CALCULATION OF POSITION AND INDICES OF LAUE SPOTS ON LAUE-PHOTOGRAPHS

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ABSTRACT

Formulae are derived for the calculation of the polar-coordinates and indices of the Laue spots on the film for the perfect crystal lattice. This calculated Laue pattern can be used as a reference pattern for any deviation of the experimental one. The formulae are given for the cases where the incident beam is perpendicular to (001), (010), (100) or (111). An example of the evaluation of a group of Laue spots of the Laue pattern of a muscovite mica will be given and discussed.

INTRODUCTION

It has been recognized for a long time that the application of the Laue method can serve as a powerful tool for the studies of stacking disorder in the growth of layer structures, polymorphism, intergrowth and order-disorder in alloys. Furthermore, the application to problems such as thermal or elastic agitation of crystals, thermal decomposition with simultaneous nucleation and oriented growth of new phases, will only be mentioned here. The advantages of the Laue method over the conventional rotating crystal methods for a study of such problems are its greater resolution power of the reflections, the non-destruction of the specimen crystal, and the much shorter exposure time of the photographic film. For a thorough study of these problems an evaluation of the Laue pattern in respect to position and indices of the spots is necessary.

In the Laue method the crystal and the photographic film are held stationary while the crystal is irradiated by a fine beam of *x*-rays, comprised of a wide range of wavelengths. Variables can be provided by the range of wavelengths or by the setting of the crystal in respect to the incident beam. Two techniques, the transmission- (for glancing angles $<45°$) and the back-reflection (for glancing angles$>45°$) technique can be distinguished. The Laue spots from perfect crystals are sharp but from imperfect or deformed crystals they are diffuse or elongated. Also, a change in position and intensity, the existence of extra spots, or a change in the symmetry of the pattern are indications of imperfections, and represent very important experimental data for the solution of special problems and fundamental research.

The graphical methods for determining the indices of Laue spots on Laue photographs involve no difficulties as long as the crystal is free from imperfections and belongs to one of the orthogonal systems. In other than these cases, a numerical control of at least some of the indices is a necessity. For that reason and since a completely calculated Laue pattern can be used as a reference pattern for the interpretation of any deviation of the experimental one, it was decided to establish practical useful formulae for the calculation of the position and indices of Laue spots. E. Schiebold[1] in a monograph on the Laue method, published 20 years ago, has derived a formula for the general case where any crystallographic direction of a triclinic crystal can be chosen as the direction of incidence for the *x*-ray beam. Therefore, his formula contains too many variables and is not practical. Besides this, many extra calculations and controls, with the help of the stereographic projection, have to be included. The purpose of this paper is to present the results obtained from calculations leading to simple formulae for the calculation of positions and indices of Laue spots if the incident beam is perpendicular to one of the principal planes (001), (100), (010), or perpdendicular to the (111) plane.

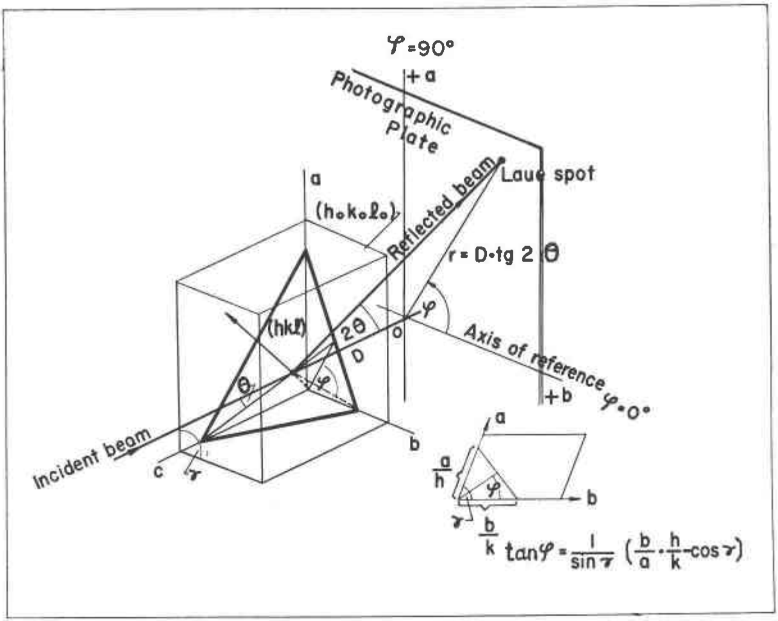

FIG. 1. Geometrical relationship between the position of the Laue spot and the reflecting plane for the case that the incident *x*-ray beam is perpendicular to a principal crystal plane. In the figure the (001)-plane is chosen as the plane of incidence.

