Propagating electroconvection in nematic liquid crystals under DC excitation

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On the basis of a simple one-dimensional linear model we show for the first time that a propagating electrohydrodynamic instability (PI) of a nematic liquid crystal occurs at the threshold of DC excitation if the symmetry of a homogeneously aligned cell is changed by introducing a small (~ 0.01 rad) pre-tilt of the director. Flexoelectric torques, which are $\pi/2$ out of phase with those produced by other couplings, provide the driving mechanism for the PI mode, whose direction depends on that of the applied field. We also report an experimental observation of this mode.

ELECTROCONVECTION in nematic liquid crystals with dielectric anisotropy $\Delta\varepsilon \lesssim 0$ and subjected to a DC or low-frequency AC electric field is well known[1-3]. The director **n** of the anisotropic fluid ensures that the convective instability occurs in the form of rolls rather than more complicated structures at the threshold, and the aspect ratios can be large (~ 1000). At low frequencies of excitation *oblique* rolls have been found whose wave vector **q** makes a non-zero angle with $\mathbf{n}_0$, where the subscript '0' indicates the rest state[4]. This is now well understood as arising from the flexoelectric properties of the nematic[5-7]. Very recently, a travelling-wave (TW) mode has been discovered near the threshold in the normal rolls which occur at higher frequencies in the conduction regime, either only near the 'cut-off' frequency (f_c) of this regime when the charge relaxation time becomes comparable to the director relaxation time[8], or, in some compounds, throughout the conduction regime[9]. Joets and Ribotta[8], who have found a localized TW instability near f_c, have speculated that dispersive effects in phase dynamics as well as nonlinearities may become important even near the onset in such cases.

The possibility of oscillatory or TW electroconvection in nematics under DC excitation has been discussed by a few authors earlier. Ioffe, and Matyushichev and Kovnatskii[10] included flexoelectric terms in the electrohydrodynamic (EHD) equations. In the former work the x-component of the bulk force due to the flexoelectric effect was neglected and in the latter the boundary conditions were not properly taken into account. These authors therefore incorrectly got oscillatory solutions to the EHD equations[11]. Laidlaw[12] argued that, for materials with $\Delta\varepsilon > 0$ and negative conductivity anisotropy ($\Delta\sigma = \sigma_\parallel - \sigma_\perp$), an oscillatory convective instability may be found at DC voltages much above the Freedericksz threshold for static deformation of the director field, but there is no experimental confirmation of this result. Penz[13] suggested that, for materials with $\Delta\varepsilon > 0$ and $\Delta\sigma > 0$, a TW convective instability could set in if the sample thickness is rather small, but again at DC voltages above the Freedericksz threshold. He also noted that, as the Freedericksz threshold pre-empts the TW solution, it may not be possible to observe the latter using the usual experimental geometries. Inclusion of flexoelectric terms in this problem suppresses the TW solution[11]. Thus, up to now, there appears to be no theoretical model that can give rise to TW solutions in experimentally realizable situations.

In this article we demonstrate that it is possible to obtain a propagating solution at the threshold of DC excitation in an appropriate geometry. We show this by a linear one-dimensional analysis for materials with $\Delta\varepsilon = 0$ oriented in a cell with a small pre-tilt angle β of ~ 0.01 rad. The propagation is caused by flexoelectric torques that are $\pi/2$ out of phase with respect to the hydrodynamic and elastic torques that arise in the convecting liquid. The propagation direction depends on the sign of the field. We also report experimental observations confirming this prediction. We must however point out that our model cannot explain the TW instabilities studied by earlier experimenters[8,9].

