Cholesteric–nematic texture transition in liquid crystals

V. L. Golo
M. V. Lomonosov Moscow State University, 119899, Moscow

E. I. Kats
L. D. Landau Institute of Theoretical Physics, Russian Academy of Sciences, 142432, Chernogolovka

(Submitted 3 February 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **55**, No. 5, 275–280 (10 March 1992)

The cholesteric–nematic phase transition which occurs in a thin layer of a liquid crystal upon a change in the pitch of the helix or the thickness of the layer is analyzed. A geometric analog of the Ritz–Galerkin variational method is proposed for calculating the energy of various textures of a liquid crystal. Contours in order-parameter space which describe the configuration of one-dimensional textures of a liquid crystal are used as trial functions. Several metastable states exist (there is a multistability). There are also regions in which the various states of a cholesteric spiral remain nearly constant (at the accuracy level of the variational method used here). The physical meaning of this result is an additional degeneracy of the system.

1. In thin layers of cholesteric liquid crystals with a strong orientational coupling at the boundaries of the sample, there will be various texture conversions associated with a competition between bulk and surface effects. These texture transitions hold promise for applications, so they have been the subject of a fairly long list of studies.

 0021-3640/92/050273-07$03.00

In this connection we note the work by Press and Arrot (Ref. 1; see also Refs. 2–4), who studied a nematic-cholesteric mixture in a wedge-shaped cell. By taking that approach, they were able to vary the pitch of the spiral ($p_0 \equiv 2\pi/q_0$) and the thickness of the liquid crystal (L) independently. At small thicknesses ($L < 0.5p_0 \sim 5\ \mu$m), Press and Arrot[1] observed a uniform nematic structure. At intermediate thicknesses (5–10 μm) they observed a highly distorted cholesteric structure. Finally, in thick samples ($L \geqslant 20\ \mu$m) they observed a cholesteric structure.

Zel'dovich and Tabiryan[5] carried out a theoretical study of the one-dimensional problem of the structure of a cholesteric liquid crystal in a thin layer with homeotropic boundary conditions. They found an exact solution in terms of elliptic functions.

In the present paper, guided by the experimental situation,[1–4] we wish to describe the nematic–cholesteric transition in thin films. We also propose a convenient method for calculating the energies of various textures of a liquid crystal. This method is a geometric analog of a variational method, in which the trial functions are specially selected closed loops in order-parameter space.

To offer a graphic description of the state of the liquid crystal, it is convenient to use a geometric representation of the order-parameter space.[6] For both a nematic and a cholesteric, the degeneracy space is a sphere S^2 with identified antipodal points. Any state of a nematic or a cholesteric (a texture) can be described by certain points or lines on this surface. For example, a uniformly ordered nematic corresponds to a point on S^2, while an ideal cholesteric corresponds to an equatorial circle.

Upon a change in the external conditions (the temperature, the composition of the mixture, or the thickness of the layer of liquid crystal), a nematic–cholesteric phase transition can occur. In the course of this transition, there should be a certain region of a nonuniform orientation (represented by some curve on S^2). An instability of the nematic liquid crystal (of the point on S^2) with respect to the nonuniformly deformed region (a curve which emerges from this point) can occur in either a soft fashion (i.e., continuously) or a hard fashion (i.e., as a first-order phase transition), depending on the boundary conditions and on the elastic anisotropy. A particular case of this instability, corresponding to an imaging point starting at the "north pole" of S^2, was analyzed in Ref. 5 and labeled a "field-free Fréedericksz transition."

2. Both the cholesteric structure and the nematic structure of a liquid crystal can be described as extrema of a Frank free-energy functional[7]

$$F = \frac{1}{2}\int d^3r[K_1(\mathrm{div}\vec{n})^2 + K_2(\vec{n}\cdot\mathrm{curl}\,\vec{n} + q_0)^2 + K_3(\vec{n}\times\mathrm{curl}\,\vec{n})^2]. \qquad (1)$$

Here $\vec{n}$ is the director; K_1, K_2, and K_3 are elastic moduli; and q_0 is a parameter. At $q_0 = 0$, the ground state of (1) corresponds to a nematic (a uniform state, with $\vec{n} = \mathrm{const}$); with $q_0 \neq 0$, it corresponds to a cholesteric. In the latter case, there is a nonuniform ground state with a one-dimensional twisting of the field $\vec{n}$:

$$\vec{n}\cdot\mathrm{curl}\,\vec{n} = -q_0.$$

In principle, the ground state of (1) can have a nonzero twisting under the

condition $K_2 < 0$, even if $q_0 = 0$. If (1) is to be positive definite in this case, gradients of higher order must of course be considered; these gradients limit the growth of the twisting. The region $K_2 < 0$ describes a cholesteric, while at $K_2 > 0$ the ground state corresponds to a nematic liquid crystal. The point $K_2 = 0$ is the "critical Lifshitz point." A critical behavior of this type has been studied thoroughly for magnetic materials and certain other systems (Ref. 8, for example).

