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Brillouin Scattering in Anisotropic Media: Calcite

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We have used comparative Brillouin scattering (reference scatterer: toluene) to measure the nine photoelastic tensor elements of calcite (eight elastically symmetric, one elastically antisymmetric) in both magnitude and sign. Our measurement of the recently predicted antisymmetric component agrees in magnitude to 5% with that theoretically predicted. By adopting the theoretical sign for this component, the signs of all other components are determined from the data. The 5% agreement gives a measure of the *absolute* accuracy of the results. The presence of the antisymmetric component causes p_{2323} and p_{2332}, previously believed to be equal, to differ by more than a factor of 10. The formulas needed to analyze Brillouin scattering in anisotropic media are derived using a new constitutive relation and a new Green's function for radiation in an anisotropic medium. Inclusion of boundary effects (solid angle expansion, scattering-volume demagnification, etc.) makes the formulas valid *outside* the scattering medium. The convenience of comparative Brillouin scattering for the numerical determination of the photoelastic tensor components, particularly in low-symmetry crystals, is emphasized.

I. INTRODUCTION

We report here measurements of the photoelastic tensor elements of calcite made by comparative Brillouin scattering using toluene as a reference scatterer. All nine elements (eight symmetric upon interchange of the tensor indices associated with the measure of elastic deformation; one antisymmetric upon the interchange) were measured in both magnitude and sign using Brillouin scattering alone.

The purposes of this work are fivefold: (i) to measure the recently predicted[1,2] *antisymmetric* tensor element that arises from the coupling of *rotations* (in contrast to strains) to the scattering process (we find agreement between the predicted and measured magnitudes of this element to within 5%); (ii) to demonstrate that the algebraic *signs* of all the tensor elements can be determined from the theoretically known sign of the antisymmetric tensor element; (iii) to point out that measurement of the antisymmetric tensor element can be used to determine the *absolute* calibration of the scattered light intensity; (iv) to show that comparative Brillouin scattering is a method as accurate in determining absolute values of photoelastic tensor elements as acousto-optic diffraction or static-load-induced birefringence and is more convenient for low-symmetry crystals; and (v) to present Brillouin-scattering formulas valid for crystals of any symmetry and of any optical or acoustic anisotropy in any orientation, for any angle of scattering, and for rather general orientations of the exit surface. A derivation of the latter is presented that is based on a new constitutive relation derived by an *ab initio* calculation[2,3] and on a new Green's function[4] for radiation in an anisotropic medium.

Calcite was chosen for this study because (a) it is expected to possess a very large antisymmetric photoelastic tensor element resulting from its large optical birefringence, (b) it is nonpiezoelectric and nonferroelectric, (c) it is not optically active, (d) it has lower symmetry (trigonal, $\bar{3}\,m$) than rutile[5] (the only other crystal whose antisymmetric photoelastic tensor element has been measured), and (e) it is readily available in large high-optical-quality crystals. Calcite has added interest for this study because it was the only strongly birefringent crystal chosen by Pöckels for study[6] to demonstrate the correctness of his phenomenological formulation[7] of the photoelastic effect.

The Pöckels formulation assumed that the strain completely characterized the elastic deformation that caused the photoelastic effect. Since Pöckels measured refractive index changes caused by static *homogeneous* deformations, the rotation associated with the elastic deformation was simply a rigid-body rotation. As such, it was of trivial interest, and Pöckels attempted to eliminate its effect on his measurements by alterations of his experimental technique.[6] During the past half-century the photoelastic interaction has often been studied by the deflection of light by elastic waves created thermally (Brillouin scattering[8]) or coherently by transducers (acousto-optic diffraction[9,10]). All workers in these fields until very recently[1,2,5] assumed that the Pöckels formulation applied equally well to these forms of measurement of the photoelastic interaction. This is not so because in these measurements the wavelength of the elastic wave is typically very small compared to the dimensions of the medium studied. Thus, the elastic deformation is *inhomogeneous* and the associated rotation is not a rigid-body rotation and cannot be avoided experimentally. The contribution of rotations to the photoelastic interaction in these cases is thus as intrinsic as that arising from strain.

The effect of rotations on the photoelastic interaction can be derived by a phenomenological argument[1] or a fundamental calculation[2] with similar results. The change in the inverse dielectric tensor $\Delta(\overleftrightarrow{\kappa}^{-1})$ is found to be given (omitting for the moment the indirect photoelastic effect) by

$$\begin{aligned}(\Delta\kappa^{-1})_{(ij)} &= p_{(ij)(kl)}S_{(kl)} + p_{(ij)[kl]}\tilde{R}_{[kl]}\\ &= p_{(ij)kl}\,u_{k,l}\ , \qquad (1.1)\end{aligned}$$

where the summation convention has been used and the infinitesimal strain and mean rotation are defined, respectively, by

$$S_{(kl)} \equiv \tfrac{1}{2}(u_{k,l}+u_{l,k}) \qquad (1.2)$$

and

$$\tilde{R}_{[kl]} \equiv \tfrac{1}{2}(u_{k,l}-u_{l,k})\ . \qquad (1.3)$$

Parentheses enclosing subscripts indicate symmetry upon interchange of the subscripts, while brackets indicate antisymmetry upon interchange. The gradient of the displacement vector is denoted by $u_{k,l} \equiv \partial u_k/\partial x_l$. In Eq. (1) the Pöckels tensor is denoted by $p_{(ij)(kl)}$. An explicit expression for the new antisymmetric part $p_{(ij)[kl]}$ of the total photoelastic tensor $p_{(ij)kl}$ is given by[1,2]

$$p_{(ij)[kl]} = [\delta_{i[k}(\kappa^{-1})_{l]j} + \delta_{j[k}(\kappa^{-1})_{l]i}]\ . \qquad (1.4)$$

The elements of this tensor allowed by symmetry for the various crystal classes have been discussed elsewhere.[1,2] (These requirements are more specific than those for a general fourth-rank tensor symmetric on the first pair of indices and antisymmetric on the second pair.) Suffice it to say here that for calcite the only nonzero elements are *predicted* by Eq. (1.4) to be

$$p_{(23)[23]} = p_{(13)[13]} = \tfrac{1}{2}(n_e^{-2} - n_o^{-2}) = +0.0450 \qquad (1.5)$$

for light of 5145-Å wavelength. Here n_o and n_e are the principal ordinary and extraordinary refractive indices. Note that when these indices are known, the *sign* and *magnitude* of the antisymmetric part of the photoelastic tensor can be calculated. (The sign is positive for a negative uniaxial crystal and vice versa.) The sign can be used in conjunction with Brillouin scattering (or acousto-optic diffraction) measurements *alone* to determine the algebraic signs of *all* of the photoelastic tensor elements. The predicted magnitude can be used as an absolute standard or calibration for such measurements. In this work, however, we will use the light scattering from toluene as an absolute standard in order to show that the measured magnitude of the antisymmetric tensor element agrees with the prediction of Eq. (1.5).

