Table 8. *Intermolecular contacts (less than* 3·6 Å)

Atoms	Distance	Atoms	Distance
Q–O2 (I)	3·343	*G*–*H* (III)	3·557
O1–*E* (II)	3·444	*I*–*J* (III)	3·556
C–*B* (III)	3·554	*K*–*L* (III)	3·520
E–*F* (III)	3·511	*P*–*N* (III)	3·514
F–O2 (III)	3·574	*R*–*Q* (III)	3·559

(I) refers to the molecule at $\frac{1}{2}+x, -y, -z$;
(II) $\frac{1}{2}+x, \frac{1}{2}-y, -\frac{1}{2}+z$;
(III) $x, y, 1+z$.

The mean value for the four single bonds in ring II is 1·491 Å but while pairs of bonds which are on opposite sides of the ring [*GH* and *AS*; *HI* and *AB*] have almost identical lengths, the adjacent bonds [*GH* and *HI*; *AS* and *AB*] have different values. The difference (0·016 Å) from the mean is, however, not significant. There is a significant lengthening of the aromatic bond *IJ* (1·472 Å) due to steric interference between O2 and the hydrogen attached to atom K. The three aromatic rings have the following mean bond lengths, I (1·394), III (1·405), IV (1·413).

The short length of the *c* axis means that molecules at each end of this axis are already fairly close and the inclination of the molecular plane brings certain atoms in neighbouring molecules even closer. There are some other short intermolecular distances between the molecules related by the screw axis. Those intermolecular contacts (not including hydrogen atoms) which are less than 3·6 Å are listed in Table 8.

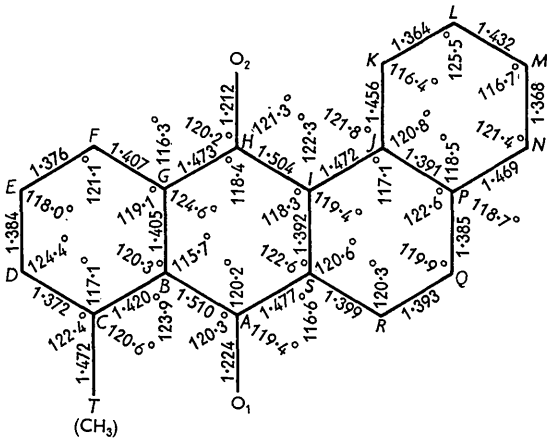

Fig. 6. Bond lengths (Å) and bond angles.

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Dynamical Diffraction Theory of Waves in Distorted Crystals. I. General Formulation and Treatment for Perfect Crystals

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A dynamical diffraction theory for distorted crystals is formulated for the Laue case based on a 'lamellar crystal' method originally given by C. G. Darwin (*Phil. Mag.* 1914, **27**, 325 and 675). In each lamellar crystal a Born approximation is assumed. Relations among two-dimensional Fourier transforms of wave functions on the successive boundaries between lamellae are obtained in terms of generalized matrix multiplication. In the two-beam case of a perfect crystal, results coincide with those of the ordinary Laue–Bethe theory.

The theory is applicable to asymmetrical cases with a large Bragg angle; namely to the general cases of X-rays and neutrons as well as electrons.

1. Introduction

Standard dynamical theories for diffraction phenomena in crystals are based on the Laue–Bethe formulation (Laue, 1931; Bethe, 1928). We can, however, treat the problem in an entirely different way which we may call 'lamellar crystal' approach. It seems

necessary to do this for the purpose of extending dynamical theories to elucidate diffraction phenomena in distorted crystals. Since the Laue–Bethe theory is based on a Bloch wave which is only permissible in perfect crystals, in the case of distorted crystals it seems to be almost inapplicable.

The idea of the lamellar crystal approach was originally given by Darwin (1914*a*, *b*) in the simple Bragg case. Recently Cowley & Moodie (1957, 1958, 1959) have presented a more extensive theory along this line for the Laue case. A short description of a similar theory was also given by Sturkey (1957).

