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THE OSCILLATION METHOD OF X-RAY ANALYSIS OF CRYSTALS

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ABSTRACT

In the oscillation method of crystal analysis a crystal is mounted on an axis and turned back and forth through an angle of about 30° from preferably a known position. A beam of monochromatic X-rays normal to the axis of rotation strikes the crystal and is diffracted. The spectra are recorded on a photographic plate. The coordinates for each spot or spectrum are measured. These data are used for the construction of a projection of a new lattice called the reciprocal one with respect to the crystal space lattice. In the projection the indices of each plane producing a spectrum can be read off directly. The advantages of this method especially in the case of crystals with low symmetry are mentioned below. It is also possible to distinguish with it members of groups like that of the feldspars, which is extremely difficult, if possible at all, with the Laue and powder methods.

As comparatively few students will be able to understand the method thoroughly by merely reading this paper, therefore, the application of the method will be described shortly by a detailed example, that of the determination of the space group of analcite. For a proper understanding of this method the student should have a working knowledge of the simplest formulas of spherical trigonometry and of analytical geometry.

INTRODUCTION

Since the discovery of the diffraction of X-rays by crystals in 1912, probably no method of crystal-analysis has promised greater success for the future than the rotation method by Seemann[1] and Polanyi,[2] and the oscillation method by Schiebold.[3] While in the rotation method the crystal is rotated 360° about an axis, in the oscillation method the crystal is turned back and forth about an axis through a limited number of degrees (usually 30).

Since this method has been described but briefly[4] in America, and only one paper of any length has appeared in English[5] an

[1] Seemann, H.; *Phys. Zeitschr.*, vol. **20**, p. 169 (1919).

[2] Polanyi, M.; *Die Naturwissenschaften*, vol. **9**, p. 337 (1921).

[3] Schiebold, E.; *Fortschritte der Mineralogie, Krist. und Petrog.*, vol. **11**, pp. 113–280 (1927).

[4] Wyckoff, R. W. G.; The structure of crystals, *New York* (1924).

[5] Bernal, J. D.; *Proc. Royal Soc. London*, vol. **113A**, p. 116.

explanation and discussion of the method with one or two detailed examples may be of value.

For the following reasons the oscillation method can be used to great advantage where single crystals, though they may be small, are available: (1) The lengths of the edges of the unit cell of a crystal, as well as the number of molecules in it, can be determined with great precision, regardless of the system of crystallization. Neither the powder nor the Laue method can give us this information in crystals of low symmetry. (2) If the crystal is oscil-

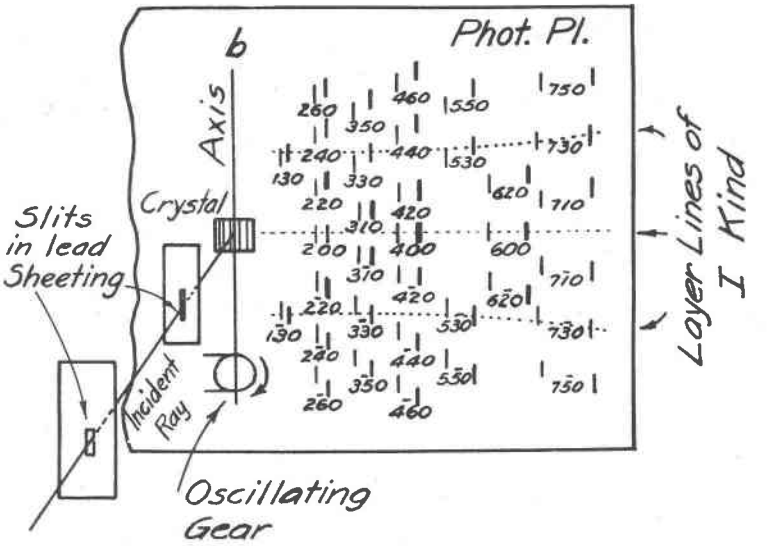

Figure 1

The relation of the crystal mounted on the axis of rotation with respect to the spectra produced on the photographic plate. The spectrogram is that of adularia oscillated about the crystallographic b axis. Each spectrum is represented by a *pair* of short lines. The line nearer the axis is the β, the one farther away the α line of the K radiation of molybdenum. Usually only the α line is used. Some of the layer lines of I kind (hyperbolas) are indicated by dotted lines. Layer lines of II kind, parallel to the axis, are sufficiently prominent to be seen without further indications. The widths of the lines of the spectra in the diagram are supposed to show intensity of reflection. (After Schiebold.)

lated in a sufficient number of directions the space group can be determined reliably. The Laue method and, where the symmetry is lower than hexagonal, the powder method, are not to be depended upon for this information. (3) There are often sufficient X-ray

spectra recorded on the plates to make important deductions as to parameters of the structure. At present the Laue method is the one most widely used for this purpose.

Bragg's[6] ionization method is and will of course continue to be of great importance in special investigations, but ordinarily it would require much more time than could be given to a problem.

It is not intended to give descriptions of apparatus and the technique necessary for the application of the method, but an

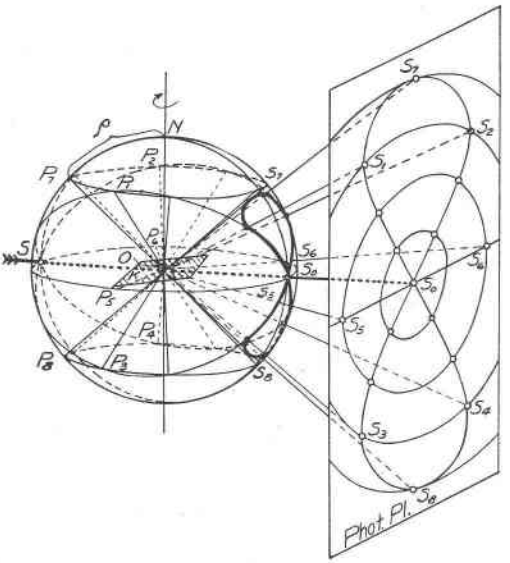

Figure 2. (After Schiebold).

explanation of the principles and especially of the graphical methods used in the interpretation of spectrograms. This the writer studied at the Mineralogische Institut at Leipzig under Prof. E. Schiebold and Prof. F. Rinne, to whom he is greatly indebted.

