The pertinent intensity observations all reduced to an arbitrary scale on which $P_{1c}=814$ are listed in Table 3. For the 200 and 220 reflections one has $A_3/A_1=1{\cdot}09$, $\bar{T}_3/\bar{T}_1=0{\cdot}90$, $L_1^*/L_1=1{\cdot}46$, $p_{11}(3)/p_2=0{\cdot}97$ giving $a=2{\cdot}04$. On absolute scale one calculates $Q_1=29{\cdot}1\times10^{-3}$, $Q_3=0{\cdot}54\times10^{-3}$, and $\bar{T}_1=1{\cdot}81\times10^{-2}$ cm.

Table 3. *Initial* (*A*) *and final* (*B*) *intensities*

Reflection	*A*	*B*	Calculated
200	489	807	814
220	16·3	16·2	16·4
220*	31·2	49·1	—

Insertion of the data from Table 3 in equations (6) gives

$$x_A=2{\cdot}5\times10^{-2} \qquad y_A=0{\cdot}61$$
$$x_B=1{\cdot}9\times10^{-2} \qquad y_B=1{\cdot}00\ (1{\cdot}01)\ .$$

The corresponding values of the secondary extinction coefficient g are $g_A=46$ and $g_B=36$. Equation (3) suggests that $t_0\simeq6\times10^{-4}$ cm for the "virgin" crystal.

In other words the data show that the secondary extinction in the specimen is small, *i.e.* that there is poor alignment of the mosaic blocks. The initial large primary extinction in the specimen is practically eliminated in the course of the irradiation, *i.e.* the dimensions of the coherently scattering regions in the crystal are greatly reduced as a consequence of the radiation disorder.

Clearly, the practical usefulness of the proposed method for distinguishing between primary and secondary extinction ought to be given further tests.

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Multiple Diffraction in Imperfect Crystals

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It is shown that many experimental diffraction intensities used for structure determination have been obtained under conditions of multiple diffraction, and it is pointed out that some of the intensities measured under such circumstances may be appreciably in error.

The intensity effect has been studied theoretically for frequently encountered cases of double, triple and quintuple diffraction in imperfect (mosaic) crystals. Exact solutions have been obtained for a plane parallel plate and approximate formulas are given for crystals of arbitrary shape.

The results of the theoretical study show that multiple diffraction may double or triple the extinction correction for strong reflections and, in some cases, increase the intensity of weak reflections manifold.

The first objective of this article is to demonstrate that multiple diffraction due to symmetry is a much more common phenomenon in crystal structure studies by X-rays, electrons or neutrons than is generally realized.

The second purpose is to give the results of a theoretical study of the intensity effects due to multiple diffraction in an imperfect crystal.

Multiple diffraction due to symmetry

Most intensity measurements reported in the literature, and used for structure determination, have been made under conditions of multiple diffraction. The classical example of symmetry-caused simultaneous diffraction is a Laue photograph taken with the incident beam parallel to a symmetry axis or plane. Every Laue spot of an equivalent set is produced at the same time by the same wave length component of the incident radiation, and as many as twelve reflections may occur simultaneously.

A more important example of double diffraction (case *A*) is illustrated in Fig. 1. The Laue–Bragg equation $\mathbf{k}_i-\mathbf{k}_0=\mathbf{H}_i$, where $\mathbf{k}_i=\lambda^{-1}\mathbf{u}_i$, is simultaneously satisfied for two reciprocal lattice vectors $\mathbf{H}_1$ and $\mathbf{H}_2$ for which, by symmetry, $|\mathbf{H}_1|=|\mathbf{H}_2|$. In Fig. 1, O is the origin of the reciprocal lattice, $\mathbf{H}_1$ and $\mathbf{H}_2$ lie in the plane of the paper, and the circle is the intersection of this plane with the sphere of reflection. The situation shown in Fig. 1 is bound to occur for any wave-length when the normal beam Weissenberg technique is used with a non-triclinic crystal rotating about [010] (and for any crystal of orthorhombic or higher symmetry rotating about [100] or [001]). Reflections *HKL* and $H\bar{K}L$ occur simultaneously, *i.e.* all intensity data for non-zero layer lines will have been obtained under the double diffraction conditions of case *A*.

A special situation (however, of no particular interest) arises if, in Fig. 1, $|\mathbf{H}_1|=|\mathbf{H}_2|=|\mathbf{H}_1-\mathbf{H}_2|$. The center of the sphere of reflection will in that case lie on a threefold axis.

