

Field control of the surface electroclinic effect in chiral smectic-A liquid crystalsJoseph E. MacLennan,¹ David Muller,¹ Ren-Fan Shao,¹
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The surface electroclinic effect, which causes an azimuthal deviation of the layer normal from the surface rubbing direction in cells of chiral smectic-A liquid crystals, can be eliminated (and even reversed) by applying an electric field during cooling from the isotropic phase. The observed dependence of layer orientation on field strength leads to a model in which the surface electroclinic tilt results from an effective surface electric field. The experiments suggest a general method for controlling the azimuthal layer alignment of chiral smectic cells.

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I. INTRODUCTION

The electro-optic properties of liquid crystal (LC) devices depend on the uniformity of the molecular alignment, which is typically determined by rubbed polymer coatings on the surfaces of the device. While nematics are readily oriented in this way, the alignment of chiral smectics can be more challenging. For example, when the smectic A (Sm-A) phase of an achiral liquid crystal appears on cooling from the isotropic or nematic phase in a cell that is rubbed to induce parallel (homogeneous) alignment of the molecules, the smectic layers form with their layer normal \hat{z} along the rubbing direction \hat{R} , the direction favored by the molecules. In chiral Sm-A materials, however, \hat{z} generally makes a finite angle ψ with the rubbing direction because of the surface electroclinic (SEC) effect [1–6]. The magnitude and sign of the induced tilt depend both on the material properties of the liquid crystal and on the surface treatment [1] and ψ can be quite large in some cases. For example, the well-known Tokyo mixture T3 [7] grows in with the layer normal deviated by $\psi = +20^\circ$ on a nylon surface [8], and in the chiral compound W415 [9], the SEC effect is even bigger, with $\psi = -24^\circ$. In both cases, the layers grow in from the two bounding plates of a parallel-rubbed test cell with markedly different orientations [10]. Uniform layering can be obtained in cells of such materials by careful cross rubbing of the alignment coatings.

II. BACKGROUND

By analogy with the bulk electroclinic effect, in which an externally applied electric field induces a tilt of the Sm-A* director [11], Xue and Clark proposed that the angular deviation of the Sm-A* layer normal from the rubbing direction is a manifestation of a surface electroclinic effect [2], with (1)

the director tilted by θ_s ; (2) the appearance of an induced polarization; and (3) the liquid crystal undergoing a transition to a Sm-C*-like state at the surface of the cell. On cooling from the isotropic phase, the liquid crystal molecules align preferentially along the rubbing direction and the smectic layers grow in uniformly rotated so that at the surface the layer normal makes an angle ψ with the director. This layer orientation is maintained into the interior of the cell, while the LC director relaxes back to the Sm-A* orientation, the induced molecular tilt $\theta(x)$ becoming rapidly smaller with distance from the cell surface and vanishing in the bulk.

Applied electric fields have long been used to modify the layer *tilt* in cells in the chiral Sm-C* phase, for example, to transform chevron to bookshelf-type layering [12]. In materials with large surface electroclinic effect in the Sm-A* phase, on the other hand, we find that electric fields applied during cooling from the isotropic phase to the Sm-A* have a dramatic influence on the SEC tilt, allowing direct control of the layer *azimuth*. Similar behavior is observed in both W415 and T3, although T3 cells are somewhat less responsive to field treatment. The effect is described in more detail below.

III. EXPERIMENT

W415, which has the structure shown in Fig. 1, has the bulk phase sequence

$$\text{Isotropic} \xleftrightarrow{34.3^\circ\text{c}} \text{Sm-A}^* \xleftrightarrow{24.1^\circ\text{c}} \text{Sm-C}^* \xleftrightarrow{<-20^\circ\text{c}} \text{Crystal}.$$

ITO-glass cells, typically around $2 \mu\text{m}$ thick and prepared with nylon alignment layers (Elvamide 8023R), are filled by capillarity in the isotropic phase then cooled slowly (typically at $-0.1^\circ\text{C}/\text{min}$) into the Sm-A* phase. In these cells, rubbed only on the bottom plate, the smectic layers nucleate first at the rubbed surface then grow into and eventually fill the cell interior [10]. When no field is applied during cooling, the layers grow in rotated counterclockwise from the rubbing direction, as shown in Fig. 2(a), with an intrinsic layer deviation $\psi = -24^\circ$, comparable to the bulk electroclinic

