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Electrostatics and the electro-optic behaviour of chiral smectics C: 'block' polarization screening of applied voltage and 'V-shaped' switching

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We present a new model for the physics of thresholdless switching in chiral smectics. In the limit of high polarization, the electro-optics of chiral smectic C liquid crystals are dominated by two distinct electrostatic effects. Complete (surface to surface) 'stiffening' of the polarization field by its charge self-interaction causes the polarization to orient as a uniform block. Complete screening of applied electric field by polarization charge leads to voltage-induced orientation where the electric field in the liquid crystal is exactly zero, These effects, both operative during the 'V-shaped' portion of thresholdless transmission vs. voltage curves, combine to produce 'V-shaped' switching.

1. Introduction

An important theme of current research on chiral smectic liquid crystals (LCs) is understanding the origin of the high contrast 'V-shaped' or 'thresholdless' analogue electro-optic effect found in some homologues of the antiferroelectric LC MHPOBC [1], and more recently in other high polarization chiral smectics $\lceil 2 \rceil$. 'V-shaped' switching cells are planar-aligned and surfacestabilized (helix free), as sketched in figure 1 (a), exhibiting electric field-induced continuous reorientation of a spatially uniform optic axis, which, with light incident perpendicular to the plates, appears to be along the layer normal z for applied voltage V = 0. The spatial uniformity of the optic axis is evidenced by the excellent extinction achievable with these cells between crossed polarizer and analyser with polarizer along z, the transmission exhibiting the characteristic field-induced 'V-shape' shown in figure 1(b).

Results, first presented at the 17th International Liquid Crystal Conference in 1998 [3] and based on the prediction of thresholdless electro-optic behaviour [4], have demonstrated that this effect is a feature of the ferroelectric smectic C* (SmC*) phase [5], and that the rotation of the optic axis is, in fact, due to the field-induced azimuthal molecular reorientation on the smectic C* tilt cone. The SmC* spatial optic axis

uniformity is electrostatic in origin, a result of the polarization charge self-interaction, which becomes important with materials having sufficiently large spontaneous polarization, **P**. This means that thresholdless behaviour is due to a field-excited SmC* Goldstone mode and not the 'Langevin mode' of a random smectic phase [1]. This collective picture has since been independently confirmed [6].

In this note we discuss the electrostatics of planaraligned SmC* cells, focusing on polarization charge self-interaction and on a second electrostatic effect, polarization charge screening of the applied electric field. We illustrate their respective roles in determining electrooptic behaviour (V-shaped vs. bistable) of ion-free cells and demonstrate the key role of insulating alignment layers in the absence of ions. We find that with high polarization materials, electrostatics dominate the electrooptic behaviour in a way completely new for liquid crystals, and, for that matter, ferroelectric materials. The high P regime proves to be quite elegant: electrostatic energies are large, surface interactions can be ignored, and the polarization responds only to electric field, its equilibrium state being that for which the liquid crystal has zero field! As a result, insulating layers on the cell electrodes play a crucial role in the electro-optics, which are described by a single simple equation involving only a single LC parameter (P), a surprising result given the complexity of the problem and the large number

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Figure 1. (a) Bookshelf cell geometry of a tilted chiral smectic, where the liquid crystal is a dielectric in a transparent ITO-glass capacitor with the smectic layers more or less normal to the plates. Shown are the director **n**, the tilt cone angle θ relative to the layer normal **z**, the polarization **P**, the **c** director projection of **n** onto the layer plane, the azimuthal orientation ϕ of **c**-**P**, and the layer direction **y**. 'V-shaped' switching is observed with normally incident light between crossed polarizer/analyser. (b) 'V-shaped' electro-optic response of the Tokyo mixture T3 (pink dots); $T(V)/T_{SAT}$ calculated from equation (3) (black curve). Voltage across LC layer (blue curve) is zero for $0 < \phi < \pi$ (yellow region).

of elastic and surface interaction parameters typically required in the modelling of SSFLC cells. The model accounts for key aspects of the phenomenology of 'V-shaped' switching, and illustrates electrostatic effects which are *unavoidable* at high **P**.

