MEASUREMENT OF THE THREE PRINCIPAL INDICES OF REFRACTION IN MICACEOUS MINERALS BY IMMERSION ON A TILTING STAGE

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ABSTRACT

An accurate measurement of the refractive index for the vibration direction normal or nearly normal to the plane of cleavage in micaceous minerals is easily obtained by immersion measurements on a tilting stage. The simple theory of the method is developed and the application of the method to muscovite is described. Material from Mattawan township, Ontario, gave $\alpha=1.567$ (tilting method), 1.566_5 (total reflectometer, M. E. Jefferson), and by immersion on the horizontal stage, $\beta=1.606$, $\gamma=1.611$, in sodium light.

The micas and related platy minerals (brittle micas, chlorites, vermiculites) are pseudo-hexagonal in structure and form, and consequently pseudo-uniaxial in optical behaviour with the acute bisectrix nearly normal to the plane of eminent cleavage. The refractive indices for the two principal optical directions which are practically in the cleavage plane are conveniently measured by immersion on a horizontal stage; on the other hand the measurement of the refractive index for the third principal optical direction, which is nearly normal to the cleavage, is difficult, since it involves setting the plate on edge, and it is not likely to be very accurate, since the conditions are unfavourable for the Becke effect and the observation may have to be made very rapidly while the plate moves through the upright position.

In the course of a study of muscovite from a recently developed Canadian deposit one of us (R. B. F.) was faced with the problem of obtaining an accurate measurement of the index of refraction nearly normal to the cleavage. Noting that Jefferson (in Hendricks, 1939) had recently obtained measurements of this index on numerous micas using the total reflectometer, this method was tried, but the available instruments did not give good shadow-edges, even with the arrangements devised specially for muscovite by Viola (1900, p. 118). The problem was finally solved by making immersion measurements on a tilting stage, which gave us numerous consistent values for the desired principal index. Subsequently Dr. Jefferson was good enough to measure this index on our material using the total reflectometer. His result agrees with ours, showing that the tilting method gave the correct value. Since the method is also convenient and applicable to the wide range of platy minerals, the present account of the method and its application to muscovite may be of interest.

Theory of the Method

Figure 1 represents a crystal cleavage place C, with greatly exaggerated thickness, mounted in an immersion medium I with refractive index ρ, between glass hemispheres GG with refractive index μ. We assume the special optical orientation which is almost realized in micaceous minerals, namely one principal vibration direction, in this case $OX=\alpha$, normal to the cleavage plate. The other two principal vibration directions, $OY=\beta$,

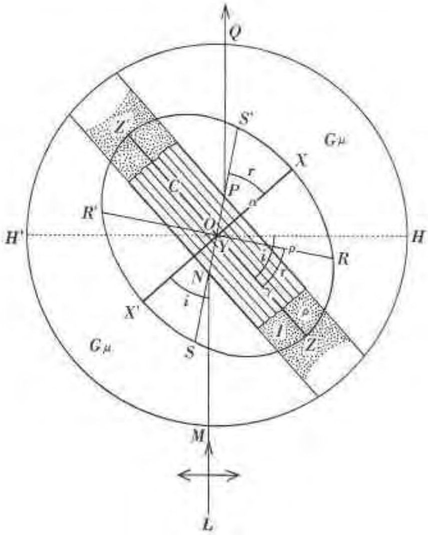

Fig. 1

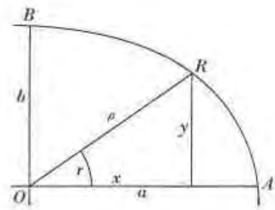

Fig. 2

and $OZ=\gamma$, are thus in the plane of the plate which has been arranged to tilt about OY which is normal to the plane of the drawing. The ellipse $XZX'Z'$ is a principal section of the indicatrix of the crystal. The value of γ is known from previous immersion measurement on the horizontal stage; α is to be found. The refractive index ρ of the liquid has been chosen less than γ and presumably greater than α. Vertically propagated polarized light enters at M and the system has been tilted from the horizontal HOH' through an angle i, in which position the apparent refractive index of the crystal plate equals ρ, the refractive index of the liquid.

