Verification of expressions for Raman and Brillouin scattering in anisotropic media and effects on previously measured elasto-optic constants of rutile

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A simple experiment is proposed and carried out to verify a prediction of light scattering in birefringent media derived by Neslon, Lazay, and Lax. In the light of this theory, previous measurements of the elasto-optic constants of rutile are reviewed leading to a disappearance of the claimed $p_{ijkl} \neq p_{jikl}$, which has been recently challenged by Nelson and Lazay.

I. INTRODUCTION

Although the theory of Brillouin scattering in optically isotropic materials is well known[1] its generalization to anisotropic media is by no means straightforward. In 1972 Nelson, Lazay, and Lax[2] derived a general expression for scattering in a birefringent medium, which differs from all previous derivations,[3–7] and applied it to Brillouin scattering in calcite. The expression derived by the above authors is valid for general propagation directions. Unfortunately the important differences which exist even for scattering geometries of high symmetry have been overlooked.[8,9]

In this paper we propose and carry out a simple experiment to verify an important prediction of the scattering expression from Ref. 2. It is possible that this verification is implicit in the application of the equation to Brillouin scattering in calcite.[2] However, because of the widespread use of Brillouin scattering techniques, we feel that an explicit verification of one of the predictions of the theory of Ref. 2 is worthwhile.

In the light of the above mentioned theory, the inequality of the elasto-optic components $p_{ijkl} \neq p_{jikl}$ claimed for rutile in Ref. 8, and recently challenged by Nelson and Lazay,[10] can be dismissed.

II. THEORETICAL PREDICTIONS

The result for the ratio of the incident (Θ) and scattered (ϕ) powers (P) as derived in Ref. 2 and valid outside the medium, is given by

$$\frac{P^{\phi}}{P^{\Theta}} = \frac{\omega^4 k T l_D \Delta\Omega_D \tau^{\phi}\tau^{\Theta}}{8\pi^2 c^4 \rho v^2 (\sin\Theta_s) n^{\phi} n^{\Theta} \cos\delta^{\phi} \cos\delta^{\Theta}} \times |e_m^{\phi} \chi_{mnkl}^{\mathrm{eff}} e_n^{\Theta} b_k a_l|^2 , \quad (1)$$

where ω is the frequency of the scattered light, k Boltzmann's constant, T the temperature, l_D is the apparent length of the scattering volume normal to the observation direction seen by the detector, $\Delta\Omega_D$ is the solid angle subtended by the detector, τ are transmission coefficients at the surface, c is the velocity of light, ρ the density, v the sound velocity, Θ_s the scattering angle, n the refractive index, δ the angle between the propagation direction and Poynting's vector, $\hat{e}$ a unit vector parallel to the electric field of the radiation, and χ is the change in polarizability induced by a phonon traveling along $\hat{a}$ and polarized along $\hat{b}$.

We now rewrite Eq. (1) for the case of 90° scattering with incident and scattered directions along principal axes of the dielectric tensor. In this case $\sin\Theta_s = 1$ and $\delta^{\phi} = \delta^{\Theta} = 0$. It has recently been shown[10] that the transmission factors *should* be set equal to approximately unity for certain scattering geometries. This being the case for the geometry to be discussed, we shall omit the factors τ. For the above conditions Eq. (1) can be written

$$\frac{P^{\phi}}{P^{\Theta}} = \frac{\omega^4 k T l_D \Delta\Omega_D}{8\pi^2 c^4 \rho v^2 n^{\phi} n^{\Theta}} |e_m^{\phi} \chi_{mnkl} e_n^{\Theta} b_k a_l|^2 = K \frac{\Delta\Omega_D}{n^{\phi} n^{\Theta}} |\hat{e}^{\phi} \cdot \underline{T} \cdot \hat{e}^{\Theta}|^2 . \quad (2)$$

The tensors $\underline{T}$ are defined in Ref. 8 and the second form of Eq. (2) applies also to the case of Raman scattering with the appropriate changes in K and $\underline{T}$.

Assuming that the scattered light leaves the sample normal to the surface, the solid angle subtended by the detector can be written [using Eq. (2.32) Ref. 2] as

$$\Delta\Omega_D = (n^{\phi})^2 \Delta\Omega , \quad (3)$$

where $\Delta\Omega$ is the solid angle inside the medium; with

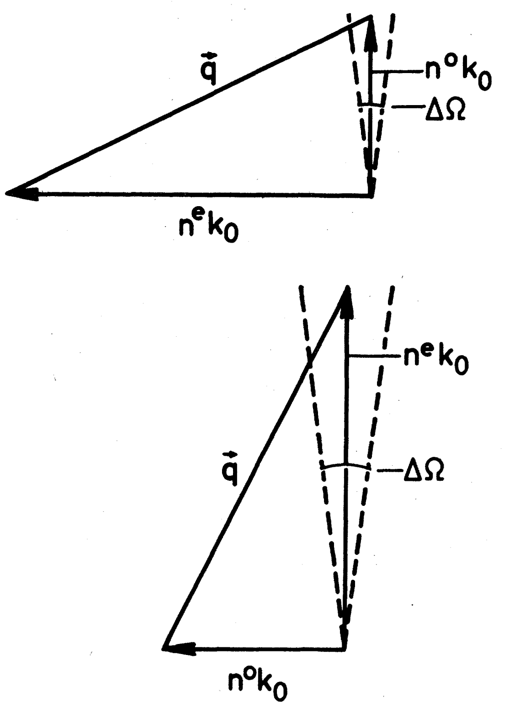

FIG. 1. Schematic diagram illustrating the origin of the inequality of the intensities of Raman or Brillouin scattering in an anisotropic medium.

