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Friedericksz Threshold Field in Bipolar Nematic Droplets with Strong Surface Anchoring

O. O. Prishchepa, A. V. Shabanov, V. Ya. Zyryanov*, A. M. Parshin, and V. G. Nazarov

Kirensky Institute of Physics, Siberian Division, Russian Academy of Sciences, Akademgorodok, Krasnoyarsk, 660036 Russia

** e-mail: zyr@iph.krasn.ru*

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A numerical method has been developed for calculating the director configuration in ellipsoidal droplets of a nematic liquid crystal with strong tangential anchoring in a uniform magnetic field of an arbitrary orientation. A relation has been obtained for determining the Friedericksz threshold corresponding to the beginning of the reorientation of the central region of a droplet when the field is orthogonal to the bipolar axis. The effect of the breaking of the orthogonal condition on the threshold character of the orientation process is considered. The reorientation of the ensemble of bipolar droplets of the 5CB nematic liquid crystal dispersed in polyvinyl butyral has been studied by the magneto-optical method. Comparative analysis of calculation data and measured values of the threshold field has been performed.

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INTRODUCTION

Polymer-dispersed liquid-crystal films [1, 2] attract the attention of researchers owing to wide possibilities of their use in optoelectronics and display engineering. These films are based on a polymer matrix, where the liquid crystal is dispersed. In comparison with homogeneous liquid-crystal layers, composite films are characterized by a much more complex manifestation of various orientation effects induced by external fields (electric, magnetic, etc.).

In the case of the tangential anchoring of liquid-crystal molecules with the polymer surface, a bipolar director configuration with two surface point defects (boojums) is formed in nematic droplets [3]. In an external field, two scenarios are possible for transforming the bipolar structure depending in on the energy of the surface anchoring between nematic molecules and the polymer matrix. When anchoring is weak, the orientation transition in a liquid-crystal droplet is accompanied by the displacement of poles (boojums) [3]. In spherical droplets, this process is thresholdless. However, droplets in real structures have the ellipsoidal shape, which leads to the threshold character of their reorientation. The relation for determining the threshold field in nematic droplets with movable poles was derived in [3, 4].

For strong surface anchoring, the rigid fixation of poles is characteristic, and the Friedericksz transition is manifested in the reorientation of the central part of a droplet with a change in the symmetry of its bipolar structure [5, 6]. Such a transformation occurs in a threshold manner even in spherical droplets. The threshold field for this case was calculated in [6], but comparison with the experimental data obtained from electro-optical measurements shows that they differ from each other by a factor of about 2. The cause of this discrepancy was the imperfection of the computer model, which does not involve the ellipsoidal shape of real liquid-crystal droplets. Moreover, the interpretation of experiments concerning the reorientation of droplets by the electric field involves an unsolved problem of the correct inclusion of a complex distribution of the field strength in a heterophase medium with a large anisotropy of the dielectric constant of the liquid-crystal component. The problem can be simplified by using the magnetic field, because it is uniform due to the smallness of the gradient of the diamagnetic susceptibility χ at the interfaces of used components, as well as to the anisotropy of $\Delta\chi$ of nematic liquid crystals, as compared to the mean χ value.

The aims of this work are to develop the procedure for calculating the orientation structure of ellipsoidal nematic droplets with strong surface anchoring in a uniform external field, to calculate the threshold field for nematic droplets in real polymer-dispersed liquid-crystal samples, to measure the threshold characteristics of the Friedericksz transition by the magneto-optical

