Chapter 6

Influence of flexoelectricity on static distortions in nematics induced by an external electric field

CHAPTER 6

INFLUENCE OF FLEXOELECTRICITY ON STATIC DISTORTIONS IN NEMATICS INDUCED BY AN EXTERNAL ELECTRIC FIELD

6.1 INTRODUCTION

As discussed in the introductory chapter, nematic liquid crystals, in general, have non-zero dielectric and diamagnetic anisotropies. Therefore an external electric or magnetic field can be used to induce static distortions of the director field in a uniformly aligned sample. When the destabilizing field is applied normal to the initial orientation of the director, the deformation sets, in at a critical value of the field strength if the anchoring at the walls is strong. This is known as the Freedericksz transition [1]. In nematics with positive dielectric or diamagnetic anisotropy, the Freedericksz transition can be conveniently studied in three geometries [2,3]. In geometry 1 the sample is aligned homogeneously and the destabilizing field is applied normal to the bounding glass plates (Fig. la). At the threshold of the transition a splay distortion of the director develops in the medium (Fig. 1b). If the field is increased beyond the critical value, the distortion of the director field increases and is a combination of bend and splay. In geometry 2 the sample is again aligned homogeneously and the field is applied normal to the undistorted director, parallel to

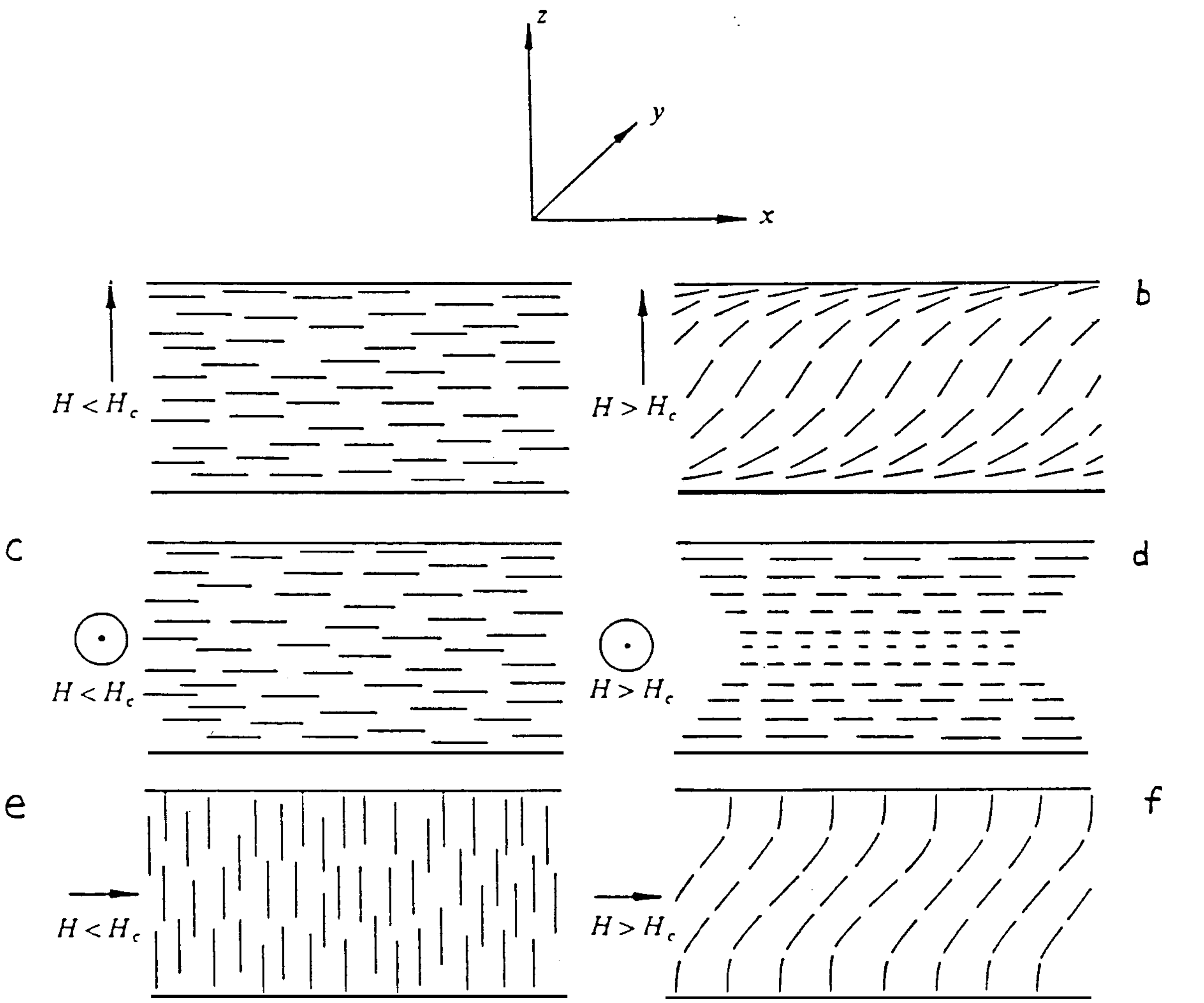

Fig.1. The three principal types of Freedericksz deformation.

