Acta Cryst. (1989). A**45**, 679-686

Multiple Reflections in a Plane-Parallel Mosaic Crystal

BY R. CANNATA, S. MARTELLI AND G. MAZZONE

ENEA, Dipartimento Tecnologie Intersettoriali di Base, CRE Casaccia, Roma, Italy

AND F. SACCHETTI

Dipartimento di Fisica, Università di Perugia, Perugia, Italy

(*Received* 18 *April* 1988; *accepted* 9 *May* 1989)

Abstract

The transfer equations for multiple reflections of X-rays in a mosaic crystal have been derived from the classical theory of electromagnetism. Unless all the beams are contained in the same plane (coplanar case), it is shown that: (i) for a generic polarization of the incident beam the process of diffraction mixes the σ and π components of the electric field in such a way that the transfer equations become non-linear; (ii) in the case of unpolarized incident radiation the transfer equations can be linearized if the electric field of each beam is split into two perpendicular components so that in the n-beam case a system of $2n$ linear differential equations has to be solved. For neutrons (and X-rays in the coplanar case) reasonably simple solutions for the three- and four-beam cases have been derived. The solutions for the three-beam case are approximate but cover most of the situations which occur in practice, while the solution for the four-beam case is exact but applies to a special, although very common, case. Numerical calculations covering realistic situations have been performed in order to study quantitatively the effect of multiple reflections on both the peak and the integrated power of the primary diffracted beam.

Introduction

The intensity of the diffraction of X-rays from a mosaic crystal in the two-beam case has been discussed by Zachariasen (1945, 1963) and in more general terms by Werner (1974). Partial extensions to the multiple-beam case have been described by, among others, Moon & Shull (1964) for neutrons, Zachariasen (1965), Caticha-Ellis (1969), Prager (1971), Unangst & Melle (1975) and Chang (1982) for X-rays. A computer program which corrects X-ray integrated intensities for the effect of multiple reflections using the formulas quoted by Prager (1971) and deduced from the approximate treatment of Moon & Shull (1964) has been published by Rossmanith (1985).

All of the above multiple-beam studies suffer from at least one of the two following shortcomings: (i) the transfer equations are solved approximately; (ii) the effect of X-ray polarization is ignored altogether or dealt with incorrectly. In view of these limitations we have decided to re-examine the secondary-extinction problem for a plane-parallel mosaic crystal in the two-beam and in the many-beam case. In accordance with previous studies, no attempt has been made in this paper to introduce the effect of primary extinction. This means that the distances travelled by the n beams in each coherent domain must be much smaller than the corresponding extinction lengths.

The polarization factor

According to classical theory, the phenomenon of polarization introduces in the intensity of diffraction from the ith to the jth beam a factor $p_{ij} = \sin^2 \varphi$, where φ is the angle between the electric field $\mathbf{E}_i$ of the ith beam and the propagation vector $\mathbf{k}_j$ of the jth beam:

$$p_{ij} = |\hat{\mathbf{k}}_j \times (\hat{\mathbf{k}}_j \times \hat{\mathbf{E}}_i)|^2 = \sin^2 \varphi, \qquad (1)$$

where the notation $\hat{\mathbf{v}}$ denotes a unit vector parallel to $\mathbf{v}$.

In accordance with (1) the direction of the electric field $\mathbf{E}_{ij}$ of the radiation diffracted along $\hat{\mathbf{k}}_j$ is given by:

$$\hat{\mathbf{E}}_{ij} = -\hat{\mathbf{k}}_j \times (\hat{\mathbf{k}}_j \times \hat{\mathbf{E}}_i)/|\hat{\mathbf{k}}_j \times (\hat{\mathbf{k}}_j \times \hat{\mathbf{E}}_i)| \qquad (2)$$

and the total electric field of the jth beam is written as

$$\mathbf{E}_j = \sum_i \mathbf{E}_{ij}, \qquad (3)$$

where the term with $i = j$ contains the contributions arising both from transmission and from forward diffraction.

Of course, since the incident electromagnetic field is a stochastic quantity, in order to derive from (1) and (3) the average value of the energy flowing along $\hat{\mathbf{k}}_j$, suitable ensemble averages have to be performed. To this purpose it must be borne in mind that, although the n-beam interaction takes place within each crystallite, all processes occurring within a single block are described in the present model by means of kinematical theory. In addition, in each layer of blocks and for each angular position of the crystal, the domains contributing significantly to the process

0108-7673/89/100679-08$03.00

of scattering are distributed at random. As such, in a mosaic crystal the transfer equations are written for the intensities using reflectivities calculated according to kinematical theory. A further point concerns the phase difference between the two mutually perpendicular components ($\mathbf{E}_{0\sigma}$ and $\mathbf{E}_{0\pi}$) of the incident electromagnetic field. The correlation between these two components has a direct bearing upon the scattered intensities since the polarization factors have to be calculated, in whatever model, from the polarization vector of the electromagnetic fields. In the case of spontaneous emission of radiation (line spectrum of an X-ray tube) and if the observation is performed on a time scale which comprises a large number of elementary processes of photon emission, one has

$$\langle E_{0\sigma} E^*_{0\pi} \rangle = 0, \tag{4}$$

where $E_{0\sigma}$ and $E_{0\pi}$ are the complex amplitudes of $\mathbf{E}_{0\sigma}$ and $\mathbf{E}_{0\pi}$ and the angular brackets denote ensemble-average values. It is important to realize that (4) is not an inherent property of electromagnetic radiation so that in the case of synchrotron radiation or the white spectrum of an X-ray tube the correlation between the two components of the electromagnetic field has to be taken into account. Actually these correlations are the elements of the density matrix of the photon field or, in classical language, they are directly related to the Stokes parameters of the field (Shurcliff, 1962).

In order to obtain further information from (1), (2) and (3), consider one of the n beams. As this beam travels a distance dt within the crystal, its electric field apart from absorption is subjected to a number of infinitesimal changes due to scattering to and from the other beams. The directions of the electric fields of all these infinitesimal changes are different and, in general, they are also different from the direction of the electric field of the beam as a whole. As a result, the state of polarization of each beam after travelling a distance dt within the crystal suffers an infinitesimal change and all the polarization factors become a function of distance. An explicit expression for the instantaneous value of p_{ij} may be obtained from (1) if we set

$$\hat{k}_j = l_j \hat{\mathbf{x}} + m_j \hat{\mathbf{y}} + n_j \hat{\mathbf{z}}, \tag{5a}$$

$$\mathbf{E}_i = E_{iy} \hat{\mathbf{y}} + E_{iz} \hat{\mathbf{z}}. \tag{5b}$$

Then,

$$p_{ij} = 1 - (m_j E_{iy} + n_j E_{iz})^2 / |\mathbf{E}_i|^2. \tag{6}$$

Equation (6) means that since the polarization factor depends separately upon E_{iy} and E_{iz} each of these quantities has to be taken as an independent variable. Moreover, the transfer equations are not linear.

