## An analysis of the movements of shadow-edges on the refractometer in the case of biaxial gemstones.

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THE study and identification of the gem minerals present difficulties which are not to be resolved by the ordinary methods of mineralogy. The commercial value of the material forbids recourse to chemical reagents and the blowpipe, and when faceted and mounted, the criteria of crystal form and specific gravity are also inapplicable. Optical phenomena then constitute the sole means of discrimination, and the refractometer, spectroscope, polariscope, and dichroscope become the only arbiters between gemstones of similar appearance but different species. The function of the polariscope is to narrow the field of inquiry by differentiating between isotropic and anisotropic gemstones; the verdict of the spectroscope is unequivocal in gemstones characterized by absorption spectra, which, unfortunately, form a minority; dichroism, when present, can be regarded as supplementary evidence only; the refractometer, an instrument of wider application than the spectroscope, and capable of yielding information which is usually equally unambiguous, thus assumes a role of primary importance. The method of minimum deviation, while furnishing refractive index readings which are far more critical, is attended by difficulties which outweigh this advantage, since gem material in the rough is usually too valuable to be ground into 60° prisms, and after it has passed through the lapidary's hands is seldom found to have a pair of facets which are sufficiently plane and suitably inclined to be employed for this purpose. Moreover, the accuracy of spectrometric refractive index determinations is largely nullified when, as in anisotropic faceted gemstones, optical orientation is difficult or impossible to establish, whereas with the refractometer, maximum and minimum readings are obtainable from any facet, provided these values lie within the range of the instrument. For ordinary identification purposes it is sufficient when using the refractometer to combine a reasonable degree of manipulative skill with a knowledge of this fact, ignoring the apparently fortuitous behaviour of the shadow-edges in their movements between the two limiting values. However, a theoretical explanation of the shadow-edge movements is essential when anything beyond an everyday routine use of the instrument is proposed, and it is intended in the present paper to describe techniques employing the methods of analytical geometry by which these movements can be investigated. Some points of practical importance which are relevant to this analysis will also be discussed.

The theory of the refractometer postulates a hemisphere or prism which is optically denser than the stone whose refractive indices it is desired to read. In

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the earlier forms of the instrument,<sup>1</sup> the dense-glass hemisphere had a refractive index lower than those of several important gemstones, which were in consequence beyond its range, but in later models<sup>2</sup> a greater range has been provided by the employment of types of glass having very high refractive indices, and, still more recently,<sup>3</sup> isotropic crystalline materials have been used still further to extend this range. Another practical difficulty of a similar kind arises from the necessity of effecting optical contact between the stone and the hemisphere by means of a liquid of high refractive index, but here again recent years have seen the introduction of new liquids for this purpose, which have effectively widened the range of the instrument.<sup>3</sup>

For the identification of precious stones, the primary function of refractometers of this type, no very exacting standards of accuracy are ordinarily required, and a tolerance of  $\pm 0.005$  is usually permissible. For an experienced observer, using a specially calibrated instrument, the limits consistently possible are approximately  $\pm 0.001$ . Data of this kind would be hopelessly inadequate for the purpose of the present analysis, and their development from empirically determined results is impossible. It has therefore been developed theoretically from an initial consideration of the fundamental principle common to all types of the instrument now available. Reference to any extended applications of the refractometer suggested by an analysis of this kind would need a full discussion of the possibilities and limitations of the instrument, a discussion which is beyond the scope of the present paper, and the only incursion into this field which has been included is that of the determination of  $\beta$  in biaxial gemstones. It is probable that, given an instrument of a precision character, such as the Abbe-Zeiss, the absence of observational crudities would justify further research, and that it might be possible, for instance, to develop a technique for the ascertainment of the optical orientation of any facet of a gemstone from a set of refractive index readings derived from it. Further to pursue these interesting possibilities also lies beyond the scope of the present inquiry.

A refractive index reading, or pair of readings, obtained from a facet of a gemstone on the refractometer, corresponds to a direction at right angles to the normal to that facet. In the Tully refractometer, the hemisphere can be rotated to give values corresponding to all directions at right angles to this normal, and in other models with a fixed stage, the stone itself can be rotated with the same result. In terms of the optical indicatrix, the refractive indices for any direction through a crystalline medium are graphically represented by the lengths of the semi-axes of the ellipse formed by the intersection of the indicatrix and that plane through its centre which has this direction as normal. Thus the values given in a rotation through two right angles of a gemstone are represented by the centre of the indicatrix and sharing a common line of intersection, which is the normal to the facet being employed.

