Pis'ma v ZhETF, vol.71, iss.2, pp.123 - 127

THEORY OF SPHERULITIC DOMAINS IN CHOLESTERIC LIQUID CRYSTALS WITH POSITIVE DIELECTRIC ANISOTROPY

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Submitted 15 December 1999

New localized axisymmetric nonsingular solutions minimizing the Frank functional have been found for a cholesteric layer with homiotropic anchoring. They have a continuous distribution of the director field and are characterized by convex shape. These solutions describe the so called spherulitic domains which have been observed in a large-pitch cholesteric near the unwinding transition.

PACS: 61.30.-v, 64.60.-i

Inhomogeneous localized structures (*topological defects or localized states*) are the focus of attention in many fields of modern physics. Since late 1960s soliton-like solutions of nonlinear field equations have been studied in condensed matter physics and biophysics, in particle and nuclear physics, in astrophysics and cosmology [1]. In particular, spontaneous pattern formation during symmetry breaking phase transitions is intensively studied in particle physics, cosmology, and many regions of condensed matter physics. The idea to model cosmological scenarios in condensed matter systems [2] has given new directions to the intra-disciplinary collaboration in physics of topological defects.

It is characteristic of liquid crystals to have a rich variety of phases and phase transitions between them [3]. Due to optical anisotropy of liquid crystals, inhomogeneous states can be observed directly with a polarizing microscope; material and external parameters can be easily varied in a wide range; as a rule, experiments are carried out at room temperature. All these advantages make a liquid crystal a convenient object for the modeling and investigation of different inhomogeneous states. Recently numerous experimental and theoretical results on pattern formation [4] and solitons [5] have been obtained for liquid crystals.

During last decades, many experiments have been devoted to topological defects in a large-pitch cholesteric near the unwinding transition (see bibliography in Ref. [6]). In these experiments a thin layer of a liquid crystal is sandwiched between two parallel electrodes which anchor the molecules perpendicularly to the surfaces (homeotropic anchoring). Depending on value and frequency of an applied electric field textures consisting of elongated domains (*fingers*), loops, spirals, bubble-shaped objects (named *cholesteric bubbles* or *spherulitic domains*) have been observed in many samples, and their static and dynamic properties have been investigated. Spherulitic domains are generated by quenching of electrohydrodynamic turbulence and at the cholesteric-isotropic transition. They exist either as isolated localized states or form regular lattices. Up to now several models of spherulitic domains have been proposed [7, 8]. In these contributions the director field in spherulitics has one or more singular lines (disclinations).

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The aim of this Letter is to propose a nonsingular model for spherulitic domains in cholesterics with positive dielectric anisotropy. It is shown that localized rotationally symmetric structures with a continuous distribution of the director field are among solutions of the equations minimizing the Frank functional for the cholesteric layer with homeotropic boundary alignment. These static solutions are stabilized by an energy contribution linear in the first order spatial derivatives of the director, which arise from enantiomorphy of cholesteric materials. Convex shape of spherulitic is due to homeotropic anchoring.

The possible distributions of the director $\mathbf{n}(\mathbf{r})$ of a bulk cholesteric in an applied electric field $\mathbf{E}$ are determined by minimizing the Frank free energy [3]

$$\Delta F = \int f dV = \int [\frac{K_1}{2}(\mathrm{div}\mathbf{n})^2 + \frac{K_2}{2}(\mathbf{n}\cdot\mathrm{rot}\mathbf{n}+q_0)^2 + \frac{K_3}{2}(\mathbf{n}\times\mathrm{rot}\mathbf{n})^2 -$$

$$-\frac{\varepsilon_a}{2}(\mathbf{n}\cdot\mathbf{E})^2 - f_0]dV, \qquad (1)$$

where K_i and q_0 are elastic constants; ε_a is the dielectric anisotropic constant (we consider materials with $\varepsilon_a > 0$); $f_0 = K_2/2 - \varepsilon_a E^2/2$ is a free energy density for the homogeneous state with $\mathbf{n}||\mathbf{E}$. At zero electric field the ground state of a cholesteric corresponds to a helical structure with a pitch $p = 2\pi/|q_0|$. With increasing electric field perpendicular to the helix axis the pitch gradually increases. Finally, in the critical field

$$E_0 = \frac{\pi q_0}{2}\sqrt{\frac{K_2}{\varepsilon_a}} \qquad (2)$$

the system transforms into a homogeneous states with $\mathbf{n}||\mathbf{E}$.

