

## Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl19>

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Version of record first published: 24 Sep 2006

To cite this article: P. Sixou, C. Gautier & H. Villanova (2001): Nematic and Cholesteric PDLC Elaborated under Shear Stress, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 364:1, 679-690

To link to this article: <http://dx.doi.org/10.1080/10587250108025036>

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## Nematic and Cholesteric PDLC Elaborated under Shear Stress

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The properties of nematic and cholesteric Polymer Dispersed Liquid Crystal (PDLC) with ellipsoidal droplets are reported. The shape of the droplets are modified by a shear stress during the preparation. Different values of the shear stress were used which permit an ellipticity ratio modification by a factor 10. The electro-optical properties are measured for different types of samples and compared with theoretical previsions: transmission or reflection under voltage, threshold voltage, response times. We also describe the preparation of the sample, the experimental apparatus used for the shear stress, and the expected theoretical quantities like response times. When the liquid crystals are nematics, we obtain an increase of the threshold voltages, as predicted. The relaxation time decreases with increasing deformation but the diminution is not as great as predicted. When the liquid crystals are cholesterics, the high deformations induce polygonal fields, which strongly modify the electro-optical properties of the micro-composite. The threshold voltages and response time at applied voltage decrease whereas the relaxation time is increasing.

**Keywords:** PDLC; nematic; cholesteric; shear stress

## INTRODUCTION

Nematic and cholesteric PDLC materials were widely studied. Now, theoretical predictions, taking into account the shape of the droplets [1-3], indicated that electro-optical properties shall be improved by modification of droplets shape [4;5]. In this paper, we present results of PDLC elaborated under shear stress in order to obtain ellipsoidal droplets. The morphology of the film is then observed by microscopy and scanning electron microscopy (SEM). The measured electro-optical properties are presented and compared with theoretical predictions.

## EXPERIMENTAL

The polymerisation-induced phase separation (PIPS) method was used to elaborate PDLC film [6]. A special apparatus was created in order to shear PDLC films. It contained a vibrating part and an axis was fixed on it. This axis moved away from its equilibrium position to a distance, which is proportional to the applied voltage. Then, a sinusoidal voltage allowed to generate a shearing movement at the same frequency. One of both glass substrates was placed at the end of the vibrator axis and the second substrate was fixed on a support.

Rapidly, it appears that the viscosity of polymer/liquid crystal mixture was an important parameter in order to obtain a good shear stress. In some case, the viscosity was increased by a preliminary treatment corresponding to a partial polymerisation of the mixture before shearing and gelation treatment.

Both nematic and cholesteric PDLC mixtures were studied. The morphology of shear stress films was observed by microscopy and SEM. The electro-optical properties were measured using a motor-driven Jobin-Yvon monochromator equipped with a photo-multiplier. The photo-multiplier response was monitored either by a HP34401A multimeter or a HP54601B oscilloscope.

## RESULTS AND DISCUSSION

The morphology of sheared PDLC was observed by scanning electron microscopy (SEM). Figure 1a presents two cross sections of the film according to different directions of shear stress. The modification of the

droplets shape appears clearly. An enlargement of a liquid crystal droplet (Figure 1b) allows to estimate an ellipticity ( $L/l$ ) about 2.

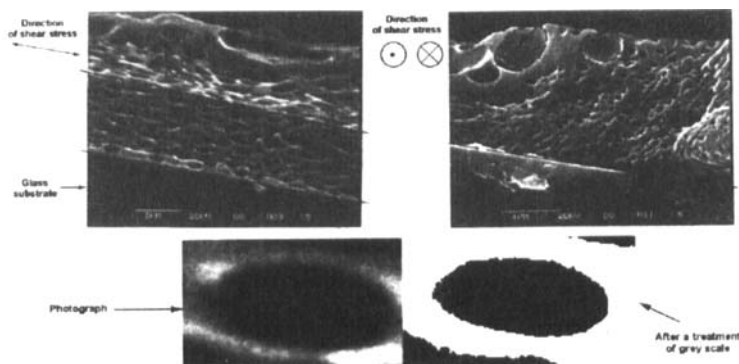


FIGURE 1 Morphology of PDLC elaborated under shear stress observed by scanning electron microscopy : (a) cross section, (b) enlargement of a liquid crystal droplet (ellipticity  $\sim 2$ ).

