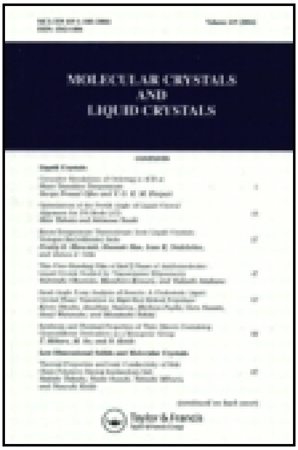

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INVESTIGATION OF REFRACTIVE INDICES OF FREE-STANDING FILMS BY ELLIPSOMETRY

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INVESTIGATION OF REFRACTIVE INDICES OF FREE-STANDING FILMS BY ELLIPSOMETRY

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The refractive indices of free-standing films (FSFs) of an antiferroelectric liquid crystal MHPOBC and a conventional smectic liquid crystal 8CB were studied by means of transmission and reflection ellipsometry. When the FSF thickness of MHPOBC is thicker than 260 nm, the refractive indices of FSFs appear to be the same as those of bulk smectic layers. In the case of thin FSFs of 8CB with a small numbers of layers, birefringence could not be obtained clearly by means of reflection ellipsometry.

Keywords: antiferroelectric liquid crystal; ellipsometry; free standing film; refractive index

We would like to thank J. A. Woollam Japan Co. Ltd for providing an opportunity to use VASE in our experiments.

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INTRODUCTION

Free Standing Films (FSFs) can be varied in thickness between hundreds and only two molecular layer. FSFs provide thus an ideal experimental system to study the influence of surface interactions on physical properties such as phase transitions, and FSF are useful systems to study the layer structure of smectic liquid crystal by means of ellipsometry.

We simulated the phase difference Δ between the p-polarized and s-polarized light outgoing from the FSFs of MHPOBC [1], so as to investigate the layer structure of FSFs in smectic phase, where refractive indices were assumed to be the same as those of the bulk sample ($n_e = 1.64$, $n_o = 1.50$ [5]). Because the FSFs is composed of a tremendous numbers of molecules in each layer of FSFs, therefore the physical concept of refractive index may hold even in such thin films. Under such assumption, previously we reported that these film thickness of MHPOBC were determined to be 5.0 nm(N = 2), 7.5 nm(N = 3), 10.0 nm(N = 4), 12.5 nm (N = 5), 15.0 nm(N = 6), 20.0 nm(N = 8) respectively (N represents the numbers of smectic layers in FSFs), by fitting the experimental results with numerical simulation. These discrete FSF thickneses showed that the each smectic layer thickness is assumed to be 2.5 nm, which is much smaller than the molecular length. This result may be interpreted to mean that the molecules in thin FSFs from interdigitated structures between the layer [2]. In this paper, we will report the behavior of the refractive indices of FSFs.

EXPERIMENTALS AND SIMULATIONS

FSF was formed between the two stainless steel plates as a frame, which is placed inside the thermostatic oven to control the temperature. The stainless steel plates also act as electrodes to apply an electric field (~ 9.0 V/mm) parallel to the smectic layer of FSF. The phase difference Δ of the outgoing light between the p-polarized and s-polarized light, was measured by means of transmission ellipsometry system which is composed of photoelastic modulator, He-Ne laser and a pair of Glan-Thompson prisms [3]. The phase difference Δ and the angle of amplitude ratio Ψ of the reflected light was measured by the variable angle spectroscopic ellipsometry (VASE, J. A. WOOLLAM). The AFLC substance used in the study was 4-(1-methylheptyloxy carbonyl)phenyl 4′-octyloxybiphenyl-4-carboxylate (MHPOBC)[4], which exhibits several kind of phase and subphases such as SmA, SmC^*_α, SmC^*, SmC^*_γ and SmC^*_A in the bulk state. Another smectic substances used in the study was 4′-Octyl-4-biphenylcarbonitrile(8CB). By fitting the incidence angle (θ_i) dependence of Δ with the simulation based on the 4×4 matrix method, the layer structure in the FSF was considered.

