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Transformation of Director Configuration upon Changing Boundary Conditions in Droplets of Nematic Liquid Crystal

O. O. Prischepa, A. V. Shabanov, and V. Ya. Zyryanov*
Kirenskiĭ Institute of Physics, Siberian Division, Russian Academy of Sciences,
Akademgorodok, Krasnoyarsk, 660036 Russia
**e-mail: zyr@iph.krasn.ru*

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The transformation of the director configuration upon changing boundary conditions from planar to homeotropic for bipolar nematic droplets dispersed in a polymeric matrix was studied. The characteristic textural patterns are presented for droplets with different concentrations of homeotropic surfactants, and their orientational structure is identified. The scenario predicted earlier by G.E. Volovik and O.D. Lavrentovich (ZhETF 85, 1997 (1983)) for the transformation of orientational structure of nematic droplets from bipolar to radial, without the formation of additional disclinations, is revealed. It is shown that, by using the computational method of minimization of the director elastic-strain energy in the droplet bulk and by introducing the inhomogeneous boundary conditions, one can obtain orientational structures that are analogous to the observed ones.© *2004 MAIK "Nauka/Interperiodica".*

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INTRODUCTION

Topological analysis is a rather efficient tool for studying spatially inhomogeneous structures in orientationally ordered systems, including various director and disclination configurations in liquid crystals (LCs). The classification of the topologically stable defects in nematic LC droplets has shown [1, 2] that the planar (tangential) orientation of nematic molecules at the interface is characterized by the bipolar direction configuration, with two point defects (boojums) arranged at the opposite sides of the droplet surface. In the case of a normal (homeotropic) nematic anchoring, the radial director ordering with a point defect (hedgehog) at the droplet center is the equilibrium structure, in accordance with the experimental observations [3, 4].

The theoretical analysis [2] of nematic droplets with varying boundary conditions suggests two possible scenarios for the interconversion of the bipolar and radial configurations. In the first case, one of the boojums in the bipolar structure gradually disappears and another transforms to a hedgehog, which subsequently breaks away from the surface and moves to the droplet center. In the second case, both boojums undergo changes in a similar manner, but the structure interconversion is accompanied by the formation of additional, including linear, disclinations.

In [2], a nematic with a lecithin impurity was dispersed in glycerol and taken for the experimental study. The surface anchoring of LC molecules in this system could be varied from homeotropic to planar by varying temperature in the range of nematic phase. In this case, the second structure-transformation scenario was realized with the formation of additional disclinations. However, considering that this composite consists of spherical LC droplets dispersed in an isotropic liquid matrix [2], one cannot assert that the observed scenario for the interconversion of orientational structures is universal for other objects as well, e.g., for the films of polymer-dispersed liquid crystal (PDLC) films that have been intensively studied in the last years. At the same time, a change in the boundary conditions by varying temperature is not the only approach to the study of this problem; a direct method is also possible. Namely, one can fabricate a set of samples with different concentrations of the required surface-active material and perform comparative analysis of their structural organization. This work was aimed at the detailed study of the director configurations in nematic droplets dispersed in a polymeric matrix, with the boundary condition varying as a result of varying concentration of the corresponding surfactant.

EXPERIMENTAL

The well-known 4-*n*-pentyl-4'-cyanobiphenyl (5CB) nematic liquid crystal with the transition temperatures "crystal $\xleftrightarrow{22°\mathrm{C}}$ nematic $\xleftrightarrow{35°\mathrm{C}}$ isotropic liquid" was chosen for the study. At T = 22°C and λ =