Consider a thin layer of liquid crystal placed between two parallel glass plates. Let the z axis be perpendicular to the glass plates. The layer has thickness D and is infinite in x and y directions. The liquid crystal is constrained to be perpendicular to the boundary surfaces (homeotropic anchoring) and an electric field $\mathbf{E}$ is applied along the z direction. This model corresponds to geometry of the experiments on spherulitic domains.

For simplicity an approximation of equal elastic constants will be used ($K_1 = K_2 =$ $= K_3 = K$). It is convenient to express the director vector in terms of spherical coordinates

$$\mathbf{n} = (\sin\theta\cos\psi, \sin\theta\sin\psi, \cos\theta),$$

and the spatial variable in cylindrical coordinates $\mathbf{r} = (\rho, \varphi, z)$. In these variables the energy density f (1) assumes the following form:

$$f = \frac{K}{2}\left\{\left(\frac{\partial\theta}{\partial z}\right)^2 + \left(\frac{\partial\theta}{\partial\rho}\right)^2 + \frac{\sin^2\theta}{\rho^2}\left(\frac{\partial\psi}{\partial\varphi}\right)^2 + \right.$$

$$\left. +2q_0\left[\left(\frac{\partial\theta}{\partial\rho}\right) + \frac{\sin\theta\cos\theta}{\rho}\left(\frac{\partial\psi}{\partial\varphi}\right)\right]\sin(\psi-\varphi)\right\} + \frac{\varepsilon_a E^2\sin^2\theta}{2} + \frac{\beta}{2}\sin^2\theta\delta(z\pm z_0) \qquad (3)$$

The last term in (3) describes the interaction between the liquid crystal and the confining surfaces ($z = \pm D/2$), for homeotropic anchoring $\beta > 0$; $\delta(x)$ is the Dirac function.

The variational problem for the energy functional (1) with (3) has rotationally symmetric solutions $\theta(\rho, z)$, $\psi(\varphi) = \varphi + \pi/2$. Substituting this solution into (3) and integrating the energy (1) with respect to φ, one obtains:

$$\Delta F = \pi K \int_{-D/2}^{D/2} dz \int_0^\infty \rho d\rho \{ \left(\frac{\partial\theta}{\partial z}\right)^2 + \left(\frac{\partial\theta}{\partial\rho}\right)^2 + \frac{\sin^2\theta}{\rho^2} - \frac{2q_0}{K}\left(\frac{\partial\theta}{\partial\rho} + \frac{\sin\theta\cos\theta}{\rho}\right) +$$

$$+ \frac{\varepsilon_a E^2}{K}\sin^2\theta + \frac{\beta}{K}\sin^2\theta\,\delta\,(z \pm z_0)\,\}. \tag{4}$$

The Euler equation for the functional (4)

$$\frac{\partial^2\theta}{\partial z^2} + \frac{\partial^2\theta}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial\theta}{\partial\rho} - \frac{\sin\theta\cos\theta}{\rho^2} - \frac{2q_0}{K}\frac{\sin^2\theta}{\rho} - \frac{\varepsilon_a E^2}{K}\sin\theta\cos\theta = 0 \tag{5}$$

with boundary conditions: $\theta(0,z) = 0$, $\theta(\infty, z) = \pi$,

$$\left(\frac{\partial\theta}{\partial z} + \frac{\beta}{K}\sin\theta\cos\theta\right)_{z=\pm D/2} = 0 \tag{6}$$

describes the director field in a nonsingular localized axisymmetric structure.

