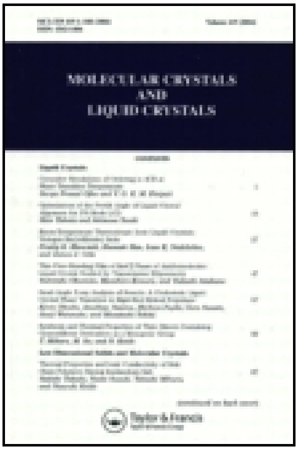

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Bipolar Nematic Droplets with Rigidly Fixed Poles in the Electric Field

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We have studied theoretically the reorientation process induced by the electric field inside a bipolar nematic droplet with rigidly fixed poles. Computer simulation has revealed the threshold director reorientation only in that part of a droplet, where the LC director is orthogonal to the applied field, and the nonthreshold reorientation in the rest of the droplet volume. A formula has been proposed to estimate the threshold field. The data obtained have allowed us to calculate a PDLC film transmittance in the framework of the anomalous diffraction approach and to interpret the oscillating behavior of the volt-contrast curve observed experimentally.

Keywords: polymer dispersed liquid crystals; nematics

INTRODUCTION

Reorientation of bipolar droplets of the nematic liquid crystal (LC) in the electric field was investigated by many authors[1,2]. The reorientation process consists in the moving of poles on the surface of a droplet (see Figures 1a, 1b) without disturbing the uniaxial symmetry of its volume, and according to the theoretical estimations[3,4] should have a threshold character.

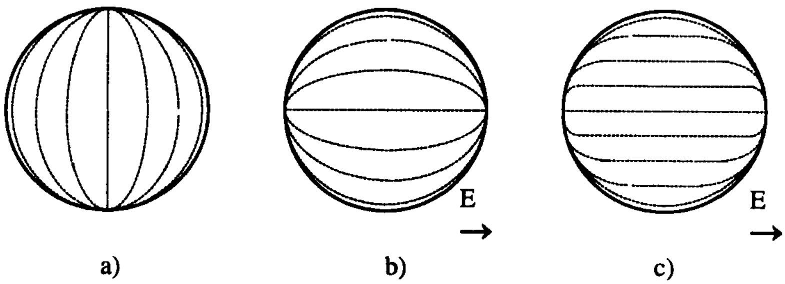

FIGURE 1 Reorientation process of bipolar nematic droplet with moving poles in the electric field.

The value of a threshold field is calculated in[3] in the one-constant approximation by the formula:

$$E_c = \frac{A}{D}\left(\frac{2\varepsilon_p + \varepsilon_{lc}}{3\varepsilon_p}\right)\left(\frac{K}{\varepsilon_0 \Delta\varepsilon}\right)^{1/2} \tag{1}$$

where K is the constant of elasticity; $D = (6V/\pi)^{1/3}$ is the diameter of a droplet, V is the droplet volume; parameter $A^2 = 100\,(l^2 - 1)\,/\,(l^2 + 1)$ takes into account the form of a droplet; $l = a/c$ is the ratio of the maximum axis a of the ellipsoid of the droplet to the minimum axis c; the factor $(2\varepsilon_p + \varepsilon_{lc})\,/\,3\varepsilon_p$ is the correction to calculate the electric field inside the LC droplet.

The formula obtained in the model[4] specially for small LC droplets

$$E_c = \frac{(l^2 - 1)^{1/2}}{a}\left(\frac{2\varepsilon_p + \varepsilon_{lc}}{3\varepsilon_p}\right)\left(\frac{K}{\varepsilon_0 \Delta\varepsilon}\right)^{1/2} \tag{2}$$

differs from (1) only in the first term.

The reorientation process at this stage (see Figure 1b) is not completed. A further increase of voltage results in a more homogeneous orientation of the director inside LC droplets (see Figure 1c). Such transformation of the director configuration (the electric field is parallel to the symmetry axis of a bipolar nematic droplet and there is a rigid tangential anchoring of LC molecules at the interface) was considered by computer simulation in[5].