Referring from now on to ensemble average values (for which we drop the angular brackets), if (4) holds for $\mathbf{E}_i$, we have

$$p_{ij} = 1 - (m_j^2 E_{iy}^2 + n_j^2 E_{iz}^2) / |\mathbf{E}_i|^2 \tag{7}$$

so that with E_{iy}^2 and E_{iz}^2 $(i = 1, 2, \ldots, n)$ chosen as independent variables, it is possible to write a set of linear differential equations.

Although the above argument is perfectly general, when all the beams are contained in the same plane (coplanar case) a suitable choice of coordinates allows the transfer equation to be linearized, irrespective of whether (4) holds or not. We will work out explicitly the formulas for the two-beam case where this choice can always be made.

Defining

$$\hat{\mathbf{k}}_0 = \hat{\mathbf{x}}, \quad \hat{\mathbf{k}}_1 = \cos 2\theta \hat{\mathbf{x}} + \sin 2\theta \hat{\mathbf{y}}, \quad \mathbf{E}_0 = E_{0y} \hat{\mathbf{y}} + E_{0z} \hat{\mathbf{z}},$$

where 2θ is the angle between $\hat{\mathbf{k}}_0$ and $\hat{\mathbf{k}}_1$, we have for a plane-parallel crystal in symmetrical transmission or reflection:

$$\mathrm{d}|\mathbf{E}_0|^2/\mathrm{d}t = -\mu_0 |\mathbf{E}_0|^2 - p_{01} r_{01} |\mathbf{E}_0|^2 + p_{i0} r_{i0} |\mathbf{E}_1|^2, \tag{8a}$$

$$\pm \mathrm{d}|\mathbf{E}_1|^2/\mathrm{d}t = -\mu_0 |\mathbf{E}_1|^2 - p_{i0} r_{i0} |\mathbf{E}_1|^2 + p_{01} r_{01} |\mathbf{E}_0|^2, \tag{8b}$$

$$p_{01} = 1 - E_{0y}^2 \sin^2 2\theta / |\mathbf{E}_0|^2, \tag{9a}$$

$$p_{10} = 1 - E_{1x}^2 / |\mathbf{E}_1|^2, \tag{9b}$$

where μ_0 is the linear absorption coefficient. The upper sign in (8b) refers to symmetrical transmission and the lower sign to symmetrical reflection. For centrosymmetric crystals, $r_{10} = r_{01} = r$, so that defining

$$I_{0\sigma} = cE_{0y}^2, \quad I_{0\pi} = cE_{0z}^2, \quad I_{1\sigma} \cos^2 2\theta = cE_{1y}^2,$$

$$I_{1\sigma} \sin^2 2\theta = cE_{1x}^2, \quad I_{1\pi} = cE_{1z}^2$$

(where c is a constant which we need not detail here) and substituting in (8a,b) and (9a,b), we have

$$\begin{aligned} \mathrm{d}(I_{0\sigma} + I_{0\pi})/\mathrm{d}t = &-\mu_0 (I_{0\sigma} + I_{0\pi}) \\ &- r(I_{0\sigma} \cos^2 2\theta + I_{0\pi}) \\ &+ r(I_{1\sigma} \cos^2 2\theta + I_{1\pi}) \end{aligned} \tag{10a}$$

$$\begin{aligned} \pm \mathrm{d}(I_{1\sigma} + I_{1\pi})/\mathrm{d}t = &-\mu_0 (I_{1\sigma} + I_{1\pi}) \\ &- r(I_{1\sigma} \cos^2 2\theta + I_{1\pi}) \\ &+ r(I_{0\sigma} \cos^2 2\theta + I_{0\pi}). \end{aligned} \tag{10b}$$

The system (10a), (10b) can be split into two systems, one for the parallel component (σ) and the other for the perpendicular component (π). For the σ component we can write

$$\mathrm{d}I_{0\sigma}/\mathrm{d}t = -(\mu_0 + r \cos^2 2\theta) I_{0\sigma} + r \cos^2 2\theta I_{1\sigma} \tag{11a}$$

$$\pm \mathrm{d}I_{1\sigma}/\mathrm{d}t = -(\mu_0 + r \cos^2 2\theta) I_{1\sigma} + r \cos^2 2\theta I_{0\sigma}, \tag{11b}$$

while for the other component we have the same equations without the factor $\cos^2 2\theta$. This factor, then, can be interpreted as the polarization factor of the σ component, according to the well known elementary treatment of X-ray polarization (Warren, 1969). The system (11a), (11b) has the same structure as the system of equations originally written by Zachariasen (1945). Setting $t = x/\gamma$, where γ is the modulus of the direction cosine of the incident beam relative to

the normal of the crystal plate, we have in symmetrical transmission the following solution for the diffracted beam:

$$I_{1\sigma}(x) = I_{0\sigma}(0)\exp(-\mu_0 x/\gamma)\exp(-r\cos^2 2\theta x/\gamma) \times \sinh(r\cos^2 2\theta x/\gamma) \quad (12a)$$

$$I_{1\pi}(x) = I_{0\pi}(0)\exp(-\mu_0 x/\gamma)\exp(-rx/\gamma) \times \sinh(rx/\gamma). \quad (12b)$$

If we set $I_{0\sigma}(0) = I_0/(1+K)$ and $I_{0\pi}(0) = KI_0/(1+K)$, where K is the usual parameter defining the degree of polarization of the incident beam and I_0 is its power at $x=0$, adding (12a) to (12b) we obtain

$$I_1(x) = (1+K)^{-1} I_0 \exp(-\mu_0 x/\gamma) \times [\exp(-rx/\gamma)\sinh(rx/\gamma) + K\exp(-r\cos^2 2\theta x/\gamma) \times \sinh(r\cos^2 2\theta x/\gamma)] \quad (13)$$

and, upon expansion of (13) up to second-order terms,

$$I_1(x) = (1+K)^{-1} I_0 \exp(-\mu_0 x/\gamma) \times [rx(1+K\cos^2 2\theta)/\gamma - r^2x^2(1+K\cos^4 2\theta)/\gamma^2]. \quad (14)$$

