Director Structures in Achiral Smectic C Liquid Crystal Cells: Field Induced Twist Domain Nucleation

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*The electro-optic response of two achiral smectic C liquid crystals, **W479** and **$\overline{8}$S5**, in thin surface stabilized cells has been studied using polarized light microscopy. In their virgin (field free) state, **W479** cells have "bookshelf" structure, with the smectic layers uniformly tilted with respect to the bounding glass plates, while **$\overline{8}$S5** cells have bent or "chevron" layering. Upon first cooling into the smectic C phase, both materials exhibit domains of uniform optic axis orientation. Subsequent application of an electric field leads to a Freedericksz transition and to the nucleation and growth of non-uniform domains in which the optic axis is twisted. Models for the virgin and field-induced director structures are proposed.*

I. Introduction

Researchers have made significant inroads into understanding the electro-optic behavior of chiral tilted smectics, probing the director response induced by field coupling to the ferroelectric polarization. There has been comparatively little study of the dielectric-based response of achiral tilted smectics, although there are many good reasons to explore this area. For example, as well as governing the electro-optic behavior of achiral smectics, the field-induced dielectric response is important in the chiral case at fields large enough that the ferroelectric and dielectric field-induced torques are comparable. In addition, in surface stabilized ferroelectric liquid crystal (SSFLC) cells, a.c. field coupling to dielectric torques can lock-in a molecular orientation [1] with the director stabilized either normal or parallel to the cell plates, depending on the sign of dielectric biaxiality, the former case being useful for obtaining analog electro-optic behavior in tilted smectics [2]. Understanding the dielectric response of achiral tilted smectics thus provides information relevant to a variety of smectic electro-optic applications, motivating the present study.

We have investigated two achiral smectic C (SmC) materials, **W479** and **$\overline{8}$S5**, shown in Figure 1. The temperature dependence of the smectic layer spacing of these materials through the SmA-SmC transition results in cells with different SmC layer structures: uniformly tilted layers in **W479** and chevron layers in **$\overline{8}$S5**. Both materials have positive dielectric anisotropy, so while the cell is treated to align the molecular director parallel to the bounding glass plates, the field-preferred director orientation is normal to the glass, leading to a variety of interesting field-induced orientational transitions.

II. Experimental

Optical experiments were performed with the liquid crystal in 4 µm thick, transparent capacitor cells of ITO glass, filled in the isotropic phase. The surface alignment layers were polyimide, rubbed antiparallel to give planar alignment and tilted layers in the SmC phase with a slight pretilt of the director [3, 4]. The materials studied were **W479** and **$\bar{8}$S5** [5], with the phase diagrams and layer spacings shown in Figure 1.

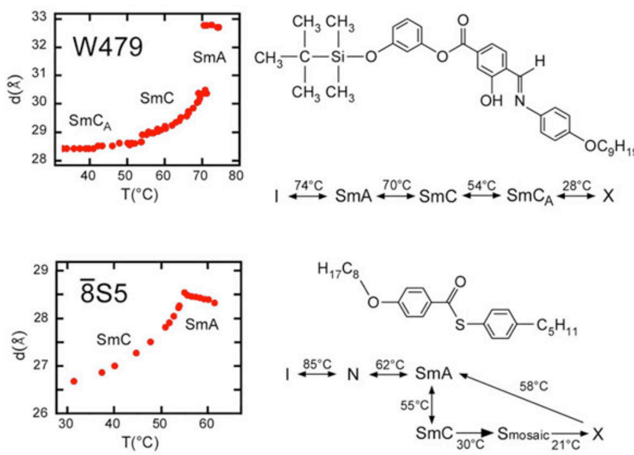

Figure 1. Structures, phase diagrams, and layer spacing dependence on temperature of W479 and $\bar{8}$S5.

Layer spacing was measured by x-ray diffraction on powder samples contained in 1-mm-diameter glass capillaries in an Instec STC200 hotstage, carried out on beamline X10A of the National Synchrotron Light Source at Brookhaven National Laboratory. **W479** exhibits a first-order SmA - SmC transition, with large jumps in the layer spacing d(T) at the transition [6], and uniformly tilted bookshelf layers. **$\bar{8}$S5**, on the other hand exhibits the classic d(T) behavior associated with a second-order SmA - SmC transition, with a continuous shrinkage of the layers as the molecules tilt from the layer normal in the SmC phase. The layers tilt to accommodate this shrinkage, leading to the formation of a chevron layer structure [7].

W479 and **$\bar{8}$S5** showed changes in texture, domain structure, and net birefringence under the influence of changing temperature T and a.c. and d.c. electric fields E. The experiments were performed on a Zeiss polarizing microscope with the samples in an Instec STC200 hotstage. Switching time measurements, made by monitoring the 10% to 90% risetime of transmitted laser intensity with a photodiode, were measured on a Tektronix 2215A oscilloscope: the optical response time was found to scale as $1/E^2$ in both liquid crystal materials, confirming that the switching is dielectric, as expected for the achiral SmC phase.

III. Results

3.1 W479: Induced Twist States in Bookshelf Cells

W479 exhibits very smooth SmC textures, and aligns well, especially when cooled slowly (~ -0.1°C / min) from the isotropic to the SmA phase.

