Twist-energy-driven Helfrich modulations in cholesteric liquid crystals illustrated in the transient-planar to planar transition

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The complete time evolution of a Helfrich-type instability that is driven by twist energy in a cholesteric liquid crystal is presented. The modulation is investigated using an initially uniform planar director configuration of a cholesteric liquid crystal (with helical axes uniformly aligned along the cell normal) whose pitch is substantially longer than the equilibrium value.

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I. INTRODUCTION

Liquid crystals with layered structures in nonequilibrium configurations may under certain conditions undergo sinusoidal modulations of the layer structure in order to lower the free energy of the system. Electric-field-driven periodic modulations of cholesteric liquid crystals were first described by Helfrich [1]. In such instabilities, the cholesteric layers are seen to form sinusoidal patterns that tend to alleviate the dielectric free energy while increasing the bend (K_{33}) free energy. Hurault [2] and Chigrinov [3] have provided more thorough analyses of the threshold fields involved. The resultant director configurations involved in this problem were further clarified through use of perturbation-theory techniques in the early 1990s [4]. An identical effect has been observed when a magnetic field is employed. These phenomena are well understood, and are explained thoroughly in the literature [5]. Similar distortions have been predicted in smectic liquid crystals, but are not generally observed due to the high threshold field necessary for such a modulation to occur [6]. On increasing field strength in field-induced Helfrich instabilities in cholesteric liquid crystals, the wavelength of the modulation increases until a nematic state is acquired.

A second example of Helfrich-type modulations has been observed in smectic, columnar, and cholesteric systems in which the thickness of the sample is physically increased through application of tension to the glass plates between which the liquid crystal is contained. In such a situation, the nonequilibrium layer spacing of the material seeks to achieve a lower-energy configuration through a buckling of the layers, resulting in locally tilted layers with a somewhat tighter spacing. This effect was discovered in smectic liquid crystals by Delaye, Ribott, and Durand [7] and Clark and Mery [8]. Similar effects resulting from physically changing the thickness of the sample have been observed in columnar systems [9] and cholesteric systems [6]. The expected final state on increasing the amplitude of a tension-driven Helfrich modulation is significantly different from that of a field-induced modulation, due to the fact that under tension, the sample desires to increase the number of layers.

In cholesteric liquid crystals, a nonequilibrium layer spacing occurs during relaxation from the homeotropic configuration, a state in which the natural twist of the material has been completely removed through application of a voltage. When the applied voltage is removed, a relaxation process occurs that results in the formation of the ''transient-planar'' (TP) state, which is a grandjean state, i.e., all helical axes are aligned perpendicular to the substrates, with a cholesteric pitch significantly longer than the equilibrium pitch [10]. The pitch of a cholesteric liquid-crystal system is defined as the distance over which the liquid-crystal director twists 360°. The TP pitch is roughly equal to $(K_{33}/K_{22})P_0$, where P_0, is the equilibrium pitch of the material, and K_{22} and K_{33} are the twist and bend elastic constants of the material, respectively [11]. The occurrence of the nonequilibrium pitch in the TP is a result of the fact that the degree of twist in the material is determined while the director is still roughly normal to the cell plane. For cases such as this in which the director is not perpendicular to the twist axis, the degree of azimuthal twist corresponding to the lowest-energy state is less than the equilibrium value of twist. It is expected that such a state with a nonequilibrium cholesteric layer spacing may exhibit a Helfrich modulation in order to reduce the high twist free energy of the configuration. In this paper, we will investigate the dynamics of such a system, and show how a transformation between states of unequal twist may occur via a Helfrich modulation.

II. PREVIOUS EXPERIMENTAL FINDINGS

In earlier studies, we have experimentally investigated the dynamics of modulation of the TP state in a simple cholesteric liquid-crystal system. This was discussed in our previous work [12], and consisted of a 15-μm cell with rubbed planar surfaces, filled with a mixture of Merck liquid-crystal ZLI-4792 and 3% CB15 chiral additive. This resulted in a d/P (cell thickness over pitch) ratio for the system of about 2.8.

