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# Voltage Controlled Thermo-optical Effect in Polymer Dispersed Liquid Crystals

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**Abstract.** Polymer dispersed liquid crystals (PDLC) are dispersions of liquid crystal micro-droplets in a polymeric binder. Droplets appear as optically uniaxial spheres randomly oriented so that the material is optically isotropic. The application of an external electric field results in a reorientation of the liquid crystal and therefore in a switching of the sample from an opaque to a transparent state. Sample transmittance is also a function of the temperature. If the liquid crystal refractive index in the isotropic phase is equal to the one of the polymer, after the Nematic Isotropic transition the material is transparent. Therefore PDLCs show both thermo-optical and electro-optical effects. In this paper, we present a mathematical model able to describe the dependence of the sample transparency on the temperature, as a function of the applied voltage. We show the model accuracy by comparison with new experimental data that are briefly discussed.

## INTRODUCTION

In Polymer Dispersed Liquid Crystals (PDLC), a dispersion of nematic liquid crystal micro-droplets in a polymeric binder, each droplet appears as an optically uniaxial sphere. The refractive index mismatch between droplets and surrounding medium produces a strong light scattering when droplet size is close to visible light wavelength. Owing to the temperature dependence of the refractive indexes and in particular to the liquid crystal one, sample scattering and transmittance are functions of the temperature. If the refractive index of the polymeric binder is similar to the refractive index of the liquid crystal in the isotropic phase switching from a translucent to a transparent state can be induced by sample heating. PDLC are used to realize electro-optical devices. Liquid crystals, and therefore droplets, are optically and dielectrically anisotropic. The application of a high intensity low frequency external electric field results in a reorientation of droplet directors and of molecular directors

inside the droplets. The refractive index mismatch between droplets and polymeric matrix can be controlled by an applied voltage and PDLC films can be switched from an opaque to a transparent state [1]. Possible applications of these materials range from large-scale flexible displays to windows with controlled transparency to thermal sensors and devices for optical processing. In this paper, we study the coupling of thermo-optical and electro-optical effects in these materials. A mathematical model able to describe PDLC films transmittance for normally incident light is introduced. Theoretical predictions are validated by comparison with experimental results.

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## THE MODEL

We assume that there is no light absorption either in the polymer or in the liquid crystal. The light scattering due to the liquid crystal droplets controls the light transmission. We consider the case of a liquid crystal in the nematic phase having bipolar molecular orientation inside each droplet. PDLC behavior can be described by means of three order parameters [2-4]. The usual molecular order parameter $S = \langle P_2(\hat{n} \cdot \hat{l}) \rangle$ where $P_2$ is the second order Legendre polynomial, $\hat{l}$ is the molecular axis, $\hat{n}$ is the nematic director and the <> parentheses denotes the spatial average. The droplet order parameter $S_d = \langle P_2(\hat{N}_d \cdot \hat{n}) \rangle_{droplet}$ where $\hat{N}_d$ is the droplet director (i.e. the mean value of $\hat{n}$ inside each droplet) and the average is taken over each droplet. The sample order parameter $S_s = \langle P_2(\hat{E} \cdot \hat{N}_d) \rangle_{sample}$, where $\hat{E}$ is the direction of the applied electric field and the average is taken over the whole sample.

The transmitted light intensity ratio is

$$I_t / I_0 = \exp(-\rho_N d_s \sigma_s)$$

Where $I_t$ is the transmitted light intensity, $I_0$ is the incident light intensity, $\rho_N$ is the droplet number per unit volume and $d_S$ is the sample thickness. The average scattering cross section $\langle\sigma\rangle$ is given by [5]:

$$\langle\sigma\rangle = \frac{\pi R_d^2}{2}\left(\frac{4\pi}{\lambda}R_d\right)^2\left(\frac{n_{de}-n_{do}}{n_p}\right)^2\left[\left(\frac{n_p-n_{do}}{n_{de}-n_{do}}\right)^2 - \frac{2}{3}\frac{n_p-n_{do}}{n_{de}-n_{do}}(1-S_s)+\frac{4}{105}(7-10S_s+3\tilde{S}_s)\right]$$

