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## Liquid Crystals

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# Light shutters from antiferroelectric liquid crystals of bent-shaped molecules

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We present novel scattering-type displays using antiferroelectric smectic phases of liquid crystals of bent-shaped molecules. There can be two distinct states ('racemic' and 'chiral') that work in opposite ways. The racemic structure is scattering in the OFF state and is optically clear under electric fields. The chiral structure is transparent at zero fields and is scattering in the field ON state. These two structures may be reversibly interchanged, implying their use in devices that consume energy only during switching from one stable state to the other.

## 1. Introduction

Scattering-type liquid crystal devices have been known since the 1930s when Fréedericksz and Tsetkov observed dynamic light scattering in highly conductive nematics [1]. The first liquid crystal displays in wristwatches were developed on this basis [2]. Scattering can also occur in insulating liquid crystals; examples include polymer dispersed liquid crystals (PDLCs) [3], polymer network liquid crystals (PNLCs) [4], smectic A (SmA) [5] and SmC\* [6] materials. Very recently scattering mode displays were proposed also for antiferroelectric SmC<sub>A</sub>\* liquid crystals of rod-shape molecules with a 45° director tilt angle [7].

In this paper we present completely new types of antiferroelectric smectic displays of bent-shaped molecules, where the scattering effect is based on spontaneous symmetry breaking effects. These novel displays have important advantages over the previous examples.

It has been known since 1996 that bent-shaped molecules form polar smectic layers [8]. The layers lose reflection symmetry, i.e. each layer becomes chiral [9] if, in addition to polar packing, the molecules also adopt a uniform tilt about the layer polarization. The layer and director structures of this SmCP phase (P stands for polar) are represented in figure 1. The structure is called *racemic* if the chirality alternates in the adjacent layers, and is called *chiral* if the neighbouring layers have the same handedness in macroscopic ranges. Most of the known SmCP phases have antiferroelectric (AFE)

ground states, but the AFE arrangement can be switched to ferroelectric (FE) by external electric fields. The *racemic* AFE phase is synclitic, i.e. the molecules of adjacent layers tilt in the same direction. The racemic FE state has an anticlinic structure in which the optical axis is parallel to the layer normal independent of the sign of the external field.