Microphotographic evidence suggested that the cell relaxed to a TP state with 1 twist in about 15 ms. It remained in that state for about 200 ms, after which a bulk modulation with a striped appearance began to distort the texture (see Fig. 1). Over the next 100 ms, the modulation became more pronounced, until the equilibrium texture was seen to appear between stripes. Stripes grouped into pairs, allowing larger

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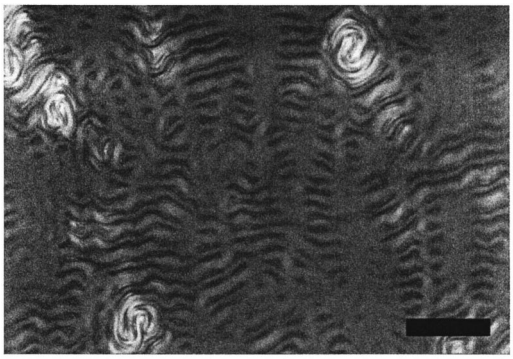

FIG. 1. Striped modulation observed during the transient-planar to planar director configuration change. The black bar is 50 μm long.

regions of equilibrium texture to form. Eventually, the stripes were seen to go away through continued grouping into pairs, as well as through shrinking of the stripes due to defect motion in the third dimension. Because of the appearance and dimensions of these stripes, we proposed that they were a similar structure to those seen in a two-dimensional Helfrich deformation. In this paper we attempt to demonstrate through computer simulation that such a modulation is energetically favorable.

III. SIMULATION TECHNIQUE

For our modeling of the liquid-crystal director, we used the same technique we discussed in a previous publication concerning the homeotropic to focal conic transition in cholesteric liquid crystals [13]. The expression for the free-energy density of a liquid crystal system in terms of the director **n**, is given by Eq. (1) [6].

$$f=\tfrac{1}{2}K_{11}(\vec{\nabla}\cdot\hat{n})^2+\tfrac{1}{2}K_{22}(\hat{n}\cdot\vec{\nabla}\times\hat{n}+q_0)^2 + \tfrac{1}{2}K_{33}|(\hat{n}\times\vec{\nabla}\times\hat{n})|^2-\tfrac{1}{2}\vec{D}\cdot\vec{E}. \quad (1)$$

Here, K_{11}, K_{22}, and K_{33} are the elastic constants for splay, twist, and bend, respectively, $\vec{D}$ is the electric displacement and $\vec{E}$ is the electric field.

Ignoring flow of the material, the dynamics of the director reorientation can be calculated by setting the functional derivatives of the free-energy density with respect to each director component (the elastic torque) equal to the rotational torque on that component as shown in Eq. (2) [14].

$$\gamma_1\frac{dn_i}{dt}=-\frac{\delta f}{\delta n_i}+\lambda n_i, \quad i=x,y,z, \quad (2)$$

where γ_1 is the rotational viscosity. In two dimensions,

$$\frac{\delta f}{\delta n_i}=\frac{\partial f}{\partial n_i}-\frac{d}{dx}\left[\frac{\partial f}{\partial(dn_i/dx)}\right]-\frac{d}{dz}\left[\frac{\partial f}{\partial(dn_i/dz)}\right], \quad i=x,y,z. \quad (3)$$

In Eq. (2), the Lagrange multiplier λ, is used to maintain the unit length of the director. However, we cannot simultaneously solve this equation for the Lagrange multiplier and a numerical update formula for n_i. Therefore, the λ term is dropped and n is renormalized to have unit length after each time step i.e., each director component is divided by the total length [15].

In our calculations, we assume that our starting point corresponds to a time shortly after the voltage has been removed; thus the voltage is assumed to be zero over the entire grid at all times, and is not taken into account. In order to calculate the dynamics of the system, the new director field on the entire computational grid must be calculated before any variables are updated. Note that this model does not take flow effects and effects of ionic contaminants into account. Additionally, such random effects as thermal fluctuations are not considered in the model. Thus, in order to avoid situations where the director acquires a state of equilibrium, which in reality would be short lived or unstable, it is occasionally necessary to add some small perturbation to the director so that the calculation can proceed to a lower-energy state.

Because the experimentally observed modulation appears to be two-dimensional, calculations for our cholesteric system were based on a two-dimensional calculation grid of dimensions 394×197 in the x and z dimensions, respectively, with the x axis parallel to the substrates and the axis perpendicular. The lattice dimensions were set such that the horizontal size of the grid was 50 μm, while the vertical size (cell gap) was 15 μm. Each lattice point thus represented a rectangle of dimensions 0.126 μm wide by 0.0765 μm tall. Fixed homogeneous boundary conditions were assumed with a 5° pretilt. Because of the low concentration of chiral additive used in our experimental mixture, the elastic constants and dielectric anisotropy of the host (Merck ZLI-4792) were used in the calculation.