A common and important case (*B*) of triple diffraction is shown in Fig. 2(*a*). In this example $\mathbf{H}_1 \,.\, \mathbf{H}_2=0$, and the Laue–Bragg equation is also satisfied for $\mathbf{H}_3=\mathbf{H}_1+\mathbf{H}_2$. This type of triple diffraction occurs inevitably for any wave-length when the equi-inclination Weissenberg technique is used with a non-triclinic crystal rotating about [010] (or with crystals of orthorhombic symmetry rotating about [100] or [001]). If the inclination angle is set for the *K*th layer, the reflections *H*0*L*, 0*K*0 and *HKL* will be produced at the same time. In other words, all intensity data for non-zero layer lines correspond to triple diffraction conditions. This fact in respect to the equi-inclination Weissenberg technique was first recognized implicitly by Fankuchen & Williamson (1956) and explicitly by Yakel & Fankuchen (1962).

Fig. 2(*b*) demonstrates the special case (*B*′) of triple diffraction for which $\mathbf{H}_1 \,.\, \mathbf{H}_2=0$ and $|\mathbf{H}_1|=|\mathbf{H}_2|$. This particular situation is thus possible for tetragonal or cubic crystals, and will occur for rotation axes [010] and $[1\bar{1}0]$.

An interesting case (*C*) of quintuple diffraction, possible for hexagonal or cubic crystals, is illustrated in Fig. 3. An example is a hexagonal crystal with [010] vertical, and crystal and counter set to measure the (200) reflection in the horizontal plane. The four additional reflections (010), $(1\bar{1}0)$, (110) and $(2\bar{1}0)$ are produced at the same time.

The examples discussed above demonstrate that multiple diffraction is a frequent phenomenon with commonly used experimental techniques. Indeed, it is

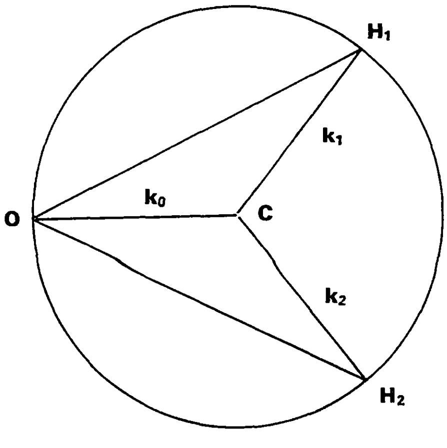
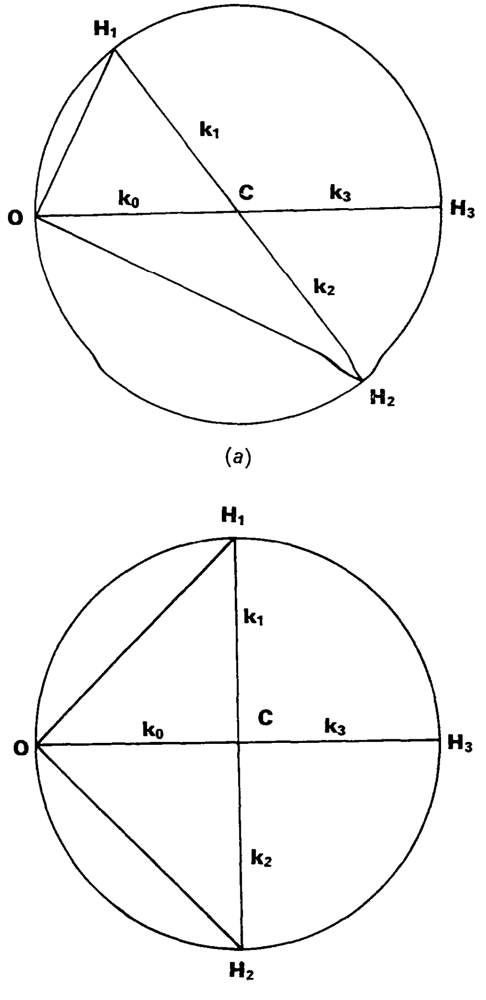


Fig. 1. Double diffraction (case A). *O* is the origin of the reciprocal lattice. The vectors $\mathbf{H}_1$ and $\mathbf{H}_2$ lie in the plane of the paper and are given by OH_1 and OH_2, where $OH_1=OH_2$. The center of the sphere of reflection is at *C*, at a height $\lambda^{-1}\cos\varphi$ above the paper. The three wave vectors $\mathbf{k}_i$ are *CO*, CH_1 and CH_2.



(*a*)

(*b*)

Fig. 2. (*a*) Case *B* of triple diffraction with $\mathbf{H}_1 \,.\, \mathbf{H}_2=0$ and $\mathbf{H}_3=\mathbf{H}_1+\mathbf{H}_2$. (*b*) The special case *B*′ of triple diffraction for which $\mathbf{H}_1 \cdot \mathbf{H}_2=0$ and $|\mathbf{H}_1|=|\mathbf{H}_2|$.

