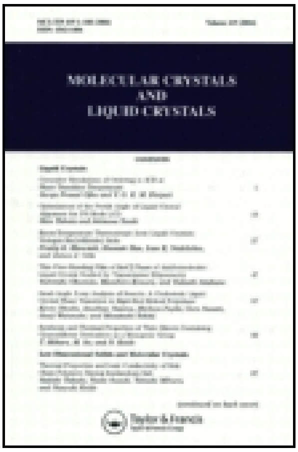

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THE SCATTERING EFFICIENCY OF THE DICHROIC DYE CONTAINING POLYMER DISPERSED LIQUID CRYSTAL FILMS

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Abstract. Electro-optical characteristics of the dye-containing polymer dispersed nematic liquid crystals (PDLC) are investigated by two methods: numerically and experimentally. Light transmission of the dyed PDLC films was calculated using the anomalous diffraction approximation and the dye dichroic absorption characteristics. The effect of varying the refractive index of LC, the polymer and LC resistivity and the dye fraction dissolved in a LC droplet on the PDLC transmission was numerically studied. Each of these parameters taken separately has little effect on PDLC transmission. The experiment showed the effect of the dye on electro-optical characteristics of the PDLC film to be more intricate. Two anthraquinone dyes with different molecular structure were used. The PDLC film transmission and scattering efficiency depend on the order parameter, the dichroic ratio and the solubility of the dyes in LC droplet.

INTRODUCTION

Coloured polymer dispersed liquid crystals (PDLC) are formed by incorporation of dye-containing liquid crystals (LC) into the polymer[1]. The dye dissolved in a nematic droplet possesses dichroic properties. When a critical electric field is applied to a LC cell, the LC molecules (host) together with the dichroic dye molecules (guest) with positive dichroism are re-oriented parallel to the field. The contrast of a dye-containing PDLC film is determined by the optical PDLC densities in the ON and OFF states, which are dependent on the dye fraction dissolved in the binder and droplet. Even when the dye has a low order parameter and its dichroic number is small, an essential effect is observed due to its incorporation into PDLC. This may be ascribed to the increase in dye-containing PDLC absorption in the OFF state due to the increasing efficient path length of light passing through the film because of its multiple reflection on the droplet borders[2]. In the case of a dichroic dye, the effect will be more pronounced if all dye dissolves in a LC droplet. The "guest-host" effect in PDLC films is more intricate. The dye affects the PDLC droplet morphology, changes the physico-chemical properties of the binder and LC, and the optical PDLC film density in the ON and OFF states.

The influence of the indicated factors on electro-optical performance of dye-containing PDLC films are experimentally and numerically studied in the present paper.

CALCULATIONS

To describe the transmission characteristics of light scattering PDLC film with bipolar director orientation in droplets for the incident polarized monochromatic light wave, we used the Bouguer-Lambert law:

$$I / I_0 = \exp(-\mu d) \tag{1}$$

where I_0 and I are the intensities of the incident and attenuated waves, respectively, μ is the attenuation factor, d is the PDLC film thickness. The attenuation factor is proportional to the number of droplets, distributed in the unit volume of a polymer binder, multiplied by the total extinction cross-section of a separate droplet (σ).

In the external electric field a group of droplets with the radius larger than the critical one (R_{cr}) is reoriented along the field (dielectric anisotropy is positive) while a group of droplets with smaller radius conserves the initial configuration. Therefore, taking into account the droplet polydispersion described by the size distribution function P(R) of the droplets, the attenuation factor may be presented as:

$$\mu = \int_0^{R_{cr}} \sigma_1(R)P(R)dR + \int_{R_{cr}}^{\infty} \sigma_2(R)P(R)dR \quad (2)$$

where the first term describes the light attenuation by the droplets retaining the initial configuration and the second one describes that by the droplets which changed their structure.

To determine the extinction cross-section of a droplet we used the anomalous diffraction approximation developed for nematic droplets in[3].

