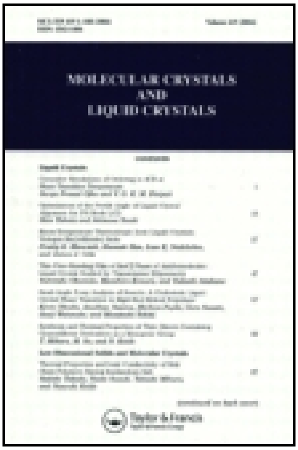

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Field-Induced Orientational Transitions in a Nematic with Chiral Dopant

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In the nematic cell with chiral dopant and homeotropic anchoring can exist the next one-dimensional distributions of the director: 1) homeotropic, with the director normal to the plates; 2) twisted with tilted orientation of the director; 3) twisted with the director parallel to the surfaces. Stability analysis shows that the transitions might occur between these distributions. The dimensionless parameters controlling the director field are the ratio of the pitch of the twisted nematic structure to the cell thickness and ratio of the cell thickness to the extrapolation length of the nematic material.

Keywords: liquid crystals; stability; chiral nematic

INTRODUCTION

Study of the director distributions in cholesteric liquid crystals (LCs) and doped with chiral agent nematic LCs with finite anchoring energy on the boundary surfaces still remains the actual problem for both the theoretical description and experimental investigations.

The simplest geometries of the director distributions in the chiral nematic LCs such as *homeotropic* (director is normal to the bounded plates) and *twisted planar* (director rotates parallel to the bounded surfaces) were investigated because of their practical interest in the realization of LC's displays[1]. B.Zel'dovich, N.Tabiryan[2] shown that the homeotropic structure in the cell with rigid homeotropic anchoring is unstable above the definite concentration of the chiral dopant. The transition from the homeotropic to twisted alignment was found to be the second-order in the one-elastic-constant

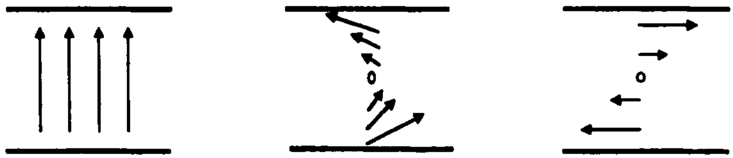

FIGURE 1 Possible one-dimemsional director distributions of chiral nematic

approximation and the first order for the typical LC's elastic constants. More recently it was proved experimentally that bubbles of cholesteric phase appear in the nematic matrix[3].

The subject of our consideration is one-dimensional distributions of the director in the cell with chiral nematic LC and finite *homeotropic* anchoring. On the list of problems one has to clarify, number one stands the problem of classification of distributions and investigation of their stability. The solution of this problem is essential to understand properties and behavior of the chiral nematic structures in the real experimental conditions. Until now the answer has been known only for two limiting cases, namely, of strong and weak anchoring. Herein we present the extended solution of the problem valid for an arbitrary anchoring strength.

The situation with the finite anchoring energy results in the qualitatively new effects in comparison with structural transitions in the cell with infinite anchoring. The matter is that under the rigid homeotropic anchoring twisted distribution with the director parallel to the bounded surfaces is forbidden because of the homeotropic boundary conditions. At the same time this configuration is possible in the case of finite anchoring energy and, moreover, can be stabilized by the external electric or magnetic field. This complicates the diagram of possible orientational structures giving one more region which corresponds to the planar twisted distribution of director.

EQUATIONS FOR THE DIRECTOR

Let us formulate the problem we are dealing with in more detail. We consider a nematic LC in the magnetic field doped with chiral agent and placed between two bounding plates. The plates are able to orient or "anchor" the adjacent liquid crystal. The degree of that anchoring is characterized by the surface energy density F_s, that is, the anisotropic part of the surface tension for a given pair of the solid substance and liquid crystal. To describe it, we shall use a simple model expression, first proposed by Rapini[4], written as

$$F_s = -\tfrac{1}{2} W \cos^2 \gamma$$

where W is the characteristic energy of the surface director perturbation, γ is the deviation of actual director orientation from the direction demanded by the surface free energy. As is well known, W may be, and often is, easily modified by special treatment of the solid surfaces[5].

Since we consider not the pure nematic LC, but doped with chiral molecules, the infinite specimen of it tends to have the twisted structure with the pitch p, which depends on the twisting power of chiral molecules and their concentration. In the small dopant concentration limit the pitch of this structure is defined as $p = 2\pi / t = (4\pi\beta c)^{-1}$, where t is the chirality of the dopant, c is its concentration, β is the microscopic twisting power[6].