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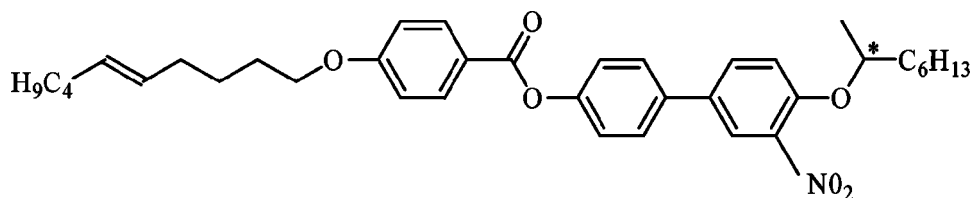


FIG. 1. Chemical structure of W415.

saturation tilt angle. In the absence of external fields, the smectic layer orientation does not change below the isotropic–Sm-A* transition, i.e., ψ is constant over the entire Sm-A*–Sm-C* temperature range. W415 has a negative spontaneous polarization in the Sm-C* phase.

We now consider the effect on the layer orientation of applying fields to the cell while cooling into the Sm-A* phase [13]. A negative dc field (pointing up, and hence aligned with the polarization associated with the native surface electroclinic tilt) only slightly increases the amount of layer rotation, an indication that the native surface tilt is near saturation. A positive field, on the other hand, reduces it. Indeed, for a sufficiently large positive field ($E_c \geq 14 \text{ V}/\mu\text{m}$) the sign of the SEC tilt is even reversed, the layers growing in deviated clockwise from the rubbing direc-

tion, as indicated in Fig. 3(a) [14]. In this way, external electric field can be used to control the layer rotation induced by the SEC effect over the range $-24^\circ < \psi < +24^\circ$. When only moderate fields are applied, the uniformity of field-treated regions of the cell is not significantly different from the virgin texture, as illustrated in Fig. 3(b). Above E_c , however, the layer orientation becomes increasingly inhomogeneous and the global optical properties of the cell (e.g., the contrast) become progressively worse, a trend indicated qualitatively by the upper curve in Fig. 3(a).

The average director orientation close to the rubbed cell surface $\langle \theta_s \rangle$ was determined in a depolarized total internal reflection (TIR) experiment, in which an evanescent wave probes primarily the first 100 nm of liquid crystal and a transmitted beam simultaneously measures the bulk optic axis orientation [15]. In a virgin W415 cell cooled with no external field, where $\psi = -24^\circ$, the average surface optic axis in the absence of applied field is found to be oriented at $\langle \theta_s \rangle = -7^\circ$ from the layer normal (and from the bulk optic axis), rotated toward the rubbing direction as shown in Fig. 3(c). In a strong positive applied field ($E = +15 \text{ V}/\mu\text{m}$), the bulk optic axis rotates to $\theta_B = +26^\circ$ under the action of the electroclinic effect, whereas the average surface optic axis only rotates to $\langle \theta_s \rangle = +23^\circ$. A negative field ($E = -15 \text{ V}/\mu\text{m}$) rotates the optic axis both in the bulk and near the surface to $\theta_B = \theta_s = -26^\circ$, approximately parallel to the rubbing direction. Optically, this behavior is consistent either with strong surface anchoring of the director parallel to the rubbing direction [with a rapid but smooth elastic relaxation of $\theta(x)$ to the bulk orientation [2]], or with a thin layer of immobile molecules adsorbed on the surface and oriented parallel to the rubbing direction (with the director jumping discontinuously to the bulk orientation θ_B) [16].

If a positive dc field is applied to the cell while cooling, reducing the layer deviation from the rubbing direction, both the bulk and apparent surface optic axes are found to be rotated from their virgin positions, as expected. Interestingly, though, their *relative orientations* are essentially unchanged, i.e., the apparent surface optic axis is still offset by $\langle \theta_s \rangle = -7^\circ$ from the bulk layer normal, itself now at $\psi = -11^\circ$ relative to the rubbing direction [17], as can be seen from Fig. 3(d). The effect of the applied field is thus to reorient the entire layer-director field of the LC about the electric field direction. The TIR experiments reveal no evidence of any molecular pretilt, i.e., the apparent optic axis is in the plane of the surface, independent of field treatment.