When the dye is added into a LC film, it dissolves both in LC droplets and polymer binder. Denoting the dye fraction dissolving in a LC droplet as x, we write the refractive index of the droplet in the form $m = n_{LC} - i\chi x$ where the imaginary part of the refractive index describes the light absorption by LC droplet ($n = n_o$ or n_e; $\chi_{LC} = \chi_o$ or χ_e). In turn, the light absorption coefficient of the polymer binder may be written[4] as

$$\alpha = \frac{4\pi\chi_{(i)}(1 - x)}{\lambda} \quad (3)$$

where $\chi_{(i)} = \frac{2\chi_o + \chi_e}{3}$. Then the light transmission by a dye-containing PDLC film may be determined as

$$T = \frac{I}{I_0} = \exp(-\mu d)\exp(-\frac{4\pi\chi_i(1 - x)}{\lambda} d) \quad (4)$$

When the dye is introduced into a PDLC in small quantities, it changes the refractive and conductivity indices of LC and polymer binder insignificantly. The microscopic studies show that the droplet morphology also changes, i.e. their dimensions and density. The impact of these parameters on the light transmission of a dye-containing PDLC film is numerically studied.

The following indices were used for the calculations: $n_e = 1.74$, $n_o = 1.54$, $n_p = 1.47$, $d = 20\mu m$, $R = 1.0$, $P(R)=0.0009$, $\chi_o = 0.000055$, $\chi_e = 0.00072$, $\lambda = 0.633nm$. Figure 1 shows the transmission as a function of the dye fraction dissolved in a droplet. The larger dye fraction in the droplet, the less the driving voltage. The difference in the driving voltage for the samples with different dye contents in the droplet is essential. We noticed a tendency for the transmission increase in the ON state with the growing dye fraction in the droplet.

The measurements show that the dye has a greater influence on the LC conductivity than on the polymer binder conductivity. The ratio ρ_p/ρ_{LC} usually increases. Figure 2 shows that this increase in ρ_p/ρ_{LC} for small voltage impairs noticeably the transmission of the PDLC film. At high voltages the elecro-optical performance is nearly the same. Besides, note the effect of the refractive indices (n_o) which change when the dye is incorporated. The electro-optical characteristics remain nearly the same.

A greater effect on the PDLC film characteristics is exerted by the change in the droplet dimensions. It is especially strongly revealed in the saturation region (Fig.3). The computations show that in the OFF condition the transmission characteristics of a dye-containing PDLC film differ little from those of a dye-free PDLC.

Only one parameter affecting the transmission was varied during the present study. Therefore, it is of interest to compare the calculations with the experiment where PDLC films containing various dyes with simultaneous effect on all characteristics are studied.

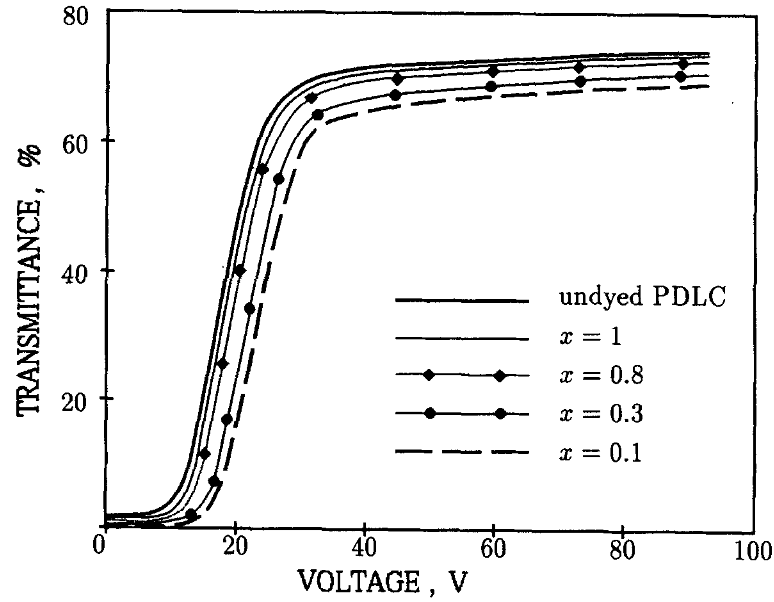


FIGURE 1. The transmittance of the PDLC films with the different dissolved portion of a dye in the LC droplet as function of voltage applied across the cell.