### Nematic PDLC

In a general way, the observations of various samples indicate that a sinusoidal shearing leads to a dislocation of the droplets, in spite of a suitable mixture viscosity. The morphology of PDLC films containing NOA65/N2 mixture polymerised after several shearing pass is presented in Figure 2a, showing the dislocation of droplets.

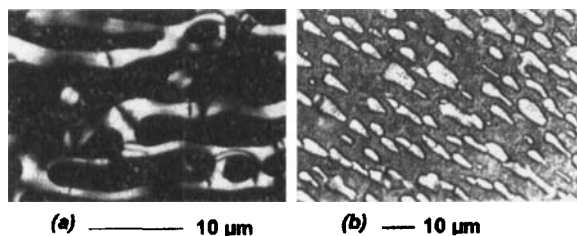


FIGURE 2 Morphology of various PDLC elaborated under shear stress observed by microscopy : (a) case of NOA65/N2 mixture after several shearing passes, (b) case of NOA81/N1 mixture after one single shearing pass. See Color Plate XXVII at the back of this issue.

On the other hand, one single shearing pass doesn't destroy the structure but induces homogeneous ellipsoidal droplets (Figure 2b).

In order to estimate the optical anisotropy, the cells were placed between two parallel polarizers. The transmission intensity was measured versus the angle  $\theta$  defined by the both directions of polarizers and shear stress (Figure 3a). Figure 3b presents the results obtained for two PDLC mixtures. The transmission intensity indicates a clear optical anisotropy in the cases of sheared samples in comparison with no

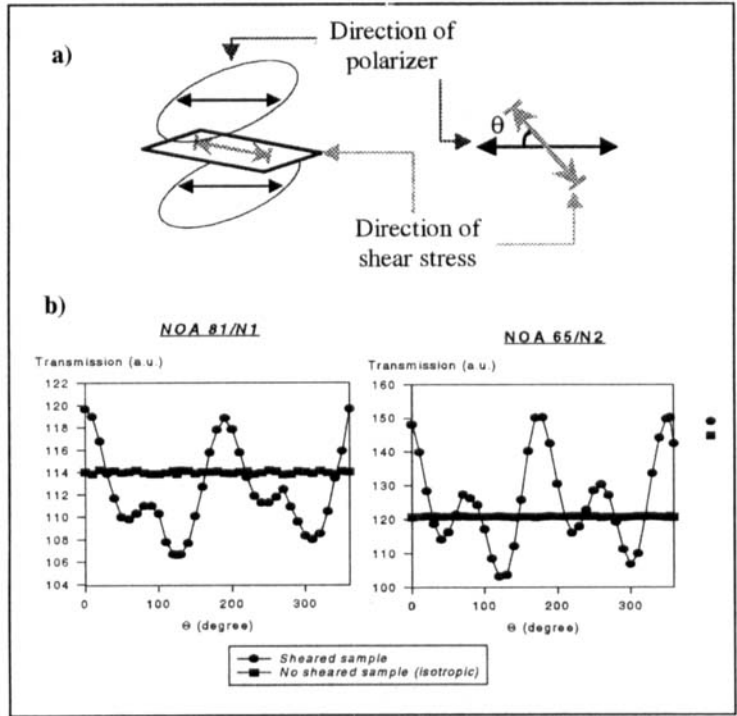


FIGURE 3 (a) Schematic representation defining the angle  $\theta$  between the directions of polarizer and shears stress, (b) Variation of transmission measurements versus  $\theta$  in the cases of sheared and no sheared samples for two PDLC mixture.

sheared films, due to the anisotropy of the matrix structure, on the one hand, and the alignment of the liquid crystal molecules in the direction of the droplets long axis, on the other hand.

Then, the transmission intensity was measured according to applied voltage (Figure 4a). It appears that the transition from the OFF-state to the ON-state needs higher voltage when the ellipticity increases.

