

Indicatrix Orientation for Barium Feldspars Referred to Crystallographic Axes by Computer Using X-Ray and Extinction Data

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Abstract

A spindle stage method has been used to obtain optical directions accurately referred to crystallographic axes for three monoclinic barium feldspars, giving indicatrix orientation angles with errors of less than 0.5° . The present results show that $Z \parallel b$ for 37.5% Cn but $Y \parallel b$ for 91.9% and 97.8% Cn, confirming the results of Roy (1965). Present values of $2V$ agree well with those of Roy.

The crystal to be studied is mounted on a spindle stage which is designed to be positioned both on an X-ray camera and a polarizing microscope. The crystal is first accurately aligned in a specific crystallographic orientation. The stage is then transferred to the microscope, and extinction data is measured as the crystal is rotated through 360° about the spindle axis. The computer program of Bloss and Riess (1973) has been modified so as to give each extinction reading equal weight and also has been extended so as to calculate the indicatrix orientation with respect to the crystallographic axes.

Introduction

Knowledge of the nature and orientation of the optical properties of a biaxial crystal can be of use in determining composition or structural state. For example, in evaluating the composition of feldspar minerals, both $2V$ and indicatrix orientation angle (angle between a principal vibration direction and a crystallographic axis) have been used in evaluating the composition of feldspar minerals. Sometimes the first indication that a structural difference exists between two crystals is given by a change in optic orientation, as in potassium-barium feldspars (Roy, 1965).

Measurements of indicatrix orientation angles in crystals require a correlation between crystallographic and optical directions. The existing techniques for obtaining this correlation (identification of cleavage planes in the microscope or the use of drawings or photographs of a crystal already aligned by X-ray methods) can be subject to an experimental error of several degrees, although a skilled experimenter can obtain reliable results. The present paper describes a method whereby the crystallographic directions of a single crystal may be determined quite precisely by X-ray diffraction, and the optical directions may be determined without having to remount the crystal, using an adaptation of the method of Bloss and Riess (1973). The X-ray photographs and

optical measurements form a permanent record of the experiment, enabling the results to be verified later if necessary.

A similar method has been developed independently by Louisnathan and Bloss (in preparation).

Experimental Methods

Apparatus

The spindle stage, spindle, and miniature goniometer head used were designed and built in this laboratory and are shown in Figures 1 and 2. The needle is carried on the miniature goniometer head, which allows $4\text{--}6^\circ$ angular adjustment and 2 mm linear traverse. The spindle can be rotated 360° around its axis, and its position is read to 0.1° using a vernier scale.

Figure 1 shows a Unicam oscillation camera fitted with a flat back-reflection film holder and a mounting specially built to hold the spindle stage with its axis horizontal and accurately perpendicular to the X-ray beam.

A polarizing microscope (Fig. 2) is fitted with another mounting specially built to hold the spindle stage horizontal on the microscope stage. The crystal rotates in a small removable oil cell which is clipped to the mounting.

The crystals used should be large enough to give a

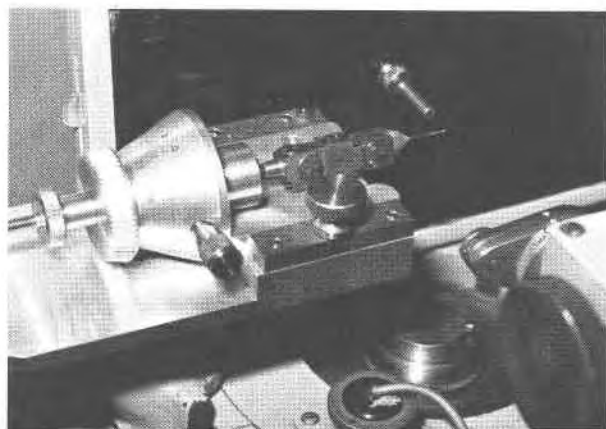


FIG. 1. Spindle stage with miniature goniometer head mounted on an X-ray camera fitted with a flat back-reflection film holder.

clear X-ray photograph and small enough to fit inside the oil cell. Those described in Table 1 were 0.4 mm in diameter.

