Dynamical theory of x-ray diffraction in flat, focusing, and distorted crystals by Abelés's matrix method

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Results of the standard dynamical theory of x-ray diffraction by Bragg planes parallel to a crystal surface are reproduced using Abelés's 2×2 matrix method for finding reflection and transmission by layered isotropic structures. In addition to giving a somewhat different insight into the problem this technique can be used to compute reflection and transmission in crystals with varying lattice spacing such as might be formed by thermal or impurity gradients between the surfaces. The method is also used to obtain reflection in point-to-point focusing x-ray monochromators made with ideal elastically curved perfect crystals.

INTRODUCTION

Abelés[1] described a 2×2 matrix method to compute reflection and transmission of light by plane layered media in which the refractive index is isotropic and varies only in the direction normal to the layers. The method can be used to find solutions for the dynamical theory of x-ray diffraction if variation of x-ray refractive index normal to any sets of Bragg planes of interest is known. This variation can be obtained from the complex x-ray structure factor, the real part of which is largely a measure of a Fourier component of electron density in a direction normal to the Bragg planes. The imaginary part is a measure of local average linear absorption coefficient. If local optical anisotropy were appreciable it would be possible to use the 4×4 matrix generalization of the Abelés method,[2,3] but this is unlikely to be necessary for x rays.

Since the refractive index for x rays within a crystal varies by a very small amount, a highly accurate closed-form solution for the matrix relating the electromagnetic field vectors on opposite sides of one cycle of the periodic structure can be obtained from a Fourier analysis of the complex refractive index or the structure factors. This greatly speeds numerical solution of the problem and makes it quite practical to consider the problem of millions of layers with slowly varying periodicity, as in a crystal with a thermal gradient or an impurity gradient.

The technique as described is not suited to the problem of x-ray diffraction by Bragg planes that are not parallel to the crystal surface. However, for small tilt angles, the solutions are known to be very nearly the same as for zero tilt. By a simple transformation, the results can also be applied to the problem of reflection and transmission by thin crystal wafers used in ideal elastically curved point focusing x-ray monochromators.[4,5]

RESUMÉ OF THE ABELÉS METHOD

If variation of optical parameters of a layered medium in the x and y directions can be neglected, field components of plane waves that are incident in the x-z plane must be of the form

$$f_j=\psi_j(z)\exp[i(k_x x-\omega t)]. \tag{1}$$

If, in addition, the complex refractive index n at frequency ω is everywhere isotropic, and if the magnetic susceptibility is negligible, Maxwell's equations reduce to the form

$$\frac{\partial}{\partial z}\begin{pmatrix}E_x\\H_y\end{pmatrix}=i\frac{\omega}{c}\begin{pmatrix}0 & 1-(1/n^2)(k_xc/\omega)^2\\ n^2 & 0\end{pmatrix}\begin{pmatrix}E_x\\H_y\end{pmatrix} \tag{2}$$

and

$$\frac{\partial}{\partial z}\begin{pmatrix}E_y\\-H_x\end{pmatrix}=i\frac{\omega}{c}\begin{pmatrix}0 & 1\\ n^2-(k_xc/\omega)^2 & 0\end{pmatrix}\begin{pmatrix}E_y\\-H_x\end{pmatrix}, \tag{3}$$

where E_i and H_j are electric and magnetic field components. Note that k_xc/ω is $\cos\theta_b$, where θ_b is the angle between the wave normal in vacuum and the layer surfaces. Both equations can be written

$$\frac{\partial}{\partial z}\begin{pmatrix}\psi_1\\\psi_2\end{pmatrix}=i\frac{\omega}{c}\begin{pmatrix}0 & a^2\\ b^2 & 0\end{pmatrix}\begin{pmatrix}\psi_1\\\psi_2\end{pmatrix}$$

or

$$\frac{\partial}{\partial z}\vec{\psi}=i\frac{\omega}{c}\underline{D}\vec{\psi}. \tag{4}$$

If n varies with z, then $\underline{D}$ will be a function of z.

