffraction pattern due to a (perfect) single block.

Assumption 1 is normally used in connection with mosaic crystals in the absence of a more realistic model appropriate to the sample tested. Eventually, however, one obtains information showing that $W(\Delta)$ varies with direction or with some other parameter. Assumption 2 is fulfilled through careful experimental technique and it implies that even that the beams are slightly divergent we shall make use of the approximation that plane waves are travelling in the crystal, their intensities being functions of position. Fulfilment of assumption 3 depends entirely on the sample under study; it does not apply to a nearly perfect or perfect crystal, *i.e.* it amounts to assuming that in the crystal used primary extinction is negligible.

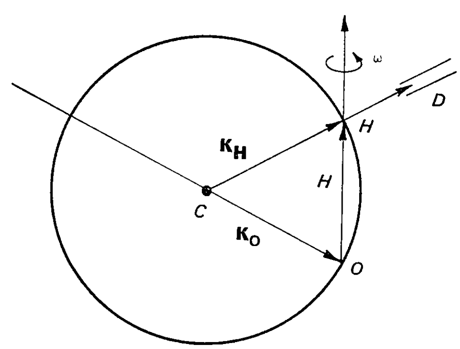

Fig. 1. The crystal is oriented so that the RELP H is under reflecting conditions. It is turned around the diffraction vector **H** and the intensity measured by the detector D is recorded as a function of the angle of rotation ω.

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Intensity solutions for multiple reflections in a crystal plate

Let us designate with subscripts $i, j, k, \ldots$, the different beams as well as the RELP's that originate them. The subscript o is reserved for the incident beam.

The total change in power of beam i owing to absorption and to the simultaneous reflections by n other RELP's as it traverses a layer of thickness dx at depth x below the surface is given by (Zachariasen, 1965)

$$\frac{\mathrm{d}P_i}{\mathrm{d}x} = -\frac{P_i}{\gamma_i}\mu + \sum_{j=o}^{n} \bar{Q}_{ij}\left(\frac{P_j}{\gamma_j} - \frac{P_i}{\gamma_i}\right), \qquad (1)$$

where γ_i and γ_j are the direction cosines of beams i and j relative to the normal to the plate surface. $\bar{Q}_{ij}$ is the effective reflectivity of plane $(i-j)$ and P_i the power of beam i.

In equations (2), (3) and (4), which apply respectively to the incident beam, to the primary diffracted beam (subscript 1), *i.e.* that corresponding to the RELP which is maintained on the Ewald sphere during the rotation, and to an arbitrary reflection i, the contributions of the incident and primary diffracted beams have been explicitly introduced.

$$\frac{\mathrm{d}P_o}{\mathrm{d}x} = -\frac{P_o}{\gamma_o}(\mu + \bar{Q}_{o1} + \sum_j \bar{Q}_{oj}) + \frac{P_1}{\gamma_1}\bar{Q}_{1o} + \sum_j \frac{P_j}{\gamma_j}\bar{Q}_{jo}\,, \qquad (2)$$

$$\pm\frac{\mathrm{d}P_1}{\mathrm{d}x} = \frac{P_o}{\gamma_o}\bar{Q}_{o1} - \frac{P_1}{\gamma_1}(\mu + \bar{Q}_{1o} + \sum_j \bar{Q}_{1j}) + \sum_j \frac{P_j}{\gamma_j}\bar{Q}_{j1}\,, \qquad (3)$$

$$\pm\frac{\mathrm{d}P_i}{\mathrm{d}x} = \frac{P_o}{\gamma_o}\bar{Q}_{oi} + \frac{P_1}{\gamma_1}\bar{Q}_{1i} - \frac{P_i}{\gamma_i}(\mu + \bar{Q}_{io} + \bar{Q}_{i1} + \sum_{\substack{j=2\\ j\neq i}}\bar{Q}_{ij}) + \sum_{\substack{j=2\\ j\neq i}} \frac{P_j}{\gamma_j}\bar{Q}_{ji}\,. \qquad (4)$$

Equations (2), (3) and (4) were first given by Moon & Shull (1964). The plus sign on the left side applies to the transmitted beams and the minus sign to reflected beams. The interaction coefficients $\bar{Q}_{ij}$, which will be discussed later, are related to the reflectivity per unit volume of a small crystallite Q_{ij} and to the mosaic distribution function $W(\Delta)$:

$$\bar{Q}_{ij} = Q_{ij}\,.\,W(\Delta)\,. \qquad (5)$$

We are interested in the intensity changes in beam 1 when other RELP's are brought into contact with the Ewald sphere. Moon & Shull (1964) have given approximate intensity solutions for the case where all the beams are transmitted through the crystal. They used a Taylor's series expansion of $P_1(x)$ about the point $x=0$.

The simultaneous differential equations will next be solved for the double-beam and the triple beam cases.

Double-beam case

$$\dot{P}_o = -A_oP_o + BP_1\,,$$
$$-\dot{P}_1 = CP_o - A_1P_1\,,$$

where the dots indicate derivation with respect to x. When the beam 1 is reflected:

$$A_o = \frac{1}{\gamma_o}(\mu + \bar{Q}_{o1})\,, \qquad B = \bar{Q}_{1o}/\gamma_1\,,$$
$$A_1 = \frac{1}{\gamma_1}(\mu + \bar{Q}_{o1})\,, \qquad C = \bar{Q}_{1o}/\gamma_o\,.$$

If the beam 1 is transmitted one has to change the signs of A_1 and C.

