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Optical switching has been studied in epoxy-based polymer dispersed liquid crystal films. A 'self-transparency' effect is observed due to refractive index variations thermally induced either by heating the sample with a hot stage, or by absorption of the incident laser power. The switching phenomena were studied for different liquid crystal contents in the composites, and experimental evidence of light modulation by a probe beam is also reported. The thermo-optical behaviour of the material is correlated with the morphology of the samples. The results obtained verify the possibility of employing this type of material in optical devices based on the thermo-optical switching effect.

1. Introduction

Modulation of a cw laser beam by another light beam depends fundamentally on the optical properties of suitable materials employed in properly designed devices in order to enhance and exploit this effect. Research efforts to modulate light by light have been greatly stimulated by applications in optical computing and optical telecommunications [1–3]. Liquid crystals (LCs) and their related technology have attracted great interest during recent decades due to their unique optical properties which make them useful for the realization of fast response and low cost devices. Polymer dispersed liquid crystals (PDLCs) play an important role in LC technology applications because they combine the advantages of polymers with the electro-optical response of liquid crystals [4]. Basically, what makes them more attractive compared to other existing technologies is their ease of preparation; no need for alignment layers and no need of polarizers, resulting in a higher transmission in the transparent state. These unique properties compared with other existing LC techniques offer great potential in applications of electro-optical modulation, LC display techniques, and shutters for infrared cameras [5–9].

A PDLC film can be prepared by phase separation, LC emulsification or LC impregnation [10–12], where LCs are dispersed in a polymer matrix as micro-sized droplets. The morphology will influence the electro-optical and thermo-optical properties of the films as well as the final device performance.

The morphology of the PDLC sample depends on both the preparation technique used and the LC content in the matrix, and it can be controlled and optimized by modifying parameters during the sample preparation. Two major types of phase separated morphologies have been observed which depend on the amount of LC employed: the 'swiss cheese' morphology and the 'sponge-like' morphology. At higher concentrations of LC (up to 40 wt %) the 'swiss cheese' morphology is dominant; it consists of spherical or ellipsoidal LC droplets surrounded by the polymeric matrix; at higher concentrations of LC the 'sponge-like' morphology prevails where there are two continuous phases, a liquid crystal phase and a polymeric one.

In phase separation preparations, several techniques are employed: polymerization-induced phase separation (PIPS), solvent-induced phase separation (SIPS) and thermally-induced phase separation (TIPS). In the PIPS method, the polymerization occurs either thermally or photochemically. Phase separation occurs by droplet nucleation and growth. The droplets continue to grow until polymer gelation locks in the droplets' morphology. Either thermal or photochemical curing affects the kinetics of phase separation, thus influencing the droplets' size and shape.

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One of the most attractive properties of PDLC films is their capability to be switched from an opaque state to a transparent one either electrically or thermally [13].

The PDLC film can be electrically switched from the 'off-state' to an 'on-state' which has a high degree of transparency by applying an electric field in the case of a LC with a positive dielectric anisotropy. Due to the random distribution of LC directors in the droplets which effects the mismatch of refractive indices between the LC droplets and the polymer matrix, the incident light is strongly scattered by the film which appears opaque (off-state). When an electric field is applied, the LC directors will be aligned along the electric field direction to make the film transparent as a result of index matching between the aligned LC and the polymer matrix. The degree of transparency is determined by optimized index matching between the LC droplets and the polymer matrix.

A similar switching effect can also be achieved when the LC inside the droplets undergoes a change from the nematic phase to the isotropic state. This phase transition can be thermally induced by directly heating the sample or by exploiting the non-linear thermal properties of LCs. In the latter case, the incident light induces thermal heating leading to the phase transition of the LC droplets to the isotropic state due to the absorption of the power of the incident light. The optically induced change of the refractive index of the material acts as a feedback system to modulate the input light. This effect is the basis of many promising applications in light modulation and temperature sensing.

Here we present a detailed experimental study of light modulation in epoxy-based PDLC films, switching a weak He-Ne laser beam by an Ar laser pump beam. The influence of the LC concentration in the polymer matrix on the properties of the PDLC films is investigated. We demonstrate how the thermal- and optical properties of our samples are strongly affected by the LC concentration.

