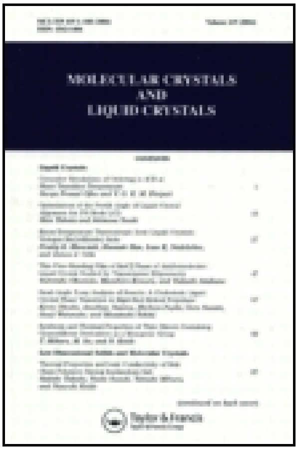

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Order Parameter Theory for Switching in Antiferroelectric Liquid Crystals

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Order Parameter Theory for Switching in Antiferroelectric Liquid Crystals

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In order to model the field induced switching of AFLC materials we have employed a bilayer order parameter theory which includes changes in the distribution of molecules around the smectic cone. The free energy is a function of the azimuthal director angle and the molecular order about this director in the odd and even layers. By minimizing the free energy the effective optic axis may be calculated as a function of applied voltage. This theory is able to produce not only hysteretic tristate switching typical of AFLC materials but also the pre-transitional electroclinic-like behaviour at low field strengths and the frequency dependence of the double hysteresis loop.

Keywords: antiferroelectric; switching; order parameter theory

INTRODUCTION

Interest in AFLCs has grown due to their characteristic tri-state switching which may be useful in display devices[1,2]. The interaction between the spontaneous polarization and an applied electric field switches the material to one of the two possible FLC states depending on the direction of the field. One of the main factors limiting the exploitation of AFLCs is the pre-transitional regime observed at low fields which affects the achievable contrast and hence prevents the reproduction of a large number of grey levels. The nature of this regime is still not understood fully, although there is strong evidence that it is not a pure soft mode electroclinic behaviour[3].

In this paper we extend a previous order parameter theory[4], which was used to explain the behaviour of this pre-transitional regime, to allow switching from the AFLC state to the FLC state. Instead of the usual description of smectic materials, solely in terms of the mean molecular direction within a layer, this model includes information about the *variation* of the distribution of molecules from this mean (see Fig. 1). This variation, or spread, of the distribution, is a measure of the order within the layer. The free energy is written in terms of the director angles, ϕ_o and ϕ_e, and

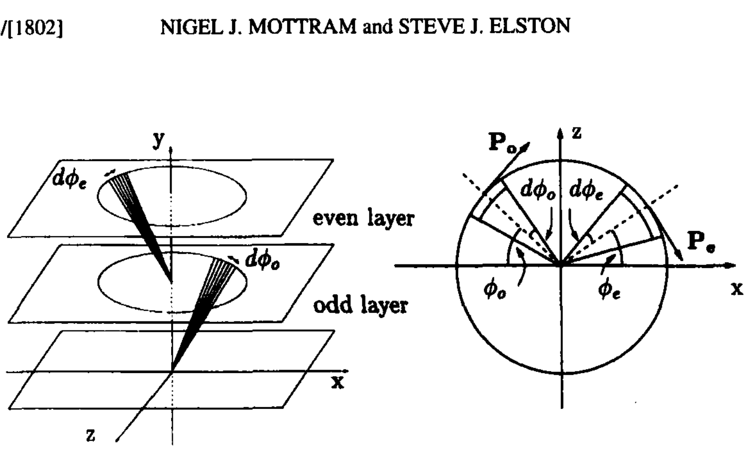

Figure 1: Molecular distributions in the odd and even layers described by the director angles ϕ_o and ϕ_e and the spreads $d\phi_o$ and $d\phi_e$ around the smectic cone.

the spreads, $d\phi_o$ and $d\phi_e$, in the odd and even layers of the AFLC material. The energy is assumed to consist of contributions from the thermodynamic potential in each layer, the interaction between molecular dipoles *within* each layer and *between* each layer, and the interaction between the dipoles and an electric field applied in the z direction. The dynamic governing equations are solved numerically, for an AC applied electric field, to obtain the characteristic double hysteresis loop of AFLC switching. We then investigate the form of the hysteresis loop as the applied field frequency varies.