By differentiating the differential equations we obtain after a few manipulations:

$$\ddot{P} + (A_o - A_1)\dot{P} + (BC - A_oA_1)P = 0\,,$$

where P stands for either P_o or P_1. Then

$$P_1 = F_1 \exp k_1x + F_2 \exp k_2x\,,$$
$$P_o = F_3 \exp k_1x + F_4 \exp k_2x\,,$$

where k_1 and k_2 are the roots of the characteristic equation.

For a reflected beam the boundary conditions are:

$$P_o|_o = P_o(0)$$
$$A_1\dot{P}_o(0) - B\dot{P}_1(0) = (BC - A_oA_1)P_o(0)$$
$$P_1(T) = 0$$
$$C\dot{P}_o(T) - A_o\dot{P}_1(T) = 0\,,$$

where T stands for the thickness of the crystal plate. Accordingly, the constants of integration are

$$F_1 = -b/a\,.\,F_2$$

$$F_2 = P_o(0)\,\frac{aC(D - A_1k_1)(bk_2 - ak_1) + a^2k_1CA_1(k_2 - k_1)}{BC(bk_1 - ak_2)(bk_2 - ak_1) + abA_oA_1(k_2 - k_1)^2}$$

$$F_3 = P_o(0) - F_4$$

$$F_4 = \frac{-ak_1CP_o(0) + bA_o(k_2 - k_1)F_2}{C(bk_2 - ak_1)}\,,$$

with $a = \exp(k_1T)$, $b = \exp(k_2T)$ and $D = BC - A_oA_1$. The intensity value we are interested in is $p_1 = P_1(0)/P_o(0)$:

$$p_1 = (a-b)\,\frac{C(D - A_1k_1)(bk_2 - ak_1) + ak_1CA_1(k_2 - k_1)}{BC(bk_1 - ak_2)(bk_2 - ak_1) + abA_oA_1(k_2 - k_1)^2}\,. \qquad (6)$$

For a symmetric reflection equation (6) reduces to:

$$p_1 = \bar{Q}\cdot\frac{\frac{a+b}{a-b}\sqrt{\mu^2+2\mu\bar{Q}}-(\mu+\bar{Q})}{\left(\frac{2}{a-b}\right)^2(\mu^2+2\mu\bar{Q})-\bar{Q}^2}\,. \tag{7}$$

If absorption is negligible, setting $l_o = T/\gamma_o$, we obtain

$$\lim_{\mu=0} p_1 = \frac{\bar{Q}l_o}{1+\bar{Q}l_o}\,, \tag{8}$$

a well known result.

Triple beam case

The differential equations are

$$\begin{aligned}\dot{P}_o &= a_oP_o+b_oP_1+c_oP_2\,,\\ \dot{P}_1 &= a_1P_o+b_1P_1+c_1P_2\,,\\ \dot{P}_2 &= a_2P_o+b_2P_1+c_2P_2\,.\end{aligned}$$

If beam 1 is reflected and beam 2 is transmitted the coefficients are

$$\left.\begin{aligned} a_o &= -\frac{\mu+\bar{Q}_{01}+\bar{Q}_{02}}{\gamma_o}\,,\quad b_o=\frac{\bar{Q}_{01}}{\gamma_1}\,,\quad c_o=\frac{\bar{Q}_{02}}{\gamma_2}\,,\\ a_1 &= -\frac{\bar{Q}_{01}}{\gamma_o}\,,\quad b_1=\frac{\mu+\bar{Q}_{10}+\bar{Q}_{12}}{\gamma_1}\,,\quad c_1=-\frac{\bar{Q}_{21}}{\gamma_2}\,,\\ a_2 &= \frac{\bar{Q}_{02}}{\gamma_o}\,,\quad b_2=\frac{\bar{Q}_{21}}{\gamma_1}\,,\quad c_2=-\frac{\mu+\bar{Q}_{20}+\bar{Q}_{21}}{\gamma_2}\,.\end{aligned}\right\} \tag{9}$$

If both beams are reflected one has to change the signs of a_2, b_2 and c_2, while if both beams are transmitted one has to change the signs of a_1, b_1 and c_1.

By differentiating twice the differential equations we arrive after some manipulations at:

$$\dddot{P}+E\ddot{P}+F\dot{P}+GP=0\,, \tag{10}$$

where P stands for either P_o, P_1 or P_2, and

$$\left.\begin{aligned} E &= -(a_o+b_1+c_2)\,,\\ F &= a_ob_1-a_1b_o+b_1c_2-b_2c_1+a_oc_2-a_2c_o\,,\\ G &= a_o(b_2c_1-b_1c_2)+a_1(b_oc_2-b_2c_o)+a_2\\ &\quad\times(b_1c_o-b_oc_1)\,.\end{aligned}\right\} \tag{11}$$

Then

$$\left.\begin{aligned} P_o &= F_1\exp k_1x+F_2\exp k_2x+F_3\exp k_3x\\ P_1 &= F_4\exp k_1x+F_5\exp k_2x+F_6\exp k_3x\\ P_2 &= F_7\exp k_1x+F_8\exp k_2x+F_9\exp k_3x\,.\end{aligned}\right\} \tag{12}$$