2. Experimental details

PDLC samples were prepared by phase separation of the LC component from a solution containing epoxy prepolymer (PIPS method). Epoxy prepolymer, the diglycidyl ether of bisphenol A (DGEBA), (Epikote 828 from Shell Italia S.p.A.) was mixed at 70°C with the liquid crystal component, the commercially available material E7 (a eutectic mixture of four liquid crystals components), to form a homogenous solution. 1 wt % of initiator, 2,4,6-tris(dimethylaminomethyl)phenol (DMP-30), and an appropriate hardener, methyl-5-norbornene-2,3-dicarboxylic anhydride (MNA) were added to the solution. The hardener and the initiator (both supplied by Aldrich) were used without further purification. The resulting solution was deposited on a glass substrate, then baked in an oven up to 130°C at an appropriate heating rate, followed by cooling. By this method, thin films ($\sim$55 µm thick) with 35, 40 and 50 wt % concentrations of E7 and good phase separation were obtained. The nematic–isotropic transition temperature of the E7 was at 58°C, as measured by differential scanning calorimetry (DSC).

The experimental set-up is shown in figure 1. Samples were heated inside a programmable oven (FP900 Thermosystem from Mettler-Toledo) which allows temperature control to $\pm$0.1°C and heating rates as low as 0.1°C min^{-1}. The actual sample temperature T_s (background temperature) inside the cell was measured and monitored using a thermocouple connected to a data-acquisition system. The output light from the sample ($\lambda = 632.8$ nm from a He-Ne laser) was detected as a function of the temperature at a heating rate of 4°C min^{-1}. A similar measurement was performed using a cw Ar laser beam ($\lambda = 514$ nm) as pump beam, incident normally on the sample, and a He-Ne laser used to probe the thermal effect induced by the pump beam. The amplitude of the input pump beam was modulated by a Pockels cell driven by a function generator. As a
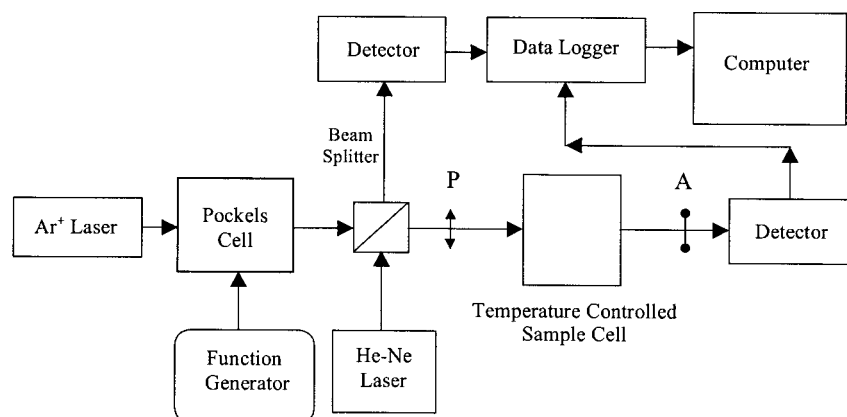

Figure 1. Experimental set-up; P: Polarizer, A: Aperture.

consequence, temperature variations of the sample were obtained by increasing and decreasing the input light power ranging from 0 to 300 mW, driven by a triangular waveform at a frequency of 0.01 Hz. An interference filter was employed to block the 514 nm light and detect only the He-Ne output light. The pump beam was continuously monitored with respect to the reference beam. Light powers of both He-Ne and Ar lasers were calibrated with a Coherent LM-E power meter. The optical response to the thermal effect induced by the pump beam with a square driving waveform, was measured by a Tektronix TDS 784D digital phosphor oscilloscope. In order to get a true measure of the on-state transmittivity (avoiding the collection of light scattered out of the specular beam), an aperture (1.5 mm in diameter) was placed behind the sample cell to generate a small collecting angle of 2.5°) [14].

3. Results and discussion

Experiments were initially carried out to measure the temperature dependence of the transmitted light from the epoxy-based PDLC samples. The samples with several concentrations of E7 (35, 40 and 50 wt %), heated by the programmable oven, were illuminated by a He-Ne beam (<1 mW) at normal incidence. A clear switching effect was observed as the temperature was increased in the range from 35–75°C as shown in figure 2. The samples exhibit an 'off-state' with a lower transmission below the transition temperature T_{th} (around 60°C). Their transmission increases gradually, and the transition to the 'on-state' is quite sharp within a narrow temperature range (~ 5°C) when T approaches T_{th}. The observed switching effect is completely reversible after

cooling the samples and then re-heating them. It is worth noting that the changes in the transition temperatures of the PDLCs (see figure 2) is generally attributed to either contamination of the LC or to a non-homogeneous phase separation of the four different components of E7.