If n is independent of z, then the integral of Eq. (3) for a finite slab of thickness h is[6]

$$\begin{pmatrix}\psi_1\\\psi_2\end{pmatrix}_{z+h}=\begin{pmatrix}\cos qh & i(a/b)\sin qh\\ i(b/a)\sin qh & \cos qh\end{pmatrix}\begin{pmatrix}\psi_1\\\psi_2\end{pmatrix}_z,$$

where $q=ab\omega/c$, or

$$\vec{\psi}(z+h)=\underline{M}(n,h)\vec{\psi}(z). \tag{5}$$

It is useful to note that q is the same for π-, as for σ-polarized radiation.

To find the matrix $\underline{M}$ across a medium of varying refractive index, matrices for short intervals across which the variation is negligible may be multiplied together.

$$\underline{M} = \prod_{j=1}^{N} \underline{M}(n_j, h_j). \tag{6}$$

Similarly, if the matrix across one cycle of a periodic medium is $\underline{M}_1$, then the matrix across N such cycles is the Nth power of $\underline{M}_1$.

$$\underline{M}_N = \underline{M}_1^N. \tag{7}$$

COMPUTATION OF REFLECTANCE AND TRANSMITTANCE

Regardless of what media are adjacent to the layers for which $\underline{M}$ is computed, the reflectance can be defined as the ratio of the mean value of the component of Poyntings vector normal to the surface for the reflected ray to that for the incident ray. Similarly, transmittance is the ratio of the mean normal component of Poynting's vector exiting from the opposite side to that for the incident ray. By our definition of ψ [Eq. (4)], Poynting's vector along z is just the product $\psi_1\psi_2$. Using the subscripts I, R, and T to designate incident, reflected, and transmitted waves, Eq. (5) may be written

$$\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}_T = \underline{M}\left[\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}_I + \begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}_R\right]. \tag{8}$$

If the refractive index of the medium on the side of the incident and reflected ray is n_I and on the other side is n_T, and if θ_b is the angle between the plane of the surface and the incident ray when it is in vacuum (assuming no prisms in the system), then we may represent the incident and reflected rays by the vectors

$$\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}_I = \begin{pmatrix}1\\ \rho_I\end{pmatrix}, \tag{9}$$

$$\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}_R = r\begin{pmatrix}1\\ -\rho_I\end{pmatrix}, \tag{10}$$

and the transmitted ray by

$$\begin{pmatrix}\psi_1\\ \psi_2\end{pmatrix}_T = t\begin{pmatrix}1\\ \rho_T\end{pmatrix}, \tag{11}$$

wherein, for σ-polarized radiation

$$\rho_k = (n_k^2 - \cos^2\theta)^{1/2}, \tag{12}$$

and for π-polarized radiation,

$$\rho_k = n_k^2(n_k^2 - \cos^2\theta_B)^{-1/2}. \tag{13}$$

Thus, Eq. (8) becomes

$$t\begin{pmatrix}1\\ \rho_T\end{pmatrix} = \underline{M}\left[\begin{pmatrix}1\\ \rho_I\end{pmatrix} + r\begin{pmatrix}1\\ -\rho_I\end{pmatrix}\right]. \tag{14}$$

These two simultaneous linear equations may be solved for the two unknown constants r and t. The transmittance is given by the expression

$$T = |t|^2 \mathrm{Re}(\rho_T)/\mathrm{Re}(\rho_I), \tag{15}$$

where Re denotes the real part of a possibly complex number, and reflectance is

$$R = |r|^2. \tag{16}$$

$\underline{M}_1$ WHEN n VARIES SLIGHTLY

Suppose we write

$$\underline{M}_1 \approx \prod_{j=1}^{L}\left[\underline{M}\left(\bar{n}, \frac{h}{L}\right) + (\bar{n} - n_j)\frac{\partial \underline{M}}{\partial n}\left(n, \frac{h}{L}\right)\Big|_{\bar{n}}\right], \tag{17}$$

where $\bar{n}$ is some approximate average value of refractive index, and $\underline{M}$ is defined by Eq. (5). The first two terms in the binomial expansion for M_1 are

$$\underline{M}_1 \approx \underline{M}^L\left(\bar{n}, \frac{h}{L}\right) + \sum_{j=1}^{L}(n - n_j)\underline{M}^{j-1}\left(\bar{n}, \frac{h}{L}\right)\frac{\partial M(n, h/L)}{\partial n}\Big|_{\bar{n}}\underline{M}^{L-j}\left(\bar{n}, \frac{h}{L}\right),$$