The 9 constants of integration can be found by using the appropriate boundary conditions. In the case when beam 1 is reflected and beam 2 is transmitted, the following set applies:

$$\begin{aligned} &P_o|_o = P_o(0)\\ &P_2|_o = 0\\ &b_2\dot{P}_1(0)-b_1\dot{P}_2(0)=(b_2a_1-b_1a_2)P_o(0)\\ &b_2\dot{P}_o(0)-b_o\dot{P}_2(0)=(b_2a_o-b_oa_2)P_o(0)\\ &P_1(T)=0\\ &\frac{c_1\dot{P}_o(T)-c_o\dot{P}_1(T)}{a_oc_1-a_1c_o}=\frac{c_2\dot{P}_1(T)-c_1\dot{P}_2(T)}{a_1c_2-a_2c_1}\\ &\ddot{P}_o(0)=a_o\dot{P}_o(0)+b_o\dot{P}_1(0)+c_o\dot{P}_2(0)\\ &\ddot{P}_1(0)=a_1\dot{P}_o(0)+b_1\dot{P}_1(0)+c_1\dot{P}_2(0)\\ &\ddot{P}_2(0)=a_2\dot{P}_o(0)+b_2\dot{P}_1(0)+c_2\dot{P}_2(0)\,,\end{aligned}$$

and one arrives at the following set of 9 simultaneous equations (13):

	F_1	F_2	F_3	F_4	F_5
1	1	1	1	–	–
2	–	–	–	–	–
3	–	–	–	b_2k_1	b_2k_2
4	b_2k_1	b_2k_2	b_2k_3	–	–
5	–	–	–	a	b
6	ak_1	bk_2	ck_3	ak_1M	bk_2M
7	$k_1(a_0-k_1)$	$k_2(a_0-k_2)$	$k_3(a_0-k_3)$	b_0k_1	b_0k_2
8	a_1k_1	a_1k_2	a_1k_3	$k_1(b_1-k_1)$	$k_2(b_1-k_2)$
9	a_2k_1	a_2k_2	a_2k_3	b_2k_1	b_2k_2

(13)

	F_6	F_7	F_8	F_9	2nd member
1	–	–	–	–	$P_o(0)$
2	–	1	1	1	0
3	b_2k_3	$-b_1k_1$	$-b_1k_2$	$-b_1k_3$	$(b_2a_1-b_1a_2)\,.\,P_o(0)$
4	–	$-b_0k_1$	$-b_0k_2$	$-b_0k_3$	$(b_2a_0-b_0a_2)\,.\,P_0(0)$
5	c	–	–	–	0
6	ck_3M	ak_1N	bk_2N	ck_3N	0
7	b_0k_3	c_0k_1	c_0k_2	c_0k_3	0
8	$k_3(b_1-k_3)$	c_1k_1	c_1k_2	c_1k_3	0
9	b_2k_3	$k_1(c_2-k_1)$	$k_2(c_2-k_2)$	$k_3(c_2-k_3)$	0

where

$$\left.\begin{aligned} a &= \exp k_1T,\ b = \exp k_2T,\ c = \exp k_3T, \\ M &= c_2/c_1 \,.\, N - c_o/c_1 \,, \\ N &= \frac{a_1c_o - a_oc_1}{a_1c_2 - a_2c_1} \,. \end{aligned}\right\} \quad (14)$$

The computations involved can be easily programmed and the exact solution for the three-beam case obtained to any approximation. In the general case of n beams the treatment would be similar and one would end up with a set of n^2 linear simultaneous equations in the n^2 constants of integration.

We have sought approximate solutions for the multiple beam case.

Multiple beam case, second order approximation

When P_1 is a reflected beam, the boundary conditions are:

At $x=0$:

$$P_o = P_o(0),$$
$$P_1 = P_1(0) \neq 0$$
$$P_i = P_i(0) \begin{cases} =0 \text{ for transmitted beams} \\ \neq 0 \text{ for reflected beams} \end{cases}$$

At $x=T$:

$$P_1(T) = 0,$$
$$P_i(T) = 0 \text{ for all reflected beams.}$$

From the Taylor's series expansion of $P_1(x)$ about $x=0$ taken up to the second order and using the condition $P_1(T)=0$, one obtains:

$$P_1(0) = -\dot{P}_1(0) \,.\, T - \tfrac{1}{2}\ddot{P}_1(0) \,.\, T^2.$$

$\dot{P}_1(0)$ and $\ddot{P}_1(0)$ are calculated from (2), (3) and (4). In the following, reflected beams are identified by a subscript r and transmitted beams by a subscript t.

