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## Optical Textures and Orientational Structures of Nematic and Cholesteric Droplets with Heterogeneous Boundary Conditions

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*Brief review of the orientational structures within the droplets of chiral as well as achiral nematics with the heterogeneous boundary conditions is reported. The modification of boundary conditions in the 5CB/polyvinylbutyral and LN-396/cholesterylacetate/polyvinylbutyral compositions with the planar anchoring at the interface is created by addition of the homeotropic surfactant (lecithin). Sequences of the novel stable and non-stable orientational structures being realized within the droplets of achiral nematic are presented. For the LN-396 nematic droplets doped by cholesterylacetate the formation of a spherulite structure has been shown when the tangential surface anchoring is modified to the homeotropic one.*

**Keywords:** director configuration; interfaces; nematic and cholesteric droplets; surfactant.

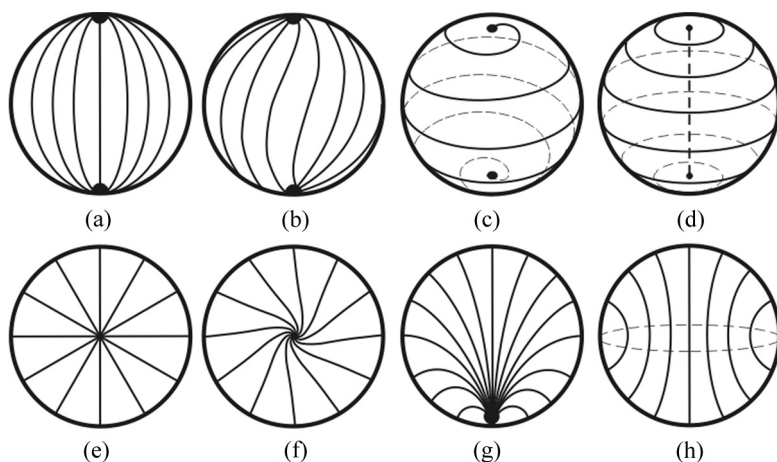
## INTRODUCTION

Polymer dispersed liquid crystals (PDLC) are of great interest due to their application in the advanced optical devices such as large flexible displays, switchable windows, paper-like displays for electronic books etc. The electrooptical performances of PDLC materials depend on such factors as the content of liquid crystal (LC), the morphology of the films, the dielectric properties of the composite in electric fields,

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and the orientational structure of LC in the droplets. The latter factor mainly depends on the anchoring conditions of LC molecules at the polymer interface. As early as XIX century Lehmann [1] revealed two types of orientational arrangement of optical axis within LC droplets dispersed in viscous matrices: a bipolar structure (Fig. 1a) for the tangential anchoring and a radial one (Fig. 1e) for the homeotropic boundary conditions. The bipolar configuration (Fig. 1a) is characterized by two surface point defects (boojums [2]) located at the opposite ends of long droplet axis. In the radial structure the director is aligned along the radius all over the droplet's volume forming the point defect in the droplet's center. The bipolar and radial structures are formed more frequently in the composite LC materials. However, today a number of other director configurations within LC droplets dispersed in solid matrices [3,4] are known. Generally, the orientational LC structure depends not only on the boundary conditions, but also on the ratio of elastic constants, the size and form of droplet as well as external fields. So, when  $K_{11} \geq K_{22} + 0.431 K_{33}$  ( $K_{11}$ ,  $K_{22}$  and  $K_{33}$  are the elastic constants corresponding to bend, torsion and splay deformations, respectively) the twisted-bipolar nematic structure is formed (Fig. 1b) [2,5]. A decrease of  $K_{33}$  comparatively  $K_{11}$  module may in principle result in the formation of the toroidal configuration (Fig. 1d) [6,7] in which there is only a bend director



**FIGURE 1** Director configurations within nematic droplets, (a) bipolar, (b) twisted bipolar, (c) super-twisted bipolar, and (d) toroidal structures for tangential boundary conditions; (e) radial, (f) twisted radial, (g) escaped radial, and (h) axial configurations for homeotropic anchoring.

deformation. However, as shown in [8], in the real composites with such a ratio of elastic modules a super-twisted-bipolar configuration (Fig. 1c) is realized, but not a toroidal one.