The SEC effect can similarly be controlled by applying unipolar, pulsed fields during cooling, the effect of the field on the layer orientation increasing with the positive duty cycle, as indicated in Fig. 4 [18].

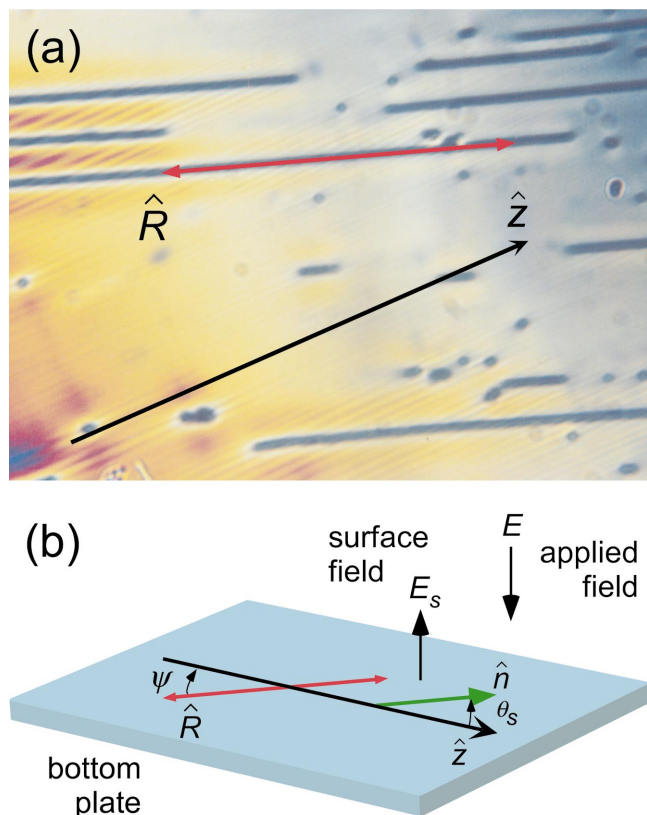


FIG. 2. (Color) (a) Polarized photomicrograph showing smectic layer formation at the isotropic–Sm-A* transition in a W415 cell (of thickness $d \sim 3.5 \mu\text{m}$) rubbed on the bottom surface only. The layers grow in with the layer normal \hat{z} , which is parallel to the ripples in this texture, deviated counterclockwise from the direction of rubbing \hat{R} , defined by the dark scratches left in the nylon alignment layer. This corresponds to $\psi < 0$, as indicated in (b).

