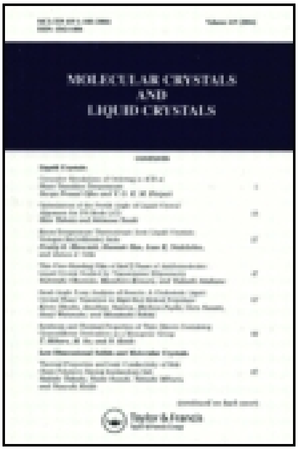

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Low Frequency Relaxation Processes in a PDLC Film

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Based on complex electro-optical and dielectric measurements, the processes of low frequency relaxation in PDLC (Polymer Dispersed Liquid Crystals) films have been studied. Experimental data are presented to investigate PDLC structure and its evolution with increase in LC concentration. These results are found to be in good agreement with observational data taken from polarization microscopy.

Keywords: PDLC; electro-optics; director relaxation

INTRODUCTION

Investigations of liquid crystals (LC) in dispersed states (porous membranes and polymer matrix) first started in the 1970's[1,2], however interest in them is still high. Practical interest in such systems arises because of their characteristics which match the requirements of certain display devices (for example, large screen and projective displays) for which orientated LC layers are less well suited.

The distinction of PDLC's and other heterogeneous systems is not only their novel operating principle, which is electrically controlled light scattering, but also their better dynamic characteristics, when compared with conventional LC systems (about 1-10 ms, i.e. ten times faster than the corresponding time for a LC layer). The dynamic characteristics of the PDLC have been studied by a number of workers. It was established that the characteristic times of the electro-optical response depend on the elasticity, viscosity [3] and dielectric anisotropy of the LC[4], the resistance of both the LC and the polymer, the relative concentration of LC and various anchoring parameters[5,6]. Droplet characteristics (size, shape, director distribution[6]) have also been shown to have a great influence on

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these dynamic processes[9,10]. A study of high frequency relaxation in PDLC has been performed by Bloisi et al [11].

The present work is devoted to the study of some new aspects of low frequency relaxation in PDLC films, and is based on complex electrooptical and dielectric measurements. The results obtained will be used to investigate the structure of PDLCs.

OBJECTS OF INVESTIGATION AND EXPERIMENTAL METHODS

Nematic LC BL001(Merck) and a commercial photoadhesive polymer (NISSAN, Japan) were used to prepare mixtures. LC droplets were formed as a result of using the method of photo-induced phase separation[12]. For this purpose, the LC-polymer mixture (the proportion of LC was varied from 30 to 80 weight %) was placed between two glass substrates with transparent ITO electrodes on the inner sides. The LC layer thickness fixed at 10 μm, by the use of a small quantity of spacers. The cells were irradiated with a Hg-lamp, the intensity of the radiation and exposure time being 5 mW/cm^2 and 30 min respectively. Investigation of the cells using polarization microscope showed the presence of a droplet structure, with characteristic droplet diameter close to 1 μm. The large droplets were observed to have a bipolar structure.

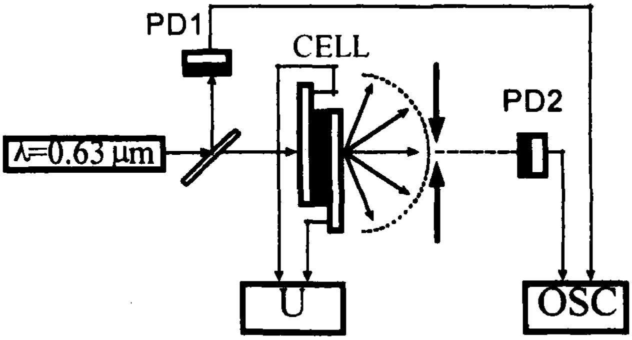

FIGURE 1. Experimental setup scheme.