Since, however, Cowley & Moodie used a Huyghen's principle and a couple of *ad hoc* assumptions, their theory appears to be applicable only to cases of small scattering angle such as electron diffraction problems. In the present paper the author intends to show that a lamellar crystal theory can be established on a more satisfactory base and the above-mentioned assumptions can be avoided. This opens the way for applying the present formalism also to X-ray and neutron cases.*

2. General formulation

(*a*) *Scattering by a crystal slice*

As illustrated in Fig. 1, we divide a bulk crystal into parallel-sided slices, their thickness being of a few unit cells. First we consider the scattering process in a crystal slice. The front and rear surfaces are specified by suffixes α and β respectively. A wave function on the α-surface is written in terms of its Fourier transform as follows, taking x- and y-axes in the α-surface:

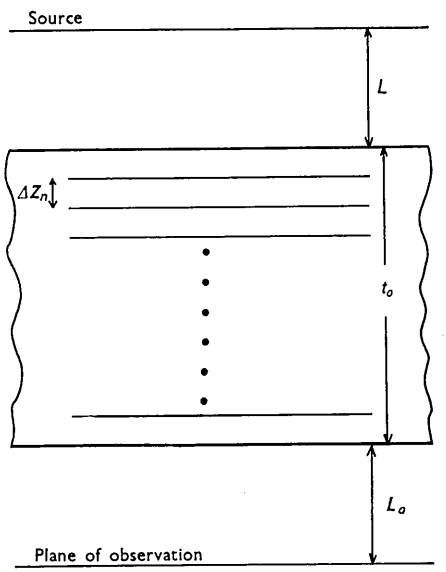

Fig. 1. Illustration of dividing a bulk crystal into parallel slices and the geometrical relation among a wave source, the crystal and a plane of observation.

$$f_\alpha(x, y)=(1/2\pi)\int F_\alpha(\xi, \eta)\exp i(\xi x+\eta y)d\xi d\eta.\dagger \quad (1)$$

The axes ξ and η are reciprocal to the axes of x and y respectively. A similar expression will be given to a function f_β and its Fourier transform F_β.

Our first problem is to find a relation between $F_\alpha(\xi, \eta)$ and $F_\beta(\xi, \eta)$ based on a Born approximation. Since the thickness Δz of a slice is thin enough, this can be justified in usual cases. For simplicity, we consider first a scalar wave function f which satisfies Schrödinger's equation

$$\Delta f+(8\pi^2m/h^2)(E-v)f=0 \quad (2)$$

where h is Planck's constant; E and v are the total energy of an incident particle of mass m and the potential energy of a crystal respectively. A well-known integral form equivalent to equation (2) is (see, for example, Mott & Massey, 1949)

$$f(x, y, z)=f^0(x, y, z)+(1/4\pi)\times\int(\exp iKr/r)g(x', y', z')dx'dy'dz' \quad (3)$$

where

$$r^2 = (x-x')^2+(y-y')^2+(z-z')^2$$
$$g = u(x', y', z')f(x', y', z') \quad (4)$$
$$K^2 = 8\pi^2mE/h^2 \quad (5)$$
$$u = -(8\pi^2m/h^2)v(x', y', z')\,. \quad (6)$$

Moreover f^0 may express the undisturbed wave which coincides with $f_\alpha(x, y)$ at the α-surface, namely

$$f^0(x, y, z)=(1/2\pi)\int F_\alpha(\xi, \eta)\exp i(\xi x+\eta y+\zeta z)d\xi d\eta \quad (7)$$

where
$$\xi^2+\eta^2+\zeta^2=K^2. \quad (8)$$

Remembering the two-dimensional Fourier integral form of a spherical wave

$$\exp iKr/r=(i/2\pi)\int\frac{\exp i\zeta(z-z')}{\zeta}\times\exp i\{\xi(x-x')+\eta(y-y')\}d\xi d\eta \quad (9)$$

we have the following Fourier transform of $f_\beta = f(x, y, \Delta z)$ from equation (3), in which f in the integrand is replaced by f^0:

$$F_\beta(\xi, \eta)=\exp i\zeta\Delta z\times\{F_\alpha(\xi, \eta)+(i/2\pi)(K/\zeta)G(\xi, \eta; \Delta z)\} \quad (10)$$

where

$$G=(1/2K)\int_0^{\Delta z}dz'\int g(x', y', z')\times\exp -i(\xi x'+\eta y'+\zeta z')dx'dy'\,. \quad (11)$$

* By entirely different approaches, Fujiwara (1959) and Fujimoto (1959) also showed that some results of Cowley & Moodie were obtained without using their approximations.

† Hereafter $\iint_{-\infty}^{+\infty}d\xi d\eta$ is abreviated to $\int d\xi d\eta$.

