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Electrohydrodynamic Solutions for Nematic Liquid Crystals with Positive Dielectric Anisotropy

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Electrohydrodynamic Solutions for Nematic Liquid Crystals with Positive Dielectric Anisotropy†‡

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Abstract—The steady state electrohydrodynamic equations of motion have been solved for nematic liquid crystals which are characterized by a positive dielectric anisotropy and possess a parallel homogeneous texture when contained between transparent electrodes. It is found that a periodic distortion of the director is possible above a critical voltage which is a function of the material constants. The optical texture should be similar to that observed in the Williams domain mode. The wavelength of the distortion is a function of voltage, and the relationship is very similar to the dispersion relation calculated for a negative dielectric anisotropy material in the homeotropic texture. These calculations demonstrate that the numerous experimental observations of domain structures have a common explanation in terms of the continuum theory. A physical explanation is given for the double valued solutions observed in the parallel homogeneous negative dielectric anisotropy problem.

1. Introduction

Electrohydrodynamic instabilities are known to exist in nematic liquid crystals which possess a negative dielectric anisotropy and exhibit a parallel homogeneous texture.[1,2] The fundamental physics of these effects is now reasonably well understood.[3,4] Recently several authors have observed a domain structure in positive dielectric anisotropy material in the same geometry.[5,6,7] Some question has arisen in the literature as to whether conduction induced torques can produce domains in positive material. de Gennes[8] has shown that domains will *not* appear in positive material

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in the *homeotropic* geometry. de Jeu *et al.*[5] extended Helfrich's theory to show that domains should be observed in the parallel homogeneous case with positive material. Reference 5 implies that de Gennes predicts domains only for negative material. A more accurate description of the de Gennes prediction is given above. Both the sign of the dielectric anisotropy and the boundary conditions are necessary to characterize an experiment.

Penz and Ford[4] have demonstrated that important additional information can be obtained from a rigorous solution of the boundary value problem in two dimensions. We have varied the magnitude and sign of the dielectric anisotropy in the electrohydrodynamic problem to determine the influence on the type of theoretical solutions possible. Section 2 gives a description of the method used to solve the problem, and Sec. 3 gives a physical interpretation of the method. Section 4 gives the results of computer solutions of the problem. A discussion of the results is presented in Sec. 5.

2. Theoretical Method

The technique for solving the boundary value problem associated with the parallel homogeneous geometry has been extensively discussed in Ref. 4. We assume the experiments are governed by Maxwell's equations and the normal equations of hydrodynamics. We adopt linear constitutive equations, e.q., the five viscosities developed by Leslie[9] are employed as is Ohm's Law. We assume that the fluid is incompressible. The coordinate system, relative to the experimental geometry, is shown in Fig. 1. The director is assumed to have a spatial dependence of the form $\mathbf{n} = (1, 0, \theta_1 \exp i[q_x x + q_z z])$, where the x direction is defined by rubbing the capacitor plates and the z direction is parallel to the applied electric field E_0. The response electric field $\mathbf{E}_1$, the fluid velocity $\mathbf{v}_1$ and the pressure changes p_1 are all assumed to have a similar two dimensional spatial variation. These trial functions are substituted into the differential equations, and all second order terms in E_1, v_1, θ_1, and p_1 are dropped; i.e., the problem is linearized. All time derivatives are assumed to be zero; we treat the steady state problem. The resultant secular equation is an eighth order equation in $S \equiv q_z/q_x$ as a function of E_0/q_x. For any given q_x and E_0, there are eight plane

