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B. J. Frisken[a b]; P. Palffy-muhoray[a]
[a] Liquid Crystal Institute, Kent State University, Kent, Ohio, U.S.A. [b] Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada

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Effects of a transverse electric field in nematics: induced biaxiality and the bend Fréedericksz transition

by B. J. FRISKEN† and P. PALFFY-MUHORAY
Liquid Crystal Institute, Kent State University, Kent, Ohio 44242, U.S.A.

We have studied the effects of a transverse electric field on director fluctuations in the nematic liquid crystal 5CB (4-n-pentyl-4′-cyanobiphenyl) in the bend Fréedericksz geometry. The sample was homeotropically aligned by surface treatment of the glass cell walls and an additional magnetic field was applied perpendicular to the walls. An electric field was then applied parallel to the walls; below the bend Fréedericksz transition, director fluctuations parallel to the electric field are enhanced. This field-induced biaxiality was observed and studied by monitoring the intensity of light transmitted by the sample placed between crossed polarizers. Landau theory for 5CB predicts the electric field induced bend transition to be first order. Our observations of the transmitted intensity are consistent with this prediction. We have also observed that this transition is to a modulated rather than to a uniform phase.

1. Introduction

In a simple continuum model, nematic liquid crystals are characterized by a director field which defines the local principal axis of a second rank tensor property. Thermal fluctuations of this director field are opposed by elastic torques and may be influenced by external magnetic and electric fields (for a recent review, see [1]). These fields couple to the director fluctuations via the anisotropic susceptibilities; for a material of positive susceptibility anisotropy a field along the director will quench the fluctuations, whereas one applied perpendicular to the director will enhance fluctuations parallel to the field. In the latter case the uniaxial symmetry of the system is broken and the sample becomes biaxial. For a material of negative susceptibility anisotropy a field applied along the director will enhance the fluctuations in the plane perpendicular to the director, whereas one applied perpendicular to the director will enhance fluctuations in the direction which is perpendicular to both the director and the applied field, breaking the uniaxial symmetry. Recently, field-induced biaxiality has been studied in materials of both negative diamagnetic anisotropy [2] and negative dielectric anisotropy [3].

A nematic liquid crystal, aligned by surface torques, can be reoriented by an external field when the field is sufficiently strong to overcome the bulk elastic torques [4]. In the case of reorientation by a magnetic field this Fréedericksz transition is generally second order [5]. Second order transitions are characterized by the divergence of fluctuations; however, director fluctuations in the vicinity of the Fréedericksz transition have not received much attention. Two recent works [6, 7] report observations of these critical fluctuations but no attempt was made to characterize the nature of the divergence.

† Permanent address: Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 2A6.

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We have studied the nematic liquid crystal 5CB (4-*n*-pentyl-4′-cyanobiphenyl) in the geometry of the electric field-induced bend transition. In this geometry the sample is aligned homeotropically so that the director is perpendicular to the cell windows. In addition, we have applied a D.C. magnetic field perpendicular to the windows. An electric field is applied parallel to the cell windows, and if sufficiently intense, will give rise to a bend deformation.

First we describe the effect of the competing electric and magnetic fields on thermal fluctuations of the director, using simple continuum theory which is appropriate for small deformations. These anisotropic director fluctuations give rise to a bulk biaxiality of the sample. We then use Landau theory to describe the Fréedericksz transition in this geometry, deriving an expression for the divergence of the fluctuations at the transition. Landau theory indicates that the transition for 5CB in this geometry should be first order. Information about biaxiality due to anisotropic director fluctuations is obtained from measurements of intensity of light transmitted by the sample between crossed polarizers. The results of these measurements are compared with the predictions of theory.