Since the integral with respect to x' and y' can be interpreted as 2π times the two-dimensional Fourier transform of g, it follows that

$$G=(1/2K)\int_0^{\Delta z}dz'\int F_\alpha(\xi',\eta')\times\exp i\zeta' z'.U(\xi-\xi',\eta-\eta';z')\exp -i\zeta z'\,d\xi'\,d\eta' \quad (12)$$

where

$$U(\xi,\eta;z)=(1/2\pi)\int u(x,y,z)\exp -i(\xi x+\eta y)\,dx\,dy\,. \quad (13)$$

In simple cases where u is quasi-periodic, U may be approximated by a perfectly periodic function with respect to z within a thin slice. Then U can be expanded as

$$U(\xi,\eta;z)=(2K)\sum_{g_3}Q_{\alpha\beta}(\xi,\eta;g_3)\exp 2\pi i g_3 z \quad (14)$$

where g_3 is a reciprocal lattice vector of this one-dimensional lattice.*

The suffix $\alpha\beta$ in $Q_{\alpha\beta}$ is used in order to emphasize the fact that the Fourier coefficient $Q_{\alpha\beta}$ depends on the individual slice with which we are concerned. Under this approximation

$$G=\int F_\alpha(\xi',\eta')\tilde{Q}_{\alpha\beta}(\xi-\xi',\eta-\eta')\,d\xi'\,d\eta' \quad (15)$$

where

$$\tilde{Q}_{\alpha\beta}=\sum_{g_3}Q_{\alpha\beta}(\xi-\xi',\eta-\eta',g_3)\{1-\exp -2i\varphi\Delta z\}/2i\varphi \quad (16)$$

and

$$\varphi=\tfrac{1}{2}(\zeta-\zeta'-2\pi g_3)\,. \quad (17)$$

In X-ray cases, Maxwell's equation can be reduced to

$$\Delta\mathbf{d}+K^2\mathbf{d}+\text{rot rot}\,(\chi\mathbf{d})=0 \quad (18)$$

for a displacement vector $\mathbf{d}$ and the polarizability χ of a crystal (see, for example, v. Laue, 1960). No essential difficulty arises even in vector fields to obtain a relation between Fourier transforms $\mathbf{D}_\alpha$ and $\mathbf{D}_\beta$ of wave functions on the α and β surfaces. Corresponding to equation (10) we have

$$\mathbf{D}_\beta(\xi,\eta)=\exp i\zeta\Delta z\{\mathbf{D}_\alpha(\xi,\eta)+(i/2\pi)(K/\zeta)\mathbf{G}(\xi,\eta;\Delta z)\} \quad (19)$$

where

$$\mathbf{G}(\xi,\eta)=\int\mathbf{D}_{\alpha[\xi,\eta,\zeta]}(\xi',\eta')\tilde{Q}_{\alpha\beta}(\xi-\xi',\eta-\eta')\,d\xi'\,d\eta'. \quad (20)$$

In equation (20) $\mathbf{D}_{[\xi,\eta,\zeta]}$ means a component of $\mathbf{D}$ on the plane perpendicular to a direction $[\xi,\eta,\zeta]$. The symbol $Q_{\alpha\beta}$ in $\tilde{Q}_{\alpha\beta}$ is now related to χ by

$$\chi=(1/2\pi)(2/K)\int\sum_{g_3}Q_{\alpha\beta}(\xi,\eta;g_3)\times\exp i(\xi x+\eta y+2\pi g_3 z)\,d\xi\,d\eta\,. \quad (21)$$

* In general, g_3 depends on ξ and η.

Moreover, in deriving equations (19) and (20), the direction $[\xi,\eta,\zeta+2\pi g_3]$ is approximated by $[\xi,\eta,\zeta]$, since we are concerned with such cases where the Bragg condition is almost satisfied.

Clearly equations (19) and (20) can be reduced to a set of scalar equations such as equations (10) and (15) taking into account proper factors of polarization. Therefore we consider only scalar waves in the following with no violation of generality.

