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Thresholdless Switching in Homogeneous Cells and Antiferroelectric Conoscopic Figures in Free-Standing Films as Observed in A Binary SC*-Like Mixture

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THRESHOLDLESS SWITCHING IN HOMOGENEOUS CELLS AND ANTIFERROELECTRIC CONOSCOPIC FIGURES IN FREE-STANDING FILMS AS OBSERVED IN A BINARY SC*-LIKE MIXTURE

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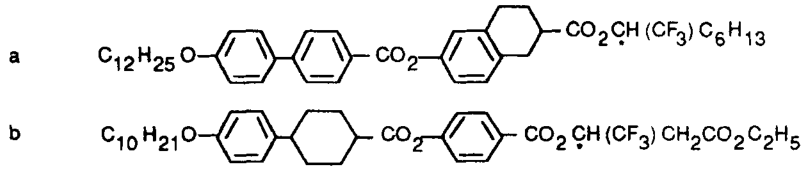
Abstract To elucidate the V-shaped switching reported by Inui *et al.*, we have studied another binary mixture. In its free-standing film, several subphases were observed in the ordinary sequence, indicating frustration between ferro- and antiferro-electricity; in particular, the antiferroelectric subphase exists in a temperature range as wide as 22 °C and the helical pitch is longer than 2.5 μm. In a homogeneous cell, however, only two liquid crystal phases were distinguished; one is SA and the other is a phase characterized by the nearly V-shaped switching. We have concluded that the substrate interfaces induce tilting randomization, breaking the reduced intrinsic tilting correlation in adjacent layers.

INTRODUCTION

Antiferroelectric liquid crystals (AFLCs) are commonly characterized by the tristable switching attributable to the electric-field-induced transition between antiferroelectric and ferroelectric phases. The switching has characteristic DC threshold and hysteresis.[1-3] On the other hand, Inui *et al.*[4,5] quite recently reported the thresholdless, hysteresis-free, V-shaped switching in an apparently AFLC mixture. The switching is very promising in realizing attractive liquid crystal displays.[6] They introduced the concept of thresholdless antiferroelectricity (TLAF) and speculated that diminished tilting correlation in adjacent layers causes tilting randomization and Langevin type alignment from layer to layer which are considered to be fundamental to the V-shaped switching.[4-7] Because of its extremely limited sample amount available at that time, Inui *et al.* did not confirm the thresholdless, hysteresis-free character as its bulk property by using a free-standing film. In this paper, we report a new liquid crystal mixture that shows the V-shaped switching in homogeneous cells but is antiferroelectric in free-standing films. The thresholdless behavior is considered to be realized by virtue of the substrate interfaces that induce the tilting randomization, breaking the reduced intrinsic tilting correlation in adjacent layers.

EXPERIMENTAL

The investigated sample was the MITSUI liquid crystalline mixture, MLC-0076, which consists of the following two compounds with a mixing ratio in weight of a / b = 6 / 4.[8]



About 100 μm thick free-standing films were prepared by the method described in Ref. 9. Transition temperatures were determined and phases were identified almost unambiguously by observing the electric field dependence of conoscopic figures.[10] We investigated the helicoidal structure as a function of temperature by measuring the characteristic reflection band for normal or 20° oblique incidence with a Hitachi U-3410 spectrophotometer and the optical rotatory power (ORP) at 514.5 nm with an Ar ion laser; the layer thickness as a function of temperature was also studied with a Rigaku RU-200 X-ray diffractometer following the method reported previously.[11]

Homogeneous cells were prepared by sandwiching the samples between two ITO glass susbstrates with 2.7 μm thick polyester films; the substrates were coated with Toray SP-510 polyimide and only one of them was rubbed with cloth. With an optical microscope, textures were observed for determining the phase sequence and transmittance measurements under crossed polarizers were performed by applying an electric field; the polarizer axes were set parallel and perpendicular to the smectic layer normal and the field applied was of 5 mHz ~ 1 Hz, ± 2 ~ 3 V_{pp} μm^{-1} triangular waveform.

RESULTS AND DISCUSSION

The phase sequence together with transition temperatures determined in free-standing films is:

$$\text{cry} — \text{SI*} \xleftrightarrow{32°C} \text{ferri} \xleftrightarrow{34°C} \text{AF} \xleftrightarrow{56°C} \text{FI} \xleftrightarrow{59°C} \text{SC*} \xleftrightarrow{79°C} \text{SA} \xleftrightarrow{100°C} \text{iso}$$

Conoscopic figures show the electric filed dependence as illustrated in Figure 1. It clearly indicates the existence of two ferroelectric (SC* and SI*), two ferrielectric (FI and ferri), and one antiferroelectric (AF) phases. The standard phase sequence so far established by studying various compounds and mixtures suggests the above identification. The high temperature ferroelectric phase is ordinary fluid SC* and the low temperature one must be hexatic SI*. The temperature change of the smectic layer thickness shown in Figure 2 supports this assignment; an increase in layer thickness with decreasing temperature is typical for the appearance of SI*, although we could not confirm the hexatic order by observing the X-ray Laue pattern thus far.[12,13] The antiferroelectric phase must not be SC_A* but AF, because it emerges above a ferrielectric phase.[3,14] The emergence of such a ferrielectric phase between antiferroelectric AF and ferroelectric SC* was recently reported by Hatano *et at.*[15] and O'Sullivan *et al.*[16] The ferrielectric phase below AF may be assigned to either of FI_L, SC_γ*, or FI_H;[3,14] since it was not easy to identify the phase more specifically, however, we just designate as "ferri".

