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Article

Studies of the structure and properties of polymer dispersed liquid crystal films to create a polarizer

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Abstract: A novel composite material based on polymers (polyvinyl alcohol, polyvinyl butyral) and liquid crystal (4-n-pentyl-4'-cyanobiphenyl) has been developed and studied. Configuration transformations of point defects in nematic droplets under the influence of an electric field, caused by localized changes in the concentration of NLC within the polymer matrix, have been discovered and analyzed. The boundary conditions necessary for achieving a nematic structure with homogeneous alignment of the director both within the droplet and at its surface have been established, optimizing the anisotropy of light transmission in polymer-dispersed liquid crystal (PDLC) films. Additionally, polarization effects inside nematic droplets under the application of an electric field have been identified.

Keywords: polyvinyl alcohol; polyvinyl butyral; polarizers; liquid crystal; surfactant; refractive index; matrix; planar and homeotropic alignments; interfacial anchoring; droplets; textures

1. Introduction

Film polarizers (polaroids) based on polymer films are widely used across various industries, ranging from instrumentation and industrial electronics to consumer and medical equipment. Most polarizers are designed for the visible spectrum and are commonly used in liquid crystal display devices, such as LC indicators and LC displays. Additionally, film polarizers serve as light filters in numerous optical and optoelectronic devices. They are indispensable in specialized fields such as polarization microscopy, magnetometry, spectrophotometry, ellipsometry, and electrical signal measurement [1–3]. Recently, the application of film polarizers has expanded to include devices for detecting hidden images, providing robust protection against forgery for trademarks, securities, and confidential documents. In all these applications, the primary function of film polarizers is to convert incident light into polarized light.

For polaroids, dichroism is due to the introduction of special additives into the polymer matrix or the anisotropy of the intrinsic electronic absorption bands of the macromolecules of the polymer used. However, in laser and optical technologies, polaroid films are applicable only in the case of low radiation intensity, since light absorption can lead to heating and subsequent thermal destruction of the polymer matrix.

Polymer compounds, primarily polyvinyl alcohol (PVA), serve as the main matrix (substrate) material in polarizer films [4]. The PVA polarizer film is a uniaxially oriented polymer containing a dichroic agent. The uniaxial stretching of the film contributes to the orientation of the PVA macromolecules in a specific direction, which ultimately produced a dichroic agent. In this case, the electrical energy is converted into thermal energy. The component of the light radiation polarized orthogonally to the iodine chains usually passes through the PVA polarizer [4]. PVA films are easily subjected to uniaxial orientation, after which they become optically anisotropic and partially polarize the transmitted light radiation. To maximize light transmission, the study [5] investigated the reorientation process of cholesteric liquid crystal, induced by electric-controlled ionic modification of surface anchoring. It was established that in the initial state, the homeotropic orientation of the director was realized in the liquid crystal cell, and when exposed to a constant electric field, a twisted home planar structure of the cholesteric was formed.

In the transformation of the director configuration was investigated when the boundary conditions were changed from planar to homeotropic for bipolar nematic droplets dispersed in a polymer matrix [6]. The authors presented characteristic texture patterns for droplets with different concentrations of homeotropic surfactants and determined their orientational structure. They demonstrated that by using a computational method for minimizing the elastic deformation energy of the director within the droplet and introducing non-uniform boundary conditions, it is possible to obtain orientational structures necessary for maximizing light transmission. However, the main drawback of most traditional and new methods for producing polarizers is the inability to achieve high concentrations of fillers in the final solutions, as well as the complexity of implementation and the use of aggressive substances [7]. Moreover, many methods for producing a new class of polarizers require thorough and labor-intensive purification of the resulting product.

Therefore, in the present work, a new polymer composite material, called PDLC, is proposed, based on the nematic liquid crystal 5CB and polymer matrices (PVA and PVB), which eliminates this drawback.

