Substitution of (42) into (38) now gives s and s_0 which, in this case, immediately produce the n_i through (24) since R^{-1} is the identity operation. Because reflection to a given point on the film occurs twice, corresponding to the $\pm$ ambiguity in equation (42), both diffraction conditions must be considered in an absorption correction.

I would like to express to Prof. B. C. Carlson my appreciation for his several, valuable discussions concerning coordinate systems.

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A Theoretical Study of Pendellösung Fringes. Part I. General Considerations

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The assumption of an incident plane wave is shown to be not adequate for single-crystal experiments of X-ray diffraction (Laue case). A dynamical theory of diffraction is formulated for a general type of monochromatic incident wave. Fundamental aspects of wave behavior are discussed in terms of wave-bundle considerations. Diffraction phenomena are classified by $\Delta\theta$ (an angular width of single-crystal reflection) and Ω_0 (a width in which the angular spectrum of an incident coherent wave takes an appreciable value). If $\Delta\theta \gg \Omega_0$, a plane-wave assumption is adequate. This is usually the case for electron diffraction. If $\Delta\theta \ll \Omega_0$, a spherical wave assumption is more appropriate and most of X-ray cases fall under this alternative. Furthermore, a criterion is given to distinguish between Fresnel and Fraunhofer diffraction in a crystalline medium. 'Pendellösung' fringes of X-rays (Kato & Lang, 1959) can be interpreted as Fraunhofer diffraction, while those of electrons are observed in a Fresnel diffraction region. The essential features of section patterns, particularly 'hook-shaped' fringes, can now be explained.

1. Introduction

In previous papers the first observations of X-ray Pendellösung fringes were reported (Lang, 1959; Kato & Lang, 1959). In addition, new types of diffraction fringes were obtained in section topographs under the experimental conditions fully described. These fringes are essentially due to interference between two kinds of crystal waves which correspond to different branches of the dispersion surface. Thus they have to be explained by a dynamical theory of diffraction.

'Pendellösung' interference effects were discovered first in electron-microscope experiments and could be well explained by dynamical theory.* Thus it seemed quite natural to apply this theory to X-rays because it is generally accepted that the theory is essentially the same for both electron and X-ray diffraction. However, as shown briefly in the previous paper (Kato & Lang, 1959), section patterns cannot be interpreted in a straightforward manner by the usual dynamical theory. In fact, they imply that we have to consider a divergent coherent wave instead of an

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* A detailed historical survey is given in the previous paper (Kato & Lang, 1959).

ideal plane wave as the incident wave (Kato, 1960b). The same is true for the general X-ray case, as shown in Section 2 by a simple argument. Hence, we must formulate the dynamical theory for a general type of incident monochromatic wave (Section 3). This is the main object of the present paper. In the following sections, only fundamental aspects of wave behavior are discussed on the basis of wave bundle considerations. A further development of the theory and detailed discussion of 'Pendellösung' phenomena will be reserved for the next paper.

2. Preliminary considerations

The usual dynamical theory may be summarized as follows. First, we assume a plane wave as an incident wave (PW assumption). As crystal waves we consider a sort of Bloch wave function. This is a general type of wave in a medium of periodically distributed scatterers. The incident wave and the crystal waves are connected by boundary conditions including the tangential continuity of wave vectors at the surfaces of the crystal (TC assumption). In the surrounding vacuum, we

obtain waves which correspond to the observed transmitted and diffracted waves.

We are here concerned with electron-microscope experiments and X-ray section topograph experiments. The main purpose of this section is to criticize the PW and TC assumptions in both cases.

(a) Plane wave assumption

If we pick up a coherent wave in an incident beam it is obvious that such a wave is a single spherical wave modified by a collimation system in the X-ray case, and a modified plane wave produced by a condenser lens in the electron case. The problem is to specify to what extent these waves may be approximated by an ideal plane wave. Two conditions must be satisfied for such an approximation. The first is

$$\varDelta\theta \gg \varOmega_0 , \tag{1}$$

where $\varDelta\theta$ is an angular width of reflection based upon the ordinary dynamical theory and $\varOmega_0$ is an angle in which the angular spectrum of the coherent incident wave takes an appreciable value. If condition (1) does not apply, we are no longer restricted to considering only plane waves making a given angle of incidence upon the crystal.

$\varOmega_0$ can be approximated by the geometrical angular width of the incident beam, w, if it is not extremely small. In the electron case $\varDelta\theta \simeq 10^{-2}$ rad. and w may be less than 10^{-3}. On the other hand, in the X-ray case $\varDelta\theta \simeq 10^{-5}$ and $w \ll 10^{-4}$ under ordinary experimental conditions of single-crystal diffraction. Thus (1) is in general satisfied nicely for electrons but not for X-rays.

