measurements for studies of local order in alloys is again emphasized. In the low-angle region the Compton scattering, the thermal diffuse scattering, atomic size effects and thermal attenuations are all at their smallest, so that with low-angle measurements one can obtain the greatest possible accuracy of interpretation.

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Ultrasonic Methods of Determining Elasto-Optic Constants of Uniaxial and Biaxial Crystals*

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Ultrasonic methods of studying the photoelastic behaviour of crystals, suggested by Mueller in 1938, have heretofore been applied only to glasses and cubic crystals. The method has been applied to uniaxial and biaxial crystals. Both theory and experiment are given. The ultrasonic methods replace the more difficult and less accurate interferometric methods for determining the elasto-optic constants. Results obtained on calcite, quartz, and barite are presented.

Introduction

Studies on the photoelastic behavior of solids have as their aim the determination of elasto-optic and piezo-optic constants (the p's and q's of Pockels (1906)) for the given solid. In an actual experiment, one determines the small changes in refractive index in different directions when a stress of known magnitude is applied in certain convenient directions. The bulk of the results obtained for p's and q's is based on the optical measurements in which one obtains relative path retardations using any one of the well known interferometers. The discovery of ultrasonics and the effect of a sound field on a transparent crystal through which polarized light is passing, led Bergmann & Fues (1936) to indicate the possibility of studying the photo-elastic constants of glasses by observing the polarized diffraction spectra from a point source. The situation very much improved when Hiedemann & Hoesch (1936) showed how easily line diffraction patterns could be obtained. The theory of diffraction of light in solids given by Nath & Mueller (1938) was experimentally verified by Hiedemann (1938). Subsequently, Mueller (1938) suggested in detail various methods of obtaining the elasto-optic constants of glasses and cubic crystals by studying the polarized diffraction spectra. Using the procedure suggested by Burstein *et al.* (1948), Vedam (1950) and Iyengar (1953) obtained all the photo-elastic constants of some glasses and cubic crystals by combining the relative path retardation methods using the Babinet compensator with the ultrasonic method due to Mueller (1938). It may be pointed out that the ultrasonic methods yield results to the same order of accuracy as the Babinet compensator method, whereas the interferometric methods often yield results differ-

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ing by as much as 50% from the Babinet measurements.

For non-cubic systems, however, all the photoelastic constants of quartz, calcite and beryl by Pockels (1906) topaz by Eppendahl (1920), and barite by Vedam (1951) have been measured by optical methods, i.e., by Babinet and interferometric methods. The interferometric methods are in several cases not very accurate and hence it is desirable to develop another method which will be a more reliable supplement to the Babinet observations.

Although Mueller (1938) has indicated that his method is also applicable to uniaxial crystals like quartz and calcite, no attempts seem to have been made to use it to study these crystals. The purpose of this paper is to apply Mueller's method to the uniaxial crystals quartz and calcite, and the biaxial crystal barite.

Theoretical considerations

If a mechanical disturbance is propagated in an arbitrary direction in a crystal then in general it will be in the form of quasi longitudinal and quasi transverse waves. In certain special directions in the crystal the waves will be pure, as shown by Borgnis (1955). In our present investigations, we are interested only in the purely longitudinal waves and the diffraction pattern they produce. Therefore, we consider only waves propagated along the axes of a uniaxial crystal.

Consider a longitudinal wave propagated along the Z axis of a uniaxial crystal such as calcite. This wave produces longitudinal strains along the z axis only. We shall deliberately avoid the formation of shear waves in the crystal; hence their influence on the optical ellipsoid is eliminated. For the particular direction of observation parallel to the y axis, before the crystal is strained, the cross-section of the optical ellipsoid is represented by

$$B_{11}X^2+B_{33}Z^2=1\ . \tag{1}$$

Under the influence of the longitudinal waves, and consequent longitudinal strains, every volume element of the crystal becomes birefringent. Hence the cross section of the optical ellipsoid now becomes

$$B'_{11}X^2+B'_{33}Z^2+2B_{31}ZX=1\ . \tag{2}$$

Then according to laws of photoelasticity, the polarization constants B_{ik} for calcite are given by

$$\begin{aligned} B'_{11}-B_{11}&=p_{11}x_x+p_{12}y_y+p_{13}z_z+p_{14}y_z\\ B'_{33}-B_{33}&=p_{31}x_x+p_{31}y_y+p_{33}z_z\\ B'_{31}\quad&=p_{41}x_x-p_{41}y_y\qquad\quad+p_{44}y_z\ . \end{aligned}$$