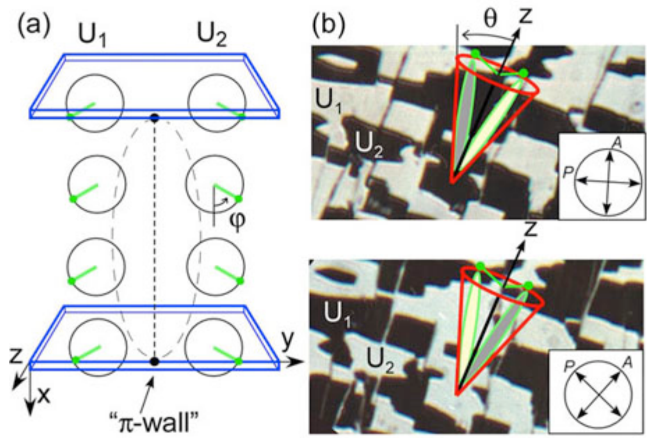

Figure 2. (a) Model for uniform planar states in tilted bookshelf SmC cell. The circles represent the open ends of the tilt cone, and the green lines the c-director orientation. The intersections of the tilt cone with the substrates mark the orientations where there is planar alignment. Between the two uniform domains is a tilted layer π-wall equivalent, where the opposite states meet. (b) DTLM images of W479 showing the same area of the cell with the crossed polarizers rotated through 2θ, producing extinction alternately in the U_2 and U_1 domains. The black (white) lines running approximately parallel to z are zigzag walls. Their internal texture is not visible at this magnification.

SmC cells, as shown in Figures 2 and 3, have (i) bookshelf layering with the layer normal z along the rubbing direction; (ii) two domains with apparently uniform but distinct optic axis orientations; and (iii) zigzag wall defect lines reminiscent of the zigzags often seen in chevron cells [8].

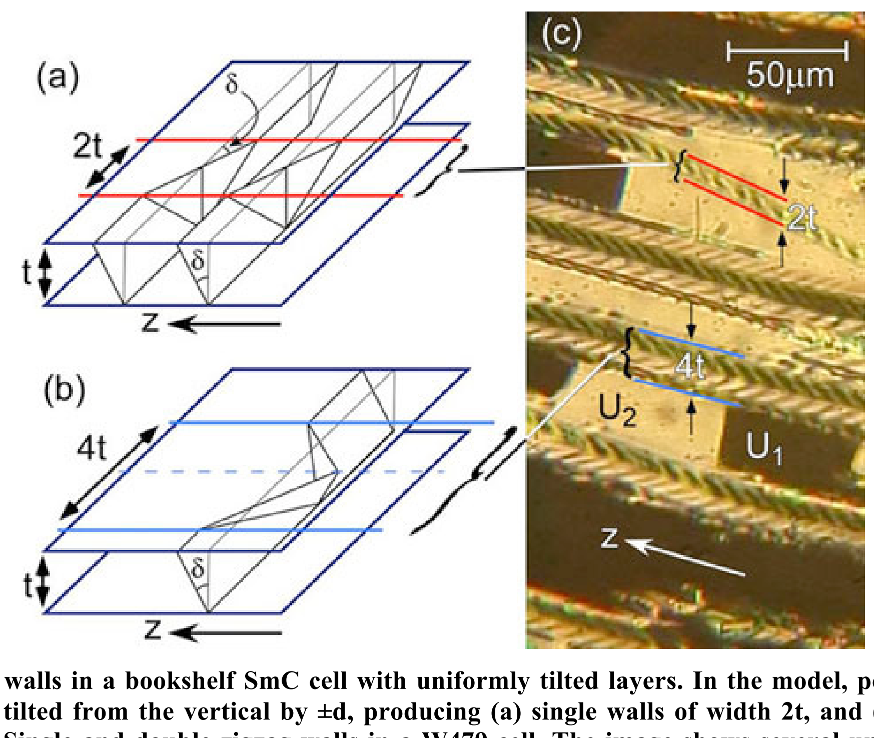

Figure 3. Zigzag walls in a bookshelf SmC cell with uniformly tilted layers. In the model, polygonal wall layer segments are tilted from the vertical by ±d, producing (a) single walls of width 2t, and (b) double walls of width 4t. (c) Single and double zigzag walls in a W479 cell. The image shows several uniform domains, U_1 and U_2, of approximately equal area, confined between zigzag walls. The internal striations of the zigzag walls are currently not understood.

These zigzag defects, as illustrated in Figure 3, appear both as single walls (marked in red) and as double walls (marked in blue). The zigzag defects mediate a change in sign of the layer tilt and are a signature of tilted bookshelf cells. The observed widths w of the zigzag defects (w=2t for a single wall and w=4t for a double wall, where t is the thickness of the cell) confirm unambiguously that the layers are uniformly tilted in the SmC phase of **W479** [9].

