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The Nature of the Specular Reflexion of Electrons from a Crystal Surface

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In electron diffraction patterns from smooth cleavage surfaces of single crystals, a sharp reflexion having a specular relation to the incident beam appears for all glancing angles of the incident electrons. Its intensity is especially enhanced when the reflexion happens to fall on a Kikuchi line.

This reflexion has formerly been considered as a two-dimensional diffraction effect caused by the high absorption of electrons within solids. The present authors propose an interpretation according to the dynamical theory of diffraction, neglecting absorption; they take account of the fact that enhancement of the specular reflexion occurs when a Bragg reflexion in a side direction takes place inside the crystal. It is shown that the regular total reflexion of the incident electrons by the surface may take place even when the Bragg condition is not satisfied for the lattice plane parallel to the surface, and the phenomenon of enhancement can be explained as the result of Bragg reflexion on a side plane. The total reflexion of the interior wave at the surface plays an important role in producing enhancement. It is noted that the proper choice of the wave points in reciprocal space is essential in this problem.

The influence of the weak constituents of the internal wave field on the intensity of the specular reflexion is also discussed.

1. Introduction

In the electron diffraction pattern obtained by reflecting electrons incident at an arbitrary angle on the fresh cleavage plane of a well grown crystal, there appear many Kikuchi lines, some Bragg spots and, besides, several circular arrays of reflexions which are as sharp as the spot of the incident ray. The circles are most easily noticed when the azimuth of the incident beam deviates a little from some important zone axis of the crystal contained in the surface (Fig. 1).* The specular reflexion spot, which is geometrically the regular reflexion of the incident beam by the surface, is a member of the circular group corresponding to the innermost circle passing through the incident spot, and it possesses a well discernible intensity at any azimuthal and glancing angle of incidence.

On the basis of the kinematical theory of diffraction, Kirchner & Raether (1932), Raether (1932), and Kikuchi & Nakagawa (1933*a*, *b*) regarded the circular group of spots as the result of the two-dimensional diffraction caused by the small penetrating power of electrons into the crystal. The positions of the spots are actually found to be in accordance with the geometrical relation given by the two Laue conditions for the two lattice rows lying in the plane parallel to the surface:

$$(\mathbf{s}-\mathbf{s}_0 . \mathbf{a}_1) = h_1\lambda, \quad (\mathbf{s}-\mathbf{s}_0 . \mathbf{a}_2) = h_2\lambda, \qquad (1)$$

where $\mathbf{a}_1$ and $\mathbf{a}_2$ are the vectors of the fundamental translations of the crystal lattice lying in the boundary surface, $\mathbf{s}_0$ and $\mathbf{s}$ the unit vectors of the directions of the incident and diffracted rays, λ the wave length of the electron, and h_1 and h_2 integers.

It seems, however, difficult to explain by this simple theory the following phenomenon which was found by Kikuchi & Nakagawa (1933*b*), and was called by them 'the anomalous phenomenon of the second kind of rotation spectra': the specular reflexion is anomalously enhanced to an intensity comparable to that of Bragg reflexions when the spot is traversed by a Kikuchi line; this occurs under suitable conditions of incidence. Fig. 2 shows an example of this effect on the cleavage face of zincblende. A similar example is contained in the paper of Kikuchi & Nakagawa. As will be explained in § 2, this phenomenon of enhancement is known to be correlated with the occurrence of a Bragg reflexion inside the crystal. Since such interaction between the specular reflexion and the Bragg reflexion cannot be treated by any kinematical theory, it is suggested that an explanation of the specular reflexion, and of the circular group of spots

* We are indebted to Dr. G. Honjo for Fig. 1.

in general, must be sought in the dynamical theory.

In fact, the geometrical condition (1) is in accordance also with the dynamical theory. This theory assumes:

(i) there exist within the crystal many diffracted waves whose wave number vectors $\mathbf{k}_h$ are related to that of the primary wave $\mathbf{k}_0$ by the relation

$$\mathbf{k}_h = \mathbf{k}_0 + \mathbf{h} \ , \tag{2}$$

where $\mathbf{h}$ is the vector representing the point (h_1, h_2, h_3) of the reciprocal lattice, namely,

$$\mathbf{h} = h_1\mathbf{b}_1 + h_2\mathbf{b}_2 + h_3\mathbf{b}_3 ; \tag{3}$$

(ii) each of these waves is joined to a corresponding wave in free space so as to satisfy the condition of continuity of the tangential components of the relevant wave vectors. The relations (i) and (ii) immediately result in the condition (1) (Bethe, 1928; Thomson & Cochrane, 1939; Laue, 1948). We can expect, therefore, that the intensity of the reflexions satisfying (1) may be calculated by the usual procedure of the dynamical theory. The ordinary Bragg reflexions, then, are considered as particular cases of these reflexions.

The interpretation of the circular group of spots on the dynamical theory does not, however, seem to have been attempted previously. The present paper is intended to explain the nature of these reflexions, especially of the specular reflexion, on the basis of this theory.

2. Some experimental facts

(a) Geometrical condition of enhancement

As mentioned in § 1, the intensity enhancement of the specular reflexion takes place when it is traversed by a Kikuchi line. An important physical implication follows.

In diffraction photographs taken with the reflexion method, the pattern is in general limited towards the lower angle by the so-called *shadow edge*, which corresponds to the intersection of the surface plane of the crystal with the photographic plate. The lower half below the shadow edge cannot be seen on account of the absorption of electrons within the crystal, but we can imagine the pattern which would be there if the crystal were transparent for electrons. Then, the pattern under the condition of the enhancement will be as shown schematically in Fig. 3, where the refraction of electrons at the boundary surface is neglected for the sake of simplicity.

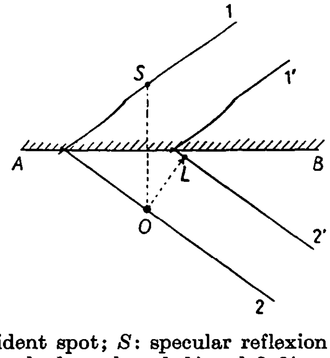

Fig. 3. O: incident spot; S: specular reflexion; L: Bragg reflexion; AB: shadow edge; 1, 1′ and 2, 2′: pairs of Kikuchi lines.