We will not discuss that possibility here. We instead consider phase transitions which occur in thin layers of a liquid crystal upon a variation of q_0 (or L), since this is the conversion which is realized experimentally.[1-4]

To describe these transitions, we write the energy in (1) for a thin layer of a liquid crystal. We assume that the configuration of this liquid crystal is uniform in the plane of the layer. In other words, we consider only the dependence on the coordinate in the direction orthogonal to the layer (the z axis). We then find from (1)

$$F = \frac{1}{2}\int_0^L dz[K_1\dot{n}_3^2 + K_2(\epsilon_{\mu\nu}n_\mu\dot{n}_\nu + q_0)^2 + K_3 n_3^2\dot{\vec{n}}^2]. \tag{2}$$

The superior dot here means differentiation with respect to z; L is the thickness of the layer of liquid crystal; μ, $\nu = 1$, 2; and $\epsilon_{\mu\nu}$ is the antisymmetric tensor ($\epsilon_{12} = -\epsilon_{21} = 1$).

The problem which arises here is equivalent, even with $q_0 = 0$, to a study of geodesics on an ellipsoid. With $q_0 \neq 0$, it is an even more complex matter to find an exact solution, since (2) contains a contribution which is linear in the twisting (and which is formally analogous to a magnetic field[6]). As a result, a minimization of F in the case $q_0 \neq 0$ establishes the best compromise between the length of the contour on S^2 and the flux of the vector field through the area bounded by this contour.

This discussion can be reworded more precisely. The part of the energy in (2) which is quadratic in the gradients, and which is given by the integral

$$\int_0^L dz[K_1\dot{n}_3^2 + K_2(\epsilon_{\mu\nu}n_\mu\dot{n}_\nu)^2 + K_3 n_3^2\dot{\vec{n}}^2],$$

is greater than the square of the length of a geodesic[1]:

$$I = \frac{1}{L}\left[\int_0^L dz[K_1\dot{n}_3^2 + K_2(\epsilon_{\mu\nu}n_\mu\dot{n}_\nu)^2 + K_3 n_3^2\dot{\vec{n}}^2]^{1/2}\right]^2.$$

We thus find the following estimate of the energy:

$$F \geq Lq_0^2 + I + K_2 q_0 \Delta, \tag{3}$$

where Δ is the area of the contour in degeneracy space (i.e., on a sphere of unit radius).

If the characteristic size of the contour on S^2 is r, then we have $\Delta \sim r^2$ and $I \sim K_{1,3} L^{-1} r^2$.

The first two contributions in (3) are positive; the third is not of fixed sign. This contribution will have to be rendered negative in order to reduce F. It describes the flux of the vector field through the region bounded by the contour. At small values of $|Lq_0|$, the rough estimate in (3) leads to the following condition for the stability of a uniform nematic structure (i.e., of a point in comparison with a contour on S^2):

$$L \leq \frac{K_{1,3}}{K_2} \frac{1}{q_0}. \tag{4}$$

3. In light of the discussion above, we will use a Ritz–Galerkin variational method to minimize the functional in (2). Our method makes use of the specific geometry of the problem. The functional in (2) can be specified on contours in order-parameter space, which has the symmetry SO(3). As trial functions we accordingly select a class of curves on S^2 which describe the configuration of a cholesteric. The optimum contours for minimizing (2) are circles, since (for a fixed length) circles bound the largest area on a sphere. Geodesics as such do not solve our minimization problem, since they give us only the extremum of the length.

