

# Terahertz time-domain spectroscopy studies of the optical constants of the nematic liquid crystal 5CB

Tsong-Ru Tsai, Chao-Yuan Chen, Ci-Ling Pan, Ru-Pin Pan, and Xi-Cheng Zhang

The optical constants of a nematic liquid crystal, 4'-n-pentyl-4-cyanobiphenyl (5CB), in the frequency range 0.3–1.4 THz were determined by terahertz (THz) time-domain spectroscopy. The real parts of the extraordinary refractive index  $n_e$  and the ordinary refractive index  $n_o$  of 5CB varied from 1.74 to 2.04 and from 1.59 to 1.83, respectively. Liquid-crystal 5CB exhibits a relatively small absorption loss in this frequency range. The birefringence of 5CB was found to be as large as 0.21. The experimental results indicate that liquid-crystal 5CB is potentially useful for device applications in the THz frequency range.

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OCIS codes: 160.3710, 160.4760, 300.6270, 320.7150, 300.6530.

## 1. Introduction

During the past two decades, terahertz (THz) technology has undergone remarkable growth, with intense interest in, e.g., time-domain far-infrared spectroscopy,<sup>1,2</sup> THz imaging,<sup>3</sup> THz ranging,<sup>4</sup> and space communication.<sup>5</sup> A THz electro-optical modulator and phase shifters are required for further advances in many of the above applications. An optically controllable THz filter and phase shifter was previously demonstrated by use of mixed type I-type II GaAs/AlAs multiple quantum wells.<sup>6</sup> A He–Ne laser was used to modify the carrier density and thereby to shift the phase of the THz wave by 30°. Kersting *et al.*<sup>7</sup> previously demonstrated an electronically controllable THz phase modulator by using GaAs/AlGaAs parabolic quantum wells. They used electronic control of the carrier density within the parabolic quantum wells.

The birefringence ( $\Delta n = n_e - n_o$ ) of liquid crystals (LCs) is well known and has been extensively utilized for the modulation of visible, infrared, and millimeter-wave beams. In the visible range, the

birefringence of LCs has been investigated by many groups of researchers.<sup>8–12</sup> The refractive indices and other optical properties of LCs have also been reported for the infrared region.<sup>13–16</sup> Lim *et al.*<sup>17</sup> showed that in the millimeter-wave range many LCs have comparatively large birefringence with approximate values of 0.1–0.18 at 30 GHz. Furthermore, the birefringence of LCs varied only slightly in the 15–94-GHz range. Based on these results, LC-based waveguide-type and microstrip-type phase shifters in millimeter-wave regions have been demonstrated.<sup>17–19</sup> For application of LCs in the THz frequency range, knowledge of the dielectric anisotropy and electro-optic properties of LCs in this frequency range is most important. However, knowledge of the dielectric and electro-optical properties of LCs in the THz frequency range is still incomplete.

In the THz frequency range the refractive index and the transmission losses for some nematic LCs, including 4'-n-pentyl-4-cyanobiphenyl (5CB), were measured by Nose *et al.*<sup>20</sup> at three discrete wavelengths (118, 215, and 435  $\mu\text{m}$ ): The refractive indices of the LCs for ordinary ( $n_o$ ) and extraordinary ( $n_e$ ) rays in the submillimeter wave region were found to be slightly larger than those in the visible range. A large birefringence, comparable with that in the visible range, was also reported.<sup>20</sup> Recently the refractive index of the nematic LC 4-(*trans*-4'-pentylcyclohexyl)-benzotrile (PCH-5) in the frequency range 0.1–0.8 THz (approximately 375–3000  $\mu\text{m}$ ) was studied by THz time-domain spectroscopy.<sup>21</sup> The birefringence of PCH-5 at 0.1–0.8 THz

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Received 17 September 2002; revised manuscript received 16 December 2002.