The second condition is

$$f \gg s , \tag{2}$$

where f is a width in which the phase of the incident wave along a plane may be considered approximately constant, and s is an effective crystal size. In electron diffraction f is of the order of 200μ (the size of a diaphragm placed in the condenser lens) and s is of the order of 10^3 Å. Hence condition (2) is easily satisfied.

In the X-ray case, f should be considered to be of the order of the diameter of the first Fresnel zone of a spherical wave, i.e., $2\sqrt{(\lambda L)}$, where λ is the wave length and L is the distance between the X-ray source and the specimen. The effective crystal size s should be of order $L\varDelta\theta$. Thus, it turns out that the condition (2) may be expressed as

$$2\sqrt{(\lambda/L)} \gg \varDelta\theta . \tag{2'}$$

If we assume a set of reasonable figures, $\lambda = 10^{-8}$ cm., $L = 30$ cm. and $\varDelta\theta \simeq 10^{-5}$ rad., condition (2') is barely satisfied.

(b) Tangential continuity assumption

The TC assumption is equivalent to assuming an infinitely extended crystal surface. A limited lateral size s causes diffraction at the surface. The angular broadening of wave vectors due to this diffraction effect may be estimated crudely as λ/s according to well-known optical principles. For this broadening to be neglected it is necessary that

$$\varDelta\theta \gg \lambda/s . \tag{3}$$

In the electron case, the wave length is of the order of 0·05 Å. Thus, again condition (3) is well satisfied. In the X-ray case, again estimating s as $L\varDelta\theta$, it turns out that condition (3) is equivalent to

$$\varDelta\theta \gg \sqrt{(\lambda/L)} . \tag{3'}$$

This is the reverse condition of (2') except for a numerical factor of no great significance. As seen above, in most X-ray cases the TC assumption is not fulfilled.

Actually, the TC assumption loses its meaning in a case where the PW assumption is not adequate. Further, equation (3) or (3') imply that even if the geometrical aperture w can be reduced to less than $\varDelta\theta$ by a narrow slit system, we have to give up the TC assumption because s should then be estimated by Lw. We will discuss this point again in Section 6.

3. General formulation

(a) Incident waves

Firstly, we consider a scalar wave field such as is applicable to electrons. We take as incident wave

$$\varPhi = \iint_{-\infty}^{+\infty} F(\hat{\mathbf{K}}) \exp i(\mathbf{K}\cdot\mathbf{r})dK_x dK_y , \tag{4}$$

where K_x and K_y are x and y components of the wave vector $\mathbf{K}$, and $F(\hat{\mathbf{K}})$ is a weight function expressing an angular spectrum of the wave field $\varPhi$. Equation (4) is the most general form for waves traveling along the positive z direction which satisfies the Helmholtz wave equation. (See Stratton, 1941a.) Three examples of F may be considered:

(i) $$F_p(\hat{\mathbf{K}}) = \delta(\hat{\mathbf{K}} - \hat{\mathbf{K}}_e) . \tag{5}$$

In this case, obviously, equation (4) means a plane wave. With such, the present theory becomes equivalent to the ordinary plane-wave theory.

(ii) $$F_L(\hat{\mathbf{K}}) = \iint_S \exp i\{(\mathbf{K} - \mathbf{K}_e)\cdot\mathbf{r}_s\}d\mathbf{r}_s , \tag{6}$$

where S means the slit aperture or the incident surface and the range of integration covers the area S. This formalism becomes essentially the same as that of the dynamical theory for a polyhedral crystal in the electron case. (See Kato, 1952a.)

(iii) The spherical wave, $\varPhi_s$. Using the standard Fourier representation of a spherical wave we can show that

$$\Phi_s = \exp iKr/4\pi r \qquad (7)$$

$$= \frac{i}{8\pi^3} \iiint_{-\infty}^{+\infty} \frac{\exp i(K_x x + K_y y + K_z z)}{K_x^2 + K_y^2 + K_z^2 - K^2} \, dK_x dK_y dK_z$$

$$= \frac{1}{8\pi^2} \iint_{-\infty}^{+\infty} \frac{\exp i(\mathbf{K} \cdot \mathbf{r})}{K_z} \, dK_x dK_y \; . \qquad (8)$$

Since we are only concerned with a traveling wave of finite amplitude along the $+z$ direction, K_z is assumed to be real positive or imaginary positive. Comparing with equation (4), it turns out that for a spherical wave

$$F_s(\widehat{\mathbf{K}}) = i/(8\pi^2 K \cos (\mathbf{K} \wedge \mathbf{z})) \; . \qquad (9)$$