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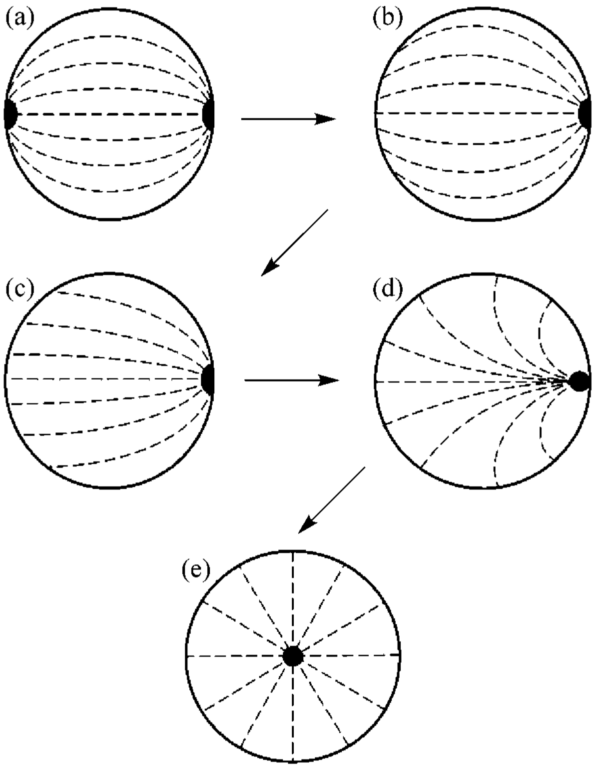

Fig. 1. The sequence of director configurations in nematic droplets with different lecithin content. The arrows are directed toward the increase in the surfactant content. (a) Bipolar droplet; (b) droplet with a destructed left boojum; (c) monopolar droplet; (d) surface hedgehog structure; and (e) radial structure.

0.589 μm, the refractive indices of 5CB are $n_{\|} = 1.725$ and $n_{\perp} = 1.534$ [5]. Poly(vinyl butyral) (PVB) of the 1PP type was used as a polymeric matrix. This polymer is transparent in the visible region and provides planar anchoring to the molecules of mesomorphic alkylcyanobiphenyl derivatives [6]. The refractive index of PVB is $n_p = 1.492$ at $T_c = 22°\text{C}$ and $\lambda = 0.589$ μm.

The homeotropic LC orientation at the interface was produced using lecithin—surface-active substance relating to the class of phospholopids. In the LC droplets with lecithin impurity, the long axes of surfactant molecules are arranged perpendicular to the surface in such a way that their polar groups are directed toward the interface, while the nonpolar fragments (flexible alkyl chains) are directed toward the bulk of the LC. As a result of such structural ordering, the lecithin molecules provide the homeotropic orientation of LC molecules at the interface.

Using a common solvent (ethyl alcohol) for all components, a set of samples with an LC content of 55% were prepared by the solvent-induced phase-separation method with the PVB and lecithin concentrations varying within 41.5–45.0 and 0–3.5 wt %, respectively. The ethanol evaporation rate was controlled so as to provide the same morphological parameters for the samples of composite film studied. The LC droplets formed a monolayer with a size dispersion of 4–16 μm.

The textural patterns of PDLC films were studied using a polarizing microscope both in the geometry of crossed polarizers and in the linearly polarized light with a switched-off analyzer. Observations showed that the textures of all droplets in a composite film without lecithin are typical for the bipolar director configuration (Fig. 1a). In the crossed polarizers, two symmetrically arranged hyperbola-shaped extinction bands are seen (Fig. 2a) that emanate from the droplet poles (point defects) and are gradually expanded. In the geometry with one polarizer, point defects are clearly seen as dark spots because of a strong optical inhomogeneity and, hence, intense local light scattering near the defects for any light polarization. For the same reason, the sections of droplet boundaries, where the light polarization coincides with the local director orientation and the gradient of refractive index $n_{\|} - n_p$ is large, are also clearly seen. By contrast, the boundary sections with the orthogonal arrangement of director and light polarization are seen least distinctly, because the gradient of refractive index $n_{\perp} - n_p$ is minimal in this region.

