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## Liquid Crystals

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## Theory of an electric field induced periodic phase in a nematic film

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The elastic theory of nematic liquid crystals is used to determine whether it gives support for recent experimental observations of a novel periodic phase occurring in homeotropically aligned films when an electric field is applied in the plane of the film. The director remains in the plane defined by the film normal and the field, and the wavevector of the periodicity is parallel to the field. It is found that the theory is consistent with experiment.

Although static periodic director configurations have been known to be induced in thin nematic films by applied magnetic fields for many years [1], new phenomena continue to be discovered. In particular, Frisken and Palffy-Muhoray [2] have recently reported the observation of a hysteretic (first order) phase transition from an undistorted homeotropically aligned nematic film to a periodic state. An electric field ( $\mathbf{E}$ ) was applied in the plane of the film, and a magnetic field ( $\mathbf{H}$ ) perpendicular to the plane of the film (parallel to the initial director alignment). Since both the magnetic and dielectric susceptibility tensors of the sample had positive anisotropy, the magnetic field tended to stabilize the configuration while the electric field favoured a realignment parallel to  $\mathbf{E}$ . The magnetic field improved the homogeneity of the periodicity, but was not essential and will be ignored in all further discussion. The most striking feature of the periodic phase, beside the fact that the phase transition was first order, was that the wavevector of the modulation was parallel to  $\mathbf{E}$ . In this paper, using instability analysis, we show that the elastic theory of liquid crystals [3], coupled with applied field terms, gives theoretical support to these experimental observations.

Before beginning the analysis, a brief review will serve to point out the similarities as well as the differences between the periodic phase of interest here and other known periodic configurations. There are six experimental situations to consider: the splay Freedericksz geometry having homogeneous planar director alignment with either  $\mathbf{H}$  or  $\mathbf{E}$  normal to the film; the twist Freedericksz geometry in which there is again planar homogeneous alignment but  $\mathbf{H}$  or  $\mathbf{E}$  is in the plane of the film and normal to the director; and finally the bend Freedericksz geometry where there is homeotropic alignment and  $\mathbf{H}$  or  $\mathbf{E}$  is in the plane of the film. The physics is potentially different for  $\mathbf{E}$  and  $\mathbf{H}$  because local field effects [4, 5] are important for  $\mathbf{E}$ , but not for  $\mathbf{H}$ . Periodic phases have been observed for applied magnetic fields in the bend and splay geometries under appropriate conditions [1, 6, 7]. Theoretical explanations have also been given [7-10]. In each of these cases the director configuration of the periodic phase has a component coming *out of the plane* defined by the field and the initial surface

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alignment. Large differences among the splay, twist and bend elastic constants ( $K_1$ ,  $K_2$  and  $K_3$ ) are necessary in order for this out-of-plane component to be favourable. In contrast, for the new phenomena observed in [2], the director configuration remains in the field-initial alignment plane and the periodic phase may apparently occur for the relatively small differences between  $K_1$  and  $K_3$  ( $K_1/K_3 \lesssim 1$ ) which are typical of thermotropic nematics.

No attempt is made here to derive the exact solution from the Euler–Lagrange equation, though this would prove rigorously that the observed structure indeed follows from theory. Instead, we pursue the much more modest goal of demonstrating that a model form of the periodic configuration has lower free energy than the non-periodic Freedericksz distortion.

The analysis of the bend geometry case with  $\mathbf{E}$  in the film plane starts by expressing the director as

$$\hat{\mathbf{n}} = \cos \phi \hat{\mathbf{z}} + \sin \phi \hat{\mathbf{x}}, \quad (1)$$

where  $\hat{\mathbf{z}}$  is the film normal and  $\hat{\mathbf{x}}$  is the direction of the applied electric potential gradient. The free energy density (in units of  $K_3(\pi/d)^2$ , where  $d$  is the film thickness) is given by

$$F = \frac{1}{2} \int d^3 \mathbf{r} [k(\text{div } \hat{\mathbf{n}})^2 + (\hat{\mathbf{n}} \times \text{curl } \hat{\mathbf{n}})^2 - e^2(1 - p \sin^2 \phi)]. \quad (2)$$