As (14) has already been reported by Zachariasen (1963), who stated explicitly that the system of differential equations has to be integrated separately for the two components, the results of the two-beam case may appear trivial. However, we note that the choice of two components, one parallel and the other perpendicular to the plane of diffraction, allows one to linearize and decouple the transfer equations. The physics corresponding to this choice is that the process of diffraction does not mix these two components which are effectively independent. It is clear that the same choice is not possible in the multiple-beam case unless all the beams happen to be in the same plane. Needless to say, a change of coordinate system after the first scattering process does not eliminate this difficulty. In the n-beam coplanar case, on the other hand, it is easy to show that the solution of the transfer equations may be obtained in analogy with (10a) and (10b) if one sets up two systems of n equations, one for the perpendicular component with $p_{ij}=1$, and another for the parallel component with $p_{ij}=\cos^2 2\theta_{ij}$. In order to derive the transfer equations for the many-beam case, it is convenient to describe the effect of polarization on the intensity of diffraction from a field $\mathbf{E}_0$ to a field $\mathbf{E}_1$ by means of the factor $|\hat{\mathbf{E}}_0\cdot\hat{\mathbf{E}}_1|^2$. This formulation of the effect of polarization is entirely equivalent to (1). We will specialize in what follows to the case of three beams having propagation vectors $\mathbf{k}_0$, $\mathbf{k}_1$ and $\mathbf{k}_2$. For each beam we have chosen two perpendicular components of the electromagnetic field as detailed below:

$$\hat{\mathbf{k}}_0 = \hat{\mathbf{x}};\ \hat{\mathbf{E}}_{0\sigma} = \hat{\mathbf{y}};\ \hat{\mathbf{E}}_{0\pi} = \hat{\mathbf{z}} \quad (15a)$$

$$\left.\begin{aligned}\hat{\mathbf{k}}_1 &= \cos 2\theta_{01}\hat{\mathbf{x}} + \sin 2\theta_{01}\hat{\mathbf{y}},\\ \hat{\mathbf{E}}_{1\sigma} &= -\sin 2\theta_{01}\hat{\mathbf{x}} + \cos 2\theta_{01}\hat{\mathbf{y}};\\ \hat{\mathbf{E}}_{1\pi} &= \hat{\mathbf{z}},\end{aligned}\right\} \quad (15b)$$

$$\left.\begin{aligned}\hat{\mathbf{k}}_2 &= l\hat{\mathbf{x}} + m\hat{\mathbf{y}} + n\hat{\mathbf{z}};\\ \hat{\mathbf{E}}_{2\sigma} &= -m/(m^2+l^2)^{1/2}\hat{\mathbf{x}} + l/(m^2+l^2)^{1/2}\hat{\mathbf{y}};\\ \hat{\mathbf{E}}_{2\pi} &= ln/(m^2+l^2)^{1/2}\hat{\mathbf{x}} + mn/(m^2+l^2)^{1/2}\hat{\mathbf{y}} - (m^2+l^2)^{1/2}\hat{\mathbf{z}}.\end{aligned}\right\} \quad (15c)$$

In order to simplify the algebra, $\mathbf{k}_0$ and $\mathbf{k}_1$ have been placed in the xy plane as the σ component of all beams. The π component has been chosen along the z axis for $\mathbf{E}_0$ and $\mathbf{E}_1$, but owing to the other constraints it is impossible to do the same for $\mathbf{E}_2$ as well.

As a result $\mathbf{E}_{2\pi}$ is not orthogonal to $\mathbf{E}_{0\sigma}$ and $\mathbf{E}_{1\sigma}$ so that one has mixing of the σ and π components of the three beams.

If the source of electromagnetic radiation is such that (4) holds, it is possible to neglect in the transfer equations all cross terms of the type $\langle E_{i\sigma}E^*_{i\pi}\rangle$ and to retain only the squares of $E_{i\sigma}$ and $E_{i\pi}$ ($i=0,1,2$). However, because the polarization factors $|\hat{\mathbf{E}}_{0\sigma}\cdot\hat{\mathbf{E}}_{2\pi}|^2$ and $|\hat{\mathbf{E}}_{1\sigma}\cdot\hat{\mathbf{E}}_{2\pi}|^2$ are not zero it is impossible to split the system of $2n$ linear differential equations in two systems, each consisting of n equations. The polarization factors can be calculated at once from their definition and equations (15a), (15b), (15c). Having defined a new set of reflectivities

$$r_{i\alpha j\beta} = r_{ij}|\hat{\mathbf{E}}_{i\alpha}\cdot\hat{\mathbf{E}}_{j\beta}|^2 \quad (i\neq j=0,1,2;\ \alpha,\beta=\sigma,\pi), \quad (16)$$

one has the following transfer equations in the three-beam case:

$$\left.\begin{aligned}\mathrm{d}I_{0\sigma}/\mathrm{d}t &= -(\mu_0 + r_{0\sigma1\sigma} + r_{0\sigma2\sigma} + r_{0\sigma2\pi})I_{0\sigma} + r_{1\sigma0\sigma}I_{1\sigma} + r_{2\sigma0\sigma}I_{2\sigma} + r_{2\pi0\sigma}I_{2\pi},\\ \mathrm{d}I_{0\pi}/\mathrm{d}t &= -(\mu_0 + r_{0\pi1\pi} + r_{0\pi2\pi})I_{0\pi} + r_{1\pi0\pi}I_{1\pi} + r_{2\pi0\pi}I_{2\pi},\end{aligned}\right\} \quad (17a)$$

$$\left.\begin{aligned}\pm\mathrm{d}I_{1\sigma}/\mathrm{d}t &= r_{0\sigma1\sigma}I_{0\sigma} - (\mu_0 + r_{1\sigma0\sigma} + r_{1\sigma2\sigma} + r_{1\sigma2\pi})I_{1\sigma} + r_{2\sigma1\sigma}I_{2\sigma} + r_{2\pi1\sigma}I_{2\pi},\\ \pm\mathrm{d}I_{1\pi}/\mathrm{d}t &= r_{0\pi1\pi}I_{0\pi} - (\mu_0 + r_{1\pi0\pi} + r_{1\pi2\pi})I_{1\pi} + r_{2\pi1\pi}I_{2\pi}\end{aligned}\right\} \quad (17b)$$

$$\left.\begin{aligned}\pm\mathrm{d}I_{2\sigma}/\mathrm{d}t &= r_{0\sigma2\sigma}I_{0\sigma} + r_{1\sigma2\sigma}I_{1\sigma} - (\mu_0 + r_{2\sigma0\sigma} + r_{2\sigma1\sigma})I_{2\sigma},\\ \pm\mathrm{d}I_{2\pi}/\mathrm{d}t &= r_{0\sigma2\pi}I_{0\sigma} + r_{0\pi2\pi}I_{0\pi} + r_{1\sigma2\pi}I_{1\sigma} + r_{1\pi2\pi}I_{1\pi} - (\mu_0 + r_{2\pi0\sigma} + r_{2\pi0\pi} + r_{2\pi1\sigma} + r_{2\pi1\pi})I_{2\pi},\end{aligned}\right\} \quad (17c)$$

with obvious generalization to the case of n beams. From this discussion we deduce that, even when (4) holds, the approximate solution obtained by Moon & Shull (1964) for neutrons by means of an expansion up to second order of the intensities of the n beams as a function of crystal thickness cannot be modified for X-rays with the simple addition of single- and double-diffraction polarization factors. Moreover, for X-rays one has the further problem that an expansion up to second order may not be sufficiently accurate. Unfortunately, even neglecting the problem of polarization, the exact solution of the multiple-reflection problem is of considerable difficulty unless the diffracted beams are symmetry related. If this is not the case, the most likely occurrence of multiple reflections concerns the three- and four-beam cases for which we will derive some relatively simple analytical solutions valid approximately for neutrons and in the coplanar case also for X-rays.