II. THEORY OF BRILLOUIN SCATTERING IN ANISOTROPIC MEDIA

Calculation of the Brillouin-scattered light intensity requires a solution of the driven or inhomogeneous wave equation

$$\begin{aligned}&\overleftrightarrow{\alpha}(-i\vec{\nabla}, \omega_B)\cdot\vec{\mathrm{E}}(\vec{\mathrm{r}}, \omega_B)\\ &\quad\equiv\left[\left(\frac{c}{\omega_B}\right)^2[\vec{\nabla}\vec{\nabla} - \overleftrightarrow{1}(\vec{\nabla}\cdot\vec{\nabla})] - \overleftrightarrow{\kappa}(\omega_B)\right]\cdot\vec{\mathrm{E}}(\vec{\mathrm{r}}, \omega_B)\\ &\quad= \vec{\mathcal{P}}(\vec{\mathrm{r}}, \omega_B)/\epsilon_0\ . \qquad (2.1)\end{aligned}$$

Here $\vec{\mathrm{r}}$ is the position vector, ω_B is the scattered light frequency (either the Stokes frequency $\omega_O - \omega_A$ or the anti-Stokes frequency $\omega_O + \omega_A$, where ω_O is the input optical frequency and ω_A the acoustic frequency), c is the velocity of light in vacuum, $\vec{\nabla}$ is the gradient operator, $\overleftrightarrow{\kappa}(\omega_B)$ is the dielectric of free space (we choose to work in mks units). $\vec{\mathcal{P}}$ is the nonlinear driving polarization and $\vec{E}$ the electric field. The Fourier expansion of any of these fields (denoted by Z) follows the general notation

$$Z(\vec{r},t)=\tfrac{1}{2}[Z(\vec{r},\omega_B)e^{-i\omega_B t}+Z^*(\vec{r},\omega_B)e^{+i\omega_B t}] + \text{terms at other frequencies} . \quad (2.2)$$

When plane waves are used, the notation will conform to

$$Z(\vec{r},t)=\tfrac{1}{2}[Z(\omega_B)e^{i(\vec{k}_B\cdot\vec{r}-\omega_B t)}+Z^*(\omega_B)e^{-i(\vec{k}_B\cdot\vec{r}-\omega_B t)}] + \text{other terms}, \quad (2.3)$$

so that $Z(\omega_B)$ is the amplitude of the ω_B Fourier component of $Z(\vec{r},t)$.

In all previous treatments of Brillouin scattering in an anisotropic medium[11–16] the nonlinear driving polarization has been determined from the Pöckels phenomenlogical formulation of the interaction. Here we use the form[2] derived from our *ab initio* theory of electrodynamics in anisotropic elastic dielectrics[3]:

$$\vec{\mathcal{P}}_i(\vec{r},\omega_B)=\epsilon_0\left[\chi_{ijkl}-\frac{2d_{ijr}a_r a_s e_{skl}}{\epsilon_0 a_p \kappa_{pq}(\omega_A)a_q}\right] \times E_j(\vec{r},\omega_O)u_{k,l}(\vec{r},\omega_A) , \quad (2.4)$$

where

$$\chi_{ijkl}=-\tfrac{1}{2}\kappa_{im}(\omega_O)p_{(mn)kl}\,\kappa_{nj}(\omega_O) , \quad (2.5)$$

$$2d_{ijk}=-\tfrac{1}{2}\kappa_{il}(\omega_O)r_{lmk}\kappa_{mj}(\omega_O) . \quad (2.6)$$

Here e_{skl} is the piezoelectric tensor, that when summed over the strain yields the polarization; r_{lmk} is the conventional clamped electro-optic tensor, that when summed over the low-frequency electric field yields the change in the inverse dielectric tensor; $2d_{ijk}$ is the clamped electro-optic tensor expressed in the form conventional for the optical mixing tensor; $\vec{a}$ is a unit vector in the direction of propagation of the acoustic wave. The derivation[2] of Eq. (2.4) yields constitutive expressions for χ_{ijkl}, d_{ijk}, e_{jkl}, and κ_{ij} including their expected frequency dependence.

The second term of Eq. (2.4) represents the indirect photoelastic effect, that is, the succession of the piezoelectric and electro-optic effects. It is numerically significant in strongly piezoelectric crystals such as $LiNbO_3$, $LiTaO_3$, α-HIO_3, etc. This effect was first calculated by Chapelle and Taurel[17] though their work had been forgotten[18] when the first photoelastic measurements were made on these materials.

In a typical geometry used to observe Brillouin scattering the nonlinear polarization $\vec{\mathcal{P}}$ is nonzero over a small but finite region in the medium under study. Outgoing-plane-wave solutions are inappropriate for such a geometry. Hence Eq. (2.1) is best solved by a Green's-function technique. Formally, the solution is

$$\vec{E}(\vec{r},\omega_B)=\int\overleftrightarrow{G}(\vec{r},\vec{r}\,')\cdot\vec{\mathcal{P}}(\vec{r}\,',\omega_B)\,d\vec{r}\,'/\epsilon_0 , \quad (2.7)$$

where the dyadic Green's function is given by

$$\overleftrightarrow{G}(\vec{r},\vec{r}\,')=\int_{-\infty}^{+\infty}\overleftrightarrow{\alpha}^{-1}(\vec{k},\omega_B)e^{i\vec{k}\cdot(\vec{r}-\vec{r}\,')}\,d\vec{k}/(2\pi)^3 . \quad (2.8)$$

The Green's function of Eq. (2.8), which is valid for arbitrary anisotropy of the medium, has been derived,[4] that is, $\overleftrightarrow{\alpha}^{-1}$ calculated and the integral of Eq. (2.8) evaluated in the far field. Using this form[4] of $\overleftrightarrow{G}$ and taking the optical and acoustic driving fields that produce $\vec{\mathcal{P}}(\vec{r})$ as plane waves over the source volume which we assume for simplicity is a rectangular parallelepiped, we find the radiated electric field to be

$$\vec{E}^{\varphi}(\vec{r},\omega_B)=\frac{V_s\Phi\cos\delta^{\varphi}}{4\pi\epsilon_0 r(K^{\varphi})^{1/2}}k_B^{\varphi}(\vec{s}_0,\omega_B) \times\vec{\mathcal{E}}^{\varphi}(\vec{s}_0,\omega_B)\,\vec{\mathcal{E}}^{\varphi}(\vec{s}_0,\omega_B)\cdot\vec{\mathcal{P}}(\omega_B)e^{i\vec{k}_0^{\varphi}\cdot\vec{r}}e^{i\mathfrak{p}} , \quad (2.9)$$

where V_s is the scattering volume, r is the distance from the volume to the surface, $\vec{s}_0$ is a unit wave vector for the wave whose Poynting vector is in the direction of observation $\vec{r}$, $k_B^{\varphi}(\vec{s}_0,\omega_B)$ is the magnitude of the wave vector of this wave, K^{φ} is the Gaussian curvature in $\vec{k}$ space of the $\omega(\vec{k})$ surface for the φ polarization at the point where the surface normal is parallel to the direction of observation $\vec{r}$, δ^{φ} is the angle between the Poynting vector and the wave vector (or equivalently, between $\vec{\mathcal{E}}^{\varphi}$ and $\vec{\mathfrak{D}}^{\varphi}\equiv\overleftrightarrow{\kappa}\cdot\vec{\mathcal{E}}^{\varphi}$), and $\mathfrak{p}$ is a numerical phase which we need not specify here. Φ is defined by

$$\Phi=\frac{1}{V_0}\int e^{i\Delta\vec{k}^{\varphi}\cdot\vec{r}}\,d\vec{r}=\prod_{i=1}^{3}\frac{\sin(\frac{1}{2}\Delta k_i^{\varphi}l_i)}{\frac{1}{2}\Delta k_i^{\varphi}l_i} , \quad (2.10)$$

where

$$\Delta\vec{k}^{\varphi}\equiv\vec{k}_O+\vec{k}_A-\vec{k}_B^{\varphi}(\vec{s}_0,\omega_B) \quad (2.11)$$

and l_1, l_2, and l_3 are the lengths of the sides of the parallelepiped. From Eq. (2.10) it can be seen that a significant electric field occurs only near

$$\Delta\vec{k}^{\varphi}=0 , \quad (2.12)$$

which is called the phase matching or Bragg scattering condition. $\vec{\mathcal{E}}^{\varphi}(\vec{s}_0,\omega_B)$ is a real eigenvector[3] of the homogeneous wave equation

$$(\overleftrightarrow{1}-\vec{s}_0\vec{s}_0)\cdot\vec{\mathcal{E}}^{\varphi}=[1/n^{\varphi}(\vec{s}_0,\omega_B)]^2\,\overleftrightarrow{\kappa}(\omega_B)\cdot\vec{\mathcal{E}}^{\varphi} \quad (2.13)$$

normalized according to

$$\vec{\mathcal{E}}^{\varphi}\cdot\overleftrightarrow{\kappa}\cdot\vec{\mathcal{E}}^{\theta}=\delta_{\varphi\theta} . \quad (2.14)$$

In Eq. (2.13), n^{φ} is the refractive index and φ denotes the polarization state.