We assume that strong-coupling conditions hold at the boundaries:

$$\vec{n}|z = 0| = \vec{n}(z = L) = \vec{n}_0 = \text{const}. \tag{5}$$

Boundary conditions (5) determine a point on S^2. We choose the contour (circle) mentioned above in the following way. We denote by $\vec{m}$ a vector drawn from the center of the sphere S^2 to the center of the circle. The contour must of course also satisfy boundary conditions (5); i.e., the circle with center at $\vec{m}$ must pass through the point $\vec{n}_0$. Such a circle is conveniently parametrized by means of the orthogonal vec-

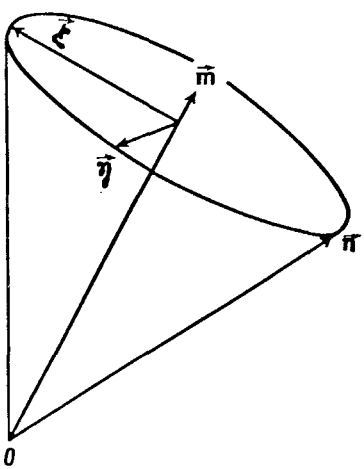

FIG. 1.

tors $\vec{\xi}$ and $\vec{\eta}$, which, along with $\vec{m}$, constitute an orthogonal but not normalized basis (Fig. 1):

$$\vec{\xi} = \vec{n}_0 - (\vec{m}\vec{n}_0)\vec{m}; \quad \vec{\eta} = \vec{m} \cdot \vec{n}_0 ,$$
$$\vec{\eta}^2 = \vec{\xi}^2 = 1 - (\vec{m}\vec{n}_0)^2. \tag{6}$$

In this basis, an arbitrary point $\vec{n}$ of the contour is specified by

$$\vec{n} = (\vec{m} \cdot \vec{n}_0)\vec{m} + \vec{\xi}\cos z + \vec{\eta}\sin z. \tag{7}$$

Substituting (7) into (2), and integrating over z, we find

$$F = \frac{1}{2}[1-(\vec{m}\vec{n}_0)^2]\{(K_1+K_3)+(K_2-K_3)(\vec{m}\vec{n}_0)^2$$
$$-m_3^2[(K_1+K_3)+(K_2-3K_3)(\vec{m}\vec{n}_0)^2]\}$$
$$+K_2\{q_0+[1-(\vec{m}\vec{n}_0)^2]m_3\}^2. \tag{8}$$

In the class of contours which we have selected, the problem of minimizing functional (2) (i.e., the problem of solving a system of nonlinear partial differential equations) has been reduced to the algebraic problem of minimizing the function in (8) with respect to the parameters $y \equiv \vec{m}\vec{n}_0$ and m_3.

We can replace the variables y, m_3 by m_1, m_3, since we can always choose $\vec{n}_0$ in the form

$$\vec{n}_0 = (\sin\alpha, 0, \cos\alpha) ,$$

so we can always write

$$y = \sin\alpha m_1 + \cos\alpha m_3$$

(the angle α specifies the orientation at the boundary of the sample).

Since $\vec{m}$ is a unit vector, we must seek a minimum of (8) in circle K:

$$m_1^2 + m_3^2 \leq 1.$$

If the minimum lies on the boundary of the circle ($m_1^2 + m_3^2 = 1$), then there exists a trial circle which is perpendicular to the plane passing through $\hat{z}$ and $\vec{n}_0$ ($\hat{z}$ is a vector along the normal to the plane layer of the liquid crystal). In other words, the solution is invariant under reflection (R) in the plane which is parallel to the boundaries and which passes through the middle of the layer [these solutions are considered in (1) for the particular case $\alpha = 0$].

Our numerical analysis of the function in (8) revealed no local minima inside K. In other words, equilibrium (and even metastable) textures of the liquid crystal should be invariant under R.

The energy in (8) takes its simplest form in the particular case of equal elastic moduli. Also noting that all the minima of (8) occur on the boundary of our circle K,

and specifying the orientation of $\vec{m}$ by means of an angle $\beta(m_1 = \sin\beta,\ m_3 = \cos\beta)$, we find from (8)

$$F = K[q_0^2 + \sin^2(\alpha - \beta)(1 + 2q_0\cos\beta)]. \tag{9}$$

We postpone a detailed analysis of the texture transitions described by the functions in (8) [or (9)] to a separate paper. We restrict the present letter to the results of that analysis.

At values $|q_0L|$ smaller than a certain critical κ_c (which depends on K_2/K_1, K_3/K_1, and α), the original nematic structure remains stable. In other words, a point $\vec{n}_0$ on S^2 corresponds to a minimum. At $|q_0L| > \kappa_c$, some new minima appear. They correspond to a distorted nematic or cholesteric structure. These minima are reached on the circular contours described above at various values of the angle β. Figure 2 illustrates the situation with a plot of F versus β for the values $q_0 = -0.5$, $K_1 = K_3$, $K_2 = 1.6\,K_1$, and $\alpha = 0.47$ rad. We see a plateau with an angular width of about 25°, on which F remains constant within 0.1%. For the specified values of the elastic moduli, a one-dimensional texture is, according to Ref. 1, preferred to a two-dimensional texture from the energy standpoint.

