

Measurement of Birefringence with Brillouin Spectroscopy

Sukmock Lee* and Chang Kwon Hwangbo

Dept. of Physics, Inha University, Incheon 402-751, KOREA

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An alternative way to determine the birefringence for uniaxial crystals with Brillouin scattering experiments equipped with a Fabry-Perot interferometer is presented. The value of the minimum cavity spacing of the interferometer to observe the birefringence was found, and it is shown that the experimental error of the birefringence could be reduced by increasing the cavity spacing. For a single crystal of α -LiIO₃, the birefringence was found to be 0.1481 ± 0.0007 at 514.5 nm.

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I. INTRODUCTION

Birefringences ($n_o - n_e$, the difference between the extraordinary and ordinary refractive indices) for uniaxial crystals are fundamental properties and it is always of fundamental interest how to measure them precisely and accurately. One method is to measure the two indices of refraction separately by minimum deviation angle measurements with a wedged prism and the other is to measure directly by double refraction, conical refraction [1] or by using polarized light interference [2]. Recently, Chey et al. showed that Brillouin scattering (BS) experiments could be another alternative to determining the birefringence directly [3]. Thus, this paper describes the details of measurements of the birefringence by using BS, and shows that this method has an additional ability to magnify the birefringence in terms of a single Fabry-Perot interferometer's properties.

BS is inelastic scattering of light from thermal acoustic phonons and has been a well-known technique to determine the elastic properties of materials [4]. In BS, there are two measurable quantities, the acoustic phonon wavevector q determined from the scattering geometry and the angular frequency of the phonon $\Omega (= 2\pi f)$ 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 f and the interferometer is well established for measuring the frequency shift [4]. 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

$$q = \frac{4\pi n}{\lambda} \quad (1)$$

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

$$v = \frac{\Omega}{q} = \frac{\lambda f}{2n} \quad (2)$$

where f is the frequency shift of a corresponding Brillouin peak.

Since Eq. (2) can be rewritten in $f = (2/\lambda)nv$, the differential of f is contributed by both differentials of n and v , given by

$$\frac{\Delta f}{f} = \frac{\Delta n}{n} + \frac{\Delta v}{v} \quad (3)$$

When the index of refraction n is constant for optically isotropic media, e.g. glasses or cubic symmetry structure crystals, the refractive index is used as a given parameter to determine the sound velocity by using the frequency shift for each Brillouin peak with Eq. (2), which is the case of most BS experiments. In this case, each peak in a Brillouin spectrum corresponds to an acoustic phonon mode. Since there are generally three phonon modes available, one longitudinal and two transverse, there are three peaks in a spectrum.

In contrast, if there are two values of index of refraction available, double refraction appears. Each of two refracted waves is scattered from the phonons, generating a Brillouin spectrum consisted of a number of double-peaks. Each double-peak corresponds to one of the phonon modes and the difference between the two peaks of each double-peak is related to the difference between two refractive indices. When the

incident plane is perpendicular to the optic axis of a uniaxial medium, the two indices of refraction are the extraordinary and ordinary refractive indices. In addition, the refracted 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. If incident light is a sum of P- and S-polarizations in equal amount, there are two refracted waves with indices of refraction of n_o and n_e , for any angle of incidence.

The two indices of refraction mean two different directions of propagation of the phonon mode probed, which results in two different sound velocities. Usually, the sound velocity varies as a function of direction. But the changes of the sound velocity due to the two indices of refraction are typically as small as a few percent of the sound velocity. In particular, when the medium has a hexagonal symmetry structure, the plane is elastically isotropic [5], i.e. $\Delta v = 0$. So Eq. (3) can be rewritten as

$$\Delta n = n_o - n_e = n_o \frac{f_P - f_S}{f_P} \quad (4)$$

where f_P (f_S) is the frequency shift of the Brillouin peak in P (S)-polarized incidence. So we can determine the birefringence (Δn) from the difference in the two frequency shifts directly.

In order to resolve the two frequency shifts, the difference must be larger than equal to the full width at half maximum (FWHM), according to the Rayleigh's criterion, as

$$f_P - f_S \geq FWHM = \frac{FSR}{F} = \frac{c}{2LF} \quad (5)$$

for a single Fabry-Perot interferometer (FPI) with a finesse F [6] and the free spectral range ($FSR = c/2L$), where L is the cavity spacing and c is the speed of light in vacuum. Then we can combine the Eqs. (4) and (5) to determine the minimum cavity spacing to resolve the two frequency shifts as

$$L_{min} = \frac{150n_o}{F\Delta n f_P} \quad (6)$$

where the frequency shift of a Brillouin peak in P-polarization incidence is given in unit of GHz and the cavity spacing is in unit of mm.

Furthermore, the uncertainty of the birefringence $\delta(\Delta n)$ can be obtained by differentiating the Eq. (4) with respect to the frequency shift and is given by

$$\frac{\delta(\Delta n)}{\Delta n} \simeq \left| \frac{\delta f_P}{f_P} \right| \quad (7)$$

where δf_P can be equal to the FWHM of a Brillouin peak whose frequency shift is f_P . With a little algebra, the $\delta(\Delta n)$ can be rewritten as

$$\delta(\Delta n) = \frac{150\Delta n}{LF f_P} \quad (8)$$

So, the uncertainty can be reduced by increasing the cavity spacing with a given finesse F of the interferometer.