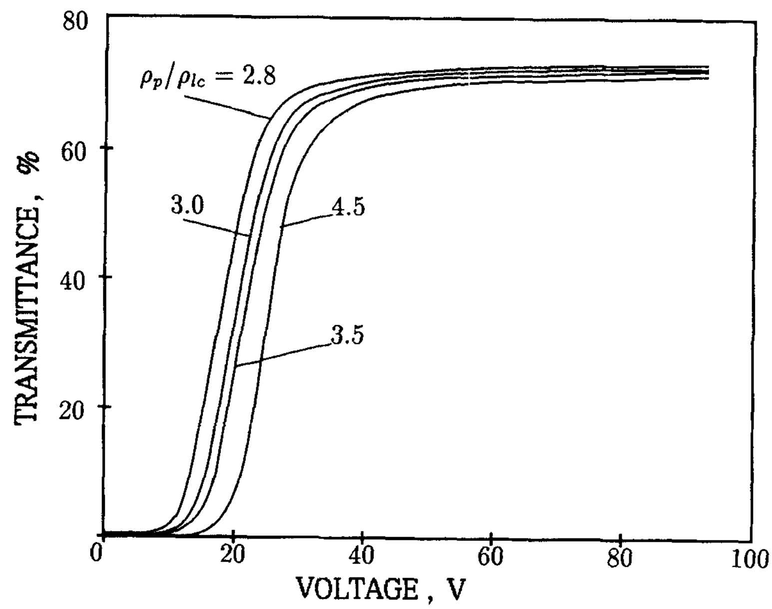


FIGURE 2. The transmittance of the PDLC films with the different ratio of the polymer resistivity to the LC resistivity (ρ_p/ρ_{lc}) as function of voltage applied across the cell.

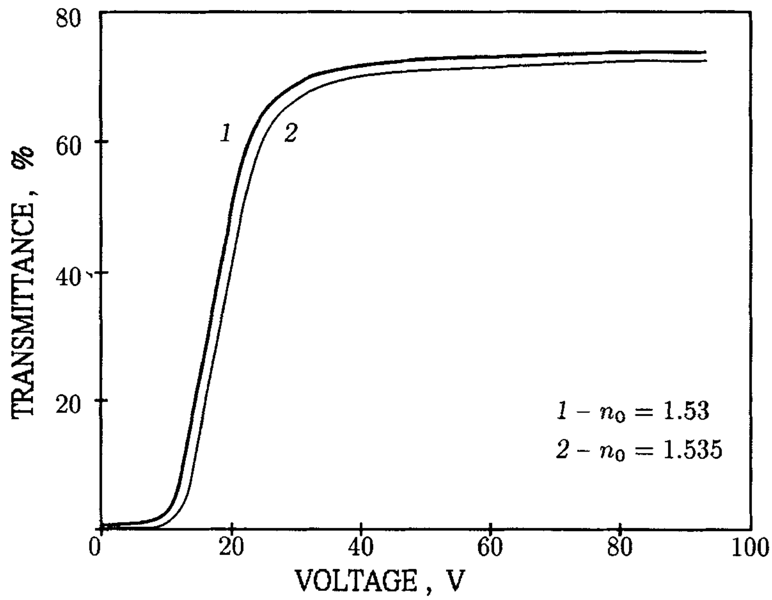


FIGURE 3. The transmittance of the PDLC films with the different ordinary refractive indeces (n_0) as function of voltage applied across the cell.