The use of uniaxially stretched films dispersed with polymer-dispersed liquid crystals (PDLC) that exhibit anisotropic light scattering as polarizing elements allows for a significant increase in the maximum luminous flux power [8]. PDLC films effectively polarize radiation across the entire transparency range of their components (visible and near-infrared), whereas traditional polaroids operate only within the dichroic absorption band of the material or its impurities. Furthermore, the anisotropy of light transmission in LC films can be controlled by applying an electric field. PDLC films are especially promising for use in laser devices, as the collimated nature of the radiation allows for easy removal of scattered light with a diaphragm. Doping the liquid crystal with a suitable surfactant, followed by uniaxial stretching of the composite film, offers a promising approach to overcoming this challenge, enabling the selection of optimal technological conditions for producing highly efficient film polarizers based on the anisotropy of light scattering.

2. Research objects and experimental methods

The nematic liquid crystal (NLC) 4-n-pentyl-4'-cyanobiphenyl (5CB), from the alkyl biphenyl series, was used to prepare and investigate polymer-dispersed liquid crystal (PDLC) films, a type of composite material. Two thermoplastic polymers were utilized as the matrix: polyvinyl alcohol (PVA) and polyvinyl butyral (PVB).

These polymers, after dissolution in appropriate solvents and subsequent drying, do not undergo chemical transformations.

The components of the studied materials were in the proportional ratios of 1:33.03:1.04:0.003 for NLC 5CB:PVA:Gly:CTAB and 1:33.2:1.17 for NLC 5CB:PVB:CTAB, respectively. For PVB, there was no need to add glycerin to provide elasticity, as NLC 5CB simultaneously acts as a plasticizer. The physical parameters and coefficients required for studying light transmission, contrast, and texture characterization of nematic droplets are presented in **Table 1**.

Table 1. Physical parameters of the research objects.

Materials	The temperature of enlightenment, °C	Refractive index coefficient		Wavelength, µm
NLC 5CB	34	1.717	1.531	0.633
PVA	22	1.52		0.633
PVB	22	1.488		0.633

Polymer matrices (PVA, PVB) have one refractive index equal to n_p 1.52. The liquid crystal 5CB, due to optical anisotropy, has two refractive indices: n parallel and n perpendicular. In the initial state, i.e., before the start of the study of the PDLC films, the refractive indices of the polymer (n_p = 1.52) and the parallel component of the refractive index of LC 5CB (n_{II} = 1.531) are equal to each other, but, conversely, the refractive indices of the polymer (n_p = 1.52) and the perpendicular component of the refractive index of LC 5CB are very different ($n_{\perp}$ = 1.717). This fact is better known as optical anisotropy, based on the phenomenon of double refraction. In order for the equality conditions ($n_{\perp} = n_p$) to be fulfilled, we needed to apply an electric field to the LC cell. Then the LC 5CB molecules are reoriented along the direction of the electric field strength and the above equation is fulfilled. This equality contributes to the maximum light transmission of PDLC films.

As shown by the numerical values in **Table 1**, the condition ($n_{\perp} = n_p$) is well satisfied for both PVA and 5CB. For pure PVB, the refractive index is approximately 0.04 lower than the $n_{\perp}$ value of 5CB. However, during phase separation in the encapsulation process, a portion of the liquid crystal remains dissolved in the polyvinyl butyral matrix, leading to an increase in the absolute refractive index (n_p) of the polymer, aligning it more closely with the refractive index of the nematic [9]. Both polymers set the planar (tangential) orientation of the LC 5CB molecules. To modify the boundary conditions on the surface of the polymer matrix from tangential to homeotropic, two homeotropic surfactants were selected [10]: cetyltrimethylammonium bromide (CTAB) $C_{19}H_{42}NBr$, belonging to the cationic type, and phosphatidylcholine (lecithin). Their concentration relative to the nematic varied in the range of 0%–7% by weight.

Samples of PDLC films were produced by emulsifying NLC in an aqueous polymer solution for composition with polyvinyl alcohol and using the SIPS (solvent induced phase separation) method using PVB [11]. Glycerin was added to the composition to reduce the hardness of the PVA. The resulting films had a thickness of 25–85 µm with sufficiently evenly distributed nematic droplets of varying size in the volume and on the surface of the polymer matrix.