After cooling from the SmA phase, virgin **W479** cells exhibit two uniform SmC domain structures, U_1 and U_2, of equal area, stabilized by the preference of the director, **n**, to be planar aligned, approximately parallel to the glass at both surfaces. While in the bulk the director can adopt any azimuthal orientation φ about **z**, as shown in Figure 4, planar alignment at the surface is obtained, when $\delta < \theta$, producing two degenerate, uniform domains at only two positions on the tilt cone with opposite signs of φ (U_1, U_2 in Figure 2).

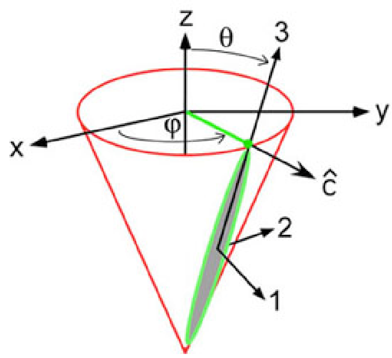

Figure 4. Smectic C tilt cone geometry. The director in the SmC phase tilted from the layer normal z, by an angle q, and has a projection c onto the smectic layer plane. With the directions of the principal components of the dielectric permittivity tensor as shown, positive dielectric anisotropy occurs when $\varepsilon_1, \varepsilon_2 < \varepsilon_3$.

The apparent tilt of the optic axis from the layer normal in this virgin condition is 16°. The uniform states are separated by π-walls [10], stabilized at the surfaces by the planar alignment condition [11]. The cells are apparently thin enough that twisted states, which would be metastable in the case of sufficiently strong surface anchoring [9], do not appear.

Application of small electric fields to the cell has little effect, but above a threshold (E ~ 3 V/μm), there is a decrease in the birefringence [12]. This is characteristic of a Freedericksz transition, confirming that the dielectric anisotropy Δε is positive, i.e., $\varepsilon_3 > \varepsilon_2, \varepsilon_1$ (see Figure 4) [13, 14]. Since the preferred director orientation is along the applied field **E**, the bulk realigns (with φ → 0, 180°), producing the distorted states D_1 and D_2 (distorted from U_1, U_2 by the alignment of the bulk with the applied field) shown in Figure 5(c)-(f), from the initial uniform states shown in Figure 2(a) and in Figure 5(b).

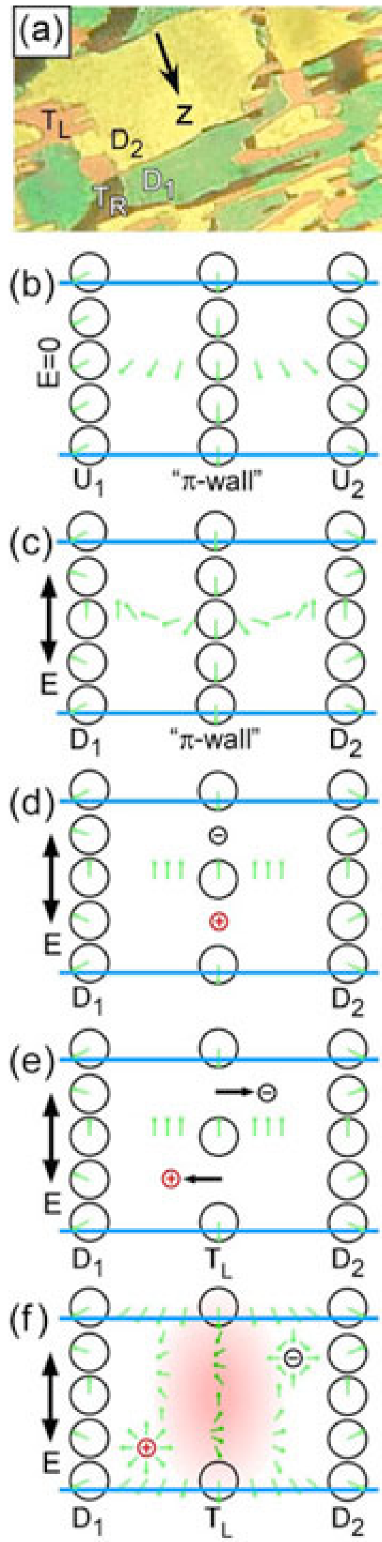

Figure 5. Field-induced twist domains in a bookshelf cell. (a) DTLM image of W479 after an electric field-induced change in layer geometry, which results in the elimination of the zigzag walls. Twisted T_R (brown) and T_L (orange) domains (which appear only temporarily at the boundaries between uniform states upon the application of an electric field in the virgin tilted bookshelf geometry), are stable in the upright bookshelf geometry shown here, remaining even after the removal of the applied field. (b)-(f) Proposed scenario for the appearance of twisted states at the D_1, D_2 boundaries in the tilted bookshelf geometry. (b) Uniform states U_1

and U_2 in the absence of field. (c) As the uniform states become deformed, the director orientation from D_1 to D_2, across the π-wall, remains continuous. (d) As the applied electric field is increased, two line defects of opposite strength (red and black circles) running parallel to the glass plates are nucleated in the director field, creating a region in the middle of the cell with ideal n alignment. (e) As the applied field is increased further, the line defects start to move laterally, introducing twist domains. (f) As the line defects move away from the initial U_1, U_2 boundary, the area between the defects possesses the twisted director field (shaded in red). It is not required that both line defects make a lateral movement, for the twist domain forms in the same way when only one defect moves; all that is required for the formation of the twist domain is that the line defects not be in vertical alignment.

