A Dynamical Theory of Diffraction for a Distorted Crystal

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(Received January 11, 1969)

A dynamical theory of X-ray and electron diffraction for a distorted crystal which has been briefly explained elsewhere (Acta cryst. **15** (1962) 1311) is presented in more detail. The fundamental equations for both the X-ray and electron cases are deduced from the Maxwell or the Schrödinger equations, respectively, inside crystalline media which may contain distortions. A method of solution of the equations using the Riemann function is given together with the following general results; (1) the intensities and integrated intensities, with respect to the incident angle, of the transmitted and diffracted rays at a point on the exit surface are given by integrals over the entrance surface, (2) a theoretical basis for the column approximation in the electron case is provided, (3) the intensities of the X-ray projection topographs (traverse pattern) is not dependent upon the type of the incident waves but upon the total intensity to which the crystal is exposed. The limit of the applicability of the present theory to distorted crystals is given compared with the other theories.

§1. Introduction

A dynamical theory of X-ray and electron diffraction which may be applied to distorted crystals as well as perfect ones has been briefly presented elsewhere.[1] In the present paper the theory will be described in more detail together with a method of solution and with some general results as to diffraction phenomena inside perfect and distorted crystals.

Several theories have already been published. A lamellar theory of Cowley and Moodie[2,3] may also be applied to distorted crystals. In their theory the crystal is divided into a set of thin lamellae parallel to the surface, and the relation between the wave functions at successive bounda-ry surfaces of the lamellae has been derived on the basis of the wave optical principles applied to each lamella. Iteration of these relations gives the relation of the wave functions at the entrance and the exit surfaces. In principle this relation can be applied to crystals with any kind of distor-tion, but this application involves many iterations of complicated convolutions and the calculation seems impractical. Cowley and Moodie[4] have deviced a simpler method of approximation (the phase grating approximation), but this can only be applied to very thin crystals.

Kato[5,6] has developed a similar theory. His relation between the wave functions at successive surfaces of the lamellae, given in terms of the

two-dimensional Fourier transforms, is also quite general in principle. In practice, however, he confined the application to the case where the Fourier transform has significant values at only two points in the two dimensional reciprocal space. This means that each lamella has the same perfectly periodic structure which may be displaced parallel to each other. To remove this restriction, one has to consider the Fourier transform to be a continuous function in the reciprocal space, but again this will make the calculation very complicated.

Howie and Whelan[7−9] have developed another theory based upon the method of Darwin[10] originally applied to the symmetrical Bragg case. They applied this method to the Laue case, and obtained a set of differential equations for the transmitted and the diffracted waves which are assumed to be varying in the z-direction, the direction perpendicular to the surface. Here again the assumption of the lamellar structure is made use of.

Penning and Polder [11] have developed another theory which deals with the refraction of X-rays inside a strained crystal. They consider that the X-rays can be split into beams narrow enough that within the width of a beam the crystal is almost perfect but on the other hand wide enough that the corresponding uncertainty in k-value is much smaller than the difference corresponding to the width of the Bragg reflection. This difference is of the same order of magnitude of the distance between the two apices of the dispersion surface. This means that each elementary beam is wider than the extinction distance which is just the reciprocal of the distance between the apices, and still within this distance the distortion is almost homogeneous. Therefore this theory is applicable to crystals with very slight distortion in which any part with dimension of the extinction distance can be regarded as almost perfect. Kato[12] and Kambe[13] have independently given theoretical deductions of the theory from the fundamental wave equations. This does not mean, however, that the restriction of the theory to slightly distorted crystals is removed.

These theories except Penning and Polder's are mainly for electron diffraction and deal with lamellar structures. Howie and Whelan[7−9] have introduced the column approximation[14] to apply this kind of theory to actual crystal with more complicated distortions. The crystal sliced in many lamellae is again divided into a set of columns perpendicular to the lamellae and thin enough that any part of each lamella within a column can be regarded as perfect. An assumption is then made use of that the wave function at the exit surface of a column can be obtained by applying the theory to each column individually. The result of the present theory shows that this assumption is good enough for most of the images in electron-micrographs, as will be discussed below and in § 8.

When a plane wave is incident on a crystal so that one Bragg reflection is excited, the wave function at a point P (Fig. 1) on the exit surface
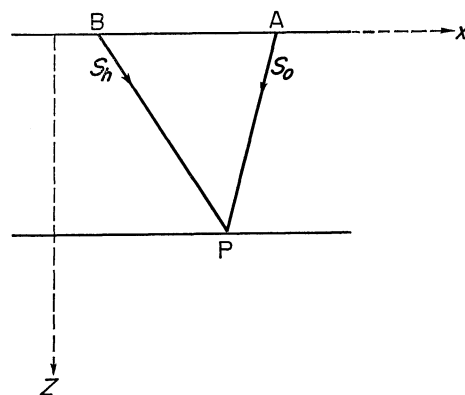

Fig. 1. Showing the triangle PAB which determines the waves at P on the exit surface, A and B being on the entrance surface, s_0 and s_h represent unit vectors in the directions of the incident and diffracted beams, respectively.

of a crystal is determined by the wave field inside the triangle PAB, where A and B are the points on the entrance surface such that $\overline{\mathrm{AP}}$ and $\overline{\mathrm{BP}}$ are parallel to the incident and the diffracted beams, respectively. This fact may be seen from considerations of the energy flow, but the exact proof will be given in § 8. The shape of the triangle PAB in the X-ray and the electron cases are very different as a re schematically shown in Fig. 2(a) and (b). The order of magnitude of the quantities involved in usual experimental conditions in each case is shown in Table I, where λ is the wavelength, θ_B the Bragg angle, D the thickness of the crystal and $2l$ the distance $\overline{\mathrm{AB}}$. The range of the effective strain field around a dislocation for both the electron micrograph and the X-ray diffraction topograph is of the order of the extinction distance in each case.[14] This range is shown schematically by dotted circles in Fig. 2(a) and (b). In the electron case where $\overline{\mathrm{AB}} \approx 10\mathrm{A}$ the triangle reduces to a thin column which penetrates through the
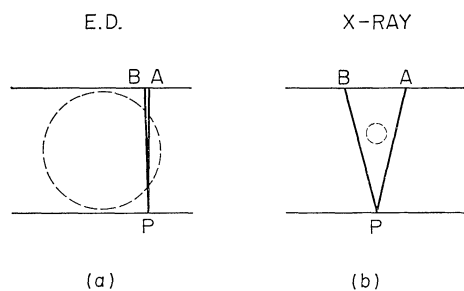
Fig. 2. Schematic figures showing the shape of the triangle PAB in (a) electron and (b) X-ray cases, respectively. Dotted circles represent the range of effective strain field around a dislocation.

Table I.

	λ	$2\theta_B$	D	$2l$
Electron case	$\sim 0.05\,\text{A}$	$\sim 1°$	$\sim 500\,\text{A}$	$\sim 10\,\text{A}$
X-ray case	$\sim 1.0\,\text{A}$	$\sim 25°$	$\sim 500\,\mu$	$\sim 200\,\mu$

effective strain field (Fig. 2(a)), whereas in the X-ray case where $\overline{AB} \approx 200\,\mu$ the whole effective field is embedded in the triangle PAB (Fig. 2(b)). The column approximation can be applied to the former case, but not to the latter.

In a previous short note[1] an attempt to avoid the column approximation and the assumption of the lamellar structure has been briefly presented. The fundamental equations which include Howie and Whelan's equations as a special case and which are applicable to crystals with any kind of small distortion have been obtained directly from the wave equation in crystal by a simple assumption as to the slow variation of the amplitudes and phases of the incident and diffracted waves. Two methods of solution, one that of Riemann and the other a numerical have also been pointed out in the note. Analytical solutions for a perfect crystal by the method of Riemann have been reported.[15] The results coincide with those of the Ewald-Laue theory and those of Kato's theory.[16] Uragami and the present author have numerically calculated the intensity distribution of the images of a dislocation.[17] These results will be described in more detail in subsequent papers. Taupin[18] has developed a theory which is essentially the same as the present one and has calculated the intensities of dislocation images. Authier *et al.*[19] have also calculated those of similar patterns based upon the fundamental equation of the present theory. These calculations differ somewhat from each other in the condition of diffraction, but each of them has explained some of the important features of dislocation images obtained experimentally. Recently Schlangenoto[20] has developed another theory which is also the same as the present one. He has not, however, yet given the solution of the fundamental equations.

Wilkens[21–23] has developed a theory similar to that of Howie and Whelan. He has expanded the wave function in a series of modulated Bloch waves instead of modulated plane waves used in the present theory. His theory is good for the electron case, as is Howie and Whelan's, because of the column approximation.

