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THERMAL DIFFRACTIVE PROPERTIES OF TRANSMISSION GRATINGS RECORDED IN NEAR INFRARED SENSITIVE LIQUID CRYSTAL PHOTOPOLYMERIZABLE MATERIALS

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We present thermal features of holographic gratings recorded in near infrared sensitive polymer dispersed liquid crystal films. Experimental results indicate that the diffraction efficiency of our gratings can be smoothly controlled by monitoring the sample's temperature. Diffraction efficiencies versus temperature for both s- and p-polarized probe and transmitted probe's polarization are studied and qualitative explanations are provided.

INTRODUCTION

Photopolymer materials have significant advantages making them suitable for applications as optical interconnection [1], optical data storage [2], holographic interferometry [3], acoustic wave detection [4], etc. Polymer dispersed liquid crystal (PDLC) materials have been shown [5,6] to be very promising for the electrical control of light in displays applications.

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More recently the holographic fabrication of gratings that use liquid crystal-polymer spatially periodic dispersions has been reported [7–16]. The holographic PDLC's (H-PDLC's) thus formed, although permanent, can be electrically switchable if the liquid crystal (LC) content exceeds a certain weight percentage (for example, the LC content is 30 wt.% in Ref. [10]). The resulting system is usually divided into polymer-rich and LC-rich regions, thanks to the mixible LC-monomer and unmixible LC-polymer compounds chosen. The bright zones have higher polymer density and the effective refractive index there is near to the polymer refractive index. The LC density is higher in the dark zones, the effective refractive index there being thus determined mainly by the LC orientation. In fact the molecular orientational distribution in each of the LC droplet is controlled by elastic energy considerations associated with the anchoring conditions at the droplet walls, the curvature constraint of the droplet geometry and droplet size. When a suitable external voltage is applied to these gratings, the LCs became reoriented, causing a matching of the refractive index of the polymer and the LCs. As a result, the diffraction intensity declines abruptly.

In this paper we describe an alternative way of controlling the diffraction of holographic gratings recorded in near infrared sensitive liquid crystal photopolymerizable materials. Our experimental results indicate that the diffraction efficiency of the grating can increase significantly (from 30–40% up to 95%) by increasing the temperature of the sample up to the phase transition temperature of the LC. It means that this composition can be used to create as a switchable grating. This work will describe such experiments along with corresponding qualitative explanations.

EXPERIMENTAL

A photopolymerizable solution was used, which consisted (see details in Ref. [14]) of monomer DPEPA (Di-Penta-Erithriol-Penta-Acrylate), near infrared dye with a maximum of absorption at 818 nm, electron donor, initiator EDMAB_{zt} (Ethyl-di Methyl-Amino-Benzoate), nematic liquid crystal (NLC) E7 (with refractive indexes for ordinary and extraordinary waves, respectively $n_{LC}^o = 1.5183$, $n_{LC}^e = 1.7371$ at $\lambda = 633$ nm) and a second monomer 2EEEA (2-Ethoxy- Ethoxy-Ethyl Acrylate Ester). The homogeneously mixed LC-monomer compounds were sandwiched between two microscope glass slides (25 mm \times 25 mm \times 1 mm), separated by a $d = 25$ μ m thick plastic spacer. A CW Ti-Sa laser (operating at 823 nm) was used as a recording light source providing a beam polarized perpendicular to the plane of incidence (s-waves). The experimental setup was a typical holographic one, the same as in [17]. The two beams were

overlapped at symmetric incidence to interfere on the photopolymer film at a cross-angle of 48.6° , which results in a sinusoidal pattern with a spacing of $\Lambda = 1 \mu\text{m}$, previously shown to be the optimal one for this composition and intensity range [17].

The diffraction efficiency ($I_1/I_0 + I_1$) was measured using a He-Ne laser (operating at $\lambda = 633 \text{ nm}$) impinging at Bragg angle ($\alpha = 18.3^\circ$). The sample in the completely polymerized state was placed in a temperature chamber to test its thermal behavior. The intensities of the diffracted and transmitted beams were measured to determine the holographic diffraction efficiency. Note that for the used geometrical parameters (d, Λ, λ) we have a hologram geometric factor $Q = 2\pi\lambda d(\Lambda^2 n_0) = 63$, where $n_0 = 1.6$ is the estimated average refractive index of the grating. Still there is another physical factor, $\rho = 2\lambda^2/(\Lambda^2 n_0 n_1)$ (where $n_1 = n_{LC} - n_{pol}$ is the modulation of the refractive index and $n_{pol} \approx 1.5$ is the polymer refractive index), that must also be considered to define the hologram to be “thick” (Bragg regime) or “thin” (Raman-Nath regime). In our case we estimate $\rho = 5$ for the p-polarization and $\rho = 16.5$ for the s-polarization. An exact definition of the thick grating has been given in [18] and the condition to be fulfilled is $\rho > 20$ for $Q = 63$. Thus we are in a diffraction regime, between Bragg and Raman-Nath regimes, closer to the Bragg regime. This is confirmed by the fact that we observe only one diffracted beam near to the Bragg angle.

