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Vladimir P. Dick; Valery A. Loiko

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Model for coherent transmittance calculation for polymer dispersed liquid crystal films

VLADIMIR P. DICK and VALERY A. LOIKO*

Institute of Physics, Academy of Sciences of Belarus, F. Scaryna Avenue 68, Minsk 220072, Belarus

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This paper analyses the methods of calculating the coherent (direct) transmittance of polymer dispersed liquid crystal (PDLC) films which are polymer-based films with embedded liquid crystal droplets. By comparison with experimental data it is shown that at a high concentration of LC droplets, Beer's law, which is frequently used, leads to large errors in calculations of the transmittance of PDLC films. To calculate coherent transmittance, it is expedient to use the interference approximation which takes into account the interference of waves scattered by individual LC droplets and leads to much more accurate results.

1. Introduction

Layers of polymer dispersed liquid crystals (PDLC films) represent one of the classes of LC materials which is being actively investigated today [1–3]. The interest in PDLC films is because they possess a variety of practically important properties which follow from their physical peculiarities. For instance, PDLC films possess great mechanical strength and can be relatively easily made in the form of large size screens. They can also be made in the form of flexible films and applied to surfaces of arbitrary form. Additionally, PDLC films make it possible to do without polarizers, which simplifies device construction and increases the brightness of the image transferred by them.

PDLC film is a polymer-based film with embedded liquid crystal droplets of sizes ranging, as a rule, from fractions to units of microns. Because of the difference in refractive indices between the polymer and the LC such a film is light scattering: when light is incident on it, the light is partially scattered by the film and only part of the incident light passes through the film retaining its original properties. The intensity ratio of the light scattered by the PDLC film to that passing through it depends on the orientation of the molecules in the LC droplets. Therefore, a change in molecular orientation in the LC droplets, e.g. by applying an external electric field, permits control of the intensity of light passed through and scattered by the film.

To develop a device on the basis of PDLC films for optical image transfer and optical light modulation, it is necessary to know the dependence of the optical parameters of the PDLC film on its microstructure. One of the most important parameters of a PDLC film is the transmission coefficient T which characterizes the attenuation of the parallel light beam by the film and is given by the relation

$$T = I/I_0 \qquad (1)$$

where I_0 is the incident light intensity and I is the intensity of the attenuated incident light. In the radiation transfer theory [4, 5], T is termed the 'direct transmittance', and in the theory of multiple scattering it is referred to as the coherent transmittance [6, 7].

The dependence of the transmittance T of PDLC films on their structure has been the subject of a large number of publications, both experimental and theoretical; see, for example, [8–11] and references therein. In theoretical studies, the Bouguer law

$$T = \exp(-\varepsilon l) \qquad (2)$$

is used for calculating T. Here ε is the light extinction coefficient in the PDLC film, l is the path length of the non-scattered light in the film; ε is often calculated in accordance with the relation

$$\varepsilon = n\sigma_{\text{ext}} \qquad (3)$$

which is known as Beer's law, where n is the number of droplets in the unit volume and σ_{ext} is the extinction cross section of an individual droplet [4, 5]. However, the use of Beer's law in investigating PDLC films is not correct. Indeed, the peculiarity of PDLC films is the relatively high concentration of LC droplets in the films.

*Author for correspondence; e-mail: loiko@dragon.bas-net.by

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As a rule, the volume concentration of LC droplets, c_v, (the portion of the film volume occupied by LC droplets) amounts to a few tens per cent. In the theory of scattering, such media are identified as densely packed and, they have been actively investigated over the last few decades [7, 12]. These investigations have shown that during the interaction of light with densely packed media, a number of specific peculiarities show up. In particular, it has been found [13–16] that Beer's law (3) only holds at small concentrations of inhomogeneities; c_v not exceeding some value c_v^{max} depending on the parameters of the particles and not exceeding a few percent. At large c_v a calculation by equation (3) leads to either underestimated or overestimated values of the extinction coefficient ε.