§2. Modulated Waves in Crystal

In the Ewald-Laue theory for a perfect crystal both the incident and diffracted waves are expressed as composed of two or more plane waves with slightly different wave vectors so that each of them shows a kind of beat effect or an amplitude and phase modulation. This is called the Pendellösung effect.[24,25] When a plane wave is incident, the direction of these modulations is perpendicular to the surface. The period of the modulation, called the extinction distance, is of the order of several hundred angstroms in the electron case and several tens of microns in the X-ray case. These values are, respectively, 10^4 and 10^5 times as large as the wave-length. In fact their variation occurs only in macroscopic dimensions. Hereafter, we shall refer to this kind of slow variation as a macroscopic variation, in contrast with the microscopic variation of the carrier wave $\exp(-2\pi i k_0 \cdot r)$, with the period which is of the order of or less than the atomic dimension. When the crystal is distorted the direction of the modulation of the incident and diffracted waves will no longer be perpendicular to the surface and the variation will become much more complicated. In any case, however, their variation will be much slower than that of the carrier wave itself, so long as the distortion is slight.

Thus in a general case where the number of strong diffracted waves is not necessarily restricted to one, the wave function in a distorted crystal may be expressed by a sum of modulated waves

$$\phi(r) = \sum_g \phi_g(r) \exp(-2\pi i k_g \cdot r) , \qquad (1)$$

where each k_g is related to k_0 by

$$k_g = k_0 + g , \qquad (2)$$

g being a reciprocal lattice vector. Each $\phi_g(r)$ and

its gradient are of macroscopic variation, and $\phi_g(r)$ represents all possible modulations of the g-th diffracted wave.

However, a more convenient expansion for a distorted crystal is that based upon a " local reciprocal lattice " and is given by

$$\phi(r) = \sum_g \phi'_g(r) \exp(-2\pi i S_g(r)) , \qquad (3)$$

with

$$S_g(r) = K_g \cdot r - g \cdot u(r_0) . \qquad (4)$$

Here $u(r)$ is a continuous function of position representing the displacement of the atom at r, and r_0 is the position vector of the point where the atom which is displaced to r by the distortion is originally situated. Thus the relation between r and r_0 is given by

$$r = r_0 + u(r_0) . \qquad (5)$$

$S_g(r)$ in (3) is the characteristic function of Hamilton of the g-th component wave, and the vector

$$k'_g = \text{grad } S_g(r) = k_g - \text{grad } (g \cdot u) , \qquad (6)$$

can be regarded as the wave vector of this component wave. As is shown in Appendix I, a local reciprocal lattice vector defined in the vicinity of r in the distorted crystal is given by

$$g' = g - \text{grad } (g \cdot u(r_0)) , \qquad (7)$$

so that k'_g is related to k_0 by

$$k'_g = k_0 + g' . \qquad (8)$$

The expansions (1) and (3) are based upon the reciprocal lattices of the perfect and the distorted crystals, respectively. $\phi'_g(r)$, grad $\phi'_g(r)$ and $(g \cdot u)$ in (3) and (4) are assumed to be of macroscopic variation.

That $\phi_g(r)$ and $\phi'_g(r)$ are now variable allows an ambiguity in the definition of k_0, because the effect of any slight change Δk_0 in k_0 can be compensated by multiplying a factor $\exp(2\pi i \Delta k_0 \cdot r)$ to $\phi_g(r)$ or $\phi'_g(r)$ without affecting the macroscopic character or their variation so long as Δk_0 is sufficiently smaller than any reciprocal lattice vector. Thus the value of k_0 is left to an appropriate initial choice. A convenient choice is that, (i), the magnitude of k_0 is given by

$$|k_0| = k = nK , \qquad (9)$$

where n is the mean refractive index, and, (ii), k_0 satisfies the continuity condition for the tangential component of the wave vector at the entrance surface. In this paper, however, k_0 will be taken as always satisfying (i), but not always (ii). In § 10, in fact, k_0 will be defined as always satisfying the Bragg condition even if the direction of incidence deviates from it.

§ 3. Crystalline Field inside a Distorted Crystal

In the present paper any function of position inside a crystal will be called a crystalline field and designated by $\chi(r)$. For the present purpose the following two functions are important: (i) The electrostatic potential, $V(r)$, inside a crystal divided by the accelerating voltage, E, of the incident electron, in electron diffraction, i.e.

$$\chi_e(r) = V(r)/E^* , \qquad (10)$$

and (ii) 4π times the polarizability of the crystal, in the case of X-rays, given by

$$\chi_x(r) = -\frac{e^2 \lambda^2}{\pi m c^2} n(r) , \qquad (11)$$

where e is the electronic charge, m the electronic mass, c the velocity of light, λ the wavelength of X-rays and $n(r)$ the density of electrons at r.

When the crystal is perfect each of $\chi(r)$ is obviously expanded in a Fourier series,

$$\chi(r) = \sum_g \chi_g \exp(-2\pi i g \cdot r) , \qquad (12)$$

where g is a reciprocal lattice vector. In a slightly distorted crystal the displacement of atoms is represented by $u(r)$ in (5). The positions of an atom r_0 and r before and after the distortion is introduced, respectively, are connected to each other by (5). This equation represents not only the relation between the positions of atoms but that between the corresponding positions of any point in the distorted and undistorted crystals. If the distortion is slight, the value of χ in the distorted crystal, which is designated by $\chi'(r)$, can be put equal to the value of χ in the undistorted crystal at the corresponding point r_0 given by (5), i.e.

$$\chi'(r) = \chi(r - u(r_0)) . \qquad (13)$$

Substitution of (12) into (13) gives

$$\chi'(r) = \sum_g \chi_g \exp(-2\pi i (g \cdot r - g \cdot u)) . \qquad (14)$$

As before grad $(g \cdot u)$ is assumed to be of macroscopic variation.

§ 4. Fundamental Equations for Electron Diffraction

The Schrödinger equation for an electron in an

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* χ_e may also be written as $\chi_e = V(2me/h^2)\lambda^2$, in terms of the electronic mass, m, and the wavelength, λ. If the relativistic effect[26)] has to be taken into account, m should be replaced by the relativistic mass and the relativistic relation between λ and E should be used. In this case eq. (9) should be replaced by

$\chi_e = V(2me/h^2)\lambda^2(1 + (\lambda_c/\lambda)^2)^{1/2} = (V/E)(1 + 2\alpha)/(1 + \alpha)$

where $\lambda_c = h/mc$ and $\alpha = eE/2mc^2$, m being the rest mass of electron.

electrostatic potential (10) can be written as

$$\nabla^2\phi(r)+4\pi^2 K^2(1+\chi'(r))\phi(r)=0 , \qquad (15)$$

where K, given by

$$K^2=(2me/h^2)E^* , \qquad (16)$$

is the wave number of the electron in vacuum. Substitution of (3) and (14) into (15) gives

$$\sum_g \{\nabla^2\phi_g'(r)+2\pi i\nabla^2(g\cdot u)\phi_g'(r)-4\pi i(k_g'\cdot\text{grad }\phi_g'(r))$$
$$+4\pi^2[K^2(1+\chi_0)-k_g^2]\phi_g'(r)$$
$$+4\pi^2 K^2\sum_{g'\neq g}\chi_{g-g'}\phi_{g'}'(r)\}$$
$$\times\exp(-2\pi i[k_g\cdot r-g\cdot u])=0 . \qquad (17)$$

where k_g', given by (8), is the wave vector of the g-th wave based upon the local reciprocal lattice vector. Here the following two assumptions are made; (i), the first and the second terms in the curly bracket of (17) can be neglected on account of the macroscopic character of the variations in $\phi'(r)$ and in $(g\cdot u)$, and (ii), the remaining terms are also of macroscopic variation and can be taken out of the integration when (17) is multiplied by $\exp(2\pi i[k_h\cdot r-h\cdot u])$ and integrated with respect to r over a unit cell where h is a reciprocal lattice vector. The integral of the exponential function vanishes except for $g=h$, since u is almost constant within a unit cell, and the following equation is obtained for each unit cell.

$$(k_h'\cdot\text{grad }\phi_h'(r))=-i\pi[K^2(1+\chi_0)-k_h'^2]\phi_h'(r)$$
$$-i\pi K^2\sum_{h'\neq h}\chi_{h-h'}\phi_{h'}'(r) . \qquad (18)$$

For significant reflections $|k_h'|\doteqdot K$, the difference between them being of the order of $10^{-4}\times K$, and (18) becomes

$$(s_h\cdot\text{grad }\phi_h'(r))=i2\pi K\beta_h'(r)\phi_h'(r)$$
$$-i\pi K\sum_{h'\neq h}\chi_{h-h'}\phi_{h'}'(r) , \qquad (19)$$

where s_h is the unit vector in the direction of k_h, and

$$\beta_h'(r)=(k_h'(r)^2-k^2)/2K^2 , \qquad (20)$$

k being the " mean wave number " in the crystal given by (8) or

$$k=K(1+\chi_0)^{1/2}=nK . \qquad (21)$$

The directions of k_h and of k_h' are slightly different, but the difference being very small, s_h can be regarded as representing the direction of k_h' as well.