Figure 3(a) shows the temperature dependence of the peak wavelength of the selective reflection for 20° oblique incidence: Both in SI* and SC*, the selective reflection bands were observed. The band in SI* and the longer band in SC* disappeared for normal incidence and hence are so-called full-pitch bands.[1-3] The helical pitch in SI* is so short that the ordinary selective reflection band does not appear in the transparent region of the sample. In ferri, AF, and FI, on the other hand, the characteristic reflection

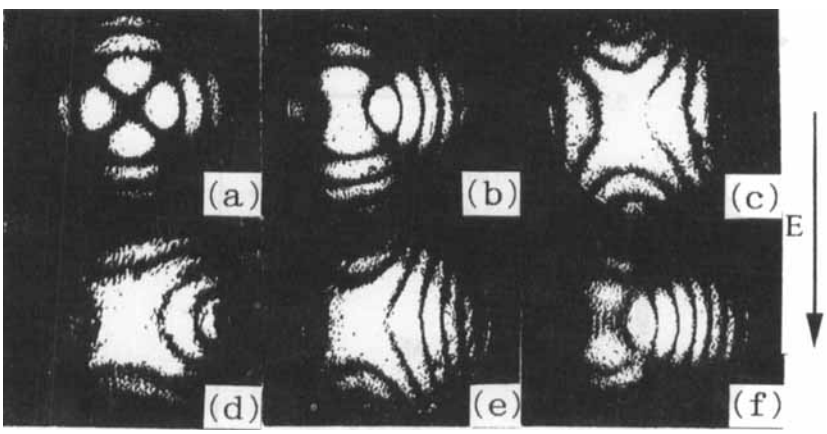


FIGURE 1. The field dependence of conoscopic figures at several temperatures. (a) SA and 0 Vmm^{-1}, (b) SC* (77.8 °C) and 30 Vmm^{-1}, (c) FI (56 °C) and 469 Vmm^{-1} (d) AF (41 °C), and 498 Vmm^{-1}, (e) ferri (34° C) and 752 Vmm^{-1}, (f) SI* (25 °C) and332 Vmm^{-1}.

bands were not observed in the wavelength region transparent and shorter than 2.5 μm. The ORP temperature variation given in Figure 3(b) indicates that the helical pitch in ferri, AF, and FI is long and that its handedness changes at the SI*-ferri and FI-SC* phase transitions. In this way, six liquid crystal phases are observed and their properties are quite normal in free-standing films.

In homogeneous cells, on the other hand, we were able to distinguish only two liquid crystal phases; one is the ordinary SA phase and the other is a phase considerably influenced by the substrate interfaces. This interface-induced phase is designated as SX* for the present. After the transition from SA to SX*, the texture changes slightly with decreasing temperature but does not show any indications which suggest the phase transitions observed in free-standing films. Figure 4 illustrates the electrooptic responses in SX* under crossed polarizers with the axes parallel and perpendicular to the smectic layer normal. At all the frequencies and temperatures studied, the nearly V-shaped switching is observed. In particular, even at 47 °C where antiferroelectric AF stably emerges in free-standing films, any DC threshold does not exist at a frequency as low as 5 mHz; the transmittance stays almost zero after the field is turned off. The switching differs from the ideal V-shaped one in two respects: (1) it shows some hysteresis and looks like W-shaped near the minimum (arrow 3); and (2) some steps (arrow 1, 2) are observed rather distinctly before complete switching to ferroelectric SC* or indistinctly after starting to decrease down to the minimum. At higher temperatures and lower frequencies, the hysteresis becomes smaller, the steps are more indistinct, and the switching looks almost V-shaped.

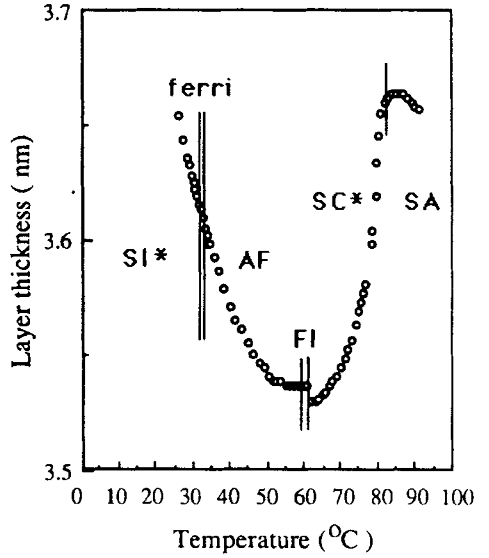


FIGURE 2. Temperature dependence of the smectic layer thickness.