The three-beam case

In principle the exact solution of the transfer equations in the three-beam case is always possible; however, if one uses the formula for the roots of a cubic equation, the solution becomes awkward and difficult to handle (Mazzone, 1981). A fairly simple approximate solution may be obtained for a crystal in the shape of a plate which is large compared with the incident-beam cross section. The transfer equations at a depth x below the crystal are written as follows in the notation of Moon & Shull (1964):

$$\mathrm{d}P_0/\mathrm{d}x = -(\mu_0 + r_{01} + r_{02})P_0/\gamma_0 + r_{10}P_1/\gamma_1 + r_{20}P_2/\gamma_2, \quad (18a)$$

$$\pm\mathrm{d}P_1/\mathrm{d}x = r_{01}P_0/\gamma_0 - (\mu_0 + r_{10} + r_{12})P_1/\gamma_1 + r_{21}P_2/\gamma_2, \quad (18b)$$

$$\pm\mathrm{d}P_2/\mathrm{d}x = r_{02}P_0/\gamma_0 + r_{12}P_i/\gamma_1 - (\mu_0 + r_{20} + r_{21})P_2/\gamma_2, \quad (18c)$$

where P_0, P_1 and P_2 are the powers of the incident, primary and secondary beam, respectively, with appropriate boundary conditions. Restricting our attention to the primary beam in symmetrical transmission ($\gamma_0 = \gamma_1$), we have two cases T and R depending on whether the secondary beam is in transmission or in reflection. We define: $\mu = \mu_0/\gamma_0$, $r_1 = r_{10}/\gamma_0 = r_{01}/\gamma_0$, $r_2 = r_{20}/\gamma_0 = r_{02}/\gamma_0$, $r_3 = r_{12}/\gamma_0 = r_{21}/\gamma_0$ and $\Gamma = \gamma_0/\gamma_2$ with the understanding that in the case of X-rays the three beams have to be coplanar and that each r_i contains the appropriate polarization factor. Following Zachariasen (1965) the solution of the system may be written as $P_i = \sum_j A_{ij} \exp \lambda_j x$, where the coefficients A_{ij} are determined from the boundary conditions and the λ_j's are the roots of the secular equation. In practice the algebra is considerably simplified using Laplace transforms (Mazzone, 1981). Defining $\sigma = \sum_i r_i$ and $\tau = \sum_{i>j} r_i r_j$, we have the following secular equation:

$$\lambda^3 + \lambda^2[\sigma(1 \pm \Gamma) + r_1(1 \mp \Gamma) + \mu(2 \pm \Gamma)] + \lambda[\mu^2(1 \pm 2\Gamma) + \mu\sigma(1 \pm 3\Gamma) + \mu r_1(1 \mp \Gamma) + \tau(1 \pm 2\Gamma)] \pm \mu\Gamma(\mu^2 + 2\mu\sigma + 3\tau) = 0 \quad (19)$$

with the upper sign applying to the T case and the lower sign to the R case (this convention will be used throughout this section). The algebra of the problem is particularly simple if one root of (19) happens to be known. For instance, in the T case, one root of (19) is $\lambda_1 = -\mu$ if one sets $\Gamma = 1$ everywhere in (18a), (18b), (18c) or, alternatively, if μ_0/Γ is substituted for μ_0 in the right-hand side of (18c). This second change, although much less drastic than the first, still leads to a cubic equation substantially different from (19). One is therefore led to look for a minor modification of (19) which allows one to determine immediately one of its roots. Writing (19) in terms of a new variable $\alpha = \lambda + \mu$, we have

$$\alpha^3 + \alpha^2[2\sigma - (1 \mp \Gamma)(\mu + \sigma - r_1)] + \alpha[\tau(1 \pm 2\Gamma) - \mu(1 \mp \Gamma)(\sigma + r_1)] = \mu\tau(1 \mp \Gamma). \quad (20)$$

If the quantity on the right-hand side of (20) was zero, it is clear that one solution of (19) would be $\lambda_1 = -\mu$. For this to happen we have to modify the last term of (19) as follows:

$$\mu\Gamma(\mu^2 + 2\mu\sigma + 3\tau) \rightarrow \mu\Gamma(\mu^2 + 2\mu\sigma + 2\tau \pm \tau/\Gamma). \quad (21)$$

Since in an actual experiment $\Gamma \geq 0{\cdot}5$, it may be shown that modification (21) is substantially negligible if the absorption coefficient is either much larger or much smaller than the crystal reflectivities. These two conditions correspond to what usually happens with X-rays ($\mu \gg r_i$) and with neutrons at the peak of the reflectivity ($\mu \ll r_i$). One root of (19) having been determined approximately, the remaining algebra becomes fairly simple. The expression for $P_1(x)$, if l is the crystal thickness and $\mathscr{P}_0$ is the power of the incident beam at $x = 0$, is

$$P_1(x) = (\mathscr{P}_0/A)\exp(-\mu x)\{B - \exp(-Cx/2) \times [B\cosh(\Delta x/2) + (1/\Delta)(BC - AD) \times \sinh(\Delta x/2)]\} \quad (22)$$

where

$$A=(\sigma+r_1)\mu(\pm\Gamma-1)+(1\pm2\Gamma)\tau,$$
$$B=(\mu r_1+\tau)(\pm\Gamma-1)+\tau(1+V\Gamma),$$
$$C=(\mu+\sigma-r_1)(\pm\Gamma-1)+2\sigma,$$
$$D=2r_1+2r_3V\Gamma,$$
$$\Delta=(C^2-4A)^{1/2},$$
$$V(\text{case } T)=0;$$
$$V(\text{case } R)=\{\tau\Delta[\exp(Cl/2)-\cosh(\Delta l/2)]+(2Ar_2-\tau C)\sinh(\Delta l/2)\}\times\{\tau\Delta\exp(Cl/2)+\Delta(A-\tau)\times\cosh(\Delta l/2)+[2A(\sigma+r_1)-C(\tau+A)]\sinh(\Delta l/2)\}^{-1}.$$

Concerning the validity of (22), we notice that the solution of the system (18) has to satisfy certain boundary conditions which in the T case are relative to $\mathscr{P}_0$, $P_1(0)$ and $P_2(0)$. Inserting these conditions in (18a), (18b), (18c), one obtains further conditions for the first (and second) derivatives of P_0, P_1 and P_2 at $x=0$. All these conditions are used to determine the coefficients A_{ij} so that, in the T case, the approximate solution (22) is exact together with its first and second derivative at $x=0$. As a consequence, the validity of (22) depends also upon the depth of penetration, becoming less and less accurate as x increases.

