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In this study we investigate anisotropic transmission properties of the membrane films impregnated with nematic LC materials in the THz frequency region. Typical experimental result shows that transmittance of membrane film without LC material is almost 100% in THz region. Phase change of the THz wave is observed but the transmittance does not decrease so much after impregnating LC material. Refractive index anisotropy increases after impregnating LC material which is related to the molecular orientation in the membrane film. These results show that the membrane film has a potential application to the LC device components in the THz region.

Keywords membrane film; molecular orientation; nematic liquid crystal; THz time-domain spectroscopy

Introduction

THz wave has some unique properties such as penetrate through various materials (paper, plastic, cloth etc.) and finger-print spectrum which means the specific absorption band appears depending on materials. Then it attracts attention for medical imaging, security check and other nondestructive testing [1–3]. However, it was difficult to measure the fundamental properties of materials and to study application devices in this frequency region, due to the lack of good light source and sensitive detector. Recently there has been remarkable progress in THz wave generation and detection technologies such as photoconductive switch, electro-optic sampling and etc. [4–6].

THz time-domain spectroscopy (TDS) [7,8] is the most popular technique of the quantitative investigation for imaging application and material analysis,

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since complex refractive indices can be calculated from the measured coherent electromagnetic pulse as absorption properties.

Liquid crystal (LC) has a large optical anisotropy and sensitive to an external stress such as an electric field, a magnetic field and so on. In the past years, there has been an extensive effort to measure the refractive indices and transmission losses for some LCs in THz region [9–14]. In this frequency region, LC materials show relatively large refractive index anisotropy and they can have a potential application to some tunable devices with low drive voltage and low power consumption similar to the optical applications. Recently, birefringence and extinction coefficient of 5CB have been measured by Pan *et al.* at 0.2–1.0 THz region using THz time-domain spectroscopy [14]. Furthermore, electrically tunable LC THz phase shifter using two cells (cell thickness is 1.012 mm) have been reported and maximum phase shift of 362.6° was achieved at 1.054 THz [15]. THz frequency is much lower than that in optical region, so extremely thick LC layer is necessary in this frequency region because of the longer wavelength. But increase in the thickness of LC layer makes LC molecular alignment difficult, and causes fatal disadvantage of response speed degradation, which especially appears in the recovery time. Simple solution for the problem is increase of wall effects which can be easily introduced by porous materials such as membrane films. In the millimeter wave region, Kuki *et al.* reported that response time of LC variable delay line becomes faster by using membrane film [16]. In previous work, we studied the relationship between microscopic structure of membrane films and LC molecular orientation by capacitance measurements. It is confirmed that the LC molecules are aligned along the fine fiber aligning structure of membrane film [17].

In this work we adopt membrane films to support LC molecules in extremely thick LC layer and investigate the transmission properties of the membrane/LC composite film in THz frequency region by using THz TDS system.

Experimental

Membrane films are usually used for filtering acid/alkaline solution or microbe, and there are many commercially-available products. Figure 1 shows the scanning electron microscope (SEM) image of the membrane film used in our experiments (Millipore, Omnipore Membrane), where the nominal pore size is 1 μm. It is seen that thick fibers elongated to the horizontal direction are suspended by many fine fibers between them. Nominal pore size means the ability of filtering size and corresponding to the mean spacing between the fine fibers. The material of membrane film is hydrophilic polytetrafluoroethylene. The porosity rate of membrane films was 80% and thickness of films is about 85 μm. From our previous study, LC materials penetrate through the membrane film well and it is observed that LC molecules tend to align along the direction parallel to the fine fiber direction. Furthermore, LC molecular orientation direction can be changed by the mechanically stretching of the membrane films.