In Fig. 1, the geometrical relationship between the position of the Laue spot and the reflecting crystal plane is shown for the case where the in-

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[1] Schiebold, E., Die Laue Methode, Leipzig (1932).

cident x-ray beam is perpendicular to one of the principal crystal planes and also perpendicular to the photographic film. For the determination of the position of the Laue spot, a polar-coordinate system in the plane of the photo-plate with its pole at the center of the Laue pattern is chosen. The azimuthal angle φ is measured from an axis, which corresponds to the direction of one of the crystal axes, parallel to the photographic plate. In this figure, the b-axis direction is chosen as the reference axis. A Laue spot on the photographic film is then determined by the radius vector r and the azimuthal angle φ. The following formulae were derived to correspond to Laue patterns as seen on the film in the direction of the incident beam. The distance r of a Laue spot from the center of the pattern is related to the corresponding Bragg angle θ by

$$r = D \cdot \tan 2\theta \tag{1}$$

where D is the distance between the crystal and the photographic plate. In the Laue method, using a white radiation, the angles φ and θ are independent of the wavelength and can be expressed in terms of the unit cell dimension, the indices of the plane of incidence ($h_0k_0l_0$), and reflecting plane (hkl) only.

DETERMINATION OF THE AZIMUTHAL ANGLE φ

If the incident beam is chosen perpendicular to the (001)-plane, the azimuthal angle φ of a Laue spot corresponds to the angle between the b-axis and the projection of the normal of the reflecting plane (hkl) on the (001) plane on the crystal, denoted by ($h_0k_0l_0$). From the figure it can be seen that the intersection of the (hkl)-plane and the ($h_0k_0l_0$)-plane cuts on the a- and b-axes the section a/h and b/k. The relation between the angle φ and these sections is given by

$$\tan \varphi = \frac{1}{\sin \gamma}\left(\frac{b}{a} \cdot \frac{h}{k} - \cos \gamma\right) \tag{2}$$

where γ is the crystallographic angle between the a-and b-axes. Similar expressions can be obtained if the plane of incidence chosen is the (010) or (100)-plane. From equation 2, it can be seen that Laue spots with the same h/k-ratio of their indices, for example, spots originating from the ($21l$), ($42l$), ($63l$). . . -planes lie on a straight line drawn from the center of the pattern.

DETERMINATION OF THE REFLECTION ANGLE θ

If the incident beam is perpendicular to a crystal plane ($h_0k_0l_0$), the reflection angle θ from a plane (hkl) can be derived from the angle between these planes, i.e. from the angle betweeen their normals, as shown

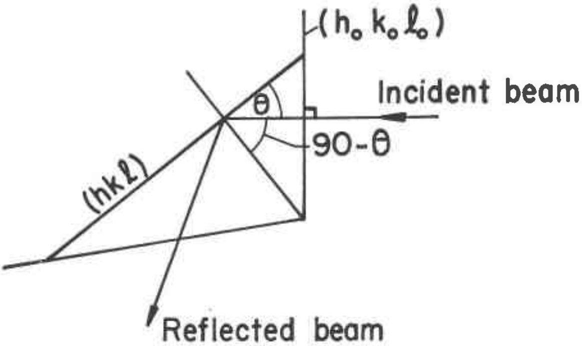

FIG. 2. Relationship between Bragg angle θ and the angle between the normals of reflecting plane and plane of incident. $(h_0k_0l_0)$ is the plane perpendicular to the incident beam, (hkl) is the reflecting plane, θ the Bragg angle, $(90-\theta)$ the angle between the normals of (hkl) and $(h_0k_0l_0)$.

in Fig. 2 Using the reciprocal lattice space the normals are vectors represented by

$$\begin{aligned} r_{hkl}^* &= a^*h + b^*k + c^*l \\ r_{h_0k_0l_0}^* &= a^*h_0 + b^*k_0 + c^*l_0. \end{aligned} \tag{3}$$