A twisted-radial structure (Fig. 1f) [9–11] appears when  $K_{11} > K_{33}$ . Under action of the electric or magnetic field a number of various structures such as an escaped radial configuration (Fig. 1g) [12,13] and an axial structure (Fig. 1h) [10,12,14] can be formed.

The nematic droplets with the variable boundary conditions have been considered in [2]. By means of topological analysis two different scenarios of transformation of the bipolar configuration into the radial one caused by change of the planar surface anchoring into the homeotropic one have been predicted. One of them has been demonstrated in the experiment [2] with the LC nematic droplets dispersed in the isotropic liquid. This transformation is characterized by the formation of additional disclinations within the transitional structures.

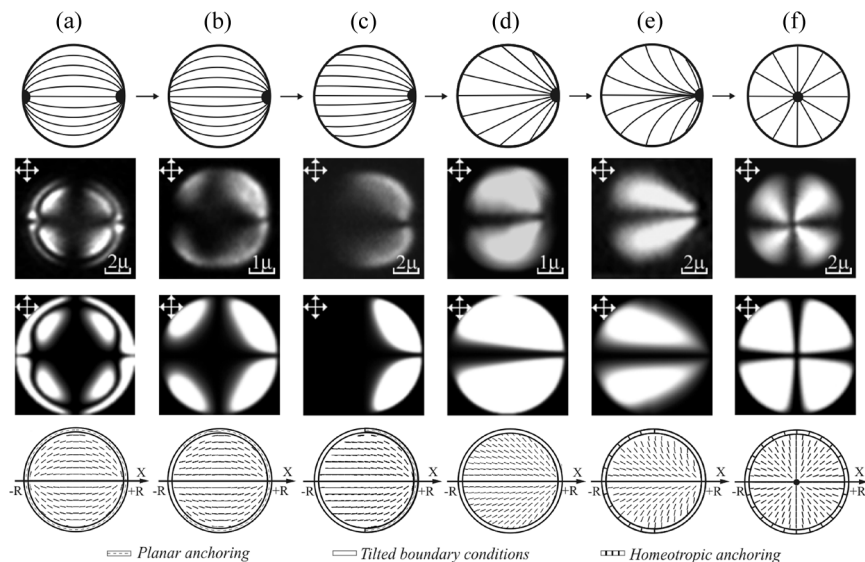
A possibility of bipolar-to-radial structural transition for the 5CB nematic droplets dispersed in water resulted from the cationic surfactant CTAB concentration change was considered in [15].

In this paper we present the stable and non-stable orientational structures within polymer dispersed achiral and chiral nematic droplets which are formed under the change of the planar surface anchoring into the homeotropic one due to addition of lecithin.

## 1. Stable orientational structures within nematic droplets doped by lecithin

The polymer dispersed nematic liquid crystal (PDNLC) films based on the polyvinylbutyral (PVB) and 4-n-pentyl-4'-cyanobiphenyl (5CB) were prepared for studying. The clearing temperature of 5CB is  $T_c = 35^\circ\text{C}$ , the refractive indices of 5CB and PVB are  $n_{||} = 1.725$ ,  $n_{\perp} = 1.534$  and  $n_p = 1.492$ , respectively. The optical textures were tested by means of polarizing microscope POLAM P-113 in crossed polarizers. We have carried out the analysis comparing the orientational structures inside the droplets of the same size.

In this composite PVB provides the tangential anchoring on the whole droplet surface. As mentioned above, the bipolar director configuration (Fig. 2a) is formed in PDNLC films with homogeneous tangential boundary conditions. The addition of lecithin results in the creation of a number of novel structures (Fig. 2b–e) [16]. These structures are essentially different from the well-known ones (see Fig. 1) by the presence of a single point surface defect. For example, in the PDNLC sample doped by 0.08%lecithin (see Fig. 2b) the droplets with one destructing boojum are formed along with a small number



**FIGURE 2** Sequence of the orientational structures (the first row) being formed within nematic droplets under changing of boundary conditions from the tangential to homeotropic ones. (a) bipolar structure, (b) droplet with one destructing boojum, (c) monopolar configuration, (d) sunset structure, (e) pre-radial structure, (f) radial configuration. Microphotos of corresponding droplets in crossed polarizers are shown in the second row. Optical textures calculated in the same geometry are presented in the third row. Director configurations calculated at the boundary conditions similar to the experimental ones are shown in the fourth row.