RESULTS AND DISCUSSIONS

Figure 1 shows the experimental and simulated results of the dependence of Δ on the incidence angle θ_i, where the measurements were carried out at the temperature of 145°C so as to be assumed that the phase of FSF is SmA. In this experiment, thick FSFs were prepared and then the film thickness was analyzed by substituting the refractive indices of bulk smectic layer, as shown in Figure 1(a). It was found that the simulated results of Δ are not fitted on the measurement results at all. However, by adjusting refractive index, the simulated result can be well fitted on the experimental results as shown in Figure 1(b), where the resultant refractive indices are $n_e = 1.522$ and $n_o = 1.424$, respectively. This result suggests that the refractive indices of thin FSFs are different from those of bulk sample.

Figure 2 shows the experimental results of the dependence of the refractive indices on the film thickness. We tried to analyze of refractive indices the the thick FSFs by the simulation in order to determine the refractive indices. 150 sample films were prepared and analyzed. It was found that the refractive indices of thick FSFs where the thickness is more than 1000(nm) do not exhibit the clear thickness dependence. In the case

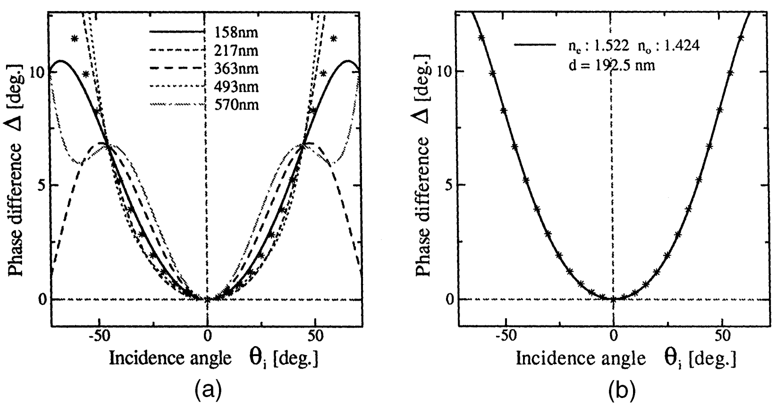


FIGURE 1 The dependence of Δ on the incidence angle θ_i under the DC applied electric field (±2.0 V/mm), where the measurements were carried out at the temperature of 145°C. (a) The curves are simulationed results for the various thicknesses of FSFs substituting the refractive indices of the bulk sample ($n_e = 1.64$, $n_o = 1.50$). (b) The curve is the result when the refractive index is adjusted.

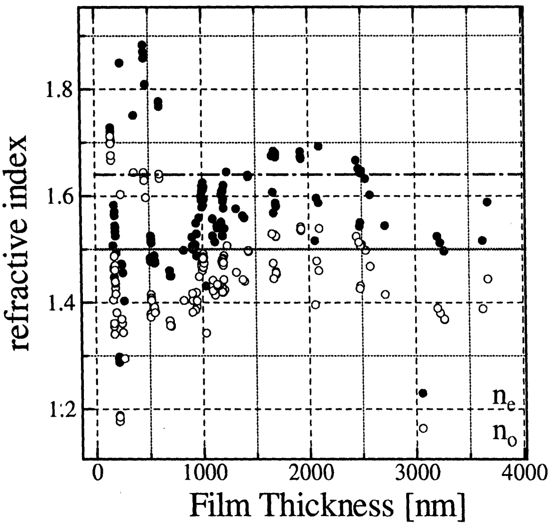


FIGURE 2 The dependence of the refractive indices on the film thickness. The open circle represent the results of n_e and the closed circle represent those of n_o. The lines represent the refractive index of the bulk sample (solid line: n_o, dash dot dash line: n_e).

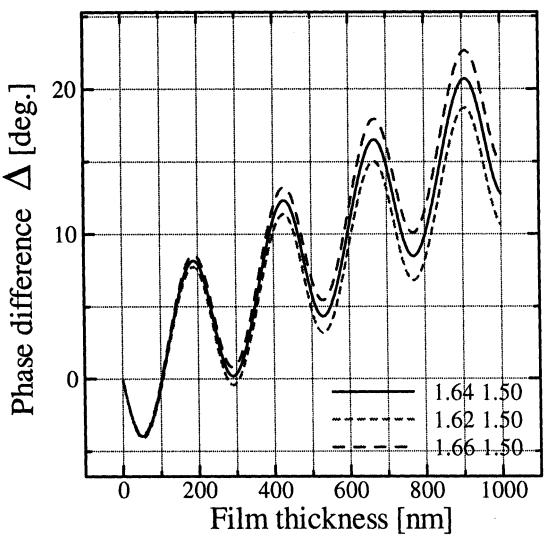


FIGURE 3 The dependence of Δ on the film thickness of the FSF. The curves represent simulated results ($\theta_i = 45°$).

