THz Optical Constants of the Liquid Crystal MDA-00-3461

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Published online: 30 Jun 2011.

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We have measured the far infrared optical constants of MDA-00-3461 at 25°C by using terahertz time-domain spectroscopy. The extraordinary and ordinary refractive indices of MDA-00-3461 are $n_e \approx 1.74$ and $n_o \approx 1.54$, or a birefringence of 0.20 from 0.3 to 1.4 THz. The extinction coefficient of MDA-00-3461 is relatively small. There are no absorption peaks in the frequency range investigated.

Keywords Absorption; birefringence; liquid crystal; refractive index; terahertz; time-domain spectroscopy

1. Introduction

The knowledge of the frequency dependence and the magnitude of the refractive indices of liquid crystals (LCs) is important for fundamental studies and practical applications of LCs. Applications of LCs in many fields are well-known. Following rapid development of terahertz science and technology in the past decade, terahertz optics and functional elements are in great demand. Recognizing the need for tunable LC terahertz photonic elements, we explored the optics, electro-optic and magneto-optics of LC in the terahertz frequency range [1]. Using terahertz time-domain spectroscopy (THz-TDS) [2], we have determined the complex indices of refraction of nematic liquid crystals, 5CB, PCH5 and E7 from 0.2 to beyond 1 THz [3–8]. Significantly, the birefringences of 5CB [3] and E7 [8] at terahertz frequencies are found to be comparable to their values in the visible regime, while the absorption is negligible. These works paved the way for the realization of functional LC tetrahertz optical elements such as 0–2π tunable tetrahertz phase shifters [9], control of enhanced transmission [10], polarizers [11], phase gratings [12], broadly tunable birefringent filters of the Lyot [13] and Solc types [14], etc.

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MDA-00-3461 is a relatively new liquid crystal by Merck, intended to replace currently widely-used LC mixture, E7. According to Merck's data sheet, the new LC exhibits a clearing point of 92°C, visible birefringence of ~0.26 at 20°C [15].

In this work, we employ THz-TDS to determine the refractive indices of LC MDA-00-3461 from 0.3 to 1.4 THz, for the first time to our knowledge.

2. Experimental Methods

We employed two types of cells, a homogeneously aligned LC cell and a reference cell, in this work. They are schematically shown in Figure 1. The LC cell was prepared by sandwiching MDA-00-3461 (Merck) between two fused silica windows as substrates. The thickness of the LC layer was controlled by Mylar spacers and was measured by subtracting the total thickness of substrate windows from the total thickness of the LC cell. The total thickness of substrate windows of the LC cell is $6348 \pm 1\,\mu m$. The thickness of the LC layer is $255 \pm 1\,\mu m$. Homogeneous alignment was achieved by spin coating the polyimide films on inner surfaces of the substrates of the LC cell, followed by mechanical rubbing [16]. The reference cell was fabricated by two fused silica windows and in contact with each other. The thickness of the reference cell is $6334 \pm 1\,\mu m$. The measurement temperature was set at room temperature ($25 \pm 0.1°C$).