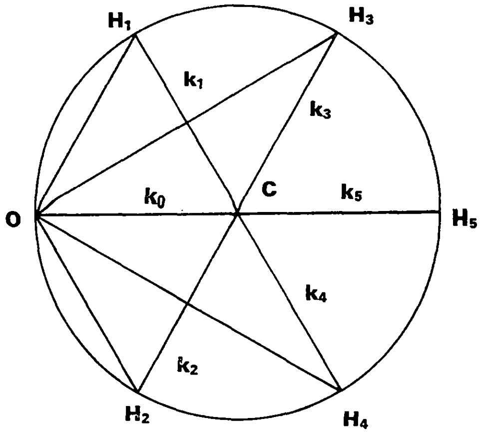


Fig. 3. The quintuple diffraction case *C*. For this case the points $OH_1H_3H_5H_4H_2$ form a hexagon. $|\mathbf{H}_1|=|\mathbf{H}_2|$ and $|\mathbf{H}_3|=|\mathbf{H}_4|$. $\mathbf{H}_3=2\mathbf{H}_1+\mathbf{H}_2$, $\mathbf{H}_4=\mathbf{H}_1+2\mathbf{H}_2$ and $\mathbf{H}_5=2\mathbf{H}_1+2\mathbf{H}_2$.

in general necessary to take special precautions in order to avoid making intensity measurements under simultaneous diffraction conditions.

Intensity solutions for a plane parallel plate

It will be assumed that the crystal specimen under study is of mosaic type with moderate or small extinction. Accordingly the half width of the homogenous and isotropic distribution function W describing the misalignment of the mosaic blocks is presumed to be much greater than that of the diffraction pattern due to a single block.

When the Laue–Bragg equation $\mathbf{k}_i-\mathbf{k}_0=\mathbf{H}_i$ is satisfied for two or more lattice planes $\mathbf{H}_i$, it follows that $\mathbf{k}_j-\mathbf{k}_i=\mathbf{H}_j-\mathbf{H}_i$, *i.e.* the equation is also satisfied for $\mathbf{H}_j-\mathbf{H}_i$ with $\mathbf{k}_i$ now serving as incident beam. Hence, multiple diffraction affects the intensities in two ways. First, the absorption due to scattering (*i.e.* extinction) will be greatly enhanced. Second, the radiation scattered in a direction $\mathbf{u}_j$ consists not only of the incident beam reflected by $\mathbf{H}_j$, but also of each diffracted beam $\mathbf{k}_i$ reflected by $\mathbf{H}_j-\mathbf{H}_i$. In this manner incident and diffracted beams form a coupled system, the intensity of one beam depending upon the intensities of all the others.

For the sake of convenience let the crystal be a plate of thickness T_0 parallel to the mean vectors $\mathbf{H}_1$ and $\mathbf{H}_2$. In the cases A, B, C to be studied each beam makes the same angle φ with the normal to the plate, and the path length through the crystal is $T_0/\cos\varphi$. Consider a sharply defined direction of incidence, deviating by a small angle Δ from the ideal direction.

Let $P_i(\Delta, T)$ be the power of the ith beam at a distance of travel T into the crystal plate and $\dot{P}_i\equiv dP_i/dT$. The power losses due to absorption and to diffraction by the jth lattice plane are respectively $\dot{P}_i=-\mu_0P_i$ and $\dot{P}_i=-\sigma_jP_i$. Similarly the power gain due to diffraction of the jth beam by the kth lattice plane in the ith scattering direction is $\dot{P}_i=\sigma_kP_j$. If there is n-fold diffraction one gets accordingly a system of $n+1$ coupled equations of the form $\dot{P}_i=\Sigma a_{ij}P_j$. The solutions will be of the type $P_i=\Sigma A_{ij}e^{Z_jT}$ where the Z_j's are the roots of the secular equation of degree $n+1$ and where the coefficients A_{ij} must be determined from the boundary conditions. The latter are $P_i(0)=0$ for $i\neq 0$ and $P_0(0)\equiv\mathscr{P}_0$, the total incident power at the crystal surface. Since absorption represents the only real power loss, one must have $\Sigma\dot{P}_i=-\mu_0\Sigma P_i$ and hence $\Sigma P_i=\mathscr{P}_0e^{-\mu_0T}$ with $T=T_0/\cos\varphi$. It is further obvious that $e^{-\mu_0T}$ must be a particular integral of the differential equations.