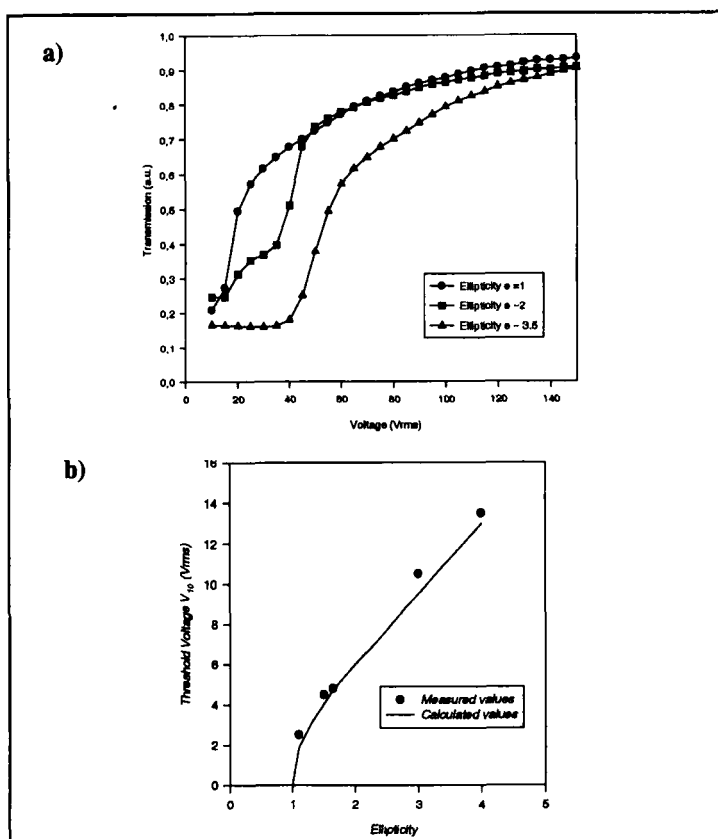


FIGURE 4 (a) Transmission measurements versus applied voltage for several cases of ellipticity, (b) threshold voltage  $V_{10}$  versus ellipticity.

The variation of threshold voltage  $V_{10}$  versus ellipticity is presented in Figure 4b. For spherical droplets, the model predicted an increase of  $V_{10}$  versus ellipticity as indicated in Equation (1) [7].

$$V = \frac{d}{3L} \left( \frac{1}{\epsilon_0} \frac{e(e^2 - 1)}{3L} \frac{K}{\Delta\epsilon} \right)^{1/2} \cdot \left( \frac{\rho_p + 2}{\rho_{lc}} \right) \tag{1}$$

The variation of  $V_{10}$  (Figure 4b) is in good agreement with theoretical previsions, indicating an increase of organisation difficulty in ellipsoidal droplets under applied voltage. Then, the threshold voltage increases with increasing ellipticity.

Then, we have studied the influence of shear stress in PDLC on the relaxation time versus applied voltage for sheared ( $e \sim 3$ ) and no sheared samples (Figure 5).

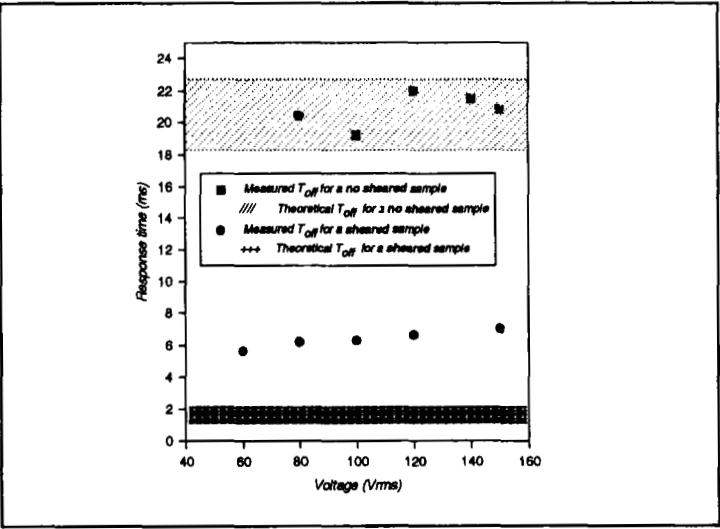


FIGURE 5 Variation of relaxation time  $t_{OFF}$  versus applied voltage for sheared and no sheared samples.



