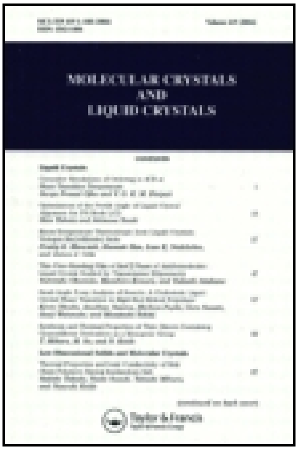

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Electrooptical Response in Free Standing Films of Ferroelectric Liquid Crystals

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Electrooptical Response in Free Standing Films of Ferroelectric Liquid Crystals

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By using free standing film of ferroelectric liquid crystals, we can realize a system with finite size along helical axis. The dynamics of free-standing film of FLCs has been studied by electrooptical measurement under weak sinusoidal electric field with dc bias field not to occur the switching of directors. We have measured the electrooptical spectra by varying the thickness of film L and the ratio of L to helical pitch P by changing temperature. The obtained spectra make good agreement with those predicted theoretically by Urbank and Zekš. In most cases, two relaxation modes are observed in the spectra. The relaxation frequency of the faster mode slightly varies around 1kHz and that of the slower mode varies largely with the ratio L/P.

Keywords: ferroelectric liquid crystal; free standing film; electrooptical spectra; finite size system; phase mode

INTRODUCTION

Smectic liquid crystal form free standing films whose thickness can be varied from only two to several thousands molecular layers. Thus, free standing film provide an ideal experimental system to study the influence of spatial dimensionality and surface interaction on physical properties.

Many experimental and theoretical studies have been performed on this unique system concerning to their structures, fluctuations and phase transitions [1-7]. In the bulk sample of ferroelectric liquid crystals (FLCs), the constituent chiral molecules form the helical structure whose pitch is as long as hundreds of smectic layers in the SmC* phase and and it varies largely with temperature. In free standing film of FLC, smectic layers stack parallel to the surfaces of film and the axis of helical structure is perpendicular to smectic layers. By preparing free standing film of FLCs of a few helical pitch thick, we can realize the system with finite size along helical axis. In the SmC* phase, there are two collective relaxation modes of the directors. One is soft mode, which is the fluctuation in the magnitude of tilt angle an the other is Goldstone mode, which is the fluctuation in azimuthal angle of director around helical axis. Except at the vicinity of the SmA-SmC* phase transition, Goldstone mode is predominant in the SmC* phase. Although the electrooptic response of FLCs in sandwich cells have been intensively studied, only few attempts have been made to investigate that of free standing films of FLCs[8,9].

In this study, we have studied the dynamics of FLCs in the free standing film whose thickness L is about a few times as long as the pitch P of helix by electrooptical relaxation spectroscopy. The electrooptical spectra have been studied by varying the thickness of film L and the ratio of L to P by changing temperature.

EXPERIMENT

The ferroelectric liquid crystal used in this study was p-decyloxybenzylidene-p'-amino-2-methylbuthylcinnamate(DOBAMBC). A free standing film was prepared across two metal plates which were also used as electrodes to apply a uniform electric field to the film and the distance between the electrodes was 3.5mm. Two PET sheets were set between the metal plates. The sample was loaded into the square-free area surrounded by the metal plates and PET sheets in the SmA phase (106°C). One of the PET sheets can be moved between the electrodes in order to expand the film and we can vary the thickness of the film. The film thickness was measured in the SmA phase by ellipsometry[10]. Then, the prepared free standing film is cooled into the SmC* phase at a rate of 0.1K/min.

For the electrooptical measurement, the beam of He-Ne laser (λ=632.8nm) after passing through polarizer and $\lambda/4$ compensator impinged on the free standing film with an incident angle of 45° and perpendicular to the applied field. The optical axis of the compensator was fixed at the angle of 45° with respect to the incident plane of the free standing film. The transmitted light after passing through analyzer was detected and converted into voltage signal by a high-speed PIN photodiode module (Hamamatsu S3387). We adjusted the directions of polarizer and analyzer in the SmA phase to increase the S/N ratio in the ac components of the electrooptical response by minimizing the transmission intensity. The electrooptical response was amplified and decomposed into frequency components by FFT analyzer (Advantest R9211).