Alignment of the Spindle Axis

The spindle axis must be aligned perpendicular to the X-ray beam. This can be accomplished by the use of double exposure Laue photographs. A crystal is affixed to the needle with a prominent zone axis approximately parallel to the spindle axis, and is rotated through 180° between exposures. Corrections can be estimated and applied simultaneously to the spindle mounting and goniometer head until the two traces of the zone coincide in a straight line down the middle of the photograph.

Preliminary Orientation of the Crystal

The crystal is first attached to a glass fiber using a glue (e.g., Durofix) which is soluble in amyl acetate and insoluble in water. The fiber is mounted on a standard X-ray goniometer head, and the crystal is aligned with a zone axis vertical using standard X-ray techniques.

The goniometer head is then transferred to the modified camera, and the crystal is positioned with a known crystallographic plane perpendicular to the X-ray beam.

The spindle stage is mounted on the modified camera and a second glue (e.g., Seccotine) soluble in water and insoluble in both amyl acetate and refractive index oils is applied to the spindle needle. The crystal is moved onto the needle using the horizontal traverses of the standard goniometer head. When the second glue has dried, the first can be dissolved.

If a spindle stage accepting a standard goniometer

head were available, the above procedure could be simplified.

Final Orientation

The crystal alignment usually requires a slight adjustment after it is transferred to the spindle needle. The corrections are estimated from double exposure Laue photographs, the crystal being rotated through 180° about the spindle axis between exposures. When the crystal is perfectly set the horizontal line of spots is straight and shows no doubling.

Optical Measurements

The spindle stage is now transferred to the polarizing microscope. Extinction readings are taken at 10° intervals as the crystal is rotated about the spindle axis. A Nakamura plate has been used to improve the accuracy of extinction readings, which were measured in white light. The spindle reading S and the extinction reading $M(S)$ are recorded over a 360° range of S . Following Bloss and Riess (1973), the *reference azimuth* is the microscope reading when the spindle axis lies East-West, and this position is found by averaging pairs of readings $M(S)$ and $M(S+180)$.

Time of Measurement

The time taken from the initial mounting of a crystal to the completed set of extinction measurements might be 2 or 3 days, but the time is variable: one run was completed in one day, but other crystals proved troublesome. Because most of this time is taken up by X-ray exposures (5 or 6 are usually needed), the experiment does not require continuous attention.

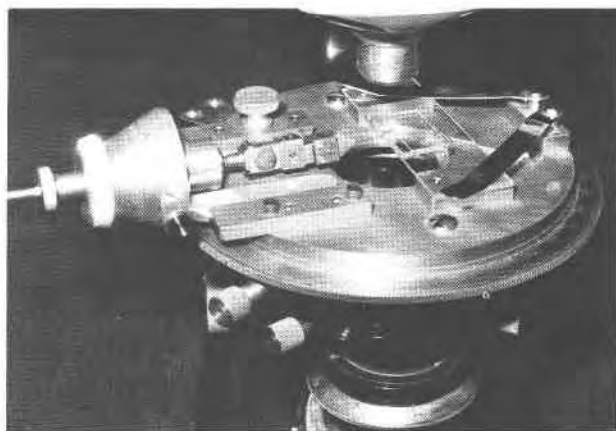


FIG. 2. Spindle stage with miniature goniometer head mounted on a polarizing microscope. The tip of the spindle needle is situated in a small oil cell.

TABLE 1. Preliminary Indicatrix Orientations and Optic Angles for Potassium Barium Feldspars

Sample ⁺	wt%Cn	Indicatrix orientation angles (degrees) [*]				2V _α [*]		
			present	Roy ^{**}		present ^{***}	present	Roy ^{**}
16	37.5	X:a=	-16.27(13)	-18.2	Z:b=	0.17(13)	78.28(18)	78.2
25	91.9	Z:a=	21.71(48)	23.5	Y:b=	0.48(48)	92.49(61)	92.4
28	97.8	Z:a=	21.95(16)	25.1	Y:b=	0.14(16)	96.86(23)	95.9

* Parenthesised figures represent the estimated standard deviation (esd) in terms of least units cited for the value to their immediate left, thus 0.14(16) means 0.14 esd 0.16.