2. Polarization space charge effects

We make our points with the aid of figure 2, which shows schematically a series of different planar-aligned SmC* cell geometries. The SmC* is viewed with the layers parallel to the plane of the page, i.e. layer normal z towards the reader. The SmC* is characterized by the distribution of $\phi(x)$, the azimuthal orientation about z of **P** (black arrows) on the tilt cone (circles), and by the



Figure 2. Sketches of the polarization-director (P-black arrow, c-green tee) structure obtained in SSFLC cells under various conditions of polarization magnitude, applied field, and surface interaction, using the following symbols: white +,- are free charges; black and red +,- are polarization charges; blue arrows are electric field. (a, b) Up and down SSFLC states, stabilized by the surface energies sketched in (a). (c) Splayed polarization state for low \mathbf{P} $(\xi_{\mathbf{P}} > d)$. (d) Splayed polarization state for high **P** $(\xi_{\mathbf{P}} < d)$. (e-i) Uniform polarization states where the polarization charge self-interaction forces P to reorient as a 'block'. (h, i) Cells with insulating surface layers. 'V-shaped' switching is illustrated in (i): with large polarization ϕ is determined electrostatically by the condition that $\mathbf{E} = 0$ in the LC. The cell voltage is that required to produce a surface charge $\mathbf{P}\cos\phi$ on the insulating layers. See text for a detailed discussion.

nematic-like director (projected onto the xy plane as green T's) locally in the plane normal to **P** and tilted from **z** through the cone angle. The cells sketched are composed of SmC* (blue) bounded by conducting electrodes (grey), which in (h) and (i) are coated with an electrically insulating layer (pink) of thickness t. Several

of the cells [(a)-(f), and (h)] are electrically shorted, as indicated by the vertical black 'wire' connecting the upper and lower electrodes.

Figures 2(a) to 2(c) show respectively the UP and DOWN states identified in the initial SSFLC devices [7], and the polarization SPLAYED state found shortly thereafter [8]. The basic physics of these classic SSFLC states can be understood in low polarization materials in terms of the combined action of bulk nematic director deformation energy, which is lowest when P(x) is uniform, and of an effective surface energy, which is lowest when **P** at the surface is directed out of the cell and P(x)is consequently splayed. Typically the surface interaction can be modelled with polar and non-polar surface energy terms, $U(\phi) = U_{p}(\mathbf{x} \ \mathbf{s}) \cos(\phi_{s}) - U_{np} \cos^{2}(\phi_{s})$, where **s** is a surface normal directed into the LC and ϕ_s is the orientation at the surface [9]. The polar (U_p) and nonpolar (U_{np}) interaction coefficients are such that $U(\phi)$, sketched in figure 2(a), is double-welled, with minima when **n** is parallel to the surface (due to the nematic-like surface interaction), that have differing energy, depending on the orientation of P relative to s (due to the polar nature of the surface). For the purposes of this discussion we can assume that the surface preferred orientation has **P** directed *out* of the LC, as indicated in figures 2(c) and 2(d). Assuming isotropic elasticity $U_{elas} = 1/2K(\phi_{xx})^2$, the SPLAYED state is characterized by a linear variation of $\phi(x)$ across the cell. It is the stable ground state generally when the surface interaction is strong enough to overcome the bulk elasticity, i.e. when U_p , $U_{np} > K/d$ [9].

In the physics of the SSFLC states of low **P** materials, electrostatics plays a role only when an external electric field is applied to produce ferroelectric/dipolar $\mathbf{P} \times \mathbf{E}$ torque, affecting discrete transitions among the states or producing continuous deformation within them. These effects can be understood on the basis of solutions of field equations describing the balance of elastic, surface and $\mathbf{P} \times \mathbf{E}$ torque [9]. However, as the polarization is increased, this situation changes, such that, in the limit of large **P**, only the $\mathbf{P} \times \mathbf{E}$ torque is relevant, and it behaves in a rather non-intuitive way.