The theoretical model[6], where the reorientation of a nematic in rectangular cell having the sizes $d_x \times d_y \times d_z$ is studied, is also of interest for our analysis. Here, initially, the director is oriented parallel to the axis Z in the entire volume, anchoring of the LC molecules at the boundaries is rigid. Then the value of the threshold field directed along the axis X, is calculated by the formula:

$$E_C = \pi \left(\frac{1}{\varepsilon_0 \Delta \varepsilon} \right)^{1/2} \left(\frac{K_{11}}{d_x^2} + \frac{K_{22}}{d_y^2} + \frac{K_{33}}{d_z^2} \right)^{1/2} \tag{3}$$

where K_{ii} ($i = 1,2,3$) are the coefficients of elasticity for S-, T- and B-deformations, respectively.

In papers[7,8], it was found experimentally, that during the reorientation process of bipolar droplets of nematic 5CB dispersed in polyvinylbutyral, the poles of a droplets remain rigidly fixed in the initial state. The possibility of occurrence of such situation was predicted earlier in[3].

In the films of polymer dispersed liquid crystals (PDLC) with large droplets, the effect of oscillation of the volt-contrast characteristic (VCC)[7,8] was also observed which needs a special theoretical consideration. The complexity of such analysis consists in the necessity to calculate the scattering cross-section of a LC droplet, which depends not only on its size and form, but also on its internal orientational structure.

As is known[9], the light passing through a scattering medium, containing N spherical particles in unit volume, is weakened, if there is no absorption and multiple light scattering, as

$$J = J_0 \exp(-N\sigma d) \quad . \tag{4}$$

Here σ is the scattering cross-section of a particle, d is the sample thickness. For the analysis of large droplets of a nematic ($kR >> 1$, R is the radius of a droplet, $k = 2\pi/\lambda$, λ denotes the light wavelength) in[5,10] it is proposed to use the anomalous diffraction approach, where the scattering cross-section

$$\sigma = 2\sigma_0\left[1-\frac{2}{\nu}\sin\nu+\frac{2}{\nu^2}(1-\cos\nu)\right] ; \tag{5}$$

$\sigma_0 = \pi R^2$ is the geometrical cross-section of a spherical droplet, the parameter

$$\nu = 2kR\left(\frac{n_{eff}}{n_p}-1\right) \tag{6}$$

where n_{eff} is the effective refractive index of LC, n_p is the refractive index of polymer.

Formula (5) is obtained[9] for the case of an optically isotropic particles. For an anisotropic LC droplets the problem becomes more complicated. However, as shown in[5,10], the correct determination of n_{eff} yields a good agreement between the calculated transmittance of the PDLC film and the experimental data.

We present here the results of computer simulation of the transformation of the director configuration in a bipolar nematic droplet with rigidly fixed poles, where the electric field is directed perpendicularly to the initial symmetry axis of a droplet. We use these data to calculate the VCC's of PDLC films and to interpret the experimental data.

THEORY

Similar to[5,11-13], the director configuration inside a nematic droplet in the electric field can be calculated in the one-constant approximation from the condition of minimum free energy

$$F=\frac{1}{2}\int\{K[(\mathrm{div}\ \underline{\mathbf{n}})^2+(\mathrm{rot}\ \underline{\mathbf{n}})^2]-\varepsilon_0\Delta\varepsilon(\underline{\mathbf{n}}\ \underline{\mathbf{E}})^2\}dV \tag{7}$$

where $\underline{\mathbf{n}}(\underline{\mathbf{r}})$ is the LC director, K is the coefficient of elasticity; $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$, where $\varepsilon_{\parallel}$, $\varepsilon_{\perp}$ are the dielectric permeability components, which are parallel and perpendicular to the director, $\underline{\mathbf{E}}$ is the vector of the electric field strength, which was considered as homogeneous in our simulation.

