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Flexoelectric origin of oblique-roll electrohydrodynamic instability in nematics

N V MADHUSUDANA, V A RAGHUNATHAN and
K R SUMATHY
Raman Research Institute, Bangalore 560080, India

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Abstract. We develop the theory of electrohydrodynamic instability in nematic liquid crystals by incorporating the flexoelectric terms. Using a one-dimensional linear analysis of the problem for an applied DC field, we demonstrate that for the usual materials the rolls have an oblique orientation as has been found experimentally. We also provide an experimental evidence for the strong flexoelectric influence on the director profile in the rolls.

Keywords. Nematic liquid crystals; flexoelectricity; oblique roll instability; electrohydrodynamics.

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An electrohydrodynamic (EHD) roll instability is exhibited by nematic liquid crystals with either negative or weakly positive dielectric anisotropy ($\Delta\varepsilon$) above a DC or low frequency AC threshold voltage (Blinov 1983; Chandrasekhar 1977). The first theoretical analysis of the problem was given by Helfrich (1969) for an applied DC field. It was later extended by the Orsay group (Dubois-Violette *et al* 1971; Smith *et al* 1975) to AC fields. The conductivity anisotropy ($\Delta\sigma$) of the medium gives rise to space charge densities under the action of the external field if there are bend fluctuations in a planar aligned sample. The electric force on the space charge density gives rise to EHD instabilities. The one-dimensional theories mentioned above considered only the possibility of rolls whose wavevector is along the initial undistorted orientation of the director $\mathbf{n}_0$. Recently, however, there have been a few experimental observations (Ribotta *et al* 1986; Hirata and Tako 1982) of *oblique* rolls, in which the wavevector makes an angle (α say) with $\mathbf{n}_0$, in both DC and very low frequency AC fields. Zimmermann and Kramer (1985) made a three-dimensional, linear analysis of the EHD problem. However, unrealistically, they assumed 'stress-free' boundary conditions for simplifying the analysis. They found that oblique rolls result for a certain range of material parameters, and also noted that an one-dimensional analysis would *not* lead to oblique rolls.

We must however point out that the deformation in the director field in the EHD structures gives rise to a flexoelectric polarization **P** of the medium which was not considered in the above model. The action of the external field on **P** could be expected to influence the roll structure. Some early Russian work (Ioffe 1975; Matyushichev and Kovnatskii 1975) incorporating the effect of **P** in the theory of EHD predicts the possibility of an oscillatory instability, the wavevector of the rolls remaining parallel to $\mathbf{n}_0$. In the present communication, we demonstrate that the flexoelectric terms lead to oblique rolls in an one-dimensional model itself.

In his original paper on flexoelectric properties of nematics, Meyer (1969) envisaged that only pear-shaped molecules with longitudinal dipoles or banana-shaped molecules with lateral dipoles could give rise to **P**. Prost and Marcerou (1977) have subsequently shown that the quadrupole density of the medium makes a very important contribution to **P** and hence flexoelectricity is a universal property of all nematics.

We consider a nematic taken in a sandwich cell, with $\mathbf{n}_0$ lying parallel to the conducting plates, along the x axis, say (Figure 1). With an applied DC field E_z, we assume that the EHD instability gives rise to oblique rolls whose wavevector $\mathbf{q}$ lies along ξ making an angle α with $\mathbf{n}_0$. In the deformed state, $\mathbf{n}$ makes polar angles θ and ϕ with the xyz system, so that the components of $\mathbf{n}$ in the $\xi\eta z$ system are $[\cos\theta\cos(\alpha-\phi), -\cos\theta\sin(\alpha-\phi), \sin\theta]$. It is well known that the one-dimensional model does not predict a nonzero value of q, but following Helfrich (1969) we can assume on physical grounds that the sample thickness d determines the width of the rolls. As in the earlier work of Smith *et al* (1975), the one-dimensional model not only brings out all the essential features of the problem, but also leads to a simple physical interpretation of the results. We assume that only the vertical component of the velocity (v_z), is nonzero, and that v_z, θ and ϕ are functions of ξ only. In the present communication we confine our attention to DC fields. We have then a stationary director profile which satisfies the torque balance equations

$$\Gamma^i_{\text{elastic}} + \Gamma^i_{\text{flexo}} + \Gamma^i_{\text{dielectric}} = \Gamma^i_{\text{hydrodyn}}, \qquad i = y, z, \tag{1}$$

