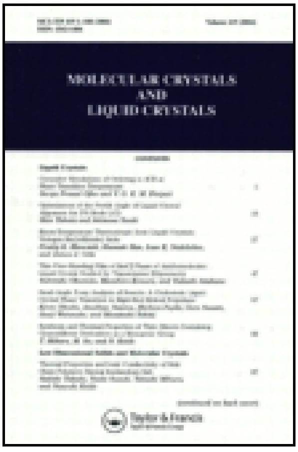

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Propagating Electrohydrodynamic Instabilities in Nematics

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Propagating Electrohydrodynamic Instabilities in Nematics

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We show that a propagating electrohydrodynamic instability occurs in a nematic liquid crystal at the threshold of DC excitation if the symmetry of a planar aligned cell is changed by introducing a small pretilt of the director. Flexoelectric torques, which are $\pi/2$ out of phase with those produced by other couplings, provide the driving mechanism for the propagating mode, whose direction depends on that of the applied field. Some experimental observations on this mode are also presented.

Keywords: electrohydrodynamic instabilities, nematics

I. INTRODUCTION

Nematic liquid crystals with negative or weakly positive dielectric anisotropy ($\Delta\varepsilon$) exhibit an electrohydrodynamic (EHD) instability under the action of a DC or AC electric field.[1] Due to the simplicity of the experimental set up this system has gained a lot of importance in the study of pattern formation.[2,3] The orientational ordering in the nematic ensures that the convective instability sets in the form of cylindrical rolls rather than more complicated structures. At low frequencies of excitation oblique convective rolls have been found where the wave-vector $\bar{q}$ makes a non-zero angle with $\hat{n}_o$, the initial orientation of the director.[4] This is now well understood as arising mainly from the flexoelectric property of the nematic.[5,6] Recently, a traveling wave (TW) instability has been observed in the conduction regime in thin samples.[3,7] However, all the theoretical studies so far have failed to give solutions corresponding to such a TW mode in experimentally realizable situations.

The possibility of oscillatory or TW electro-convection in nematics under DC excitation has been discussed by a few authors in the past. Ioffe[8] and Matyushichev and Kovnatskii[9] included the flexoelectric terms in the EHD equations and incorrectly got oscillatory solutions. In Reference 8 the flexoelectric terms in the equation of motion and in the torque balance equation are incorrect and in Reference 9 the boundary conditions are not properly taken into account. Laidlaw[10] showed that materials with $\Delta\varepsilon > 0$ and negative conductivity anisotropy ($\Delta\sigma$) could exhibit an oscillatory EHD instability under DC excitation. However, as the threshold for this instability is much higher than that for the Freedericksz transition, it cannot be experimentally observed. Penz[11] suggested that in materials with $\Delta\varepsilon > 0$ and $\Delta\sigma > 0$ a TW EHD instability could set in if the sample thickness is very small, but again at DC voltages above the Freedericksz threshold. In section 2 below we extend the calculations of Penz by including the flexoelectric terms. It is found that the inclusion of these terms suppresses the TW solutions.

In section 3 we demonstrate the existence of a propagating EHD instability at the threshold of DC excitation in samples having a small pretilt of the director at the bounding plates. The propagation is caused by the flexoelectric torques which are $\pi/2$ out of phase with respect to the hydrodynamic and elastic torques which arise in the convecting nematic. The propagation direction depends on the sign of the applied electric field. Some experimental observations on this new type of instability are also reported. A preliminary report on the one-dimensional model has been reported earlier.[12]

II. TRAVELING WAVE INSTABILITY

As mentioned earlier Penz[11] showed that it is possible to get oscillatory solutions to the EHD equations under DC excitation for materials with $\Delta\varepsilon > 0$ and $\Delta\sigma > 0$, at voltages slightly above the Freedericksz threshold. Such solutions exist over a narrow voltage range and only for very thin samples. We have extended these calculations by including flexoelectricity to see its influence on the domain of existence of the oscillatory solutions in the parameter space.