For further description we used the system of cartesian coordinates. The vector $\underline{\mathbf{n}}$ in this geometry can be represented as:

$$\underline{\mathbf{n}} = \underline{\mathbf{i}}\,X + \underline{\mathbf{j}}\,Y + \underline{\mathbf{k}}\sqrt{1-X^2-Y^2} \tag{8}$$

where the X and Y projections of the vector $\underline{\mathbf{n}}$ (the Z projection is replaced with an identical expression) generally depend on all three cartesian co-ordinates x, y, z. For this geometry, the unit volume is

$$dV = dx\,dy\,dz \quad . \tag{9}$$

In our case, the vector of the electric field strength $\underline{\mathbf{E}}$ is parallel to the X axis, then:

$$\underline{\mathbf{n}}\,\underline{\mathbf{E}} = n_x E_x + n_y E_y + n_z E_z = EX \tag{10}$$

The resultant expression for free energy will as:

$$\mathrm{F} = \frac{KR}{2}\iiint\limits_{V}\{2Z^2(Y_x X_y - X_x Y_y) + \ldots + E_n^2 X^2 Z^2\} / Z^2 dxdydz$$

$$Z^2 \equiv 1 - X^2 - Y^2 \tag{11}$$

where R is the radius of a spherical droplet of the liquid crystal, the indexes x, y, z - differentiation on these coordinates. The variational equations for the director configuration have the form:

$$\frac{\partial}{\partial x}\left(\frac{\partial F}{\partial X_x}\right) + \frac{\partial}{\partial y}\left(\frac{\partial F}{\partial X_y}\right) + \frac{\partial}{\partial z}\left(\frac{\partial F}{\partial X_z}\right) = \frac{\partial F}{\partial X}\,, \tag{12}$$

$$\frac{\partial}{\partial x}\left(\frac{\partial F}{\partial Y_x}\right) + \frac{\partial}{\partial y}\left(\frac{\partial F}{\partial Y_y}\right) + \frac{\partial}{\partial z}\left(\frac{\partial F}{\partial Y_z}\right) = \frac{\partial F}{\partial Y}\,. \tag{13}$$

With (11), we obtain from (12) and (13) the equations for the orientational state of the director configuration in a more detailed form:

$$X_{rr}XYZ^2 + Y_{rr}Y^2Z^2 + Y_{rr}Z^4 + Y^3Y_r^2 + Y_r^2YZ^2 - \\ - X_r^2Y^3 + X_r^2Y + 2XY^2R_r = 0 \tag{14}$$

$$E_n^2XZ^4 - X_{rr}Y^2Z^2 + X_{rr}Z^2 + Y_{rr}XYZ^2 + XY_r^2Y^2 + \\ + Y_r^2Z^2X - X_r^2XY^2 + X_r^2X + 2X^2YR_r = 0 \ , \tag{15}$$

where the following terms are used:

$$\begin{gathered} Z \equiv \sqrt{1 - X^2 - Y^2} \ , \\ X_{rr} = X_{xx} + X_{yy} + X_{zz} \ , \ Y_{rr} = Y_{xx} + Y_{yy} + Y_{zz} \ , \\ R_r = X_xY_x + X_yY_y + X_zY_z \ , \\ X_r^2 = X_x^2 + X_y^2 + X_z^2 \ , \ Y_r^2 = Y_x^2 + Y_y^2 + Y_z^2 \ , \\ E_n = \sqrt{\frac{\varepsilon_0 \Delta\varepsilon}{K}} R \cdot E = \frac{E}{E_0} \ , \ E_0 = \frac{1}{R}\sqrt{\frac{K}{\varepsilon_0 \Delta\varepsilon}} \ . \end{gathered} \tag{16}$$

In Figure 2, the results of computer simulation of the director configuration in bipolar nematic droplets for different values of E_n are shown. Here the symmetry axis of the initial structure is directed along the Z axis. The changes in the director reorientation for characteristic points inside a droplet (see Figure 3) are exhibited more obviously by the curves shown in Figures 4-6. In the field-off state, the orientational structure of a nematic droplet has a cylindrical symmetry $D_{\infty h}$. In the powered state, the droplet symmetry goes down to D_{2h}. In the range 0÷3.3 of E_n values, the director orientation at the Z axis and in the XOY plane does not vary (see Figure 4). The director in the rest of the droplet volume begins to turn along the field (see Figures 5,6). At point *C* the director projection not only on the X, Z axes, but also on the Y axis changes. Similar transformation of the director configuration should be occurred inside bipolar droplets with moving poles[3,4] in the range $0<E<E_c$ (see formulas (1),(2)).