The sequence of director configurations arising in the nematic droplets upon an increase in the fraction of lecithin in the composite is schematically illustrated in Fig. 1. In the PDLC films with 0.08% lecithin, one of the boojums disappears in most (about 70%) of droplet ensemble. This is clearly seen in the bottom region of the droplet in Fig. 2b. The extinction band near the decaying boojum expands rather than narrows. In the remaining region of the droplet, the textural pattern has a form similar to the bipolar structure. The director-field distribution in these droplets can be represented by the configuration shown in Fig. 1b. It should be noted that the boundary conditions in this case are inhomogeneous. Here, the director in the bottom region of the droplet is oriented homeotropically. When moving along the surface to the remaining point defect, the director orientation becomes tilted and, finally, planar.

In samples with a higher lecithin content, the region of homeotropic and tilted director orientations increases and the director lines further straighten out (Fig. 1c), so that the structure, in essence, becomes monopolar. In [2], such a structure transformation was treated as "a continuous defect destruction accompanied by turning the size of its nucleus to infinity." In the crossed polarizers with the geometry presented in Fig. 2c, only one extinction band is seen in the droplet with such a director configuration. It emanates at the top of the point defect, strongly expands, and fills almost the entire lower half of the droplet. The texture of the upper half of the droplet is also analogous to the bipolar structure.

As the lecithin concentration increases, droplets with the surface hedgehog structure (Fig. 1d) and radial

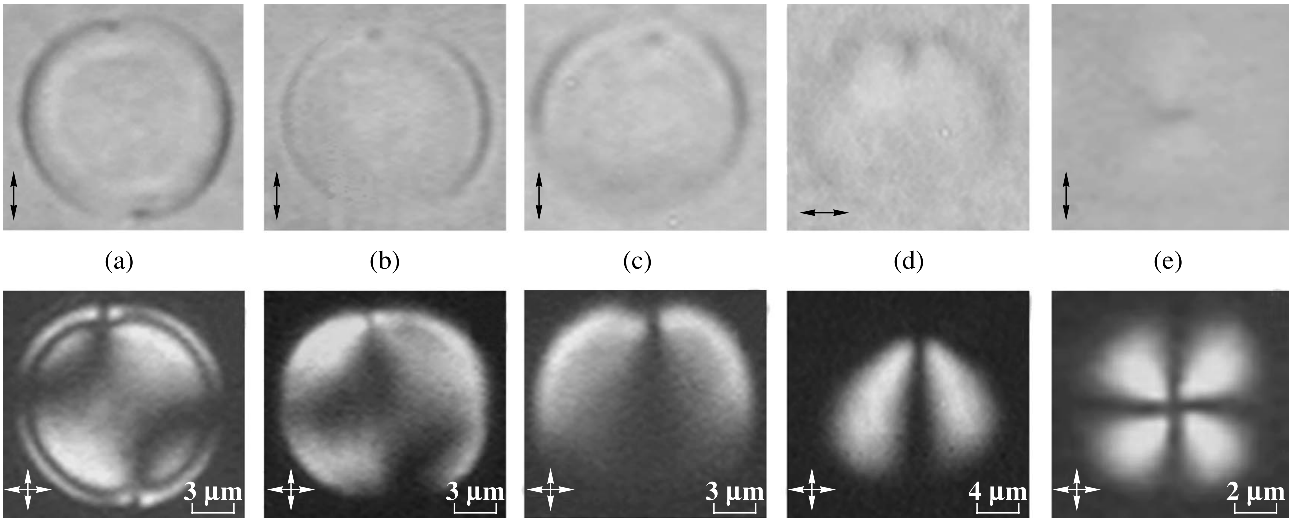

Fig. 2. Textural patterns of the 5CB LC nematic droplets dispersed in poly(vinyl butyral) with different lecithin concentrations C_{lec}. (bottom) Microphotographs in the geometry of crossed polarizers (shown by white arrows) and (top) photographs in the polarized light (the polarizer direction is shown by black arrows). (a) Bipolar droplet with symmetry axis directed at $\alpha = 11°$ with the polarizer, $C_{\text{lec}} = 0\%$; (b) droplet with a destructed bottom boojum, $\alpha = 11°$ and $C_{\text{lec}} = 0.08\%$; (c) monopolar droplet, $\alpha = 0°$ and $C_{\text{lec}} = 0.1\%$; (d) droplet with the surface hedgehog structure, $\alpha = 0°$ and C_{lec} 2.0%; and (e) droplet with the radial structure, $C_{\text{lec}} = 2.6\%$.