As L approaches infinity, we get

$$\underline{M}_1 \approx \underline{M}(\bar{n}, h) + \int_0^h [n(z) - \bar{n}]\,\underline{M}(\bar{n}, z)\frac{\partial^2 \underline{M}(n, z)}{\partial z\,\partial n}\Big|_{\bar{n}, 0}\underline{M}(\bar{n}, h - z)\,dz. \tag{18}$$

It is easy to show from Eq. (5) that

$$\frac{\partial^2 \underline{M}(n, z)}{\partial z\,\partial n}\Big|_{\bar{n}, 0} = \frac{\partial q}{\partial n}\begin{pmatrix}0 & i(a/b)\\ i(b/a) & 0\end{pmatrix} + q\left(\frac{(\partial a/\partial n)b - a(\partial b/\partial n)}{ab}\right)\begin{pmatrix}0 & i(a/b)\\ -i(b/a) & 0\end{pmatrix} \equiv f^+\underline{Q}^+ + f^-\underline{Q}^-. \tag{19}$$

Furthermore, $\underline{Q}^+$ commutes with $\underline{M}$, while $\underline{Q}^-$ does not. Hence the integral

$$\underline{S}^+ = \int_0^h [n(z)-\bar{n}]\underline{M}(\bar{n},z)f^+\underline{Q}^+\underline{M}(\bar{n},h-z)\,dz$$
$$= \underline{Q}^+\underline{M}(\bar{n},h)f^+\int_0^h [n(z)-\bar{n}]\,dz \qquad (20)$$

is just a correction to be added to $\underline{M}(\bar{n},h)$ in case the value chosen for $\bar{n}$ is not exactly the mean refractive index. It is the term

$$\underline{S}^- = f^-\int_0^h [n(z)-\bar{n}]\underline{M}(\bar{n},z)\underline{Q}^-\underline{M}(\bar{n},h-z)\,dz \qquad (21)$$

that leads to Bragg reflection and other phenomena that result from the variation of n.

Direct matrix multiplication shows that

$$\underline{Q}^+\underline{M}(\bar{n},h) = \begin{pmatrix} -\sin qh & i(a/b)\cos qh \\ i(b/a)\cos qh & -\sin qh \end{pmatrix} \qquad (22)$$

and

$$\underline{M}(\bar{n},z)\underline{Q}^-\underline{M}(\bar{n},h-z) = \begin{pmatrix} \sin q(2z-h) & i(a/b)\cos q(2z-h) \\ -i(b/a)\cos q(2z-h) & -\sin q(2z-h) \end{pmatrix}. \qquad (23)$$

If we make the Fourier expansion

$$n(z)-\bar{n} = c_0 + \sum_{j=1}^{\infty} c_j\cos\frac{2j\pi z}{h} + s_j\sin\frac{2j\pi z}{h}, \qquad (24)$$

(where all constants are generally complex numbers), and insert this and (23) into (21) and integrate, we get

$$\underline{S}^- = \frac{f^- qh\sin qh}{2}\begin{pmatrix} -A & -i(a/b)B \\ i(b/a)B & A \end{pmatrix},$$

where

$$A = \sum_{j=1}^{\infty} s_j\left(\frac{1}{j\pi-qh}+\frac{1}{j\pi+qh}\right) \qquad (25)$$

and

$$B = \sum_{j=0}^{\infty} c_j\left(\frac{1}{j\pi-qh}-\frac{1}{j\pi+qh}\right).$$

The factor $\sin qh$ prevents divergence of the matrix $\underline{S}^-$ when $qh = \pm j\pi$. Bragg reflection occurs in the neighborhood of these values of qh.