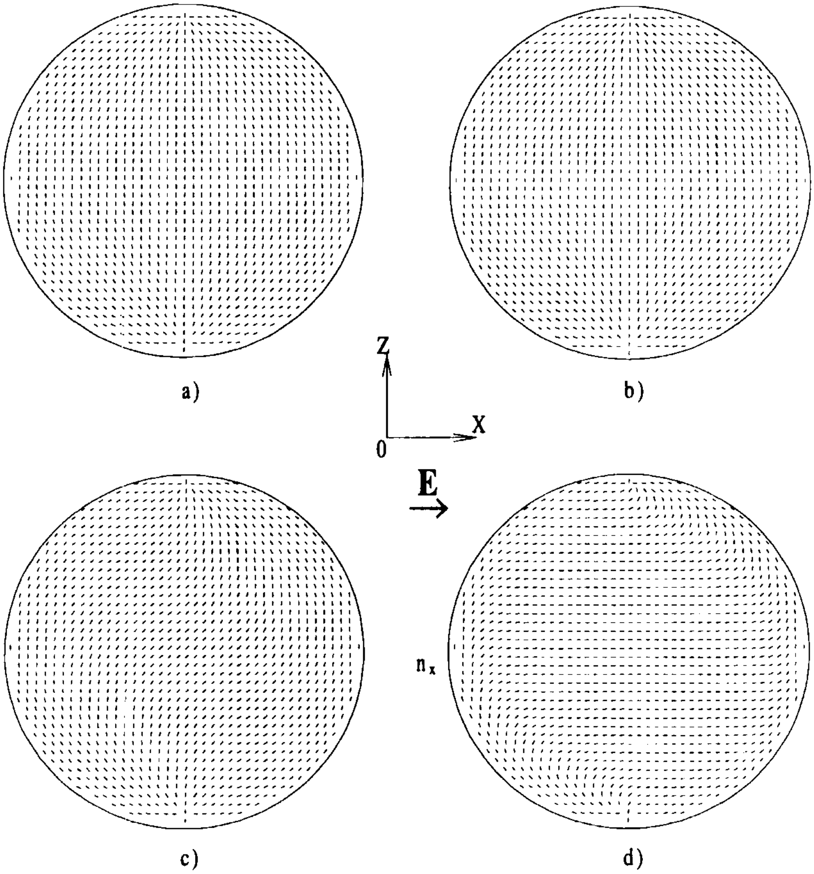

FIGURE 2 Transformation of the director configuration in a bipolar nematic droplet with rigidly fixed poles, where the electric field is directed perpendicularly to the initial symmetry axis of a droplet. a) E_n=0; b) E_n=3.2; c) E_n=3.7; d) E_n=10.

The director configuration essentially varies for $E_n > 3.3$ (see figure 2c,2d). The director in the center of a droplet, at the Z axis and in the XOY plane begins to turn. It results in the droplet symmetry going down to C_{2h}. The resultant director configuration in some features is similar to the one obtained in [6] for a rectangular cavity.

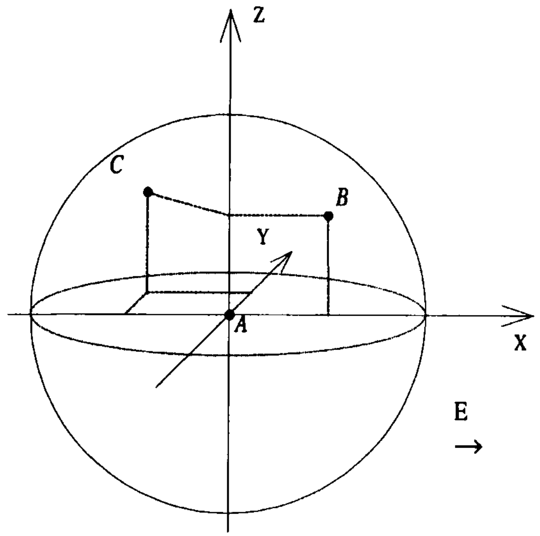

FIGURE 3 The arrangement inside the droplet of characteristic points *A, B, C*, in which the director reorientation is shown in the next figures.