Under the assumed conditions the reflecting power σ will be $\sigma_j=W(\Delta)Q_j$, with $Q_j=|e^2F_j/mc^2V|^2\lambda^3L_j$ where L_j is the Lorentz factor. For the present polarization effects will be ignored.

The solution of the system of equations gives the quantities $P_i(\Delta)$ while the observable powers are $\bar{P}_i=\int P_i(\Delta)d\Delta$. To obtain suitable expressions for $\bar{P}_i$ the exponentials e^{Z_jT} can be expanded in series, and one encounters integrals of the form $\int\sigma_jd\Delta=Q_j$, $\int\sigma_i\sigma_jd\Delta=gQ_iQ_j$, where $g=\int W^2d\Delta$.

The differential equations and their solutions are given below for the cases A, B, C of Figs. 1–3. The results for the special case of Fig. 2(*b*) are obtained if for case B one sets $\sigma_1=\sigma_2$ and $Q_1=Q_2$. The well known formulas for the single diffraction case are included for the sake of comparison.

Single diffraction

$$\begin{array}{r|cc} & P_0 & P_1 \\ \hline \dot{P}_0= & -\mu_0-\sigma_1 & \sigma_1 \\ \dot{P}_1= & \sigma_1 & -\mu_0-\sigma_1 \end{array} \qquad (1a)$$

$$\begin{aligned} P_0&=\tfrac{1}{2}\mathscr{P}_0e^{-\mu_0T}[1+e^{-2\sigma_1T}] \\ P_1&=\tfrac{1}{2}\mathscr{P}_0e^{-\mu_0T}[1-e^{-2\sigma_1T}] \end{aligned} \qquad (1b)$$

$$\begin{aligned} \bar{P}_0&=\mathscr{P}_0e^{-\mu_0T}[1-Q_1T+gQ_1^2T^2+\ldots] \\ \bar{P}_1&=\mathscr{P}_0Te^{-\mu_0T}[Q_1-gQ_1^2T+\ldots] \end{aligned} \qquad (1c)$$

Double diffraction, case A

$$\begin{array}{r|ccc} & P_0 & P_1 & P_2 \\ \hline \dot{P}_0= & -\mu_0-2\sigma_1 & \sigma_1 & \sigma_1 \\ \dot{P}_1= & \sigma_1 & -\mu_0-\sigma_1-\sigma_3 & \sigma_3 \\ \dot{P}_2= & \sigma_2 & \sigma_3 & -\mu_0-\sigma_1-\sigma_3 \end{array}$$

σ_3 is the reflecting power of $\mathbf{H}_1-\mathbf{H}_2$. (2*a*)

$$\begin{aligned} P_0&=\tfrac{1}{3}\mathscr{P}_0e^{-\mu_0T}[1+2e^{-3\sigma_1T}] \\ P_1=P_2&=\tfrac{1}{3}\mathscr{P}_0e^{-\mu_0T}[1-e^{-3\sigma_1T}] \end{aligned} \qquad (2b)$$

$$\begin{aligned} \bar{P}_0&=\mathscr{P}_0e^{-\mu_0T}[1-2Q_1T+3gQ_1^2T^2+\ldots] \\ \bar{P}_1&=\mathscr{P}_0Te^{-\mu_0T}[Q_1-\tfrac{3}{2}gQ_1^2T+\ldots]. \end{aligned} \qquad (2c)$$

Triple diffraction, case B

$$\begin{array}{r|cccc} & P_0 & P_1 & P_2 & P_3 \\ \hline \dot{P}_0= & -\mu & \sigma_1 & \sigma_2 & \sigma_3 \\ \dot{P}_1= & \sigma_1 & -\mu & \sigma_3 & \sigma_2 \\ \dot{P}_2= & \sigma_2 & \sigma_3 & -\mu & \sigma_1 \\ \dot{P}_3= & \sigma_3 & \sigma_2 & \sigma_1 & -\mu \end{array} \qquad (3a)$$

$$\mu=\mu_0+\sigma_1+\sigma_2+\sigma_3$$

$$\begin{aligned} P_0&=\tfrac{1}{4}\mathscr{P}_0e^{-\mu_0T}[1+ \\ &\qquad e^{-2(\sigma_1+\sigma_2)T}+e^{-2(\sigma_1+\sigma_3)T}+e^{-2(\sigma_2+\sigma_3)T}] \\ P_1&=\tfrac{1}{4}\mathscr{P}_0e^{-\mu_0T}[1- \\ &\qquad e^{-2(\sigma_1+\sigma_2)T}-e^{-2(\sigma_1+\sigma_3)T}+e^{-2(\sigma_2+\sigma_3)T}] \\ P_2&=\tfrac{1}{4}\mathscr{P}_0e^{-\mu_0T}[1- \\ &\qquad e^{-2(\sigma_1+\sigma_2)T}+e^{-2(\sigma_1+\sigma_3)T}-e^{-2(\sigma_2+\sigma_3)T}] \\ P_3&=\tfrac{1}{4}\mathscr{P}_0e^{-\mu_0T}[1+ \\ &\qquad e^{-2(\sigma_1+\sigma_2)T}-e^{-2(\sigma_1+\sigma_3)T}-e^{-2(\sigma_2+\sigma_3)T}] \end{aligned} \qquad (3b)$$

