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PHYSICAL OPTICS

Polarization of Light by a Polymer Film Containing Elongated Drops of Liquid Crystal with Inhomogeneous Interfacial Anchoring

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Abstract—An optico-mechanical model describing the coherent (directed) transmittance and the degree of polarization of forward-transmitted light by a polymer film with elongated liquid-crystal (LC) drops has been developed. This model, based on the Foldy–Twersky and anomalous-diffraction approximations, makes it possible to analyze the optical response of a film under extension as a function of the film thickness, refractive index of the polymer, the sizes and anisometry parameters of liquid-crystal drops, their concentration, internal structure, polydispersity, and orientation of optical axes. The model is verified based on the comparison of numerical and experimental data for the inverse modification of interfacial anchoring by an ion-forming surfactant. The internal drop structure is determined by solving the problem of minimizing the volume free energy density. A comparative analysis of the calculated transmittance and degree of polarization for films with uniform homeotropic and modified inhomogeneous interfacial anchoring is performed. The spectral polarization characteristics of a film with elongated LC drops and single-domain internal structure, formed under mechanical extension with the aid of surfactants, are investigated.

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INTRODUCTION

Polymer films with anisotropy of light absorption have become very popular light-polarizer materials [1, 2].

Their dichroism is caused by the introduction of special additives into the polymer matrix or the anisotropy of the intrinsic absorption of polymer macromolecules. The advantages of these polarizer films are their technological efficiency, compactness, and low cost. However, these films can be used in laser and optical devices only when the light intensity is sufficiently low, because the absorption of light may lead to heating and subsequent fracture of polymer matrix.

The maximum allowable power of incident light flux can be significantly increased using uniaxially elongated polymer-encapsulated liquid-crystal LC (PELC) films [3–5], where oriented ellipsoidal LC drops are introduced into an adhesive polymer matrix. These films, the operation of which is based on the scattering effect, efficiently polarize light in the entire transparency range of the components in use (visible and near-IR regions), whereas polaroids can polarize light in only the dichroic band of intrinsic or impurity absorption. In addition, their characteristics can be controlled by applying an electric field affecting the structure of the director field (local optical axes) in LC drops. PELC films are promising for laser devices, because the scattered light can easily be cut off by a diaphragm in the case of a collimated laser beam.

A new method has recently been proposed to control the electro-optical response of PELC films in the light-scattering mode, which is based on the local Freedericksz effect [6–8]. The essence of this effect is as follows: the structure of the director field in drops changes due to the inhomogeneous interfacial anchoring at the LC drop–polymer interface. The anchoring inhomogeneity is provided by surfactants. This way of controlling the internal structure of LC drops makes it possible to form a close-to-uniform orientation of director field in PELC films under extension. As a result, the light-polarization efficiency can be significantly increased [9, 10] in comparison with the techniques based on uniform interfacial anchoring.

The orienting effect of surfactant depends on its concentration at the interface in the PELC film. For example, when the concentration of surfactant cations is low (about 0.08%), they are arranged into long alkyl chains oriented generally parallel to the interface and set planar (tangential) anchoring of LC molecules on this interface. In this case, the direct modification of

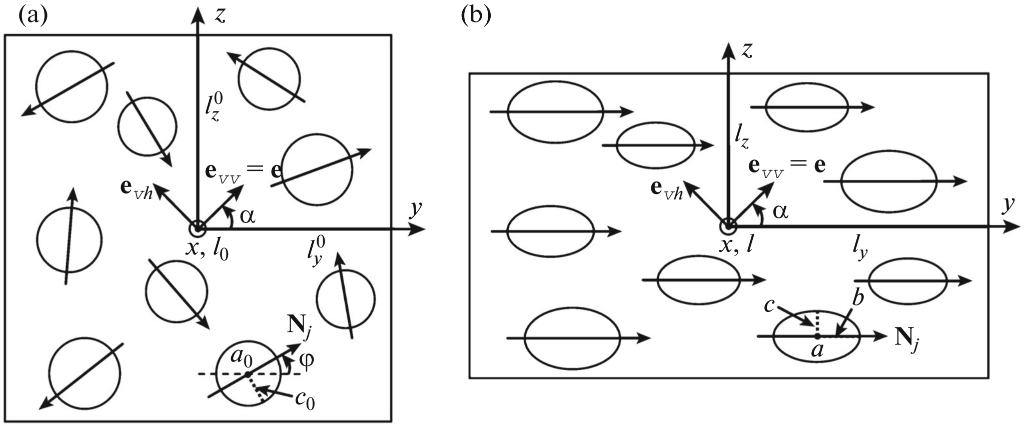

Fig. 1. Schematic diagram of the PELC layer structure. (a) Random orientation of directors (optical axes) $\mathbf{N}_j$ of spheroidal LC drops with semiaxes a_0 and c_0 before the extension and (b) uniformly oriented structure of directors $\mathbf{N}_j$ and elongated ellipsoidal drops with semiaxes a, b, and c after unidirectional extension along the y axis of the laboratory coordinate system (x, y, z). The (y, z) plane coincides with the layer-front surface; the x axis is directed along the normal to the layer (the direction of incidence of a linearly polarized plane wave with unit polarization vector $\mathbf{e}$); $\mathbf{e}_{vv}$ and $\mathbf{e}_{vh}$ are the unit vectors, along which the vv and vh components of the forward-transmitted wave are polarized (correspondingly, parallel and perpendicular to the plane of polarization $(x, \mathbf{e})$ of the incident wave); φ is the orientation angle of the drop optical axes $\mathbf{N}_j$; and l_0, l_y^0, l_z^0 and l, l_y, l_z are, respectively, the linear sizes of the PELC layer along the x, y, and z axes before and after the extension.

interfacial anchoring is implemented in the film, where the initial drop structure is bipolar [9]. At a high concentration of surfactant cations (about 1.6%), their alkyl chains are oriented perpendicular to the polymer surface and provide homeotropic (normal) boundary conditions. This case corresponds to the inverse interface modification with the initial radial structure of LC drops [10].

In this study, we consider uniaxially elongated PELC films. An optico–mechanical model is developed to describe the coherent (directed) transmittance of a PELC film and the degree of polarization of forward-transmitted light. An ensemble of polydisperse LC drops shaped as spheroids (or spheres) with optical axes randomly oriented before the film extension and an ensemble of oriented LC drops shaped as elongated ellipsoids under uniaxial extension are considered. The mean field and coherent transmittance of a PELC layer are analyzed within the Foldy–Twersky approximation [11, 12]. The anomalous diffraction approximation [12–14] is used to analyze the strictly forward scattering from an individual LC drop. The distribution of local optical axes in a drop was determined by solving the problem of minimizing the volume free-energy density [12, 15]. The model is experimentally verified by an example of a PELC film with inverse modification of interfacial anchoring and normal boundary conditions before the extension. It is shown that the modification of interfacial anchoring by ion-forming surfactants under extension makes it possible to significantly increase the transmittance of PELC films and the polarization efficiency of forward-transmitted light in comparison with the films that were not subjected to surfactant modification (with homogeneous interfacial anchoring on the surface of LC drops).

AN OPTICO–MECHANICAL MODEL FOR DESCRIBING THE COHERENT TRANSMITTANCE OF A PELC LAYER AND THE POLARIZATION OF FORWARD-TRANSMITTED LIGHT

Let a PELC layer be illuminated along the normal (the x axis of laboratory coordinate system (x, y, z)) by a linearly polarized plane wave with unit-polarization vector $\mathbf{e}$ (Fig. 1). The (y, z) plane in Fig. 1 coincides with the layer-front surface; $\mathbf{e}_{vv}$ and $\mathbf{e}_{vh}$ are the unit-polarization vectors of the vv and vh components of the forward-transmitted wave, which are polarized, respectively, parallel and perpendicular to the plane of polarization $(x, \mathbf{e})$ of the incident wave.

