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Determination of Birefringence by Brillouin Spectroscopy

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We have combined Brillouin scattering experiments and fundamental optics theory to determine the birefringence of single crystals. Double refraction in anisotropic materials give rise to doublet peaks in Brillouin spectra. With the incident plane orthogonal to the optic axis, the birefringence of the materials can be determined from one spectrum. We present the Brillouin spectra to confirm this fact for a single crystal $PbMoO_4$.

I. INTRODUCTION

When light is incident from air on the plane surface of an optically anisotropic medium, there are two transmitted waves, so-called double refraction. The two transmitted waves propagate in two different directions in the medium with different phase velocities, governed by the principal refractive indices, n_x, n_y, n_z of the medium. There are many methods to determine these indices: from measurements of the angle of deviation, from total reflection in a prism, or by immersion in a series of liquids of graded refractive index. In this paper, we present another method, which uses Brillouin scattering. Brillouin scattering is the inelastic scattering of light by thermally excited acoustic phonons [1] and has been a well-known technique for determining the elastic properties of various materials, from single crystals to thin films of metals, semiconductors, or organics.

The two transmitted waves produced by double refraction can be scattered by acoustic phonons and the two signals in the Brillouin spectra can be separated because of the differences both in indices and in acoustic properties. According to the detailed calculations described below, if the incident plane is orthogonal to the optic axis and the incident light is S-polarized, it is possible to determine the differences between the two principal indices directly. Therefore, it is our objective to introduce Brillouin scattering as a technique to determine the birefringence of single crystals. The ordinary index of refraction for uniaxial crystals usually can be determined, for example by Brewster angle measurement. Then, the extraordinary index can be calculated from the birefringence.

II. THEORETICAL BACKGROUND

In Brillouin scattering (BS), there are two measurable quantities, the acoustic phonon wavevector q determined from the scattering geometry and the angular frequency of the phonon $\Omega(=2\pi\nu)$ measured by a high contrast interferometer. The scattered light is shifted in frequency due to the Doppler effect by as much as the acoustic phonon's frequency. Interferometers are well established for measuring the frequency shift [2]. In 180° backscattering geometry, the direction of the acoustic phonon wavevector is parallel to that of the transmitted wave and the magnitude of the wavevector is given by

$$q = \frac{4\pi n}{\lambda} \tag{1}$$

where λ is the wavelength of the light in air and n is the index of refraction for the light. Then the sound velocity of the phonon can be determined by

$$v = \frac{\Omega}{q} \tag{2}$$

and can be calculated from the frequency shift ν of the signal in the spectrum if the index of refraction is known.

The sound velocity is also determined by the elastic properties of the medium. For a homogeneous, nonpiezoelectric material without external forces, the sound velocity in the xy-plane of tetragonal symmetry is determined by [3]

$$v_L = \sqrt{\frac{c_{11} + c_{66} + \sqrt{(c_{11} + c_{66})^2 - 4C}}{2\rho}} \tag{3}$$

for the quasi-longitudinal wave,

$$v_{T1} = \sqrt{\frac{c_{11} + c_{66} - \sqrt{(c_{11} + c_{66})^2 - 4C}}{2\rho}} \tag{4}$$

for the quasi-transverse wave, and

$$v_{T2} = \sqrt{\frac{c_{44}}{\rho}} \tag{5}$$

for the pure-transverse wave, where

$$\begin{aligned} C = & \ (c_{11}\cos^2\phi + c_{66}\sin^2\phi + c_{16}\sin 2\phi) \\ & \times (c_{11}\sin^2\phi + c_{66}\cos^2\phi - c_{16}\sin 2\phi) \\ & -(c_{16}\cos 2\phi + (c_{12}+c_{66})\sin\phi\cos\phi)^2 \end{aligned} \tag{6}$$

and ϕ is the angle from the x-axis. The c_{ij} are the elastic constants and ρ is the density of the medium. If the elastic properties are known a priori, in principle, the index of refraction can be deduced. However, in typical cases the elastic properties determined by several independent coefficients as shown above have to be determined. Thus, the index of refraction should be known and cannot be directly determined by BS. Nonetheless, we have found a way to determine the difference between the principal indices, birefringence, $n_e - n_o$.