Consider a planar aligned nematic layer of thickness d with the director $\hat{n}_o$ along the x-axis and subjected to a DC electric field E_o applied along z. A perturbation in the director field, described by the angle θ, creates a space charge density Q in the medium due to the conductivity anisotropy. This space charge density in turn gives rise to the transverse electric field E_x. The force on the space charges due to E_o creates a flow in the medium described by the velocity field $\vec{v}$, if E_o is sufficiently large. The linearized EHD equations describing the system are[1]
Poisson equation:

$$4\pi Q - \varepsilon_{\parallel} \frac{\partial E_x}{\partial x} - \varepsilon_{\perp} \frac{\partial E_z}{\partial z} - \Delta\varepsilon E_o \frac{\partial \theta}{\partial x} - 4\pi(e_1 + e_3) \frac{\partial^2 \theta}{\partial x \partial z} = 0 \qquad (1)$$

where e_1 and e_3 are the flexoelectric coefficients.
Charge conservation equation:

$$\frac{\partial Q}{\partial t} + \sigma_{\parallel} \frac{\partial E_x}{\partial x} + \sigma_{\perp} \frac{\partial E_z}{\partial z} + \Delta\sigma E_o \frac{\partial \theta}{\partial x} = 0 \qquad (2)$$

Torque balance equation:

$$\gamma_1 \frac{\partial \theta}{\partial t} - K_3 \frac{\partial^2 \theta}{\partial x^2} - K_1 \frac{\partial^2 \theta}{\partial z^2} - \frac{\Delta\varepsilon}{4\pi} (E_o^2 \theta + E_o E_x)$$

$$+ (e_1 + e_3) \frac{\partial E_x}{\partial z} + \alpha_3 \frac{\partial v_x}{\partial z} + \alpha_2 \frac{\partial v_z}{\partial x} = 0 \qquad (3)$$

where K_3 and K_1 are the bend and splay elastic constants respectively, and $\gamma_1 = \alpha_3 - \alpha_2$. The α_i are the Leslie viscosity coefficients.
Equations of motion:

$$-\frac{\partial p}{\partial x} + \alpha_3 \frac{\partial^2 \theta}{\partial z \partial t} + \eta_1 \frac{\partial^2 v_x}{\partial x^2} + \eta_2 \frac{\partial^2 v_x}{\partial z^2} = 0 \tag{4}$$

where p is the pressure,

$$\eta_1 = \alpha_1 + \alpha_5 + \frac{1}{2}(\alpha_4 + \alpha_3 + \alpha_6)$$

and

$$\eta_2 = \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6);$$

$$-\frac{\partial p}{\partial z} + \alpha_2 \frac{\partial^2 \theta}{\partial x \partial t} + \eta_3 \frac{\partial^2 v_z}{\partial z^2} + \eta_4 \frac{\partial^2 v_z}{\partial x^2} + E_o Q = 0 \tag{5}$$

where $\eta_3 = 1/2\,(\alpha_4 - \alpha_5 - \alpha_2)$

and $\eta_4 = 1/2\,(\alpha_4 + \alpha_5 - \alpha_2)$

Substituting solutions of the form

$$\theta = \theta_1 \exp i(\bar{q} \,.\, \bar{r} - \omega t), \text{ etc.},$$

where $\omega = W_R - iW_I$ in Equations (1–5), we get a polynomial of degree 8 in q_z. There are eight boundary conditions to be satisfied: $\theta = E_x = v_x = v_z = 0$ at $z = \pm d/2$. These boundary conditions result in a set of linear homogeneous equations. The solution of the boundary value problem requires that the roots of the above mentioned polynomial satisfy these equations. For applied fields greater than or equal to a threshold value, the boundary value problem can be satisfied only for particular values of ω and q_x.

Figure 1 shows the variation of W_I with $2\pi d/\lambda$ obtained at a voltage slightly above the Freedericksz threshold. For all the solutions shown the boundary value problem is solved only if $W_R \equiv 0$, i.e., these solutions correspond to a stationary instability. However, there is a gap in the $2\pi d/\lambda$ values for which no solutions are shown. In this gap a non-zero value of W_R is required to solve the boundary value problem. Thus in this gap oscillatory solutions to the EHD equations exist. In curves a, b and c we have shown the results of the calculations with $(e_1 - e_3) = 0$, -2.0×10^{-4} and -4.0×10^{-4} cgs units, respectively. It is clear from the figure that flexoelectricity *suppresses* the range of existence of the oscillatory solutions. We