Figure 2 shows the THz Time-Domain Spectroscopy system which is consists of a femtsecond laser, a photoconductive THz emitter, and a photoconductive detector. The THz emitter and detector are photoconductive switches fabricated on low-temperature grown InAs and GaAs, respectively. Laser pulses from the femtsecond laser are split into pump and probe beams. The pump beam impinges on a photoconductive switch to excite THz radiation. The emitted THz pluses are

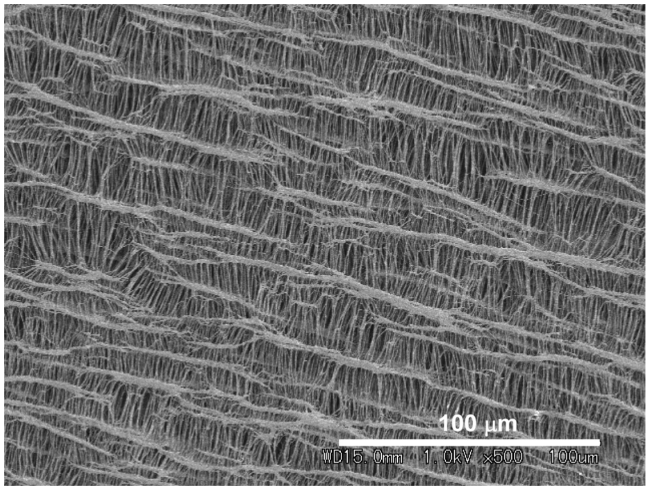


Figure 1. SEM image of membrane film whose nominal pore size is 1 μm.

guided with parabolic mirrors through the sample and onto the detector. Scanning delay line allows the waveform of THz electric field to be recorded via photoconductive THz detector.

Transmission properties of membrane films with and without LC materials were measured by the THz time-domain spectroscopy system from 0.2 to 2.0 THz. Transmittance and refractive indices are calculated from the measured data.

Figure 3(a) shows the measured pulse shapes for the cases without any sample (reference), membrane film with and without LC material (5CB). The nominal pore size of membrane film is 1 μm and parallel/perpendicular means that the sample setting of the fine fiber direction in the membrane film is parallel/perpendicular to the polarization direction of THz pulse. In the case without LC materials, we observed almost the same results with reference. On the other hand, some pulse delay

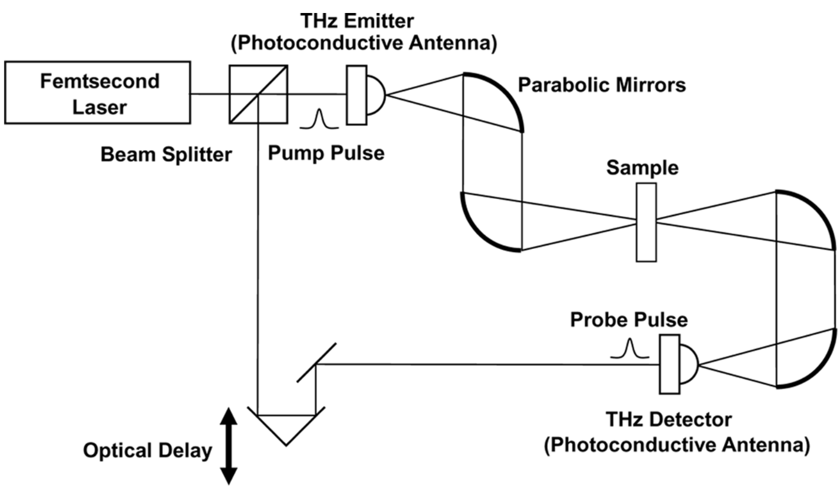
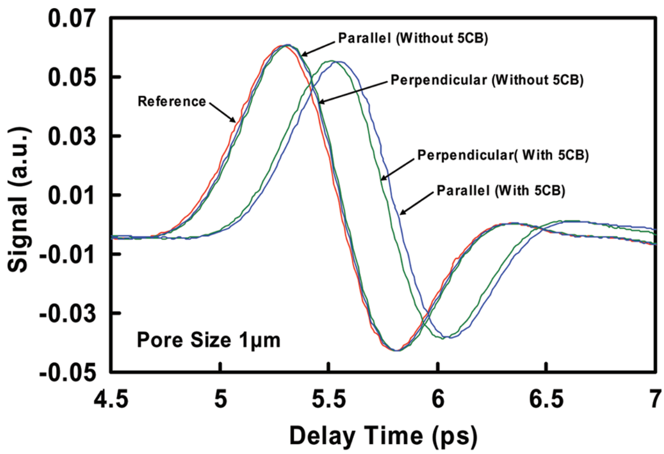
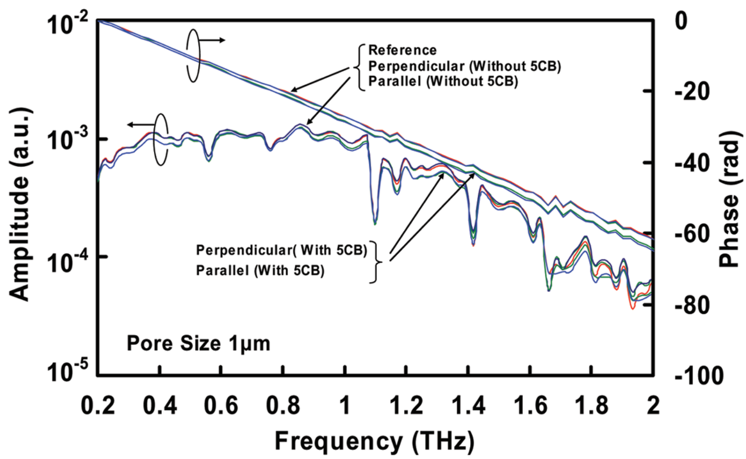


