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## EFFECT OF THE EXTERNAL TWIST ON MOLECULAR ORIENTATION IN ANTIFERROELECTRIC LIQUID CRYSTALS

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**Abstract** We report on microscopic observations of molecular orientation and layer structure induced by an external twist in antiferroelectric liquid crystals (AFLCs). In a twisted geometry, the smectic layers are formed perpendicular to the surface normal of the cell such that the molecular director experiences twist by twice of intrinsic molecular tilt on going from one surface to the other. It is found that this external twist significantly influences the uniformity of molecular alignment and the characteristics of the double hysteresis inherent to the antiferroelectric order. The resultant electro-optic effect is described within the framework of the field-induced antiferroelectric-ferroelectric phase transition.

### INTRODUCTION

Since the surface stabilized structure of ferroelectric liquid crystals<sup>1</sup> (SSFLCs) has been developed, much effort has been made for a basic understanding of surface phenomena as well as the bistable nature of the electro-optic (EO) effect. In such SSFLC structure, it is difficult to achieve gray scales because of the intrinsic bistability. One of the approaches to the realization of the gray scales in FLCs is to construct a twisted FLC (TFLC) structure, which turns out to exhibit indeed analog gray scale and fast response.<sup>2,3</sup> This TFLC is capable of producing a continuous optical transmission as a function of the electric field.

Recently, a novel biaxial smectic phase showing the antiferroelectricity has been observed in 4-(1-methylheptyloxycarbonyl) phenyl 4'-octyloxybiphenyl-4-carboxylate (MHPOBC).<sup>4</sup> This antiferroelectric (AF) phase,  $\text{Sm C}_A^*$ , may be suitable for use in novel display applications because of its wide viewing characteristics, fast response, sharp dc threshold, and durability against mechanical shocks. However, an aligning technique for producing a uniform structure and a driving scheme of achieving the gray scales are a prerequisite for new AFLC-based displays.

In this paper, we present experimental results for the molecular orientation

and the layer structure induced by an external twist in twisted AFLCs (TAFLCs) which adopt a similar concept used for TFLCs. It is found that TAFLC produces much improved uniformity of the alignment than a planar AFLC. Moreover, variable EO transmittance is achieved with varying the electric field. The effect of twist is described within the framework of the field-induced antiferroelectric-ferroelectric phase transition.

## EXPERIMENTAL

The material used in this study was CS4000 of Chisso Petrochemical Co. It has a wide temperature range of the Sm C<sub>A</sub><sup>\*</sup> phase between  $-10^{\circ}\text{C}$  and  $82^{\circ}\text{C}$  and the tilt angle of  $27^{\circ}$  at room temperature. The sample cells were made with indium-tin-oxide coated glasses which were treated with polyimide. The thickness of the polyimide layer was about  $300\text{ \AA}$ , and the surface was unidirectionally rubbed. The cells were assembled such that two rubbing axes on the substrates make an angle of  $0^{\circ}$  for a planar cell and  $54^{\circ}$  for a twisted one. The nominal cell thickness is  $3\mu\text{m}$ . The twist angle is exactly twice of the tilt angle as for the TFLC case. In this configuration, the molecular director in smectic layers will stay on one side of the cone at the top surface and on the other side at the bottom surface. The molecules in two adjacent smectic layers will then form the herringbone arrangement characteristic to the AF order.

The material was filled in the isotropic state and cooled down into the mesophase at a rate of  $0.1^{\circ}\text{C}/\text{min}$ . The EO transmittance through the cell, placed under crossed polarizers, was monitored with a photodiode and a digitizing storage oscilloscope. An arbitrary waveform generator was used to apply a voltage with variable amplitude to the cell. A He-Ne laser of  $632.8\text{ nm}$  was used as a light source. All the measurements were carried out at room temperature.

## RESULTS AND DISCUSSION

We first discuss the effect of an external twist on the uniformity of molecular orientation and layer structure in TAFLCs. Figs. 1(a) and 1(b) show microscopic textures of a planar AFLC cell and a TAFLC one, observed under crossed polarizers at room temperature, respectively. Clearly, the texture of the  $0^{\circ}$  twisted (planar) cell exhibits multidomains because of the herringbone structure of AFLC. The  $54^{\circ}$  twisted cell, however, produces a well aligned monodomain as shown in Fig. 1(b).

This indicates that a gradual twist of smectic layers is essential to produce uniform alignment. Among various treated cells, for example,  $126^\circ$  twisted and/or only one surface rubbed, the best alignment was achieved for the  $54^\circ$  twisted cell.