Defining,

$$\left.\begin{aligned} A_o &= \mu + \bar{Q}_{o1} + \Sigma\, \bar{Q}_{ot} \,, \\ A_1 &= \mu + \bar{Q}_{1o} + \Sigma\, \bar{Q}_{1t} \\ \text{and} \quad A_r &= \mu + \bar{Q}_{ro} + \bar{Q}_{r1} + \sum_{j\neq r} \bar{Q}_{rj} \,, \end{aligned}\right\} \quad (15)$$

the following result is obtained:

$$\begin{aligned} p_1[1 + A_1l_1 &+ \tfrac{1}{2}(A_1^2l_1^2 - \bar{Q}_{1o}^2l_ol_1) + \tfrac{1}{2}\sum_r \bar{Q}_{1r}^2l_1l_r \\ - \tfrac{1}{2}\sum_t \bar{Q}_{t1}^2l_1l_t] &= \bar{Q}_{o1}l_o[1 + \tfrac{1}{2}(A_1l_1 - A_ol_o)] + \sum_r p_r\bar{Q}_{r1}l_r \\ &+ \tfrac{1}{2}\sum_r p_rl_r(\bar{Q}_{ro}\bar{Q}_{o1}l_o + A_1\bar{Q}_{r1}l_1) \\ &- \tfrac{1}{2}\sum \bar{Q}_{r1}l_r(\bar{Q}_{or}l_o - p_rA_rl_r + \sum_{j\neq r} p_j\bar{Q}_{jr}l_j) \\ &+ \tfrac{1}{2}\sum_t \bar{Q}_{t1}l_t(\bar{Q}_{ot}l_o + \sum_r p_r\bar{Q}_{rt}l_r) \,, \quad (16) \end{aligned}$$

where $p_j = \dfrac{P_j(0)}{P_o(0)}$, and the l's are the path lengths for the different beams.

Equation (16) will now be applied to two particular cases:

Case I: *Two diffracted beams*

Beam 1, reflected; beam 2, transmitted.

$$\begin{aligned} P_2(0) &= 0, \\ \bar{Q}_{1i} &= 0 \text{ for all } i \neq 0, 2. \\ A_o &= \mu + Q_{o1} + \bar{Q}_{o2} \,, \\ A_1 &= \mu + \bar{Q}_{1o} + \bar{Q}_{12} \,. \end{aligned}$$

The following result obtains:

$$p_1 = \frac{\bar{Q}_{o1}l_o + \tfrac{1}{2}\bar{Q}_{o1}l_o(A_1l_1 - A_ol_o) + \tfrac{1}{2}\bar{Q}_{o2}l_o \,.\, \bar{Q}_{21}l_2}{1 + A_1l_1 - \tfrac{1}{2}(-A_1^2l_1^2 + \bar{Q}_{1o}^2\,l_ol_1 + \bar{Q}_{12}^2l_1l_2)}. \quad (17)$$

Case II: *Two diffracted beams*

Both beams reflected: $p_1 \neq 0$, $p_2 \neq 0$.

One obtains the result:

$$p_1 = (a_1p_2 + b_1)/c_1 \,, \quad (18)$$

where:

$$\left.\begin{aligned} a_1 &= \bar{Q}_{12}l_2 + \tfrac{1}{2}(\bar{Q}_{o1}\bar{Q}_{o2}l_ol_2 + A_1\bar{Q}_{21}l_1l_2 + A_2\bar{Q}_{21}l_2^2) \,, \\ b_1 &= \bar{Q}_{o1}l_o[1 + \tfrac{1}{2}(A_1l_1 - A_ol_o)] - \tfrac{1}{2}\bar{Q}_{2o}\bar{Q}_{21}l_ol_2, \\ c_1 &= 1 + A_1l_1 + \tfrac{1}{2}(A_1^2l_1^2 - \bar{Q}_{1o}^2l_ol_1) + \tfrac{1}{2}\bar{Q}_{12}^2l_1l_2 \,. \end{aligned}\right\} \quad (19)$$

An equation similar to (18) holds for p_2:

$$p_2 = (a_2p_1 + b_2)/c_2 \,, \quad (18')$$

where a_2, b_2 and c_2 can be obtained from equations (19), interchanging subscripts 1 and 2. Finally

$$\left.\begin{aligned} p_1 &= \frac{a_1b_2 + b_1c_2}{c_1c_2 - a_1a_2} \,, \\ p_2 &= \frac{a_2b_1 + b_2c_1}{c_1c_2 - a_1a_2} \,. \end{aligned}\right\} \quad (20)$$

Since the preceding intensity solutions include only up to the second order terms in the series development, they are applicable only when conditions for the rapid convergence of the series are met, *i.e.* $\bar{Q}_{ij}l_i \ll 1$ and $\mu l_i \ll 1$. In the examples studied here the first of them was generally quite well satisfied, the values of $\bar{Q}_{ij}l_i$ being of the order of 0·01 to 0·10 (with a maximum of 0·13). However, in most cases, the values of the l_i's are determined entirely by the absorption of the crystal and then μl_i is close to 0·5 as shown below (Appendix 2). Higher order terms need then to be considered.

We give next the third order approximation only for the case where the second diffracted beam is transmitted through the crystal.