In general, a radius of the indicatrix and the (rectangular) semi-axes of the normal central elliptic section give, respectively, the wave direction of light transmitted in a biaxial crystal and the two corresponding vibration directions and their refractive indices. In the present special case the direction of transmission is in the plane XOZ and we are concerned only with the vibrations in this plane. A beam of light L, polarized in the plane normal to HOH', passes into the lower hemisphere at M without refraction and enters the crystal plate at N with the angle of incidence i. Within the crystal plate the wave direction is NOP which is inclined to the plate normal at the angle of refraction r. At P the beam enters the upper hemisphere at the angle i, and leaves the system at Q with negligible refraction, if the crystal plate is thin compared to the radius of the glass sphere.

With reference to the section XOZ of the indicatrix, the wave direction is NOP or SOS', with the angle $XOS'=r$, and the vibration direction is ROR', with the angle $ZOR=r$. The apparent refractive index of the plate is ρ, and therefore OR, the refractive index associated with the wave-direction SOS', equals ρ. We may therefore write

$$\sin i : \sin r = \rho : \mu, \text{ or}$$
$$\sin r = (\mu \sin i)/\rho \qquad (1).$$

Thus r is obtained at once from the known values of μ, ρ, i, and we have the direction r and the length ρ of a radius of the ellipse whose major axis γ is known. Similar relations hold when OZ is taken as the tilting axis of the system.

Given one semi-axis of an ellipse, one radius, and the included angle, the other semi-axis is readily obtained from the equation of the ellipse. In Fig. 2 OA, OB, are the semi-axes of the ellipse with lengths a, b; R is the point x, y. We have

$$x^2/a^2 + y^2/b^2 = 1, \text{ or}$$
$$b^2 = y^2/(1-x^2/a^2).$$

Corresponding to Fig. 1, $a=\gamma$ (the known semi-axis), $b=\alpha$ (the unknown semi-axis), $OR=\rho$ (the determined radius), $x=\rho \cos r$, $y=\rho \sin r$. Substituting these values in the equation of the ellipse we obtain

$$\alpha^2 = (\gamma^2\rho^2 \sin^2 r)/(\gamma^2-\rho^2 \cos^2 r) \qquad (2),$$

or with OZ as the tilting axis

$$\alpha^2 = (\beta^2\rho^2 \sin^2 r)/(\beta^2-\rho^2 \cos^2 r) \qquad (2').$$

Thus each measurement of ρ and i (giving r), and the known γ or β, gives an independent value for the unknown α, and therefore a sufficient number of observations should yield a reliable average for the unknown refractive index.

The value of α having been determined in this way, it is of interest to compare the measured indices ρ with the calculated indices given by the experimental angle r together with α and γ or β. For this purpose equations (2) and (2′) may be rearranged to give

$$\rho^2 = \alpha^2\gamma^2/(\alpha^2 \cos^2 r + \gamma^2 \sin^2 r) \quad (3),$$
$$\rho^2 = \alpha^2\beta^2/(\alpha^2 \cos^2 r + \beta^2 \sin^2 r) \quad (3').$$

Similarly, to compare the experimental angles r with the angles calculated from the indices ρ together with α and γ or β, we have

$$\cos^2 r = \gamma^2(\rho^2 - \alpha^2)/\rho^2(\gamma^2 - \alpha^2) \quad (4),$$
$$\cos^2 r = \beta^2(\rho^2 - \alpha^2)/\rho^2(\beta^2 - \alpha^2) \quad (4').$$

Application of the Method to Muscovite

The tilting immersion method was applied to muscovite from Mattawan township, Ontario, which has been studied in some detail by one of us (R. B. F.) with a view to a fuller description to be given elsewhere. This material gives perfectly plane colourless cleavage sheets in which the angle between X (acute negative bisectrix) and the cleavage normal was found to be less than 1°, by exact measurement with a special device. The special optical orientation assumed in the theoretical section is thus practically attained. Careful measurements on the horizontal stage, with sodium light and a new set of immersion liquids, gave $\beta = 1.606$, $\gamma = 1.611$.

It was found that the best Becke effects were obtained on torn edges of thin sheets rather than on the straight ribbon edges given by separation along $(h0l)$ glide planes. Such torn sheets were mounted between glass hemispheres with refractive index $\mu = 1.554$ in liquids with refractive indices ρ ranging from 1.605 to 1.580 in steps of 0.005. The mounts were adjusted on the universal stage to bring first OY then OZ into the principal left-right axis of the stage. All the observations were made with the sodium vapour lamp and the polarizing nicol set to give light vibrating in the plane normal to the tilting axis. The angles of tilt, i, were read to the nearest half degree, the recorded values being averages of several readings taken with the plate tilted to the front and to the back. This procedure eliminated the small error introduced by the small inclination of X to the cleavage normal.