Upon this basis it is possible, given the values of the principal axes of the indicatrix, and the optical orientation of the normal to the facet, to calculate all the refractive indices which will be obtained during the complete rotation of the

<sup>&</sup>lt;sup>1</sup> G. F. Herbert Smith, Min. Mag., 1905, vol. 14, pp. 83-86; 1907, vol. 14, pp. 354-359.

<sup>&</sup>lt;sup>2</sup> B. J. Tully, ibid., 1927, vol. 21, pp. 324-328.

<sup>&</sup>lt;sup>8</sup> B. W. Anderson, C. J. Payne, and J. Pike, ibid., 1940, vol. 25, pp. 579-583.

stone on the refractometer. For this purpose it is useful to consider the question as a problem in analytical geometry, using the conventions and terminology of that subject, rather than those of mineralogy.<sup>1</sup>

The semi-axes of the ellipses formed by the intersection of any plane with an ellipsoid are given by the following expression:

$$\frac{a^{2l^2}}{a^2 - r^2} + \frac{b^2 m^2}{b^2 - r^2} + \frac{c^2 n^2}{c^2 - r^2} = 0. \qquad \dots \qquad \dots \qquad \dots \qquad (i)$$

This equation is a biquadratic in r; l, m, and n, and a, b, and c have their usual significances as direction-cosines of the plane, and the principal semi-axes of the ellipsoid, respectively. Applying to it the familiar rule for the solution of quadratic equations, two roots in  $r^2$  are given, the square roots of which are the required values of the semi-axes. (An alternative and less clumsy method of solution is obtainable by manipulating the expressions for the sum and product of the roots.) To obtain values of the direction-cosines for successive positions during the rotation in terms of those of the normal to the facet, the latter is written in the form of the intersection of two planes L and  $L_1$ . Then  $L + \lambda L_1$  is the equation of a plane having a common line of intersection with these two planes, and by giving lambda all possible values, the equations of all planes intersecting in the normal to the facet are obtained. Writing down the equations of a sufficient number of these planes, the angles between which are given by the relationship  $ll_1 + mm_1 + nn_1 = \cos \theta$ , and using equation (i) to calculate the corresponding refractive index values, the necessary data are obtained for the construction of curves graphically representing the movements of the shadow-edges seen on the refractometer scale during a complete rotation.

A specific instance may be given in illustration. A set of principal values for a topaz from Minas Geraes has been recorded as  $\alpha 1.6294$ ,  $\beta 1.6308$ , and  $\gamma 1.6375$ , and may be regarded as sufficiently typical for this mineral. A genstone is cut from this material, and refractive index readings are made on the refractometer, using for this purpose the table facet, which happens to be equally inclined to each of the principal optical directions. It is required to construct a pair of curves showing the variation in these readings during a rotation with or on the refractometer stage. The equation of the normal to the facet may be written as x = y = z, or, choosing one of the infinitely many ways of writing this straight line in the form of the intersection of two planes, as

$$3x-2y-z=0 \\ x+y-2z=0$$

The equations of the planes which intersect in this straight line are of the form

$$3x-2y-z+\lambda(x+y-2z) = 0.$$
 ... ... (ii)

<sup>1</sup> When algebraic methods are used to investigate the behaviour of anisotropic gemstones on the refractometer, the retention of the symbols a, b, and c of the ellipsoid is justified on the ground of consistency, since those for the direction-cosines are lower-case members of the same alphabet. When the results are used to interpret biaxial phenomena, a, b, and c are equated to  $\alpha$ ,  $\beta$ , and  $\gamma$  respectively. Mathematically, an analysis of the properties of uniaxal minerals follows similar lines, the appropriate modification being introduced that two of the principal semi-axes are equal in length, and the final results are restated in terms of the omega and epsilon of mineralogy. From equation (i) the following results are obtained:

$$egin{aligned} r_1^2 + r_{11}^2 &= rac{a^{2l^2(b^2+c^2)} + b^2m^2(a^2+c^2) + c^2n^2(a^2+b^2)}{a^{2l^2} + b^2m^2 + c^2n^2} \ r_1^2 r_{11}^2 &= rac{a^{2b^2c^2}}{a^{2l^2} + b^2m^2 + c^2n^2}. \end{aligned}$$