where $\Gamma = \mathbf{n} \times \mathbf{h}$, $\mathbf{h}$ being the relevant molecular field (de Gennes 1975). The anisotropy of conductivity $\Delta\sigma$ gives rise to a transverse field which by symmetry has only the ξ component E_ξ. The electric displacement is given by

$$\mathbf{D} = \varepsilon_\perp \mathbf{E} + \Delta\varepsilon(\mathbf{n}\cdot\mathbf{E})\mathbf{n} + 4\pi(e_1\mathbf{n}\,\mathrm{div}\,\mathbf{n} + e_3\,\mathrm{curl}\,\mathbf{n}\times\mathbf{n}), \tag{2}$$

$$\Delta\varepsilon = \varepsilon_\parallel - \varepsilon_\perp,$$

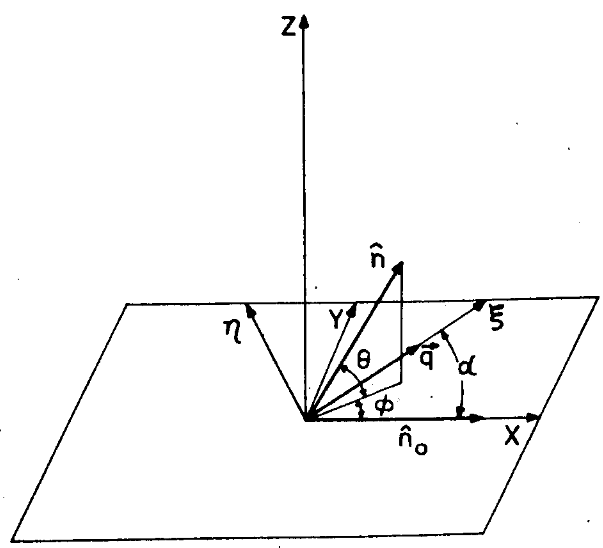

Figure 1. Illustration of the coordinate system and the definitions of angles used in the text.

where $\varepsilon_\parallel$, $\varepsilon_\perp$ are the principal dielectric constants, and e_1, e_3 the flexoelectric coefficients. The action of E_z on the total space charge density (div $\mathbf{D}/4\pi$) is used to calculate the shear stress which in turn is used to eliminate the shear rate $\mathrm{d}v_z/\mathrm{d}\xi$ from $\mathbf{h}_{\mathrm{hydrodyn}}$.

Retaining only the linear terms, equations (1) yield

$$(k_2\sin^2\alpha+k_3\cos^2\alpha)\frac{\mathrm{d}^2\theta}{\mathrm{d}\xi^2}+HE_z^2\theta+\{(e_1-e_3)$$

$$-(e_1+e_3)(F+G)\cos^2\alpha\}\sin\alpha\,E_z\frac{\mathrm{d}\phi}{\mathrm{d}\xi}=0, \tag{3}$$

$$\{(e_1+e_3)F\cos^2\alpha-(e_1-e_3)\}\sin\alpha\,E_z\frac{\mathrm{d}\theta}{\mathrm{d}\xi}$$

$$+(k_1\sin^2\alpha+k_3\cos^2\alpha)\frac{\mathrm{d}^2\phi}{\mathrm{d}\xi^2}=0, \tag{4}$$

along y and z respectively, where

$$F=\Delta\sigma/(\sigma_\perp+\Delta\sigma\cos^2\alpha),\quad G=-2\alpha_2/[\alpha_4+(\alpha_5-\alpha_2)\cos^2\alpha],$$

$$H=[\Delta\varepsilon(F\sigma_\perp/\Delta\sigma)^2-\{\Delta\varepsilon-F(\varepsilon_\perp+\Delta\varepsilon\cos^2\alpha)\}\,G\cos^2\alpha]/4\pi, \tag{5}$$

α_2, α_4, α_5 are the viscosity coefficients and k_1, k_2, k_3 the curvature elastic constants (de Gennes 1975). The coupled equations (3) and (4) admit solutions of the form

$$\theta=\theta_0\sin q\xi,\qquad \phi=\phi_0\cos q\xi, \tag{6}$$

in which the relative signs of θ_0 and ϕ_0 depend on those of E_z and α. The threshold for deformation is given by

$$E_{\mathrm{th}}=q(k_2\sin^2\alpha+k_3\cos^2\alpha)^{1/2}[H+\sin^2\alpha\{(e_1-e_3)^2$$
$$+(e_1+e_3)^2F(F+G)\cos^4\alpha-(e_1^2-e_3^2)$$
$$(2F+G)\cos^2\alpha\}/(k_1\sin^2\alpha+k_3\cos^2\alpha)]^{-1/2}. \tag{7}$$

