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Liquid Crystals

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Online publication date: 06 August 2010

To cite this Article Mottram, N. J.(1999) 'Order parameter theory of the pre-transitional behaviour in antiferroelectric liquid crystals', *Liquid Crystals*, 26: 3, 457 – 464

To link to this Article: DOI: 10.1080/026782999205254

URL: <http://dx.doi.org/10.1080/026782999205254>

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Order parameter theory of the pre-transitional behaviour in antiferroelectric liquid crystals

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(Received 16 July 1998; in final form 9 October 1998; accepted 17 October 1998)

We propose an order parameter theory as a possible description of the pre-switching behaviour of antiferroelectric liquid crystals (AFLCs). Assuming that the antiferroelectric state is preserved so that the directors in adjacent layers are on opposite sides of the smectic cone, we consider changes in the molecular order about these directors. Our theory incorporates interlayer and intralayer dipole–dipole interactions, the dipole–electric field interaction and the thermodynamic potential. The effective optic axis as a function of applied voltage can then be calculated and is found to be in qualitative agreement with experimental observations which show a superlinear dependence.

1. Introduction

The symmetry of chiral smectic C (SmC*) liquid crystals allows for the existence of a spontaneous polarization which causes them to exhibit ferroelectric properties [1]. In this ferroelectric liquid crystal (FLC) phase the liquid crystal molecules form two dimensional layers in which the average molecular direction is tilted at an angle to the layer normal. In the antiferroelectric liquid crystal (AFLC) phase, which was discovered 15 years after the ferroelectric phase [2, 3], the director is tilted in opposite directions in adjacent layers. The polarizations of adjacent layers are therefore antiparallel and the net polarization is zero.

Interest in AFLCs has grown due to their characteristic tristate switching which may be useful in display devices. Two FLC states may be accessed from the AFLC ground state upon application of an electric field. 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 [4].

Although theories have been proposed [4–10], this switching has not been understood fully. One region of particular interest is the low field, pre-transitional regime where the effective optic axis tends to move away from the layer normal as the field increases. This variation in the effective optic axis orientation causes a change in the transmission through the device when placed between crossed polarizers. In turn this affects the achievable contrast by reducing the quality of the black state in the AFLC ground state and thus prevents the reproduction

of a large number of grey levels. Although this pre-switching regime is not understood clearly, there is strong evidence that it is not a pure soft mode as in the smectic A electroclinic behaviour [11, 12]. It has been shown that theoretical models which allow for variations of the layer directors across the cell may exhibit this pretransitional behaviour [6, 7, 9]. In such models the competition between surface and bulk effects causes an unstable bulk state to be stabilized by the surface anchoring. However, very little is known about the surface alignment and anchoring in an AFLC cell or in fact what state the liquid crystal is in at the surfaces whilst switching occurs. Choosing a realistic surface interaction energy is therefore difficult. Free-film conoscopic studies suggest [13] that there exists a region of helix unwinding before switching from the AFLC state to the FLC state occurs and it has been suggested that if the helix is present in the bulk of an AFLC cell, this could be the mechanism behind the pretransitional effect. Both of these effects may contribute to the pretransitional behaviour in AFLCs. In this paper we propose an order parameter theory which gives an alternative explanation for this regime. By fully understanding the effect of changes in order in AFLCs we intend to include this behaviour together with helix and surface effects in future work.

Experimental evidence of such changes in order has not been presented, although it is likely that it will be difficult to separate this effect from other effects, such as helix unwinding and surface stabilization, with standard experimental techniques. It is however possible that guided mode experiments [14] will be able to determine any changes in order of the system through their ability

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to determine the optic tensor components accurately. It is hoped that such techniques will be applied to AFLC materials in the future.

Order parameter theories have been effectively applied to nematic liquid crystal materials for some time (see for example [15–17]). They consider not only the average direction of the molecules but the degree of order around the direction. In regions of high distortion such as near defects, surfaces or in any region where more than one competing alignment field occurs, this change in order may be important.

We will use these ideas to construct an order parameter theory for AFLC materials. It is possible to define two independent order parameters in a tilted smectic state associated with the zenithal molecular tilt angle and the azimuthal molecular angle. The first is related to the distribution of molecules about the tilt angle whilst the second is the distribution *around* the smectic cone. Experimental evidence [11] suggests that the pretransitional regime contains no zenithal (soft mode) variation of the directors and we will thus neglect changes in zenithal order and only consider changes in azimuthal (Goldstone mode) order. With this assumption we will describe the AFLC material in terms of the usual average molecular direction within a layer and the *spread around the smectic cone* of molecules about this direction. This spread is then a measure of the order within the layer.

Given the above assumptions, the interaction of an applied electric field with the permanent molecular dipoles will tend to move molecules towards the preferred side of the smectic cone. The electric field will therefore cause an increase in the spread (a decrease in order) if the layer director is *not* on the preferred side of the smectic cone and cause a decrease in the spread (an increase in order) if the layer director *is* on the preferred side of the smectic cone.

