Fast Electro-Optical Switching of Dichroic Dye-Doped Antiferroelectric Liquid Crystals Without Polarizers

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We investigate alignment and electro-optical properties of a room temperature dye-doped antiferroelectric liquid-crystal mixture. We achieve uniform alignment on a macroscopic scale of thin cells with the combination of proper surface alignment and electric field treatment. We also successfully demonstrate that two films of dye-doped antiferroelectric liquid crystals in their anticlinic chiral smectic C phase can be used to switch the transmitted light intensity between dark and bright states without the need for polarizer filters. We also show that one can get either normally dark or bright states. Normally dark states can be useful in a number of applications such as in privacy windows or smart refrigerators. A normally transparent display has applications in a plethora of other areas, such as navigation systems, built-in windshields, in goggles, or smart windows.

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I. INTRODUCTION

It was realized by Heilmeier *et al*. [1,2] at the end of the 1960s, that pleochroic dyes align along the liquid-crystal (LC) director, thus leading to color change when the LC director is switched by applied electric fields. This switching required only one polarizer instead of the two polarizers that are usually used in field-effect LCDs without dye. An improved guest-host system presented by White and Taylor [3] utilized unpolarized light and dichroic azodyes, which permit the device to operate in the reflective mode with very good brightness in the clear state and an acceptable contrast ratio. Further improvement was the use of black dyes allowing black-and-white switching [4]. Since then, dye-doped guest-host nematic (N) [1,4–8], cholesteric (N*) [9–12], and polymer-dispersed liquid crystals (PDLC) [13–17] have been extensively used for polarizer-free electro-optical devices. In all these displays, the director is switched between random, helical, or planar (the director is parallel to film surface) and homeotropic (the director is perpendicular to the substrate) alignments. In planar alignment, the electric field component of unpolarized light is parallel to the dichroic dye molecules and is absorbed by the dye molecules, resulting in a dark image. In homeotropic alignment, the average orientation of the dyes is perpendicular to the electric vector of light; consequently, they do not absorb light, leading to a bright image. In practice, due to director fluctuations and partial director order (order parameter, $S < 0.8$), the contrast is limited to about 10:1. This is about the same as the contrast of a printed text, so it is acceptable for various applications. The limiting factor is rather the switching time that is limited by the nematic rotational viscosity to typically well above 1 ms.

Liquid crystals that can switch below 1 ms are the ferroelectric (chiral rod-shape SmC* [18], achiral bent-shape $\mathrm{SmCP_F}$ [19] and $\mathrm{SmAP_F}$ [20]), and antiferroelectric (chiral rod-shape $\mathrm{SmC_A}$* [21] or bent-shape $\mathrm{SmCP_A}$ [22,23]) tilted smectic LC materials. Due to constant layer spacing requirements of smectic liquid crystals, the director, however, can only rotate on a tilt cone and for both positive and negative fields, the director is basically parallel to the substrate. Furthermore, fine optical properties rely on good alignment of smectic materials, which is very

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difficult. In spite of this, guest-host effects in SmA and SmC* LC have been studied, but mainly to understand the effect of dyes on thermal effects [24,25], surface anchoring [26], switching properties (between crossed polarizers) [27], and dielectric properties [28–31]. To the best of our knowledge, electro-optical switching of ferroelectric (FE) and antiferroelectric (AFE) dye-doped liquid crystals has not been experimentally studied for polarizer-free electro-optical devices.

In this paper, we demonstrate electro-optical switching of a room temperature AFE LC mixture, both in single-layer configurations with the use of one polarizer, and in double-layer configurations without the need for any polarizer. Although the contrast is not yet optimized due to the relatively low tilt angle, we are able to achieve perfect alignment in the macroscopic scale and show that the switching under an ac field is below 1 ms, which should significantly broaden the applications of guest-host-mode LC displays.