Using the Helfrich condition $q=\pi/d$ we get a voltage threshold. The angle α is selected to minimise V_{th}. If $\alpha=0$, the flexoelectric contribution vanishes completely. Note that our expression for V_{th} (for $\alpha=0$) is slightly different from that given by Helfrich (1969) since, in his model, the dependence of E_ξ on θ is ignored in calculating $\mathbf{h}_{\mathrm{dielec}}$. If $\alpha=\pi/2$, all the hydrodynamic contributions are absent, and the flexoelectric effect can cause a periodic static distortion of $\mathbf{n}$ if $\Delta\varepsilon$ is sufficiently small. Such flexoelectric distortions have been studied earlier both experimentally (Barnik *et al* 1978) and theoretically (Bobylev and Pikin 1977).

e_1 and e_3 have been measured only for a couple of compounds. In MBBA (4-methoxybenzylidene-4′-butylaniline) $e_1-e_3\simeq1{\cdot}2\times10^{-4}$ e.s.u. (Dozov *et al* 1982) and $e_1+e_3\simeq-7\times10^{-4}$ e.s.u (Madhusudana and Durand 1985). Using $\varepsilon_\parallel=4{\cdot}5$, $\varepsilon_\perp=5$, $k_1=6{\cdot}1\times10^{-7}$ dyne, $k_2=4\times10^{-7}$ dyne, $k_3=7{\cdot}3\times10^{-7}$ dyne, $\alpha_2=-77{\cdot}5$ cP,

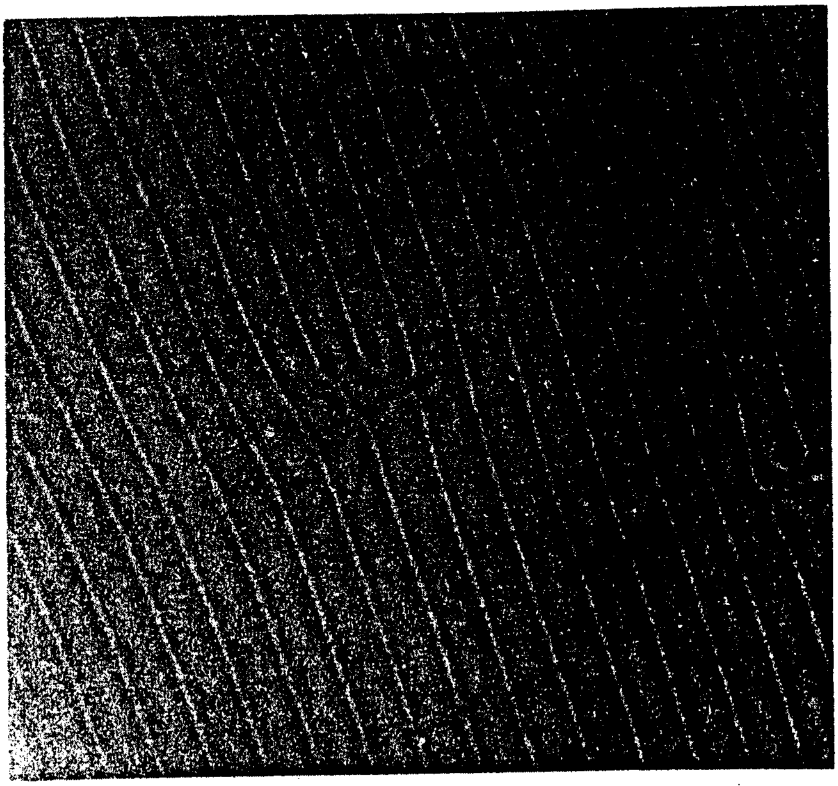

Figure 2. Photograph of the EHD pattern in a room temperature nematic. $\mathbf{n}_0$ is horizontal. Note that the edge dislocation corresponds to the addition of one optical domain, which has two hydrodynamic cells of opposite vorticity as explained in the text. Sample thickness $d \simeq 15\ \mu$m. Magnification $\times 250$.