THEORY

In a previous theory by the present authors[4] it was assumed that the director within each layer was fixed so that the system remained in the AFLC state (equivalent to $\phi_o = 0 = \phi_e$ in Fig. 1) but the distribution of molecules about the director was allowed to change as an electric field was applied. It was thus possible to model the pre-transitional regime of the AFLC hysteresis loop. We now extend this model, allowing the director to rotate in order to switch between the AFLC and FLC states. In this way it is hoped that the present model will exhibit switching *and* the pre-transitional effect. A similar argument to the previous model is used in order to determine the free energy of the system as a function of the extent of the molecular spread *and* the director angle, in both the odd and even layers. The free energy is

$$
\begin{aligned}
F \;=\; & A\left((d\phi_o - d\phi)^2 + (d\phi_e - d\phi)^2\right) \\
& - B_1\left(\cos(\phi_o) + \cos(\phi_e)\right) - B_2\left(\cos^2(\phi_o) + \cos^2(\phi_e)\right) \\
& + \frac{P^2}{2\chi}\left(\left(\frac{\sin(d\phi_o)}{d\phi_o}\right)^2 + \left(\frac{\sin(d\phi_e)}{d\phi_e}\right)^2\right) \\
& - \frac{P^2}{\epsilon\epsilon_0}\left(\frac{\sin(d\phi_o)\sin(d\phi_e)}{d\phi_o d\phi_e}\cos(\phi_o + \phi_e)\right) \\
& + \frac{EP}{2}\left(\frac{\sin(d\phi_o)}{d\phi_o}\cos(\phi_o) - \frac{\sin(d\phi_e)}{d\phi_e}\cos(\phi_e)\right) \qquad (1)
\end{aligned}
$$

where A, B_1 and B_2 are positive constants. It is worth noting that when the directors are fixed in the AFLC state ($\phi_o = 0 = \phi_e$) eq. (1) reduces to the free energy of the previous order parameter theory discussed above[4] and when the spread is fixed at the equilibrium value ($d\phi_o = d\phi = d\phi_e$) eq. (1) reduces to the free energy used by Nakagawa[5] (and later by the present authors[6]).

The first term in eq. (1) is the thermodynamic potential which models the excluded volume effect in each layer and exhibits a minimum at the equilibrium values, $d\phi_o = d\phi = d\phi_e$. The simplest form of such a potential is quadratic and is essentially the first term in the Taylor expansion of the actual thermodynamic potential function. Consequently it is only an approximation. By estimating the amount of spread in a nematic liquid crystal close to the smectic phase transition we set $d\phi = 0.4$ rad.

The second term of eq. (1) is a polar energy term which has a minimum when $\phi_o = 0,\ 2\pi\ldots$ and $\phi_e = 0,\ 2\pi\ldots$. The third term is a non-polar energy term which has a minimum when $\phi_o = 0,\ \pi,\ 2\pi\ldots$ and $\phi_e = 0,\ \pi,\ 2\pi\ldots$. These two terms are included in order to prescribe a preferred orientation for the mean polarization vectors. The second term can be thought of as the effect of a surface which is tending to orient the polarization vectors in the odd and even layers of the bulk material in fixed directions whilst the third term simply breaks the degeneracy of the model and prescribes the xy-plane as the plane within which the directors in the odd and even layers would prefer to lie.