In the both kinds of sample, the model predicts that the relaxation time is independent of applied voltage (Equation 2 [7]).

$$t_{OFF} = \frac{\gamma L^2}{K(e^2 - 1)} \quad (2)$$

The experimental results are in good agreement with the predicted variation and with the  $t_{OFF}$  value in the case of no sheared sample.

However, the predicted  $t_{OFF}$  value in the case of sheared sample is clearly lower than the measured one, indicating that another phenomenon has an effect, which is not included in Equation 2.

Moreover, Equation (2) gives the value of the relaxation time, which is inversely proportional to ellipticity. Figure 6 illustrates this phenomenon by a decrease of  $t_{OFF}$  versus ellipticity.

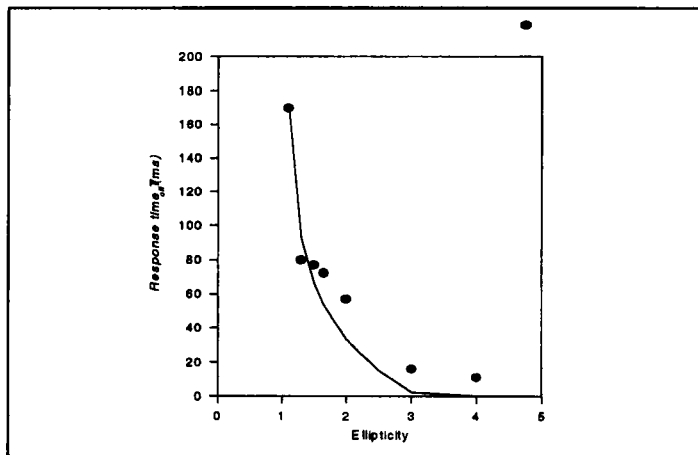


FIGURE 6 Variation of the relaxation time  $t_{OFF}$  versus ellipticity in the case of nematic PDLC.

However, a weak disagreement is observed between experiments and theoretical model from an ellipticity of 2. In fact, the

model predicted a relaxation time up to ten times as low to those observed in the experiments.

### Cholesteric PDLC

The morphology of cholesteric PDLC was observed by microscopy.

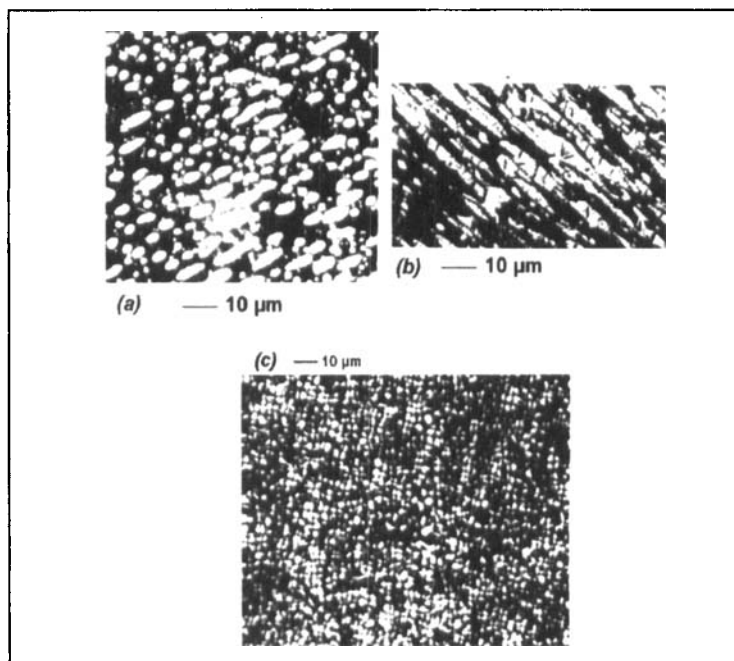


FIGURE 7 Morphology of NOA65/CI cholesteric PDLC elaborated under shear stress observed by microscopy : (a) case of ellipticity  $\sim 2$ , (b) case of ellipticity  $\sim 9$ , (c) polygonal fields.