the glass plates (Fig. 1c). A twist distortion develops in the medium as the field strength is increased beyond the critical value (Fig. Id). In geometry 3 the sample is aligned homeotropically and the field is applied parallel to the plates (Fig. 1e). A bend distortion is created in the medium at the threshold (Fig. If). Beyond the threshold there is a combination of bend and splay. An expression for the critical value of the field can be obtained by equating the elastic and the dielectric or diamagnetic torques. For an external magnetic field the threshold or critical values in the three geometries are given by [2,3]

$$H_c^i = (\pi/d)\,(K_i/\chi_a)^{1/2} \quad , \ i=1,2,3 \qquad (1)$$

where H_c^1, H_c^2, H_c^3 are the critical values in geometries 1,2 and 3, respectively, d is the sample thickness, χ_a the volume diamagnetic anisotropy and K_1, K_2 and K_3 are the splay, twist and bend elastic constants, respectively. The corresponding relations for an applied electric field are [2.3]

$$E_c^i = (\pi/d)\,(4\pi K_i/\epsilon_a)^{1/2} \quad , \ i=1,2,3 \qquad (2)$$

where ϵ_a is the dielectric anisotropy. It is clear from the above relations that if the critical values of the applied field can be measured in the three geometries the elastic constants of a nematic can be determined. Thus Freedericksz transition offers a convenient method to measure the elastic constants of a nematic and is widely used for this purpose.

Recently Lonberg and Meyer [4] reported a new type of transition in geometry 1, induced by a magnetic field in a polymeric nematic. Unlike the classical Freedericksz transition, the deformed state in this case is characterized by a periodic splay - twist distortion of the director field and leads to the appearance of domains with their axes parallel to the undistorted director $\hat{n}_o$. They also showed that such a distortion is preferred to a uniform splay distortion if the ratio of the twist and splay elastic constants $R = K_2/K_1$ is less than a critical value $R_c \approx 0.303$.

It was later pointed out by Kini [5] and Oldano [6] that static periodic distortions should also be observed in geometry 2 under a magnetic field. The equations describing the distorted director field in this case and in the case of geometry 1 are isomorphic under the transformation [5]

$$(K_1\ ,\ K_2\ ,\ \theta,\ \Phi,\ H_y\ ) \longrightarrow (K_2\ ,\ K_1\ ,\ \Phi,\ -\theta,\ H_z\ ) \qquad (3)$$

It therefore follows that in geometry 2 the static periodic distortion is preferred to the uniform twist distortion if the ratio $R' = K_1/K_2$ is less than the critical value $R_c' \approx 0.303$.

Static periodic distortions in a nematic in geometry 1, induced by an external electric field have been the subject of many experimental [7] and theoretical studies [8,9]. The theory of Bobylev and Pikin [8] explains these periodic distortions as arising from the flexoelectric effect [10]. Neglecting the elastic anisotropy they showed that these distortions can be obtained if the dielectric anisotropy satisfies the inequality

$$|\epsilon_a| < (4\pi/K)(e_1 - e_3)^2 \quad ,$$

where e_1 and e_3 are the flexoelectric coefficients. This model was later extended by Bobylev et al. [11] by including the elastic anisotropy. It may be noted here that the three dimensional analysis presented in chapter 4 reduces to the model of Bobylev et al. when the wavevector of the distortion is normal to $\hat{n}_o$, in which case all the hydrodynamic terms drop out of the problem.

The fact that the static periodic distortion observed under a magnetic field follows from the model of Bobylev et al. appears not to have been appreciated in the recent literature. We have therefore calculated the threshold field of the static distortions in geometry 1 as a function of the ratio $R = K_2/K_1$. When $\epsilon_a = 0$, the periodic distortion is caused by flexoelectricity and can be observed for any value of R. When $\epsilon_a > 0$, the periodic distortion is obtained upto a critical value R_c of R. For $R > R_c$, the classical splay Freedericksz transition is obtained and the flexoelectric terms do not contribute to the solutions. As $|e_1 - e_3| / \epsilon_a$ is decreased, the critical value R_c decreases and when $[e_1 - e_3] / \epsilon_a = 0$, $R_c \approx 0.303$. When a magnetic field is used to induce the distortion the flexoelectric terms are naturally absent and as found by Lonberg and Meyer [4], $R_c \approx 0.303$.