Since several subphases are observed in free-standing films, the tilting correlation in adjacent layers must be reduced considerably but not diminish to zero. Suppose the

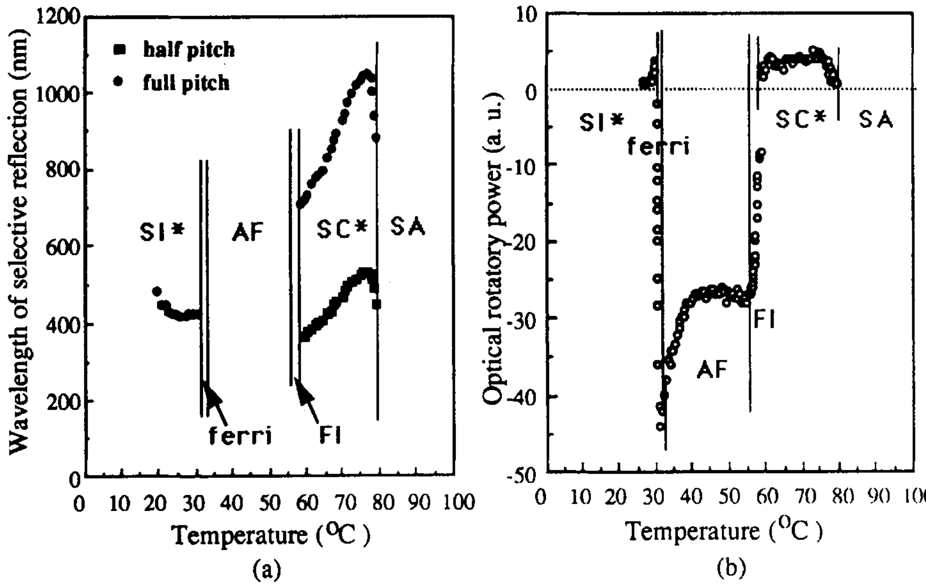


FIGURE 3. Temperature dependence of the selective refrection peaks and the optical rotatary power.

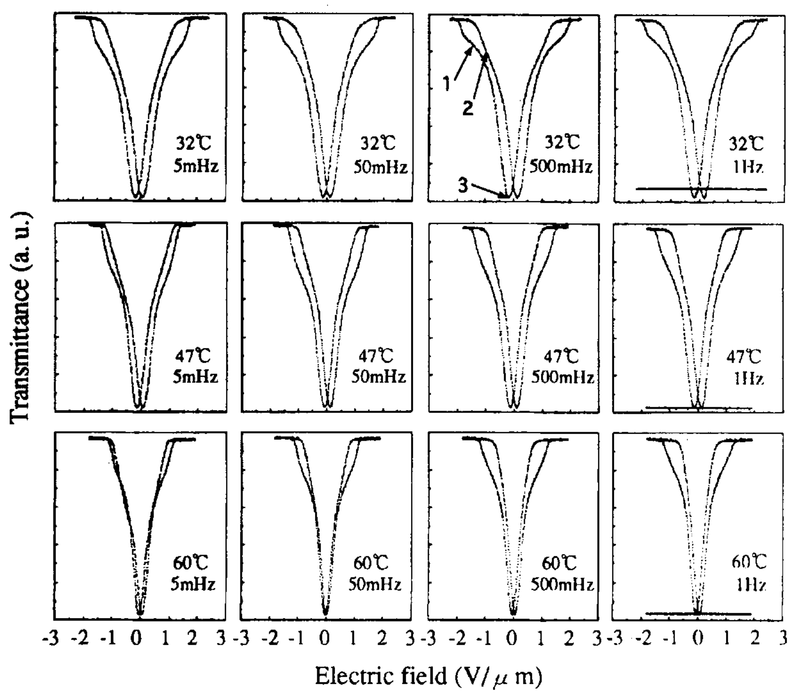


FIGURE 4. Transmittance vs electric field curve obtained in a homogeneous cell under crossed polarizers by applying a field of triangular waveform.

polar interaction between the sample and the substrate interfaces also very much decreases. The interfaces may induce the tilting randomization, breaking the reduced intrinsic tilting correlation of the sample and forcing the molecules to tilt randomly to the right or to the left from layer to layer. Such an interface-induced type of TLAF must be responsible for the present nearly V-shaped switching. The incomplete breaking results in the stabilization of some ferrielectric orderings which are observed as steps and in the W-shaped switching due to the slower randomization process; in a possible intrinsic type TLAF the randomization process is considered to be complete and fast.[5-7]

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