The orientation structures in the liquid crystal droplets and their optical textures were studied using the polarizing optical method with a POLAR-2 microscope, in the geometry of crossed polarizers, as well as with the analyzer turned off. This technique enables a reliable determination of the director configuration both within the volume of the droplet and at its boundary with the polymer.

The study of the electro-optical orientation effect feature was carried out on a typical configuration, illustrated in **Figure 1**. Laser light from a He-Ne laser (Linos) with an occurrence of λ = 633 nm was applied as a power cause. The light transmission coefficient of the PDLC films was measured using a light-sensor photodetector. The signal from the photodetector was recorded by a digital multimeter. The radiation scattered on the studied samples was slowed by a diaphragm with, which made it possible to register only directly transmitted light. An fluctuate potential with a periodicity of 1 kHz was applied to the PDLC cell from the signal generator.

The morphology of the samples and the visual textures of the nematic droplets were investigated using a polarizing optical microscope POLAR-2 equipped with a MyScope 500 M digital video camera (Webbers), which made it possible to photograph and videotape the ongoing processes (**Figure 1**). The studies were carried out in the geometry of crossed polarizers and with the analyzer turned off [12]. For microscopic analyzed of the droplet answer to an electrical field, samples with a spacer thickness of 30 μm and an average droplet size of 7.2 μm were made. At the concluding phase of the preparatory process, the sample was cooled to 23 ℃ for 60 minutes. It should be noted that for this purpose, the composite film samples must be made so that the liquid crystal droplets in the field of view do not overlap each other and are sufficiently large [13].

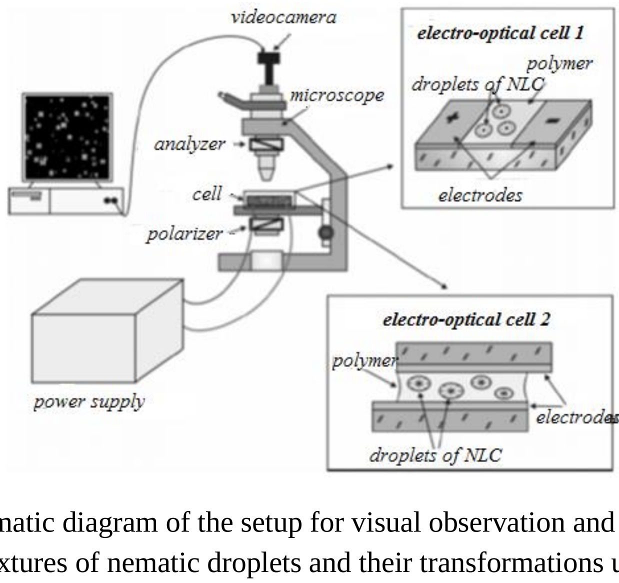

Figure 1. Schematic diagram of the setup for visual observation and image recording of the optical textures of nematic droplets and their transformations under the influence of an electric field. In electro-optical cell 1, the field is directed parallel to the substrate plane, while in cell 2, the field is directed perpendicular to the substrates.

The design of one of the electro-optical cells made it possible to observe changes in optical textures for the example when the electric applied is directed aligned to the area of the cell (electro-optical cell 1 in **Figure 1**). Such geometry allows to determine with a sufficient degree of accuracy the boundary conditions formed as a consequence of the exposure of the electric field [14].

The distance between the electrode strips ranged from 100 to 300 µm. To observe optical textures and their transformations under the influence of an external factors directed perpendicular to the cell surface; an electro-optical cell 2 was used. In this case; the distance between the electrodes was 70 µm. The cells were supplied with a constant voltage from a direct current electrical supply B5-49 with a voltage of up to 100 V. The pulse duration depended on the experimental conditions and varied from 1 sec to tens of secs [7,15]. To obtain higher voltages; a rectangular pulse signal source with a duration of 1 second and a variable amplitude of up to 1000 V was used.