RESULTS AND DISCUSSION

Figure 1 shows the temperature induced variations in the diffraction efficiencies for the s- and p-polarized probe beams. Both diffraction efficiencies return to their initial value when decreasing the temperature and the process could be repeated without any change in behavior. We do not observe any hysteresis phenomena neither. For the s-polarized probe beam, a strong increase of diffraction starts from temperatures above 40°C and reaches its maximum at 60°C . At the meantime, the diffraction efficiency for the p-polarized probe first increases with the temperature and then drops abruptly near the 55°C . Both diffractions decrease monotonically above 60°C . We can infer that such an effect is attributed mainly to the temperature dependence of LC refractive indexes for ordinary and extraordinary waves. Indeed, as we know from Kogelnik’s theory [19], the diffraction efficiency of transmission gratings changes in an oscillatory way with refractive index modulation depth n_1 (we shall call *positive* (negative) the region where this oscillatory curve has a positive (negative) slope). This means that a decrease in modulation depth does not necessarily imply a loss in diffraction efficiency.

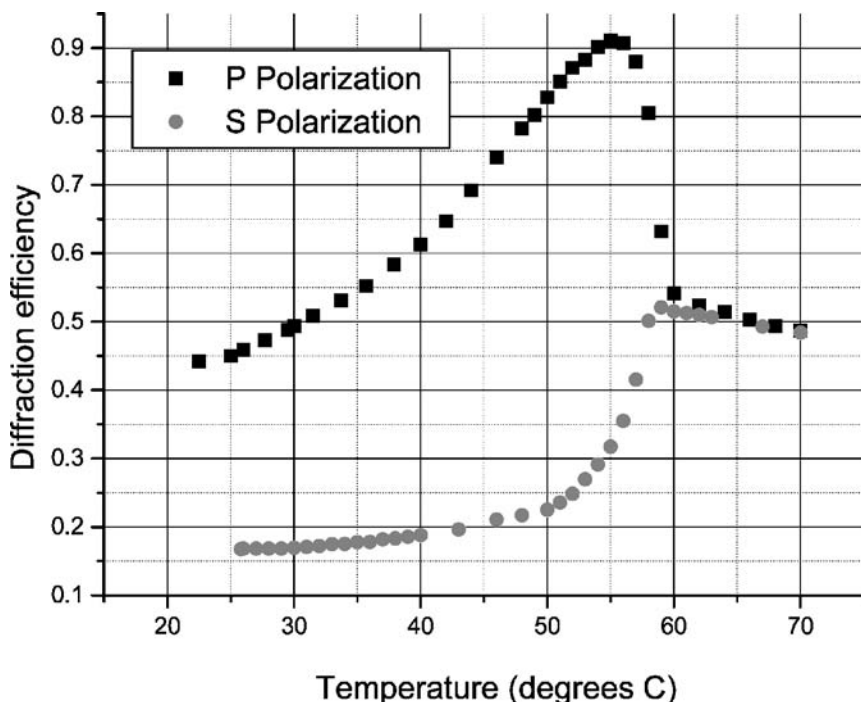


FIGURE 1 Temperature dependence of diffraction efficiency for one micron period grating for s- and p-polarized probe beam. Abs Error: 0.01; $\lambda_{\text{read}} = 633 \text{ nm}$, sample thickness $d = 25 \mu\text{m}$ $\Lambda = 0.94 \mu\text{m}$.

Previous studies on the polarization properties for these gratings [20] showed that during recording (thus increasing modulation depth) the diffraction efficiency of the p-polarized probe increases up to a maximum of 90% and then decreases to a stable final value. At the same time, the s-polarized probe's diffraction grows and stabilizes without such oscillations.

To understand the thermal behavior of our gratings we need first to determine whether the selected polarization of the probe yields a diffraction efficiency value that is in the positive or negative slope region. Another key factor is the question if the refractive indexes (n_o and n_e) of LC for ordinary and extraordinary waves will become closer or farther from the refractive index (n_p) of the polymer with increasing the temperature. The grating diffraction efficiency will then increase or decrease as a resulting combination of these two conditions.