An antenna-based THz-TDS system with a collimated terahertz beam at the sample position [17] had been used. A schematic of the THz-TDS system is shown in Figure 2. The mode-locked Ti: Sapphire laser beam in the THz-TDS system is divided into two beams, a pump and a probe. Terahertz pulses, generated by femtosecond-laser-excited dipole antenna fabricated on low-temperature-grown GaAs, were collimated by an off-axis paraboloidal mirror and propagated through the sample at normal incidence. The terahertz fields of extraordinary wave (e-wave) and ordinary wave (o-wave) are parallel and perpendicular to the rubbing direction of LC cell, respectively. The transmitted terahertz pulses were focused on another dipole-like antenna and oriented to detect terahertz waves polarized parallel to the incident terahertz wave polarization. The beam size of the terahertz wave through the sample is about 0.8 cm in diameter.
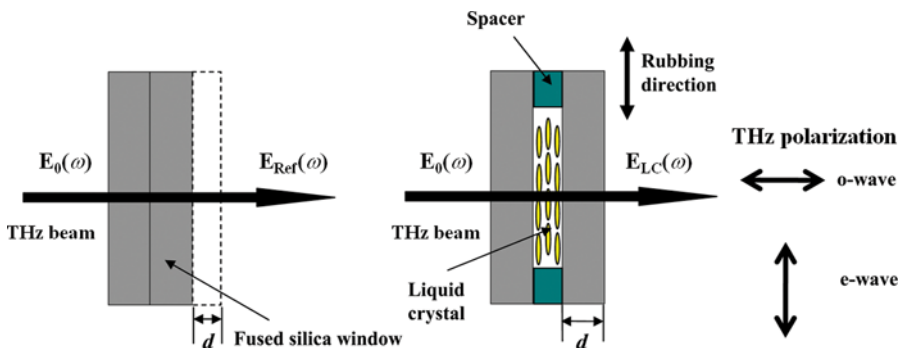
Figure 1. Sketches of (a) the reference cell and (b) the LC cell. The substrates are fused silica window and the alignment of LC cell is homogeneous and its geometry respect to the incident terahertz wave. (Figure appears in color online.)
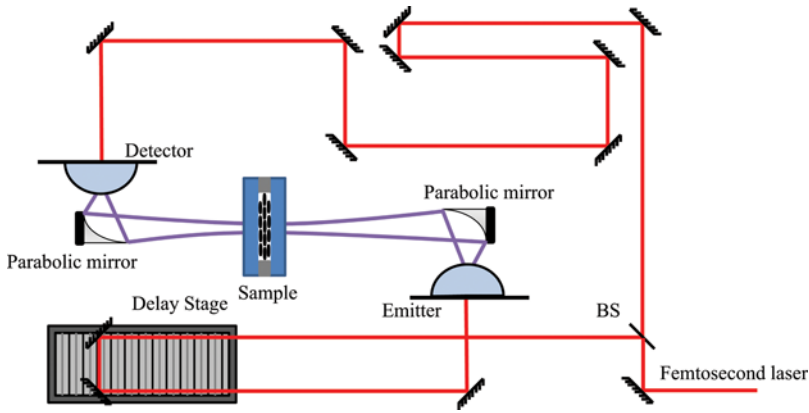
Figure 2. The schematic experimental setup for THz-TDS. BS: beam splitter. (Figure appears in color online.)
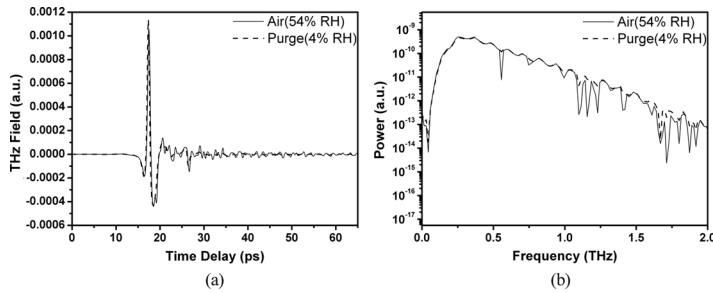
Figure 3. (a) The terahertz time-domain signals before (solid line) and after (dash line) purging. (b) The power spectra of the terahertz signals before (solid line) and after (dash line) purging. Several absorption lines of water vapor appear before purging evidently. The relative humidities before and after purging are 54% and 4%, respectively.

Several absorption lines of water vapor in the frequency range of 0.2–1.4 THz were observed [18]. Our THz-TDS system could be purged with nitrogen and kept the relative humidity at $(4.0 \pm 0.5)\%$. The terahertz time-domain signals and frequency signals before and after purging are shown in Figures 3(a) and 3(b), respectively.