The calculation of the extinction coefficient at large c_v requires, strictly speaking, a solution of the problem of light diffraction by a system of many particles. No exact solution of this problem has been found. Therefore, at present various approximate solutions are being used in calculating ε for media with a high concentration of particles. The simplest of these that can be used in the case of weakly scattering particles is the so-called interference approximation (ITA) [17–20]. In accordance with this approximation, for a medium consisting of identical spherical particles

$$\varepsilon = n\sigma_{\text{ext}} - n\sigma_{\text{sca}} \int_{4\pi} [1 - S(|\hat{\mathbf{e}}_s - \hat{\mathbf{e}}_i|)]p(\hat{\mathbf{e}}_s, \hat{\mathbf{e}}_i)\, d\Omega \quad (4)$$

where $\hat{\mathbf{e}}_i$ is the unit vector in the direction of the incident light propagation, $\hat{\mathbf{e}}_s$ is the unit vector in the direction of the scattered light propagation, and $S(|\hat{\mathbf{e}}_s - \hat{\mathbf{e}}_i|)$ is the structure factor [21]; $p(\hat{\mathbf{e}}_s, \hat{\mathbf{e}}_i)$, σ_{ext} and σ_{sca} are, respectively, the single-particle phase function ($\int_{4\pi} p(\mathbf{e}_s, \hat{\mathbf{e}}_i)\, d\Omega = 1$), the extinction cross section and the scattering cross section of a single particle [4, 5] and $d\Omega$ is the element of the solid angle.

The ITA has a simple physical meaning. Indeed, the characteristic feature of densely packed dispersion media is the correlation in the disposition of the individual particles. Correlation leads to the appearance of the effects from interference of waves scattered by individual particles. Due to this interference, the total amount of the scattered light decreases and, accordingly, the coherent transmittance increases [12, 20]. The interference phenomena for a medium consisting of identical spherical particles are taken into account easily enough by replacing the phase function of an individual particle $p(\hat{\mathbf{e}}_s, \hat{\mathbf{e}}_i)$ with the phase function $S(|\hat{\mathbf{e}}_s - \hat{\mathbf{e}}_i|)p(\hat{\mathbf{e}}_s, \hat{\mathbf{e}}_i)$, where the structure factor $S(|\hat{\mathbf{e}}_s - \hat{\mathbf{e}}_i|)$ depends on the particle concentration and the pattern of their spatial arrangement. In a polydisperse system one has to use partial structure factors [21].

We note that the possibility of using equations (3) and (4) for media with a high concentration of particles has only been explored for cases where the medium is formed from homogeneous and isotropic particles [13–16, 20]. The object of the present work is to analyse the possibility of using these equations for media with a high concentration of anisotropic LC droplets. We will compare the results of the calculations by equations (3) and (4) with experimental data and show that Beer's law is not suitable for calculating the light transmittance by PDLC films with a high concentration of LC droplets. In those cases where the scattering cross section of an individual particle σ_{sca} is not too large, it is expedient to use ITA for calculating ε.

2. Method

The assessment of the applicability of Beer's law and ITA was carried out by comparing the results of the calculation of PDLC film transmittance by equations (2) and (3) and (2) and (4) with the experimental data presented in [22]. The latter were obtained by measuring the PDLC film transmittance for various angles of incidence, ϑ_i, of linearly polarized light from a He-Ne laser ($\lambda = 632.8$ nm). Measurements were made in two cases: where the polarization plane of the laser radiation was parallel or perpendicular to the plane of incidence. The sample investigated was a polymer film with thickness $d = 20$ μm placed between glass plates with transparent electrodes. In the polymer film there were LC droplets of radius $r = 1.2$ μm. Their concentration was $n = 7 \times 10^{16}\ \text{m}^{-1}$ (the volume concentration of LC in the film was $c_v = 4\pi r^3 n/3 = 0.507$). The LC material represented a composition with refractive indices $n_0 = 1.511$ (for the ordinary beam) and $n_e = 1.736$ (for the extraordinary beam). In the measurements, an electric voltage $V = 125$ V was applied to the film. Under this (far from threshold) voltage, the long axes of the LC molecules were oriented perpendicular to the film surface.