2. Theory

The direction of the average orientation of the symmetry axis of the molecules in a nematic is described by the director field $\hat{\mathbf{n}} = (n_x, n_y, n_z)$. We consider a sample with its average director along $\hat{\mathbf{z}}$. Thermal fluctuations correspond to non-zero x and y components of the director. First we consider the effect of fields below the Fréedericksz transition in the regime where the fluctuations are small and $n_z \gg n_x, n_y$. In the continuum model the free energy of the system due to elastic and external field effects is [5]

$$F = \frac{1}{2}\int_v d^3\mathbf{r}\,[K_1(\nabla\cdot\hat{\mathbf{n}})^2 + K_2(\hat{\mathbf{n}}\cdot\nabla\times\hat{\mathbf{n}})^2 + K_3(\hat{\mathbf{n}}\times\nabla\times\hat{\mathbf{n}})^2 - \mathbf{D}\cdot\mathbf{E} - \mathbf{B}\cdot\mathbf{H}], \tag{1}$$

where $\hat{\mathbf{n}} = (\cos\phi\sin\theta,\ \sin\phi\sin\theta,\ \cos\theta)$, ϕ and θ being the azimuthal and polar angles describing the orientation of the director in the laboratory frame, K_1, K_2 and K_3 are the splay, twist and bend elastic constants, respectively, v is the sample volume, $\mathbf{D}$ and $\mathbf{E}$ are the electric displacement and field, and $\mathbf{B}$ and $\mathbf{H}$ are the magnetic induction and field. The orientation dependent part of the magnetic field term is

$$F_B = -\tfrac{1}{2}\chi_a B^2 n_z^2/\mu_0, \tag{2}$$

where χ_a is the diamagnetic anisotropy. Following [8], we obtain the electric contribution to the free energy density

$$F_E = -\frac{1}{2}\frac{\varepsilon_0\varepsilon_\perp E_x^2}{1 - un_x^2}, \tag{3}$$

where $u = (\varepsilon_\| - \varepsilon_\perp)/\varepsilon_\|$, $\varepsilon_\|$ and $\varepsilon_\perp$ being the principal components of the dielectric tensor, and $E_x = V/d$, d being the width of the cell along $\hat{\mathbf{x}}$. For small fluctuations

$$F_E \approx -\tfrac{1}{2}\varepsilon_0\varepsilon_\perp uE_x^2 n_x^2. \tag{4}$$

The spatial variation of the director can be expressed in terms of Fourier components [5]

$$\left.\begin{aligned} n_\alpha(\mathbf{q}) &= \frac{1}{v}\int_1 n_\alpha(\mathbf{r})\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{r})\,d^3\mathbf{r}, \\ n_\alpha(\mathbf{r}) &= \frac{1}{(2\pi)^3}\int n_\alpha(\mathbf{q})\exp(-\mathrm{i}\mathbf{q}\cdot\mathbf{r})\,d^3\mathbf{q}. \end{aligned}\right\} \quad (5)$$

Assuming, for simplicity, that the elastic constants are all equal, the component of the mean squared amplitude of the normal mode of the director fluctuations perpendicular to **B** and (*a*) along **E** is

$$\langle n_x^2(\mathbf{q})\rangle = \frac{kT}{2\pi^2}\frac{1}{Kq^2 + \chi_a B^2/\mu_0 - \varepsilon_0\varepsilon_\perp uE^2} \quad (6a)$$

and (*b*) perpendicular to **E** is

$$\langle n_y^2(\mathbf{q})\rangle = \frac{kT}{2\pi^2}\frac{1}{Kq^2 + \chi_a B^2/\mu_0}, \quad (6b)$$

where K is the average elastic constant and q is the amplitude of the wavevector.