Using this model we can formulate the free energy of a bilayer of the AFLC material. Designating the spreads in odd and even layers as $d\phi_o$ and $d\phi_e$, respectively, the energy is assumed to consist of contributions from the thermodynamic potential in each layer, the interaction between dipoles within each layer and between each layer, and the interaction between the molecular dipoles and the applied electric field parallel to the layers. In the next section we discuss each of these terms and then find the field dependence of $d\phi_o$ and $d\phi_e$, and hence the effective optic axis, from energy minimization.

2. Theory

In the pre-switching regime we will assume that the average molecular direction within each layer remains the same as in the zero field AFLC state. Therefore

the molecules in the odd layers are distributed on the opposite side of the smectic cone to the molecules in the even layers (figure 1). The mean of the distribution of molecules will remain constant, whilst the extent of the spread changes with applied field. Assuming a uniform distribution of molecules between the angles $\pi - d\phi_o$ and $\pi + d\phi_o$ (measured from the x -axis) in the odd layers and between $-d\phi_e$ and $+d\phi_e$ in the even layers, so that the spreads are $2d\phi_o$ and $2d\phi_e$, each term in the free energy outlined above can be calculated. The three energy terms corresponding to the thermodynamic potential, dipole–dipole interaction and the dipole–electric field interaction term will now be discussed in turn.

The thermodynamic potential models the excluded volume effect and should exhibit a minimum at the equilibrium values of $d\phi_o$ and $d\phi_e$. We will assume that there is no fundamental difference between the odd and even layers so that the equilibrium values of $d\phi_o$ and $d\phi_e$ are the same. The simplest form of this potential is quadratic

$$F_{\text{therm}} = A(d\phi_o - d\phi)^2 + A(d\phi_e - d\phi)^2 \quad (1)$$

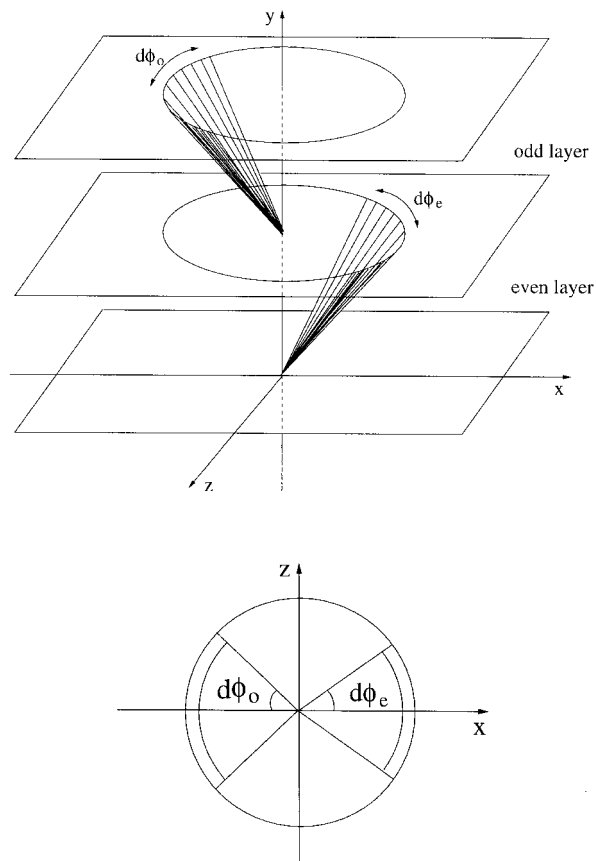


Figure 1. Molecular distributions in the odd and even layers described by the spreads $d\phi_o$ and $d\phi_e$ around the smectic cone.

This is essentially the first term in the Taylor expansion of the actual thermodynamic potential function and as such is only an approximation. For distributions that vary a great deal from $d\phi_o = d\phi = d\phi_e$ this approximation is not valid. This gives a restriction to the parameter regime where our theory may be applied, which will be discussed later. In a nematic phase, the distribution of molecules around the director is described by the order parameter $S = 1/2 \langle 3 \cos^2 \psi - 1 \rangle$ where ψ is the distribution of angles of the molecules from the average value [18]. Using the typical value of $S = 0.8$ the average molecular angle relative to the director is approximately $\langle \psi \rangle = 0.374$. Since a smectic phase is essentially a set of nematic layers stacked together, we will assume that the nematic-like order parameter *within* each layer is similar to that in the bulk nematic phase. We will therefore take the equilibrium half-spread around the smectic cone as $d\phi = 0.4$ radians.