Let us make the following assumptions: (i) each drop in the layer is characterized by a director (optical axis) $\mathbf{N}_j$, where $j = \overline{1, N}$ and N is the number of drops in the layer (vector $\mathbf{N}_j$ characterizes the drop-volume-averaged orientation of the long axes of LC molecules [16–18]); (ii) in the absence of extension, the layer consists of an ensemble of polydisperse spheroidal LC drops with semiaxes a_0 and c_0 and random orientation of the optical axes $\mathbf{N}_j$ in the (y, z) plane (Fig. 1a); and (iii) after the layer extension along the y axis, the LC drops acquire a shape of elongated ellipsoids (with semiaxes a, b, and c), and the structure of their directors becomes uniform and y-oriented (i.e., vectors $\mathbf{N}_j$

are directed along the y axis, Fig. 1b). The semiaxes c_0, b, and c of LC drops lie in the (y, z) plane, and the semiaxes a_0 and a are directed along the x axis (Fig. 1). Note that, when the semiaxes a_0 and c_0 have equal lengths ($a_0 = c_0$), the initially unstrained PELC layer consists of spherical LC drops. Before the extension, the layer linear sizes along the x, y, and z axes of the laboratory coordinate system are, respectively, l_0, l_y^0, and l_z^0. After the extension, the linear layer sizes along the x, y, and z axes are, respectively l, l_y, and l_z. The layer thickness before and after the extension is l_0 and l, respectively.

Using the Foldy–Twersky approximation [11, 12] for the mean (coherent) field [19], we can write the expressions for the vv and vh components (T_a^{vv} and T_a^{vh}) of amplitude transmittance T_a of the normally illuminated PELC layer:

$$T_a^{vv} = t_2 \cos^2 \alpha + t_1 \sin^2 \alpha, \tag{1}$$

$$T_a^{vh} = (t_2 - t_1) \sin \alpha \cos \alpha, \tag{2}$$

$$t_{2,1} = \exp(ikl) \exp(-i\varphi_{2,1}) \exp(-\gamma_{2,1} l/2), \tag{3}$$

$$\varphi_{2,1} = q \operatorname{Im} \left\langle S_{2,1}^0 \right\rangle l, \tag{4}$$

$$\gamma_{2,1} = N_v \langle \sigma_{2,1} \rangle, \tag{5}$$

$$\langle \sigma_{2,1} \rangle = \frac{4\pi}{k^2} \operatorname{Re} \left\langle S_{2,1}^0 \right\rangle, \tag{6}$$

where φ_2 and φ_1 are the changes in phases for the y and z polarizations of a forward-transmitted wave within a layer of thickness l; γ and γ_1 are the corresponding attenuation coefficients of the layer; $\langle \sigma_2 \rangle$ and $\langle \sigma_1 \rangle$ are the mean cross sections of attenuation of an individual LC drop for the y and z polarizations, which are determined according to the optical theorem [20, 21]; $q = 2\pi k^{-2} N_v$; $k = 2\pi n_p/\lambda$; n_p is the refractive index of the polymer matrix; λ is the incident-light wavelength; N_v is the number of LC drops per unit volume; and $\left\langle S_{2,1}^0 \right\rangle$ are the diagonal elements of the amplitude scattering matrix of an individual LC drop at a zero scattering angle, which are averaged over the sizes of drops and orientations of their directors $\mathbf{N}_j$ (these elements must be found in the laboratory coordinate system xyz.

To determine $\left\langle S_{2,1}^0 \right\rangle$, we will consider an auxiliary coordinate system related to the main plane (x, $\mathbf{N}_j$) of an individual LC drop. In this system, amplitude matrix $\underline{\underline{S}}^0$ of an individual nematic LC drop for the strictly forward scattering direction is diagonal [13]:

$$\underline{\underline{S}}^0 = \begin{pmatrix} S_e^0 & 0 \\ 0 & S_o^0 \end{pmatrix}, \tag{7}$$

where S_e^0 and S_o^0 are the amplitude scattering functions for the extraordinary and ordinary waves, respectively. In the laboratory coordinate system, matrix $\underline{\underline{S}}^0$ is converted into matrix $\underline{\underline{S_{\text{lab}}^0}}$ according to the expression

$$\underline{\underline{S_{\text{lab}}^0}} = \begin{pmatrix} S_2^0 & 0 \\ 0 & S_1^0 \end{pmatrix} = \underline{\underline{R}}^{-1}(\varphi) \underline{\underline{S}}^0 \underline{\underline{R}}(\varphi), \tag{8}$$

$$\underline{\underline{R}}(\varphi) = \begin{pmatrix} \cos(\varphi) & \sin(\varphi) \\ -\sin(\varphi) & \cos(\varphi) \end{pmatrix} \tag{9}$$

is the transition matrix from the coordinate system related to the optical axis $\mathbf{N}_j$ of an individual drop to the laboratory basis (x, y, z), and $\underline{\underline{R}}^T(\varphi)$ is the transposed matrix $\underline{\underline{R}}(\varphi)$; φ is the azimuthal angle made by drop optical axis $\mathbf{N}_j$ with the y axis of the laboratory coordinate system (Fig. 1). Then, based on relations (7)–(9), we find

$$S_2^0 = S_e^0 \cos^2 \varphi + S_o^0 \sin^2 \varphi, \tag{10}$$

$$S_1^0 = S_e^0 \sin^2 \varphi + S_o^0 \cos^2 \varphi. \tag{11}$$

Using relations (5), (6), (10), and (11), we obtain the following expressions for attenuation coefficients γ_2 and γ_1:

$$\gamma_2 = \frac{4\pi}{k^2} N_v \left\langle \operatorname{Re} S_e^0 \cos^2 \varphi + \operatorname{Re} S_o^0 \sin^2 \varphi \right\rangle, \tag{12}$$

$$\gamma_1 = \frac{4\pi}{k^2} N_v \left\langle \operatorname{Re} S_e^0 \sin^2 \varphi + \operatorname{Re} S_o^0 \cos^2 \varphi \right\rangle. \tag{13}$$

Energy coefficient T_c^p of coherent (directed) transmittance of the PELC layer for linearly polarized incident light in the absence of analyzer is determined from the expression

$$T_c^p = T_c^{vv} + T_c^{vh}, \tag{14}$$

where T_c^{vv} and T_c^{vh} are the coherent transmittances determined, respectively, in parallel and crossed polarizer and analyzer:

$$T_c^{vv,vh} = \left| T_a^{vv,vh} \right|^2. \tag{15}$$

It follows from relations (1), (2), (14), and (15) that

$$T_c^p = \exp(-\gamma_2 l) \cos^2 \alpha + \exp(-\gamma_1 l) \sin^2 \alpha. \tag{16}$$

To determine coherent transmittance T_c^{tr} of the layer illuminated by unpolarized light, it is necessary to average (16) over polarization angle α. Then we arrive at

$$T_c^{tr} = \frac{T_\parallel + T_\perp}{2}, \tag{17}$$

$$T_{\parallel,\perp} = T_c^p(\alpha = 0, \pi/2) = \exp(-\gamma_{2,1} l), \quad (18)$$

these being the layer transmittances, which are determined in parallel polarizer and analyzer oriented, respectively, along the y axis of the laboratory coordinate system ($\alpha = 0$) and perpendicularly to it ($\alpha = \pi/2$).