The lengths of these vectors are equal to the reciprocal of the spacings of the corresponding planes

$$|r_{hkl}^*| = \frac{1}{d_{hkl}}; \qquad |r_{h_0k_0l_0}^*| = \frac{1}{d_{h_0k_0l_0}}.$$

The angle between the vector r_{hkl}^* and $r^*_{h_0k_0l_0}$ can be found from the vector product

$$r_{hkl}^* \cdot r_{h_0k_0l_0}^* = |r_{hkl}^*| \cdot |r_{h_0k_0l_0}^*| \cdot \cos(90-\theta) = \frac{1}{d_{hkl} \cdot d_{h_0k_0l_0}} \cdot \cos(90-\theta)$$

$$\cos(90-\theta) = \sin\theta = d_{hkl} \cdot d_{h_0k_0l_0} \cdot (a^*h + b^*k + c^*l)(a^*h_0 + b^*k_0 + c^*l_0).$$

Multiplying and replacing the reciprocal unit cell dimension by the unit cell dimensions gives for a triclinic crystal:

$$\sin\theta = d_{hkl} \cdot d_{h_0k_0l_0} \cdot \frac{Q}{b^2 \cdot S} \sin\beta \tag{4}$$

where

$$Q = \frac{b^2 \sin^2\alpha}{a^2 \sin^2\beta} hh_0 + kk_0 + \frac{b^2 \sin^2\gamma}{c^2 \sin^2\beta} ll_0 + \frac{b^2 \sin\alpha \cdot \sin\gamma}{ac \sin^2\beta} \cos B(hl_0 + lh_0)$$
$$+ \frac{b \cdot \sin\alpha}{a \cdot \sin\gamma} \cos C(hk_0 + kh_0) + \frac{b \cdot \sin\gamma}{c \cdot \sin\beta} \cos A(kl_0 - lk_0)$$

with

$$\cos A = \frac{\cos\beta\cdot\cos\gamma - \cos\alpha}{\sin\beta\cdot\sin\gamma}$$

$$\cos B = \frac{\cos\alpha\cdot\cos\gamma - \cos\beta}{\sin\alpha\cdot\sin\gamma}$$

$$\cos C = \frac{\cos\alpha\cdot\cos\beta - \cos\gamma}{\sin\alpha\cdot\sin\beta}$$

$$S = 1 - \cos^2\alpha - \cos^2\beta - \cos^2\gamma + 2\cos\alpha\cdot\cos\beta\cdot\cos\gamma.$$

In order to obtain the distance of the Laue spots from the center of the pattern one has to use equation (1). In order to reduce the time in converting the sin θ-value into a tan 2 θ-value, it is useful to use a conversion graph such as shown in Fig. 3.

AZIMUTHAL ANGLE φ AND REFLECTION ANGLE θ IF INCIDENT BEAM $\perp$ TO (001), (010), (100) OR (111)

The equation for sin θ is very much simplified if the plane of incident is one of the principal planes of the crystal as shown in Table 1 where, for the different crystal systems and for different cases of incidence, the corresponding formulae for tan φ and sin θ are tabulated. For each of the three cases the reference axis, from which the angle φ has to be drawn, is included. The simplicity of this set of formulae is striking and the procedure in applying it for calculation or evaluation of Laue pattern is similar to that of evaluating powder or rotation diffraction patterns.

However, one important point might be mentioned here: Because of the absence of the wavelength term in these "Laue"-formulae, no distinction between higher orders of reflection can be made; they are superimposed in the same spot. However, after having determined the indices of the Laue spot and knowing the wavelength range of the continuous radiation of the ray source, the number of reflections of higher orders contributing to the intensity in that Laue spot can be determined.

It might also be mentioned that considerations about the symmetry of the Laue pattern to be expected for each case can be predicted from the formulae.

In Table 2 the formulae for tan ϕ and sin θ are given, if the incident beam is perpendicular to the (111)-plane. The table includes only the cubic, tetragonal, and the rhombohedral systems because the Laue photographs show symmetry in the pattern only for these; a threefold symmetry for the cubic and rhombohedral and a twofold symmetry for the tetragonal. The direction corresponding to the [011]-axis in the Laue pattern is chosen as the axis from which azimuthal angle ϕ of a Laue spot is measured.

