Sound Propagation in 5CB Liquid Crystals Homogeneously Confined in a Planar Cell

Jae-Hyeon Ko^{*a}, Yoon Hwae Hwang^b, and Jong-Hyun Kim^{*c}

Abstract

The Brillouin spectrum of 4'-n-pentyl-4-cyano-biphenyl (5CB) liquid crystals homogeneously confined in a planar liquid crystal (LC) cell was measured using a 6-pass tandem Fabry-Perot interferometer. By adopting a special right-angle scattering geometry, the sound velocity of 5CB was estimated from the Brillouin shift without knowing the refractive index. The sound velocity of the longitudinal wave propagating along the direction of the directors aligned parallel to the glass plates of the LC cell was 1784 ± 7 m/s at 300 K. The attenuation coefficient α was estimated to be approximately 1.9×10^6 m⁻¹, which is about twice as large as that of the longitudinal sound wave propagating along the direction perpendicular to the directors. The present method may be very useful in the evaluation of the elastic properties of the materials used in display devices, whose refractive indices are not known.

Keywords: 5CB liquid crystal, Brillouin scattering, sound velocity, attenuation coefficient, homogeneous alignment

1. Introduction

Among the important physical properties necessary for the engineering design of display materials are their elastic properties, which are characterized based on the complete determination of the elastic-stiffness coefficients, their corresponding sound velocities, and the attenuation coefficients of the acoustic waves. The exact knowledge of these parameters is a great help in designing display devices using various materials, among them liquid crystals.

Sound propagation in liquid crystals can be investigated using several experimental methods, such as the ultrasonic technique and Brillouin light scattering. Using these methods, the sound velocity and the attenuation coefficient of the propagating density fluctuations can be estimated. In the case of the ultrasonic method, it is necessary to measure the time needed by a sound pulse to travel over a known distance [1]. In Brillouin light scattering, thermally excited density fluctuations give rise to thermal sound waves, resulting in the Brillouin doublet in the inelastically scattered light spectrum, in which the unshifted Rayleigh central peak will also appear [2]. By combining these two methods, the acoustic properties of liquid crystals can be obtained in a wide frequency range, from a few MHz to a few hundreds of GHz. The advent of laser light sources in the 1960s, and the invention of a single and tandem multipass FPI, made it possible to use FPI as a very high-contrast and high-resolution interferometer [3, 4], which has been applied to various condensed matters [5, 6]. Thanks to the recent development of the nonscanning-type Brillouin spectrometer, it became possible to investigate the acoustic properties of condensed matters within a very short time (a few seconds) [7-9].

4'-n-pentyl-4-cyano-biphenyl (5CB) is one of the typical and most extensively studied nematic liquid crystals. 5CB shows a nematic phase within the temperature range between 297 and 308.3 K. The changes in the acoustic properties during the isotropic-nematic-phase transition in 5CB have been studied using the ultrasonic and Brillouin methods [10-14]. Vaughan estimated the sound velocity of 5CB with the scattering vector aligned perpendicular to the directors in the hypersonic regime for the first time, using a special right-angle scattering geometry [11]. Later, Brad-

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berry and Clarke discovered a small hypersound anisotropy in the nematic phase of 5CB [12]. This hypersound anisotropy begins to appear at the isotropic-nematic-phase transition temperature upon cooling. More detailed Brillouin scattering studies have been carried out by Krüger et al. on 5CB and other liquid crystals, and the temperature dependences of the sound velocities propagating both parallel and perpendicular to the directions of the directors have also been investigated [13, 14]. The acoustic properties of 5CB, however, including the absolute value of its attenuation coefficient, have not yet been completely tabulated.

The present paper reports the measurement of the sound propagation in 5CB liquid crystals homogeneously aligned in a liquid crystal (LC) cell via Brillouin scattering at 300 K. The sound velocity and attenuation coefficient of the sound waves propagating along the direction of the directors were obtained accurately and were compared with the other values that were previously reported. A detailed investigation of the temperature dependence of the sound velocity and attenuation coefficient will be reported elsewhere.