In this chapter the influence of flexoelectricity on the static distortions induced by an electric field in geometry 2 is also calculated. In this geometry the flexelectric terms are found to decrease the critical value R_c' of $R' = K_1/K_2$ upto which periodic distortions are observed. For all values of $R' > R_c'$ a new type of transition is favoured which has a lower threshold than the twist Freedericksz transition. This transition is caused by the flexoelectric terms and the distorted state which is non-periodic is characterized by a non-planar deformation of the director field, described by two polar angles 8 and Φ.

In geometry 3 flexoelectricity does not lead to any linear terms in the equations describing the system. Hence the classical bend Freedericksz transition is obtained in this case.

6.2 ANALYSIS OF GEOMETRY 1

Taking $\hat{n}_o$ along the X-axis and the applied electric field $\vec{E}$ along Z, Bobylev et al.[11] obtain the following equations, which describe the response of the system to an external DC electric field. As mentioned earlier, these equations can be obtained from the electrohydrodynamic equations of chapter 4 by setting a = $\pi/2$.

$$K_1(\partial^2\theta/\partial Z^2) + K_2(\partial^2\theta/\partial Y^2) + (e_1 - e_3)E_z(\partial\Phi/\partial Y) + (\epsilon_a E_z^2/4\pi)\,\theta + (K_1 - K_2)(\partial^2\Phi/\partial Y\partial Z) = 0 \qquad (4)$$

$$K_1(\partial^2\Phi/\partial Y^2) + K_2(\partial^2\Phi/\partial Z^2) - (e_1 - e_3)E_z(\partial\theta/\partial Y) + (K_1 - K_2)(\partial^2\theta/\partial Y\partial Z) = 0 \qquad (5)$$

where θ is the angle made by $\hat{n}$ with the XY plane and Φ the angle between the X-axis and the projection of $\hat{n}$ on the XY plane. Assuming

$$\theta = \theta_o \ \exp[i(q_z Z + q_y Y)]$$
$$\Phi = \Phi_o \ \exp[i(q_z Z + q_y Y)] \qquad (6)$$

the condition for the existence of non-trivial solutions leads to the following polynomial in $S = q_z / q_y$.

$$S^4 + [2 - \{\epsilon_a/(4\pi K_1)\} (E_z/q_y)^2] S^2$$
$$-(E_z/q_y)^2 [\epsilon_a/(4\pi K_2) + (e_1 - e_3)^2 /(K_1 K_2)] + 1 = 0 \qquad (7)$$

The boundary conditions are:

$$\theta (Z = \pm d/2) = \Phi (Z = \pm d/2) = 0 \qquad (8)$$

As discussed in detail in chapter 4 , these boundary conditions lead to a boundary value determinant. **It** is given by

$$D_{ij} = 0 \qquad , i,j = 1,4 \qquad (9)$$

The elements of the determinant are:

$D_{1j} = \cos(S_j \delta)$, $D_{2j} = \sin(S_j \delta)$, $D_{3j} = B_j D_{1j}$ and $D_{4j} = B_j D_{2j}$.

Where S_j are the roots of Eq.(7), $\delta = q_y d/2$ and

$$B_j = [(K_1 - K_2)S_j + i\,(e_1 - e_3)(E_z/q_y)]/(K_1 + K_2 S_j^2) .$$

Eqs.(7) and (9) form a characteristic value problem and the method of solution is similar to that described in chapter 4.

The threshold voltage V_{th} and the wavevector q_y of the distortion are shown in Fig.2, for different values of ϵ_a. The standard MBBA values of the other material parameters were used in the calculations. The dashed lines in the figure correspond to the uniform splay Freedericksz transition, the dotted lines to the periodic distortion obtained in the absence of the flexoelectric terms and the continuous lines to the periodic distortion obtained when the flexoelectric terms are taken into account. It is clear from the figure that when $\epsilon_a = 0$, the periodic distortion is caused by flexoelectricity and is obtained for all values of R. When $\epsilon_a > 0$, the periodic distortion is found upto a critical value R_c of R. For R > R_c the classical splay Freedericksz transition is obtained. Further, as $|e_1 - e_3|/\epsilon_a$ is decreased the critical value R_c decreases and when $[e_1 - e_3]/\epsilon_a = 0$, $R_c \approx 0.303$.

