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# Investigation of Transmittance and Small-Angle Light Scattering by Monolayer of Liquid Crystal Droplets with Modified Boundary Conditions

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*A numerical method for modelling transmittance and angular distribution of light scattered by a Polymer Dispersed Liquid Crystal (PDLC) film, containing droplets with inhomogeneous boundary conditions, is proposed. It is based on the anomalous diffraction approximation and an interference approximation. The internal structures of nematic LC droplets are calculated on the basis of the free energy minimization problem solution using the relaxation method. The results for a monolayer of spherical LC droplets with modified boundary conditions caused by the local increase of the concentration of surface-active ions at the polymer–LC interface are discussed.*

**Keywords** Light modulation; light scattering; polymer dispersed liquid crystal; optical transmittance

## 1. Introduction

Polymer dispersed liquid crystal (PDLC) films are subject of active research because they offer new opportunities for creating display systems and optical information processing [1–5]. It is possible to create multi-functional devices with controllable (tunable) parameters on the basis of these films, such as: intensity and phase modulators of light radiation, polarizers and polarization converters of light, lenses, filters, reflectors, flexible displays, etc. [6–14].

Recently a new method was proposed and implemented, for controlling the structure of LC droplets [15] in a polymer matrix by an electric field. It is based on a modification of the boundary conditions by ionic surfactants. This method creates heterogeneity of the boundary conditions and reduces the value of switching voltage of the electric field in comparison with ordinary PDLC films.

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In this paper we consider a model to describe coherent transmission and the intensity of light scattered by monolayers of oriented spherical and spheroidal droplets, with modified boundary conditions. The amplitude-phase screen model and the interference approximation of multiple wave scattering [9,12,16] are used as the basis for light propagation consideration via such a layer. The anomalous diffraction approximation to describe the scattering of radiation by individual ellipsoidal droplets with non-uniform surface adhesion has been developed. Numerical analysis of coherent transmission coefficients and the scattered light intensities were performed for a monolayer of spherical droplets of nematic LCs with uniform [1–3] and non-uniform [17–19] boundary conditions at the interface. The internal structure of LC droplets was calculated by the relaxation method [20].

## 2. Basic Relations

Consider a monolayer of oriented non-absorbing LC droplets of ellipsoidal shape, dispersed in a non-absorbing polymer matrix. Choose a laboratory coordinate system (*x, y, z*), where the x axis coincides with the normal to the PDLC monolayer. The centers of the droplets are in the (y, *z*) plane. The layer is illuminated by a linearly polarized normally incident plane wave. The long axes of the LC droplets in the layer are oriented along the axis *y*. We also assume that the optical axes (directors) of droplets coincide with their long axes in the absence of an external electric field.

We consider components of the scattered wave with parallel (*vv*-component) and perpendicular (*vh*-component) polarization, with respect to the plane of polarization of the incident wave. The *vv*- and *vh*-components correspond to the geometries of parallel and crossed polarizer and analyzer. Using a model of amplitude-phase screen [9,12], coherent transmittance for *vv*- ($T_c^{vv}$) and *vh*- ($T_c^{vh}$) components can be written as follows:

$$T_c^{vv} = \left|T_a^{vv}\right|^2 = 1 - \frac{4\pi}{k^2\sigma}\eta \mathrm{Re} f_{vv}^0 + \frac{4\pi^2}{k^4\sigma^2}\eta^2 \left|f_{vv}^0\right|^2, \tag{1}$$

$$T_c^{vh} = \left|T_a^{vh}\right|^2 = \frac{4\pi^2}{k^4\sigma^2}\eta^2 \left|f_{vh}^0\right|^2. \tag{2}$$

Here, $\eta$ is the filling factor of the PDLC monolayer, which is numerically equal to the ratio of the section of all LC droplets to the area of distribution, $k = 2\pi n_p / \lambda$ is the wave vector, $n_p$ is the refractive index of the polymer matrix, $\lambda$ is the wavelength of incident light, $f_{vv,vh}^0$ are the *vv*- and *vh*- components of the vector amplitude of the scattering function [21] in the forward direction ($\theta_s = 0$), and $\sigma$ is the cross section of a droplet.