Since the sound velocity varies much more slowly than the index of refraction for the two refracted waves, the sound velocity of the acoustic phonon can be assumed to be constant for the direction of propagation (we will check the validity of this assumption). The two transmitted waves see two different indices of refraction and the wavevectors of the phonon probed by BS have two different values according to Eq.(1). Then Eq.(2) shows that there is a difference in frequency shift between the two Brillouin signals, which is a direct indication of difference between the two indices of refraction. Furthermore, if there is a scattering geometry which gives the two principle index of refraction, we can directly measure the birefringence, $n_o - n_e$, by BS.

In a uniaxial medium, the two indices of refraction n_1 and n_2 are given by

$$n_1^2 = n_o^2 \tag{7}$$

$$\frac{1}{n_2^2} = \frac{1}{n_o^2}\cos^2\theta + \frac{1}{n_e^2}\sin^2\theta \tag{8}$$

where θ is the angle between the wavevector of the transmitted wave and the optic axis of the medium and n_o and n_e are the ordinary and extraordinary indices of refraction, respectively. When the optic axis is perpendicular to the surface, the angle θ is the same as the angle of refraction. Since the refracting angles θ_1 and θ_2 of the two transmitted wave are related with the angle of incidence θ_i as

$$\sin\theta_i = n_1 \sin\theta_1 = n_2 \sin\theta_2 \tag{9}$$

the angle of refraction θ_1 can be determined easily whereas the refracting angle θ_2 can be obtained by combining Eqs.(8) and (9). After some algebra, the corresponding index of refraction can be rewritten in terms of the angle of incidence as

$$n_2^2 = n_o^2 + \left(1 - \frac{n_o^2}{n_e^2}\right)\sin^2\theta_i. \tag{10}$$

Since the index is known, the refracting angle θ_2 can be obtained by the Snell's law, Eq.(9).

If the optic axis lies in the surface, it is necessary to specify the refracting direction with respect to the optic axis. Thus we use (abc)-rectilinear coordinates for the crystal with respect to the laboratory (xyz)-coordinates, as shown in Fig. 1 and the c-axis is assumed to be along the optic axis. Furthermore, we assume that the surface normal is the a-axis and the scattering plane is rotated by an angle of ϕ from the b-axis. Thus, the wavevector of the transmitted wave is given as

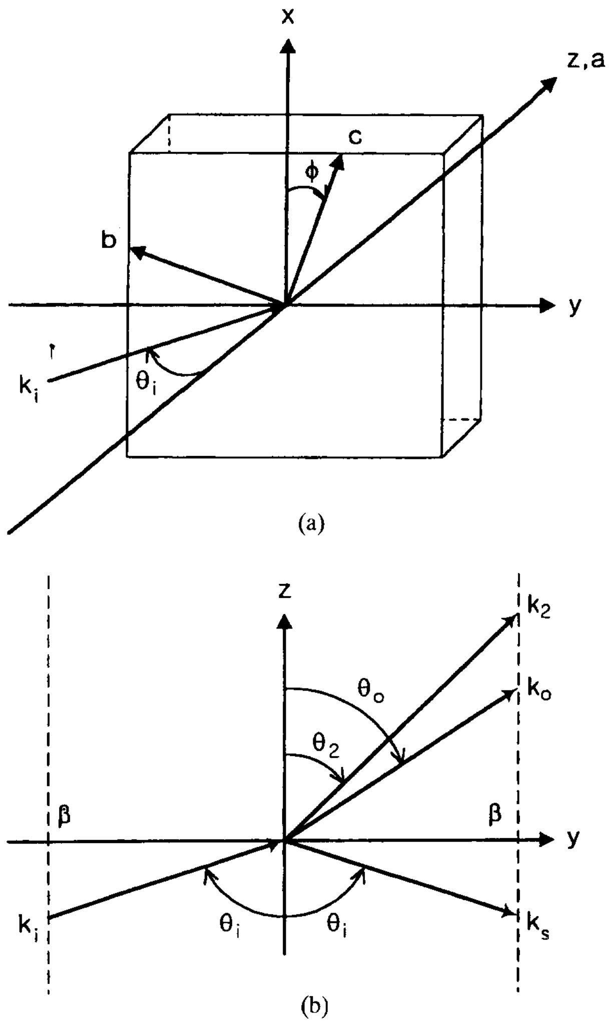

FIG. 1. (a) (xyz)- and (abc)- rectilinear coordinate system. Incident wavevector k lies in yz-plane and the c-axis of crystal lies in xy plane; (b) reflection and refraction at the crystal surface.