To determine the angle of reorientation (α) of the LC molecules inside the droplet; a schematic image of the PDLC film was used (**Figure 2**). Here the plane of the Cartesian system (*xoy*) coincides with the plane of the PDLC film. The drip directors are oriented mainly along the *ox* axis. The angles φ and θ determine the deviation of the director relative to the axis *ox*; respectively; in the *xoy* and *xoz* planes. The schematic distribution of the droplet directors at the angles of maximum reorientation (φ_{max} and θ_{max}) is shown in **Figure 3**.

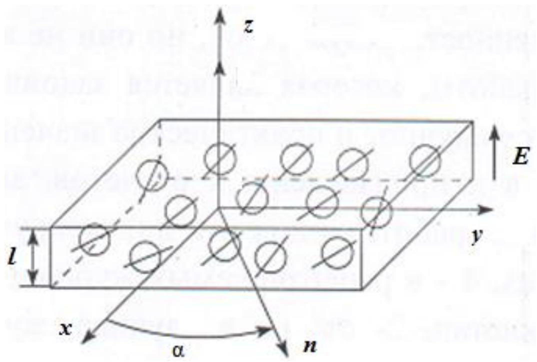

Figure 2. Schematic representation of the PDLC films in the microscope lens: *E* is the electric field strength vector, α is the angle of rotation (reorientation) of the nematic molecule inside the droplet, *l* is the film thickness, *n* is the preferred orientation (director) of the LC molecules.

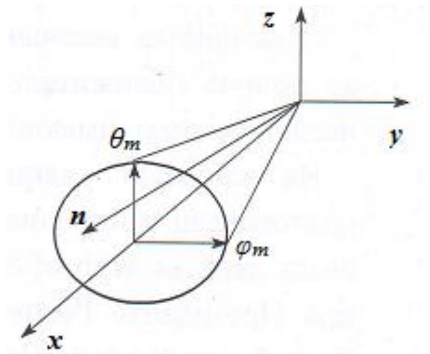

Figure 3. Orientation of the LC droplet directors: *n* corresponds to the orientation of the director of a single droplet, *φm* and *θm* are the maximum deflection angles of the droplet director, respectively, in the *xoy* and *xoz* planes.

3. Experiment

According to [16] PVB defines homeotropic barrier determines for both the nematic mixture E7 and the cholesteric blend based on the nematic amalgam EN18 [17]. In the PDLC cells based on NLC 5CB and PVB studied by us, the droplet textures (**Figure 4**) vary considerably from those characteristic of both homeotropic anchoring: (radial, axial, flowing radial structures) and planar anchoring: (bipolar, twist-bipolar, toroidal configurations).

Figure 5 presents the droplet textures (**Figure 4a**) at small angles of rotation of the specimen relative to the polarizers. Four topological particularities split the circumference of the droplet into four equal arcs are investigated in the absence of an analyzer (**Figure 5a**, below).

The inherent scheme for an orientation configurates with two boojums (black semicircles) and defects in the annular surface are sensible in the right part of **Figure 5** under the symbol (d). The bipolar axis is oriented horizontally, as in the case for **Figure 5a**. In the schematic representation, the symbol α indicates the angle the slope between the local orientation of the director and the normal to the surface. In all observed patterns, the droplet size is 10 µm.

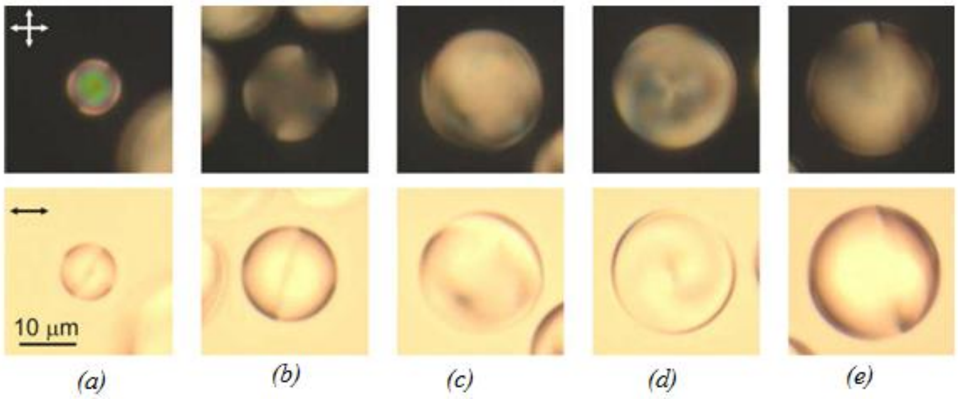

Figure 4. Pictures of specifically droplet optical textures **(a)–(e)** investigated in 5CB and PVB in stricken polarizers (upper range) and without an analyzer (lower range).