The dipole-dipole interaction term is the sum of the energy density due to the electric potential between the dipoles within a layer and between layers. If the local molecular polarization is P , the polarization vector for each molecule is $\mathbf{P}_m = P(\cos \Phi, 0, \sin \Phi)$ where Φ is the angle between the dipole and the x -axis. Due to the reflective symmetry of the spread about the x -axis (see figure 1) the only contribution to the net polarization in a layer comes from the x component of \mathbf{P}_m . Thus integrating $P \cos \Phi$ over all molecules within the spread gives the net polarization within the even and odd layers as $P_e = P \sin(d\phi_e)/d\phi_e$ and $P_o = P \sin(d\phi_o)/d\phi_o$, respectively. From [19] the self-energy (density) of a polarization P_1 within a material of susceptibility χ is given as $P_1^2/(2\chi)$. Thus the intralayer energy density is

$$F_{\text{ddintra}} = \frac{P^2}{2\chi} \left[\left(\frac{\sin(d\phi_o)}{d\phi_o} \right)^2 + \left(\frac{\sin(d\phi_e)}{d\phi_e} \right)^2 \right]. \quad (2)$$

Since minimum self interaction energy occurs when the polarization is zero, the molecular distribution tends to spread around the cone. Consequently the intralayer term (2) is a maximum when $d\phi_o = d\phi_e = 0$ and a minimum when $d\phi_o = d\phi_e = \pi, 2\pi, 3\pi \dots$. The interaction energy (density) of two polarizations P_1 and P_2 in a material of dielectric permittivity ϵ is $-P_1 P_2 / (\epsilon \epsilon_0)$. Thus the interlayer interaction energy density is

$$F_{\text{ddinter}} = - \frac{P^2}{\epsilon \epsilon_0} \frac{\sin(d\phi_o) \sin(d\phi_e)}{d\phi_o d\phi_e}. \quad (3)$$

This term is minimum when $d\phi_o = 0 = d\phi_e$ so that the molecules in the odd layer lie on the opposite side of the smectic cone to those molecules in the even layer.

The electric field term is due to the interaction between the polarization in each layer and an applied electric field along the z -axis.

$$F_{\text{elec}} = \frac{EP}{2} \left(\frac{\sin(d\phi_o)}{d\phi_o} - \frac{\sin(d\phi_e)}{d\phi_e} \right). \quad (4)$$

The total energy density is therefore

$$F = A [(d\phi_o - d\phi)^2 + (d\phi_e - d\phi)^2] + \frac{P^2}{2\chi} \left[\left(\frac{\sin(d\phi_o)}{d\phi_o} \right)^2 + \left(\frac{\sin(d\phi_e)}{d\phi_e} \right)^2 \right] - \frac{P^2}{\epsilon \epsilon_0} \frac{\sin(d\phi_o) \sin(d\phi_e)}{d\phi_o d\phi_e} + \frac{EP}{2} \left(\frac{\sin(d\phi_o)}{d\phi_o} - \frac{\sin(d\phi_e)}{d\phi_e} \right). \quad (5)$$

Since the susceptibility and the permittivity are of the same order, we will assume $\chi = \epsilon \epsilon_0$ and the free energy density becomes

$$F = A [(d\phi_o - d\phi)^2 + (d\phi_e - d\phi)^2] + \frac{P^2}{2\epsilon \epsilon_0} \left(\frac{\sin(d\phi_o)}{d\phi_o} - \frac{\sin(d\phi_e)}{d\phi_e} \right)^2 + \frac{EP}{2} \left(\frac{\sin(d\phi_o)}{d\phi_o} - \frac{\sin(d\phi_e)}{d\phi_e} \right). \quad (6)$$

This assumption will subsequently be used to enable us to obtain simple analytic solutions. It will also be used when solving the system numerically in order to be able to compare the analytic and numeric solutions. We are able numerically to solve the system without the constraint $\chi = \epsilon \epsilon_0$, but since the qualitative behaviour remains unchanged for $\chi \neq \epsilon \epsilon_0$, we will use the free energy density expression (6).

The energy density (6) may be non-dimensionalized to

$$\mathcal{F} = [(d\phi_o - d\phi)^2 + (d\phi_e - d\phi)^2] + \beta \left(\frac{\sin(d\phi_o)}{d\phi_o} - \frac{\sin(d\phi_e)}{d\phi_e} \right)^2 + \gamma \left(\frac{\sin(d\phi_o)}{d\phi_o} - \frac{\sin(d\phi_e)}{d\phi_e} \right) \quad (7)$$

where $\mathcal{F} = F/A$, $\beta = P^2/(2A\epsilon\epsilon_0)$ and $\gamma = EP/(2A)$. The free energy is then the integral of the free energy over a fixed volume which, since we are considering the bulk of the material and all variables are spatially homogeneous, is simply proportional to the free energy density. For statically stable solutions, the governing equations are thus obtained by minimizing the free energy density

with respect to the two variables $d\phi_o$ and $d\phi_e$,

$$0 = \frac{d\bar{f}}{d(d\phi_o)} \quad (8)$$

$$0 = \frac{d\bar{f}}{d(d\phi_e)}. \quad (9)$$

The free energy density consists of terms favouring spreading (F_{ddintra} and F_{elec}), no spreading (F_{ddinter}) and a fixed amount of spreading (F_{therm}). It is the relative magnitudes of these terms that will determine how an applied electric field changes $d\phi_o$ and $d\phi_e$. An increase in β will tend to increase both spreads $d\phi_o$ and $d\phi_e$,

whilst an increase in γ will tend to increase $d\phi_o$ and decrease $d\phi_e$. Variations in the third equation parameter $d\phi$ change the equilibrium solution at zero field ($\gamma = 0$).