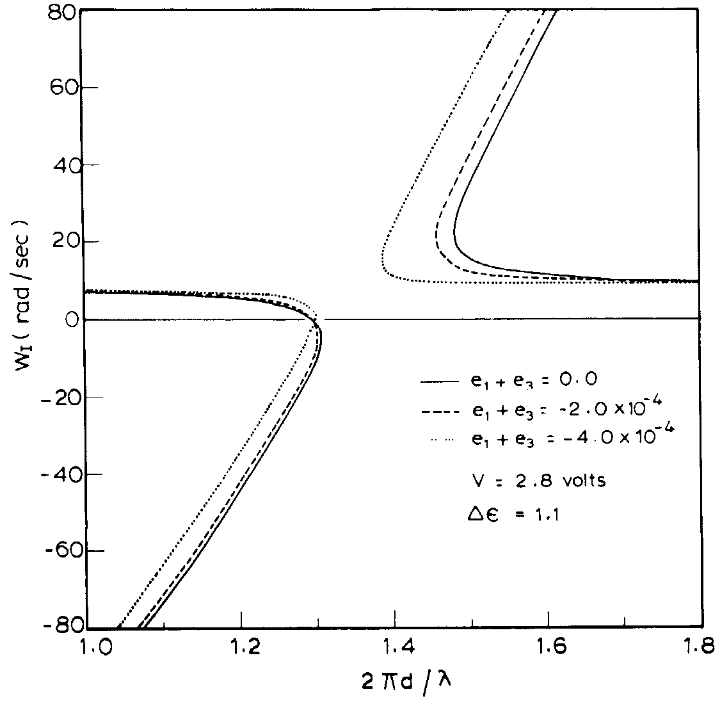

FIGURE 1 The variation of W_i with $2\pi d/\lambda$ obtained from the calculations. Note that the gap in the figure corresponding to oscillatory solutions decreases when the magnitude of $(e_1 + e_3)$ is increased.

have repeated these calculations for a few positive values of $\Delta\varepsilon$ and have obtained similar results.

III. PROPAGATING INSTABILITY

It is well known that some of the techniques commonly used to produce planar alignment of the director in a nematic layer produces a small pretilt of the director at the bounding surfaces.[13] We show below that the EHD instability obtained in such a sample under DC excitation is not stationary but propagates in a direction normal to the roll axis. Unlike in the TW the two possible propagation directions are not equivalent and the particular direction chosen is determined by the signs of the applied field, the tilt angle and that of the combination $(e_1 + e_3)$ of the flexoelectric coefficients.

(a) The EHD equations

The geometry considered is shown in Figure 2. For the sake of a clear understanding of the underlying mechanism, we make the following simplifications in the problem: (1) the dielectric anisotropy is set equal to zero; (2) the pretilt angle β is small enough to retain only linear terms in β; and (3) even though the flexoelectric effect

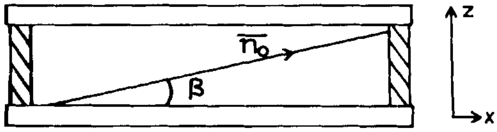

FIGURE 2 The experimental geometry considered in section 3. β is the pretilt angle.

tends to produce oblique rolls under DC excitation,[5,6] we assume that the convection rolls are normal to $\hat{n}_o$. Under these approximations, the linearized EHD equations are:

The Poisson equation:

$$4\pi Q - \varepsilon\left(\frac{\partial E_x}{\partial x} + \frac{\partial E_z}{\partial z}\right) - 4\pi(e_1 + e_3)\frac{\partial^2\theta}{\partial x\partial z} + 4\pi\beta(e_1 + e_3)\left(\frac{\partial^2\theta}{\partial x^2} - \frac{\partial^2\theta}{\partial z^2}\right) = 0 \tag{6}$$

The charge conservation equation:

$$\frac{\partial Q}{\partial t} + \sigma_{\parallel}\frac{\partial E_x}{\partial x} + \sigma_{\perp}\frac{\partial E_z}{\partial z} + \Delta\sigma E_o\frac{\partial\theta}{\partial x} + 2\beta\Delta\sigma\left(\frac{\partial E_z}{\partial x} + E_o\frac{\partial\theta}{\partial z}\right) = 0 \tag{7}$$