Thus, the electric field breaks the rotational symmetry of the sample by enhancing director fluctuations in the x direction. The effect of the fluctuations on the intensity of light transmitted through the sample can be calculated in two ways. The dielectric tensor is given by

$$\varepsilon_{\alpha\beta} = \varepsilon_\perp\delta_{\alpha\beta} + (\varepsilon_\parallel - \varepsilon_\perp)n_\alpha n_\beta. \quad (7)$$

The bulk dielectric tensor is calculated by averaging over the fluctuations of the local order in the sample, so a difference in the mean square amplitudes of the modes leads to an induced birefringence for light propagating along the magnetic field. Alternatively, the intensity can be calculated from light scattering theory as the scattering of light is governed by fluctuations of the dielectric tensor. With the sample between crossed polarizers at 45° to **E** the transmitted intensity is given by

$$I \propto \left[\int_{q_{min}}^{q_{max}} (\langle n_x^2(\mathbf{q})\rangle - \langle n_y^2(\mathbf{q})\rangle)\,d^3\mathbf{q}\right]^2. \quad (8)$$

The integral extends over a volume in q space bounded by q_{min} and q_{max}. q_{min} is fixed by the sample dimension, and may be taken to be zero for a macroscopic sample. q_{max} can be related either to the distance scale that defines local order, or to a fixed number of fluctuation modes [1]. Both result in a cut-off wavelength on the order of the intermolecular distance. Fluctuations in the direction of the applied electric field increase as the electric field strength increases, and equation (6) predicts a divergence of the voltage at $V_R = Bd\,(\chi_a/\mu_0\varepsilon_0\varepsilon_\perp u)^{1/2}$. Since the derivation of equation (6) assumes small fluctuations, we do not expect it to provide a good description of the behaviour near the transition.

To investigate the behaviour in this region, we consider distortions in the plane of the field only, i.e. $\hat{\mathbf{n}} = (\sin\theta, 0, \cos\theta)$ and assume a director field of the form

$$\theta = \theta_m \sin\left(\frac{\pi z}{l}\right), \quad (9)$$

where l is the thickness of the cell along $\hat{\mathbf{z}}$, and θ_m is the maximum deformation angle ($0 < \theta_m < \pi/2$) and may be regarded as an order parameter. To investigate the order of the transition, the free energy is expanded to sixth order in terms of the maximum deformation angle, to obtain the dimensionless Landau expansion

$$\mathscr{F} = a\theta_m^2 + \frac{b}{2}\theta_m^4 + \frac{c}{3}\theta_m^6, \tag{10}$$

where

$$\left.\begin{aligned} a &= 1 - (1/\pi^2)(e - h), \\ b &= \tfrac{1}{2}[-\kappa - (1/\pi^2)(h - e + 3eu)], \\ c &= \tfrac{1}{8}[\kappa + (1/\pi^2)(\tfrac{2}{3}h - 15eu^2 + 10eu - \tfrac{2}{3}e)], \end{aligned}\right\} \tag{11}$$

with

$$\left.\begin{aligned} \kappa &= (K_3 - K_1)/K_3, \\ e &= (\varepsilon_0\varepsilon_\perp uV^2/K_3)(l/d)^2 = \pi^2V^2/V_0^2, \\ h &= \chi_a l^2 B^2/\mu_0 K_3 = \pi^2 B^2/B_0^2. \end{aligned}\right\} \tag{12}$$

Minimizing $\mathscr{F}$ with respect to θ_m results in an expression for θ_m in terms of the coefficients of the free energy expression:

$$\theta_m^2 = \frac{-b \pm (b^2 - 4ac)^{1/2}}{2c} \tag{13}$$

The transition is second order for $b, c > 0$, and occurs when $a = 0$ at a voltage $V = V^*$. The transition is first order if $b < 0$ and $c > 0$, and occurs at a voltage $V = V_{th}$ given by $a = 3b^2/16c$. At the first order transition the value of the order parameter is given by

$$\theta_m = (-3b/4c)^{1/2}. \tag{14}$$

Below the transition $\langle\theta_m\rangle = 0$. Fluctuations in θ_m are given by [9]

$$\begin{aligned} \langle\theta_m^2\rangle &= kT\left(\frac{\partial^2\mathscr{F}}{\partial\theta_m^2}\right)^{-1}\Bigg|_{\theta_m=0} \\ &= \frac{kT}{4lK_3}\frac{V_0^2}{V^{*2}}\left(1 - \frac{V^2}{V^{*2}}\right)^{-1}. \end{aligned} \tag{15}$$