The **P** UP and DOWN uniform states of figures 2(*a*) and 2(*b*) have zero electrostatic energy. With no insulating layers at the cell surfaces, free charge (white) can flow to cancel exactly the surface polarization charge (black), resulting in electric field **E** = 0 everywhere. This is not the case for the SPLAYED state of figure 2(*c*) in which the linear variation of $\phi(x)$ produces negative volume polarization space charge $\rho_{\rm P} = -\nabla$ **P**, separated from surface polarization charge, resulting in an internal electric field directed toward the cell centre (blue), and associated electrostatic energy $U_{\rm elec}$. Since $U_{\rm elec} \sim {\bf P}^2$, as **P** is increased there will be an increasing tendency for $\phi(x)$ to adjust somehow to reduce U_{elec} [10, 11, 4, 12]. Qualitatively, U_{elec} is reduced if the positive and negative black charges can be brought closer together. If the surface orientations are kept fixed then the system response to reduce U_{elec} is shown in figure 2(d): $\rho_{\rm P}$ (and therefore ∇ **P**), tends to be expelled from the bulk LC to the surfaces, rendering $\phi(x)$ uniform away from the surfaces and moving the negative $\rho_{\rm P}$ nearer to the surfaces. Minimization of the sum of U_{elec} and the orientational elastic energy with pinned surfaces generates near each surface a splayed **P**-twisted **n** surface domain, a 'polarization-stabilized kink' (PSK) [4] of characteristic thickness $\xi_{\rm P} = (\varepsilon_{\rm LC} K/P^2)^{1/2}$, where $\varepsilon_{\rm LC}$ is the bare LC dielectric constant (excluding effects of P). Polarization charge effects thus become important when $\xi_{\rm P} < d$, in which limit the resulting 'electrostatic stiffening' of the polarization field produces nearly uniform molecular orientation away from the surfaces and thus good optical extinction (high contrast) between crossed polarizers. For normally incident light, this uniform orientation has its apparent optic axis along z for applied voltage V = 0 and responds to applied voltage by reorienting in a continuous fashion, as indicated in Figure 2(j). Calculations of transmission vs. voltage, T(V), using the Berreman 4×4 method show the characteristic high contrast 'V-shaped' response, the PSK model providing an excellent description of the T(V) data [5].

But what are the implications of this PSK picture for surface energetics? In the structure of figure 2(d) the separation of positive and negative polarization charge near the surface is held entirely by $U(\phi)$, which must apply a surface torque $\tau_{\rm PSK} \sim K/\xi_{\rm P} = (K{\bf P}^2/\varepsilon_{\rm LC})^{1/2}$ to maintain the polarization-stabilized kinks. For a typical high-polarization FLC ($\mathbf{P} \sim 100 \text{ nC cm}^{-2} = 1 \text{ mC m}^{-2}$), we have, with $K \sim 5 \times 10^{-12} \text{ J m}^{-1}$, and $\varepsilon_{\text{LC}} \sim 10\varepsilon_{\text{o}}$, $\xi_{\rm P} \sim 20 \,\rm nm \ and \ \tau_{\rm PSK} \sim 2 \times 10^{-8} \,\rm J \ m^{-2} = 2 \ erg \ cm^{-2}$. This value of **P** is that of the Tokyo T3 'V-shaped' switching mixture [1], but is considerably smaller than that of W415 [2]. This estimated τ_{PSK} exceeds by an order of magnitude the typical surface torques, $\tau_s \sim U_p$, U_{np} available from the polar [13] and non-polar [14] surface interactions, U_p , $U_{np} \sim 0.1 \ erg \ cm^{-2}$, respectively. Thus, for $P \gtrsim 20 \text{ nC cm}^{-2}$ the surfaces become too weak to hold the polar surface orientation. Thus we appear to have in T(V) strong experimental support for the PSK model, which turns out to be physically impossible!

3. Polarization surface charge effects

This conundrum can be resolved by a new picture which releases the polarization from the surface, and at the same time introduces insulating layers which serve to separate polarization charge from free charge, a key, but heretofore unappreciated, role of the surface domains in the PSK model. Thus, in high P materials such as T3, P(x) becomes spatially uniform throughout the liquid crystal, as figure 2(e) shows, the bulk nematic elasticity completely dropping out of the problem. This behaviour has been probed for the mixture T3 by comparing the surface and bulk electro-optic response measurements obtained using polarized total internal reflection and transmission, respectively. The surface and bulk optic axis orientation are found to be identical during 'V-shaped' switching, confirming the complete uniformity of the **P-n** couple in the cell [2, 15]. Although electrostatics overwhelmingly dominate the internal structure of P(x)in this cell, they have no effect on the average orientation of **P** when the cell is shorted (V = 0) with no insulating layers. As figure 2(f) indicates, an overall rotation of **P** (produced say by a strong magnetic field coupling to **n**) is accompanied by the appearance of polarization charge (black) on the surfaces; but this is exactly cancelled by the flow of free charge (white). The overall orientation of **P** is thus still controlled by the surface interactions. ϕ at the upper and lower surfaces is now identical (with the lower surface flipped by π relative to the upper) so that the net surface energy is the sum $U_{tot}(\phi) =$ $U_{top}(\phi) + U_{bottom}(\phi) = U(\phi) + U(\phi - \pi)$, shown as the blue curve in figure 2(a). Thus the V = 0 high-P cell with no insulating layers at the surfaces will be bistable.