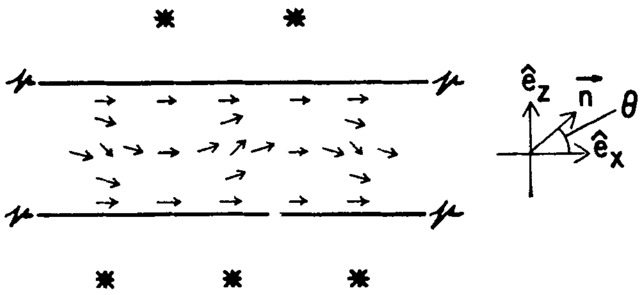

Figure 1. Director Orientation. The figure shows a cross section of the experimental geometry: a capacitor filled with liquid crystal. The electric field is applied in the z direction ($\hat{e}_z$), and the electrodes are rubbed to promote orientation of the director in the x direction ($\hat{e}_x$). The director **n** is indicated by the small arrows, and its orientation relative to the x-axis is measured by the angle θ. We seek solutions of the electrohydrodynamic equations which exhibit this pattern. The domain lines are indicated by stars above and below the sample. It can be shown (Ref. 1) that the domain lines are images of a light source below the sample as seen by an observer above the sample.

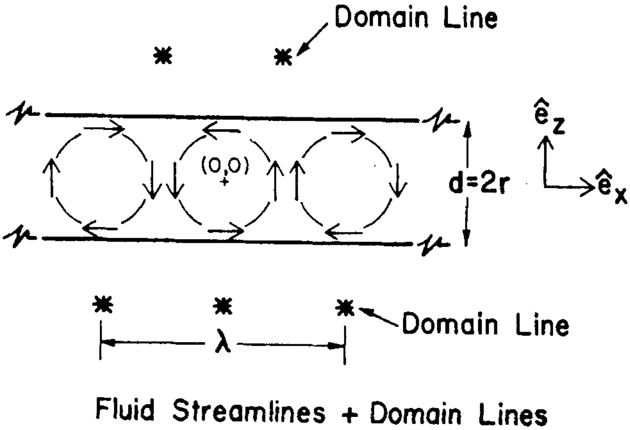

Figure 2. Flow Pattern. The fluid flow pattern is a series of vortices with the vorticity antiparallel in adjacent cells. The periodicity of the fluid motion and the director distortion is specified by $q_x = 2\pi/\lambda$. d is the thickness of the sample.

waves which satisfy the infinite medium differential equations. (See Eq. (30) in Ref. 4.)

There are eight boundary conditions associated with the problem. The director is constrained by surface forces to lie parallel to the surface at both surfaces. The capacitor faces are conductors, and so $\mathbf{E}_1$ must have zero x-component at both faces. The fluid velocity pattern is shown in Fig. 2. Both the x and z components of $\mathbf{v}_1$ must be zero at both boundaries. These eight boundary conditions lead to a second secular equation which is an implicit relationship between q_x and E_0. Thus only certain distortions q_x are possible given an applied electric field. Due to the complexity of the algebra, the problem must be solved by digital computer. The results will be presented in Sec. 4 after the relevant physical processes are sketched.

3. Physical Model

Consider the physical mechanisms at work in the processes described in Figs. 1 and 2. There must be conservation of linear momentum, i.e., the space charge induced force $(\nabla \cdot \mathbf{D})E_0$ must be balanced by viscous forces $\eta q^2 v_1$, where $\mathbf{D}$ is the displacement field, q is some representative wave vector, and η is a normal viscosity. Thus:

$$\eta q^2 v_1 + q \epsilon_0 \epsilon E_0 E_1 = 0 \tag{1}$$

where ϵ_0 is the permittivity of free space and ϵ is a relevant dielectric constant. Charge must also be conserved; i.e., $\nabla \cdot (\underset{\approx}{\sigma}\mathbf{E}) = 0$ where $\underset{\approx}{\sigma}$ is a conductivity tensor containing an element describing the conductivity parallel to the director $\sigma_\parallel$ and perpendicular $\sigma_\perp$. This condition yields:

$$E_1 = -\frac{\sigma_\parallel - \sigma_\perp}{\sigma_\parallel} E_0 \theta_1. \tag{2}$$

Finally, the three relevant torques must balance to give a steady state angular momentum. There is the dielectric torque $\mathbf{P} \times \mathbf{E}$, where $\mathbf{P}$ is the polarization vector; the elastic torque $Kq^2\theta_1$ where K is an appropriate Franck elastic constant; the shear torque $\alpha q v_1$ where α is an appropriate Leslie coefficient which describes the torque on the director when a liquid crystal experiences a shear qv_1. The torque balance is:

$$\alpha q v_1 - \epsilon_0(\epsilon_{\parallel} - \epsilon_{\perp})E_0{}^2\theta_1 + q^2 K\theta_1 = 0. \tag{3}$$