If the anchoring at the surfaces disappears ($\beta \to 0$) Eq. (5) transforms to an ordinary equation:

$$\frac{d^2\theta}{d\rho^2} + \frac{1}{\rho}\frac{d\theta}{d\rho} - \frac{\sin\theta\cos\theta}{\rho^2} - \frac{2q_0}{K}\frac{\sin^2\theta}{\rho} - \frac{\varepsilon_a E^2}{K}\sin\theta\cos\theta = 0 \tag{7}$$

for localized structures homogeneous along the z axis. Eq. (7) has the same functional form as those for localized states in noncentrosymmetric ferromagnets [9] and chiral liquid crystals [10]. Numerical integration shows that Eq. (7) has metastable localized solutions $\theta(\rho)$ for all $E > E_0(2)$. The functions $\theta(\rho)$ decay exponentially at large distances, and usually the profiles $\theta(\rho)$ have arrow-like core that can be approximated by a linear ansatz:

$$\theta(\rho) = \pi\frac{\rho}{\rho_0} \quad (0 < \rho < \rho_0), \quad \rho_0 = \frac{4}{\pi^2 q_0}\left(\frac{E_0}{E}\right)^2. \tag{8}$$

The localized structures are compressed by increasing electric field but they do not collapse even at high field. It is clear, however, that there should be an upper boundary field where the core size becomes of the order of the molecular length and the elastic approach can not be applied. Near the critical field E_0 the profile has an extending core with $\theta \approx 0$ separated from the outside by a narrow domain wall. As the field goes to its critical value the localized solution transforms to the homogeneous state by unlimited expansion of the core.

It is important to mention that solutions under discussion are stabilized by the energy contribution linear in the first spatial derivatives of the director (the "chiral" term $\mathbf{n}\cdot\mathrm{rot}\mathbf{n}$). This energy term is specific for noncentrosymmetric cholesteric phases. In contrast, two-dimensional localized states are radially unstable and collapse spontaneously in centrosymmetric materials (e.g. in nematics or regular ferromagnets). The stability of two-dimensional localized static structures in nonlinear field models has been considered from general point of view in Ref. [11]).

Now consider the influence of the homeotropic anchoring on the two-dimensional localized states. The solutions of Eq.(5) can be treated as certain deformations of a homogeneous along the z axis profile $\theta(\rho)$ by the surface anchoring:

$$\widetilde{\theta}(\rho, z) = \theta(\rho) + \sum_{n=1}^{\infty} a_n(z) \sin[n\theta(\rho)], \tag{9}$$

In particular, deformations of type

$$\widetilde{\theta}(\rho, z) = \theta[\rho/R(z)] \tag{10}$$

describing profile compression ($R < 1$) or expansion ($R > 1$) are of importance for our problem ($R(z)$ is an arbitrary positive number and characterizes the size of the profile core). Note that the main energy contribution in (4) represented by the first three terms do not change under the scale transformation (10). Thus one can assume that (10) gives the main contribution to the optimal deformation of the profile under the influence of the surface anchoring. The validity of this assumption has been proved numerically.

Integrating (4) with the solution (10) with respect to ρ results in

$$W = \int_{-D/2}^{D/2} dz \left\{ \left(\frac{dR}{dz}\right)^2 + A\frac{\varepsilon_a E^2}{K} R^2 - B\frac{2q_0}{K} R + \frac{\beta}{K} A R^2 \delta(z \pm z_0) \right\}, \tag{11}$$

where

$$A = I_1/I_0, \quad B = I_2/I_0, \tag{12}$$

$$I_0 = \int_0^{\infty} \left(\frac{d\theta}{d\rho}\right)^2 \rho d\rho, \quad I_1 = \int_0^{\infty} \sin^2\theta \rho d\rho, \quad I_2 = \int_0^{\infty} \left(\frac{d\theta}{d\rho} + \frac{\sin\theta\cos\theta}{\rho}\right) \rho d\rho. \tag{13}$$

The Euler equation for the functional (11) with the boundary conditions

$$\left(\frac{dR}{dz}\right)_{z=\pm D/2} = \mp \frac{\beta}{K} A R \tag{14}$$

has an analytical solution:

$$R(z) = R_0 \left[\frac{1 + \gamma \tanh(\alpha)}{1 + \gamma \coth(2\alpha)} - \frac{2\gamma \sinh^2(2\alpha z/D)}{\sinh(2\alpha) + \gamma \coth(2\alpha)}\right], \tag{15}$$

where

$$R_0 = \frac{B}{A}\frac{K}{\varepsilon_a}\frac{q_0}{E^2}, \quad \alpha = \frac{DE}{4}\sqrt{\frac{A\varepsilon_a}{K}}, \quad \gamma = \frac{\beta\sqrt{A}}{\varepsilon_a E^2}. \tag{16}$$