$$\begin{aligned} \bar{P}_0&=\mathscr{P}_0e^{-\mu_0T}[1-(Q_1+Q_2+Q_3)T \\ &\quad +g(Q_1^2+Q_2^2+Q_3^2+Q_1Q_2+Q_1Q_3+Q_2Q_3)T^2+\ldots] \\ \bar{P}_1&=\mathscr{P}_0Te^{-\mu_0T}[Q_1+ \\ &\qquad g(Q_2Q_3-Q_1^2-Q_1Q_2-Q_1Q_3)T+\ldots] \end{aligned}$$

$$\bar{P}_2=\mathscr{P}_0Te^{-\mu_0T}[Q_2+ g(Q_1Q_3-Q_1Q_2-Q_2^2-Q_2Q_3)T+\ldots]$$

$$\bar{P}_3=\mathscr{P}_0Te^{-\mu_0T}[Q_3+ g(Q_1Q_2-Q_1Q_3-Q_2Q_3-Q_3^2)T+\ldots] \qquad (3c)$$

Quintuple diffraction, case C

$$\begin{array}{r|cccccc}
 & P_0 & P_1 & P_2 & P_3 & P_4 & P_5 \\
\hline
\dot{P}_0= & -\mu & \sigma_1 & \sigma_1 & \sigma_3 & \sigma_3 & \sigma_5 \\
\dot{P}_1= & \sigma_1 & -\mu & \sigma_3 & \sigma_1 & \sigma_5 & \sigma_3 \\
\dot{P}_2= & \sigma_1 & \sigma_3 & -\mu & \sigma_5 & \sigma_1 & \sigma_3 \\
\dot{P}_3= & \sigma_3 & \sigma_1 & \sigma_5 & -\mu & \sigma_3 & \sigma_1 \\
\dot{P}_4= & \sigma_3 & \sigma_5 & \sigma_1 & \sigma_3 & -\mu & \sigma_1 \\
\dot{P}_5= & \sigma_5 & \sigma_3 & \sigma_3 & \sigma_1 & \sigma_1 & -\mu
\end{array} \qquad (4a)$$

$$\mu=\mu_0+2\sigma_1+2\sigma_3+\sigma_5\ .$$

$$P_0=\tfrac{1}{6}\mathscr{P}_0e^{-\mu_0T}[1+ 2e^{-3(\sigma_1+\sigma_3)T}+2e^{-(\sigma_1+3\sigma_3+2\sigma_5)T}+e^{-(4\sigma_1+2\sigma_5)T}]$$

$$P_1=P_2=\tfrac{1}{6}\mathscr{P}_0e^{-\mu_0T}[1- e^{-3(\sigma_1+\sigma_3)T}+e^{-(\sigma_1+3\sigma_3+2\sigma_5)T}-e^{-(4\sigma_1+2\sigma_5)T}]$$

$$P_3=P_4=\tfrac{1}{6}\mathscr{P}_0e^{-\mu_0T}[1- e^{-3(\sigma_1+\sigma_3)T}-e^{-(\sigma_1+3\sigma_3+2\sigma_5)T}+e^{-(4\sigma_1+2\sigma_5)T}]$$

$$P_5=\tfrac{1}{6}\mathscr{P}_0e^{-\mu_0T}[1+ 2e^{-3(\sigma_1+\sigma_3)T}-2e^{-(\sigma_1+3\sigma_3+2\sigma_5)T}-e^{-(4\sigma_1+2\sigma_5)T}] \qquad (4b)$$

$$\bar{P}_0=\mathscr{P}_0e^{-\mu_0T}[1-(2Q_1+2Q_3+Q_5)T +g(3Q_1^2+3Q_3^2+Q_5^2+4Q_1Q_3+2Q_1Q_5+ +2Q_3Q_5)T^2+\ldots]$$

$$\bar{P}_1=\mathscr{P}_0Te^{-\mu_0T}[Q_1+ g(Q_3Q_5-2Q_1^2-Q_1Q_3-Q_1Q_5)T+\ldots]$$

$$\bar{P}_3=\mathscr{P}_0Te^{-\mu_0T}[Q_3+ g(\tfrac{1}{2}Q_1^2+Q_1Q_5-2Q_1Q_3-\tfrac{3}{2}Q_3^2-Q_3Q_5)T+\ldots]$$

$$\bar{P}_5=\mathscr{P}_0Te^{-\mu_0T}[Q_5+ g(2Q_1Q_3-2Q_1Q_5-2Q_3Q_5-Q_5^2)T+\ldots] \qquad (4c)$$

Discussion of the intehsity equations

The basic formulas given in equations (1)–(4) contain a number of interesting features and the reader is urged to study them in detail.

