Freedericksz transitions in nematic liquid crystals: The effects of an in-plane electric field

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We have studied the effects of perpendicular electric and magnetic fields on a liquid-crystal sample aligned between glass plates. One field is applied to stabilize the initial alignment, and the other is applied in order to drive the transition; the field and initial alignment directions define six possible geometries. We find that, in each case in which an electric field is present parallel to the glass plates, the Freedericksz transition from a uniform to an elastically deformed state can be first order irrespective of whether the electric field drives or stabilizes the transition. Using material parameters characteristic of the liquid-crystal 4-cyano-4'-n-pentylbiphenyl (5CB), we show that two of the transitions should be first order for this material in all values of stabilizing fields and that two should exhibit a tricritical point as the stabilizing field is increased.

INTRODUCTION

A transition from a uniform to an elastically deformed state can occur in an aligned liquid crystal sample between parallel glass plates due to the action of electric or magnetic fields. This transition was first studied by Freedericksz and Zolina.[1] Three different sample geometries can be distinguished, depending on whether the initial distortion of the sample takes the form of a splay, twist, or bend deformation. This transition is generally considered to be second order[2,3] although it has been found to be first order in some instances under the influence of an optical field.[4-10] In a recent paper,[11] we have shown that the electric-field-induced bend and the electric-field-induced twist transitions can be first order due to the novel form of the electric field contribution to the free energy if the electric field is applied parallel to the glass plates. In these cases we considered the Freedericksz transition in the presence of both electric and magnetic fields, with one field applied parallel to the direction of initial alignment to stabilize the alignment and one applied perpendicular to the direction of initial alignment to drive the transition.

If we consider Freedericksz transitions involving static fields and materials of positive susceptibility anisotropies, the transition can be induced by either a magnetic or an electric field perpendicular to the direction of initial alignment and there are, in fact, six possible geometries which should be considered to complete this picture. Here we would like to present the results of Landau expansions of the free energies associated with each geometry to show the qualitative behavior of these systems.

THEORY

The six geometries under study are shown in Fig. 1. Initial alignment is either parallel or perpendicular to the glass plates and two fields are applied to the sample. The director $\hat{n}$ describes the direction of the eigenvector of

the dielectric permittivity tensor associated with the largest eigenvalue. The director is assumed to be confined to the plane of the two fields so that it can be expressed in terms of only one angle, the angle between the director and the direction of alignment at the glass cell walls. (This angle will be called θ for splay and bend transitions
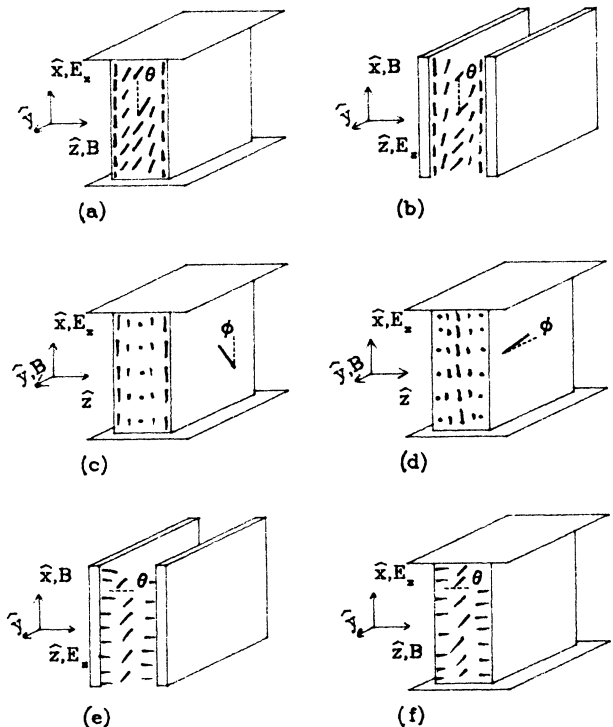

FIG. 1. Diagrams of the six geometries considered for Freedericksz transitions. (a) Magnetic-field-induced splay, (b) electric-field-induced splay, (c) magnetic-field-induced twist, (d) electric-field-induced twist, (e) magnetic-field-induced bend, and (f) electric-field-induced bend.

and ϕ for twist transitions. In the case of twist, θ should be replaced by ϕ in all of the following expressions.) It is assumed in all six cases that the distortion depends only on the coordinate z. The cell area in the x-y plane is A, the thickness (along $\hat{z}$) is l and the width (along $\hat{x}$) is d. At the center of the cell, $(z = l/2)$ the deformation angle is a maximum $(\theta = \theta_m$ or $\phi = \phi_m)$.