REFRACTIVE INDEX n FOR X RAYS

The refractive index n for x rays is a complex number whose imaginary part n_i is small and positive and whose real part n_r usually differs from unity by a small negative amount. At x-ray wavelengths shorter than and not too near the characteristic absorption wavelengths of atoms in the medium, the average value of the real part of n is[7]

$$\bar{n}_r = 1-\lambda^2 r_e\bar{\rho}_e/2\pi\,, \qquad (26)$$

where $r_e = e^2/m_ec^2$ is the classical electron radius and $\bar{\rho}_e$ is the average number of electrons per unit volume in the medium. If the electron density is Fourier analyzed in a direction normal to the surfaces, then

$$n(z) = 1-\frac{\lambda^2 r_e}{2\pi V}\left[F_0 + 2\sum_{j=1}^{\infty}\left(F_{cj}\cos\frac{2j\pi z}{h} + F_{sj}\sin\frac{2j\pi z}{h}\right)\right], \qquad (27)$$

where V is the volume of a unit cell and F_0 is the number of electrons in a unit cell. F_{cj} and F_{sj} are cosine and sine components of the structure factor of the crystal planes parallel to the surfaces for Bragg reflection in jth order, when the Bragg spacing of first order is h. The structure factor for a set of Bragg planes with Miller indices jh, jk, and jl is usually defined as

$$F = (F_{cj}^2+F_{sj}^2)^{1/2}\,. \qquad (28)$$

Comparison of Eqs. (27), (28), and (24) shows the relation between structure factors and the Fourier components of refractive index. The 2 before the sum in Eq. (27) but not in Eq. (23) is a matter of convention.

Since much of the x-ray absorption in a crystal occurs near atomic nuclei, the imaginary part of n also has Fourier components. These can be incorporated into the structure factor components by making F_{cj} and F_{sj} complex. The average value of the imaginary part of n is

$$\bar{n}_i = \mu_e\lambda/4\pi\,, \qquad (29)$$

where μ_e is the linear absorption coefficient of the medium in directions where there is no Bragg reflection or anomalous transmission. This part can easily be incorporated into a negative imaginary component of F_0.

When x-ray photon energy is near an absorption band in the crystal, a more rigorous treatment of the relation between $n(z)$ and the atomic structure of the crystal is necessary.[7]

COMPUTATIONAL SHORTCUTS

The computation of $\underline{M}^N$ can be done with a minimum number of matrix multiplications by repeated squaring of the matrix in the following way. Suppose there were 23 layers. Note that

$$M^{23} = M\times M^2\times M^{2^2}\times M^{2^4}$$
$$= M\times M^2\times(M^2)^2\times\{[(M^2)^2]^2\}^2\,. \qquad (30)$$

By squaring M, squaring the square, and squaring that twice more, followed by three more multiplications, we obtain M^{23} in seven rather than 22 multiplications. The saving is much more impres-

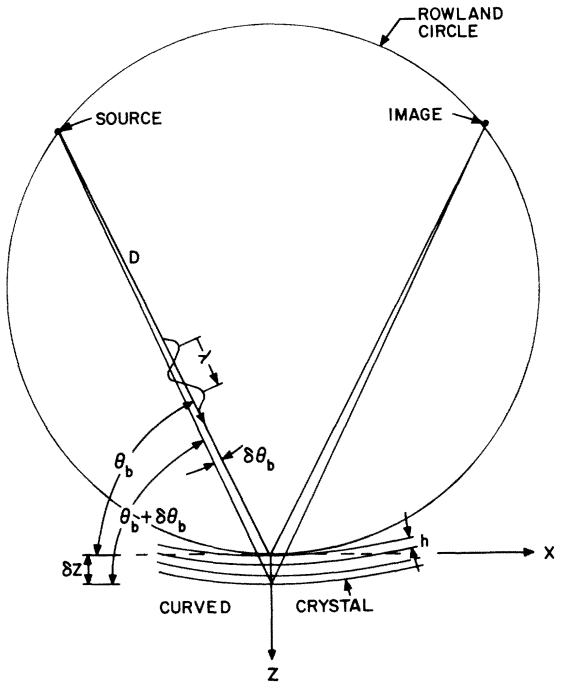

FIG. 1. Focusing curved crystal monochromator showing symbols used in formulas.

sive when several million identical layers are involved, as is usual in crystal diffraction problems.

