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ANGULAR TRANSMISSION OF POLYMER DISPERSED LIQUID CRYSTALS FILMS

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Abstract The angular transmission of Polymer Dispersed Liquid Crystals (PDLC) films has been experimentally investigated as a function of droplets dimension, refractive indices and thickness of sample. The experimental data have been interpreted in terms of the Anomalous Diffraction Approximation slightly modified in order to take into account extra-scattering phenomena generated by some of the peculiar PDLC characters. The dimension of the droplets and refractive index difference between polymer and liquid crystal are found to be the most important factors affecting the light transmission as well as extra scattering factors.

INTRODUCTION

Polymer Dispersed Liquid Crystals (PDLC) are composite materials formed with microdroplets of liquid crystal dispersed in a polymer matrix.^{1,2} PDLC are opaque as droplets scatter light for the random distribution of their directors, but they become transparent by application of an external field. Such change of states has proposed PDLC as alternative materials in opto-electronics devices.^{2,3} It is well known that one of the major problems preventing the extensive use of PDLC films is the drastic dropping of transmitted light with increasing of the viewing angle. The phenomenon is indicated in literature as "Haze".

The transmission properties in PDLC films have been studied as a function of droplet density, droplet size, wavelength and applied field in several papers over last decade.⁴⁻¹⁵ Zumer⁴ has derived the theoretical scattering cross-section for nematic droplets with different internal configurations (a uniformly oriented nematic structure, a radial structure and an isotropic droplet with a nematic boundary layer) in the Anomalous Diffraction Approximation

(ADA). O.A. Aphonin and V.F. Nazvanov⁵ have proposed an expression for bipolar droplets. Several experimental works have tested ADA. Whitehead *et al.*¹³ have measured the transmissions of PDLC films as a function of incident angle and were able to detect an oscillatory structure in their experimental data as their samples presented a narrow distribution of droplet size. ADA has been successfully used by Drzaic and coworkers⁷⁻⁹ to interpret their experiments on powered and unpowered PDLC. In particular they showed that turbidities have a quadratic dependence on the refractive index difference between the nematic liquid crystal and polymer matrix. The same authors found that turbidity has higher values in unpowered PDLC films and in films with low droplet densities. These results indicate that the scattering properties in PDLC are governed by the structure of the PDLC film beyond the liquid crystal/polymer matrix interface. We must recall that theoretical analysis has been performed involving weakly scattering and/or low density films, even if the most interesting PDLC, from a technological viewpoint, have high droplet densities and are strongly scattering. As a consequence several neglected terms (such as structure factors, multiple scattering, refractive index difference at droplet boundary, not uniform director alignment inside droplets, droplet size and shape distributions) could affect the scattering properties of PDLC as extra scattering factors. Recently, two models have been proposed for the scattering cross-section, both for bipolar⁵ and radial¹⁴ droplets in order to take into account that liquid crystal molecules of a powered PDLC are not perfectly aligned at droplet boundary. A powered droplet of radius r_0 is divided into two parts (see Fig. 1): a central part (between 0 and r), where the liquid crystal molecules are uniformly aligned to the field, and an edge part (between r and r_0), where the molecules keep the off-field configuration (bipolar or radial). The higher is the field, the larger is the value of r which must be used as a field parameter to fit experiments.

J. Kelly and W. Wu¹⁶ have developed a model in order to evaluate single and multiple scattering effects in PDLC with high droplet density. Whitehead *et al.*¹³ have considered different droplet sizes, shapes and orientations to fit their data.

In order to study the influence of the extra scattering factors on the scattering properties of PDLC films, we have investigated the light transmission across PDLC as a function of the viewing angle. The analysis of the experimental data has been performed in terms of the Anomalous Diffraction Approximation opportunely adapted to take into consideration extra scattering phenomena.

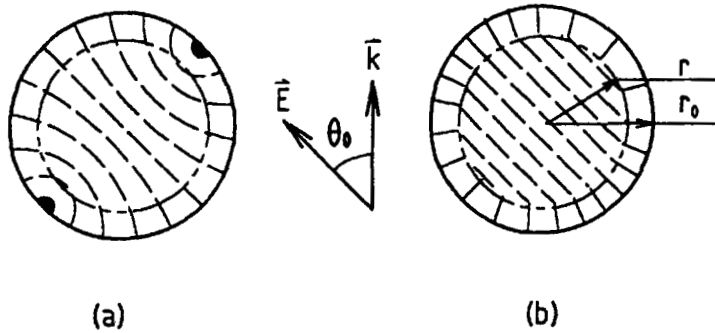


FIGURE 1 Realistic (a) and modelled (b) axial director configuration in a PDLC droplet with strong anchoring. Droplet is divided in a central part, where the nematic molecules are aligned to the field, and in an edge part, where molecules keep their off-field configuration (radial in this figure).