The temperature dependence of the LC refractive index for s-polarized wave is usually slow far from the LC-isotropic phase transition

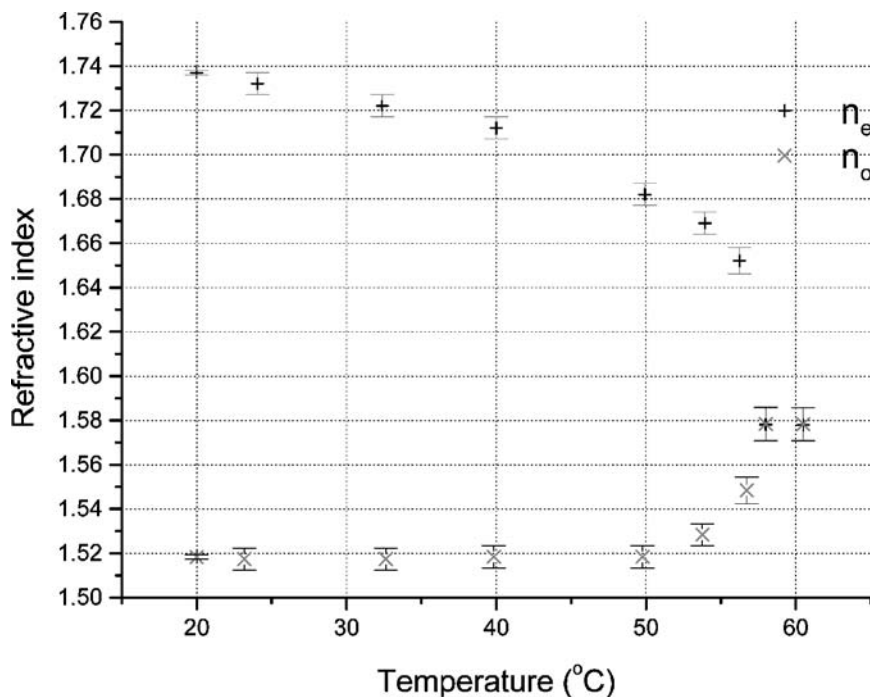


FIGURE 2 Temperature dependence of ordinary and extraordinary refractive indices for LC E7 at 633 nm.

(see Fig. 2). Therefore the s-polarized wave diffraction efficiency varies very little until it approaches the phase transition temperature. Meanwhile, the p-polarized wave diffraction efficiency reaches its maximum at $\sim 55^\circ\text{C}$ and then decreases until it coincides with the final value for the s-polarized probe. According to our previously mentioned polarization study, we believe that the s-polarized diffraction efficiency is still in the positive zone of the oscillatory curve. Considering the general thermal behavior of the ordinary refractive index for our LC [from results of the group of I.C. Khoo] and the initial values given earlier, the modulation depth will increase with temperature resulting in an increasing diffraction efficiency. The p-polarized probe's diffraction efficiency is in the first negative region due to its very large modulation depth. The extraordinary refractive index of the LC decreases with the temperature increase, thus getting closer to that of the polymer and reducing the modulation depth. The corresponding diffraction of the probe can then increase until its maximum and continue decreasing in the positive

region to reach its final state. A qualitative schematic of this result is given on Figure 3.

Although our preliminary theoretical calculations (taking into account the slopes of temperature dependence of n_o and n_e) show that this mechanism alone could not explain the obtained experimental results, the temperature dependence of the LC refractive indexes leads to the general observed behavior. There must then be other mechanisms at work to complement this effect in order to reach a good theoretical fit of diffraction efficiency. One of these mechanisms could be the formation of a reversible density grating caused by the polymer and LC expansion dependence upon temperature. It is also known that the change in temperature may affect the anchoring strength between the polymer and the LC droplets resulting in a change of director orientation [Amundson, Phys Rev E, 1996]. To clear up this question of possible LC orientational changes, we then measured the ellipticity (see Fig. 4) of the diffracted beam for various angles of the incident probe's polarization. This curve shows that no ellipticity is present