(b) Scattering by the whole crystal

In order to extend the above formulation to multi-layer cases it is convenient to use a matrix notation in a symbolic sense.* In the following a bold letter $\mathbf{F}$ stands for a vector in Hilbert space, its (ξ,η)-component being $F(\xi,\eta)$. A matrix of infinite rank $\mathbf{Q}_{\alpha\beta}$ expresses the scattering process in an individual slice. Its $(\xi,\eta;\xi',\eta')$ elements are

$$\delta(\xi-\xi',\eta-\eta')+(i/2\pi)(K/\zeta)\tilde{Q}_{\alpha\beta}\,.$$

The symbol $[\exp i(\zeta\Delta z)]$ is used for a diagonal matrix whose elements are

$$\exp i(\zeta\Delta z).\delta(\xi-\xi',\eta-\eta')\,.$$

Using these notations and equation (15), equation (10) can be written as†

$$\mathbf{F}_\beta=[\exp i(\zeta\Delta z_{\alpha\beta})]\mathbf{Q}_{\alpha\beta}\mathbf{F}_\alpha\,. \quad (22)$$

The multiplication of $\mathbf{Q}_{\alpha\beta}$ and $\mathbf{F}_\alpha$ implies a two-dimensional convolution through common variables of columns and rows. Applying this relation to all the slices successively we have the following relation between Fourier transforms $F_e(\xi,\eta)$ and $F_a(\xi,\eta)$ which refer to the incident and exit surface respectively:

$$\mathbf{F}_a=\mathbf{Q}_{ea}\mathbf{F}_e \quad (23)$$

where

$$\mathbf{Q}_{ea}=[\exp i(\zeta\Delta z_N)]\mathbf{Q}_N\ldots[\exp i(\zeta\Delta z_n)]\mathbf{Q}_n\ldots\mathbf{Q}_1 \quad (24)$$

for a crystal of N-layer. The suffixes $\alpha\beta$ is now replaced by the number n specifying the order of sequence.

(c) Consideration of incident waves

The Fourier transform F_e is determined by the incident wave concerned. It is not difficult to obtain F_e for simple cases such as a plane wave, a plane wave modified by a slit system and a spherical wave (*cf.* Kato, 1961*a*).

For example, in the last case, using the present notation,

$$F_e(\xi,\eta)=i\exp i\zeta L/\zeta \quad (25)$$

where L is the perpendicular length from a point source to the incident surface (see equation (9)).

* See, for example, Schiff (1949).

† Generally the thickness (Δz) of slices must be specified by $\alpha\beta$ since their thicknesses may differ from each other.

(d) Consideration of outgoing waves

If a Fourier transform F_a on an exit surface is given, we can calculate quantities which are relevant to particular experimental conditions. As an example we consider here an ordinary diffraction experiment. In this case we need an intensity distribution at a large distance from the exit surface. The wave function at L_a from the exit surface is given by

$$f(x, y, L_a) = (1/2\pi)\int F_a(\xi, \eta) \times \exp i(\xi x+\eta y+\zeta L_a)d\xi d\eta . \quad (26)$$

If L_a is large enough, an asymptotic form of equation (26) can be obtained by a two-dimensional stationary phase method (for example, Born & Wolf, 1959). The result is

$$f(x, y, L_a) \simeq (-i\zeta)F_a(\bar{\xi}, \bar{\eta}) \exp iKR/R , \quad (27)$$

where

$$R=(x^2+y^2+L_a^2)^{\frac{1}{2}}$$

and

$$\bar{\xi}=(x/L_a)\zeta, \quad \bar{\eta}=(y/L_a)\zeta .$$

In other words the angular distribution of an outgoing wave is proportional to the angular spectrum of the Fourier transform of the wave function at an exit surface.

3. Two-beam theory of perfect crystals

(a) Non-absorbing cases

In perfect crystals $u(x, y, z)$ is a periodic function in the x- and y-directions (lateral) as well as in the z-direction (normal). Thus $Q_n(\xi-\xi', \eta-\eta', g_3)$ is a δ-type function and can take an appreciable value only at three-dimensional reciprocal lattice points $\{g_1, g_2, g_3\}$.

For a lattice point (g_1, g_2, g_3) at which the Bragg condition is satisfied the magnitude of φt_0 must be less than or comparable to 2π where φ is defined by equation (17) and $t_0=\Sigma\Delta z_n$ is the total thickness of the crystal. For thick crystals, therefore, we can safely approximate the shape function

$$\{1-\exp-2i\varphi\Delta z_n\}/2i\varphi$$

by Δz_n. For other points $\{g_1, g_2, g_3'\}$, 2φ is almost equal to $2\pi(g_3-g_3')$. As stated below we take Δz_n as a small multiple of the lattice periodicity along the z-direction in order to keep the phase factor of Q_n constant. Therefore $2\varphi\Delta z_n$ is close to an integral multiple of 2π. Thus we have

$$\tilde{Q}_n \simeq Q_n(g_1, g_2, g_3)\Delta z_n$$

in which (g_1, g_2, g_3) refers to the particular net plane on which the Bragg reflection takes place.