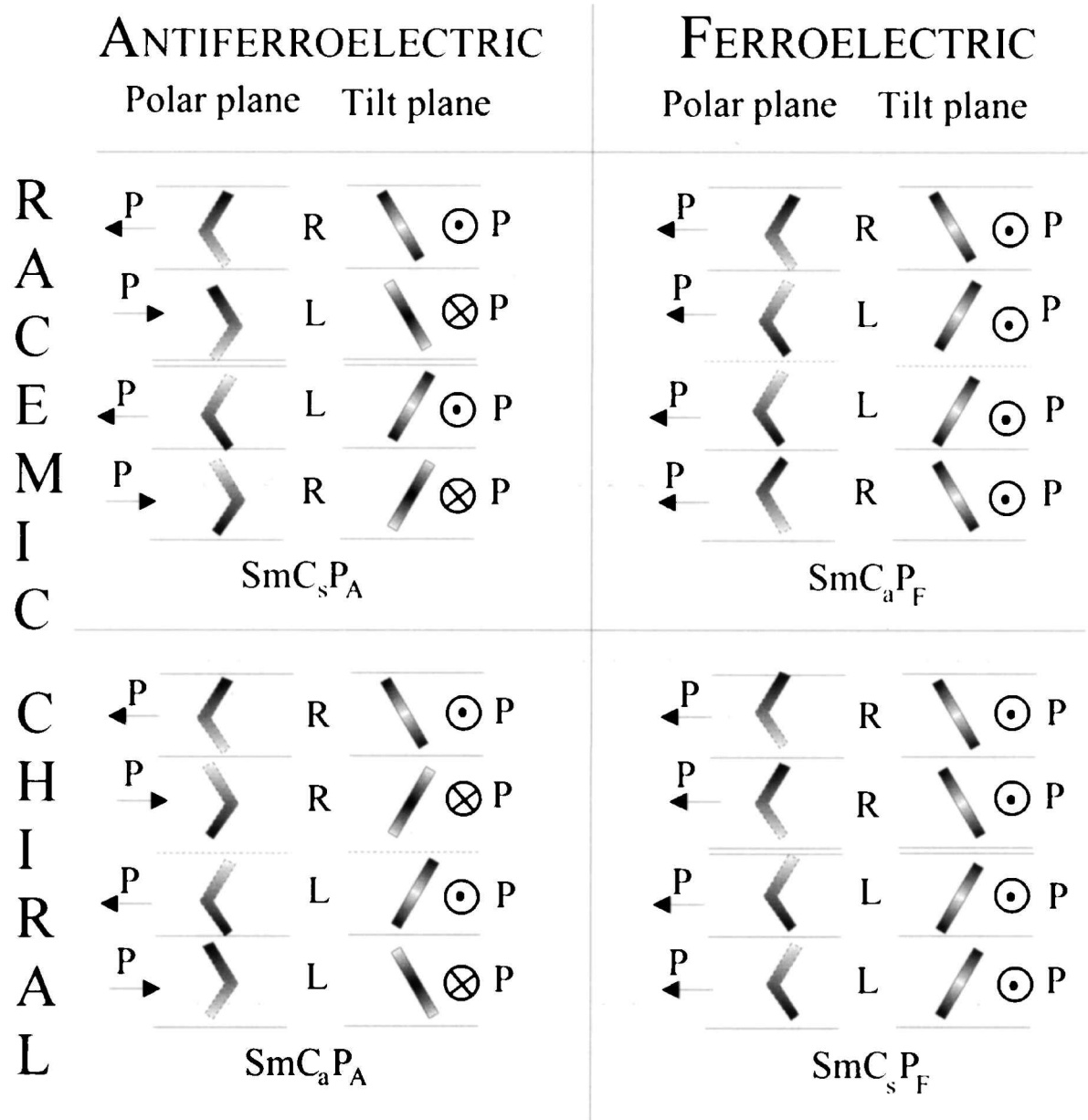
Regardless of the handedness, the chiral domains in the AFE state are anticlinic and the optical axes are parallel to the layer normal. In the chiral FE state the director structures become synclitic and, depending on the sign of the electric field, the optical axes make angles  $\pm\theta$  with the layer normal. It was assumed [9] that the chirality of the layers is conserved during the AFE  $\rightarrow$  FE transition, although under long term application of strong fields a gradual change of the layer chirality could be observed [10, 11].

This paper is based on the observation that the synclitic states (AFE state of the racemic and FE state of the chiral structures) scatter light, whereas the anticlinic states (FE state of the racemic and AFE state of the chiral structures) are transparent.

## 2. Results

We studied four materials. Two of them (R1, R2) consist of only non-chiral molecules, one (R3) is a racemic mixture with chiral dopants and one (C1) contains enantiomeric chiral dopant. R1, 1,3-phenylene bis[4-(4-tetradecylphenyliminomethyl)benzoate], [12] has a SmCP phase between 130 and 153°C. R2 is a mixture of two banana-shaped compounds and has

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$\text{SmC}_s$  : synclinic tilted smectic