If $r_2=r_3$, a root of (19) is $\lambda_1=-(\mu+2r_1+r_2)$ as one can verify immediately. Consequently, (18a), (18b), (18c) can be solved exactly. The explicit expression for $P_1(x)$ in the T case is

$$P_1(x)=\mathscr{P}_0\exp(-\mu x)\{A\exp[(-2r_1-r_2)x]-\exp[(\mu-\mu\Gamma-2r_2\Gamma-r_2)x/2]\times[A\cosh(\Delta x/2)+(1/\Delta)(AB-2r_1)\times\sinh(\Delta x/2)]\},\qquad(23a)$$

where

$$A=[(\mu r_1+\tau)(\Gamma-1)-2r_1^2+r_1r_2+r_2^2]\{\mu r_2(\Gamma-1)-(2r_1+r_2)(\mu+2r_2)(\Gamma-1)-2(r_1-r_2)]\}^{-1},$$
$$B=(\mu+2r_2)(\Gamma-1)-4r_1+r_2,$$
$$\Delta=\{[(\mu+2r_2)(\Gamma-1)+3r_2]^2-4\mu r_2(\Gamma-1)\}^{1/2}.$$

In the R case,

$$P_1(x)=(\mathscr{P}_0/2)\exp(-\mu x)\{-\exp[-(2r_1+r_2)x]+\exp[(\Gamma\mu+2\Gamma r_2-r_2)x/2][\cosh(\Delta x/2)+(1/\Delta)(4\Gamma r_2V-r_2-2\Gamma r_2-\mu-\Gamma\mu)\times\sinh(\Delta x/2)]\},\qquad(23b)$$

where

$$\Delta=\{[\mu(1-\Gamma)+r_2(1-2\Gamma)]^2+4\Gamma\mu(\mu+3r_2)\}^{1/2},$$
$$V=[2r_2\tanh(\Delta l/2)]/\{\Delta+[\mu(1+\Gamma)+r_2(1+2\Gamma)]\tanh(\Delta l/2)\}.$$

In the case of neutrons where the reflectivities are weakly angle dependent, the condition $r_2\sim r_3$ occurs rather frequently so that the use of the solutions (23) may be particularly appropriate. A final remark concerns the possibility that, because of symmetry, more than three reciprocal-lattice points (r.l.p.'s) are simultaneously, but not accidentally, on the sphere of reflection. We have considered the situation in which four r.l.p.'s are simultaneously excited with the geometric conditions that $|\mathbf{k}_0-\mathbf{k}_3|=|\mathbf{k}_1-\mathbf{k}_2|$ and $|\mathbf{k}_0-\mathbf{k}_2|=|\mathbf{k}_1-\mathbf{k}_3|$, where the $\mathbf{k}$'s are the propagation vectors of the four beams. This situation is particularly frequent in actual practice, as evidenced by experimental papers on multiple reflections. The exact solution of this case can be obtained fairly easily if the symmetry of the problem is used to split the quartic secular equation into the product of two quadratics. We quote the result for $P_1(x)$ when all beams are in transmission so that the following relations apply: $\gamma_0=\gamma_1$; $\gamma_2=\gamma_3$; $r_{10}=r_{01}=r_1$; $r_{20}=r_{02}=r_{13}=r_{31}=r_2$; $r_{30}=r_{03}=r_{21}=r_{12}=r_3$; $r_{32}=r_{23}=r_4$.

$$P_1(x)=(\mathscr{P}_0/2)\exp(-Ax/2)(\cosh(\Delta_1x/2)+[(\Gamma-1)(\mu+r_2+r_3)](1/\Delta_1)\times\sinh(\Delta_1x/2)-\exp(-r_1x-\Gamma r_4x)\times\{\cosh(\Delta_2x/2)+[(\Gamma-1)(\mu+r_2+r_3)+2(\Gamma r_4-r_1)](1/\Delta_2)\sinh(\Delta_2x/2)\}),\qquad(24)$$

where

$$A=(\Gamma+1)(\mu+r_2+r_3),$$
$$B=\Gamma[\mu^2+2\mu(r_2+r_3)],$$
$$C=A+2(r_1+\Gamma r_4),$$
$$D=\Gamma(\mu^2+2\mu\sigma+2\tau+2r_1r_4+2r_2r_3),$$
$$\Delta_1=(A^2-4B)^{1/2},$$
$$\Delta_2=(C^2-4D)^{1/2}.$$

Numerical results

The equations derived in the preceding section can be used to assess the influence of a secondary reflection on the integrated and peak power of the primary reflection. The calculation of the crystal reflectivities and of the Lorentz factors can be performed along the lines indicated by Moon & Shull (1964), who have limited their study to the case when all r.l.p.'s are simultaneously on the sphere of reflection. In practice it may happen that the peak reflectivities of two or

more reflections are separated by an angular distance which is small but not negligible with respect to the mosaic distribution halfwidth. This case, which requires a simple modification of the standard formulas, will not be considered here. Since the numerical results presented in the following refer to coplanar multiple reflections it has been possible to account correctly for the effect of X-ray polarization. Because of the number of parameters appearing in (22), (23) and (24), a systematic study of these equations is clearly impossible. Rather, we have chosen to study a few realistic cases for which we have calculated the power of the primary beam according to purely kinematical two-beam and multiple-beam theory for neutrons and X-rays. For X-rays we have considered separately the two states of polarization.

The integrated power in the multiple-beam case has been obtained by numerical integration of the primary-beam power over the angular variable of an ω scan. All examples refer to situations occurring in the cubic system. The first example reported in Table 1 concerns 115 as primary reflection in symmetrical transmission and 222 as secondary reflection in the T case. If the two scattering vectors are contained in the (110) plane of the reciprocal lattice these reflections are excited simultaneously when the radius of the sphere of reflection is 2·6383 reciprocal-lattice units. This condition is satisfied using, for instance, $a_0 = 3{\cdot}673$ Å, which is typical of copper alloys, and $\lambda = 1{\cdot}3922$ Å (Cu $K\beta$). Accordingly, we have used the physical properties of copper to calculate the reflectivities of X-rays and neutrons at this wavelength. The symbol δ appearing in Table 1 corresponds to the FWHM of the mosaic distribution function, while P_1 is the peak value of the primary-beam power and R_1 is its integrated power. All values have been normalized to the incident-beam power ($\mathscr{P}_0 = 1$). Notice that, since $2\theta_{02} \sim 82°$, the parallel X-ray component of the secondary reflection is very weak. In addition, since $\theta_{02} \sim \theta_{12}$, the condition $r_2 = r_3$ is satisfied very well for neutrons and within 10% for X-rays. This case therefore allows the possibility of performing a significant check on the limits of validity of (22).

Listed in Table 1 are the numerical results of (22) with the results of (23a) added in parentheses when they differ from the former by more than ~1%. Inspection of this table shows that significant differences between the two solutions occur only for relatively thick ($\mu_0 l = 1$) crystals when modification (21) [which amounts to neglecting the quantity $|(\Gamma \mp 1)\tau/\Gamma|$ with respect to μ^2] is not irrelevant. Actually, in the case where the two solutions show the largest difference one has at the peak of the mosaic distribution function $(1-\Gamma)\tau/\Gamma \sim 0{\cdot}35\mu^2$. It is also interesting to notice that in most cases the excitation of the secondary reflections weakens the power of the primary beam and that in no case does one observe *Umweganregung*,

Table 1. *Calculated power of the* 511 *primary reflection in the presence of the* 222 *secondary reflection*

Physical properties of copper, $\lambda = 1{\cdot}3922$ Å.