A coherent transmission coefficient is defined, for PDLC monolayer under normal illumination by linearly polarized light, in the absence of polarization devices, which represents the sum of equations (1) and (2). We write this expression as follows:

$$T_c^p = 1 - Q_p\eta + \frac{Q_p^2 L_T}{2}\eta^2. \tag{3}$$

Here $Q_p$ is the extinction efficiency factor of linearly polarized light of a single droplet:

$$Q_p = \frac{4\pi}{k^2\sigma}\mathrm{Re} f_{vv}^0, \tag{4}$$

$$L_T = \frac{1}{2}\left(1 + \frac{\mathrm{Im}^2 f_{vv}^0}{\mathrm{Re}^2 f_{vv}^0}\right)\left(1 + \frac{\left|f_{vh}^0\right|^2}{\left|f_{vv}^0\right|^2}\right). \tag{5}$$

We consider the angular structure of radiation scattered by monolayer of spherical or spheroidal droplets. The interference approximation is used for the description of light scattering by an ensemble of single-row scatterers [9,12,16]. It takes into account the interference of single scattered light in the far field. For the parallel ($I_{vv}$) and perpendicular ($I_{vh}$) components of polarization of the incident radiation intensity of incoherently (diffusely) scattered light, we obtain:

$$I_{vv}(\theta_s, \varphi_s) = C\frac{\eta}{\sigma k^2}\,|f_{vv}(\theta_s, \varphi_s)|^2\, S(\theta_s), \tag{6}$$

$$I_{vh}(\theta_s, \varphi_s) = C\frac{\eta}{\sigma k^2}\,|f_{vh}(\theta_s, \varphi_s)|^2\, S(\theta_s). \tag{7}$$

Here $C = AE_i^2/R^2$ is the normalization constant, $A$ is the area of the test section of the sample, $E_i$ is the incident wave amplitude; $R$ is the distance from the origin to the point of observation, $f_{vv}(\theta_s, \varphi_s)$ and $f_{vh}(\theta_s, \varphi_s)$ are the $vv$- and $vh$-components of the vector amplitude of the scattering function in the direction of the wave vector of the scattered wave $\mathbf{k}_s = (k\cos\theta_s, k\sin\theta_s\cos\varphi_s, k\sin\theta_s\sin\varphi_s)$, $S(\theta_s)$ is the structure factor of the monolayer [16], and $\theta_s$ and $\varphi_s$ are the polar and azimuthal scattering angles. For a statistically isotropic ensemble of droplets in the form of spheres or oblate spheroids, it does not depend on the azimuthal angle $\varphi_s$. At low concentration of LC droplets, single scattering of light is implemented. In this case the structure factor $S(\theta_s) \approx 1$ for all values of the polar scattering angle. The difference of the structure factor from unity determines the degree of influence of interference effects in angular structure of scattered light.

The components of the amplitude function in the expressions (6) and (7) are defined in terms of the amplitude scattering matrix elements $S_j$ ($j = 1,2,3,4$), as follows:

$$\begin{aligned} f_{vv}(\theta_s, \varphi_s) = {} & S_2(\theta_s, \varphi_s)\cos^2(\alpha - \varphi_s) + S_1(\theta_s, \varphi_s)\sin^2(\alpha - \varphi_s) \\ & + \tfrac{1}{2}(S_3(\theta_s, \varphi_s) + S_4(\theta_s, \varphi_s))\sin 2(\alpha - \varphi_s), \end{aligned} \tag{8}$$

$$\begin{aligned} f_{vh}(\theta_s, \varphi_s) = {} & S_3(\theta_s, \varphi_s)\sin^2(\alpha - \varphi_s) - S_4(\theta_s, \varphi_s)\cos^2(\alpha - \varphi_s) \\ & + \tfrac{1}{2}(S_2(\theta_s, \varphi_s) - S_1(\theta_s, \varphi_s))\sin 2(\alpha - \varphi_s). \end{aligned} \tag{9}$$

For light scattered in the direction of the incident wave amplitudes, the off-diagonal elements of the scattering matrix are zero ($S_3 = S_4 = 0$). In this case, the components of the vector amplitude of the scattering function can be written as follows:

$$f_{vv}^0 = S_2^0\cos^2\alpha + S_1^0\sin^2\alpha, \tag{10}$$

$$f_{vh}^0 = \frac{1}{2}\left(S_2^0 - S_1^0\right)\sin 2\alpha. \tag{11}$$

Here $S_{2,1}^0$ are the diagonal elements of the amplitude scattering matrix at zero scattering angle ($\theta_s = 0$).