If the transition is second order, fluctuations in θ_m (and hence n_x) are expected to diverge as $V \to V^*$. If the transition is first order, the fluctuations are expected to increase until $V = V_{th}$, further increase is pre-empted by a finite bulk reorientation associated with the first order transition. Using material constants for 5CB [10], we find that the transition is expected to be first order. Figure 1 shows θ_m and $\mathscr{F}$ as a function of the inverse of the voltage parameter, e, for several values of the magnetic field parameter, h. Although this Landau theory is not exact, since it neglects higher order Fourier components of the deformation (and the transition is first order), exact calculations confirm these results [11]. There has been a great deal of interest recently in first order Fréedericksz transitions. These have been predicted and observed in several other systems: in systems of large conductivity anisotropy [12], in systems

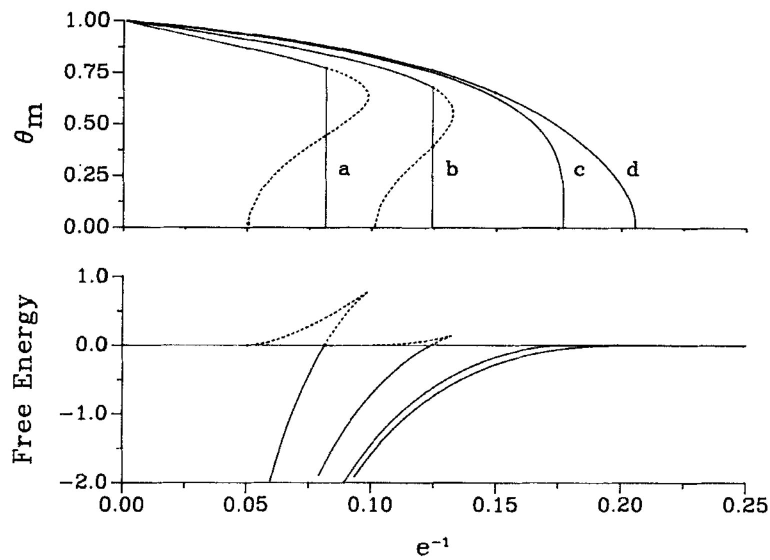

Figure 1. Calculated values of the order parameter and the dimensionless free energy for the electric field-induced bend transition as a function of $1/e$ for (*a*) $h = 10$, (*b*) $h = 0{\cdot}0$, (*c*) $h = -4{\cdot}211$, and (*d*) $h = -5{\cdot}0$, $e = \varepsilon_0\varepsilon_\perp uV^2(l/d)^2/K_3$ and $h = \chi_a B^2/(\mu_0 K_3)$. The order parameter is in reduced units.

where there are two external fields, both perpendicular to the initial director orientation at the boundaries for a limited range of physical constants [13, 14], in systems with particular physical constants where the transition is induced by optical fields [15–17] and with more general physical constants when the transition is induced by optical fields in the presence of a stabilizing magnetic or electric field [18–20].

3. Experiment

Figure 2 shows the experimental set-up used to study field effects in the nematic liquid crystal 5CB. The sample cell consists of two glass plates, 3·3 mm × 0·5 mm × 30·0 mm, separated by 0·5 mm and treated with a silane compound (Dow Corning X1-6136; 3-(trimethoxysilyl) propyldimethyloctadecyl ammonium chloride) to induce alignment perpendicular to the plates. The liquid crystal used was supplied by Dr G. S. Bates of the Chemistry Department at the University of British Columbia. A sinusoidal 1 kHz voltage was applied across two stainless steel electrodes epoxied perpendicular to the plates. The sample was placed between the poles of an electromagnet, so that the magnetic field was applied along $\hat{\mathbf{z}}$, in a temperature-controlled housing (temperature stability ± 1 mK). The beam from a 5 mW He–Ne laser was aligned so that it passed through the cell along $\hat{\mathbf{z}}$; polarizers crossed at 45° to $\hat{\mathbf{x}}$ were placed before and after the magnet.