For $K_1 = K_2 = K_3$ and $\alpha = 0$ (this case was discussed in Ref. 5), a second-order phase transition occurs right at κ_c. In general, however, the value of κ_c specifies only the boundaries of metastable structures, and the texture transitions described above

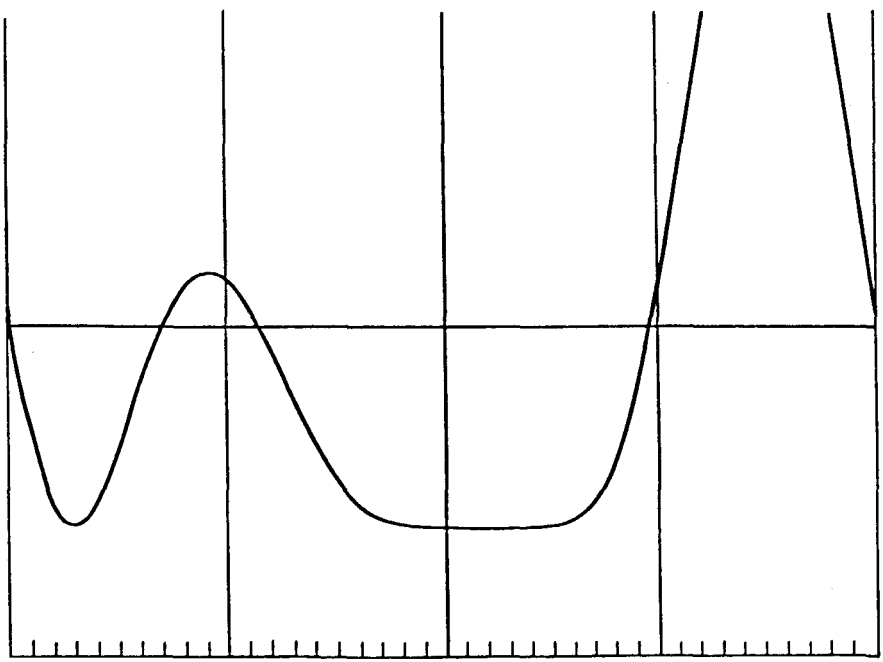

FIG. 2.

are first-order phase transitions. Consequently, the nematic-cholesteric transition in thin layers of liquid crystals should be accompanied by a variety of thermal effects, since the parameter q_0 is extremely sensitive to temperature changes in liquid crystals.

We close with one more aspect of the energies of various cholesteric textures calculated from (8) or (9) by a contour method. A numerical analysis of (8) and (9) reveals that these functions have regions of a very slow change, which can be thought of (within the accuracy of our variational approach) as regions of a constant potential. Physically, this circumstance means that there is an additional degeneracy of the ensemble of cholesteric structures in regions corresponding to the plateau of the effective potential in (8) or (9). We will not discuss this phenomenon in more detail here, but it is totally clear that this additional degeneracy should have numerous physical consequences (e.g., various multidomain structures and an increased intensity of light scattering). This effect has some promising applications.

[1)]This result follows from the Cauchy inequality $\int_0^T \dot{\vec{u}}^2(t)dt \geqslant \frac{1}{T}(\int \sqrt{\dot{\vec{u}}^2}dt)^2$.

[1]M. J. Press and A. S. Arrot, J. Phys. (Paris) **37**, 387 (1976).
[2]W. Haas and J. Adams, Appl. Phys. Lett. **25**, 535 (1974).
[3]M. Uchiyama and C. Ishii, J. Phys. Soc. Jpn. **50**, 4030 (1981).
[4]G. Porte and J. P. Jadot, J. Phys. (Paris) **39**, 213 (1978).
[5]B. Ya. Zel'dovich and N. V. Tabiryan, Zh. Eksp. Teor. Fiz. **83**, 998 (1989) [*sic*].
[6]F. Lequas, J. Phys. (Paris) **49**, 967 (1988).
[7]P. G. de Gennes, *The Physics of Liquid Crystals*, Oxford Univ. Press, New York, 1974.
[8]R. M. Hornreich, M. Luban, and S. Shtrikman, Phys. Rev. Lett. **35**, 1678 (1975).

Translated by D. Parsons