The magnetic induction $\vec{H}$ can now be calculated and the time-averaged Poynting vector

$$\tfrac{1}{2}\,\mathrm{Re}[\vec{\mathrm{E}}^{\varphi}(\vec{\mathrm{r}},\omega_B)\times\vec{\mathrm{H}}^{\varphi *}(\vec{\mathrm{r}},\omega_B)]$$

is found to be

$$\vec{\mathrm{S}}^{\varphi}=\frac{c^2[k_B^{\varphi}(\vec{\mathrm{s}}_0,\omega_B)]^3V_s^2\Phi^2\cos^2\delta^{\varphi}}{32\pi^2\epsilon_0 r^2\omega_B K^{\varphi}}\,|\vec{\mathcal{E}}^{\varphi}\cdot\vec{\mathcal{P}}(\omega_B)|^2$$

$$\times[\vec{\mathrm{s}}_0(\vec{\mathcal{E}}^{\varphi}\cdot\vec{\mathcal{E}}^{\varphi})-\vec{\mathcal{E}}^{\varphi}(\vec{\mathcal{E}}^{\varphi}\cdot\vec{\mathrm{s}}_0)]\,. \quad (2.15)$$

It is now necessary to relate the average-displacement-vector amplitude (in the mean-square sense) to the temperature of the medium. We expand the displacement vector considered as a function of position and time in terms of plane waves as

$$\vec{\mathrm{u}}(\vec{\mathrm{r}},t)=\sum_{\vec{\mathrm{k}}_A,\xi}\left(\frac{\hbar}{2\rho V\omega_A(\vec{\mathrm{k}},\xi)}\right)^{1/2}[a(\vec{\mathrm{k}}_A,\xi)e^{i(\vec{\mathrm{k}}_A\cdot\vec{\mathrm{r}}-\omega_A t)}$$

$$+a^*(\vec{\mathrm{k}}_A,\xi)e^{-i(\vec{\mathrm{k}}_A\cdot\vec{\mathrm{r}}-\omega_A t)}]\vec{\mathrm{b}}(\vec{\mathrm{a}},\xi)\,. \quad (2.16)$$

Here V is the normalization volume of the acoustic modes, ρ is the mass density of the medium, $\hbar$ is Planck's constant divided by 2π, $a(\vec{\mathrm{k}}_A,\xi)$ is an amplitude factor depending on the acoustic wave vector $\vec{\mathrm{k}}_A$ and mode type ξ ($\xi=1,2,3$) having the property $a^*(\vec{\mathrm{k}}_A,\xi)=a(-\vec{\mathrm{k}}_A,\xi)$, and $\vec{\mathrm{b}}(\vec{\mathrm{a}},\xi)$ is a real unit displacement eigenvector satisfying the acoustic propagation eigenvalue equation

$$c_{ijkl}a_ja_lb_k(\vec{\mathrm{a}},\xi)=\rho v^2(\vec{\mathrm{a}},\xi)b_i(\vec{\mathrm{a}},\xi)\,. \quad (2.17)$$

Here c_{ijkl} is the usual elastic-stiffness tensor, $\vec{\mathrm{a}}$ the unit acoustic wave vector, and $v(\vec{\mathrm{a}},\xi)$ the velocity of the ξ-type acoustic mode related to the frequency by

$$\omega_A(\vec{\mathrm{a}},\xi)=k_Av(\vec{\mathrm{a}},\xi)\,. \quad (2.18)$$

It can be shown from Eq. (2.17) that the displacement eigenvectors $\vec{\mathrm{b}}$ are orthogonal. With unit normalization we have

$$\vec{\mathrm{b}}(\vec{\mathrm{a}},\xi)\cdot\vec{\mathrm{b}}(\vec{\mathrm{a}},\eta)=\delta_{\xi\eta}\,. \quad (2.19)$$

The elastic energy of the scattering medium

$$H=\tfrac{1}{2}\int_V(\rho\dot{u}_i\dot{u}_i+u_{i,j}c_{ijkl}u_{k,l})d\vec{\mathrm{r}}\,, \quad (2.20)$$

derivable from Eq. (2.16), can be expressed in terms of the expansion of Eq. (2.16) as

$$H=\sum_{\vec{\mathrm{k}}_A,\xi}\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)|a(\vec{\mathrm{k}}_A,\xi)|^2\,. \quad (2.21)$$

It can be seen here that $|a|^2$ represents the number of acoustic excitations (phonons). It was for this reason that the expansion constants were chosen as in Eq. (2.16). By treating the phonons as noninteracting bosons the energy at thermal equilibrium can also be expressed as

$$H=\sum_{\vec{\mathrm{k}}_A,\xi}\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)(e^{\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)/kT}-1)^{-1}\,, \quad (2.22)$$

where k is Boltzmann's constant and T is the absolute temperature. Since $kT\gg\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)$ for typical experimental conditions,

$$H=\sum_{\vec{\mathrm{k}}_A,\xi}\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)\left(\frac{kT}{\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)}\right)\,. \quad (2.23)$$

Comparison of Eqs. (2.21) and (2.23) yields

$$|a(\vec{\mathrm{k}}_A,\xi)|^2=kT/\hbar\omega_A(\vec{\mathrm{k}}_A,\xi)\,. \quad (2.24)$$

The displacement-vector amplitude needed in Eq. (2.4) is then given according to the convention in Eq. (2.2) by

$$\vec{\mathrm{u}}(\vec{\mathrm{r}},\omega_A)=2\left(\frac{\hbar}{2\rho V\omega_A(\vec{\mathrm{k}}_A,\xi)}\right)^{1/2}$$

$$\times a(\vec{\mathrm{k}}_A,\xi)\vec{\mathrm{b}}(\vec{\mathrm{a}},\xi)e^{i\vec{\mathrm{k}}_A\cdot\vec{\mathrm{r}}}$$

$$=\left(\frac{2kT}{\rho V\omega_A^2(\vec{\mathrm{k}}_A,\xi)}\right)^{1/2}\vec{\mathrm{b}}(\vec{\mathrm{a}},\xi)e^{i\vec{\mathrm{k}}_A\cdot\vec{\mathrm{r}}}\,, \quad (2.25)$$

omitting an arbitrary phase factor implied by Eq. (2.24).

We now multiply the magnitude of $\vec{\mathrm{S}}^{\varphi}$ from Eq. (2.15) by $r^2\Delta\Omega_r$, where $\Delta\Omega_r$ is the solid angle (inside the medium) of Poynting vectors observed, to obtain the power $P^{\varphi}(\vec{\mathrm{k}}_A)$, in W, scattered from an acoustic wave characterized by a wave vector $\vec{\mathrm{k}}_A$. Using Eqs. (2.4), (2.15), and (2.25), we obtain

$$P^{\varphi}(\vec{\mathrm{k}}_A)=\frac{\epsilon_0\omega_B^2[n^{\varphi}(\vec{\mathrm{s}}_0,\omega_B)]^3(\cos^2\delta^{\varphi})V_s^2\Phi^2\Delta\Omega_r kT}{16\pi^2cV\rho v^2(\vec{\mathrm{a}},\xi)K^{\varphi}}$$

$$\times|\mathcal{E}_i^{\varphi}\chi_{ijkl}^{\mathrm{eff}}E_j(\omega_O)a_lb_k(\vec{\mathrm{a}},\xi)|^2$$

