Possible nature of anomalies in the physical characteristics of a liquid-crystal cell near the Fréedericskz effect threshold

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Possible reasons for the anomalies of the properties of homeotropic Fréedericskz cell near threshold are studied and it is shown that for real cell dimensions defects can make the largest contribution.

INTRODUCTION

The foundations of the theory of the basic types of orientational structural transitions in a uniformly oriented plane-parallel slab of a uniaxial nematic liquid crystal (NLC) in an external electric E (or magnetic H) field—referred to collectively as the Fréedericskz effect—were laid in the 1930s[1,2] and the theory was finally completed in the 1960s.[3,4] Later, cases of complex deformation of a liquid crystal—structures[5,6] which include jumps in the order parameter[1)] ϑ_m near the threshold field—were described. In the mid-1970s Guyon[7] drew an analogy between the Fréedericskz effect in a planar NLC slab and a second-order phase transition. The analogy is based on the fact that the tilt angle of the director at the center of the cell and the order parameter in a phase transition exhibit the same square-root dependences $\vartheta_m \simeq (E-E_*)^{1/2}$ and $\eta \simeq (T-T_*)^{1/2}$, respectively. It was recently shown[8,9] that in the problem of the Fréedericskz effect with canted orientation of the NLC in crossed electric (E) and magnetic (H) fields it is possible to determine the conditions under which both a first-order structural transition, previously observed experimentally in Ref. 10, and an isostructural transition, which still has not been observed experimentally, appear. Thus the types of structural transitions obtained from Landau's theory for systems with a single-component order parameter have been exhausted.

By analogy to phase transitions, it is of interest to investigate how fluctuations of the order parameter affect the field dependence of the order parameter near threshold. This is an important question because, as far as we know, the question of the correctness of the phenomenological theory near threshold has never been discussed in theoretical works on the Fréedericskz effect while the experimental works[3,11] do not make it possible to compare the field dependence of the order parameter $\vartheta_m(E)$ near threshold with the square-root function $(E-E_*)^{1/2}$ implied by the phenomenological theory. In addition, experimental observations[12] of anomalies of the birefringence in NLCs near the Fréedericskz transition show that taking into account only a uniform static canting of the director may be insufficient for explaining the experimental results correctly.

In the case of phase transitions in three-dimensional crystals it is often found[13,14] that anomalies in the temperature dependence of the order parameter, the intensity of light scattering, and so on are caused not so much by thermal fluctuations as by defects of the crystal lattice, which produce near the transition point a nonuniform and strongly temperature-dependent distribution of the order parameter. A liquid-crystal cell can also contain defects—volume defects (topological—disclinations, hedgehog; mechanical—foreign inclusions) and surface defects (topological—boojums; mechanical—nonuniformity of boundary conditions at the surface). In this connection it is of interest to investigate the effect of defects on the field dependence of the order parameter near the Fréedericskz threshold.

Two possible mechanisms for the anomalous behavior of the physical characteristics of a liquid-crystal cell are thermal fluctuations of the director of the NLC and distortion of the uniform field of the director by defects. These anomalies are strongest near the Fréedericskz threshold. They include a change in the critical indices of the physical quantities (order parameter, correlation length, and so on), displacement of the Fréedericskz effect threshold, etc. The competition between these two mechanisms leads to a criterion for one of them to predominate.

The object of the present work is to take into account the thermal fluctuations of the director and distortions of the uniform field of the director by defects of different types in the theory of Fréedericskz effect.

THERMAL FLUCTUATIONS OF THE DIRECTOR OF AN NLC

Consider an infinite plane-parallel slab of a homeotropically oriented NLC in a uniform electric field E oriented in the plane of the liquid-crystal slab and with rigid anchoring of the director n^0 at the boundaries. Without loss of generality, we employ the single-constant approximation of Frank's theory, making the assumption that the stationary tilting $\vartheta_0(z)$ is a function of z only, and we consider fluctuations $\delta\vartheta(x,y,z)$ of the tilt angle ϑ.

The free energy of the deformed liquid-crystal slab in an external field is

$$\mathcal{F} = \int\int_S dx\,dy \int_0^L F\,dz, \tag{1}$$

where L is the thickness of the liquid-crystal slab and S is the surface area of the cell ($S \gg L^2$). The free-energy density is

$$F = K(\mathrm{div}^2 \mathrm{n} + \mathrm{rot}^2 \mathrm{n}) - \chi_a(\mathrm{nE})^2, \tag{2}$$

where χ_a is the anisotropy of the permittivity and K is Frank's elastic constant.