Now the main application of the present theory is to the X-ray case. Thus we have to consider electromagnetic fields. The most general form of transverse vector fields can be expressed by a combination of the following functions:

$$\mathbf{M} = \iint_{-\infty}^{+\infty} F(\widehat{\mathbf{K}})[\mathbf{a} \times \widehat{\mathbf{K}}] \exp i(\mathbf{K} \cdot \mathbf{r}) dK_x dK_y$$

$$\mathbf{N} = \iint_{-\infty}^{+\infty} F(\widehat{\mathbf{K}})[[\mathbf{a} \times \widehat{\mathbf{K}}] \times \widehat{\mathbf{K}}] \exp i(\mathbf{K} \cdot \mathbf{r}) dK_x dK_y, \quad (10)$$

where $\mathbf{a}$ is an arbitrary constant vector. (Stratton, 1941b). If we use $\mathbf{M}$ for expressing a magnetic vector $\mathbf{H}$, $\mathbf{N}$ represents the corresponding electric vector $\mathbf{E}$ since $\mathbf{E}$ is proportional to rot $\mathbf{H}$.

Electromagnetic waves emitted from an atomic source can be expressed to order $1/r$ as follows (see Shiff, 1948)

$$\mathbf{H}_s = iK\Phi_s[\mathbf{J} \times \widehat{\mathbf{r}}]$$

$$\mathbf{E}_s = iK\Phi_s[[\mathbf{J} \times \widehat{\mathbf{r}}] \times \widehat{\mathbf{r}}] \; , \qquad (11)$$

where $\mathbf{J}$ is a constant vector proportional to an induced current due to a spontaneous transition of an atom. If we define operators

$$\widetilde{M} = [\mathbf{J} \times \nabla]$$

$$\widetilde{N} = 1/(iK)[[\mathbf{J} \times \nabla] \times \nabla] \qquad (12)$$

we can show that

$$\mathbf{H}_s \simeq \widetilde{M}\Phi_s$$

$$\mathbf{E}_s \simeq \widetilde{N}\Phi_s \qquad (13)$$

neglecting quantities of order $1/r^2$.
Therefore,

$$\mathbf{H}_s \simeq iK \iint_{-\infty}^{+\infty} F_s(\widehat{\mathbf{K}})[\mathbf{J} \times \widehat{\mathbf{K}}] \exp i(\mathbf{K} \cdot \mathbf{r}) dK_x dK_y$$

$$\mathbf{E}_s \simeq iK \iint_{-\infty}^{+\infty} F_s(\widehat{\mathbf{K}})[[\mathbf{J} \times \widehat{\mathbf{K}}] \times \widehat{\mathbf{K}}] \exp i(\mathbf{K} \cdot \mathbf{r}) dK_x dK_y \; . \qquad (14)$$

As shown in the Appendix, similar relations to equation (13) hold between scalar and vector wave fields which are diffracted by the same slit system provided that Kirchhoff's approximation is assumed

concerning the boundary conditions. Thus, we can easily construct the vector field from the scalar field.

In the following, for simplicity, we treat a single Bragg reflection. Therefore, we can separate the vector field into two polarization components: one parallel and the other perpendicular to the net plane concerned. Thus, each field component can be treated as a scalar wave. Including polarization factors such as $[\mathbf{J} \times \widehat{\mathbf{K}}]$ and $[[\mathbf{J} \times \widehat{\mathbf{K}}] \times \widehat{\mathbf{K}}]$ in the weight function $F(\mathbf{K})$, each polarization component has generally the form shown by equation (4). Therefore, we treat hereafter scalar wave fields only. There is no loss of generality.

(b) Waves in the crystal

As shown in equation (4), a general type of incident wave can be expressed as a superposition of plane waves. Each component wave excites a set of waves in the crystal which can be predicted correctly by the ordinary dynamical theory for a plane wave. The amplitude densities of the component crystal waves are as follows:

Transmitted waves:

$$d_0^{(1)} = C_0^{(1)} \exp i\{(\mathbf{K} - \mathbf{k}_0^{(1)}) \cdot \mathbf{r}_e\}$$

$$d_0^{(2)} = C_0^{(2)} \exp i\{(\mathbf{K} - \mathbf{k}_0^{(2)}) \cdot \mathbf{r}_e\} \qquad (15)$$