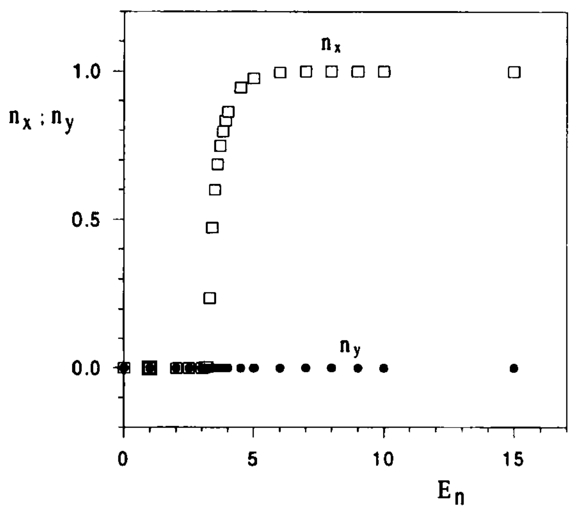

FIGURE 4 The director reorientation in the center of a droplet (point *A*, see Figure 3). n_x (□), n_y (•) are the X and Y projections of LC director. $n_z^2 = 1 - n_x^2 - n_y^2$.

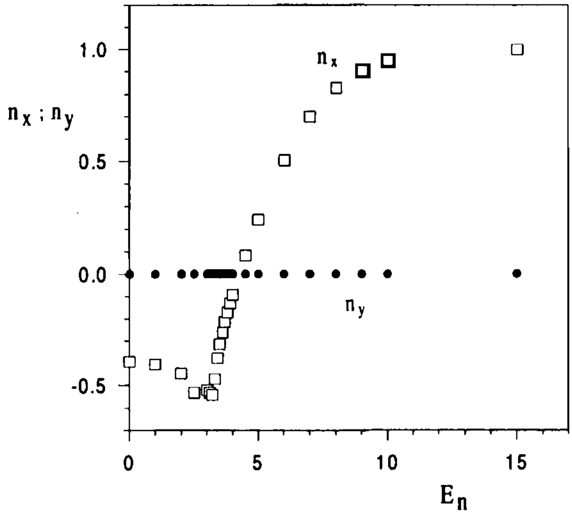

FIGURE 5 The director reorientation in the XOZ plane (point *B*, see Figure 3). n_x (□), n_y (•) are the X and Y projections of LC director. $n_z^2 = 1 - n_x^2 - n_y^2$.

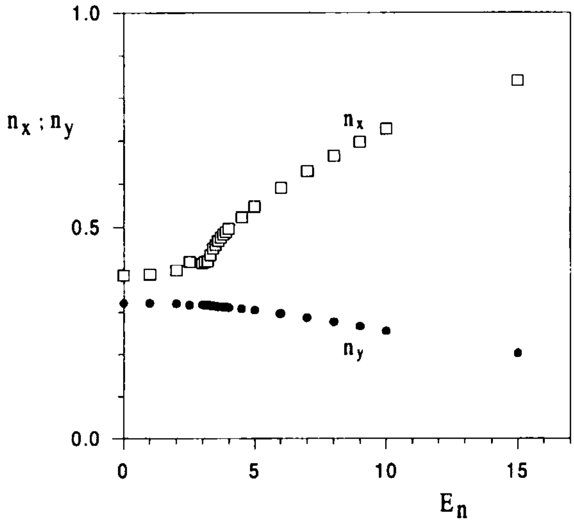

FIGURE 6 The director reorientation in the point *C* (see Figure 3). n_x (□), n_y (•) are the X and Y projections of LC director. $n_z^2 = 1 - n_x^2 - n_y^2$.