$$\times|\vec{\mathrm{s}}_0(\vec{\mathcal{E}}^{\varphi}\cdot\vec{\mathcal{E}}^{\varphi})-\vec{\mathcal{E}}^{\varphi}(\vec{\mathcal{E}}^{\varphi}\cdot\vec{\mathrm{s}}_0)|\,, \quad (2.26)$$

where χ^{eff} refers to the entire bracketed quantity in Eq. (2.4). Since the normalization condition (2.14) requires the magnitude of $\vec{\mathcal{E}}^{\varphi}$ to be $(n^{\varphi}\cos\delta^{\varphi})^{-1}$, the last factor in Eq. (2.26) is simply $[(n^{\varphi})^2\cos\delta^{\varphi}]^{-1}$. In order to obtain the total scattered power $P_{\mathrm{ins}}^{\varphi}$ inside the medium Eq. (2.26) must be integrated over $V\,d\vec{\mathrm{k}}_A/(2\pi)^3$. In performing this integration, functions of $\vec{\mathrm{k}}_A$ such as $v^2(\vec{\mathrm{a}},\xi)$ and $\vec{\mathrm{b}}(\vec{\mathrm{a}},\xi)$ can be removed from the integrand since they are slowly varying functions of k_A compared to Φ^2. After this removal $\vec{\mathrm{a}}$ should be interpreted as being a unit vector in the direction of $\vec{\mathrm{k}}_B-\vec{\mathrm{k}}_O$. With this approximation and with the use of the integral expression for Φ given in Eq. (2.10) the integral I which must be evaluated is simply

$$I\equiv\frac{V}{(2\pi)^3V_s^2}\iint_{V_s}\int_{V_s}e^{i\Delta\vec{\mathrm{k}}^{\varphi}\cdot(\vec{\mathrm{r}}-\vec{\mathrm{r}}\,')}d\vec{\mathrm{r}}\,d\vec{\mathrm{r}}\,'d\vec{\mathrm{k}}_A=\frac{V}{V_s^2}\int_{V_s}\int_{V_s}\delta(\vec{\mathrm{r}}-\vec{\mathrm{r}}\,')e^{i(\vec{\mathrm{k}}_O-\vec{\mathrm{k}}^{\varphi})\cdot(\vec{\mathrm{r}}-\vec{\mathrm{r}}\,')}d\vec{\mathrm{r}}\,d\vec{\mathrm{r}}\,'=\frac{V}{V_s^2}\int_{V_s}d\vec{\mathrm{r}}=\frac{V}{V_s}\,. \quad (2.27)$$

Thus, we obtain

$$P^{\varphi}_{\text{ins}}=\frac{\epsilon_0\omega_B^2 n^{\varphi}(\vec{s}_0,\ \omega_B)(\cos\delta^{\varphi})V_s\Delta\Omega_r kT}{16\pi^2 c\rho v^2(a,\ \xi)K^{\varphi}}\,\left|\mathcal{E}^{\varphi}_i\chi^{\text{eff}}_{ijkl}E^{\theta}_j(\omega_O)b_k(\vec{a},\ \xi)a_l\right|^2$$

$$=\left(\frac{\omega_B^2V_s\Delta\Omega_r kT}{8\pi^2c^2\rho v^2}\right)\frac{S^{\theta}}{n^{\theta}n^{\varphi}(\cos\delta^{\varphi})(\cos\delta^{\theta})K^{\varphi}}\left|e^{\varphi}_i\chi^{\text{eff}}_{ijkl}e^{\theta}_jb_ka_l\right|^2$$

$$=\left(\frac{\omega_B^2V_s\Delta\Omega_r kT}{32\pi^2c^2\rho v^2}\right)\frac{(n^{\varphi})^3(n^{\theta})^3(\cos\delta^{\theta})(\cos\delta^{\varphi})S^{\theta}}{K^{\varphi}}\left|d^{\varphi}_m p^{\text{eff}}_{mnkl}d^{\theta}_nb_ka_l\right|^2, \tag{2.28}$$

where $\vec{e}^{\,\varphi}$ and $\vec{e}^{\,\theta}$ are unit vectors in the direction of the scattered and incident electric fields, $\vec{d}^{\,\varphi}$ and $\vec{d}^{\,\theta}$ are unit vectors in the direction of the scattered and incident electric displacement fields ($\vec{d}\equiv\overleftrightarrow{\kappa}\cdot\vec{e}/|\overleftrightarrow{\kappa}\cdot\vec{e}|$), and p^{eff} is related to χ^{eff} by

$$p^{\text{eff}}_{ijkl}\equiv-2(\kappa^{-1})_{im}(\kappa^{-1})_{jn}\left(\chi_{mnkl}-\frac{2d_{mnr}a_ra_se_{skl}}{\epsilon_0a_p\kappa_{pq}(\omega_A)a_q}\right)$$

$$=p_{(ij)(kl)}+p_{(ij)[kl]}-\frac{r_{ijr}a_ra_se_{skl}}{\epsilon_0a_p\kappa_{pq}(\omega_A)a_q}\ . \tag{2.29}$$

The intensity (W/m^2) of the incident light beam in the medium is

$$S^{\theta}=\tfrac{1}{2}\epsilon_0cn^{\theta}|E^{\theta}|^2\cos\delta^{\theta}\ . \tag{2.30}$$

Equation (2.28) applies *within* the scattering medium. Previous theories of Brillouin scattering have left the supposedly simple calculation of coupling the scattered radiation from the inside to the outside of the crystal to the experimenter. For many simple commonly used geometries the problem is, in fact, trivial; for general orientations of low-symmetry crystals and general orientations of the exit surface the problem is far from trivial. Besides transmission factors, source demagnification and solid-angle expansion must be considered. Consideration of these effects is complicated in the general case by the noncollinearity of wave vectors and Poynting vectors and by astigmatism introduced by the planar exit surface. Solution of this general coupling problem has recently[4] been obtained. The solid angle expansion is given by

$$\Delta\Omega_r=\left(\frac{d\Omega_r}{d\Omega_k}\right)\left(\frac{d\Omega_k}{d\Omega_D}\right)\Delta\Omega_D$$

$$=\left(\frac{K^{\varphi}(k^{\varphi})^2}{\cos\delta^{\varphi}}\right)\left(\frac{\cos\alpha\cos\delta^{\varphi}}{(n^{\varphi})^2\cos\beta}\right)\Delta\Omega_D\ , \tag{2.31}$$

where α and β are the angles between the Poynting vector and the surface normal outside and inside the surface, respectively, $\Delta\Omega_D$ is the (small) solid angle in free space subtended by the detector, and $d\Omega_k$ is an element of solid angle of the wave vectors inside the medium which corresponds to the element $d\Omega_r$ of solid angle of Poynting vector directions. The scattering volume can be expressed as

$$V_s=Al_s=\frac{Al_D\cos\beta}{\sin\theta_s\cos\alpha}\ , \tag{2.32}$$

where A is the cross-sectional area of the incident light beam inside the medium, l_s is the length of scattering volume observed, l_D is the apparent length of the scattering volume normal to the observation direction seen by the detector (equal to the field-stop diameter if the detector lens system has unit magnification), and θ_S the scattering angle between the incident and scattered Poynting vectors inside the medium. Equation (2.32) is valid when the incident and scattered Poynting vectors and the exit surface normal are coplanar. The incident and scattered powers measured outside the medium, P^{θ} and P^{ϕ}, respectively, are given by

$$P^{\theta}=S^{\theta}A/\mathcal{T}^{\theta}\ , \tag{2.33}$$

$$P^{\varphi}=\mathcal{T}^{\varphi}P^{\varphi}_{\text{ins}}\ , \tag{2.34}$$

where $\mathcal{T}^{\theta}$ and $\mathcal{T}^{\varphi}$ are the transmissivities of the input and output surfaces. Inserting Eqs. (2.31)–(2.34) into Eq. (2.28), we obtain

$$\frac{P^{\varphi}}{P^{\theta}}=\frac{\omega^4kTl_D\Delta\Omega_D(n^{\varphi}n^{\theta})^3\cos\delta^{\varphi}(\cos\delta^{\theta})\mathcal{T}^{\varphi}\ \mathcal{T}^{\theta}}{32\pi^2c^4\rho v^2\sin\theta_s}\left|d^{\varphi}_mp^{\text{eff}}_{mnkl}d^{\theta}_nb_ka_l\right|^2$$

$$=\frac{\omega^4kTl_D\Delta\Omega_D\mathcal{T}^{\varphi}\mathcal{T}^{\theta}}{8\pi^2c^4\rho v^2(\sin\theta_s)n^{\varphi}n^{\theta}\cos\delta^{\varphi}\cos\delta^{\theta}}\left|e^{\varphi}_m\chi^{\text{eff}}_{mnkl}e^{\theta}_nb_ka_l\right|^2\ . \tag{2.35}$$