** Data from Roy (1965, *Mineralog. Mag.*, vol. 35, page 508).

*** Expected value 0.00.

⁺ Sample	Designation	Location	Source
16	Hy BM84765	Binnenthal, Switzerland	Miss J. M. Sweet, British Museum
25	Cn 10062	Jakobsberg, Sweden	F. H. S. Vermaas, Pretoria, S.A.
28	Cn 100978	Big Creek, California	S. O. Agrell, Cambridge

Computing Method

When light passes through a biaxial crystal, the transverse plane containing the two vibration directions intersects the optical indicatrix in an ellipse. The

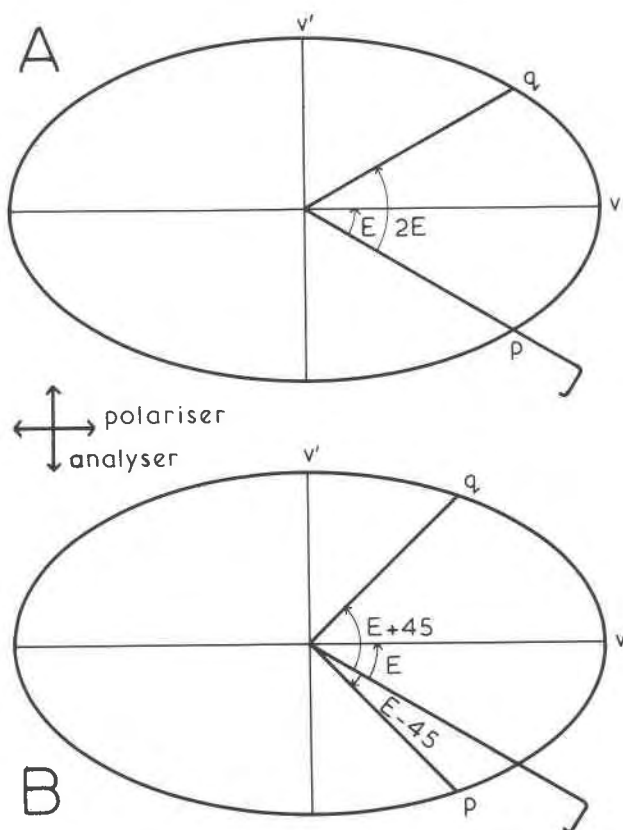


FIG. 3. Section of the indicatrix showing permitted vibration directions v, v' and alternative methods for generating equivibration directions p and q: A. by doubling extinction angle E; B. keeping p and q at right angles by adding and subtracting 45°.

permitted vibration directions are the major and minor axes of this ellipse; thus when one of these directions corresponds to the polarizer direction in the microscope, the crystal is in the extinction position. Since the indicatrix section is an ellipse, there are directions of equal refractive index, called equivibration directions (p and q in Figs. 3a and 3b), symmetrically disposed about the axes of the ellipse. Joel (1965) derived the following equation relating the directions p and q to optic axes a₁ and a₂ (all vectors are of unit length):

$$(p \cdot a_1)(p \cdot a_2) - (q \cdot a_1)(q \cdot a_2) = 0 \quad (1)$$

This equation can be used in a least-squares refinement of optic axis positions, given enough pairs of p and q vectors. The programs of Joel (1965) and of Bloss and Riess (1973) use extinction angles measured on a spindle stage to generate p and q as shown in Figure 3a. Vector p is taken along the spindle axis. The extinction angle E(S) is doubled so that [S, 2E(S)] are the polar coordinates of q. Thus p and q are chosen with an angle 2E(S) between them.

Equation 1 becomes trivial for p parallel to q and gives much less information about the positions of the optic axes for small values of 2E(S) than for 2E(S) near 90°. This means that refinement is carried out as if a weighting factor of sin 2E(S) were included in the data (see appendix). The presence of this arbitrary and un-normalized weighting means that readings with extinction angle close to 0° or 90° do not contribute as they should to the refinement, and estimated standard deviations (esd's) are calculated incorrectly (since the programming assumes unit weighting throughout).

It is, however, possible to choose different

equivibration directions separated by 90° (Fig. 3b), and in the present work this has been done. The polar coordinates of \mathbf{p} and \mathbf{q} are then $[S, E-45]$ and $[S, E+45]$. The rest of the program has been modified since \mathbf{p} is no longer always parallel to the spindle axis.