As it follows from the above said, the value of the critical field for a spherical nematic droplet is determined by the formula

$$E_c = E_{nc} E_0 = 3.3\ E_0 = 3.3\ \frac{1}{R}\sqrt{\frac{K}{\varepsilon_0 \Delta\varepsilon}}\ . \qquad (17)$$

It is interesting, that the Kilian's model[6] for a cubic cavity of 2R×2R×2R size in the one-constant approximation ($K_1 = K_2 = K_3 = K$) yields a threshold field of nearly the same value,

$$E_c = \pi\sqrt{3}/2\, E_0 = 2.7\, E_0 = 2.7\frac{1}{R}\sqrt{\frac{K}{\varepsilon_0 \Delta\varepsilon}}\ , \qquad (18)$$

which is approximately 1.2 times less, than the critical field (17), determined for a spherical droplet, inscribed in the cubic cavity.

It should be noted, that the process of reorientation of spherical nematic droplets with moving poles[3,4] is nonthreshold (E_c, calculated by formulas (1), (2) are equal to 0).

EXPERIMENT

We dispersed the nematic 5CB in polyvinylbutyral (PVB) in ratio 1:1 from the solution in ethanol. As is known[14], a rigid tangential anchoring of LC molecules and polymer is ensured in this composition. Refractive indexes of 5CB at $T = 22^0$C ($\lambda = 0.589\ \mu$m) $n_\perp = 1.534$, $n_\parallel = 1.725$, the refractive index of pure polyvinylbutyral $n_{PVA} = 1.492$. In the process of phase separating, part of 5CB remains in the polymer matrix, increasing its refractive index up to $n_p = 1.515 \div 1.535$ in dependence on the condition of the sample preparation.

The LC droplets had a round shape and were arranged in one layer in the film. The size of droplets was controlled by variation of the speed of ethanol evaporation. The distance between the droplets was approximately 1.5÷2.0

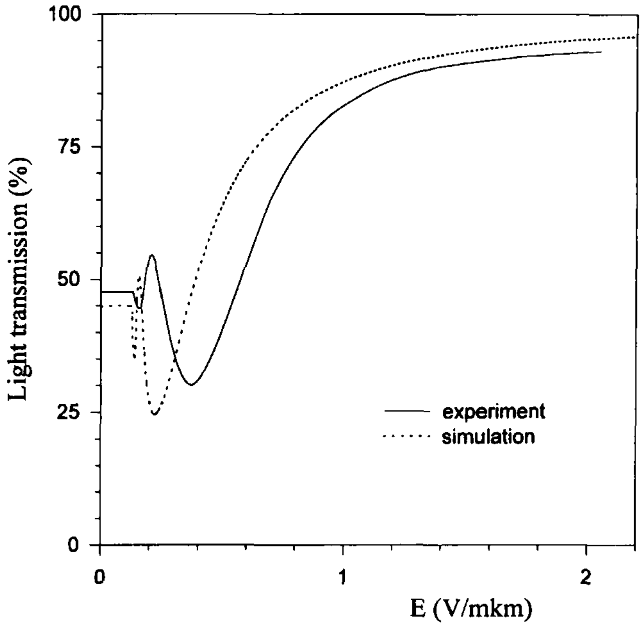

FIGURE 7 Volt-contrast characteristics of PDLC film with average size of droplets 19 μm. The values of parameters used in simulation: $N=10^5\ mm^{-3}$; $n_p=1.528$.

times larger than their diameter. Such ratio of structural parameters permits to ignore the effects of multiple light scattering in the interpretation of the experimental data[9]. The symmetry axis of the bipolar structure in all droplets lies in the film plane; thus, the condition of orthogonality of the electric field and the symmetry axis in the samples under study was satisfied.

As the observations with use of a polarizable microscope have shown, at small voltage no changes inside a droplet are visually detected. Apparently, it is due to the weak resolution of the human eye; and the instrument technique of the study of light scattering effects will allow to detect these changes.