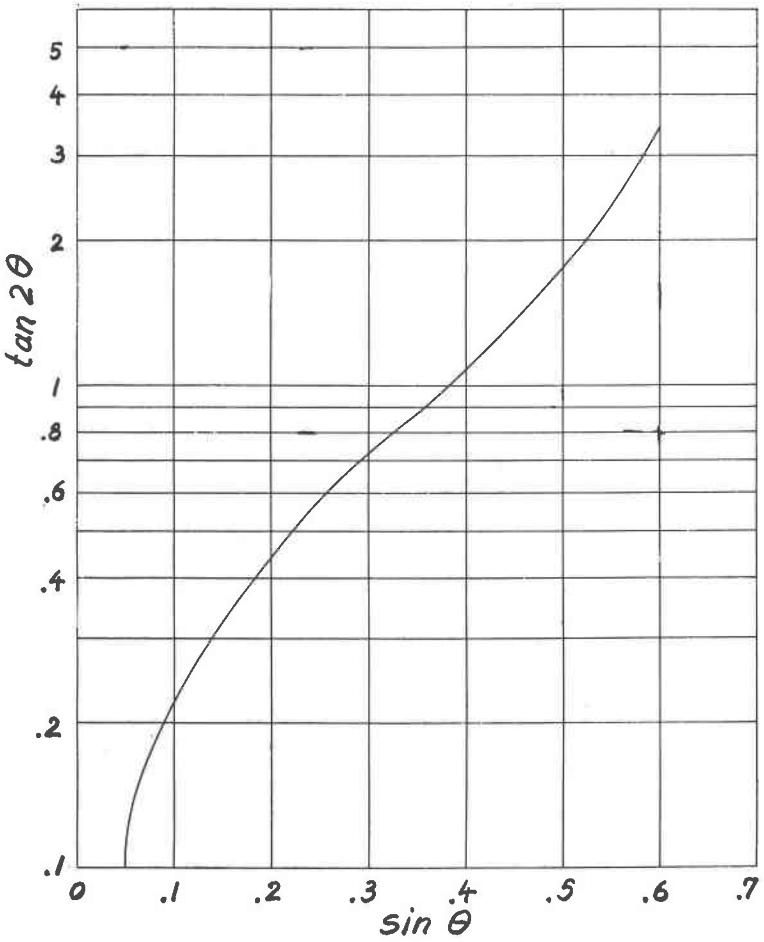

FIG. 3. Conversion of sin θ into tan 2θ.

PROCEDURE OF INDEXING LAUE PATTERN

The application of the formulae, given in Tables 1 and 2, will be demonstrated by indexing one group of Laue spots of a muscovite mica Laue pattern taken with the incident beam perpendicular to (001) and a crystal-photoplate distance $D = 58.3$ mm., Fig. 4. For the calculation, the Laue spots are chosen whose indices have a h/k-ratio equal to 3, i.e. spots with the indices $(31l)$, $(62l)$, etc. As mentioned before, these Laue spots will lie on a straight line drawn from the center of the pattern. With the unit cell dimension of muscovite $a = 5.18$ Å, $b = 9.02$ Å, $c = 20.04$ Å $\beta = 95° 30'$, the azimuthal angle φ of these spots using the correspond-