The degree of polarization P of forward-transmitted light will be defined as

$$P = \frac{T_\perp - T_\parallel}{T_\perp + T_\parallel}. \quad (19)$$

Note that formulas (1)–(4), (12), (13), and (16)–(19) allow one to determine the coherent transmittance and Stokes parameters [21] for a PELC layer exposed to linearly polarized light, the coherent transmittance and degree of polarization of light when the latter is unpolarized, the linear and circular dichroisms, etc. To this end, one must know the elements of amplitude scattering matrix S_e^0 and S_o^0 of an individual LC drop (see expressions (12) and (13)) and take into account the layer structural characteristics when averaging over the sizes of drops and orientations of their optical axes $\mathbf{N}_j$. The problem of scattering from an individual LC drop can be solved using different methods [20, 21]: the dipole [11], Rayleigh–Gans [22, 23], anomalous diffraction [12–14, 24], Wentzel–Kramers–Brillouin [25], and other approximations.

We will analyze the light scattering from an individual nematic LC drop using the anomalous-diffraction approximation [20]. Within this approach, the light field scattered by a large, optically soft drop is determined as the result of the diffraction from an equivalent flat amplitude-phase screen with a complex transmittance matrix, specified in its main cross section $\sigma = \pi bc$ in the layer plane yz. The elements of the amplitude scattering matrix $S_{e,o}^0$ entering into expressions (12) and (13) are determined as [12–14]

$$S_{e,o}^0 = \frac{k^2}{2\pi}\int_\sigma \left(1 - T_{2,1}(y,z)\right) \mathrm{d}y\mathrm{d}z, \quad (20)$$

where $T_{2,1}(y, z)$ are the diagonal elements of the Jones matrix $\underline{\underline{T}}(y,z)$ of the equivalent screen,

$$\underline{\underline{T}}(y,z) = \prod_{x=x_{\rm inp}(y,z)}^{x_{\rm out}(y,z)} R^T(x)P(\Delta x)R(x). \quad (21)$$

Here, $x_{\rm inp}$ and $x_{\rm out}$ are, respectively, the input and output coordinates of the wave front on the LC drop surface, which depend on the coordinates y and z: $x_{\rm inp,out} = \mp a\sqrt{1 - y^2/b^2 - z^2/c^2}$, $P(\Delta x)$ is the matrix determined by the local phase delays for the extraordinary and ordinary waves within an elementary drop volume with longitudinal size Δx, and $R(x)$ and $R^T(x)$ are the coordinate transformation matrices along the local basis path. Matrices $P(\Delta x)$, $R(x)$, and $R^T(x)$ depend on the orientational structure of the local director in the LC drop volume [12–14].

Let us consider the transition from the initial disordered state of the PELC layer to the state of orientational ordering of the optical axes of LC drops caused by the layer extension along the y axis of the laboratory coordinate system (Fig. 1). Assuming that the extension does not change the layer volume, we can write the dependence of the layer linear sizes, l, l_y, and l_z, in the form [4]

$$l = l_0 p^{-B}, \quad l_y = l_y^0 p, \quad l_z = l_z^0 p^{-A}, \quad (22)$$

where $p = l_y/l_y^0$ is the extension ratio, which is equal to the ratio of the lengths of the layer portion under consideration in the deformed ($p \neq 1$) and initial ($p = 1$) states, and A and B are constants determined by the mechanical properties of the polymer; note that $A + B = 1$.

We assume that the drops in the initial state are shaped as triaxial ellipsoids with semiaxes a_0, b_0, and c_0; the semiaxes b_0 and c_0 are oriented parallel to the (y, z) plane, whereas the semiaxes a_0 are directed along the normal to the layer (i.e., along the x axis). It is also assumed that the drop optical axis $\mathbf{N}_j$ is directed along the major semiaxis b_0 and makes an angle φ_0 with the y axis. Then the semiaxes a, b, and c of the drop and orientation angle φ of its optical axis $\mathbf{N}_j$ are determined by the relations [4]

$$a = a_0 p^{-B}, \quad (23)$$

$$b = \frac{\sqrt{2} b_0 p}{\sqrt{K_2 - \sqrt{K_1^2 + M^2}}}, \quad (24)$$

$$c = \frac{\sqrt{2} c_0 p}{\sqrt{K_2 + \sqrt{K_1^2 + M^2}}}, \quad (25)$$

$$\varphi = \frac{1}{2}\arctan\frac{M}{K_1}, \quad (26)$$

$$K_{2,1} = (p^{2(A+1)}\varepsilon_o^2 \pm 1)\cos^2\varphi_0 \pm (\varepsilon_o^2 \pm p^{2(A+1)})\sin^2\varphi_0, \quad (27)$$

$$M = (\varepsilon_o^2 - 1)p^{A+1}\sin 2\varphi_0, \quad (28)$$

where $\varepsilon_0 = b_0/c_0$ is the anisometry parameter of the drop shape in the (y, z) plane before the layer extension.

It follows from formulas (23)–(25), (27), and (28) that an increase in the extension ratio p leads to a monotonic increase in the major (longitudinal) semiaxis (b) and a monotonic decrease in the small semiaxes (a and c). For an initially ellipsoidal LC drop subjected to extension, the lengths and orientations of its semiaxes b and c depend on the initial orientation of angle φ_0 of the optical axis $\mathbf{N}_j$. Transverse semiaxis length a is independent of φ_0 (see expression (23)).

Figure 2 illustrates the dependence of orientation angle φ of the drop optical axis $\mathbf{N}_j$ on extension ratio p at different values of anisometry parameter ε_0 before the extension. One can see that, the closer the ratio of semiaxes $\varepsilon_0 = b_0/c_0$ to unity, the smaller p the values that are required to reach the same φ values.

At $\varepsilon_0 = 1$ (initially spherical or spheroidal LC drops, see Fig. 1a), angle φ is a step function:

$$\varphi = \begin{cases} \varphi_0, & p = 1 \\ 0, & p > 1 \end{cases}, \tag{29}$$

where φ_0 is a random value, uniformly distributed in the range from 0° to 360°. Then, as follows from expressions (23)–(25), (27), and (28), the semiaxes a, b, and c of an individual LC drop depend on extension ratio p of the layer in the same way as its linear sizes (relation (22)):

$$a = a_0 p^{-B}, \quad b = c_0 p, \quad c = c_0 p^{-A}, \tag{30}$$

where $a_0 = c_0$ for spheres and $a_0 < c_0$ for spheroids. If the LC drops are spherical in the initial state, $A = B = 0.5$. For initially spheroidal drops, $A \approx 0.4$ and $B \approx 0.6$ [4].