$\text{SmC}_a$  : anticlinic tilted smectic

$\text{P}_A$  : polar antiferroelectric

$\text{P}_F$  : polar ferroelectric

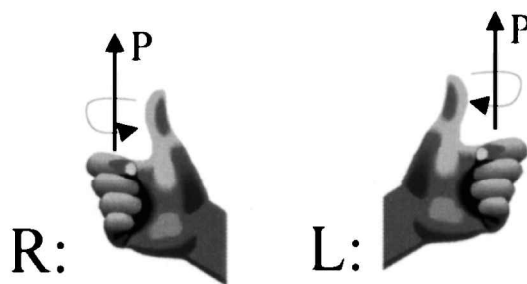


Figure 1. Orthogonal views of the SmCP phase of non-chiral banana-shaped molecules in antiferroelectric (AFE) and ferroelectric (FE) states (left and right columns). The ‘Polar plane’ contains the layer normal and the layer polarization ( $P$ ), whereas the ‘Tilt plane’ is perpendicular to  $P$ . The molecular plane is tilted with respect to the layer normal. The shading illustrates the bend and tilt of the molecules (brighter parts are closer to the reader).  $R(L)$  is the chirality descriptor corresponding to Right(Left)-handed layer conformations.  $\cdots\cdots$  indicates synclinic interfaces in anticlinic state;  $\equiv$  represents defect walls separating synclinic layers with opposite tilt directions.

a wide SmCP phase range from 130°C to room temperature. The first component of R2 is 1,3-phenylene bis-[4-(3-fluoro-4-*n*-decyloxyphenyl)iminomethyl]benzoate [13] having a 'B7' phase between 119 and 159°C ('B7' forms helical filaments cooling from the isotropic melt). The other component [14] has a nematic phase between 72 and 124°C. R3 is the same as R1, but doped with 0.75 wt % of ZLI 811 and 0.75 wt % of ZLI 3786 (both from E. Merck Germany), which are left-, and right-handed chiral dopants, respectively. The chemical structures of the dopants are identical except that they are optical antipodes. Material C1 is the same as R3, except that it contains only 1.5 wt % of the left-handed dopant ZLI 811.

The studies were carried out in commercially available sample cells (4  $\mu\text{m}$  cells from Displaytech; 5  $\mu\text{m}$  and 10  $\mu\text{m}$  thick cells from EHC, Japan) with rubbed polyimide alignment coatings. In films of R1, R2 and R3 a racemic AFE structure appears on cooling from the isotropic melt, i.e. the textures are not sensitive to the sign of the external electric fields. At zero fields the films are opaque. At increasing fields the transmitted light intensity is increasing, especially where it switches to the racemic FE state. At higher fields the transmittance decreases and the film becomes opaque again. By simultaneous observation of the electro-optical switching between crossed polarizers under square wave electric fields, we verified that during these changes the racemic FE structure eventually transfers to the chiral FE state. Such a transformation has already been observed for R1 [10] and was understood as the preference for synclinc interlayer interactions [11]. At decreasing fields the transmittance increases as the texture switches back to the chiral AFE state. In R1 and R3 the chiral FE state can be transformed back to the racemic FE state in a few seconds by changing the waveform from square wave to triangular, whereas R2 remains chiral. An example of the field dependence of the transmittance of R2 is shown in figure 2.

As the scattering in the chiral AFE and racemic FE states is negligible these results indicate that the bent-core materials can be used for light shutters. It is demonstrated in figure 3, where a black-and-white text is shown behind a 10  $\mu\text{m}$  film being in the chiral state. The text is fully visible in virtually any directions in the chiral AFE (OFF) state and completely blurred at the chiral FE (ON) state. The switching time increases as the temperature decreases. The temperature dependence of the switching time for  $E = 12 \text{ V } \mu\text{m}^{-1}$  fields is shown in figure 4. It is remarkable that the switching time is below 100  $\mu\text{s}$ , even more than 50°C below the clearing point. In contrast to observations on R1, R2 and R3, scattering is negligible in C1, which is uniformly chiral due to the chiral dopant. This proves that the scattering