structure (Fig. 1e) appear. Droplets with the new structure appear and gradually increase in number. In this case, droplets with different director configurations can be observed simultaneously in the same PDLC film sample (Figs. 1a–1e), likely because of the inhomogeneous lecithin distribution over the film volume and due to some other factors, such as droplet shape, structure of the transition layer at the droplet surface, etc.

In the sequence of orientational structures from monopolar (Fig. 1c) to the surface hedgehog (Fig. 1d), the director field transforms in a smooth manner. One should identify these structures with care, because their textural patterns are, on the whole, similar. The distinctions in these textures are most clearly seen in the observation geometry shown in Figs. 2c and 2d. Here, only one extinction band is seen in the monopolar droplet. Three extinction bands are observed in the droplet with a surface hedgehog (Fig. 2d). The central band goes along the droplet symmetry axis, but it is much narrower than in the monopolar structure. Two side bands emanate from the point defect at an angle of approximately 50° to the symmetry axis on both its sides. As a result, such droplets are cone-shaped in the crossed polarizers (photographs of such textures were presented earlier in [7]), although they are, in fact, circular. The appearance of the three aforementioned extinction bands can be understood from the consideration of the corresponding director configuration shown in Fig. 1d. For the switched-off analyzer, the droplet boundaries adjacent to the defect are clearly seen for the monopolar structure if the light polarization is parallel to the symmetry axis (Fig. 2c). By contrast, the boundaries in the droplet with a surface hedgehog are seen most distinctly for the light polarized perpendicular to the symmetry axis (Fig. 2d).

CALCULATION OF DIRECTOR CONFIGURATION

The problem of determining the director configuration through the minimization of the orientational part of free energy

$$F = \frac{1}{2}\int K[(\nabla\cdot\mathbf{n})^2 + (\nabla\times\mathbf{n})^2]dV \tag{1}$$

of the LC volume with a given boundary conditions can be solved analytically for some simple geometries, e.g., for the plane-parallel cells. Here, $\mathbf{n}$ is the nematic director and K is the elastic constant. For the droplet structures, the problem becomes more complicated, so the director field in this case is calculated numerically [8].

The transition structures described above (Figs. 1b–1d) were calculated within the framework of a three-dimensional model using the method developed in [9, 10] for the study of Friedericksz transitions in bipolar nematic droplets with rigidly fixed poles. The problem was solved in a one-constant approximation with the elastic modulus $K = (K_{11} + K_{22} + K_{33})/3$. The K_{ii} values were taken from [11]. The minimum of F in Eq. (1) was found by the variational method in the Cartesian coordinate system using the experimentally observed boundary conditions.

For example, to determine the director configuration for the texture shown Fig. 2b, the anchoring was assumed to be rigid and planar for all surface points

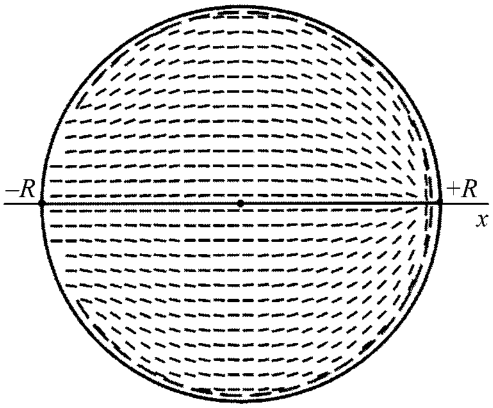

Fig. 3. The director configuration in the diametrical cross section passing through the symmetry axis of a spherical nematic droplet, as obtained by theoretical calculation within the framework of a 3D model with inhomogeneous boundary conditions. The dashed line near the surface shows the region with a specified planar director orientation.