IV. SIMULATION RESULTS

The initial configuration in experiment was a TP state with a single 360° twist. In simulation, such a state would appear stable because the ideal TP state has spatial dependencies in one dimension. Thus, it is necessary to provide an initial perturbation in order to allow the striped modulation that had experimentally been observed to form. To this end, a sinusoidal Helfrich-type modulation was added to the TP state. Chigrinov *et al.* determined that the expected wavelength of a sinusoidal distortion caused by the presence of an external field should be

$$\lambda=\sqrt{PD\sqrt{3K_{33}/2K_{22}}},$$

where P is the pitch of the material and D is the thickness of the sample [3]. For our system, we expect the periodicity to be roughly equal to this λ, although we must take into account the fact that the formula was derived based on the assumption that the state being distorted was the zero-field equilibrium state. Considering $P=5.4\,\mu$m, $D=15\,\mu$m and $K_{33}/K_{22}=2.815$, we may expect λ to be about 12.9 μm. Measurement of the periodicity in Fig. 1 yields a wavelength of 15.6 μm, agreeing well with the Chigrinov-Helfrich model. We chose to use this experimentally observed wavelength in our simulation [Fig. 2(a)]. After the modulation was inserted, the simulation was allowed to continue. As can be seen in Fig. 2(b), the modulation that we had seeded

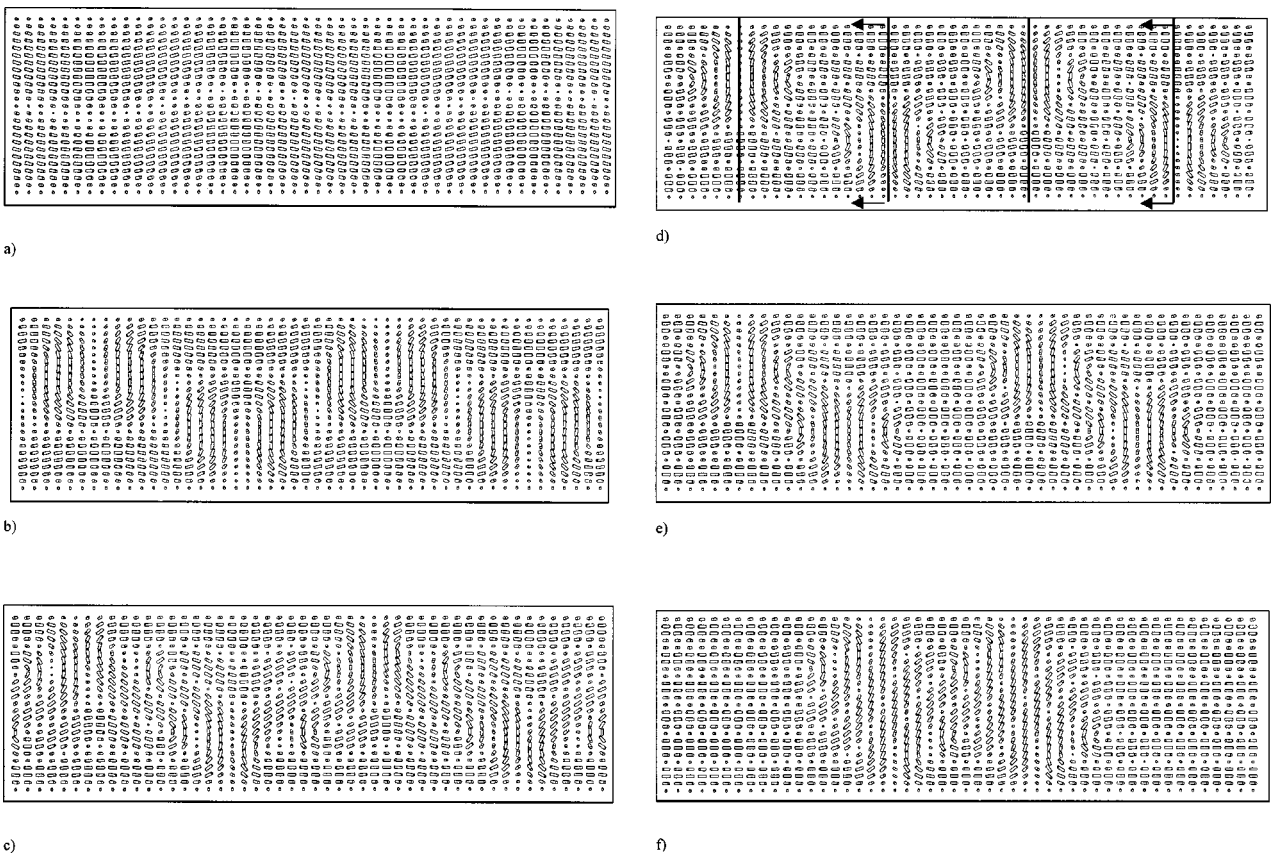

FIG. 2. Drawings of the simulated director configurations throughout the transformation process. The substrates are assumed to be on the top and bottom of the figures. The vertical lines in (d) show the axes of symmetry of the structure. The arrows indicate how alternating regions of equilibrium pitch were increased or decreased.

increased in amplitude with increased time. The modulation at this time no longer exhibits a sinusoidal appearance, but rather appears to exhibit fingers that grow from the surfaces in a spatially alternating fashion.