A plate mounted in this way in an oil of index ρ (between α and γ) shows in the horizontal position a strong Becke effect, mineral > liquid. On tilting the stage the Becke effect becomes weaker and at some angle of tilt the edge under observation vanishes. On further tilting of about 4° the edge reappears, giving the effect, mineral < liquid with increasing strength.

To determine more closely the angle of tilt at which the apparent index of the plate is equal to the index of the liquid, the stage was tilted to approximately the mid position of the invisible range and the angle of the revolving stage of the microscope was noted. This stage was then turned until the edge reappeared, in one direction and then in the other, and the readings were noted in each case. If necessary the tilt was changed until equal angles of rotation of the microscope stage, to the right and to the left, gave reappearances of the edge. When this condition was reached it was assumed that the correct angle of tilt had been found. In this way a series of measurements of i and ρ were made with OY and with OZ as the tilting axes.

Table 1 gives the refractive indices of the liquids used (ρ) and the observed angles of tilt (i), together with the angles of refraction (r) obtained by equation (1) and the values of the unknown index of refraction (α) given by equations (2) and (2′). The average value of α is 1.567. The extreme variation is $+0.005$ to -0.003, the largest variation occurring at the smallest tilt when, as might be expected, the measurement is least accurate. Between 30° and 50° of tilt the variation is ± 0.001 which is less than the limit of accuracy usually claimed for the immersion method.

To verify the value of α obtained by the tilting method we sent specimens of the mica to Dr. Merrill E. Jefferson who kindly tested a series of cleavage plates, including seven from the actual piece which we had used, on the total reflectometer. In his reply Dr. Jefferson stated that the best value he obtained for α was 1.566_5 which is practically identical with our value 1.567.

These independent observations show that the tilting immersion method gives results quite as accurate as those that can be obtained on a horizontal stage and that a single good observation at a tilting angle of about 45° will give a good value for the unknown index of refraction. Furthermore it will be noted in the table that i and r do not differ greatly even though ρ and μ differ considerably. If therefore the refractive indices of the liquid and the glass are nearly alike no considerable error will be introduced by assuming that r equals i.

Comparison of the Calculated and Experimental Values of ρ and r

Given the three refractive indices, α, β, γ, the value of ρ for a given value of r may be calculated from equations (3) and (3′), or the value of r for a given value of ρ may be obtained from equations (4) and (4′). The results of such calculations are given in Tables 2 and 3. From these we see that the greatest difference between the calculated and experi-

TABLE 1. REFRACTIVE INDEX α COMPUTED FROM IMMERSION MEASUREMENTS ON TILTED PLATES OF MUSCOVITE
($\beta=1.606$, $\gamma=1.611$, $\mu=1.554$)

			Tilting axis *OY*				
ρ	1.605	1.600	1.595	1.590	1.585	1.580	
i	$23\frac{1}{2}°$	31°	37°	$44\frac{1}{2}°$	50°	55°	
r	22°43′	30°01′	35°54′	43°14′	48°41′	53°41′	
α	1.572	1.568	1.566	1.567	1.566	1.564	Av. 1.567
			Tilting axis *OZ*				
ρ		1.600	1.595	1.590	1.585	1.580	
i		26°	32°	40°	46°	53°	
r		25°12′	31°05′	38°55′	44°51′	51°46′	
α		1.571	1.566	1.567	1.566	1.564	Av. 1.567

mental values of ρ is 0.002 while the mean difference is less than 0.001. The greatest difference between the calculated and experimental values of r, which is nearly equal to the tilting angle i, is nearly 3° while the mean difference is 1.3°. The same facts are expressed in Figs. 3 and 4 which give the graphs of equations (3) and (3′), respectively, and points corresponding to the experimental values of ρ and r.