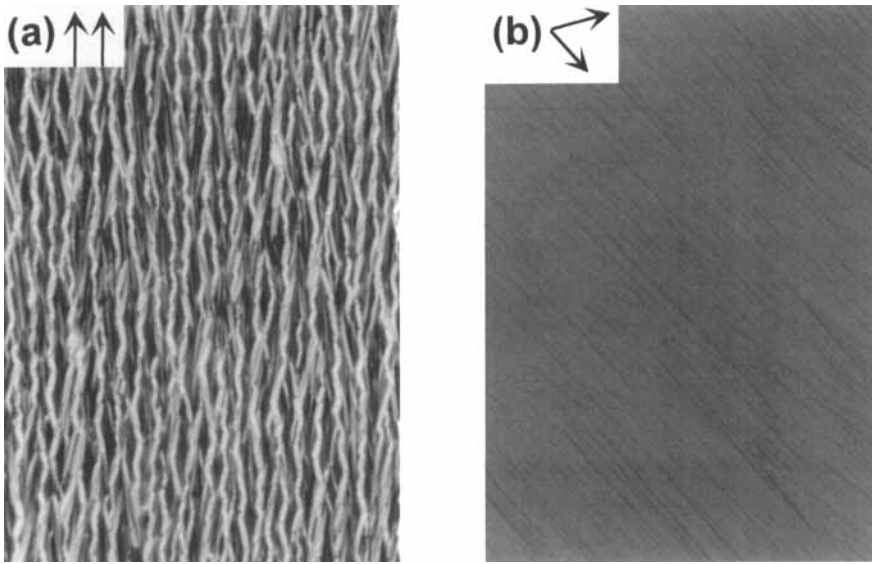


Figure 1: Typical textures of (a) the planar and (b) the  $54^\circ$  twisted AFLC cells, observed under crossed polarizers. (See Color Plate I).

We now discuss the resultant EO transmittance through the  $54^\circ$  twisted cell together with that through the planar one. Since gradual unwinding of the twisted structure is expected, variable transmittance through TAF LC is produced with varying the electric field, thereby giving some degree of gray scale. Fig. 2 shows the transmittance through the planar AFLC and TAF LC as a function of the field. For the EO measurements, a pulsed voltage at 30Hz was applied to the cell in which one of crossed polarizers coincides with one of two rubbing axes on the surfaces. The TAF LC has about 20% larger transmittance than AFLC. For AFLC, the transmittance increases sharply at  $E_{th} = 11\text{V}/\mu\text{m}$  where the field-induced antiferroelectric (AF) to ferroelectric (FO) phase transition takes place. Above  $E_{th}$ , the FO phase produces the maximum transmittance. For TAF LC, however, the maximum transmit-

tance is obtained in the absence of the field since TAFLC experiences waveguiding, and the polarization of the incident light becomes to rotate by  $54^\circ$ . In the low field regime ( $E < E_{th}$ ), the twisted state becomes gradually transformed into a uniform state by the unwinding process, and the resultant transmittance changes continuously with changing the field. In the high field regime ( $E > E_{th}$ ), the field-induced AF-FO phase transition occurs, so that the minimum transmittance is produced.

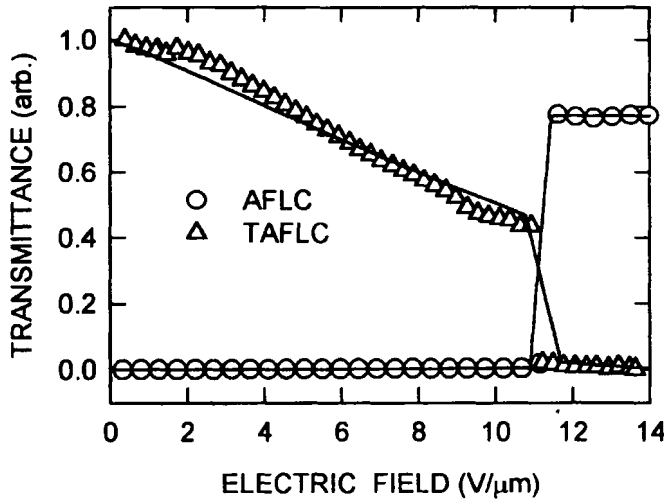


Figure 2: The EO transmittance as a function of the electric field. Open circles and triangles represent the experimental data for AFLC and TAFLC, respectively. The solid lines are theoretical fits to numerical results.