We can analytically investigate the behaviour around the equilibrium solution, $d\phi_o = d\phi = d\phi_e$, for small values of the field parameter γ . For higher field strengths we can find exact solutions numerically.

3. Analysis

Since we know the solution for $\gamma = 0$ we can perturb around this point assuming that the field remains small. Therefore using the perturbation parameter $\gamma \ll 1$ we

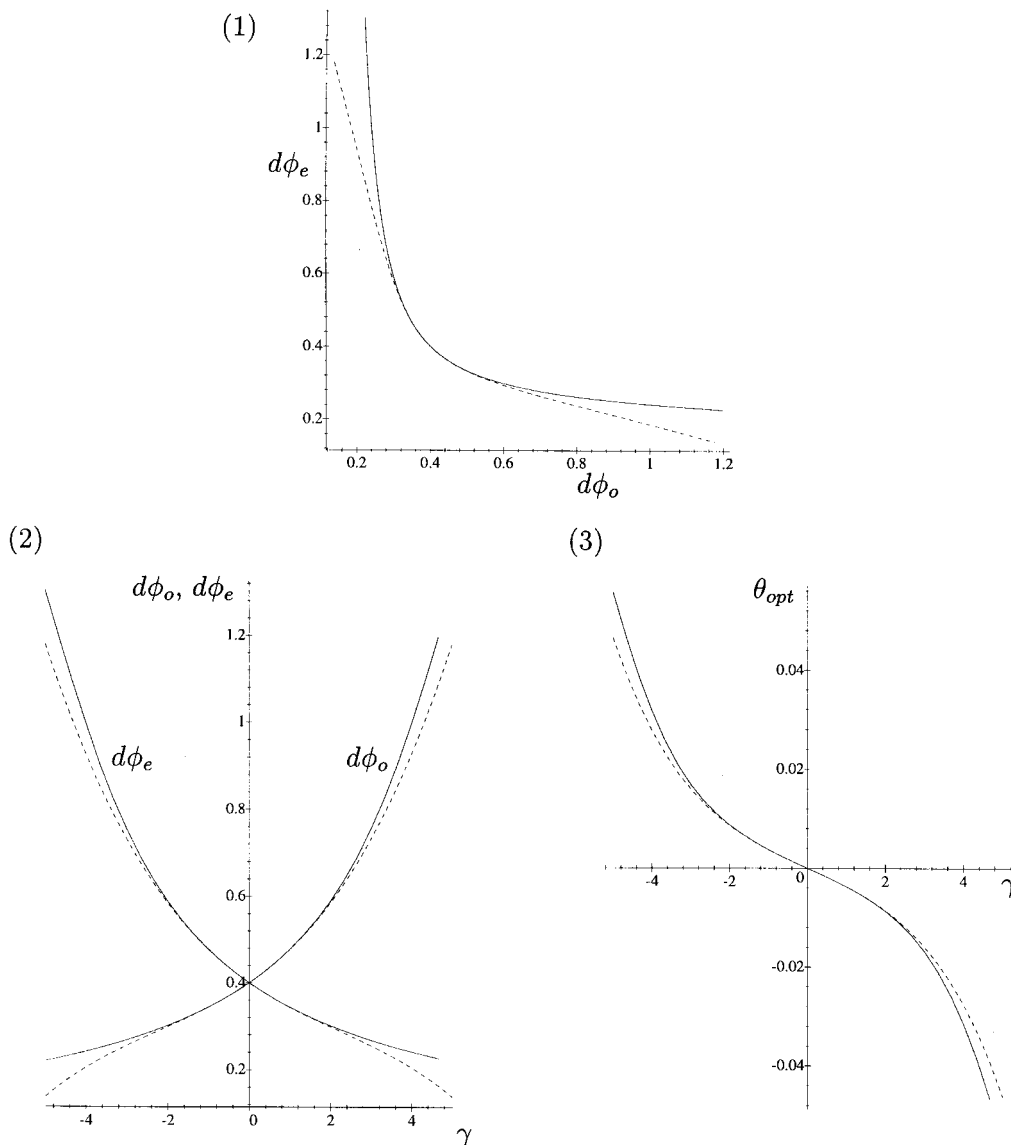


Figure 2. Comparison between numerical and analytical solutions for $\beta = 0.1$, $d\phi = 0.4$. The dashed lines are analytical solutions and the solid lines are numerical solutions. (1) $d\phi_e$ versus $d\phi_o$ as the field parameter γ varies; (2) $d\phi_o$, $d\phi_e$ versus γ ; (3) optic axis tilt angle θ_{opt} versus γ .