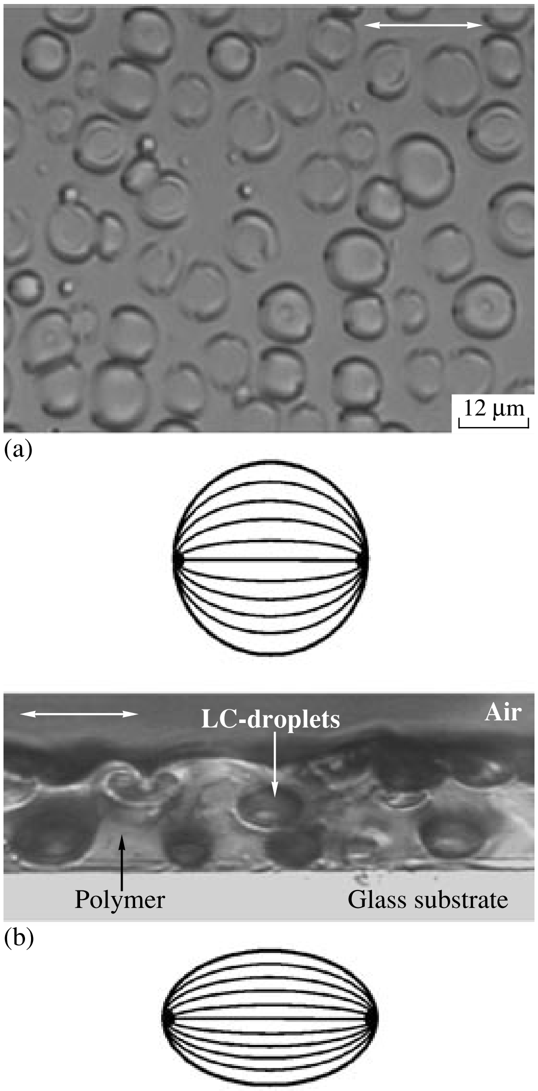

Fig. 1. (a) Texture of the polymer-dispersed liquid-crystal film in polarized light (the polarization direction is shown by the arrow) and the scheme of the bipolar director configuration in the nematic droplets in the film plane. (b) Microphotograph of the cross section of the film with the multilayer arrangement of the liquid-crystal droplets and the scheme of the corresponding director configuration.

method, and to comparatively analyze the theoretical and experimental data.

EXPERIMENT

Polymer-dispersed liquid-crystal film samples were prepared by solution technology [1, 2]. The mixture of the liquid crystal and polymer in the 1 : 1 weight ratio was solved in ethanol. The widely known 4-n-penthyl-4'-cyanobiphenyl (5CB) nematic liquid crystal with the clearing temperature T_c = 34°C is used. The polymer component is polyvinyl butyral, which ensures strong tangential anchoring with 5CB molecules [5]. The obtained homogeneous solution was poured out on a glass substrate. The further evaporation of the solvent leads to the phase separation of components with the formation of an ensemble of liquid-crystal droplets in a polymer film. Varying the content of the composition, film thickness, and ethanol evaporation rate, one can change the morphology of the polymer-dispersed liquid-crystal structure.

A certain spread in the sizes of the droplets exists in each sample under consideration (Fig. 1a). As seen, the liquid-crystal droplets have a circular shape in the film plane. Texture patterns characteristic of the bipolar director configuration are observed inside the droplets. The poles of the structure (point defects) are localized on opposite sides of the visible circle of the dots and are manifested in the form of dark dots when observed by means of a polarization microscope without analyzer (Fig. 1a).

In order to determine the transverse size of liquid-crystal droplet, the samples are cooled in liquid nitrogen and are broken by means of bending. Figure 1b shows the characteristic pattern of the cross section of the film with the multilayer arrangement of liquid-crystal droplets observed by using an optical microscope. The comparison of Figs. 1a and 1b shows that liquid-crystal droplets have the form of ellipsoids with the axes $a < b = c$ that are oblate in the film plane. For the samples being studied, the ratio of the shortest axis a of the droplets to the longest axis c is equal to $a/c = 0.7$ on average. In the samples with the monolayer arrangement of the droplets, the longest ellipsoid axes are predominantly oriented in the film plane. For samples with the multilayer droplet ensemble, the deviation of the longest axes from the film plane becomes noticeable (Fig. 1b). The location of the poles in the droplets under consideration agrees with the known condition of achieving the minimum elastic energy when the bipolar axis coincides with the longest axis of the ellipsoid cavity [3].