Putting  $\lambda = 0$  in equation (ii), which then becomes 3x-2y-z = 0, it is possible, on inserting the values of a, b, c, l, m, and n in the equations written above, to calculate  $r_1$  and  $r_{11}$ , which prove to be 1.6304 and 1.6370. Similarly for  $\lambda = 1$ , equation (ii) becomes 4x-y-3z = 0, and the refractive indices are 1.63075 and 1.6347. The angle between this plane and that for  $\lambda = 0$  is 27°. Repeating this process, taking a number of values of  $\lambda$ , selected at random, and choosing the plane for  $\lambda = 0$  as giving the initial point from which degrees of rotation are measured, these results are obtained:

	А	igre			
from					
λ.	$\lambda = 0.$		Equation.	$r_1$ .	r <sub>11</sub> .
0	0	<b>)</b>	3x-2y-z=0	1.6304	1.6370
1	27		4x - y - 3z = 0	1.63075	1.6347
2	40	54'	x-z=0	1.6308	1.6334
6	58	17	9x + 4y - 13z = 0	1.6305	1.63255
<u> 20</u> -	70	54	x+y-2z=0	1.6300	1.6327
-1	-19	6	x-y=0	1.6301	1.6375
-i	-38	14	2x - 3y + z = 0	1.6300	1.6368
2	-65	14	x - 4y + 3z = 0	1.6295	1.6352
3	-79	8	y-z=0	1.6294	1.63405

From these figures the curves shown in fig. 1 can then be plotted.



It may be noted that the ordinates of this graph are evenly spaced, whereas the divisions of the scale of the refractometer become increasingly widely spaced with increase in refractive index. It is easily seen that this compression of the refractometer scale at the 'low' end is imposed by the fundamental principle of the instrument; it can be verified that the spacing is a function of the cotangent of the critical angle. Fortunately, there are few gemstones having refractive indices less than 1.5, so that in practice this tends to be an advantage rather than a demerit, permitting greater accuracy in the region where it is of most use. In the graphs of shadow-edge curves accompanying this paper, it would be pointless to mirror this practical exigency, which is therefore ignored. It is scarcely necessary to add that fourth-figure accuracy, which is easily attainable in the calculated values employed for the construction of these curves, is unfortunately impossible of achievement in practice, except with the more elaborate and costly total reflectometers of the Abbe or Pulfrich type.

There are four turning-points in a typical pair of biaxial shadow-edge curves, three of which are coincident with the principal values for the mineral under examination. These are the a, b, and c of the ellipsoid, or, in mineralogical terms,  $\alpha$ ,  $\beta$ , and  $\gamma$ . The analysis makes it evident that any facet of a biaxial gemstone must give readings of  $\alpha$ ,  $\beta$ , and  $\gamma$  during a rotation through two right angles.<sup>1</sup> It has been seen that the equation of the normal to the facet may be written in the form of the joint equation of two planes L and  $L_1$ , and it will be evident that, whatever the coefficients of x, y, and z in these two planes, it is always possible to find a plane passing through their intersection in whose equation the x-term is missing. This is effected by assigning a suitable value to  $\lambda$  in  $L + \lambda L_1$ , the equation representing the pencil of planes intersecting in the normal to the facet. For example, when  $\lambda = -3$ , equation (ii) reduces to y-z=0. All such planes whose equations lack an x-term intersect in the x-axis, and hence in their intersection with the ellipsoid form ellipses all of which share a common axis, of length  $2a^2$ . Thus at one point during the rotation of any biaxial gemstone on the refractometer, a reading of  $\alpha$  must be given, and, by a similar reasoning, readings of  $\beta$  and  $\gamma$  are also furnished.