II. MATERIALS AND METHODS

For our studies, a well-known room temperature AFE LC mixture CS4000 from Chisso Corporation (now, Japan New Chisso) doped with either a cyan dichroic dye (G-472 from Hayashibara Chemicals) or black dichroic dye (S-428 from Mitsui Chemicals) is used. The black dye comprises a mixture of a red azo dye, a yellow azo dye, and a blue anthraquinone dye in 16 wt %, 7 wt %, and 77 wt % quantities, respectively. The phase sequence, alignment properties, tilt angle, polarization, and switching time measurements of CS4000 have already been published by Le Bourhis *et al.* [32]. The phase sequence of the pure CS4000 was reported as Isotropic (98.5 °C) SmA, (83 °C) SmC$_\alpha$*, (81.2 °C) SmC*, (79.9 °C) SmC$_\gamma$*, (79 °C) SmC$_A$*, (−10 °C) Crystal.

The dyes are mixed in CS4000 by dissolving a proper amount of dye and liquid crystal in chloroform, followed by complete evaporation of chloroform. No phase separation is found either in 2 wt % G-472 or up to 5 wt % S-428 mixtures, and the transition temperatures are found to be suppressed only by less than 1 °C. The mixtures of blue and black dye-doped AFE LC are then capillary filled into 3, 5, and 12 μm thick sandwiched cells in a vacuum oven at 120 °C. The glass substrates are treated with transparent ITO, spin-coated with polyimide (PI-2555), and rubbed unidirectionally to promote planar alignment. As a control, 3 and 12 μm thick pure CS4000 samples are also prepared and studied.

III. RESULTS

Although the director structure and optical properties of AFE liquid crystals have been explained in a number of textbooks [33,34], to understand our results, first, we briefly summarize the most important features in Fig. 1. There we illustrate the director structure and their optics looking from the top on a film with a bookshelf alignment (smectic layers are normal to the substrate) when no field is applied, and under sufficiently large fields applied in opposite directions. In this view, lines illustrate the smectic layers. At zero-applied fields, the average orientation of the molecules (illustrated by elongated ellipses) is tilting alternatingly from one layer to the other and the spontaneous polarizations points in and out of the plane of the drawing. The effective birefringence of this structure is small, and the index ellipsoid is fat with the long axis along the layer normal (vertical direction in the drawing). In fact, the AFE state will appear optically isotropic if the tilt angle is

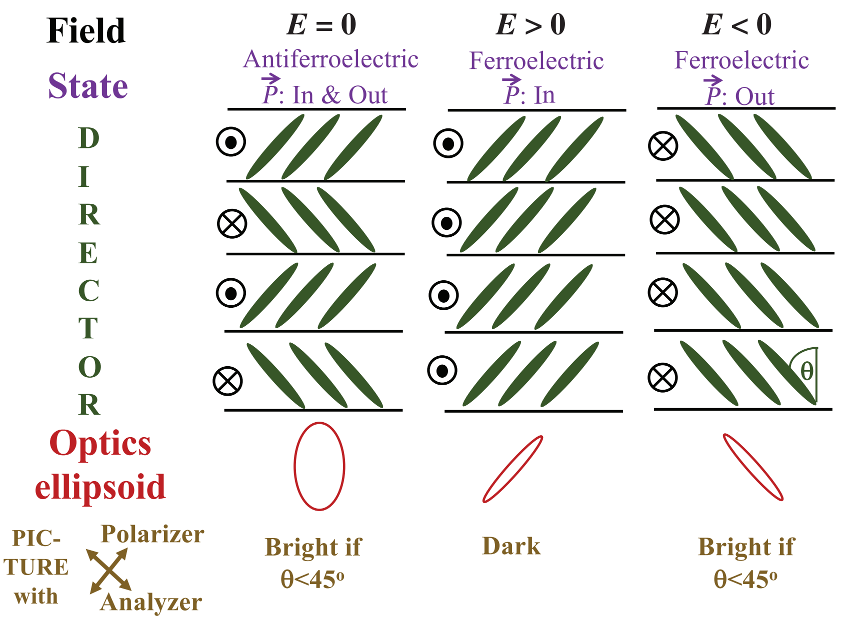

FIG. 1. Illustration of the structure of the AFE SmC$_A$* liquid crystals viewed from the direction normal to the plane of drawing.