This transition is due to the phase changing in the LC droplets from the nematic state to the isotropic state upon heating. Since the isotropic refractive index of the LC is closer to that of the polymer matrix than its nematic refractive index, a better index matching condition is reached at the transition temperature T_{th} (nematic–isotropic phase transition), leading to a huge reduction of scattering and an abrupt increase in transmission (on-state) [15, 16].

The measured T_{th} of the samples at different concentrations are very close to that of E7 liquid crystal. This indicates that most of the dispersed liquid crystal component is separated from the polymer matrix. This is confirmed by scanning electron microscopy (SEM) images of the PDLC as shown in figure 3 (*a–c*), where samples with 35, 40 and 50 wt % concentrations, respectively, are presented. From figures 3 (*a*) and 3 (*b*) we can see that the samples are clearly phase-separated, the LC component forming sphere-like droplets uniformly distributed in the epoxy matrix. It is apparent from figure 3 (*c*) that at a concentration of 50 wt %, the polymer forms a continuous phase with a sponge-like structure, and the LC domains, which are more or less curved and stratified, are continuously embedded in the polymer matrix.

Employing software-based image analysis [17], the distribution of average diameters of the LC droplets appears broad, as shown in figure 4, ranging from 0.5 to 3.5 μm for 40% and from 0.3 to 1.0 μm for 35% concentration. However, the main size of the LC droplets (exceeding 67% of the droplets) for 40 wt % concentration lies between 1.5 and 2.5 μm, and over 70% for the 35 wt % concentration lie between 0.4 and 0.85 μm. The SEM image for the 50 wt % concentration is not clear enough for morphological analysis. However, observing the samples by polarizing optical microscopy, the characteristic birefringence of the droplets disappears when the sample temperature is above T_{th} and the LC enters the isotropic phase.

The existence of a transition temperature suggests the possibility of obtaining an optical modulation based on the nematic to isotropic phase transition of the PDLC samples. This possibility was explored in a pump and probe geometry, with the pump beam power ranging from 0 to 300 mW, and changing at a rate of 4 mW s^{-1}. By modulating the pump beam, at a fixed temperature around the transition temperature T_{th}, the probe beam is modulated. Figures 5 (*a*) and 5 (*b*) depict the results
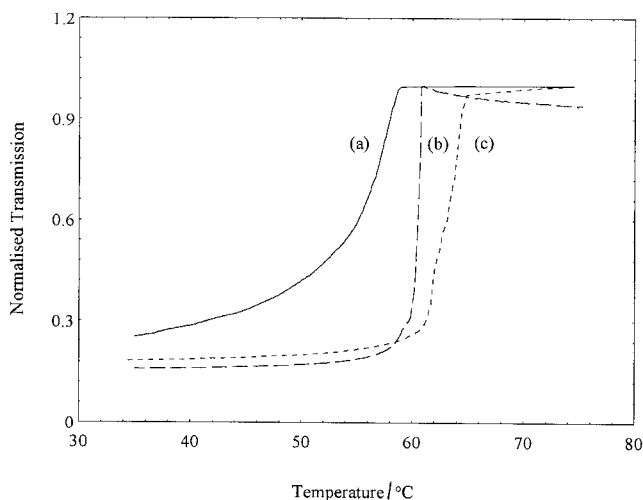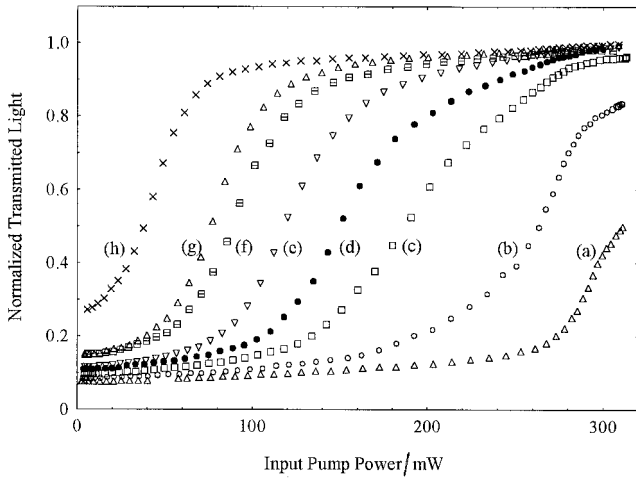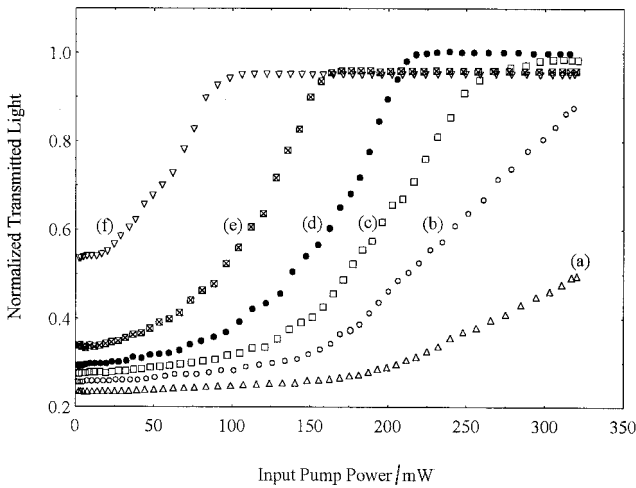