Being maxima and minima, the turning-points at  $\gamma$  and  $\alpha$  are immediately recognizable, but to distinguish between  $\beta$  and the fourth turning-point is less simple. As has been pointed out by Dr. Herbert Smith ('Gemstones.' London, 1940, p. 54 et seq.), a comparison of results obtained from two non-parallel facets will enable this distinction to be made,  $\beta$  being that median turningpoint which will be common to both. If this is impracticable, as in the case of most mounted gemstones, it is possible to calculate  $\beta$  from data provided by readings from one facet only, and a formula for this purpose may be given. When the stone on the stage of the refractometer has been rotated into the position giving the maximum reading  $\gamma$ , the other shadow-edge provides a second reading, and similarly there is an associated value seen when one edge is at the  $\alpha$  position. Calling these associated readings R and r, and the angle between the points at which they occur  $\phi$ , an approximation to the value of  $\beta$  is given by the following expression:

$$\cos^2 \phi = \frac{\alpha^2 \gamma^2}{r^2 R^2} \cdot \frac{(\beta^2 - r^2)(\beta^2 - R^2)}{(\beta^2 - \alpha^2)(\beta^2 - \gamma^2)} \dots \dots \dots \dots \dots (iii)$$

It will be seen that this method does not require considerations of the values of

<sup>1</sup> L. J. Spencer, Gemmologist, London, 1937, vol. 6, pp. 231-236.

<sup>2</sup> This can be checked by rewriting equation (i) as a straightforward biquadratic, equating l to zero, and showing that the resultant equation has a root  $r^2 = a^2$  by the factor theorem.

the median turning-points to be taken into account, but it is clear that a check is afforded by comparison with the appropriate value.

Another criterion by which  $\beta$  can be separated from the second median point in cases when the Herbert Smith test is inapplicable is furnished by observation of the behaviour of the shadow-edges when a polarizing filter is employed. Such filters are commercially available for one type of instrument, in the form of a cap for the eyepiece, but an accessory of this kind can be improvised without difficulty for use with any refractometer. When, in a doubly-refractive stone, an insufficient separation of the shadow-edges makes critical readings difficult, it is useful to make separate observations of each edge by extinguishing the other by means of a polarizing cap. This is rotated into a position which is visually judged to effect optimum extinction of the unwanted shadow-edge, a position which is variable for a biaxial stone and constant for a uniaxial-an effect which is analogous to the behaviour of minerals in thin section between the crossed nicols of a petrological microscope. In terms of the indicatrix, it will be apparent that, for successive positions during the rotation of a biaxial stone, the corresponding ellipses of section will in general have their axes occupying skew positions with regard to the principal optical directions; the skew directions will be coincident with the positions of maximum extinction. Straight extinction with respect to the principal optical directions is shown at positions in which a principal value  $(\alpha, \beta, \text{ or } \gamma)$  is given. For a uniaxial stone, the indicatrix becomes a spheroid, and the directions of maximum extinction are invariant.

Hence a way of discriminating between  $\beta$  and the other median turning-point is afforded by the following means: the stone is placed in a position to give a reading of  $\alpha$ , and the polarizing cap is adjusted to extinguish the shadow-edge giving this reading. The stone is then rotated into the position giving the lower median turning-point reading, and the angle through which the polarizing cap has to be rotated to extinguish this reading is noted. A similar procedure is followed to find the angle of rotation of the polarizing cap between the  $\gamma$  position and that affording the upper median turning-point. The angle of rotation of the cap for the median turning-point which is not  $\beta$  will differ from that which is, in being sensibly different from zero.

This second method of obtaining  $\beta$  from readings given by one facet of a biaxial gemstone is simpler in practice and theoretically preferable to the first, since the degree of error in a calculated result based upon a set of five observations which cannot be critically accurate is likely to be high. An example may be given in illustration. A step-cut peridot from the Congo gave these readings:  $\alpha$  1.643, associated value 1.665;  $\gamma 1.681$ , associated value 1.650; angular distance between the positions giving these readings, 15°. The calculated value of  $\beta$  given by formula (iii) is 1.657—probably somewhat too low. The other method, which avoids the tedium of arithmetical calculations, has proved to be fairly satisfactory in practice. Observations from a random set of eight gemstones yielded the following results: in five cases (two peridots, two topazes, and a hambergite), the test was applicable, and its correctness was confirmed; three other stones gave indeterminate results (a peridot required no appreciable shift in position of the polarizing cap for extinction at either median turning-point, and in a peridot and a diopside the extinction positions were not determinable with sufficient accuracy).