Note that the result is quite different if we start from the condition of figure 2(e), but open circuit the cell before rotating **P**. In this case (figure 2(g)) the surface polarization charge generates a field in the LC which opposes the agent turning **P**, harmonically restoring the orientation to $\phi = \pi/2$. For high **P** materials this field is quite large: for **P** ~ 100 nC cm⁻², $\varepsilon_{\rm LC} \sim 10\varepsilon_{\rm o}$, and $d = 1 \,\mu\text{m}$, an initially V = 0 open circuited cell with $\phi = \pi/2$ develops a voltage $V = -\mathbf{P}d/\varepsilon_{\rm LC} \sim -20$ V when **P** is reoriented to $\phi = 0$.

Next we consider the case central to the discussion: the cell with insulating layers coating the surfaces, (figures 2(h) and 2(i)). As in figures 2(e) and 2(f), polarization charge stabilization makes P internally rigid. However, with the cell shorted, unlike in figure 2(f), rotation of **P** from $\phi = \pi/2$ generates non-zero electric fields, **E**_{IL} and \mathbf{E}_{LC} , in the insulating layer and LC, respectively. This is because the insulator forces a separation of the polarization surface charge $\sigma_{\rm P}$ and the free charge $\sigma_{\rm F}$ so that the two cannot completely cancel. Additionally, with the cell shorted, the condition $V = \int \mathbf{E} \, d\mathbf{x} = 0$ necessitates a field in the LC: $\mathbf{E}_{LC} = -2\mathbf{E}_{IL}(t/d)$. On the upper insulating layer we find $\sigma_{\rm P} = {\bf P} \cos \phi$ and $\sigma_{\rm F} = -(1 - 2t/d)\sigma_{\rm P}$, with sign reversal on the lower insulating layer. For the typical condition that the insulating layers are thin relative to the overall cell thickness (t < d), we have $\sigma_{\rm P} \approx \sigma_{\rm F}$, $\mathbf{E}_{\rm IL} \approx \sigma_{\rm P} / \varepsilon_{\rm IL} = \mathbf{P} \cos \phi / \varepsilon_{\rm IL}$ and the electrostatic energy per unit cell area is dominated by the charge separation across the insulating layer:

$$U_{\text{elec}}(\phi) = \sigma_{\text{F}} V = (1/A) \int_{\text{IL}} \mathbf{E} \quad \mathbf{D} \, \mathrm{d}v = (2t/\varepsilon_{\text{IL}}) (\mathbf{P} \cos \phi)^2.$$
(1)

 $U_{\text{elec}}(\phi)$ is a minimum at $\phi = \pi/2$ [$U_{\text{elec}}(\pi/2) = 0$] and is a maximum at $\phi = 0$, π [$U_{\text{elec}}(0, \pi) = 2t\mathbf{P}^2/\varepsilon_{\text{IL}}$]. With $\mathbf{P} = 100 \text{ nC cm}^{-2}$ and $\varepsilon_{\text{IL}} \sim 3\varepsilon_0$ [16] we find $U_{\text{elec}}(0, \pi) \approx$ [6.6 × 10⁻⁵ t(nm)] J m⁻². Even for thin insulating layers ($t \ge 10 \text{ nm}$) $U_{\text{elec}}(0, \pi) > 7 \times 10^{-4} \text{ J m}^{-2}$, comparable to the largest known U_p or U_{np} , indicating that the electrostatic energy 'harmonically' constraining \mathbf{P} to $\phi = \pi/2$ will typically be dominant. Under this circumstance, the surface interactions drop out of the problem and \mathbf{P} , rendered into a spatially uniform block by the electrostatic polarization charge interaction, is itself controlled completely by electrostatics.

For $V \neq 0$, indicated in figure 2(*i*), this electrostatic limit is particularly interesting and elegant. If the surface interactions are ignored then **P** is responsive *only* to whatever electric field appears in the LC, and the equilibrium orientation of **P** is necessarily that for which $\mathbf{E}_{\rm LC} = 0$. Thus, ϕ is determined by the condition that **P** reorients to screen the field completely to zero in the LC. The free charge and polarization charge on opposite surfaces of the insulating layer must then be equal in magnitude, and the applied voltage is dropped entirely across the insulating layers. The results of this condition are very simple and universal relationships giving orientation in terms of applied voltage, i.e. governing 'V-shaped' switching in high-P SmC* materials:

$$\varepsilon_{\rm IL} V = 2t D_{\rm IL} = 2t \mathbf{P} \cos \phi \tag{2}$$

for $-2t\mathbf{P}/\varepsilon_{IL} < V < 2t\mathbf{P}/\varepsilon_{IL}$, or $V > 2t\mathbf{P}/\varepsilon_{IL}$: $\phi(V) = 0$, $V_{LC} = V - 2t\mathbf{P}/\varepsilon_{IL}$ (3 a)