Figure 2. Temperature dependence of light transmission at 632.8 nm wavelength for samples with different concentrations of E7: (*a*) LC content = 35 wt %, (*b*) LC content = 50 wt %, (*c*) LC content = 40 wt %.

(*a*)

(*b*)

(*c*)

Figure 3. SEM images of phase-separated PDLC samples. (*a*) 35 wt % concentration, (*b*) 40 wt % concentration, (*c*) 50 wt % concentration.

Figure 4. Distributions of LC droplets sizes at (*a*) 35 wt % and (*b*) 40 wt % concentration.

obtained with the 40 and 50 wt % concentration, respectively. Only a very weak modulation is obtained from the sample with 35 wt % concentration, though the sample has a good phase separation. We can see from figures 5(*a*) and 5(*b*) that the largest switching effect can be achieved with an optimized temperature under the same experimental conditions; the corresponding contrast ratios (90 to 10% transmission value) are 7.1 for the 40 wt % and 3.4 for the 50 wt % concentration, respectively. It is evident from figure 6 that the threshold power of the pump beam to induce the switching effect decreases linearly with a corresponding increase of the background temperature.

Further measurements were carried out by changing the modulation frequency at 59°C (below the transition temperature for the 50 wt % concentration). The pump beam was modulated with a triangular waveform as depicted in figure 7. A distinct hysteresis is observed where the loop becomes larger with increasing modulation frequency at a fixed background temperature. Due to the absorption process, which depends on the incident
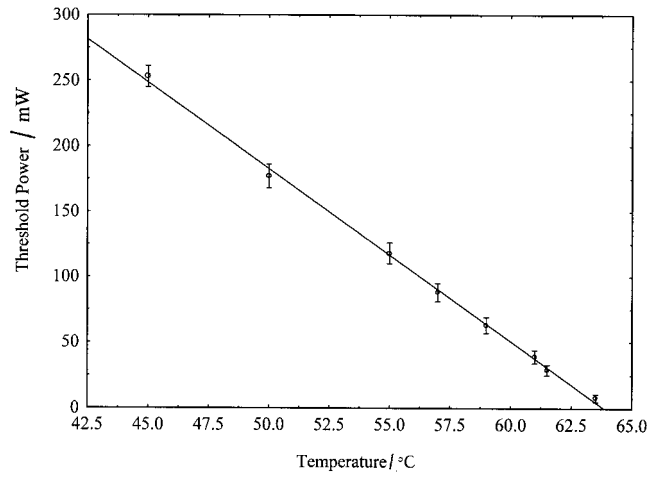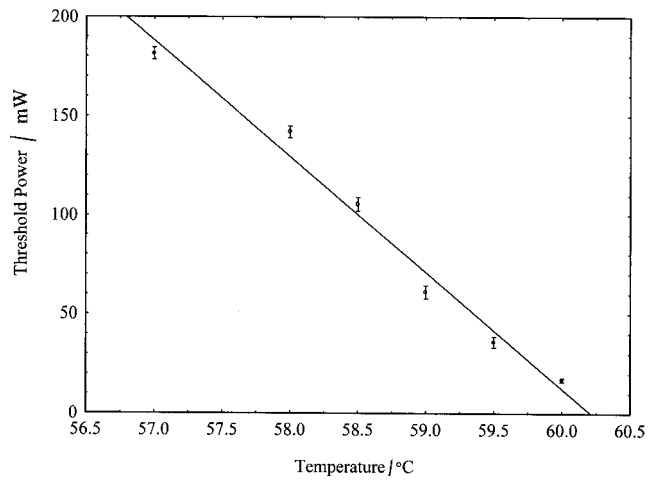
(a)

