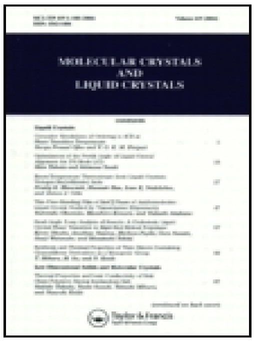

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Magnetic-Field-Induced Structural Transition in Polymer-Dispersed Liquid Crystals

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The structural transition occurring in droplets of nematic liquid crystals formed in a polymer in magnetic field is studied. The transition is caused by field-driven weakening of tangential surface anchoring and develops following two scenarios: bipolar structure/homogeneous structure or bipolar structure/radial structure. The first scenario suggests a cyclic transition occurring upon thermal fluctuations without field. The structural transition is considered within a model of the biaxial liquid crystal/polymer interface. The model describes tendency of the nematic director to align tangentially on a droplet's surface, normally under the action of flexoelectric polarization, and axially along the magnetic-field-imposed easy orientation axis.

Keywords Anchoring energy; magnetic field; polymer-dispersed liquid crystals; structural transition; temperature fluctuations

Introduction

Study of polymer-dispersed liquid crystals (PDLCs) discloses a great variety of orientational structures and their mutual transformations owing to the unique properties of a mesophase, including its high sensitivity to the effect of a confining material and external factors [1, 2]. Variation in the director configuration inside droplets leads to variation in optical characteristics of light transmitted via an ensemble of the droplets, which offers wide possibilities for practical applications of PDLC films. Conventional methods of preparation of the films, in particular, phase separation of an LC and a polymer by solvent evaporation, allow obtaining nematic droplets with both bipolar and radial structure. In this case, configuration of the director is determined by tangential or normal boundary conditions, respectively. On the other hand, when the polymer matrix used contains a surfactant at the LC/polymer interface, there are forces responsible for tangential or normal arrangement of the LC. The boundary conditions can be changed and the angle between the director and the surface normal can be gradually varied by temperature variation [3]. This leads to the mutual transformation between the bipolar and radial structures. Variation in the concentration of the surfactant added to the solution during the formation of PDLC films yields various structural transitions and, consequently, new stable configurations [4]. The

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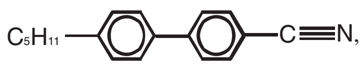
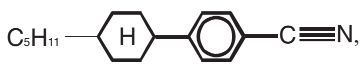
external electric or magnetic field applied upon phase separation is an additional factor affecting the formation of the interface in a PDLC film. By means of the effect of a nematic on a plastic polymer matrix, a field applied to a droplet during its formation favors the orientational structure that remains after the field is switched off. For example, in photocured polymer films prepared in electric or magnetic field, high optical transmission of a nematic was observed useful for practical applications [5]. In studies [6, 7], the authors investigated orientation of axes of the bipolar droplets prepared by phase separation of an LC and a polymer upon solvent evaporation in magnetic field and optical anisotropy of the films obtained. However, the structural transformations in nematic droplets prepared in an external magnetic field have not been studied yet. It is noteworthy that the use of a magnetic field instead of an electric field eliminates side effects resulting from possible charge accumulation in droplets and excitation of ionic conductivity in an LC. The aim of this study was to investigate the structural phase transition in PDLC films prepared in magnetic field using the solution technology.

Experiments and Results

PDLC films were prepared from the nematics 4-n-pentyl-4′-cyanobiphenyl (5CB) with the sequence of phase transitions Cr 22 N 34 Is and the chemical structure



trans-4-n-pentyl-(4′-cyanophenyl)-cyclohexane (5PCH) with the transition temperatures Cr 22 N 34 Is and the structural formula



and the polymer polyvinyl butyral (PVB). To study the effect of a magnetic field on the surface phenomena, LC molecules with a similar structure and different types of rings in the core (benzene for 5CB and cyclohexane for 5PCN) were taken.