In order to calculate the last three terms of eq. (1) we must consider the net polarization in each layer. Each molecule lying on the smectic cone contains a dipole lying within the xz-plane and perpendicular to the long axis of the molecule which causes a spontaneous polarization (see Fig. 1). We assume that the local molecular polarization is P and the polarization vector for each molecule is $\mathbf{P}_m = P(\sin\Phi,\ ,0,\ \cos\Phi)$ where Φ is the angle

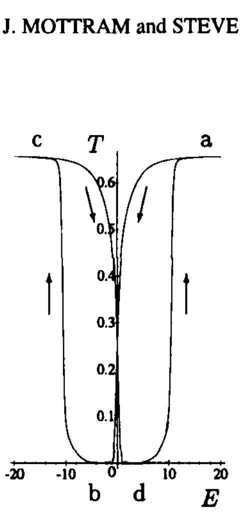

Figure 2: Transmission T versus applied electric field strength E for the parameters $f = 0.1$ and $E_{max} = 20$. The arrows show the path the system takes around the hysteresis loop.

between the dipole and the z-axis. Integration of $\mathbf{P}_m$ over all molecules within the spread gives the net polarization in the odd and even layers as

$$P_o = P\frac{\sin(d\phi_o)}{d\phi_o}[\sin(\phi_o),\ 0,\ \cos(\phi_o)] \tag{2}$$

$$P_e = P\frac{\sin(d\phi_e)}{d\phi_e}[\sin(\phi_e),\ 0,\ -\cos(\phi_e)] \tag{3}$$

From Goodby et al[7] the self-energy (density) of a polarization $\mathbf{P}_1$ within a material of susceptibility χ is given as $\mathbf{P}_1{}^2/(2\chi)$. Thus the *intra*layer energy density of the odd and even layers is the fourth term of eq. (1). Since minimum self interaction energy occurs when the polarization is zero the molecular distribution tends to spread around the cone. Consequently the intralayer energy is a maximum when $d\phi_o = d\phi_e = 0$ and a minimum when $d\phi_o = d\phi_e = \pi,\ 2\pi,\ 3\pi \ldots$.

The interaction energy (density) of two polarizations $\mathbf{P}_1$ and $\mathbf{P}_2$ in a material of dielectric permittivity ϵ is $-\mathbf{P}_1.\mathbf{P}_2/(\epsilon\epsilon_0)$. Thus the *inter*layer interaction energy density is the fifth term of eq. (1). This energy is minimum when $\phi_o = 0 = \phi_e$ and $d\phi_o = 0 = d\phi_e$ so that the molecules in the odd layer lie on the opposite side of the smectic cone to those molecules in the even layer with no spread and hence the polarizations are antiparallel.

The electric field term is due to the interaction between the polarization in each layer and the applied electric field $\mathbf{E} = [0,\ 0,\ E]$. The energy (density) between a polarization $\mathbf{P}_1$ and field $\mathbf{E}$ is $\mathbf{E}.\mathbf{P}_1/2$ which leads to

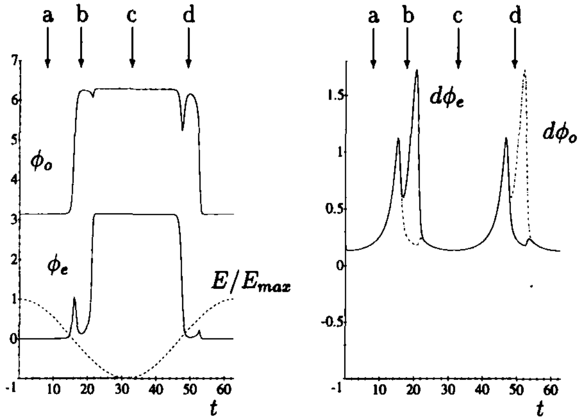

Figure 3: The director angles ϕ_o and ϕ_e and the spreads $d\phi_o$ and $d\phi_e$ as time varies. The dashed line shows the relative field strength E/E_{max} and in the second plot $d\phi_o$ is shown as a dashed-dotted line whilst $d\phi_e$ is shown as a solid line.

the last term of eq. (1). The electric field energy is therefore minimum when both polarizations align with the field, i.e. $\phi_o = \pi$, $\phi_e = 0$ when $E > 0$ and $\phi_o = 0$, $\phi_e = \pi$ when $E < 0$.