Diffracted waves:

$$d_g^{(1)} = C_g^{(1)} \exp i\{(\mathbf{K} - \mathbf{k}_0^{(1)}) \cdot \mathbf{r}_e\}$$

$$d_g^{(2)} = C_g^{(2)} \exp i\{(\mathbf{K} - \mathbf{k}_0^{(2)}) \cdot \mathbf{r}_e\} \; , \qquad (16)$$

where $k_0^{(j)}$ ($j = 1$ or 2) are the transmitted wave vectors; these satisfy the equation of the so-called dispersion surface,

$$\begin{vmatrix} (k_0^2 - K^2) & K^2 \sin \chi \psi_g \\ K^2 \sin \chi \psi_{\bar{g}} & (k_g^2 - K^2) \end{vmatrix} = 0 \qquad (17)$$
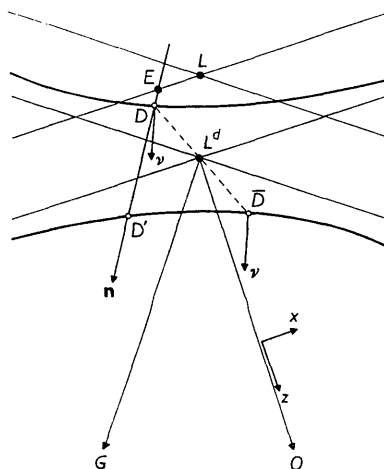

Fig. 1. Dispersion surfaces and coherent wave points. $L = $ Laue point of kinematical theory. $L^d = $ Laue point of dynamical theory. $n = $ normal of the incident surface. $\nu = $ direction of wave propagation. Interference occurs between D- and D'-wave in electron cases (ordinary plane wave theory) and between D- and $\bar{D}$-wave in X-ray cases (spherical wave theory).

with

$$\mathbf{k}_g = \mathbf{k}_0 + 2\pi\mathfrak{g} \ . \tag{18}$$

In this expression $\sin\chi$ is the polarization factor resulting from crystal reflection and ψ_g is the $\mathfrak{g}$th order Fourier coefficient of polarizability (X-ray case) or crystal potential (electron case). Equation (17) represents a surface of fourth order in $\mathbf{k}_0$ having a rotation axis $\mathfrak{g}$. Usually, however, it is approximated by a cylindrical hyperbolic surface near the vicinity of a Bragg reflection as shown in Fig. 1. The vectors $\mathbf{k}_0^{(j)}$ and $\mathbf{K}$ are connected by a TC-condition, so that $\mathbf{K} - \mathbf{k}_0^{(j)}$ is always perpendicular to the incident surface. The incident surface is specified by the radius vector $\mathbf{r}_e$. Also, $C_0^{(j)}$ and $C_g^{(j)}$ are the amplitudes of crystal waves. Their explicit expressions will be given in the next paper as functions of $\mathbf{K}$.

Thus, the crystal wave for a general type of incident wave can be expressed by a superposition of waves of types (15) and (16) as follows:

Transmitted waves:

$$\varphi_0^{(j)} = \iint_{-\infty}^{+\infty} F(\widehat{\mathbf{K}}) d_0^{(j)} \exp i(\mathbf{k}_0^{(j)} \cdot \mathbf{r}) dK_x dK_y \ . \tag{19}$$

Diffracted waves:

$$\varphi_g^{(j)} = \iint_{-\infty}^{+\infty} F(\widehat{\mathbf{K}}) d_g^{(j)} \exp i(\mathbf{k}_g^{(j)} \cdot \mathbf{r}) dK_x dK_y \ . \tag{20}$$

(c) *Vacuum waves from the exit surface of the crystal*

In a similar way we can easily write the vacuum waves which correspond to the observed transmitted and diffracted waves. The results are

$$\varPhi_0^{(j)} = \iint_{-\infty}^{+\infty} F(\widehat{\mathbf{K}}) d_0^{(j)} \exp i\{(\mathbf{k}_0^{(j)} - \mathbf{K}_0) \cdot \mathbf{r}_a\} \exp i(\mathbf{K}_0 \cdot \mathbf{r})$$
$$\times dK_x dK_y \ . \tag{21}$$

$$\varPhi_g^{(j)} = \iint_{-\infty}^{+\infty} F(\widehat{\mathbf{K}}) d_g^{(j)} \exp i\{(\mathbf{k}_g^{(j)} - \mathbf{K}_g) \cdot \mathbf{r}_a\} \exp i(\mathbf{K}_g \cdot \mathbf{r})$$
$$\times dK_x dK_y \ , \tag{22}$$

where

$$|\mathbf{K}_0| = |\mathbf{K}_g| = K \ . \tag{23}$$

The vectors $\mathbf{K}_0$ and $\mathbf{K}_g$ are related to $\mathbf{k}_0^{(j)}$ and $\mathbf{k}_g^{(j)}$ by the TC-conditions at the exit surface of the crystal, and the vector $\mathbf{r}_a$ indicates a point in the exit surface.