The case where absorption of waves takes place will be given in a later section and now the argument is confined to the non-absorbing case,

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* If the relativistic formula for $\chi_e(r)$ (see footnote on p. 1243) is to be used, K^2 should also be given by a relativistic formula $K^2=(2me/h^2)E(1+\alpha)$, $\alpha=eE/2mc^2$.

where k and k_h are real so that β_h' is, in sufficient approximation, given by

$$\beta_h'(r)=(|k_h'(r)|-k)/K , \qquad (22)$$

since $|k_h'|\doteqdot k\doteqdot K$. The geometry of β_h' is shown in Fig. 3, where $QM'=K\beta_h'$.
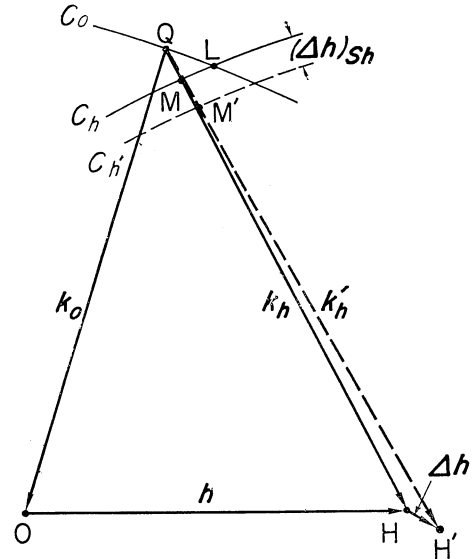

Fig. 3. Geometry of k's and β's. O is the origin of the reciprocal lattice, H and H' an ideal and a local reciprocal lattice points, respectively. c_0, c_h and c_h' are spheres of radius k drawn around O, H and H' as centres, respectively. $QM=K\beta_h$ and $QM'=K\beta_h'$. The distance between c_h and c_h' is $(\Delta h)_{sh}$.

Equation (19) is the fundamental equation of the present theory in the case of electron diffraction. It includes the equations derived from the lamellar theories as a special case. When the crystal has a lamellar structure so that u, and hence β_h', is a function of a single coordinate, z, perpendicular to the surface, and when a plane wave is incident so that the magnitude and phase of $\phi_0'(r)$ on the top surface are constant, the value of ϕ_h''s inside the crystal are obviously function of only z. Then (19) reduces to

$$\frac{d\phi_h'}{dz}=i2\pi(K/\gamma_h)\beta_h'\phi_h'-i\pi(K/\gamma_h)\sum_{h'\neq h}\chi_{h-h'}\phi_{h'}' , \qquad (23)$$

where γ_h is the cosine of the angle between k_h and the z-axis. This equation can as well be written in a tensorial form

$$\frac{d\phi'}{dz}=i\pi M\cdot\phi' , \qquad (24)$$

where $\phi'(\phi_0', \cdots, \phi_h', \cdots)$ is an N-dimensional vector, N being the number of terms in the right-hand side of (23), and M a scattering matrix defined by

$$(M)_{hh'} = 2(K/\gamma_h)\beta'_h\delta_{hh'} - (K/\gamma_h)\chi_{h-h'} \ . \qquad (25)$$

Equation (23) is equivalent to that derived by Howie and Whelan.[8]

For a perfect crystal for which M is independent of z, (24) can easily be integrated giving the relation between the values of ϕ at $z=0$ and at $z=D$, where D is the thickness of the crystal,

$$\phi(D) = \exp(i\pi M \cdot D) \cdot \phi(0) \ . \qquad (26)$$

This result has been given by Sturkey[27], Niehrs[28] and Fujimoto[29] by different methods.

§5. Fundamental Equations for X-Ray Diffraction

A wave equation for the electric displacement D induced by X-rays in a crystalline medium is given by Laue.[30]

$$\nabla^2 D + 4\pi^2 K^2 D + 4\pi \ \text{curl curl} \ P = 0 \ , \qquad (27)$$

where K is the wave number of the X-rays in vacuum and P the electric polarization given by

$$4\pi P = \chi' D \ , \qquad (28)$$

χ' being 4π times the polarizability of the crystal given by (11). The prime on χ' means that it refers to a distorted crystal. D represents the oscillatory part of D, so that it satisfies the relation

$$\text{div} \ D = 0 \ .$$

D may be expanded in either a series of modulated waves similar to (1), namely

$$D(r) = \sum_g D_g(r) \exp(-2\pi i k_g \cdot r) \ , \qquad (29)$$

where k_g is given by (2), or that similar to (3), namely

$$D(r) = \sum_g D'_g(r) \exp(-2\pi i [k_g \cdot r - g \cdot u]) \ . \qquad (30)$$

P is also expanded in a similar series as

$$\chi'(r)D(r) = 4\pi P(r) = 4\pi \sum_g P'_g(r)$$
$$\times \exp(-2\pi i [k_g \cdot r - g \cdot u]) \ , \qquad (31)$$

where $P'_g(r)$ is given by

$$4\pi P'_g(r) = \sum_{g'} \chi_{g-g'}(r)D'_{g'}(r) \ . \qquad (32)$$

The first and the third terms in (27) then turn out to be

$$\text{curl curl} \ P = \sum_g \{\text{curl curl} \ P'_g$$
$$-2\pi i [\nabla^2(g \cdot u)P'_g - (P'_g \ \text{grad}) \ \text{grad} \ (g \cdot u)$$
$$+ k'_g \cdot \text{div} \ P'_g - (k'_g \cdot \text{grad})P'_g$$
$$+ [k'_g \times \text{curl} \ P'_g]] - 4\pi^2 [k'_g \times [k'_g \times P'_g]]\}$$
$$\times \exp(-2\pi i [k_g \cdot r - g \cdot u]) \ , \qquad (33)$$

and

$$\nabla^2 D = \sum_g \{\nabla^2 D'_g + 2\pi i \nabla^2(g \cdot u)D'_g - 4\pi i(k'_g \cdot \text{grad})D'_g$$
$$- 4\pi^2 k'^2_g D'_g\} \exp(-2\pi i [k_g \cdot r - g \cdot u]) \ . \qquad (34)$$

Here as before, the assumptions are made use of that D_g, P_g and their first derivatives are of macroscopic variation. Moreover P_g's are small compared with D_g's, since χ_g's are small being of the order of $10^{-5} \sim 10^{-6}$ in the X-ray case. In Table II, the order of magnitude of the quantities

Table II.

0 th order	$K^2\|D_g\|$		
1 st order	$K\left\|\dfrac{\partial D_g}{\partial x}\right\|$ etc. , $K^2\|P_g\|$		
2 nd order	$\left\|\dfrac{\partial^2 D_g}{\partial x^2}\right\|$ etc. , $K\left\|\dfrac{\partial P_g}{\partial x}\right\|$ etc. , $\|\nabla^2(g \cdot u)\|\,\|D_g\|$ etc.		
3 rd order	$\|\nabla^2(g \cdot u)\|\,\|P_g\|$ etc.		

involved are shown. If only the first order terms are retained (33) and (34) become

$$\text{curl curl} \ P = -4\pi^2 \sum_g [k_g \times [k_g \times P'_g]]$$
$$\times \exp(-2\pi i [k_g \cdot r - g \cdot u]) \ , \qquad (35)$$

and

$$\nabla^2 D = \sum_g \{-4\pi i(k'_g \cdot \text{grad})D'_g - 4\pi^2 k'^2_g D'_g\}$$
$$\times \exp(-2\pi i [k_g \cdot r - g \cdot u]) \ . \qquad (36)$$

Substitution of (35) and (36) into (27) gives

$$\sum_g \{-4\pi i(k_g \cdot \text{grad})D'_g + 4\pi^2(K^2 - k'^2_g)D'_g$$
$$+ 4\pi^2 \sum_{g'} \chi_{g-g'}[D'_{g'}]_g\} \exp(-2\pi i [k_g \cdot r - g \cdot u])$$
$$= 0 \ , \qquad (37)$$

where

$$[D'_{g'}]_g = -\frac{1}{|k'_g|^2}[k'_g \times [k'_g \times D'_{g'}]] \ , \qquad (38)$$

signifies the component vector of $D'_{g'}$ perpendicular to k_g. Multiplication by $\exp(2\pi i [k_h \cdot r - h \cdot u])$ and integration with respect to r over a unit cell, similar to the deduction of (19) from (17), give

$$(s_h \cdot \text{grad})D'_h = i2\pi K\beta'_h D'_h - i\pi K \sum_{h' \neq h} \chi_{h-h'}[D'_{h'}]_h \ , \qquad (39)$$

where the term $h'=h$ in the summation is now included in the first term in the right-hand side, since every D'_h is perpendicular to k_h so that $[D'_h]_h = D'_h$. Equation (39), the fundamental equation in the X-ray case, has a similar form to (19) the difference being that now D'_h is a vector instead of a scalar and that the last term in the right-hand side is a linear function of $[D'_{h'}]_h$ instead of $D'_{h'}$ itself. This last fact makes the solution of (39) more difficult.