0003-6935/03/132372-05\$15.00/0

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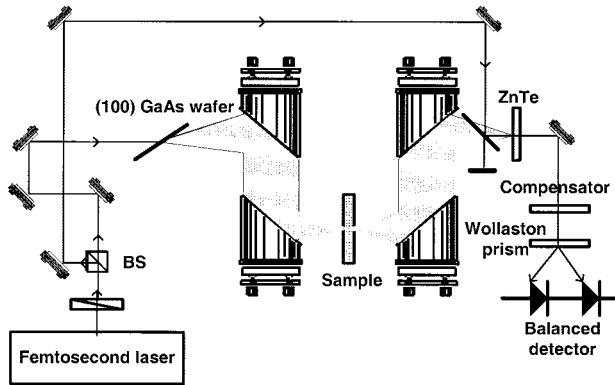


Fig. 1. Schematic of experimental setup for the THz spectrometer: BS, beam splitter.

( $\sim 0.01$ ) was found to be much smaller than that in the visible range ( $\sim 0.12$ ).<sup>22</sup>

The physical and chemical properties of 5CB have been studied extensively. The temperature range of 5CB in the nematic LC phase is 22.4–34.5 °C. Nematic 5CB exhibits nearly uniaxial molecular symmetry, high chemical stability, high positive dielectric anisotropy, and high birefringence in the visible region.<sup>23</sup> The birefringence for nematic 5CB in the visible region varies from 0.11 to 0.20, depending on the temperature and wavelength.<sup>23</sup> In this paper we describe our use of THz time-domain spectroscopy to characterize the optical constants of the nematic LC, 5CB. The optical constants of 5CB in the frequency range 0.3–1.4 THz are reported.

## 2. Experiment

We prepared the testing cell by sandwiching commercially available 5CB (Merck) LC between two fused-silica windows. The thickness of the sample was controlled by Mylar spacers with a nominal cell thickness of 25  $\mu\text{m}$ . The thicknesses of the two fused-silica windows were 1296 and 1278  $\mu\text{m}$ . We achieved homogeneous alignment of the nematic phase by coating polyimide films onto the inner surfaces of substrates.<sup>24</sup> The measurements were made at room temperature (25 °C).

Figure 1 shows a schematic of the experimental setup for THz time-domain spectroscopy, which is similar to the conventional THz system and has been described in detail elsewhere.<sup>25</sup> Briefly, a mode-locked Ti:sapphire laser ( $\lambda = 800$  nm) with 1.6-W average output power generates  $\sim 150$ -fs pulses at a repetition rate of 82 MHz. The laser beam is divided into two beams, a pump and a probe. The pump beam illuminates an inclined GaAs surface to generate the THz wave. The THz wave is collimated and focused onto the sample by a pair of off-axis paraboloidal mirrors. The THz wave transmitted through the sample is collimated and focused again onto a detector by a second pair of off-axis paraboloidal mirrors. A 2-mm-thick (110) ZnTe crystal is used in an electro-optic sampling setup for detection of the THz wave.

The electric field of the THz wave at the sample position is  $\sim 600$  V/m. Consider the dielectric anisotropy and distortion energy for 5CB; the estimated threshold electric field required for rotating the molecules of our sample is  $2.2 \times 10^4$  V/m. This is larger by 2 orders of magnitude than the THz electric field in the present experiment. Thus the THz electric field ought not to change the orientation of LC in the sample during the experiment.