This is conceptually similar to the process that occurs above the Freedericksz transition in nematic cells [14], where the director field becomes more uniform as the applied field is increased. In achiral SmC cells, however, we observed a second voltage threshold, above which twist domains, T_L and T_R, appear, growing from the π-walls that separate the two original uniform domains [15]. Under the influence of an applied field the π-walls break apart in the center of the cell, to allow the bulk to align with the field, and form two line defects moving parallel to the glass plates of the cell. These line defects can then move apart laterally with increasing field, yielding a twisted director, as shown in Figure 5(b)-(f). These twist domains disappear again after the field is removed [16].

The development of field-induced twist states is similar in regions with opposite sign of layer tilt, i.e., it is not strongly affected by the surface pretilt. However, if a very large field ($E > \sim 20$ V/μm) is applied, the zigzag walls disappear, implying that the layers have straightened up. The resulting layer structure is stable, but the induced twist domains no longer shrink when the applied field is reduced or removed.

3.2 $\bar{8}$S5: Half and Fully Twisted States in Chevron Cells

$\bar{8}$S5 has a nematic phase, which results in very good alignment in both the SmA and SmC phases. Cells of **$\bar{8}$S5** in the SmC phase have chevron layering and in their virgin state exhibit six coexisting domains, shown in Figure 6(a)-(c). Two of these are uniform (U_1 and U_2) and four are "half-twisted" (T_{Rt}, T_{Rb}, T_{Lt}, and T_{Lb}, where t and b denote whether the twist is confined to the top or the bottom part of the chevron cell, and L and R refer to the handedness) [17]. Figure 6(d) shows our model of the virgin states.

In principle, the chevron layer structure, with three bistable interfaces (two cell surfaces and the chevron interface [8]), allows eight possible states. Initially, though, only the six lowest-energy states, the uniform and half-twisted configurations, appear. The two "full-twist" states, where the director twists in both the top and bottom halves of the cell, appear only upon the application of an electric field. Thus, there are two degenerate twist domains of opposite handedness in the top half of the cell (T_{Rt} and T_{Lt}), and two degenerate domains of opposite handedness in the bottom half (T_{Rb} and T_{Lb}). The top and bottom half-twisted states are degenerate only if the cell surfaces are identical and the chevron interface is in the middle of the cell [18]. While the slight birefringence change observed across the zigzag walls indicates that the chevron interface is often not exactly in the middle of the cells [19], it is apparently close enough that twist is as likely to occur in the top half as in the bottom half of the cell.

Like **W479**, **$\bar{8}$S5** has positive dielectric anisotropy [20] and when an electric field is applied to a virgin SmC cell, here too a Freedericksz transition occurs, with the domains starting to switch at $E \sim 4$ V/μm. The initial response is different from what was observed in **W479** though. In **$\bar{8}$S5** the uniform and twisted domains respond simultaneously, sweeping across the cell to create beautiful domain patterns such as those shown in Figures 7 and 8.

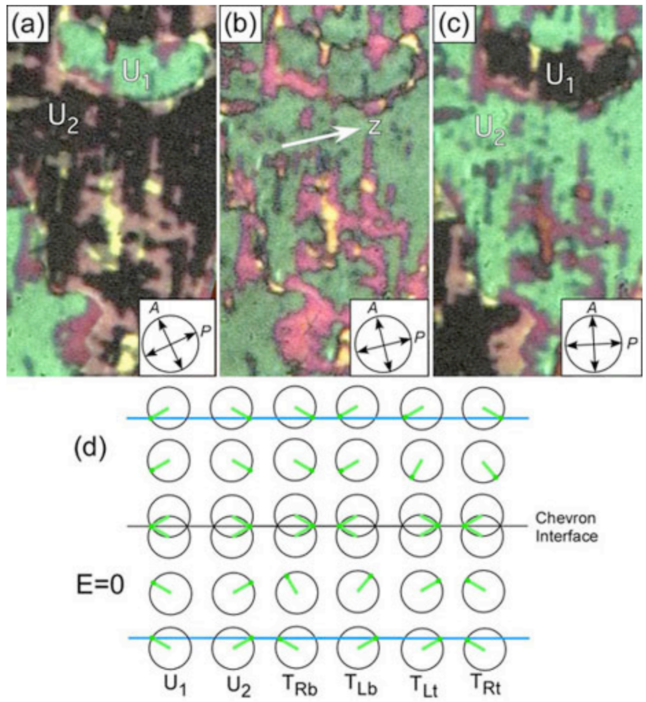

Figure 6. Uniform and twist domains in a virgin chevron cell of $\overline{8}$S5. (a)-(c) DTLM images showing the same area with differing orientations of the crossed polarizers, all in the absence of applied field. In these photomicrographs, the uniform states are green or black, while the twisted states appear in shades of pink and yellow. (a) and (c): The polarizer pair is rotated counter-clockwise and clockwise respectively from z. (b) The polarizer is along z, bisecting the tilt cone. (d) Model of director structures of six virgin domains in a chevron cell of $\overline{8}$S5.