In the anomalous diffraction (AD) approximation, the scattered field in the far zone is determined by the diffraction at the amplitude-phase screen. The complex transmission matrix of the screen is determined by the projection of the droplet onto a plane orthogonal to the direction of the incident light [21]. When illuminated along the $x$ axis, using the known results [22] for the amplitude scattering matrix elements, we get:

$$S_1(\theta_s, \varphi_s) = \frac{k^2\sigma}{2\pi}\int\limits_{\sigma}(1 - T_1(\xi))\exp(-i\mathbf{k}_s\xi)\,d\xi, \tag{12}$$

$$S_2(\theta_s, \varphi_s) = \frac{k^2\sigma}{2\pi} \cos\theta_s \int_\sigma (1 - T_2(\xi)) \exp(-i\mathbf{k}_s\xi)\, d\xi, \tag{13}$$

$$S_3(\theta_s, \varphi_s) = -\frac{k^2\sigma}{2\pi} \cos\theta_s \int_\sigma T_3(\xi) \exp(-i\mathbf{k}_s\xi)\, d\xi, \tag{14}$$

$$S_4(\theta_s, \varphi_s) = -\frac{k^2\sigma}{2\pi} \int_\sigma T_4(\xi) \exp(-i\mathbf{k}_s\xi)\, d\xi. \tag{15}$$

Here $\xi$ is the radius vector of the cross section $\sigma$, and $T_j\ (\xi)$ are the elements of the $2 \times 2$ transmittance matrix $\underline{\underline{T}}$ of the equivalent amplitude-phase screen ($j$ = 1,2,3,4). It depends on the internal structure of the LC droplet orientation [23].

Equations (12)–(15) allow calculation of the amplitude matrix elements of the scattering at homogeneous and inhomogeneous boundary conditions on the surface of the droplet, based on the configuration of the director (the distribution of local optical axis) in the droplet. The configuration of the director in the droplet with given boundary conditions [17,18,20] was found by solving the problem of minimizing the bulk free energy density [24].

## 3. Results and Discussion

The director configuration of the LC droplet is calculated in a Cartesian coordinate system by finding the distribution of directors of the elementary volumes of LC in the droplet using the relaxation method of the bulk free energy density minimization problem solution [20,24]. Textures of the considered LC droplets with inhomogeneous boundary conditions obtained using crossed polarizers in the absence of an applied field are displayed in Fig. 1.

To characterize the fraction of surface droplet with normal and tangential boundary conditions we introduce the parameter W. The value of W is determined by the ratio of the height of segment surface of the droplet with normal boundary conditions, to droplet diameter. The parameter values W = 0% and W = 100% correspond to a fully homogeneous tangential and normal surface adhesion at the interface LC-polymer, respectively. When the parameter W = 0%, there is a cylindrically symmetric bipolar configuration of the

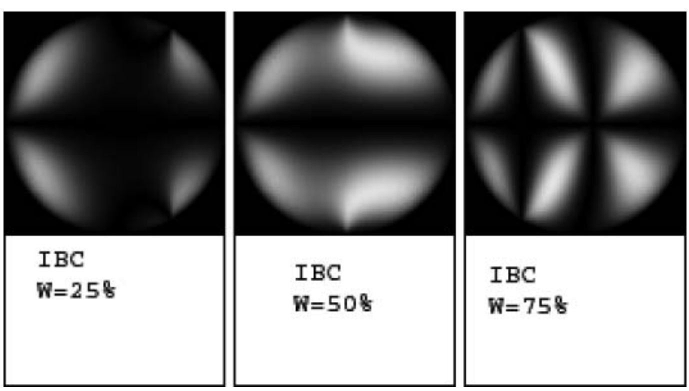


**Figure 1.** Textures of LC droplets with inhomogeneous boundary conditions (IBC), obtained using crossed polarizers at different values of the parameter W, in the absence of an applied electrical field.