The effect of this change differs for different orientations of the crystal. The orientation can be favorable, when the solution is well determined, or unfavorable when it is badly determined. An example of an unfavorable orientation is when a principal vibration direction is within 10° of the spindle axis.

For a favorable orientation the results from both programs usually agree to within the esd's. $2V$ seems to be most affected—in the case of sample 16 (Table 1) $2V$ changed from 77.61° esd 0.27 using the previous program to 78.28° esd 0.18 using the present version. Roy's (1965) value is 78.2° (measured by direct observation of the crystal on a spindle stage).

For an unfavorable orientation the results from the two programs can differ more, and esd's calculated by the new program are in fact much greater (nearly trebled in one case). There are a number of factors which contribute to this increase, but because in practice such unfavorable orientations are undesirable, and the crystal should be repositioned in these cases, it is perhaps sufficient to point out that the new program indicates more insistently than the previous version the need to change the crystal orientation.

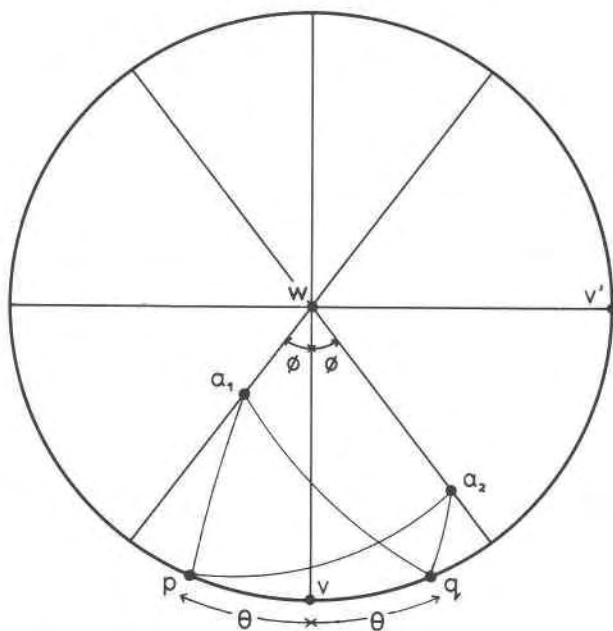


FIG. 4. Stereogram showing relationship between permitted vibration directions v, v' ; wave normal w ; optic axes a_1, a_2 ; and equivibration directions p and q .

The change towards higher esd's indicates that the incorrect weighting causes the effect of experimental errors to be underestimated. In one case the difference between the two methods was sufficiently large to prevent the new program from converging altogether, although the old program did converge with the same data. It is possible to choose the zone axis and plane used for alignment to avoid an unfavorable orientation.

The results of the least-squares refinement are the direction cosines of the optic axes. From these, the programs calculate $2V$ and the principal vibration directions and express all vectors in terms of polar coordinates $[S, E]$ as well as direction cosines. They calculate standard deviations for all these values.

The program has been extended to calculate the direction cosines of the crystallographic axes, using the matrix method of Bond (1946), from the cell dimensions and the crystallographic directions used in the alignment. It tabulates all possible indicatrix orientation angles—*i.e.*, all $(\mathbf{a}, \mathbf{b}, \mathbf{c}):(\mathbf{X}, \mathbf{Y}, \mathbf{Z})$. It plots a stereogram of all the vectors (on the line printer), transforms them so that the crystallographic axes are in the conventional position, and plots a second stereogram.

Results

This method has been used to investigate the optical properties of monoclinic potassium-barium feldspars. Preliminary results are shown in Table 1. The alignment along the diad axis is consistent with the estimated standard deviations. The results show that the principal vibration direction parallel to \mathbf{b} is \mathbf{Z} for 37.5% Cn, but changes to \mathbf{Y} for 91.9% and 97.8% Cn, confirming the results of Roy (1965). This radical difference in optical orientation for the two ranges of composition strongly suggests a difference in the feldspar structures, possibly related to the necessary change in Si-Al ordering pattern from 1:3 in KAlSi_3O_8 to 1:1 in $\text{BaAl}_2\text{Si}_2\text{O}_8$. Further measurements in this series are planned.