Assuming that the PELC layer in the unstrained state consists of spherical or spheroidal LC drops ($\varepsilon_0 = 1$) and carrying out independent averaging over their sizes and orientations of optical axes (which is valid at $\varepsilon_0 = 1$, as follows from relations (24)–(28)), we obtain the attenuation coefficients (see relations (12) and (13)) in the form

$$\gamma_2 = \frac{3}{4} c_{\mathrm{v}} \frac{\langle a^2 \rangle}{\langle a^3 \rangle} \left\{ \langle Q_{\mathrm{e}} \rangle \frac{1 + S_{2\mathrm{f}}}{2} + \langle Q_{\mathrm{o}} \rangle \frac{1 - S_{2\mathrm{f}}}{2} \right\}, \tag{31}$$

$$\gamma_1 = \frac{3}{4} c_{\mathrm{v}} \frac{\langle a^2 \rangle}{\langle a^3 \rangle} \left\{ \langle Q_{\mathrm{e}} \rangle \frac{1 - S_{2\mathrm{f}}}{2} + \langle Q_{\mathrm{o}} \rangle \frac{1 + S_{2\mathrm{f}}}{2} \right\}, \tag{32}$$

$$\langle Q_{\mathrm{e,o}} \rangle = \frac{4 \operatorname{Re} \langle S^0_{\mathrm{e,o}} \rangle}{k^2 \varepsilon_y \varepsilon_z \langle a^2 \rangle}, \tag{33}$$

$$S_{2\mathrm{f}} = 2\overline{\cos^2 \varphi} - 1 = \begin{cases} 0, & p = 1 \\ 1, & p > 1 \end{cases}, \tag{34}$$

where $c_{\mathrm{v}} = N_{\mathrm{v}} \langle V \rangle$ is the volume-filling factor of the layer, $\langle V \rangle$ is the mean drop volume; $\langle Q_{\mathrm{e,\,o}} \rangle$ are the drop-size-averaged attenuation efficiency factors for the extraordinary and ordinary waves, respectively; $S_{2\mathrm{f}}$ is the 2D order parameter [5] of the PELC layer (the bar above in expression (34) indicates averaging over orientation angle φ_0); $\varepsilon_y = b/a = (c_0/a_0)p^{1+B}$ and $\varepsilon_z = c/a = (c_0/a_0)p^{B-A}$ are the drop-shape anisometry parameters under extension; and $a = a_0 p^{-B}$.

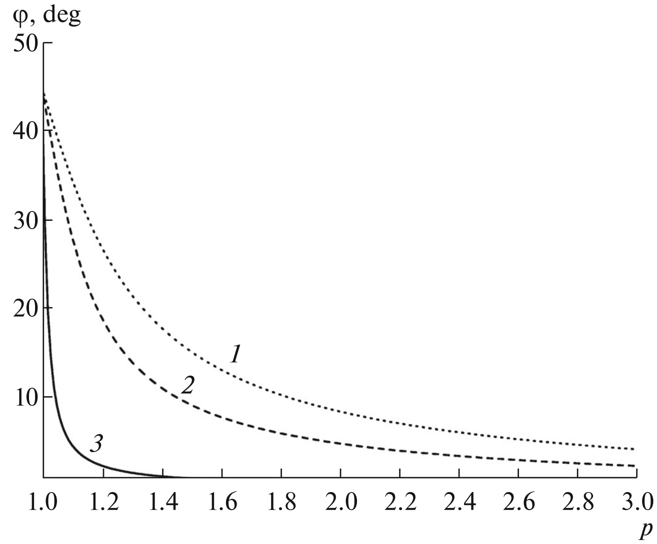

Fig. 2. Dependence of optical-axis orientation angle φ for an individual LC drop in a PELC layer on extension ratio p at different axis ratios ε_0 in the initial unstrained state. The optical-axis orientation angle before the extension is $\varphi_0 = 45°$; $\varepsilon_0 =$ (*1*) 1.5, (*2*) 1.25, and (*3*) 1.025.

Thus, coherent transmittances $T_{\mathrm{c}}^{\mathrm{tr}}$, $T_{\parallel}$, and $T_{\perp}$ of a layer of polydisperse drops and degree of polarization P of forward-transmitted light can be found using relations (17)–(19), where l must be taken in the form $l = l_0 p^{-B}$, and attenuation coefficients γ_2 and γ_1 are given by relations (31) and (32), respectively.

An analysis of expression (20) [12] at identical anisometry parameters ε_y and ε_z and the same internal drop structure shows that the amplitude scattering functions $S^0_{\mathrm{e,o}}$ depend on only the transverse semiaxis $a = a_0 p^{-B}$:

$$S^0_{\mathrm{e,0}} = S^0_{\mathrm{e,o}}(a). \tag{35}$$

Then, using the mean-value theorem [12], we obtain the following expressions for attenuation coefficients γ_2 and γ_1 of the PELC layer (see relations (31) and (32)):

$$\gamma_2 = \frac{3 c_{\mathrm{v}}}{4 a_{\mathrm{ef}}} \left\{ Q_{\mathrm{e}}(a_{\mathrm{ef}}) \frac{1 + S_{2\mathrm{f}}}{2} + Q_{\mathrm{o}}(a_{\mathrm{ef}}) \frac{1 - S_{2\mathrm{f}}}{2} \right\}, \tag{36}$$

$$\gamma_1 = \frac{3 c_{\mathrm{v}}}{4 a_{\mathrm{ef}}} \left\{ Q_{\mathrm{e}}(a_{\mathrm{ef}}) \frac{1 - S_{2\mathrm{f}}}{2} + Q_{\mathrm{o}}(a_{\mathrm{ef}}) \frac{1 + S_{2\mathrm{f}}}{2} \right\}, \tag{37}$$

$$Q_{e,o}(a_{\mathrm{ef}}) = 2 \operatorname{Re} \int\limits_{\sigma = \pi b c} (1 - T_{2,1}(a_{\mathrm{ef}}))\, d\sigma, \tag{38}$$

where

$$a_{\mathrm{ef}} = \langle a^3 \rangle / \langle a^2 \rangle \tag{39}$$

is the effective length of semiaxis a.

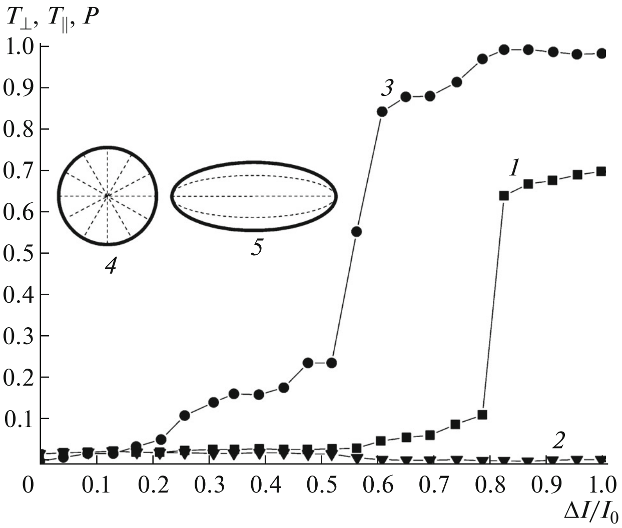

Fig. 3. Experimental dependences of coherent transmittances (*1*) $T_\perp$ and (*2*) $T_\parallel$ and (*3*) degree of polarization P of forward-transmitted light on elongation factor $\Delta l/l_0$ of a PELC film with modified interfacial anchoring; (*4*) and (*5*) are, respectively, schematic diagrams of a spherical LC drop in a layer with radial structure before the extension and an elongated ellipsoidal drop in the extended film. The LC is 5CB ($n_\perp = 1.53$, $n_\parallel = 1.717$ at $\lambda = 0.633$ μm). Before the extension, the film thickness is $l_0 = 45$ μm and the mean drop radius is $\langle a_0 \rangle = 2$ μm. The volume-filling factor of the film is $c_v = 0.143$.