To obtain the expression for the free energy of the LC cell, one should sum up both the volume and surface contributions of the orientational-elastic origin and integrate them over the LC specimen volume and the alignment surface, respectively. Further we consider one-dimensional distributions, that is, they assumed to be homogeneous along x and y coordinates and therefore depend only on z coordinate.

Let the z axis be normal to the aligning plates which provide homeotropic anchoring with strength W and the magnetic field $\vec{H}$ is applied parallel to the z axis. The orientational field can be written in the form $\vec{n} = (\cos\varphi \sin\theta, \sin\varphi \sin\theta, \cos\theta)$, $\varphi = \varphi(z), \theta = \theta(z)$, which transforms the cell free energy into[6,7]

$$F = F_b + F_s + F_H = \int g dV, \qquad (1)$$

$$F_b = \tfrac{1}{2} S \int_0^L \left\{ K_1 \dot\theta^2 \sin^2\theta + K_2 \left(\dot\varphi \sin^2\theta - t \right)^2 + K_3 \cos^2\theta \left(\dot\theta^2 + \dot\varphi^2 \sin^2\theta \right) \right\} dz,$$

$$F_s = -\tfrac{1}{2} W \int \cos^2\theta dS, \quad F_H = -\tfrac{1}{2} \chi_a H^2 S \int_0^L \cos^2\theta dz$$

Here $\dot\theta = \partial\theta / \partial z, \dot\varphi = \partial\varphi / \partial z$, χ_a is a magnetic susceptibility.

Minimization of the functional (1) with respect to the angles θ and φ gives the following equations for the director distribution and the boundary conditions

$$\begin{cases} \ddot\theta f_{13}(\theta) - \sin 2\theta \left[\tfrac{1}{2}(K_3 - K_1)\dot\theta^2 + \tfrac{1}{2}\{K_3 - 2(K_3 - K_2)\sin^2\theta\}\dot\varphi^2 - K_2 t \dot\varphi \right] - \\ - \tfrac{1}{2}\chi_a H^2 \sin 2\theta = 0 \\ \ddot\varphi f_{23}(\theta) \sin^2\theta + \dot\theta \dot\varphi \left[K_3 - 2(K_3 - K_2)\sin^2\theta \right] \sin 2\theta - \dot\theta K_2 t \sin 2\theta = 0 \end{cases} \qquad (2)$$

$$\begin{cases} f_{13}(\theta)\dot\theta \pm W\cos\theta\sin\theta\big|_{z=L,0} = 0 \\ f_{23}(\theta)\dot\varphi - K_2 t\big|_{z=L,0} = 0 \end{cases} \tag{3}$$

i.e. the non-linear second-order equations accompanied with the specific boundary conditions. Here $f_{ij}(\theta) = K_i \sin^2\theta + K_j \cos^2\theta$.

The second bulk equation for the azimuthal part of director distribution (2) can be easily integrated (the free energy density g in (1) does not depend explicitly on the azimuthal director angle, thus $\partial g/\partial\dot\varphi = const$) and gains the form

$$\dot\varphi = \frac{A + tK_2 \sin^2\theta}{f_{23}(\theta)\sin^2\theta}. \tag{4}$$

Taking into account the boundary conditions (3) we obtain that $A = 0$.

Thus, the symmetry of the boundary conditions together with the symmetry of bulk equations gives the opportunity to eliminate the azimuthal director angle from the consideration. The azimuthal part of the orientational field is determined now by the polar part and can be obtained from (4) if the boundary problem for polar director angle is solved. Further we pay the special attention to the polar part of the orientational field.

Substituting the integral (4) to the first bulk equation (2) we obtain the boundary problem for the polar director angle θ

$$f_{13}(\theta)\ddot\theta - \tfrac{1}{2}\dot\theta^2(K_3 - K_1)\sin 2\theta + K_3(K_2 t)^2 \frac{\sin 2\theta}{2 f_{23}(\theta)^2} - \tag{5}$$
$$- \tfrac{1}{2}\chi_a H^2 \sin 2\theta = 0$$

$$f_{13}(\theta)\dot\theta \pm W\cos\theta\sin\theta\big|_{z=L,0} = 0 \tag{6}$$

Solutions of the boundary problem (5,6) give possible one-dimensional distributions of director in the cell. Note, that (5,6) does not necessarily lead to stable solutions, and hence, to the experimentally realized distributions of director. The analysis of stability should be the next step after the solution has been derived. In what follows we perform such an analysis.