One-dimensional model for DC excitation

For the sake of a clear understanding of the underlying mechanism, we make several simplifications in the problem: (a) The dielectric anisotropy is set equal to zero. (b) The angle β between $\mathbf{n}_0$ and the X-axis (see Figures 1, *b* and 2) is small enough to retain only linear terms in β. (c) We solve a 'one-dimensional' problem in which the boundary conditions are ignored, and, from physical considerations[14], q_x is set equal to π/d where d is the sample thickness. The physical processes become transparent in a 1-D model, and a full 3-D model, which needs numerical solutions, is expected to make only quantitative differences[3,14]. (d) Even though the flexoelectric effect tends to produce oblique rolls under DC excitation[5], for the sake of simplicity we assume 'normal' roll solutions here. (We have confirmed that similar propagating solutions are found in the case of oblique rolls also[11].) Under these approximations, the linearized EHD equations are[3]:

(a) the Poisson equation

$$4\pi Q - \varepsilon(\partial E_x/\partial x) + 4\pi\beta(e_1 + e_3)(\partial^2\theta/\partial x^2) = 0, \qquad (1)$$

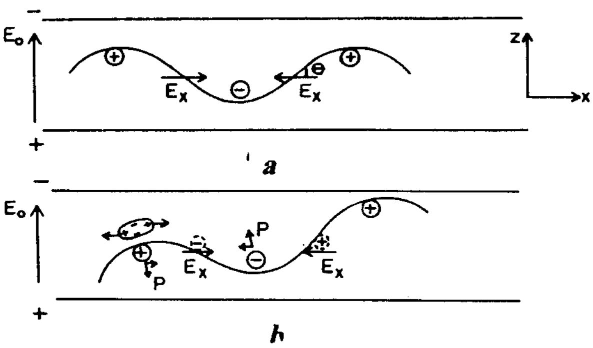

Figure 1. *a*, $\beta = 0$; The space charges (shown in full circles) arising from the conductivity anisotropy cause EHD instability. *b*, $\beta \neq 0$; the quadrupoles have an out-of-phase torque due to the horizontal field gradients, and additional charges (in dotted circles) are collected owing to the horizontal gradient in the flexoelectric polarization **P**.

(b) the charge continuity equation

$$(\partial\theta/\partial t) + \sigma_1(\partial E_x/\partial x) + \Delta\sigma E_0\,(\partial\theta/\partial x) = 0, \qquad (2)$$

(c) the torque balance equation

$$\gamma_1(\partial\theta/\partial t) + \alpha_2(\partial \upsilon_z/\partial x) - k_{33}(\partial^2\theta/\partial x^2) - \beta(e_1 + e_3)(\partial E_x/\partial x) = 0, \qquad (3)$$

and (d) the equation of motion along the Z-axis

$$\alpha_2(\partial^2\theta/\partial x\,\partial t) + \eta(\partial^2\upsilon_z/\partial x^2) + E_0 Q = 0, \qquad (4)$$

where Q is the charge density, k_{33} the bend elastic constant, θ the angle of deviation of the director from $\mathbf{n}_0$, $(e_1 + e_3)$ the sum of the flexoelectric coefficients, α_2, α_4, α_5, $\gamma_1 = \alpha_3 - \alpha_2$, and $\eta = (\alpha_4 + \alpha_5 - \alpha_2)$ the viscosity coefficients, t the time, and X and Z are defined in Figure 1. The external DC field E_0 is applied along the Z-axis and E_x arises from the space charge densities (see Figure 1). It is clear that under the approximations made in the present model, only the flexoelectric terms couple to the tilt angle. Using solutions of the form

$$\theta = \theta_1 \exp i\,(q_x x - \omega t), \text{ etc.}, \qquad (5)$$

the conditions for the onset of the instability are given by

$$4\pi k_{33} q_x^2 + E_{th}^2\,\varepsilon\alpha_2\Delta\sigma/(\eta\sigma_\parallel) - \varepsilon\omega^2\,(\gamma_1 - \alpha_2^2/\eta)/\sigma_\parallel = 0, \qquad (6)$$

and

$$\omega = \beta(e_1 + e_3) q_x E_{th}(\Delta\sigma/\sigma_\parallel + \alpha_2/\eta)\ \{(\gamma_1 - \alpha_2^2/\eta)\,(1 + \tau/T)\}, \qquad (7)$$

where the charge relaxation rate

$$\tau^{-1} = 4\pi\sigma_\parallel/\varepsilon \qquad (8)$$

and the director relaxation rate

$$T^{-1} = k_{33} q_x^2/(\gamma_1 - \alpha_2^2/\eta). \qquad (9)$$

If ω is zero, eq. (6) is just the threshold condition derived by Helfrich for the onset of stationary EHD instability[14]. From eq. (7), the EHD pattern always propagates for $(e_1 + e_3) \neq 0$. Further, since the third term of eq. (6) has a negative sign, the propagating solution with $\omega \neq 0$ in fact has a lower threshold than the stationary instability which is obtained for $\beta = 0$.