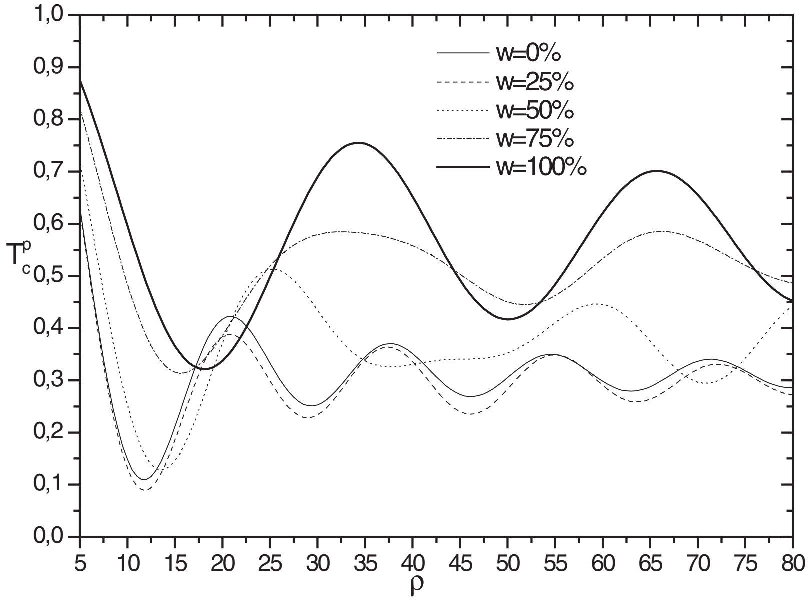


**Figure 2.** Coherent transmission coefficient $T_c^p$ of a monolayer of spherical LC droplets on the diffraction parameter $\rho$. The polarization angle is $\alpha = 0$, the refractive indices of the LC are $n_o = 1.531$, $n_e = 1.717$, the wavelength of incident light is $\lambda = 0.633$ $\mu$m, the refractive index of the polymer is $n_p = 1.53$, and the filling factor of the monolayer is $\eta = 0.5$.

LC droplet. At W = 100%, the internal structure of the LC droplet orientation is radially symmetric.

The dependence of the coherent light transmission $T_c^p$ for the layer consisting of spherical droplets of LC, on the diffraction parameter $\rho = ka$, where $a$ is the radius of the droplets, is shown in Fig. 2. The calculations are performed in the absence of an external electric field, and the polarization angle of the incident light is $\alpha = 0$. The refractive indices of the LC are $n_o = 1.531$, $n_e = 1.717$, the wavelength of incident light is $\lambda = 0.633$ $\mu$m, the refractive index of the polymer is $n_p = 1.53$, and the monolayer filling factor is $\eta = 0.5$. The internal structure of the LC droplet orientation was calculated in a Cartesian coordinate system by finding a local vector field with director **n**, using the relaxation method for solving the problem of minimizing the bulk free energy density [20].

The results displayed in Fig. 2 show that when illuminated with linearly polarized light with polarization angle $\alpha = 0$, the monolayer with radial internal structure of droplets in a layer (W = 100%) has large values of the coefficient of the coherent transmittance in the entire range of considered values of diffraction parameter $\rho$ (from 5 to 80), compared to the bipolar configuration of LC droplets (W = 0%). When illuminated by a monolayer of linearly polarized light with polarization angle $\alpha = \pi/2$, the situation is reversed: the values of $T_c^p(\rho)$ for layer with tangential boundary conditions (and bipolar configuration of the LC droplets) are greater than the $T_c^p(\rho)$ values for the layer with normal boundary conditions (and radial configuration of the internal droplet structure).