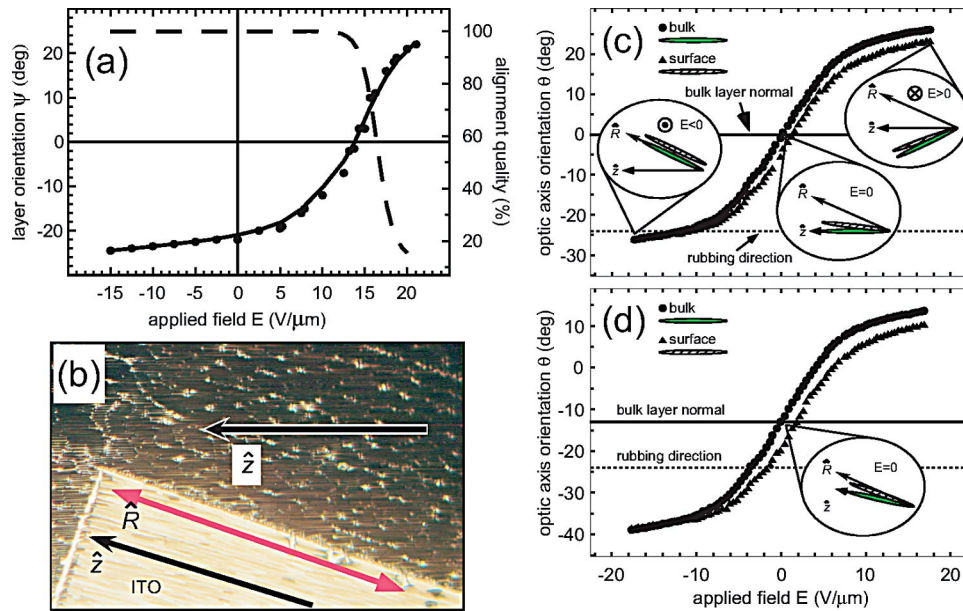


FIG. 3. (Color online) Cooling W415 with dc applied fields. (a) Final layer orientation induced by dc field while cooling (lower curve) and overall alignment quality of the cell (upper curve). (b) Typical Sm-A* cell texture after cooling with dc field. In this case, a field of $E = +13 \text{ V}/\mu\text{m}$ applied to the bright region produces a layer orientation $\psi \approx -2^\circ$. The dark area (where there are no electrodes) shows the virgin layer orientation ($\psi = -24^\circ$). The horizontal dimension of the image is about $200 \mu\text{m}$. (c) and (d) Bulk optic axis orientation measured by transmission polarized light microscopy (circles) and apparent surface optic axis orientation measured by TIR (triangles) as a function of increasing applied field. (c) In the virgin cell ($\psi = -24.5^\circ$), the bulk optic axis reorients smoothly from $\theta_B = -26^\circ$ to $\theta_B = +26^\circ$ as the field increases from $-15 \text{ V}/\mu\text{m}$ to $+15 \text{ V}/\mu\text{m}$, due to the bulk electroclinic effect. The surface optic axis is parallel to the saturated bulk at $-15 \text{ V}/\mu\text{m}$, but attains only a smaller angle ($\langle \theta_S \rangle = +23^\circ$) at $+15 \text{ V}/\mu\text{m}$. (d) When an electric field is applied during cooling, the layer deviation in the smectic-A phase is reduced ($\psi = -11^\circ$). The orientation of the surface optic axis relative to the bulk is, however, unchanged. The layer normal orientation was determined by averaging the orientations of the bulk optic axis in large positive and negative fields. This cell was cooled from the isotropic at $-0.2^\circ\text{C}/\text{min}$ and its optical properties studied at 29.9°C .

IV. DISCUSSION

These experiments demonstrate unequivocally that the SEC tilt can be controlled with an applied electric field, an effect which we explain as follows. When the liquid crystal cell is cooled from the isotropic, the material closest to the rubbed cell surface undergoes the phase transition into the Sm-A* first, with the director preferentially oriented parallel to the glass. In the absence of applied fields during cooling, the azimuthal orientation of the layers is then determined as follows.

- (1) If the cell surface was rubbed then the director orients along \hat{R} .
- (2) The LC near the surface is subject to a localized surface field which induces a surface tilt θ_s (hence a Sm-C*-like state at the surface), resulting in turn in an angular deviation of the layer normal from the rubbing direction [see Fig. 2(b)].

Although the surface field, a basic manifestation of the polar nature of the interface, may be chemical, steric, and/or electrostatic in origin, its effect on the director field may be modified by an applied electric field, similar to the bulk electroclinic effect. It is thus both convenient and natural to model the surface field as an effective local electric field E_S .

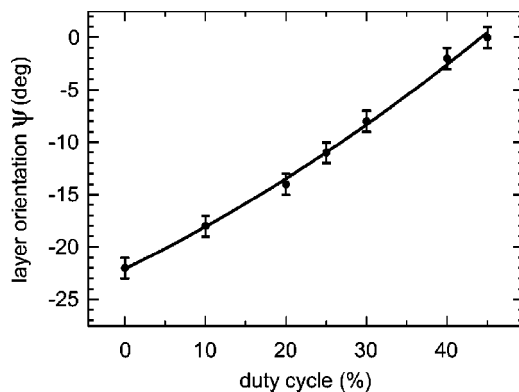


FIG. 4. Cooling W415 while applying pulsed electric fields. The layer orientation that appears in the course of the isotropic-Sm-A* phase transition depends on both the amplitude of the applied pulse train and the duty cycle, shown here. In this cell, a 2 Hz applied field of amplitude $E = +13 \text{ V}/\mu\text{m}$ achieves a layer orientation along the rubbing direction ($\psi = 0$) for a positive duty cycle of only 45%, the same modification of the SEC effect obtained when cooling in a dc field of the same magnitude. In all experiments, the applied field was removed as soon as the first typical smectic features were observed.