4. Wave bundle considerations

According to equations (19) and (20) the crystal waves have the form

$$\varphi = \iint_{-\infty}^{+\infty} H(\widehat{\mathbf{K}}) \exp i(\mathbf{K} \cdot \mathbf{r}_e) \exp i\{\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}_e)\} dK_x dK_y \ . \tag{24}$$

where $H(\widehat{\mathbf{K}})$ stands for either $F(\widehat{\mathbf{K}})C_0^{(j)}$ or $F(\widehat{\mathbf{K}})C_g^{(j)}$ and $\mathbf{k}$ stands for $\mathbf{k}_0$ or $\mathbf{k}_g$. Generally this is a very complicated wave field which, however, becomes simple at positions deep inside the crystal. In order to see

this, consider a wave bundle whose wave vectors are specified by wave points distributed within a region δK_x around a point D (see Fig. 1). Since the dispersion surface can be approximated by a cylindrical surface, the crystal wave may also be approximated essentially by a cylindrical wave. Thus, for the present, we consider the problem in the x–z plane which is perpendicular to the cylindrical axis. If we take δK_x sufficiently small, we may approximate as follows:

$$H(\widehat{\mathbf{K}}) = H(D)$$
$$(\mathbf{K} \cdot \mathbf{r}_e) = \{\mathbf{K}(E) \cdot \mathbf{r}_e^*\}$$
$$\mathbf{k} = \mathbf{k}(D) + \mathbf{\tau} \ , \tag{25}$$

where E is the wave point of an incident plane wave connected with D by the TC condition. The vector $\mathbf{r}_e^*$ is that particular vector $\mathbf{r}_e$ which is parallel to $\mathbf{K}(E)$. The vector $\mathbf{\tau}$ lies in the tangential plane of the dispersion surface at D. Thus, the wave bundle due to δK_x is

$$\delta\varphi = H(D) \exp i\{\mathbf{K}(E) \cdot \mathbf{r}_e^*\} \exp i\{\mathbf{k}(D) \cdot (\mathbf{r} - \mathbf{r}_e^*)\}$$
$$\times \int_{\delta K_x} \exp i\{\mathbf{\tau} \cdot (\mathbf{r} - \mathbf{r}_e^*)\} dK_x \ . \tag{26}$$

This integral takes an appreciable value only when $\mathbf{r} - \mathbf{r}_e^*$ is nearly parallel to the normal $\mathbf{\nu}$ of the $\mathbf{\tau}$-plane. Thus, the wave bundle $\delta\varphi$ propagates in the direction $\mathbf{\nu}$. This result is essentially the same as that discussed previously for electron diffraction (Kato, 1952b).

The lateral width, g, of this wave bundle defined by equation (26) is given crudely by $1/\delta K_x$ in accord with usual optical-diffraction principles. With increase of δK_x, however, approximations (25) no longer hold, and the wave bundle becomes broader due to bending of the dispersion surface. Actually, if we neglect the diffraction effect (broadening due to limitation of δK_x) the geometrical beam size g' would be determined by $l = |\mathbf{r} - \mathbf{r}_e^*|$ times the angular range W of directions $\mathbf{\nu}$ within the region δK_x. In fact, we have always a certain minimum size g_m determined by balancing the geometrical beam size g' and diffraction width $1/\delta K$, i.e.

$$g_m = l W_m \equiv l W(\omega_m) = 1/(K\omega_m) \ , \tag{27}$$

where ω_m stands for that particular value of the angular width $(\delta K_x / K)$ which satisfies condition (27).

The waves outside ω_m do not essentially interfere with the waves inside ω_m; thus, g_m gives an order of magnitude of the lateral dimension of coherence of a wave bundle which propagates in a given direction.

Since g and g' depend upon K and l, and the functional form of W depends upon the point D, g_m also depends upon l, K and D. It should be expected that g_m increases with l and λ, and that it is a maximum at the apex point of the dispersion surface. In Fig. 2, some examples of g and g' are shown. They are calculated on the assumptions $\Delta\theta = 10^{-6}$ rad. and 10^{-5}

rad., $K = 2\pi \times 10^8$ cm.$^{-1}$, $\theta_B = 10^{-1}$ and D being an apex point. It will be seen that the minimum values, g_m, are larger than a few microns in regions deeper than 0·1 mm. within the crystal.
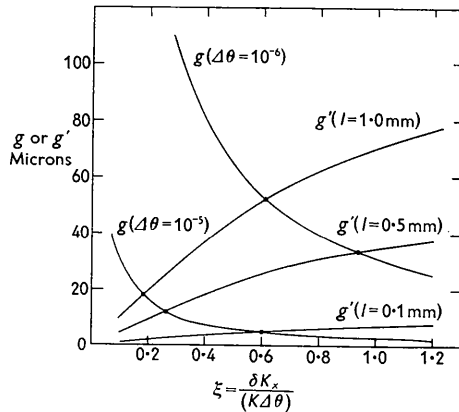