Figures 7a and 7b show the ellipsoidal droplets in the case of cholesteric PDLC (NOA65/CI) for two ellipticity values about 2 and 9. The variation of ellipticity is obtained by modifying the distance  $D$  of the vibrator axis. When the shape of droplets is spherical or weakly

sheared ( $e \leq 3$ ), the cholesteric liquid crystal get into a classic helicoidal configuration. Beyond this ellipticity, a particular texture appears, similar to polygonal fields. This texture, widely studied by Y. Bouligan et al. [8], contains several distinct domains. Inside these domains, the cholesteric layers are nested all in all and they are called "lines of flare" with reference to their trumpet shape. The liquid crystal alignment under applied voltage occurs in a different manner in comparison with the helicoidal texture. The reflection spectra are summarised in Figure 8 for several ellipticity values.

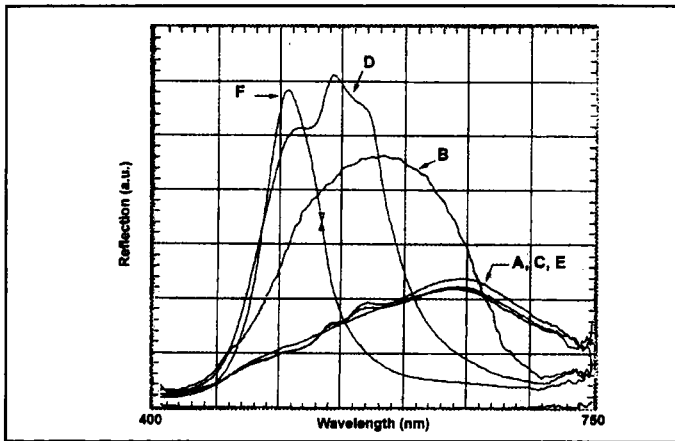


FIGURE 8 Reflection measurements in various cases : (A, C, E) OFF-state with respectively  $e \sim 1.1$ ,  $e \sim 3$  and  $e \sim 9$ , (B, D, F) ON-state with respectively  $e \sim 1.1$ ,  $e \sim 3$  and  $e \sim 9$ .

In OFF-state, the large and weak peaks are centred on 650 nm whatever the ellipticity value may be. This result traduces the multiple orientations of cholesteric axis. In ON-state, the ellipticity influence the reflection properties that leads to a decrease of the peak width and a shift of the reflection maximum towards the low wavelengths. This phenomenon is explained by the decrease of the reflective angle due to the slope and the ellipsoidal shape of droplets. These results are summarised in a schematic representation given in Figure 9.

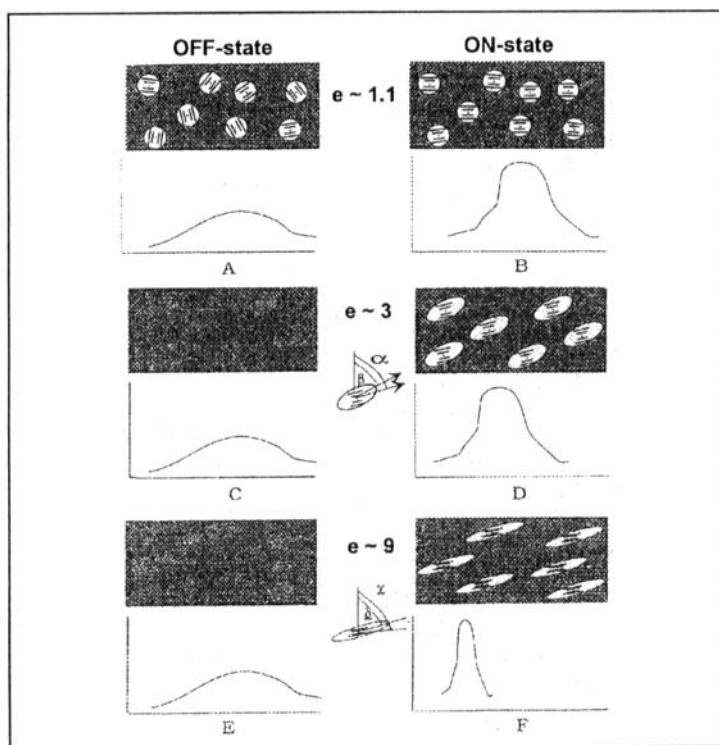


FIGURE 9 Correlation between liquid crystal configuration in ellipsoidal droplets and reflection measurements.