In an applied electric field eight domains are now observed, the field supplying the torque necessary to allow simultaneous twist in both sides of the chevron interface [21]. Figure 7 shows the evolution of the twist domains as the applied field is increased. Figure 8 shows the final texture of the eight domains, as well as proposed models of the director field. Once the field is removed the cell returns to its virgin domain structure.

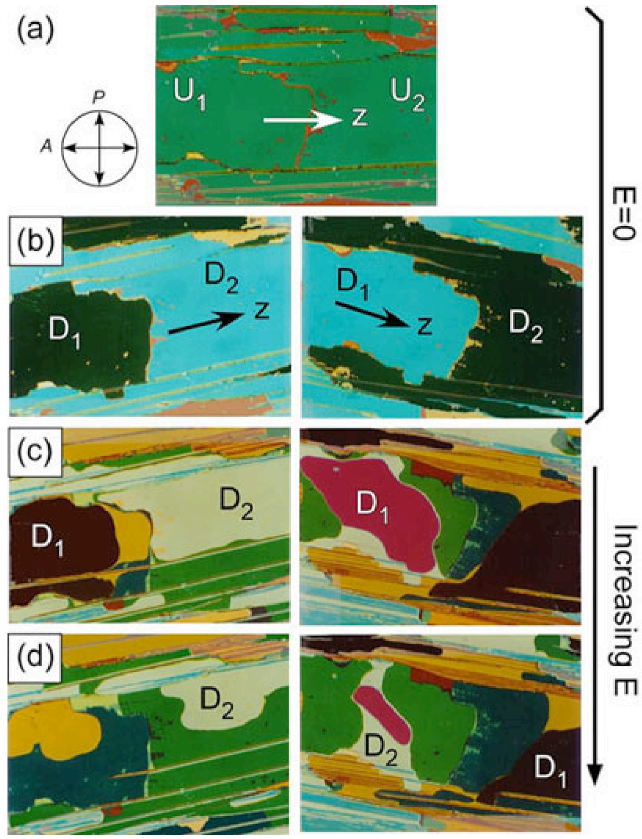

Figure 7. Optical micrographs of a 4 mm thick chevron cell of $\overline{8}$S5 in the SmC phase. (a) With no applied electric field, with z along the analyzer, the twisted (pink) and uniform (green) states appear different. (b) The cell can be rotated to extinguish either of the uniform states. (c) and (d): As the field is increased, twist domains grow into the previously uniform areas.

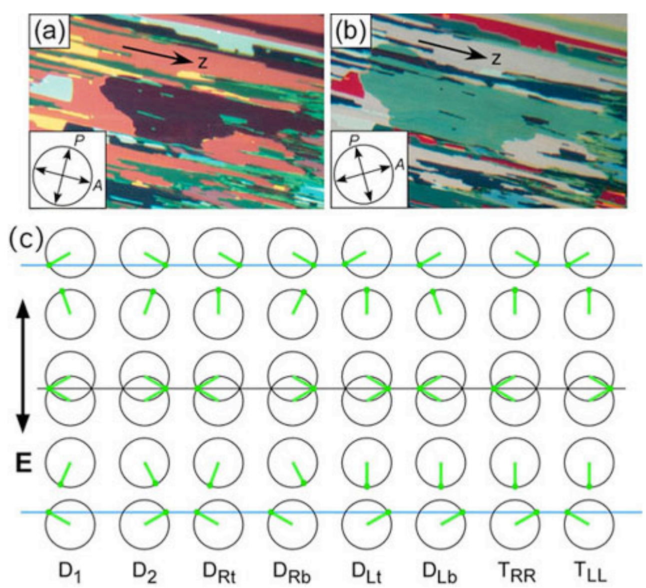

Figure 8. Domain behavior of $\overline{8}$S5 under the influence of an electric field. (a) and (b): DTLM images of a chevron cell with an applied electric field, showing eight distinct domains. (c) Model for the eight coexisting domains observed in a chevron cell of $\overline{8}$S5 in an applied electric field. The director structures labeled as

distorted, (e.g. D_1, D_{Lt}) are derived from the zero-field structures shown in Figure 6(d), with the bulk aligning with the electric field. The fully twisted domains, T_{RR} and T_{LL}, only appear when an electric field is applied.

When a large electric field (E ~ 15 V/μm) is applied, the chevroned smectic layers straighten out and a tilted bookshelf structure is obtained. In the absence of applied field, the cell then shows two uniform domains as with **W479** described above. A switching threshold of E ~ 6 V/μm is then required to grow twist domains, illustrated in Figure 9. In general, bookshelf $\overline{\mathbf{8}}$**S5** cells behave in a manner equivalent to **W479**, with the twist domains growing in from the boundaries between two uniform domains. In bookshelf cells of $\overline{\mathbf{8}}$**S5,** the exact twist domain boundary locations are determined by the sign of the applied field, meaning that for one sign of field the domain boundaries occur in different locations than for the opposite sign of field, but for a given sign of field the boundaries always reappear in the same place.