The liquid crystals and the polymer were mixed in the ratio 2:3, dissolved in purified ethyl alcohol, and poured onto a glass substrate. The sample was placed in a gap between electromagnet poles, and magnetic field H was applied in the substrate plane for several hours until the solvent was completely evaporated and the PDLC film was formed. Then, the films were taken out from the magnet and placed in a thermostatically controlled cell. The thicknesses of the films lied within $d \cong 20 \div 30\ \mu$m. Optical textures of the LC droplets were studied on a polarized-light microscope at crossed polarizers. It was observed that the films contain ensembles of droplets with the ellipsoid form in the film plane with the average ratio of lengths of the semimajor and semiminor axes $l = a/b \cong 1.1$ and the size $2a \cong 5 \div 15\ \mu$m. Comparison of the droplets formed without H and at the values of H used in the experiment showed that the magnetic field does not affect l.

The 5CB nematic droplets formed in the magnetic field $H < 4$ kOe have a classical bipolar structure (Fig. 1(a)) typical of tangential surface anchoring at the 5CB/PVB interface. Bipolar axes of the droplets are oriented randomly in the plane of the composite film and do not correlate with the field direction. When the bipolar axis coincides with the direction of one of the polarizers, the texture of the droplets contains two crosslike

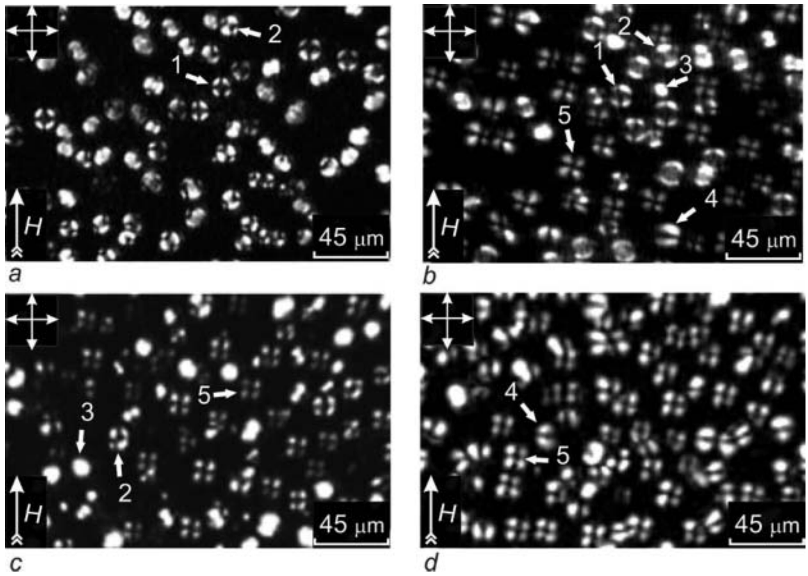


Figure 1. Ensemble of 5CB nematic droplets prepared at the temperature $t = 24°C$ in orienting magnetic field H: (a) $H = 3$ kOe, (b) $H = 4$ kOe, (c) $H = 6$ kOe, and (d) $H = 15$ kOe.

extinction bands (droplet 1 in Fig. 1(a)). When the bipolar axis is deviated from the direction of the polarizer, one can observe two dark hyperbolic arcs (droplet 2 in Fig. 1(a)). A narrower part of the extinction bands passes to boojums, i.e., point surface defects that represent poles of the bipolar configuration. In the fields $H \geq 4$ kOe, in the PDLC film, apart from the bipolar configurations (droplets 1 and 2 in Figs. 1(b) and (c)), one can also see the structures that look light when the droplet axis is located at an angle to the polarizer (droplet 3 in Figs. 1(b) and (c)). The image of such a droplet is completely darkened when the droplet axis coincides with one of the polarizers, which can be observed by rotating the microscope stage. The absence of disclinations in these droplets allows us to conclude that their orientational structure is almost homogeneous, i.e., the director is oriented in the same preferable direction in all points of a droplet volume. Such a structure is most close to the axial configuration [2] without circular equatorial disclinantion and with high degree of order of the director in a droplet volume. It should be noted that the homogeneous structures are unstable and gradually pass into the bipolar structures. In addition, there are droplets with the monopolar configurations (droplet 4 in Figs. 1(b) and (d)) that were found and reported for the first time in [4] and the radial structures (droplet 5 in Figs. 1(b)–(d)). In the radial structures, one can see four extinction bands coinciding with the directions of the polarizers and forming a cross. The bands narrow toward the droplet center where a point hedgehog defect is located. The cross does not change its location relative to the polarizers upon rotation of the sample. With increasing field H, a number of the droplets with the radial configuration of the director grow.