An external electric field E applied during cooling modifies the net field at the surface, amplifying, reducing or even reversing the sign of the surface tilt $\theta_s(E)$. In this case, the director initially orients along the rubbing direction as before. However, once the sample is cooled, the smectic layers are well established [with a corresponding deviation $\psi(E)$], and the applied field is removed, the surface director reorients under the influence of the surface field alone, relaxing toward the preferred native tilt θ_s . Since the smectic layer orientation is now fixed, the surface director in this case abandons the easy axis defined by the rubbing [24]. While this result was unexpected, it is not too surprising since it is known that, unlike nematics, the director in tilted smectics does not necessarily align with the rubbing direction anyway, for example, below the Sm-A–Sm-C phase transition [12].

V. THEORETICAL MODEL

The similarity of the bulk and surface electroclinic responses plotted in Figs. 3(c) and 3(d) strongly suggests using a theoretical approach which couples the surface and bulk electroclinic effects to obtain an expression for $\theta_s(E)$. For small electric field E , the induced electroclinic tilt in the bulk $\theta_B(E)$ is linear in E , and the mean-field density of the Sm-A* phase may be written in terms of the polarization P and tilt angle θ [5,10,11,25] as

$$F = F_A + \frac{A'}{2}\theta^2 - tE\theta + \frac{K_2}{2}\left(\frac{d\theta}{dx}\right)^2. \quad (1)$$

The Euler-Lagrange equation governing the director field,

$$K_2 \frac{d^2\theta}{dx^2} = A'\theta - \chi tE, \quad (2)$$

has the solution

$$\theta(x,E) = [\theta_s(E) - \theta_B(E)]\exp(-x/\xi) + \theta_B(E), \quad (3)$$

where $\theta_s(E)$ is the tilt at the cell surface ($x=0$), $\xi = (K_2/A')^{1/2}$ is the bulk Sm-A* penetration length, and $\theta_B(E) = tE/A'$. At the surface we assume the same restoring energy as in the bulk and postulate that the surface electroclinic tilt established at the phase transition results from an effective surface electric field E_S . The surface energy/area may then be expressed, using the notation of Eq. (1), as

$$f_s(\theta_s,E) = \left(\frac{A'}{2}\theta_s^2 - t\theta_s E_S\right)w + t\theta_s E_S v, \quad (4)$$

where w and v are the effective thicknesses over which the surface and bulk forces act. The surface and bulk tilts are constrained by the requirement of torque balance at the cell surface:

$$K_2 \left. \frac{\partial \theta}{\partial x} \right|_{x=0} = \frac{\partial f_s(\theta_s,E)}{\partial \theta_s}, \quad (5)$$

which yields the relation

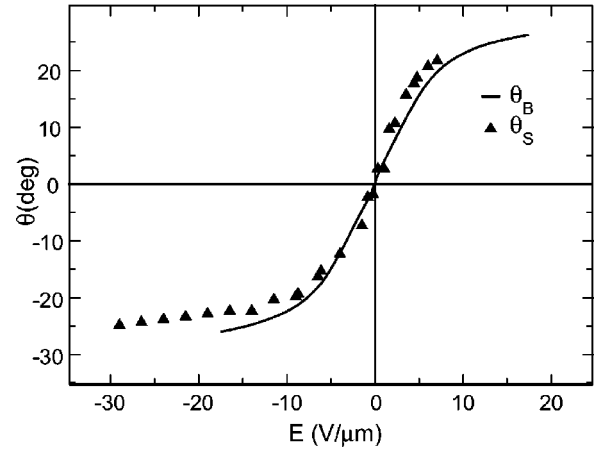


FIG. 5. Bulk (—) and surface electroclinic tilt (\blacktriangle) in a W415 cell in the Sm-A* phase. The bulk tilts are the same as in Fig. 3(c) and the shifted surface angles correspond to the data in Fig. 3(a) offset by a field of $+14 \text{ V}/\mu\text{m}$. These curves overlap quite well, with the bulk and the surface demonstrating the same linear susceptibility to small (net) electric fields.