Fig. 1 shows diagrammatically the arrangement of the axis of rotation upon which the crystal is definitely oriented with reference to the incident X-rays. The spectra produced on the photographic plate, which is normal to the incident ray, are shown also. As far as possible the same letters will be used for corresponding points in all the diagrams accompanying this paper.

[6] Bragg, W. H., and Bragg, W. L.; X-rays and crystal structure. *London* (1925).

Geometrical Relations in the Oscillation Method

Fig. 2 illustrates how reflection occurs in the rotating crystal method, of which the oscillation method is a special case. SOS_0 is the incident monochromatic X-ray which meets a plane K of a crystal in O, the center of the spherical projection. ON is the axis about which the crystal is rotated. For the sake of convenience we make it normal to the incident ray SOS_0. Points of reflection are recorded on the photographic plate (Phot. Pl.), which is normal to the ray SOS_0. Reflection by the plane K depends on α[6a], the glancing angle between incident ray and plane K, and upon ρ, the angle between the normal upon the plane K and the axis ON. The normals P_1, P_2, P_3, P_4, to the plane K, when at various positions during rotation through 360° form a conic surface whose vertex angle is 2 ρ. In general a plane K during a complete rotation about ON fulfills the well known Bragg's equation,[7]

$$n\lambda = 2d \sin \alpha, \text{ four times.} \qquad (1)$$

This gives rise to four spots of the same order on the plate, for example S_1, S_2, S_3, S_4. If the plane K with indices hkl happens to be in the zone $[uvw]$ parallel by construction to the axis ON, ($hu+kv+lw=0$ is the equation of the plane),[8] the vertex angle of the cone is 180°. Therefore, each two of four reflections will coincide and only two will occur (for example S_5, and S_6). These two lie in the so-called principal spectrum (*Hauptspektrum*). Other special positions are possible but improbable.[9]

For the understanding of the angular relations and the interpretation of the spots on the photographic plate Fig. 3 has been drawn. AN is the axis of rotation, SAO^1 is the incident ray which lies in the plane of the principal meridian SNO. SA is normal to AN and the photographic plate (Phot. Pl.). K is the plane of reflection. AP is normal to it. AR is the reflected ray which meets the plate at R^1. ρ_P is the angle NP between the normal to the plane K and the axis AN. $\rho_R = NR$ is the angle between the reflected ray and the axis AN. ϕ[9a] is the angle OQ_P in the equatorial

[6a] α is called Θ by the Braggs. The reader is referred to pp. 6–15 and 73–103 of their elementary book (*op. cit.*).

[7] *Op. cit.*, p. 13.

[8] See for example: Dana's Textbook of Mineralogy, 3d ed., p. 46 (1922).

[9] Schiebold, *op. cit.*, p. 119.

[9a] The greek letter ϕ in the drawings looks slightly different from the printed one.

plane, or in other words the meridian angle of the pole P. $\phi_R = OQ$ is the meridian angle of the pole R. $\bar{\phi} = NSP = Z^1O^1R^1$ is the angle between the great circle $SPRO$ and the principal meridian. The coordinate system x, z on the plate is right-angled. $\bar{\phi}$ and $\bar{r}$ are the polar coordinates of R^1. The points O and O^1 could have been made to coincide except for the sake of clearness

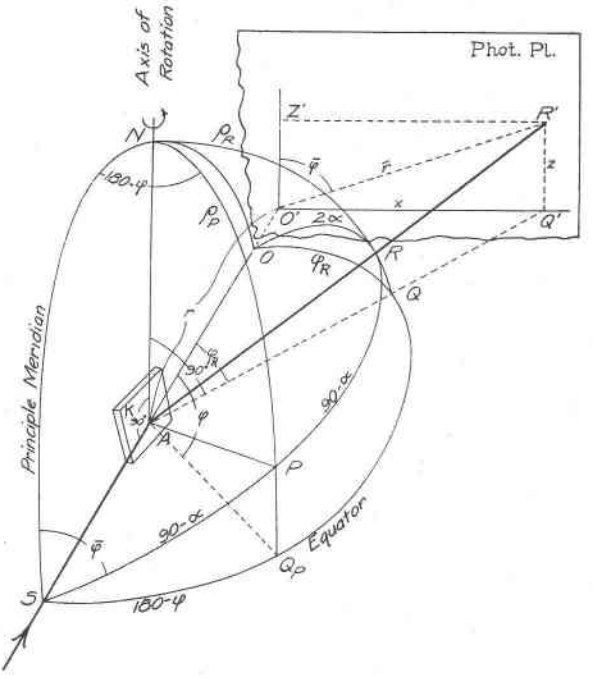

Figure 3.

of the drawing. $r = AO^1$ is the distance of the plane K to the photographic plate.