In Figs. 2(c) and 2(d), it may be seen that the ends of the fingers spread out horizontally, and eventually overlap each other. This results in small regions of the equilibrium pitch (3 twists) alternating with regions of horizontal twist. At this point, the simulation again stopped, having acquired a geometrical symmetry, as is indicated by the vertical lines in Fig. 2(d). This geometrical symmetry, which was present in the initial conditions, is again only an artifact of the mathematics of the model and is not expected to occur in a real system. The experimental data had suggested that at this point in the relaxation process, the stripes exhibited significant lateral fluctuations, and formed into pairs when they made contact. These lateral fluctuations break the geometric symmetry of the director structure. To approximate this in simulation, alternating regions of the equilibrium pitch were increased or diminished in size in order to approximate the randomness of the real system as shown in Figure 2(d). This resulted in a horizontal translation of the horizontal-twist regions of the display [Fig. 2(e)]. After moving these horizontal-twist regions, the simulation program was restarted to allow the system to smooth out any regions of locally high strain energy, which may have resulted from the modifications of the director. Because the energy density reduction resulting from the lateral movement of horizontal-twist regions is roughly two orders of magnitude smaller than that caused by the early increases of amplitude of the modulation, in simulation it is expected to move about 100 times slower. Due to the extremely long calculation time that would be necessary to complete this translation, the abovementioned manual translation process was repeated several times until the horizontal-twist regions became close enough that they could combine [Fig. 2(f)]. In all cases, modifications to the director structure decreased the free energy of the system. These events would therefore be expected to occur in the experimental system, seeded by random dynamics in the display, such as flow effects and thermal fluctuations, combined with imperfections in the cell surfaces and the presence of spacers. At the time of Fig. 2(f) the four horizontal-twist regions of Fig. 2(d) have been reduced to two. Allowing these regions to combine also results in a significant reduction of free energy, while at the same time increasing the volume of equilibrium cholesteric texture.

Table I shows the splay, twist, bend, and total free energies for each director configuration given in Fig. 2. We can clearly see that the total free energy decreases as the transition progresses. It may also be noted that the twist energy is alleviated at the cost of increased bend energy, and, later in the process, slightly increased splay energy.

Our model does not allow for the scalar order parameter S to vary in amplitude, so in simulation, defects could not form. In order to verify that this assumption is valid, we compared the free-energy density observed in the simulation with that of a defect. By using a method described by De-

TABLE I. Splay, twist, bend, and total (splay+twist+bend) free energies during the transition from the transient-planar state to the planar state. The energies given correspond to the director configurations shown in Fig. 2.

	Splay energy density ($J\,m^{-1}$)	Twist energy density ($J\,m^{-1}$)	Bend energy density ($J\,m^{-1}$)	Total energy density ($J\,m^{-1}$)
Fig. 2(a)	1.57×10^{-6}	1.06×10^{-4}	1.75×10^{-6}	1.09×10^{-4}
Fig. 2(b)	8.52×10^{-7}	6.20×10^{-5}	1.47×10^{-5}	7.75×10^{-5}
Fig. 2(c)	6.30×10^{-7}	1.22×10^{-5}	1.77×10^{-5}	3.05×10^{-5}
Fig. 2(d)	6.10×10^{-7}	6.43×10^{-6}	1.60×10^{-5}	2.30×10^{-5}
Fig. 2(e)	6.15×10^{-7}	6.40×10^{-6}	1.58×10^{-5}	2.28×10^{-5}
Fig. 2(f)	5.84×10^{-7}	5.34×10^{-6}	9.92×10^{-6}	1.58×10^{-5}

Gennes and Prost [6], the energy density of a defect can be estimated to be roughly $75\,000\,J\,m^{-3}$ [13]. Figure 3 (a) shows a plot of the energy density and the director configuration corresponding to Fig. 2(d). Figure 3(b) shows a close up of one of the high-energy regions. We can note that this energy density is almost 10 000 times lower than that needed to form a defect [13].