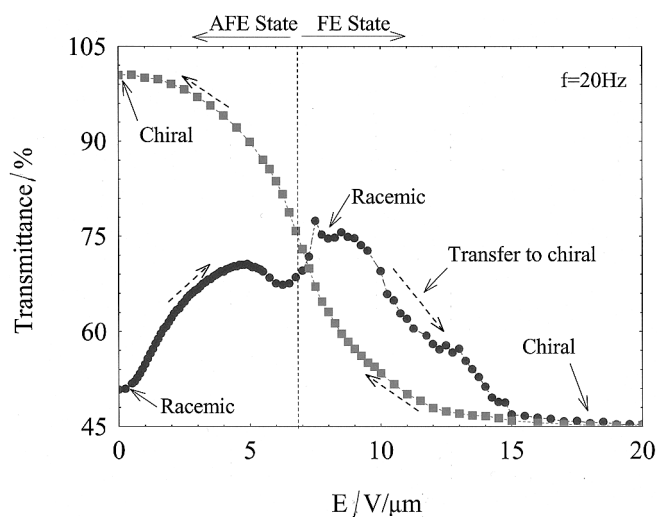


Figure 2. Electric field (square waveform from  $f = 20 \text{ Hz}$ ) dependence of the transmitted light intensity of R2 ( $d = 4 \mu\text{m}$ ,  $T = 70^\circ\text{C}$ ,  $\lambda = 450 \text{ nm}$ ). The transmittance is normalized so that the maximum intensity is 100%; actually this intensity is 96% of the light intensity transmitted in the isotropic phase.

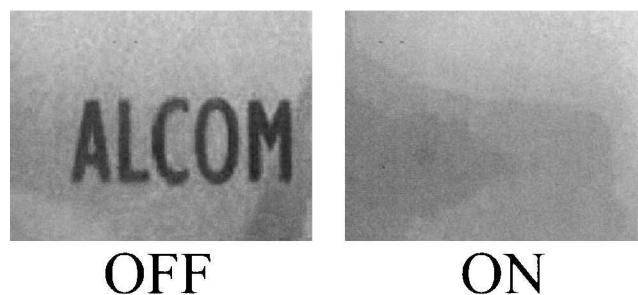


Figure 3. Electrically switchable light shutter in the chiral SmCP phase of non-chiral banana-shaped molecules. Pictures illustrate 1  $\text{cm}^2$  areas of a 10  $\mu\text{m}$  thick film of R2 at room temperature. At zero fields the film is transparent and the text 'ALCOM' placed behind the cell is visible. At fields  $E > 8 \text{ V } \mu\text{m}^{-1}$  the film is opaque and the text blurred.

is connected with the coexistence of left- and right-handed domains. Chiral textures of C1 and R3 are presented in figure 5. Textures of C1 are smooth both in the ferroelectric and in the antiferroelectric states, figure 5(a), whereas textures of R3 contain small domains in the ferroelectric state, figures 5(b) and 5(c). Films are opaque when the director structures are synclinc (racemic AFE and chiral FE states), while the structures are transparent if the director structures are anticlinc (chiral AFE and racemic FE states). This means that the differently tilted synclinc domains are separated by defects, which are observable even without polarizers, figure 5(c). Defects cause light scattering because the refractive index of the defect is different from the uniform

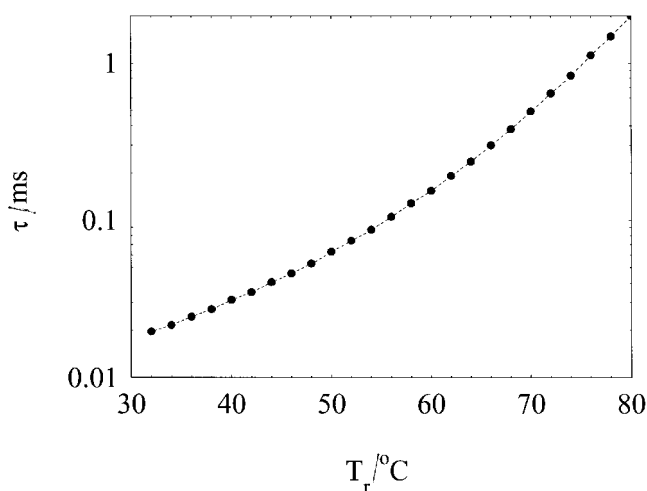


Figure 4. Switching time  $\tau$  as a function of relative temperature  $T_r$  at  $\mathbf{E} = 12 \text{ V } \mu\text{m}^{-1}$  fields.  $T_r = T_c - T$ , where  $T_c$  is the isotropic-SmCP transition temperature.