	Three beams	Two beams	Kinematical
Neutrons			
	$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}01$		
P_1	$2{\cdot}54\times10^{-1}$	$4{\cdot}31\times10^{-1}$	$1{\cdot}16$
R_1	$1{\cdot}83\times10^{-3}$	$2{\cdot}54\times10^{-3}$	$5{\cdot}06\times10^{-3}$
	$\delta = 1°$ $\mu_0 l = 0{\cdot}01$		
P_1	$1{\cdot}88\times10^{-1}$	$2{\cdot}16\times10^{-1}$	$2{\cdot}89\times10^{-1}$
R_1	$3{\cdot}75\times10^{-3}$	$4{\cdot}13\times10^{-3}$	$5{\cdot}06\times10^{-3}$
	$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}001$		
P_1	$1{\cdot}02\times10^{-1}$	$1{\cdot}08\times10^{-1}$	$1{\cdot}22\times10^{-1}$
R_1	$4{\cdot}71\times10^{-4}$	$4{\cdot}90\times10^{-4}$	$5{\cdot}33\times10^{-4}$
	$\delta = 1°$ $\mu_0 l = 0{\cdot}001$		
P_1	$2{\cdot}91\times10^{-2}$	$2{\cdot}95\times10^{-2}$	$3{\cdot}05\times10^{-2}$
R_1	$5{\cdot}16\times10^{-4}$	$5{\cdot}22\times10^{-4}$	$5{\cdot}33\times10^{-4}$
X-rays parallel			
	$\delta = 0{\cdot}25°$ $\mu_0 l = 1$		
P_1	$1{\cdot}21\times10^{-3}$	$1{\cdot}30\times10^{-3}$	$2{\cdot}67\times10^{-3}$
R_1	$6{\cdot}78\times10^{-6}$	$7{\cdot}06\times10^{-6}$	$1{\cdot}17\times10^{-5}$
	$\delta = 1°$ $\mu_0 l = 1$		
P_1	$5{\cdot}41\times10^{-4}$	$5{\cdot}46\times10^{-4}$	$6{\cdot}69\times10^{-4}$
R_1	$1{\cdot}01\times10^{-5}$	$1{\cdot}02\times10^{-5}$	$1{\cdot}17\times10^{-5}$
	$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}1$		
P_1	$4{\cdot}32\times10^{-2}$	$4{\cdot}33\times10^{-2}$	$4{\cdot}70\times10^{-1}$
R_1	$1{\cdot}94\times10^{-4}$	$1{\cdot}94\times10^{-4}$	$2{\cdot}06\times10^{-4}$
	$\delta = 1°$ $\mu_0 l = 0{\cdot}1$		
P_1	$1{\cdot}15\times10^{-2}$	$1{\cdot}15\times10^{-2}$	$1{\cdot}18\times10^{-2}$
R_1	$2{\cdot}03\times10^{-4}$	$2{\cdot}03\times10^{-4}$	$2{\cdot}06\times10^{-4}$
X-rays perpendicular			
	$\delta = 0{\cdot}25°$ $\mu_0 l = 1$		
P_1	$5{\cdot}72\times10^{-5}$ ($1{\cdot}78\times10^{-3}$)	$1{\cdot}36\times10^{-3}$	$3{\cdot}03\times10^{-3}$
R_1	$3{\cdot}49\times10^{-6}$ ($8{\cdot}58\times10^{-6}$)	$7{\cdot}57\times10^{-6}$	$1{\cdot}33\times10^{-5}$
	$\delta = 1°$ $\mu_0 l = 1$		
P_1	$5{\cdot}35\times10^{-4}$ ($5{\cdot}94\times10^{-4}$)	$6{\cdot}03\times10^{-4}$	$7{\cdot}58\times10^{-4}$
R_1	$1{\cdot}05\times10^{-5}$ ($1{\cdot}11\times10^{-5}$)	$1{\cdot}13\times10^{-5}$	$1{\cdot}33\times10^{-5}$
	$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}1$		
P_1	$4{\cdot}49\times10^{-2}$	$4{\cdot}86\times10^{-2}$	$5{\cdot}33\times10^{-2}$
R_1	$2{\cdot}07\times10^{-4}$	$2{\cdot}18\times10^{-4}$	$2{\cdot}33\times10^{-4}$
	$\delta = 1°$ $\mu_0 l = 0{\cdot}1$		
P_1	$1{\cdot}28\times10^{-2}$	$1{\cdot}30\times10^{-2}$	$1{\cdot}33\times10^{-2}$
R_1	$2{\cdot}26\times10^{-4}$	$2{\cdot}29\times10^{-4}$	$2{\cdot}33\times10^{-4}$

that is a primary-beam power higher than that calculated in the kinematical approximation. Equivalent calculations have been performed exchanging the role of the two sets of planes, that is setting 222 as primary reflection and 115 as secondary reflection in the R case. The main feature of this calculation is the systematic occurrence of *Umweganregung* for the parallel X-ray component, owing to the very low reflectivity of the primary reflection for this radiation. For neutrons and the other X-ray component, the effect of the perturbation is definitely smaller than that observed in the preceding calculation for equal crystal thicknesses. The results of (22) relative to this case are reported in Table 2. Notice that in this setting, because of the change of geometry, $(1+\Gamma)\tau/\Gamma$ is much smaller than μ^2. The accuracy of the solution (22) can be checked quite easily since for the perpendicular X-ray component, which shows the largest

Table 2. *Calculated power of the* 222 *primary reflection in the presence of the* 511 *secondary reflection*

Physical properties of copper, $\lambda = 1{\cdot}3922$ Å.