This is the power in W *outside* the medium scattered into one Brillouin frequency component ω_B into the detector solid angle $\Delta\Omega_D$ in a polarization state φ from an incident light beam of power P^{θ} and polarization state θ by an acoustic mode of type ξ that propagates close to the direction $\vec{k}_B-\vec{k}_O$. Note the reciprocity in Eq. (2.35) between characteristics of the incident and scattered light waves.

Note also that Eq. (2.35) applies to any crystal symmetry, any orientation of the crystal, any scattering angle, and rather general exit surface orientations (specified above).

We have ignored here the finite phonon lifetime and hence the linewidths in frequency of the Brillouin-scattered components. The scattered power in Eq. (2.35) thus refers to the scattered power per unit frequency integrated over the experimental line shape of a particular Brillouin frequency component.

The formulas of this section can be used as follows: (i) Choose the input optical frequency ω_O, the input and output polarization, and propagation directions; (ii) find $\vec{k}_A = k_A \vec{a}$ from Eq. (2.12) and one of the frequency conditions $\omega_B = \omega_O \pm \omega_A$; (iii) find the three acoustic velocities $v(\vec{a}, \xi)$ and their associated displacement eigenvectors $b(\vec{a}, \xi)$ from Eq. (2.17), the latter with a normalization determined by Eq. (2.19); (iv) find the acoustic mode frequencies (Brillouin splittings) by Eq. (2.18); (v) find the electric field unit vector $\vec{e}^{\varphi}$ or the electric displacement field unit vector $\vec{d}^{\varphi}$ for the chosen output propagation direction, and polarization state φ from Eq. (2.13); (vi) find $\vec{e}^{\theta}$ from the known input propagation and polarization directions from the wave equation (2.13); (vii) find the nonzero components of the tensors in χ^{eff} from the point-group symmetry of the scattering medium; and (viii) find the expected scattered power from Eq. (2.35).

We have compared our Eq. (2.35) for the scattered optical power with several other workers' results with boundary coupling added to their work. If we specialize Eq. (2.35) to liquids, we find that we are in agreement with the result of Benedek and Greytak.[19] For cubic crystals we find agreement with Loudon[20] and Chandrasekharan.[21] We agree with Born and Huang[15] provided the additional numerical[22] and dimensional[23] factors previously pointed out by Loudon are included in their equations. We find that our Eq. (2.35) is at variance with the predictions of Benedek and Fritsch[24] on some orientations in cubic crystals. It appears that their Eq. (25) will follow from Eq. (23) only if p_{44} is replaced by $2p_{44}$ in both places it appears in their Eq. (25). The same replacement should also be made in their Eq. (36). We also find our Eq. (2.35) disagrees with Chandrasekharan[21] in birefringent crystals. His treatment of anisotropic crystals appears to confuse the role of electric and electric displacement vectors.

The most thorough previous attempt to discuss Brillouin scattering in an anisotropic crystal is the work of Motulevich.[11] Several remarks should be made concerning her work. First, Motulevich did not concern herself with the surface coupling effects discussed above. Second, she did not distinguish between the solid angle of propagation vectors and the solid angle of Poynting vectors as done in Eq. (2.31). Third, she did not distinguish between the crystal volume and the scattering volume. Fourth, her introduction of the average index of refraction (which appears to the eighth power in his final result) rather than appropriate dielectric tensor components in transforming the photoelastic susceptibility to the Pöckels tensor is a needless approximation. Last, her final result is the component of the Poynting vector along the propagation direction, not the magnitude of the Poynting vector. Cummins and Schoen[25] have recently given expressions for Brillouin scattering in a great variety of simple but useful scattering geometries in crystals of different symmetries. They give a comparison of their formulas with ours.

III. EXPERIMENTAL TECHNIQUE

The light source used in these experiments was an argon-ion laser which produced a linearly polarized output of 0.2 W when operated at 5145 Å. A prism in the laser cavity caused exclusive oscillation on the 5145-Å line. A temperature-controlled etalon in the cavity led to oscillation on a single cavity mode. Stable operation on this mode was maintained by controlling the position of one cavity mirror with a servocontrol mechanism. The error signal used to operate this mechanism was derived from the output laser beam at a frequency at which this cavity mirror was being piezoelectrically oscillated. A half-wave plate in the output beam was used to obtain the orthogonal polarization. A 25-cm focal-length lens focused the beam into the calcite crystal which was at room temperature (23 °C). The calcite crystals used had polished faces perpendicular to the orthogonal crystallographic axes. Two selected schlieren-grade calcite crystals were studied, one of Mexican origin and the other of South African origin, with consistent results. A pressure-scanned Fabry-Perot interferometer which had collection optics with an $f/6.3$ aperture, an operating finesse of 40, and a spacer chosen to give a 69-GHz free spectral range was used to analyze the Brillouin scattered light. A scattering angle of 90° outside the crystal was used in all measurements. The containing vessel of the Fabry-Perot had windows with slightly wedged surfaces and with antireflective coatings. Following a 0.5-mm-diam field stop of the interferometer were a narrow band filter (20 Å between wavelengths of half-transmission), used to discriminate against the Raman scattered light and fluorescence, and a Polaroid polarizer. Initially, the light was detected by a low-dark-current EMI No. 6256S photomultiplier tube whose anode current was amplified and recorded on a strip chart. Later, the light was detected by a Bendix Channeltron phototube (S-11 photocathode) followed by amplification, pulse-height selection, counting, and strip-chart recording.

Absolute values of the photoelastic tensor elements were obtained by comparing the scattered light signal with that from toluene in the same geometry and at the same input laser power. The photoelastic coefficient of toluene is well known[26] ($n^4 p = 1.60$). Since the output polarizations of the light must be perpendicular to the scattering plane for Brillouin scattering in toluene, while an output polarization parallel to the scattering plane is also possible with crystals, we checked that the detection system (apart from the Polaroid polarizer) was insensitive to the state of optical polarization. This showed that the comparison to toluene of both output polarizations of scattered light from calcite was valid.

The phonons responsible for the scattering were identified by their Brillouin splittings calculated from Eq. (2.15) with the refractive index values of Gifford[27] and the elastic constants of Dandekar.[28] The splittings were also calculated using the elastic constants listed in Ref. 29 but the Dandekar values gave better agreement with the observed Brillouin splittings (average absolute deviation 2.6 times smaller). Thus the latter were used in the evaluation of the data, that is, for the calculation of $v(\hat{a}, \xi)$ and $\vec{b}(\hat{a}, \xi)$ appearing in Eq. (2.35). Though it is algebraically complicated in low-symmetry crystals, the Brillouin splittings can be used to determine the elastic constants. Because of the good agreement of the measured and calculated splittings (from Dandekar's values, see Table I), however, this was not done here.

