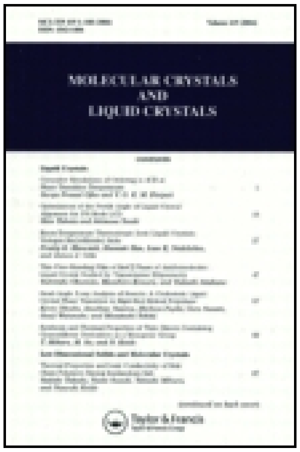

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Nonlinear Optical Effects in Polymer Dispersed Liquid Crystals

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NONLINEAR OPTICAL EFFECTS IN POLYMER DISPERSED LIQUID CRYSTALS

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Abstract We report the observation of optical nonlinearities induced by thermal effects in polymer dispersed liquid crystals. Experimental data about optical switching of a low power laser beam are presented and discussed

INTRODUCTION

Remarkable scientific interest has grown recently about Polymer Dispersed Liquid Crystals (PDLC): new plastic materials which contain nematic microdroplets.[1-8] Starting from a homogeneous solution of polymer or prepolymer, different processes can be used to get a phase separation with droplets formation and polymer solidification; liquid crystal droplets are then formed inside a solid matrix.[1] Several are the advantages over conventional microincapsulation technique and among them we can mention: ability to form droplets of uniform size, ability to adjust the droplet size over a broad range (from 0.01 μm to 20 μm), a wide choice of useful polymers, simplicity and cheapness of the method.

The nematic liquid crystal can exhibit several different director configurations within the droplet[3] with tangential or normal surface alignment. Moreover it is generally possible to find a simmetry direction for the director configuration, thus defining the simmetry axis of the droplet.

The optical properties of these physical systems are dominated by the strong light scattering which is induced by the droplets dispersion. Important parameters which affect such phenomenon are the liquid crystal refractive indexes n_o and n_e and the polymer refractive index n_p.[3] It is possible to define an effective refractive index n_{eff} of the droplet as:

$$n_{eff} = \frac{n_{od}\, n_{ed}}{(n_{od}^2 \sin^2\alpha + n_{ed}^2 \cos^2\alpha)^{1/2}} \tag{1}$$

where α is the incidence angle of the light with respect to the symmetry axis of the droplet while n_{od} and n_{ed} are respectively its ordinary and extraordinary refractive indexes. They depend on the director configuration inside the droplet. In fact n_{ed} can be defined as an average of the local refractive index performed along the **z** axis of the light wavevector inside the droplet :

$$n_{ed} = \frac{1}{l_1} \int_0^{l_1} \frac{n_o n_e}{[n_o^2 \sin^2\beta(z) + n_e^2 \cos^2\beta(z)]^{1/2}}\, dz \tag{2}$$

where $\beta(z)$, being the angle between the liquid crystal director and the light wavevector, depends on the local orientation of the director itself; l_1 is the length of the droplet along the **z** axis. In a similiar way n_{od} can be defined by averaging along a direction perpendicular to **z**.

The light scattering strongly depends[2] on the quantity $n_{eff}^2 - n_p^2$, i.e. if $n_{eff} \gg n_p$ we are in the high scattering and low transmission state; on the other hand, if $n_{eff} \approx n_p$, we get low scattering and high transmission. Therefore,by choosing $n_p \approx n_o$ and by exploiting an electric field alignment of the molecules inside the droplets,[2] it is possible to switch from low transmission to a high transmission state. In fact, in the absence of an applied external field, the optic axes of the droplets are randomly oriented thus producing strong light scattering (OFF STATE). When an electric field is applied, the optic axes of the droplets align themselves along the field direction. If in the same direction a light beam is impinging on the sample, it will be transmitted as far as $n_p \approx n_{od}$ (ON STATE) and the value of n_{od} approaches n_o when the molecules are aligned by the electric field. The light polarization does not affect the onset of the optical switching, but determines the film transparency as well as the angle of incidence does.

The same kind of switching behavior can be obtained by heating the sample, because the refractive index of the liquid crystal in the isotropic state n_i is close to n_o. It is possible to forecast that similiar switching effects could be induced by means of the impinging light itself, by optical reorientation or optical heating of the liquid crystal molecules.