The intensity of light transmitted by the cell increases gradually as the applied voltage is increased until the threshold voltage for the transition is reached, at which point the intensity increases abruptly. Figure 3 shows the intensity of light transmitted through the cell as a function of the applied voltage for a magnetic field of 0·33 T. The sweep rate of the applied voltage was 50 V h^{-1}. The abrupt jump in the transmitted intensity at the threshold voltage is consistent with the predicted first order nature of the transition. The full curve is the prediction of equation (8), where

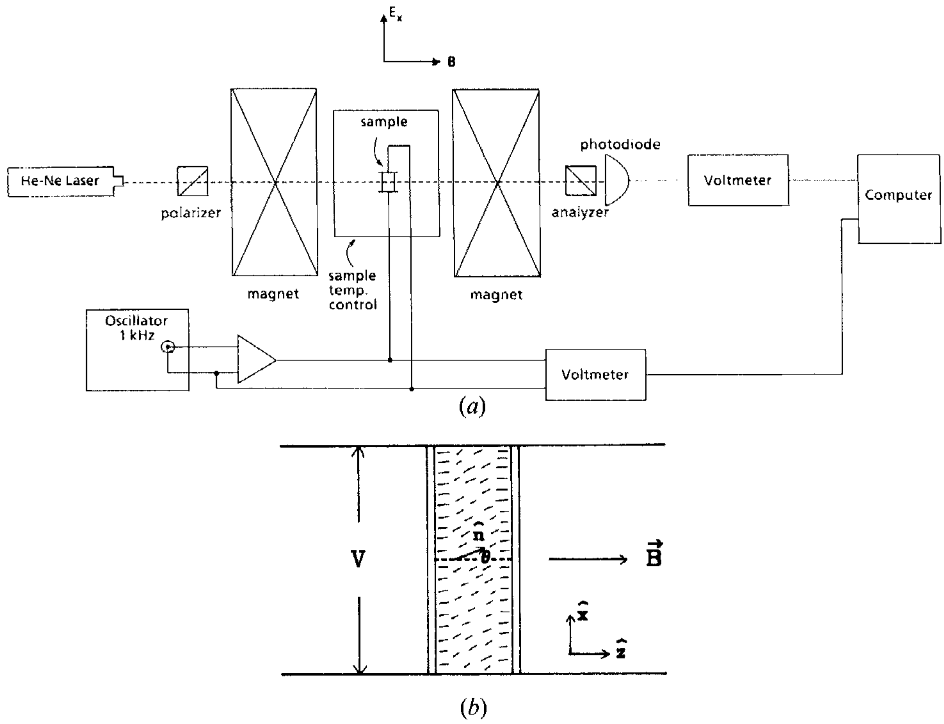

Figure 2. (*a*) Experimental set-up for intensity measurements. (*b*) Cell geometry. The glass walls are treated to give homeotropic alignment (along $\hat{\mathbf{z}}$) of the director. Magnetic field **B** is applied along $\hat{\mathbf{z}}$ and voltage V is applied across the electrodes, which are parallel to $\hat{\mathbf{z}}$.