The total free energy of the system contains three terms associated with elastic distortions and with interaction of the sample with the two fields; that is,

$$F = F_f + F_e + F_m . \tag{1}$$

F_f is the Frank free energy[12] given by

$$F_f = \tfrac{1}{2} \int [K_1(\nabla \cdot \hat{n})^2 + K_2(\hat{n} \cdot \nabla \times \hat{n})^2$$
$$+ K_3(\hat{n} \times \nabla \times \hat{n})^2] d\mathbf{r} . \tag{2}$$

K_1, K_2, and K_3 are the splay, twist, and bend elastic constants of the liquid crystal and the integral is over the sample volume.

Because of the large difference in the magnitudes of the elastic and magnetic susceptibilities of liquid crystals, the two field energy terms have different forms. In the case of the magnetic field, the diamagnetic susceptibility is small $(\chi \simeq 10^{-7})$ so that the sample does not significantly perturb the applied field. In the case of the electric field, both the permittivity and the permittivity anisotropy are large $(\epsilon \simeq \Delta\epsilon \simeq 10\epsilon_0)$ so that the electric field and the displacement can be significantly altered by the presence of the sample. The free energy due to the magnetic field is given by

$$F_m = -\tfrac{1}{2} \int \mathbf{B} \cdot \mathbf{H} \, d\mathbf{r} . \tag{3}$$

The orientation-dependent free energy associated with the magnetic field can be written[13] as

$$F_m = -\tfrac{1}{2} \frac{\Delta\chi}{\mu_0} \int (\mathbf{B} \cdot \hat{n})^2 d\mathbf{r} , \tag{4}$$

where μ_0 is the permeability of free space, $\mathbf{B} \simeq \mu_0 \mathbf{H}$, and $\Delta\chi$ is the anisotropy of the diamagnetic susceptibility.

The free energy due to the electric field is of the form

$$F_e = -\tfrac{1}{2} \int \mathbf{D} \cdot \mathbf{E} \, d\mathbf{r} . \tag{5}$$

The application of a voltage across the sample results in an electric displacement $\mathbf{D}$,

$$D_\alpha = \epsilon_0 \epsilon_\perp E_\alpha + \epsilon_0(\epsilon_\parallel - \epsilon_\perp) n_\alpha n_\beta E_\beta , \tag{6}$$

where ϵ_0 is the permittivity of free space and $\epsilon_\perp$ and $\epsilon_\parallel$ are the principal values of the bulk dielectric susceptibility tensor, transverse and parallel to the director. The sample volume is enclosed by one pair of conducting surfaces across which the voltage is applied, and two pairs of nonconducting surfaces. Assuming that $\mathbf{D}$ and $\mathbf{E}$ vary only in the $\hat{z}$ direction, i.e., the direction perpendicular to the glass plates which enclose the sample, $\nabla \cdot \mathbf{D} = 0$ implies that D_z is a constant. The requirement that $\nabla \times \mathbf{E} = 0$ implies that $\mathbf{E}$ can be expressed as the gradient of some potential U. When the voltage is applied across the glass plates, E_z is the only nonzero component of $\mathbf{E}$ as $U = U(z)$. The free energy due to the field is then[2,3]

$$F_e = -\frac{A}{2} \int_0^l D_z E_z \, dz = -\frac{A}{2} D_z \int_0^l E_z \, dz = -\frac{A}{2} D_z V . \tag{7}$$

D_z can be found from the constitutive relation [Eq. (6)]. In this case, the electric field term does not depend on the thickness l of the sample. When the voltage is applied to conductors perpendicular to the glass plates, D_z is zero if there are no free charges. The surviving term in the free energy is given by

$$F_e = -\frac{A}{2} \int_0^l D_x E_x \, dz . \tag{8}$$

Here E_x is a constant and is equal to V/d and D_x is a function of z. It is interesting to note that the electric field term is now dependent on the ratio of l/d. This result was first obtained by Arakelyan, Karayan, and Chilingaryan[14] to describe the free energy of the electric-field-induced bend transition. The detailed forms of these expressions for specific geometries are given in Table I.