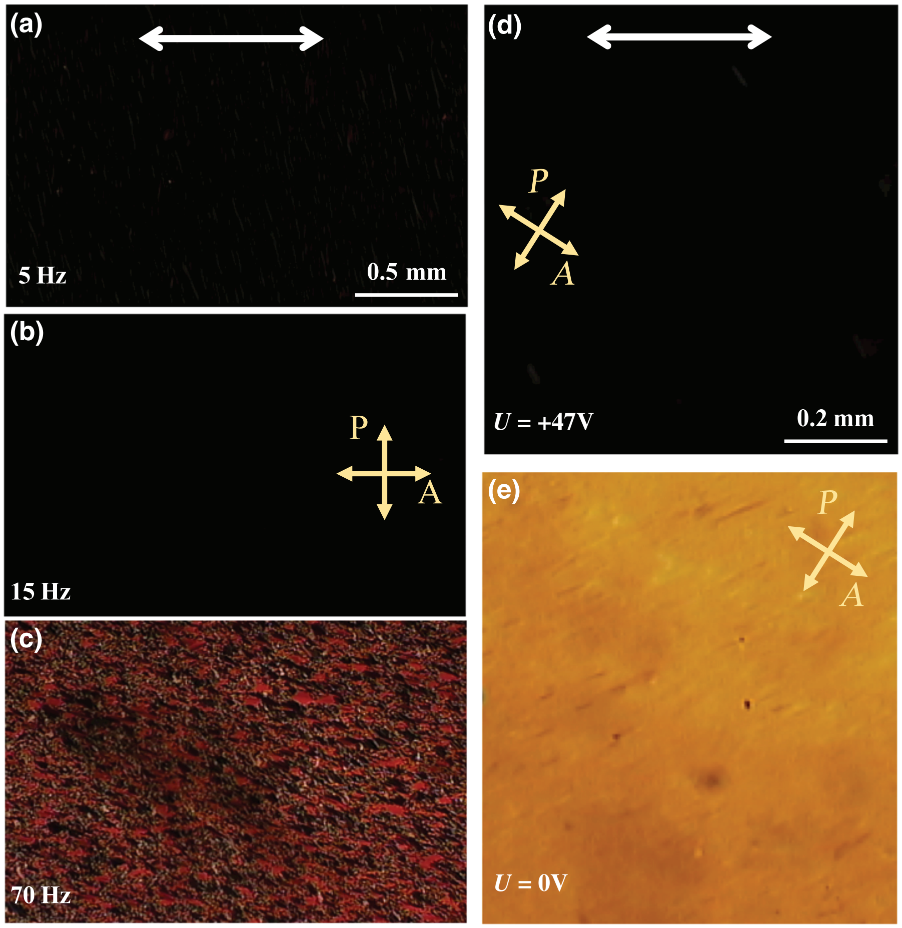

FIG. 2. Illustration of the combined rubbed surface and ac electric field treatment during cooling a 3-μm CS4000 + 5 wt% S-428 sample from the I-SmA phase transition through the SmC* down to the $\mathrm{SmC_A}$* phase. (a)–(c) Illustration of the effect of $U = 90\ V_{p-p}$ square-wave field at different frequencies. Pictures are taken at zero fields at room temperature rotating the crossed polarizers to find the darkest image. (a) $f = 5$ Hz; (b) $f = 15$ Hz; (c) $f = 70$ Hz. (d),(e) Effect of $U = 94\ V_{p-p}$ $f = 45$ Hz square-wave field. (d) In the FE phase, while $U_{\mathrm{dc}} = 47$ V is applied and one of the crossed polarizers is aligned along the director; (e) in the AFE state with the same crossed polarizers alignment as in (d).

precisely 45°. Such a structure is called orthoscopic and is promising for fast displays with a large viewing angle [35]. When large enough fields are applied in the viewing direction pointing in or out of the plane of the drawing, the anticlinic structure becomes synclinic, in which the director tilts uniformly left or right, depending on the direction of the applied field. In these two states, the directions of the optical axes make an angle $\pm\theta$ with respect to the layer normal. Turning the crossed polarizers so that the polarizer or analyzer is parallel to one of the optic axes (in Fig. 1 to the $E > 0$ state), this state appears black, whereas the state with the opposite FE polarization direction appears bright when $0 < \theta < 45°$. The brightest state appears when $\theta = 45°/2 = 22.5°$, whereas the state remains dark when $\theta = 45°$.