In order to perform magneto-optical measurements, a sample was placed in a constant magnetic field perpendicular to the polymer-dispersed liquid-crystal film plane. The setup makes it possible to vary the field strength in a range of 0–24 kOe. A He–Ne laser is used as a light source, whose beam is normally incident on the sample and is partially scattered, and its directly transmitted component is detected by a photodiode.

The measurements were performed at a temperature of T = 28°C. At this temperature, the anisotropy of the diamagnetic susceptibility of the 5CB liquid crystal is $\Delta\chi = 1.085 \times 10^{-7}$ [7] and the elastic-modulus components are $K_{11} = 4.7 \times 10^{-12}$ N, $K_{22} = 2.45 \times 10^{-12}$ N, and $K_{33} = 6.05 \times 10^{-12}$ N [8]. The refractive indices of the 5CB liquid crystal for light polarized along and across the detector are $n_\parallel = 1.711$ and $n_\perp = 1.537$, and the refractive index of the pure polyvinyl butyral polymer is $n_p = 1.490$ ($\lambda = 0.589$ μm).

In the magnetic field, the liquid-crystal director tends to be oriented along the field lines, i.e., perpendicular to the plane of the composite film. For the ratio of the refractive indices of the liquid crystal and polymer, this weakens the scattering of light in the composite film and, therefore, increases the intensity of directly passed light.

THEORY

The equilibrium director configuration in the nematic droplet in the magnetic field is calculated using the known procedure of minimizing the free energy of the liquid crystal [9] written in the one-constant approximation

$$F = \frac{1}{2}\int\{K[(\mathrm{div}\mathbf{n})^2 + (\mathrm{curl}\mathbf{n})^2] - \Delta\chi(\mathbf{n}\cdot\mathbf{H})^2\}dV. \quad (1)$$

Here, **n** is the unit vector describing the director field and **H** is the magnetic field vector. The free energy F is minimized in the three-dimensional model of the liquid-crystal droplet in the Cartesian coordinate system. The elastic constant is determined as the average value of three components, $K = (K_{11} + K_{22} + K_{33})/3$. The condition of the strong anchoring of the liquid-crystal molecules with the capsule surface is used.

The X axis coincides with the short axis a of the droplet ellipsoid, and the Z axis is directed along the bipolar axis coinciding with the longest axis c (Fig. 2). The axis-length ratio is taken to be equal to $a/c = 0.7$ that is the experimental value. The magnetic field vector **H** generally lies in the XZ plane at the angle φ to the X axis. The Y axis is perpendicular to the figure plane and the coordinate origin coincides with the center of the droplet. The director lines on the capsule surface coincide with meridians passing through two poles of the droplet. Such a choice of the boundary conditions and the field direction corresponds to the experiment carried out. Since the field vector **H** in the experiment is perpendicular to the film plane, the angle φ specifies deviations of the bipolar axes of the droplets from the film plane, which are noticeable for samples with the multilayer arrangement of the droplets (Fig. 1b).

The director field is determined at the points of a three-dimensional lattice separating the droplet volume into 100 equal intervals along the X, Y, and Z axes. Figure 2 shows the director configuration with a smaller number of intervals.

RESULTS

Calculations show that the bipolar director configuration is formed in the bulk of the ellipsoidal droplet in the absence of the field (Fig. 2a). The symmetry of the orientational structure of the droplet holds when the value of the field H, which is perpendicular to the bipolar axis ($\varphi = 0$), is small. The orientation of the director in the XY plane, as well as along the Z axis, remains perpendicular to the vector **H**. However, in other regions of the droplet bulk, where the director is initially nonorthogonal to the field, its angle to the YZ plane increases.