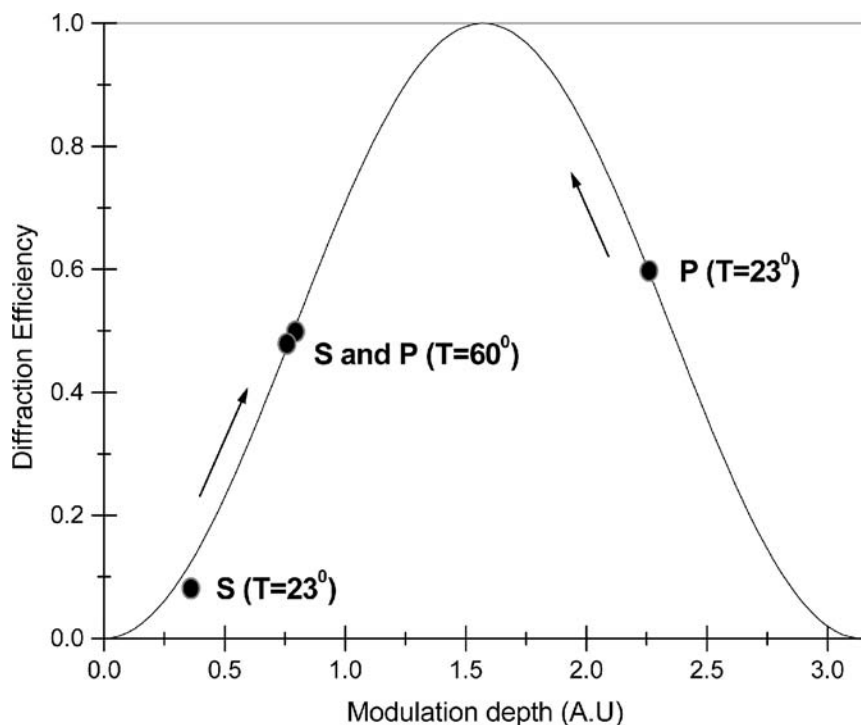


FIGURE 3 Qualitative representation of the diffraction efficiency thermal behaviour in term of modulation depth.

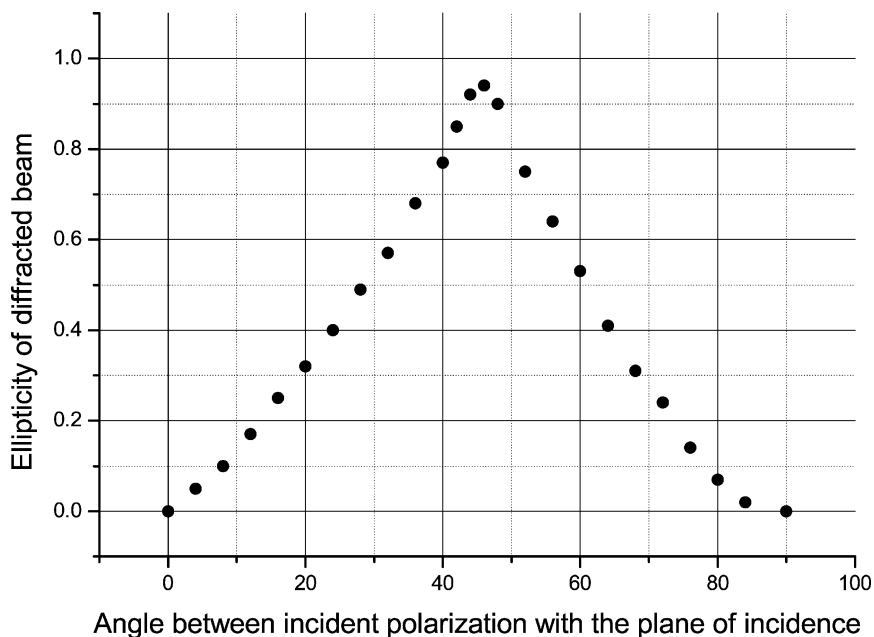


FIGURE 4 Ellipticity of the diffracted beam (measured as the minor axis divided by the major axis) depending on the angle (in degrees) between the linear polarization of the incident probe beam and the plane of incidence at the temperature 23°C. Abs. Err: 0.01

for pure s- or p-polarized incident probes. This means that the director remains either perpendicular or parallel to the incident polarization. This result was also found to be temperature independent.

CONCLUSION

Important temperature induced variations of diffraction efficiencies for s- and p-polarized light were measured. Furthermore, the magnitude of the thermal effect for p-polarized measurements indicates that for different initial conditions (reading wavelength, thickness) the diffraction efficiency could be tuned from zero to its maximum. Further measurements showed that there was no significant change in director orientation since the s- or p-polarizations of the probe were preserved. Finally, measurement of long-term reliability indicates that these gratings do not age for at least one year and that they therefore have possible practical uses.

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