The specular spot S is located at the position symmetrical to the incident spot O with respect to the shadow edge AB. From Fig. 3 it is seen at once that when a Kikuchi line 1 traverses the specular spot, another Kikuchi line 2, symmetrical to line 1 with respect to the shadow edge, passes through the incident spot, provided that the surface is a mirror plane of the crystal *lattice*—not necessarily of the crystal *structure*. This condition is fulfilled by the cleavage surface of many crystals.

The condition that the incident spot lies on a Kikuchi line is the same as the condition for the Bragg reflexion of the incident beam by the corresponding lattice plane. Therefore enhancement of the specular reflexion can occur if the incident electrons suffer a Bragg reflexion on a certain lattice plane, provided that the boundary surface is a mirror plane of the crystal lattice. In general the lattice plane concerned is one which is not parallel to the surface, so that the corresponding reflexion may be called hereafter the *Bragg reflexion in a side direction*. This reflexion gives rise to a spot L on another Kikuchi line 2′, which is parallel to the line 2, the two lines together forming a pair of black and white Kikuchi lines corresponding to the lattice plane concerned. The spot L may lie either above or below the shadow edge.

Experimentally, enhancement is especially remarkable when the wave of the Bragg reflexion in a side direction is expected to travel nearly parallel to the crystal surface, so that L lies close to the shadow edge. Such a Bragg reflexion, however, will not necessarily appear in the actual diffraction pattern, even if L (drawn by ignoring the refraction effect) happens to lie above the shadow edge, because the refractive index of crystals for electrons is in general larger than unity (see § 5).

Though the enhancement of the specular reflexion came first to our attention as the effect of its coincidence with Kikuchi lines, the role of the Kikuchi lines in the present phenomenon seems to be only to furnish a very convenient indication of the geometrical relation between the incident electron beam and the crystal, and no more.

(b) Intensity variation of the specular reflexion with the change of glancing angle

When a crystal is rotated in an electron diffraction camera about an axis contained in the surface and perpendicular to the incident electron beam, the specular spot is found to move smoothly on a fluorescent screen or photographic plate with changing glancing angle. It becomes especially bright at positions when Bragg reflexions occur on the lattice planes parallel to the surface.

The specular reflexion is in general fairly weak in angular ranges between two successive Bragg reflexions, unless it is located close to the Bragg positions; its intensity varies only monotonously in the middle angular ranges so long as no phenomenon of the enhancement takes place. On photographs of the rotation spectra, we note a continuous faint streak joining successive Bragg reflexions of different orders, and this can be regarded as manifesting the intensity distribution of the specular reflexion with the glancing angle. The photographs show that the intensity is asymmetric with respect to every Bragg spot: its decrease with distance from the Bragg spot is much slower for increasing than for decreasing angles. This is seen in Fig. 4, which is obtained by subtracting

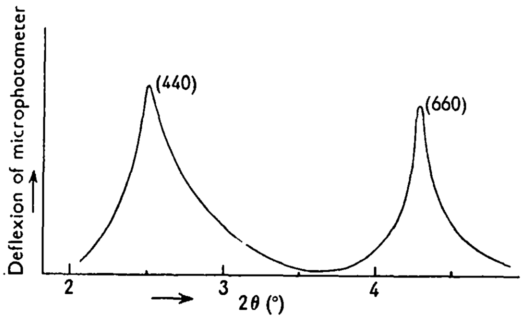

Fig. 4. Micro-photometer curve of the intensity distribution of the specular reflexion from a cleavage face (110) of zincblende, the general background being subtracted. Accelerating voltage, 46 kV. θ is the glancing angle.

the background on a micro-photometric curve of a rotation spectrum obtained on a cleavage face of zincblende. The same trend is also noticed in the intensity curve for a diamond crystal (Beeching, 1935).

3. General procedure of the dynamical theory

A solution of the Schrödinger equation for an electron in a crystalline periodic potential $V(\mathbf{r})$ with the energy E,

$$\nabla^2\psi+\frac{8\pi^2 m}{h^2}(E+eV(\mathbf{r}))\psi=0\,, \tag{4}$$

is given by the Bloch function

$$\psi(\mathbf{k}_0)=\sum_h u_h(\mathbf{k}_0)\exp\left[-2\pi i(\mathbf{k}_h.\mathbf{r})\right], \tag{5}$$

where h indicates the triple indices corresponding to the reciprocal-lattice point $\mathbf{h}$, and the relation between $\mathbf{k}_0$ (the wave number vector of the primary wave within the crystal) and the $\mathbf{k}_h$'s is as already given by (2). Each Bloch function, then, is determined corresponding to a given wave point in the reciprocal space. The coefficients u_h's are determined, in their ratios, by the fundamental equations of diffraction:

$$(\varkappa_0^2-\mathbf{k}_h^2)u_h+\sum_{h'}{}' v_{h'}\,u_{h-h'}=0, \tag{6}$$

where

$$\varkappa_0^2=\frac{2m}{\hbar^2}(E+eV_0),\;\; v_n=\frac{2me}{\hbar^2}V_n\,; \tag{7}$$

V_0 is the mean inner potential and V_n the Fourier coefficient of the periodic potential $V(\mathbf{r})$ (Bethe, 1928).

The dispersion surface, or the surface of constant energy in reciprocal space, is determined by the compatibility relation of the above homogeneous equations. The surface is generally a hypersurface of infinite degree, being periodic in reciprocal space.