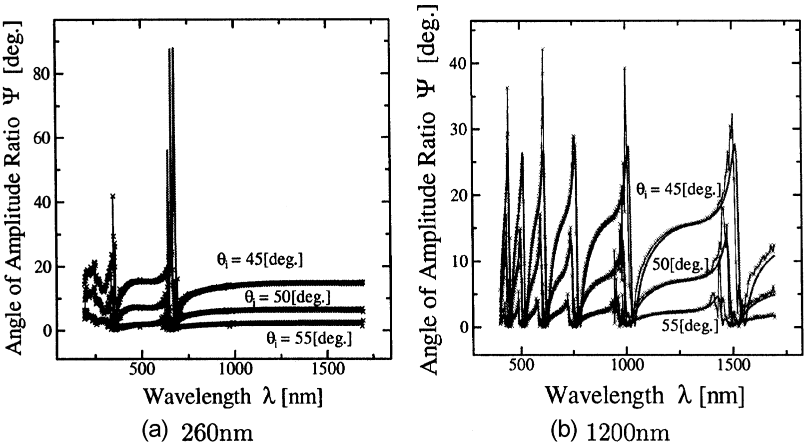


FIGURE 4 The wavelength dependence of the angle of amplitude ratio Ψ, where the measurements were carried out at the temperature of 145°C. The curves represent simulated results for the SmA structure.

of relatively thin FSFs where the thickness is less than 1000(nm), it was found that the amount of scatter in measured values with decreasing the film thickness. Figure 3 shows the simulated results of the dependence of Δ on the film thickness of the FSF. In the case of the thin film thickness region (viz. thinner than 10 layers), the effect of the change of refractive indices on Δ is small. Because, Δ corresponds to the birefringence $(n_e - n_o)d$, as we know, it is quite difficult to determine the film thickness and n_e and n_o separately by means of the transission ellipsometry.

Next we carried out the investigation of the film thickness dependence of the refractive indices by means of the variable angle spectroscopic refraction ellipsometry (VASE). Figure 4 show the experimental results of the wavelength dependence of the angle of amplitude ratio $\Psi(\lambda)$. The curves represent a numerical fittings, where the SmA structure was assumed. VASE can provide the film thickness and n_e and n_o, simultaneously [6]. In the case of the FSFs with thickness more than 160 nm, numerical fitting curves were well fitted on the experimental result and a remarkable change of Ψ which implies the existence of anisotropy of the refractive indices (e.g. birefringence Δn) can be recognized. Figure 5 shows the obtained wavelength dependence of the refractive indices in various film thickness, and Table 1 shows the refractive indices in the various film thickness at $\lambda = 632.55$ nm. It was found that the birefringence of thick FSFs whose thickness is more than 200 nm is almost the same as those of bulk sectic layers.