If the Bragg spacing changes slowly, results can be made practically the same whether $\underline{M}_1$ for each cycle is made different or whether the crystal is divided into several zones, each zone being approximated by some number m of identical cycles. A suitable criterion for choosing m for one zone is to require that no Bragg plane within the zone differs in position from the correct position by more than one or two percent of one Bragg spacing h. By averaging the spacing in one zone to be approximated, the outermost layers of a perfectly periodic approximate zone can be set exactly in register with those of the approximated zone. It usually saves computation time if m is made equal to the largest integer power of two that is larger than the minimum value of m, even though this may require up to twice the number of zones required by a particular phase error limitation.

APPLICATION TO CURVED CRYSTAL MONOCHROMATORS

A crystalline wafer that is elastically bent to the proper curvature may reflect x rays in one plane from one point to another point. In a symmetrical system the curved Bragg planes near the center of the wafer are parallel to the surface. θ_b, the complement of the angle of incidence upon the Bragg planes near the top surface is slightly smaller than that deeper into the crystal (see Fig. 1). The effect on Bragg reflection for monochromatic rays diverging from the source is equivalent to the effect of a flat crystal with linearly varying Bragg spacing on monochromatic plane waves. The equivalent linear variation in Bragg spacing with depth for a flat crystal can be found by differentiating Bragg's law

$$j\lambda = 2h \sin\theta_b \ , \tag{31}$$

and inserting the variation of Bragg angle with depth in the curved crystal, as follows.

To obtain equivalent variation by the changing either h alone or θ_b alone on the right side of Bragg's law, we must have

$$\delta h \sin\theta_b = h \cos\theta_b \delta\theta_b \ . \tag{32}$$

If z is measured from the first surface into the crystal, and if the source is at distance D from the first surface of the curved crystal on the Rowland circle of foci, then the change in angle θ_b with depth in the crystal is

$$\delta\theta_b = \cos\theta_b \delta z / D \tag{33}$$

(see Fig. 1). Setting $\delta z = h$ and combining Eqs. (32) and (33), we get

$$\delta h/h = h \cos^2\theta_b/(D \sin\theta_b). \tag{34}$$

Figure 2, using the abscissa scale at the bottom, shows the computed Darwin reflection band at fixed angle as a function of wavelength for plane waves around the middle of the Cu $K\alpha_1$ line incident on a thick flat quartz crystal with the (502) planes parallel to the surface. The larger band is

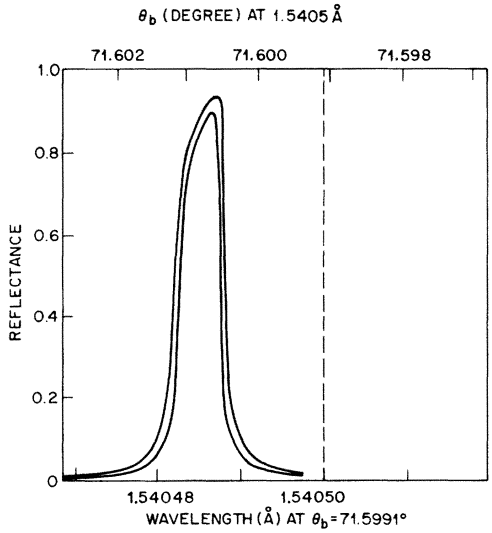

FIG. 2. Darwin reflection band of ideal thick, unstrained quartz from (502) planes in the neighborhood of the Cu $K\alpha_1$ line, as a function of wavelength at fixed angle, and as a function of angle at fixed wavelength.

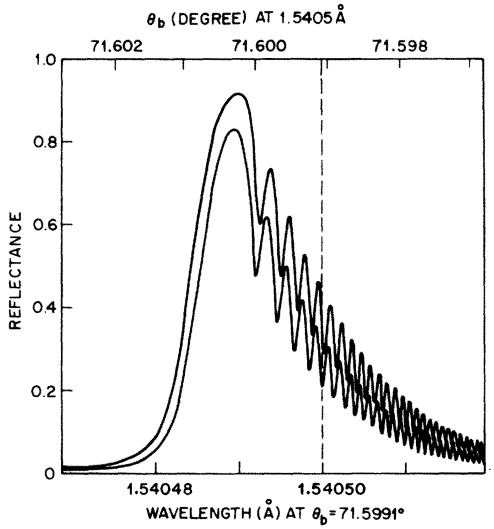

FIG. 3. Darwin band of a quartz wafer as in Fig. 2, bent to an arm length D of 1 m.

for σ-polarized and the smaller for π-polarized radiation. We set the incident (Bragg) angle in vacuum θ_b at 71.5991°. The terms in Eq. (24) were $\bar{n} = 1 - 10^{-7}\,(84.4 - 1.1 \times i)$ and $c_1 = 10^{-7}\,(17.0 - 1.0 \times i)$. All other values of c and s were set equal to zero. Only c_0 would affect the results appreciably anyway.