	Three beams	Two beams	Kinematical
Neutrons			
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}01$	
P_1	$1{\cdot}16\times10^{-1}$	$1{\cdot}18\times10^{-1}$	$1{\cdot}35\times10^{-1}$
R_1	$5{\cdot}32\times10^{-4}$	$5{\cdot}37\times10^{-4}$	$5{\cdot}90\times10^{-4}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}01$	
P_1	$3{\cdot}26\times10^{-2}$	$3{\cdot}26\times10^{-2}$	$3{\cdot}37\times10^{-2}$
R_1	$5{\cdot}76\times10^{-4}$	$5{\cdot}76\times10^{-4}$	$5{\cdot}90\times10^{-4}$
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}001$	
P_1	$1{\cdot}35\times10^{-2}$	$1{\cdot}35\times10^{-2}$	$1{\cdot}36\times10^{-2}$
R_1	$5{\cdot}91\times10^{-5}$	$5{\cdot}91\times10^{-5}$	$5{\cdot}97\times10^{-5}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}001$	
P_1	$3{\cdot}40\times10^{-3}$	$3{\cdot}40\times10^{-3}$	$3{\cdot}41\times10^{-3}$
R_1	$5{\cdot}96\times10^{-5}$	$5{\cdot}96\times10^{-5}$	$5{\cdot}97\times10^{-5}$
X-rays parallel			
		$\delta = 0{\cdot}25°$ $\mu_0 l = 1$	
P_1	$1{\cdot}02\times10^{-3}$	$9{\cdot}76\times10^{-4}$	$9{\cdot}80\times10^{-4}$
R_1	$4{\cdot}42\times10^{-6}$	$4{\cdot}28\times10^{-6}$	$4{\cdot}29\times10^{-6}$
		$\delta = 1°$ $\mu_0 l = 1$	
P_1	$2{\cdot}48\times10^{-4}$	$2{\cdot}45\times10^{-4}$	$2{\cdot}45\times10^{-4}$
R_1	$4{\cdot}33\times10^{-6}$	$4{\cdot}28\times10^{-6}$	$4{\cdot}29\times10^{-6}$
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}1$	
P_1	$3{\cdot}31\times10^{-4}$	$3{\cdot}23\times10^{-4}$	$3{\cdot}23\times10^{-4}$
R_1	$1{\cdot}44\times10^{-6}$	$1{\cdot}41\times10^{-6}$	$1{\cdot}41\times10^{-6}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}1$	
P_1	$8{\cdot}12\times10^{-5}$	$8{\cdot}07\times10^{-5}$	$8{\cdot}07\times10^{-5}$
R_1	$1{\cdot}42\times10^{-6}$	$1{\cdot}41\times10^{-6}$	$1{\cdot}41\times10^{-6}$
X-rays perpendicular			
		$\delta = 0{\cdot}25°$ $\mu_0 l = 1$	
P_1	$3{\cdot}51\times10^{-2}$ ($3{\cdot}72\times10^{-2}$)	$4{\cdot}26\times10^{-2}$	$5{\cdot}14\times10^{-2}$
R_1	$1{\cdot}74\times10^{-4}$ ($1{\cdot}79\times10^{-4}$)	$1{\cdot}97\times10^{-4}$	$2{\cdot}25\times10^{-4}$
		$\delta = 1°$ $\mu_0 l = 1$	
P_1	$1{\cdot}18\times10^{-2}$	$1{\cdot}23\times10^{-2}$	$1{\cdot}29\times10^{-2}$
R_1	$2{\cdot}12\times10^{-4}$	$2{\cdot}17\times10^{-4}$	$2{\cdot}25\times10^{-4}$
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}1$	
P_1	$1{\cdot}65\times10^{-2}$	$1{\cdot}66\times10^{-2}$	$1{\cdot}70\times10^{-2}$
R_1	$7{\cdot}29\times10^{-5}$	$7{\cdot}32\times10^{-5}$	$7{\cdot}42\times10^{-5}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}1$	
P_1	$4{\cdot}21\times10^{-3}$	$4{\cdot}22\times10^{-3}$	$4{\cdot}24\times10^{-3}$
R_1	$7{\cdot}38\times10^{-5}$	$7{\cdot}39\times10^{-5}$	$7{\cdot}42\times10^{-5}$

Table 3. *Calculated power of the* 200 *primary reflection in the presence of the* 311 *and* 11$\bar{1}$ *secondary reflections*

Physical properties of copper, $\lambda = 1{\cdot}7903$ Å.

	Four beams	Two beams	Kinematical
Neutrons			
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}01$	
P_1	$2{\cdot}93\times10^{-1}$	$2{\cdot}57\times10^{-1}$	$3{\cdot}63\times10^{-1}$
R_1	$1{\cdot}44\times10^{-3}$	$1{\cdot}25\times10^{-3}$	$1{\cdot}59\times10^{-3}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}01$	
P_1	$9{\cdot}46\times10^{-2}$	$8{\cdot}29\times10^{-2}$	$9{\cdot}07\times10^{-2}$
R_1	$1{\cdot}65\times10^{-3}$	$1{\cdot}49\times10^{-3}$	$1{\cdot}59\times10^{-3}$
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}001$	
P_1	$3{\cdot}81\times10^{-2}$	$3{\cdot}54\times10^{-2}$	$3{\cdot}67\times10^{-2}$
R_1	$1{\cdot}65\times10^{-4}$	$1{\cdot}56\times10^{-4}$	$1{\cdot}60\times10^{-4}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}001$	
P_1	$9{\cdot}28\times10^{-3}$	$9{\cdot}08\times10^{-3}$	$9{\cdot}17\times10^{-3}$
R_1	$1{\cdot}62\times10^{-4}$	$1{\cdot}59\times10^{-4}$	$1{\cdot}60\times10^{-4}$
X-rays parallel			
		$\delta = 0{\cdot}25°$ $\mu_0 l = 1$	
P_1	$2{\cdot}81\times10^{-2}$	$3{\cdot}17\times10^{-2}$	$3{\cdot}53\times10^{-2}$
R_1	$1{\cdot}31\times10^{-4}$	$1{\cdot}43\times10^{-4}$	$1{\cdot}55\times10^{-4}$
		$\delta = 1°$ $\mu_0 l = 1$	
P_1	$8{\cdot}27\times10^{-3}$	$8{\cdot}59\times10^{-3}$	$8{\cdot}83\times10^{-3}$
R_1	$1{\cdot}47\times10^{-4}$	$1{\cdot}52\times10^{-4}$	$1{\cdot}55\times10^{-4}$
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}1$	
P_1	$9{\cdot}79\times10^{-3}$	$9{\cdot}82\times10^{-3}$	$9{\cdot}93\times10^{-3}$
R_1	$4{\cdot}30\times10^{-5}$	$4{\cdot}31\times10^{-5}$	$4{\cdot}35\times10^{-5}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}1$	
P_1	$2{\cdot}47\times10^{-3}$	$2{\cdot}48\times10^{-3}$	$2{\cdot}48\times10^{-3}$
R_1	$4{\cdot}33\times10^{-5}$	$4{\cdot}34\times10^{-5}$	$4{\cdot}35\times10^{-5}$
X-rays perpendicular			
		$\delta = 0{\cdot}25°$ $\mu_0 l = 1$	
P_1	$6{\cdot}77\times10^{-2}$	$9{\cdot}01\times10^{-2}$	$1{\cdot}33\times10^{-1}$
R_1	$3{\cdot}65\times10^{-4}$	$4{\cdot}43\times10^{-4}$	$5{\cdot}82\times10^{-4}$
		$\delta = 1°$ $\mu_0 l = 1$	
P_1	$2{\cdot}77\times10^{-2}$	$3{\cdot}00\times10^{-2}$	$3{\cdot}33\times10^{-2}$
R_1	$5{\cdot}12\times10^{-4}$	$5{\cdot}41\times10^{-4}$	$5{\cdot}82\times10^{-4}$
		$\delta = 0{\cdot}25°$ $\mu_0 l = 0{\cdot}1$	
P_1	$3{\cdot}67\times10^{-2}$	$3{\cdot}59\times10^{-2}$	$3{\cdot}74\times10^{-2}$
R_1	$1{\cdot}62\times10^{-4}$	$1{\cdot}59\times10^{-4}$	$1{\cdot}64\times10^{-4}$
		$\delta = 1°$ $\mu_0 l = 0{\cdot}1$	
P_1	$9{\cdot}32\times10^{-3}$	$9{\cdot}26\times10^{-3}$	$9{\cdot}36\times10^{-3}$
R_1	$1{\cdot}63\times10^{-4}$	$1{\cdot}63\times10^{-4}$	$1{\cdot}64\times10^{-4}$

difference between the two-beam and three-beam results, one has $r_2 = r_3$ almost exactly. The results of (23*b*) are the same as those of (22), with only one exception reported in parentheses in Table 2. We can summarize the results of the three-beam case by saying that unless the reflectivity of the primary reflection is much weaker than the others, in general the effect of the secondary reflection is to weaken the primary beam. The magnitude of this effect depends in a complicated way upon all experimental parameters, but in addition to an increase of this effect with the crystal reflectivities there is an increase with μl.