Figures 1 and 2 show the results of the coherent film transmittance measurements plotted against the angle of incidence of the light for various states of polarization. For convenience of comparison with the calculated data, the experimental results have been corrected by the value for Fresnel reflection from the PDLC film boundaries in accordance with the relation

$$T_{\text{PDLC}} = T_{\text{film}} / T_{\text{glass}} \quad (5)$$

where T_{film} is the measured PDLC film transmittance, and T_{glass} is the measured film transmittance in the absence of LC in the film, i.e. the film transmittance caused by the Fresnel reflection from its boundaries.

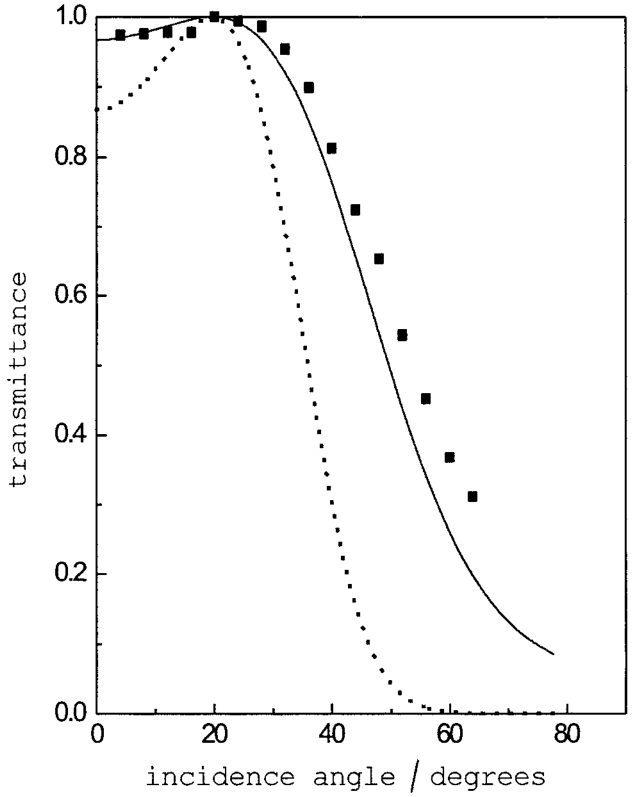

Figure 1. Transmittance of the PDLC film in a strong electric field versus the angle of incidence of light polarized in the plane of incidence. Filled squares: experimental data of Bloisi, Ruocchio, Terrecuso and Vicari (Ref. [22]); solid curve: results of the calculation by formulaes (2), (4); dashed curve: results of the calculation by formulaes (2), (3).

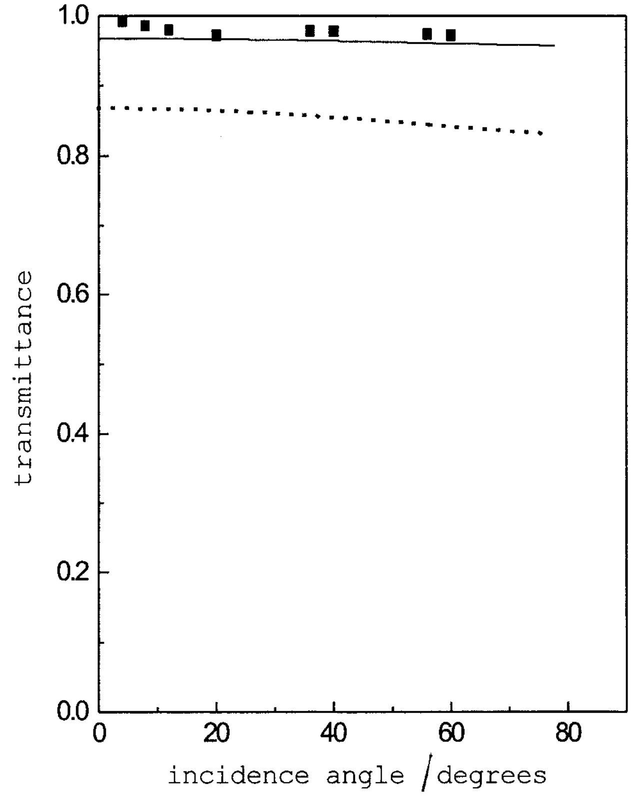

Figure 2. Same as Figure 1, but for light polarized perpendicular to the plane of incidence.