Let us now assume that an electron wave with the wave vector $\mathbf{K}_0^i$ and the amplitude Ψ_0 is incident on the upper boundary surface of a crystal slab of infinite area and of finite thickness, where

$$|\mathbf{K}_0^i|=\frac{\sqrt{(2mE)}}{h}. \tag{8}$$

Then the possible wave points are given as the points of intersection of the dispersion surface with a line normal to the entrant surface and at a distance from the origin of this space equal to the tangential component of $\mathbf{K}_0^i$. This line will be called hereafter the *ν-normal.*

We discriminate the different wave points determined by the above procedure by the index N (N = I, II, . . .); the quantities relevant to the N-th wave point will also be indexed by N, e.g. u_h^N, $\mathbf{k}_0^N$, $\mathbf{k}_h^N$ etc. The amplitude $\Psi_{h_1h_2}$ of the reflected wave of indices (h_1h_2), emerging from the upper surface according to the law (1) with the wave number vector $\mathbf{K}_{h_1h_2}$, is expressed by

$$2\Gamma_{h_1h_2}\Psi_{h_1h_2}=\Psi_0\sum_N\sum_{h_3}(\Gamma_{00}-\gamma^N_{h_1h_2h_3})u^N_{h_1h_2h_3}\,, \tag{9}$$

where Γ_{00} and $\Gamma_{h_1h_2}$ are, respectively, the normal components of $\mathbf{K}_0^i$ and $\mathbf{K}_{h_1h_2}$, assumed to be positive, and $\gamma^N_{h_1h_2h_3}$ is the normal component of $\mathbf{k}^N_{h_1h_2h_3}$, assumed positive when the vector is directed downwards (Bethe, 1928; Laue, 1948). Equation (9) serves to determine the intensities of the spots in the circular group in general, the Bragg reflexions being particular cases. The reflexion (00) corresponds to the specular reflexion.

In the ordinary treatment of the dynamical theory the intensity of a Bragg reflexion, say $(h_1'h_2'h_3')$, is calculated by assuming all of the waves $\Psi_{h_1h_2}$ (except $\Psi_{h_1'h_2'}$) to be vanishingly weak. In the present study, however, the wave Ψ_{00} should always be retained, even when it is fairly weak, since this wave is our main object.

4. Choice of wave points

As is well known, the greater part of the dispersion surface can be approximated by a group of spheres of radius $\varkappa_0$ around every reciprocal point; it deviates appreciably from the spheres only in the regions near

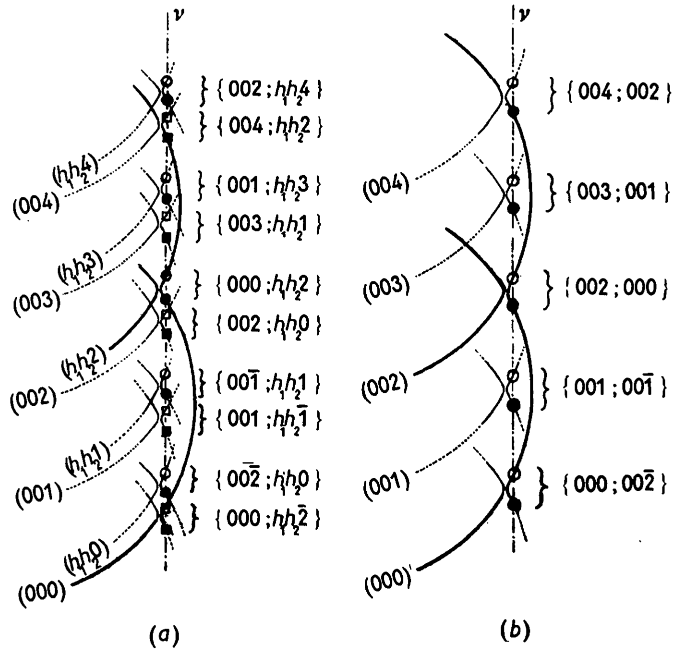

Fig. 5. The intersection of the dispersion surface and the ν-normal. (*a*) The case $h_1 \neq 0$, $h_2 \neq 0$ and $h_3 = 2$. (*b*) The case $h_1 = 0$, $h_2 = 0$ and $h_3 = 2$. Each of the symbols ○, ●, □ and ■ represents the equivalent wave points.

the intersections of two spheres. We denote each of these regions by $\{h; h'\}$, where h and h' represent the triple indices of the reciprocal-lattice points at which the two spheres have their centres. For the sake of simplicity, we disregard triple or multiple intersections of the spheres.

The number of possible wave points determined as the intersecting points of the dispersion surface with the ν-normal is in general infinite. For instance, when the ν-normal passes through the neighbourhood of the $\{000;\ h_1h_2h_3\}$ region, the two real or complex wave points are determined there, but since the reciprocal-lattice rows parallel to $\mathbf{b}_3$, which run perpendicular to the surface of the crystal, are parallel to the ν-normal, this line passes also through regions $\{00n;\ h_1,h_2,h_3+n\}$ and $\{00n;\ h_1,h_2,\bar{h}_3+n\}$ ($n = \pm 1, \pm 2, \ldots$), and determines two wave points in every region (Fig. 5).

As discussed by Lamla (1938*a*, *b*, *c*), the Bloch functions corresponding to these infinitely many wave points are not independent of one another. Since, as is seen from (5), the Bloch functions $\psi(\mathbf{k}_0)$ and $\psi(\mathbf{k}_0+\mathbf{h})$ represent the same state, it is apparent that the number of independent Bloch functions corresponding to the wave points on the ν-normal passing through the $\{000; h_1h_2h_3\}$ region is confined to only four (Fig. 5(*a*)). When, especially, $h_1 = 0$ and $h_2 = 0$, this number becomes two (Fig. 5(*b*)). For a rigorous treatment, we have to make full use of all the independent Bloch functions. When, for instance, the Bragg reflexion in a side direction $(h_1h_2h_3)$ takes place, we have to choose four wave points properly on the ν-normal passing through the $\{000; h_1h_2h_3\}$ region. The most convenient choice in this case may be such as shown in Fig. 6, in which the wave points A^{I} and A^{II} are selected at the $\{000; h_1h_2h_3\}$ region, and A^{III} and A^{IV} at the $\{000; h_1h_2\bar{h}_3\}$ region; O, H and H' are the reciprocal-lattice points (000), $(h_1h_2h_3)$ and $(h_1h_2\bar{h}_3)$ respectively.