## 3. Calculating the Optical Constants

In this study a 5CB LC layer of thickness  $L$ , referred to as medium 2, is sandwiched between two fused-silica substrates (referred to as media 1 and 3). The THz wave transmits through the cell from medium 1 to medium 3. Assuming that the THz wave is a plane wave, the electric field of the THz wave transmitted through the LC cell can be written as

$$E_{\text{sample}}(\omega) = T_{12}(\omega)P_2(\omega, L)T_{23}(\omega) \times \sum_{k=0}^{\infty} [R_{23}(\omega)P_2^2(\omega, L)R_{21}(\omega)]^k E(\omega), \quad (1)$$

where  $E(\omega)$  is the incident electric field of the THz wave,  $R_{ij}$  is the reflection coefficient at the  $i$ - $j$  interface,  $T_{ij}$  is the transmission coefficient from medium  $i$  to medium  $j$ , and  $P_2$  is the propagation coefficient in medium 2 over a distance  $d$ . The definitions for these coefficients are

$$R_{ij} = (n_i - n_j)/(n_i + n_j),$$

$$T_{ij} = 2n_i/(n_i + n_j),$$

$$P_2(\omega, d) = \exp(in_2\omega d/c),$$

with the complex refractive index of medium 2,  $n_2(\omega)$ . Equation (1) is equivalent to the following form found in optics texts<sup>26</sup>:

$$E(\omega) = T_{12}(\omega)T_{23}(\omega)\exp(in_2\omega d/c)E(\omega)/[1 + R_{21}(\omega)R_{23}(\omega)\exp(2in_2\omega d/c)]. \quad (2)$$

The electric field of the terahertz wave transmitted through a vacant cell is given by

$$E_{\text{ref}}(\omega) = T_{1\text{air}}(\omega)P_{\text{air}}(\omega, L)T_{\text{air}3}(\omega) \times \sum_{k=0}^{\infty} [R_{\text{air}3}(\omega)P_{\text{air}}^2(\omega, L)R_{\text{air}1}(\omega)]^k E(\omega). \quad (3)$$

One can obtain the complex transmission coefficient  $T(\omega)$  of the LC by dividing the signal  $E_{\text{sample}}$  obtained with the LC by  $E_{\text{ref}}$  obtained without the LC:

$$T(\omega) = E_{\text{sample}}(\omega)/E_{\text{ref}}(\omega). \quad (4)$$

In our case,  $n_1$  and  $n_3$  are indices of refraction of the fused-silica substrate, which were measured separately. The optical constants of LC can be obtained by solution of Eq. (4) numerically.<sup>27</sup>

Two principal refractive indices of nematic LC can be defined in terms of the propagation vector of the

linearly polarized THz beam both along the LC director and perpendicular to it. The measurements proceeded as follows: First, a reference temporal THz waveform was obtained by introduction of a vacant cell between the THz transmitter and the receiver. Subsequently the vacant cell was replaced by the filled cell and a second set of waveforms was taken. Because a nematic LC can be viewed as a uniaxial crystal,<sup>28</sup> the information for  $n_e$  and  $n_o$  of LC can be obtained by rotation of the cell to adjust the alignment direction of the LC to be parallel and perpendicular to the polarization direction of THz wave, respectively.

#### 4. Results and Discussions

To obtain the optical constants of LC by solving Eq. (4) we also need to know the refractive indices of the fused-silica substrates used in the LC cell. These constants were also measured with the same apparatus. We found that in the range 0.3–1.4 THz the real index of fused silica extends from 1.947 to 1.952 and the imaginary indices of fused silica are small. Our results are closed to the values reported by Grischkowsky *et al.*<sup>2</sup>

Figure 2 shows the measured time-domain THz waveforms transmitted through the cell. It is clear that the LC 5CB exhibits low loss in this frequency range. The THz wave spectrum can be obtained by application of a fast Fourier transform to the time-domain waveform. A typical THz spectrum is shown in Fig. 3. The signal-to-noise ratio of the data was roughly 1000:1, and the dynamic range of the spectrometer exceeded 1000. The spectral resolution of the system depends on the length of total scan time. In our case, the total scan range was roughly 16 ps, as displayed in Fig. 2(a). The spectral resolution was thus  $\sim 0.06$  THz. Dividing the spectrum obtained with the LC cell by the spectrum obtained with the vacant cell yielded the magnitude and phase difference of the complex amplitude transmission function of the 5CB layer. The optical constants of 5CB for both the  $o$  ray and the  $e$  ray were then calculated from Eq. (4). The results are shown in Fig. 4. Clearly, 5CB exhibits a positive birefringence ( $n_e > n_o$ ), which is consistent with the reported results in the visible range. There is no sharp resonance in the 0.3–1.4-THz range. Both  $n_e$  and  $n_o$  increase with increasing frequency. For comparison, we also plot the discrete values reported in Ref. 20. There is a discrepancy between our results and the values reported in Ref. 20. Both  $n_e$  and  $n_o$  reported in this paper are somewhat smaller than those of Ref. 20.