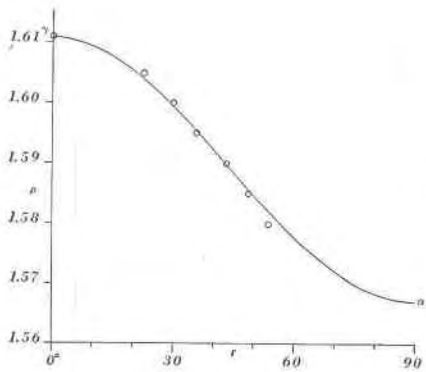

FIG. 3

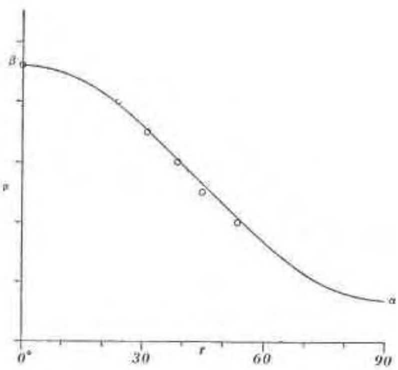

FIG. 4

SPECIALIZATION AND GENERALIZATION OF THE TILTING METHOD

The theory given for the tilting immersion method applies directly and strictly to orthorhombic crystals with cleavage parallel to any pinakoid and to monoclinic crystals with cleavage parallel to the symmetry plane. In the case of cubic crystals and uniaxial crystals with prismatic cleavage, n, ω, ϵ, are measured on the horizontal stage. In

TABLE 2. CALCULATED AND MEASURED INDICES ρ AT THE EXPERIMENTAL ANGLES r
($\alpha=1.567$, $\beta=1.606$, $\gamma=1.611$)

	Tilting axis OY						
r exper.	22°43′	30°01′	35°54′	43°14′	48°41′	53°41′	
ρ calc.	1.6042	1.5997	1.5955	1.5899	1.5857	1.5820	
ρ exper.	1.605	1.600	1.595	1.590	1.585	1.580	
$\Delta\rho$	+0.0008	+0.0003	−0.0005	+0.0001	−0.0007	−0.0020	Av. 0.0007
	Tilting axis OZ						
r exper.	25°12′	31°05′	38°55′	44°51′	51°46′		
ρ calc.	1.5987	1.5953	1.5903	1.5862	1.5816		
ρ exper.	1.600	1.595	1.590	1.585	1.580		
$\Delta\rho$	+0.0013	−0.0003	−0.0003	−0.0012	−0.0016	Av. 0.0009	

TABLE 3. CALCULATED AND EXPERIMENTAL ANGLES r AT THE MEASURED INDICES ρ
($\alpha=1.567$, $\beta=1.606$, $\gamma=1.611$)

	Tilting axis OY						
ρ	1.605	1.600	1.595	1.590	1.585	1.580	
r calc.	21°12′	29°31′	36°33′	43°07′	49°41′	56°33′	
r exper.	22°43′	30°01′	35°54′	43°14′	48°41′	53°41′	
Δr	+1°31′	+0°30′	−0°39′	+0°07′	−1°00′	−2°52′	Av. 1.1°
	Tilting axis OZ						
ρ	1.600	1.595	1.590	1.585	1.580		
r calc.	22°52′	31°43′	39°23′	46°42′	54°17′		
r exper.	25°12′	31°05′	38°55′	44°51′	51°46′		
Δr	+2°20′	−0°38′	−0°28′	−1°51′	−2°31′	Av. 1.5°	

uniaxial crystals with basal cleavage ω is obtained on the horizontal stage and ϵ by tilting about any direction in the cleavage plane. In the case of monoclinic crystals with cleavage in the zone [010] the index of refraction for the vibration direction [010] is measured on the horizontal stage. If the plate is then set to tilt about [010] the orientation of the indicatrix is at once obtainable and two tilting immersion measurements will suffice to determine the refractive indices for the two vibration directions in the symmetry plane.

The general case is presented by monoclinic crystals with prismatic cleavage and triclinic crystals with any cleavage. In such cases it will be seen that, by tilting about any principal axis of the indicatrix, three tilting immersion measurements will be needed to determine the lengths and orientation of the other two axes of the indicatrix. In practice the tilting immersion method is most likely to prove useful in the case of crystals with micaceous cleavage which is generally associated with a layer structure and pronounced uniaxial optical pseudo-symmetry.

References

Hendricks, S. B., Polymorphism of the micas. With optical measurements by M. E. Jefferson: *Am. Mineral.*, **24,** 729–771 (1939).

Viola, C., Optische Studien über italienische Mineralien: *Zeits. Kryst.*, **32,** 113–124 (1900).