Using Eqs. (1) and (2) to eliminate v_1 from Eq. (3), one gets:

$$\theta_1 \left\{ \left[-\frac{\alpha}{\eta}\epsilon \frac{(\sigma_{\parallel} - \sigma_{\perp})}{\sigma_{\parallel}} + (\epsilon_{\parallel} - \epsilon_{\perp}) \right] \epsilon_0 E_0{}^2 - q^2 K \right\} = 0. \tag{4}$$

Equation (4) is a simplified version of Helfrich's original electrohydrodynamic expression.[3] We can see the origin of all the torques, including the shear torque shown in the first term. Anisotropic conductivity leads to a space charge which leads to a shear flow which in turn leads to a shear torque. Note that the distortion amplitude θ_1 is a factor of Eq. 4, a feature common to all linear analysis. Aside from the trivial solution $\theta_1 = 0$, a linear analysis will not predict the distortion amplitude. There are three possible regimes for the solution of Eq. (4); E_0 small, medium and large. The case of E_0 small is uninteresting when we are concerned with electric field induced effects. For E_0 intermediate in size, a solution of the form $E_0 \propto q$ should be expected. As the electric torques are increased, the elastic torques can only increase by decreasing the period of the distortion.

The possibility of a qualitatively different mode at large E_0 can only be seen by referring to the complete analysis of Ref. 4:

$$\left[-\frac{(\alpha_2 - S^2\alpha_3)}{(\eta_2 + S^2\eta_1)}\epsilon_{\perp}\left(\frac{\sigma_{\parallel}}{\sigma_{\perp}} - \frac{\epsilon_{\parallel}}{\epsilon_{\perp}}\right) + (\epsilon_{\parallel} - \epsilon_{\perp})(1 + S^2) \right] \epsilon_0 E_0{}^2 - q_x{}^2 \times (k_{33} + S^2 k_{11})\left(\frac{\sigma_{\parallel} + S^2\sigma_{\perp}}{\sigma_{\perp}}\right) = 0 \tag{5}$$

Note that when E_0 is large, the solutions for S in Eq. (5) will be determined primarily by the terms inside the square brackets. Since E_0 is a factor, the problem will be relatively independent of E_0. The coefficient of $E_0{}^2$ in Eq. (5) contains the torques due to conduction anisotropy and dielectric anisotropy. For a $\epsilon_{\perp} > \epsilon_{\parallel}$ and $\sigma_{\parallel} > \sigma_{\perp}$, these torques are of opposite sign. In the one dimensional analysis of Eq. (4) (Note: generally $|\alpha_2| \gg |\alpha_3|$, $\alpha_2 < 0$), an accidental balance would require very special material constants. In a two-dimensional analysis, however, there are several possible plane wave solutions. The net torque on a small volume of fluid will be the sum of the torques due to all the plane waves; it can in fact be zero. Thus, it is possible to have a solution where q_z/q_x is not a function of E_0.

The boundary conditions will in general fix a minimum value for q_z. The distortions cannot have a larger wavelength than the thickness of the sample. This semi-quantitative analysis would then indicate two types of solutions above a minimum $q = \sqrt{q_x^2 + (\pi/d)^2}$; i.e., a minimum voltage. The first solution would show $q_x \propto E_0$ and the other solution would have q_x relatively independent of E_0. In the first case, the torques associated with the electric field drive the instability and the elastic torques oppose it. In the second case, the conduction induced torque drives the instability and the dielectric torque opposes it ($\epsilon_\perp > \epsilon_\parallel$).