For a fixed field strength the variables ϕ_o, ϕ_e, $d\phi_o$ and $d\phi_e$ can then be calculated by minimizing the free energy (eq. 1). The dynamic equations are found by assuming that the rate of change of any of the variables is proportional to the gradient of the free energy at that point,

$$\eta_1 \frac{\mathrm{d}(\phi_o)}{\mathrm{d}t} = -\frac{\mathrm{d}F}{\mathrm{d}(\phi_o)}, \qquad \eta_1 \frac{\mathrm{d}(\phi_e)}{\mathrm{d}t} = -\frac{\mathrm{d}F}{\mathrm{d}(\phi_e)} \tag{4}$$

$$\eta_2 \frac{\mathrm{d}(d\phi_o)}{\mathrm{d}t} = -\frac{\mathrm{d}F}{\mathrm{d}(d\phi_o)}, \qquad \eta_2 \frac{\mathrm{d}(d\phi_e)}{\mathrm{d}t} = -\frac{\mathrm{d}F}{\mathrm{d}(d\phi_e)} \tag{5}$$

where η_1 is the viscosity of the rotation of the distribution around the smectic cone and η_2 is the viscosity of the spreading motion of the distribution. After solving eqs. (4) and (5) we can calculate the effective optic axis and estimate the transmission through a cell placed between crossed polarizers. The optic axis for a single molecule is taken as the projection of that molecule on the cell plane (the xy-plane). The macroscopic optic axis, ψ, is then the average molecular optic axis fo all molecules in the distributions described by ϕ_o, ϕ_e, $d\phi_o$ and $d\phi_e$. The transmission through crossed polarizers is estimated using $T = \sin^2(2\psi)$ which assumes that any

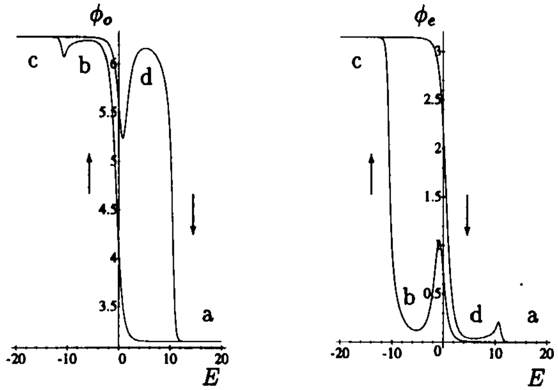

Figure 4: The director angles ϕ_o and ϕ_e as a function of applied electric field. The arrows show the path the system takes around the hysteresis loop.

changes in optical anisotropy are negligible.

Figures 2-5 show the solution to eqs. (4-5) when the electric field $E = E_{max}\cos(2\pi f t)$ is applied for the parameter values $d\phi = 0.4$, $A = 1.0$, $B_1 = 1.0$, $B_2 = 1.0$, $P = 2.0$, $\chi = 1.0$, $\epsilon\epsilon_0 = 1.0$, $\eta_1 = 1.0$, $\eta_2 = 1.0$, $E_{max} = 20.0$ and $f = 0.1$. As the field cycles from $+E_{max}$ to $-E_{max}$ and back to $+E_{max}$ the system goes from point a to point b to point c to point d and then back to point a. Figure 2 shows the transmission versus applied electric field strength, Figure 3 shows the spreads, $d\phi_o$ and $d\phi_e$, and director angles, ϕ_o and ϕ_e, as time varies and Figures 4 and 5 show the director angles and spreads as a function of field strength.