As can be seen from Fig 1, the examined cell was illuminated using a He-Ne laser beam, some of whose intensity was then lost due to scattering. The part of laser beam that was not scattered (or was only scattered within the angle of 2°) was registered by the photodiode PD$_2$ and sent through amplifier to one of the channels of oscilloscope OSC. An AC voltage was applied to the studied cell from the sound frequency generator U, the applied voltage signal also being sent to the second channel of the oscilloscope. This enabled simultaneous observation of the transmitted beam intensity and the applied voltage signal.The photodiode PD_1 was used to establish the stability of the laser beam intensity. The cell transmittance was defined as $T=(I_{out}/I_{in})*100\%$, where I_{in} and I_{out} are beam intensities before and after cell, respectively.

The dependencies of ΔT, and its alternating ΔT_a and direct ΔT_d components on the frequency of the applied field as well as kinetic curve of ΔT relaxation after switching off the field were the main characteristics measured[13]. Measurements of the real ε' and imaginary ε'' components of permittivity for both the PDLC film and the pure LC were also performed. The dielectric method used was described in detail in [14]. The morphology of the PDLC films were also observed in polarization microscope.

EXPERIMENTAL RESULTS AND DISCUSSION

Dependence of the transmittance change on the frequency of the applied field.

Both the total transmittance change ΔT and its two components ΔT_a and ΔT_d are shown to depend on the field frequency. Typical dependencies for $\Delta T(f)$, $\Delta T_a(f)$ and $\Delta T_d(f)$ are shown in Fig.2. Curve $\Delta T(f)$ increases sharply in the area of several Hertz, reaches its maximum and then gradually goes down towards a limiting value ΔT_s.

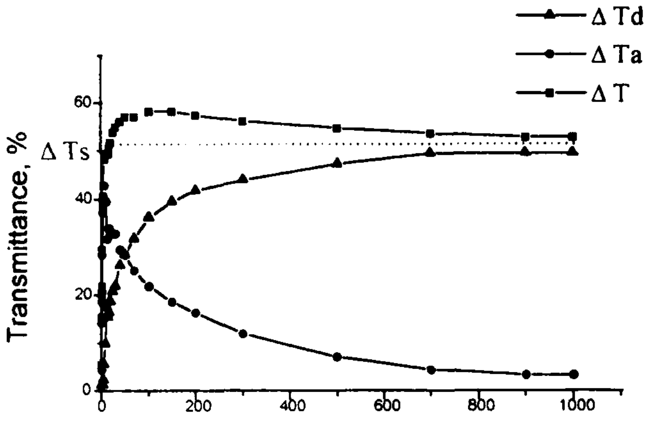

FIGURE 2. Dependence of the transmittance change ΔT and its components ΔT_a and ΔT_d on frequency of applied field.

The initial sharp increase of the curve $\Delta T(f)$ can be explained as resulting from an increase in the effective field inside the cell when the frequency is increased. The reason for such an increase could be the relaxation of the surface polarization at the PDLC-conductive substrate as well as at the edge of each LC drop. Existence of such relaxation processes at f<10 Hz are confirmed by dielectric measurements of PDLC films (Fig.3).

On the other hand, the gradual decrease of the $\Delta T(f)$ curve can not be explained in terms of a dielectric relaxation process. Indeed, no dielectric relaxation was observed in the region f= 50-20000 Hz. The decrease in $\Delta T(f)$ can, however, be explained by considering the relaxation processes in LC drops and taking into account difference between the refractive index of the polymer, n_p, and the ordinary index of the LC n^o. Let the bipolar LC drop be located in the electrical field applied in the vertical direction. Let us also denote the direction of the preferred orientation of the LC director in the bipolar drop by the vector $\mathbf{d_0}$ which goes through the poles of the drop.