V. SUMMARY

For this system, the simulation is seen to agree very well with the observed texture. The uniform TP texture was seen to be deformed by a spatial Helfrich-like modulation that increased in amplitude, eventually resulting in the appearance of the equilibrium state. The simulation showed that the peaks of this modulation spread out horizontally after a short time, resulting in small regions of equilibrium twist alternating with regions of horizontal twist. It was seen in the simulation that having two regions of horizontal twist meet each other and join, would result in a significant lowering of the free energy, and also result in the appearance of larger regions of equilibrium texture. These events are also seen to occur in experiment, as we have reported earlier [7].

In this paper we have confirmed through computer simu-

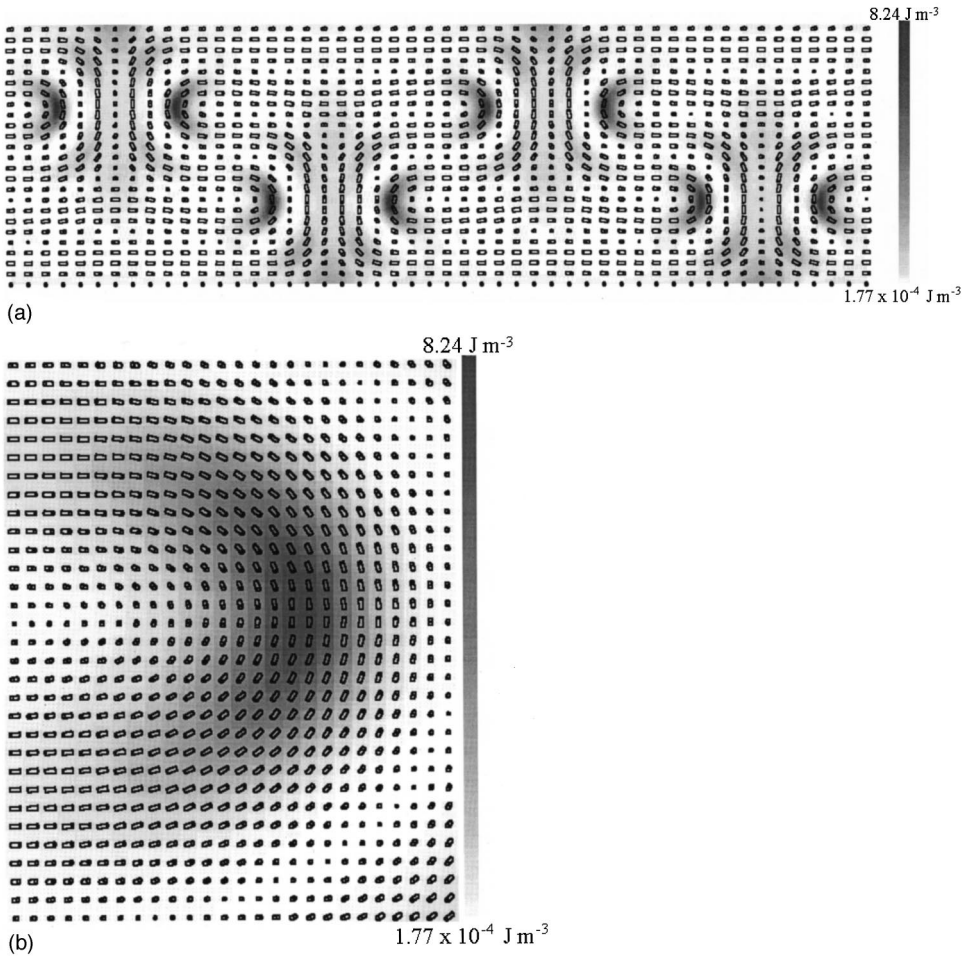

FIG. 3. (a) Total free-energy density plot showing the regions of high energy. The director configuration is the same as in Fig. 2(d). (b) Close up of one of the high-energy regions from (a). All calculated directors are shown. Note the almost pure bend distortion in this region. The director configuration is the same as in (d).

lations that, once seeded, a sinusoidal modulation in a nonequilibrium twist state of a cholesteric liquid-crystal system can be expected to grow in amplitude. This agrees well with experimental evidence presented in previous reports. The numerical simulation and energy analysis confirms that the basic mechanism of transition between the transient-planar and equilibrium states of cholesteric liquid-crystal systems begins with a Helfrich modulation and proceeds in a defect-free, continuous manner. We can therefore conclude that tension-driven Helfrich modulations in cholesteric liquid crystals are well described by this model.

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