Figure 2. Experimental set up for THz TDS system.



(a)



(b)

Figure 3. (a) Transmission pulse of reference (without any sample), membrane film with and without LC material. (b) FFT spectrum. The nominal pore size of membrane film is 1 μm. Parallel/perpendicular means that fine fiber direction is parallel/perpendicular to the polarization direction of THz pulse.

was observed in the case of membrane films with LC materials, and the pulse delay of the parallel case becomes larger than that of the perpendicular case. The time delay corresponds to the refractive index of the test sample. It is seen that the refractive index anisotropy of LC material is detectable in this system, although the sample thickness is only 85 μm.

Figure 3(b) shows that the frequency spectra of amplitude and phase obtained by the first Fourier transform (FFT) of the time domain data (Fig. 3(a)). The several absorption lines observed in all scans at 056, 0.76, and 1.10 THz etc. are caused by the water vapor. These results show that transmittance of membrane film without LC material is as high as 100%. When the LC materials are introduced into the membrane film, transmittance decreases to be about 90%. We believe that the transmittance of membrane film with LC is high enough to apply to various THz controlling LC devices.

Here, we assume that THz pulse is passing through the sample in the air, and the complex transmittance $T_{obs}(\omega)$ can be written as [18]

$$T_{obs}(\omega) = \frac{E_{sample}(\omega)}{E_{reference}(\omega)} = t_{sample\rightarrow air}(\omega)\frac{p_{sample}(\omega, d)}{p_{preference}(\omega, d)}t_{air\rightarrow sample}(\omega) \tag{1}$$

where $E_{sample}(\omega)$ and $E_{reference}(\omega)$ are the transmission pulse with and without sample. $t_{sample\rightarrow air}$ and $t_{air\rightarrow sample}$ are the complex Fresnel transmission coefficients from sample to air and that from air to sample, respectively. $p_{sample}(\omega, d)$ and $p_{reference}(\omega, d)$ are propagation coefficients in the sample region with and without sample, respectively. Assuming that complex refractive index of air is $n_{air} = 1$, the complex Fresnel transmission coefficients and propagation coefficients can be written as

$$t_{sample\rightarrow air}(\omega) = \frac{2}{1 + \tilde{n}_{sample}(\omega)} \tag{2}$$

$$t_{air\rightarrow sample}(\omega) = \frac{2\tilde{n}_{sample}(\omega)}{1 + \tilde{n}_{sample}(\omega)} \tag{3}$$

$$p_{sample}(\omega, d) = \exp\left(i\frac{\omega d\tilde{n}_{sample}(\omega)}{c}\right) \tag{4}$$

$$p_{reference}(\omega, d) = \exp\left(i\frac{\omega d}{c}\right) \tag{5}$$

where ω is the angular frequency, d is the thickness of sample, c is the light speed. Complex refractive index is written as

$$\tilde{n}_{sample}(\omega) = n_{sample}(\omega) + i\kappa_{sample}(\omega) \tag{6}$$

where $n_{sample}(\omega)$ and $\kappa_{sample}(\omega)$ are refractive index and extinction ratio, respectively. From the experimentally measured amplitude and phase data, we can calculate the $n_{sample}(\omega)$ and $\kappa_{sample}(\omega)$ by solving Eqs. (1)–(6).