3. Determination of Optical Constants

We assume that the terahertz signal is a monochromatic plane wave passing through the cell at normal incidence. The electric field of the terahertz wave transmitted through the reference cell at an angular frequency ω, can be written as

$$E_{\mathrm{Ref}}(\omega) = E_0(\omega) \cdot \eta(\omega) \cdot P_{Air}(\omega, d), \tag{1}$$

where $E_0(\omega)$ is the electric field of the incident THz wave. The parameter $\eta(\omega)$ takes into account the Fabry-Perot effect due to the Fresnel reflection of the tetrahertz

wave at the window-air interfaces and its propagation in fused silica. The propagation coefficient in air over a distance d is denoted by $P_{Air}(\omega, d) = \exp[(-i\tilde{n}_{Air}\omega d)/c]$, where $\tilde{n}_{Air}$ is the complex refractive index of air and the value of $1 + 0i$ is assumed in this work. The length d is chosen to be the same as that of the LC layer (see Fig. 1). Similarly, the electric field of the THz wave transmitted through the LC cell can be written as

$$E_{LC}(\omega) = E_0(\omega) \cdot \eta(\omega) \cdot T_{W-LC}(\omega) \cdot P_{LC}(\omega, d) \cdot T_{LC-W}(\omega) \cdot FP_{LC}(\omega, d), \quad (2)$$

where $T_{W-LC}(\omega) = (2 \cdot \tilde{n}_W)/(\tilde{n}_W + \tilde{n}_{LC})$ and $T_{W-LC}(\omega) = (2 \cdot \tilde{n}_{LC})/(\tilde{n}_{LC} + \tilde{n}_W)$ are the transmission coefficients of the window-LC and LC-window interfaces, respectively; $\tilde{n}_W$ is the complex refractive index of fused silica ($\tilde{n}_W = 1.951 + 0i$ in the frequency range of $0.2 - 1.4 \, \text{THz}$) and $\tilde{n}_{LC}$ is either the ordinary index ($\tilde{n}_o = n_o - i\kappa_o$) or extraordinary index ($\tilde{n}_e = n_e - i\kappa_e$) of the LC layer. $P_{LC}(\omega, d) = \exp[(-i\tilde{n}_{LC}\omega d)/c]$ and $FP_{LC}(\omega, d)$ are propagation and Fabry-Perot coefficients in the LC layer with a thickness of d. The coefficient, $\eta(\omega)$ for the LC cell is the same as that for the reference cell because of the thicknesses of the fused silica windows used are the same. The complex transmittance $T(\omega)$ of the LC layer can be obtained by dividing $E_{LC}(\omega)$ by $E_{Ref}(\omega)$:

$$T(\omega) = \frac{E_{LC}(\omega)}{E_{Ref}(\omega)} = \frac{4 \cdot \tilde{n}_{LC} \cdot \tilde{n}_W}{(\tilde{n}_{LC} + \tilde{n}_W)^2} \cdot \exp\left[-i(\tilde{n}_{LC} - \tilde{n}_{Air})\frac{\omega \cdot d}{c}\right] \cdot FP_{LC}(\omega, d), \quad (3)$$

For optically thick samples, such as ours, the echoes of tetrahertz waves from the multiple reflections of the sample are temporally well separated from the main signal. These can be easily removed without affecting accuracies of the measurements. Thus we can set $FP_{LC}(\omega, d) = 1$ in Eq. (3) [19] without considering the Fabry-Perot effect [20]. We set $FP_{LC}(\omega, d) = 1$ when we calculated the optical constants. In this work, $T(\omega)$ is experimentally measured. On the right of Eq. (3), only $\tilde{n}_{LC} = n_{LC} - i\kappa_{LC}$ is an unknown parameter, which can be easily calculated with a given trial set of (n_{LC}, κ_{LC}). Approximate values of n_{LC} and κ_{LC} were obtained by setting $FP(\omega) = 1$ in Eq. (3) and then this set was used as the initial trial values. After equalizing Eq. (3), the optical constants n_{LC} and κ_{LC} are determined for any angular frequency ω.