A. Alignment

To achieve the "bookshelf alignment," an electric field (square waveform) is applied onto the dye-doped AFLC cells while cooling from the SmA phase to the $\mathrm{SmC_A}$* phase at a rate of 1.0 °C/min. It is found that the quality of the alignment strongly depends on the strength and frequency of the applied square-wave field, and to a less extent, on the cooling rate. For 3-μm cells, it is found that $f = 15$ Hz is the optimal frequency. This can be seen by comparing the images of the textures obtained after applying a $U = 90\ V_{p-p}$ square-wave voltage at (a) $f = 5$ Hz; (b) $f = 15$ Hz; (c) $f = 70$ Hz frequencies [see Figs. 2(a)–2(c)], respectively. Similar results are found for 5 μm thick cells and for both dyes. Pictures are taken at zero fields at room temperature (AFE state) by rotating the crossed polarizers parallel and perpendicular to the rubbing direction to find the darkest image. The darker the image, the better is the alignment (see Fig. 1 for explanation). We can judge that the alignment obtained at 70 Hz [Fig. 2(c)] is much worse than that obtained under 5 Hz [Fig. 2(a)] and 15 Hz [Fig. 2(b)]. Careful inspection of Fig. 2(a) (5 Hz) reveals the presence of defect lines, which are completely missing in Fig. 2(b) (15 Hz).

The quality of the electro-optical switching between crossed polarizers can be judged by looking at Figs. 2(d) and (e). In these cases, the crossed polarizers are rotated by about 25° to find the darkest state when +47 V (dc) is applied on the cell. This angle is slightly smaller than the tilt angle $\theta = 27°$ measured in pure CS4000 [32]. Such a decrease in the tilt angle has been observed for dye-doped FE LCs, as well [36]. We think there are two possible reasons for the decrease of the tilt angle. (i) The slight decrease of the SmA-SmC phase-transition temperature

(T_{AC}), which therefore makes the distance from the phase transition $\Delta T = T_{AC} - T$ lower, and thus the tilt angle that is increasing with ΔT becomes slightly smaller; (ii) if the dye molecules are smaller than the average LC molecules, the dye molecules should have a smaller tilt angle with respect to the layer normal to effectively fill the layer spacing. This, therefore, will lead to an effectively smaller tilt angle. We note that the alignment is practically perfect in the macroscopic (over mm) scale, which is a significant improvement compared to the alignments published on pure CS4000 with similar methods, but with different alignment layers and different voltages applied on thicker samples [32]. Also, it is significant that no stripes are observable in contrast to the striped bookshelf textures achieved by Itoh *et al*. under the application of triangular ac fields at constant temperatures in the SmC* and SmA phases of two other AFE materials [37].

The physical mechanism of the combined electric field treatment and surface alignment is not clear yet. Since the spontaneous polarization is restricted along the layers, switching it between up and down along the field across the substrates will definitely straighten the layers along the electric field. However, the field alone does not provide a bookshelf layer structure. Additionally, the presence of a rubbing direction will distinguish the directions normal to the field, but in a tilted SmC* phase, it will still allow two equally possible orientations of the layers, namely with angle $\pm\theta$ with respect to the rubbing direction. The absence of these two directions is possibly due to the alternating flow induced by the switching of the director on a cone while switching the direction of the FE polarization [38,39]. This flow aligns the smectic layers in one direction by sweeping away domains in the wrong layer directions. Having only a small bias toward one direction (possibly due to surface pretilt induced by unidirectional rubbing) may also facilitate the complete disappearance of the unfavorable layer direction.