It is obvious that $OR = 2\alpha$ when reflection occurs (see equation (1)). Since AP is normal to K, $SP = 90° - \alpha$ and $PR = 90° - \alpha$. In $\Delta\ SPN$ by the sine law,

$$\frac{\sin(90°-\alpha)}{\sin(180°-\phi)} = \frac{\sin\rho_P}{\sin\bar{\phi}} = \frac{\cos\alpha}{\sin\phi}.$$

By the cosine law,

$\cos(90°-\alpha) = \cos 90° \cdot \cos\rho_P + \sin 90° \cdot \sin\rho_P \cdot \cos(180°-\phi)$ or, $\sin\alpha = \sin\rho_P \cdot \cos(180°-\phi)$ (2)

Suppose x and y or $\bar{r}$ and $\bar{\phi}$ to be given:

$$\bar{r}=r \cdot \tan 2\alpha$$

$$\frac{\bar{r}}{r}=\tan 2\alpha$$

$$x=\bar{r}, \sin \bar{\phi}$$

$$z=\bar{r} \cdot \cos \bar{\phi}$$

Substituting, $x=r \cdot \tan 2\alpha \cdot \sin \bar{\phi}$

$$z=r \cdot \tan 2\alpha \cdot \cos \bar{\phi}$$

By construction,

$$x=r \cdot \tan \phi_R$$

$$z=AQ' \cdot \cot \rho_R=\frac{r}{\cos \phi_R} \cdot \cot \rho_R \tag{4}$$

By the cosine law in ΔNRO,

$$\cos 2\alpha=\cos 90^\circ \cdot \cos \rho_R+\sin 90^\circ \cdot \sin \rho_R \cdot \cos \phi_R$$

or, $$\cos 2\alpha=\sin \rho_R \cdot \cos \phi_R$$

and, $$\cos \rho_R=\cos 90^\circ \cdot \cos 2\alpha+\sin 90^\circ \cdot \sin 2\alpha \cdot \cos \bar{\phi} \tag{5}$$

or, $$\cos \rho_R=\sin 2\alpha \cdot \cos \bar{\phi}=2 \sin \alpha \cdot \cos \alpha \cdot \cos \bar{\phi} \tag{6}$$

In ΔNPO by the cosine law,

$$\cos \rho_P=\cos 90^\circ \cdot \cos(90^\circ-\alpha+2\alpha)+\sin 90^\circ \cdot \sin(90^\circ-\alpha+2\alpha) \cdot \cos \bar{\phi}$$

or, $$\cos \rho_P=\sin (90^\circ+\alpha) \cdot \cos \bar{\phi}=\cos \alpha \cdot \cos \bar{\phi} \tag{7}$$

Substituting from (7) $\cos \alpha=\dfrac{\cos \rho_P}{\cos \bar{\phi}}$

we have another form for $\cos \rho_R$,

$$\cos \rho_R=2 \sin \alpha \cdot \cos \rho_P \tag{8}$$

The Reciprocal Space Lattice

It would be difficult to explain the use of the oscillation method especially the graphical interpretations without introducing the conception of the reciprocal lattice, used first by Ewald.[10]

If we consider a crystal as a simple space lattice we may choose three directions or vectors $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$, in it as primitive translations, (Fig. 4). These may serve also as axes[11] when they will be marked

[10] *Zeitschr. f. Kristallographie*, vol. **56**, p. 129 (1921).

[11] It is customary to use German letters for vectors or directed magnitudes and corresponding Latin ones for absolute lengths. In Fig. 4, then, $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$, imply not only the lengths of the edges of the parallelopiped but also their directions. a, b, c, on the other hand, are used either just for the lengths of the edges or to designate the three crystallographic axes. A primitive translation is the vector from an atom to the nearest identical atom in the direction of the vector. See f. example Wyckoff, R. W. G., *op. cit.*, pp. 56–58.

a, b, c to distinguish them from the translations $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$. A plane in such a lattice may be designated by Miller indices h, k, l, where the intercepts of such a plane are a/h, b/k, c/l, on a, b, c, respectively.

The origin of the unit cell or parallelopiped is at 0. The planes which have as indices integers prime to each other are farthest

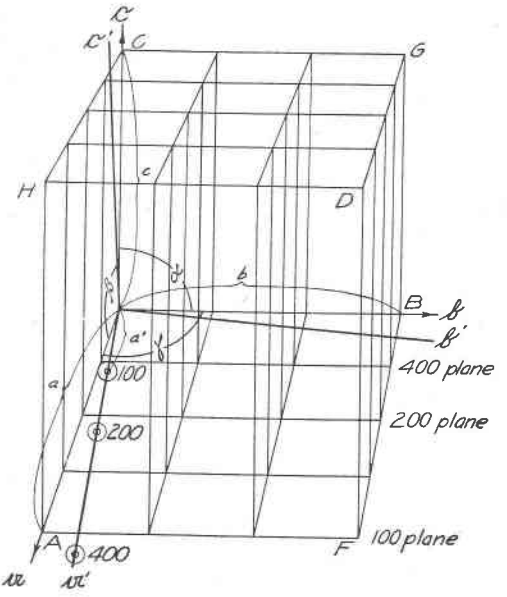

Figure 4.

away from the origin in the cell. They are responsible for the I order reflections. Planes which are sub-multiple distances of the first mentioned planes away from the origin have indices that are multiples of the indices consisting of integers prime to each other. These intermediate planes produce the higher orders of reflection.[12] In Fig. 4, considering the pinacoid, for example, plane (100) would be farthest away from the origin in the unit cell $OAFBCHDG$, and would produce I order reflections. The plane (200) would be half way between and would cause II order reflections and so forth.

"Now from the planes of the lattice, as so defined (quoting Bernal[13]) another lattice can be built up each point of which lies

[12] Bragg, op. cit., p. 13.