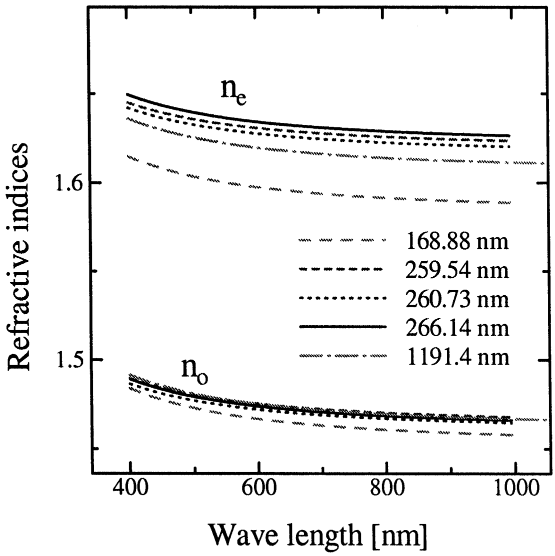
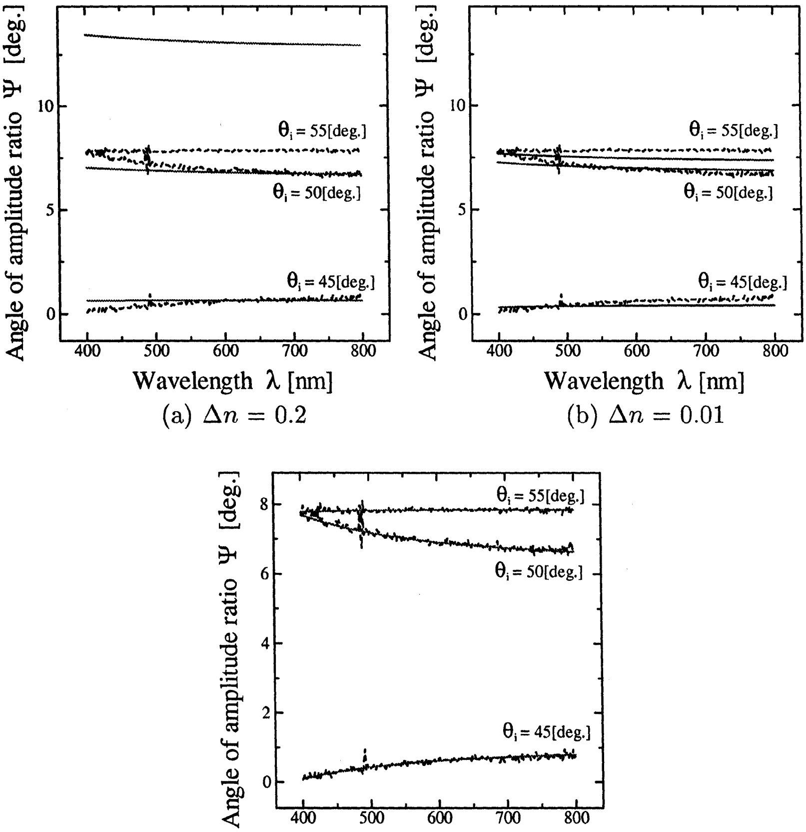


FIGURE 5 The dependence of the refractive indices on the film thickness.

To verify the thickness dependence with thin FSFs, we used 4′-Octyl-4-biphenylcarbonitrile (8CB) which can be formed as FSF only when the temperature is in the SmA temperature range. Figure 6 represents the experimental results for 8CB thin FSFs and its numerical fitting curves, where the birefringence was assumed to be 0.20, 0.01 and 0.00, respectively. The numbers of layers prepared in these experiment are assumed to be less than 10. As shown in Figure 6, the numerical simulation with assuming the birefringence $\Delta n = 0.00$ provided a best fitting curves. That is, this result implies that the thin FSFs were *isotopic* though it is unreasonable. As shown in Figure 2, it is difficult to determine n_e and n_o for such a thin FSFs, so we think that further consideration concerning the error analysis is required. Also another experiment such as Infrared -

TABLE 1 Refractive Indices ($\lambda = 632.55\,\text{nm}$)

d(nm)	n_e	n_o
1191.4	1.6185	1.4740
266.14	1.6330	1.4727
260.73	1.6265	1.4709
259.54	1.6296	1.4742
168.88	1.5962	1.4655
bulk	1.64	1.50



(a) $\Delta n = 0.2$ (b) $\Delta n = 0.01$ (c) $\Delta n = 0.0$

FIGURE 6 The wavelength dependence of the angle of amplitude ratio Ψ of 8CB, where the measurements were carried out at the room temperature. The dash line represent measurement results, the solid line represent simulated results. The refractive index anisotropy of FSF used with the simulation was supposed as (a) 0.20, (b) 0.01 and (c) 0.00.

ray experiment and the simulation of Molecular Dynamics are under progress and will be shown in the near future.

CONCLUSION

We examined the refractive indices of FSFs of MHPOBC and 8CB by means of transmission and reflection ellipsometry. When the FSF thickness is thicker than 260 nm, the refractive indices of FSFs seems not to be the same as those of bulk smectic layers. In the case of thin FSFs of 8CB with a small numbers of layers, birefringence could not be obtained by means of reflection ellipsometry. Anyway the absolute values of refractive indices is quite important to determine the layer structure. Another numerical approach and experiments will be demonstrated and will be shown in the near future.

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