(b)

Figure 5. Experimental results on light transmission (probe beam) as a function of the incident modulation power (pump beam) at different background temperatures of the samples: (*a*) sample at 40 wt % (a) 45°C, (b) 50°C, (c) 55°C, (d) 57°C, (e) 59°C, (f) 61°C, (g) 61.5°C, (h) 63.5°C. (*b*) sample at 50 wt % (a) 57°C, (b) 58°C, (c) 58.5°C, (d) 59°C, (e) 59.5°C, (f) 60°C.

(a)

(b)

Figure 6. Threshold power changes as a function of temperature: values obtained from figure 5. (*a*) 40 wt % concentration, $y = -13.2x + 842$; (*b*) 50 wt % concentration, $y = -58.5x + 3524$.

light power, on the modulation frequency, and of course also on the heat capacity of the materials, the hysteresis finds an explanation in the pumping dynamics. Once the laser power is high enough to switch on the sample thermally, the sample is nearly transparent and can mantain this state for a short time before reversing back if the temperature of the sample declines. Since a higher laser power is needed to turn the sample on at a higher modulating frequency (on the way up), when the laser power is quickly reduced (on the way down) from its maximum, the short on-state allows the sample to main-

tain a comparatively high transmission until a considerable decrease in the laser power occurs. As a result, a larger hysteresis loop is achieved at a higher modulating frequency. Conversely, a similar reason leads to a smaller hysteresis at a lower modulating frequency. This effect was also investigated for the 40 wt % concentration (for details see [18]).

The temporal response of the probe beam on the input pump beam (square waveform) at different modulating frequencies was studied in detail. The corresponding input and output waveforms at a background temperature of 59°C and a frequency of 0.2 Hz are shown in figure 8. A rise time of 390 ms is obtained for the 50 wt % concentration at 59°C and 0.2 Hz. Similarly, for the 40 wt % liquid crystal content the shortest rise time (140 ms) is measured when the sample exhibits its best
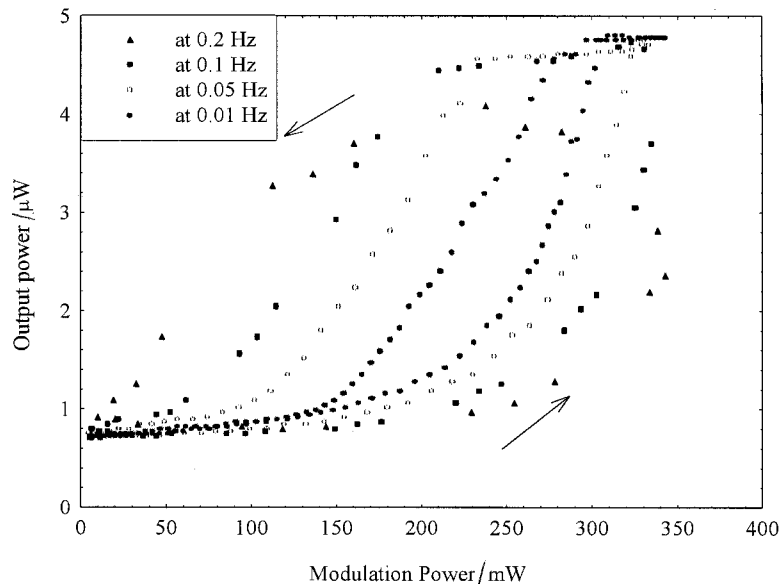
Figure 7. Light transmission (probe beam) as a function of the input power (pump beam, triangular waveform) at different modulating frequencies (0.01 to 0.2 Hz) at a fixed background temperature (59°C).
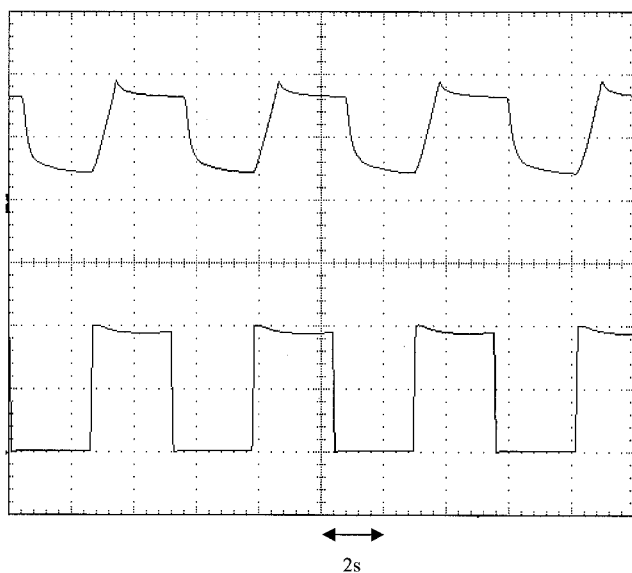

