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Dynamical Theory of Electron Diffraction for a Finite Polyhedral Crystal. I

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The multiplet structure of Debye–Scherrer rings and the splitting of Laue spots were observed recently in electron diffraction by several authors (Sturkey & Frevel, 1945; Hillier & Baker, 1945; Honjo, 1947; Cowley & Rees, 1947; Sturkey, 1948). Although these phenomena were explained by the refraction of the electron beam on the surface of micro-crystals of polyhedral shape, it may require the dynamical theory of diffraction to understand them completely. The general formulation of the theory for a finite crystal, however, is very difficult (Ekstein, 1942). One of us (Kato, 1949) previously extended Bethe's theory of electron diffraction (Bethe, 1928) to the case of a wedge-shaped crystal with an infinite lateral extension, and discussed the refraction effects. We recently tried to extend the theory further to a finite crystal of polyhedral shape. In the present note we give a summary of our results.

Diffraction by a wedge-shaped infinite crystal

Let us first consider the case where a primary beam, whose wave function is expressed by

$$\Phi_e(\mathbf{r})=\Psi_e \exp 2\pi i(\mathbf{K}_e, \mathbf{r}), \qquad (1)$$

impinges on a wedge-shaped infinite crystal, and is reflected by a net plane represented by $\mathbf{g}$. If we assume, for the sake of simplicity, the Laue case which is important in electron diffraction by micro-crystals, the waves in the crystal and in vacuum are determined by the boundary conditions at the entrance and the exit surface, as was shown in the previous paper (Kato, 1949). Their wave-number vectors can be obtained graphically as follows. In Fig. 1, O is the origin of the reciprocal lattice and G is the reciprocal-lattice point which corresponds to the net plane $\mathbf{g}$; L_k and L_d are the kinematical and the dynamical Laue points respectively, and the hyperbola represents the dispersion surface. When the wave-number vector of the incident wave $\mathbf{K}_e=\overrightarrow{EO}$ is given, two wave points of the crystal wave, $\bar{D}'$ and $\bar{D}''$, are determined by the condition of the tangential continuity of the wave-number vectors on the entrance surface, where $\overrightarrow{\bar{D}'\bar{D}''}$ is the direction of the normal of the surface of entrance of the incident wave. The wave-points of the diffracted waves in vacuum, $\bar{A}'$ and $\bar{A}''$, are obtained similarly by the condition on the exit surface with normal $\mathbf{n}_a$. These wave points determine the wave-number vectors as follows:

Primary waves in crystal:

$$\bar{\mathbf{k}}_0'=\overrightarrow{\bar{D}'O}, \quad \bar{\mathbf{k}}_0''=\overrightarrow{\bar{D}''O}.$$

Diffracted waves in crystal:

$$\bar{\mathbf{k}}_g'=\bar{\mathbf{k}}_0'+\mathbf{g}=\overrightarrow{\bar{D}'G},$$

$$\bar{\mathbf{k}}_g''=\mathbf{k}_0''+\mathbf{g}=\overrightarrow{\bar{D}''G}.$$

Diffracted waves in vacuum:

$$\bar{\mathbf{K}}_g'=\overrightarrow{\bar{A}'G}, \quad \bar{\mathbf{K}}_g''=\overrightarrow{\bar{A}''G}.$$

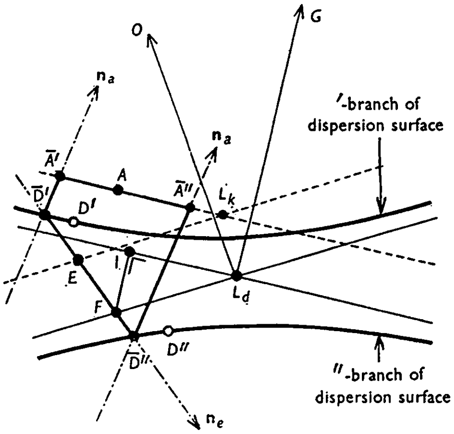

Fig. 1. Schematic diagram of dispersion surfaces and wave points. (All the vectors and points in this diagram are their projections on OEG-plane.)

$\mathbf{n}_e$: normal of the incident surface;
$\mathbf{n}_a$: normal of the exit surface.
$\overline{OL_k}=K$, $\overline{GL_k}=K$;
$\overline{OL_d}=\sqrt{\{K^2+v_0-V_{11}\}}$, $\overline{GL_d}=\sqrt{\{K^2+v_0-V_{22}\}}$;
$\overline{FI}=u=-wW$, where v_0, V_{11}, V_{22}, w and W are the Bethe's notations, except a numerical factor $1/4\pi^2$ in v_0, V_{11} and V_{22}, and $1/2\pi$ in w.