$$\mathbf{k_2} = \cos\theta_r\hat{a} - \sin\theta_r\cos\phi\hat{b} + \sin\theta_r\sin\phi\hat{c} \tag{11}$$

where θ_r is the angle of refraction as shown in Fig. 1. Since the angle between the wavevector of the transmitted wave and the optic axis is given by $\cos\theta = \mathbf{k_2}\cdot\hat{c}$, Eq.(8) for the transmitted wave can be rewritten as

$$\frac{1}{n_2^2} = \frac{\sin^2\theta_r\sin^2\phi}{n_o^2} + \frac{1-\sin^2\theta_r\sin^2\phi}{n_e^2} \tag{12}$$

whereas Eq.(7) can still be applied for the other. The angle of refraction θ_r then is determined by Snell's law with the angle of incidence θ_i as

$$n_2 = \frac{\sin\theta_i}{\sin\theta_r}. \tag{13}$$

Having substituted Eq.(13) into Eq.(12), we can rewrite the index of refraction in terms of scattering parameters, after some algebra, as

$$n_2^2 = n_e^2 + \left(1 - \frac{n_e^2}{n_o^2}\right)\sin^2\theta_i\sin^2\phi. \tag{14}$$

Since we can determine the value of index from the scattering parameters only, Eq.(14) is more useful than Eq.(12). Once the value of index n_2 is determined, the angle of refraction can be calculated by Eq.(13), as well.

The combination of either of Eq.(10) or (14) and Eq.(7) shows that one of the two transmitted waves always has the same index of refraction while the other varies as the scattering parameters change. In particular, when $\phi = 0°$, i. e. the scattering plane is perpendicular to the optic axis, the second term of Eq.(14) vanishes and we get

$$n_2 = n_e \tag{15}$$

which indicates that the two possible indices of refraction, no matter what the angle of incidence is, are the two principal indices of refraction. That is, the transmitted wave for P-polarized incident light is governed by the ordinary index of refraction, while that for S-polarization is governed by the extraordinary index. The two polarizations are the two eigen-polarizations. If incident light is in between S- and P-polarizations, the incident light can be considered to be a sum of two eigen-polarizations. The magnitude of each polarization is not of concern in this paper, so it will be neglected. Rather, it is important to note that there are generally two transmitted waves, and the two indices of refraction are n_o and n_e under the condition that the scattering plane is perpendicular to the optic axis and that the polarization of the incident light is in between S- and P-polarizations. Furthermore, it should be noted that this scattering plane is just the xy-plane where sound velocities can be determined analytically using Eqs. (3)-(5).

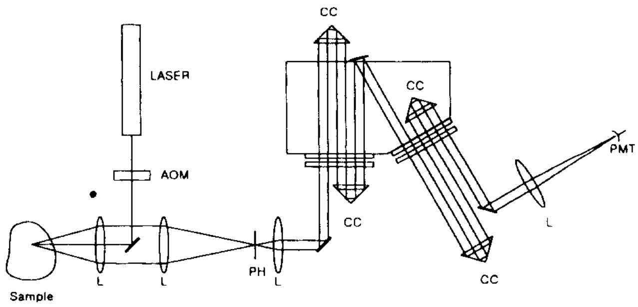

FIG. 2. Schematic diagram of Brillouin scattering setup. AOM : acousto-optic modulator, L : lens, PH : pinhole, CC : corner cube, PMT : photomultiplier tube.