Several effects due to geometric optics in the calcite crystal and the toluene reference cell must be accounted for. First, owing to differing refractive indices the transmission coefficients of the input and output surfaces will be different for the two specimens. Second, when the unscattered laser beam leaves the sample perpendicular to a surface, the reflected beam can produce Brillouin-scattered light which can also enter the detection optics. For the toluene sample and for X-Z and X-Y (incident-scattered) propagation directions in calcite the reflected-beam Brillouin-scattered light will have the same Brillouin splittings as the forward beam has. For Y-Z geometries in calcite the splittings will differ and the effect can be easily explored and a small correction to the data justified. If the input and exit surfaces for the laser beam form a Fabry-Perot interferometer for the unscattered light, these effects will be enlarged. This, however, is easily prevented by a slight tilt of the crystal relative to the laser beam. Third, when the scattered light propagates within a finite solid angle about the optic axis, polarization mixing can occur *along the exit path* of the scattered light because of the finite aperture effect.[30] Strain birefringence can add to this effect. The result of this can be the appearance of forbidden Brillouin scattered lines. These effects can be minimized by minimizing the exit path of the scattered light, that is, by bringing the scattering volume as close to the exit surface as practicable (~ 0.3 mm in our experiments) and limiting the detection solid angle.[30] A correction in the measured intensities can then be made to account for the small residual effect. Fourth, the difference of the indices of refraction of the test sample and the reference scatterer requires a small change in position of the scattered-light-collection optics to compensate for the altered optical path.

The final test of the accuracy of the absolute values of the photoelastic components by the comparative scattering technique is furnished by measuring components known accurately from other work and comparing the results. Previously, we have reported measurements of certain components in rutile[5] which agreed to within 3% of measurements of the same quantities obtained by acousto-optic diffraction experiments. We have also measured the two independent photoelastic components in fused silica (Homosil) and find $|p_{12}| = 0.275$ and $|p_{44}| = 0.078$. These compare favorably in magnitude with the values $p_{12} = +0.270$ and $p_{44} = -0.075$ found by Primak and Post[31] by static-load-induced refractive-index variations and with the values $p_{12} = +0.265$ and $p_{44} = -0.077$ found by Vedam, Schmidt, and Roy[32] by hydrostatic-pressure-induced refractive-index variations.

We wish to emphasize the convenience of the above-described method of determining numerical values for the photoelastic components. The convenience of the method increases for low-symmetry crystals relative to acousto-optic diffraction. This is due to the difficulty of generating the mixed-character acoustic waves in low-symmetry crystals by transducer techniques. These waves are present in a crystal by virtue of its thermal content in sufficient quantity for the Brillouin-scattering technique.

IV. RESULTS AND DISCUSSION

A summary of the experimental orientations studied in calcite is given in Table I. The scattering geometry is given in the first column. The notation $A(bc)D$ means that the incident beam propagated along A and was polarized (electric field direction) along b, and that the scattered light propagated along D and was polarized along c. As can be seen, right-angle scattering was used in all cases. The second column lists the unit displacement vectors $\vec{b}$ found from Eq. (2.17) when the unit propagation vector $\vec{a}$, given in the third column, is used. In the fourth column the calculated value of a constant C, which contains only linear acoustic and optic properties, defined by

$$C = (n^{\varphi} n^{\theta})^3 \cos\delta^{\varphi} \cos\delta^{\theta} / \rho v^2 \quad , \tag{4.1}$$

TABLE I. Summary of Brillouin-scattering measurements. Column 1, scattering geometry; column 2, unit acoustic displacement vector; column 3, unit acoustic propagation vector; column 4, calculated values of C_s [see Eq. (4.1)]; column 5, calculated value of acoustic velocity; column 6, calculated value of Brillouin frequency splitting; column 7, measured value of Brillouin frequency splitting; column 8, measured value of $(FC)_S$ [see Eq. (4.2)]. Modes whose scattered power is indicated with an asterisk were observed but weak. Because of possible feedthrough from the stronger modes in the orthogonal polarization the measured scattered powers were regarded as untrustworthy.