We can estimate E_{th} from eq. (6) by setting ω, which is expected to be small, to zero and then use eq. (7) to calculate ω. The factor $(e_1 + e_3)$ arises from the aligned quadrupoles of the molecules[15], and has a non-zero value in general. The velocity of propagation ($= \omega/q_x$) is $\propto q_x$. For a given sign of β and $(e_1 + e_3)$, the sign of ω depends on that of E_0, i.e. the propagation reverses direction when the field is reversed.

In the context of the 1-D model, if $\Delta\varepsilon < 0$ it is clear that the dielectric torque $\beta\Delta\varepsilon E_0^2/4\pi$ would rotate the director to the $\beta = 0$ configuration for any non-zero value of E_z. Actually, however, in a real sample with rigid boundary conditions, β will become Z-dependent near the threshold for convective instability. One could assume that at the threshold the dielectric torque is just balanced by the elastic torque to produce an 'average' value β in the sample. Using this physical approximation the first two terms in eq. (6) would define the Helfrich threshold corresponding to the $\Delta\varepsilon < 0$ case. Similarly, the director relaxation time T (eq. 9) would have a dielectric contribution. In other words, a DC field would produce a propagating EHD instability even for this case. Detailed two-dimensional calculations taking into account the boundary conditions are now under way, and the preliminary results confirm the fact that a non-zero pre-tilt of the director at the aligning surfaces produces a propagating EHD pattern, with the direction of propagation reversing with the sign of the applied electric field.

The physical mechanism for the propagation of the instability can be understood by referring to Figure 1. The transverse field gradient is large in the region in which the charge density is high, When $\beta = 0$ (Figure 1, *a*), this cannot produce any torque on the quadrupoles of the medium. On the other hand, when $\beta \neq 0$ (Figure 1, *b*), the field gradient does produce a torque on the director which is clearly $\pi/2$ out of phase with the torques produced by other physical mechanisms. Also, when $\beta \neq 0$ the flexoelectric polarization has a divergence which gives rise to charge densities that are out of phase with those collected by the coupling of the conductivity anisotropy with the curvature in the medium (Figure 1, *b*). The force produced by E_0 on this additional charge gives rise to hydrodynamic torques that are again out of phase with the main contributions responsible for the EHD instability. As a consequence of these out-of-phase torques, we get a slow propagation of the instability in the medium. Since the coupling responsible for the propagating solution is flexoelectric in origin, the velocity depends on the sign of the applied field.

Experimental studies

Igner and Freed[16] studied EHD instabilities in *N*-(4-methoxybenzylidene)-4-butylaniline (MBBA) under DC excitation using glass plates with SiO coated at grazing incidence. In this case the tilt angle at the surface is ~20°, and they got complicated two-dimensional EHD patterns. On the other hand it is known that with polyimide-coated and unidirectionally rubbed plates the nematic orients with a tilt angle of ~2°. We found in our earlier studies[5] that a mixture consisting of three chemically stable components, viz. CE-1700, CM-5115 and PCH-302 of Roche Chemicals, forms a room-temperature nematic which is well suited for DC studies on EHD instabilities. Charge injection does not play an important role in this mixture[5]. We have chosen this material doped with ~2% of pentylcyanobiphenyl (5CB) to get $\Delta\varepsilon \simeq 0$. The conductivity of the sample was quite low, $\simeq 10^{-11}\ \Omega^{-1}\ \mathrm{cm}^{-1}$, giving a cut-off frequency of ~15 Hz for the conduction regime. The two unidirectionally rubbed plates of the cell were aligned in a mutually 'antiparallel' orientation so that $\mathbf{n}_0$ is aligned with a constant angle β (Figure 2). We found that when the coating was made