Figure 4 shows the obtained (a) refractive index and (b) extinction ratio of membrane film with and without LC material. In our calculations, we set the thickness of membrane film 85 μm of which nominal pore size is 1 μm. In Figure 4(a), we can see the refractive index anisotropy both cases without and with LC materials. In the case without LC material, refractive index of "parallel" which means that the fine fibers are parallel to the THz pulse polarization, becomes smaller than that of "perpendicular". We think that this is because the influence of thick fibers in the membrane film is effective to the THz pulse. In the case with LC material, refractive index of "parallel" becomes bigger than that of "perpendicular". This means that LC molecules tend to orient parallel to the direction of fine fibers and this fact corresponds to the result obtained by the capacitance measurements in Ref. [9]. From the results of Figure 4(b), extinction ratios become smaller than 0.1 for both case without and with LC material in the almost all measurement frequency range. We believe that membrane film with LC material is applicable in the THz frequency region.

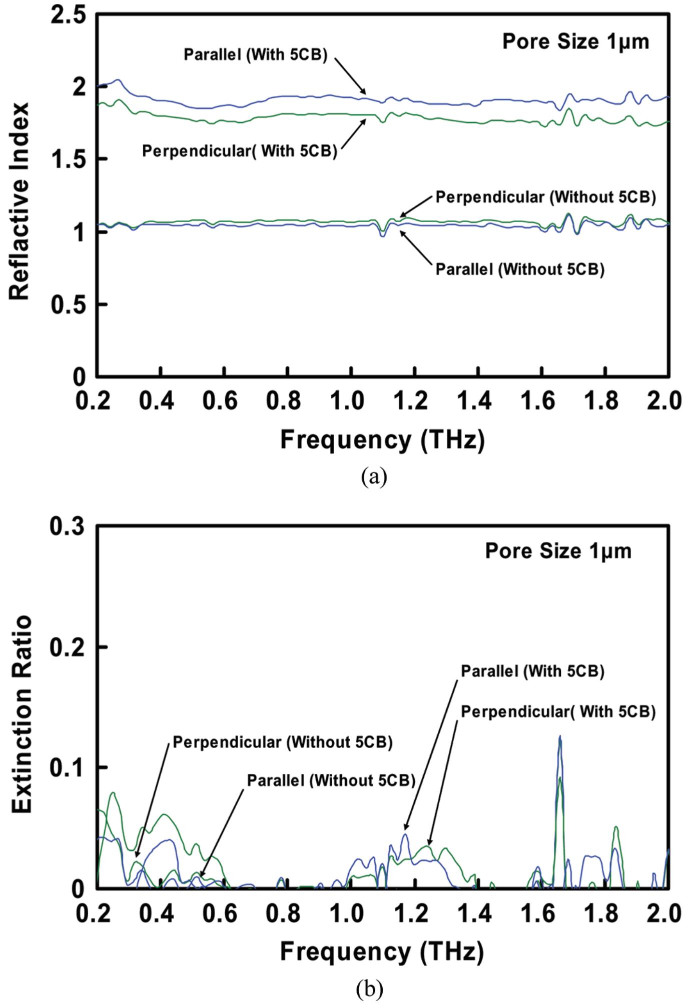


Figure 4. Obtained (a) refractive index and (b) extinction ratio of membrane film with and without LC material by solving Eq. (1).

Figure 5(a) shows the relationship between the nominal pore size of membrane films and refractive indices at 1 THz. THz transmission through the pulses of membrane films with 0.2 and 10 μm pore size were measured by the same way in Figure 3. Here, we set the thickness of membrane films 65 and 85 μm whose nominal pore size are 0.2 and 10 μm, respectively. Furthermore, refractive indices for the case with LC material are calculated by using the data without LC materials as reference data and they are shown in Figure 5(a) by closed markers (■, ▲).