Sinusoidal electric field generated from the synthesizer in the frequency range of 6.5Hz to 20kHz is applied to a film. The amplitude of the applied field is E=1.0V/3.5mm and this is small enough not to cause a switching behavior. We also applied the dc bias electric field to make the optical axis of the film uniform and fixed the favorable direction of the total polarization in order to distinguish the Goldstone mode from the switching.

RESULTS AND DISCUSSION

We measured the electrooptical spectra of free standing films of DOBAMBC thicker than one helical pitch. The temperature dependence of the complex spectrum (imaginary part I'') of transmitted light intensity obtained for the film of 1280 layers is shown in Figure 1. The SmA-SmC* transition temperature T_c was about 96.0℃. Except at 95.0℃ and 83.0℃, we found two relaxation modes in the spectra. The relaxation frequency of the faster mode is at about 1kHz and is not so sensitive with the variation of temperature. On the contrary, the frequency of the slower mode gradually decreases with cooling.

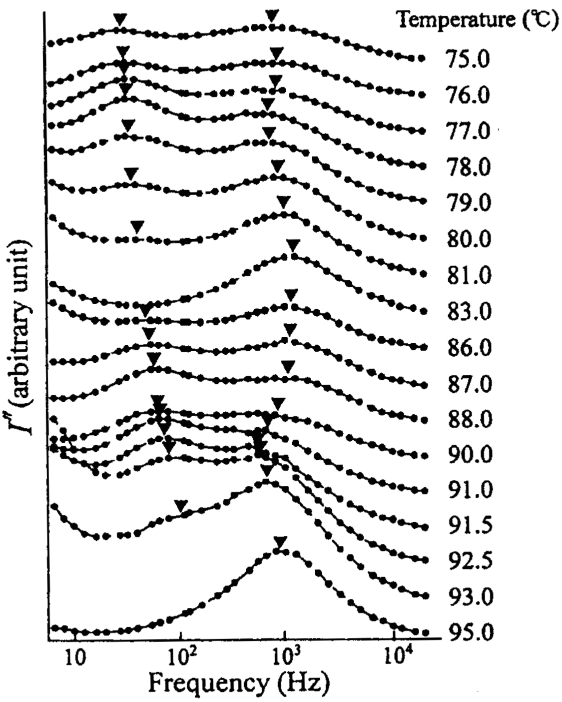

FIGURE 1 The temperature dependence of the electrooptical spectra (imaginary part) of free standing film of 1280 layers.

Recently, Urbanc and Žekš have studied the dynamics of phase mode in a system with finite size along helical axis theoretically[11]. They predicted the existence of the system-size mode other than usual Goldstone mode observed in sandwich cells. The former mode relates to the local reorientation of tilt and leaves the pitch of helix constant. The latter mode relates to the winding-unwinding motion of helical structure as a whole system. In their theory, dielectric response of a system with finite size along the helical axis was investigated. This system polarizes unless the length L is exactly a multiple of helical pitch. We denote the total dipole moment of the sample by $\vec{p}$ and here we assume that the directors at the surface of a film can move freely.

They discussed the dielectric response of a system to sinusoidal electric field of angular frequency ω, $\delta\vec{E} = \delta\vec{E}_0 \cos(\omega t)$, applied perpendicular to the helical axis. The real and imaginary part of susceptibility of the linear response to a time-dependent field applied parallel to the total dipole moment ($\delta\vec{E} // \vec{p}$) $\chi^{\parallel}_{\mathrm{Re}}$ and $\chi^{\parallel}_{\mathrm{Im}}$ are given as

$$\chi^{\parallel}_{\mathrm{Re}} = \frac{P_s}{2E_c} l^4 \cos^2\left(\frac{\pi l}{2}\right) \sum_{k=0}^{\infty} \frac{(2k+1)^2}{\left[(2k+1)^4 + l^4\Omega^2\right]\left[(2k+1)^2 - l^2\right]^2}, \quad (1)$$