Figure 8. The characteristics of the input (lower curve, square waveform) and output (upper curve) waveforms for 50 wt % concentration, at a background temperature of 59°C, and at a modulating frequency of 0.2 Hz. The time scale is 2 s cm^{-1}.

of the sample (refractive index, birefringence, order parameter, viscosity and elastic constants, etc.) and the LC isotropizes. As a result, switching occurs at the phase transition, and light modulation is achieved.

According to our experimental results, the sample with 40 wt % concentration exhibits the best light modulation effect, while in the case of a 35 wt % concentration, only weak switching is found even if the sample morphology shows a good phase separation. This suggests that there exists a critical concentration for obtaining an optically-induced transition. For the 50 wt % concentration, reasonable light modulation is obtained. However, good phase separation is very hard to achieve at this concentration and above, which is perhaps the reason why light modulation at this concentration is poorer than that with the 40 wt % concentration.

Experimental data show that better light modulation is observed near the isotropic state of the samples, e.g. at 61.5°C (background temperature) for the 40 wt % concentration and at 59°C for the 50 wt % concentration. At the same time, since the background temperature preheats the whole sample and reduces the index difference between the liquid crystal droplets and the polymer matrix, a lower power of the incident light is sufficient to induce the transition. This results in the shift to low values of the threshold power. At 40 wt % concentration, the shift in the threshold values shows a very good linear dependence with the increase in the background temperature (figures 5 and 6). Furthermore, the actual local temperature of the samples can be varied by changing the frequency of the pump beam [19]. Our data show (figure 7) that to induce switching more power is required for the input laser beam at higher frequencies, because the local temperature rise is lower than with a

switching effect at 61.5°C and 0.2 Hz. The rise time decreases on increasing the temperature or on reducing the driving frequency.

In order to discuss properly the thermo-optical behaviour of the PDLC samples investigated, it seems important to highlight the thermal phenomena occurring during the laser–material interaction. The temperature rise in the local area of the samples is due to the absorbed light power. The temperature increase in the local area leads to a change in several parameters

lower frequency. Therefore, a larger hysteresis loop is achieved when the power of the input pump is increased linearly at higher frequencies.

4. Conclusions

The optical properties of epoxy-based PDLC films have been studied, and thermally and optically induced changes by both heating and laser irradiation have been explored. Optically induced light modulation based on refractive index change near the threshold temperature for the nematic to isotropic phase transition has been demonstrated. Results show that the light modulation effect depends strongly on the LC concentration in the sample system; 60/40 wt% (Epoxy/E7) is the optimized ratio for this PDLC composition to achieve the best light modulation. Increasing and reducing the LC component in the system will influence the energy exchange between the pump beam and the samples, and will also alter the morphology of the samples, which in turn affects their light modulation properties.

Our experiments demonstrate that the optical switching effect can also be obtained by changing the frequency of the pump beam. A large thermally induced hysteresis loop is obtained at a frequency of 0.2 Hz near the isotropic state (61.5°C) for the 40 wt% LC concentration. In this situation we measured the corresponding rise time to be 140 ms. According to these results, we believe that an optimal light modulation can be achieved by choosing an appropriate combination of parameters (background temperature, input light power and frequency). Furthermore, a different background temperature gives rise to a linear shift of the threshold power for light modulation. In conclusion, our experimental results show that the material studied is promising for use as the active element in thermo-optical devices. The possibility of modulating the incident light all-optically makes such a material very suitable for controlling the power levels inside an optical system.

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