The vector connecting a wave point to any of the reciprocal-lattice points represents the wave number vector of a plane wave to be contained in the relevant

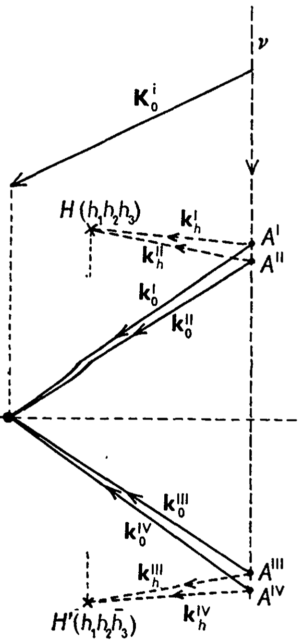

Fig. 6. Ray construction in reciprocal space.

Bloch function. These waves within the crystal are joined to the entrant and emerging waves in the free spaces so as to satisfy the boundary condition, and, only when the wave points are chosen reasonably in the above-mentioned way, the amplitudes of all the waves are uniquely determined by the boundary relations and the fundamental equations (6), as proved by Lamla.

From (6), it is expected that only the waves for which $\varkappa_0^2-\mathbf{k}_h^2 \approx 0$ will have appreciable amplitudes, so that when a wave point, say A^{I} in Fig. 6, is in the closest neighbourhood of the $\{000; h_1h_2h_3\}$ region, only the two waves corresponding to $\overrightarrow{A^{\mathrm{I}}O} \equiv \mathbf{k}_0^{\mathrm{I}}$ and $\overrightarrow{A^{\mathrm{I}}H} \equiv \mathbf{k}_h^{\mathrm{I}}$ need to be retained as the strong waves relating to A^{I}. In Fig. 6, only the wave vectors of such strong rays for the four wave points are inscribed. The reciprocal-lattice points H and H' are not in general in the plane determined by O and the ν-normal, so that the corresponding vectors are drawn as broken lines.

The ray relation for the crystal slab can alternatively be represented as shown schematically in Fig. 7, where

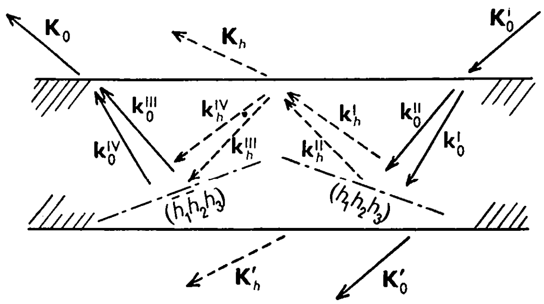

Fig. 7.

$\mathbf{K}_h$ and $\mathbf{K}_h'$ correspond to the (h_1, h_2) reflexions emerging from the upper and lower surfaces respectively, $\mathbf{K}_0'$ to the transmitted wave in the lower space, and $\mathbf{K}_0$ to the specular reflexion by the upper surface of the incident wave $\mathbf{K}_0^i$. The amplitudes of waves $\mathbf{K}_h$ and $\mathbf{K}_0$ are given by (9).

5. Physical consideration of the process of enhancement

When the crystal surface is the mirror plane of the crystal *lattice*, the reciprocal-lattice points H and H' are symmetrically situated with respect to the plane which passes the origin O and is parallel to the crystal surface, so that the wave vectors $\mathbf{k}_0^{\mathrm{I}}$ and $\mathbf{k}_0^{\mathrm{IV}}$, $\mathbf{k}_0^{\mathrm{II}}$ and $\mathbf{k}_0^{\mathrm{III}}$, $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{IV}}$, $\mathbf{k}_h^{\mathrm{II}}$ and $\mathbf{k}_h^{\mathrm{III}}$, are at least approximately in the relation of the regular reflexion with respect to the surface of the crystal slab. When, in addition, the surface is the mirror plane of the crystal *structure*, the forms of the dispersion surface near the $\{000; h_1h_2h_3\}$ and $\{000; h_1h_2\bar{h}_3\}$ regions are the mirror image of each other, so that the relation of the above-mentioned regular reflexion becomes complete.

Though all the waves within the crystal are actually the result of their dynamical interference, the essential process of the phenomenon of the enhancement can be now understood by a qualitative reasoning as follows.

The waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ are interpreted respectively as the Bragg reflexions by the $(h_1h_2h_3)$ plane of the waves $\mathbf{k}_0^{\mathrm{I}}$ and $\mathbf{k}_0^{\mathrm{II}}$, so that the amplitudes of the reflected waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ are large so long as the Bragg conditions for the relevant waves are satisfied fairly well. When the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ travel upwards, impinging on the upper boundary of the crystal slab (the condition of the Bragg case), and the angles of their ray directions to the boundary surface, β_{I} and β_{II} respectively, are very small, then these waves are reflected efficiently by the boundary, resulting in the waves $\mathbf{k}_h^{\mathrm{IV}}$ and $\mathbf{k}_h^{\mathrm{III}}$. Since the surface is assumed to be a mirror plane of the lattice, the reflected waves $\mathbf{k}_h^{\mathrm{IV}}$ and $\mathbf{k}_h^{\mathrm{III}}$ will necessarily receive in turn the Bragg reflexion by the $(\bar{h}_1\bar{h}_2h_3)$ plane, generating the waves $\mathbf{k}_0^{\mathrm{IV}}$ and $\mathbf{k}_0^{\mathrm{III}}$. As the result of these processes it is to be expected that, under the assumed conditions, the amplitudes u_{000}^{I}, u_{000}^{II}, u_{000}^{III} and u_{000}^{IV} will be of comparable order of magnitude with one another. It is then apparent that, under the conditions of enhancement, the amplitude Ψ_{00} of the specular reflexion $\mathbf{K}_0$ is almost entirely formed by the waves $\mathbf{k}_0^{\mathrm{III}}$ and $\mathbf{k}_0^{\mathrm{IV}}$. Analytically the amplitude Ψ_{00} follows from (9) as

$$\Psi_{00} = \Psi_0 \frac{1}{2\Gamma_{00}} \{(\Gamma_{00}-\gamma_{000}^{\mathrm{I}})u_{000}^{\mathrm{I}}+(\Gamma_{00}-\gamma_{000}^{\mathrm{II}})u_{000}^{\mathrm{II}} +(\Gamma_{00}-\gamma_{000}^{\mathrm{III}})u_{000}^{\mathrm{III}}+(\Gamma_{00}-\gamma_{000}^{\mathrm{IV}})u_{000}^{\mathrm{IV}}\},$$

where $\Gamma_{00} \approx \gamma_{000}^{\mathrm{I,II}}, -\gamma_{000}^{\mathrm{III,IV}}$, and the last two terms of the curly bracket, are the important ones.