§6. Two-Wave Approximation

In the case of two strong waves, i.e. the case where the transmitted and only one diffracted

waves have appreciable amplitude, each of eqs. (19) and (39) reduce to a set of two equations with two unknown functions. Three cases are to be distinguished: (i) the electron case, (ii) the X-ray case where both D'_0 and D'_h are perpendicular to the plane of incidence, the plane determined by k_0 and k_h, and (iii) the X-ray case where both D'_0 and D'_h are parallele to it. The fundamental equations for each of these three cases are written in a common form:

$$\left.\begin{aligned} \frac{\partial \psi'_0}{\partial s_0} &= -i\pi KC\chi_{-h}\psi'_h \ , \\ \frac{\partial \psi'_h}{\partial s_h} &= -i\pi KC\chi_h\psi'_0 + i2\pi K\beta'_h\psi'_h \ , \end{aligned}\right\} \quad (40)$$

where ψ'_0 and ψ'_h stand, respectively, for D'_0 and D'_h in the X-ray case, C is the polarization factor given by

$$C = \begin{cases} 1 \ , & \text{for the case (i) and (ii)} \ , \\ \cos 2\theta_B & \text{for the case (iii)} \ , \end{cases}$$

θ_B being the Bragg angle, and s_0 and s_h the oblique coordinates parallel to k_0 and k_h, respectively. The differential operator has the meaning $\partial/\partial s_0 = (s_0 \cdot \mathrm{grad})$ etc.

Equations (40) are based upon the expansion (3) or (30). If the expansion (1) or (29) is adopted, equations for ϕ_h's or D_h's are obtained. These equations are also got by the substitution of

$$\left.\begin{aligned} \psi'_0(r) &= \phi_0(r) \ , \\ \psi'_h(r) &= \phi_h(r) \exp(-2\pi i(h \cdot u)) \ . \end{aligned}\right\} \quad (41)$$

into (40), namely

$$\left.\begin{aligned} \frac{\partial \phi_0}{\partial s_0} &= -i\pi KC\chi'_{-h}\phi_h \ , \\ \frac{\partial \phi_h}{\partial s_h} &= -i\pi KC\chi'_h\phi_0 + i2\pi K\beta_h\phi_h \ , \end{aligned}\right\} \quad (42)$$

where

$$\left.\begin{aligned} \chi'_{-h} &= \chi_{-h} \exp(-2\pi i(h \cdot u)) \ , \\ \chi'_h &= \chi_h \exp(2\pi i(h \cdot u)) \ , \end{aligned}\right\} \quad (43)$$

and

$$\left.\begin{aligned} \beta_h &= \beta'_h + \frac{1}{K}\frac{\partial}{\partial s_h}(h \cdot u) \ , \\ &= \beta'_h - \frac{1}{K}(\Delta h)_{sh} \ . \end{aligned}\right\} \quad (44)$$

χ'_h's may be considered as the Fourier components of $\chi'(r)$ based upon the reciprocal lattice of the undistorted crystal, because $\chi'(r)$ can be written as

$$\chi'(r) = \sum_h \chi'_h(r) \exp(-2\pi i h \cdot r) \ , \quad (45)$$

if (43) is substituted into (14). The vector Δh in (44) is given by

$$\Delta h = h' - h = -\mathrm{grad}(h \cdot u) \ ,$$

where h' and h are the local and the normal reciprocal lattice vectors, respectively (see Fig. 3 and Appendix I). Figure 3 shows the geometry of k_h and k'_h. It is seen from the figure that

$$(\Delta h)_{s_h} = |k'_h| - |k_h| \ .$$

Substitution of this relation and (22) into (44) gives

$$\beta_h = (|k_h| - k)/K \ ,$$

which shows that β_h is the value of β'_h defined by the reciprocal lattice of the undistorted crystal and is constant all over the crystal. In (40) β'_h is a function of position and χ_h and χ_{-h} are constant, whereas in (42) β_h is constant and χ'_h and χ'_{-h} given by (43) are functions of position.

Finally, it is of interest to note that the second term in the second equation of (40) can be eliminated altogether by the other transformation

$$\left.\begin{aligned} \psi''_0 &= \phi_0 \ , \\ \psi''_h &= \phi_h \exp(-2\pi i\beta_h k_h \cdot r) \ . \end{aligned}\right\} \quad (46)$$

Since $\partial(k_h \cdot r)/\partial s_h = |k_h| \doteq K$, the fundamental equations become

$$\left.\begin{aligned} \frac{\partial \psi''_0}{\partial s_0} &= -i\pi KC\chi''_{-h}\psi''_h \ , \\ \frac{\partial \psi''_h}{\partial s_h} &= -i\pi KC\chi''_h\psi''_0 \ , \end{aligned}\right\} \quad (47)$$

where

$$\left.\begin{aligned} \chi''_{-h} &= \chi_{-h} \exp(2\pi i(\beta_h k_h \cdot r - h \cdot u)) \ , \\ \chi''_h &= \chi_h \exp(-2\pi i(\beta_h k_h \cdot r - h \cdot u)) \ . \end{aligned}\right\} \quad (48)$$

From (1) and (46) the wave function of the h-th wave can be expressed as $\psi''_h \exp(-2\pi i(1-\beta_h)k_h \cdot r)$, with the wave vector $(1-\beta_h)k_h = k \cdot k_h/|k_h|$. This wave vector with the same direction as k_h and with the magnitude k is just the same as that used in the kinematical theory, and this choice of the wave vectors gives simpler forms of fundamental equations. However, for getting analytical and numerical solution eqs. (40) are much more convenient, and in the following we are concerned with these equations. The partial equations (40), (42) and (47) are generalized forms of the corresponding sets of ordinary differential equations given by Howie and Whelan (ref. 8), eqs. (18a and b); (20a and b); ref. 7), eqs. (1) and (2)).

§7. Boundary Conditions

The incident wave is considered as "quasi-plane" which is expressed as

$$\Psi(r) = \Psi_0(r) \exp(-2\pi i K \cdot r) \ , \quad (49)$$

where K is the wave vector in vacuum and $\Psi_0(r)$,

the complex amplitude, is a function of macroscopic variation. $\Psi_0(r)$ is constant only when the incident is a plane wave with infinite lateral extent. In practice, however, it is a spherical wave or a wave with finite width etc. and so, $\Psi_0(r)$ is a function of position which can be considered as of macroscopic variation in usual experimental conditions.

The crystal surface may be either plane of curved in the macroscopic sense. The incident wave is assumed not to make a small angle with any part of the surface so that the reflection at the surface can be neglected. Thus only the continuity of the wave function itself need be considered and in the case of two strong waves, this can be written as

$$\Psi_0(r_e) \exp(-2\pi i K \cdot r_e) = \phi_0'(r_e) \exp(-2\pi i k_0 \cdot r_e)$$
$$+ \phi_h'(r_e) \exp(-2\pi i [k_h \cdot r_e - h \cdot u]) , \qquad (50)$$

where r_e is the position vector of any point on the entrance surface. If $\Phi_0(r)$ defined by

$$\Phi_0(r) = \Psi_0(r) \exp(-2\pi i [K - k_0] \cdot r) , \qquad (51)$$

is introduced, (50) can be written as

$$\{\phi_0(r_e) - \Phi_0(r_e)\} + \phi_h'(r_e) \exp(-2\pi i h[r_e - u]) = 0 . \quad (52)$$

From the microscopic point of view, the surface is composed of many tiny facets with fairly simple crystallographic indices, and we can take a unit cell of the two dimensional lattice on the surface sufficiently small that the functions of macroscopic variation, $\Phi(r)$, $\phi_0'(r)$ and $\phi_h'(r)$, do not vary appreciably over each unit cell. Integration of (52) over any of such two dimensional unit cells gives

$$\phi_0'(r_e) = \Phi_0(r_e) , \qquad \qquad \bigg\} \quad (53)$$
$$\phi_h'(r_e) = 0 .$$

The difference between these boundary conditions and those in the Ewald-Laue theory is that here both sides of (53) are functions of position so that any type of incident wave is allowed, whereas there both sides are constant. $\Phi_0(r_e) = \Psi_0(r_e)$ only when the wave vectors of the waves in vacuum and in crystal are equal to each other. However, if the boundary surface is a plane and k_0 is so chosen that it has the same tangential component as K, the distinction between Φ_0 and Ψ_0 is not important, since then the difference between them is just a constant phase factor all over the surface. Otherwise this distinction is important.

Similar conditions hold also for the amplitude functions ϕ_0 and ϕ_h defined by (1), namely:

$$\phi_0(r_e) = \Phi_0(r_e) , \qquad \qquad \bigg\} \quad (54)$$
$$\phi_h(r_e) = 0 .$$

In the next section a method of solution of eqs.