Here we assume that a wave of wave vector $\{\xi,\eta,\zeta\}$ excites only one diffracted wave having a wave vector $\{\xi', \eta', \zeta'\}$, where

$$\xi-\xi'=2\pi g_1, \quad \eta-\eta'=2\pi g_2 . \quad (29)$$

The components ζ and ζ' are given by equation (8).

Under this condition and also if the incident wave is a plane wave it is sufficient to consider only two components $F(\xi, \eta)$ and $F(\xi', \eta')$ of a vector $\mathbf{F}$. Also continuous matrices $[\exp i(\zeta\Delta z_n)]$ and $\mathbf{Q}_n$ can be reduced to the following two-by-two matrices respectively.

$$\zeta_n = \begin{pmatrix} \exp i\zeta\Delta z_n & 0 \\ 0 & \exp i\zeta'\Delta z_n \end{pmatrix} \quad (30)$$

$$\mathbf{Q}_n = \begin{pmatrix} 1+i(K/\zeta)Q_0^n\Delta z_n & i(K/\zeta)Q_{-g}^n\Delta z_n \\ i(K/\zeta')Q_g^n\Delta z_n & 1+i(K/\zeta')Q_0^n\Delta z_n \end{pmatrix} \quad (31)$$

where Q_0^n and Q_g^n stand for the 0th and gth Fourier coefficients of $u(x, y, z)$ in the nth slice.

So far we took the origin of the coordinates for each slice at an arbitrary point on a line perpendicular to the slice. Therefore $\{Q_g^n\}$ differ by a phase factor from each other. If we divide the bulk crystal by parallel planes passing through equivalent points on a zone axis in the Bragg-reflecting net plane concerned and take the origin at the equivalent point for each slice, $\{Q_g^n\}$ do not depend upon n.

As a result of changing the origin, however, we must change the phase factor of the Fourier transform of the wave function at the $(n+1)$th surface relative to that at the nth surface by

$$\mathbf{d}_n = \begin{pmatrix} \exp i(\xi p_n+\eta q_n) & 0 \\ 0 & \exp i(\xi' p_n+\eta' q_n) \end{pmatrix} \quad (32)$$

where p_n and q_n are the relative displacements of the origin in the x- and y-directions on going from the nth to the $(n+1)$th slice. Thus we have the relation

$$\mathbf{F}_{n+1}=(\mathbf{d}_n\zeta_n\mathbf{Q}_n)\mathbf{F}_n \quad (33)$$

as a special case of equation (22). Obviously, in perfect crystals, displacements $\{p_n, q_n\}$ and the thickness Δz_n can be considered as constant throughout the crystal. Thus we need not specify them by the suffix n.

In order to make the matrix product $(\mathbf{d\zeta Q})$ symmetrical we use the following notations:

$$(K/\zeta)Q_0 = \phi_t, \quad (K/\zeta')Q_0 = \phi_r \quad (34)$$

$$a = \exp \tfrac{1}{2}i(\phi_t-\phi_r)\Delta z \quad (35)$$

$$b_{12}= (K/\zeta)Q_{-g} \exp -\tfrac{1}{2}i(\phi_t+\phi_r)\Delta z$$
$$b_{21}= (K/\zeta')Q_g \exp -\tfrac{1}{2}i(\phi_t+\phi_r)\Delta z . \quad (36)$$

In practice we can neglect the term $\exp -\frac{1}{2}i(\phi_t+\phi_r)\Delta z$ for both of X-rays and electrons since the magnitude of Δz is reducible to a distance of a few atoms.

Using these relations, we have

$$\mathbf{d\zeta Q} = \exp i\Phi.\mathbf{R} \quad (37)$$

where

$$\Phi = (\xi p+\eta q+\zeta\Delta z)+\tfrac{1}{2}(\phi_t+\phi_r)\Delta z-\varphi\Delta z \quad (38)$$

and

$$\mathbf{R}=\begin{pmatrix}\exp i\varphi\Delta z & 0\\ 0 & \exp -i\varphi\Delta z\end{pmatrix}\begin{pmatrix}a & ib_{12}\Delta z\\ ib_{21}\Delta z & a^*\end{pmatrix}. \quad (39)$$

In deriving this we use the relation

$$g_1p+g_2q+g_3\Delta z=0\,. \quad (40)$$

Thus our problem is reduced to a mathematical problem of calculating a matrix product $\mathbf{R}^N$. A problem of this type appears frequently in various fields of physics; for example, four-terminal-circuit-theories, multilayer optical films and the Ising model of lattice statistics.