[13] *Op. cit.*, p. 118.

on the normal from the origin to a plane and at a distance h[14] from the origin, h and the spacing d of the plane being related by

$$hd = k^2 \tag{9}$$

where k is a constant. In other words, each point in the new lattice is the reciprocal polar of a plane in the old lattice in a sphere of radius k." The new lattice is called the reciprocal lattice (reziprokes Gitter) of the old. For every plane in one may be found a corresponding point in the other as shown in Fig. 4 for the planes (100), (200), and (400). The reciprocal lattice has the same origin as the old lattice but is referred to the three vectors $\mathfrak{a}^1$, $\mathfrak{b}^1$, $\mathfrak{c}^1$. With respect to $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$,[15]

$$\mathfrak{a}^1 = \frac{k^2 bc}{V} \sin\alpha, \text{ and is normal to the plane } \mathfrak{bc},$$

$$\mathfrak{b}^1 = \frac{k^2 ca}{V} \sin\beta, \text{ and is normal to the plane } \mathfrak{ca},$$

$$\mathfrak{c}^1 = \frac{k^2 ab}{V} \sin\gamma, \text{ and is normal to the plane } \mathfrak{ab},$$

a, b, c are the lengths of the vectors $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$ respectively, α, β, γ, are the angles between the primitive translations. V is the volume of the unit cell or parallelopiped. With these new vectors $\mathfrak{a}^1$, $\mathfrak{b}^1$, $\mathfrak{c}^1$ whose lengths are a^1, b^1, c^1, respectively, we build the reciprocal lattice in which h, k, l are the indices of a point which corresponds to a plane (hkl) in the original lattice. The reciprocal lattice has the following important properties: (1) the vector $\mathfrak{h}$ (Fig. 6) to a point P_{hkl} of the reciprocal lattice is the normal to the corresponding plane (hkl) of the space lattice. (2) The length h of the vector $\mathfrak{h}$ to the first point encountered in a given direction in the reciprocal lattice is equal to $1/d_{(hkl)}$. d is the distance between equivalent planes in the space lattice. (3) The coordinates of a point P_{hkl} in the reciprocal lattice are proportional to the indices of the corresponding plane (hkl) in the space lattice, or in other words if x^1, y^1, z^1 are the coordinates, $x^1 = a^1 h$, $y^1 = b^1 k$, $z^1 = c^1 l$. (4) The volumes of the unit cells of the two lattices are reciprocal to each other $V = 1/V^1$. V^1 is the volume of the unit of the reciprocal lattice.

[14] I substituted h for Bernal's ρ because ρ is used for angles in my paper. This h has no relation to the index h, but is the length of the vector $\mathfrak{h}$ used further on. d is always the spacing between equivalent planes.

[15] Ewald, *op. cit.*

Bernal says,[16] "It is difficult to obtain a physical picture of the phenomena in the reciprocal lattice corresponding to the diffraction of X-rays in crystals. By assuming Bragg's law, however, we can obtain a geometrical one." If the angle between the incident ray and the reflecting plane in the space lattice satisfies the equation (1) reflection occurs. In the reciprocal lattice α is the angle between the incident ray and the plane which is normal to the radius vector $\mathfrak{h}$ connecting the origin O with the point P_{hkl} which

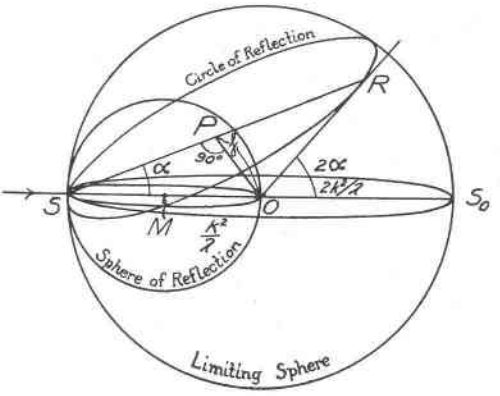

Figure 5. (After Bernal.)

corresponds to the plane (hkl) in the space lattice. (See Fig. 5.) By equation (9)

$$h=\frac{k^2}{d} \text{ or, } d=\frac{k^2}{h} \tag{10}$$

Substituting (10) in equation (1), we get (assuming for the present $n=1$)

$$\sin\alpha=\frac{\lambda h}{2k^2} \tag{11}$$

Fig. 5 (after Bernal) is an attempt to present reflection in the reciprocal lattice diagrammatically. SOS_0 is the incident ray. A sphere with a radius $2k^2/\lambda$ is drawn around O as center. The point P in the sphere is the point of the reciprocal lattice corresponding to the reflecting plane of the space lattice. OP is the radius vector $\mathfrak{h}$ which is normal at P to a plane SR which cuts the

[16] *op. cit.* p. 120.

sphere in the circle marked "circle of reflection." SPR is a straight line in that plane. Then

$$\sin OSP = \frac{h}{\frac{2k^2}{\lambda}} = \frac{\lambda h}{2k^2} \text{ which is equation (11) Bragg's equation.}$$

h is the length of the vector $\mathfrak{h}$ in this equation.

The incident ray SO, therefore, will be reflected along OR by a plane in the space lattice which corresponds to the point P in the reciprocal lattice. Therefore the angle $ROS_0 = 2\alpha$. Further all rays that can be reflected at all by the plane corresponding to P lie along the surface of the cone OSR, *and the incident and reflected rays are diametrically opposite generators of this cone.* A study of Fig. 5 and equation (11) shows that no point of reflection can lie outside of the sphere with $2k^2/\lambda$ as radius, for if h were longer than $2k^2/\lambda$, sin α would be greater than 1. The sphere with $2k^2/\lambda$ as radius, therefore, is called the "limiting sphere," (Begrenzungskugel).