For a layer consisting of monodisperse LC drops, coherent transmittances $T_\parallel$ and $T_\perp$ can be written in the form

$$T_{\parallel,\perp} = \exp(-\tau_{\parallel,\perp}), \tag{40}$$

$$\tau_\parallel = \tau_\perp = \frac{3}{8} c_v \frac{l_0}{a_0} \{Q_e + Q_o\}, \quad p = 1, \tag{41}$$

$$\tau_{\parallel,\perp} = \frac{3}{4} c_v \frac{l_0}{a_0} Q_{e,o}, \quad p > 1, \tag{42}$$

$$Q_{e,o} = \frac{4}{\varepsilon_y \varepsilon_z k^2 a^2} \operatorname{Re} S^0_{e,o}, \tag{43}$$

where $\tau_\parallel$ and $\tau_\perp$ are the layer optical densities for light beams polarized parallel and perpendicular to the extension direction.

EXPERIMENTAL VERIFICATION AND ANALYSIS OF THE RESULTS

To compare the results obtained within the developed model with the measurement data, we used the experimental dependences of transmittances $T_\parallel$ and $T_\perp$ in the inverse mode of interfacial anchoring modification [10] for a composite film based on nematic 5CB. Its ordinary ($n_\perp$) and extraordinary ($n_\parallel$) refractive indices are $n_\perp = 1.53$ and $n_\parallel = 1.717$ at a wavelength of $\lambda =$ 0.633 μm. The sample was prepared by emulsifying the nematic LC in an aqueous solution of a mixture of polymer, glycerol, and surfactant, with subsequent solvent evaporation. The polymer was polyvinyl alcohol (PVA); its refractive index is $n_p = 1.532$. The n_p value of the sample changes in the range from 1.49 to 1.53 due to the glycerol added. Cetyltrimethylammonium bromide (CTMB) was used as a cationic surfactant. The component weight ratio was PVA : glycerol : 5CB : CTMB = 1 : 0.3 : 0.2 : 0.006. A recalculation to volume-filling factor c_v of LC drops in the film yields $c_v = (0.2 + 0.006)/(1 + 0.3) \approx 0.158$. The drop concentration in the prepared sample is reduced to $c_v = 0.143$ due to the partial LC dissolution in the polymer and the presence of residual water. The used amount of surfactant (3 wt % with respect to the LC) provides normal boundary conditions and radial configuration of LC drops before the extension. The PVA–glycerol–5CB–CTMB heterogeneous mixture was formed using rotation in a special agitator and deposited on the surface of a glass substrate with subsequent drying in air. The thus-obtained composite film was subjected to unidirectional extension.

The results of measuring the light transmission anisotropy (transmittances $T_\parallel$ and $T_\perp$) and the degree of light polarization for the PELC film under extension are presented in Fig. 3. Film-elongation factor $\Delta l/l_0 = (l - l_0)/l_0$, which is related to extension ratio p by the expression $p = 1 + \Delta l/l_0$, is plotted on the abscissa axis. In the absence of extension, the drops were spherical, with a mean radius of $\langle a_0 \rangle = 2$ μm and a small size dispersion. The film thickness was $l_0 =$ 45 μm. A polarization microscopic analysis revealed that elongation factor $\Delta l/l_0 = 1$ ($p = 2$) corresponds to drops elongated in the extension direction, shaped as ellipsoids of revolution with respect to the extension axis with a defect-free homogeneously oriented (single-domain) structure of the local director. The physical nature of the formation of defect-free single-domain structures of drops under extension and in the presence of surfactant consists in the occurrence of inhomogeneous surface anchoring of LC molecules at the drop–polymer interface, which changes in the meridional direction from tangential (on the equator) to homeotropic (on the poles) [9].

Figure 4 shows the dependences of $T_\parallel$ and $T_\perp$ on refractive index n_p of the polymer matrix, calculated for an unstrained PELC film at $p = 1$ (elongation factor $\Delta l/l_0 = 0$) with the initial radial structure of LC drops and for a stretched film with a single-domain internal structure of drops at an extension ratio of $p =$ 2 (elongation factor $\Delta l/l_0 = 1$). The calculations were based on relations (18) and (36)–(39). For an extension ratio of $p = 2$, the drop-shape anisometry parameters are $\varepsilon_y = b/a \approx 2.83$ and $\varepsilon_z = c/a = 1$. For initially spherical drops, $\varepsilon_y = \varepsilon_z = 1$. Under twofold extension ($p = 2$), the film thickness is $l = l_0/\sqrt{2} \approx 32$ μm.

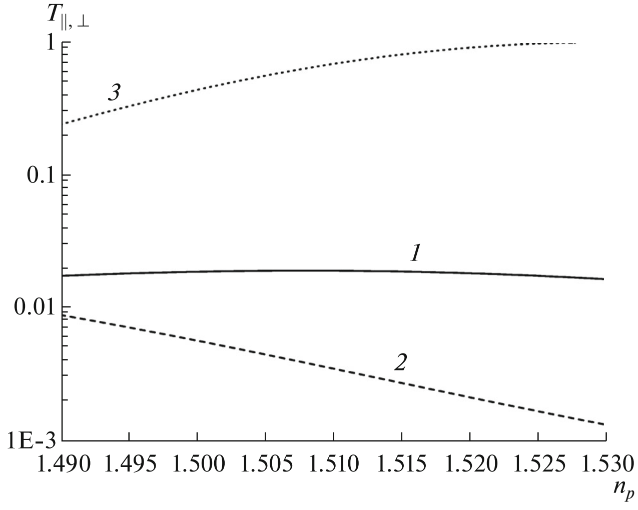

Fig. 4. Calculated dependences of transmittances $T_\|$ and $T_\perp$ on refractive index n_p of the polymer matrix. (*1*) $T_{\perp,\|}$ for an unstrained PELC film ($p = 1$, $l_0 = 45$ μm, $c_v = 0.143$, $a_{ef} = 2$ μm, $\varepsilon_y = \varepsilon_z = 1.0$). (*2*) $T_\|$ and (*3*) $T_\perp$ for a deformed film at an extension ratio of $p = 2$ ($l = 32$ μm, $c_v = 0.143$, $a_{ef} = 1.4$ μm, $\varepsilon_y = 2.83$, $\varepsilon_z = 1.0$).

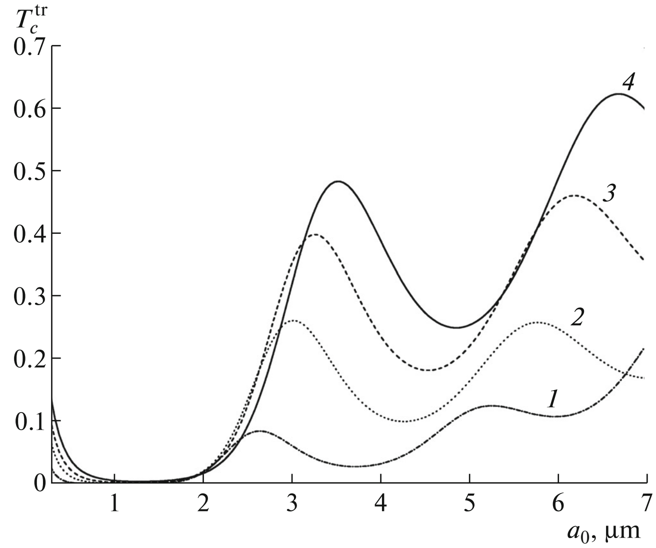

Fig. 5. Dependences of coherent transmittance T_c^{tr} of a PELC layer before the extension ($p = 1$) on drop radius a_0 at different values of refractive index n_p of the polymer matrix: $n_p =$ (*1*) 1.49, (*2*) 1.51, (*3*) 1.52, and (*4*) 1.53. Radial structure of LC drops; $n_\perp = 1.53$, $n_\| = 1.717$ ($\lambda = 0.633$ μm), $c_v = 0.143$, $l_0 = 45$ μm, and $\varepsilon_y = \varepsilon_z = 1.0$.