The ellipse (2) has its axes in the directions which make angles θ and $(\theta+90°)$ with the Z axis where $\tan 2\theta=2B_{31}/(B_{11}-B_{33})$. These directions do not vary with time and are the same for every volume element.

For a longitudinal wave along the Z axis the only strain that remains is z_z.

$$\begin{aligned} B'_{11}-B_{11}&=p_{13}z_z\\ \text{and}\qquad&\qquad (3)\\ B'_{33}-B_{33}&=p_{33}z_z\ . \end{aligned}$$

Hence there is no tilt of the axes of the optical ellipsoid.

The lengths a_{I} and a_{III} of the new or the transformed axes of the co-ordinate system (which in the present case coincide with the original axes, i.e., the axes of the crystal) vary periodically and we can write for a progressive wave, following Mueller (1938)

$$\begin{aligned} a_{\mathrm{I}}&=(1/n_x^2)-i_{\mathrm{I}}2\pi\nu Ak/\varrho\\ &\qquad\times\sin 2\pi\nu(t-(z\cos\varphi+y\sin\varphi)/v)\\ a_{\mathrm{III}}&=(1/n_z^2)-i_{\mathrm{III}}2\pi\nu Ak/\varrho\\ &\qquad\times\sin 2\pi\nu(t-(z\cos\varphi+y\sin\varphi)/v)\ , \end{aligned} \tag{4}$$

where ν, A, k, ϱ, t, φ and v have the usual significance.

Thus the quantities i_{I} and i_{III} are proportional to the amplitudes of variation of the principal axes of the index ellipse.

Further it may be noted that a_{I} represents B'_{11} and a_{III} represents B'_{33} at any particular instant.

From equations (4) we have

$$i_{\mathrm{I}}/i_{\mathrm{III}}=(a_{\mathrm{I}}-1/n_x^2)/(a_{\mathrm{III}}-1/n_z^2)=p_{13}z_z/p_{33}z_z=p_{13}/p_{33}\ . \tag{5}$$

Now, if μ_{I} and μ_{III} are the amplitudes of index variations along the X and Z directions, respectively, then

$$a_{\mathrm{I}}=1/(n_x+\mu_{\mathrm{I}})^2\approx(1/n_x^2)-(2\mu_{\mathrm{I}}/n_x^3) \tag{6}$$

and similarly

$$a_{\mathrm{III}}\approx(1/n_z^2)-(2\mu_{\mathrm{III}}/n_z^3)\ . \tag{6a}$$

Hence from equation (4)

$$\mu_{\mathrm{I}}=n_x^3.i_{\mathrm{I}}.K$$

and

$$\mu_{\mathrm{III}}=n_z^3.i_{\mathrm{III}}.K\ ,$$

where K is a constant.

Thus the amplitudes of refractive index variation are proportional to i_{I} and i_{III} which are the amplitudes of variation of the principal axes of the index ellipse. Hence

$$\begin{aligned} \mu_{\mathrm{I}}/\mu_{\mathrm{III}}&=(n_x^3/n_z^3)\times(i_{\mathrm{I}}/i_{\mathrm{III}})\\ &=(n_0^3/n_e^3)\times(p_{13}/p_{33})=R_1\ . \end{aligned} \tag{7}$$

Further, these amplitudes of refractive indices parallel to and perpendicular to the acoustical wave normals cause corrugations of two different degrees on the emergent optical wave front. From the Raman-Nath theory, each of these corrugated wave fronts causes a diffraction pattern. There are then two diffraction patterns, one with its electric vector parallel to the sound wave normals and the other perpendicular to them. The intensity of each pattern of a particular order depends upon the μ causing the pattern.