The 5PCH nematic droplets formed in the magnetic fields $H < 5$ kOe have the bipolar structure with the axes randomly oriented relative to field H (droplets 1 and 2 in Fig. 2). In the fields $H \geq 5$ kOe, the droplet axes tend to orient along the field. Homogeneous structures are also observed in the droplet ensemble (droplet 3). The bipolar structures (droplets 1 and 2) contain broadened boojums. No monopolar or radial structures were observed in the

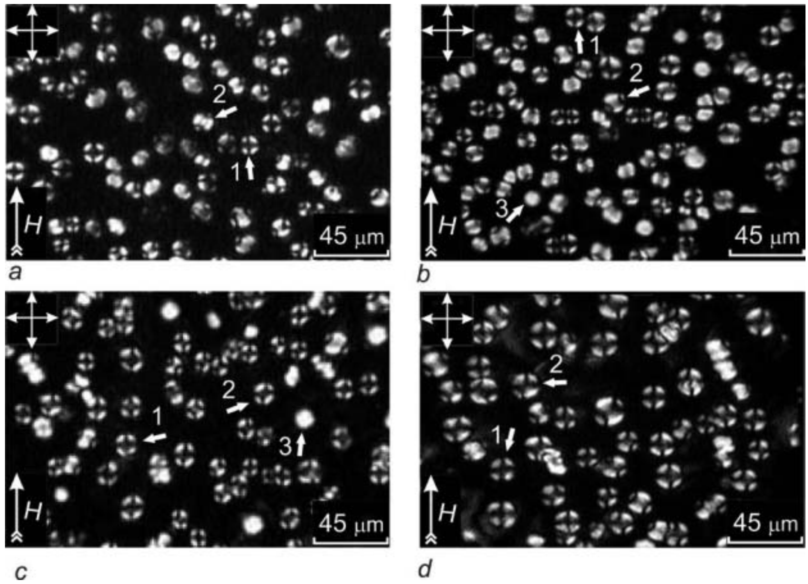


Figure 2. Ensemble of 5PCH nematic droplets prepared at the temperature $t = 32°C$ in orienting magnetic field H: (a) $H = 4$ kOe, (b) $H = 8$ kOe, (c) $H = 15$ kOe, and (d) $H = 30$ kOe.

5PCH nematic droplets. Typical textures and corresponding orientational structures of the nematic droplets are presented in Fig. 3.

Within the respective temperature ranges $t = (24 \div 34)°C$ and $t = (32 \div 54)°C$, the 5CB and 5PCH nematic droplets undergo a cyclic transition between the bipolar configuration of the director and the homogeneous structure. The transition occurs spontaneously at constant

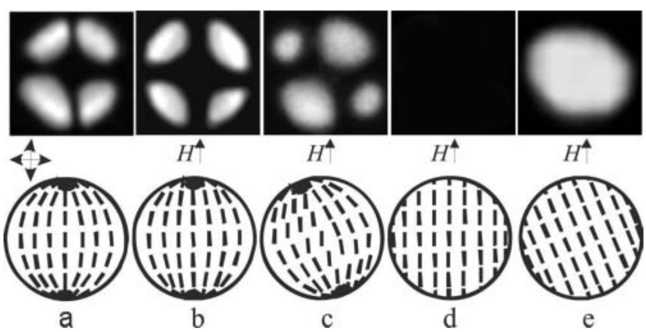


Figure 3. Textures (on the top) and orientational structures (in the bottom) of nematic droplets in a polymer matrix that are observed in the polarized-light microscope at the crossed polarizers: (a) bipolar configuration of the droplets formed without field, (b) bipolar configuration of the droplets with the broadened boojums (the droplet axis coincides with the direction of magnetic field H), (c) bipolar configuration of the droplets located at the angle $\theta \approx 30°$ to the magnetic field direction, (d) homogeneous structure formed in the magnetic field (the droplet axis coincides with the field direction), and (e) homogenous structure formed in the magnetic field (the droplet axis is located at the angle $\theta \approx 30°$ to the direction of magnetic field H). The directions of the polarizers are indicated by double arrows.