In order to clearly understand the influence of flexoelectricity in this geometry, let us consider the

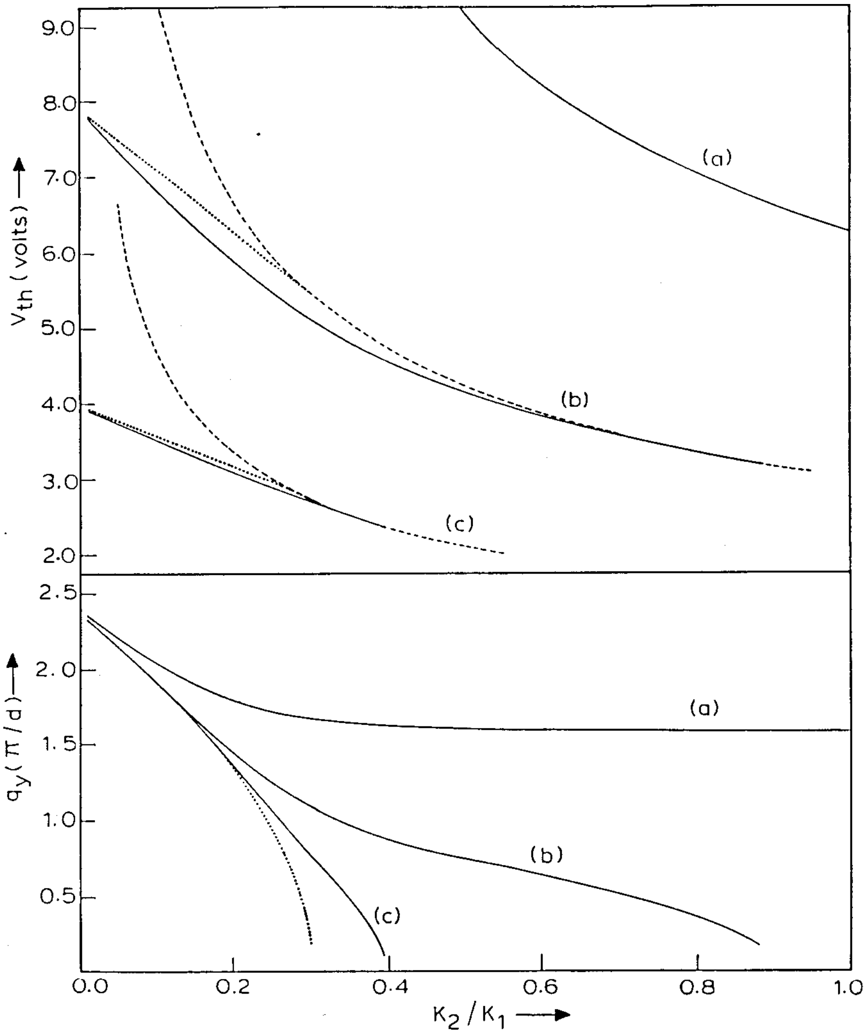

Fig.2. Variation of the threshold voltage and the wavevector of the distortion at the threshold, with the ratio R = K_2/K_1 in geometry 1. The curves labeled (a), (b) and (c) correspond to ϵ_a = 0, 0.1 and 0.5, respectively. Dashed lines: uniform splay Freederiksz transition. Dotted lines: periodic distortion in the absence of flexoelectricity. Solid lines: periodic distortion with flexoelectricity. Note that when the flexoelectric terms are neglected, q_y goes to zero at $R_c \approx 0.303$ for all positive values of ϵ_a.

flexoelectric energy density given by

$$f^{fl} = -\vec{P}.\vec{E} = -E_z[e_1\theta(\partial\Phi/\partial Y) + e_3\Phi(\partial\theta/\partial Y)] \qquad (10)$$

Here we have neglected the surface terms as the anchoring of the director at the two surfaces is assumed to be strong. It is clear from Eq. (10) that flexoelectricity favours a periodic distortion of the director. Further f^{fl} is minimized if $\theta(y)$ and $\Phi(y)$ are 90' out of phase with each other. In the absence of the flexoelectric terms the periodic distortion is obtained only if R is less than about 0.303. Since the flexoelectric terms also favour such a distortion, the critical value R_c increases when they are taken into account.

6.3 ANALYSIS OF GEOMETRY 2

Taking $\hat{n}_o$ along the X-axis and the applied field $\vec{E}$ along Y, the linearized torque balance equations are:

$$K_1(\partial^2\Phi/\partial Y^2) + K_2(\partial^2\Phi/\partial Z^2) + (K_1 - K_2)(\partial^2\theta/\partial Y\partial Z) + (\epsilon_a E_y^2/4\pi)\,\Phi + (e_1 - e_3)E_y(\partial\theta/\partial Z) = 0 \qquad (11)$$

$$K_1(\partial^2\theta/\partial Z^2) + K_2(\partial^2\theta/\partial Y^2) + (K_1 - K_2)(\partial^2\Phi/\partial Y\partial Z) - (e_1 - e_3)E_y(\partial\Phi/\partial Z) = 0 \qquad (12)$$

Note that Eqs.(11) and (12) and Eqs.(4) and (5) are not isomorphic under the transformation (3) with $\vec{E}$ instead of $\vec{H}$, due to the presence of the flexoelectric terms. However, these two sets of equations are isomorphic under the transformation

$$(\theta, \Phi, Y, Z, E_y) \longrightarrow (\Phi, \theta, Z, Y, E_z) \qquad (13)$$