Since the angle SPO must always be 90°, all points P will lie on the surface of a sphere with a radius k^2/λ whose center M lies half way between S and O. This sphere is called the "sphere of reflection" (Ausbreitungskugel by Ewald and Schiebold). It is exceedingly useful in finding the indices of the spots in the photograph, for only those planes can reflect whose points P in the process of rotation about O *pass through the surface* of the sphere of reflection. Therefore the number of reflections is limited by the angle of rotation or oscillation, respectively.

The Evaluation of Reflections in the Photograms

The plotting of reflections and the determination of their indices resemble the gnomonic projection. (See Fig. 11.) If in equation (9) we choose $k=1$ we have

$$h_{hkl} \cdot d_{hkl} = 1, \text{ or } h_{hkl} = \frac{1}{d_{hkl}}$$

d_{hkl} = the distance between equivalent planes (hkl). In order to find h_{hkl}, the length of vector $\mathfrak{h}_{hkl}$, we construct the auxiliary figure 7. With two arbitrary lengths a^1, b^1, we draw a reciprocal lattice in two dimensions. With the radius $k_2/\lambda = 1/\lambda$ (λ here is also arbitrary) we draw the circle marked "sphere of reflection in

Fig. 5, so that O will lie on its surface. Any other point P_{hkl}, of the lattice that happens to be on the surface is a possible point of reflection. $OP_{hkl}=h_{hkl}$, therefore. Since OMP is an isosceles triangle, the normal from M upon OP bisects the angle $2\alpha=OMP$.[16a]

$$OL=PL=OM\cdot\sin\alpha$$

therefore, $\frac{1}{2}h=1/\lambda\cdot\sin\alpha$, and since $h_{hkl}=1/d_{hkl}$

$$h_{hkl}=\frac{1}{d_{hkl}}=\frac{2}{\lambda}\sin\alpha_{hkl} \tag{12}$$

which is equation (1) in a slightly different form.

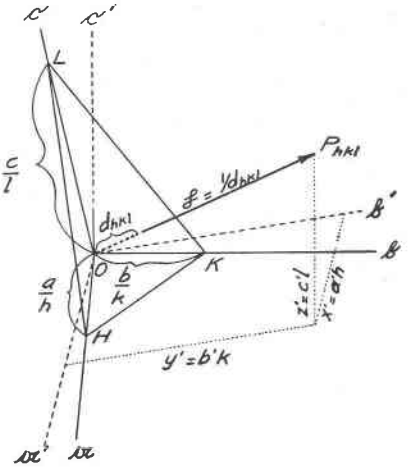

Figure 6

Axial relations of space lattice to reciprocal lattice. $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$ are primitive translations and axes of space lattice. $\mathfrak{a}^1$, $\mathfrak{b}^1$, $\mathfrak{c}^1$ are primitive translations of reciprocal lattice. $\mathfrak{a}^1$ normal to plane $\mathfrak{bc}$, $\mathfrak{b}^1$ normal to plane $\mathfrak{ca}$, $\mathfrak{c}^1$ normal to plane $\mathfrak{ab}$.

But the length of $\mathfrak{h}$ is one of our polar coordinates in the reciprocal lattice corresponding to $\bar{r}$ on the spectrogram of the space lattice (Fig. 3), as will be seen more clearly in Fig. 11. For plotting the length of $\mathfrak{h}$ we need a factor which will make our projections of a convenient size. We choose $R_0\lambda/2$ ($Ro=500$ mm. usually is convenient). Multiplying both sides of equation (12) by this factor $R_0\lambda/2$ we get

$$\frac{R_0\lambda}{2d_{hkl}}=Ro\cdot\sin\alpha_{hkl}. \tag{13}$$

[16a] The incident ray is parallel to MO and not as shown in Fig. 7.

The radius of the sphere of reflection $1/\lambda$ when multiplied by the same factor becomes $Ro/2$, or 250 mm. if we choose $Ro = 500$ mm. To locate a point P_{hkl} in the projection of the reciprocal lattice corresponding to a plane (hkl) we use the polar coordinates $Ro \cdot \sin \alpha_{hkl}$ and $\rho_{P_{hkl}}$ which we compute with the aid of equation (7).

An examination of the spots in a photogram taken by the oscillation method (for example Fig. 1) shows that they are arranged in distinct lines or curves at right angles to the trace of the

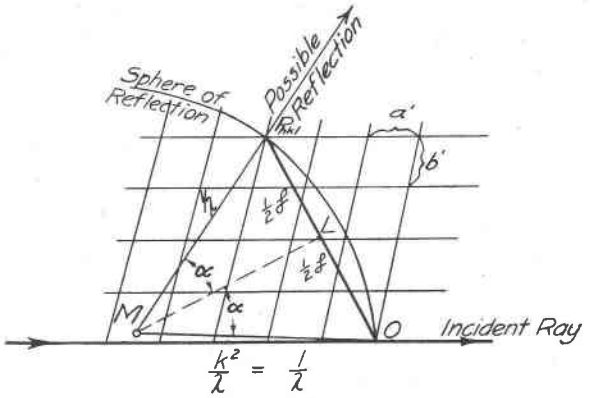

Figure 7.[16a]

axis of rotation provided the axis is parallel to a prominent crystallographic direction. These lines or curves are called "layer lines of *I* kind" (Schichtlinien I. Art). In order to explain these lines we go back to the conception of the one dimensional lattice by Ewald.[17] We *imagine* the space lattice to be made up of three one-dimensional ones. Each of them is parallel to one of the primitive translations $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$. A one-dimensional lattice consists of a line on which atomic centers are spaced at equal intervals. We orient the lattice so that the one-dimensional one parallel to $\mathfrak{c}$ is also parallel to the axis of rotation. In Fig. 8 the one-dimensional lattice with the direction $[u_3v_3w_3]$ of the c axis and of the axis of rotation is shown. (Any other direction or line on which atoms occur at regular intervals could have been chosen.) Neighboring atomic centers A_0, A_1 are a primitive translation $T_{[u_3v_3w_3]}$ apart.