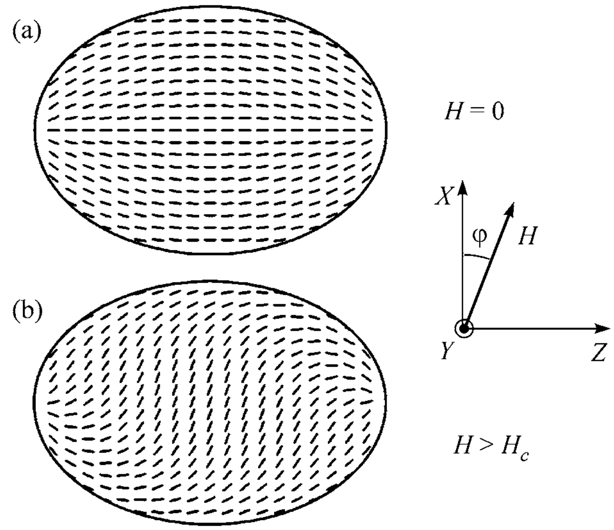

Fig. 2. Director-field distribution calculated in the central section XZ of the bipolar nematic droplet (a) in the initial state and (b) in the magnetic field exceeding the threshold value $H > H_c$.

When the field reaches the threshold value

$$H_c = \frac{A}{R}\sqrt{\frac{K}{\Delta\chi}} \quad (2)$$

the reorientation of the director in the XY plane and along the Z axis begins (see Fig. 2b) with a change in the symmetry of the orientational structure of the droplet. In this case, the characteristic S-shape bend of the director lines appears. The radius R determines the droplet size in the film plane, $R = c/2$. The parameter A determined from the calculations depends strongly on the anisometry of the droplet.

The threshold character of the reorientation process is seen in Fig. 3 for the dependence of the angle θ between the director at the droplet center and the Z axis. The abscissa axis shows the normalized magnetic field

$$A = HR\sqrt{\frac{\Delta\chi}{K}}. \quad (3)$$

It is seen that the director orientation at the droplet center remains unchanged as the field increases to $A = 3.64$. When the field exceeds the threshold value, the sharp reorientation of the director begins along the field with the saturation for $\theta \longrightarrow 90°$ in the region $A > 9$. Thus, according to the calculations, the value $A = 3.64$ determines the threshold field H_c given by Eq. (2) for oblate ellipsoidal droplets with the axis ratio $a/c = 0.7$.

For comparison, Fig. 3 also shows the dependence $\theta(A)$ calculated for the spherical droplet ($a/c = 1.0$).

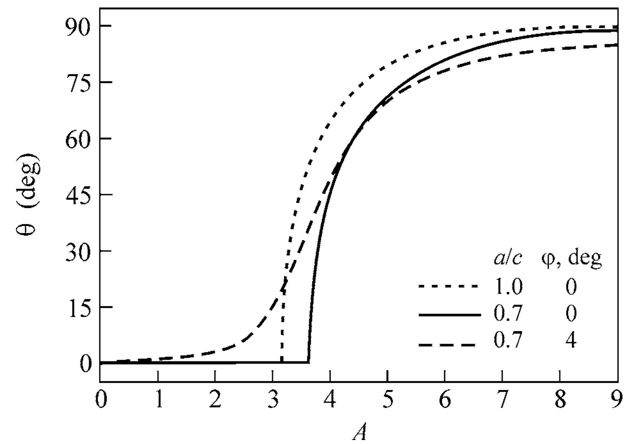

Fig. 3. Director orientation angle θ calculated at the center of the nematic droplet vs. the normalized field A for (dotted line) spherical droplets and for ellipsoidal droplets for the bipolar-axis deviation angle φ = (solid line) 0° and (dashed line) 4°.

In this case, the threshold field is determined by the value $A = 3.16$. Therefore, disregarding the ellipsoidal shape of the liquid-crystal droplets leads to the underestimation of the threshold field by approximately 13%.

Figure 4 shows light transmission as a function of the magnetic field for the polymer-dispersed liquid-crystal films with different sizes of droplets. For three samples under investigation (see Figs. 4a–4c), these dependences have a pronounced threshold form, which allows one to estimate the threshold field with an accuracy of 1 kOe. When analyzing experimental data, it is necessary to take into account that the threshold field is inversely proportional to the size parameter of the liquid-crystal droplets. Therefore, the lowest field is required for the reorientation of the largest droplets in the samples under investigation. Thus, the beginning of the increase in light transmission curves (which corresponds to experimental threshold field values) corresponds to the droplets with the maximum radius R_{max}.