Third order approximation

Two diffracted beams: beam 1, reflected; beam 2, transmitted.

$$p_1 = \frac{\mathcal{N}}{\mathcal{D}} \,,$$

with

$$\begin{aligned}\mathcal{N} = {} & \bar{Q}_{o1}l_o + \tfrac{1}{2}[\bar{Q}_{o1}l_o(A_1l_1 - A_ol_o) + \bar{Q}_{o2}\bar{Q}_{21}l_ol_2] \\ & + \tfrac{1}{6}[-A_o^2\bar{Q}_{o1}l_o^3 - A_oA_1\bar{Q}_{o1}l_o^2l_1 - A_o\bar{Q}_{o2}\bar{Q}_{12}l_o^2l_2 \\ & + A_1^2\bar{Q}_{o1}l_ol_1^2 - \bar{Q}_{o1}^3l_o^2l_1 + \bar{Q}_{o1}\bar{Q}_{o2}^2l_o^2l_2 - \bar{Q}_{12}^2\bar{Q}_{o1}l_ol_1l_2 \\ & + A_1\bar{Q}_{21}\bar{Q}_{o2}l_ol_1l_2 - A_2\bar{Q}_{21}\bar{Q}_{o2}l_ol_2^2]\,, \end{aligned} \tag{21}$$

and

$$\begin{aligned}\mathcal{D} = {} & 1 + A_1l_1 + \tfrac{1}{2}(A_1^2l_1^2 - \bar{Q}_{01}^2l_ol_1 - \bar{Q}_{12}^2l_1l_2) \\ & + \tfrac{1}{6}[(A_ol_o - 2A_1l_1)\bar{Q}_{01}^2l_ol_1 - 2\bar{Q}_{o1}\bar{Q}_{o2}\bar{Q}_{12}l_ol_1l_2 \\ & + A_1^3l_1^3 - 2A_1\bar{Q}_{12}^2l_1^2l_2 + A_2\bar{Q}_{12}^2l_1l_2^2]\,. \end{aligned} \tag{21'}$$

Reflectivities

It has been shown by Moon & Shull (1964), that the expectation value of the reflectivity is given by:

$$\bar{Q}_{ij} = \frac{Q_{ij}^o}{\eta\sqrt{2\pi}} \exp\left[-\frac{(K_{ij}^\varepsilon\,.\,\Delta\varepsilon)^2}{2\eta^2}\right], \tag{22}$$

when the crystal is rotated about an arbitrary axis. Q_{ij}^o is the integrated reflectivity for a rotation around an axis normal to the plane of incidence, K_{ij}^ε a geometric factor which relates the rotation ($\Delta\theta_{ij}$) around this normal to that around the arbitrary axis ($\Delta\varepsilon$) (Zachariasen, 1945):

$$\Delta\theta_{ij} = K_{ij}^\varepsilon\,.\,\Delta\varepsilon\,.$$

Equation (22) holds when the mosaic distribution function has a breadth which is large by comparison with that of the diffraction pattern due to a (perfect) single block (assumption 3).

For the peak intensity, $\Delta\varepsilon = 0$, and

$$\bar{Q}_{ij}^{\text{peak}} = \frac{Q_{ij}^o}{\eta\sqrt{2\pi}}\,. \tag{23}$$

Determination of mosaic breadth

The graph recorded in the experiment described at the beginning shows a more or less uniform trace with either negative or positive peaks superimposed (Fig. 2). The magnitudes

$$\overset{\text{single}}{p_1^s = \frac{P_1(0)}{P_o(0)}} \quad\text{and}\quad \overset{\text{double}}{p_1^d = \frac{P_1(0)}{P_o(0)}}\,,$$

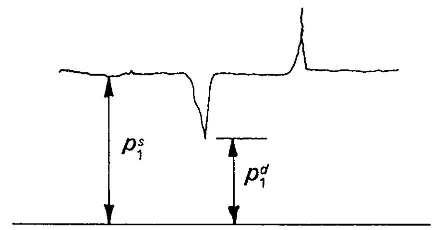

Fig. 2. Scheme of a multiple reflection pattern. The intensities, $p_1{}^s$ due to a single reflection, and $p_1{}^d$ due to a double reflection, are recorded at the same scale.