Fig. 2. The diffraction size g and geometrical size g' of a wave bundle. Curves are based upon equations: $g = 2/(K\Delta\theta\xi)$, $g' = l\xi \tan \theta_B/(1+\xi^2)^{\frac{1}{2}}$, and values $\tan \theta_B = 0\cdot 1$, $K = 2\pi \times 10^8$ cm.$^{-1}$. Here l is the distance of wave propagation in the crystal. Minimum beam sizes are given by the ordinates of the intersections of the g and g'-curves.

Next we consider a conjugate wave bundle which corresponds to the conjugate point $\bar{D}$ of D with respect to the Laue point in Fig. 1. This wave bundle propagates in the same direction as the bundle at D. The difference between the centers of both wave bundles may be estimated as

$$|\mathbf{r}_e^* - \bar{\mathbf{r}}_e^*| \sim L\Delta\theta . \tag{28}$$

As explained in Section 2, it is less than a few microns for the X-ray case. Thus, in most parts of the crystal we find

$$|\mathbf{r}_e^* - \bar{\mathbf{r}}_e^*| < g_m . \tag{29}$$

This means that we can expect interference only between the conjugate bundles. Equation (26) and similar formulae for the conjugate bundle show that fringe separations in a direction of propagation $\mathbf{v}$ are given by

$$\Lambda \simeq 2\pi/\{(\mathbf{k} - \bar{\mathbf{k}}) \cdot \mathbf{v}\} \tag{30}$$

neglecting the difference between $\mathbf{r}_e^*$ and $\bar{\mathbf{r}}_e^*$. The above theory describes in their essentials the hook-shaped fringes which were obtained in section patterns (Kato & Lang, 1959). Detailed discussion will be given in the subsequent paper.

5. Fraunhofer diffraction in a crystalline medium

In a vacuum, the diffracted waves due to a slit can be written as follows:

$$\Phi = \iint_{-\infty}^{+\infty} F(\hat{\mathbf{K}}) \exp i(\mathbf{K} \cdot \mathbf{r}_s) \exp i\{\mathbf{K} \cdot (\mathbf{r} - \mathbf{r}_s)\} dK_x dK_y , \tag{31}$$

where $F(\hat{\mathbf{K}})$ is given by either equation (A–2) or equation (6).

The angular width Ω_0 within which $F(\hat{\mathbf{K}})$ has an appreciable value is related to the slit width S_0 as follows, if the slit width S_0 is small enough,

$$S_0 \simeq 1/K\Omega_0 . \tag{32}$$

The condition of Fraunhofer diffraction is given by

$$R\Omega_0 \gg S_0 , \tag{33}$$

where R is the distance between slit and observation point. Under this condition the size of the slit can be neglected compared with the diffraction pattern, and the angular distribution of $F(\hat{\mathbf{K}})$ corresponds to the angular distribution of $\Phi(\mathbf{r})$.

If we compare equation (24) with equation (31), we can consider the crystal waves as analogous to diffraction waves due to a slit which is put on the incident surface. Difference are the form of the weight function and the shape of the wave surface.† In a crystalline medium the condition (33) should be generalized as

$$lW(\omega_0) \gg 1/K\omega_0 , \tag{34}$$

where ω_0 is the angular width in which $H(\hat{\mathbf{K}})$ of equation (24) has an appreciable value, because in a crystalline medium waves propagate in the direction of $\mathbf{v}$ instead of $\mathbf{k}$. Comparing this with equation (27), the Fraunhofer condition can be expressed also as

$$\omega_0 > \omega_m \tag{35}$$

since $W(\omega_0)$ is an increasing function of ω_0. The reverse condition implies the Fresnel diffraction condition, in a wider sense.

It is worthwhile here to notice that the Fraunhofer condition is satisfied at a much smaller distance l in a crystalline medium than in vacuum. The reason is that $W(\omega_0) > \omega_0$ in a crystal instead of $W(\Omega_0) = \Omega_0$ in a vacuum. In the vicinity of the exact Bragg condition we see that

$$W(\omega_0) \simeq \frac{\tan \theta_B}{\Delta\theta} \omega_0 . \tag{36}$$

Therefore, in a crystal the Fraunhofer condition is satisfied at a distance which is only 10^{-4} of that for a vacuum in X-ray cases.