Switching to the tilt angle $\vartheta(x,y,z)$ of the director n away from the initial homeotropic orientation $\mathbf{n}^0$ and drop-

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ping terms of order higher than ϑ^4 in the power series expansion we obtain

$$F/K = (\nabla\vartheta)^2 - q^2(E/E_*)\vartheta^2 + q^2(E/E_*)^2\vartheta^4/3, \qquad (3)$$

where $q=\pi/L$ and $E_* = q[(4\pi K/\varepsilon_a)]^{1/2}$ is the threshold field of the Fréedericskz effect. Expanding $\vartheta(x, y, z)$ in a Fourier series taking into account the boundary conditions we have

$$\vartheta(x,y,z) = \sum^{\infty} \sin(nqz)\frac{1}{\sqrt{S}}\sum_{\mathbf{k}} \vartheta_{n,\mathbf{k}}\exp(i\mathbf{k}\boldsymbol{\rho}), \qquad (4)$$

where $\boldsymbol{\rho} = (x,y)$. In the quadratic approximation in $\vartheta_{n,\mathbf{k}}$

$$F = \frac{\pi^2 K}{2L}\sum_{n=1}^{\infty}\sum_{\mathbf{k}} [n^2 - (E/E_*)^2 + \mathbf{k}^2/q^2]\,|\vartheta_{n,\mathbf{k}}|^2. \qquad (5)$$

With the help of Eq. (5) we find the mean-square fluctuation $\langle|\vartheta_{n,\mathbf{k}}|^2\rangle$:

$$\langle|\vartheta_{n,\mathbf{k}}|^2\rangle = \frac{1}{q^2[n^2 - (E/E_*)^2 + \mathbf{k}^2/q^2]}\,\frac{2T}{KL}. \qquad (6)$$

In r space we have the correlation function

$$G(z, z_0, \boldsymbol{\rho}-\boldsymbol{\rho}_0) = \langle\vartheta(\mathbf{r})\vartheta(\mathbf{r}_0)\rangle = \frac{4T}{KL}\sum_{n=0}^{\infty}\sin(nqz)\sin(nqz_0)K_0\left(\frac{|\mathbf{x}-\mathbf{x}_0|}{\xi_n}\right), \qquad (7)$$

where

$$\xi_n = \frac{L}{\pi(n^2 - E^2/E_*^2)^{1/2}} \qquad (8)$$

is the correlation length for the nth mode and $K_0(x)$ is a modified Bessel function.

Near the Fréedericskz transition the mode $n=1$, for which the correlation radius becomes infinite at $E=E_*$, makes a significant contribution to $G(\mathbf{r}, \mathbf{r}_0)$. Confining our attention to this mode $\vartheta_1 = \vartheta$, Landau's theory is applicable for $\langle(\vartheta - \langle\vartheta\rangle)^2\rangle \ll \langle\vartheta\rangle^2$, which can be written as

$$\frac{2T}{KL}\,|\ln|1-(E/E_*)^2||\ll 1-(E/E_*)^2. \qquad (9)$$

At temperature $T\sim300$ K and for a liquid crystal with the elastic constant $K\sim10^{-12}$ N and thickness $L\sim10^{-6}$ m we have

$$1-(E/E_*)^2 \gg f(2T/KL), \qquad (10)$$

where $f(x)$ is the function inverse to $x/|\ln x|$. In this case the quantity $f(2\tau/KL)$ plays the role of the Ginzburg number Gi, which for the chosen parameters of the liquid crystal is $\sim10^{-3}$–10^{-2}. Thus the critical fluctuations of the director must be taken into account when $1-(E/E_*)^2 \leqslant$ Gi. In this range only the critical mode $n=1$ need be included. For it Landau's potential, up to terms of order ϑ^4, has the form

$$F = \frac{KL}{2}\int d^2x\{(\nabla\vartheta)^2 + q^2[1-(E/E_*)^2]\,\vartheta^2 + q^2(E/E_*)^2\vartheta^4/4\}. \qquad (11)$$

Introducing the normalization $\vartheta = \phi/\sqrt{(KL/T)}$ we have for $H=F/T$ the standard expression of the fluctuation theory

$$H = \frac{1}{2}\int d^2x[\tau\phi^2 + (\nabla\phi)^2 + g\phi^4], \qquad (12)$$

where $\tau = q^2[1-(E/E_*)^2] = \xi^{-2}$ and $g = q^2(E/E_*)^2(T/KL)$. In the first ε approximation[15] we obtain the exponent $\nu = 2/3$ for the correlation radius and $\beta = 1/6$ for the order parameter:

$$\xi \sim |E-E_*|^{-\nu}, \qquad \phi\sim|E-E_*|^{\beta}.$$

We note that the fluctuation region for thin samples, even though it is narrow, may still be wide enough to be observed experimentally.