The incidence of the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ towards the surface corresponds to the case of the incidence from a dense medium to a less dense medium; this makes the reflexion of these waves strong when the angles β_{I} and β_{II} become smaller than a certain critical value.

Since we are dealing with the Bragg case there will be a narrow angular range close to the Bragg condition wherein all waves in the crystal possess complex wave vectors, each of which corresponds to either an exponentially decreasing or increasing wave amplitude. For a slab of infinite thickness, however, only the decreasing waves, say $\mathbf{k}_0^{\mathrm{I}}, \mathbf{k}_h^{\mathrm{I}}, \mathbf{k}_0^{\mathrm{III}}$ and $\mathbf{k}_h^{\mathrm{III}}$, should be retained in that angular range. All the energies of the waves $\mathbf{k}_0^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{III}}$ are then transferred to the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_0^{\mathrm{III}}$, respectively. Moreover, if the angle β_{I} between $\mathbf{k}_h^{\mathrm{I}}$ and the surface is less than the critical value, all the incident energy of the wave $\mathbf{K}_0^i$ is transferred to the wave $\mathbf{K}_0$ in the end, and here we see a total specular reflexion of the incident wave by the surface, even if its glancing angle is not especially small (Fig. 8). Though the total reflexion is only the extreme case of the enhancement of the specular reflexion, this occurrence is not rare, because the critical angle β_c, which is given approximately by

$$\beta_c = \sqrt{(eV_0/E)}\,, \tag{10}$$

is usually as large as of the order of 1°. The experimentally observed intensity enhancement of the spec-

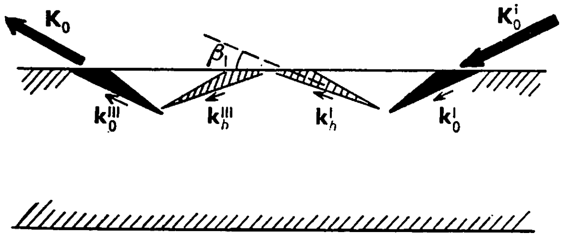

Fig. 8.

ular reflexion is then explained as a result of such total, or nearly total, reflexion.

6. Further consideration of the phenomenon of enhancement

In the usual calculation of the intensity of a Bragg reflexion (e.g. Bethe, 1928; Thomson & Cochrane, 1939), only two wave points, for instance A^{I} and A^{II}, are taken into account, and the other two wave points, A^{III} and A^{IV}, are neglected. Such a treatment may be legitimate when the reflecting lattice plane is parallel to the boundary surface. In other cases it is a poor approximation unless the angles between the ray directions of the waves within the crystal and the surfaces are fairly large. For the purpose of calculating the intensity of the specular reflexion, however, it is imperative to make use of the four wave points because, otherwise, the waves such as $\mathbf{k}_0^{\mathrm{III}}$ and $\mathbf{k}_0^{\mathrm{IV}}$ which contribute most to the intensity of the specular reflexion cannot be introduced into the theory.

The above explanation of the phenomenon of the enhancement, however, assumes that: (i) the crystal surface is a mirror plane of the lattice, and the Fourier coefficients for $(h_1h_2h_3)$ and $(h_1h_2\bar{h}_3)$ are finite: (ii) we are dealing with the Bragg case, namely the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ travel towards the upper surface: (iii) the angles between the ray direction of these waves and the surface are sufficiently small; and (iv) only the waves relating to the reciprocal-lattice points O (000), H $(h_1h_2h_3)$ and H' $(h_1h_2\bar{h}_3)$ are retained as the strong waves.

Concerning these assumptions, it should be noted that the condition (ii) is in general not always fulfilled. But even for the Laue case, where the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ travel downwards and all the wave points are always real, strong reflexions of these waves are expected to take place at the lower boundary surface; hence the phenomenon of the enhancement may appear in a similar way to that in the Bragg case. It is nevertheless quite doubtful if such consideration for the Laue case could retain any physical meaning compatible with the experimental condition. For, in the first place, the crystal used in the actual experiment, or each of the mosaic blocks in it, is by no means the parallel slab assumed in the present theory. In the second place, the absorption of the electron waves which is neglected in the present theory will weaken the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ sufficiently by the time they reach the lower boundary, and the phenomenon of enhancement cannot arise. These difficulties in the Laue case are, however, met by the following consideration.

The assumption (iv) will not be valid if the condition (iii) is satisfied and the structure amplitude for the first-order reflexion by the lattice plane parallel to the surface (whose indices will be denoted by $(00s)$ or simply by s) is large, and if furthermore the corresponding spacing is not too small, which is usually the case. As pointed out by Artmann (1947), we must assume, under these conditions, the coexistence of new strong waves relating to the reciprocal-lattice points $G(h_1h_2h_3\pm s)$ and G' $(h_1,h_2,\bar{h}_3\pm\bar{s})$, where the sign $+$ or $-$ is taken when the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ travel downwards or upwards respectively (Fig. 9). Qualitatively

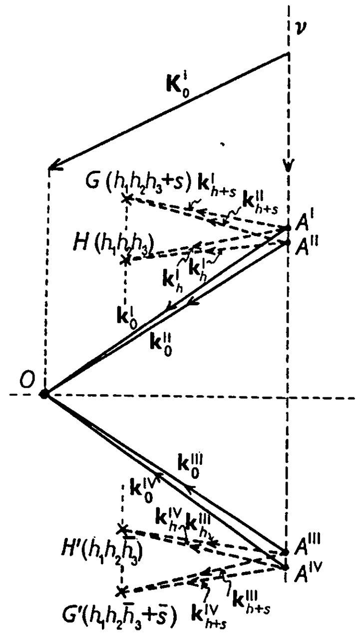

Fig. 9. Ray construction in reciprocal space.

speaking, the new waves can be interpreted as the reflexions of the waves $\mathbf{k}_h^{\mathrm{I}}$, $\mathbf{k}_h^{\mathrm{II}}$, $\mathbf{k}_h^{\mathrm{III}}$ and $\mathbf{k}_h^{\mathrm{IV}}$ by the lattice plane $(00s)$.