(40) will be given in which the boundary conditions for the first order derivatives of ϕ_0' and ϕ_h' are also to be taken into account. Two of these conditions are readily obtained from (53) and (40), namely:

$$\frac{\partial \phi_0'}{\partial s_0}(r_e) = 0 , \qquad \qquad \bigg\} \quad (55)$$
$$\frac{\partial \phi_h'}{\partial s_h}(r_e) = -i\pi K C \chi_h \Phi_0(r_e) .$$

Differentiation of (53) with respect to the curved coordinate, ξ, along the intersection of the boundary surface and the plane of incidence (Fig. 4) gives

$$\frac{\partial \Phi_0}{\partial \xi}(r_e) = \frac{\partial \phi_0'}{\partial \xi}(r_e) = \frac{\partial s_0}{\partial \xi} \frac{\partial \phi_0'}{\partial s_0} + \frac{\partial s_h}{\partial \xi} \frac{\partial \phi_0'}{\partial s_h} , \bigg\}$$

and

$$0 = \frac{\partial \phi_h'}{\partial \xi}(r_e) = \frac{\partial s_0}{\partial \xi} \frac{\partial \phi_h'}{\partial s_0} + \frac{\partial s_h}{\partial \xi} \frac{\partial \phi_h'}{\partial s_h} .$$
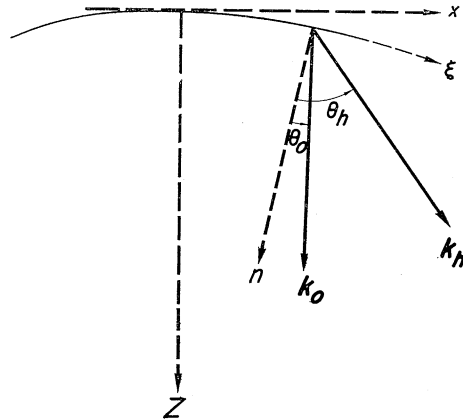$$(56)$$

Fig. 4. Showing the relation between the curved coordinate along the surface, ξ, surface normal n, and the rectangular coordinates x and z.

The other two conditions are obtained from (55) and (56), namely:

$$\frac{\partial \phi_0'}{\partial s_h} = (\sin 2\theta_B / \gamma_0(r_e)) \frac{\partial \Phi_0}{\partial \xi}(r_e) , \bigg\}$$
$$\frac{\partial \phi_h'}{\partial s_0} = -i\pi K C \chi_h (\gamma_0(r_e)/\gamma_h(r_e)) \Phi_0(r_e) , \bigg\} \quad (57)$$

where $\gamma_0(r_e)$ and $\gamma_h(r_e)$ are the cosines of the angle θ_0 and θ_h (Fig. 4) which k_0 and k_h, respectively, make with the inward normal to the entrance surface at r_e.

It is worth noticing that the boundary conditions of the present theory are for the functions of macroscopic variation. It is therefore plausible that the boundary is considered to be a mathematical surface. In the Ewald-Laue theory, on

the contrary, the boundary conditions is for the wave function itself which is of microscopic variation. It is not at all obvious that the boundary surface with extrusions of atomic dimensions can be considered as a mathematical surface.

§8. Solution of the Two-Wave Equations for the Laue Case by the Method of Riemann

Each set of the two-wave eqs. (40), (42) and (47) can be solved by converting them into second order differential equations. However, only the solutions of (40) will be given which have the most convenient form for the solution. The other equations may be solved similarly. Differentiation of the first equation of (40) with respect to s_h and elimination of ϕ'_h and its derivative gives

$$\frac{\partial^2\phi'_0}{\partial s_h \partial s_0} - i2\pi K\beta'_h \frac{\partial\phi'_0}{\partial s_0} + \pi^2 K^2 C^2 \chi_h \chi_{-h} \phi'_0 = 0 , \qquad (58)$$

and similarly

$$\frac{\partial^2\phi'_h}{\partial s_0 \partial s_h} - i2\pi K\beta'_h \frac{\partial\phi'_h}{\partial s_0}$$
$$+ \left(\pi^2 K^2 C^2 \chi_h \chi_{-h} - i2\pi K \frac{\partial\beta'_h}{\partial s_0}\right)\phi'_h = 0 . \qquad (59)$$

These equations have the hyperbolic form and can be solved by the method of Riemann.[31] Let P be any point inside or on the exit surface and let A and B be those on the entrance surface shown in Fig. 1. Here the pure Laue case is assumed that each of the lines AP and BP intersects the entrance surface at one point (A or B) and the direction of each is the same as and not opposite to the direction of k_0 or k_h, respectively. This means that at no point on the entrance

surface the incident or the diffracted wave emerges from the crystal. The solutions at P can then be expressed in a single form

$$\phi'_i(P) = \frac{1}{2}\left\{\phi'_i(A)v_i(A) + \phi'_i(B)v_i(B)\right\}$$
$$+ \frac{1}{2}\int_{AB}\left\{\left(\frac{\partial\phi'_i}{\partial s_0}v_i - \phi'_i\frac{\partial v_i}{\partial s_0}\right)ds_0\right.$$
$$\left. - \left(\frac{\partial\phi'_i}{\partial s_h}v_i - \phi'_i\frac{\partial v_i}{\partial s_h} - i4\pi K\beta'_h\phi'_i v_i\right)ds_h\right\}$$
$$i=0 \text{ or } h, \quad (60)$$

where the suffix i stands for 0 or h, the integral extends over the entrance surface from the points A to B and $\phi'_i(A)$, $\phi'_i(B)$ etc. signify the value of the relevant functions at A or B. The function v_0 and v_h in (60) are the Riemann functions for ϕ'_0 and ϕ'_h, respectively, given by the following conditions.

(1) v_0 and v_h satisfy the equations

$$\frac{\partial^2 v_0}{\partial s_0 \partial s_h} + i2\pi K\beta'_h \frac{\partial v_0}{\partial s_0}$$
$$+ \left(\pi^2 K^2 C^2 \chi_h \chi_{-h} + i2\pi K\frac{\partial\beta'_h}{\partial s_0}\right)v_0 = 0 , \quad (61)$$

or

$$\frac{\partial^2 v_h}{\partial s_0 \partial s_h} + i2\pi K\beta'_h \frac{\partial v_h}{\partial s_0} + \pi^2 K^2 C^2 \chi_h \chi_{-h} v_h = 0 , \qquad (62)$$

(2) $\qquad\qquad v_i = 1 , \qquad i = 0, h , \qquad (63)$

on PA, including the point P, and

(3) $\qquad \dfrac{\partial v_i}{\partial s_h} = -i2\pi K\beta'_h v_i , \quad i = 0, h, \qquad (64)$

on PB.

Equations (60) can be reduced to simpler forms because of the boundary conditions (53), (55) and (57). For $\phi'_0(P)$, (60) becomes

$$\phi'_0(P) = \frac{1}{2}\left\{\Phi_0(A)v_0(A) + \Phi_0(B)v_0(B)\right\}$$
$$- \frac{1}{2}\int_{BA}\left\{\left[-\frac{\partial v_0}{\partial s_0}\frac{\partial s_0}{\partial\xi} + \left(\frac{\partial v_0}{\partial s_h} + i4\pi K\beta'_h v_0\right)\frac{\partial s_h}{\partial\xi}\right]\Phi_0 - \frac{\sin 2\theta_B}{\gamma_0(\xi)}v_0\frac{\partial s_h}{\partial\xi}\frac{\partial\Phi_0}{\partial\xi}\right\}d\xi .$$

Since the contribution to the integral of the last term, which is equal to $v_0 \partial\Phi_0/\partial\xi$, can be calculated by partial integration as

$$\frac{1}{2}\int_{BA}v_0\frac{\partial\Phi_0}{\partial\xi}d\xi = \frac{1}{2}\left\{\Phi_0(A)v_0(A) - \Phi_0(B)v_0(B)\right\} - \frac{1}{2}\int_{BA}\Phi_0\left(\frac{\partial s_0}{\partial\xi}\frac{\partial v_0}{\partial s_0} + \frac{\partial s_h}{\partial\xi}\frac{\partial v_0}{\partial s_h}\right)d\xi ,$$

and since $v_0(A) = 1$ from (63), $\phi'_0(P)$ turns out to be

$$\phi'_0(P) = \Phi_0(A) - \frac{1}{\sin 2\theta_B}\int_{BA}\left\{\frac{\partial v_0}{\partial s_h}(\xi) + i2\pi K\beta'_h(\xi)v_0(\xi)\right\}\gamma_0(\xi)\Phi_0(\xi)d\xi . \qquad (65)$$

The expression for $\phi'_h(\mathrm{P})$ has a simpler form since $\phi'=0$ on BA. It is found to be

$$\phi'_h(\mathrm{P})=-\frac{i\pi KC\chi_h}{\sin 2\theta_B}\int_{\mathrm{BA}}\gamma_0(\xi)v_h(\xi)\Phi_0(\xi)d\xi . \qquad (66)$$

Equations (65) and (66) show that each value of ϕ'_0 and ϕ'_h at P is given by an integral over a portion BA of the entrance surface, the integral involving the Riemann function associated with P and its derivatives. Since this Riemann function is determined by the distortion inside the triangle PAB the wave field at P is entirely determined by the distortion inside the triangle PAB and the incident wave falling between B and A. This results provides a mathematical basis for the column approximation in the electron case where this triangular area reduces to a thin column because of the small Bragg angle involved, as has been pointed out in § 1. We have seen there that $\overline{\mathrm{AB}}\approx 10\mathrm{A}$ in a typical experimental condition in electron diffraction. This means that the column approximation is applicable up to a part of the crystal where $\partial/\partial s_h(\boldsymbol{h}\cdot\boldsymbol{u})$ and $\partial^2/\partial s_0\partial s_h(\boldsymbol{h}\cdot\boldsymbol{u})$ are still almost constant within an interval 10A in length parallel to the surface. This condition would be satisfied at a region quite close to a dislocation.