HOMEOTROPIC DIRECTOR DISTRIBUTION

There are some apparent solutions to the boundary problem (5,6). The first one, $\theta = 0$, corresponds to the homeotropic alignment of the director and is natural for the homeotropic anchoring. It can be destabilized by the twist torque of the chiral dopant. Stability of this solution was discussed earlier[7] in the infinite anchoring limit. It was found that homeotropic structure is stable if

$tL < \pi K_3/K_2$ i.e. if the spiral pitch of the twisted nematic structure is less than the cell thickness. In the opposite case the tilted twisted distribution is realized in the cell. Transition between these distributions was found to be the first order with hysteresis if $K_1 < 3(K_3 - K_2)$.

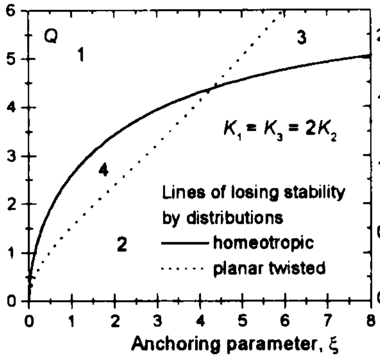

FIGURE 2 Typical LCs: 1 - twisted planar; 2- homeotropic region; 3 - tilted twisted structure; 4 - bistable homeotropic - twisted planar region

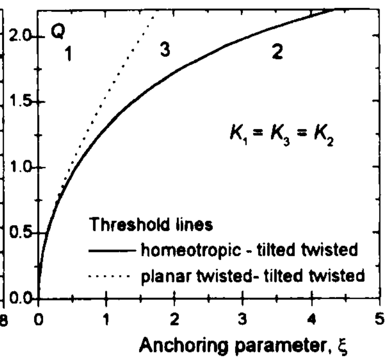

FIGURE 3 One-elastic-constant approximation: 1 - twisted planar; 2 - homeotropic region; 3 - region with tilted twisted structure

Stability analysis in the cell with finite anchoring can be carried out by investigating the small director deviations. The equations and boundary conditions (5,6) for the small deviations of the director from the homeotropic alignment $\theta = 0 + \delta\theta$ take the form

$$\begin{cases} K_3\delta\ddot{\theta} + \left\{(tK_2)^2/K_3 - \chi_a H^2\right\}\delta\theta = 0 \\ K_3\delta\dot{\theta} \pm W\delta\theta\big|_{z=L,0} = 0 \end{cases}$$

Seeking the non-trivial solution as $\delta\theta = C_1\sin(\lambda z) + C_2\cos(\lambda z)$ we obtain the equation for the transition line, where the homeotropic alignment becomes unstable

$$\xi^h = Q\tan(Q/2) \qquad (7)$$

Here $Q^2 = (K_2/K_3)T^2 - \chi_a H^2 L^2/K_3$, and we introduce the dimensionless ratios $\xi = WL/K_3$ (anchoring parameter) and $T = tL$ (chiral parameter). The dependence (7) is shown in Fig.2 for the typical LCs $(K_1 = K_3 = 2K_2)$.

In the case of the strong anchoring ($\xi^h \to \infty$) the critical chiral parameter which makes the homeotropic distribution unstable is given by expression $Q = \pi$ and is in full agreement with the results obtained in[7].

TWISTED PLANAR DISTRIBUTION

The other apparent solution of the boundary problem (5,6), $\theta = \pi / 2$, is absent under the infinite anchoring which constrains director to be perpendicular to the boundaries. The director distribution, correspondent to this solution, is similar to that one realized in the twisted nematic cell with the planar anchoring, with director parallel to the bounded surfaces and linear dependence of azimuthal director angle φ on z coordinate, $\varphi = tz + \varphi_0$.

Clearly, this distribution is not stable in the absence of magnetic field: rotation of the LC bulk on a small angle does not change the bulk free energy (the director distribution remains unchanged) but decreases the surface free energy, because we partially satisfy the boundary conditions. Nevertheless, we can stabilize this distribution applying suitable electric or magnetic field.

Thus, further we assume that the magnetic field $\vec{H}$ is applied parallel to the z axis (normal to the bounding plates) and the LC possesses a magnetic susceptibility $\chi_a < 0$. In this case, the magnetic field orients LC molecules parallel to the cell boundaries, stabilizing the planar twisted director distribution. Rotation of the LC bulk on an angle α leads to the increase of the cell free energy due to term $F_H = -\frac{1}{2}\chi_a H^2 \sin^2\alpha V$ of the magnetic origin and decrease due to term $F_S = \frac{1}{2} W \sin^2\alpha S$ of the surface origin. Here V and S are the cell volume and square of the bounding surface correspondingly. The rotation is energetically unfavorable, if $F_H > F_S$, i.e. only the fields $H > \left(W/|\chi_a| L\right)^{1/2}$ can stabilize the planar twisted distribution. Increasing of anchoring makes this distribution unstable and leads to the threshold reorientation to the alignment with tilted twisted orientation of director or directly to the homeotropic alignment.