TABLE 1

Crystal lattice	Incident beam perpendicular to:		
	(001)	(100)	(010)
Cubic	$\sin\theta = d_{hkl}\cdot\frac{l}{a}$ $\tan\varphi = \frac{h}{k}$	$\sin\theta = d_{hkl}\cdot\frac{h}{a}$ $\tan\varphi = \frac{l}{k}$	$\sin\theta = d_{hkl}\cdot\frac{k}{a}$ $\tan\varphi = \frac{l}{h}$
Tetragonal	$\sin\theta = d_{hkl}\cdot\frac{l}{c}$ $\tan\varphi = \frac{h}{k}$	$\sin\theta = d_{hkl}\cdot\frac{h}{a}$ $\tan\varphi = \frac{a}{c}\cdot\frac{l}{k}$	$\sin\theta = d_{hkl}\cdot\frac{k}{a}$ $\tan\varphi = \frac{a}{c}\cdot\frac{l}{h}$
Orthorhombic	$\sin\theta = d_{hkl}\cdot\frac{l}{c}$ $\tan\varphi = \frac{b}{a}\cdot\frac{h}{k}$	$\sin\theta = d_{hkl}\cdot\frac{h}{a}$ $\tan\varphi = \frac{b}{c}\cdot\frac{l}{k}$	$\sin\theta = d_{hkl}\cdot\frac{k}{b}$ $\tan\varphi = \frac{a}{c}\cdot\frac{l}{h}$
Monoclinic	$\sin\theta = d_{hkl}\cdot\frac{1}{\sin\beta}\left(\frac{l}{c}-\frac{h}{a}\cos\beta\right)$ $\tan\varphi = \frac{b}{a}\cdot\frac{h}{k}$	$\sin\theta = d_{hkl}\cdot\frac{1}{\sin\beta}\left(\frac{h}{a}-\frac{l}{c}\cos\beta\right)$ $\tan\varphi = \frac{b}{c}\cdot\frac{l}{k}$	$\sin\theta = d_{hkl}\cdot\frac{k}{b}$ $\tan\varphi = \frac{1}{\sin\beta}\left(\frac{a}{c}\cdot\frac{l}{h}-\cos\beta\right)$
Hexagonal	$\sin\theta = d_{hkl}\cdot\frac{l}{c}$ $\tan\varphi = \frac{1}{3}\sqrt{3}\left(2\frac{h}{k}+l\right)$	$\sin\theta = d_{hkl}\cdot\frac{1}{a\sqrt{3}}(2h+k)$ $\tan\varphi = \frac{a}{c}\cdot\frac{l}{k}$	$\sin\theta = d_{hkl}\cdot\frac{1}{a\sqrt{3}}(2k+h)$ $\tan\varphi = \frac{a}{c}\cdot\frac{l}{h}$
	Angle φ measured from [010]	Angle φ measured from [010]	Angle φ measured from [100]

TABLE 2

Crystal lattice	Incident beam perpendicular to (111)
Cubic	$\sin\theta = d_{hkl}\cdot\frac{1}{3}\sqrt{3}\left(\frac{h+k+l}{a}\right)$ $\tan\varphi = \frac{1}{3}\sqrt{3}\left(2\,\frac{l-h}{l-k}-1\right)$
Tetragonal	$\sin\theta = d_{hkl}\cdot\frac{(h+k)+\frac{a^2}{c^2}\,l}{a\sqrt{\frac{a^2}{c^2}+2}}$ $\tan\varphi = \frac{1}{\sqrt{2\,\frac{c^2}{a^2}+1}}\left(\sqrt{2}\cdot\sqrt{\frac{c^2}{a^2}+1}\right)\cdot\left(\frac{l-h}{l-k}-1\right)$
Rhombohedral	$\sin\theta = d_{hkl}\cdot\frac{1}{3}\sqrt{3}\cdot\sqrt{1+2\cos\alpha}\cdot\frac{(1+\cos\alpha)^2}{(1-3\cos^2\alpha+2\cos^3\alpha)}\cdot\left(\frac{h+k+l}{a}\right)$ $\tan\varphi = \frac{1}{3}\sqrt{3}\left(2\,\frac{l-h}{l-k}-1\right)$

Angle φ measured from [011].

ing formula in Table 1, is calculated to be 79°09′. The b-axis direction in the Laue pattern from which the angle φ will be measured can be found easily from the symmetry of the pattern due to the fact that the symmetry axis is along the a-axis direction. At a right angle to it the b-axis direction is found. The radius vector drawn under an angle of 79°09′ from the b-axis direction, passes through five reflections whose indices now will be determined. The equation for the reflection angle θ takes the form (Table 1):

$$\sin\theta = d_{hkl}\cdot 1.005(0.0555k + 0.0499l)$$

where

$$d_{hkl} = \frac{1}{\sqrt{0.351k^2 + 0.00251l^2 + 0.0056kl}} \qquad \text{for } h = 3k$$

and therefore

$$\sin\theta = \frac{0.0558k + 0.0501l}{\sqrt{0.351k^2 + 0.00251l^2 + 0.0056kl}}\,. \tag{5}$$

The next step is to calculate some values for $\sin\theta$ as a function of l for $k = 1, 2, \cdots$. After a conversion of the $\sin\theta$-values into $\tan 2\theta$-values us-