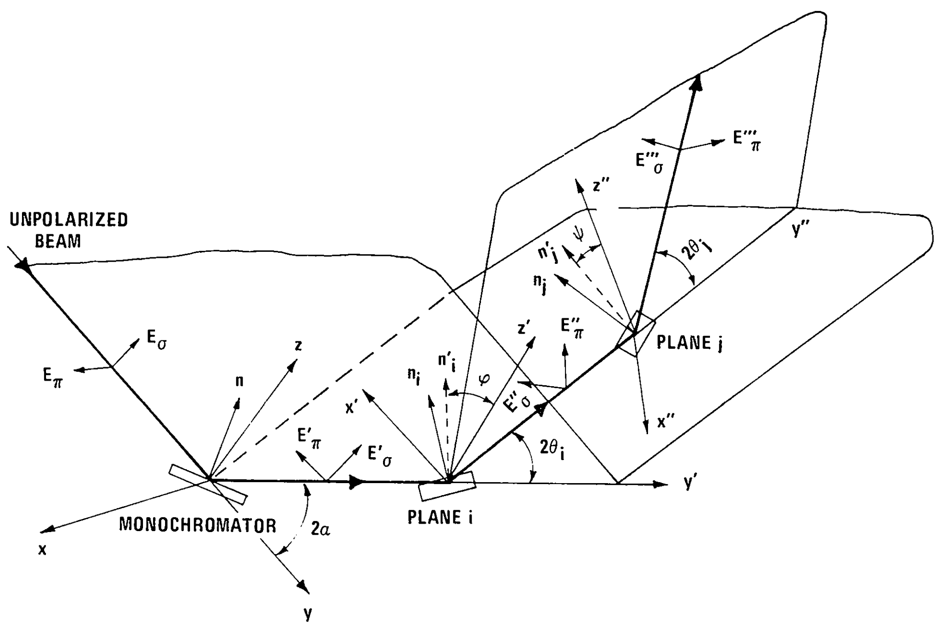

Fig. 3. Illustrating the nomenclature used in the calculation of polarization factors. E_σ indicates the electric field component in the plane of incidence and E_π the component normal to it. Planes i and j in the scheme are respectively the primary and the secondary diffracting planes. The Z axis is in the plane of incidence and normal to the incident beam, n'_i is the projection of the normal n_i to the plane i onto the $x'z'$ plane. The angle φ depends on the experimental set up, while ψ can be expressed as a function of $2\theta_i$, $2\theta_j$ and $2\theta_k$.

marked in Fig. 2, represent to a certain scale the values of p_1^s and p_1^d. The former is given by equation (7) and the latter by equation (17) or (19) and (20) according to whether the second diffracted beam is transmitted or reflected, or by the exact solution, equations (9), (11), (12) and (13) after substituting for the peak values of $\bar{Q}_{ij}$, equation (23). The fact that both magnitudes are recorded at the same scale shows that there is no need to measure absolute intensities. If we write

$$p_1^d = R \, . \, p_1^s \, ,$$

where the value of R is determined from the experiment for a given double reflection, we can substitute for p_1^s and p_1^d the expressions given by equations (7) and (17) or (21), or(7) and (20), thus obtaining an equation in the mosaic breadth η.

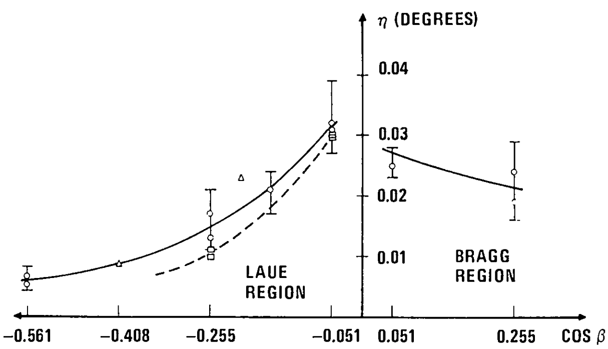

Fig. 4. Germanium plate. Some results from preliminary measurements made with Mo $K\alpha$. ○ Primary reflection: 111; values calculated without polarization correction for secondary reflecting planes diffracting in a direction forming an angle β with the normal to the plate. □ Values for the same secondary reflecting planes taking polarization into account. △ Primary reflection: 222. Each point was obtained from a pair of planes diffracting in directions which form the same angle with the normal. The bars across the experimental points indicate the uncertainty arising from the estimated error in the measured value of R.

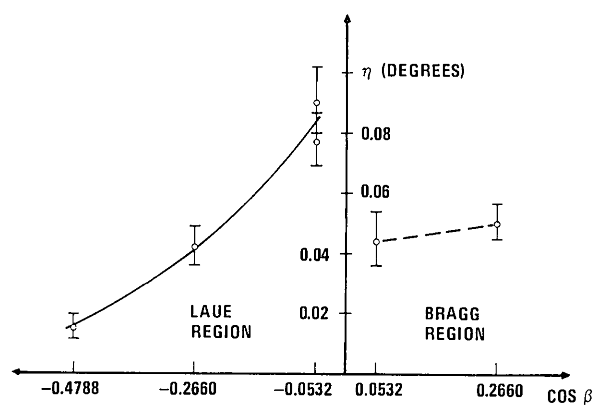

Fig. 5. Silicon plate. Some results from preliminary measurements made with Mo $K\alpha$. Primary reflection: 111. The branch of the curve in the Bragg region has the wrong slope; this is probably due to experimental error.

The method just described cannot be used in the case where **1** is a 'forbidden' reflection, since p_1^s is nil. However, the intensity of doubly diffracted beams can be quite considerable. Equation (17) assumes then the simpler form:

$$p_1 = \frac{\frac{1}{2}\bar{Q}_{02} l_0 \bar{Q}_{21} l_2}{1 + A_1 l_1 + \frac{1}{2}(A_1^2 l_1^2 - \bar{Q}_{12}^2 l_1 l_2)} \, . \qquad (24)$$

The numerator of equation (24) is of the type ky^2 where y is a magnitude inversely proportional to the mosaic breadth (we have used in most computations $y = 10^{-3}/\eta\sqrt{2\pi}$). The ratio between the peak intensities for two double-reflected beams can then be used; equation (24) leads then to a second order equation in y. Equations (21) and (21′) give a better approximation leading to a third degree equation in y.