4. Numerical Results

The solutions to the parallel homogeneous boundary value problem for various values of the dielectric anisotropy are presented in Fig. 3. The other material constants used in the calculations were those measured for MBBA at 25 °C. For the exact values, see Ref. 4. In particular, it was assumed that $\sigma_\parallel/\sigma_\perp = 1.5$. We realize that it would be very difficult to change only $\epsilon_\parallel/\epsilon_\perp$ in any mixing process, e.g., PEBAB in MBBA. Our approach is expedient theoretically and can be expected to yield qualitative results. Figure 3 presents the dispersion relations relevant to the fundamental solution for each $\epsilon_\parallel/\epsilon_\perp$, i.e., one layer of vortices in the sandwich. The higher order harmonics,[4] two, three layers, etc., have been suppressed for clarity.

The ordinate of Fig. 3 is $\pi d/\lambda$ or $q_x d/2$. Given an applied voltage V_0, only certain values of $\pi d/\lambda$ are permitted by the boundary value problem. Consider the case $\epsilon_\parallel/\epsilon_\perp = 0.9$, appropriate to pure MBBA. Above a critical voltage $V_c = 6.9$ V, two types of solutions are possible. One branch exhibits $\pi d/\lambda$ approximately proportional to V_0. This is the $q_x \propto E_0$ solution predicted in the previous section and has been identified with the mode observed by Greubel and Wolff.[2] The lower branch of the $\epsilon_\parallel/\epsilon_\perp = 0.9$ solution becomes insensitive to V_0 above 8 V. This qualitative behavior was foreseen

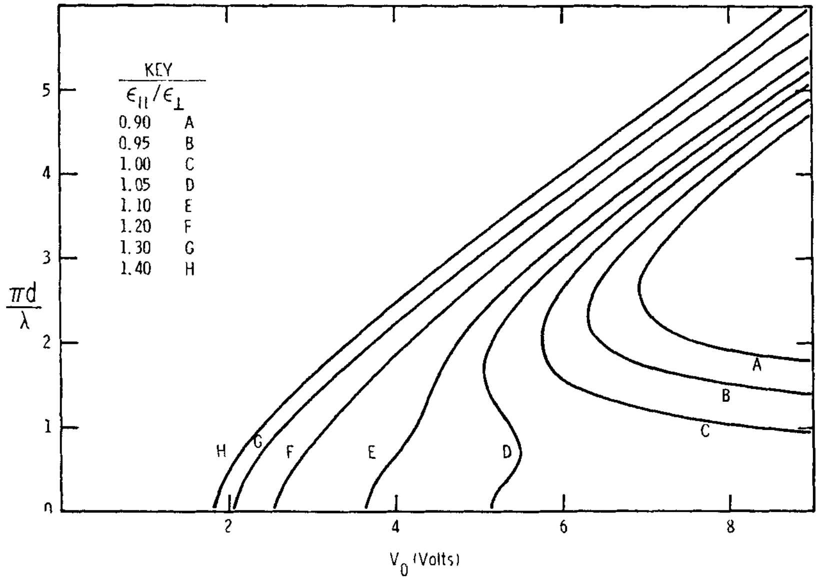

Figure 3. Solutions as a Function of Applied Voltage. As described in the text, the steady state, linear boundary value problem can be solved for unique combinations of $\pi d/\lambda$ and V_0. The figure shows the solutions for single layer vortex motion with dielectric anisotropy as a parameter. For $\epsilon_{\parallel}/\epsilon_{\perp} < 1$, the solutions are double valued as a function of voltage (MBBA: $\epsilon_{\parallel}/\epsilon_{\perp} = 0.9$). The lower branch results from a balance between the conduction induced torque and the dielectric torques. The upper branch results from conduction induced plus dielectric torques opposed by elastic torques. For positive dielectric material, the conduction induced and dielectric torques are parallel, and the voltage independent mode disappears. The variable grating type mode continues to exist.

in the physical discussion above. Experimental observations have shown that the domain wave vector increases linearly with the voltage in thin samples.[2] This also seems to be the case in thick samples, although dynamic scattering makes measurement difficult. Thus we tentatively identify the upper branch with the experimental observations.