$$\chi^{\parallel}_{\mathrm{Im}} = \frac{P_s}{2E_c} l^6 \Omega \cos^2\left(\frac{\pi l}{2}\right) \sum_{k=0}^{\infty} \frac{1}{\left[(2k+1)^4 + l^4\Omega^2\right]\left[(2k+1)^2 - l^2\right]^2}, \quad (2)$$

where E_c is critical electrical field of the SmC*-SmC phase transition, P_s is spontaneous polarization, $l = 2L/p_0$ is dimensionless length of the system and $\Omega = \gamma\omega / K_3 q_0^2$ is dimensionless frequency. The predicted relaxation frequency of the slowest mode in eqs. (1) and (2) is $f = K_3\pi^2 / \gamma L^2$, where K_3 is elastic constant and γ is rotational viscosity. In sandwich cells, the corresponding system size L is so large that the relaxation frequency becomes very low and cannot be observed in the measured spectrum. But, in the case of the free standing film whose thickness is about a few times as long as the helical pitch, we can observe this mode in the spectrum. We notice that the term of k=0 and the k-th harmonic term which takes the value of $(2k+1)$ nearest to l becomes much larger than other harmonic terms when L is not a multiple of the pitch of helix. This is the reason why there are two relaxation modes observed in most cases. Strictly speaking, these two modes are not independent each other and relate via value L/P which is characteristic value in a system with finite size along helical axis.

The linear elctrooptical relaxation spectrum I^* obtained for the film of 1280 layers at 76.0℃ is shown in Fig. 2. The best-fitted curve of eqs. (1) and (2) is drawn as a solid line. The temperature dependence of the best fitted values of L/P (=l/2) obtained for 1280 layers film is shown as filled circles in Fig. 3. It is found that the value P varies from about 850 layers to about 400 layers. The empty circles are the values of L/P calculated from the tilt angle and the pitch P measured for sample in sandwich cell. The calculated value of L/P saturates below T_c-T≈10℃. This difference is probably due to the influence of surfaces of glass substrates.

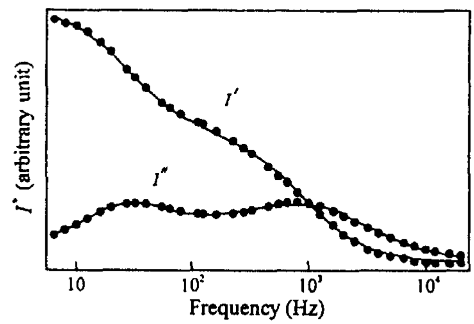

FIGURE 2 The linear elctrooptical relaxation spectrum $I^*(=I'-iI'')$ obtained for the free standing film of 1280 layers at 76.0℃

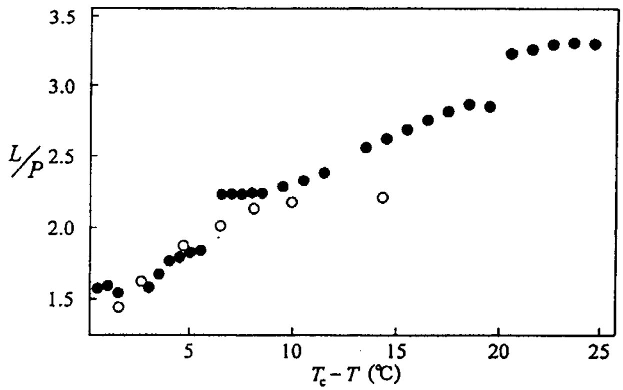

FIGURE 3 The temperature dependence of *L/P* obtained for free standing film of 1280 layers (●) and calculated ones (○) from the reported values in a sandwich cell.

The value of *L/P* shows jump at around integer value of *L/P*. Additionally, fitting curves slightly deviates from the obtained spectra just above integer value of *L/P*. These unexpected behaviors are partly due to the suppression and promotion of helical winding of dc bias field.

CONCLUSION

We have measured erectrooptical response in free standing films of FLCs of a few helical pitches thick. The obtained spectra make good agreement with those predicted theoretically by Urbank and Žekš on a system with a finite size along helical axis. The difference between the free standing film and the bulk that is observed emperature dependence of *L/P* seems to be due to the influence of surfaces of glass substrates.

Acknowledgements

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