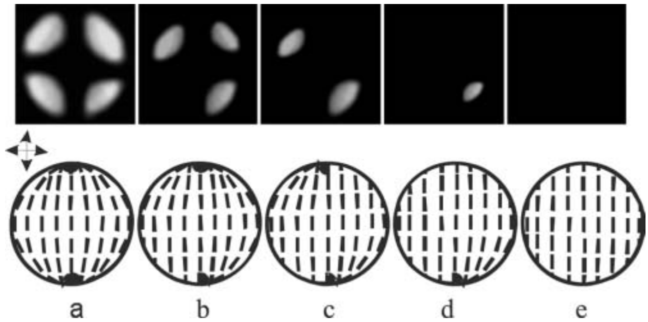


Figure 4. Dynamics of the spontaneous structural transition in the 5CB nematic droplet with a size of 15 μm. The sample was prepared upon phase separation in the magnetic field $H = 4$ kOe. During the formation, the droplet axis approximately coincided with the field direction. The directions of the polarizers are indicated by double arrows.

temperature without any external fields. To study the transition thoroughly, we used video recording. If the droplet axis coincided with the direction of magnetic field H during the formation of the droplet, then the transition follows the structural transformations shown in Fig. 4, where the droplet axis is parallel to one of the microscope polarizers. The transition starts with broadening the boojums of the bipolar structure (Fig. 4(a)). Further, the four light regions in the droplet (Figs. 4(b)–(d)) alternately extinguish. In the last phase, the light regions extinguish completely in the crossed polarizers, which imply the formation of the homogeneous structure (Fig. 4(e)). After the half-period is finished, the bipolar structure is returned in the reverse sequence.

If the droplet axis was located initially at an angle to field H, then the spontaneous transition follows the scenario illustrated in Fig. 5. The periods of appearance and disappearance of the light regions are different and depend on parameters of the droplet.

In the droplet prepared in field H, the local regions appear and disappear in the crossed polarizers with different frequency, independent of one another. Some regions of the droplet may have the stable homogenous configuration, while the other regions vary cyclically. In

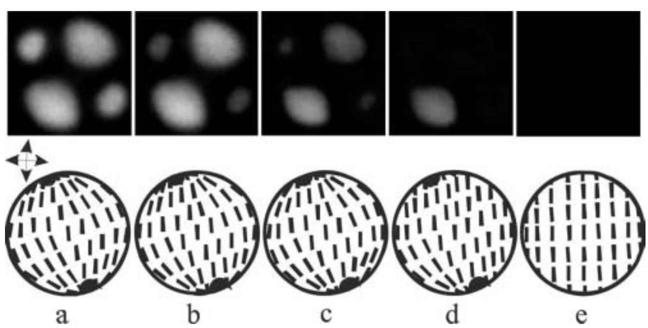


Figure 5. Dynamics of structural phase transition, proceeding in the nematic drop of the 5CB with the diameter $\sim$15 μm in the absence of magnetic field. Sample was prepared during the phase separation under influence of the magnetic field $H = 4$ kOe. The drop axis in the formation process approximately was located at an angle to the direction of the magnetic field. The directions of polarizers are shown by double arrows.

any case, if the mutual transformations occurred, then they stably repeated for many months of observations. The cycle period for the nematic local regions with the size $r \cong (2.5 \div 7.5)\ \mu$m is $\tau \cong (0.5 \div 3.5)$ s.

Discussion

The magnetic field applied during the formation of the PDLC film arranges a volume of the nematic whose surface layer interacts with the surface layer of the formed polymer matrix. Configuration of the nematic director field in a droplet is determined by a balance between the bulk and surface energies. In the case of a solidified PBV matrix, anchoring of the 5CB LC with the surface is sufficiently strong ($W = 0.8 \times 10^{-2}$ erg cm^{-2} [8]) and a deformed structure arises in which the tangential boundary conditions work on the surface, while in the sample volume the director lines are extended along the magnetic field direction [1]. If the droplet is formed in the magnetic field, the energy of anchoring of the nematic with the uncured polymer is much lower ($W = 5.3 \times 10^{-4}$ erg cm^{-2} [6]). Under these conditions, one may expect deviation of the nematic director from the surface under the action of magnetic field H applied in the experiment.