The torque balance equation:

$$\gamma_1\frac{\partial\theta}{\partial t} - K_3\frac{\partial^2\theta}{\partial x^2} - K_1\frac{\partial^2\theta}{\partial z^2} + (e_1 + e_3)\frac{\partial E_x}{\partial z} + \alpha_3\frac{\partial v_x}{\partial z} + \alpha_2\frac{\partial v_z}{\partial x} + \beta\left[(e_1 + e_3)\left(\frac{\partial E_z}{\partial z} - \frac{\partial E_x}{\partial x}\right) - 2(K_3 - K_1)\frac{\partial^2\theta}{\partial x\partial z} - 2\gamma_2\frac{\partial v_x}{\partial x}\right] = 0 \tag{8}$$

where $\gamma_2 = \alpha_3 + \alpha_2$

The equations of motion:

$$-\frac{\partial p}{\partial x} + \alpha_3\frac{\partial^2\theta}{\partial z\partial t} + \eta_1\frac{\partial^2 v_x}{\partial x^2} + \eta_2\frac{\partial^2 v_x}{\partial z^2} - \beta\left[\gamma_2\frac{\partial^2\theta}{\partial x\partial t} + \eta_5\frac{\partial^2 v_z}{\partial z^2} - \eta_6\frac{\partial^2 v_z}{dx^2}\right] = 0 \tag{9}$$

where

$$\eta_5 = (2\alpha_1 + 2\alpha_5 - \alpha_6)$$

and

$$\eta_6 = \alpha_1 + \alpha_6$$

$$-\frac{\partial p}{\partial z} + \alpha_2 \frac{\partial^2 \theta}{\partial x \partial t} + \eta_4 \frac{\partial^2 v_z}{\partial x^2} + \eta_3 \frac{\partial^2 v_z}{\partial z^2}$$

$$+ E_o Q + \beta \left[\gamma_2 \frac{\partial^2 \theta}{\partial z \partial t} + \eta_7 \frac{\partial^2 v_x}{\partial x^2} + \alpha_6 \frac{\partial^2 v_x}{\partial z^2} \right] = 0 \tag{10}$$

where $\eta_7 = (\alpha_1 + \alpha_6 - 2\alpha_5)$.

(b) 1-Dimensional Calculations

In order to get a physical understanding of the problem let us first study the stability of the system by neglecting the boundary conditions. Using solutions of the form

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

the conditions for the onset of the instability are given by

$$4\pi K_3 q_x^2 + E^2 \varepsilon \frac{\alpha_2}{\eta_4} \frac{\Delta\sigma}{\sigma_\parallel} - \frac{\varepsilon \omega^2}{\sigma_\parallel} \left(\gamma_1 - \frac{\alpha_2^2}{\eta_4} \right) = 0 \tag{11}$$

$$\omega = \frac{\beta (e_1 + e_3) q_x E \left(\frac{\Delta\sigma}{\sigma_\parallel} + \frac{\alpha_2^2}{\eta_4} \right)}{\left(\gamma_1 - \frac{\alpha_2^2}{\eta_4} \right) \left(1 + \frac{\tau}{T} \right)} \tag{12}$$

where $\tau^{-1} = 4\pi\sigma_\parallel/\varepsilon$ is the charge relaxation frequency and $T^{-1} = k_3 q_x^2/(\gamma_1 - \alpha_2^2/\eta_4)$ is the director relaxation frequency.

The sum $(e_1 + e_3)$ of the flexoelectric coefficients arises from the aligned quadrupoles of the molecules and has a non-zero value in general.[14] Therefore, it is clear from Equation (12) that the convective rolls obtained at the threshold of the instability should propagate normal to their axes. Further, 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 direction reverses when the applied electric field is reversed. For the standard MBBA values of the material parameters from Equations (11) and (12) the velocity of propagation $(V = \omega/q_x)$ is found to be about 0.1 μm/sec.

The physical mechanism responsible for the propagating instability can be understood by referring to Figure 3. The transverse electric field gradient in the medium is large in the region where the charge density is high. When $\beta = 0$ (Figure 3a) this cannot produce any torque on the quadrupoles of the medium. On the other hand, when $\beta \neq 0$ (Figure 3b), the field gradient produces a torque on the director