Pursuing the scheme adopted in Sec.3 we can obtain the linearized equation and boundary conditions for director deviations near the twisted planar structure ($\theta = \pi / 2 + \delta\theta$)

$$\begin{cases} K_1 \delta\ddot{\theta} - \left(K_3 t^2 - \chi_a H^2\right)\delta\theta = 0 \\ K_1 \delta\dot{\theta} \pm W \delta\theta\big|_{z = L, 0} = 0 \end{cases}$$

Now we must try the non-trivial solution as $\delta\theta = C_1\exp(\lambda z) + C_2\exp(-\lambda z)$ and the final equation for the transition line from twisted planar distribution is (Fig.2)

$$\xi^p = Q\tanh(Q/2), \qquad (8)$$

where $Q^2 = (K_3/K_1)T^2 - \chi_a H^2 L^2/K_1$. Thus, if the planar twisted distribution is the distribution realized in the cell, it becomes unstable if the anchoring parameter exceeds that one, defined by expression (8): $\xi > \xi^p(Q)$.

Comparison of the asymptotic behaviors of the stability lines of the homeotropic and twisted planar alignments shows, that for the typical LC elastic constants ($K_1 = K_3 = 2K_2$) there is the range of anchoring and chiral parameters where both homeotropic and twisted planar configurations are stable (two-phase region, Fig.2). Hence, the first - order transition between these states can exist.

TILTED TWISTED DISTRIBUTION. CHARACTER OF TRANSITIONS

To continue the analysis of the possible one-dimensional configurations in doped nematic LC let find the distribution with tilted orientation of director on the surfaces and investigate its stability. To simplify our results, we shall use a one-elastic-constant approximation setting $K_1 = K_2 = K_3 = K$.

In this frame the equation and the boundary conditions (5,6) for the polar director angle are simplified to

$$\begin{cases} K\ddot{\theta} + \left(Kt^2 - \chi_a H^2\right)\sin\theta\cos\theta = 0 \\ K\dot{\theta} \pm W\cos\theta\sin\theta\big|_{z=L,0} = 0 \end{cases} \qquad (9)$$

The first integral of this boundary problem can be obtained from the free energy density (1), since the latter does not depend explicitly on z coordinate and permits, in accordance to Noether theorem, the integral of 'energy': $\dot{\theta}\,\partial g/\partial\dot{\theta} + \dot{\varphi}\,\partial g/\partial\dot{\varphi} - g = const$. As a result we have

$$L^2\left(\frac{\partial\theta}{\partial z}\right)^2 = \left(\xi\frac{\sin 2\theta_L}{2}\right)^2 + T^2\left(\sin^2\theta_L - \sin^2\theta\right), \qquad (10)$$

where $\xi = WL/K$ is the anchoring parameter, $\theta_L = \theta(z = L)$ is the tilt angle.

Analysis of the equation (10) can be performed in several ways. The usual procedure needs the evaluation of director distribution in the cell as the solution to equation (10) and further integration of the free energy (1) which corresponds to this distribution. Investigation of the obtained free energy as a function of chiral and anchoring parameters allows to determine stability of considered solution. Unfortunately it is difficult to realize this scheme in practice, since solutions of equation (10) are the special functions and their integration is troublesome.

Another way is to obtain the parameters of director distribution such as tilt angle, polar director angle in the middle of the cell, and study their behavior under the change of anchoring and chiral parameters. This way is less intuitive but more convenient for us because we operate only with the analytical form of solutions to (10). Thus, further we follow this strategy in the analysis of solutions.

It should be mentioned that considered symmetric cell can have several distributions of polar director angle, because the boundary conditions have already been satisfied in the first integral (10), but it still remains the first-order differential equation and has the undefined constant of integration. Thus, further specification of solution should be performed manually. We assume that the polar director angle θ changes within the range $[\theta_L, \theta_{max}]$ and the maximum polar distortion θ_{max} is reached in the middle of the cell $z = L/2$. This assumption is dedicated by the choice of the distribution with the lowest free energy.

Thus, integrating (10) from $z = 0$ to $L/2$ and observing that $d\theta/dz = 0$, where θ takes on its maximum value, we obtain the implicit equation for determination of the tilt director angle θ_L and the angle of the maximum distortion in the middle of the cell θ_{max}

$$\int_{\theta_L}^{\theta_{max}} \frac{1}{\sqrt{A^2 - T^2 \sin^2\theta}} d\theta = \frac{1}{2} \qquad (11)$$

where $A^2 = \sin^2\theta_L(\xi^2 \cos^2\theta_L + T^2)$, $\sin\theta_{max} = A/T$.