In the case without LC material, refractive indices of 0.2 μm pore size membrane film show higher value compared to the 1 and 10 μm pore size. This result is due to the difference of fiber concentration as shown in Figure 5(b) (i.e., there are dense fibers compared to the Figs. 1 and 5(c)). Furthermore, it is seen that refractive indices of 1 and 10 μm pore size show almost the same value but the relationship between the value of parallel and perpendicular becomes inverse. This is because that mean

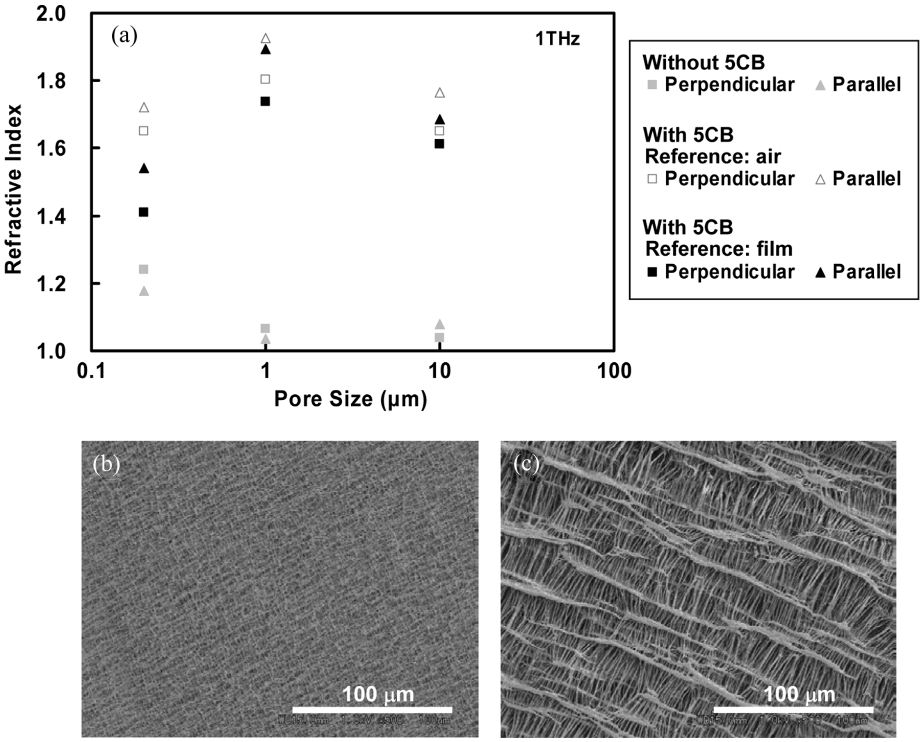


Figure 5. (a) Relationship between the nominal pore size of membrane films and refractive index at 1 THz. SEM images of membrane films whose nominal pore sizes are (b) 0.2 µm and (c) 10 µm.

concentration of fibers in two films is almost the same but relationship of thick fibers and fine fibers is different due to the difference of pore size.

On the other hand, all the data with LC materials show the large refractive index anisotropy and these results are corresponding to the Ref. [9]. From the calculated refractive indices using the data without LC materials as a reference, it is seen that refractive index anisotropy becomes lager than that using air (without any sample) as a reference in 0.2 and 1 µm pore size membrane films. In contrast, it becomes smaller in the 10 µm pore size membrane film. These results were probably caused by the difference of refractive index relationship between parallel and perpendicular in the membrane films without LC materials.

From Figure 5(a), the obtained refractive index anisotropy of 5CB at 1 THz is 0.08–0.16 and it is a little smaller than the value of 0.20 ± 0.02 measured by Pan *et al.* in Ref. [6]. This fact means that 5CB may not be perfectly orientated in our membrane films. On the other hand, obtained refractive index of 5CB at 1 THz is 1.41–1.89 and it is not in good agreement with the previous results (ordinary and extraordinary indices of refraction is 1.58 and 1.77) measured by Pan *et al.* in Ref. [6]. If we vary the sample thickness 10% in calculation, obtained refractive indices will change 0.04–0.05 in spite of the change of refractive index anisotropy is only 0.01.Then, there may be several mismatch of the sample thickness in calculations and experiment. Further investigation is in progress.

Summary

In this study, transmission properties of membrane films with and without LC material are measured by THz time-domain spectroscopy system from 0.2 to 2.0 THz. Transmittance of membrane film without LC material is as high as almost 100%. After impregnating LC material, phase change of the THz wave is observed and transmittance keeps about 90%. Refractive index anisotropy increases after impregnating LC material which is related to the molecular orientation in the membrane film. From these results, we believe that the membrane films have a potential application to various LC device components in THz region.

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