$$-2t\mathbf{P}/\varepsilon_{\mathrm{IL}} < V < 2t\mathbf{P}/\varepsilon_{\mathrm{IL}}: \ \phi(V) = \cos^{-1}(\varepsilon_{\mathrm{IL}}V/2t\mathbf{P}), \ V_{\mathrm{LC}} = 0$$
(3 b)

$$V < -2t\mathbf{P}/\varepsilon_{\mathrm{IL}}: \qquad \phi(V) = \pi, \qquad V_{\mathrm{LC}} = V + 2t\mathbf{P}/\varepsilon_{\mathrm{IL}}$$
(3 c)

where $V_{\rm LC}$ is the voltage across the LC layer. **P** can screen the field in the LC only as long as $|V| < V_{\rm SAT} \equiv$ $|2t\mathbf{P}/\varepsilon_{\rm IL}|$. For larger voltage magnitudes the orientation is saturated at $\phi = 0$ or π , depending on the sign of V. For $|V| < V_{\rm SAT}$ the electrostatic energy binding ϕ to the value in equation (2) is comparable to that of the V = 0case of equation (1). The optical transmission through crossed polarizers, T(V), of an SSFLC cell governed by equation (3) is readily calculated, with the result shown in figure 1(*b*), expressed as the contrast $C = T(V)/T_{\text{SAT}}$, where T_{SAT} is the high voltage saturation T(V). We use typical SmC* parameters (tilt angle $\theta = 25^{\circ}$, birefringence $\Delta n = 0.15$), with the parameter $\alpha = 2t\mathbf{P}/\varepsilon_{\text{IL}}$ adjusted to fit the data. The result, with $2t\mathbf{P}/\varepsilon_{\text{IL}} = 4.2$ V, is an essentially perfect match for the data. Taking $\varepsilon_{\text{IL}} = 3\varepsilon_0$ gives t = 63 nm, comparable to the alignment layer thickness.

Equation (3) makes several specific and rather striking predictions: (i) the width V_{SAT} of the 'V-shaped' switching curve is linear in the alignment layer thickness (such behaviour is observed qualitatively [16] and recently quantitatively [17] in systematic studies of the effects of alignment layer thickness); (ii) the width of the 'V-shaped' switching curve is independent of the LC thickness; (iii) the width of the 'V-shaped' switching curve is proportional to **P**. Predictions (ii) and (iii) are counter to the conventional intuition that switching should occur at lower voltage in higher **P** materials and thinner cells. They are directly a result of the polarization screening and a sure sign that this effect is operative.

4. Concluding remarks

This electrostatic behaviour also has implications for the effects of ionic charges in 'V-shaped' switching cells (figure 2(*i*)). As equations (3) and figure 1(*b*) show, for applied voltage in the range $|V| < |2t\mathbf{P}/\varepsilon_{IL}|$, the voltage on the liquid crystal layer $V_{LC} = 0$. Thus, if V is maintained in the range $|V| < |2t\mathbf{P}/\varepsilon_{IL}|$ the ions in the LC layer become irrelevant and only ions in the insulating layers affect electro-optic behaviour. Thus, charge separation in the insulating layer produces hysteresis and it is the mobility and concentration of ions in the insulating layer that governs hysteretic behaviour and dynamics. A V = 0 (shorted) cell containing ions will eventually relax to the SSFLC state, uniform or splayed, of lowest energy. If it is the splayed state, a gradual loss of extinction between crossed polarizers is observed [2].

These considerations should also apply to chevron layer SSFLC cells, in which the chevron interface acts as an additional interface constraining the LC director orientation [18]. Typically there are two states of opposite P_x stabilized at the chevron interface, yielding bistable SSFLC cells even with fixed surface orientations. The orientational pinning at the chevron interface tends to be quite weak, however, the chevron interface typically switching at lower voltages than the surfaces. Thus, polarization self-interaction stiffening is sufficient to eliminate the bistable states at the chevron interface in favour of a state of uniform P_x , even in rather low polarization materials [12]. For high **P**, chevron cells will exhibit 'block **P**' switching, which is slightly more complicated than that in the bookshelf cells. With V = 0, **P** will be parallel to the cell plates, as in the bookshelf case, but in opposite directions above and below the chevron interface [19, 12], and will rotate in opposite directions with applied field.

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