Data from the $I_{vv}$ component of the scattered light intensity for a monolayer of spherical LC droplets at $\alpha = 0$, as function of the polar scattering angle $\theta_s$, are shown in Figs. 3 and 4. The calculations are presented in relative units at a value of the normalization constant

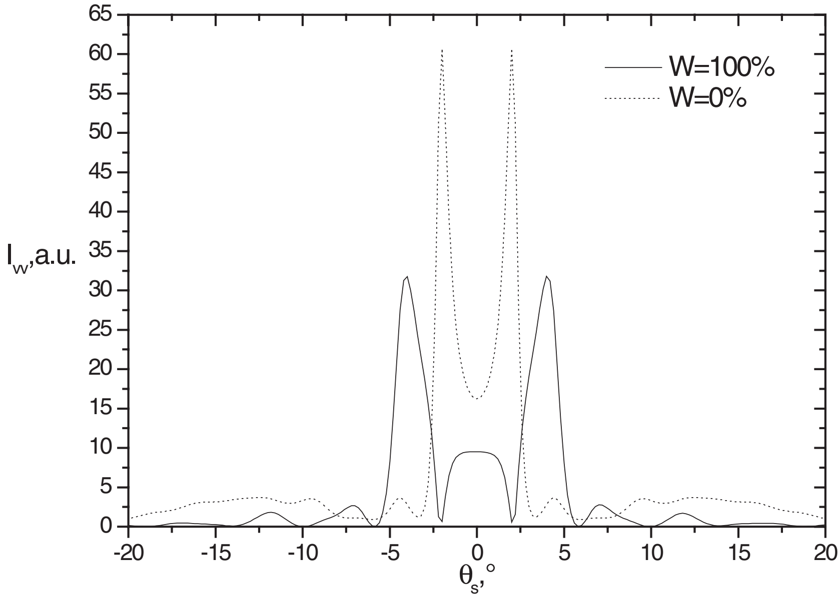


**Figure 3.** Dependence of the $I_{vv}$-components of the light intensity, scattered by a monolayer of spherical LC droplets with homogeneous tangential (W = 0%) and normal (W = 100%) boundary conditions, as function of the polar scattering angle $\theta_s$, with a value of the azimuthal scattering angle $\varphi_s = 0$. The other parameters are the same as in Fig. 2.