If the incident light is polarized in any arbitrary direction, its two components of vibrations of amplitudes parallel and perpendicular to the acoustical wave normals, i.e., parallel to the principal axes, will each produce a diffraction pattern whose amplitude in the mth order is given by

$$E_{m\mathrm{I}} = E_{\mathrm{I}} J_m(v_{\mathrm{I}})$$
$$E_{m\mathrm{III}} = E_{\mathrm{III}} J_m(v_{\mathrm{III}}) , \qquad (8)$$

where $v_{\mathrm{I}}=2\pi\mu_{\mathrm{I}}L/\lambda$ and $v_{\mathrm{III}}=2\pi\mu_{\mathrm{III}}L/\lambda$; J_m is the mth order Bessel function, L the width of the sound field, and λ the wave length of light. The two components are diffracted to the same extent and have the same frequency and phase. For small values of m and v

$$E_{m\mathrm{I}}/E_{m\mathrm{III}} = E_{\mathrm{I}}(v_{\mathrm{I}})^m/E_{\mathrm{III}}(v_{\mathrm{III}})^m$$
$$= (E_{\mathrm{I}}/E_{\mathrm{III}})(\mu_{\mathrm{I}}/\mu_{\mathrm{III}})^m = R_1{}^m. \qquad (9)$$

If further $E_{\mathrm{I}}=E_{\mathrm{III}}$, which will be the case of the incident light has its electric vector inclined at an angle of 45° to the sound field, then

$$R_1{}^m = (n_o{}^3 p_{13})/(n_e{}^3 p_{33}) . \qquad (10)$$

The ratio of intensities in the components of the mth diffraction order is

$$B_m = [E_{m\mathrm{I}}/E_{m\mathrm{III}}]^2. \qquad (11)$$

Then, assuming incident light polarized at 45° and extrapolation to zero sound amplitudes, one has for both progressive and standing waves

$$B_m{}^o = R_1{}^{2m}. \qquad (12)$$

In most cases one measures the intensity of only the first diffraction orders for which $m=1$.

Since the ratio of intensities is the quantity experimentally obtained, the sign of R_1 cannot be determined from the ultrasonic measurements. However, this sign is readily obtained from optical measurements.

Experimental details

The method employed by the previous authors consists in allowing a plane polarized beam of light to pass through the experimental crystal prism in a direction perpendicular to the direction of ultrasonic wave normals (the plane of the polarization of the beam of light making an angle of 45° with them). An analyzer is used to study the state of polarization of the diffracted orders in the Hiedemann pattern. This method is suitable only for glasses and cubic crystals. For uniaxial and biaxial crystals which are birefringent (even in the unexcited state), the method of passing natural light through a double image prism and examining it with an analyzer, suggested by Mueller, may be employed. An analogous method was used by Bergmann & Fues (1936) and by Gates & Hiedemann (1956) to study glasses.

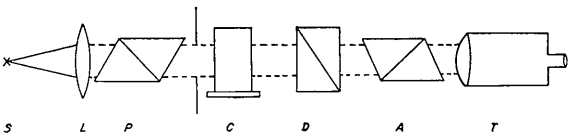

Fig. 1. Optical arrangement for ultrasonic studies.

S—Source
L—Lens
P—Polarizer
C—Crystal specimen with attached transducer
D—Double image prism
A—Analyzer
T—Telescope