There is a clear trend for the critical voltage to decrease as $\epsilon_{\parallel}/\epsilon_{\perp}$ is increased. By critical voltage we mean that voltage at which any value of $\pi d/\lambda$ becomes a solution of the problem. This definition is necessary due to the fact that the dispersion relation exhibits a qualitatively different form as $\epsilon_{\parallel}$ becomes greater than $\epsilon_{\perp}$, i.e., positive material. The voltage independent branch disappears, and the linear $\pi d/\lambda$ versus V_0 branch survives. This behavior can be understood using the same phenomenological arguments discussed previously. The voltage independent mode results from the balance of the dielectric torque and the conduction induced torque. For $\epsilon_{\perp} > \epsilon_{\parallel}$ and $\sigma_{\parallel} > \sigma_{\perp}$, these torques act in opposition; a solution is possible. For $\epsilon_{\parallel} > \epsilon_{\perp}$ and $\sigma_{\parallel} > \sigma_{\perp}$, the torques are parallel; no V_0 independent solution exists.

Objections have been raised regarding our interpretation of the two steady state solutions for negative dielectric anisotropic material in terms of the experimentally observed domains. Helfrich[10] contends that the line $\pi d/\lambda$ vs. V_0 only bounds a region of solutions possible in the time dependent problem. It is not possible to directly answer this objection without performing a time dependent analysis. Such an analysis is under way at the present time.

5. Discussion of the Freedericksz Transition

The parallel homogeneous geometry with positive material is most often used in a field effect mode. The general observation is uniform ($\lambda = \infty$) tipping of the director above a critical voltage. This effect has become known as the Freedericksz transition.[11,12] In order to investigate the relationship between the present calculation, which includes conductivity and dielectric effects, and the Freedericksz formula, the critical voltages in Fig. 3 are plotted as a function of $\epsilon_{\parallel}/\epsilon_{\perp}$ in Fig. 4. This dependence has been observed by Flint and Carr[7] and by de Jeu *et al.*[13] For $\epsilon_{\parallel}/\epsilon_{\perp} > 1.2$, our calculations agree with the Freedericksz expression $V_c = \pi(k_{11}/\epsilon_0(\epsilon_{\parallel} - \epsilon_{\perp}))^{1/2}$. For sufficiently positive dielectric anisotropy, the critical voltage is primarily determined by dielectric vs. elastic torques, even when $\sigma_{\parallel} \neq \sigma_{\perp}$.

The agreement of the dielectric versus dielectric plus conduction torque calculations can be seen in two ways. First consider that a Freedericksz transition is characterized by $q_x = 0$. In this limit, Eq. (5) reduces to an equation for q_z. To satisfy the boundary conditions $q_z = \pi/d$. Under these conditions, Eq. (5) is identical with the Freedericksz formula. A different method of expressing the same idea is to note that neither $\nabla \cdot \mathbf{D}$ nor $\nabla \cdot \mathbf{J}$ can be different