The thickness of the LC cell must be measured as accurately as possible, because this quantity enters directly into the determination of the refractive index. In our case the thickness of the LC layer was measured by an interferometric method.<sup>29</sup> The thickness of the fused-silica window was measured with a universal measuring machine (Société d'Instruments de Physique, Model SIP-1002M) with a precision better than 0.1  $\mu\text{m}$ . Maximum uncer-

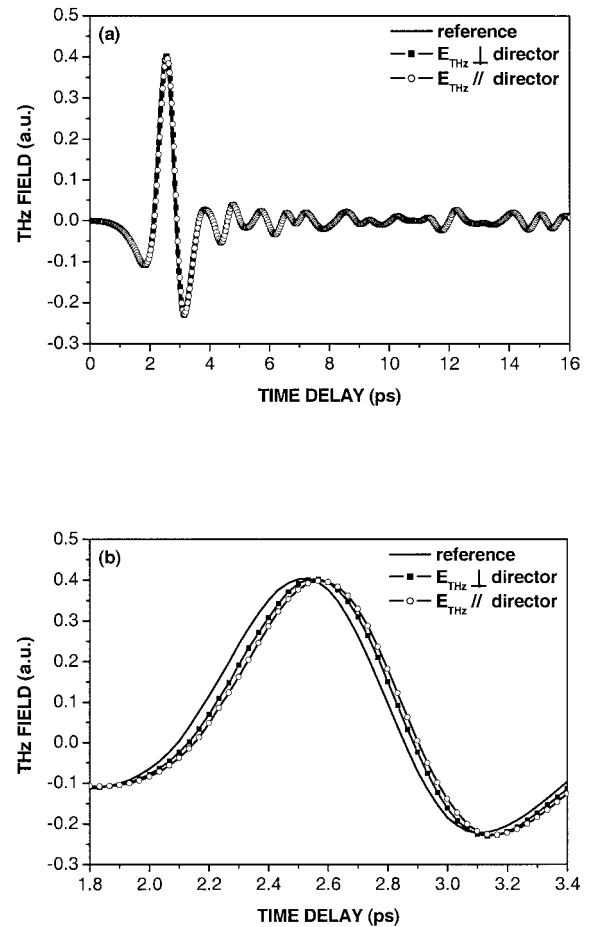


Fig. 2. (a) Temporal THz waveforms transmitted through a vacant reference cell and a LC cell. The LC director is either parallel or perpendicular to  $E_{\text{THz}}$ , the direction of polarization of the incident THz wave. (b) Expanded view of (a) in the time window 1.8–3.4 ps.

tainties are  $\delta n = \pm 0.01$  and  $\delta \kappa \pm 0.005$ . Preliminary data were also taken for a 125- $\mu\text{m}$ -thick 5CB cell. The variation in  $n_e$  or  $n_o$  extracted from the experimental data of 5CB cells with different thicknesses was less than 3%. The extinction coefficients  $\kappa_e$  and  $\kappa_o$  of 5CB are small in this frequency range.