It should be noted that, as follows from figure 1, at the angle of incidence $\vartheta_i \approx 20°$ for light polarized parallel to the incidence plane, the transmission coefficient of the PDLC film $T_{PDLC} = 1$. This fact has been used by us to determine the refractive index of the surrounding matrix of the PDLC, n_p. Indeed, the condition $T_{PDLC} = 1$ points to the absence of scattering in the PDLC film and can only take place when the scattering cross section of the LC droplets, σ_{ext}, is equal to zero. For spherical LC droplets in a strong electric field, the condition $\sigma_{ext} = 0$ can only be realized at a refractive index of the polymer, n_p, determined by the relation [23]

$$n_p = \left(\frac{\cos^2 \vartheta_p}{n_0^2} + \frac{\sin^2 \vartheta_p}{n_e^2} \right)^{-1/2} \tag{6}$$

where ϑ_p is the angle between the direction of the LC droplet and the direction of propagation of the incident wave; we shall refer to this as the angle of incidence of light on the droplet. Since in the case of a film in a strong electric field the angle of incidence of light on the droplet, ϑ_p, is related to the angle of incidence of light on the film, ϑ_i, by the apparent relation

$$n_p \sin \vartheta_p = \sin \vartheta_i \tag{7}$$

we can write

$$n_p = n_0 \left[1 + \sin^2 \vartheta_i \left(\frac{1}{n_0^2} - \frac{1}{n_e^2} \right) \right]^{1/2} \tag{8}$$

At $\vartheta_i = 20°$, $n_0 = 1.511$ and $n_e = 1.736$, it follows from equation (8) that $n_p = 1.520$. We have used this value of n_p to calculate the PDLC film transmittance.

3. Peculiarities of the calculation

The PDLC film in question was formed from relatively large LC droplets (the diffraction parameters of the droplets $x = 2\pi r n_p / \lambda = 18.3$). In calculating the light scattering parameters of such droplets, the anomalous diffraction approach (ADA) is normally used. In accordance with this approach, the scattering matrix $\boldsymbol{S}$ of an LC droplet placed in a strong electric field is of the form [23]

$$\boldsymbol{S} = \begin{pmatrix} S_2(\theta) & 0 \\ 0 & S_1(\theta) \end{pmatrix} \tag{9}$$

where

$$S_1(\theta) = k^2 r^2 \int_0^{\pi/2} \{1 - \exp[\mathrm{i}2kr(n_0/n_\mathrm{p} - 1)\sin\tau]\} \times J_0(kr\sin\theta\cos\tau)\cos\tau\sin\tau\,\mathrm{d}\tau \quad (10)$$

$$S_2(\theta) = k^2 r^2 \int_0^{\pi/2} \{1 - \exp[\mathrm{i}2kr(n_\mathrm{e}(\vartheta_\mathrm{p})/n_\mathrm{p} - 1)\sin\tau]\} \times J_0(kr\sin\theta\cos\tau)\cos\tau\sin\tau\,\mathrm{d}\tau \quad (11)$$

and

$$n_\mathrm{e}(\vartheta_\mathrm{p}) = \left[\frac{\cos^2\vartheta_\mathrm{p}}{n_0^2} + \frac{\sin^2\vartheta_\mathrm{p}}{n_\mathrm{e}^2}\right]^{-1/2}. \quad (12)$$

Here θ is the scattering angle, $k = 2\pi n_\mathrm{p}/\lambda$ and J_0 is the zero order Bessel function. In the present paper, to calculate the light scattering parameters of individual particles, we used another method of calculation which has wider limits of applicability. This is the Mie solution which assumes that an individual spherical LC droplet in a strong electric field scatters like a homogeneous isotropic sphere of the same size. It is noteworthy that the refractive index of this isotropic sphere $n_\mathrm{eqv} = n_0/n_\mathrm{p}$ for light polarized perpendicular to the plane of incidence and $n_\mathrm{eqv} = n_\mathrm{e}(\vartheta_\mathrm{p})/n_\mathrm{p}$ for light polarized parallel to the plane of incidence.