[17] Ewald, P. P., Kristalle und Röntgenstrahlen, *Berlin*, **1923**, p. 42.

The parallel beam of monochromatic rays S_1S_2 meets the lattice and is diffracted. Since each atom sends out spherical wave impulses[18] the diffracted beams lie on a cone whose vertex angle is equal to 2γ. The axis of the cone is the line $[u_3v_3w_3]$ as shown in Figs. 8 and 9. Diffraction (or reflection) occurs only in those directions where the phase difference between impulses from adjacent atoms is an integral multiple of the wave length λ. In other words, in Fig. 8

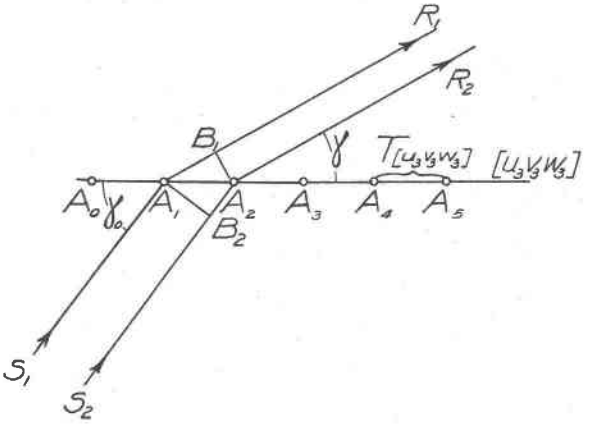

Figure 8.

$$A_1B_1 - A_2B_2 = 0,\ 1,\ 2,\ 3,\ \cdots,\ l'\lambda$$

$$A_1B_1 = T_{u_3v_3w_3} \cdot \cos\gamma$$

$$A_2B_2 = T_{u_3v_3w_3} \cdot \cos\gamma_0$$

$$A_1B_1 - A_2B_2 = l'\lambda = T_{u_3v_3w_3} \cdot (\cos\gamma - \cos\gamma_0) \qquad (14)$$

This equation and two other exactly similar ones to be mentioned later are called the Laue equations. l' is the order of reflection in the one dimensional lattice just as n is the order in the three dimensional lattice in equation (1).

In Fig. 9 we assume a special case, that of the incident ray S being normal to the axis of rotation which is parallel to a prominent crystallographic direction. The cones of reflection of the different orders cut the photographic plate in hyperbolas. The O order will be a special case, namely a straight line. In the figure we must

[18] These spherical waves can be explained by Huygens' principle. Only waves which are exactly in phase lie on the surface of the cone.

imagine the cones to extend also downward from the *O* order. All spots, therefore, have these hyperbolas or layer lines for one of their loci. They become straight lines normal to the axis of rotation in the projection of the reciprocal lattice. Figs. 3 and 8 show that the angle ρ_R and γ are identical. Since we are dealing with

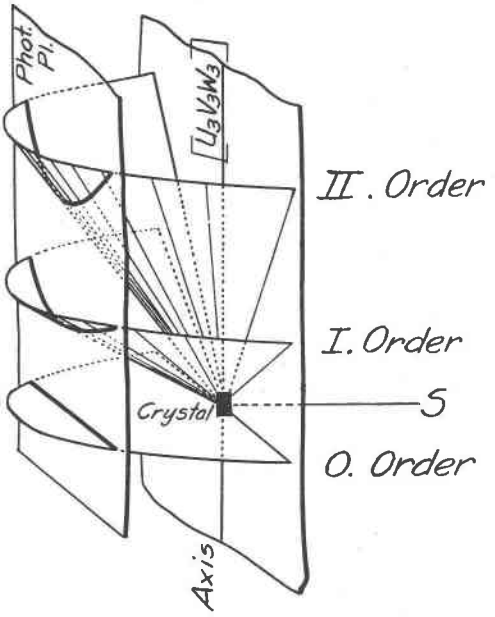

Figure 9. (After Ewald.)

an incident ray *normal* to the direction $[u_3v_3w_3]$ the cosine of γ_0 becomes 0. We may substitute in (14) ρ_R and get

$$l'\lambda = T_{u_3v_3w_3} \cdot \cos \rho_R \qquad (14')$$

By equation (6) and (8) $\cos \rho_R = \sin 2\alpha \cdot \cos \phi = 2 \sin \alpha \cdot \cos \rho_P$. Substituting in (14′) we get

$$\sin 2\alpha \cdot \cos \bar{\phi} = 2 \sin \alpha \cdot \cos \rho_P = \frac{l'\lambda}{T_{u_3v_3w}}$$

$$= \text{a constant for each layer line} \qquad (15)$$

Equation 15 may be used for calculating the primitive translation $T_{u_3v_3w_3}$ by taking the mean value of $T_{u_3v_3w_3}$ calculated from all the spots in the photograph. A shorter and more accurate way is

to use the reflections of the principal spectrum along the 0 layer line, if we know the orientation of the crystal with respect to the principal meridian and to the incident ray. The principal spectrum consists of reflections from the faces that lie in the zone of the axis of rotation. Therefore it is at right angles to the direction of the axis (See Fig. 1) and to $[u_3v_3w_3]$ which by construction is parallel to the axis. The advantage of taking a prominent crystallographic zone or direction for the axis of rotation is obvious now, for it insures prominent reflections in the principal spectrum. These can be evaluated by equation (1) if we know the approximate T from equation (15), as will be shown in a paper on the structure of analcite. l' in (14) and (15) is an integer. It tells us how many times we must take the distance $T_{u_3v_3w_3}$ in the direction $[u_3v_3w_3]$ to arrive at a certain point. It is the index along $[u_3v_3w_3]$. l' must remain a constant for each layer line as is obvious from equation (14) and (15). (Note the resemblance to the gnomonic projection in this respect.)