Only terms up to T^2 are specifically given in the expansions; but higher order terms can be added as needed.

One important point about the formulas of equations (3c) and (4c) for the triple and quintuple diffractions should be given special mention. It is seen that $\bar{P}_i \neq 0$ even when $Q_i=0$. In other words, multiple diffraction can cause otherwise "absent" reflections to appear. Renninger (1937) was the first to observe and explain the appearance of such spurious reflections.

The only purpose of the discussion to follow is to make realistic estimates as to the magnitude of the intensity effects, and to this end it is convenient to assume special, simplifying situations which will not affect the general validity of the conclusions.

Disregarding terms in T^3 and higher powers all expressions $\bar{P}_i$ for diffracted beams can be given in the form

$$\bar{P}_i \simeq I_0Q_iT_{e}^{-\mu_0T}[1+\alpha_i]$$

where the factor outside the bracket represents the integrated intensity in the limit of zero extinction. The quantity α_i is proportional to gT with the proportionality factor known from equations (1c)–4(c). Specifically for the single diffraction case one has the well known result $\alpha_1=-Q_1gT$. It is clear that $100\alpha_i$ measures the percentage correction due to combined extinction and repeated reflection.

A realistic value of g for a crystal of small to moderate extinction is 2×10^2 which corresponds to a half width for W of 0·08° if W is an error function. For an average crystal one has in practical situations $QT=10^{-3}$ for a strong and $QT=5\times10^{-5}$ for a weak reflection. The single diffraction extinction correction is thus twenty per cent for a strong and one per cent for a weak reflection. Since $gQ^2T^2 \ll 1$ and $QT \ll 1$, the series expansions used to obtain equations (1c)–(4c) are justifiable.

Table 1 gives the theoretical values of α_i for various selected cases using $g=2\times10^2$ and $QT=10^{-3}$ or 5×10^{-5} for strong or weak reflections respectively.

Table 1. *Multiple diffraction corrections*

Type of multiple diffraction		Reflections Strong $gQT=0{\cdot}20$	 Weak 0·01	Relations between $\bar{P}_i$'s	α_i
Single		Q_1		$\bar{P}_1$	−0·20
Double		Q_1		$\bar{P}_1$	−0·30
Triple					
	(a)	Q_1, Q_2, Q_3		$\bar{P}_1=\bar{P}_2=\bar{P}_3$	−0·40
	(b)	Q_1, Q_2		$\bar{P}_1=\bar{P}_2$	−0·40
			Q_3	$\bar{P}_3$	3·59
Quintuple					
	(a)	Q_1, Q_3, Q_5		$\bar{P}_1=\bar{P}_3=\bar{P}_5$	−0·60
	(b)	Q_1, Q_3		$\bar{P}_1=\bar{P}_3$	−0·60
			Q_5	$\bar{P}_5$	7·39

When all reflections involved in multiple diffraction are equally strong, the extinction correction is increased by a factor of 1·5 for double, 2 for triple and 3 for quintuple diffraction as compared with single diffraction. Thus for our standard strong reflection the correction is increased from −20 to −60 per cent.

As shown in Table 1 the intensity effect becomes spectacular in triple and quintuple diffraction if one reflection is weak and the others strong. As a striking example consider triple diffraction with $Q_1=Q_2$, $Q_3=0$. One finds

$$\bar{P}_3/\bar{P}_1=gQ_1T/1-2gQ_1T \qquad (5)$$

so that $\bar{P}_3/\bar{P}_1=\frac{1}{3}$ if $gQ_1T=0{\cdot}2$. (It must be remembered that $gQ_1T\ll 1$ is a condition which must be fulfilled in using the first order approximations).

Although the above discussion is both sketchy and crude it serves to show that it is not normally permissible to neglect the effect of multiple diffraction in quantitative intensity work. The obvious way to circumvent such complications is to avoid making intensity measurements under simultaneous diffraction conditions. However, the considerations of the preceding section of this paper show that multiple diffraction (due to symmetry and chance) is a common phenomenon. Hence, before making any one intensity measurement, one needs to make certain that the experimental conditions are not such as to produce multiple diffraction.