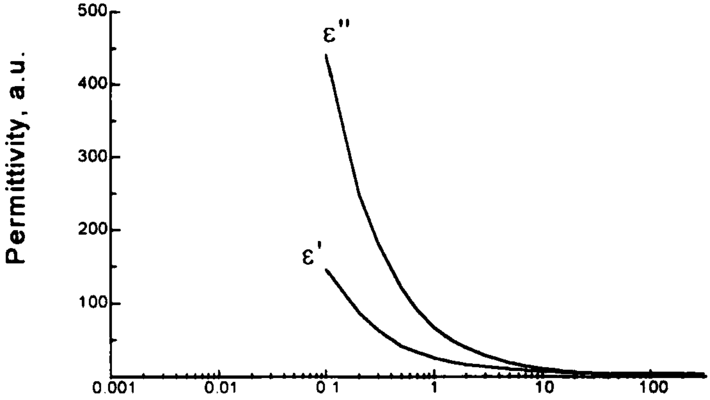

FIGURE 3. Dielectric constants of PDLC layer.

At low frequencies the droplet director orientation, obviously, completely follows the field oscillations. When the field frequency increases, the vector $\mathbf{d_0}$ can deflect within a cone 1 with top angle α_l. Let the condition $n^o<n_p<n^e$ be valid for the investigated polymer-LC mixture (this is the case for most LC-polymer mixtures used in the preparation of PDLCs). Such a condition means that the best concordance between refractive indexes of polymer and LC, which corresponds to maximum transmittance, would not take place when drop is fully oriented along field direction, but when vector $\mathbf{d_0}$ is

oriented at an angle α_2 to the field. When the field frequency increases, the vector $\mathbf{d_0}$ can deflect only to the angle α_3, which is smaller than the concordance angle α_2. This means that for such frequencies, the maximum transmittance of the layer cannot be achieved. The bigger the frequency of the field, the smaller the angular deflection of the drop vector $\mathbf{d_0}$ and the smaller its projection on the direction of maximum correlation of refractive indices. This causes the observed gradual decrease in the $\Delta T(f)$ curve. The asymptotic value ΔT_s corresponds to the field frequencies at which the drop vector $\mathbf{d_0}$ can no longer respond. The value of ΔT_s is, therefore, determined by the extent of correlation of the refractive indices n_p and n^o.

As shown in Fig.2, the alternating component of the transmittance change, ΔT_a, decreases as the field frequency increases. $\Delta T_a(f)$ goes down to zero at frequency $f_{rel} \approx 100$ Hz (in practice, this frequency was found to correspond to a factor of 10 decrease). The same relaxation processes in the PDLC caused the frequency dependencies of the parameters ΔT and ΔT_a. One of these processes, described above, related to the decreasing oscillation amplitude of the drop orientation vector with increasing field frequency. Another factor which influences the character of the frequency relaxation of ΔT and ΔT_a could be size distribution of the drops, since the frequency of the drop relaxation depends on the drop size [15]:

$$\tau_{decay} = \frac{1}{f_{rel}} \approx \frac{\gamma \cdot d^2}{K} \qquad (1)$$

where γ is the orientational viscosity index, d is the drop diameter and K is a Frank elastic constant. There are some reasons to believe that the size distribution of the drops in our case is not too wide. This is confirmed by direct observations of the PDLC structure by polarization microscope as well as by the shape of the $\Delta T(t)$ relaxation curve (Fig.4). One can see that the $\Delta T(t)$ curves for the PDLCs with different LC content consist of two exponents. According to [9,16] the exponent with the shorter time τ_{decay1} corresponds to LC director relaxation in the drop core, while the exponent with the longer time τ_{decay2} relates to relaxation of the droplet periphery. If a wide size distribution of drops were present then a more complicated exponential form of $\Delta T(t)$ curve would have been obtained. So in our case dependence $\Delta T(t)$ confirms a narrow size distribution of LC drops.

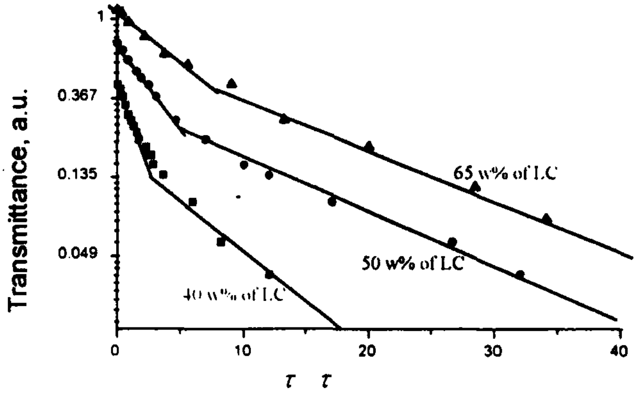

FIGURE 4. Ln-dependence of transmittance on the frequency of the applied field.