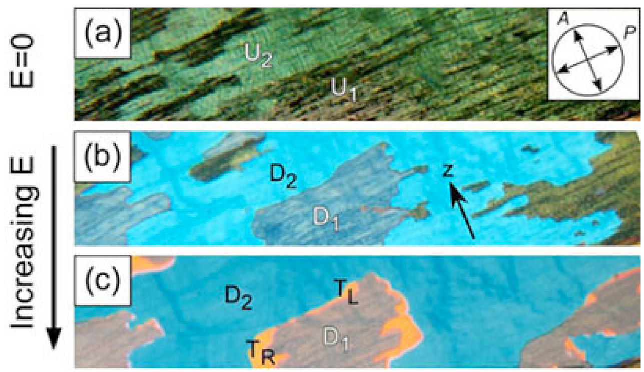

Figure 9. Evolution of uniform domains and nucleation of twist domains in a tilted bookshelf cell of $\overline{8}$S5 in an increasing applied electric field. (a) When no field is applied, two distinct uniform domains are visible, the extinguished (black) and bright (green) regions. (b) Above the Freedericksz transition the blue domains, where the surface orientations are the same, are almost uniform. (c) At higher E, twist domains (orange) appear. Here the surface orientations are different, giving both right- and left-handed director structures.

IV. Discussion

Dielectric switching of liquid crystal material with Δε > 0 in SmC cells with tilted layers may be described in simple terms as follows [22]: in an applied electric field, molecules reorient to the top or the bottom of the tilt cone, depending on the sign of the local layer tilt δ, in order to minimize the dielectric energy. When δ = 0, the top and bottom orientations are degenerate but when the layers are tilted, one orientation is preferred, with the other becoming metastable, as indicated in Figure 10. The difference in the energy of these states grows as the layer tilt increases. In chevron cells with the geometry sketched in Figure 8(c), the molecules above the chevron interface rotate up, while below it the molecules rotate down.

The free energy density of a smectic C liquid crystal with uniform layering may be expressed as

$$f = \frac{K}{2}(\nabla\phi)^2 - \frac{\Delta\epsilon}{8\pi}(\mathbf{n}\cdot\mathbf{E})^2 , \qquad (1)$$

where K is an elastic constant. When the cell boundaries are taken to be elastic (rather than infinitely strong) then the surface contribution to the free energy takes the form [18]

$$f_s \sim \gamma_1 \sin^2 \psi, \qquad (2)$$

where γ_1 represents the anchoring anisotropy and ψ is the elevation angle of the director above the plane of the glass. Here it is implicit that the surface energy is minimized when the director is oriented parallel to the glass.

In bookshelf cells of either **W479** or $\mathbf{\overline{8}S5}$ we observe that field-induced twist domains nucleate at the uniform domain boundaries. The π-walls that separate the uniform domains in the bulk are surface stabilized to form sharp line defects at (or very near) the surfaces, where the molecules experience strong planar anchoring (even after the application of E > 20 V/µm the surfaces do not change substantially from their planar orientation). In our model, line defects nucleate in the bulk and move laterally under the influence of the applied field, causing one surface stabilized orientation to effectively expand, growing past the orientation stabilized by the opposite surface. In this way, molecules on the top substrate oriented on the side of the tilt cone corresponding to D_1 appear to invade the D_2 domain, causing a twist in the bulk director field. The handedness of the twist is determined by the direction in which the defect moves. In chevron cells of $\mathbf{\overline{8}S5}$, the domain boundaries are able to move quite easily, the energy barrier for reorientation being much lower at the chevron interface than at the cell surfaces.