In this circumstance, there are three strong waves relating to each wave point, so that the treatment becomes a little more complicated than for the problem of two strong waves, though, as is clear from the discussion given in § 4, the number of the necessary wave points remains four. It is important to note here the fact that the distinction of the Laue and Bragg cases is now lost, because whether the waves $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_h^{\mathrm{II}}$ travel upwards or downwards, their reflected waves travel correspondingly downwards or upwards (the waves $\mathbf{k}_{h-s}^{\mathrm{I}}$ and $\mathbf{k}_{h-s}^{\mathrm{II}}$ and $\mathbf{k}_{h+s}^{\mathrm{I}}$ and $\mathbf{k}_{h+s}^{\mathrm{II}}$ respectively). But, in general, either of the two corresponding waves, for instance $\mathbf{k}_h^{\mathrm{I}}$ and $\mathbf{k}_{h+s}^{\mathrm{I}}$, may be the stronger one according to the given conditions; we call the two cases the quasi-Laue or quasi-Bragg case respectively, according to whether the stronger wave travels downwards or upwards. The problem discussed in § 5 on assuming the simple Bragg case, then, should now be treated again on the assumption of the quasi-Bragg case; but it can be shown that the general conclusion remains unaltered.

There is, however, a more fundamental difference between the results for the simple Laue case and the quasi-Laue case, because it can be proved that an angular range wherein all of the four wave points are imaginary may result also from the quasi-Laue case, not only from the quasi-Bragg case.

Fig. 10 shows the ray scheme for the condition

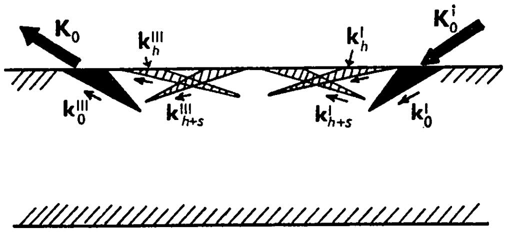

Fig. 10.

when all the wave points are complex, so that there remain only damped waves within the crystal. From this figure and a consideration similar to that given in the previous section, we can readily understand how total specular reflexion can take place in the quasi-Laue case as well as in the quasi-Bragg case. The assumption (ii), therefore, can be removed from the necessary condition for the phenomenon of enhancement, when the role of the s-plane is taken into account.

7. Influence of weak reflexions on the specular reflexion

Though there exist within the crystal many weak waves besides the strong waves, the neglect of the weak waves seems to be harmless as far as the phenomenon of enhancement is concerned. For, as shown, the enhanced specular reflexion owes its intensity almost entirely to some strong waves travelling upwards. The enhancement, however, takes place only exceptionally, so that the specular reflexion is usually rather weak. When the primary wave is the only strong wave in the crystal, and hence the specular reflexion is weak, its intensity might depend more on the presence of the weak waves.

In order to study this, we will discuss a simplified model crystal whose structure is one-dimensionally periodic in the direction perpendicular to the boundary surface. The structure of this kind can be obtained from an arbitrary crystal by averaging the potential field in the direction parallel to the surface.

We assume as before that the crystal is a parallel slab of thickness H. For the simplified crystal there exist only reciprocal-lattice points $(00n)$ $(n = \ldots, -1, 0, 1, 2, \ldots)$ corresponding to the lattice planes parallel to the boundary surface, whose spacing, of the lowest order, is d. The dispersion surface is then approximated by the circles around the reciprocal-lattice points $(00n)$, and, following the considerations in § 4, we may now choose only two wave points. The most convenient choice will be A^{I} and A^{II} (Fig. 11),

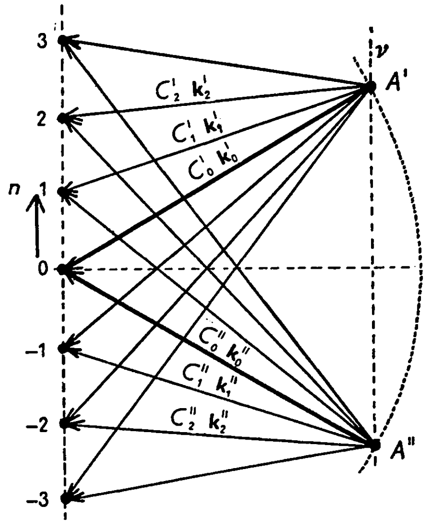

Fig. 11. Ray construction in reciprocal space.

corresponding to the points of intersection of the ν-normal with the circle around the origin (000). The wave vectors permitted within the crystal are

$$\mathbf{k}_n^{\mathrm{I}} = \mathbf{k}_0^{\mathrm{I}} + n \cdot \mathbf{i}/d\,, \quad \mathbf{k}_n^{\mathrm{II}} = \mathbf{k}_0^{\mathrm{II}} + n \cdot \mathbf{i}/d\,, \tag{12}$$

where $\mathbf{k}_0^{\mathrm{I}}$ and $\mathbf{k}_0^{\mathrm{II}}$ correspond to the primary waves relating to the wave points A^{I} and A^{II}, and $\mathbf{i}$ is the unit vector normal to the lattice plane, or the boundary surface.