Systematic Errors

Transfer errors resulting from mis-alignment of the spindle stage would be revealed in measurements on crystals where the optical indicatrix is fixed by symmetry. If monoclinic crystals are being investigated, then the deviation of one principal vibration direction from the \mathbf{b} axis will indicate the magnitude of such transfer errors. Table 1 suggests that transfer errors are not contributing significantly to the total error in orientation, in the present apparatus.

If such errors were indicated they could be most easily measured using an orthorhombic crystal. Either the mechanical arrangements could be improved, or a correction matrix could be written into the program..

Acknowledgments

I wish to thank Dr. M. G. Bown for stimulating my interest in this method, and for his help during its development. I am grateful to Dr. P. Gay for allowing me access to his collection of the rare natural specimens of barium feldspars. Dr. Bown, Dr. Gay, and Dr. M. E. Fleet kindly read through the manuscript, and their helpful comments are much appreciated. The technical assistance of Mr. W. Nunn has been indispensable. I thank the Natural Environment Research Council for financial support.

Appendix: Mathematical Discussion

Let permitted vibration directions \mathbf{v}, \mathbf{v}' and wave normal \mathbf{w} of parallel light travelling through the biaxial crystal define orthonormal axes such that:

$$x = \mathbf{v} \quad y = \mathbf{v}' \quad z = \mathbf{w} \quad (\text{see Fig. 4})$$

and let optic axes $\mathbf{a}_1, \mathbf{a}_2$ and equivibration directions \mathbf{p} and \mathbf{q} have direction cosines as follows (all vectors are of unit length):

$$\mathbf{a}_1 = (l_1, m_1, n_1) \quad \mathbf{a}_2 = (l_2, m_2, n_2)$$

$$\mathbf{p} = (c, -s, 0) \quad \mathbf{q} = (c, s, 0)$$

where $c = \cos\theta$, $s = \sin\theta$, and 2θ is the angle between \mathbf{p} and \mathbf{q} .

According to the Biot-Fresnel construction, the optic axes lie in vertical planes (parallel to \mathbf{w}) at equal angles ϕ on either side of the plane \mathbf{wv} . If this is so, then:

$$\frac{-m_1}{l_1} = \tan \phi = \frac{m_2}{l_2}$$

but the direction of \mathbf{v} is subject to experimental error, and \mathbf{a}_1 and \mathbf{a}_2 may not be correctly refined, so the relationship is better written:

$$l_1 m_2 + m_1 l_2 = \lambda. \quad (2)$$

Similarly the equation of Joel (1965) can be written:

$$(\mathbf{p}_i \cdot \mathbf{a}_1)(\mathbf{p}_i \cdot \mathbf{a}_2) - (\mathbf{q}_i \cdot \mathbf{a}_1)(\mathbf{q}_i \cdot \mathbf{a}_2) = \lambda_i \quad (3)$$

for the i th pair \mathbf{p}, \mathbf{q} . The least-squares refinement minimizes the sum:

$$F = \sum_i (\lambda_i^2) \text{ over all measurements.}$$

Expansion of Equation (3) gives:

$$(l_1 c - m_1 s)(l_2 c - m_2 s) - (l_1 c + m_1 s)(l_2 c + m_2 s) = \lambda$$

$$(l_1 m_2 + l_2 m_1) 2sc = \lambda$$

$$(l_1 m_2 + l_2 m_1) \sin 2\theta_i = \lambda_i \quad (4)$$

which is similar to Equation (2).

The amount of information which the i th reading can give to the program about any solution variable (the contribution to F and its partial derivatives) is related to λ_i^2 and its partial derivatives. Since $\sin 2\theta$ is a common factor in the latter, a small value of $\sin 2\theta$ will cause the i th measurement to be almost excluded from the analysis. The value θ is entirely dependent on the method used to compute \mathbf{p} and \mathbf{q} . If \mathbf{p} and \mathbf{q} are generated at right angles then $\sin 2\theta = 1$ and all measurements are allowed to play their correct part in the refinement.

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Manuscript received, January 27, 1975; accepted for publication, July 9, 1975.