Experimental

Preliminary experiments were performed on Ge and Si single-crystal plates; these crystals were saw cut and accordingly had considerable surface damage. The intensity diffracted by the planes (111), (222) and (333) was then recorded as a function of the angle of rotation around the diffraction vector. Experimental values for the ratio R were then obtained from the graphs for several pairs of double reflections. Each of such pairs when treated by the method explained above yielded a value for the mosaic spread η. In our case the primary diffracted beams were actually reflected (Bragg case) and the average path-lengths were determined by absorption (Appendix 2). For a crystal whose surface is considerably damaged one would expect to find important variations in the value obtained for η as one considers, for a given primary diffracted beam, secondary beams which bear a different inclination with respect to the normal to the crystal surface. For example, in a limiting case, if a secondary diffracted beam makes an angle of 90° to the normal it will give information regarding mostly the superficial layers; a beam more inclined with respect to the normal will go deeper into the crystal and should provide some information about deeper layers. One would expect then to obtain a maximum for η at 90° and then increasingly lower values as the angle between the secondary diffracted beam and the normal to the plate increases over 90° (Laue case) or as it decreases from 90° (Bragg case), but the curve is not expected to be symmetric about 90°. Also, for a given inclination of the secondary beam one should expect increasingly lower values of η as the primary beam departs more from the surface of the crystal, *i.e.* η for the primary reflection 333 should be smaller than for the 111 when secondary reflections with the same inclination are considered. These predictions are indeed confirmed by the experiment as shown in Figs. 4 and 5, where the results of the analysis have been plotted. For these preliminary measurements the second approximation was used in the calculations except in the case of the primary reflection 222 (Fig. 4) where the third approximation was used.

Since the depth to which a given beam penetrates below the surface is given by $x = l \,.\, \cos\beta$, where l is the average path-length and β is the angle with the normal, we have plotted η against $\cos\beta$ for a given primary reflection. The bars across the experimental points indicate the uncertainty arising from the estimated error in the measured value of R. This error was in general of the order of 1%. It is clear then that in order to obtain accurate values for η it is necessary to measure R to a few tenths of one per cent.

The value of η plotted should not be attributed to the layer located at the depth x below the surface, it is rather some average over the volume of the crystal between the surface and x. Different points on the same abscissae correspond to independent secondary planes having the same inclination to the normal; they agree well within the limits of the experimental error.

The fact that reflections bearing the same inclination to the normal, but actually going in different directions, may yield different values for η, could eventually be used to test the anisotropy of the mosaic spread of the sample. However, more accurate data are needed to perform such an analysis.

APPENDIX 1

Polarization correction for double reflection preceded by a crystal monochromator

In the experiments performed, a plane monochromator of silicon was used. Consequently, polarization corrections were needed for double reflections in the crystal preceded by reflection of the primary beam in the monochromator.

Azaroff (1955) has worked out the polarization correction for a single reflection in the crystal preceded by a reflection monochromator. Also, polarization corrections for double reflections when the incident beam is not polarized have been calculated by Zachariasen (1965). Neither of them applied to our case.

The polarization factors were calculated following a procedure similar to the one used by Azaroff (see Fig. 4). The electric field in the double diffracted beam, *i.e.* after three reflections, one in the monochromator (with Bragg angle α) and then in planes i and j (Bragg angles θ_i and θ_j respectively) is found to be given by:

$$\begin{aligned}\bar{E}'''^2 = \tfrac{1}{2}k^2k_i^2k_j^2\bar{E}_0^2[(\cos^2 2\theta_j \,.\, \cos^2\psi + \sin^2\psi)\cos^2 2\theta_i \\ \times(\cos^2 2\alpha\cos^2\varphi + \sin^2\varphi) \\ + (\cos^2 2\theta_j \sin^2\psi + \cos^2\psi) \\ \times(\cos^2 2\alpha \sin^2\varphi + \cos^2\varphi)]\,,\end{aligned} \tag{1·1}$$

where $\bar{E}_o$ refers to the electric field in the primary non-polarized beam and the k's include the reflectivities and other factors which are irrelevant here; the angles φ and ψ are defined in Fig. 4, φ coincides with Azaroff's angle ϱ and depends on the experimental setup. ψ can be calculated for each pair of planes i and j using a relation given by Zachariasen (1965):

$$\cos\psi = \frac{\cos 2\theta_k - \cos 2\theta_i \,.\, \cos 2\theta_j}{\sin 2\theta_i \sin 2\theta_j}\,, \tag{1·2}$$

where $2\theta_k$ is the angle between the beam reaching the crystal and the doubly diffracted beam. Substituting for ψ in equation (1·1) the following expression is found for the polarization factor:

$$\begin{aligned}p_{ij}(k) = \tfrac{1}{2}\Big\{\Big[1 - \frac{(\cos 2\theta_k - \cos 2\theta_i \,.\, \cos 2\theta_j)^2}{\sin^2 2\theta_i}\,.\Big] \\ \times\cos^2 2\theta_i(1 - \sin^2 2\alpha \,.\, \cos^2\varphi) \\ + \Big[\frac{(\cos 2\theta_k - \cos 2\theta_i \cos 2\theta_j)^2}{\sin^2 2\theta_i} \\ + \cos^2 2\theta_j\Big] \,.\, (\sin^2 2\alpha \cos^2\varphi + \cos^2 2\alpha)\Big\}\,.\end{aligned} \tag{1·3}$$

In our case, $\varphi = \pi/2$ and then

$$\begin{aligned}p_{ij}(k) = \tfrac{1}{2}\Big\{\frac{(\cos 2\theta_k - \cos 2\theta_i \cos 2\theta_j)^2}{\sin^2 2\theta_i} \\ \times(\cos^2 2\alpha - \cos^2 2\theta_i) + \cos^2 2\alpha\cos^2 2\theta_j + \cos^2 2\theta_i\Big\}\end{aligned} \tag{1·4}$$

It is readily seen that equation (1·3) contains as particular cases both Zachariasen's and Azaroff's formulae. In fact, when there is no monochromator, setting $\alpha = 0$ one reobtains Zachariasen's result [his equation (6)].