The writer has extended the study to include polarization as well as crystals of arbitrary shape. The results of these further investigations are given in the two appendices.

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APPENDIX

I. Polarization factors

Polarization effects can be included in the equations in the following manner. Assuming the incident beam to consist of unpolarized X-rays, the electric vectors can be resolved into components along two conveniently chosen, mutually perpendicular vibration directions (a) and (b). The differential equations for the P_i's can then be set up separately for each of the two vibration directions using the appropriate expressions $\sigma_i^{(a)}=WQ_i^{(a)}$ and $\sigma_i^{(b)}=WQ_i^{(b)}$. Thus one gets distinct solutions $\bar{P}_i^{(a)}$, $\bar{P}_i^{(b)}$ and the sought result is $\bar{P}_i=\frac{1}{2}(\bar{P}_i^{(a)}+\bar{P}_i^{(b)})$. Although the procedure is straightforward the expressions become complicated if one goes beyond the quadratic terms in the series expansion.

The polarization factor p_i for a single deflection is defined by $Q_ip_i=[Q_i^{(a)}+Q_i^{(b)}]/2$, and one finds readily the familiar result $p_i=(1+\cos^2 2\theta_i)/2$.

Next consider a doubly reflected X-ray beam. The incident beam is first reflected by the lattice plane $\mathbf{H}_i$ in scattering direction $\mathbf{u}_i$ such that $\mathbf{u}_0 \,.\, \mathbf{u}_i=\cos 2\theta_i$. The reflected beam is again reflected by a lattice plane $\mathbf{H}_j=\mathbf{H}_k-\mathbf{H}_i$. The angle of scattering relative to the once-reflected beam is $2\theta_j$; but the scattering direction is $\mathbf{u}_k$. The two scattering angles $2\theta_i$ and $2\theta_j$ do not uniquely determine the direction $\mathbf{u}_k$ and hence one needs to specify a third angle $2\theta_k$, given by $\mathbf{u}_0 \,.\, \mathbf{u}_k=\cos 2\theta k$, which measures the angle between the incident and the twice-reflected beam.

Accordingly one must use the notation $p_{ij}(k)$ for the polarization factor of a twice-reflected beam. The indices i, j designate the scattering angles for the two reflections and the index k the resultant scattering angle relative to the incident beam.

The analysis of the problem gives as result

$$p_{ij}(k)=\tfrac{1}{2}[\cos^2 2\theta_i+\cos^2 2\theta_j \\ +(\cos 2\theta_k-\cos 2\theta_i\cos 2\theta_j)^2]\,. \quad (6)$$

Azaroff (1955) considered the same problem for another purpose. Instead of $2\theta_k$ Azaroff used an auxiliary angle ϱ which is not directly observable, but which is related to $2\theta_k$ in the following manner

$$\cos 2\theta_k=\cos 2\theta_i\cos 2\theta_j+\sin 2\theta_i\sin 2\theta_j\cos\varrho\,.$$

In other words ϱ is the angle opposite $2\theta_k$ in the spherical triangle of which $2\theta_i$, $2\theta_j$ and $2\theta_k$ are sides. It is readily verified that Azaroff's equation (14) gives the correct expression for the quantity $p_{12}(k)/p_1$.

It is obviously true from equation (5) that $p_{ij}(k)=p_{ji}(k)$. In the multiple diffraction situations with which this paper is concerned it is evident that

$$p_{ii}(0)=(1+\cos^4 2\theta_i)/2 \quad (7)$$

since the incident beam twice reflected by the same lattice plane will be in the direction of incidence. However, it is important to note that the polarization factor is not given by equation (7) if the two reflecting planes are equivalent but not identical. In this latter case one has

$$p_{ii}(k)=[2\cos^2 2\theta_i+(\cos 2\theta_k-\cos^2 2\theta_i)^2]/2\,. \quad (8)$$

Products Q_iQ_j and Q_i^2 occur in the first order expansion terms, and by definition

$$p_{ij}(k)Q_iQ_j=\tfrac{1}{2}[Q_i^{(a)}Q_j^{(a)}+Q_i^{(b)}Q_j^{(b)}]\,. \quad (9)$$

The multiple diffraction cases A, B' and C are degenerate in the sense that $Q_1=Q_2$ (A, B', C) and $Q_3=Q_4$ (C), and one must accordingly exercise care in deducing the appropriate polarization factors for the terms Q_1^2 and Q_3^2.

By way of illustration the intensity formulas, with polarization factors, are given in detail for the non-degenerate (B) and the degenerate (B') cases of triple diffraction.