Effective size a_{ef} (relation (39)) was assumed to be equal to the mean value of semiaxis a: $a_{ef} = \langle a_0 \rangle = 2$ μm at $p = 1$ and $a_{ef} = \langle a_0 \rangle / \sqrt{2} \approx 1.4$ μm at $p = 2$. The local director distributions in drops were determined by solving the problem of minimizing the volume free-energy density [12, 15].

The experimental and calculated values of the $T_\perp$ and $T_\|$ components of unstrained PELC film at $\Delta l/l_0 = 0$ and $p = 1$ (Figs. 3, 4, respectively) are equal. The reason is that the attenuation coefficient of the layer containing initially spherical LC drops with a radial structure is independent of the polarization of the incident light. In the case of an unpolarized incident light beam, the transmitted light remains unpolarized (the degree of polarization P is zero). For an elongation factor $\Delta l/l_0 = 1$, the degree of polarization P reaches a value of 0.96 (Fig. 3); i.e., the forward-transmitted light is strongly polarized. This effect is due to the formation of defect-free homogeneous structures of drops in the presence of surfactant. The directed transmittance coefficient T_c^{tr} of the layer in the case of unpolarized incident light at $\Delta l/l_0 = 1$ is 0.35.

A comparison of the experimental data (Fig. 3) and calculation results (Fig. 4) revealed the correspondence between the theory and experiment to be the best at the polymer matrix refractive index $n_p = 1.51$. The experimental and theoretical $T_\perp$ and $T_\|$ values for $n_p = 1.51$ are listed in the table. One can see that they are in good correspondence. The difference is maximum for coherent transmittance $T_\|$; it does not exceed 12.2%.

The refractive index of the polymer matrix, $n_p = 1.51$, significantly differs from the ordinary LC refractive index, $n_\perp = 1.53$. The circumstance explains the fact that the measured coherent transmittance of the film illuminated by unpolarized light, $T_c^{tr} = 0.35$, does not reach the limiting value of 0.5 [2].

The calculated transmittances and degrees of polarization of forward-transmitted light are shown in Figs. 5–7. The calculations were based on relations (17)–(19) and (40)–(43) for films with monodisperse LC drops. The layer thickness was $l_0 = 45$ μm before extension ($p = 1$) and $l = 32$ μm after the twofold extension ($p = 2$).

Figure 5 shows the dependence of coherent transmittance T_c^{tr} of the initial layer (unstrained, $p = 1$) of spherical drops with randomly oriented optical axes on drop radius a_0 at different refractive indices n_p of the polymer matrix. Degree of polarization P of the light transmitted through this layer is zero. Figure 6 presents the dependence of layer transmittance T_c^{tr} and degree of polarization P of the forward-transmitted light on the length of the semiaxis transverse to the extension direction, $a = a_0/\sqrt{2}$, at twofold extension ($p = 2$) under conditions wherein the surfactant redistribution over the drop surface leads to the formation of single-domain internal structures.

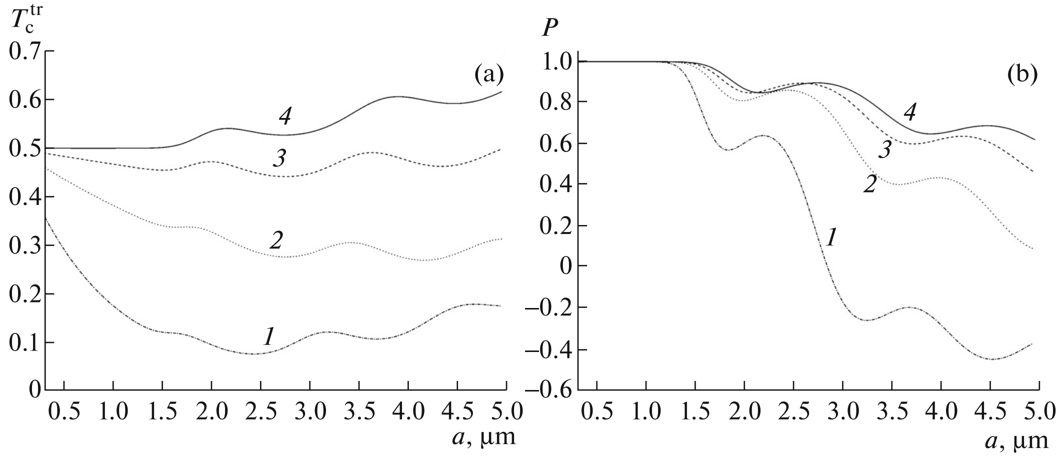

Fig. 6. Dependences of the (a) coherent transmittance T_c^{tr} of a PELC layer with modified interfacial anchoring and (b) degree of polarization P of forward-transmitted light on the transverse semiaxis a of drops at different values of refractive index n_p of the polymer matrix: n_p = (*1*) 1.49, (*2*) 1.51, (*3*) 1.52, and (*4*) 1.53. The layer-extension ratio is $p = 2$. Single-domain structure of LC drops; $n_\perp = 1.53$, $n_\parallel = 1.717$ ($\lambda = 0.633$ μm), $c_v = 0.143$, $l = 32$ μm, $\varepsilon_y = 2.83$, and $\varepsilon_z = 1.0$.

Figure 7 shows dependences $T_c^{tr}(a)$ and $P(a)$ at $p = 2$ under conditions of constant homeotropic interfacial anchoring without a surfactant.

It can be seen in Fig. 6 that, at $n_p = n_\perp$, there is an interval of drop sizes $a \in [0.3–1.15$ μm] (which corresponds to the interval $a_0 \in [0.42–1.63$ μm]), where the characteristics of the PELC film with modified interfacial anchoring reach limiting values: $T_c^{tr} = 0.5$ and $P = 1$. It follows from Fig. 7 that, in the same interval, at $n_p = n_\perp$, transmittance T_c^{tr} of the film with uniform normal interfacial anchoring is much less than 0.5 and the absolute value of the degree of polarization P does not exceed 0.47.

One can see in Figs. 6 and 7 that an increase in coherent transmittance T_c^{tr} above 0.5 for films with modified and uniform interfacial anchoring leads to a significant decrease in the absolute value of degree of polarization of light P. Note that degree of polarization P is negative (Figs. 6b, 7b) at $T_\parallel > T_\perp$ (see relation (19)).

It follows from the performed analysis that the main parameters determining the limiting values of the degree of polarization of light ($P = \pm 1$) and the transmittance of the PELC film ($T_c^{tr} = 0.5$) under extension are as follows: (i) refractive index of the polymer matrix n_p, which should be equal to LC ordinary refractive index $n_\perp$ (or extraordinary refractive index $n_\parallel$), and (ii) transverse size a of LC drops. The limiting values $T_c^{tr} = 0.5$ and $P = \pm 1$ depend on incident-light wavelength λ.

Figures 8 and 9 present spectral dependences $T_c^{tr}(\lambda)$ and $P(\lambda)$ for a deformed PELC layer of monodisperse 5CB LC drops with an oriented structure of optical axes (Fig. 8) and a layer of polydisperse drops with allowance for the misorientation of their optical axes (Fig. 9). The drops have a single-domain internal structure.