(approximately 30%) of bipolar droplets. If the surfactant content is higher (0.1%), the appearance of a monopolar director configuration is observed (Fig. 2c). This structure also has only a single surface defect. The comparative analysis of the structures presented in Figure 2a–c shows that the increase of lecithin concentration in this range  $0 \div 0.1\%$  causes the straightening of director lines. Consequently, such a tendency can result in the formation of the structure (Fig. 2d), where the director lines are practically straight and originate from the boojum as the rays of sunset. When the lecithin concentration is increased up to 1% and more, the angle between LC molecules and the interface changes significantly at the whole droplet's surface including the surroundings of residual boojum (Fig. 2d,e).

According to the topological analysis [2], the change of anchoring to the homeotropic state causes the transformation of such a boojum into the bulk point defect-hedgehog near the surface (Fig. 1g). The latter

structure is unstable and further the defect-hedgehog separates from the surface and moves to the droplet's center (Fig. 2f). This fact allows us to name the configuration shown in Figure 2e a pre-radial structure.

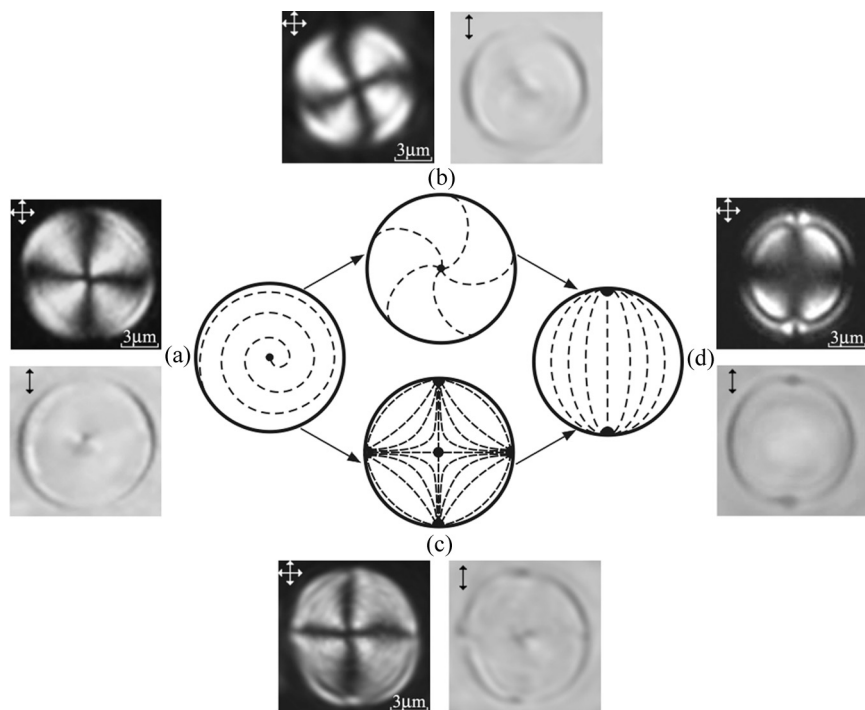
To prove theoretically the possibility of novel structures formation we carried out the numerical simulation of the discovered director configurations by means of the elastic energy minimization procedure of LC volume with the boundary conditions corresponding to the experimental ones. Detailed description of the used simulation method has been presented in [16]. After that, we simulated the proper texture patterns of the nematic droplets in crossed polarizers. The obtained results are presented in Figure 2 (the fourth and third rows), respectively. One can see, the textures of real droplets (Fig. 2, the second row) agree well with calculated ones (Fig. 2, the third row).

Therefore the novel stable orientational structures (Fig. 2b–e) within nematic LC droplets dispersed in polymer can be formed when the boundary conditions are transformed from the tangential (Fig. 2a) to homeotropic ones (Fig. 2f) due to the addition of a suitable surfactant. Such a consequence of stable orientational structures corresponds to the scenario of transformation of topological defects without the formation of additional disclinations inside the volume and at the surface of LC droplets [2].