Fig. 1 indicates the experimental arrangement. A plane parallel beam of monochromatic light is passed through a polarizing nicol P and is polarized at 45° with the vertical. This plane polarized beam passes through the crystal specimen C and then through the double image prism D. The double image prism is mounted with its axis parallel to the direction of observation and is so adjusted that the two images are seen one above the other. The analyzer A can be rotated about the optic axis. It is initially set in such a way that the plane of polarization of light passing through it is horizontal. The double image prism is then rotated through a right angle, if necessary, to make the upper image disappear. This means that the lower image has its plane of vibration horizontal and the upper one vertical. The analyzer is then rotated through 45°, the two images now having equal intensity. Now the ultrasonic power is applied to the experimental crystal along one of its axes and the Hiedemann pattern is observed. The ultrasonic power is next adjusted, so that only first orders are excited. The images of the first orders for the two perpendicularly vibrating components differ in their intensity. The analyzer is then rotated to equalize the intensities of the first order components. Then if α is the angle of rotation of the analyzer from the zero position, $\tan\alpha$ gives directly R_1 from which the ratio R of the corresponding elasto-optic constants can be calculated. The corrections for the finite amplitude of the ultrasonic power, suggested by Mueller (1938), and for optical activity, used by Vedam & Ramachandran (1951), are made here.

Uniaxial crystals

Results for quartz

Measurements were made on three rectangular prisms of quartz. The largest sample had dimensions of 10·7 × 10·3 × 9·7 mm. Prisms of different dimensions were used both to check the observations in the several cases and to see whether the lateral dimensions influence the observed values of R. No difference could be observed for lateral dimensions between 5 and 10·3 mm.

The values of α after allowing for the finite amplitude of the ultrasonic power and the optical activity are indicated in Table 1.

Table 1. *Ratio of elasto-optic constants of low-temperature quartz*

Direction of ultrasonic excitation	Direction of observation	α	Expression for the ratio R	Numerical value of R: Ultrasonic method	Numerical value of R: Optical method
X	Z	$45+12°$	p_{12}/p_{11}	1·57	1·67
X	Y	$45+19$	p_{31}/p_{11}	2·00	1·78
Z	X	$45+25$	p_{13}/p_{33}	2·81	2·67

The numerical values of R, the ratios of the elasto-optic constants, obtained by the ultrasonic method are in fairly good agreement with those obtained by Pockels (1906), who determined them by optical methods. Thus one has an independent check on the optical measurements.

Results for calcite

Prisms of calcite having approximately the same dimensions as the quartz samples were used in these measurements. The results are given in Table 2.

Table 2. *Ratio of elasto-optic constants of calcite*

Direction of excitation	Direction of observation	Expression for R	Numerical value of R: Ultrasonic method	Numerical value of R: Optical method
X	Z	p_{12}/p_{11}	1·90	2·0
X	Y	p_{31}/p_{11}	2·79	3·0
Z	X	p_{13}/p_{33}	2·40	2·6

The data under R are those calculated from Pockels' values for calcite and those obtained from the ultrasonic studies. The agreement is fairly good.

Biaxial crystals

The expressions collected in Table 3 can be derived for orthorhombic crystals. The derivation is very similar to that given for the uniaxial crystal calcite.

Table 3. *Ratio of elasto-optic constants for orthorhombic crystals*

Direction of excitation	Direction of observation	R, the ratio of elasto-optic constants
X	Y	p_{31}/p_{11}
X	Z	p_{21}/p_{11}
Y	Z	p_{12}/p_{22}
Y	X	p_{32}/p_{22}
Z	X	p_{23}/p_{33}
Z	Y	p_{13}/p_{33}

Results for barite

The elasto-optic constants were measured for several samples of barite by the ultrasonic method. The results are given in Table 4.

Table 4. *Ratio of elasto-optic constants for barite*

R	Numerical value of R: Ultrasonic method	Numerical value of R: Optical method
p_{31}/p_{11}	0·73	0·76
p_{21}/p_{11}	1·28	1·19
p_{12}/p_{22}	1·33	1·41
p_{32}/p_{22}	1·04	0·79
p_{23}/p_{33}	0·81	0·89
p_{13}/p_{33}	0·75	0·71

It is seen that the values obtained by studies of the ultrasonic diffraction pattern are in good agreement with the values obtained by the optical measurements by Vedam (1951) in almost all cases.

Conclusions

The ultrasonic method can be used to study both uniaxial and biaxial crystals. Its accuracy depends primarily on the size of the angle of polarization of light. In the present measurements there can be inaccuracies up to 10%.

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