$\overrightarrow{E\bar{D}'}=\mathbf{d}_e'$, $\overrightarrow{E\bar{D}''}=\mathbf{d}_e''$; $\overrightarrow{\bar{D}'\bar{A}'}=\mathbf{d}_a'$, $\overrightarrow{\bar{D}''\bar{A}''}=\mathbf{d}_a''$;
$\overrightarrow{\bar{A}'A}=\mathbf{\Delta}_d'$, $\overrightarrow{\bar{A}''A}=\mathbf{\Delta}_d''$.

The appearance of two primary and diffracted waves in the crystal implies the so-called double refraction; this effect, however, is not observable when the crystal is bounded by two parallel planes where $\bar{\mathbf{K}}_g'$ and $\bar{\mathbf{K}}_g''$ coincide with each other. In the case of a wedge-shaped crystal, they are separated and the effect should be actually observable. Putting terms such as $\sqrt{(\Gamma/\gamma)}$, where Γ is the normal component of $\mathbf{K}_e$ and γ is that of

$\bar{\mathbf{k}}_0'$, $\bar{\mathbf{k}}_0''$, $\bar{\mathbf{k}}_g'$ or $\bar{\mathbf{k}}_g''$, approximately equal to unity, the amplitudes of the two waves are

$$\left.\begin{aligned}\bar{\Psi}_g' &= \Psi_e\{\bar{c}'\bar{c}''/(\bar{c}''-\bar{c}')\}\exp 2\pi i(\mathbf{K}_e-\bar{\mathbf{k}}_0', \mathbf{R}_e)\\ &\qquad\times\exp 2\pi i(\bar{\mathbf{k}}_g'-\bar{\mathbf{K}}_g', \mathbf{R}_a),\\ \bar{\Psi}_g'' &= \Psi_e\{-\bar{c}'\bar{c}''/(\bar{c}''-\bar{c}')\}\exp 2\pi i(\mathbf{K}_e-\bar{\mathbf{k}}_0'', \mathbf{R}_e)\\ &\qquad\times\exp 2\pi i(\bar{\mathbf{k}}_g''-\bar{\mathbf{K}}_g'', \mathbf{R}_a),\end{aligned}\right\}\quad(2)$$

where $\bar{c}'$ (or $\bar{c}''$) is the ratio of the amplitude of the diffracted wave $\mathbf{k}_g'$ (or $\mathbf{k}_g''$) to that of the primary wave $\mathbf{k}_0'$ (or $\mathbf{k}_0''$), which is determined by the position of the wave point $\bar{D}'$ (or $\bar{D}''$); and $\mathbf{R}_e$ and $\mathbf{R}_a$ have the meaning shown in Fig. 2.

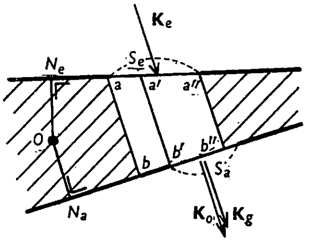

Fig. 2. Two-dimensional schematic diagram of the wedge-shaped crystal with the diaphragms explained in the text.

S_e: aperture on the incident surface.
S_a: aperture on the exit surface.
O: origin of the co-ordinate (arbitrarily chosen).
$\mathbf{K}_e$: incident wave in vacuum.
$\mathbf{K}_0$: transmitted wave in vacuum.
$\mathbf{K}_g$: diffracted wave in vacuum.
$\overrightarrow{ON_e}=\mathbf{R}_e$, $\overrightarrow{ON_a}=\mathbf{R}_a$.
$ab \parallel a'b' \parallel a''b'' \parallel \mathbf{K}_e$.

Effect of diaphragm

Let us consider, secondly, a case where both surfaces of the wedge are covered by opaque screens, each of which has an aperture of arbitrary shape and position. The crystal wave in this case is expressed by the general expression which is a superposition of plane waves as follows:

$$\begin{aligned}\phi(\mathbf{r}) &= \int \psi_0'(\mathbf{k}_0')\{\exp 2\pi i(\mathbf{k}_0', \mathbf{r})+c'\exp 2\pi i(\mathbf{k}_g', \mathbf{r})\}\,d\mathbf{k}_0'\\ &\quad+\int \psi_0''(\mathbf{k}_0'')\{\exp 2\pi i(\mathbf{k}_0'', \mathbf{r})+c''\exp 2\pi i(\mathbf{k}_g'', \mathbf{r})\}\,d\mathbf{k}_0'',\end{aligned}\quad(3)$$