In order to clearly understand the influence of flexoelectricity in this geometry, let us simplify the problem by taking $\epsilon_a = 0$. Assuming the solutions

$$\theta = \theta_o \exp[i(q_z Z + q_y Y)] \text{ and } \Phi = \Phi_o \exp[i(q_z Z + q_y Y)],$$

we get the following relation between the electric field and the wavevector:

$$E_y = (K_1 K_2)^{1/2} (q_y^2 + q_z^2)/[q_z |e_1 - e_3|] \qquad (14)$$

It is clear from this equation that E_y is minimized for $q_y = 0$. Thus in this geometry flexoelectricity does not lead to a periodic distortion of the director field when $\epsilon_a = 0$, but gives rise to a uniform distortion at a critical value of the field. However, unlike the uniform twist Freedericksz transition this new transition is characterized by both θ and Φ distortion angles in the director field. Neglecting the Y dependence in Eqs.(11) and (12) the following solutions can be obtained.

$$\theta = a\,(K, \;/K_1\;)^{1/2}\,\sin(2\pi Z/d)$$
$$\Phi = \pm\, a\,\{\, \cos(2\pi Z/d) - 1 \,\} \qquad (15)$$

where a is an arbitrary constant., The relative signs of θ and Φ depend on the signs of $(e_1 - e_3)$ and E_y. The critical value of the field is given by

$$E_{y_c} = (2\pi/d)\;(K_1 K_2)^{1/2}\;/\;|e_1 - e_3| \qquad (16)$$

When $\epsilon_a \neq 0$ the problem can be solved numerically as in the previous section. The variation of the threshold field and the wavevector q_y at the threshold obtained from the calculations are shown in Figs.3 and 4, for a few values of ϵ_a. The standard MBBA values of all the other parameters were used in the calculations. In the absence of flexoelectricity, as found by Kini [5] and Oldano [6] for the analogous magnetic case, a periodic distortion is favoured for $R' < 0.303$ for any positive value of ϵ_a. For $R' > 0.303$ the uniform twist Freedericksz transition occurs. When the flexoelectric terms are included the critical value R'_c of R' up to which the periodic