To take into account the dependence of the LC ordinary $n_\perp$ and extraordinary $n_\parallel$ refractive indices on incident-light wavelength λ in the calculations, we used the Cauchy formula:

$$n_{\perp,\parallel} = A_{\perp,\parallel} + \frac{B_{\perp,\parallel}}{\lambda^2} + \frac{C_{\perp,\parallel}}{\lambda^4}. \quad (44)$$

Experimental and calculated values of coherent transmittances $T_\perp$ and $T_\parallel$ of a PELC film for incident light polarized, respectively, perpendicular and parallel to the extension axis. The extension leads to interface modification in the inverse mode: LC drops with initially radial structure become single-domain

Extension ratio p	$T_\perp$ (experiment)	$T_\perp$ (theory)	$T_\parallel$ (experiment)	$T_\parallel$ (theory)
1.0	0.0178	0.0194	0.0178	0.0194
2.0	0.7	0.687	0.0041	0.0036

$n_\perp = 1.53$, $n_\parallel = 1.717$ ($\lambda = 0.633$ μm), $n_p = 1.51$, $c_v = 0.143$. Before extension ($p = 1$): $S_{2f} = 0$, $l_0 = 45$ μm, $a_{ef} = 2$ μm, and $\varepsilon_y = \varepsilon_z = 1$. Under twofold extension ($p = 2$): $S_{2f} = 1$, $l = 32$ μm, $\varepsilon_y = 2.83$, $\varepsilon_z = 1.0$, and $a_{ef} = 1.4$ μm.

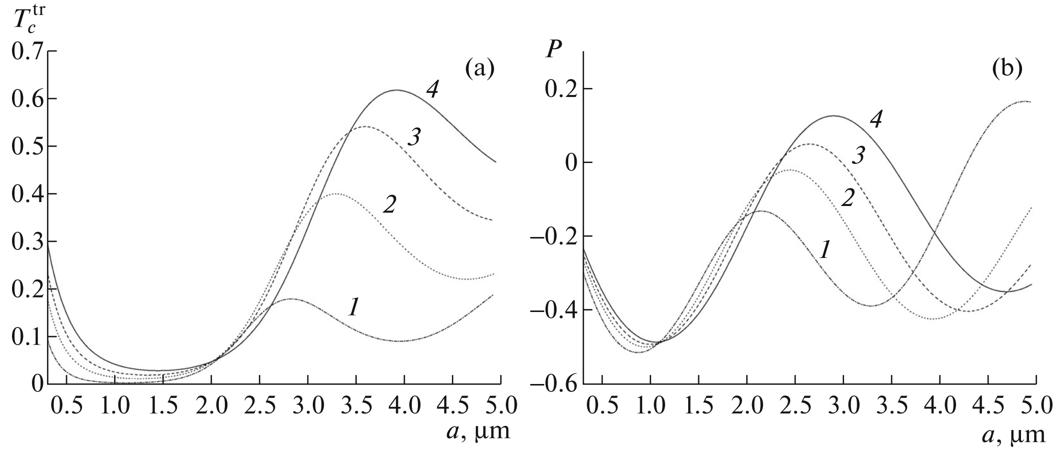

Fig. 7. Dependences of (a) coherent transmittance T_c^{tr} of a PELC layer with uniform homeotropic interfacial anchoring and (b) degree of polarization P of forward-transmitted light on the transverse semiaxis a of drops at different values of refractive index n_p of the polymer matrix: n_p = (*1*) 1.49, (*2*) 1.51, (*3*) 1.52, and (*4*) 1.53. The layer-extension ratio is $p = 2$. Radial structure of LC drops; $n_\perp = 1.53$, $n_\parallel = 1.717$ ($\lambda = 0.633$ μm), $c_v = 0.143$, $l = 32$ μm, $\varepsilon_y = 2.83$, and $\varepsilon_z = 1.0$.

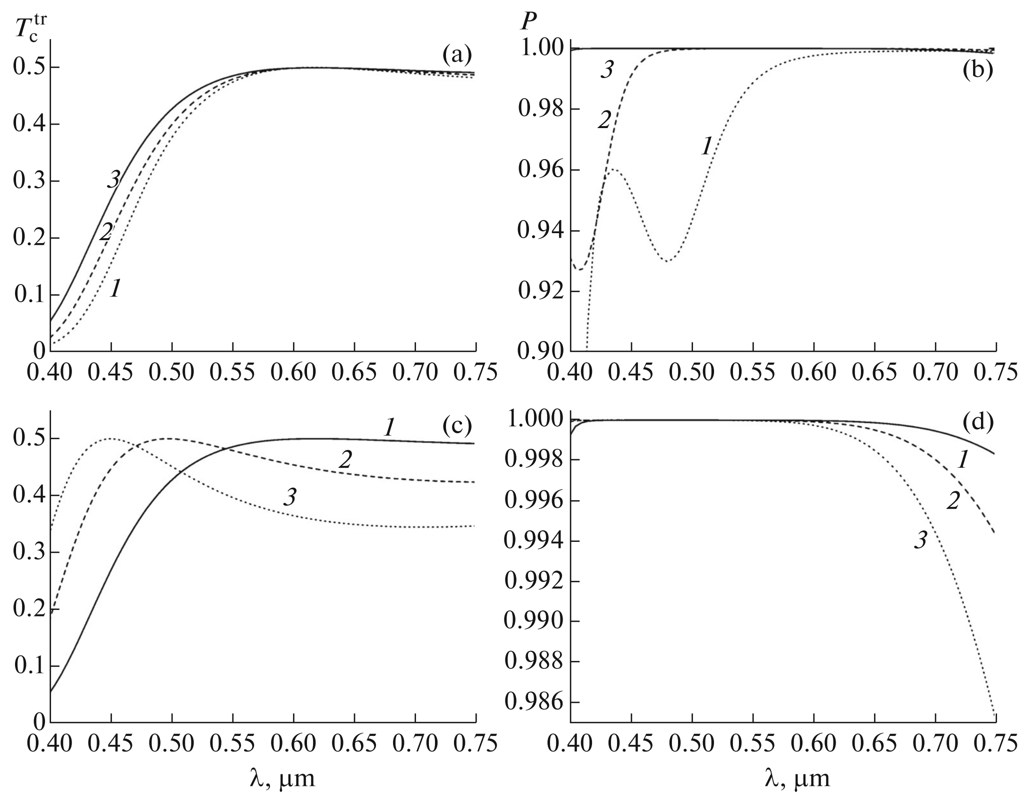

Fig. 8. Spectral dependences of (a, c) coherent transmittance $T_c^{tr}(\lambda)$ and (b, d) degree of polarization $P(\lambda)$ of forward-transmitted light at (a, b) different lengths of the drop transverse semiaxis a and (c, d) different refractive indices n_p of the polymer matrix; $p = 2$, $l = 32$ μm, $c_v = 0.143$, $\varepsilon_y = 2.83$, and $\varepsilon_z = 1.0$. Monodisperse drops ($D_a/\langle a\rangle = 0$) with a single-domain structure of LC 5CB and oriented optical axes (film order parameter $S_{2f} = 1$). (a, b): $n_p = n_\perp = 1.533$ ($\lambda = 0.62$ μm), a = (*1*) 1.4, (*2*) 1.0, and (*3*) 0.7 μm. (c, d): $a = 0.7$ μm, $n_p = n_\perp$ = (*1*) 1.533 ($\lambda = 0.62$ μm), (*2*) 1.548 ($\lambda = 0.5$ μm), and (*3*) 1.559 ($\lambda = 0.45$ μm).