The value of the magnetic field at which the nematic director deviates from the droplet's surface is given by [8]

$$\sin(\theta_0 - \theta_s) = \frac{\sqrt{K\Delta\chi}\,H}{2W}, \tag{1}$$

where $K = (K_{11} + K_{22} + K_{33})/3$ is the nematic modulus of elasticity, i.e., the arithmetic mean of the constants of splay, torsion, and bend distortions of the director, respectively; $\Delta\chi$ is the anisotropy of magnetic susceptibility; θ_0 and θ_s are the initial (at $H = 0$) and resulting angles between the direction of H and the nematic director on the droplet's surface.

The structural transition upon weakening of tangential anchoring can be schematically presented in two variants (Fig. 6): bipolar structure/homogeneous structure (b/h) transition and bipolar structure/radial structure (b/r) transition.

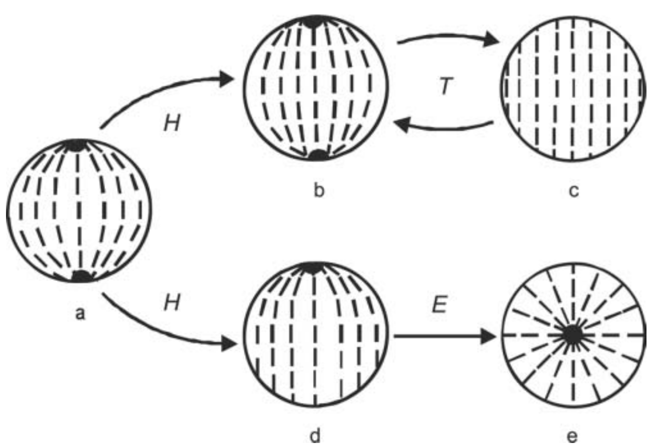


Figure 6. Schematic of the structural transition in the nematic droplets formed in magnetic field H: (a) bipolar structure, (b) bipolar configuration with broadened boojums, (c) homogeneous structure, (d) monopolar configuration, and (e) radial structure. T is a temperature of the cyclic transition. E is an electric field caused by flexoelectric polarization.

b/h Transition

Assuming $\theta_0 = \pi/2$ near a boojum and using the data at the temperature $t = 24°C$ ($K_{11} = 6.42 \times 10^{-7}$ dyn, $K_{33} = 8.6 \times 10^{-7}$ dyn [9], $K_{22} = 3 \times 10^{-7}$ dyn [10], $K = 6 \times 10^{-7}$ dyn, $\Delta\chi = 1.16 \times 10^{-7}$ [9], and $W = 5.3 \times 10^{-4}$ erg cm^{-2}), we obtain from expression (1) that, for 5CB, $\theta_s = 0$ at $H = 4.1$ kOe. Using the data at $t = 32°C$ ($K = 7 \times 10^{-7}$ dyn [11] and $\Delta\chi = 0.5 \times 10^{-7}$ [12]), we obtain that, for 5PCH, $H = 5.7$ kOe. At these values of H, the tangential orientation of the nematic director, on the surface near the boojum, should change for the normal one. The value of H is consistent with the experimentally observed field value corresponding to the beginning of the structural transformation, i.e., $H \cong 4$ kOe for 5CB and $H \cong 5$ kOe for 5PCH, when the magnetic field coincides with the droplet axis. In this case, the boojums may vanish following the scenario described in [2] with the parameter $A = \sin^2(\alpha/2)$, where α is the equilibrium angle between the nematic director on the surface and the surface normal. If the axis of the formed droplet is located at an angle to magnetic field H, reorientation of the director inside the droplet depends on a degree of boojum pinning. If the latter is weak, the boojums easily move over the droplet's surface under the action of an external field, so that the bipolar axis reorients along the field lines without symmetry variations in the structure and tangential ordering remains over the entire droplet's surface [2]. If the boojums are pinned rigidly [13], then the projection of the magnetic field $H = H \cos \beta$ (β is the angle between the direction of H and the droplet axis) favoring boojum degradation will work along the droplet axis direction. At $H = 6$ kOe and $\beta = 30°$, the value $H = 5.2$ kOe exceeds the field of the beginning of the structural transformation $H = 4$ kOe, which leads to homogeneous ordering of the nematic with the director located at an angle to the forming field. The similar scenario is implemented in the 5PCH droplets, where at $H = 8$ kOe and $\beta = 30°$ the value $H = 6.9$ kOe is higher than $H = 5$ kOe. In addition, under the action of magnetic field H, the boojums will move over the droplet surfaces depending on the degree of their pinning; in the 5PCH droplets, they move easier than in the 5CB droplets because of different energies of their tangential anchoring with the polymer. On the droplet's surface modified by LC molecules in magnetic field H, the easy orientation axis is formed that corresponds to the equilibrium state of the nematic. This axis is fixed after polymer cure. After the field is switched off, the surface energy contains two energy minima; one of them corresponds to this equilibrium state, the other is caused by the orientational anisotropy related to nonsphericity of the droplet.