substitute the approximate solutions

$$d\phi_o = d\phi + \gamma d\phi_{o1} + \gamma^2 d\phi_{o2} + \gamma^3 d\phi_{o3} + \dots \quad (10)$$

$$d\phi_e = d\phi + \gamma d\phi_{e1} + \gamma^2 d\phi_{e2} + \gamma^3 d\phi_{e3} + \dots \quad (11)$$

into equations (8, 9) to obtain a set of equations for $d\phi_{oi}$ and $d\phi_{ei}$. The first order solutions are

$$d\phi_{o1} = \frac{1}{2} \frac{d\phi^2 (\sin(d\phi) - \cos(d\phi)d\phi)}{d\phi^4 + \beta(\sin(d\phi) - \cos(d\phi)d\phi)^2} \quad (12)$$

$$d\phi_{e1} = d\phi_{o1}. \quad (13)$$

The higher order terms may be calculated analytically but are too lengthy to present here. Figure 2 shows the $d\phi_o$ and $d\phi_e$ solutions and the effective optic axis orientation θ_{opt} which were calculated using the first four terms of the expansion (10, 11) and the parameter

values $\beta = 0.1$, $d\phi = 0.4$. Figures 2(1) and 2(2) show the relationship between the $d\phi_o$ and $d\phi_e$ as γ varies. For positive γ the value of $d\phi_o$ increases as $d\phi_e$ decreases, whilst for negative γ the opposite occurs. As a first approximation to the effective optic axis orientation we have averaged the angle between the y -axis and the projection of each molecule onto the xy -plane for all the molecules in each layer assuming that the smectic cone angle was $\theta = 25^\circ$. The optic axis as a function of the applied field is shown in figure 2(3).

4. Numerics

The numerical solutions to equations (8, 9) are shown in figure 2 for the parameter values $d\phi = 0.4$, $\beta = 0.1$ with γ varying between ± 5 . Figure 2 shows the good agreement between the analytical solutions of the