Equations (65) and (66) give another important result that each of $\phi'_0(\mathrm{P})$ and $\phi'_h(\mathrm{P})$ is given by an integral in real space and is linear in $\Phi_0(\boldsymbol{r})$. This means that if the incident wave $\Phi_0(\boldsymbol{r})$ is made up of elementary component waves, i.e. $\Phi_0=\sum_i\Phi_0^{(i)}$, the resultant wave at P, $\phi'_0(\mathrm{P})$ and $\phi'_h(\mathrm{P})$, are expressed as a sum of the contributions $\phi_0'^{(i)}$ and $\phi_h'^{(i)}$ from each of the elementary components $\Phi_0^{(i)}$ as $\phi'_0=\sum_i\phi_0'^{(i)}$ etc. A convenient choice of the elementary wave in the present case is an infinitely narrow incident wave. Equations (65) and (66) can be written as

$$\phi'_i(\mathrm{P})=\int\varphi_i(\mathrm{P},\xi)\Phi(\xi)K\gamma_0(\xi)d\xi , \qquad (67)$$

where $i=0$ or h and

$$\varphi_0(\mathrm{P},\xi)=\frac{\delta(\xi_A-\xi)}{K\gamma_0(\xi)}$$
$$-\frac{1}{K\sin 2\theta_B}\left[\frac{\partial v_0}{\partial s_h}(\xi)+i2\pi K\beta'_h(\xi)v_0(\xi)\right], \qquad (68)$$

$$\varphi_h(\mathrm{P},\xi)=-i\frac{\pi C\chi_h}{\sin 2\theta_B}v_h(\xi) . \qquad (69)$$

$\varphi_0(\mathrm{P},\xi)$ and $\varphi_h(\mathrm{P},\xi)$ mean, respectively, the amplitudes of the transmitted and diffracted waves at P when an infinitely narrow wave

$$\Phi(\eta)=\frac{\delta(\eta-\xi)}{K\gamma_0(\xi)}$$

is incident at point Q(η) on the entrance surface. The expression (67) is important for the discussion of the integrated intensity as will be seen in § 10.

§ 9. Absorbing Crystal

The absorption of waves in the crystal can be treated by the well known methods of assigning complex values to $\chi(\boldsymbol{r})$.[30,32-34] The formulae derived in the previous sections are applicable also to this case, provided that χ_h's, ϕ_h's and other relevant quantities should now assume complex values. However, some comments may be helpful.

The real and imaginary parts of $\boldsymbol{k}_0$ are called $\boldsymbol{k}_{0,r}$ and $\boldsymbol{k}_i$, respectively. So that

$$\boldsymbol{k}_0=\boldsymbol{k}_{0,r}+i\boldsymbol{k}_i . \qquad (70)$$

From (2) and (8), $\boldsymbol{k}_h$ and $\boldsymbol{k}'_h$ can then be written as

$$\boldsymbol{k}_h=\boldsymbol{k}_{h,r}+i\boldsymbol{k}_i , \quad \boldsymbol{k}'_h=\boldsymbol{k}'_{h,r}+i\boldsymbol{k}_i , \qquad (71)$$

since $\boldsymbol{h}$ and $\boldsymbol{h}'$ are real vectors. The magnitude of $\boldsymbol{k}_0$ is still given by (9) which can be written

$$(1+\chi_0)K^2=\boldsymbol{k}_0^2=|\boldsymbol{k}_{0,r}|^2+i2(\boldsymbol{k}_{0,r}\cdot\boldsymbol{k}_i) , \qquad (72)$$

where $\boldsymbol{k}_i^2$ is neglected, since $\boldsymbol{k}_i$ is usually very small compared with $\boldsymbol{k}_{0,r}$. Similarly $\boldsymbol{k}'^2_h$ is given by

$$\boldsymbol{k}'^2_h=\boldsymbol{k}'^2_{h,r}+i2(\boldsymbol{k}'_{h,r}\cdot\boldsymbol{k}_i) . \qquad (73)$$

In the current dynamical theories $\boldsymbol{k}_i$ is taken as perpendicular to the entrance surface. This is necessary in order to connect the wave in the crystal to a plane wave in vacuum. Here, however, this restriction is not necessary because our incident wave can have any amplitune function provided that it should be of macroscopic variation. A convenient choice in the two-wave case is that $\boldsymbol{k}_i$ is parallel to the lattice plane associated with $\boldsymbol{h}'$, or

$$(\boldsymbol{h}'\cdot\boldsymbol{k}_i)=0^* , \qquad (74)$$

so that

$$(\boldsymbol{k}'_{h,r}\cdot\boldsymbol{k}_i)=(\boldsymbol{k}_{0,r}\cdot\boldsymbol{k}_i)=K^2\chi_{0,i}/2 , \qquad (75)$$

where $\chi_{0,i}$ is the imaginary part of χ_0. The imaginary part of β'_h, defined by (20) with (21) then vanishes and its real part still has the meaning given by (22). From (74) and (75) $|\boldsymbol{k}_i|$ is given by

$$|\boldsymbol{k}_i|=K|\chi_{0,i}|/2\cos\theta_B , \qquad (76)$$

where $|\boldsymbol{k}_{0,r}|=K$ is assumed and a small deviation in the angle between $\boldsymbol{k}_{0,r}$ and the lattice plane from the Bragg angle θ_B is neglected.

It should be remembered that the value of $|\boldsymbol{k}_i|$ given by (76) is just obtained from the definition

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* Since the difference between $\boldsymbol{h}'$ and $\boldsymbol{h}$ is very small, $\boldsymbol{k}_i$ is considered as perpendicular to $\boldsymbol{h}$.

of k_i as satisfying (72) and (74), and does not have physical meaning. The true absorption effect can only be obtained after the amplitudes ϕ_0' and ϕ_h' are calculated from the fundamental equations with complex χ_h. Analytical solutions of ϕ_0 and ϕ_h for perfect crystals can be obtained, as will be explained in a subsequent paper. It is shown that, for thick crystals, both ϕ_0 and ϕ_h may become very large, much larger than $\varPhi_0$ itself, when imaginary part is introduced into χ_h. This fact represents the anomalous transmission effect partly compensating the effect of diminuation in intensity due to the factor $\exp(-2\pi k_i \cdot r)$.

§ 10. Integrated Intensity

The integrated intensity of diffraction is usually referred to the total intensity of a particular reflection from a whole crystal integrated with respect to the incident angle over a range sufficient to cover a Bragg reflection. Here, however, it is referred to the intensity of a particular reflection emerging from a point P on the exit surface, integrated with respect to the incident angle, and is given by

$$\mathscr{I}_h(\mathrm{P}) = A\int|\psi_h'(\mathrm{P})\exp(-2\pi i[k_h \cdot r(\mathrm{P})$$
$$-h\cdot u(\mathrm{P})])|^2 d(\varDelta\varTheta)\,, \qquad (77)$$

where A is a scale factor, $r(\mathrm{P})$ the position vector of the point P and $\varDelta\varTheta$ is the angle of deviation of the incident beam from the Bragg incidence. The photographic density at each point on an X-ray diffraction topograph just corresponds to this quantity. Since the integrand of (77) is appreciable only when $\varDelta\varTheta$ is very small, the variable of integration can be transformed into the component $K_\perp$ of K normal to K_B which denotes the vector K at the Bragg incidence. Substitution of (66) and (71) into (77) gives

$$\mathscr{I}_h(\mathrm{P}) = A\frac{\pi^2 KC^2|\chi_h|^2}{\sin^2 2\theta_B}\int_{-\infty}^{\infty}\left\{\iint_{\mathrm{BA}}\gamma_0(\xi)\gamma_0(\xi')v_h(\xi)v_h^*(\xi')\varPhi_0(\xi)\varPhi_0^*(\xi')d\xi\,d\xi'\right\}\exp(4\pi k_i \cdot r(\mathrm{P}))dK_\perp\,. \qquad (78)$$