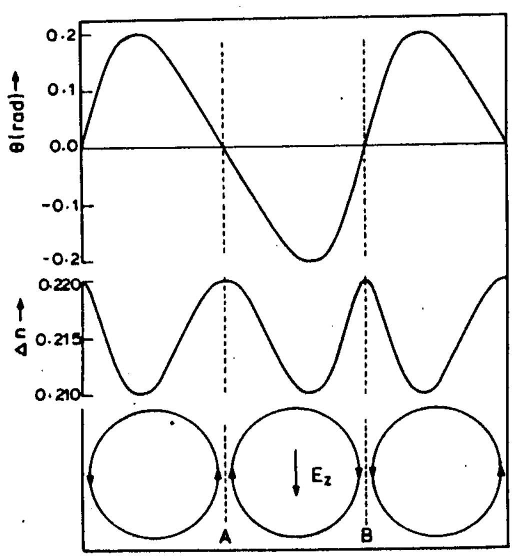

Figure 3. The non-sinusoidal θ profile obtained as a solution of equation (8) (top). The resulting variation of the effective Δn with ξ ($n_e = 1{\cdot}769$, $n_0 = 1{\cdot}549$) (middle). The disposition of the hydrodynamic rolls (bottom) agree with the observed dust particle motion, regions B corresponding to the bright lines of figure 2. A reversal of the field shifts the bright line to A.

$\alpha_4 = 83{\cdot}2$ cP, $\alpha_5 = 46{\cdot}3$ cP and $\Delta\sigma/\sigma_\perp = 0{\cdot}3$, which are typical values for MBBA at room temperature, we get $\alpha \simeq 47°$ and $V_{th} \simeq 1{\cdot}9$ V. Experimentally, Hirata and Tako (1982) find $\alpha \simeq 30°$ for MBBA. Ribotta *et al* (1986) also find oblique rolls in MBBA. (However the model of Zimmermann and Kramer (1985) does not lead to oblique rolls for the standard values of the material parameters of MBBA.)

We have studied the EHD patterns in a three-component mixture (containing CE-1700, CM-5115 and PCH 302 of Roche Chemicals) which is a stable room temperature nematic with $\Delta\varepsilon \simeq -0{\cdot}1$, and the values of $(e_1 + e_3)$ and $(e_1 - e_3)$ comparable to those of MBBA. We have verified by various tests that under a DC field, there is no influence of charge injection on the EHD rolls. Figure 2 shows a photograph of the EHD domains at a voltage slightly above V_{th} ($\simeq 3{\cdot}6$ V). We see oblique rolls with $\alpha \simeq 20°$, and more interestingly, the *optical* domain width, i.e., separation between two bright lines $\simeq 2d$. Indeed dust particle motion indicates that two rolls of opposite vorticity are contained in one optical domain, which corresponds to a full spatial period. Obviously, the director profile is not sinusoidal but the curvature in the region of the bright lines is much stronger than in the region midway between two bright lines (see figure 3). Further, when the voltage is reversed, the bright lines are shifted by half an optical domain width, showing that the distortion of the profile is polarity-dependent. Nonlinear terms are needed to account for the distortion of the director profile, and when $\alpha = 0$, up to second order terms equation (1) gives along y

$$K_3\frac{\mathrm{d}^2\theta}{\mathrm{d}\xi^2} + H_0E_z^2\theta + G_0(e_1 + e_3)E_z\theta\frac{\mathrm{d}\theta}{\mathrm{d}\xi} = 0, \tag{8}$$

where the subscript 0 signifies that $\alpha = 0$. The lone quadratic term in (8) arises from the action of E_z on ρ_e, the flexoelectric contribution to the space charge density. Equation (8) can be solved graphically by the phase plane technique. The resulting nonsinusoidal θ profile and the effective birefringence Δn are shown in figure 3.

From (3) and (4), it is clear that a reversal of the sign of E_z can either change the sign of θ or ϕ but not both if the sign of α is fixed. Experimentally, in the material used by us, it is seen that θ does not reverse its sign with E_z and hence we conclude that ϕ changes sign with E_z. (Note that in the absence of flexoelectric terms, signs of θ and ϕ do not depend on that of E_z). Consequently, in an AC field, the flexoelectric contribution to the nonzero value of α should be ineffective beyond a frequency corresponding to a typical director relaxation rate under the given E_z. On the other hand, in the *dielectric* regime ($q \gg \pi/d$), θ oscillates with E_z (Dubois-Violette *et al* 1971; Smith *et al* 1975), and the flexoelectric contribution can again dominate. The large values of α seen in the 'chevron patterns' (Blinov 1983; de Gennes 1975) of the dielectric regime are then a natural consequence of our model. A detailed analysis of the AC regimes is underway.

When flexoelectric terms are included, the solutions (1) of Zimmermann and Kramer (1985) are not applicable in a simple three-dimensional analysis. Thus, a direct comparison of the present model with their theory is not possible. A full numerical analysis of the 3-dimensional problem is also underway.

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