Let us denote the amplitudes of the primary waves $\mathbf{k}_0^{\mathrm{I}}$ and $\mathbf{k}_0^{\mathrm{II}}$, which are assumed to be the only appreciable ones within the crystal, by c_0^{I} and c_0^{II} respectively; then, from (6), the amplitudes of the weak waves can be expressed in terms of c_0^{I} and c_0^{II} as follows (Bethe, 1928):

$$c_n^{\mathrm{I}} = -\frac{v_{-n}}{\varkappa_0^2 - k_n^{\mathrm{I}2}}\, c_0^{\mathrm{I}}, \quad c_n^{\mathrm{II}} = -\frac{v_{-n}}{\varkappa_0^2 - k_n^{\mathrm{II}2}}\, c_0^{\mathrm{II}}\,. \tag{13}$$

Ψ_0 and Ψ_{00} (the amplitudes of the impinging incident wave and of the specular reflexion, respectively) are related to the amplitudes of waves within the crystal by the ordinary boundary conditions (Bethe, 1928; Laue, 1948) at the upper surface as follows (the second equation corresponds to (9)):

$$\left.\begin{aligned}\Psi_0 &= \frac{1}{2\Gamma_{00}}\sum_n \{(\Gamma_{00}+\gamma_n^{\rm I})c_n^{\rm I}+(\Gamma_{00}+\gamma_n^{\rm II})c_n^{\rm II}\}\,,\\ \Psi_{00} &= \frac{1}{2\Gamma_{00}}\sum_n \{(\Gamma_{00}-\gamma_n^{\rm I})c_n^{\rm I}+(\Gamma_{00}-\gamma_n^{\rm II})c_n^{\rm II}\}\,,\end{aligned}\right\} \quad (14)$$

where $\gamma_n^{\rm I}$ and $\gamma_n^{\rm II}$ are the normal components of $\mathbf{k}_n^{\rm I}$ and $\mathbf{k}_n^{\rm II}$, respectively. Since at the lower boundary no waves enter the crystal, we have the boundary relation

$$\sum_n \{(\Gamma_{00}-\gamma_n^{\rm I})c_n^{\rm I}\exp[2\pi i\gamma_n^{\rm I}H] + (\Gamma_{00}-\gamma_n^{\rm II})c_n^{\rm II}\exp[2\pi i\gamma_n^{\rm II}H]\} = 0\,. \quad (15)$$

In (14) and (15) the index n takes all negative and positive integers including zero.

Now, we assume $\Gamma_{00} \approx \gamma_0$ (where $\gamma_0 \equiv \gamma_0^{\rm I} \equiv -\gamma_0^{\rm II}$), so that $|\Gamma_{00}-\gamma_0|$ is small compared with $\Gamma_{00}+\gamma_0$ or $2\Gamma_{00}$. Also the $\left|\frac{v_{-n}}{\varkappa_0^2-k_n^2}\right|$'s are assumed to be small compared with unity. Neglecting the second-order small quantities, we find from (13), (14) and (15) the ratio r of the amplitudes of the incident wave and of the specular reflexion:

$$r \equiv \frac{\Psi_{00}}{\Psi_0} = (1-\exp[4\pi i\gamma_0 H])\frac{\Gamma_{00}-\gamma_0}{\Gamma_{00}+\gamma_0} - \sum_n{}'(1-\exp[4\pi i\gamma_0 H+2\pi i(n/d)H])\frac{\Gamma_{00}-\gamma_n^{\rm I}}{\Gamma_{00}+\gamma_0}\frac{v_{-n}}{\varkappa_0^2-k_n^{\rm I2}}\,, \quad (16)$$

where Σ' means to drop $n=0$ from the summation.

The ratio of the intensities of the two waves becomes, after averaging over varying H,

$$|r|^2 = \left(\frac{\Gamma_{00}-\gamma_0}{\Gamma_{00}+\gamma_0} - \sum_n{}'\frac{\Gamma_{00}-\gamma_n^{\rm I}}{\Gamma_{00}+\gamma_0}\frac{v_{-n}}{\varkappa_0^2-k_n^{\rm I2}}\right)^2 + \left(\frac{\Gamma_{00}-\gamma_0}{\Gamma_{00}+\gamma_0}\right)^2 + \sum_n{}'\left(\frac{\Gamma_{00}-\gamma_n^{\rm I}}{\Gamma_{00}+\gamma_0}\frac{v_{-n}}{\varkappa_0^2-k_n^{\rm I2}}\right)^2. \quad (17)$$

In order to see the general trend of (17), it is convenient to express it as a function of $\sqrt{E}\,.\sin\Theta$, where Θ is the glancing angle of the incident ray in free space. Γ_{00}, γ_0 and $\gamma_n^{\rm I}$ are given in terms of $\sqrt{E}\,.\sin\Theta$ in the following way:

$$\Gamma_{00} = K\sin\Theta = \sqrt{\alpha}\,.\sqrt{E}\,.\sin\Theta\,,$$
$$\gamma_0 = \varkappa_0\sin\theta = \sqrt{\alpha}\,.\sqrt{(E\sin^2\Theta+eV_0)}\,,$$

$$\gamma_n^{\rm I} = \sqrt{\alpha}\,.\sqrt{(E\sin^2\Theta+eV_0)} - n/d\,,$$
$$\varkappa_0^2-k_n^{\rm I2} = (n/d)(2\sqrt{\alpha}\,.\sqrt{(E\sin^2\Theta+eV_0)}-n/d)\,,$$

where θ is the glancing angle of the primary ray within the crystal and $\alpha = 2m/h^2$.

Fig. 12 exemplifies the result of a numerical calculation of (17), wherein the values of d and the Fourier coefficient V_n, and consequently v_n, are taken from the data concerning the cleavage surface (220) of zincblende crystal, namely we put

$$d = d_{220} \text{ of zincblende} = 1{\cdot}92\ \text{Å},$$
$$v_n = \alpha e V_{2n,2n,0} \text{ of zincblende.}$$

The values of $V_{2n,2n,0}$ of zincblende are given in Table 1 in volts, calculated by using the atomic factors of zinc and sulphur atoms. V_0, the mean inner potential, is put as 12 volts according to the experimental value

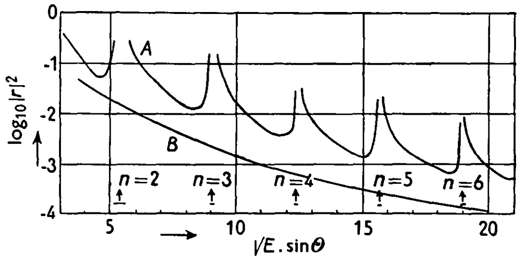

Fig. 12. Calculated intensity distribution of specular reflexion.