THE MODEL

When a sufficiently high external field is applied to a PDLC droplet the nematic director can be considered uniformly aligned in the field direction all over the droplet volume except close to the surface. If light absorption is not taken into account the "on state" transparency of the PDLC depends on droplet scattering characters, scatterer density and sample thickness. According to ADA^{4,7} the scattering character is expressed by the following cross-section:

$$\sigma_s = 2 \sigma_0 [\cos^2 \alpha_0 H(v_0, 0) + \sin^2 \alpha_0 H(v_0, 0)]$$

where

$$H(v, 0) = 1 - \frac{2}{v} + \frac{2}{v^2} (1 - \cos v)$$

and

$$v_e = 2kR \left[\frac{n_{\text{eff}}(\theta)}{n_p} - 1 \right], \quad v_o = 2kR \left[\frac{n_o}{n_p} - 1 \right], \quad n_{\text{eff}}(\theta) = \frac{n_o}{\left[1 - \left(\frac{\sin \theta_o}{n_p} \right)^2 \left(1 - \left(\frac{n_o}{n_e} \right)^2 \right) \right]^{\frac{1}{2}}}$$

being α_0 the polarization angle, θ_0 the incident angle in air, n_p the index of refraction of the surrounding polymer, n_o and n_e the ordinary and extraordinary refractive indices of liquid crystal, $n_{\text{eff}}(\theta)$ the effective refractive index of the liquid crystal droplet (corrected for the refraction at the air/polymer interface¹⁰) at the viewing angle θ , respectively. k is the module of wave vector of incident light and σ_0 is the average geometric cross-section of the droplets.

The angle dependence of transmission, when the collecting angle is very small, can be obtained according to:^{13,17}

$$\frac{I}{I_0} = \frac{1}{2} \left[\exp(-\beta \sigma_v d) + \exp(-\beta \sigma_h d) \right]. \quad (1)$$

Here, light is regarded as the sum of V and H polarized components. I_0 and I are the incident and transmitted light intensities, respectively. σ_v and σ_h are total diffraction cross-sections with respect to V and H directions. β is the droplets' density and d is the thickness of the cell. The change of the incident angle and length of the path in PDLC film, due to the refraction at air/polymer interface, is given by (see Fig. 2):

$$\sin(\theta) = \frac{\sin(\theta_0)}{n_p} \quad \text{and} \quad d = \frac{d_0}{\cos(\theta)} = \frac{d_0}{\cos \left[\sin^{-1} \left(\frac{\sin(\theta_0)}{n_p} \right) \right]}$$

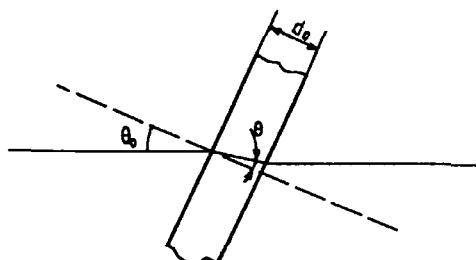


FIGURE 2 Change of the incident angle and path length in PDLC cell.