Here k_0 is defined as satisfying the condition (9) and the Bragg condition at the point where $(h\cdot u)=0$, and its imaginary part, k_i, the condition (74). Then β_h defined by (44) is always zero and

$$\beta_h' = \frac{1}{K}\frac{\partial}{\partial s_h}(h\cdot u)\,, \qquad (79)$$

does not depend on the angle of incidence. The effect of the choice of k_0 is compensated by the exponential factor in $\varPhi_0(\xi)$ given by (51). Thus $\gamma_0(\xi)$ and $v_h(\xi)$ which depend on k_h or β_h' are also independent of the angle of incidence. The factor $\varPhi_0(\xi)\varPhi_0^*(\xi')$ in (78) is given by

$$\varPhi_0(\xi)\varPhi_0^*(\xi') = \varPsi_0(\xi)\varPsi_0^*(\xi')\exp(-2\pi i[K_\perp(r_e-r_e')_\perp + K(r_e-r')_{s_0}])\exp(2\pi i[k_0\cdot r_e - k_0^*\cdot r_e'])\,, \qquad (80)$$

where $(r_e-r_e')_\perp$ means the component of (r_e-r_e') parallel to the plane of incidence and perpendicular to s_0 and $(r_e-r_e')_{s_0}$, the component parallel to s_0. The order of integration is now changed and that with respect to $K_\perp$ is carried out first, assuming that $\varPsi_0(\xi)$ does not involve $K_\perp$. This is valid for a plane wave but, in general, not for a spherical wave. However, the dependence of $\varPsi_0(\xi)$ on $K_\perp$ can be neglected, if the width of the incident beam is much smaller than $\sqrt{R_0 t_h}$, where R_0 is the radius of curvature of the wave front and t_h the extinction distance associated with the h-th reflection, as is shown in Appendix II. Since k_0 does not depend on the angle of incidence, the only factor in the integrand of (78) that depends on $K_\perp$ is $\exp(-2\pi i K_\perp(r_e-r_e')_\perp)$ which on integration gives $\delta(\xi-\xi')/\gamma_0(\xi)$, where $\delta(\xi-\xi')$ is Dirac's delta function. Thus the integral are easily carried out giving the result

$$\mathscr{I}_h(\mathrm{P}) = A\frac{\pi^2 KC^2|\chi_h|^2}{\sin^2 2\theta_B}\int_{\mathrm{BA}}\gamma_0(\xi)|v_h(\xi)|^2|\varPsi_0(\xi)|^2\exp(-\mu_m[r(\mathrm{P})-r_e(\xi)])d\xi\,, \qquad (81)$$

where

$$\mu_m = -4\pi k_i\,. \qquad (82)$$

The magnitude of μ_m is given by (76) and (82), its direction is parallel both to the lattice plane and to the plane of incidence.

Equation (81) can also be written as

$$\mathscr{I}_h(\mathrm{P}) = A\int|\varphi_h(\mathrm{P},\xi)|^2|\varPsi_0|^2\exp(-\mu_m\cdot[r(\mathrm{P})-r_e(\xi)])K\gamma_0(\xi)d\xi\,, \qquad (83)$$

where $\varphi_h(\mathrm{P}, \xi)$ is given by (69). This equation has a simple physical meaning. The amplitude of the diffracted wave given by (66) can be interpreted as a sum of contributions of the infinitely narrow elementary waves as is given in (67). The phase of $\Phi_0(\xi)$ given by (51) is, in general, different at different points on the entrance surface, since k_0 is now a constant vector and the tangential components of K and k_0 are not equal to each other even if the surface is plane, so that the effect of each elementary wave should be summed up with appropriate phase factor which varies with the angle of incidence. However, if the latter covers a sufficient range of angle, the values of phase angle will be distributed homogeneously over possible values, so that each elementary wave acts, in effect, as if they were incoherent with each other. Therefore the intensities rather than the amplitudes due to these elementary waves should be summed up, as is the case in (83).

The integrated intensity of the transmitted wave, $\mathscr{I}_0(\mathrm{P})$, is sometimes referred to. The integral obviously diverges. However, it has a practical meaning for crystals which show marked Borrmann effect, because in this case the transmitted intensity is negligible outside the range of the Bragg reflection. $\mathscr{I}_0(\mathrm{P})$ may be defined as the integral of the excess intensity from its normal value outside the range of the Bragg reflection, namely,

$$\mathscr{I}_0(\mathrm{P})=A\int(|\phi_0'(\mathrm{P})|^2-|\Phi_0(\mathrm{A})|^2)|\exp(-2\pi i k_0\cdot r(\mathrm{P}))|^2 d(\varDelta\Theta) \ . \tag{84}$$

Substitution of (65) into (84) and a similar derivation as before gives

$$\mathscr{I}_0(\mathrm{P})=A\int_{\mathrm{BA}}\left\{|\varphi_0(\mathrm{P},\xi)|^2-\left|\frac{\delta(\xi_A-\xi)}{K\gamma_0(\xi)}\right|^2\right\}|\Psi_0|^2\exp(-\mu_m[r(\mathrm{P})-r_e(\xi)])K\gamma_0(\xi)d\xi \ , \tag{85}$$

where $\varphi_0(\mathrm{P}, \xi)$ is given by (68).

§11. Intensity of X-Ray Projection Topograph

Equation (81) gives another important result concerning the intensity of the X-ray projection topograph,[35,36] *i.e.* the integrated intensity at a point P again integrated with respect to the position of the incident beam which sweeps the crystal surface. Total energy of the diffracted beam which emerges from the point P (Fig. 1) is given by

$$E_h=\int_{\mathrm{BA}}A\frac{\pi^2 K C^2|\chi_h|^2}{v\sin^2 2\theta_B}\left[\int_{\mathrm{BA}}A\gamma_0(\xi)|v_h(\xi)|^2|\Psi_0(\xi-\xi')|^2\exp(-\mu_m(r(\mathrm{P})-r_e(\xi)))d\xi\right]\frac{dx}{d\xi'}d\xi' \ ,$$

where v is the sweeping velocity and x is the coordinate parallel to the direction of sweeping (Fig. 4).

$$\mathscr{I}_h^{tr}=E_h v \ , \tag{86}$$

will be called the traverse intensity of the diffracted beam. By changing the order of integration, this turns out to be

$$\mathscr{I}_h^{tr}=\frac{\pi^2 K C^2|\chi_h|^2}{\sin^2 2\theta_B}\mathscr{I}_0\int_{\mathrm{BA}}\gamma_0(\xi)|v_h(\xi)|^2\exp(-\mu_m(r(\mathrm{P})-r_e(\xi)))d\xi \ , \tag{87}$$

where

$$\mathscr{I}_0=A\int_{\mathrm{BA}}|\Psi_0|^2 dx \ .$$

Equation (87) shows that the traverse intensity of the diffracted beam is proportional to the total energy of the incident X-rays which falls on the region between B and A (Fig. 1), and not dependent upon their intensity distribution, nor the shape of the wave front. Comparison of (87) with (81) shows that the intensity at a point in the projection topograph is the same as the integrated

intensity at the point due to an appropriate incident wave with the same total intensity. For the calculation of the intensity of a projection topograph, we have only to consider a plane incident wave, the simplest one.

This conclusion can be derived from the two conditions: (1) the wave function at a point on the exit surface is linear in the wave function of the incident wave. (2) the intensity at a point on the X-ray diffraction topograph is the integrated intensity: *i.e.* the angular width of the incident

beam at a point on the entrance surface is sufficiently wide as to cover the whole range of a Bragg reflection. (1) is always satisfied and (2) in usual experimental conditions. The conclusion is valid quite generally: it is valid *e.g.* for crystals with distortions which is not very small, for those with discontinuities such as stacking faults and for those which give moiré patterns.

§ 12. Discussion

The condition for the validity of the assumptions used in deriving the fundamental equations (19) in the electron case and (39) in the X-ray case are now considered quantitatively.

(i) In the electron case $\nabla^2\phi_g$ and $\nabla^2(g\cdot u)\phi_g$ are neglected in deriving (19), compared with the other terms in the curly bracket of (17). If the variation in ϕ_g is sinusoidal, so that it is represented by

$$\phi_g = B\exp(-2\pi i\boldsymbol{\kappa}\cdot\boldsymbol{r}) ,$$

$\nabla^2\phi_g$ is given by

$$\nabla^2\phi_g = -4\pi^2\kappa^2\phi_g .$$

In order that this should be negligible compared with *e.g.* the last term in the curly bracket in (17), κ should satisfy

$$|\kappa|^2 \ll K^2|\chi_h| , \qquad (88)$$

where χ_h is the largest Fourier coefficient of χ in the summation over g'. This condition can be written

$$\Lambda = \frac{1}{|\kappa|} \gg l , \qquad (89)$$

where

$$l = 1/K\sqrt{\chi_h} \approx \sqrt{\lambda t_h} , \qquad (90)$$

t_h being the extinction distance for the $\boldsymbol{h}$-th reflection. If the variation in ϕ_g is of more general type, it may be expanded in a Fourier-like series

$$\phi_g = \sum_n B_n\exp(-2\pi i\boldsymbol{\kappa}_n\cdot\boldsymbol{r}) , \qquad (91)$$

where $\boldsymbol{\kappa}_n$ is a wave vector not necessarily satisfying the condition (2) for k_n. $\nabla^2\phi_g$ can then be neglected if

$$\Lambda_m \gg l , \qquad (92)$$

where Λ_m is the smallest value among $\Lambda_n = 1/|\kappa_n|$. Equation (92) shows that $\nabla^2\phi_g$ can be neglected if the value of ϕ_g is almost constant over a range l given by (90).