It can be seen that a dark line called the straight quenching branches, the positions of which do not change in the vertical and horizontal planes, connects the upper and lower defects. Under the action of an electric field in the nematic droplets, the textures change: a solid quenching disk occupying the boundary areas of the droplet projection replaces the quenching cross. This evidences that the molecules LC is oriented mainly parallel to the polarizer. A perpendicular thin attenuation band connecting the upper and lower defects at the boundary, and a horizontal attenuation band combining two other defects are also visible in crossed polarizers (**Figure 5a**, upper row). Furthermore, four attenuation districts appear, located close to each other in a circle in the drop and located approximately the same distance from the perpendicular and horizontal lines.

Rotating the cell by 45° clockwise relative to the polarizers completely qualitative transformation the optical texture observed both in stricken polarizers and without an analyzer (**Figure 5c**). An interference color configuration appears in crossed polarizers in the absence of a fade band. Four identical sections are also visible along the border of the drop without an analyzer. When moving the droplet clockwise, maximum darkening is observed in the characteristic sections. Then the image clearness of the brink slowly decreases to the next image.

The optical structure shown in **Figure 5a** is similar to the picture obtained without an analyzer (**Figure 5c**). Consequently, when the analyzer is turned off, the boundaries of the droplet lose their clarity. When comparing the extinction band (**Figure 5**, upper row), taking into account the predominant orientation of the director at the boundary of the drop (**Figure 5**, lower row), it is possible to determine the angle of inclination α between the n director and the N perpendicular to the plane (**Figure 5d**). The predominant direction of nematic molecules can be parallel to any horizontal or vertical polarizer in the attenuation region located at the bottom left (**Figure 5a**, top row).

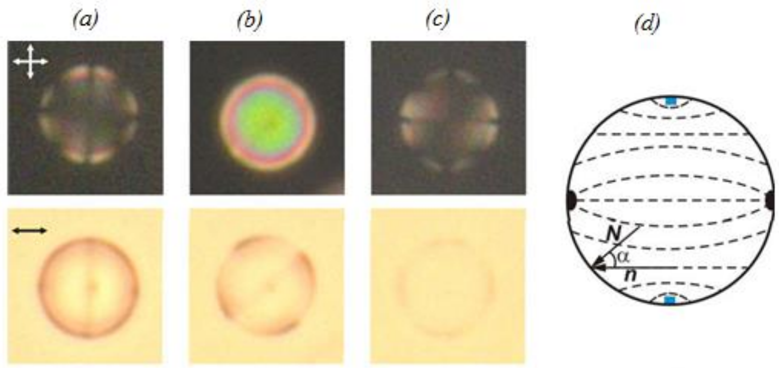

Figure 5. Images of nematic droplets under crossed polarizers (upper row) and without an analyzer (lower row). In **(a)**; the lines connecting diametrically opposite defects at the boundary are aligned with the polarizers. The cell is rotated clockwise by 45° relative to the polarizers in **(b)** and by 90° in **(c)**. The configuration of the director **(d)** corresponding to the textures **(a–c)**: the dashed lines inside the droplets show the local direction of the director.

Nevertheless, observation of the droplet absent an analyzer showed that the priority direction here is oriented almost horizontally (**Figure 5d**). The angle determined in this way at various points of the interface, with the exception of the surroundings of the objects, is $\alpha = 40^{o} \pm 4^{o}$.

The invariance of the angle α along the droplet's circumference allows for a schematic illustration of the orientation structure in the most significant region of the droplet on the plane of the film (**Figure 5d**). This pattern of the director field distribution corresponds to a structure with two point topological defects—boojums—and an annular topological defect, which has been previously observed in polymer matrices [18–20]. The segment of the annular defect on the film surface appears as two attractions at the droplet boundary. The bipolar axis connecting the boojums serves as the symmetry axis for the droplet's orientation structure.