TABLE I. Free energy terms for geometries (a)–(f). The geometries are shown in Fig. 1. $u = \Delta\epsilon/\epsilon_\parallel$, $w = \Delta\epsilon/\epsilon_\perp$, $\kappa = 1 - K_1/K_3$, $\gamma = 1 - K_3/K_1$, $h = \Delta\chi B^2 l^2/\mu_0 K_i \pi^2$, and the free energies are expressed in units of $\pi^2 AK_i/4l$, where $i = 1$ for geometries (a) and (b), $i = 2$ for geometries (c) and (d), and $i = 3$ for geometries (e) and (f).

Geometry	$\hat{n}$	F_f	F_m	F_e	e
(a)	$(\cos\theta, 0, \sin\theta)$	$\int (1 - \gamma \sin^2\theta)\theta'^2$	$-\int h \sin^2\theta$	$-\int \dfrac{e/w}{1 + w \sin^2\theta}$	$\dfrac{\epsilon_0 w V^2 l^2 \epsilon_\parallel}{K_1 \pi^2 d^2}$
(b)	$(\cos\theta, 0, \sin\theta)$	$\int (1 - \gamma \sin^2\theta)\theta'^2$	$-\int h \cos^2\theta$	$-\dfrac{e/w}{\int \dfrac{1}{1 + w \sin^2\theta}}$	$\dfrac{\epsilon_0 \Delta\epsilon V^2}{K_1}$
(c)	$(\cos\phi, \sin\phi, 0)$	$\int \phi'^2$	$-\int h \sin^2\phi$	$-\int \dfrac{e/w}{1 + w \sin^2\theta}$	$\dfrac{\epsilon_0 w V^2 l^2 \epsilon_\parallel}{K_2 \pi^2 d^2}$
(d)	$(\sin\phi, \cos\phi, 0)$	$\int \phi'^2$	$-\int h \cos^2\phi$	$-\int \dfrac{e/u}{1 - u \sin^2\phi}$	$\dfrac{\epsilon_0 u V^2 l^2 \epsilon_\perp}{K_2 \pi^2 d^2}$
(e)	$(\sin\theta, 0, \cos\theta)$	$\int (1 - \kappa \sin^2\theta)\theta'^2$	$-\int h \sin^2\theta$	$-\dfrac{e/u}{\int \dfrac{1}{1 - u \sin^2\theta}}$	$\dfrac{\epsilon_0 \Delta\epsilon V^2}{K_3}$
(f)	$(\sin\theta, 0, \cos\theta)$	$\int (1 - \kappa \sin^2\theta)\theta'^2$	$-\int h \cos^2\theta$	$-\int \dfrac{e/u}{1 - u \sin^2\theta}$	$\dfrac{\epsilon_0 u V^2 l^2 \epsilon_\perp}{K_3 \pi^2 d^2}$

A Landau free energy can be constructed by assuming a deformation of the director field of the form

$$\theta = \theta_m \sin\frac{\pi z}{l} \tag{9}$$

and expanding the free energy F in terms of the order parameter θ_m. In order to investigate whether the transition is first or second order, the expansion must include powers of θ_m up to sixth order. In this way, the dimensionless Landau expansion obtained is

$$\mathcal{F} = \mathcal{A}\,\theta_m^2 + \frac{\mathcal{B}}{2}\theta_m^4 + \frac{\mathcal{C}}{3}\theta_m^6 \ , \tag{10}$$

where the free energy units and the coefficients are given explicitly in Table II for the six cases considered. Minimizing $\mathcal{F}$ with respect to θ_m results in three solutions for θ_m, whether or not they have physical meaning depends on the parameters of the system. Minimizing $\mathcal{F}$ yields an expression for θ_m in terms of the coefficients of the free energy expression,

$$\theta_m^2 = \frac{-\mathcal{B}\pm[\mathcal{B}^2 - 4\mathcal{A}\,\mathcal{C}]^{1/2}}{2\mathcal{C}} \ , \tag{11}$$

and the null solution ($\theta_m = 0$) is always an extremum. The transition is second order for $\mathcal{B}, \mathcal{C} > 0$ and the transition occurs when $\mathcal{A} = 0$. A first order transition will occur for $\mathcal{B} < 0$, $\mathcal{C} > 0$ when $\mathcal{A} = 3\mathcal{B}^2/16\mathcal{C}$. At the first-order transition, the value of the order parameter is given by

$$\theta_m = \sqrt{-3\mathcal{B}/4\mathcal{C}} \ . \tag{12}$$

The third solution corresponds to a local maximum of the free energy and occurs when $\mathcal{A} = \mathcal{B}^2/16\mathcal{C}$.