## 2. Non-stable director configurations within nematic droplets

In PDNLC films under study without surfactant the non-stable director configurations may appear along with the above-described bipolar structures. The typical textures of such droplets of LC 5CB dispersed in PVB matrix by the solution method are shown in Figure 3a,b,c. At first sight, the texture of droplet displayed in Figure 3a in crossed polarizers is similar to the radial one (Fig. 2f). However, the detailed analysis of texture without analyzer indicates that they essentially differ. In this geometry the right and left droplet's borders (Fig. 3a) are seen more distinctly. It means that in these sections the director is approximately tangential to the droplet's surface and parallel to the picture plane. In our case, when  $K_{11} < K_{33}$ , the toroidal structure (Fig. 1d) cannot be formed because it is not realized even for  $K_{11} \geq K_{33}$  [8]. Consequently, the director configuration within such droplets has a super-twisted bipolar ordering (Fig. 3a).

Observing the super-twisted-bipolar structures for several months we have revealed that they are transformed into an ordinary bipolar configuration. At that the samples under study were stored in the closed box at room temperature. Two various schemes of transformation are

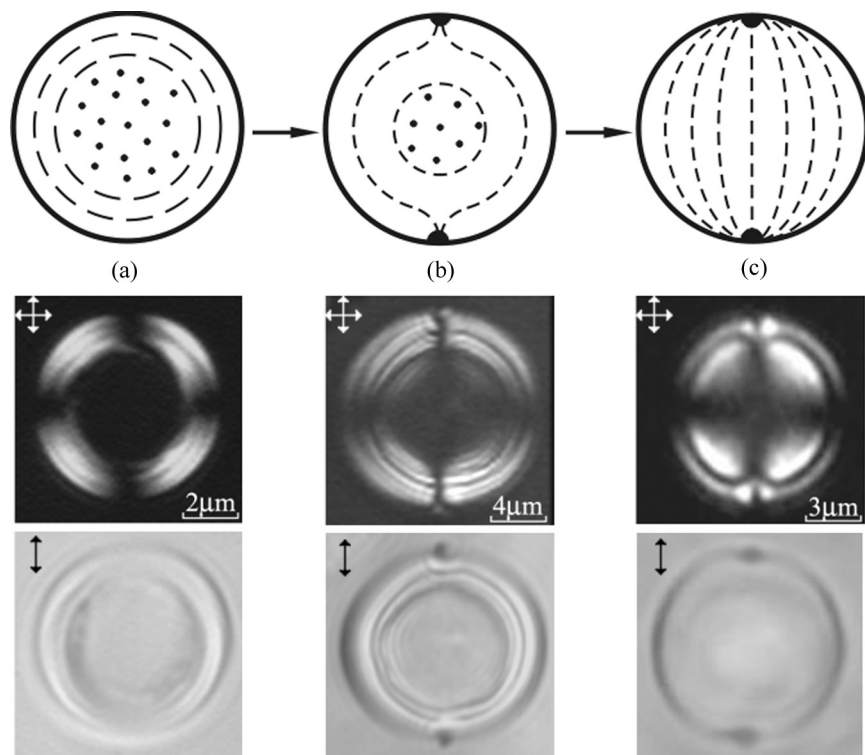


**FIGURE 3** Schematic presentation of two variants of transformation of the super-twisted-bipolar configuration (a) into the bipolar one (d). The first variant is the transition (a)→(d) through the twisted-bipolar structure (b). The second variant is the transition through the multi-defects structure (c). Next to the director configurations their corresponding textures in crossed polarizers and without analyzer are shown.

possible: either through the twisted-bipolar structure (Fig. 3b, transition (a) → (b) → (d)) or through the multi-defects structure shown in Fig. 3c (transition (a) → (c) → (d)). The two transitions are allowed from the point of view of the conservation law of summarized topological charge.

Another kind of non-stable structures has been found in the PDNLC films (PVB + 5CB) doped by lecithin. For example, in the sample with 0.24% lecithin the droplets (Fig. 4a) with the peripheral field similar to the super-twisted bipolar texture (Fig. 3a) are formed. But the central part of these droplets in crossed polarizers is dark and doesn't change at the turn of the sample (see Fig. 4a, photo in crossed polarizers). It means that in the center of droplet the director is perpendicular to the film plane. Moreover, in the equatorial section the toroidal or close





**FIGURE 4** Schematic presentation of the transformation process of the twisted-toroidal structure (a) into the bipolar one (c) through the intermediate bipolar structure with two line disclinations (b). The corresponding textures in crossed polarizers (second row) and without analyzer (third row) are shown under the director configurations.

to the toroidal structure is realized. The director is twisted removing from the surface to the droplet center aligning here along the line of sight. The described orientation structure named a twisted-toroidal one is displayed in Figure 4a (top). This configuration as well as the super-twisted-bipolar are non-stable and can transform into the bipolar one.