In the PDLC samples studied, the optical textures demonstrate in **Figure 5** were discovered only inside droplets of 10 µm in size and smaller. A similar structure with two boojums and an annular defect on the surface was found in larger droplets

(**Figure 4b**), however, attenuation bands in the central part of the droplet are absent at any orientation of the bipolar axis relative to stricken polarizers. This is typical for a twisted orientation structure inside a droplet. The angle of rotation, defined as the azimuthal angle α between the orientation of the bipolar axis and the orientation of the director at the interface of the droplet surface in the equatorial surface, was determined by the method presented in detail in [21].

When the refractive indices of the polymer and liquid crystal are equal, the light passing through the droplet under crossed polarizers will scatter synchronously if the following conditions are met:

1) the bipolar axis of the droplet is oriented perpendicular to the direction of observation;
2) the polarizer is aligned either perpendicular or parallel to the projection of the director on the lower surface of the droplet;
3) the analyzer is aligned either parallel or perpendicular to the projection of the director on the upper surface of the droplet.

The angle of rotation at which the orientation of the director in the central segment of the drop is parallel to the bipolar axis consider be selected from two practicable ones. For such droplets with two boojums and an annular defect, this means that the bipolar axis must be in the plane of the cell and the surface of the annular defect must be normal to the plane of the film. The twist angle measured using this manner (**Figure 6**) for different droplets arranging the above positions depends on the size of the droplet. In this figure, the bipolar axis of the droplet is normal to the polarizer, and the angle between the polarizer and the analyzer is 90° (a). In the second case, the bipolar axis is rotated 20° counterclockwise correlative to the polarizer, and the angle between the polarizer and the analyzer is 130° (b).

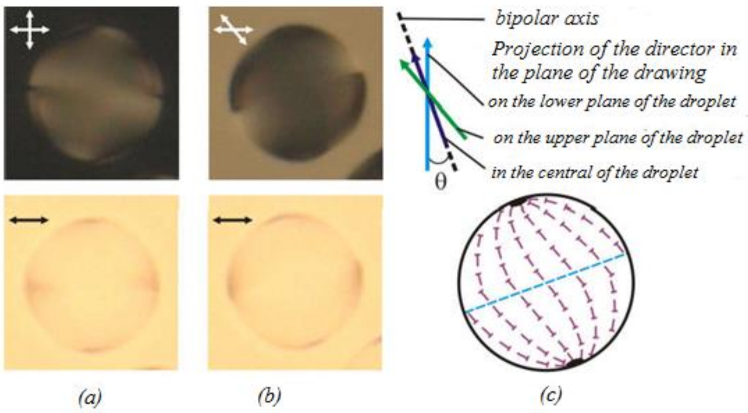

Figure 6. Images of nematic droplets in crossed polarizers when the angle between the polarizer and the analyzer is 90°. **(a)** turned counterclockwise by 20° relative to the polarizer; **(b)** calculated configuration of the LC director; **(c)** double arrows show the orientation of the polarizers.

The **Figure 6** shows in a separate view on the right a diagram of the considerable orientation of the bipolar axis of the drop, the planning of the director on the image planes along the lower and upper edges (top), as well as the director's field above the upper surface of the drop (bottom) (c). For all the paintings we observed and the micrographs depicted in **Figure 6**, the drop size was 17 µm. Thus, a comparative analysis of the obtained microscopic images suggests that for droplets with a diameter less than 10 µm, the twist angle is $\theta = 12^o \pm 3^o$, and for droplets with a diameter greater than 21 µm, $\theta = 27^o \pm 3^o$.

In addition to the above-described droplet textures, other types of paintings based on micrography are also observed, illustrated in **Figure 6c**. The rotation of the sample correlative to the polarizers leads to a complex transformation in the texture of droplets both in crossed polarizers and without an analyzer. These results are shown in **Figure 7** For all observed patterns, the drop size is 21 µm.