2. Experiment

5CB liquid crystals were homogeneously aligned in an LC cell with a cell gap (d) of approximately 60 μ m, using a thin polyimide film (AL3046) as an alignment layer. The rubbing directions of both the upper and lower substrates were the same; hence, the direction of the directors, denoted as **n**, of the 5CB liquid crystals was the same as the rubbing direction. A tandem 6-pass Fabry-Perot interferometer was used to measure the Brillouin spectrum. This interferometer is characterized by a high contrast ratio and a wide frequency range [4, 5], which is very suitable for rejecting strong elastic scattering arising from the surfaces of the LC cell. This equipment is the standard instrument for the investigation of the acoustic properties of condensed matters over the GHz frequency range. The mirror spacing was 10 mm; hence, a free spectral range (FSR) of 15 GHz was obtained. An Ar+ laser at a 488 nm wavelength was used as a probe light. The laser power was maintained at less than 100 mW to avoid the local heating of the sample cell. A special right-angle scattering geometry was used to estimate the sound velocity without knowing the refractive index [5]. Fig. 1 shows a schematic diagram of the scattering geometry that was used in the present investigation. In this scattering geometry, the scattering wavevector \mathbf{k} is aligned in the



Fig. 1. A schematic diagram of the right-angle scattering geometry adopted in the present investigation. V and H denote the vertical and horizontal polarization directions, respectively.

cell along the rubbing direction, and is thus parallel with **n**. The Brillouin frequency shift $v_{\rm B}$ is given by the sound velocity V of the longitudinal waves propagating along **n**, the wavelength of the laser λ , and the refractive index of the air, which is set to be 1, as follows:

$$v_{B} = 2V \sin 45^{\circ} / \lambda = \sqrt{2V} / \lambda . \tag{1}$$

Therefore, measuring only the Brillouin frequency shift is enough for the determination of the sound velocity in LC cells, and there is no need to know the ordinary refractive index of liquid crystals. This method was successfully applied in the measurement of the acoustic properties of biological molecules and polymers [15, 16]. If the material is isotropic and nondispersive, the refractive index can be obtained by comparing the measurements of the Brillouin shift in the forward and backward directions [5].

3. Results and Discussion

The longitudinal waves propagating along **n** of 5CB were probed at 300 K at the scattering geometry described in Fig. 1. Fig. 2 shows the measured Brillouin spectrum. The spectrum consists of a Brillouin doublet caused by the longitudinal acoustic waves and a strong Rayleigh peak. Due to the fluidity and continuously broken rotational symmetry in the nematic phase, there is no shear stiffness, and there are thus no transverse acoustic modes in the Brillouin spectrum of 5CB. The small side peaks located near 10 GHz came from the sound waves propagating in the glass plates comprising the LC cell. To obtain the peak po-



Fig. 2. A Brillouin spectrum of 5CB liquid crystals homogeneously confined in a cell measured at 300 K. The solid line depicts the best-fitted result, as described in the text.

sition and phonon width, a combination of the response functions of the damped harmonic oscillator for the longitudinal acoustic mode, and of a single Debye relaxator for the central peak, was used to fit the measured spectrum I(v)as a function of the frequency v, which is given by the following equation:

$$I(\nu) = C_1 \frac{\nu_B^2 \Gamma_B}{\left(\nu^2 - \nu_B^2\right)^2 + \nu^2 \Gamma_B^2} + C_2 \frac{1}{1 + \nu^2 \Gamma_{CP}^2} + I_b$$

$$\approx C_1 \frac{1}{\left(\nu - \nu\right)^2 + \left(\Gamma_B / 2\right)^2} + C_2 \frac{1}{1 + \nu^2 \Gamma_{CP}^2} + I_b$$
(2)

Here, C_1 , C_2 , and C'_1 are proportional constants; I_b is the constant background; v_B and Γ_B are the Brillouin frequency shift and the full-width at half-maximum (FWHM) of the Brillouin doublet, respectively; and Γ_{CP} is the FWHM of the central Rayleigh component. Since the instrumental function of the Fabry-Perot interferometer is Gaussian, the actual fitting function was a convolution between the Gaussian and Lorentzian functions (the so-called "*Voigt function*").

As seen in Fig. 2, the best-fitted result represented by the solid line well explains the measured data. The curve fitting of the spectrum based on a response function of the damped harmonic oscillators gives the Brillouin frequency shift $v_{\rm B}$ of 5.17±0.02 GHz and the half-width $\Gamma_{\rm B}$ of 1.09±0.06 GHz. A sound velocity of 1784±7 m/s is derived from $v_{\rm B}$ via eq. (1). As for the central peak, quantitative analysis was difficult due to the strong elastic peak overlapping on the quasi-elastic central peak. Since liquid crystals can be considered a uniaxial system, the sound velocity of the longitudinal waves propagating along **n** is related to the elastic stiffness coefficient C_{33} in the following equation:

$$C_{33} = \rho V^2 = \rho v_B^2 \lambda^2 / 2, \qquad (3)$$

where ρ is the mass density of the liquid crystals. The calculated C_{33} is 3.25 ± 0.03 GPa considering $\rho = 1.022$ g/cm³ at 297 K [17]. On the other hand, $\Gamma_{\rm B}$ in relation to the sound velocity and attenuation coefficient α is as follows:

$$\Gamma_B = \alpha V / \pi \,, \tag{4}$$

where the estimated α is approximately $1.9 \times 10^6 \text{ m}^{-1}$.