where $\mathbf{k}_0'$ and $\mathbf{k}_g'$ (or $\mathbf{k}_0''$ and $\mathbf{k}_g''$) are the wave-number vectors of the primary and the diffracted waves respectively in the crystal determined by a wave point D' (or D'') on the dispersion surface, and c' (or c'') is the ratio of the amplitudes, as mentioned above. The amplitudes, $\psi_0'(\mathbf{k}_0')$ and $\psi_0''(\mathbf{k}_0'')$, in the integrals are determined by the boundary conditions at the entrance aperture which state:

$$\left.\begin{aligned}&\left.\begin{aligned}&\Phi_e(\mathbf{r})=\phi(\mathbf{r})\\ &\operatorname{grad}_{\mathbf{n}_e}\Phi_e(\mathbf{r})=\operatorname{grad}_{\mathbf{n}_e}\phi(\mathbf{r})\end{aligned}\right\}\quad\text{on the aperture } S_e,\\ &\phi(\mathbf{r})=0\qquad\qquad\qquad\qquad\text{on the diaphragm,}\end{aligned}\right\}\quad(4)$$

where $\mathbf{n}_e$ means the outward normal of the entrance surface. By a simple calculation of Fourier integral, they turn out to be

$$\left.\begin{aligned}\psi_0'(\mathbf{k}_0') &= \Psi_e\{c''/(c''-c')\}\,L(\mathbf{K}_e-\mathbf{k}_0'; S_e),\\ \psi_0''(\mathbf{k}_0'') &= \Psi_e\{-c'/(c''-c')\}\,L(\mathbf{K}_e-\mathbf{k}_0''; S_e),\end{aligned}\right\}\quad(5)$$

where the function L is the diffraction function of the aperture S and is expressed as

$$L(\mathbf{K}-\mathbf{k}; S)=\int_S \exp 2\pi i(\mathbf{K}-\mathbf{k}, \mathbf{r})\,d\mathbf{r}.\quad(6)$$

By repeating the similar procedure at the exit aperture S_a, we derive the diffracted wave in vacuum as

$$\Phi_g(\mathbf{r})=\int\{\Psi_g'(\mathbf{K}_g)+\Psi_g''(\mathbf{K}_g)\}\exp 2\pi i(\mathbf{K}_g, \mathbf{r})\,d\mathbf{K}_g,\quad(7)$$

where $\mathbf{K}_g$ is $\overrightarrow{AG}$ shown in Fig. 1, and

$$\left.\begin{aligned}\Psi_g'(\mathbf{K}_g) &= \Psi_e\int\{c'c''/(c''-c')\}\\ &\qquad\times L(\mathbf{K}_e-\mathbf{k}_0'; S_e)\,L(\mathbf{k}_g'-\mathbf{K}_g; S_a)\,d\mathbf{k}_0',\\ \Psi_g''(\mathbf{K}_g) &= \Psi_e\int\{-c'c''/(c''-c')\}\\ &\qquad\times L(\mathbf{K}_e-\mathbf{k}_0''; S_e)\,L(\mathbf{k}_g''-\mathbf{K}_g; S_a)\,d\mathbf{k}_0''.\end{aligned}\right\}\quad(8)$$

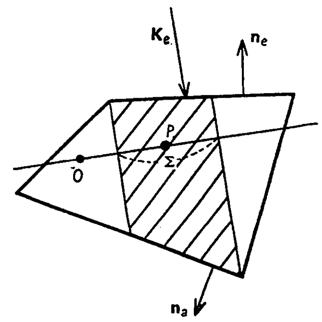

Fig. 3. Two-dimensional schematic diagram of a polyhedral crystal.

O: origin of the co-ordinate (arbitrarily chosen).
Σ: common region of the projection of S_e and S_a on the plane perpendicular to $\mathbf{K}_e$ passing through O.
P: an arbitrary point on Σ and $\mathbf{s}=\overrightarrow{OP}$.

The formula (7) means that the diffracted beam is not perfectly sharp, but it is diffuse around the direction of $\bar{\mathbf{K}}_g'$ and $\bar{\mathbf{K}}_g''$, and the behaviours are given by (8). The expressions in (8) reduce to those of (2) when the apertures are infinitely extended, because the function L becomes a δ-type function which vanishes except at the points $\bar{D}'$ and $\bar{D}''$, and (7) is reduced to two plane waves corresponding to doubly refracted beams of perfect sharpness. When the apertures have finite extensions, L does not vanish at all points on the dispersion surface. However, in the case of electron diffraction where the wave-length is very short (~ 0·05 A.), it is vanishingly small except in the neighbourhood of $\bar{D}'$ and $\bar{D}''$ (angular variation of wave vectors ~ 30′) even for a quite small aperture (~ 10 A.). Since the Bragg angle is also small (< 5°) in this case, the wave function in the crystal almost vanishes in the shaded region in Fig. 2, and the wave which departs from the aperture S_a depends only on the wave function in the region $a'b'b''a''$.