More quantitatively, the EO transmittance through TAFLC described above can be understood within the framework of the field-induced AF-FO transition provided that the external twist is imposed by the surface anchoring.<sup>5</sup> For numerical simulations, the following expression for the free energy density is used.

$$F = \sum_i^N [F_{FLC}(\phi^i) + (J_1/2)\{\cos(\phi^{i+1} - \phi^i) + \cos(\phi^{i-1} - \phi^i)\} + (J_2/2)\{\cos(\phi^{i+2} - \phi^i) + \cos(\phi^{i-2} - \phi^i)\} - \gamma_1 \cos^2(\phi^i) - \gamma_2 \cos(\phi^i)] , \quad (1)$$

where  $\phi^i$  denotes the azimuthal angle of the molecule in the  $i$ -th layer.  $F_{FLC}$  represents the free energy density of the FO phase.  $J_1$  and  $J_2$  are the coupling constants of the nearest and the next nearest neighboring layer pairs, respectively.<sup>5</sup> The last

two terms are the nonpolar ( $\gamma_1$ ) and polar ( $\gamma_2$ ) anchoring energies.<sup>6</sup> The boundary conditions used are (i)  $\phi_t = 0$  and  $\phi_b = 0$  for AFLC, and (ii)  $\phi_t = \pi$  and  $\phi_b = 0$  for TAFLC, where the subscript  $t$  and  $b$  represent the top and bottom surfaces, respectively. The transmittance can be computed using the  $2 \times 2$  Jones matrix method once the director profiles are obtained. The solid lines in Fig. 2 are theoretical fits performed with numerical results. For both AFLC and TAFLC, the fitted parameters are  $J_1 = 7.0 \times 10^3 \text{ J/m}^3$ ,  $J_2 = 10 \text{ J/m}^3$ ,  $\gamma_1 = 5.0 \times 10^3 \text{ J/m}^3$ , and  $\gamma_2 = 1.0 \times 10^2 \text{ J/m}^3$ .

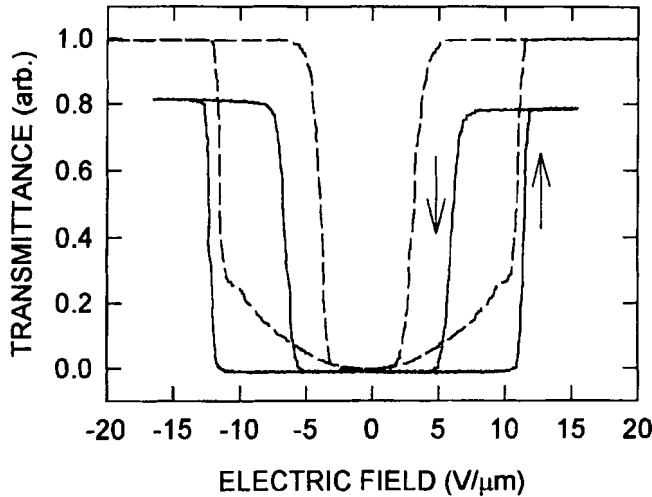


Figure 3: The double hysteresis in the EO transmittance measured with a triangular wave voltage at 0.5Hz. Solid and dashed lines represent AFLC and TAFLC, respectively.

Let us examine the effect of the external twist on the characteristics of the double hysteresis. Fig. 3 shows the double hysteresis in the EO transmittance as a function of the electric field. The measurements were performed with a triangular wave voltage at 0.5 Hz. Note that in both the AFLC and TAFLC cases, the hysteresis loops are very symmetric with respect to the field, meaning that no appreciable bias fields at the surfaces are present. The difference in the threshold for the AF to FO transition between AFLC and TAFLC is relatively small while that for the FO to AF transition is noticeable. It is suggested that the hysteresis loop is influenced by not only microscopic environment of the molecules but also externally imposed conditions such as the cell thickness and surface anchoring energy.<sup>7,8</sup> Moreover, any

finite degree of twist is expected to renormalize the surface anchoring energy in a twisted geometry.<sup>9</sup>

### CONCLUDING REMARKS

We have studied the effect of an external twist on molecular orientation and layer structure in twisted antiferroelectric liquid crystals (TAFLCs). This external twist was found to significantly influence the uniformity of molecular alignment and the characteristics of the double hysteresis inherent to the antiferroelectric order. The width of the hysteresis loop for TAFLC is larger than that for planar AFLC. Furthermore, variable EO transmittance was achieved with varying the electric field. The twist effect on the dynamical properties of TAFLC remains to be explored.

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