We have performed a Brillouin scattering experiment for a α -LiIO₃ single crystal to test the expectations. The single longitudinal mode of light ($\lambda=514.5$ nm) from an Ar⁺-ion laser was incident on the sample which is properly aligned. The incident light was a sum of P- and S- polarization in about equal amount and the angle of incidence 70°. The diffusely scattered light was analyzed by a 3-pass FPI and the experimental apparatus is described in detail elsewhere [4]. The reflectivity and flatness of the mirrors is 92% and over $\lambda/200$, respectively.

Fig. 1 shows three Brillouin spectra measured. The symmetric appearance of the spectra about the 0 frequency shift is characteristic of the Brillouin scattering, and it is due to the Stokes ($\Delta v < 0$) and Anti-Stokes ($\Delta v > 0$) scatterings, respectively. For the three spectra, all experimental parameters are fixed, except that the cavity spacing of the FPI is changed

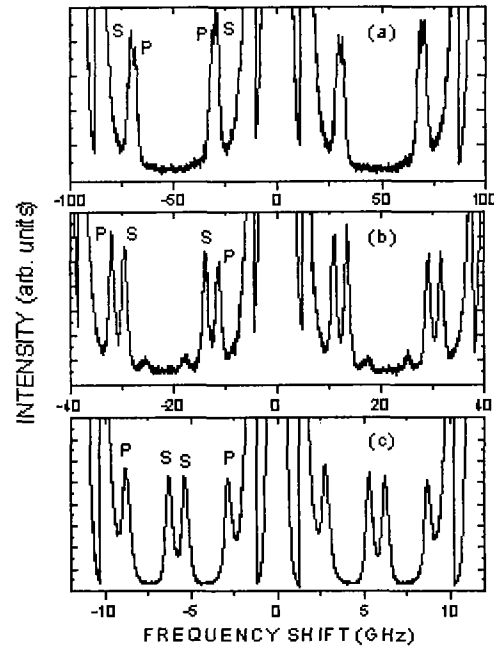


FIG. 1. Three Brillouin spectra measured for a-cut α -LiIO₃. The incident plane is perpendicular to the optic axis and the polarization of the incident light is a sum of P and S. The cavity spacing of the interferometer was 1.5 (a), 3.5 (b) and 13.0 mm (c). P(S) denotes that the corresponding peaks show up when the incident light is P(S)-polarization. Note the different scales in frequency shift.

to 1.5, 3.5, and 13.0 mm. The finesse of the interferometer was determined experimentally from the spectra as a ratio of the FSR and the FWHM of the 0 frequency shift peak at center, and the value of about 80 was observed throughout this study. The minimum cavity spacing calculated by Eq. (6) with the reference values of $n_o = 1.898$, $\Delta n = 0.147$, $f_p = 31.76$ GHz, and $F = 80$ was $L_{min} = 0.76$ mm. Because of the experimental limit, however, we tested the cavity spacing at 1.5 mm (Fig. 1(a)), instead. The FWHM of the peaks was 1.71 GHz and the frequency shifts of the longitudinal Brillouin peak were 31.6 and 29.2 GHz. Their separation (2.4 GHz) is large enough to be well resolved. The birefringence is then 0.144 and the uncertainty becomes 0.006. For the spectrum of 3.5 mm cavity spacing (Fig. 1(b)), the two peaks (denoted in P and S) are well separated and the birefringence was 0.149 ± 0.003 . When the cavity spacing is 13.0 mm (Fig. 1(c)), the birefringence of crystal α -LiIO₃ at 514.5 nm is 0.1481 ± 0.0007 .

The peaks at about 70(-70) GHz in Fig. 1(a) are characteristic of the interferometer and they correspond to the higher (lower) interference mode. As the cavity length increase, the FSR becomes smaller and the spectral position of Brillouin peaks moves away from the 0 frequency shift. Thus, the two real peaks of the 30 GHz frequency shift now are far from the center, whereas the higher (lower) interference mode on the contrary moves in close to the center as in Fig. 1(b). When the FSR becomes smaller than the frequency shift, as in Fig. 1(c), where FSR = 11.54 GHz, the two real peaks could not be shown in the original spectrum. Instead, the higher (lower) interference mode can be displayed. So the peaks at 8.7 and 6.2 GHz are the higher mode of the real peaks in Fig. 1(c). It is the property of FPI and is the main advantage of using FPI to determine the birefringence in conjunction with BS. Once the Brillouin peaks are identified, it is only required to increase the cavity spacing to increase the precision of the birefringence.

All experimental data agrees well with the theoretical expectations. The birefringence can be determined when the cavity spacing is larger than the minimum

value and when the cavity spacing is increased ten times, the uncertainty of the birefringence can be reduced into a tenth of the original value.

In summary, we presented a detailed calculation to show that Brillouin scattering experiments equipped with a single Fabry-Perot interferometer can be used to determine the birefringence of uniaxial crystals. In order to determine it, the cavity spacing must be larger than the minimum value given in Eq. (7). Furthermore, in order to increase the precision, the cavity spacing can be increased as much as allowed in the experimental apparatus.

As a closing remark, the FPI used in this work is a plane FPI, which limits the possible cavity spacing up to a few tens of mm and thus limits the precision. In order to increase the precision further, it is necessary to use a different type of FPI, such as a confocal FPI, and this is the subject of future work.

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*Corresponding author : smlee@inha.ac.kr.

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