The time of drop relaxation can be evaluated from Fig.4. It is about 10 ms. This value correlates well with the value of the relaxation frequency $f_{rel}\approx 100$ Hz, thus confirming that both measurements refer to the same relaxation processes. Placing the values of critical frequency $f_{rel}\approx 100$ Hz, $\gamma=1$ P and $K=10^{-6}$ dyne into equation (1) one can evaluate the size of nematic drops to be $d\approx 1$ μm. Such an evaluation is in order of magnitude agreement with measurements made by polarization microscope.

Dependence of relaxation processes on LC concentration in the system.

To investigate this problem, LC-polymer mixtures with different concentrations of LC were used in the preparation of the PDLC films. Observation of the prepared dispersions by polarization microscope showed that a droplet structure (in particular a PDLC structure) could be observed at a LC concentration of 35-70 weight %. At concentrations of 35-60 weight % only one type of droplet was observed with a size of about 0.5-1.0 μm. At weight concentrationsof 65-70 weight % one more type of droplet appeared with a size of about 5-10 μm. At much higher LC concentrations, the binary LC-polymer mixture separated under irradiation into macrodomains of LC and polymer. Such evolution of structure with increase in LC concentration in a dispersion was observed earlier in [9].

Relaxation curves $\Delta T(t)$ for dispersions with different LC concentrations are shown in Fig.4. We can see that, firstly, appearance of the new drop type does not change essentially the shape of the relaxation curve. This could be explained by the fact that the big drops do not bring a significant contribution to the light scattering. Secondly, the relaxation time of the exponential components of $\Delta T(t)$ monotonically increases with increase in LC concentration. The time τ_{decay1} of the faster process, associated with LC reorientation in the droplet core, increases more rapidly. It is possible to plot the concentration dependence of the drop size d calculated from (1) using the value of $\tau_{decayf1}$ (Fig.5).

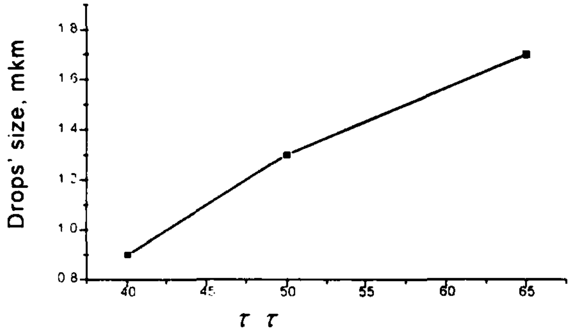

FIGURE 5. Dependence of droplet size on LC concentration.

As we see, in our case increasing the LC concentration in the dispersion leads not only to the appearance of a new type of droplet of bigger size but also to an increase in the size of the smaller droplets.

CONCLUSION

Thus, low frequency and time dependencies of electrically controlled light transmittance of the PDLC films are studied. The non-monotonic behaviour of the $\Delta T(f)$ curve is explained. Low frequency dielectric relaxation and correlation between the refractive indexes of the polymer and LC lead to its shape. The curve of the relaxation kinetic $\Delta T(t)$ corresponds to a narrow distribution of LC droplet sizes. It consists of two exponents corresponding respectively to relaxation of the core part of the LC drops and their peripheries. Using the characteristic relaxation time obtained from the measurements of $\Delta T(f)$ and $\Delta T(t)$ curves the size of LC droplets $d \approx 1$ μm was evaluated. Increase of LC concentration leads to the appearance of a new sort of LC droplet with a size of 5-10 μm. Additionally, the size of the smaller droplets also increases. These results, obtained from electroopical curves, were confirmed by direct observations made using a polarization microscope. They show that electro-optical measurements can be useful tool for PDLC structure investigation.

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