At a certain voltage E_c (experimental) the change of brightness begins, which at first appears sharply in the center of a droplet, and then gradually extends in the entire volume. For large droplets, oscillation of brightness is observed with the increasing voltage, the brightness of a droplet alternately goes through minima and maxima. It should be noted, that no additional disclination inside a droplet appears in the field of oscillations.

The dependence of the light transmission of the PDLC film on the applied voltage was measured similar to[8]. For droplets with a diameter of less than 4 μm, VCC has a typical [1,2] S-form. For PDLC films with droplets of a greater size, there are one or more extrema (minima and maxima) on VCC[7,8]. The typical volt-contrast curve of PDLC films with great size of droplets are shown in Figure 7. The dependence of light transmission on the applied voltage clearly reveals a threshold behavior.

DISCUSSION

The VCC's calculated with the use formulas (4-6) are also shown in Figures 7. We used the expression

$$2R\, n_{\text{eff}} = \int_{-R}^{+R} n_{\text{lc}}(R)\, dR \tag{19}$$

as the effective value of the optical path for a light beam passing along the droplet diameter and parallel to the X axis (this beam determines mainly the intensity of the directly passing light).

It is clear, that in this case n_{lc} begins to vary only upon reaching the threshold field

$$E_c = \frac{\varepsilon_{lc}/\varepsilon_p + d_{lc}/d_p}{1 + d_{lc}/d_p} \cdot 3.3 \frac{1}{R} \sqrt{\frac{K}{\varepsilon_0 \Delta\varepsilon}} \tag{20}$$

obtained from (17) in view of the correction for the electric field inside a droplet. The correction factor in formula (20) was calculated also, like in a layered system, where the line of the electric field strength alternately passes through a polymer layer of $d_p/2$ thickness, the LC layer with thickness

$d_{lc} = 2R$, the polymer layer $d_p/2$. $d_p = d - 2R$, where d is the thickness of the PDLC film. We have taken for calculation the values: $K = (K_{11}+K_{22}+K_{33})/3$, $K_{11} = 6.2\times10^{-12}$ N, $K_{22} = 3.1\times10^{-12}$ N, $K_{33} = 8.3\times10^{-12}$ N [15], $\Delta\varepsilon = 11.8$ [16] and the experimentally measured ratio $\varepsilon_{lc} / \varepsilon_p = \varepsilon_{\perp} / \varepsilon_{lc} = 1.46$ [7,8].

As is seen, the calculated and experimental VCC's are qualitatively in good agreement with each other. For more exact simulation it is necessary to take into account the real distribution of droplets sizes and the ellipsoidal form of a droplet.

CONCLUSION

We have carried out a computer analysis of the transformation of the orientational structure inside bipolar nematic droplets with rigidly fixed poles in the electric field directed perpendicularly to the initial symmetry axis of the droplet. Simulation of the director configuration has revealed a complex behavior of the reorientation process. The threshold reorientation occurs only in that part of a droplet, where the LC director is orthogonal to the applied field. In the rest of the droplet, the director reorientation is nonthreshold. However at a low voltage ($E<E_c$), the transformation of the orientational structure does not give any noticeable contribution to the change of the light transmission of PDLC films, as the main contribution to this characteristic is given by the light beam passing along the droplet diameter.

A critical value of voltage was theoretically estimated, at which a dramatic change of the orientational ordering inside a droplet occurs causing the change in the light transmission of PDLC films. This value is in a good agreement with the experimental measurements, but for a more correct calculation it is necessary to take into account the unsphericity and difference in the size of real nematic droplets, and also to calculate correctly the value of the electric field inside a droplet. However the latter problem can be eliminated by using the magnetic measurements.

We used the results of computer simulation of a director configuration inside a droplet for calculation of VCC's within the framework of anomalous diffraction approach. The results of the calculation which has shown an oscillating behavior of VCC's for PDLC films with large droplets, are a convincing theoretical interpretation of detected earlier experimentally[7,8] effects.

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