A standard technique for calculating the matrix product is to find the eigenvalue $\lambda_\pm$ and the eigenvector $\mathbf{X}$ which are defined by

$$\mathbf{R}=\mathbf{X}\begin{pmatrix}\lambda_+ & 0\\ 0 & \lambda_-\end{pmatrix}\mathbf{X}^{-1}. \quad (41)$$

In our particular problem, it turns out that

$$\begin{aligned}\lambda_\pm&=\{\cos\varphi_D\pm i\,(\sin^2\varphi_D+B^2)^{\frac{1}{2}}\}\Delta z\\ &\simeq\exp\pm i(\varphi_D^2+B^2)^{\frac{1}{2}}\Delta z\end{aligned} \quad (42)$$

where

$$\varphi_D=\varphi+\tfrac{1}{2}(\phi_t-\phi_r) \quad (43)$$

and

$$B^2=(K/\zeta)(K/\zeta')Q_gQ_{-g}\,. \quad (44)$$

Here φ_D is a parameter which indicates the deviation from the exact Bragg condition in a dynamical sense. The eigenvector matrices are given in the Appendix.

Thus, writing

$$\mathbf{K}\equiv\mathbf{R}^N=\mathbf{X}\boldsymbol{\lambda}^N\mathbf{X}^{-1} \quad (45)$$

we have the matrix elements of $\mathbf{K}$ as

$$\begin{aligned}K_{11}&=\tfrac{1}{2}(\varphi_D^2+B^2)^{-\frac{1}{2}}\{(\varphi_D+(\varphi_D^2+B^2)^{\frac{1}{2}})\exp i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\\ &\quad-(\varphi_D-(\varphi_D^2+B^2)^{\frac{1}{2}})\exp -i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\}\\ K_{12}&=\tfrac{1}{2}b_{12}(\varphi_D^2+B^2)^{-\frac{1}{2}}\{\exp i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\\ &\quad-\exp -i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\}\\ K_{21}&=\tfrac{1}{2}b_{21}(\varphi_D^2+B^2)^{-\frac{1}{2}}\{\exp i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\\ &\quad-\exp -i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\}\\ K_{22}&=\tfrac{1}{2}(\varphi_D^2+B^2)^{-\frac{1}{2}}\{(-\varphi_D+(\varphi_D^2+B^2)^{\frac{1}{2}})\exp i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\\ &\quad+(\varphi_D+(\varphi_D^2+B^2)^{\frac{1}{2}})\exp -i(\varphi_D^2+B^2)^{\frac{1}{2}}t_0\}\,.\end{aligned} \quad (46)$$

Here t_0 is the thickness of the crystal, namely $N\Delta z$. Thus, finally, the transmitted and reflected waves (Fourier transforms of wave functions) are given by

$$\begin{pmatrix}T\\ R\end{pmatrix}=\exp iN\Phi\begin{pmatrix}K_{11}\\ K_{21}\end{pmatrix}. \quad (47)$$

Spherical wave cases

In X-ray cases we need to consider a spherical wave as an incident wave (Kato, 1960, 1961*a*, *b*). According to the present formulation, wave functions at the exit surface can be given immediately as

$$\begin{pmatrix}f_a^0\\ f_a^g\end{pmatrix}=\frac{1}{2\pi}\int\begin{pmatrix}T\exp i(\xi x+\eta y)\\ R\exp i(\xi' x+\eta' y)\end{pmatrix}F_e(\xi,\eta)\,d\xi\,d\eta \quad (48)$$

where f_a^0 and f_a^g correspond to the transmitted and reflected waves on the exit surface respectively and $F_e(\xi,\eta)$ is given by equation (25).

(*b*) *Absorbing crystals*

We can derive a phenomenological theory for absorbing crystals assuming $v(x,y,z)$ is complex. According to this formal extension we may predict easily ordinary absorption effects as well as the Borrmann effect. The former can be described by the imaginary part of $(\phi_t+\phi_r)$ in equation (38) and the latter by the imaginary part of B^2, and of $(\phi_t-\phi_r)$ of φ_D in asymmetrical cases.