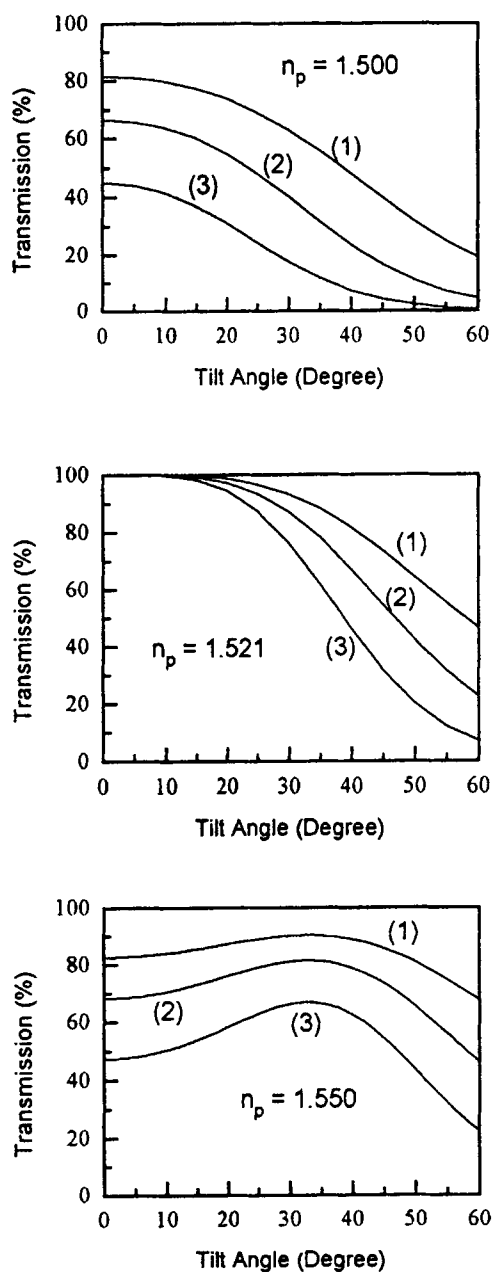


FIGURE 3 Angular on-state transmission of PDLC for different values of polymer refractive index, n_p , and droplets' radius, r_0 : (1) $r_0 = 1 \mu\text{m}$, (2) $r_0 = 2 \mu\text{m}$, (3) $r_0 = 4 \mu\text{m}$.

We calculated the on-state angular transmissions of PDLC with three values of refractive indices of matrix and droplets' radii, to see how the transmission changes when the refractive index of the matrix n_p is lower, equal and higher than the ordinary refractive index of liquid crystal. n_o and n_e were set equal to 1.521 and 1.746, which are the measured refractive indices of E7 nematic liquid crystal. n_p was taken equal to 1.500, 1.521 and 1.550 respectively. Results are shown in figure 3. Theoretical calculations predict a strong dependence from refractive indices mismatch and droplets' size.

SAMPLES AND EXPERIMENTAL SET UP

PDLCs by Poly-Methyl MethAcrylate (PMMA) or Poly-Iso Butyl MethAcrylate (PIBMA) were made by Thermal Induced Phase Separation.¹⁵ PDLCs by Epon 815 and Capcure 800 were made by Solvent Induced Phase Separation.¹⁵

Eutectic nematic mixtures, E7 and E49, were used as liquid crystals. The transmission of samples was measured using the simple set up shown in figure 4.

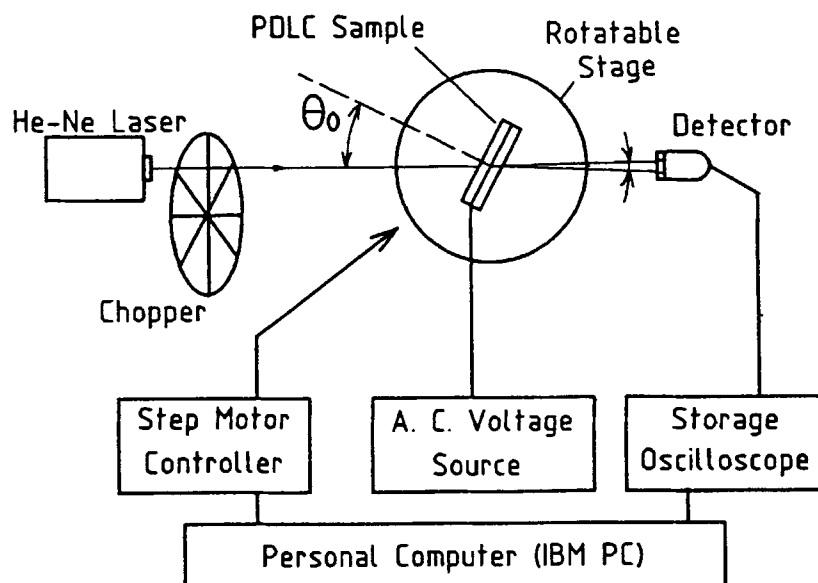


FIGURE 4 Experimental set up.