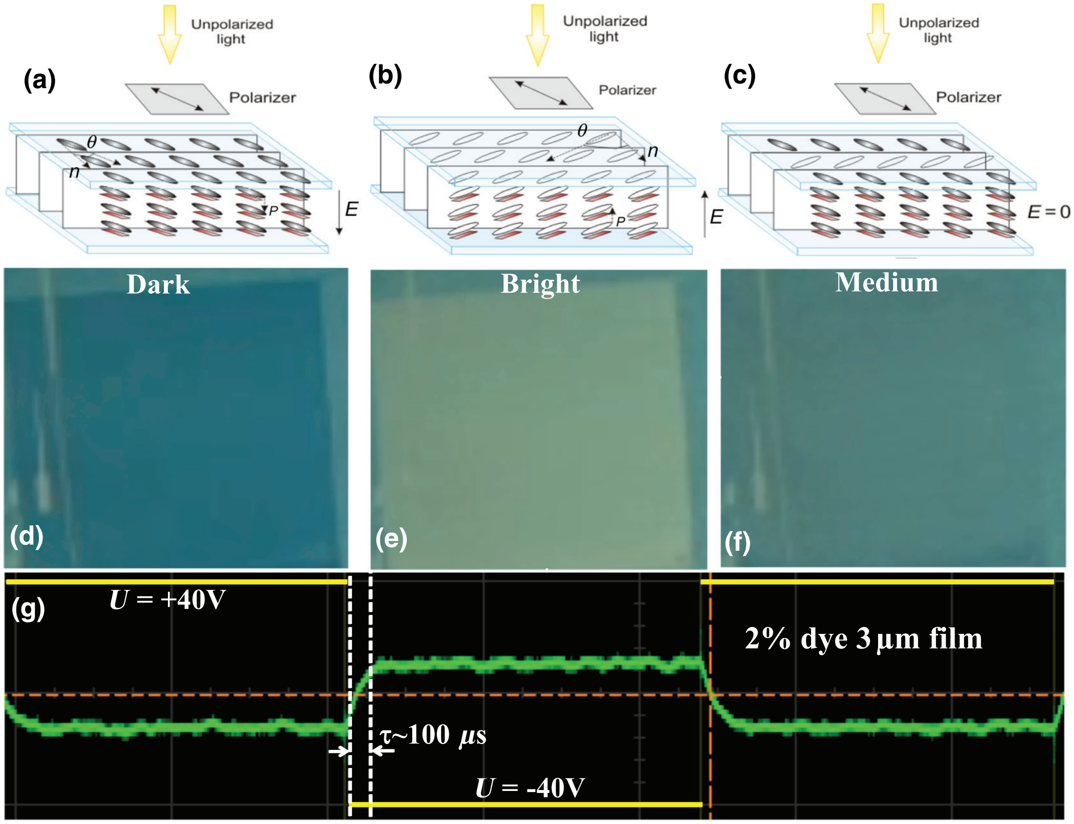

FIG. 3. Explanation of the switching principle of AFLC guest-host system using one polarizer. (a)–(c) Sketch of the director structures when fields applied across the film (FE states) and when the field is turned off (AFE state). (a) Applying field in the direction where the director and the dye molecules tilt along the polarizer direction. (b) Reversing the field, the director and the dye molecules make an angle equal to twice the tilt angle (2θ). Note this state would be the brightest when $2\theta = 90°$, i.e., $\theta = 45°$. (c) When the field is turned off, the average director and the average orientation of the dye molecules make angle θ with respect to the polarizer. (d)–(f) Typical texture of 3 μm thick CS4000 with 2 wt % G-472 dye molecules in the darkest (d), brightest (e), and medium brightness (f) states corresponding to the directors shown in (a)–(c), respectively. (g) An oscillogram trace illustrating the switching between dark and bright states of CS4000 with 2 wt % G-472 at room temperature.