Eq. (15) describes a convex shape of the localized structure. The parameter R_0 characterizes the profile size and the parameters α and γ describe the structure convexity. The largest size reaches in the center of the layer ($z = 0$)

$$R_{max} = R_0 \frac{1 + \gamma \tanh(\alpha)}{1 + \gamma \coth(2\alpha)}, \tag{17}$$

and the smallest values

$$R_{min} = \frac{R_0}{1 + \gamma \coth(2\alpha)} \tag{18}$$

correspond to the layer surfaces ($z = \pm D/2$). The profile size and convexity decrease with increasing field. At high field the ratio

$$R_{max}/R_{min} = 1 + \gamma \tanh(\alpha) \tag{19}$$

goes to unity i.e. the structure tends to be homogeneous along the z axis.

The angle between the tangent to the line (15) and the z axis

$$\xi = \arctan\left[-\frac{4\alpha\gamma R_0}{D}\frac{\sinh(4\alpha z/D)}{\sinh(2\alpha) + \gamma\coth(2\alpha)}\right] \tag{20}$$

equals zero for $z = 0$ and reaches the highest value at the surfaces.

In the strong anchoring limit ($\beta \to \infty$) the localized structure has zero size at the surfaces ($R_{min} = 0$) but the angle ξ has a finite value at the surfaces

$$(\xi)_{z=\pm D/2} = \mp \arctan\left[\frac{4\alpha R_0}{D}\tanh(2\alpha)\right]. \tag{21}$$

In summary, two-dimensional axisymmetric nonsingular localized structures can exist in cholesterics with positive dielectric anisotropy above the unwinding critical field E_0 (2). In layers with homeotropic anchoring these localized states have a convex shape. They can be used as a model for the spherulitic domains observed in cholesteric layers with positive dielectric anisotropy. It is shown that such localized structures can exist not only near the critical field E_0 but also at higher fields. Their stability is due to the chiral energy contributions of cholesteric materials.

Spherulitic domains also occur in cholesteric thin layers with negative dielectric anisotropy and strong homeotropic anchoring (see, e.g. Refs.[7, 12]). There are no localized axial structures in bulk materials for this case. The formation and stability of such spherulitics are completely determined by surface anchoring. Theory of these localized states will be presented in a separate contribution.

The idea of this work arose during the Seminar on Topological defects in Non-Equilibrium Systems and Condensed Matter. I grateful to L. Kramer for the invitation to the seminar and P. Fulde for hospitality and support. I also thank P. Cladis, M. Kléman, L. Kramer, O. Lavrentovich, and A. Shestakov for helpful discussions and D. Garanin for critically reading the manuscript.

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1. C.Rebbi and G.Soliani,*Solitons and Particles*, World Scientific Pub. Co., Singapore, 1984.
2. W.H.Zurek, Phys. Rep. **276**, 177 (1996).
3. P.G. de Gennes and J.Prost, *The Physics of Liquid Crystals*, Clarenton, Oxford, 1993.
4. *Pattern Formation in Liquid Crytals*, Eds. A. Buka and L. Kramer, Springer-Verlag, New York, 1996.
5. *Solitons in Liquid Crystals*, Eds. L.Lam and J. Prost, Springer-Verlag, New York, 1992.
6. J.Baudry, S.Pirkl, and P.Oswald, Phys. Rev. **E59**, 5562 (1999).
7. A.E.Stieb, J. Physique **41**, 961 (1980).
8. S.Hirata, T.Akahane, and T.Tako, Mol. Cryst. Liq. Cryst. **75**, 47 (1981).
9. A.N.Bogdanov and D.A.Yablonsky, Sov. Phys. JETP **68**, 101 (1989); A.Bogdanov and A.Hubert, J. Magn. Magn Mater. **195**, 182 (1999).
10. A.N.Bogdanov and A.A.Shestakov, JETP **86**, 911 (1998).
11. A.Bogdanov, JETP Lett. **62**, 247 (1995); **68**, 317 (1998).
12. T.Nagaya, H.Hotta, H.Orihara, and Y.Ishibashi, J. Phys. Soc. Japan **60**, 1572 (1990).