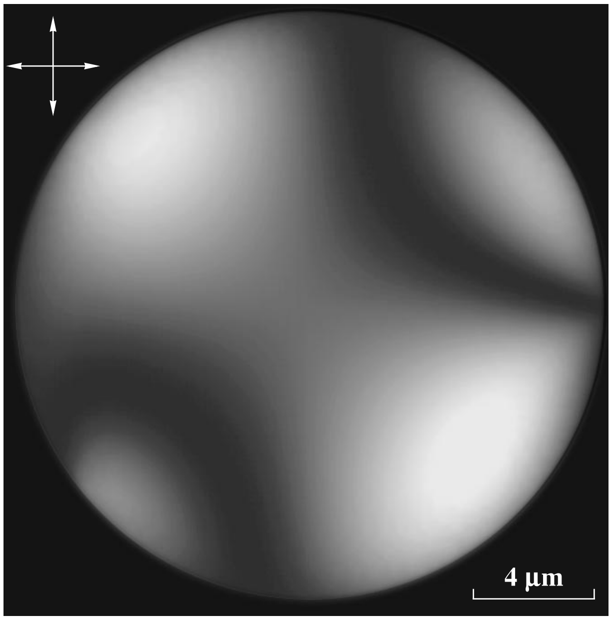

Fig. 4. Theoretically calculated texture of a spherical nematic droplet in crossed polarizers for the director configuration shown in Fig. 3 and an angle of 11° between the droplet symmetry axis and polarizer.

whose X coordinates fell within the interval $-0.8R \le X \le +R$. The azimuthal director direction in this surface region corresponded to the bipolar configuration with defects at the points $-R$ and $+R$ on the X axis. The boundary conditions were not specified for the rest of the surface with the coordinates $-R \le X < -0.8R$; i.e., the director orientation in this region was determined, as in the droplet volume, from the condition that the free energy be minimal. The resulting distribution of director field (Fig. 3) was similar to the configuration presented in Fig. 1b. In this distribution, the surface director orientation is also homeotropic near the point $-R$. When moving away from the symmetry axis, the orientation becomes tilted and then smoothly changes to planar.

Next, this information was used to calculate the corresponding textural patterns in crossed polarizers by applying the theoretical approach described in [12]. One can see (Fig. 4) that the theoretically calculated texture agrees well with the microphotograph of nematic droplet (Fig. 2b), taken for the same angle of inclination α of the symmetry axis to the polarizer.

CONCLUSIONS

Thus, a set of orientational structures intermediate between the bipolar and radial director configurations have been revealed for nematic droplets in PDLC film samples with various lecithin content. The data obtained are evidence that the change of surface anchoring from planar to homeotropic results in a gradual transformation of the bipolar structure of nematic droplets into the radial structure through a sequence of equilibrium director configurations without the formation of additional disclinations, as was predicted previously by the topological analysis in [1, 2]. Clearly, the approach described above allows the observation of only the equilibrium director configurations in different samples, whereas the temperature-induced variations of boundaries [2] allow one to trace the droplet restructurization dynamics. However, it should be remembered that not only the boundary conditions change in the second case but so do the elastic moduli that strongly influence the distribution of director field, thereby hampering the interpretation of the experimental results.

It should be emphasized that the surface anchoring in nematic droplets with intermediate configurations is inhomogeneous. Due to the use of the realistic inhomogeneous boundary conditions in our numerical calculation, the obtained director configurations and textural patterns of nematic droplets proved to be analogous to the experimentally observed ones.

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