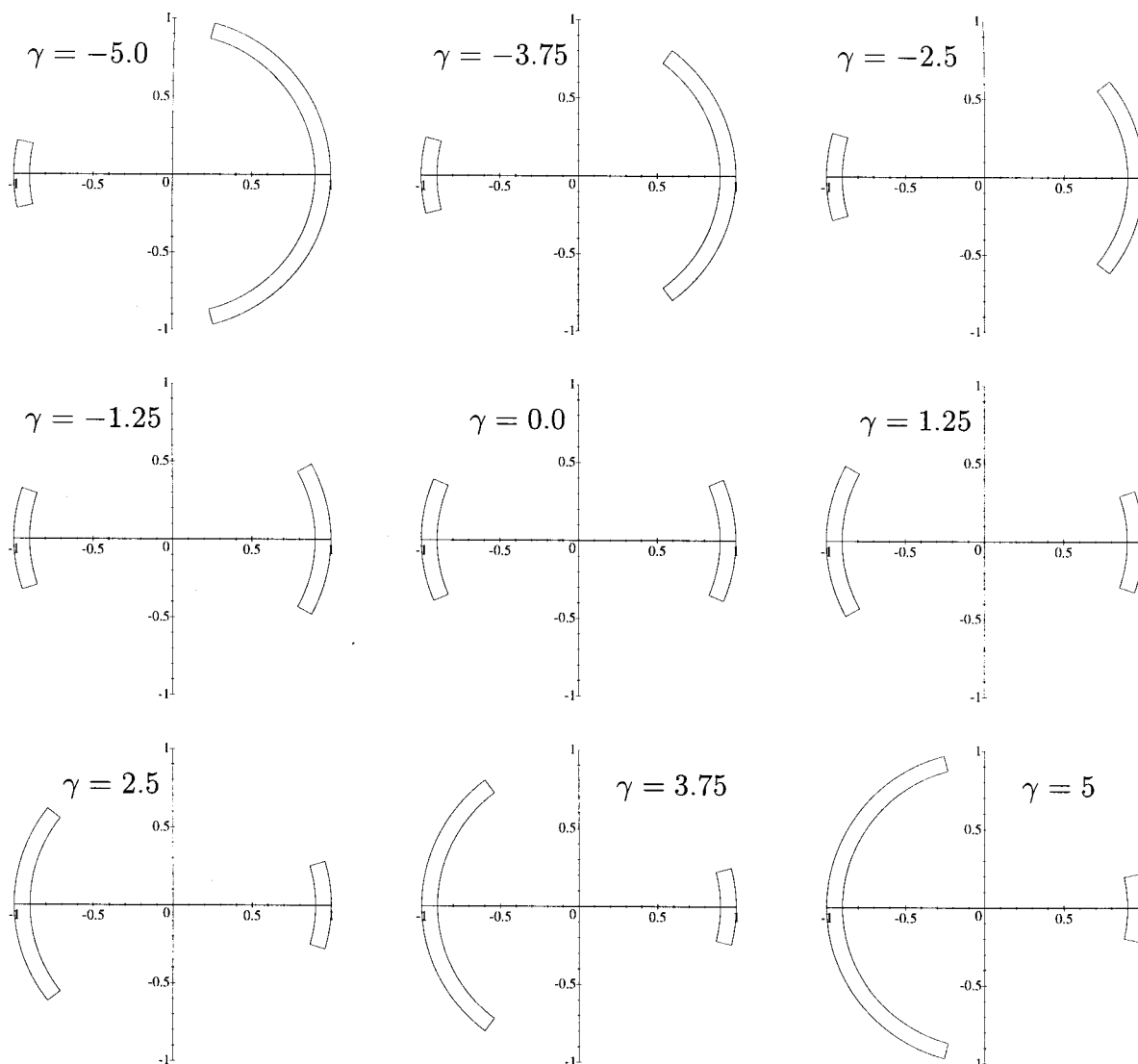


Figure 3. Variation of $d\phi_o$ and $d\phi_e$ for $\beta = 0.1$ as the field parameter γ varies from -5 to $+5$.

previous section and the numerical solution. For $\gamma \ll 1$, the analytic solution is in exact agreement with the numerical solution, as is expected, but we see that for relatively large values of γ (up to $\gamma \approx 2$) the agreement is still extremely good.

Figure 3 shows the change in the spreads $d\phi_o$ and $d\phi_e$ around the smectic cone as γ increases from -5 to 5 . We clearly see the spreading of one layer and the compression of the other. The odd layer is becoming less ordered and the even layer more ordered. At a high enough field strength, the odd layers will be forced to switch around the cone to the opposite side forming the FLC state. However, in the present model there is no continuous transformation from the AFLC state to the FLC state. In order to form the FLC state, the system would have to undergo a first order transition in which, for example, the spread of molecules splits and reforms on the other side of the cone or the mean position of the spread, i.e. the director angle, rotates around the cone. This is consistent with experimental observations which show that the AFLC–FLC transition is first order.

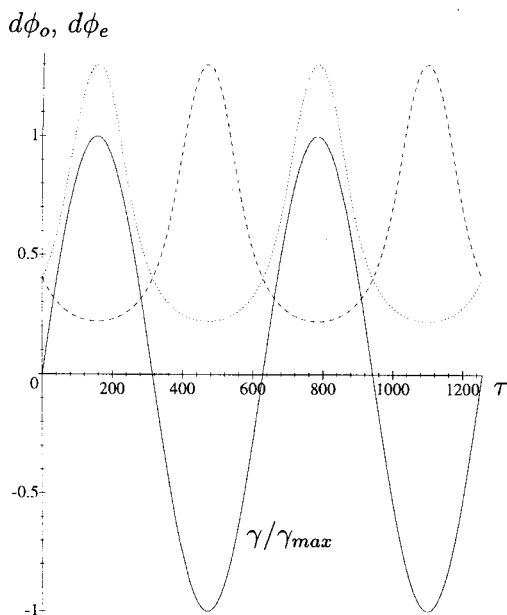
We can also model the dynamics of the pretransitional regime when a sinusoidal field

$$\gamma = \gamma_{\max} \sin \omega t \quad (14)$$

is applied using the equations,

$$\frac{\lambda}{A} \frac{d(d\phi_o)}{dt} = - \frac{d\mathcal{F}}{d(d\phi_o)} \quad (15)$$

$$\frac{\lambda}{A} \frac{d(d\phi_e)}{dt} = - \frac{d\mathcal{F}}{d(d\phi_e)} \quad (16)$$



where λ is the viscosity of the spreading motion. The rate of change of the variables $d\phi_o$ and $d\phi_e$ is then proportional to the gradient of the free energy at that point. Non-dimensionalizing time in equations (15, 16) with λA leads to

$$\frac{d(d\phi_o)}{d\tau} = - \frac{d\mathcal{F}}{d(d\phi_o)} \quad (17)$$

$$\frac{d(d\phi_e)}{d\tau} = - \frac{d\mathcal{F}}{d(d\phi_e)} \quad (18)$$

where $\tau = At/\lambda$ and the applied electric field is $\gamma = \gamma_{\max} \sin \Omega\tau$ where $\Omega = \omega\lambda A$.

Figures 4, 5, 6 and 7 show the response of $d\phi_o$, $d\phi_e$ and the effective optic axis to the sinusoidal field for frequencies $\Omega = 0.01, 0.1, 0.5$ and 1.0 and maximum field strength $\gamma_{\max} = 5$. As the frequency of the applied field increases, the response *lags* behind, creating a dynamic hysteresis loop. With increasing frequency, the maximum achievable effective optic axis tilt is reduced and the loop becomes wider as the phase difference between the applied electric field and the response increases.