B. Electro-optics with one polarizer

Looking at the $E = 0$, $E > 0$, and $E < 0$ states shown in Fig. 1 with no polarizer, there would be no difference in the transmitted light intensities. However, placing the film behind (or in front of) a linear polarizer, we see contrast (see Fig. 3). The image is the darkest [Fig. 3(d)] when the polarizer is parallel to the director in one of the FE states [see Fig. 3(a)]. This is because the dye molecules, parallel to the electric vector of light coming through the linear polarizer, absorb the light. Note that the dark state is bluish in transmission using white LED backlight, because G-472 in LC solvents absorbs in the red range [40]. In this polarizer direction, the image becomes bright [Fig. 3(e)] when the polarity of the FE state is switched to the opposite direction, because now the dye molecules make an angle of 2θ with respect to the electric vector of the incoming light, and only the projection of the electric vector parallel to the dye molecules will be absorbed [Fig. 3(b)]. One can realize that this state is the brightest when the tilt angle is 45°, i.e., the director (and the dye molecules) will be orthogonal to the incoming electric vector. At zero electric field, when the director relaxes back to the anticlinic configuration, the average dye direction will be parallel to the layer normal, i.e., it will make an angle θ with respect to the electric vector of the incoming light. Since the projection of the electric vector to the average dye direction is larger, the image will be darker [Fig. 3(f)] than in Fig. 3(e). When $E = 0$, the image is brighter than shown in Fig. 3(e), consequently the brightness in Fig. 3(f) lies between the brightest and darkest states, hence the label "medium" in Fig. 3(f). Figure 3(g) shows the oscillogram trace of the switching between (a) and (b) states of CS4000 with 2 wt % G-472 at room temperature under ±40 V applied. The switching time is about 100 μs.

C. Electro-optics with two cells and no polarizer

Note that in the above examples, the dye-doped AFLC film behaves as a switchable polarizer, implying that the polarizer may be substituted by another dye-doped AFLC film. The expected switching behaviors are sketched in Fig. 4. In the top row of Fig. 4, we show the director configurations when the smectic layers (and rubbing directions) in the cells are oriented perpendicular to each other. The bottom row illustrates the situation when the smectic layers