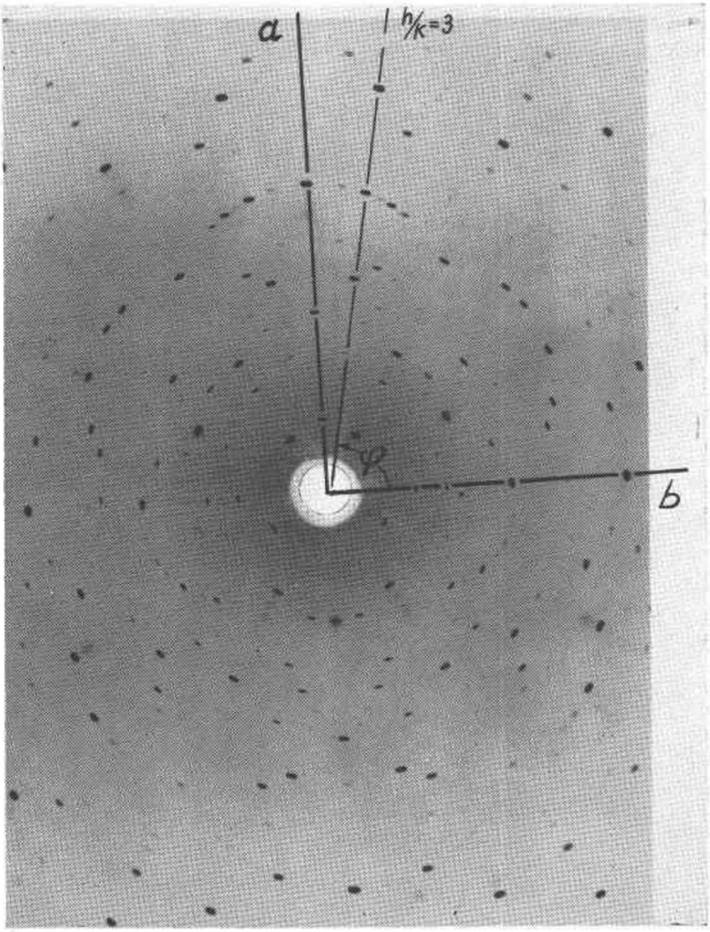

FIG. 4. Laue-photograph of muscovite mica with the incident beam perpendicular to (001). Distance crystal—photoplate $D=58.3$ mm. Radiation: Copper target.

ing the graph in Fig. 3, $r=D\cdot\tan 2\,\theta$ is plotted as a function of l, for reflections having the indices $(31l)$ and $(62l)$, as shown in Fig. 5. From this graph the indices of the five measured Laue spots, knowing their distances r from the center of the pattern, can be read off directly. Table 3 shows the results, including the possible number of higher orders of the reflections superimposed on the same Laue spot.

As a second example, the results from a complete evaluation of the Laue-pattern of a phlogopite mica are shown in Fig. 6. Here the value of a complete indexing of a Laue pattern can be recognized by the fact that extra spots (marked with a cross), which do not belong to the calculated

TABLE 3

Laue spots measured r [*mm*]	(*hkl*)	$n \cdot \lambda_{\mathrm{min.}}$	n
21.5	311	0.593	1
33.2	312	0.844	1, 2
47.0	313	1.06	1, 2, 3
63.2	314	1.25	1, 2, 3, 4
73.0	629	0.666	1, 2