6. General remarks and conclusions

(a) Relation between the light source and the crystal

The three inequalities of Section 2 can now be represented more systematically in reciprocal space. Based upon the general expression of the incident wave, equation (4), one extreme case of diffraction conditions is given by the relation (1). If the weight of the coherent plane waves on the wave surface

† In a crystalline medium the wave surface is called the dispersion surface.

|**K**| $= K$ has an appreciable value only within a very small region, the plane-wave approximation of the ordinary theory is appropriate. This is what happens in electron diffraction. In the alternative extreme case, in which $\Delta\theta \ll \Omega_0$, we can assume that the angular spectrum of the incident wave is homogeneous over the effective angular range of $\Delta\theta$. A spherical wave approximation, assuming $F_s(\hat{\mathbf{K}})$ for the weight function, is then appropriate.

Next, we consider in more detail the case in which an original incident wave is a spherical wave. This corresponds to the general situation of X-ray diffraction. If we modify a spherical wave by a slit system, we obtain a sort of diffraction wave. The angular width of such a diffraction wave cannot be reduced to roughly less than a certain value Ω_m. By a similar argument to that used in obtaining the minimum beam size g_m (see equation (27)), we see that

$$\Omega_m = 1/(KS_m) = S_m/L \tag{37}$$

or

$$\Omega_m = \sqrt{(1/(KL))} , \tag{38}$$

where L is the distance between the light source and the slit and S_m is an optimum slit width in order to obtain a minimum angular width of the weight function. According to equation (38) Ω_m depends upon L. For a given distance between the light source and the crystal Ω_m is at an absolute minimum if we place the slit at the crystal surface. Therefore, if

$$\Delta\theta < \Omega_m , \tag{39}$$

where L is the distance between the light source and crystal, the plane-wave approximation is fundamentally inadequate. This statement is equivalent to that given in Section 2 regarding condition (3′).

Remembering that $\Delta\theta$ is almost linearly proportional to λ at low Bragg angles we see that for a large wave length and a large distance L it is possible for the reverse condition of (39) to be realized. But then we have another difficulty in using the usual dynamical theory. In the usual theory the wave surface $|\mathbf{K}_0| = K$ is approximated by a tangential plane in the vicinity of the Laue point.* The necessary condition for this approximation is

$$KL\Omega_0^2 < 1 . \tag{40}$$

Otherwise the phase difference between the true wave and the approximated wave after traveling a distance L becomes larger than 2π. This condition cannot be satisfied in general with a spherical wave since Ω_0 cannot be reduced to less than Ω_m. If, however, condition (39) is realized, $\Delta\theta$ should be used in equation (40) as the effective width of Ω_0, and the approximation of neglecting the bending of the wave surface is permissible. With this substitution condition (40) becomes equivalent to condition (2′) above.

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* This is equivalent to the approximation of assuming the dispersion surface to be a hyperbolic surface (see equation (17)).

As discussed there, most of X-ray cases fall under this category.

(b) *Relation between the crystal and the observation point*

Consider the wave field at the exit surface of the crystal. In electron cases ω_0 is quite small as long as we use a crystal large enough compared with the wave length. According to inequality (35), therefore, the condition for Fresnel diffraction is realized in most regions of the crystal. Moreover, according to the plane-wave approximation, there result two plane waves **DO** and **D′O** as transmitted waves and two plane waves **DG** and **D′G** as diffracted waves. Thus we can expect sinusoidal interference fringes in both transmitted and diffracted waves.

On the other hand, in the case of X-rays, complicated wave fields are excited in the crystal close to the incident surface because Ω_0, and accordingly ω_0, are large compared with $\Delta\theta$. In the X-ray case, however, ω_m is quite small for most parts of the crystal so that the diffraction pattern is simplified by the Fraunhofer condition. In other words, as shown in terms of wave-bundle considerations, the component waves are separated, propagating in different directions. Thus, we have only to consider interference between waves **DO** and $\overline{\mathbf{DO}}$ for the transmitted wave fields and waves **DG** and $\overline{\mathbf{DG}}$ for the diffracted wave fields (see Fig. 1).

(c) *Effect of absorption*

There is no objection to applying the present formulation to the case of absorbing crystals. Also, the fundamental aspects of wave behavior described above are retained. Such a conclusion is based on the fact that crystal waves propagate along the direction of the normal of the dispersion surface. This is true also for absorbing crystals, as shown in the previous paper (Kato, 1960a). Of course, for the high absorption case 'Pendellösung' interference disappears because, as in the Borrmann effect, only one branch wave survives, the other branch wave being rapidly attenuated.