DEFECTS

The influence of defects is taken into account in the theory of phase transitions by introducing into the thermodynamic potential local terms (different from zero near a defect) which describe the interaction of a defect with the order parameter (OP) ϕ. Defects are usually divided into two classes depending on the character of this interaction:[16]

1. Defects of the type "local temperature"—the interaction with the OP is described by the term $A(\mathbf{r})\eta^2$, i.e., near a defect the local transition temperature $\tilde{T}_c$ is different from the temperature T_c in the bulk. For the Fréedericskz transition the external electric field **E** or magnetic field **H** plays the role of the temperature T.

2. A defect of the type "local field"—the interaction with the OP is described by the term $B(\mathbf{r})\eta$ or the value of the OP on a defect is given. In this case the value of the OP on a defect is different from zero at any temperature.

In the case of Fréedericskz transitions local-temperature defects are local changes in the interaction energy between a molecule of the liquid crystal and the surface of the cell and local-field defects are regions where the boundary conditions differ from a uniform orientation at the surface of the liquid crystal (in the case of rigid anchoring to the surface) or disclinations and other defects, which disturb the distribution of the director in the volume of the liquid crystal.

We consider first local-temperature defects. As in the preceding section, we are considering a Fréedericskz transition in a homeotropically oriented slab of a NLC. The defects are taken into account by adding to the integrand in Eq. (11) the nonuniform term $A(\boldsymbol{\rho})\vartheta^2$, where $\boldsymbol{\rho} = (x,y)$. For definiteness, for a single defect at the point $x=0$ we choose $A(\boldsymbol{\rho}) = A_D\theta(R-\rho)$, where $\rho = |\boldsymbol{\rho}|$, R is the radius of the defect, A_D is a factor characterizing the local "transition threshold" inside a defect, and $\theta(x)$ is the unit step function.

Switching to dimensionless variables

$$\eta = \vartheta(2/|1-(E_*/E)^2|)^{1/2},$$

$$\mathbf{x} = \boldsymbol{\rho}/\xi, \qquad \mathcal{F} = F\frac{\pi[1-(E_*/E)^2]}{2q},$$

we obtain

$$\mathcal{F} = \int\int d^2x\{(\nabla\eta)^2 \pm \eta^2 + \eta^4/2 + \tau_*(\mathbf{x})\eta^2\}, \qquad (13)$$

where

$$\tau_*(\mathbf{x}) = \xi^2 A(\boldsymbol{\rho}) = \tau_D\theta(a - x), \qquad (14)$$

$\tau_D = \xi^2 A_D$ and $a = R/\xi$. The " + " sign in Eq. (13) corresponds to a subthreshold field and the " − " sign corresponds to a field above threshold. We note that near threshold

$$\xi \to \infty, \quad \tau_*(x) \to \pi f\delta(x), \quad f = a^2\tau_D = R^2 A_D \qquad (15)$$

where the quantity f characterizes the strength of the defect. Below threshold only defects with $\tau_D > 0$ contribute to $\eta(\mathbf{x})$, and it is these defects that are considered. Inside the defect $(x < a)$ (15) the equation of equilibrium

$$\Delta\eta - \eta[1 - \tau_*(\mathbf{x})] - \eta^3 = 0 \qquad (16)$$

assumes the form

$$\Delta\eta + \eta(\tau_D - 1) - \eta^3 = 0. \qquad (17)$$

We consider below quite weak defects, so that a nonzero distribution $\eta(\mathbf{x})$ arises for quite large ξ, i.e., for sufficiently large $\tau_D \gg 1$. In this case, as will become evident below, not too close to threshold $\eta(\mathbf{x}) \lesssim 1$ and the term η^3 in Eq. (16) can be neglected. In this approximation

$$\eta(\mathbf{x}) = CJ_0(x(\tau_D - 1)^{1/2}), \qquad (18)$$

where $J_0(x)$ is a Bessel function.

Outside the defect $(x > a)$ the equation (15) assumes the form

$$\Delta\eta - \eta = \eta^3. \qquad (19)$$

We solve Eq. (18) by the iteration method $\eta = \eta_0 + \eta_1 + \ldots$. In the zeroth (linear) approximation $\eta_0 = UK_0(x)$, where U is a constant factor and $K_0(x)$ is a modified Bessel function of second order. In the first approximation Eq. (18) assumes the form

$$\Delta\eta_1 - \eta_1 = \eta_0^3. \qquad (20)$$

With the help of the Green's function of the left-hand side we have

$$\eta_1 = -\frac{1}{2\pi}\int\int d^2rK_0(|\mathbf{x} - \mathbf{r}|)\eta_0^3(x)$$

$$= -\frac{U^3}{2\pi}\int\int d^2rK_0(|\mathbf{x} - \mathbf{r}|)K_0^3(r). \qquad (21)$$