5. Conclusions

We have presented an order parameter theory which gives a possible explanation of the pretransitional behaviour in an AFLC under an applied electric field. Using a bilayer, bulk model and considering the field-induced change in order of the AFLC phase, we have been able to show a superlinear dependence of the effective optic axis as the field varies. Our theory

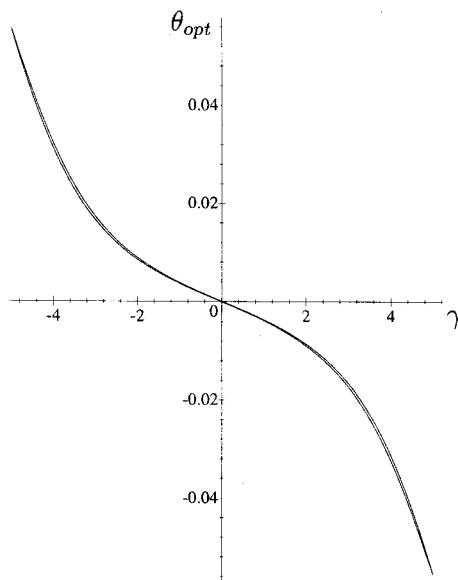


Figure 4. Dynamic response when $\Omega = 0.01$ and $\gamma_{\max} = 5$. In the first plot the solid line is the normalized field parameter γ/γ_{\max} , the dashed line is $d\phi_e$ and the dotted line is $d\phi_o$. The second plot shows the effective optic axis tilt angle θ_{opt} as a function of γ .

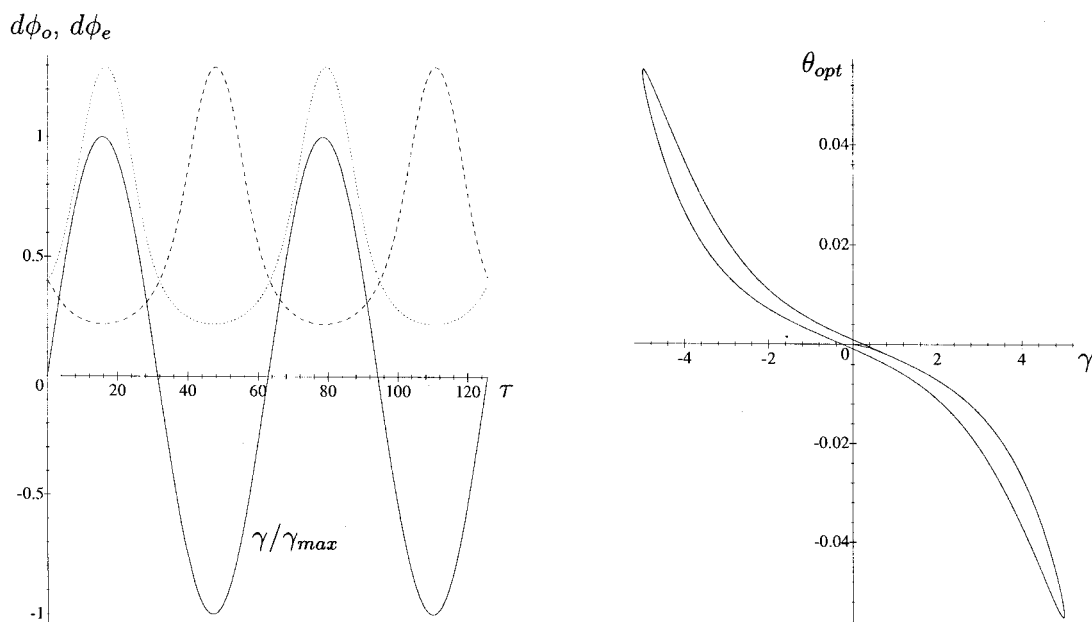


Figure 5. Dynamic response when $\Omega = 0.1$ and $\gamma_{\max} = 5$. The lines in each plot are defined as in figure 4.