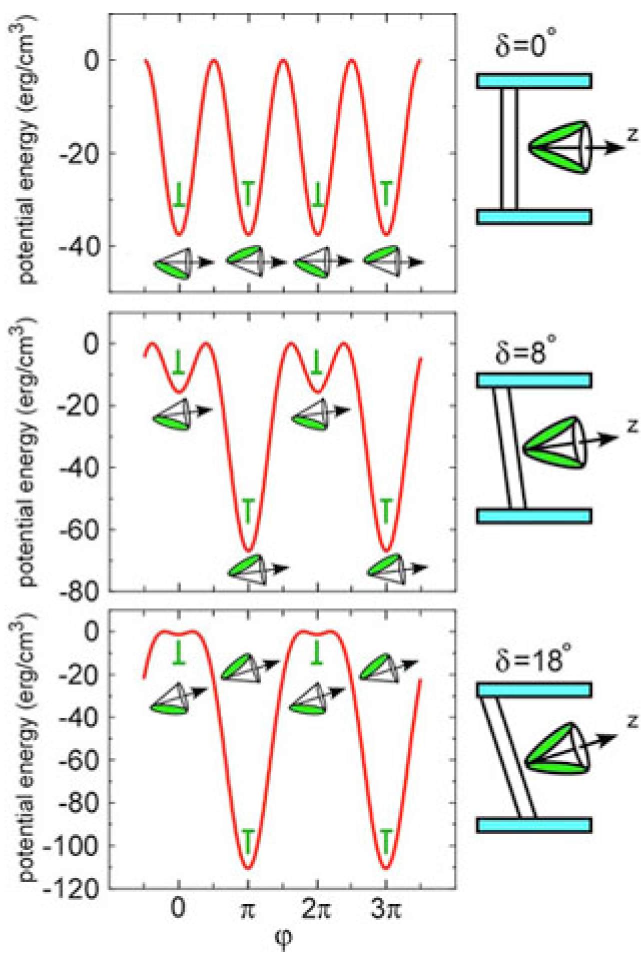

Figure 10. Dielectric potential energy density as a function of director azimuth in a bookshelf smectic C cell in an applied electric field, for different layer tilt: (a) δ = 0, (b) δ = 8°, and (c) δ = 18°. We assumed a tilt angle θ = 22°, dielectric anisotropy Δε = 0.8, and a cell thickness t = 4 µm. With a non-zero layer tilt, the director orientation at the top of the tilt cone (φ = π) is increasingly favored by the electric field.

In bookshelf cells with twisted director states, the director is oriented on opposite sides of the tilt cone at the two cell surfaces, with the director in the middle of the cell either at the top or the bottom of the tilt cone. The overall twist may be either left- or right-handed. When the smectic layers are arranged normal to the glass plates, twist around either the top or bottom of the tilt cone are equally likely, but when the layers are tilted the director field is elastically biased to follow the "short" path around the tilt cone, minimizing the change in φ and hence the distortion in **n**, as shown in Figure 11.

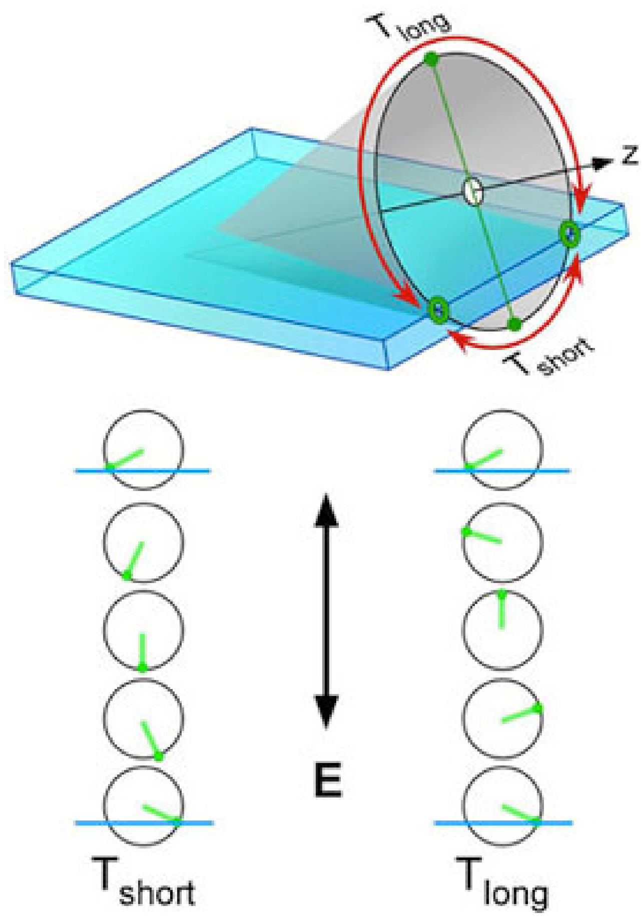

Figure 11. Schematic of the twisted director states T_{long} and T_{short}. While the T_{short} structure has less director distortion, and is therefore always energetically preferred in the absense of electric field, the T_{long} structure is favored in cells with tilted layers when a field is applied because, as shown in Figure 10, the bulk director can line up better with the field.

For example, in the geometry of Figure 2(a) this would correspond to a bulk director moving around the bottom of the tilt cone. When the layer tilt is large, this twist configuration, which we call type T_{short}, allows the director to be approximately planar throughout the cell. As we have seen before, however, in an applied electric field there is an electrostatic energy preference for the bulk director to be at the top of the tilt cone, as sketched in Figure 5(b)-(f) and in Figure 10. In this type of state, which we call T_{long}, the director precesses the "long" way around the tilt cone, at a greater cost in elastic energy, but nevertheless lowering the overall free energy because the bulk can align more closely with the applied field.