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where:

$$\widetilde{S}_s = \frac{7}{12} + \frac{5}{12}S_s - \frac{35}{32e_a^2}\left[\frac{2}{3} + \frac{\left(e_a^2-1\right)^2}{4e_a^2} + \frac{\left(e_a^2+1\right)\left(e_a^2-1\right)}{8e_a^3}\arctan\left(\frac{2e_a}{e_a^2-1}\right)\right]$$

$R_d$ is the spherical droplet radius, $\lambda$ is the wavelength of the incident light, $n_p$ is the polymer refractive index and $n_{do}$ and $n_{de}$ are the droplet ordinary and extraordinary refractive indices respectively. The sample order parameter $S_s$ and $e_a$ can be computed by the implicit relations:

$$S_s = \frac{1}{4} + \frac{3\left(e_a^2+1\right)}{16e_a^2} + \frac{3\left(3e_a^2+1\right)\left(e_a^2+1\right)}{32e_a^3}\ln\left|\frac{e_a+1}{e_a-1}\right|$$

$$e_a\left(S_s\right) = E\sqrt{\frac{3v_{lc}\varepsilon_p}{\varepsilon_{lc}+2\varepsilon_p-v_{lc}\left(\varepsilon_{lc}-\varepsilon_p\right)}\frac{\varepsilon_{\parallel}-\varepsilon_{\perp}}{K_d}}$$

$$\varepsilon_{lc}\left(S_s\right) = \varepsilon_{\perp} + \frac{1}{3}\left(1+2SS_dS_s\right)\left(\varepsilon_{\parallel}-\varepsilon_{\perp}\right)$$

where $v_{lc}$ is the liquid crystal volume fraction in the sample, $\varepsilon_p$ is the polymer dielectric permittivity, $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ are the liquid crystal dielectric permittivities, $K_d$ is an elastic constant per unit surface (units: Newton per square meter) taking into account the torque which, after the field is switched-off, produces relaxation of the droplets to their original orientation. For the droplet ordinary and extraordinary refractive indices, we use [4]:

$$n_{do} = \frac{2}{\pi}n_o\,\mathrm{F}\left(\frac{\pi}{2}, \frac{1}{n_e}\sqrt{\frac{2}{3}\left(n_e^2-n_o^2\right)\left(1-S_d\right)}\right),$$

$$n_{de} = \frac{n_o n_e}{\sqrt{\frac{2}{3}\left(n_o^2-n_e^2\right)S_d + \frac{1}{3}\left(n_o^2+2n_e^2\right)}},$$

where F(,) is the complete elliptic integral of the first kind and $n_e$ and $n_o$ are the extraordinary and ordinary liquid crystal refractive indices respectively. In a previous paper [5], it has been shown that the droplet order parameter $S_d$ is affected by the applied voltage: for low order parameters it is lowered while near the saturation it is augmented. Here we use the simple expression:

$$S_d(S_s) = S_{d0}\ [\exp(-C_1 S_s) + \exp(-C_2(S_s - 1))],$$

where $C_1$ and $C_2$ are sample-related parameters taking into account the dependence of the alignment of liquid crystal molecules inside each droplet on the value of $S_s$.

The sample temperature T affects almost all the quantities appearing in this model. It has been shown that the thermo-optical effect in absence of applied electric field can be described taking into account only the temperature dependence of the liquid crystal refractive indexes $n_e$ and $n_o$. However, this simple description did not appear accurate for the description of the effect under electric field control. To achieve an adequate mathematical model now we take into account also the temperature dependence of the liquid crystal ordinary and extraordinary dielectric permittivities $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ , of the elastic constant $K_d$ and of the order parameter S(T). All the other quantities are assumed independent on the temperature.