It is worth noting that the light transmission curves for polymer-dispersed liquid-crystal films with large droplets (see Figs. 4c and 4d) exhibit oscillating behavior caused by the interference of light waves that pass through the liquid-crystal droplets and between them. This effect was observed and studied in detail in previous works [5, 6, 10]. The saturation region of light transmission is not reached for the films under investigation due to the limited capabilities of the electromag-

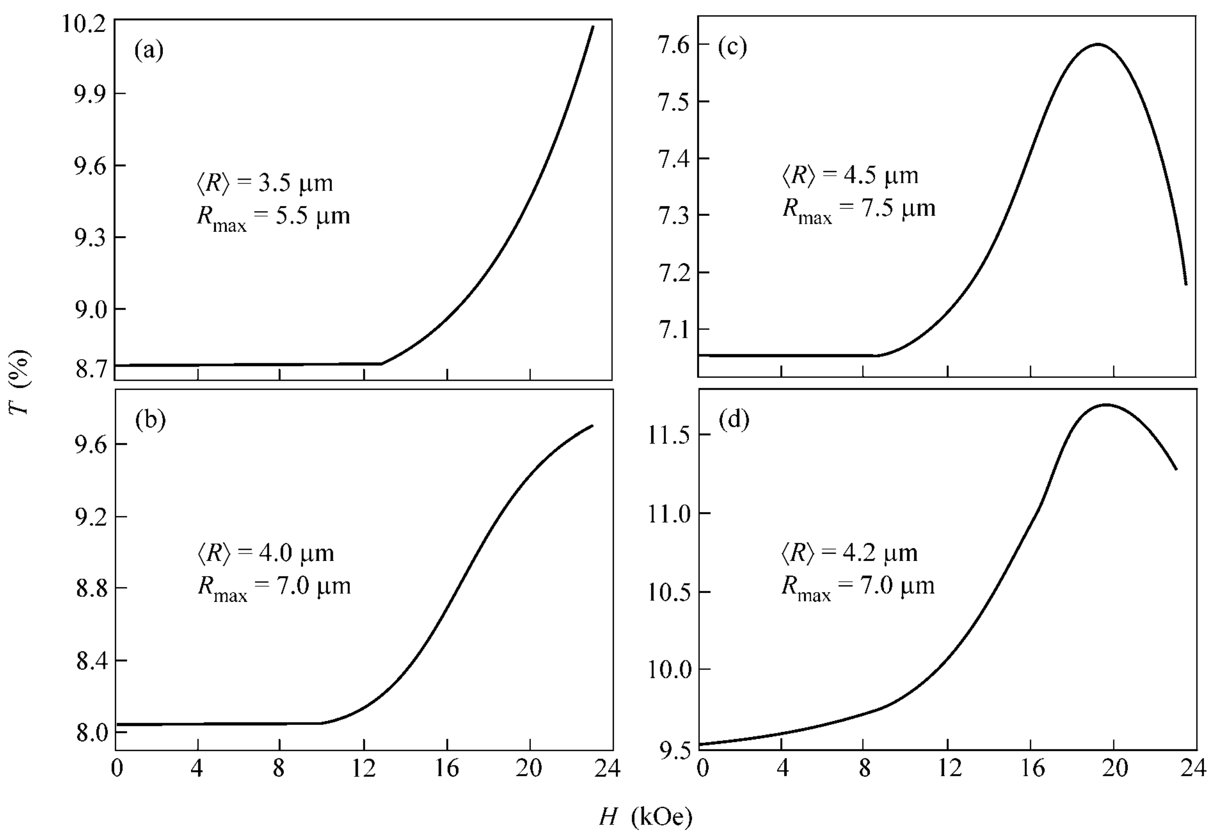

Fig. 4. Measured light transmission of polymer-dispersed liquid-crystal films vs. the magnetic field strength, where $\langle R \rangle$ is the ensemble-mean radius of the droplets in the film plane and R_{max} is the corresponding radius of the largest droplets.