Matching $\eta(x)$ and the derivative at the boundary of the defect we obtain from Eqs. (17) and (20) the following system of equations for the coefficients C and U:

$$\alpha C = \beta U - \gamma U^3, \quad \alpha_1 C = \beta_1 U - \gamma_1 U^3, \qquad (22)$$

where

$$\alpha = J_0(a(\tau_D - 1)^{1/2}), \quad \beta = K_0(a),$$

$$\gamma = \frac{1}{2\pi}\int\int d^2rK_0(|\mathbf{x} - \mathbf{r}|)K_0^3(r)|_{x=a},$$

$$\alpha_1 = J_1(a(\tau_D - 1)^{1/2}), \quad \beta_1 = K_1(a),$$

$$\gamma_1 = \frac{1}{2\pi}\int\int d^2rK_1(|x - r|)K_0^3(r)\frac{a - r_1}{|\mathbf{x} - \mathbf{r}|}\bigg|_{\mathbf{x}=(a,0)}.$$

The solution of Eqs. (22) is

$$U^2 = \frac{\alpha\beta_1 - \alpha_1\beta}{\alpha\gamma_1 - \alpha_1\gamma}, \quad C/U = \frac{\beta\gamma_1 - \beta_1\gamma}{\alpha\gamma_1 - \alpha_1\gamma}. \qquad (23)$$

Nonzero values of $\eta(x)$ near a defect appear if $\alpha\beta_1 - \alpha_1\beta < 0$ or, for weak defects $(f \ll 1)$, if $\xi > R\exp(2/f)$.

The solution obtained for Eq. (20) by the iteration method, which is applicable for $\eta \lesssim 1$, gives the threshold for the appearance of nonzero threshold $\eta(\mathbf{x})$ near the defect, but it does not permit investigating the behavior of $\langle\eta^2(\mathbf{x})\rangle$ near the threshold field E_*, i.e., in the limit $\xi \to \infty$, where $\eta(\mathbf{x}) \gg 1$ near the boundary of the defect and Eq. (20) is strongly nonlinear. In this region the term η in Eq. (20) can be neglected compared with η^3. The equation so obtained has the following asymptotic solutions $\eta(\mathbf{x})$:

$$\eta(\mathbf{x}) \approx \begin{cases} 1/x, & a < x \ll 1 \\ AK_0(x), & x \gg 1 \end{cases}. \qquad (24)$$

For $x < a$ the function $\eta(\mathbf{x})$ does not have any singularities, and for this reason when calculating $\langle\eta^2(\mathbf{x})\rangle = \int_0^\infty xdx\eta^2(\mathbf{x})$ the region $(a, 1)$, where the integral diverges logarithmically, makes the main contribution to the integral. As a result, for noninteracting defects we have

$$\langle\eta^2(\mathbf{x})\rangle \simeq -N\ln a = N\ln(\xi/R), \qquad (25)$$

where N is the two-dimensional concentration of defects.

Switching to the tilt angle ϑ we obtain the same result

$$\langle\vartheta^2(\mathbf{x})\rangle \simeq N\ln(\xi/R). \qquad (26)$$

Above the threshold E_* the equation of equilibrium has the form

$$\Delta\eta + \eta[1 + \tau_*(\mathbf{x})] - \eta^3 = 0. \qquad (27)$$

An analysis, similar to the one performed above, for the case below threshold (16) also leads to a logarithmic correction

$$\langle\Delta\vartheta^2(\mathbf{x})\rangle \simeq N\ln(\xi/R). \qquad (28)$$

For local-field defects the value of the order parameter at the boundary of a defect is given. Outside the defect the distribution of the order parameter in dimensionless variables is determined by Eq. (19). The distribution of the order parameter is given by the same equation (24) as for defects of the local-temperature type. Correspondingly, local-field defects make exactly the same contribution to the temperature dependence of $\langle \vartheta^2(x) \rangle$ as do local-temperature defects.

CONCLUSIONS

Comparing the fluctuation and defect-related contributions to the anomalies shows that fluctuations are important only in a very narrow region near threshold, though they give a power-law anomaly. Defects give logarithmic anomalies, but the region where the corrections are significant depends on the strength of the defects and could be quite wide for sufficiently strong defects. The numerical values of the input parameters depend on the technology employed to prepare the cell. The corresponding data can be estimated from optical observations near the Fréedericskz threshold. Due to the large value of the correlation radius $\xi = L[1-(E/E_*)^2]^{-1/2}$ the defects can be seen under a microscope. In this sense the Fréedericskz cell can serve as a two-dimensional model system for investigating the influence of defects on phase transitions.

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[1)]In the Fréedericskz effect, under symmetric boundary conditions, the tilt angle ϑ_m of the director at the center of the slab is used as the order parameter for describing the deformed structure.

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