Table 1 tabulates the sound velocities and attenuation coefficients obtained from the previous reports, and the values obtained from the present study. In this table, V_{\parallel} and V_{\perp} pertain to the sound velocities when the propagating direction is parallel and perpendicular to **n**, respectively, and *f* denotes the measuring frequency in the case of the ultrasonic method, and stands for $v_{\rm B}$ in Brillouin scattering. The elastic constants C_{33} and C_{11} calculated from V_{\parallel} and V_{\perp} , respectively, are also shown in the table. From the comparison in Table 1, several conclusions can be derived.

First, the sound velocities of longitudinal polarization measured in the hypersonic and ultrasonic regimes show large differences, as with the sound attenuation. These indicate that a hypersonic relaxation process is active in the nematic phase of 5CB, which is responsible for the substantial acoustic dispersion therein. Universally similar relaxation behaviors have also been observed in other nematic liquid crystals, and the thermal relaxation process and the evolution of the nematic-order parameter have been suggested as a possible origin of this common feature [13, 14].

Second, V_{\parallel} measured by different groups shows a difference in its absolute value by up to ~70 m/s at almost the same temperature. A difference of about 10 m/s in the sound velocity of 5CB has also been observed in the same group when the surface alignment condition was different [13]. This may suggest that the hypersonic sound velocity is affected by the surface interaction between the liquid crystals and the alignment layer, as well as by the sample thickness. Further investigation is required to better understand the relationship between the acoustic properties in the hypersonic regime and the surface condition of the LC cell. (Such investigation is already under way.)

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	<i>d</i> (µm)	V_{\parallel} (m/s) C_{33} (GPa) ^d	$\frac{V_{\perp} \text{ (m/s)}}{C_{11} \text{ (GPa)}^{\text{d}}}$	α (m ⁻¹)	f	Method
Present work	~60	1784 ± 5 3.25 ± 0.03	-	$1.9 \ge 10^{6}$	5.17 GHz	Brillouin
Ref. [13] ^a	~30	~1720 ~3.02	~1680 ~2.88	-	-	Brillouin
Ref. [13] ^a	~30	~1500 ~2.30	~1500 ~2.30	-	1 MHz	Ultrasonic
Ref. [12] ^b	~100	1740 3.09	1680 2.88	-	-	Brillouin
Ref. [11] ^c	~100	-	~1672 ~2.86	$1.0 \ge 10^{6}$	~ 4.77 GHz	Brillouin
Ref. [10]	-	-	-	6.6 x 10 ³	115 MHz	Ultrasonic

Table 1. Obtained Acoustic Properties of 5CB Liquid Crystals at 300 K, and Their Comparison with the Previous Data

^a The sound velocities were estimated from Fig. 6 (ref. [13]).

^b The measurement temperature was 303 K.

^c The sound velocities were estimated from Fig. 2 (ref. [11]). α is the calculated value at 4.5 GHz.

^d These values were calculated based on the reported sound velocities and the reported density of 1.022 g/cm³ at 297 K [17].

Third, the attenuation coefficient of the sound waves propagating along **n** estimated from the present work is approximately twice that of the sound waves propagating along the perpendicular direction [11]. This means that hypersonic anisotropy appears in both the sound velocity and the sound attenuation. A more detailed investigation into the dependence of this anisotropy as a function of temperature on both the homogeneous and homeotropic alignment cells is under way. The results of such investigation will be reported elsewhere.

4. Summary

In this study, hypersonic sound waves propagating along the direction of the directors of 5CB liquid crystals homogeneously confined in a planar cell were investigated via Brillouin scattering at 300 K. The accurate value of the sound velocity of longitudinal polarization was estimated to be 1784 ± 7 m/s, which is different from the previously reported values obtained at almost the same measurement temperature. This may suggest that the sound velocity is affected by the surface alignment condition in the cell as well as by the thickness. Hypersonic anisotropy appeared in both the sound velocity and the sound attenuation because the attenuation coefficient α , estimated to be approximately $1.9 \ge 10^6 \text{ m}^{-1}$ in the present study, is larger, approximately twice that of the sound wave propagating along the direction perpendicular to the directors. The present method may be very useful in the evaluation of the elastic properties of the materials used in display devices, whose refractive indices are not known.

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