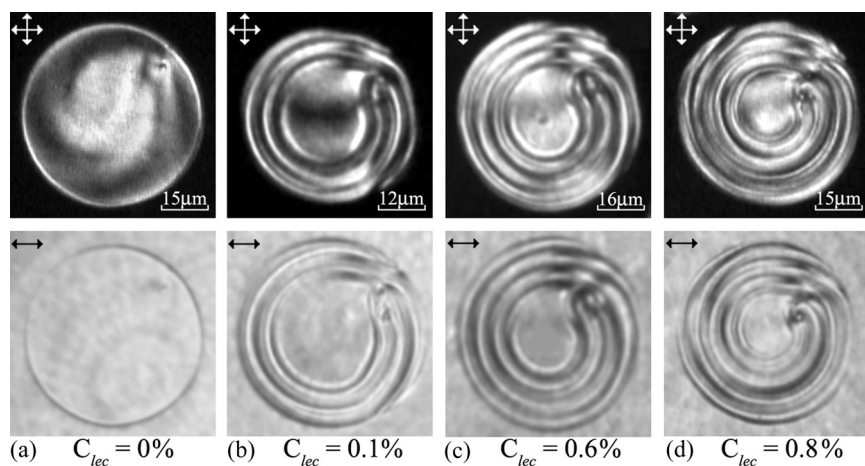
This process also takes much time, and so the continuous observation of the changes inside a droplet has not been carried out. However, the comparative analysis of the droplet textures made after some long time intervals allows us to reveal the droplets with an intermediate bipolar structure with two line disclinations (Fig. 4b). The central part of this droplet (see Fig. 4b in crossed polarizers) is dark relative to

any orientation of crossed polarizers. But two point defects-boojuims have already been formed at the surface. Then, several months later the droplets with non-stable orientational structure formed within the PDNLC film disappear and the droplet ensemble in the film become bipolar.

### 3. Orientational structures within cholesteric droplets doped by lecithin

To study the influence of homeotropic surfactant on the orientational structure of cholesteric droplets dispersed in polyvinylbutyral we have used a nematic mixture LN-396 based on the cyanobiphenyls (Belarus state university, Minsk, Belarus) doped by cholesterylacetate (X3). The clearing temperature of LN-396 is  $T_c = 64^\circ\text{C}$ . Initially the samples of nematic mixture with a different cholesterylacetate content (1.5%, 4.1%, 6.0%, 8.2%, 12.0%) were prepared. Using the well-known technique of "fingerprint" texture analysis inside thick layer placed between two glass substrates with homeotropic anchoring, the pitch of helicoid for all samples was measured. The thickness of the layer was  $200\text{ }\mu\text{m}$ , and a homeotropic orientation on the internal sides of substrates was provided due to the lecithin layer.

Then the obtained cholesterics with a different pitch of helicoid were dispersed in polyvinylbutyral. In Figure 5a the cholesteric



**FIGURE 5** Textures of droplets of nematic LN-396 mixture doped by 1.5% cholesterylacetate with the different lecithin content. Photos in the crossed polarizers are presented in the first row and without analyzer in the second row.

(LN-396 doped by 1.5% X3) droplet with the unwound helicoid and a single surface defect is shown in crossed polarizers and without analyzer. The polymer dispersed cholesteric liquid crystal (PDChLC) films doped by 1.5% of X3 are the most susceptible to the surface-active molecules in comparison with the other considered concentrations. So, changing sequentially the tangential anchoring at the LC-polymer interface in PDChLC films by adding the lecithin (from 0 to 0.8%), one can observe the transformation of optical textures of cholesteric droplets (Fig. 5a–d). Evidently the addition of lecithin results in the winding of helicoid thus forming the spherulite structure at the peripheral field and the planar ChLC texture within droplet center.

## CONCLUSION

Thus, the addition of surfactant can considerably transform the orientational structure of nematic droplets including the chiral nematic (cholesteric) with a large (15  $\mu\text{m}$ ) pitch of helicoid. The obtained data allow developing new methods to control optical properties of LC composites owing to the electrically commanded modification of the boundary conditions. For example, recently the ionic-surfactant method of surface anchoring modification has been offered in [17]. This approach has made the reversible transformation of bipolar structure into the monopolar one possible. It proves the important significance of obtained results for practical application.

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