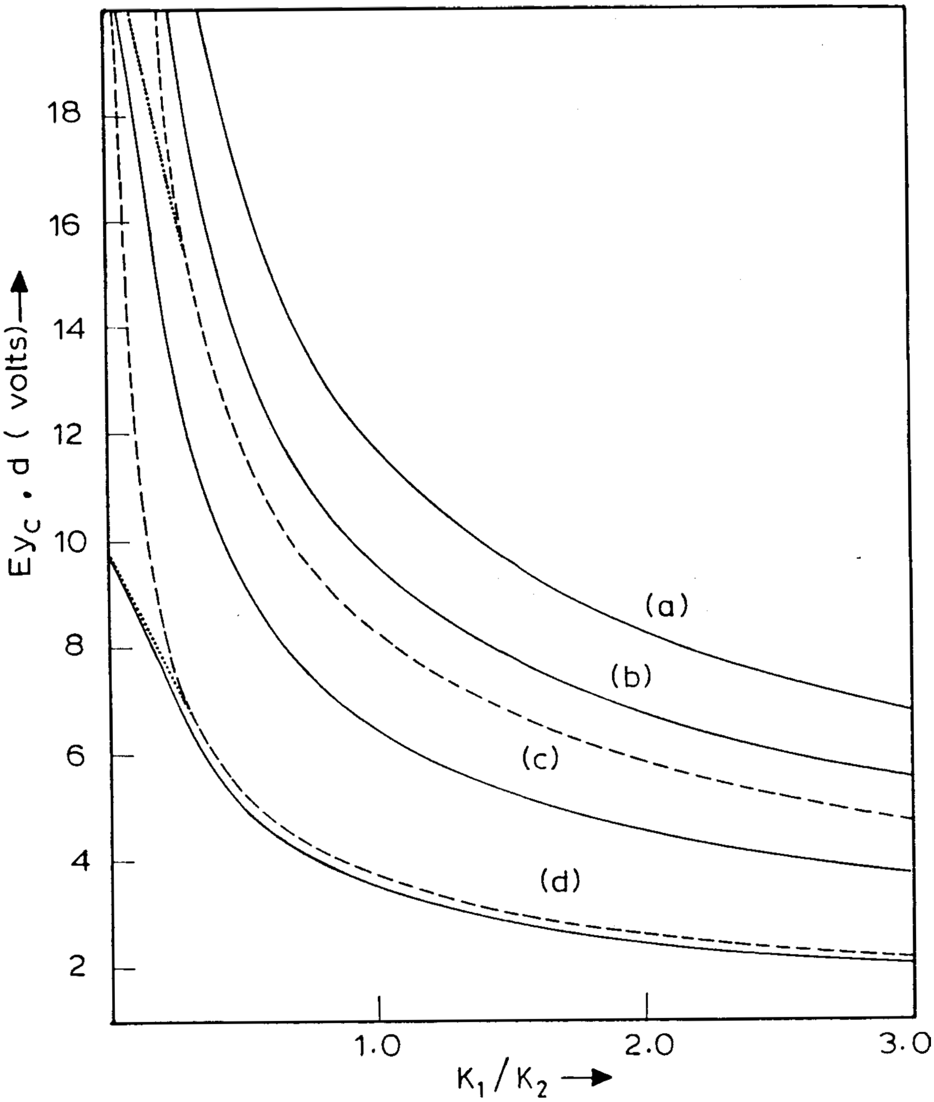

Fig.3. The product E_{y_c}.d as a function of $R' = K_1/K_2$ in geometry 2. (a) $\epsilon_a = -0.1$, (b) $\epsilon_a = 0$, (c) $\epsilon_a = 0.1$ and (d) $\epsilon_a = 0.5$. In the last two cases the dotted lines correspond to the periodic distortion and the dashed lines to the twist Freedericksz transition in the absence of flexoelectricity. The solid lines are obtained from calculations including the flexoelectric terms.

distortion is favoured decreases, the decrease being greater for larger values of $|e_1 - e_3| / \epsilon_a$. Further, for $R' > R'_c$, a uniform distortion with both polar angles θ and Φ is found to have a lower threshold than the uniform twist distortion. When $\epsilon_a = 0$, we get the new type of transition for all values of R'. As shown later, it can also be obtained for small negative values of ϵ_a less than a critical value.

In almost all nematics the ratio R' is larger than 1 and therefore in practical situations we can neglect the Y dependence in Eqs.(11) and (12). They can then be solved analytically for any value of ϵ_a to obtain the following solutions:

$$\theta = (a\, M_1/L)\, [\sin(LZ) + R\, (1 - \cos(LZ) + \{N_2 L/(M_1 M_2)\}\, Z]$$

$$\Phi = \pm\, a\, [\cos(LZ) + R \sin(LZ) - 1] \qquad (17)$$

with the condition that $\tan x = - N_2 x/(M_1 M_2)$ (18)

where $N_2 = \epsilon_a E_Y^2/(4\pi\, K_2)$; $M_i = (e_1 - e_3)E_y/K_i$, $i = 1,2$; $L = (N_2 + M_1 M_2)^{1/2}$, $x = Ld/2$ and $R = (1 - \cos Ld)/\sin Ld$.

The condition (18) gives the following expression for the

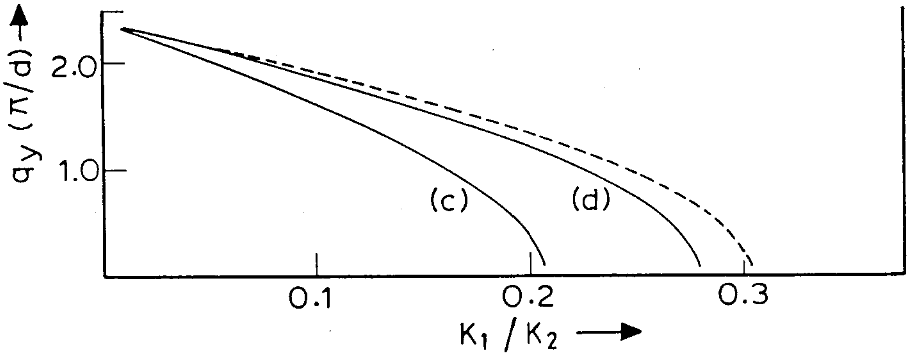

Fig.4. **Variation of q_y with R'. The dashed line is obtained when the flexoelectric terms are neglected and the solid lines are obtained when they are included.** (c) ϵ_a=0.1 and (d) ϵ_a □ **0.5. Note that $q_y \rightarrow 0$ at some R'.**