According to the Maier-Saupe theory, for the order parameter, it holds[6]:

$$S = (1 - 0.98\tau)^{0.22}$$

Therefore, S is a universal function of the reduced temperature:

$$\tau = TV^2 / T_{NI} V_{NI}^2,$$

where T is the absolute temperature, V is the volume and $T_{NI}$ and $V_{NI}$ are their values at the Nematic-Isotropic transition. The dependence of the liquid crystal ordinary and extraordinary refractive indexes $n_e$ and $n_o$ on the order parameter is well known in literature [7]:

$$n_e^2 = B_1 + 2B_2 S,$$

$$n_o^2 = B_1 - B_2 S,$$

In a previous paper, dealing with the phase shift induced on a laser beam by a PDLC[8], we have successfully extended these expressions to describe the dependence of the liquid crystal ordinary and extraordinary dielectric permittivities $\varepsilon_{\parallel}(T)$ and $\varepsilon_{\perp}(T)$

$$\varepsilon_{\parallel} = A_1 + 2A_2 S,$$

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$$\varepsilon_{\perp} = A_1 - A_2 S .$$

Here we use the same expressions to model the thermo-optical effect. Values of $B_1$, $B_2$, $A_1$, and $A_2$ can be easily computed by the knowledge of $n_e^2$, $n_o^2$, $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ for any given value of T.

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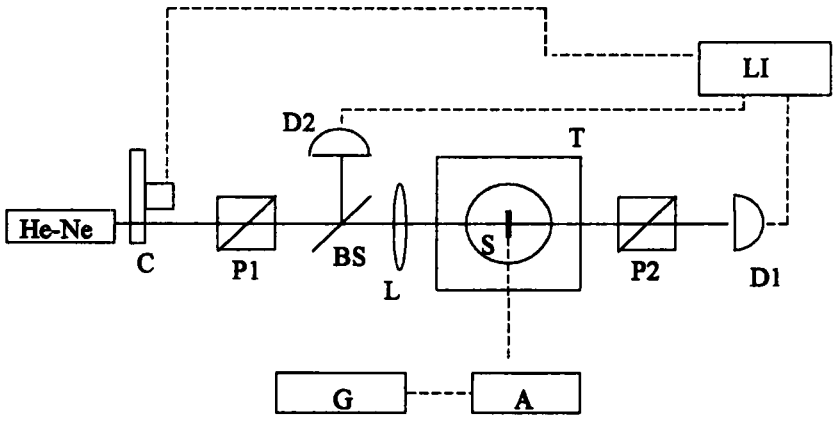


**Fig.1 Experimental apparatus**

**A:** amplifier
**BS:** beam-splitter
**C:** chopper
**D1, D2:** photodiodes
**G:** Signal generator
**He-Ne:** laser Helium-Neon
**L:** lens
**LI:** lock-in amplifier
**P1, P2:** Polarizers
**S:** Sample
**T:** thermostatic oven

## EXPERIMENT AND CONCLUSIONS.

The experimental apparatus is depicted in Fig. 1. PDLC material is achieved by Polymerization Induced Phase Separation (PIPS). The PDLC solution is

included between two glass slides coated with conducting transparent layers (ITO). The sample is cured for 16 hours in a 60°C oven. Mylar spacers 60μm thick determines the sample thickness. The average molecular weight of the polymeric molecules increases while the reaction proceeds and liquid crystals droplets separate. Droplets diameter is determined by curing rate and, for our case, is of the order of 1μm. At room temperature the sample is opaque (translucent). A 5mW He-Ne laser probe beam impinges normally on the sample. A photodiode, in a chopper and lock-in configuration, is used to measure the transmitted intensity. The sample is placed inside a thermostatic oven and temperature is measured by a Pt-100 thermo-resistance. The liquid crystal is E7 by BDH.

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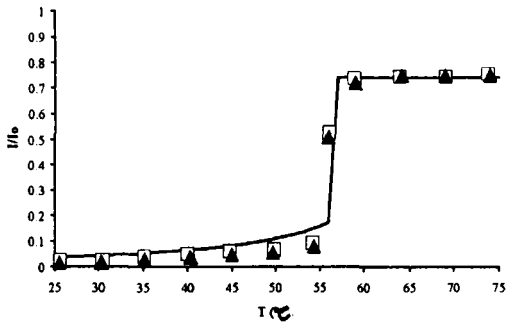


**Fig.2.** Experimental values of the sample transmission ratio Vs. temperature for two different samples are compared with theoretical results (solid line) in absence of applied voltage.

In Fig.2, we report the sample transmission ratio versus the temperature without applied voltage. Solid line represents the theoretical results while experimental results are shown by dots. Hollow squares and triangles refer to two different sets of experimental measures performed on different, though similar, samples. Errors are within dot size. We observe the usual thermo-

optical effect: transition from a scattering to a transparent state when the Nematic-Isotropic transition of the liquid crystal occurs.