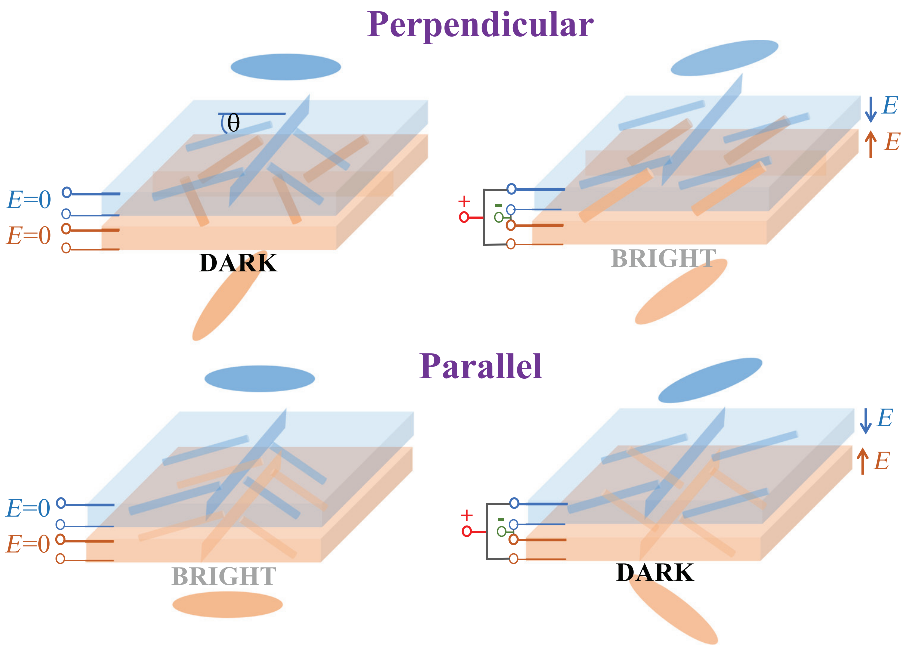

FIG. 4. Illustration of the layer and director structure of double-layer cell configurations. Top row: Smectic layers (and rubbing directions) in the cells are oriented perpendicular to each other. Bottom row: Smectic layers (and rubbing directions) in the cells are oriented parallel to each other. Left column: $E = 0$ on both cells (AFE state) with anticlinic director structures. Right column: $E > E_c$ (FE states) on both cells, but the FE polarizations point in opposite directions. Top-Left: Layers are perpendicular, and the directors are anticlinic, consequently the average direction of the dye molecules is perpendicular to each other, i.e., no light goes through (dark state). Top-Right: Layers are perpendicular to each other; the directors are synclinic in opposite direction, so the angle between the directors (direction of the dye molecules in two cells) is $90^\circ - 2\theta$. For $\theta = 45^\circ$, it would be zero resulting in maximum brightness. Bottom-Left: Layers are parallel with anticlinic director structure, the average dye directions are normal to the layers, so they are parallel to each other. Such a state is bright. Bottom right: Layers are parallel, and the directors are synclinic, but they tilt in opposite directions in the top and bottom cells. Consequently, the angle between the average dye orientations of the bottom and top cells is 2θ, which is 90° when $\theta = 45^\circ$ (darkest state).

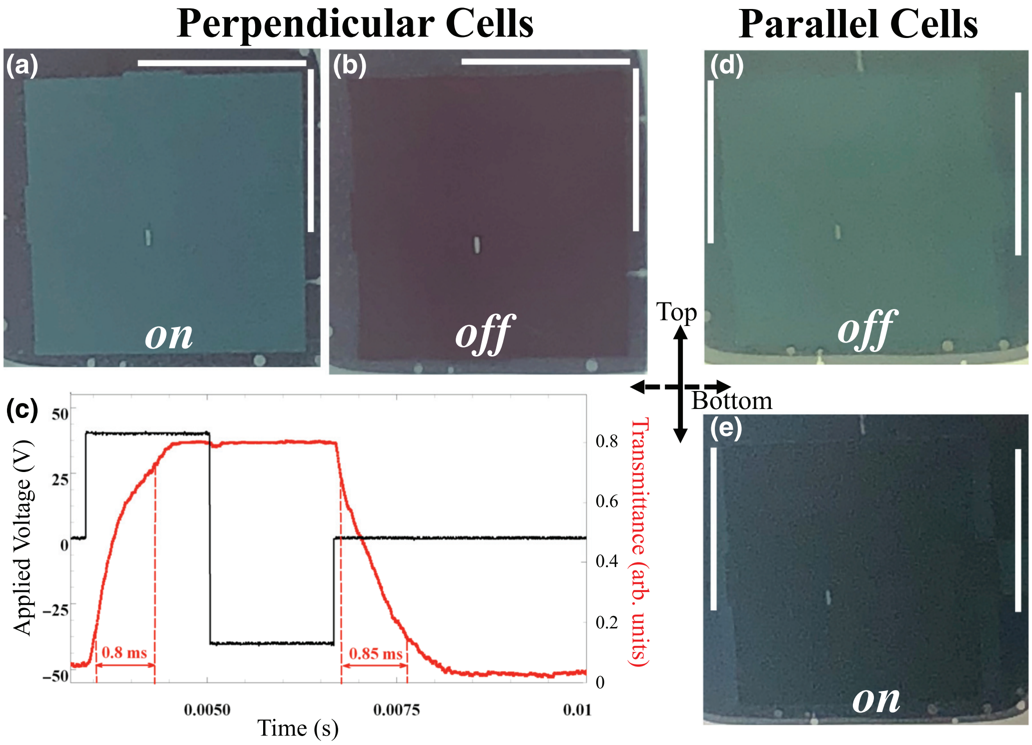

FIG. 5. Illustration of the switching of double-layered AFLC guest-host system (CS4000 + 5 wt % S-428, 5-μm gap thickness and planar alignment) without polarizers. (a) Rubbing directions of individual films make 90° with each other (along the edges of the electrodes) (a) Picture when $U = 30$ V, $f = 80$ Hz square-wave voltage is applied. (b) Field is *off.* (c) Time dependence of the applied voltage (left axis, black line) and of the transmission (right axis, red squares). (d),(e). Images of parallel cells in the antiferroelectric *off* state and (d) and in the ferroelectric *on* state (e).

(and rubbing directions) in the cells are oriented parallel to each other. The left column of Fig. 4 corresponds to AFE states in both layers ($E = 0$ on both films) with anticlinic director structures, whereas the right column illustrates the director configurations in the FE states ($E > E_c$) on both cells, but they point in opposite directions. When the smectic layers are perpendicular and the directors are anticlinic (top-left), the average directions of the dye molecules are perpendicular to each other in the top and bottom cells, consequently no light should go through (dark state). When the smectic layers are perpendicular and the directors are synclinic in opposite directions (top-right), the angle between the directors and the directions of the dye molecules in the two cells is $\alpha = 90° - 2\theta$.