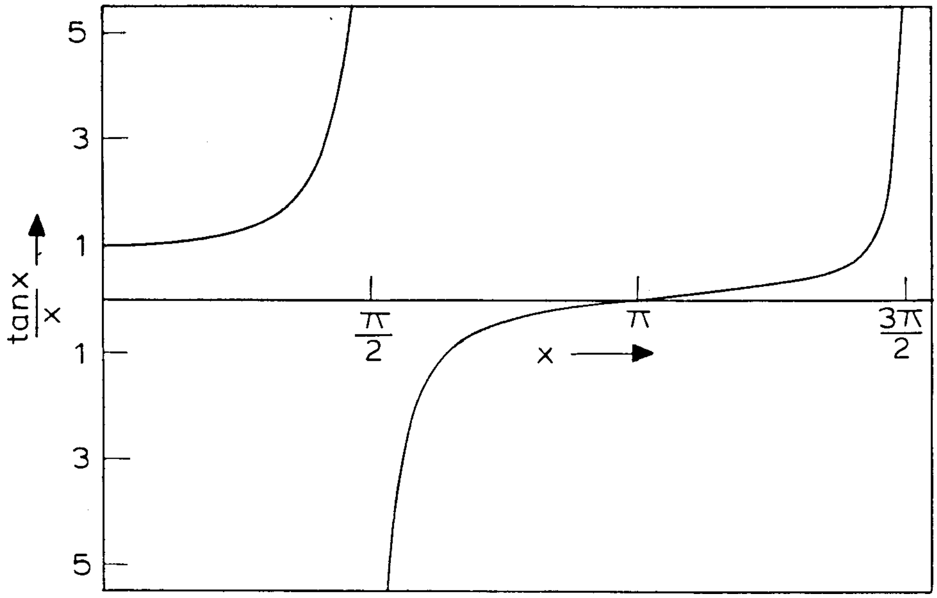

Fig.5. Variation of tan x / x with x.

threshold field:

$$E_{y_c} = (2\, x_o/d)\, [\epsilon_a/(4\pi\, K_2) + (e_1 - e_3)^2/(K_1K_2)]^{1/2} \qquad (19)$$

where x_o is the lowest non-zero value of x satisfying Eq.(18). x_o can be easily read off a plot of tanx /x (Fig.5). From Eq.(18) we see that when $\epsilon_a \rightarrow \infty$, $x_o \rightarrow \pi/2$ and Eq. (19) reduces to

$$E_{y_c} = (\pi/d)\, [\epsilon_a/(4\pi\, K_2) + (e_1 - e_3)^2/(K_1K_2)]^{1/2} \qquad (20)$$

Comparing Eqs.(20) and (2) it is clear that even in the limit of $\epsilon_a \rightarrow \infty$, the new type of transition has a slightly lower threshold due to the presence of the flexoelectric terms. From Eq.(19) we find that if ϵ_a is negative the flexoelectric transition can be observed only if

$$|\epsilon_a| < (4\pi/K_1)(e_1 - e_3)^2 \; .$$

Fig.6 shows the θ and Φ profiles in the cell just above the threshold calculated for the standard MBBA values of the material parameters. When $\epsilon_a = 0$, the amplitudes of θ and Φ are comparable and when $\epsilon_a = 5$, the amplitude of Φ is much larger than that of θ since the dielectric torque strongly favours the Φ distortion.

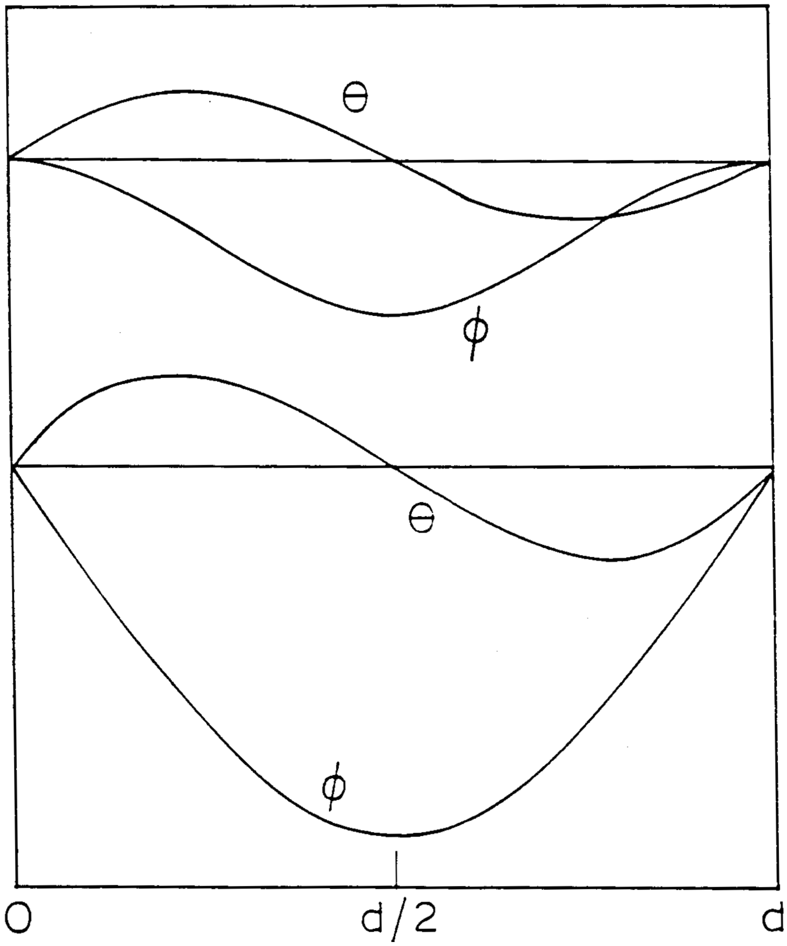

Fig.6. The θ and 9 profiles just above the threshold of the new type of deformation. Upper part: $\epsilon_a = 0$. Lower part: $\epsilon_a = 5.0$. In the latter case 9/6 is shown in the figure to save space.