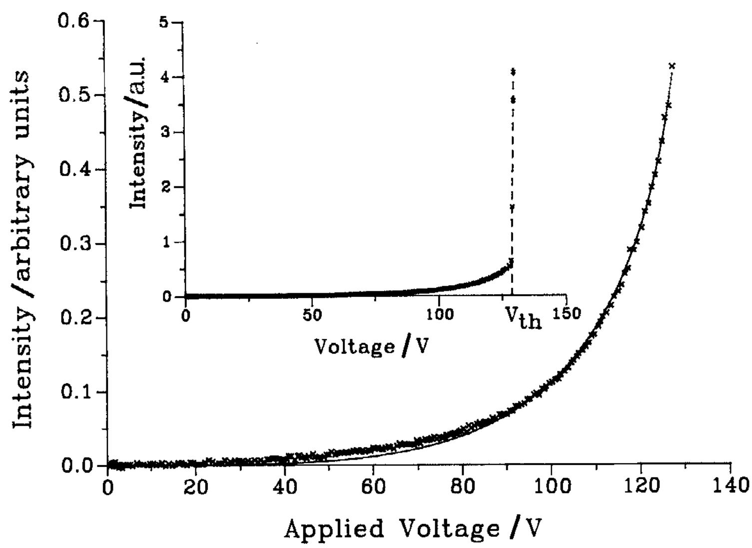

Figure 3. Intensity divergence below the electric field-induced bend Fréedericksz transition. The crosses (x) show experimental data and the full curve is calculated from theory. The inset shows the abrupt increase in the intensity which occurs at the bend Fréedericksz transition.

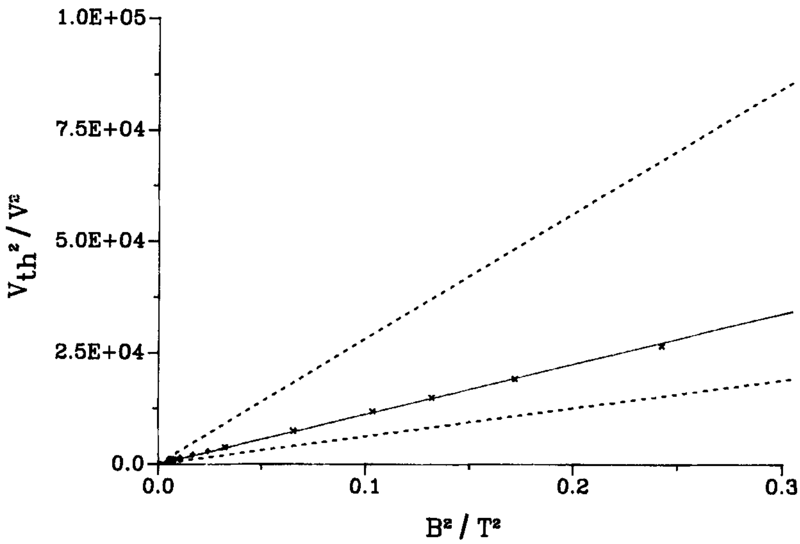

Figure 4. Threshold voltages for the electric field-induced bend transition for increasing magnetic field. The symbols (×, +) show experimental data. The full line is the line of first order transitions and the broken lines are the limits of the spinodal region as calculated from theory.

the proportionality constant has been determined from a least squares fit to the data. Since the value of V_R depends on several material parameters, and since these are not generally known with high absolute accuracy, we have fitted this parameter as well. The fitted value of V_R is 131·1 V, 2·5 V above V_{th}. No critical divergence of the transmitted intensity was observed, which is consistent with the first order nature of the transition. This is further verified by dielectric measurements; the results of these are published elsewhere [11].

The magnitude of the stabilizing magnetic field effects the threshold voltage for this transition. Figure 4 shows results for the threshold voltage measured from a number of runs at different magnetic fields. The voltage was increased at a rate of approximately 10 per cent of the threshold voltage per hour. The solid line indicates the first order transition voltages and the broken lines the stability points as calculated for 5CB using the Landau theory developed here.