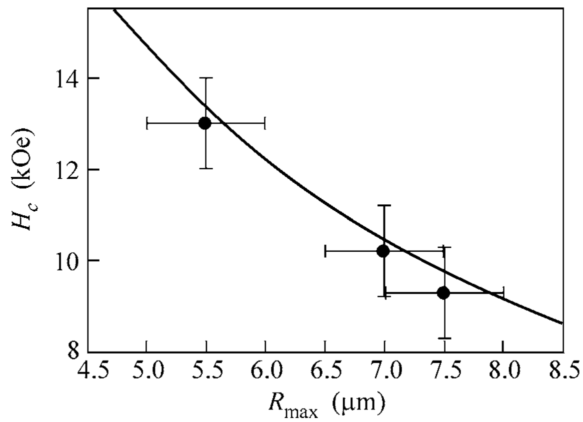

Fig. 5. Threshold field vs. the droplet radius R_{max}. The line is calculated by Eq. (2) for the value $A = 3.64$. The points indicate experimental data.

net used. For this reason, only one interference maximum is seen in Figs. 4c and 4d.

The presence of the horizontal section at the beginning of the light transmission curves is characteristic only of polymer-dispersed liquid-crystal films with the monolayer arrangement of the droplets whose long axes are parallel to the film plane (see Figs. 4a–4c). For a composite film with the multilayer arrangement of the droplets, light transmission begins to increase already for low field values (see Fig. 4d) and the break in the curve near the threshold is smoothed. Analysis of the cross section of this sample (see Fig. 1b) shows a noticeable deviation of the long axes of the droplets from the film plane by 4° on average. The dashed line in Fig. 3 shows the director orientation angle calculated for the bipolar axis deviation $\varphi = 4°$. The qualitative similarity of the shapes of the experimental (Fig. 4d) and theoretical (Fig. 3) dependences clearly indicates that the absence of the threshold character of the light transmission curve for such samples is explained by the deviation of the long axes of the droplets from the film plane. Therefore, a similar transformation of the threshold dependences can also be manifested for samples with the ideally planar orientation of the bipolar axes of all droplets when the vector **H** deviates from the normal to the film plane.

Finally, Fig. 5 shows the droplet-radius dependence of the threshold field calculated by Eq. (2) for $A = 3.64$ in comparison with the experimental H_c values taken from Figs. 4a–4c taking into account that they refer to the droplets with the maximum size R_{max}. As seen, the experimental and calculated values are in good agreement with each other within the experimental errors.

A smaller measured threshold field can be explained by the fact that the small deviations of the long axes of droplets from the film plane (smaller than 1°), which are not observable visually, exist in the samples with the monolayer droplet ensemble. However, such small deviations of the axes lead to a weak smoothing of the threshold break of the light transmission curve with its displacement towards smaller field values.

CONCLUSIONS

A numerical method has been developed in this work for determining the transformation of the director configuration in an ellipsoidal droplet of a nematic liquid crystal with allowance for both the anisotropy of the droplet shape and the deviation of the bipolar axis from the normal to the field vector. The model certainly describes the threshold character of the reorientation of real liquid-crystal films in the presence of the magnetic field, as well as explains the absence of the threshold behavior of the light transmission in films with a multilayer arrangement of nematic droplets.

It is worth noting that a similar pattern of the disappearance of the Friedericksz threshold was revealed and studied previously [11] when the homogeneous nematic layer is reoriented for the case of the nonorthogonal orientation of the field and the director. The specificity of the orientation process in the liquid-crystal dispersions under consideration is that the threshold dependence of the light transmission of the film exists only when the field is orthogonal to the bipolar axes of the droplets.

The developed approach is applicable not only for polymer-dispersed liquid-crystal films, but also for droplet liquid-crystal dispersions of other types. It is of particular importance for studying orientation processes in submicro- and nanodispersion liquid-crystal materials, because optical-microscopy observations of the transformation of the orientational structure are impossible for such structuring scales.

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