The flexoelectric free energy density due to an arbitrary non-planar director distortion in geometry 2 is given by

$$f^{fl} = - E_y[e_1\Phi\ (\partial\theta/\partial Z) + e_3\theta\ (\partial\Phi/\partial Z)] \qquad (21)$$

It is clear from this expression that flexoelectricity does not favour a periodic distortion of the director field, but a uniform distortion characterized by a non-planar deformation of the director. f^{fl} is minimized if the θ and Φ distortions are 90' out of phase. However, since these variables have to satisfy the boundary conditions we get the profiles shown in Fig.6. The director distribution in the deformed state is shown schematically as dashed lines in Fig.7. The dotted line is the projection of the director pattern on the XZ plane and the dashed and dotted line the projection on the XY plane. The variation of the Y component of the flexoelectric polarization $\vec{P}$ is also shown.

Thus it is clear that in geometry 2 the flexoelectric terms do not favour a periodic distortion of the director field. Hence when they are taken into account, the critical value R'_c of R' up to which a periodic distortion is favoured decreases. For $R'>R'_c$, however, flexoelectric-

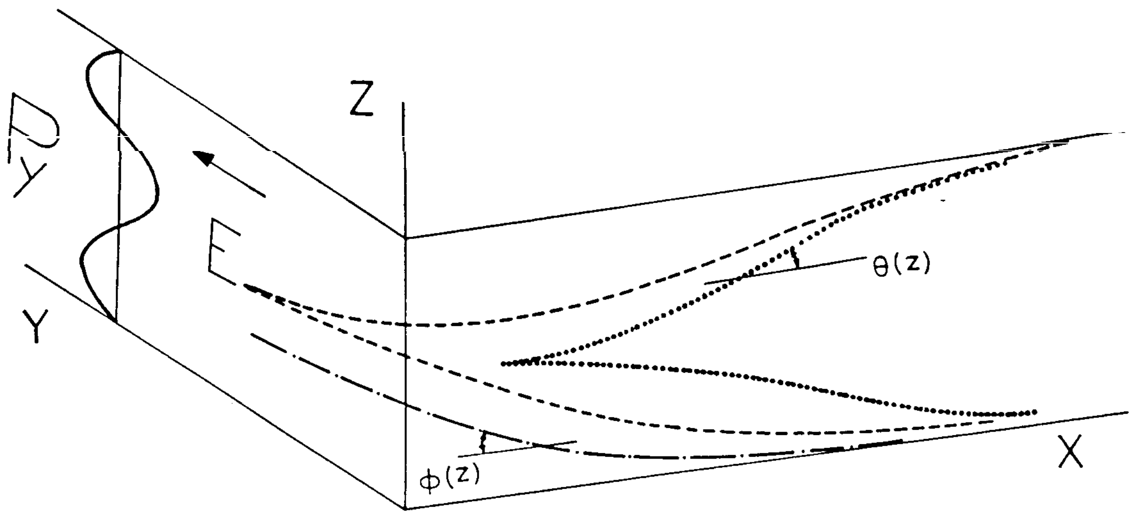

Fig.7. Schematic diagram of the director profile in the sample above the threshold of the new flexoelectric transition (dashed lines) and its projection on the XZ (dotted lines) and XY planes (dashed and dotted line). The undistorted director is along X. The variation of the Y-component of the flexoelectric polarization P across the sample thickness is also shown.

ity gives rise to a new type of transition to a deformed state with nonplanar distortion of the director field characterized by two polar angles θ and Φ.

We have detected the new type of transition in geometry 2 in a nematic mixture with $\epsilon_a \approx 0$, containing CE-1700, CM-5115 and PCH-302 of Roche chemicals. Using a quarter wave plate as compensator, we observe the onset of the θ distortion at a critical value of the applied electric field. At higher fields longitudinal domains are seen. Though such a trend is suggested by Eq. (14) a non-linear analysis is needed to clarify this point. As many of the relevant material parameters of this mixture are not known a comparison with the theory is not possible.

Thus we find that the influence of flexoelectricity on the static distortions in geometries 1 and 2 is very different. In geometry 2 it gives rise to a uniform non-planar director distortion described by two polar angles. This transition has been experimentally detected.

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