4. Discussion

(*a*) *Comparison with the theory of Darwin*

In § 3 we treated the two-beam Laue case of a perfect crystal as the simplest example of the present 'lamellar crystal' theory. In Darwin's theory (1914*a*, *b*) a recurrence formula is set up for the Fraunhofer amplitudes of transmitted and reflected waves due to lamellar slices. His treatment is not obviously understandable because the Fraunhofer amplitude is the amplitude of the scattered wave at infinity whereas we are concerned with crystal waves in individual slices.

We saw in § 2(*d*), however, that Fraunhofer amplitudes are proportional to $F(\xi,\eta)$ on the rear surface of a crystal. The argument can be applied also to the Fourier transform of any crystal slice. Thus we can see an intimate relation between a recurrence formula of Fraunhofer amplitudes and that of Fourier transforms (equation (33)). The present theory is essentially equivalent to that which may be set up for Laue cases based on the same principle as Darwin's theory. In this sense the present theory is an extension of Darwin's theory of primary extinction.

(*b*) *Comparison with the theory of Laue–Bethe*

Since Darwin's results and those of Laue and Bethe are essentially the same, it is expected that the present theory is equivalent also to the theory of Laue and Bethe in the case of perfect crystals. In fact, we can see the perfect agreement between crystal wave-functions obtained from both theories (for example, compare equations (46) and (47) with § 2 of the previous paper (Kato, 1961)). An advantage of the present theory is that it has a possibility of being extended to distorted crystals, which will be shown in the next paper.

(*c*) *Comparison with the theory of Cowley & Moodie*

The basic formulation established here has a similar form to that given by Cowley & Moodie. This can be

seen by comparing equation (24) with equation (3·2) of their paper (1958). The essential difference lies in that in their treatment the convolution is carried out in real space instead of two-dimensional Fourier space. In other words they used a Huyghens principle to connect wave functions on the front and the rear surface of a crystal slice.

Consequently, in their formulation, a quadratic approximation of an optical path length was used; namely

$$\{x^2+y^2+(\Delta z)^2\}^{\frac{1}{2}} \sim \Delta z+(x^2+y^2)/2\Delta z\,.$$

Moreover, each slice was treated as a phase grating. Thus the thickness Δz was assumed extremely small compared with atomic size. Under these conditions the convolution process of x and y over $\pm\infty$ is rather difficult to justify. It seems that their treatment is correct only when scattering angles are extremely small.

In the present theory we avoid the assumptions mentioned above. The only assumption used here is a Born approximation in a crystal slice. This is very plausible except in special cases of electron diffraction where heavy atoms are involved (Hoerni & Ibers, 1953).* Thus it is possible to apply the present theory to X-ray and neutron cases in which scattering angles are not always small. Moreover, here, a factor (K/ζ) due to obliqueness of waves is properly treated and the theory is applicable also to asymmetrical cases.

APPENDIX

The eigenvector matrix of *R*-matrix

The definition of eigenvector matrix is given by equation (41). This gives us that

$$\mathbf{X} = \begin{pmatrix} p(+)r_{12} & p(-)r_{12} \\ p(+)(\lambda_+-r_{11}) & p(-)(\lambda_--r_{11}) \end{pmatrix} \qquad \text{(A1)}$$

$$\mathbf{X}^{-1} = \begin{pmatrix} q(+)r_{21} & q(+)(\lambda_+-r_{11}) \\ q(-)r_{21} & q(-)(\lambda_--r_{11}) \end{pmatrix} \qquad \text{(A2)}$$

where r_{ij} are matrix elements of $\mathbf{R}$. $p(\pm)$ and $q(\pm)$ are normalization factors. Since only their product is to be fixed, an arbitrary factor can be chosen as

$$q(\pm)=1\,.$$

Thus

$$p(\pm)=\{(\lambda_\pm-r_{11})(2\lambda_\pm-r_{11}-r_{22})\}^{-1}. \qquad \text{(A3)}$$

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* In this case also the present formalism is applicable in principle if we use a suitable transform $\tilde{Q}_{\alpha\beta}$ in equation (15). For example, pseudo-kinematical theory (Hoerni & Ibers, 1953; Hayashi, 1960) may be used for a crystal slice.

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