When there is just a single reflection, preceded by the monochromator, we set: $\theta_j = 0$, $\theta_k = \theta_i$ thus obtaining:

$$p_{oi} = \tfrac{1}{2}(\sin^2 2\theta_i \,.\, \sin^2 2\alpha \,.\, \cos^2\varphi + \cos^2 2\theta_i + \cos^2 2\alpha]$$

In our case $\varphi = \pi/2$ and

$$p_{oi} = \tfrac{1}{2}[\cos^2 2\alpha + \cos^2 2\theta_i]\,.$$

In the common case where $\varphi = 0$, one finds the familiar result:

$$p_{oi} = \tfrac{1}{2}[1 + \cos^2 2\alpha \,.\, \cos^2 2\theta_i]\,.$$

It is to be noted that in our notation Azaroff's expression (14) is given by $\frac{p_{oi}}{p_o}$, since it is referred to the intensity of the beam after reflection on the monochromator.

APPENDIX 2

Path-lengths

The path-lengths of the different diffracted beams will now be obtained. It is easy to show that for a plane parallel plate of thickness T, the transmission factor for a reflected beam is

$$A(\mu) = \frac{\gamma}{\mu T}\left[1 - \exp\left(-\frac{\mu T}{\gamma}\right)\right]$$

where

$$\frac{1}{\gamma}=\frac{1}{\gamma_0}+\frac{1}{\gamma_i}.$$

For a diffracted beam originated at depth x below the crystal surface, the total path-length, including the distance travelled by the incident beam, is given by x/γ. The average value of x/γ is given by

$$\left\langle\frac{x}{\gamma}\right\rangle=A\frac{\mathrm{d}A^*}{\mathrm{d}\mu}$$

where $A^*=1/A$.

It can be readily shown that in this case

$$\left\langle\frac{x}{\gamma}\right\rangle=\frac{1}{\mu}-\frac{T}{\gamma}\cdot\frac{\exp(-\mu T/\gamma)}{1-\exp(-\mu T/\gamma)}.$$

Moreover,

$$\left\langle\frac{x}{\gamma}\right\rangle=\frac{\langle x\rangle}{\gamma_o}+\frac{\langle x\rangle}{\gamma_i},$$

$$\text{where }\frac{\langle x\rangle}{\gamma_o}=l_o\text{ and }\frac{\langle x\rangle}{\gamma_i}=l_i.$$

For a highly absorbing crystal (as for instance germanium with Mo $K\alpha$, $\mu=350\ \mathrm{cm}^{-1}$) we have practically:

$$\left\langle\frac{x}{\gamma}\right\rangle=\frac{1}{\mu}.$$

Then,

$$l_o=\frac{\langle x\rangle}{\gamma_o}=\frac{1}{\mu}\cdot\frac{\gamma}{\gamma_o}$$

and

$$l_i=\frac{\langle x\rangle}{\gamma_i}=\frac{1}{\mu}\cdot\frac{\gamma}{\gamma_i}.$$

When the primary reflection is symmetric: $\gamma_o=\gamma_1$, $\gamma/\gamma_o=\gamma/\gamma_1=\frac{1}{2}$ and $l_o=l_1=1/2\mu$.

Then

$$l_i=\frac{\langle x\rangle}{\gamma}-l_o=\frac{1}{\mu}-\frac{1}{2\mu}=\frac{1}{2\mu}$$

for all secondary reflections.

When the secondary beam is transmitted rather than reflected and the crystal is highly absorbing the same reasoning can be followed and one find the same effective path-lengths $1/2\mu$ for all beams.

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Quelques Propriétés des Facteurs de Structure

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The structure factor is shown to be a basis for a one-dimensional representation of the point group, and the following property of non-primitive translations of most of the space groups is derived: the sum of non-primitive translations is a primitive one.

Représentation engendrée par le facteur de structure

Rappelons qu'un facteur de structure trigonométrique $\xi(K)=\sum_\alpha\exp[-2\pi i(K.\alpha r+\tau_\alpha)]$ se transforme dans une opération $(\alpha|\tau_\alpha)$ d'un groupe d'espace G_e suivant la loi (Bertaut, 1955):

$$\xi(K\alpha)=\exp(-2\pi iK.\tau_\alpha)\xi(K). \quad (1)$$

$\xi(K)$ est donc fonction de base d'une représentation Γ_K du groupe ponctuel G d'ordre g:

$$\Gamma_K(\alpha)=\exp(-2\pi iK.\tau_\alpha). \quad (2)$$

Deux éléments $(\alpha|\tau_\alpha)$ et $(\beta|\tau_\beta)$ de G_e se multiplient suivant la relation (3), où $T_{\alpha\beta}$ est une translation réticulaire et ε l'identité: