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# Flexoelectric origin of oblique rolls with helical flow in electroconvective nematics under DC excitation

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**Abstract.** We present a 3-dimensional, linear analysis of electrohydrodynamic (EHD) roll instability in a nematic liquid crystal under DC excitation. It is shown that the flexoelectric effect leads to a new symmetry of the flow pattern, viz. a *helical* flow in *oblique* rolls. Our experimental observations agree with the theoretical predictions.

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

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EHD instability in nematics is a well-known phenomenon (de Gennes 1975; Chandrasekhar 1977; Blinov 1983). Oblique rolls, with the wavevector $\mathbf{q}$ making an angle with the undistorted director $\mathbf{n}_0$ are known to occur for DC and low frequency AC fields in the conduction regime (Ribotta *et al* 1986; Hilsum and Saunders 1980; Hirata and Tako 1981). Zimmermann and Kramer (1985) extended the earlier 2-dimensional calculations of Penz and Ford (1972) for *normal* rolls (i.e., with $\mathbf{q}$ along $\mathbf{n}_0$) to 3-dimensions using stress-free boundary conditions and showed that *oblique* rolls have a lower threshold than the normal rolls for certain range of material parameters, viz., the dielectric constants $\varepsilon_\parallel$ and $\varepsilon_\perp$ ($\parallel$ and $\perp$ are in relation to $\mathbf{n}$), the conductivities $\sigma_\parallel$ and $\sigma_\perp$, the viscosity coefficients $\alpha_i$ ($i = 1$ to 5) and the curvature elastic constants $k_{ii}$ ($i = 1$ to 3). They argued that the oblique rolls give rise to a more favourable charge density than the normal ones and also noted that a one-dimensional model would *not* lead to oblique rolls.

The deformation of the director field in the medium gives rise to a flexoelectric polarization $\mathbf{P}$ (Meyer 1969), which was not taken into account in the models mentioned above. We note that quadrupole densities make a very important contribution to $\mathbf{P}$ (Prost and Marcerou 1977) and hence flexoelectricity is a *universal* property of all nematics. If the deformation is characterized by only one polar angle, say $\theta$, flexoelectricity can influence the director field only through surface terms (Madhusudana and Durand 1985; Patel and Meyer 1987), i.e., only if the anchoring is weak (de Gennes 1975). If the deformation is more general, involving both $\theta$ and $\phi$, flexoelectricity contributes to bulk torque densities (Dozov *et al* 1982; Bobylev and Pikin 1977) and hence can be expected to favour the formation of *oblique* rolls. Indeed this result has been confirmed by us (Madhusudana *et al* 1987) on the basis of a 1-dimensional model itself. A full 3-dimensional analysis which we present in this communication shows clearly that flexoelectric terms not only make the *predominant* contribution to a tilting of the EHD rolls, but are entirely responsible for a *helical* motion of the fluid particles. We have also made experimental observations which confirm the theoretical predictions.

Let $\mathbf{n}_0$ be along the $X$-axis. With an applied DC field $E_z$, we assume that $\mathbf{q}$ of the EHD rolls lies along $\xi$ which makes an angle $\alpha$ with $\mathbf{n}_0$. In the deformed state, $\mathbf{n}$ makes polar angles $\theta$ and $\phi$ in the $XYZ$ system, so that its components in the $\zeta\eta Z$ system are [$\cos\theta \cos(\alpha-\phi)$, $-\cos\theta\sin(\alpha-\phi)$, $\sin\theta$]. We summarize below the method of calculation which is similar to that of Penz and Ford (1972). The torque balance equations are

$$\Gamma^i_{\text{elastic}} + \Gamma^i_{\text{flexo}} + \Gamma^i_{\text{dielec}} = \Gamma^i_{\text{hydrodyn}} \quad (i = y, z) \tag{1}$$

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 $\xi$ component $E_\xi$. The electric displacement is

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

where $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_\perp$, and $e_1$ and $e_3$ are the flexoelectric coefficients. The action of the total electric field on the space charge density $Q = \mathrm{div}\,\mathbf{D}/4\pi$ gives rise to an electric force on the medium and the equations of motion become (Penz and Ford 1972)

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla\right)\mathbf{v} = \mathrm{div}(\bar{\sigma} + \bar{\sigma}') + Q\mathbf{E}, \tag{3}$$

where $\mathbf{v}$ is the velocity of the fluid, $\bar{\sigma}$ and $\bar{\sigma}'$ are the elastic and viscous stress tensors respectively. We linearize the equations and ignore the inertial term. The independent variables of the problem are $\theta$, $\phi$, $E_\xi$, $v_\xi$, $v_\eta$ and $v_z$. Assuming $\theta = \theta_0 \exp(i\mathbf{q}\cdot\mathbf{r})$, etc., the solution of the set of equations (1) and (3) requires that an appropriate determinant should vanish, and this condition yields an equation of 12th degree in $s = q_z/q_\xi$. When $e_{1,3} = 0$, i.e., the flexoelectric contribution is absent, there are no odd powers in the polynomial and the roots occur in complex conjugate pairs. When $e_{1,3} \neq 0$, odd powers are present and for every root $s_n = a_n + ib_n$, we also get a root (Chandrasekhar 1954) $s_{n'} = -a_n + ib_n$. We now impose the boundary conditions $\theta = \phi = E_\xi = v_\xi = v_\eta = v_z = 0$ at $Z = \pm D/2$, where $D$ is the thickness of the sample. This results in a 'boundary value determinant (*bvd*)' which should also vanish (Penz and Ford 1972). For given values of $\alpha$, $E_z = (V/D)$ and $q_\xi$, we make calculations on a computer to obtain the roots $s_n$ ($n = 1$ to 12) and the value of the *bvd*, and adjust $q_\xi$ till the *bvd* becomes zero. We repeat the calculations for various values of $\alpha$. The lowest value of the voltage for which such solutions exist is the threshold voltage (Penz and Ford 1972). If $\alpha = \pi/2$, all the hydrodynamic contributions are absent, and the static distortion in this case has been studied earlier (Bobylev and Pikin 1977).

All the required material parameters are available for methoxybenzylidene butylaniline (MBBA) (de Gennes 1975; Penz and Ford 1972; Madhusudana and Durand 1985): $e_1 - e_3 = 1{\cdot}2\times10^{-4}$ esu, $e_1 + e_3 = -7\times10^{-4}$ esu, $\varepsilon_\parallel = 4{\cdot}7$, $\varepsilon_\perp = 5{\cdot}2$, $k_{11} = 6{\cdot}1\times10^{-7}$ dyne, $k_{22} = 2\times10^{-7}$ dyne, $k_{33} = 7{\cdot}3\times10^{-7}$ dyne, $\alpha_1 = 6{\cdot}5$ cP, $\alpha_2 = -77{\cdot}5$ cP, $\alpha_3 = -1{\cdot}2$ cP, $\alpha_4 = 83{\cdot}2$ cP, $\alpha_5 = 46{\cdot}3$ cP and $\Delta\sigma/\sigma_\perp = 0{\cdot}5$. When the flexoelectric terms are included in the calculations, we obtain oblique rolls for the standard values of the material parameters of MBBA. We find that while the flexoelectric torques are sufficient to give rise to oblique rolls, the additional space charge density resulting from the flexoelectric polarization further lowers the threshold voltage. If the flexoelectric terms are not taken into account (i.e., $e_{1,3} = 0$), as was already found by Zimmermann and Kramer (1985), oblique rolls are obtained *only* if the values of some of the material parameters are suitably adjusted. The value of $k_{22}$ listed above has been chosen to be about half of the standard experimental value for this purpose. Figure 1a clearly shows that the flexoelectric terms make the *predominant* contribution to the formation of oblique rolls even with the lowered value of $k_{22}$. When $\alpha = 0$, a non-vanishing charge density of flexoelectric origin causes the small difference in $V_{th}$ between the two cases with and without the flexoelectric terms. The axial velocity $v_\eta$ results from the oblique flow of the fluid in relation to **n**, with a vertical velocity gradient (Pieranski and Guyon 1974; de Gennes 1975). When $e_{1,3} = 0$, the polynomial in $s$ has only even powers and the profiles of the different variables have obvious symmetries about the mid-plane (figures 1b-f). On the other hand, when $e_{1,3} \neq 0$, the odd-powered terms in the polynomial give rise to asymmetries in the profiles (figures 1b-f). The strong coupling to flexoelectric terms leads to a conspicuous asymmetry in the $\phi$-profile (figure 1d) and in turn to that in the $v_\eta$-profile (figure 1f). The latter asymmetry gives rise to an *open helical* trajectory of the fluid particles. Figure 1h shows the trajectory of a particle close to the periphery of a roll. On the other hand, when $e_{1,3} = 0$, the symmetry of the $v_\eta$ profile leads to a closed trajectory of the fluid particles, even though the motion is no longer confined to the $\xi Z$ plane. Thus flexoelectricity is *essential* to produce a *helical* flow of the fluid particles in oblique rolls, though the oblique rolls themselves can be obtained even in its absence by a suitable adjustment of the material parameters. When the flow has a helical character, material conservation is ensured by opposite $\bar{v}_\eta$ (an average value of $v_\eta$) in adjacent cells (figure 1h) of opposite vorticity. For a given sense of the vorticity, $\bar{v}_\eta$ changes sign with either that of $\alpha$ or $E_z$, reflecting the flexoelectric origin of the helical flow.

Earlier studies on EHD instabilities under DC excitation were made on MBBA (Orsay Liquid Crystal Group 1971; Hirata and Tako 1981), which is chemically unstable. Charge injection complicates the pattern in this case, giving rise to a twin-domain pattern (Orsay Liquid Crystal Group 1971) and a 2-dimensional grid pattern before the oblique rolls are seen (Hirata and Tako 1981). We have studied the DC instabilities in a mixture with 3 chemically stable components, viz., CE-1700, CM-5115 and PCH-302 of Roche Chemicals. It has $\Delta\varepsilon \simeq -0{\cdot}1$ and $e_1$ and $e_3$ comparable to those of MBBA. The Carr-Helfrich mechanism can operate to produce EHD rolls only if the director relaxation time ($\propto D^2$) is longer than the charge relaxation time, and below a certain cell thickness the instability is quenched out (Smith *et al* 1975). Indeed our sample exhibits this character, demonstrating that charge injection is not significant in this case (Madhusudana *et al* 1987). If $D$ is sufficiently large we observe *oblique* rolls directly at $V_{th}$ as expected from the theory. The trajectory of dust particles could be seen only when they were sufficiently far away from the axis of the rolls, i.e. close to the periphery of the rolls. Detailed observations on the motion of these particles yield the following results. (a) The trajectory of the particle is helical. (b) $\bar{v}_\eta$ is opposite in adjacent cells of opposite vorticity and the direction of $\bar{v}_\eta$ agrees with that found in the calculations for given signs of $\alpha$, $E_z$ and the vorticity. (c) $\bar{v}_\eta$ changes sign with that of any one of the 3 parameters. (d) Often domains with $+\alpha$ and $-\alpha$ meet at a boundary forming elbow like structures. As predicted by the theory, for one sense of vorticity the fluid motion ($\bar{v}_\eta$) is towards the boundary in rolls with both $+\alpha$ and $-\alpha$ while it is away from the boundary for rolls with the opposite vorticity. Dust particle motion at the

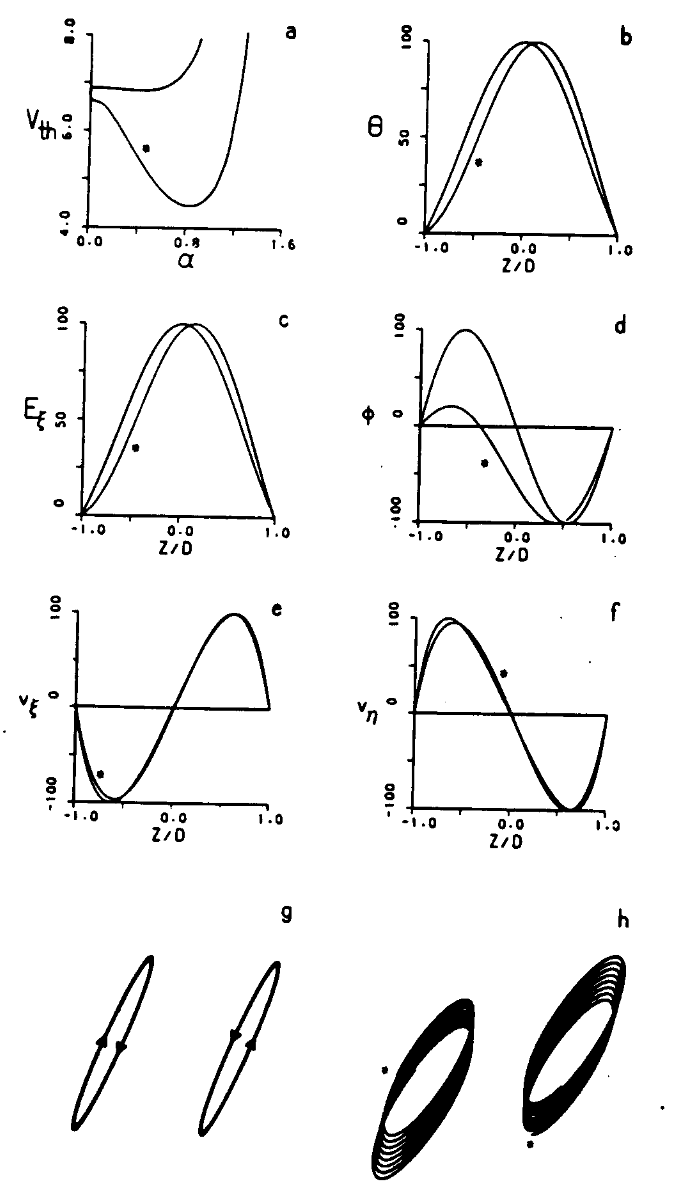


**Figure 1.** Results of the computer calculations on the EHD roll instability for MBBA (see text). $E_z$ is positive and the profiles in **b-f** correspond to the vortex cells shown on the left sides of **g** and **h** and at $V = (V_{th} + 1{\cdot}5)$ volts. In each case, the $Y$ axis is in arbitrary units, and the curve for $e_{1,3} \neq 0$ is indicated by an asterisk. (**a**) $V_{th}$ (in volts) vs $\alpha$ (in radians), (**b**) $\theta$ vs $Z/D$ for $\xi = 0$ (i.e., along the vertical line in the *centre* of the vortex cell), (**c**) $E_\xi$ vs $Z/D$ for $\xi = 0$, (**d**) $\phi$ vs $Z/D$ for $\xi = \pi/q_\xi$ (i.e., at the edge of the vortex cell), (**e**) $v_\xi$ vs $Z/D$ for $\xi = 0$, (**f**) $v_\eta$ vs $Z/D$ for $\xi = 0$, (**g**) the closed particle trajectories in two neighbouring vortices for the case with $e_{1,3} = 0$, (**h**) the corresponding helical particle trajectories when $e_{1,3} \neq 0$. In the latter case, the asterisk indicates the $t = 0$ position of the particle. In (**g**) and (**h**) the ellipticity of the trajectories arises from a steep angle of viewing. Further, the scales along $\xi$, $\eta$ and $Z$ have been chosen to be rather different for the sake of clarity.

elbows clearly shows that the material leaks from the former type of rolls to the latter, thus ensuring material conservation. The process appears to be somewhat complicated, as the boundaries are never stationary. The structure breaks up and rejoins periodically with $T \sim 1$ minute, while dislocations can be seen to move across the sample. Material transfer again takes place between rolls of opposite vorticity at the periphery of the sample. All these observations are consistent with the theory.

Thus, under the DC excitation, flexoelectricity not only makes the most important contribution to the occurrence of oblique rolls, but leads to a *qualitative* change in the symmetry of the flow pattern, viz., to a helical flow. Though a helical motion has been reported (Ribotta *et al* 1986; Hilsum and Saunders 1980) under AC excitation, its origin is not clear since our theory predicts a change in the sign of $\bar{v}_\eta$ with that of $E_z$. We are now making detailed 3-dimensional calculations for the AC case. Our one-dimensional calculations for AC excitation, to be published soon, have already shown that $\alpha$ at $V_{th}$ goes to zero at a critical frequency which is $\sim 30$ Hz in the usual samples. The dependence of $V_{th}$ on frequency is in broad agreement with the observations of Ribotta *et al* (1986).

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## References

Blinov L M 1983 *Electro-optical and magneto-optical properties of liquid crystals* (New York: Wiley)
Bobylev Yu P and Pikin S A 1977 *Sov. Phys. JETP* **45** 195
Chandrasekhar S 1977 *Liquid crystals* (Cambridge: University Press)
Chandrasekhar S 1954 *Philos. Mag.* **45** 1177
de Gennes P G 1975 *The physics of liquid crystals* (Oxford: Clarendon)
Dozov I, Martinot-Lagarde Ph and Durand G 1982 *J. Phys. (Paris)* **43** L365
Hilsum C and Saunders F C 1980 *Mol. Cryst. Liq. Cryst.* **64** 25
Hirata S and Tako T 1981 *Jpn. J. Appl. Phys.* **20** L459
Madhusudana N V and Durand G 1985 *J. Phys. (Paris)* **46** L195
Madhusudana N V, Raghunathan V A and Sumathy K R 1987 *Pramana – J. Phys.* **28** L311
Meyer R B 1969 *Phys. Rev. Lett.* **22** 918
Orsay Liquid Crystal Group 1971 *Mol. Cryst. Liq. Cryst.* **12** 251
Patel J S and Meyer R B 1987 *Phys. Rev. Lett.* **58** 1538
Penz P A and Ford G W 1972 *Phys. Rev.* **A6** 414
Pieranski P and Guyon E 1974 *Phys. Lett.* **A49** 237
Prost J and Marcerou J P 1977 *J. Phys. (Paris)* **38** 315
Ribotta R, Joets A and Lei L 1986 *Phys. Rev. Lett.* **56** 1595
Smith I W, Galerne Y, Lagerwall S T, Dubois-Violette E and Durand G 1975 *J. Phys. (Paris)* **36** C1-237
Zimmermann W and Krämer L 1985 *Phys. Rev. Lett.* **55** 403