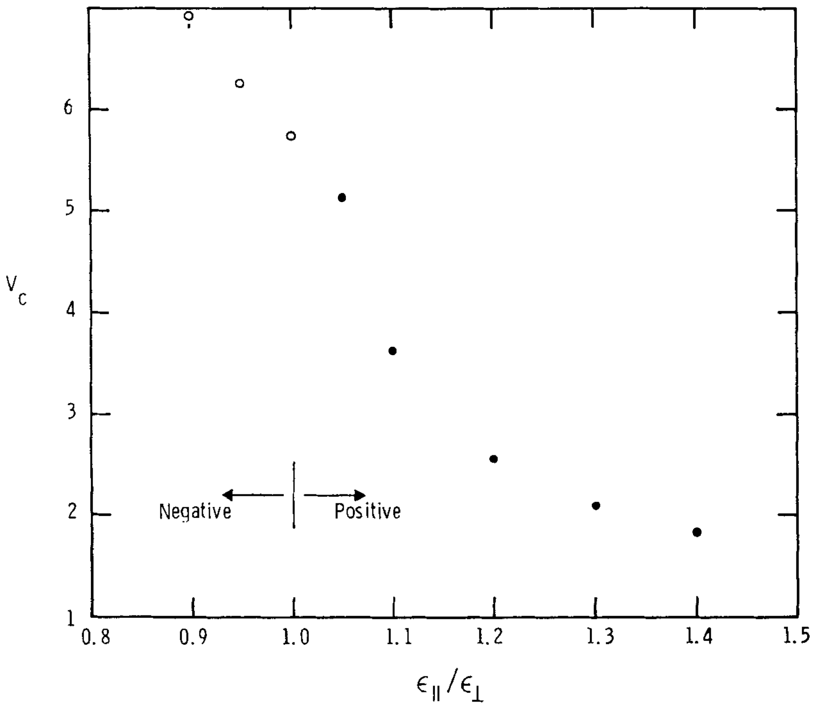

Figure 4. Critical Voltage as a Function of Dielectric Anisotropy. The minimum voltage for any type of domain formation decreases as $\epsilon_{\|}/\epsilon_{\perp}$ increases. As $\epsilon_{\|}/\epsilon_{\perp}$ crosses unity, the dielectric torque changes from a restoring torque ($\epsilon_{\|} < \epsilon_{\perp}$) to a driving torque ($\epsilon_{\|} > \epsilon_{\perp}$). The critical voltage decreases accordingly, reaching an asymptotic value of $\pi[k_{11}/\epsilon_0 (\epsilon_{\|} - \epsilon_{\perp})]^{\frac{1}{2}}$. The open circles indicate a double valued solution at threshold. The full circles indicate a single valued solution.

than zero when $q_x = 0$ and $E_1 = 0$. Since charge continuity is automatically satisfied, conductivity and viscosity will not enter the problem.

The characteristics of the problem above threshold are also interesting. For $\epsilon_{\|}/\epsilon_{\perp} = 1.4$ the $\pi d/\lambda$ versus V_0 dispersion relation is not sensitive to changes in either conductivity or viscosity. This observation suggested that domain structure can arise in an insulating liquid crystal with positive dielectric anisotropy. The boundary value problem associated with zero conductivity is much simpler than the full problem discussed above. It is possible to solve the two dimensional problem with a single sinusoidal function. The solution is qualitatively similar to the $\epsilon_{\|}/\epsilon_{\perp} = 1.3$, 1.4, etc., curves in

Fig. 3. The threshold voltage is given by the Freedericksz expression. The slope of the linear region is given by:

$$\mathrm{d}(\pi d/\lambda)/\mathrm{d}V_0 = \tfrac{1}{2}\left[\frac{(\epsilon_{\parallel} - \epsilon_{\perp})\epsilon_0}{k_{33}}\frac{\epsilon_{\perp}}{\epsilon_{\parallel}}\right]^{1/2} \quad V_0 \gg V_c,\ \sigma = 0.$$

If $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are known, k_{11} can be determined by measuring V_c and k_{33} can be obtained from the slope of the dispersion relation.

These calculations show that domains are possible *without* fluid flow. The critical voltage represents a balance between the dielectric torque and the elastic torque associated with q_z. As V_0 is increased, q_z must remain fixed at $\pi d/\lambda$ to satisfy the boundary conditions. The sample must develop a q_x distortion to balance the increased dielectric torque if the problem is restricted to linear terms in θ_1 and E_1. Domains appear to increase the elastic torques in the sample.

The solution of the linear electrohydrodynamic problem for positive dielectric material may be somewhat academic, however, as domain observation is the exception rather than the rule. This analysis shows that a uniform texture above threshold cannot be explained by the small amplitude approximation.

Aslaksen[14] has recently reported a microscopic model of the electrohydrodynamic effects. He reports that space charge cannot arise under the conditions given in this paper. The apparent paradox results from the fact that Aslaksen did not use Ohm's law (see Eq. (9), Ref. 14) for the *steady state* relationship between current density and electric. It is standard practice to define the current density in terms of the undisturbed number density of charged particles.[15] The addition of Aslaksen's ρ' term in the current density expression requires justification. We have shown here that the divergence of the displacement vector is not generally zero in a two dimensional model.

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