For $\theta = 45°$, α would be zero, thus resulting in maximum brightness. In the situation where the layers are parallel with anticlinic director structures (bottom-left), the average dye directions are normal to the layers, so they are parallel to each other, resulting in a bright state. Finally, if the smectic layers are parallel and the directors are synclinic, but they tilt in opposite directions in the top and bottom cells (bottom-right), the angle between the average dye orientations of the bottom and top cells is $\alpha = 2\theta$, resulting in a dark state. The darkest state is realized when $\alpha = 90°$, i.e., $\theta = 45°$ (orthoscopic state).

When the smectic layers in the two cells are perpendicular to each other, we expect a normally dark state that can be switched to a bright state and vice versa, and when the orientation of the rubbing directions is parallel in the two cells, we expect a normally bright state that can be switched to dark by the electric field. In both cases, the optimum contrasts can be achieved in the orthoscopic structures when the tilt angle is 45°.

Since the AFLC material CS4000 available for us has a tilt angle much smaller than 45°, we cannot expect the best contrast, but we can achieve the darkest state possible in the perpendicular anticlinic cells (top-left configuration), since that does not rely on the 45° tilt angle. We can also demonstrate the principle and can show the switching to be less than 1 ms.

Note that polarizer-free guest-host mode has already been proposed (without experimental demonstration) for two layers of FE liquid crystals with optimum contrast requiring a 22.5° tilt angle [36]. Another important difference is that two-layer dye-doped FE LC require switching between positive and negative fields, whereas AFEs switch between field *on* and *off* states, which is compatible with the usual driving schemes and consumes much less energy.

The experimental verification of the principles is shown in Fig. 5 for two 5-μm films of CS4000 + 5 wt % S-428 mixtures on top of each other with no polarizer.

The image in Fig. 5(a) represents the bright state of the perpendicular cells in the FE state when $U = 30$ V, $f = 80$ Hz square-wave voltage is applied; while Fig. 5(b) shows the dark state of the perpendicular cells in the AFE *off* state. It can be seen that the image is really dark, darker than typical guest-host nematic cells have in homeotropic alignments. This is because the order parameter of the dyes is probably larger in the smectic than in the nematic phase (another advantage of smectic guest-host systems in addition to the faster speed). Figure 5(c) shows the time dependence of the transmittance when a bipolar square-wave field is turned *on* and *off*. Although both the AFE to FE (*on*) and the FE to the AFE (*off*) switching are slower than the switching between two FE states, they are still less than 1 ms. The switching between the normally bright *off* state and the dark *on* state of parallel cells is shown in Figs. 5(d) and 5(e). The dark state is not optimal, and it appears less dark than the *off* state of the perpendicular cells. The bright *off* state is indeed brighter than the bright *on* state of perpendicular cells, although it should be less bright than what we would get for a synclinic director with a 45° tilt.

We note that the green tone of the bright cells is due to the not perfect black dye, which has a weaker absorption in the green range. Also note that the images outside the electrode areas are between the darkest and brightest states of the electrode areas, as outside the electrodes the layers are not aligned perpendicular to the film substrates.

To summarize, we investigate the alignment and electro-optical properties of a dye-doped AFE LC. There are three important achievements. (i) We obtain good uniform alignment on a macroscopic scale with the combination of proper surface alignment and electric field treatments. (ii) We successfully demonstrate that two films of AFE LC in their SmC_A* phase can be used to switch the transmitted light intensity between dark and bright states without the need for polarizer filters. (iii) We also show that one can get either normally dark or bright states. Normally dark states can be useful, for example, in privacy windows or smart refrigerators where one does not see the food inside, but can have a peak view without opening the door. The normally transparent display can be used in navigation systems built into windshields, in ski or welding goggles, or where the voltage is triggered by light, thus attenuating it within the faction of 1 ms and preventing eye damage.

ACKNOWLEDGMENTS

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