Then, we have measured the variation of the threshold voltage  $[V_{10}-V_{90}]$  as a function of ellipticity (Figure 10a). The liquid crystal behaviour under applied voltage is very different according to the ellipticity value is less or more than equal to about 4. Beyond this ellipticity value, it appears that the threshold voltage suddenly decreases. This phenomenon is attributed to the presence of polygonal fields, which leads to a different organisation process needing a minimum of energy. This phenomenon also induces a strong modification of response time  $t_{ON}$  and relaxation time  $t_{OFF}$ , the one

decreases and the other increases (Figure 10b). This result expresses a fast transition from polygonal fields to no sheared texture. However, the return to polygonal fields texture needs a long time about 1 s for an ellipticity of 9.

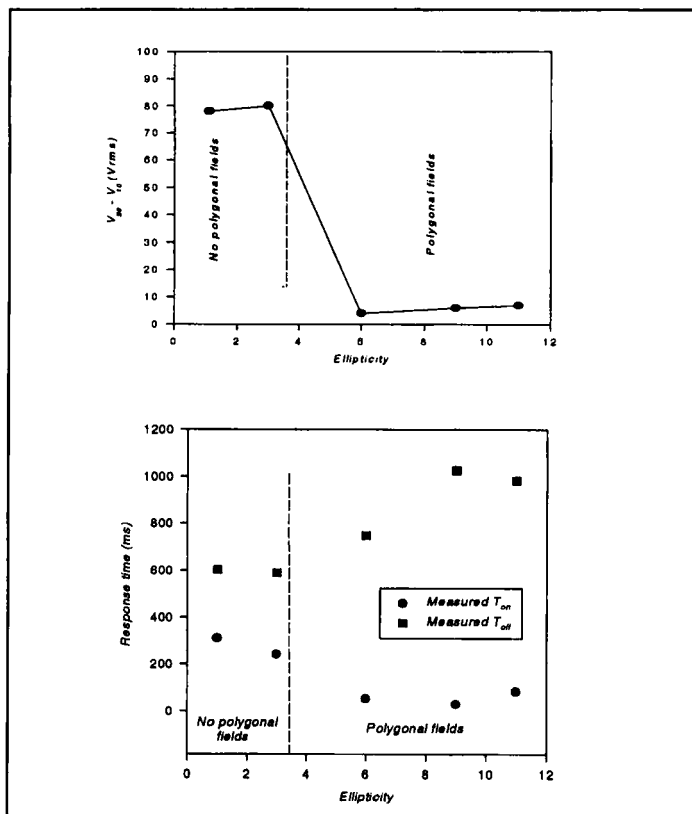


FIGURE 10 Variation of (a) the difference between threshold voltages  $|V_{90} - V_{10}|$  and (b) the response times  $t_{ON}$  and  $t_{OFF}$  versus ellipticity in the case of cholesteric PDLc.

## CONCLUSION

The morphology of PDLC elaborated under shear stress was observed by microscopy, showing the formation of ellipsoidal droplets. The various measurements indicate a modification of electro-optical properties as a function of droplet shape. Concerning nematic PDLC, the threshold voltage increases with ellipticity whereas the relaxation time  $t_{\text{OFF}}$  decreases. About cholesteric PDLC, the formation of polygonal fields induces a strong decrease of the threshold voltage and the response time  $t_{\text{ON}}$ , but a strong increase of relaxation time  $t_{\text{OFF}}$ .

## ACKNOWLEDGMENTS

The authors are grateful to M. Mitov, CEMES Toulouse and J.P. Laugier, CCMA Nice for the SEM analyses.

This work was supported by CCE under contract JOE3-CT970068

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