The degree of rotation of the polarizing cap for the extinction of a shadowedge is a function of the orientation of the normal-to-facet position with respect to the principal optical directions of the stone. If the direction-cosines of the axes of the ellipse formed by the intersection of the plane lx+my+nz = 0 and the ellipsoid  $\frac{x^2}{a^2} + \frac{y^2}{b^2} + \frac{z^2}{c^2} = 1$  are  $\lambda_1$ ,  $\mu_1$ , and  $\nu_1$ , and  $\lambda_{11}$ ,  $\mu_{11}$ , and  $\nu_{11}$ , then their ratios are expressible as follows:

$$\lambda_1 \left( \frac{a^2 - r^2}{a^2 l} \right) = \mu_1 \left( \frac{b^2 - r^2}{b^2 m} \right) = \nu_1 \left( \frac{c^2 - r^2}{c^2 n} \right), \quad \dots \quad (iv)$$

and similarly for  $\lambda_{11}$ ,  $\mu_{11}$ , and  $\nu_{11}$ . After the determination of their directioncosines, the angles which these axes make with the principal axes are calculable, and hence also the degree of rotation of the polarizing cap necessary for extinction when one of these directions is selected as the initial position.

If the selected facet of a biaxial gemstone has its normal coincident with a principal optical direction, the refractive index curves assume special forms in each of which one of the shadow-edges traces out a straight line. In practice these 'special' cases occur with an unexpected frequency, and therefore merit separate consideration.

(i) When the normal to the facet is coincident with X, one shadow-edge moves between  $\beta$  and  $\gamma$ , and the other remains constant at  $\alpha$ .

(ii) Normal to facet coincident with Y, one shadow-edge remains constant at  $\beta$ , the other moves between  $\alpha$  and  $\gamma$ , and thus coalesces with the stationary shadow-edge at two points.

(iii) Normal to facet coincident with Z, one shadow-edge remains constant at  $\gamma$ , and the other moves between  $\alpha$  and  $\beta$ .

In fig. 2 the shadow-edge curves for these 'special' orientations are sketched. A partial explanation of the relative frequency with which these 'special' cases are encountered is to be found in the lapidary's choice of a principal optical direction as the symmetry axis of the stone, to take advantage of the habit of the crystal, to display colour to best advantage, or to utilize a cleavage trend. For this reason, an additional case in which coalescence of the shadow-edges at  $\beta$  takes place is much more infrequent; it occurs when, in a gemstone whose cutting is not specially related to its principal optical directions, the median turningpoints are coincident. The degree of probability is low that, for a line chosen at random through the centre of an ellipsoid, the pencil of planes intersecting in this line should include one cutting the ellipsoid in a circular section, and here, unlike the other cases discussed in which coalescence at  $\beta$  occurs, chance operates to the exclusion of choice. In practice a simulation of this behaviour may result from the impossibility of obtaining sufficiently critical readings to differentiate between readings showing a separation of the order of 0.001. This may be illustrated by reference to fig. 1. Here a minimum separation of about 0.002 occurs, a degree of birefringence which might be overlooked on casual scrutiny of the stone on the refractometer. This might appear to indicate a close approach to an optical axial position, but in point of fact the angular distance between the plane giving this minimum birefringence and the nearest optic axial plane is about 16°. It seems reasonable to infer that cases in which apparent coalescence of this kind occurs are not infrequent.

Finally, reference must be made to the forms assumed by the shadow-edge curves of isotropic and uniaxial gemstones. The straight line traced out by the stationary shadow-edge of an isotropic medium calls for no comment. An



FIG. 2. Curves of refractometer shadow-edges on the three principal planes of a biaxial crystal.

analysis of the behaviour of the uniaxial division can follow similar lines to that described earlier in this paper, when a more general case was considered, the necessary modification being introduced that the maximum or minimum principal refractive index shall be coincident with the intermediate principal refractive index. It will be evident that the shadow-edge curves traced out assume the forms of a straight line, given by the refractive index  $\omega$ , and a curve sweeping between this line and a maximum or minimum at  $\epsilon$ . These curves are sketched in fig. 3.