Cu—Radiation, E = 40kv
$\lambda_{\mathrm{min.}} = 0.308$ Å

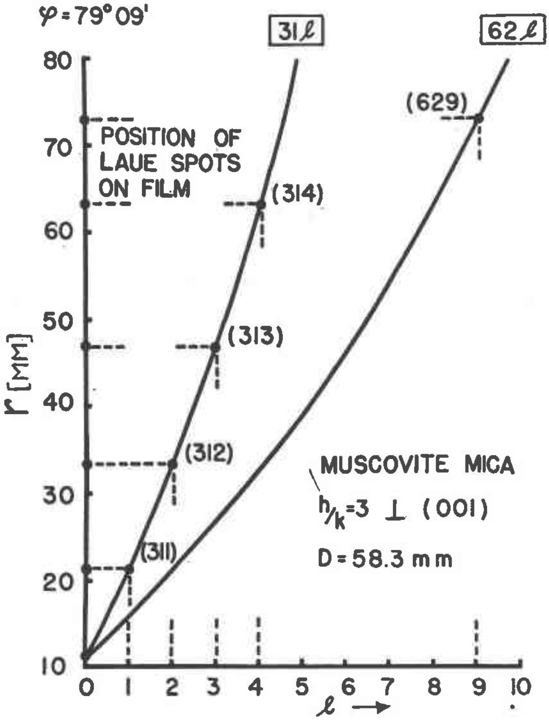

FIG. 5. Indexing of Laue spots ($h/k = 3$) on a Laue photograph from muscovite mica, Fig. 4, using the distance of the Laue spots from the center of the pattern and corresponding formula from Table 1.

pattern, could be detected. The shape of these extra spots as compared with the others are different in appearance and might be due to structure defects of this phlogopite sample, caused by stacking faults of layers.

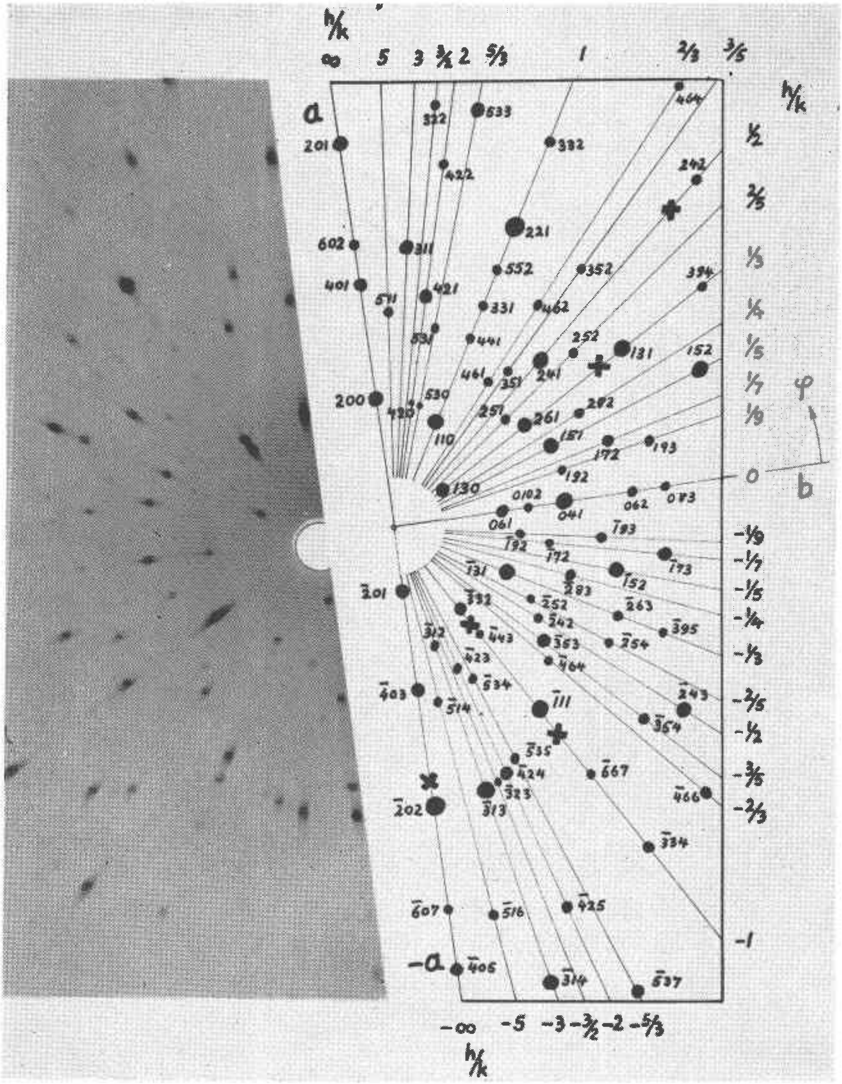

FIG. 6. Completely indexed Laue photograph of phlogopite mica with the incident x-ray beam perpendicular to (001) by the method described.

It might be worthwhile to mention that the time consumed for such a complete evaluation of a Laue pattern as shown in Fig. 6 is about 8 to 10 hours, comparable with the evaluation of crystal rotation photographs.

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