The legitimacy of using the Mie solution for calculating the light scattering parameters of LC droplets in a strong electric field is supported by the following reasoning. For large spherical particles with a small relative refractive index, calculation by the Mie formulae leads to equations (9)–(12) if the function $S_1(\theta)$ is calculated at a relative refractive index $m = n_0/n_\mathrm{p}$ and the function $S_2(\theta)$ at $m = n_\mathrm{e}(\vartheta_\mathrm{p})/n_\mathrm{p}$ [24]. For small spherical particles with small m (in the Rayleigh–Gans approximation), the Mie solution leads to the scattering matrix (9) where [25]

$$S_1(\theta) = u(\theta)(m^2 - 1) \quad (13)$$

$$S_2(\theta) = u(\theta)(m^2 - 1)\cos\theta \quad (14)$$

with

$$u(\theta) = \frac{\mathrm{i}}{[2\sin(\theta/2)]^3}\{\sin[2kr\sin(\theta/2)] - [2kr\sin(\theta/2)]\cos[2kr\sin(\theta/2)]\}. \quad (15)$$

Equation (13) coincides with the expression for the amplitude function $S_1(\theta)$ in the Rayleigh–Gans approximation for an LC droplet in a strong electric field [26] if we assume that $m = n_0/n_\mathrm{p}$. Equation (14) coincides with the expression for the amplitude function $S_2(\theta)$ of an LC droplet, in the same approximation, if we assume that $m = n_\mathrm{e}(\vartheta_\mathrm{p})/n_\mathrm{p}$ and the angle of incidence of light on the particle $\vartheta_\mathrm{p} = 0°$ or $\vartheta_\mathrm{p} = 90°$. Accordingly, the Mie solution can be used for calculating the scattering parameters of large LC droplets in a strong electric field, as well as of small LC droplets in a strong electric field, if the angles of incidence of light $\vartheta_\mathrm{p} = 0°$ and $\vartheta_\mathrm{p} = 90°$ or if the light incident on a small LC particle is polarized perpendicular to the plane of incidence.

Let us dwell upon the method of calculation of the PDLC film structural factor S featuring in equation (4). The structure factor S allows account to be taken of the influence of the processes of interference of light that occur in a system of correlated scatterers and can be calculated by the formula [21]

$$S(\theta) = 1 + 4\pi n \int_0^\infty [g(r) - 1]\frac{\sin\mu r}{\mu r} r^2\,\mathrm{d}r \quad (16)$$

where $\mu = 2k\sin(\theta/2)$, and $g(r)$ is the radial distribution function characterizing the laws of spatial arrangement of particles. No work devoted to the investigation of the laws of spatial arrangement of LC droplets in PDLC films is known to us. Therefore, we assumed that the PDLC film structure is analogous to the structure of a film formed from randomly arranged rigid spheres and that S can be calculated in the Percus–Yevick approximation [27, 28]. In accordance with this assumption, the PDLC film structure factor S was calculated by the formula

$$S(\theta) = \left\{1 - 24c_\mathrm{v}\int_0^1 c(x)\frac{\sin[4kr\sin(\theta/2)x]}{4kr\sin(\theta/2)x}x^2\,\mathrm{d}x\right\}^{-1} \quad (17)$$

where

$$c(x) = \begin{cases} -\alpha - \beta x - \delta x^3, & x < 1 \\ 0, & x > 1 \end{cases}$$