Using material parameters for 5CB, at room temperatures $\mathcal{B}$ is positive and the magnetic-field-induced bend transition and the electric-field-induced splay transition are second order. $\mathcal{B}$ is negative and the transition is predicted to be first order (1) when the bend transition is induced by an electric field; (2) when the twist transition is induced by a magnetic field in the presence of a sufficiently strong stabilizing electric field, or when the twist transition is induced by an electric field; or (3) when the splay transition is induced by a magnetic field in the presence of a sufficiently strong stabilizing electric field. The tricritical point occurs at $\mathcal{A}, \mathcal{B} = 0$ and should occur in the magnetic-field-induced splay and twist transitions as the stabilizing electric field strength is increased. The results in materials characterized by other material parameters will be different. The general requirements for the transition to be first order are given in Table II.

Several details indicate that the quantitative predictions of Landau theory may not be entirely accurate. The Landau expansion is expected to be most accurate near the tricritical point and less accurate as the transition becomes strongly first order. The value of the order parameter at the first-order transition is large, suggesting that the transition is strongly first order and that Landau theory may not provide a good description. The results are unsatisfactory in other ways; θ_m has values greater than $\pi/2$, which is unphysical but is not surprising as there are no constraints on the magnitude of the order parameter θ_m. In addition, when solving the equations describing the twist deformation, $\mathcal{C}$ becomes negative for $u > 0.6$, which indicates that the expansion should include higher-order terms. Nonetheless, numerical solutions[11] of the Euler-Lagrange equations which result from minimizing the exact free energy are in good qualitative agreement with results obtained from the simple Landau formalism.

CONCLUSIONS

We have examined the Freedericksz transition in nematic liquid crystals driven by static electric and magnetic fields using Landau theory. Explicit expressions are given for the Landau coefficients in the case of perpendicular electric and magnetic fields in all six possible geometries. We have shown that when an electric field is present parallel to the cell walls, the transitions may be first order. These results suggest that nematic cells may be useful in applications requiring bistability.

ACKNOWLEDGMENTS

TABLE II. Coefficients of the Landau expansion for geometries $a-f$. The geometries are shown in Fig. 1 and the symbols are defined in Table I.

Geometry	$\mathcal{A}$	$\mathcal{B}$	$\mathcal{C}$	Criteria for first order
a	$1 + e - h$	$\frac{1}{2}[-\gamma + h - e(3w + 1)]$	$\frac{1}{8}[\gamma\frac{2}{3}h + e(15w^2 + 10w + \frac{2}{3})]$	$e \geq \dfrac{1-\gamma}{3w}$
b	$1 + h - e$	$\frac{1}{2}[-\gamma - h + e(w + 1)]$	$\frac{1}{8}[\gamma + \frac{2}{3}h - e(3w^2 + 4w + \frac{2}{3})]$	$h \leq \dfrac{-\gamma - 1 - w}{w}$
c	$1 + e - h$	$\frac{1}{2}[h - e(3w + 1)]$	$\frac{1}{8}[-\frac{2}{3}h + e(15w^2 + 10w + \frac{2}{3})]$	$e \geq \dfrac{1}{3w}$
d	$1 + h - e$	$\frac{1}{2}[-h - e(3u - 1)]$	$\frac{1}{8}[\frac{2}{3}h - e(15u^2 - 10u + \frac{2}{3})]$	$h \geq \dfrac{1 - 3u}{3u}$
e	$1 + e - h$	$\frac{1}{2}[-\kappa + h - e(1 - u)]$	$\frac{1}{8}[\kappa - \frac{2}{3}h + e(3u^2 - 4w + \frac{2}{3})]$	$e \leq \dfrac{\kappa - 1}{u}$
f	$1 + h - e$	$\frac{1}{2}[-\kappa - h - e(3u - 1)]$	$\frac{1}{8}[\kappa + \frac{2}{3}h - e(15u^2 - 10u + \frac{2}{3})]$	$h \geq \dfrac{1 - 3u - \kappa}{3u}$

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