When the magnetic field is non-zero, we observe the unexpected result that the transition is to a modulated rather than a uniform phase. Figure 5 shows this modulated phase, viewed along the magnetic field direction ($\hat{\mathbf{z}}$) with a single polarizer parallel to the direction of the electric field ($\hat{\mathbf{x}}$). The wavevector of the stripes is primarily in the direction of the electric field, and we have observed wavelengths ranging from 0·2l to 1·1l in a variety of cells under different conditions. For voltages slightly above the threshold (by about 3–5 per cent) the stripes form slowly (in about 10 min) and persist indefinitely; the periodic structure appears to be a stable deformation. If the voltage is increased, then the stripes disappear. In some respects this pattern resembles that of Williams domains [5], but we have found no compelling evidence of electrohydrodynamic instabilities. Other work [21] shows that, for some values of physical parameters, a stable, periodic deformation minimizes the free energy. Exact calculations for both the electric field-induced bend and the electric

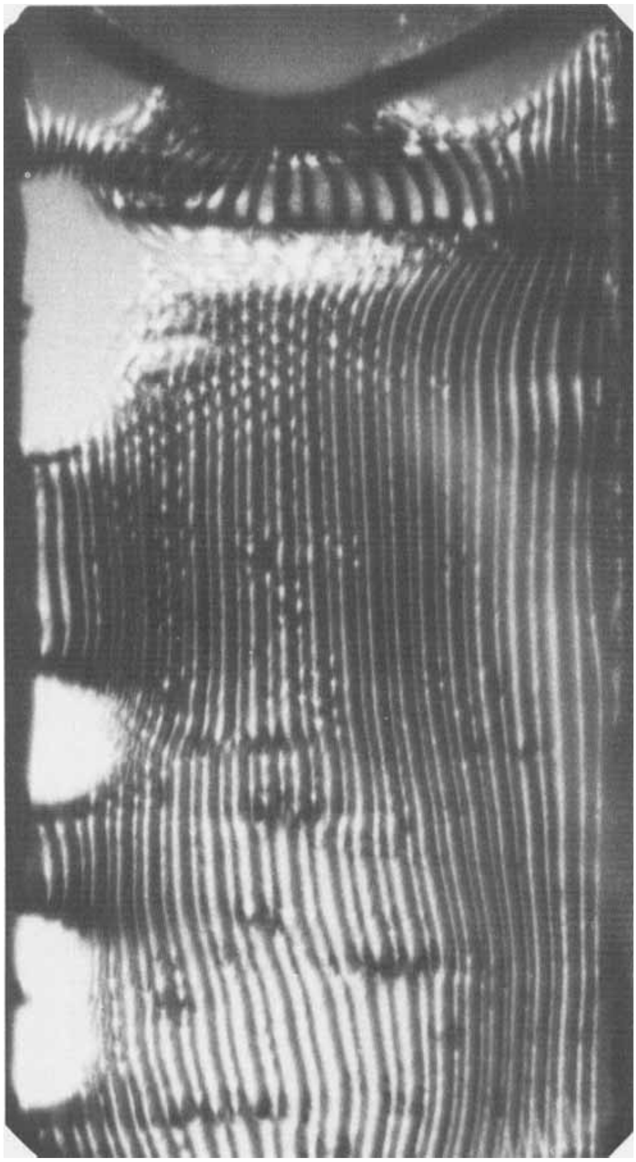

Figure 5. Periodic modulations observed at the electric field-induced bend transition as viewed along the magnetic field. The stripes are parallel to the electrodes and perpendicular to the magnetic field.

field-induced twist Fréedericksz transition as well as the results of dielectric measurements on 5CB are given in [11].

4. Conclusions

We have studied the effect of a transverse electric field on director fluctuations and alignment of the liquid crystal 5CB in the presence of a stabilizing magnetic field. Our geometry is novel since the externally applied electric field is parallel to the cell walls. For fields below the threshold of the Fréedericksz transition, we have observed field-induced biaxiality due to differential quenching of director fluctuations. The behaviour of the induced biaxiality is in good agreement with the predictions of continuum theory. The Fréedericksz transition is first order, in agreement with predictions of Landau theory. We have also found that in the presence of a competing magnetic field, the transition is to a modulated rather than to a uniform phase.

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