APPENDIX

Vector fields of diffracted waves due to a slit system

First we consider a scalar spherical wave Φ_s. The diffracted wave Φ due to the slit S is obtained by applying Kirchhoff's approximation to the boundary conditions.

$$\Phi(\mathbf{r}_s) = \Phi_s(\mathbf{r}_s) \quad \mathbf{r}_s \text{ in } S$$
$$= 0 \qquad \mathbf{r}_s \text{ outside } S , \tag{A-1}$$

where $\mathbf{r}_s$ is a radius vector to the plane of the slit. The Fourier transform of Φ with this condition gives us immediately

$$F(\hat{\mathbf{K}}) = \iint_S \Phi_s(\mathbf{r}_s) \exp\{-i(\mathbf{K} \cdot \mathbf{r}_s)\} d\mathbf{r}_s . \quad \text{(A-2)}$$

If the distance between the light source and the slit is large enough this tends to F_L (equation (6)) asymptotically.

Next we consider a vector field $\mathbf{H}_s$, which has the form of equation (13). The Fourier transform of the diffracted wave due to slit S is then

$$\mathbf{F}(\hat{\mathbf{K}}) = \iint_S \tilde{M} \Phi_s \exp\{-i(\mathbf{K} \cdot \mathbf{r}_s)\} d\mathbf{r}_s \quad \text{(A-3)}$$

using Kirchhoff's approximation again. Carrying out the operation of $\tilde{M}$:

$$\mathbf{F}(\hat{\mathbf{K}}) = -\iint_S \text{rot}\ (\mathbf{J}\Phi_s) \exp\{-i(\mathbf{K} \cdot \mathbf{r}_s)\} d\mathbf{r}_s$$

$$= -\int_C \mathbf{J}\Phi_s \exp\{-i(\mathbf{K} \cdot \mathbf{r}_s)\} d\mathbf{r}_s$$

$$+ \iint_S [\mathbf{J}\Phi_s \times i\mathbf{K}] \exp\{-i(\mathbf{K} \cdot \mathbf{r}_s)\} d\mathbf{r}_s , \quad \text{(A-4)}$$

where $\int_C$ means a line integral along the periphery of the slit, at which Φ_s should be zero according to the boundary conditions. Therefore,

$$\mathbf{F}(\hat{\mathbf{K}}) = i[\mathbf{J} \times \mathbf{K}] F(\hat{\mathbf{K}}) . \quad \text{(A-5)}$$

Accordingly,

$$\mathbf{H} = [\mathbf{J} \times \nabla]\Phi = \tilde{M}\Phi , \quad \text{(A-6a)}$$

since Φ has the form of equation (4).

The same methods can be applied also to the electric field using the operator $\tilde{N}$; thus,

$$\mathbf{E} = \tilde{N}\Phi . \quad \text{(A-6b)}$$

Repeating the same argument we can conclude that the diffracted vector waves due to an arbitrary slit system can be expressed in the form of equation (A-6a), (A-6b).

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Short Communications

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The refinement of the Co_2Al_5 structure. By J. B. NEWKIRK,* P. J. BLACK,† and A. DAMJANOVIC,‡ *Crystallographic Laboratory, Cavendish Laboratory, Cambridge, England*

The Co_2Al_5 compound has a hexagonal unit cell with dimensions:

$$a = 7{\cdot}656_0 \text{ Å}, \quad c = 7{\cdot}593_2 \text{ Å} .$$

The structure was originally determined from powder photographs taken with Fe $K\alpha$ radiation in a 19 cm. camera. Systematic absences were consistent with the

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space group D_{6h}^4–$P6_3/mmc$. There are 28 atoms per unit cell in the following positions (Bradley & Cheng, 1938):

2 Al_1 in	2(a):	0, 0, 0;
6 Al_2 in	6(h):	$x, 2x, \frac{1}{4}$ with $x = 0{\cdot}467$;
12 Al_3 in	12(k):	$x, 2x, z$ with $x = 0{\cdot}196$, $z = -0{\cdot}061$;
2 Co_1 in	2(d):	$\frac{2}{3}, \frac{1}{3}, \frac{1}{4}$;
6 Co_2 in	6(h):	$x, 2x, \frac{1}{4}$ with $x = 0{\cdot}128$.

No statement was made on the accuracy of the atomic parameters, nor was the reliability index given.

Compounds with similar structures have been reported in some other systems. The structure of $\pi(AlFeNi)$ with