Layer lines in the general direction of the axis of rotation (Fig. 1) also appear, if prominent crystallographic planes are normal to the axis. A prominent zone will then be at right angles to the axis. The reflections of such a zone lie on a curve which is almost a straight line near the principal spectrum. All the points in the reciprocal lattice corresponding to the planes in such a zone lie in a plane. Therefore, these layer lines are straight lines in the reciprocal lattice. Schiebold calls them layer lines of *II* kind. Only the index referring to the coordinate in the direction of the axis of rotation changes along these lines as shown later.

In order to make our evaluation quite general regardless of which prominent crystallographic direction is the axis of rotation we look at the portion of (15) $l'\lambda/T_{u_3v_3w_3}$. On each layer line of *I* kind the index l' is a constant. If the direction parallel to the axis of rotation is not one of the primitive translations $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$, but some other crystallographic direction, we employ the well known transformation formulas to find the indices with respect to the primitive translation $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$.

$$h' = hu_1 + kv_1 + lw_1$$
$$k' = hu_2 + kv_2 + lw_2$$
$$l' = hu_3 + kv_3 + lw$$

In these equations $[u_1v_1w_1] = \bar{\mathfrak{a}}$
$[u_2v_2w_2] = \bar{\mathfrak{b}}$
$[u_3v_3w_3] = \mathfrak{c}$

$\bar{a}$, $\bar{b}$, $\bar{c}$ are the three primitive directions or axes of a new orientation of the lattice with reference to the crystal axes a, b, c. $\bar{c}$ is the axis of rotation. h', k', l' are the indices with reference to the directions $\bar{a}$, $\bar{b}$, $\bar{c}$. h, k, l are the indices with reference to the crystal axes a, b, c, (or in other words with reference to the primitive translations $\mathfrak{a}$, $\mathfrak{b}$, $\mathfrak{c}$ of the unit cell). For every layer line of I kind we get the equation:

$$l' = s = hu + kv + lw = 0, \pm 1, \pm 2, \cdots \pm \text{ integer}$$

$[uvw]$ are the indices of the axis of rotation, and (hkl) the indices of the reflecting plane both with reference to the crystal axes

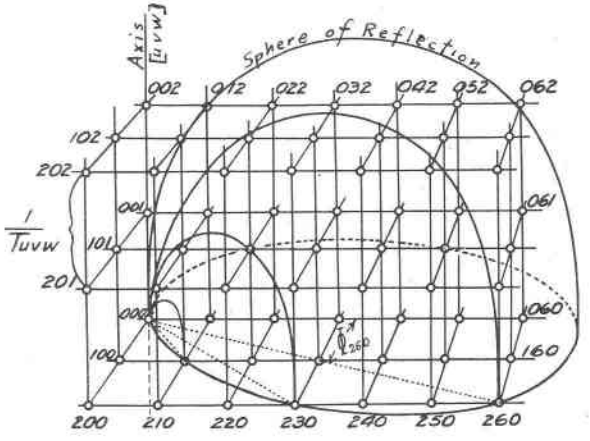

Figure 10

Sphere of reflection penetrating a portion of the reciprocal lattice. For the sake of clearness only some of the points have received indices. The small circle connecting point (000) and point (260) is the limiting circle for the vertical plane in which the two points lie. Its radius is $R_0/2 \cdot \sin \Phi_{260}$.

a, b, c. s I propose to call the index of summation (Aufzählungsindex). This equation *must* be satisfied, if the indices have been placed correctly.

The layer lines of II kind can be explained similarly. The loci for the reflection of the one-dimensional lattice parallel to the axis of rotation are cones as shown in Figs. 7 and 8. The other two primitive directions $\bar{a}, [u_1v_1w_1]$ and $\bar{b}, [u_2v_2w_2]$ of the space lattice can also be imagined as two one-dimensional lattices with loci which are cone surfaces about these directions. Their equations are then similar to (14):

$$T_{u_1v_1w_1} \cdot (\cos \alpha - \cos \alpha_0) = h'\lambda \quad (16)$$

$$T_{u_2v_2w_2} \cdot (\cos \beta - \cos \beta_0) = k'\lambda \quad (17)$$

The loci of the intersections of these two cones lie on curves of the 4th degree in the photographic plate. The curves are almost