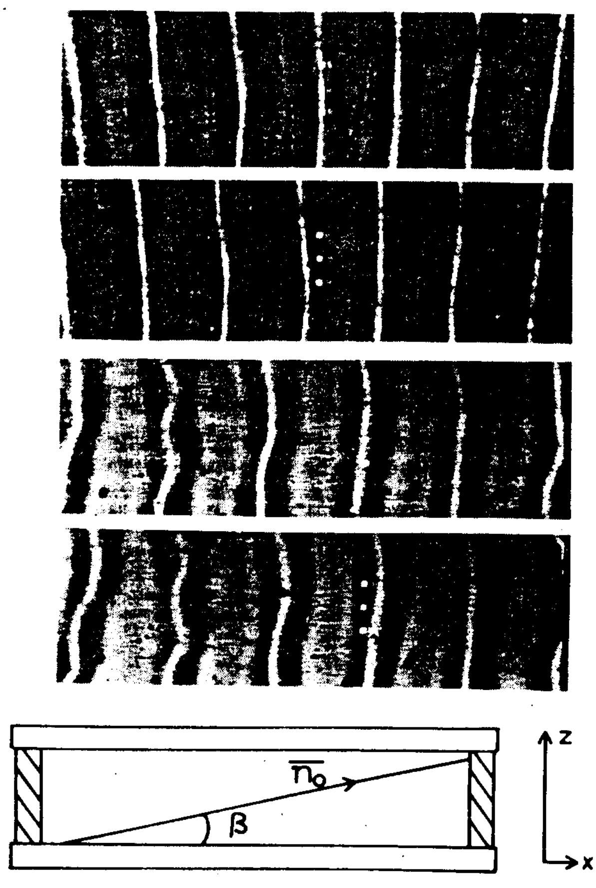

Figure 2. Propagating electroconvection in a nematic under DC excitation. The bottom drawing shows the experimental geometry. The video recordings, starting from the top, correspond to: (i) t = 0, (ii) t = 120 s for V_z = +2.4 V, (iii) t = 0, and (iv) t = 120 s for V_z = −2.4 V. Each optical domain contains two rolls of opposite vorticity. The sensitivity of the DC instability to local surface conditions causes the deviations from straight rolls (× 70).

from a solution with the standard 3% concentration of the polyimide, the EHD patterns were rather patchy and, further, the threshold varied considerably with time. Presumably a dense coating of polyimide acts as an insulating layer which reduces the field in the sample as ions collect near these layers. We reduced the concentration of the polyimide solution to about a tenth of the usual value and got better EHD patterns under DC excitation, probably due to a porous coating of the polymer. We found that the EHD pattern formation was more uniform when the sample thickness was larger. Typically we used samples with $d \simeq$ 80 μm. We obtained almost 'normal' rolls as the temperature was raised to 60°C (using a Mettler FP82 hot stage). Maintaining the voltage close to the threshold (2.2 V at 60°C), we clearly see propagating rolls whose direction of propagation *reverses when the field is reversed* (Figure 2). We measured the tilt angle in the sample using an optical technique[17] to be $\simeq 1.2°$. Using the usual values[5-7] for the viscosities and elastic constants for MBBA and the measured values of $\sigma_{\parallel}/\sigma_{\perp} \approx 1.1$, and $(e_1 + e_3)$, $\approx 10^{-4}$ cgs units, for the mixtures[5], the calculated value (at room temperature) of V_{th} is 4 V and the velocity of propagation is 0.1 μm s^{-1}. The experimental value of the velocity is 0.3 μm s^{-1} at 60°C. The higher value can arise partly from the lower viscosity at 60°C. For the experimental configuration used (Figure 2), the theory predicts that the direction of propagation is along the positive (negative) X-axis for positive (negative) E_0. Taking into account the image inversion in the microscope, the observation, which has been confirmed on several independent samples, agrees with the prediction. We must however note that the TW instabilities under AC excitation studied in earlier experiments[8,9] cannot be explained on the basis of our model.

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