Here  $k \equiv K_1/K_3$ ,  $p \equiv (\varepsilon_{\parallel} - \varepsilon_{\perp})/\varepsilon_{\parallel}$ ,  $e^2 = E_0^2 \varepsilon_{\perp}/K_3(\pi/d)^2$ ,  $E_0$  is the potential difference per unit length applied in the film plane, and the coordinates  $\mathbf{r} = (x, y, z)$  are dimensionless because they have been scaled in units of  $d/\pi$ . The principal values of the dielectric tensor are  $\varepsilon_{\parallel}$  and  $\varepsilon_{\perp}$ . An  $(\hat{\mathbf{n}} \cdot \text{curl } \hat{\mathbf{n}})$  term does not appear, because the model assumes no  $y$  dependence. The last term has been discussed by Arakelyan *et al.* [5]; it differs from  $e^2 p \sin^2 \phi$  because of local field effects.

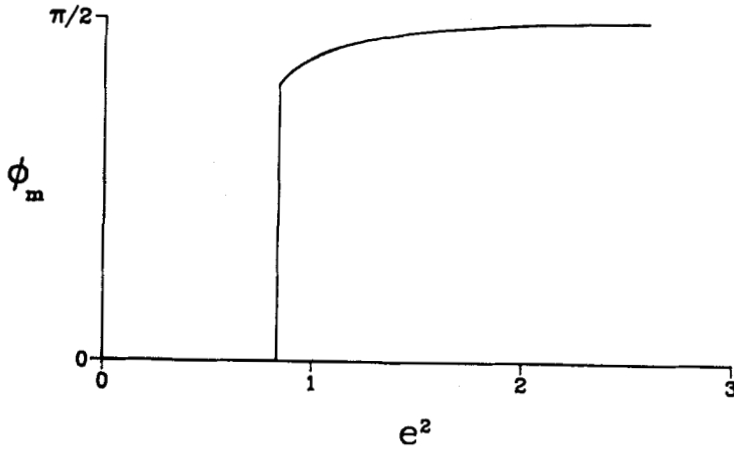
Three different forms of the function  $\phi$  are then considered where each form corresponds to one of the three anticipated states of the system: (a)  $\phi = 0$  for the undistorted uniformly oriented nematic; (b)  $\phi = \phi_0(z)$  for the non-periodic Freedericksz state; and (c)  $\phi = \phi_0(z) + \theta(x, z)$  for the periodic configuration. The form of  $\phi$  which gives the lowest free energy at given  $k$ ,  $p$  and  $e^2$  is then expected to be stable.

The non-periodic Freedericksz state has a director configuration depending only on  $z$ ;  $\phi = \phi_0(z)$ . Frisken and Palffy-Muhoray [2] minimized equation (2) with respect to the function  $\phi_0$  (ignoring the periodic state), and their results are summarized qualitatively in the figure.  $\phi_m$  is the angle  $\phi$  at  $z$  midway between the film boundary plates, and it undergoes a discontinuous jump at a critical value of  $e^2$ .  $\phi_0$  can be calculated in terms of elliptic integrals, but may also be approximated by the first Fourier component,  $\phi_0 = \phi_m \cos z$ , where the film boundary is at  $z = \pm \pi/2$ .

Finally the form of  $\phi$  for the periodic state adds a perturbation,  $\theta$ , to  $\phi_0$ ,

$$\begin{aligned} \phi &= \phi_0(z) + \theta(x, z), \\ \theta(x, z) &= \theta_0 \left[ \cos(z) \cos(Qx) + \frac{N}{2} \sin(2z) \sin(Qx) \right]. \end{aligned}$$

The amplitude  $\theta_0$  is assumed to be small, and  $N$  and the dimensionless wavenumber  $Q$  are variational parameters. This form was chosen to produce terms in equation (2) proportional to  $Q$  and  $Q^2$  so that the possibility of  $Q$  not equal to zero occurs. This form has *not* been optimized and serves only to establish that  $\phi_0$  may be less stable