It seems to us that optical reorientation should have quite a high threshold to occur, because of the small size of the droplets and of the anchoring strength of the liquid crystal molecules to the droplets wall. It is more feasible to get a nonlinear optical response from these materials by exploiting thermal effects. In particular even a small light absorption in a liquid crystal can produce a rise of the temperature which is sufficient to cause thermal indexing, that is a thermal gradient of the refractive indexes. As a consequence, these become dependent on the light intensity, thus giving an optical nonlinear response.

In the following paragraph we report experimental data about optical switching obtained in PDLCs originated by thermal nonlinearities.

EXPERIMENTS AND RESULTS

The PDLC samples were obtained by using the polymer induced phase separation method[1] starting from a homogeneous solution of nematic liquid crystal (E7 by Mark), epoxide fluid prepolymer (EPON 165 by

Shell Chemical Company), a polyamminic curing agent as the B component of the Italian Bostik and a small quantity of an orange dye (D2 by British Drug Houses). The dye was added in order to increase the light absorption and even at a microscope observation it appeared to be uniformly distributed. The mixture was sandwiched between two transparent glasses spaced about 30 μm by appropriate Mylar spacers.

The refractive indexes of the used materials were: $n_e = 1.736$, $n_o = 1.525$, $n_p = 1.55$, $n_i = 1.58$. The linearly polarized beam from an Argon Ion laser (λ = 5145 Å) was focused (f = 100 mm) on the sample at normal incidence. The transmitted optical power P_{out} was detected by a photodiode. A beam splitter placed before the sample enabled us to measure the impinging optical power P_i.

A typical experimental result is shown in Figure 1, where the ratio P_{out}/P_i vs P_i is reported.

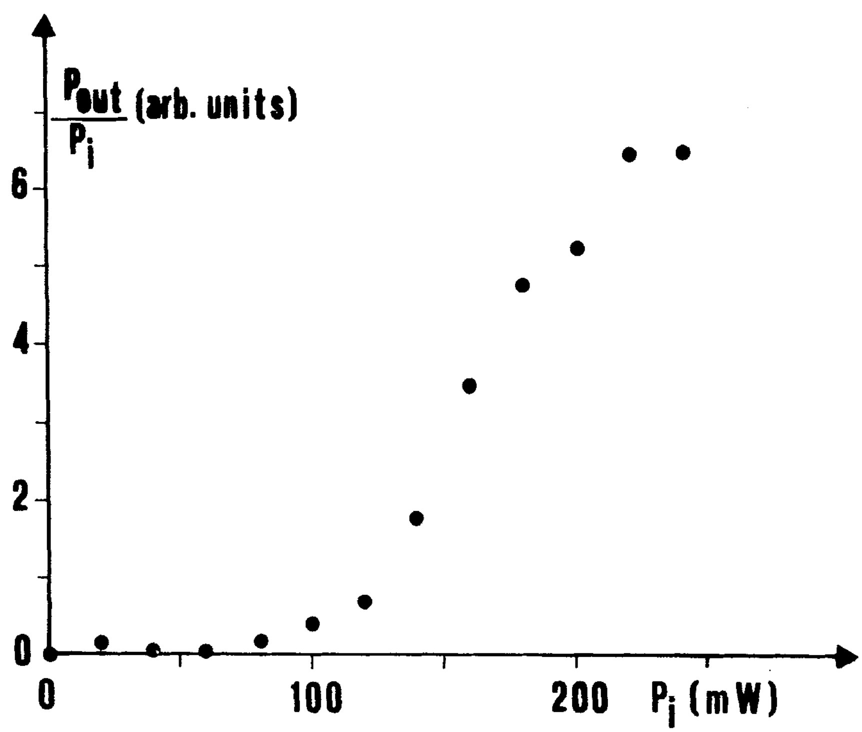

FIGURE 1. The ratio P_{out}/P_i vs P_i in arbitrary units.

A clear switching effect is observed: this behavior is completely reversible as for the case of the electrically induced switching. The effect is easily observable by naked eyes: in the OFF state one can see a typical pattern of weak diffused light after the sample, while in the ON state a clear beam is observed even if some scattered light is still present.

The effect is easily explained by the following model: increasing the incident optical power, the temperature of the sample increases due to strong light absorption. When the droplets temperature reaches the value which corresponds to the phase transition to the isotropic state, the refractive index of the droplets n_{eff} reduces to n_i and therefore $\Delta = n_{eff} - n_p$ becomes very small, thus lowering the scattered light.