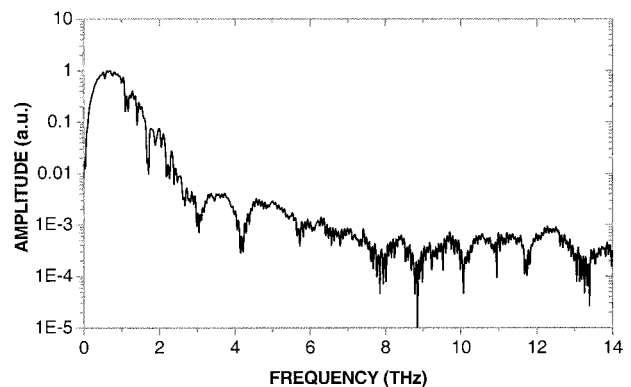


Fig. 3. Amplitude spectra of the measured THz wave after its propagation through a vacant reference cell.

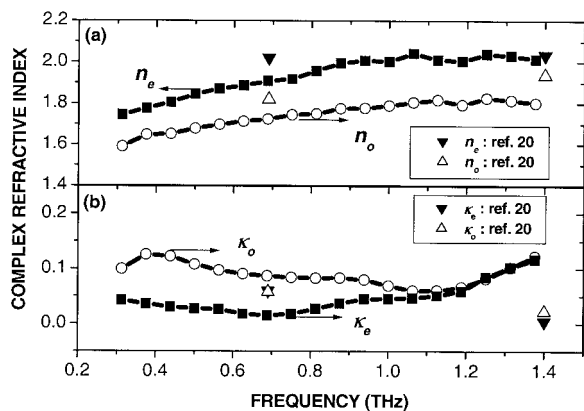


Fig. 4. Frequency dependence of (a) the real and (b) the imaginary parts of the complex refractive index of 5CB.

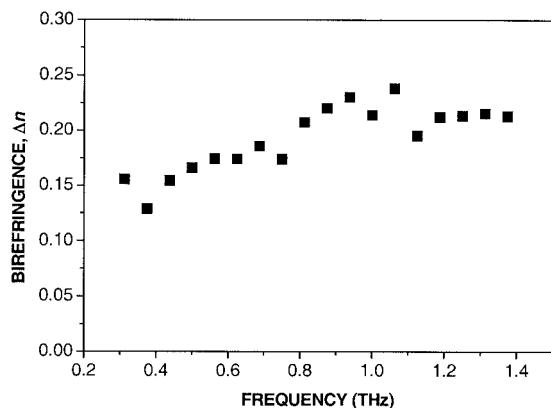


Fig. 5. Frequency dependence of the birefringence of 5CB.

The birefringence of 5CB,  $\Delta n$ , in the THz frequency range is quite large. The frequency dependence of  $\Delta n$  is shown in Fig. 5. It increases with frequency from 0.13 to 0.21. The error bars of the birefringence data are smaller than the size of a data point, so we do not show error bars. For comparison, the room-temperature (25 °C) birefringence of 5CB measured in the visible ( $\lambda = 632.8$  nm) is 0.18. This large birefringence is significant for potential applications of LC in THz devices such as phase shifters.

## 5. Conclusion

In conclusion, we have determined the THz frequency refractive index of the nematic liquid crystal 5CB at 25 °C. The ordinary and extraordinary indices of refraction of nematic 5CB are in the ranges 1.59–1.83 and 1.74–2.04, respectively. Nematic 5CB thus exhibits comparatively large birefringence, with approximate values in the 0.13–0.21 range. The extinction coefficients of nematic 5CB, however, are  $\sim 0.1$ , which indicates relatively small birefringence. These results provide the key optical properties of liquid crystals at THz frequencies and their potential device applications.

T. R. Tsai thanks Brad Ferguson and Jingzhou Xu for helpful discussions. The authors at the National

Chiao Tung University were supported in part by the National Science Council of Republic of China under grants NSC 89-2218-E-009-061, 90-2112-M-009-051, and -053. T. R. Tsai and C.-L. Pan were also supported by the Pursuit of Academic Excellence Program of the Ministry of Education of Republic of China. The study at Rensselaer Polytechnic Institute is supported in part by the U.S. Army Research Office and the National Science Foundation.

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