Triple diffraction, case B

$$\bar{P}_1\simeq\mathscr{P}_0Te^{-\mu_0T}[Q_1p_1+g\{Q_2Q_3p_{23}(1)-Q_1^2p_{11}(0) \\ -Q_1Q_2p_{12}(3)-Q_1Q_3p_{13}(2)\}T] \quad (10)$$

with analogous expressions for $\bar{P}_2$ and $\bar{P}_3$.

Triple diffraction, case B′

$$\bar{P}_1=\bar{P}_2\simeq\mathscr{P}_0T^{-\mu_0T}[Q_1p_1-g\{p_{11}(0)+p_{11}(3)\}Q_1^2T]$$

$$\bar{P}_3\simeq\mathscr{P}_0Te^{-\mu_0T}[Q_3p_3+g\{p_{11}(3)Q_1^2 \\ -2p_{11}(3)Q_1Q_3-p_{33}(0)Q_3^2\}T]\,. \quad (11)$$

II. Crystals of any shape

In the first order approximation the power of the ith beam for a plane parallel plate can be given in the form

$$\bar{P}_i \simeq I_0 v A(\mu_0)[Q_i + C_i g T]$$

where I_0 is the incident intensity, $v = ST$ (S the cross section of the incident beam) the irradiated volume and $A(\mu_0) = e^{-\mu_0 T}$ the absorption factor. C_i is the appropriate factor given in equations (1*c*)–(4*c*).

The approximate solutions for a crystal of arbitrary shape can be given in analogous form as

$$\bar{P}_i \simeq I_0 v A(\mu_0)[Q_i + C_i g \bar{T}] \tag{12}$$

where $\bar{T} = A dA^*/du_0$, $A^* = A^{-1}$ being the appropriate absorption factor.

The specific expression of equation (12) for single diffraction (with polarization included) becomes

$$\bar{P}_1 \simeq I_0 v A \left[Q_1 p_1 - g Q_1^2 p_{11}(0) A \frac{dA^*}{d\mu_0}\right]. \tag{13}$$

Apart from a difference in notation this result is identical with equation (13) of a recent paper (Zachariasen 1963).

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Extinction in Quartz

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Extinction effects in a quartz sphere have been studied with the use of Cu $K\alpha$ radiation. It is shown that extinction varies with scattering angle in accordance with the modified rather than the Darwin formula for the extinction correction.

The data indicate that the quartz specimen contained 1·5 % of material in the Dauphiné twin position, and it is suggested that it may be difficult or impossible to find quartz crystals (natural or synthetic) entirely free of such twinning.

As a by-product of the extinction study all structural parameters of quartz were obtained with greater accuracy than hitherto reported and corresponding to $R = 0{\cdot}02$.

Introduction

This investigation was undertaken to check experimentally the revised formula for the extinction correction. It was believed that quartz would be a suitable crystal for the purpose because of simple structure, great hardness and high extinction.

The study was carried out with Cu $K\alpha$ radiation and a carefully selected, seemingly flawless, natural crystal which had been ground into a nearly perfect sphere of radius $r = 0{\cdot}147$ mm, corresponding to $\mu r = 1{\cdot}35$.

In the course of the work unexpectedly large dispersion effects were observed, the intensities I_H and $I_{\bar{H}}$ differing by as much as fifty per cent for some weak reflections. For the purpose of the extinction study the dispersion effects were eliminated by neglecting the imaginary part of the dispersion corrections to the atomic scattering powers and by using the mean intensity $(I_H + I_{\bar{H}})/2$ to obtain the experimental structure factors. The observed differences $I_H - I_{\bar{H}}$ will be discussed and interpreted in the following paper.

The intensities were measured with a proportional counter to a precision of two per cent for the very weakest, one per cent or less for the strong reflections. Because of the high symmetry multiple diffraction is not uncommon, and special care was taken to avoid making the intensity measurements under such conditions.

Dauphiné twinning

According to the Dauphiné twinning law the (HKL) plane of one individual coincides with the $(HK\bar{L})$ plane of the other. Ratios $I_{HK\bar{L}}/I_{HKL}$ as low as 0·02 were observed for the pairs $10\bar{6}/106$ and $50\bar{2}/502$, and until the last stages of the investigation it was therefore believed that the specimen contained no twinned material. However, the observed structure factors for the weak components of pairs $HKL/HK\bar{L}$ were consistently larger than the calculated values. These discrepancies could not be removed by modifications of parameters, extinction or f curves; but they could be explained by the presence of a small amount of twinned material (assumed to be uniformly distributed throughout the medium for mathematical convenience).

A correction for Dauphiné twinning was accordingly applied. If I'_{HKL} and $I'_{HK\bar{L}}$ are the actually observed