III. EXPERIMENT

We performed Brillouin scattering experiments for the single crystal $PbMoO_4$ to investigate its elastic properties and birefringence. The single frequency light (λ = 514.5 nm) from an Ar^+-ion laser is incident on the sample. The angle of incidence was 70°, which was fixed for the entire experiment. The diffusely scattered light was analyzed by a Sandercock-type (3+3)-passes tandem Fabry-Perot interferometer [4], shown in Fig. 2. Preliminary studies showed that the sample surface is the (110) plane, instead of (100) [4]. Since the crystal has tetragonal symmetry (uniaxial), the analytic formulae shown above can be applied without any corrections. After careful setting up the scattering plane as the ac-plane, we obtained two spectra, shown in Fig. 3 on a logarithmic scale. Fig. 3(a) corresponds to the P-polarized incident light, whereas Fig. 3(b) corresponds to the S-polarization. Since we are concerned with only birefringence effects, we integrated the spectrum in about 20 minutes for Fig. 3(a) and 30 minutes for Fig. 3(b), so that the spectra do not show full signals, but it was long enough to observe the birefringence effect. The signals near 35 GHz in both spectra are due to the longitudinal acoustic phonons and the signal near 20 GHz results from the transverse phonons.

IV. RESULTS AND DISCUSSTION

In Fig. 3(a), there is one transmitted wave, as expected and the corresponding index is $n = n_o$ and the angle of refraction is θ_r = 22.37° for n_o = 2.469 [5] and θ_i = 70°. Therefore the signals can be identified as the acoustic phonons with the wavevector $\vec{q} = \cos 22.37°\hat{a} + \sin 22.37°\hat{b}$. If we use the fact that the surface is the (110)-plane, the wavevector is given by $\vec{q} = \cos 67.37°\hat{x} + \sin 67.37°\hat{y}$. The sound velocity of the longitudinal phonon is v_L = 3827 m/s, as determined by Eq. (3) and the corresponding frequency of the phonon is ν_L = 36.7 GHz, as determined by

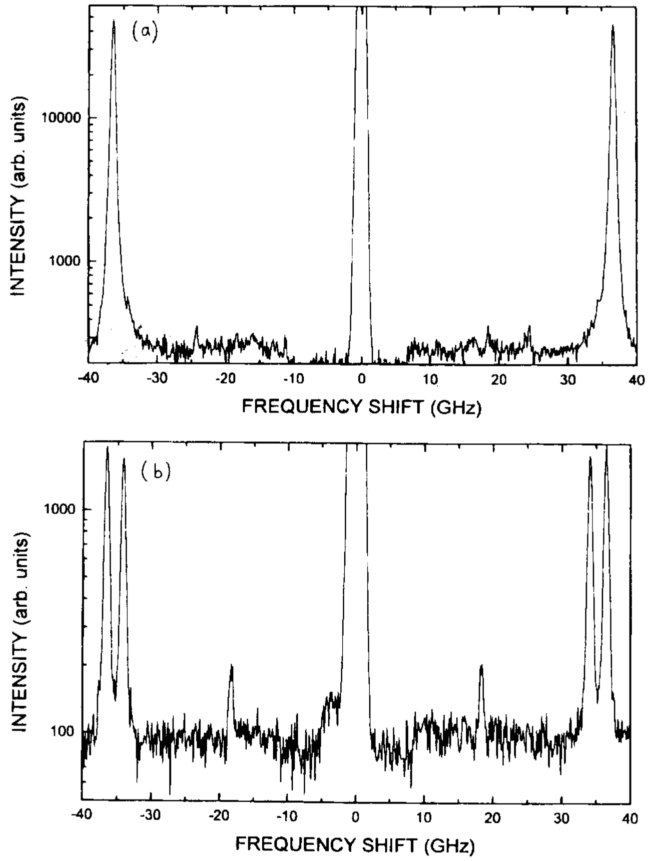

FIG. 3. Two Brillouin spectra for a single crystal of $PbMoO_4$. The incident plane is perpendicular to the optic axis and is in the xy-plane. The incident light is P-polarized (a) and S-polarized (b). The angle of incidence is 70^o for both spectra and the rest of the scattering parameters are the same.