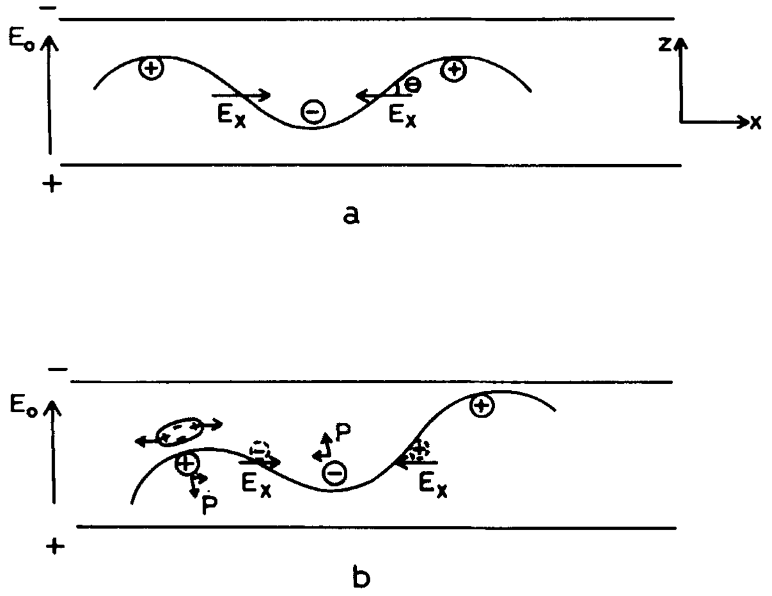

FIGURE 3 (a) $\beta = 0$, the space charges (shown in full circles) arising from the conductivity anisotropy cause EHD instability, (b) $\beta \neq 0$, the quadrupoles have an out of phase torque due to the horizontal field gradients and additional charges (in dotted circles) are collected due to the horizontal gradient in the flexoelectric polarization $\bar{p}$.

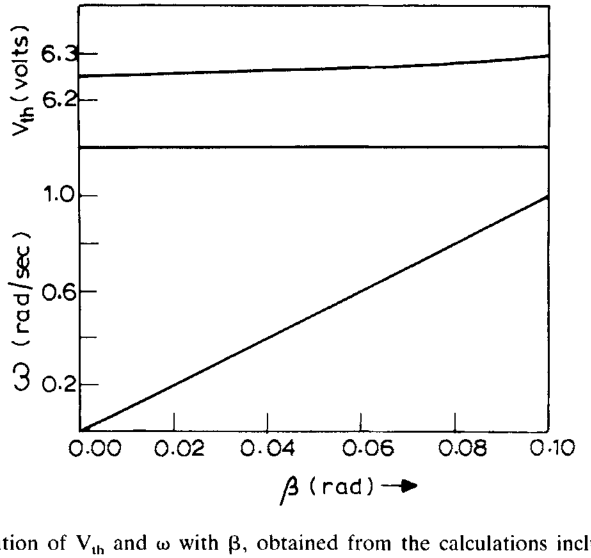

FIGURE 4 Variation of V_{th} and ω with β, obtained from the calculations including the boundary conditions for $\Delta\varepsilon = 0$ and MBBA values of the other material parameters.

which is spatially $\pi/2$ out of phase with the torques produced by other physical mechanisms. Further, when $\beta \neq 0$ the flexoelectric polarization has a divergence which gives rise to space charge densities which are $\pi/2$ out of phase with those collected by the coupling of the conductivity anisotropy with the curvature in the medium (Figure 3b). The force produced by E_o on these additional charges gives rise to hydrodynamic torques which are again $\pi/2$ 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 propagation is flexoelectric in origin, the direction of propagation depends on the sign of the applied field.

(c) Calculations Including the Boundary Conditions

The method used to solve the boundary value problem is the same as that outlined in section 2. Figure 4 shows the threshold voltage and the frequency ω as functions of the pretilt angle β, calculated using the standard MBBA values of the material parameters. For the range of β values shown the wave-vector $q_x \simeq 4/d$. The linear variation of ω with β is in agreement with Equation (12), as the threshold voltage is almost independent of β. Further, the direction of propagation is found to depend on the signs of the applied electric field, the pretilt angle β and that of $(e_1 + e_3)$, in agreement with the results of the one-dimensional calculations.

IV. EXPERIMENTAL

It is known that with polyimide coated and unidirectionally rubbed plates the nematic orients with a tilt angle of about 2°. Each sandwich cell used in our studies was made up of two such plates aligned in a mutually antiparallel orientation so as to give a uniform alignment of the director (Figure 2). The material used was a room temperature nematic mixture consisting of three chemically stable compounds, viz., CE-1700, CM-5115 and PCH-302 of Roche chemicals. When the coating on the plates was made from a solution with the standard 3% concentration of the polyimide, the EHD patterns obtained under DC excitation 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. However, on reducing the concentration of the polyimide to about 1/20th of the usual value, we got better EHD patterns under DC excitation, probably due to a porous coating of the polymer. The sample thickness was typically 15 μm. The pretilt angle β in the sample was measured using an optical technique[15] and it was found to be 1.2°.