When the microscope stage is at a rotation angle of 0° (**Figure 7a**), the droplet boundaries appear less distinct without the analyzer, and a single point defect can be observed. As the sample is rotated, the droplet boundary becomes progressively sharper (**Figure 7b, c**). The sharpest boundaries are visible at a 90° rotation angle (**Figure 7d**). By lowering the microscope stage, a single point defect (**Figure 8a**), an annular defect (**Figure 8b**), and a second point defect (**Figure 8c**) can be clearly observed.

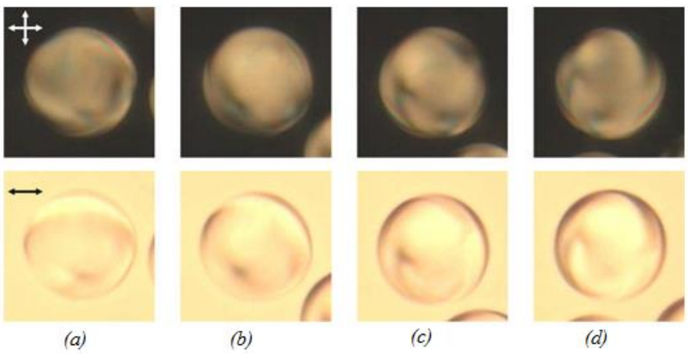

Figure 7. Images of a liquid crystal droplet under crossed polarizers (upper row) and without an analyzer (lower row) at microscope stage rotation angles of. **(a)** 0^o; **(b)** 30^o; **(c)** 60^o; **(d)** 90^o.

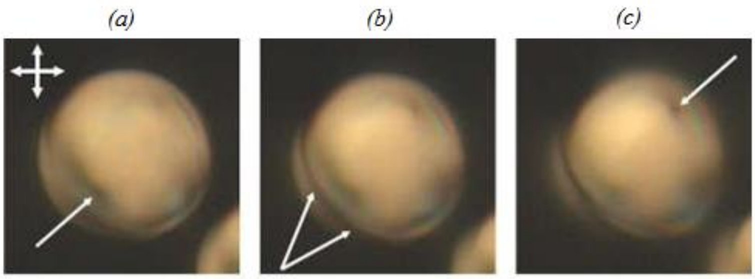

Figure 8. POM-images of a liquid crystal droplet shown in **Figure 5c** in stricken polarizers. The lens focuses on the lower defect-boojum **(a)** the annular defect; **(b)** the upper defect boojum; **(c)** single arrows indicate the location of the defects.

Compounding the obtained optical texture, it can be stated that the droplet has an orientation structure with two boojums, an annular defect and a bipolar axis inclined to the plane of the cell. The missing of attenuation bands indicates the presence of torsion deformation, as in the case of **Figure 4b** and **Figure 6**. The angle between the bipolar axis and the cell plane can vary from 0° (**Figure 4a**, **b**) to approximately 90° (**Figure 4d**) in the ensemble of droplets. If the bipolar axis is perpendicular to the cell plane, the optical texture does not depend on the rotation of the microscope stand (platform).

Let's return to the drop shown in **Figure 4e**, where there are also two boojums and a ring defect. **Figure 9** shows figures of this droplet at different angles of orientation of the bipolar axis relative to the polarizer. It should be noted that the plane of the annular defect extends from the center of the drop to one of the boojums. At the same time, torsion deformation occurs inside the droplet. Such structures are usually investigated inside large droplets (more than 15 µm). The substitution of the annular defect may be different within different droplets.

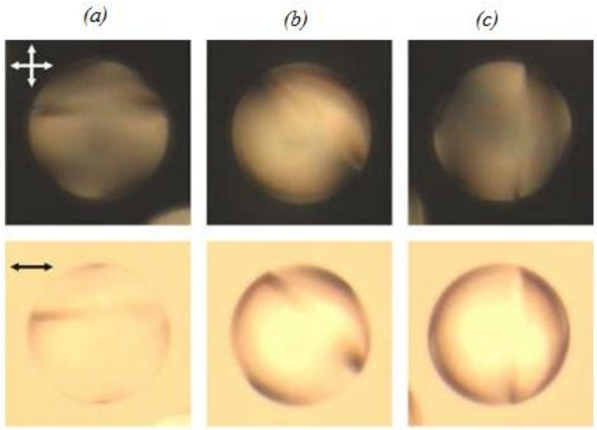

Figure 9. Pictures in stricken polarizers (upper range) and absent analyzer (lower range). **(a)** The bipolar axis is perpendicular to the polarizer; **(b)** the sample is rotated 45° clockwise relative to the polarizer; **(c)** the sample is rotated 90° clockwise.