$$\theta_s(E) = \frac{w\theta_B(E_S) + (\xi + v)\theta_B(E)}{\xi + w}. \quad (6)$$

This equation provides a model for the experimental tilt data shown in Fig. 5, in the region where θ_s is small. Here, θ_s varies linearly with field in a manner similar to the bulk, with the relative slope of the surface and bulk responses

$$R = \frac{\partial \theta_s(E)}{\partial \theta_B(E)} = \frac{\xi + v}{\xi + w} \approx 1.$$

This allows Eq. (6) to be reformulated as

$$\theta_s(E) = \theta_B(\tilde{E}_S), \quad (7)$$

implying that the director near the surface reorients linearly like the bulk but subject to the net electric field

$$\tilde{E}_S = \frac{wE_S + (\xi + v)E}{\xi + w} \approx E + \frac{wE_S}{\xi + w}. \quad (8)$$

The similarity of the bulk and surface data shown in Fig. 5 provides compelling support for the postulated model, confirming that $\theta_s(E)$ is nearly identical to the bulk electroclinic tilt $\theta_B(E)$, but shifted in field by $+14 \text{ V}/\mu\text{m}$. The magnitude of the bare surface field E_S could be determined with independent estimates of ξ and w .

Finally, we note that field control of the smectic layer orientation provides a direct means of controlling the effective birefringence of chiral Sm-A cells, suggesting possible applications in the area of electrically controlled phase plates and color filters. For example, Figs. 6(a) and 6(b) show W415 cells with stripe-patterned electrodes to which fields of differing strengths have been applied on cooling into the Sm-A* phase. Figures 6(c) and 6(d) illustrate the principle by which such cells could be field treated to form a modulated phase plate that would act as a diffractive electroclinic shutter for unpolarized light [26]. This approach provides an