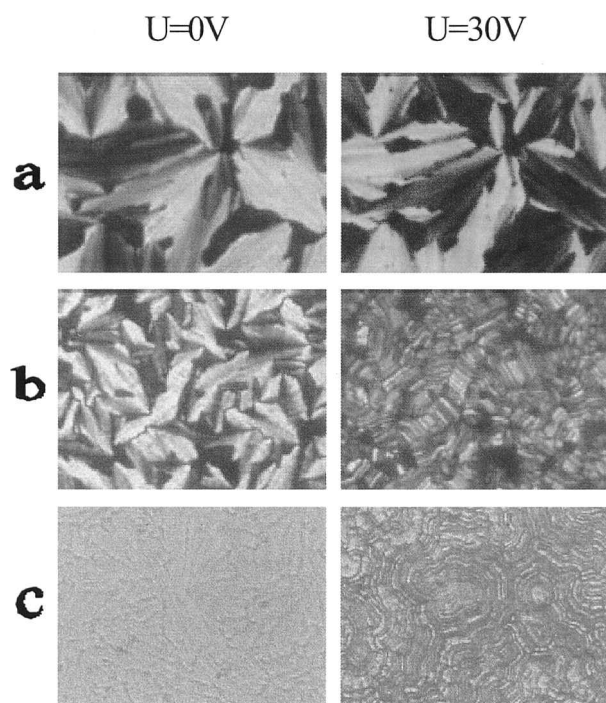


Figure 5. Textures of  $4 \mu\text{m}$  cells of R3 and Cl at  $T = 123^\circ\text{C}$  at 0 V (left-hand side) and at 30 V (right-hand side). (a) Cl between crossed polarizers; (b) R3 between crossed polarizers; (c) R3 without polarizers. The pictures represent  $100 \times 70 \mu\text{m}^2$  areas.

areas. There are no observable defects between the anticlinic domains (see figure 1) and these states are transparent. Although, due to the imperfect layer alignment, there are focal-conic type defects even in the anticlinic structures, the number of these defects is much less than of those which appear due to spontaneous symmetry breaking. Accordingly they do not cause

significant scattering and the transmittances in the anticlinic smectic states are only a few % weaker than in the isotropic phase.

The observations that the opaque racemic AFE and the transparent chiral AFE (OFF) states can be stable and can be interchanged, suggest their use in optical storage devices. For example we can envision privacy windows that consume energy only during the transformation from one state to the other. This transformation requires about the same time as needed to turn over one page in a book. Consequently such devices could be useful in future electronic newspapers, which do not use energy in the storage mode, but could also be transferred to fast switching modes.

Depending on the states where the fast switching takes place, two display modes can be distinguished, although both are due to spontaneous symmetry-breaking processes.

- (1) *Tilt Separation Mode (TSM-LCD)*. This mode exists only in the racemic state of banana-shaped molecules. The switching takes place between the synclinic racemic AFE (OFF) and anticlinic racemic FE (ON) states, i.e. the film is opaque at zero fields and transparent, in virtually any direction, under electric fields. The scattering is based on the spontaneous separation into oppositely tilted domains due to the equivalences of the tilt directions.
- (2) *Chiral Separation Mode (CSM-LCD)*. In this mode the material has chiral structure and the film is transparent at zero fields exhibiting anticlinic AFE structure and opaque under electric fields in the synclinic FE state. The scattering is due to the overall racemic nature of the phase, which results in the coexistence of left and right-handed synclinic domains separated by defect walls.

The spontaneous symmetry-breaking mechanisms that govern the domain and defect formations will be the subject of future studies. These studies may not only lead to superior and versatile displays, but will also contribute to our understanding of symmetry-breaking processes.

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