4. Results and Discussion

Figures 4(a) and 4(b) show the measured time-domain terahertz waveforms transmitted through the LC cell and reference cell. It is clear that the signal passing through the LC cell is delayed with respect to that of the reference signal. Further the delay time of the e-wave signal with respect to the reference signal is larger than that of the o-wave signal. A relatively small signal, which is the reflection of the terahertz wave from the window-LC interface, following the main terahertz signal and separating from about 8 ps is also observed. By applying the fast Fourier transform (FFT) to the terahertz time-domain signals, we obtained the amplitude and phase spectra of the terahertz wave passing through the LC cell and reference cell.
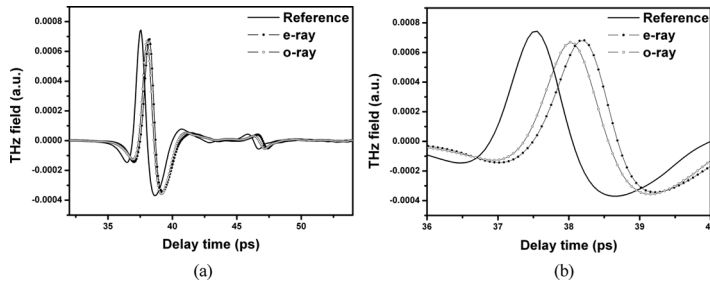
Figure 4. (a) The terahertz time-domain waveforms transmitted through a reference cell and LC cell. The LC director is either parallel or perpendicular to the direction of polarization of the incident terahertz wave. (b) The expanded view of (a) from 36 to 40 ps.

We calculated the optical constants of the LC by using the procedure described in the previous section.

Figure 5 shows the real part, imaginary part of the optical constants and birefringence of the LC MDA-00-3461 as a function of frequency at 25°C. In the 0.3 to 1.4 THz range, this LC is not dispersive. We determine that $n_e = 1.74$ and $n_o = 1.54$. The birefringence of MDA-00-3461 is thus 0.20. In comparison, the birefringence of MDA-00-3461 in the visible region is reported to be 0.26 at 589 nm and 20°C, according to the Merck data sheet. The imaginary part of the refractive indices is smaller than 0.05 in the frequency range from 0.3 to 1.4 THz. There is no apparent absorption features between 0.3 to 1.4 THz. Nematic LC E7, which is widely used in terahertz devices, exhibits $n_e = 1.71$, $n_o = 1.57$ and a somewhat smaller birefringence than those of MDA-00-3461.
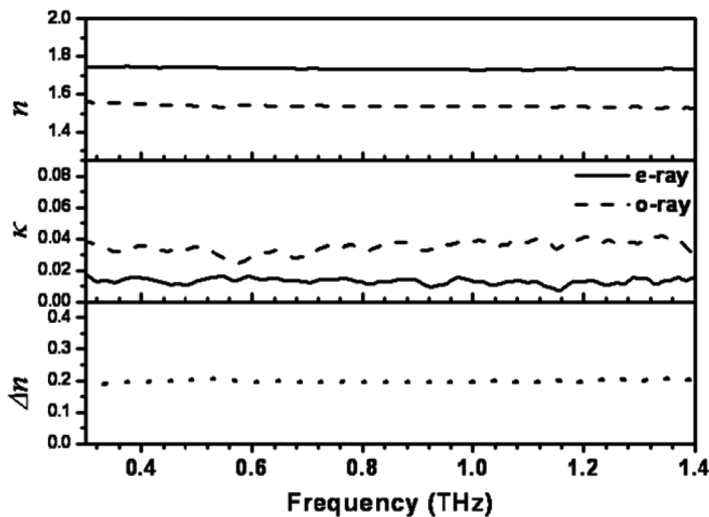
Figure 5. Room temperature terahertz optical constants, n and κ for e-ray and o-ray of MDA-00-3461 at 25°C. The solid lines are n and κ for e-ray. The dashed lines are n and κ for o-ray.

5. Conclusions

We report the far infrared complex refractive indices of MDA-00-3461 at room temperature 25°C from 0.3 to 1.4 THz by using THz-TDS system. The extraordinary and ordinary refractive indices of MDA-00-3461 are $n_e = 1.74$ and $n_o = 1.54$ at 25°C, and it cause a birefringence of 0.20 from 0.3 to 1.4 THz. The extinction coefficient of MDA-00-3461 is relatively small. There are no absorption peaks in the frequency range investigated. MDA-00-3461 exhibits somewhat larger tetrahertz birefringence than those of E7, while the imaginary indices or absorption are comparably low. The new LC MDA-00-3461 is thus a potential useful material for tetrahertz photonic devices.

Acknowledgment

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