		$\vec{b}$			$\vec{a}$		C_S	v_A	$\Delta\nu_p$	$\Delta\nu_M$	$[FC]_S$
Geometry	x	y	z	x	y	z	(10^{-10} m^2/N)	(km/sec)	(GHz)	(GHz)	(m^2/N)
$\bar{Y}(zz)\bar{X}$	0.437	0.296	−0.849	−0.707	+0.707	0	4.96	2.85	11.7	11.8	3.47×10^{-13}
$\bar{Y}(zz)\bar{X}$	0.545	0.661	0.511	−0.707	+0.707	0	1.976	4.51	18.5	18.9	1.16×10^{-13}
$\bar{Y}(zz)\bar{X}$	0.714	−0.688	0.128	−0.707	+0.707	0	0.728	7.43	30.4	30.5	3.73×10^{-12}
$\bar{Y}(zy)\bar{X}$	0.405	0.273	−0.873	−0.745	+0.667	0	6.39	2.96	12.9	13.0	4.12×10^{-12}
$\bar{Y}(zy)\bar{X}$	0.525	0.712	0.466	−0.745	+0.667	0	2.58	4.41	19.1	⋯	$<5\times10^{-15}$
$\bar{Y}(zy)\bar{X}$	0.748	−0.647	0.145	−0.745	+0.667	0	1.012	7.45	32.3	32.7	9×10^{-15}
$\bar{Y}(xy)\bar{X}$	0.437	0.296	−0.849	−0.707	+0.707	0	9.68	2.85	13.0	13.1	1.53×10^{-13}
$\bar{Y}(xy)\bar{X}$	0.545	0.661	0.511	−0.707	+0.707	0	3.85	4.51	20.6	20.7	2.4×10^{-14}
$\bar{Y}(xy)\bar{X}$	0.714	−0.688	0.128	−0.707	+0.707	0	1.419	7.43	34.0	34.3	2.71×10^{-13}
$\bar{Y}(xz)\bar{X}$	0.462	0.307	−0.832	−0.667	+0.745	0	7.48	2.74	11.9	12.0	2.82×10^{-12}
$\bar{Y}(xz)\bar{X}$	0.574	0.612	0.544	−0.667	+0.745	0	2.65	4.60	20.0	19.9	4.04×10^{-13}
$\bar{Y}(xz)\bar{X}$	0.676	−0.729	0.106	−0.667	+0.745	0	1.018	7.42	32.2	32.3	1.01×10^{-13}
$Y(xx)Z$	0	0.651	0.759	0	−0.707	0.707	6.91	3.37	15.4	15.1	8.98×10^{-13}
$Y(xx)Z$	1	0	0	0	−0.707	0.707	forb.	4.73	⋯	⋯	⋯
$Y(xx)Z$	0	−0.759	0.651	0	−0.707	0.707	2.17	6.01	27.5	27.4	6.19×10^{-12}
$Y(xy)Z$	0	0.651	0.759	0	−0.707	0.707	forb.	3.37	⋯	⋯	⋯
$Y(xy)Z$	1	0	0	0	−0.707	0.707	3.51	4.73	21.6	21.4	1.12×10^{-13}
$Y(xy)Z$	0	−0.759	0.651	0	−0.707	0.707	forb.	6.01	⋯	⋯	⋯
$Y(zx)Z$	0	0.702	0.712	0	−0.667	0.745	forb.	3.25	⋯	⋯	⋯
$Y(zx)Z$	1	0	0	0	−0.667	0.745	2.55	4.70	20.4	20.1	5.7×10^{-14}
$Y(zx)Z$	0	−0.712	0.702	0	−0.667	0.745	forb.	5.97	⋯	⋯	⋯
$Y(zy)Z$	0	0.702	0.712	0	−0.667	0.745	5.33	3.25	14.1	13.9	5.15×10^{-13}
$Y(zy)Z$	1	0	0	0	−0.667	0.745	forb.	4.70	⋯	⋯	⋯
$Y(zy)Z$	0	−0.712	0.702	0	−0.667	0.745	1.574	5.97	25.9	26.0	7.22×10^{-13}
$\bar{Y}(xx)Z$	1	0	0	0	0.707	0.707	forb.	2.62	⋯	⋯	⋯
$\bar{Y}(xx)Z$	0	−0.532	0.847	0	0.707	0.707	8.44	3.05	13.9	14.2	1.85×10^{-12}
$\bar{Y}(xx)Z$	0	0.847	0.532	0	0.707	0.707	1.462	7.33	33.5	33.6	3.07×10^{-12}
$\bar{Y}(xy)Z$	1	0	0	0	0.707	0.707	11.46	2.62	12.0	12.1	1.66×10^{-12}
$\bar{Y}(xy)Z$	0	−0.532	0.847	0	0.707	0.707	forb.	3.05	⋯	⋯	⋯
$\bar{Y}(xy)Z$	0	0.847	0.532	0	0.707	0.707	forb.	7.33	⋯	⋯	⋯
$\bar{Y}(zx)Z$	1	0	0	0	0.667	0.745	8.48	2.57	11.2	11.3	9.03×10^{-13}
$\bar{Y}(zx)Z$	0	−0.559	0.829	0	0.667	0.745	forb.	3.14	⋯	⋯	⋯
$\bar{Y}(zx)Z$	0	0.829	0.559	0	0.667	0.745	forb.	7.20	⋯	⋯	⋯
$\bar{Y}(zy)Z$	1	0	0	0	0.667	0.745	forb.	2.57	⋯	⋯	⋯
$\bar{Y}(zy)Z$	0	−0.559	0.829	0	0.667	0.745	1.084	3.14	13.6	13.8	2.67×10^{-12}
$\bar{Y}(zy)Z$	0	0.829	0.559	0	0.667	0.745	5.71	7.20	31.2	31.4	7.0×10^{-14}
$\bar{Z}(xy)\bar{X}$	0.602	−0.440	0.667	−0.707	0	0.707	7.96	3.14	14.4	14.6	1.39×10^{-13}
$\bar{Z}(xy)\bar{X}$	0.071	0.861	0.504	−0.707	0	0.707	6.05	3.60	16.5	16.6	5.42×10^{-13}
$\bar{Z}(xy)\bar{X}$	0.795	0.256	−0.549	−0.707	0	0.707	1.670	6.85	31.4	31.4	*
$\bar{Z}(yy)\bar{X}$	0.602	−0.440	0.667	−0.707	0	0.707	7.96	3.14	14.4	14.6	6.2×10^{-13}
$\bar{Z}(yy)\bar{X}$	0.071	0.861	0.504	−0.707	0	0.707	6.05	3.60	16.5	16.6	2.59×10^{-12}
$\bar{Z}(yy)\bar{X}$	0.795	0.256	−0.549	−0.707	0	0.707	1.670	6.85	31.4	31.4	4.40×10^{-12}
$\bar{Z}(xz)\bar{X}$	0.615	−0.531	0.583	−0.667	0	0.745	6.12	3.03	13.1	13.2	3.81×10^{-13}
$\bar{Z}(xz)\bar{X}$	0.161	0.809	0.566	−0.667	0	0.745	4.18	3.67	15.9	16.1	1.31×10^{-12}
$\bar{Z}(xz)\bar{X}$	0.772	0.254	−0.583	−0.667	0	0.745	1.229	6.75	29.3	29.3	2.31×10^{-13}
$\bar{Z}(yz)\bar{X}$	0.615	−0.531	0.583	−0.667	0	0.745	6.12	3.03	13.1	13.2	2.51×10^{-13}
$\bar{Z}(yz)\bar{X}$	0.161	0.809	0.566	−0.667	0	0.745	4.18	3.67	15.9	16.1	*
$\bar{Z}(yz)\bar{X}$	0.772	0.254	−0.583	−0.667	0	0.745	1.229	6.75	29.3	29.3	8.4×10^{-14}
Special	1	0	0	0	−0.247	−0.969	forb.	2.94	⋯	⋯	⋯
Special	0	−0.892	0.452	0	−0.247	−0.969	3.93	3.77	16.6	16.8	2.05×10^{-13}
Special	0	0.452	0.892	0	−0.247	−0.969	1.619	5.88	25.8	26.1	1.53×10^{-13}

is given. The fifth column gives the acoustic velocity calculated from Dandekar's elastic constants of calcite.[28] The resulting predicted Brillouin splittings $\Delta\nu_P$ in GHz are listed in the sixth column. The corresponding measured values $\Delta\nu_M$ are listed in the seventh column. The agreement between calculated and measured values (2% or better in all cases) is adjudged to be good. The eighth column lists the experimental values of $(CF)_S$ for calcite found in these experiments. F is defined by

$$F = \left| d^{\varphi}_{m} p^{\text{eff}}_{mnkl} d^{\theta}_{n} b_k a_l \right|^2 . \qquad (4.2)$$

In obtaining the values in the eighth column the longitudinal sound velocity in toluene was taken as 1.344 ± 0.001 km/sec (measured from the Brillouin splitting), the density as 866 kg/m³, the index of refraction as 1.504, and the photoelastic coefficient as 0.313.[26] These values yield $(CF\mathcal{T}_1\mathcal{T}_2)_R = 0.669\times10^{-9}$ m²/N for toluene.

It can be seen from Table I that only one allowed mode was not observed [expected at 19.1 GHz in the $Y(zy)X$ geometry]. The upper limit for the strength of this mode is sufficiently small to make the upper limit as useful as an actual measurement of it in determining the photoelastic components. Note that we have measured 36 modes from which we determine nine photoelastic tensor elements (eight symmetric, one antisymmetric). Note that all the elements except p_{33} can be obtained in calcite by 90° scattering with the incident and scattered light traveling along axes of the orthogonal crystallographic coordinate system. The one special geometry listed was used to measure p_{33}. For that geometry the incident and scattered polarizations lay in the scattering plane (extraordinary rays); the unit incident propagation vector inside the medium was [0, 0.866, 0.5] and the unit scattered propagation vector inside the medium was [0, 0.471, − 0.882].

As discussed in Sec. III, small corrections were made to the measured intensities to account for reflections of light inside the calcite crystal and the toluene reference cell. Other small corrections to account for strain birefringence and the finite aperture effect[30] were made for the YZ and $\bar{Y}Z$ geometries, also as discussed in Sec. III. The effect of strain birefringence and finite aperture is easy to evaluate in these orientations because each scattering mode is allowed in only one output polarization state as seen from Table I. For XZ geometries, however, the same scattering mode (in frequency) is allowed in either output polarization state. This makes evaluation of strain birefringence and finite aperture corrections less certain for these geometries. If ZX geometries are used, the finite aperture effect is reduced because of the small angular aperture of the incident laser beam. Depolarization from strain birefringence can be minimized by observing scattering from a volume close to the input surface in this case.

The photoelastic components for calcite were determined from a least-squares analysis by computer from the values of Table I using Eq. (4.2). The resulting values are listed in the sixth column of Table II. The corresponding photoelastic-stress coefficients Π_{ij} calculated from the symmetric part of our photoelastic-strain coefficients p_{ij} and Dandekar's elastic constants are listed in the seventh column. There cannot be a photoelastic-stress coefficient corresponding to the antisymmetric photoelastic-strain coefficient.

Several points should be made about our measured

TABLE II. Summary of photoelastic-strain tensor components p_{ij} and photoelastic-stress tensor components Π_{ij} for calcite from Pöckels's work, from Pöckels's work recalculated with Dandekar's elastic constants, and from the present work. [For reasons unspecified, Ref. 29, p. 134 quotes Pöckels's Π_{ij} values 2% higher than Pöckels did and lists $\Pi_{44} = +0.35$ rather than -3.4 (in units of 10^{-13} cm²/dyn ≡ 10^{-12} m²/N).]