The He-Ne laser beam was passed through the chopper and the beam expanding lenses. Then it was declined by a 30% attenuator to cut down the intensity into the linear range of the detector. The beam was defined to be 0.8 mm in diameter by a variable iris before entering the sample. The light coming out from the sample was collected by the detector with an acceptance angle equal to 2.5° and produced an alternating current which was opportunely amplified. The sample was mounted on a computer-controlled rotatable stage to vary the incident angle θ_0 . Reflection at the surface of the sample was corrected by $I_{\text{on}}/I_{\text{blank}}$, where I_{on} was the detected current with PDLC cell and I_{blank} was the detected current with the corresponding polymer cell at normal incidence.

RESULTS AND DISCUSSION

Some experimental transmissions (dots) are reported as a function of tilt angle in figure 5. A real PDLC film cannot reach the theoretical values of transmission because of the occurrence of extra scattering phenomena, described previously. As a complete and detailed theoretical model is complicate, let us suppose that all the extra scattering factors reduce the light transmission of a given PDLC by a constant quantity ΔT . Then the measured transmission, T^* , can be written as:

$$T^* = \frac{1}{2} [\exp(-\beta\sigma_{\text{sv}}d) + \exp(-\beta\sigma_{\text{sh}}d)] - \Delta T. \quad (2)$$

We must recall that T is the theoretical transmission, i.e. in absence of extra scattering terms and, in particular, without a boundary shell of not uniformly aligned liquid crystal molecules. Therefore an effective droplet radius,^{5,14} r (lower than the real radius, r_0) must be introduced in order to fit light scattering data. In figure 5 the experimental data are composed with the best fitting obtained from equation 2. The best fitting parameters, r , n_p and ΔT , are also shown. The introduction of extra scattering term ΔT into angular transmission equation allows to achieve a very good fit of the experimental data.

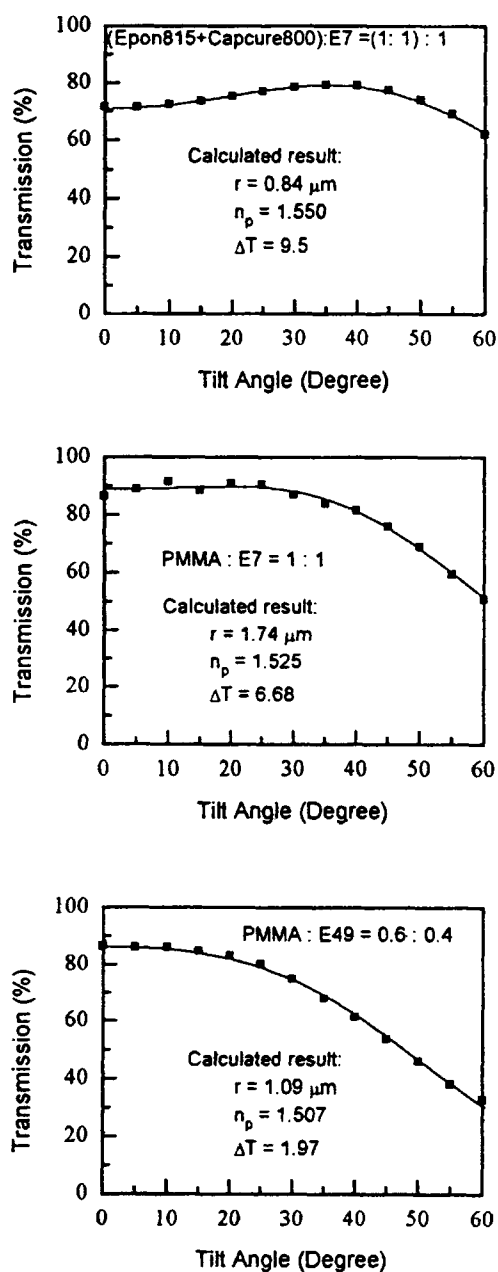


FIGURE 5 Experimental (dots) and theoretical (full lines) transmissions for different PDLC mixtures. Transmission values are corrected for reflections at the two glass-air interfaces.

CONCLUSIONS

We have showed that in order to fit light scattering experiments in PDLC system an extra scattering term must be introduced and an effective droplet radius considered. Furthermore the angular transmission is strongly affected by the droplets' sizes and the refractive index differences between liquid crystal and polymer matrix.

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