The surface energy of the biaxial boundary that describes the trend of nematic director $\mathbf{n}$ to orient tangentially in direction $\mathbf{v}$ on the droplet's surface and along easy orientation axis $\mathbf{h}$ specified by magnetic field H is expressed as [14]

$$F_s = -\frac{1}{2} W_1 (\mathbf{n} \cdot \mathbf{v})^2 - \frac{1}{2} W_2 (\mathbf{n} \cdot \mathbf{h})^2 , \tag{2}$$

where W_1 and W_2 are the anchoring energies for the tangential (along the $\mathbf{v}$ orientation axis) and axial (along the $\mathbf{h}$ orientation axis) orientation of the nematic on the surface. The energies may differently depend on temperature, since they are related to the different mechanisms of LC molecular ordering at the interface with the polymer. If $W_1 > W_2$, then the bipolar configuration with the tangential orientation of the director arises. If $W_1 < W_2$, there is a trend to the formation of the axial configuration of the nematic director. During the transition, at certain temperature T, the values of the anchoring parameters may appear close ($W_1 \approx W_2 \approx W$) and the potential barrier between the two configurations may be overcame by means of thermal fluctuations of the director, which will lead to the cyclic mutual transformations of the droplet structure.

Indeed, the energy of thermal fluctuations of nematic molecules reduced to the square, $U \sim k_b \times T/a^2$, where k_b is the Boltzmann constant and a is the size of a molecule, noticeably exceeds anchoring W. Assuming $T = 297°C$, $a = 20 \times 10^{-8}$ cm, and $k_b = 1.38 \times 10^{-16}$ erg K, we obtain that $U \sim 1$ erg cm^{-2} considerably exceeds the value $W = 0.8 \times 10^{-2}$ erg cm^{-2}.

It should be noted also that the energy of tangential anchoring increases gradually from the poles to the equator of the droplet, which can be observed upon moving the coordinate system with the axes v and u along the meridian. In order words, in the equatorial region, the surface layer of the LC is anchored more rigidly, while the region near the boojums is much more sensitive to thermal excitation, which may result in independent reorientation of local regions in the droplet. The variation in the orientation of the LC on the surface is transferred in the volume that gives rise to the cyclic structural transformations, whose period and size should depend on the time of relaxation of the nematic director and a droplet size. The expression

$$\tau = \frac{\gamma_1 a^2}{K(l^2 - 1)}, \tag{3}$$

where τ is the nematic relaxation time, l is the droplet anisometry, γ_1 is the rotational viscosity, and K is the elasticity constant, was obtained from a balance of the elasticity and viscosity moments upon director reorientation inside the droplet [15].

Taking the values $\gamma_1 = 0.82$ P [16], $K = 6 \times 10^{-7}$ dyn (5CB), $\gamma_1 = 0.97$ P [16], $K = 6 \times 10^{-7}$ dyn (5PCH), $l = 1.1$, and $\tau = (0.5 \div 3.5)$ s, we obtain switching times characteristic of the droplets with the size $a = (2.8 \div 7.5)$ μm, which are in good agreement with the experimental data.