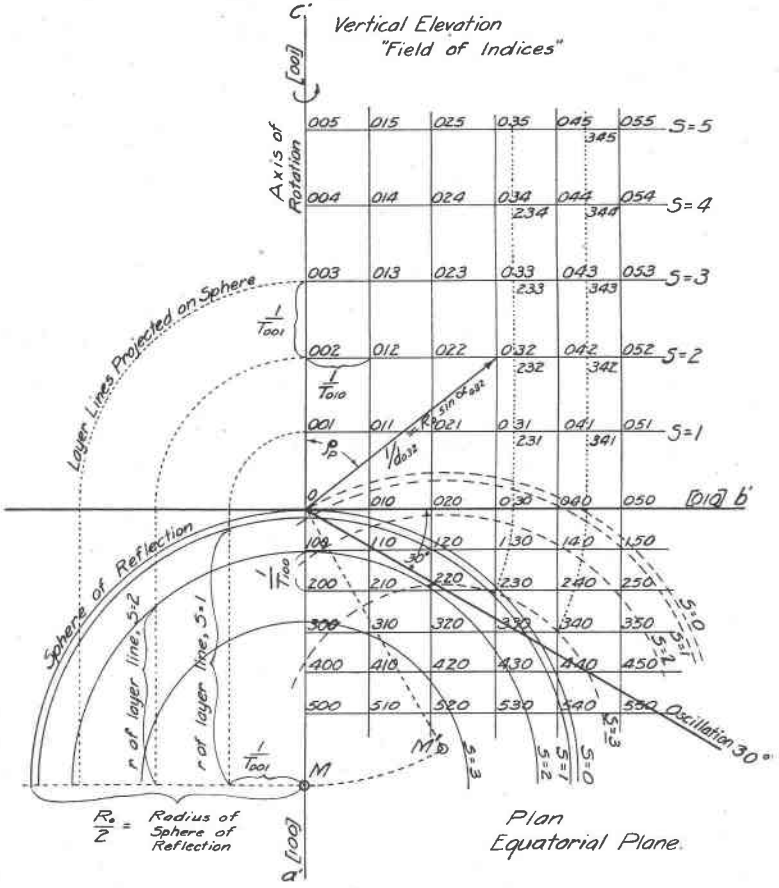

Figure 11.

Vertical elevation and plan of a right angled reciprocal lattice. Only some of the points of the plan have been projected upon the elevation. As an example of how a point is plotted the polar coordinates of point (032) are shown in the vertical projection.

straight lines near the center and near the principal spectrum. These layer lines of the *II* kind are straight lines in the reciprocal lattice as mentioned before. Every point on one must according to the transformation formulas satisfy the equations:

$$h' = hu_1 + kv_1 + lw_1 = \text{constant}$$
$$k' = hu_2 + kv_2 + lw_2 = \text{constant}$$

That means that h' and k' are constant for points that lie on the same layer line of *II* kind. Where these layer lines intersect layer lines of *I* kind all these equations (14), (15) and (17) are satisfied and real reflections are possible at those points. The chances for the surfaces of three cones to intersect in one line are very slight, however, as long as the crystal is not rotating.

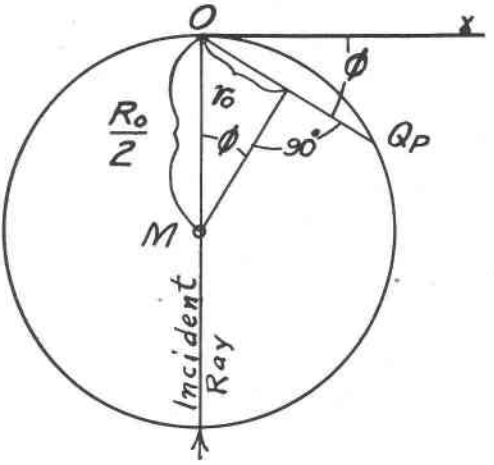

Figure 12.

In Fig. 5 we imagine the limiting sphere filled with the reciprocal lattice. The point of origin of the lattice is at *O*, the center of the sphere. It would be accidental if another point of the reciprocal lattice lay upon the surface of the "sphere of reflection," a condition necessary for reflection. However, if we turn the reciprocal lattice about *O* a definite number of degrees back and forth every point of the reciprocal lattice which passes through the surface of the sphere of reflection will be in position to reflect at that instant. Fig. 10 illustrates how the planes of the reciprocal lattice normal to the axis of rotation cut the sphere of reflection into parallel sections or slices each of which has a thickness of $1/T_{uvw}$ which is the reciprocal of the primitive translation in the direction [*uvw*] parallel to the axis of rotation. This is also shown in the upper part of Fig. 11 which represents a vertical elevation of the reciprocal lattice parallel to the photographic plate. The lower

part of Fig. 11 is the plan of the reciprocal lattice in the equatorial plane. The points in it may be projected upon the elevation as indicated. Schiebold calls this elevation the field of indices (Indicesfeld). Only a small part of the points in the plan need to be projected, however, depending upon the angle of rotation or oscillation respectively. For example, if the angle is 30° no point below the 30° line shown (Fig. 11) is projected, unless it lies within 3° to 4° of the line, for in that case there is a possibility that this point may cause reflection. But this is not the only limitation for points which might be projected. The concentric circles in Fig. 11 represent the surface of the sphere of reflection, each being a distance of $1/T_{uvw}$ above the next larger one. It is obvious that if the reciprocal lattice penetrates the sphere of reflection only through an angle of 30° by rotation about O, only the points in the plan which would go through one or more of the circles during such a rotation need to be projected. An easy way of determining these points is by the aid of a separate drawing of these concentric circles of reflection upon tracing paper. By pinning a point of the equator of the sphere to O in the reciprocal lattice we have a pivot or axis about which we can turn the sphere any desired number of degrees and watch which points of the reciprocal lattice penetrate the circles. In Fig. 11 this operation has been indicated by the dotted circles which represent the position of the sphere of reflection after turning through 30° from the starting position drawn in full lines. There is also a way of limiting the field of indices toward the top of the vertical elevation, Fig. 11. The small circles in Fig. 10 which pass through O, obviously are the lines of intersection of the sphere of reflection with vertical planes of the reciprocal lattice. No point of the lattice outside of the sphere can reflect. If we can draw these small circles on the elevation of Fig. 11, therefore, we eliminate many impossible indices from consideration. The radii of these "limiting circles" (Begrenzungskreise) are $r_0 = R_0/2 \cdot \sin \Phi_{hkl}$. $R_0/2$ is the radius of the sphere of reflection and the angle Φ_{hkl} is the angle which a pole P_{hkl} projected upon the equatorial plane as point Q_P makes with coordinate x, which is normal to the plane of the axis of rotation and the incident ray. The auxiliary construction, Fig. 12 shows how we arrive at this equation for the radii of the limiting circles.