In order that $\nabla^2(g\cdot u)\phi_g$ should be negligible u should satisfy

$$|\nabla^2(g\cdot u)| \ll K^2|\chi_h| , \qquad (93)$$

where χ_h is the same as that in (88). This condition is satisfied, if $(g\cdot u)$ is almost constant over

the range l, the same as that for ϕ_g.

In the X-ray case, if D_g and $(g\cdot u)$ satisfy the same condition as for ϕ_g' and $(g\cdot u)$ in the electron case, it can easily be shown that the estimation of the order of magnitude of the quantities shown in Table II is valid.

(ii) When ϕ_g', D_g and $(g\cdot u)$ satisfy this condition, the condition (ii), given in § 4, that remaining terms in the curly brackets in (17) and (37) are almost constant over a unit cell is clearly satisfied, because the lattice constant is much smaller than, in the X-ray case, and of the same order of, in the electron case, the distance l.

To sum up, in order that the approximation used in the present theory is valid, the term "macroscopic variatoin" should mean a slowly varying function which is almost constant within a range l given by (90).

Examples of the value of l for both the electron and X-ray cases are given in Table III. It is seen

Table III. Distance $l = 1/K\sqrt{|\chi_h|} = \sqrt{\lambda t_h}/\cos\theta_B$ for X-ray and electron cases. The value for X-rays is not dependent upon the wavelength.

	hkl	X-rays	Electron (100 kV)
Si	111	0.055$^\mu$	4.8 Å
	220	0.050	5.3
Ge	111	0.035	4.2
	220	0.031	4.3
Al	111	0.045	4.5
	220	0.050	6.2

that the value of l is much smaller than the resolving power of the X-ray diffraction topograph, and of the same order of that of the electronmicrograph. The present theory, therefore, is valid for most of the images of defects obtained by these techniques.

For the validity of the theories developed by Penning-Polder,[11] Kato[12] and Kambe[13] the intensity should be almost constant within ranges much larger than the extinction distance which is several tens of microns and several hundreds Angstrom in the X-ray and electron cases, respectively. These limits are too large for detailed explanation of dislocation images.

Applicability of the equations of Howie and Whelan and those of Wilkens is the same as the present one, provided that the column approximation should hold, because Howie and Whelan's equations are the special case of the fundamental equations of the present theory, and it is easily

shown that equivalent equations to those of Wilkens' can be derived from Howie and Whelan's equations. The validity of the column approximation, however, requires that the distortion should be almost homogeneous within ranges comparable with $\overline{AB}$ shown in Fig. 1. This requirement may not be important for thin crystals, but very serious for thick ones.

§ 13. Summary

(1) A set of partial differential equations for the amplitudes (and phases) of the transmitted and diffracted waves in electron-diffraction within distorted crystals are deduced from the Schrödinger equation by a simple assumption as to the slow variation of distortions. (eq. 19).

(2) In the case of lamellar structures and plane wave incidence these equations reduce to ordinary differential equations (eq. 23) which are equivalent to those deduced by Howie and Whelan. These equations can be written in a tensorial form (eq. 24).

(3) For perfect crystals this tensor equation gives an intensity formula for many wave cases (eq. 26) which is given by Sturkey, Niehrs and Fujimoto.

(4) A set of partial equations for X-ray case can similarly be obtained from the Maxwell equations under the same assumptions described in (1) (eq. 39).

(5) In the case of two waves, these sets of equations for X-ray and Electrons can be written in a common from (eq. 40).

(6) Three sets of equations for the two-wave case are provided: those with wave vectors based upon local reciprocal lattice (eq. 40), upon reciprocal lattice of ideal crystals (eq. 42) and wave vectors employed in the kinematical theory (eq. 47).

(7) The boundary conditions for the Laue case with two strong waves are given (eqs. 53, 55 and 57). These are valid for curved surfaces and for any type of incident waves.

(8) The fundamental equations for the two wave case common to X-ray and electron cases can be solved by the method of Riemann. The transmitted and diffracted waves at a point P on the exit surface are given by the distortion inside the triangular area PAB (Fig. 1) and the incident wave which falls upon BA. Each of them is given by an integral over the entrance surface between B and A, and is linear in the displacement of the incident wave.

(9) The validity of the column approximation in the electron case is proved.

(10) The integrated intensities at the point P is also given by an integral over the entrance surface between B and A. They are linear in the intensity of the incident wave (eqs. 81 and 85).

(11) The intensity of a projection topograph (traverse pattern) at P (Fig. 1) is given (eq. 87). It is given by the total intensity which falls upon the surface between B and A, not dependent upon the intensity distribution nor upon the type of the incident wave.

(12) The limit of the validity of the solution for both the X-ray and electron cases are given (eq. 90, Table III).

Appendix I

Let a_i be a basis vector of a unit cell situated at r_0. By a displacement represented by $u(r)$, a_i transforms into

$$a_i'(r) = \left(1 + \left(\frac{\partial u}{\partial r}\right)_{r_0}\right) a_i(r_0) ,$$

where a_i and a_i' are considered as one column matrices and $\partial u/\partial r$ is a matrix with components $(\partial u/\partial r)_{ij} = \partial u_i/\partial x_j$. The basis vectors at r of the reciprocal lattice for the distorted crystal may be defined as in the ordinary way by

$$b_j' \cdot a_i' = \delta_{ji} .$$

This relation is satisfied if b_j' is defined by

$$b_j' = b_j \left(1 + \left(\frac{\partial u}{\partial r}\right)_{r_0}\right)^{-1} ,$$

$$= b_j \left(1 - \left(\frac{\partial u}{\partial r}\right)_{r_0}\right) ,$$

where b_j and b_j' are now one row matrices, b_j being the basis vector for the undistorted state. Any b_j transforms in the same way and so does any reciprocal lattice vector. This transformation can also be written as

$$h' = h - \text{grad} \, (h \cdot u(r_0)) .$$

Appendix II

A spherical wave incident on a crystal surface may be expressed as

$$\phi = \frac{\text{Const.}}{|\boldsymbol{R}+\boldsymbol{r}|} \exp\left(-2\pi i K |\boldsymbol{R}+\boldsymbol{r}|\right),$$

where K is the wave number, $\boldsymbol{R}$ the position vector drawn from the source to the coordinate origin taken on the surface and $\boldsymbol{r}$ a position vector referred to the origin. The expansion of $|\boldsymbol{R}+\boldsymbol{r}|$ in power series gives

$$\phi = \Psi(\boldsymbol{r}) \exp\left(-2\pi i \boldsymbol{K}_0 \cdot \boldsymbol{r}\right)$$

with

$$\Psi(\boldsymbol{r}) = \text{Const.} \frac{\exp\left(-2\pi i K R\right)}{R}$$
$$\times \exp\left(-2\pi i \frac{K r^2 \cos^2 \Theta}{2R} + \cdots\right),$$

where $\boldsymbol{K}_0 = K\boldsymbol{R}/|R|$ is the wave vector at the origin and Θ the angle which $\boldsymbol{K}_0$ makes with the surface normal. The higher terms in the argument of the second term in the exponential can be neglected in usual experimental conditions. The first term is not negligible and depends upon Θ or $K_\perp$ given in the text. However, its variation with Θ is very small, if Θ varies within the width $\Delta\theta_B$ of the Bragg reflection, *i.e.*

$$\left| \Delta\left(\frac{2\pi K r^2 \cos^2 \Theta}{2R}\right) \right| = 2\pi \left| \frac{K r^2 \sin\Theta \cos\Theta}{R} \Delta\Theta \right|$$
$$\leq 2\pi \frac{r^2 \cos\Theta}{R t_h} \ll 2\pi,$$

if

$$|\Delta\Theta| < \Delta\Theta_B = 1/K t_h \sin\theta_B, \quad \text{and} \quad r \ll \sqrt{R t_h},$$

where t_h is the extinction distance. This last condition is well satisfied in usual experiments in X-ray topography; *e.q.* $\sqrt{R t_h} = 2$mm, for $R = 40$ cm and $t_h = 10\mu$.

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