our previous assumptions the solid angle of Poynting vectors and wave vectors are equal.

We consider now the case of rutile. The scattering geometry considered is $\vec{k}^{\theta} \parallel [110]$ and $\vec{k}^{\phi} \parallel [1\bar{1}0]$ and we observe the Raman E_g mode at 447 cm^{-1}. The scattering tensors for this mode are

$$\underline{\Pi}_1 = \begin{pmatrix} 0 & 0 & d \\ 0 & 0 & 0 \\ d & 0 & 0 \end{pmatrix}, \quad \underline{\Pi}_2 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & d \\ 0 & d & 0 \end{pmatrix}. \tag{4}$$

Denoting the [001] axis as vertical (V) we refer all polarizations to the horizontal (H) scattering plane. Superscripts and subscripts refer to scattered and incident polarizations. From Eqs. (2) and (4)

$${}^{H}_{V}P^{\phi}\,(\text{outside}) = {}^{V}_{H}P^{\phi}\,(\text{outside})\,, \tag{5}$$

and using Eq. (3)

$${}^{H}_{V}P^{\phi}\,(\text{inside}) = \left(\frac{n^o}{n^e}\right)^2 {}^{V}_{H}P^{\phi}\,(\text{inside})\,, \tag{6}$$

where n^o and n^e are the ordinary and extraordinary refractive indices, respectively. Previous derivations of Raman scattering predict that ${}^{H}_{V}P^{\phi}$ (inside) and ${}^{V}_{H}P^{\phi}$ (inside) must be equal.

A simple illustration of the effect leading to Eq. (6) is shown in Fig. 1. For a given solid angle the number of phonon states contributing in a given direction is proportional to $|k_{\text{scatt}}|^2 = |nK_0|^2$ leading directly to Eq. (6)

If instead of Eq. (5) the older theories are used, the ratio of intensities outside the crystal should be $(n_e/n_o)^2$. For rutile this ratio at $\lambda = 5145$ Å is 1.25, easily detected experimentally.

III. EXPERIMENT

The experiment outlined in the previous section was carried out using 5145 and 4579 Å radiation from an Ar^+ ion laser to observe the 447-cm^{-1} Raman line of rutile. Within the experimental error of ~6% the intensities measured (outside the crystal) were found to be equal, confirming the predictions of Ref. 2.

We also repeated[8] intensity measurements of the transverse Brillouin component observed in this scattering geometry and obtained identical results as in the previous paragraph. The use of Eq. (2) for the case of Brillouin scattering is not as straightforward

TABLE I. Elasto-optic constants of rutile recalculated from the results of Ref. 8 taking into account the correction factors described in the text.

	λ (Å)			
p_{ij}	6328	5145	4880	4579
p_{12}	+0.143 ± 0.010	+0.111 ± 0.009	+0.098 ± 0.008	+0.076 ± 0.006
p_{13}	−0.139 ± 0.009	−0.165 ± 0.013	−0.180 ± 0.014	−0.195 ± 0.015
p_{33}	−0.057 ± 0.009	−0.063 ± 0.010	−0.069 ± 0.011	−0.079 ± 0.011
p_{31}	−0.080 ± 0.008	−0.093 ± 0.008	−0.098 ± 0.008	−0.104 ± 0.009
p_{66}	−0.060 ± 0.005	−0.063 ± 0.004	−0.065 ± 0.006	−0.068 ± 0.006
p_{11}	+0.017 ± 0.015	+0.002 ± 0.018	+0.000 ± 0.021	−0.006 ± 0.034
$p_{4\bar{4}}$	+0.020 ± 0.003	+0.023 ± 0.001	+0.025 ± 0.001	+0.027 ± 0.002
p_{44}	−0.009 ± 0.003	−0.004 ± 0.001	−0.002 ± 0.001	+0.004 ± 0.002

as for Raman scattering because the scattering tensor $\underline{T}$ is in principle different for each phonon propagation direction (see Fig. 1). For the particular geometry chosen, however, the phonons are equivalent by symmetry and hence have the same scattering tensor.

In light of the above we have reanalyzed our previous results for the elasto-optic constants of rutile.[8] We find that we can no longer claim the inequality $p_{44} \neq p_{\bar{4}4}$ recently contested in Ref. 10. In Table I we give our recalculated values of p_{ij}'s.

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