The thermal origin of the effect is confirmed by its time response. By chopping the laser beam before the sample and using an impinging power P_i which induces the transition to the ON state, we can measure the correspondent rise time t_r of the signal S on the detector as shown in Figure 2: we find $t_r \approx 20$ msec. which is in the range of thermal effects.

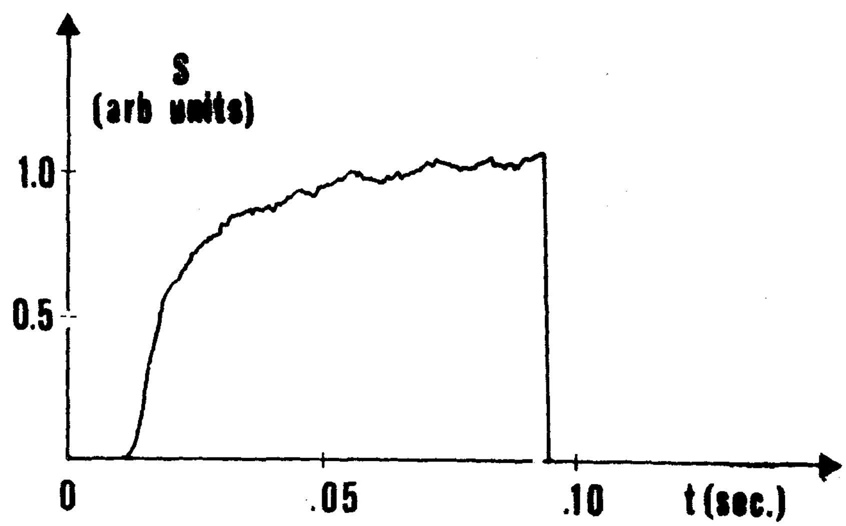

FIGURE 2. The signal S vs time when chopped light is used.

An interesting feature is observed when increasing the power under the same experimental conditions. A faster response is recorded which is evident in the spike at the beginning of the square shaped signal (Figure 3). The spike has a rise time $t_s \approx 1$ msec. The origin of the spike has been clarified by looking at the beam pattern on a screen after the sample. Right after the beam appears, a first ring is created around it due to the self induced phase modulation. The growth of the light ring corresponds to the decreasing tale of the spike. Then the ring slowly ($t \approx 20$ msec.) disappears and the signal reaches its steady state value. This effect can be due to thermal relaxation which causes uniform heating of the sample on the laser spot while, at the beginning, one gets a temperature distribution which is similiar to the light intensity distribution on the sample.

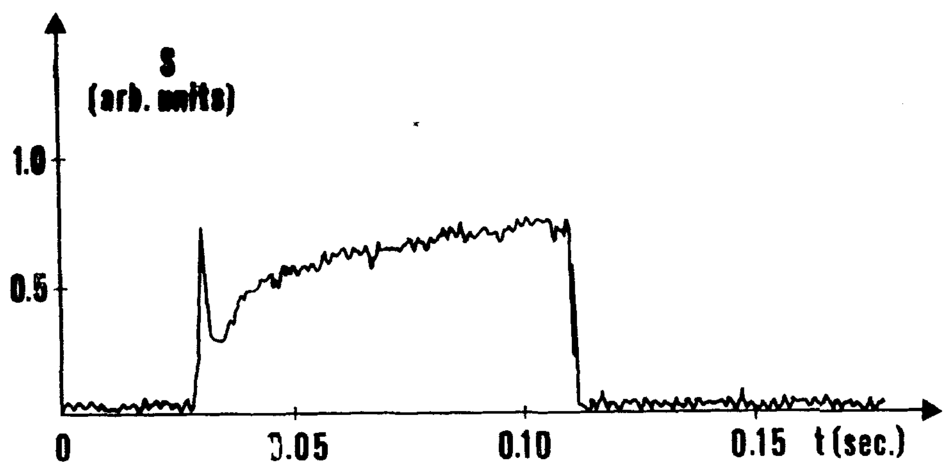

FIGURE 3. Same as Figure 2, for higher impinging power.

In conclusion we have reported the observation of optical nonlinearities of PDLC samples induced by thermal effects, using low power c.w. laser. The typical features of this phenomenon have been discussed.

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