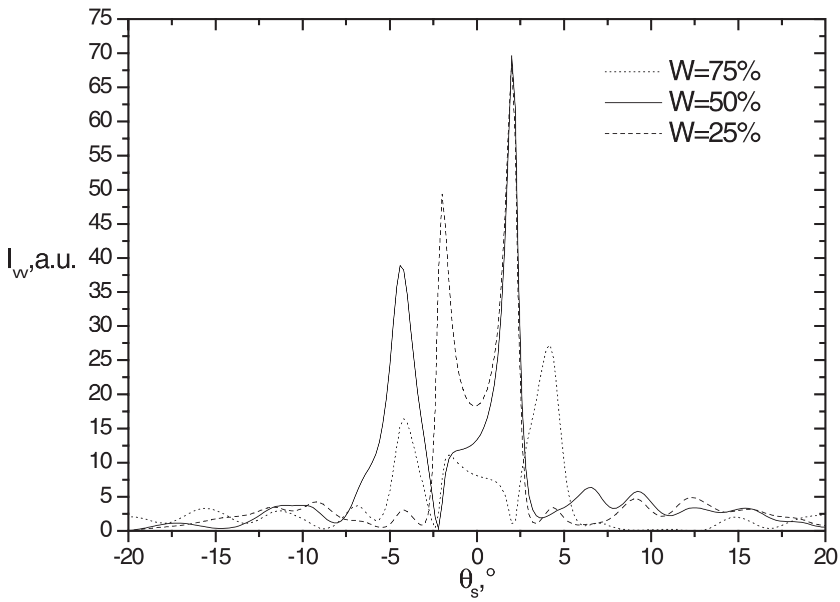


**Figure 4.** The same as in Fig. 3 for inhomogeneous boundary conditions.

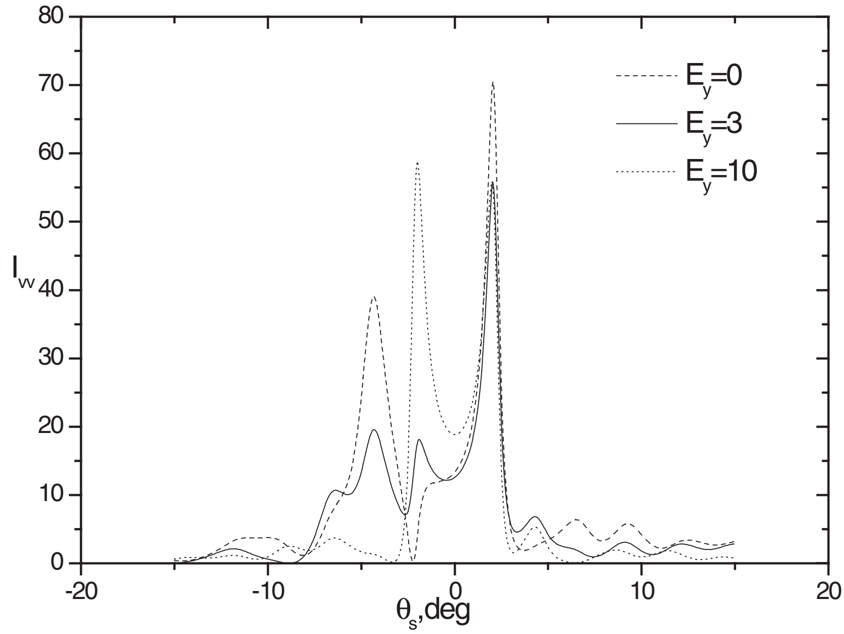


**Figure 5.** The same as in Fig. 4 for inhomogeneous boundary conditions at different values of the normalized control field $E_y$, applied to the layer plane along the optical axes of the droplets.

$C = 1$. They are performed for the droplet radius $a = 5\mu$m and the refractive index of the polymer matrix $n_p = 1.532$. The values of the other parameters are the same as for the analysis of the coherent transmission coefficient.

Fig. 3 shows the dependence of $I_{vv}$ components of the scattered light on the polar scattering angle $\theta_s$, for homogeneous boundary conditions at W = 0% and W = 100%. The data are calculated in the absence of an electrical field. The scattering plane coincides with the principal plane (*y, x*). The value of the azimuthal scattering angle is $\varphi_s = 0$, and the light incident angle is zero.

The results for intensity of scattered light $I_{vv}(\theta_s)$ in the layers containing droplets with inhomogeneous boundary conditions are presented in Fig. 4. When we deal with homogeneous boundary conditions, the values of $I_{vv}(\theta_s)$ are identical for the same deviations of the polar scattering angle $\theta_s$ in different directions, relative to the normal of the layer ($I_{vv}(\theta_s) = I_{vv}(-\theta_s)$). When we consider inhomogeneous boundary conditions, such as "tangential-normal", one observes an asymmetry of the angular structure of scattered light over the polar angle ($I_{vv}(\theta_s) \neq I_{vv}(-\theta_s)$). For the considered droplet and film parameters, this effect it is most pronounced when $w = 50\%$, i.e. with an equal share of the tangential and normal boundary conditions on the droplet surface.

Analysis of the *vh*-component of the scattered light intensity reveals that this component is also showing asymmetry of the light scattering with respect to the polar angle, but it is weaker in comparison with the *vv*-component.

The influence of the electric field (normalized to the threshold value of the electric field), applied in-plane parallel to the optical axes of the droplets, on the angular structure of light scattering at W = 50%, and at $\alpha = 0$, is shown in Fig. 5. The asymmetry of the angular structure of the scattering due to the inhomogeneity of the boundary conditions (at the considered droplet and film parameters) has a pronounced effect up to the values of the normalized control field $E_y$ equal to 3, which corresponds to the output of the electro-optical response at saturation [19]. In the strong applied field the uniformly oriented internal structure of LC droplets is formed. In this case the angular structure of transmitted light [16] is symmetric.

## 4. Conclusions

We propose a numerical method to analyze coherent transmission and small-angle light scattering by a PDLC monolayer with inhomogeneous boundary conditions on the droplet surface. In the description of coherent light transmittance of PDLC monolayers, consisting of LC droplets of spherical and oblate ellipsoids, we used the anomalous diffraction approximation and the amplitude-phase screen model. To study the angular structure of scattered light of monolayers, the interference approximation of the theory of multiple scattering of waves was used. The results of calculations of the coefficients of coherent transmission and intensity of the scattered radiation are displayed for droplets with homogeneous and inhomogeneous adhesion of LC molecules on the LC-polymer interface.

For PDLC films with inhomogeneous interphase boundary conditions, we revealed an asymmetry of the small-angle structure of scattering light, with respect to the polar angle.

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