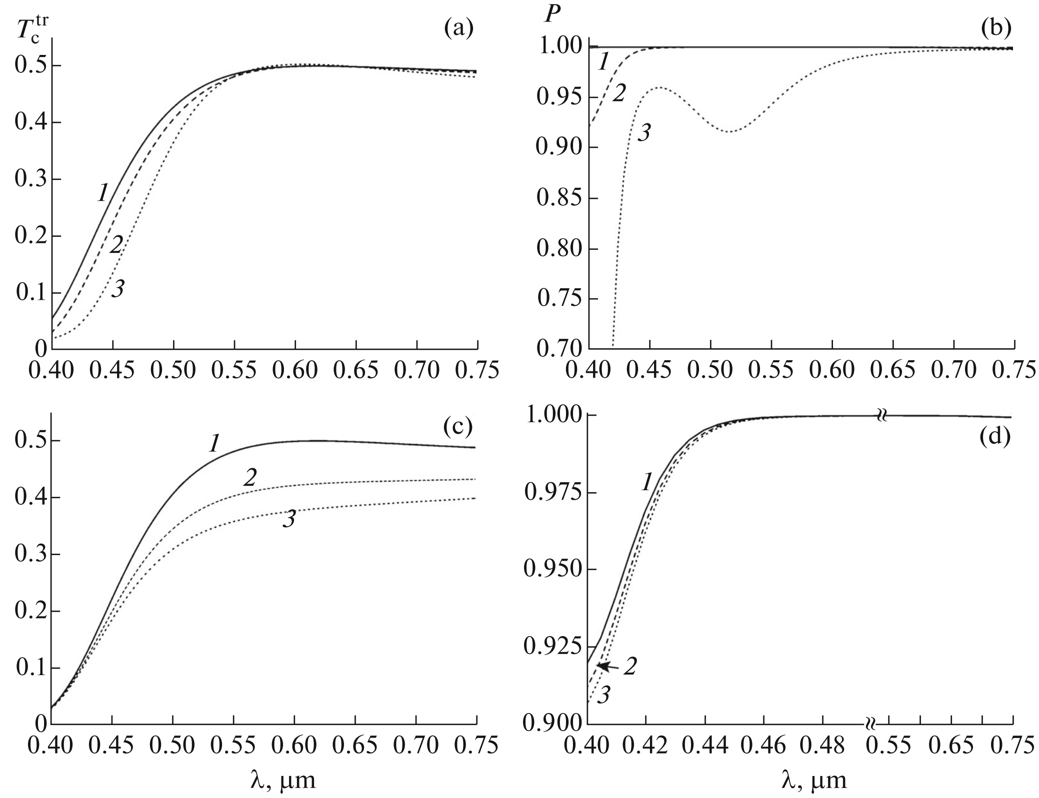

Fig. 9. Spectral dependences of (a, c) coherent transmittance $T_c^{tr}(\lambda)$ of PELC film and (b, d) degree of polarization $P(\lambda)$ of forward-transmitted light at different values of coefficient of variation $D_a/\langle a\rangle$ of the distribution of drop transverse semiaxis a and film-order parameter S_{2f}; $n_p = n_\perp = 1.533$ ($\lambda = 0.62$ μm), $p = 2$, $l = 32$ μm, $\varepsilon_y = 2.83$, $\varepsilon_z = 1.0$, and $c_v = 0.143$. The drop semiaxis mean is $\langle a\rangle = 0.7$ μm. (a, b): $S_{2f} = 1$ and $D_a/\langle a\rangle$ = (*1*) 0, (*2*) 0.4, and (*3*) 0.8. (c, d): $D_a/\langle a\rangle = 0.4$ and S_{2f} = (*1*) 1, (*2*) 0.97, and (*3*) 0.95.

The values of coefficients $A_{\perp,\parallel}$, $B_{\perp,\parallel}$, and $C_{\perp,\parallel}$ for the LC 5CB under consideration were taken from [26].

The drop polydispersity was taken into account using a Γ distribution for the a semiaxis:

$$P(a) = \frac{\mu^{\mu+1}}{\Gamma(\mu+1)}\frac{a^{\mu}}{a_m^{\mu+1}}\exp(-\mu a/a_m), \qquad (45)$$

where μ is the distribution parameter, Γ is a γ function, and a_m is the modal (most likely) size of the a semiaxis. Modal size a_m and parameter μ are related to mean value $\langle a\rangle$ and coefficient of variation $D_a/\langle a\rangle$, where D_a is the standard (rms) deviation, as follows:

$$a_m = \frac{\mu}{\mu+1}\langle a\rangle, \qquad (46)$$

$$\mu = 1/(D_a/\langle a\rangle)^2 - 1. \qquad (47)$$

In this case, effective length a_{ef} of the a semiaxis (see expression (21)) is given by the relation

$$a_{ef} = \langle a\rangle\frac{\mu+3}{\mu+1}. \qquad (48)$$

Figures 8a and 8b illustrate the influence of transverse drop size a at polymer refractive index n_p = 1.533, which is equal to ordinary refractive index $n_\perp$ of the LC at an incident-light wavelength of 0.62 μm. Figures 8c and 8d show the influence of polymer refractive index n_p (provided that $n_p = n_\perp$ at different wavelengths (λ = 0.62, 0.5, or 0.45 μm)) at size a = 0.7 μm, for which degree of polarization P of forward-transmitted light (or the polarizing ability of the PELC film) reaches close-to-limiting values ($P \approx 1$) in the entire visible spectral range (0.4–0.75 μm) (Fig. 8b). Figures 8c and 8d demonstrate that the achievement of the limiting characteristics $T_c^{tr} = 0.5$ and $P = 1$ when passing from the red-spectral region (λ = 0.62 μm) to the blue region (λ = 0.45 μm) is accompanied by an increase in polymer-matrix refractive index n_p.

An increase in the degree of drop polydispersity (coefficient of variation $D_a/\langle a\rangle$) and a decrease in the order parameter S_{2f} of the film lead to a decrease in directed film transmittance T_c^{tr} (Figs. 9a, 9c) and a change in the spectral dependence of the degree of polarization P of forward-transmitted light (Fig. 9b). The decrease in the film transmittance (with respect to a level of 0.5) is most sensitive to film-order parameter S_{2f} (the deviation of the S_{2f} values from unity, corresponding to the complete orientation of drop optical axes under extension) (Figs. 9a, 9c). The decrease in degree of polarization P with respect to unity is most sensitive to the LC-drop polydispersity (the increase in coefficient of variation $D_a/\langle a\rangle$) (Figs. 9b, 9d).

CONCLUSIONS

An optico-mechanical model was developed to analyze the transmittance and degree of polarization of light transmitted in the forward direction through a uniaxially extended PELC layer containing polydisperse LC drops shaped as spheres or spheroids in the initial unstrained state. This model is based on the Foldy–Twersky and anomalous diffraction approximations. Relations for determining the sizes and anisometry parameters of LC drops in dependence on the layer-extension ratio were derived. The model describes the optical response of a PELC film under extension as a function of the sizes and anisometry parameters of LC drops, their concentration, internal structure, polydispersity, and orientation of optical axes. It allows one to determine the film parameters that are required to attain the limiting values of the degree of polarization of forward-transmitted light and the film coherent transmittance in dependence on the incident-light wavelength.

The model was verified by comparing the theoretical and experimental results obtained for the inverse modification of interfacial anchoring.

The modification of the surface anchoring on the polymer–LC interface under extension leads to a significant increase in the coherent (directed) transmittance and the polarizing ability of the film.

The results of this study can be used to fabricate polarizer films based on polymer-encapsulated LCs with ion-surfactant modification of interfacial anchoring. These films operate in the light-scattering mode, without absorption of incident light. They are characterized by high light stability, mechanical strength, high transmittance, and good polarizing ability.

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