The system starts, at $t = 0$, in the FLC$_+$ state (point a in Figs. 2-5) with both directors on one side of the cone ($\phi_o = \pi$, $\phi_e = 0$) and a small amount of spread ($d\phi_o \simeq 0.1 \simeq d\phi_e$). As the field strength decreases the spread in both layers increases causing the optic axis to change and the transmission to decrease. Eventually the FLC$_+$ state loses stability and the system switches to the AFLC state as the spreads relax (point b in Figs. 2-5). The mode of instability is an anti-phase motion of the two polarization vectors, ϕ_o and ϕ_e both increase (Fig. 1 shows that an increase in both ϕ_o and ϕ_e is an anti-phase motion of the directors). After this initial anti-phase motion the polarization vectors then reorient to move in-phase, ϕ_o increases and ϕ_e decreases, and the AFLC state is attained. As the field strength becomes more negative the even layer spread, $d\phi_e$, increases and the odd layer spread, $d\phi_o$, decreases causing the optic axis to

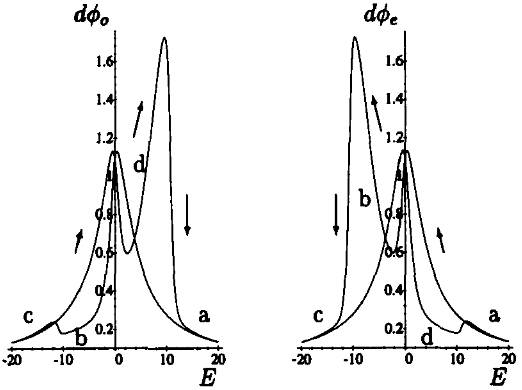

Figure 5: The spreads $d\phi_o$ and $d\phi_e$ as a function of applied electric field. The arrows show the path the system takes around the hysteresis loop.

change and the transmission to increase. This is the pre-transitional effect we aimed to model. The AFLC state loses stability as the field strength becomes sufficiently negative and the system moves to the FLC_- state with a relaxation of $d\phi_o$ (point c in Figs. 2-5). This instability is initiated by an in-phase and then an anti-phase motion. From the FLC_- state the process reverses going from point c to point d and then back to point a.

Figures 2 and 6 show the change in the hysteresis loop for applied field frequencies $f = 0.1$, 0.13 and 0.2. As the frequency increases from 0.1 to 0.13 the FLC-AFLC transition *overshoots* the AFLC state. This is seen in Fig. 6 as the sharp dip in transmission at $E = \pm 2$. The system is then *pulled back* to the AFLC state at $E \simeq \pm 5$ since it is the local minimum energy state. This overshoot increases for larger frequencies until it is large enough to overcome the energy barrier between the AFLC and FLC state. Once this has happened a return to the AFLC state is impossible and the system goes straight from FLC_+ to FLC_- (see the plot for $f = 0.2$). This overshooting behaviour has been observed experimentally[8].

CONCLUSIONS

We have presented an order parameter theory to model switching in an AFLC material. We are able to show the typical double hysteresis loop with the pre-transitional increase in transmission observed experimentally [9]. In this model the pre-transitional effect is caused by a change in order with *no* change in the director orientations whilst the switching between

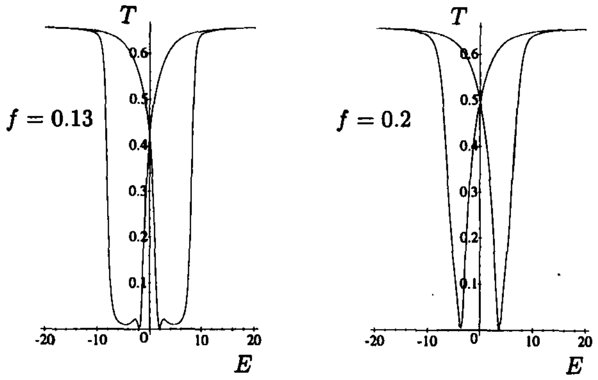

Figure 6: Transmission T versus applied electric field strength E for $E_{max} = 20$ and the frequencies $f = 0.13,\ 0.2$.

AFLC and FLC states is caused by a change in the director orientations together with a relaxation of the order.

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