Subscripts ij	Π_{ij} (Pöckels) (10^{-12} m²/N)	Π_{ij} (Pöckels recalc.) (10^{-12} m²/N)	p_{ij} (Pöckels)	p_{ij} (Pöckels recalc.)	p_{ij} (our meas.)	Π_{ij} (our meas.) (10^{-12} m²/N)
11 or 22	−0.60	−0.70	+0.095	+0.106	+0.062	−0.85
33	+0.44	+0.39	+0.178	+0.188	+0.139	+0.18
12 or 21	+0.90	+0.80	+0.189	+0.191	+0.147	+0.65
13 or 23	+2.48	+2.54	+0.215	+0.223	+0.186	+2.30
31 or 32	+1.55	+1.42	+0.309	+0.310	+0.241	+1.14
14 or (−)24 or 65	−1.09	−1.31	−0.006	−0.012	−0.011	−1.29
41 or (−)42 or 56	−0.66	−0.65	+0.010	+0.0007	−0.036	−1.10
44 or 55	−3.4	−2.9	−0.090	−0.069	−0.058	−3.18
(23)[23] or (13)[13]	⋯	⋯	⋯	⋯	+0.047	⋯

p_{ij} values: (i) The measured magnitude of the antisymmetric component

$$p_{(23)[23]} = p_{(13)[13]} = +0.047$$

agrees within 5% with the value +0.0450 calculated in Eq. (1.5). (ii) On the basis of this agreement in magnitude we feel justified in assigning the plus sign to the measured value just quoted. (iii) On the basis of this theoretically determined sign the algebraic sign of each of the other photoelastic tensor elements is determined from our Brillouin-scattering measurements alone. (iv) Combining the antisymmetric and symmetric parts we obtain

$$p_{(23)23} = p_{(13)13} = -0.011 \quad , \tag{4.3}$$

$$p_{(23)32} = p_{(13)31} = -0.105 \quad . \tag{4.4}$$

We see that these components, which for years were believed equal, differ by a factor of about 10. Thus the rotational effect in calcite is almost as dramatic as it was found to be in rutile,[5] where these components differed by a factor of about 30. (v) The 5% agreement on the magnitude of the antisymmetric component indicates the accuracy that the other measured component values are expected to have. (vi) Our measured values of the larger p_{ij} components are ~ 30% smaller than the values measured by Pöckels[6] by static-load-induced refractive-index-change measurements quoted in column 4 of Table II. Pöckels used Voigt's measurements[33] of the elastic constants of calcite in his data analysis. Since Voigt's measurements differ somewhat from modern measurements, such as Dandekar's,[28] we have recalculated photoelastic-stress components Π_{ij} and photoelastic-strain components p_{ij} from Pöckels's data[6] using Dandekar's elastic constants. These recalculated values are listed in the third and fifth columns of Table II. The elastic constants enter Pöckels's analysis in two ways: (a) The phase-change term arising from sample width changes is proportional to the elastic *compliances* and must be *subtracted* from the measured phase change. (b) The resulting photoelastic-stress components must be *multiplied* by the elastic *stiffnesses* to obtain the photoelastic-strain components. The recalculated Pöckels's p_{ij} values are in no better agreement with our values than were his originally quoted values. We thus can offer no explanation of this discrepancy. We are, however, sure that p_{41} must be of opposite sign and larger in magnitude than Pöckels's value. His measurements were very insensitive to this small coefficient.

In addition to Pöckels several other workers have studied aspects of the photoelastic effect in calcite. Recently, Kachalov[34] studied Brillouin scattering in calcite with a view to verifying our prediction[1] of an antisymmetric part of the photoelastic tensor. His procedure was to assume Pöckels's value of p_{44}, calculate $p_{(23)[23]}$ by Eq. (1.4) above, and then calculate the relative scattering strengths expected for various geometries. The values thus used by Kachalov ($p_{(23)23} = -0.045$ and $p_{(23)32} = -0.135$) are at considerable variance with those found in our work given in Eqs. (4.3) and (4.4). Kachalov finds that his experimental intensities are in considerably closer agreement with those calculated on the above basis than with those omitting the antisymmetric part of the photoelastic tensor altogether. Kachalov has measured intensities on a *relative* basis for only three scattering modes that involve $p_{(23)23}$ and $p_{(23)32}$ as well as p_{41}. Thus it is not possible to *derive* values of these components from his measurements.

Two other Brillouin-scattering studies in calcite have been performed. One[35] had the objective of observing a change in the Brillouin splitting depending on the state of polarization of the incident and scattered light. This effect, first predicted by Vladimirskii[36] and later by Chandrasekharan,[12] results from the wave-vector conservation (phase-matching) condition in the presence of birefringence. The other study[37] was aimed at measuring the anisotropy of the scattered intensity as a means of verifying the predictions of Motulevich's theory[11] of Brillouin scattering in anisotropic media.

Davis and Vedam[38] have studied the changes in the ordinary and extraordinary refractive indices of calcite under hydrostatic pressures up to 7 kbar. They point out that the phase changes induced by such large hydrostatic pressures can be measured much more accurately than the smaller phase changes to which one is limited when employing uniaxial pressures. However, to obtain the change in refractive index per unit pressure from the Davis–Vedam measurements, the dimensional changes of the crystal must be accounted for. Davis and Vedam *calculate* this; the calculated magnitudes for either polarization account for approximately 90% of their quoted result. The accuracy of the result is thus 90% dependent on the accuracy of the compressibility numbers used for the calculation. The compressibility numbers used by Davis and Vedam were measured by Bridgman[39] and are believed to be very accurate. The best and most recent set of elastic constants, Dandekar's, which we find give excellent agreement with our Brillouin splittings, agree *exactly* with Bridgman's compressibility data. Thus, we conclude that the Davis–Vedam values should be very trustworthy. They measure refractive index changes of 0.50×10^{-3}/kbar and 0.44×10^{-3}/kbar for the ordinary and extraordinary indices, respectively (see Table III). These quantities are 0.57×10^{-3}/kbar and 0.52×10^{-3}/kbar if Pöckels's original p_{ij} values and Bridgman's compressibility data are

TABLE III. Comparison of various measurements of the change of ordinary Δn_o and extraordinary Δn_e refractive indices per unit of hydrostatic pressure P.

	Pöckels–Bridgman	Recalc. Pöckels–Dandekar	Davis–Vedam	Our values
$\Delta n_o/P$ (10^{-3}/kbar)	0.57	0.60	0.50	0.48
$\Delta n_e/P$ (10^{-3}/kbar)	0.52	0.53	0.44	0.40

used for calculation. Davis and Vedam are of the opinion[38] that the discrepancy between their data and the calculated Pöckels–Bridgman values would disappear if Pöckels's values had been deduced with modern elastic constants. Using the recalculated Pöckels values of Table II (found using Dandekar's elastic constants) however, we find that the calculated Pöckels–Dandekar (Bridgman) values are 0.60×10^{-3}/kbar and 0.53×10^{-3}/kbar for the ordinary and extraordinary indices, respectively, thus increasing the discrepancy with the Davis–Vedam results. Thus we conclude that the Davis–Vedam values differ significantly from Pöckels's values, recalculated or not. Values calculated from our data differ from Pöckels's values also; our values are 0.48×10^{-3}/kbar and 0.40×10^{-3}/kbar, respectively. If we take the 5% agreement with the theoretical antisymmetric part of the photoelastic tensor in calcite as well as rutile[5] as being indicative of the accuracy of any of our p_{ij} measurements, then the former value is in agreement with Davis and Vedam while the latter is not.

Narasimhamurty[40] measured the *ratios* of several photoelastic tensor elements in calcite and believed his agreement with Pöckels to be fairly good. He apparently calculated two of the three ratios from Pöckels's values wrongly. Corrected values for these lessen the agreement markedly. His ratios are in no closer agreement with ratios obtained from our data.

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