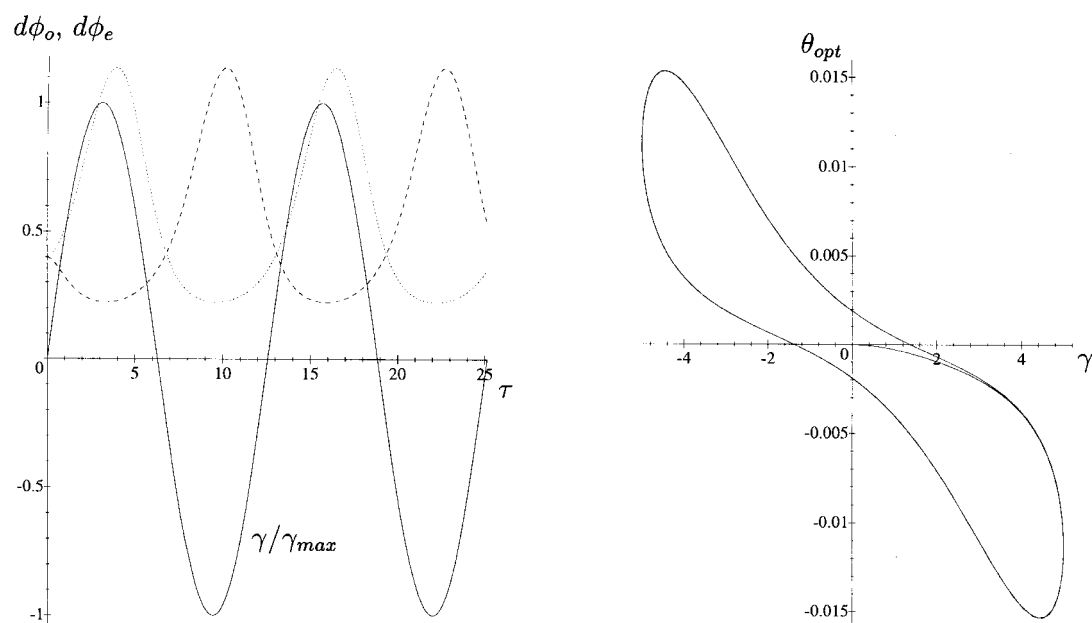


Figure 6. Dynamic response when $\Omega = 0.5$ and $\gamma_{\max} = 5$. The lines in each plot are defined as in figure 4.

therefore supports previous experimental observations which have suggested that the pretransitional regime is *not* due to soft mode motion of the director [11, 12]. In comparison to a soft mode description of the pretransitional regime, which could go from the AFLC state to the FLC state through a continuous, second order transition, it is clear from the present model that in order to form the FLC state the system would have to undergo a first order transition. This could involve

the spread of molecules splitting and reforming on the other side of the cone or the mean position of the spread, i.e. the director azimuthal angle, rotating around the cone. This is consistent with experimental observations which show that the AFLC–FLC transition is first order. Although the transition from AFLC to FLC is not allowed in the present theory, a model to include the full switching behaviour will be presented in future work.

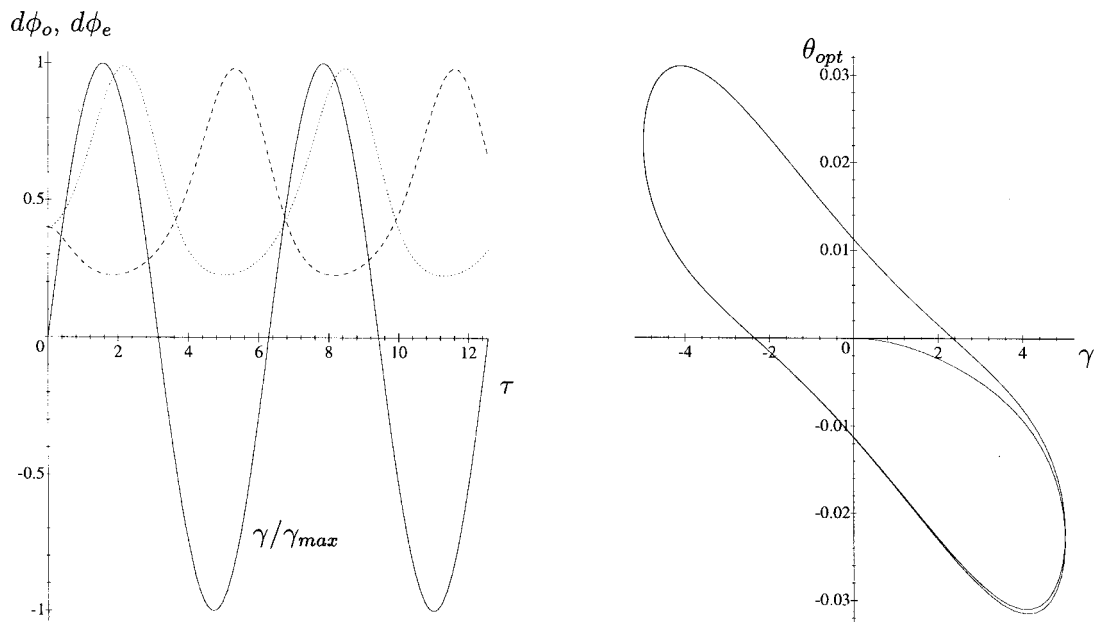


Figure 7. Dynamic response when $\Omega = 1.0$ and $\gamma_{max} = 5$. The lines in each plot are defined as in figure 4.

The authors would like to acknowledge useful discussions with Romeo Beccherelli and the financial support of Sharp Laboratories of Europe Ltd. and the EPSRC.

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