The full width at half-maximum intensity of the Cu $K\alpha_1$ line is about 0.000 47 Å,[8] which is about nine times the whole width of the plot in Fig. 2.

The scale at the top shows the reflection of monochromatic radiation of wavelength 1.5405 Å as a function of *angle*. The Bragg spacing h is 0.811 754 Å. These curves agree with the results of the standard dynamical theory.[7] The wafer is about 4×10^6 layers thick. This is a sufficiently large number that reflectance does not change appreciably with that number or with backing material. Figure 3, which was computed by the new method, shows the reflection band for a similar crystal wherein the Bragg spacing increases by a factor of 8.852×10^{-12} in each successive layer. According to Eq. (34), this is equivalent to the effect of curving the crystal planes to a radius of 105.6 cm and using point-to-point focusing with a monochromator arm length $D = 100$ cm. These Bragg planes of quartz and dimensions were used in the actual construction of a doubly curved crystal-point focusing monochromator for Cu $K\alpha_1$ radiation.[4,5]

The highest peak on the reflection band is almost as high and narrow as the Darwin band for a flat crystal in this case. It becomes lower and broader if the arm D is shorter, so that insufficient planes reflect coherently to yield nearly complete extinction. The decaying oscillating curves on the right are Bragg reflection from planes that are properly oriented below the surface, but the beam is attenuated by absorption. About half the beam is directed back and half transmitted in the interaction inside the crystal for values of strain as small, or D as large as this, as was postulated in an earlier investigation.[4] The oscillations are a complicated interference effect that depends on the amount of strain and the Darwin bandwidth. They were not anticipated and are too narrow for easy direct observation. They are not an artifact of the computational method, however, as we have shown by varying the number m of identical cycles in the computations without altering the outcome. The area under the reflection curve yields a value for reflected intensity that is consistent with rough experimental measurements,[4] but no attempt has been made to observe the curves directly.

If strain increases further, the peak reflectance falls but the exponential tail grows broader so that total reflection remains roughly the same. It is interesting to note that when the arm D is short enough that reflection is nowhere larger than about 50%, primary extinction is small enough that the theory for mosaic crystals gives a good estimate of reflected intensity. Crystal planes with smaller structure factors have narrower Darwin bands, and the effects of strain on reflection appear with less bending or variation in spacing of planes. Conversely, planes with large structure factors, such as the (011) planes of quartz, show almost unaltered main peaks and much shorter tails under similar degrees of strain at the same x-ray wavelength. When linear absorption is large, as with Al $K\alpha$ radiation with the (100) planes in quartz, the beam never gets deep enough into the crystal to sample the variations due to strain. The reflection band remains an unaltered Gaussian at very large strain levels.

We plan to submit for publication elsewhere, the results of computations for a number of different point focusing monochromator designs that will illustrate these effects in more detail.

[1]F. Abelés, Ann. Phys. (Paris) 5, 596 (1950).
[2]D. O. Smith, Opt. Acta 12, 13 (1965).
[3]D. W. Berreman, J. Opt. Soc. Am. 62, 502 (1972).
[4]D. W. Berreman, thesis (California Institute of Technology, 1955) (unpublished).
[5]D. W. Berreman, Rev. Sci. Instrum. 26, 1048 (1955).
[6]See, for example, Ref. 1 p. 629; or Ref. 3, Eqs. (4), (5), (21), (23), and (24).
[7]See, for example, W. H. Zachariasen, *Theory of X-Ray Diffraction in Crystals* (Dover, New York, 1967) pp. 95ff and 114ff.
[8]L. G. Parratt, Phys. Rev. 46, 749 (1934).