An example of symmetry-related multiple reflections in the 110 plane of the reciprocal lattice occurs when the reflection 002 is in symmetrical transmission and the sphere of reflection has a radius of 2·031 reciprocal-lattice units. In this case, one has simultaneous excitation of the reflections 113 and 11$\bar{1}$. This condition is fulfilled using $\lambda = 1{\cdot}7903$ Å (Co $K\alpha$) and $a_0 = 3{\cdot}6361$ Å ($\sim a_0$ of copper). Both secondary beams are in transmission so that (24) applies. The results of this calculation are reported in Table 3. Inspection of this table shows that the power of the primary beam calculated with (24) can be higher or lower than the corresponding two-beam and kinematical values, in an apparently unsystematic way.

A final point which has not previously been recognized concerns the behaviour of P_1 in an ω scan. While in the symmetrical two-beam case the maximum value of P_1 is always attained at the centre of the reflection, that is when the mosaic distribution function and consequently the reflectivity is at its peak value, in the three-beam case this is not necessarily so. If the reflectivity r_{ij} is written as $Q_{ij}W(\Delta)$, where Q_{ij} is the well known crystallographic function and $W(\Delta)$ is the mosaic distribution function, we can easily evaluate $\mathrm{d}P_1/\mathrm{d}\Delta$ as

$$\frac{\mathrm{d}P_1}{\mathrm{d}\Delta} = \sum_i \frac{\partial P_i}{\partial r_i}\frac{\mathrm{d}r_i}{\mathrm{d}\Delta} = \left(\sum_i Q_i \frac{\partial P_i}{\partial r_i}\right)\frac{\mathrm{d}W}{\mathrm{d}\Delta} \qquad (25)$$

and then look for those values of Δ other than 0 or ∞ which cause the right-hand side of (25) to vanish. In practice it has been observed that there is only one such value, Δ^0, which is the root, when it exists, of a transcendental equation. As a consequence, there may be a dip at the centre of the reflection and a maximum at Δ^0. Notice that the existence of the dip is not confined to the approximate solution (22) in which case it could be a mathematical artefact.

In conclusion, we can say that the present investigation has shown that, in the case of multiple scattering of electromagnetic radiation, in general there is coupling between the two states of polarization either through the amplitudes or through the intensities. Only in the coplanar case do the two X-ray components act independently and can be decoupled. The numerical examples which have been reported are the first exact or almost exact calculations of the effect of multiple reflections in a mosaic crystal within the limits of validity of the transfer equations. These calculations show that application of the kinematical approximation to crystals having reflectivities comparable to those of copper can be grossly in error (up to a factor of two) if the crystal thickness is of the order of what is used in practice ($\mu_0 l \sim 1$ for X-rays and $\mu_0 l \sim 0{\cdot}01$ for neutrons). This discrepancy can be significantly reduced using the two-beam formulas; however, if one is interested in obtaining accurate structure factors, multiple reflections have to be taken into account. Equations (22), (23) and (24) can be very useful for this purpose, particularly in the case of neutrons.

We thank V. Contini for many helpful discussions.

References

CATICHA-ELLIS, S. (1969). *Acta Cryst.* **A25**, 666-673.
CHANG, S.-L. (1982). *Acta Cryst.* **A38**, 41-48.
MAZZONE, G. (1981). *Acta Cryst.* **A37**, 391-397.
MOON, R. M. & SHULL, C. G. (1964). *Acta Cryst.* **17**, 805-812.
PRAGER, P. R. (1971). *Acta Cryst.* **A27**, 563-569.
ROSSMANITH, E. (1985). *Z. Kristallogr.* **171**, 253-254.
SHURCLIFF, W. A. (1962). *Polarized Light.* Cambridge: Harvard Univ. Press.
UNANGST, D. & MELLE, W. (1975). *Acta Cryst.* **A31**, 234-235.
WARREN, B. E. (1969). *X-ray Diffraction.* Reading, MA: Addison Wesley.
WERNER, S. A. (1974). *J. Appl. Phys.* **45**, 3246-3254.
ZACHARIASEN, W. H. (1945). *Theory of X-ray Diffraction in Crystals.* New York: John Wiley.
ZACHARIASEN, W. H. (1963). *Acta Cryst.* **16**, 1139-1144.
ZACHARIASEN, W. H. (1965). *Acta Cryst.* **18**, 705-710.

Acta Cryst. (1989). A**45**, 686-698

Maximum-Entropy Data Restoration Using Both Real- and Fourier-Space Analysis

BY DAVID M. ANDERSON,* DAVID C. MARTIN† AND EDWIN L. THOMAS‡

Polymer Science and Engineering Department, University of Massachusetts at Amherst, Amherst, MA 01003, *USA*

(*Received* 8 *February* 1988; *accepted* 15 *May* 1989)

Abstract

An extension of the maximum-entropy (ME) data-restoration method is presented that is sensitive to periodic correlations in data. The method takes advantage of the higher signal-to-noise ratio for periodic information in Fourier space, thus enhancing statistically significant frequencies in a manner which avoids the user bias inherent in conventional Fourier filtering. This procedure incorporates concepts underlying new approaches in quantum mechanics that consider entropies in both position and momentum spaces, although the emphasis here is on data restoration rather than quantum physics. After a fast Fourier transform of the image, the phases are saved and the array of Fourier moduli are restored using the maximum-entropy criterion. A first-order continuation method is introduced that speeds convergence of the ME computation. The restored moduli together with the original phases are then Fourier inverted to yield a new image; traditional real-space ME restoration is applied to this new image completing one stage in the restoration process. In test cases with various types of added noise and in examples of normal and high-resolution electron-microscopy images, dramatic improvement can be obtained from

* Current address: Physical Chemistry 1, Chemical Center, University of Lund, PO Box 124, S-221 00 Lund, Sweden.

† Current address: Department of Materials Science and Engineering, The University of Michigan, H. H. Dow Building, Ann Arbor, MI 48109, USA.

‡ Current address: Department of Materials Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, USA.

0108-7673/89/100686-13$03.00