Diffraction by a polyhedral crystal

In the case of a polyhedral crystal, we divide the crystal as shown in Fig. 3; then we can calculate the wave functions for each division from (3) to (7). Their superposition satisfies approximately the boundary condition on the whole crystal surface by the above considerations and therefore represents the wave-field for a finite crystal of polyhedral shape according to the dynamical theory. Further calculation of (8) and comparison with the kinematical theory will be given in the next note.

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Dynamical Theory of Electron Diffraction for a Finite Polyhedral Crystal. II. Comparison with the Results of Kinematical Theory

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In the previous note (Kato & Uyeda, 1951), we have shown that Bethe's dynamical theory of electron diffraction (Bethe, 1928) can be extended to the case of a finite crystal. In the present note, we shall develop the theory further and compare its results with those derived from the kinematical theory.

Dynamical and kinematical formula for a polyhedral crystal

The equation (7) of the previous note, which gives the diffracted wave due to one of the divisions of a crystal shown in Fig. 3 there, can be approximated by a more practical formula, provided the conditions described in the previous note are satisfied. The final formula for the diffracted amplitude at large distance, **R**, from the crystal turns out to be

$$\Phi^d_{\mathbf{g}}(\mathbf{R})=\frac{1}{2iR}\frac{|V_{21}|}{\sqrt{(u^2+w^2)}}\exp 2\pi iKR \times\left[\exp 2\pi i\{(\mathbf{d}'_e,\mathbf{R}_e)+(\mathbf{d}'_a,\mathbf{R}_a)\}\int_\Sigma \exp 2\pi i(\mathbf{\Delta}'_d,\mathbf{s})\,d\mathbf{s} -\exp 2\pi i\{(\mathbf{d}''_e,\mathbf{R}_e)+(\mathbf{d}''_a,\mathbf{R}_a)\}\int_\Sigma \exp 2\pi i(\mathbf{\Delta}''_d,\mathbf{s})\,d\mathbf{s}\right]; \tag{1}$$

here V_{21} is Bethe's dynamical structure factor which is approximately equal to the Fourier coefficient $v_{\mathbf{g}}$ of the crystal potential, and w stands for

$$(|V_{21}|/K)\sqrt{\{(\mathbf{k}_{\mathbf{g}},\mathbf{n}_e)/(\mathbf{k}_0,\mathbf{n}_e)\}}.$$

The notations used are explained in the legends of Figs. 1 and 3 of the preceding note.

On the other hand, the kinematical theory gives the expression of the amplitude diffracted by a finite crystal as follows:

$$\Phi_{\mathbf{g}}(\mathbf{R})=\frac{\pi}{R}v_{\mathbf{g}}\int_C \exp 2\pi i(\mathbf{K}_e+\mathbf{g}-\mathbf{K}_{\mathbf{g}},\mathbf{r})\,d\mathbf{r}, \tag{2}$$

where the integral covers the whole volume of the crystal, C. If we consider the wave due to one of the divisions as above, the integration is limited to this division, and we can rewrite (2) as follows:

$$\Phi^k_{\mathbf{g}}(\mathbf{R})=\frac{1}{2iR}\frac{v_{\mathbf{g}}}{u_0}\exp 2\pi iKR \times\left[\exp 2\pi i(\mathbf{d}_e,\mathbf{R}_e)\int_\Sigma \exp 2\pi i(\mathbf{\Delta}^e_k,\mathbf{s})\,d\mathbf{s} -\exp 2\pi i(\mathbf{d}_a,\mathbf{R}_a)\int_\Sigma \exp 2\pi i(\mathbf{\Delta}^a_k,\mathbf{s})\,d\mathbf{s}\right], \tag{3}$$

where the notations are given in Fig. 1 of this note, and the other notations are the same as those in the previous note.

Discussion by means of *Intensitätsbereich*

The interpretation of the kinematical formula (3) can be given most clearly by making use of the conception of Laue's *Intensitätsbereich* (Laue, 1936) in Ewald's construction of the wave vector. This construction can be obtained in Fig. 1 if we transfer the vector $\mathbf{K}_{\mathbf{g}}=\overrightarrow{AG}$ by parallel displacement so that the initial point A coincides with the point E. Then, the end-point G is displaced to a new point, say Q, and $\overrightarrow{GQ}=\overrightarrow{AE}$, which