At room temperature the pattern observed at the threshold under DC excitation consisted of propagating oblique rolls, whose propagation direction reversed when the field was reversed.[16] On increasing the temperature, the obliquity of the rolls decreased and at about 60°C almost normal rolls were obtained. We have made all the detailed observations at about 60°C so as to minimize the influence of the moving boundaries between domains with opposite tilts of the rolls on the propagation of the rolls themselves. The pattern was observed with a video camera mounted on a polarizing microscope, digitized with a resolution of 512 × 512 pixels of 256 grey levels and fed to a computer. Figure 5 shows the intensity profiles along a line normal to the rolls observed at intervels of 2 secs. plotted on top of one another. It is clear from the figure that the pattern propagates in opposite directions for opposite signs of the applied field, in agreement with the theory. The threshold voltage was slightly different for positive and negative voltage, probably due to some assymetry of the two electrodes. The velocity of propagation is also found to be different for the two signs of the voltage; +0.9 μm/sec. for +ve voltage slightly above threshold and −0.5 μm/sec. for −ve voltage. Under AC excitation the EHD pattern was found to drift with a velocity of about +0.2 μm/sec. just above the threshold. Hence the difference in the propagating velocity for positive and negative voltages could be arising from a small thickness gradient present in the cell which leads to a drift of the pattern in a particular direction independent of the sign of the voltage. From Figure 4 we see that for a pretilt of 1.2° the propagation velocity obtained from the theory for $\Delta\varepsilon = 0$ and the MBBA values

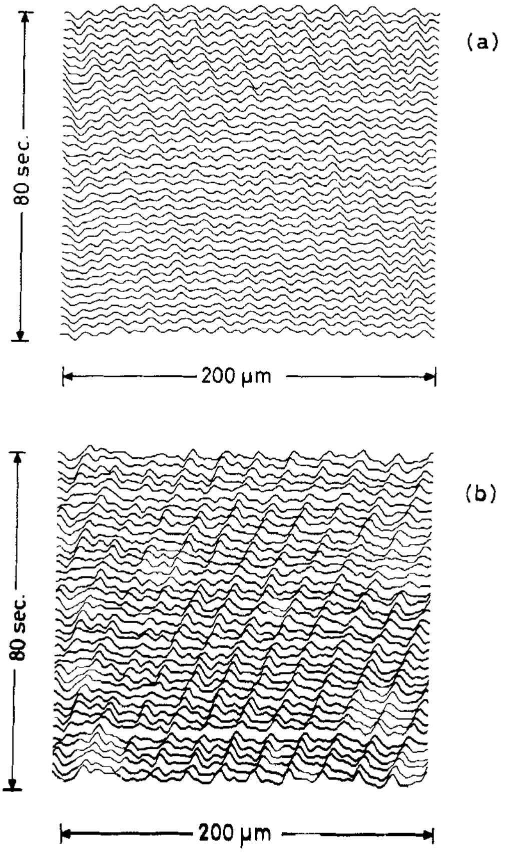

FIGURE 5 The light intensity profiles along a line normal to the rolls observed at intervals of 2 secs. plotted on top of one another. (a) V = −8.9 Volt. (b) V = +8.9 Volts. (V_{th} = 8.5 Volts). Note that the rolls propagate in opposite directions for opposite signs of the voltage.

of the other material parameters is about 0.5 μm/sec at room temperature. Thus, the observed propagation velocity is comparable to that obtained from the theory. However, as many of the material parameters of the mixture under study are not known, it is not possible to make a quantitative comparison between the theory and the experiment.

V. CONCLUSION

The oscillatory solutions of the EHD equations which exist over a narrow voltage range above the Freedericksz threshold in a planar aligned cells are suppressed by the flexoelectric effect. However, if the nematic is aligned with a small pretilt of the director at the bounding surfaces, then flexoelectricity leads to a propagating EHD instability under DC excitation. The results of our experiments on the propagating instability are in good qualitative agreement with the theoretical predictions.

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