For the scientific interpretation of the obtained textures, we determined the surface adhesion forces. In samples with inclined boundary positions, the strength of surface adhesion is established by the free field manner, which makes it possible to analyze the Bloch or Neel domain walls formed in the LC layer of the schlieren texture [22].

A schlieren texture [22] with point and linear defects, the so-called Bloch domain walls, is formed in a cell manufactured by us to conclude the surface adhesion durability for an LC film and a polymer film based on PVB (75%) + NLC 5CB (25%). If the plane of the domain wall contains optical anisotropy, then the reorientation of the director in the domains will be parallel to the wall. In this case, the LC molecules inside the droplet rotates in the plane of the wall, changing their direction to the opposite. The strength of the surface adhesion W_S to the polar surface can be determined by measuring the layer *d* of the Bloch walls [23,24]:

$$W_S\left(1-\frac{sin\,4\,\alpha}{2(\pi-2\alpha)}\right)=\frac{h}{d^2}x\frac{K_{22}}{2}x(\pi-2\alpha)^2 \quad (1)$$

where α is the angle between the preferential orientation of nematic molecules and the normal to the interface, h is the layer of the cell, K_{22} is the elastic torsion strain constant. We measured the thickness of the cell h = 13 µm and the thickness of the Bloch wall d = 3.0 µm. The angle α = 40° was taken from dimensions in liquid crystal droplets internal the polymer matrix (**Figure 3**) and K_{22}~10^{-12} (H) [23]. Using these data, the surface anchoring force to the polar surface was determined, which was equal to $W_S = 2.4\times10^6 \times K_{22}$~$10^{-6}$ (J m^{-2}).

4. Conclusions

In nematic droplets of spheroidal and ellipsoidal shape, with homogeneous anchoring transition, an inhomogeneous director field with topological point defects is always realized. Such orientation structures of NLC 5CB droplets are not optimal, especially due to the residual scattering of the orthogonal component of the incident radiation in the area of lateral defects. In order to obtain the maximum possible anisotropy of the light transmission of PDLC films, it is necessary to form nematic droplets with a uniform distribution of the director in the droplet volume. This option is possible only if inhomogeneous boundary conditions are realized, which vary in the meridian direction from tangential in the equatorial region to homeotropic near the poles of the drop of NLC 5CB. It was found that the nematic droplets have an orientation structure with two boojums and an annular topological defect. Such orientation structures are formed with uniform inclined coupling. Similar structures have previously been observed only inside liquid crystal droplets dispersed in their own isotropic phase [14], or in a liquid matrix doped with a homeotropic surfactant [11].

It is shown that the bipolar axis internal the droplets is oriented at different angles relative to the surface of the film, since the LC droplets inside the LC look like a flattened ellipsoid. This shape results in the orientation of the symmetrical axis of the bipolar droplets along the cell surface, while the symmetrical axis in the axial droplets inclines to be oriented orthogonal to the cell surface along the little axis of the ellipsoid.

It has been proved that the configuration with inclined coupling studied by us has attribute of either a bipolar structure (the central spindle-shaped part of the droplets close to the bipolar axis) and an axial one (the insignificant region of the droplets). Possibly, this feature of the orientation structure support a small difference in the energy of the LC at different orientations of the bipolar axis relative to the cell surface.

It has been established that PDLC cells with inclined boundary positions have an orientation structure combining appears of both bipolar and axial configurations [15]. Such accentuate open up opportunities for the use of these materials in the production of electro-optical devices with memory effect and low control voltage.

Conflict of interest: The author declares no conflict of interest.

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