Table. 1. *Fourier coefficients* (2*n*, 2*n*, 0) *of the inner potential for zincblende*

n	V_n (volts)
1	7·14
2	2·72
3	1·49
4	0·94
5	0·69
6	0·49
7	0·40

(Yamaguti, 1934, 1939; Kikuchi & Nakagawa, 1934; Miyake, 1935). To obtain the curve A in Fig. 12, n's from -3 to 7 were taken into the summation. The ordinate of the figure is shown on a logarithmic scale; E is measured in volts. The arrows shown above the abscissa indicate the positions of the Bragg reflexions.

Near the Bragg angles the curve A shows the increase due to nearly total reflexion ($|r|^2 = 1$). The length of the strips shown below the arrows indicates the angular ranges of the total reflexions. Of course the theoretical curve loses its meaning in the vicinity of the singular ranges on account of the approximation of the present theory; we must confine our discussion to the middle ranges between successive Bragg reflexions.

The curve B in Fig. 12 indicates

$$|r|^2 = 2\left(\frac{\Gamma_{00}-\gamma_0}{\Gamma_{00}+\gamma_0}\right)^2, \quad (18)$$

which is the intensity of reflexion expected from a parallel slab having a uniform potential V_0, and this corresponds to the specular reflexion when the effect of all weak waves is neglected. On comparing the curves A and B, it becomes clear that the dynamical effect of weak reflexions contributes to the intensity of the specular reflexion much more than the mere jump of the mean inner potential at the crystal surface, though the contribution of the latter is not negligible at smaller glancing angles.

The above result cannot be compared directly with the experiment on account of its approximate nature, but we notice the fact that Fig. 12 seems to reproduce the general trend of the experimental intensity curve shown in Fig. 4. The observed asymmetry of the intensity distribution of the specular reflexion with respect to every Bragg angle is thus explained as the dynamical effect of the weak waves due to the existence of the periodic field in the crystal.

8. General remarks

In the preceding sections it has been shown that specular reflexion can be accounted for by the dynamical theory. We believe that the whole circular group of spots may be explained along the same lines.

Though the geometrical positions of the spots can be explained in terms of the kinematical theory including absorption, this simple theory seems not to be capable of explaining the details of the intensity curve: especially, as mentioned before, the phenomenon of the enhancement can in no way be treated by the kinematical theory.

Through the above theory of the phenomenon of enhancement, it is shown that the electron wave can be totally reflected by the crystal surface, even when the Bragg condition of the lattice plane parallel to the surface is not satisfied, provided that (*a*) the Bragg reflexion in a side direction takes place, (*b*) the angle between its ray direction and the surface is smaller than the critical angle β_c, and, at the same time, is of comparable order of magnitude to the Bragg angle corresponding to (00*s*), (*c*) the surface is the mirror plane of the lattice, and (*d*) the structure amplitude of (00*s*) is sufficiently large.

The similar phenomenon, however, will never be expected to be observed for X-rays in spite of the close formal analogy of the dynamical theories for X-ray and electron waves. This is because the refractive index of crystals is smaller than unity for the X-ray wave in contrast with the case of electron, so that there can be no total reflexion of the X-ray waves *inside* the crystal at the boundary surfaces.

The present dynamical theory of the specular reflexion is developed without assuming absorption. It should therefore be regarded as a preliminary stage. The formal modification of the theory, however, will not be difficult when the phenomenological complex Fourier coefficients, as introduced by Slater (1937) and Molière (1939), are utilized. Absorption may have a considerable influence on the calculated intensity, but further detailed considerations along these lines are not given here because the theoretical basis of the absorption of the electron wave is yet not well founded. As to the phenomenon of the enhancement, the essential process, as explained in the former sections, will not be changed by the absorption effect.

The detailed calculation concerning the phenomenon of enhancement, which was not included in the present paper, will be reported elsewhere.

References

Artmann, K. (1947). *Z. Phys.* **124**, 80.
Beeching, H. (1935). *Phil. Mag.* **20**, 841.
Bethe, H. (1928). *Ann. Phys., Lpz.* **87**, 55.
Honjo, G. (1949). *J. Phys. Soc. Japan*, **4**, 352.
Kikuchi, S. & Nakagawa, S. (1933*a*). *Sci. Pap. Inst. Phys. Chem. Res. Tokyo*, **21**, 80.
Kikuchi, S. & Nakagawa, S. (1933*b*). *Sci. Pap. Inst. Phys. Chem. Res. Tokyo*, **21**, 256.
Kikuchi, S. & Nakagawa, S. (1934). *Z. Phys.* **88**, 757.
Kirchner, F. & Raether, H. (1932). *Phys. Z.* **33**, 510.
Lamla, E. (1938*a*). *Ann. Phys., Lpz.* **32**, 178.
Lamla, E. (1938*b*). *Ann. Phys., Lpz.* **32**, 225.
Lamla, E. (1938*c*). *J. reine angew. Math.* **179**, 134.
Laue, M. v. (1948). *Materialwellen und ihre Interferenzen*, chap. 5. Leipzig: Akademische Verlag.
Miyake, S. (1935). *Sci Pap. Inst. Phys. Chem. Res. Tokyo*, **26**, 216.
Molière, K. (1939). *Ann. Phys. Lpz.* **34**, 461.
Slater, J. C. (1937). *Phys. Rev.* **51**, 840.
Raether, H. (1932). *Z. Phys.* **78**, 527.
Thomson, G. P. & Cochrane, W. (1939). *Theory and Practice of Electron Diffraction*, chap. 19. London: Macmillan.
Yamaguti, T. (1934). *Proc. Phys.-Math. Soc. Japan*, **16**, 95.
Yamaguti, T. (1939). *Proc. Phys.-Math. Soc. Japan*, **21**, 375.