A schematic plot of  $\phi_m$ , the distortion angle at the centre of the film, versus  $e^2$ , the scaled field, showing a first order Fredericksz transition at  $e^2 < 1$ , where a second order transition would occur.

than  $\phi_0 + \theta$ . Equation (2) is expanded in powers of  $\theta_0$ , and the spatial integrals carried out (using  $\phi_0 \approx \phi_m \cos z$ ), giving

$$F = F(\phi_0) + \frac{\theta_0^2}{4} \left[ -e^2 p(\alpha + \beta N^2) + \frac{1}{2}(1 + N^2) - (1 - k)(\gamma + \delta N^2) - (1 - k) \frac{NQ}{4} \eta + Q^2 \left( \frac{\mu(1 - k)}{8} + \frac{k}{2} \right) + Q^2 N^2 \left( \frac{\nu(1 - k)}{8} + \frac{k}{8} \right) \right], \quad (3)$$

where the parameters  $\alpha, \beta, \delta, \eta, \mu$  and  $\nu$  are

$$\begin{aligned} c_1 &\equiv (3p - 1); & c_2 &\equiv (15p^2 - 10p + \frac{2}{3}); & c_3 &\equiv (7p^3 - 7p^2 + 7p/5 - 1/45), \\ \alpha &\equiv \frac{1}{2} + \frac{3}{4}c_1\phi_m^2 + \frac{5}{16}c_2\phi_m^4 + \frac{35}{32}c_3\phi_m^6, \\ \beta &\equiv \frac{1}{8} + \frac{1}{8}c_1\phi_m^2 + \frac{5}{128}c_2\phi_m^4 + \frac{7}{64}c_3\phi_m^6, \\ \gamma &\equiv \frac{3}{4}\phi_m^2 - \frac{5}{16}\phi_m^4 + \frac{7}{144}\phi_m^6, \\ \delta &\equiv \frac{5}{16}\phi_m^2 - \frac{5}{64}\phi_m^4 + (\frac{7}{144})(\frac{7}{40})\phi_m^6, \\ \eta &\equiv 3\phi_m - \frac{5}{3}\phi_m^3 + \frac{7}{24}\phi_m^5, \\ \mu &\equiv 3\phi_m^2 - \frac{5}{6}\phi_m^4 + \frac{7}{72}\phi_m^6, \\ \nu &\equiv \frac{1}{2}\phi_m^2 - \frac{5}{48}\phi_m^4 + \frac{7}{720}\phi_m^6. \end{aligned}$$

Here  $\phi_m$  is treated as a known function of  $e^2$  (see the figure). At values of  $e^2$  such that the coefficient of  $\theta_0^2$  in equation (3) is negative, the  $\phi = \phi_0 + \theta$  solution is more stable than  $\phi_0$ .  $F$  is minimized with respect to  $Q$  and  $N$ .

A typical result that then follows is that for  $k = 1/2$ , the coefficient is negative for  $0.682 < p < 0.688$  over a range of  $e^2$  extending above and below the critical  $e^2$  at which  $\phi_m$  undergoes its transition. This means that for  $K_1/K_3 \approx 1/2$  there is a narrow range of dielectric anisotropy such that the periodic phase will occur at the onset of the distortion, and that at a higher field the periodic phase will revert to the  $\phi_0(z)$  Fredericksz state. Similar results occur for  $0 < k < 0.8$ . The limited range of

dielectric anisotropy  $p$  for given  $k$  is not a serious restriction, because better trial functions can be expected to improve the stability of the periodic phase.

In conclusion, these theoretical results show that for a film in the bend geometry with an electric field in the film plane, the non-periodic Fredericksz state may be unstable against the formation of a periodic state having a wavevector parallel to the field. The theory has not been used to achieve quantitative accuracy, but to verify that a theoretical explanation, qualitatively consistent with experiment, is possible. Quantitative work is under way.

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