and

$$\alpha = (1 + 2c_\mathrm{v})^2/(1 - c_\mathrm{v})^4$$

$$\beta = -6c_\mathrm{v}(1 + c_\mathrm{v}/2)^2/(1 - c_\mathrm{v})^4$$

$$\delta = 0.5c_\mathrm{v}(1 + 2c_\mathrm{v})^2/(1 - c_\mathrm{v})^4.$$

4. Results and discussion

The results of the calculation of the PDLC film transmittance for various angles of incidence ϑ_i and two states of polarization of the light incident on the film, have been compared with the experimental data given in figures 1 and 2. The calculation was carried out using equation (2), and the path length l of the non-scattered light in the film was found from the relation

$$l = d\left[1 - \frac{\sin^2\vartheta_\mathrm{i}}{n_\mathrm{p}^2}\right]^{-1/2}. \quad (18)$$

The extinction coefficient ε was calculated using both Beer's law, equation (3) and ITA, equation (4). It can be seen that the use of Beer's law for calculating the extinction coefficient leads to results which appreciably differ from experiment. This is due, in our opinion, to the incorrectness of applying Beer's law to dispersion media with a high concentration of LC droplets. Beer's law does not take into account all the physical processes occurring for light propagation in dispersion media with a high concentration of scatterer. In particular, it does not take into account the processes of interference of waves scattered by individual LC droplets, processes which play an important part in the presence of ordering in the arrangement of the droplets and lead to a decrease in the extinction coefficient [20]. Furthermore, it does not take into account the fact that at a high LC concentration the distance between the LC droplets is comparable to their size and, in some cases, to the incident light wavelength as well.

Taking account of interference processes within the framework of ITA makes it possible to increase considerably the accuracy of calculation of PDLC film transmittance. As seen from figures 1 and 2, the calculations by equations (2) and (4) are in good agreement with the experimental data. Therefore, ITA can be regarded as one of the methods for calculating the transmittance of PDLC films. Besides, the good agreement of the experimental data with those calculated in the ITA points to the fact that in analysing the laws of spatial arrangement of LC droplets in PDLC films, the latter may be thought of as rigid unsqueezable spheres and the Percus–Yevick approximation can be used for calculating the radial distribution function $g(r)$. (The foregoing holds, at least, for the method of making PDLC films used in [22].)

Attention is drawn to the fact that, as follows from figure 1, at angles of incidence ϑ_i larger than 40°, when the film transmittance $T \lesssim 0.8$, the discrepancy between experimental and calculated data increases with decreasing transmittance of the film, and at $T \approx 0.3$ it reaches a few tens percent. We believe that this discrepancy is not associated with ITA errors. Indeed, as follows from [20], the use of ITA beyond its applicability limits leads to an overestimated value of the transmittance, while from figure 1 it follows that the transmittance calculated in the ITA is lower than the measured value. The observed difference between measured and ITA-calculated transmittance may be due to other causes. One of them is that in the calculations we used the PDLC parameters given in [22]. Errors in the determination of these parameters could have led to errors in the transmittance calculations. Another possible cause is associated with the measurement data. Indeed, for correct measurement of the coherent transmittance T it is necessary to measure the intensity of the attenuated incident light I. In actual experiments, however, due to the finiteness of the receiver field of view in transmittance measurements, not only attenuated incident light, but also part of the scattered light is registered. For this reason the measured transmittance is higher than the coherent transmittance T and this difference is the greater, the lesser the value of T.

5. Conclusion

The comparison of the experimental and calculated data made in this work has shown that in investigating the optical properties of PDLC films, it is necessary to take into account the specificity of the scattering processes in such films, a specificity which is due to the high concentration of LC droplets in the films. For this reason, Beer's law, which is used successfully to analyse the laws of extinction of light by dispersion media with a small concentration of scatterers, leads to large errors in calculating the coherent transmittance of PDLC films. More accurate results are obtained by using the ITA which takes into account the processes of interference of waves scattered by individual LC droplets of the PDLC film.

It is worth drawing attention once more to the fact that at low and high droplet concentrations we have exponential extinction of light. The difference for these cases is in the definition of the extinction coefficient ε. The dependence of ε on droplet concentration is linear at low concentration, equation (3) and non-linear at high concentration, equation (4).

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