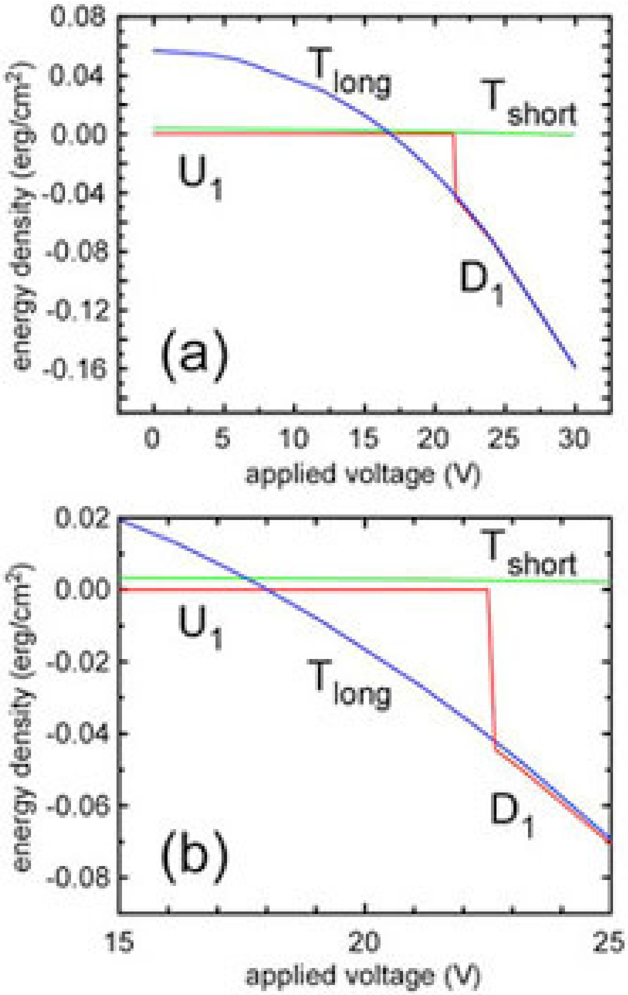

Figure 12. Free energy comparison of uniform and twisted states. The energy/area of an achiral smectic C cell was computed as a function of applied electric field assuming a smectic tilt angle $\theta = 22°$, layer tilt $\delta = 18°$, dielectric anisotropy $\Delta\varepsilon = 0.8$, Frank elastic constant $2x10^{-6}$ erg/cm, cell thickness t = 4 μm, and non-polar surface anchoring energy γ_l=0.8 erg/cm^2. (a) While the twisted T_{long} state has higher energy in weak fields (a), there is a small range of voltages over which it has lower energy than the deformed state D. (b) Above the Freedericksz transition in the uniform state (U →D), which occurs at about 21 V in this example, the D state has lower energy once again. The energy of the T_{short} state is comparatively unaffected by the field.

Applied electric fields typically induce uniform director states in nematics and ferroelectrics, the liquid crystal systems whose field response has been most widely studied. It is therefore rather curious that in non-chiral smectic C cells, non-uniform director states should be favored, at least in some range of applied field. In principle, the distorted uniform D state can change to a type T_{long} twisted state by switching the director orientation at (or at least near) one surface, as we have indicated above (c.f. Figure 5(d)-(f)). We performed 1-D molecular dynamics simulations in order to investigate under which circumstances this transition might be energetically favored, calculating the U_1, U_2 to D_1, D_2 Freedericksz-like transition (Figure 5(b) and (c)), and then compared the distorted state free energies to those of the twisted state. We assumed identical cell surfaces with a Rapini-Papoular surface energy favoring planar director orientation, and found the equilibrium director configurations using the numerical method described in Reference [18]. As expected, in weak applied fields the T_{long} state has higher energy than both the T_{short} and the U states. The simulations indicate, however, that there is a voltage region just below the Freedericksz transition in the uniform state (U $\rightarrow$D) in which the T_{long} twisted state has the lowest energy, the director in this state apparently being better aligned along the field, on the average, than in the uniform cell (see Figure 12). As the applied voltage is increased above the Freedericksz transition, the D state is found to have lower energy again. The difference in energy from the T_{long} state is small here, as expected, since in the high field limit the director field of these two states is identical in most of the cell, differing only over a small distance (of the order of the correlation length $\xi = (4\pi K/\Delta\varepsilon E^2)^{1/2}$) close to one cell surface. The T_{short} state, in contrast, has much higher energy at high fields. Since the observed domain growth and the model sketched in Figure 5 are clearly two dimensional and involve the motion of line defects, our simplified 1-D simulations should probably be regarded as a qualitative demonstration of principle, confirming that the twisted state can indeed have lower energy than the uniform state in some voltage regime.

V. Conclusion

We have studied the electric field response of two achiral SmC liquid crystals, in the bookshelf and chevron layer geometries. In both materials, the dielectric coupling to the applied electric field leads to the nucleation and growth of twisted director states in cells that initially exhibit only uniform domains. This non-intuitive effect is the opposite of the typical behavior of chiral cells, where an applied electric field always favors a uniform director orientation, suppressing twist. In $\mathbf{\overline{8}S5}$, in either chevron or field-induced bookshelf cells, the induced twist domains are not stable in the absence of field. In **W479,** on the other hand, once the tilted bookshelf smectic layers have been forced upright by the field, the induced twist domains remain even after the applied field is removed.

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VI. References

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[6] Below the SmC phase, **W479** has an anticlinic phase (SmC_A), which has a birefringence that is markedly smaller ($\Delta n = 0.047$) than in the SmC ($\Delta n = 0.23$) and that increases when a field is applied, as expected for an anticlinic phase. This molecular structure has been confirmed independently by observation of in-plane polarization parallel to the tilt plane of even layer

number freely suspended films [see D.R. Link, J.E. Maclennan, and N.A. Clark, *Phys. Rev. Lett.*, **77**, 2237, (1996)].

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[10] The π-walls discussed here are not actually π-walls in the most correct sense of the term, because in a tilted layer geometry there is not a 180° difference in the director orientation on the tilt cone. With tilted layers the opposing domain's director orientation is less than π.

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