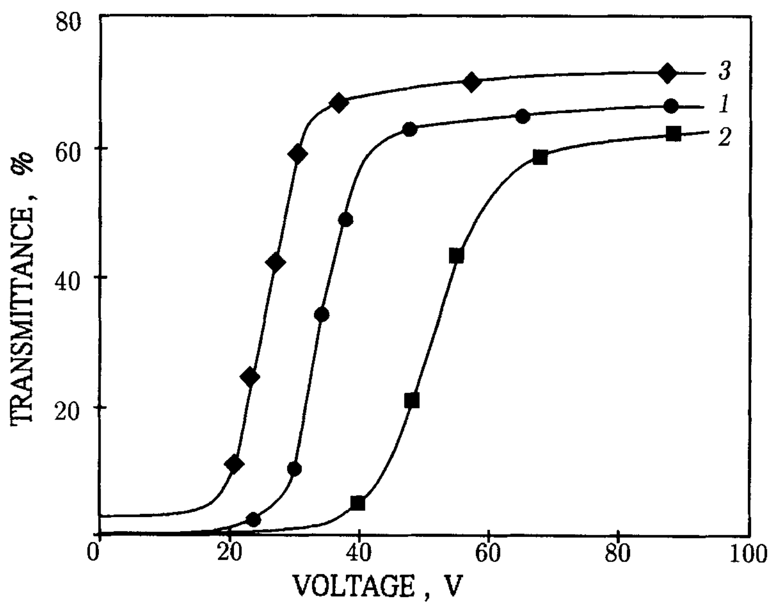


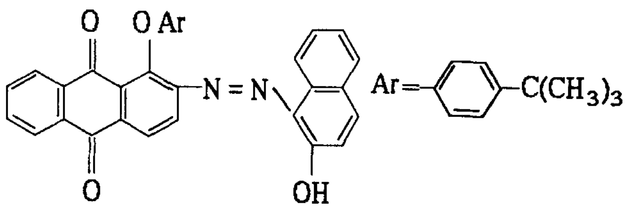
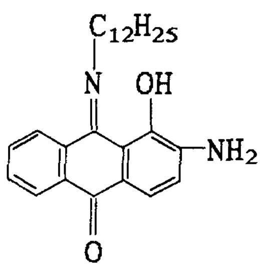
FIGURE 4. The transmittance of the undyed PDLC films (3) and the PDLC films containing 1 % dyes 1 and 2 (TABLE 2) as function of voltage applied across the cell, $f = 50$ Hz.

EXPERIMENTS AND DISCUSSION

Dye-containing PDLC films were obtained by phase separation of a LC and polymer by solvent evaporation. We used polyvinyl acetate as a polymer binder and 4-pentyl-4 -cyano-diphenyl (5CB) as a nematic. The dye concentration amounted to 1 weight % LC. Liquid crystals and dyes were obtained at the Institute of Organic Chemistry (Novosibirsk) and used without additional purification.

We studied the anthraquinone dyes. These are the dyes with positive dichroism. The structures and wavelengths corresponding to the maximum absorption are listed in Table 1.

TABLE 1 List of dyes

No	Formula	λ_{max}, nm
1		527
2		593

The parameters of the dyes (their melting points, extinction coefficients and NMR-spectroscopic data) are described in[5].

The light transmission was measured on the device similar to that described in detail in[6]. A He-Ne laser beam

5mm in diameter was incident to a PDLC film sandwiched between two glasses with transparent electrodes. The limiting aperture of the photodetector ensured the detecting angle less than 0.5^{o} . To determine the scattering efficiency of the dye (α) we used the method suggested in[1]. The efficiency is calculated using the following expression:

$$D_{OFF}/D_{ON} = \alpha/((1 - x) + 3x/(2 + R)) \qquad (5)$$

where D_{ON}/D_{OFF} is the optical density in the presence or absence of the electric field, respectively, x is the dye fraction dissolved in a LC droplet, $R = \frac{\varepsilon_{\parallel}}{\varepsilon_{\perp}}$ is the dichroic number, $\varepsilon_{\parallel}, \varepsilon_{\perp}$ are the molar absorption coefficients of the dye measured parallel and perpendicular to the LC director.

To determine the molar absorption coefficients we used the dye solution in 5CB. In the absence of the external electric field the light transmission by a cell with homogeneous (planar) alignment is determined by $D_{\parallel}$, and when the electric field is applied - by $D_{\perp}$. The optical density was measured on spectrophotometer CF-18 with an integrating sphere. The electric field was applied as a rectangular bipolar impulse with the frequency 400 Hz. The glasses were rubbed to create the homogeneous orientation.

Having measured the optical densities $D_{\parallel}$ and $D_{\perp}$of a dye-containing PDLC film, we find $\varepsilon_{\parallel}, \varepsilon_{\perp}$, R and the order parameter S . After measuring the optical densities D_{ON} and D_{OFF} of the PDLC film we get x and α.

Table 2 lists the characteristics of two PDLC films containing various dyes. Dye 1, having a longer molecular structure, is characterized by a higher dichroic number and order parameter. This dye has a higher concentration in LC droplet. This results in its higher scattering efficiency.

TABLE 2

No	$\varepsilon_{\parallel}$	$\varepsilon_{\perp}$	ε_i	R	S	α	x
	mole^{-1}cm^{-1}cm^{3}x10^{-7}						
1	0.917	0.226	0.456	4.06	0.51	2.92	0.74
2	0.543	0.454	0.484	1.20	0.10	1.36	0.50

The transmission curves of two samples with various dyes have discernable differences (Fig.4). The transmission of a PDLC film containing dye (1) (curve 1) with the higher dichroic number and order parameter is higher in the ON state. Comparison of the experimental results with the calculations shows that experimetally we observe a greater dependence of electro-optical characteristics of the films on the presence of small amounts of dye in them. Though the change in separate physical constants of dye-containing PDLC films is small, their joint impact on the PDLC film stipulates the essential change in the characteristics. Probably, the dyes affect the processes of orientation connected with the changing LC-polymer boundary conditions and the droplet morphology. The latter is especially noticeable by the slope of the transmission curve of sample 2 characterizing its larger droplet dispersion as compared with a dye-free PDLC film.

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