The model appears to give a quite accurate description of the phenomenon even if it predicts a more sharp transition. The smothering in the experimental curve can be due to a coexistence area between nematic and isotropic phases.

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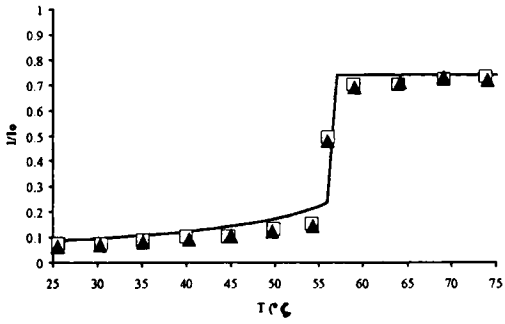


**Fig.3.** Experimental values of the sample transmission ratio Vs temperature for two different samples are compared with theoretical results (solid line). Low frequency (1 Khz) applied voltage is 5 V

In Fig. 3, we observe the effect of the application of the low frequency (1 Khz) voltage (5V). The meaning of the graphic symbols is the same of Fig.2. We can see that the transparency of the isotropic phase is the same, as it had to be expected, while the scattering of the Nematic phase increases, reducing the optical transition height. Hence the voltage can be used to control the transmittance ratio between isotropic and nematic phases of the liquid crystal. In addition, here we can see a good agreement between the model predictions and the experimental results. The parameters values are the same as before and will be the same in all the figures we present.

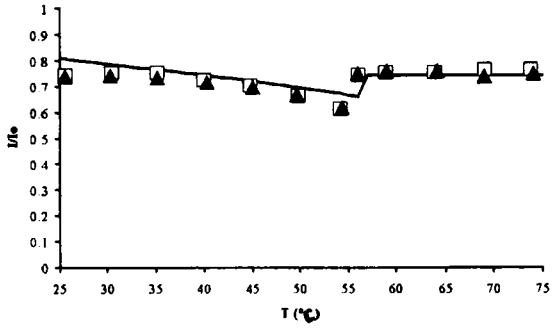


**Fig.4.** Experimental values of the sample transmission ratio vs. temperature for two different samples are compared with theoretical results (solid line). Low frequency (1 Khz) applied voltage is 20 V.

This effect can be controlled with continuity, so that in Fig.4 we see that the transparency can be made practically independent on the temperature by the appropriate voltage (20V); small variation occurring only near the phase transition. With the exception of the first two data, the model gives good results also for this curve. It is worth noting that we have assumed the droplet recall coefficient $K_d$, which express the droplets resistance to the applied torque, to be independent on the temperature. This is quite an unusual assumption since this constant is commonly believed to be proportional to the liquid crystal elastic constant that is proportional to the square of the order parameter. Nevertheless, any attempt to use this proportionality, or even a lower one, changes the lowering of the transparency with the temperature in the nematic phase. Too many parameters are involved in the phenomenon description to take this result as definitive, but this observation seems to suggest that a lower dependence of the recall coefficient on the order parameter has to be taken into consideration.

Finally, in Fig. 5 we observe that for a saturation value of the applied voltage (50V) the film transparency is lowered by the phase transition, with an

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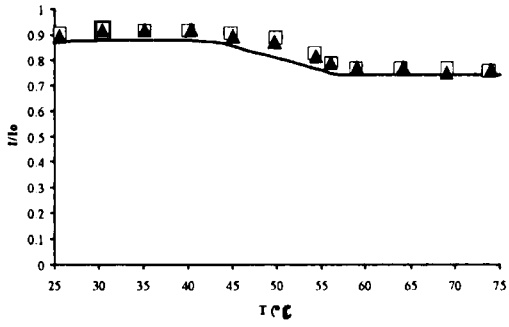


**Fig.5.** Experimental values of the sample transmission ratio vs. temperature for two different samples are compared with theoretical results (solid line). Low frequency (1 Khz) applied voltage is 56 V.

inversion of the thermo-optical effect. Even in this phase, the model seems to give a quite accurate description of the sample behavior with the assumption of constant Kd.

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