b/r Transition

One can see from Fig. 1 that the droplets are not ideally spherical or ellipsoidal and the conditions for the existence of two boojums may differ. This fact may cause the situation when one of the boojums will degrade, but critical field H will appear insufficient to overcome the force of nematic anchoring with the surface and, consequently, to destroy the other boojum. In this case, one part of the droplet will be homogeneously oriented, whereas the other will represent the director field converging to the boojum, i.e., the monopolar structure (droplet 4 in Figs. 1(b) and (d)). When the boundary conditions are modified, the boojum may transform to a hedgehog, following the scenario with the parameter $A = \cos^2(\alpha/2)$ and pass in a droplet volume [2]; then, the radial configuration arises in the droplet. It was shown, however, [3,4] that this scenario requires the normal boundary conditions. It is noteworthy that spatially separated charges may form in the deformed LC structure due to the flexoelectric effect [17]. Strains of the director field in the bipolar 5CB droplet create a spatial charge, which is maximum near the boojums, where a splay takes place, and minimum on the equator, where is a bend distortion. In particular, the electric field induced by charges in the droplets floating on the surface of an isotropic liquid favors attraction of the droplets to one another by singular points [17]. The calculation of the charge distribution in the bipolar structure makes serious difficulties, since it is necessary to obtain a consistent solution for the flexoelectric effect and the stable configuration of the droplet [1]. Nevertheless, the effect of charges on the LC structure can be estimated phenomenologically. Electric field E_u induced by the surface charges caused by flexoelectric polarization P_u is directed parallel to the droplet radius (along the u axis) and contributes to the surface energy [18]

$$F_{\mathrm{se}} = \frac{1}{2}\left[-W_e + W_0\right]\cos^2\theta_s, \tag{4}$$

where $W_e = 1/4\lambda\Delta\varepsilon E_u$, λ is the Debye length, $\Delta\varepsilon$ is the dielectric anisotropy, and W_0 is the anchoring energy caused by steric and van der Waals interactions.

Substituting this expression in Eq. (2), we obtain

$$F_s = -\frac{1}{2}W_1^*\left(\mathbf{n}\cdot\mathbf{v}\right)^2 - \frac{1}{2}W_2\left(\mathbf{n}\cdot\mathbf{h}\right)^2, \tag{5}$$

where $W_1^* = -W_e + W$ is the effective anchoring energy. If the anchoring energy $W = W_0 + W_1$ corresponds to the tangential boundary conditions, then, at $W_1^* > W_2$, the tangential orientation arises and, at $W_1^* < W_2$, the trend to radial ordering of the nematic director arises. Depending on the value of W_e, anchoring energy W_1^* may change its sign and either orientation of the nematic director arises. The dielectric anisotropy $\Delta\varepsilon = 9.9$ [19] of the 5PCH nematic is much lower than the value $\Delta\varepsilon = 13.3$ [9] of the 5CB nematic, so, for 5PCH, the electric field is insufficient for W_1^* to exceed W_2 and for the radial structure to be implemented.

Conclusions

In this paper, we have studied the structural transformations in the PDLC films formed in a magnetic field. In the experiments, the 5CB and 5PCH nematic liquid crystals and the polymer PVB were used. Despite the structural formulas of the nematics are similar, their orientational structures in droplets appeared considerably different, which is caused by modification of the liquid crystal/gel polymer interface in a magnetic field. In the 5CB, bipolar, homogeneous, monopolar, and radial structures were observed; in the 5PCH, no monopolar and radial structures were found. At certain temperatures, in both nematics a cyclic transition between the bipolar configuration of the director and a homogeneous structure was observed. This transition is spontaneous without external fields far from the nematic/isotropic liquid transition at a constant temperature. The structural transition is schematically presented in two variants: bipolar structure/homogeneous structure and bipolar structure/radial structure. The results obtained are explained on the assumption that, under the action of the magnetic field used in the experiment, the director of the nematic deviates from the surface of the gel polymer. Under this assumption, the surface energy can be written using two terms. The first term describes the trend of the director to orient tangentially in the direction of a droplet axis; the second term, along the easy orientation axis imposed by the magnetic field. Depending on which of the two terms is larger, either bipolar or homogeneous structure is implemented. If the terms are close in values, the potential barrier between two configurations can be overcome by thermal fluctuations, which ensures cyclic transformations of a drop structure. The electric field induced by flexoelectric polarization in the deformed liquid–crystal structure of a droplet may change a sign of the first term and lead to radial orientation of the nematic director. This field is sufficient to orient the 5CB nematic due to the dielectric anisotropy caused by the presence of two benzene rings in a molecule and insufficient to orient 5PCH because of the only benzene ring in a molecule.

Acknowledgments

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