Integrating (11) we have

$$T = 2(K(m) - F(\gamma, m)) \qquad (12)$$

where $F(\gamma, m) = \int_0^\gamma (1 - m^2 \sin^2 x)^{-1/2} dx$, $K(m) = F(\pi/2, m)$ is the incomplete and complete elliptic integrals of the first kind, $m = \sin\theta_{max}$, $\sin\gamma = \sin\theta_L / \sin\theta_{max}$. Using Jacobian elliptic functions equation (12) can be rewritten in the form

$$\xi = T \frac{\mathrm{sn}(\frac{1}{2}T, m)\,\mathrm{dn}(\frac{1}{2}T, m)}{\mathrm{cn}(\frac{1}{2}T, m)} = T\sqrt{\frac{1 - \mathrm{cn}(T, m)}{1 + \mathrm{cn}(T, m)}} \qquad (13)$$

$$\sin\theta_L = m \frac{\mathrm{cn}(\frac{1}{2}T, m)}{\mathrm{dn}(\frac{1}{2}T, m)} \qquad (14)$$

Let investigate the behavior of θ_L and θ_{max}, which completely define distribution in the cell, as the function of anchoring and chiral parameters.

Equation (13) defines implicit dependence $\theta_{max}(\xi, T)$. Moreover, it is seen from (13), that $\xi(m, T)$ is the monotone function of parameter m for each definite chiral parameter T, because of monotony of $\mathrm{cn}(x, m)$, and maximum and minimum of this function are reached at the boundaries $m = 0,1$. The solution for m exists for the anchoring parameter in the range $\xi(m=1) \le \xi \le \xi(m=0)$ and corresponds to the stable tilted twisted orientation of director. Out of this range only solutions $m = 0,1$ are possible. Thus the boundaries $m = 0,1$ define the critical anchoring as the function of chiral parameter and correspond to the reorientation from the tilted twisted distribution of director to the homeotropic ($m = 0$, $\theta_L = \theta_{max} = 0$) or planar twisted ones ($m = 1$, $\theta_L = \theta_{max} = \pi/2$).

Taking into account that $\mathrm{sn}(x,0) = \sin x$, $\mathrm{cn}(x,0) = \cos x$, $\mathrm{dn}(x,0) = 1$, $\mathrm{sn}(x,1) = \tanh x$, $\mathrm{cn}(x,1) = \mathrm{dn}(x,1) = \mathrm{sech}\, x$ and substituting $m = 0,1$ to (13) we obtain expressions for the transition lines $\xi = T\tan(\frac{1}{2}T)$ for the transition between homeotropic and tilted twisted distribution, and $\xi = T\tanh(\frac{1}{2}T)$ for the transition between twisted planar - tilted twisted ones. These expressions are the same to those obtained for the inverse transitions (7),(8) and prove once again the second-order character of transitions.

Therefore, in the one-elastic-constant approximation, we have three possible distributions of the director and the transitions of the second kind between them. The direct transition between twisted planar and homeotropic distributions is forbidden and can occur only through the tilted twisted distribution. Corresponding diagram of orientational states includes three regions separated by two second-kind transition lines (Fig.3).

The situation is much less intuitive for the semi-isotropic LCs. We can expect, that the anisotropy in Frank elastic constants may lead to the appearance of first-order transitions between all modes[2] but to obtain the expressions for the transition parameters is troublesome even numerically.

CONCLUSIONS

We investigated one-dimensional distributions of director in the symmetric cell with the twisted nematic LC and possible transitions between them under the variation of director anchoring energy and chirality of the dopant. Three types of distributions was found to be the most energetically favorable: homeotropic, tilted twisted and twisted planar ones. The last one is forbidden under the rigid homeotropic anchoring and give new region to the diagram of orientational states.

In the one-elastic-constant approximation transitions between these distributions were found to be second-order ones. Transition between twisted planar and homeotropic distributions can occur through the tilted twisted one.

We considered only one-dimensional distributions of director i.e. the orientational field depended only on one of the spatial coordinates. At the same time, characteristic textures, appearing in the homeotropically oriented LCs doped with chiral agents are more complicated under the large chiral parameters and not one-dimensional. Thus, the phase diagram, calculated above for the one-dimensional distributions should be completed with two and three-dimensional ones, and the final decision about the practical realization of the obtained one-dimensional structures must be given by experiment.

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