$\nu_L = \frac{v_L}{\lambda} 2n_o$. The actual frequency shift of the phonon is 36.5 GHz.

If we change the polarization of the incident light to S-polarization (Fig. 3(b)), which is another eigen-polarization for this scattering geometry, the corresponding index is n_2 and $\theta_r = 23.92°$ for $n_e = 2.318$ [5]. So the wavevector of the phonon is changed to $\vec{q} = \cos 68.92°\hat{x} + \sin 68.92°\hat{y}$, whose velocity is $v_L = 3812$m/s and the frequency is $\nu_L = 34.3$ GHz. Fig. 3(b) shows a doublet signal instead of a single signal. It is not perfectly clear why such a spectrum occurred, but we speculate on some possibilities below. Nonetheless, we can identify the signal with higher frequency shift (36.5 GHz) as associated with n_o and that with the lower (34.1 GHz) as associated with n_2.

Table 1. shows the comparison of the values of v_L obtained above with the experimental values. They agree extremely well. As it shows, the medium is elastically anisotropic and the difference in frequency shifts of the two signals depends on the difference in the sound velocities as well as on the indices of refraction for the two directions of the transmitted waves. However, the difference in the sound velocity is only 0.4 % whereas that between the indices of refraction is 6.5 % and this combined difference caused the difference in frequency shift of 7 %, indicating that it is mainly due to the difference in indices of refraction. If the sound velocities are not known, the difference between two frequency shifts can be related to the birefringence, which may have an uncertainty of about 6 %. Nonetheless, Fig. 3(b) shows clearly the difference between the indices and whether this is positive or negative uniaxial. In addition, once one of the two indices is given, the other can be determined from the birefringence value.

Now we explain why there is a doublet signal for S-polarized incident light. So far we have not considered the scattered intensities, which are a combined result of both elasto-optic interaction and transmission of a transverse electromagnetic wave at the interface between two dielectric media. Since the angle of incident as 70° is very close to the Brewster angle ($\tan^{-1} 2.318 = 66.7°$), the P-polarized light can be transmitted as much as possible, whereas more than an half of the S-polarized light reflects. So for the incident light with S-polarization, only a part of the S-polarized light can be transmitted, scattered and transmitted back to the air, giving rise to a signal. In addition, even though the incident light is S-polarized, a small part of the incident light can be P-polarized and most of it can be transmitted, scattered and transmitted back to the air, resulting in another signal. This is why there are two signals for S-polarized incident light. The intensity of the signals in Fig. 3(b) is about 1/25 of that in Fig. 3(a). Since the integration time for Fig. 3(b) is about 1.5 times of that for Fig. 3(a), the normalized intensity of the signals in Fig. 3(b) is only 2/75 of that in Fig. 3(a). In order to fully analyze this result, it is necessary to study the scattering cross section of Brillouin scattering, but it is sufficient to show

TABLE 1. The difference in various properties for the two transmitted light waves.

	Ordinary wave	Extraordinary wave	Difference (%)
n	2 469	2.318	6.5
θ	22.37°	23.92°	—
v_L	3827	3812	0.4
v_L(theory)	36.7	34.3	7.0
v_L(experiment)	36.5	34.1	7.0

that the birefringence effect can be well observed for S-polarized incident light.

Another point to note is that the directions of wavevector of the transmitted wave and of the propagating light in anisotropic media are different in general. But for the incident plane perpendicular to the optic axis, the two directions are the same [6]. Thus the directions of the acoustic phonons probed by the Brillouin scattering experiment are parallel to the transmitted lights in 180° back-scattering geometry.

V. SUMMARY

We have shown that the birefringence $(n_o - n_e)$ of a single crystal can be determined in one Brillouin scattering experiment with more than 94 % accuracy even though the crystal is elastically anisotropic. In the appropriate scattering geometry, the incident plane is perpendicular to the optic axis and the incident light is S-polarized. In conjunction with the spectrum for P-polarization, the sign of the birefringence can also be obtained.

The ordinary index can be measured easily, for example, by Brewster angle measurement. Then the extraordinary index can be determined by substituting the ordinary index into the birefringence.

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