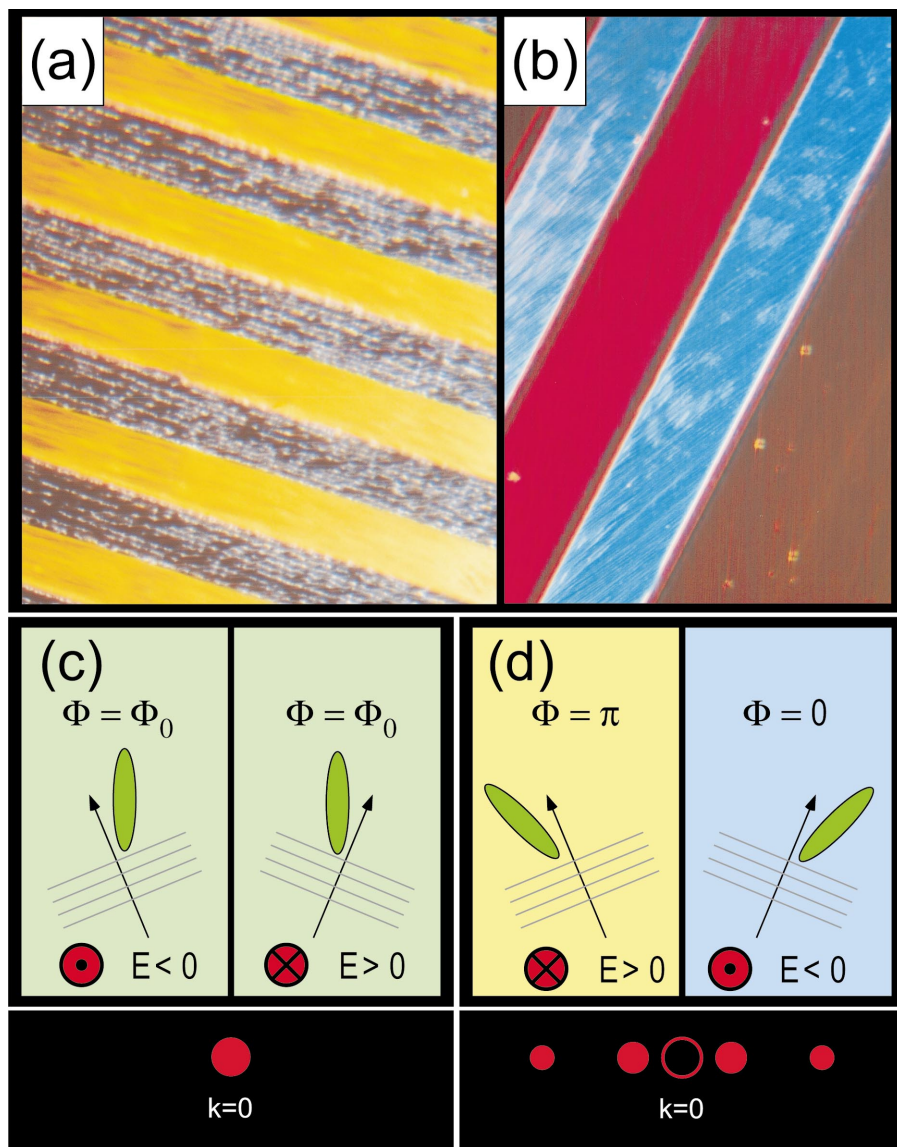


FIG. 6. (Color) (a) and (b) Polarized photomicrographs of W415 cells with stripe electrodes in the Sm-A* phase. Applying different electric fields to alternate electrodes during cooling from the isotropic phase gives a modulation of the smectic layer orientation and hence of the effective birefringence. The colors in (b) are enhanced with a phase plate. The horizontal dimension of these images is about 100 μm . (c) and (d) Proposed diffractive optical shutter for unpolarized light using a linear array of Sm-A* domains in which the layer normal orientation in adjacent pixels differs by 45° . With appropriately applied fields, the liquid crystal either causes a uniform optical phase shift $\Phi = \Phi_0$ of the transmitted beam (c), or forms a modulated phase plate, with alternating stripes providing optical phase shifts of $\Phi = 0$ and π , respectively (d). In this latter condition, the array acts as a diffraction grating, eliminating the central beam at $k=0$ as indicated.

alternative to using high tilt Sm-C* materials ($\theta \sim 45^\circ$) (which are rare and somewhat tricky to align, and require high driving voltages [27]) to achieve a 90° orientational difference of the optic axis between adjacent stripes. In general, selective control of the layer azimuth allows the design of devices with a wider range of optical tilt than is achievable by the bulk electroclinic effect on the director alone.

VI. CONCLUSION

In summary, we have demonstrated that the SEC effect determining the smectic layer orientation in cells of chiral

smectic-A liquid crystal can be controlled by applied electric fields. The magnitude of the field required to cancel the surface electroclinic effect can be directly related to an effective surface electric field localized at the liquid crystal/alignment layer interface.

ACKNOWLEDGMENTS

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- [17] The angular coordinates are consistent with the rather nonintuitive convention introduced by Nakagawa [1] and shown in Fig. 2(b): $\psi > 0$ corresponds to \hat{z} rotated clockwise from \hat{R} , while $\theta > 0$ corresponds to \hat{n} rotated counterclockwise from \hat{z} , i.e., ψ and θ have opposite senses of positive rotation.
- [18] In a fundamentally different effect, the smectic layers in W415 cells can be rotated *within* the Sm-A* phase by unipolar, pulsed fields, a phenomenon previously observed (in a different material) using asymmetric bipolar pulses [19]. In W415 the rotation can be reversed by changing the sign of E over the range $-24^\circ < \psi < +24^\circ$. In the Sm-C* phase, the layers can be rotated in either direction, depending on the bias of the applied pulsed field (the range of reorientation is limited to $-24^\circ < \delta\psi < +24^\circ$). The rotation in the Sm-C* is reversible and occurs more readily than in the Sm-A* phase but is only metastable, the layers returning to their original orientation within 10 h of the applied field being removed. Layer rotation in Sm-C* materials has been studied extensively [20–22]. It has been suggested that the origins of this effect lie in an asymmetric ratcheting of the molecules around the tilt cone [23].
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