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Improvement of Electro-Optical Properties of HPDLC Gratings by *in situ* Shearing During Holographic Recording

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We report the results of the recording of holographic polymer-dispersed liquid crystal (HPDLC) reflection gratings while applying a shear stress parallel to the film plane. The shear is transmitted through the film by moving one glass window with respect to a fixed glass window during the holographic recording in a single beam, total internal reflection geometry. The optical properties of the resulting Bragg grating are related to timing and magnitude of the stress. High diffraction efficiency for light polarized in a direction parallel to the stress is obtained with nearly zero diffraction efficiency (DE) for the perpendicular polarization. Contrary to post-recording stress-induced polarization sensitization, the in situ *process results in permanently polarized gratings. The polarization sensitivity is related to stress-induced morphology changes of liquid crystal droplets that are frozen at gelation of the thio-ene polymerization process.*

Keywords: Bragg grating; droplet alignment control; holography; polymer dispersed liquid crystal; shear; thiol-ene polymers

INTRODUCTION

The number of research groups working in HPDLC's has been increasing as the promise of holographic optical elements with electrically

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controlled diffraction efficiency using this material moves toward a reality. Indeed several applications have already been demonstrated including wearable displays [1–3], active matrix displays [4,5], video projectors [6], and fiber optical switches [7,8]. Much of the successful work toward achieving optical elements with high diffraction efficiencies (near 99%) in HPDLC's has used transmission holography to form simple holographic gratings or lenses with structures no smaller than 300 nm [9,10]. The lower size limit arises from the process which makes the HPDLC's possible, that is the complicated interaction and diffusion between monomer, liquid crystal (LC) and the polymerization wavefront during holographic exposure. At smaller periods (<300 nm) this process limits the LC volume fraction as the LC planes transitions from a near laminar structure to a plane of randomly dispersed LC droplets. Optical performance thus suffers as scatter increases off the LC droplets and a reduction in LC volume fraction lowers the index modulation [11]. This same process also limits the optical performance achievable in reflection type HPDLC gratings in the visible region which require structure sizes in the range of 220 nm (red) to 160 nm (violet), with the highest diffraction efficiency reported to date of 80% [12].

In this paper a method is presented which allows some of the limitations of the formation process to be overcome by more effectively using the birefringence of the LC through a mechanical shearing of the HPDLC film during the holographic exposure process. The incorporation of a shearing step into the exposure process has some clear advantages to the post mechanical shearing process used in HPDLC's [13,14] in the past. The technique is applied to non-slanted visible reflection grating resulting in gratings with polarization dependent DE's of $\sim$99% and reduced scatter while maintaining the electric control of the filter's DE.

EXPERIMENT METHOD

Sample cells were constructed by sandwiching a drop of syrup between two glass or ITO coated glass substrates as described previously [13]. The syrups were thiol-ene based formulations [14] with concentrations of BL037 LC ranging from 28% to 32% by weight. Glass spacer rods of 5–15 microns are mixed into the syrup to control the thickness of the sample cell. The cells were assembled inside the shear device shown in Figure 1. A cw argon ion laser operating in single-line 365 nm mode with an etalon provided the coherent light source for holographic exposure. The laser beam was expanded and collimated to give an intensity of $\sim 35\,\mathrm{mW/cm^2}$. It was directed through a right angle prism

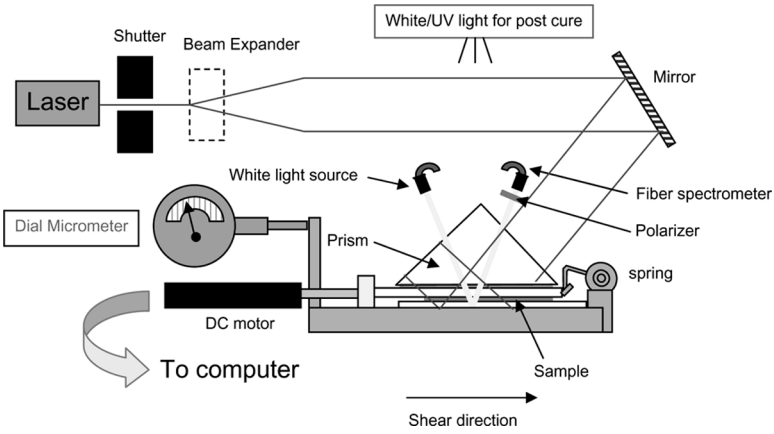
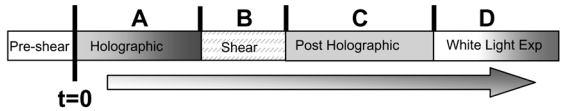


FIGURE 1 Diagram of the shear apparatus. The shutter, DC servo motor, and fiber optical spectrometer are computer-controlled. The dial micrometer gives an accurate measure of the window displacement.

at a steep enough angle to provide TIR off the rear surface of the sample. The sample was mounted with index matching fluid onto the hypotenuse of the prism. A white-light source polarized in the shear direction incident at a near normal shallow angle to avoid TIR was used to monitor the diffraction from the grating during the holographic exposure and shearing steps. A dial micrometer was used to verify the actual window displacement. Morphology of the HPDLC films was studied by low temperature cryo-SEM and TEM techniques.

All steps for in-situ shearing of an HPDLC sample are shown in Figure 2. First, the sample was sheared a small distance in a pre-shear step to remove any mechanical backlash in the DC servo motor or translation stage. A short holographic exposure (**A**) was used to start the polymerization and diffusion processes. Enough time was allowed for phase separation to begin as observed by the growth of a reflection



A – Holographically expose
B – Shutter laser and shear(1sec=>shear rate=shear dis/sec)
C – Stop shear and finish holographic exposure
D – Post cure with white light for 5 minutes

FIGURE 2 Prescription for real-time shear of HPDLC reflection grating.

notch. The laser light is then shuttered off and a shear (**B**) is applied to the cell by sliding the two substrates across each other a small distance. This was followed by a second holographic or Post-Holographic (**C**) exposure and a white-light exposure (**D**) flooding the sample fixing and curing all the remaining monomer inside and outside the grating region.

RESULTS AND DISCUSSION

As anticipated, shear creates a polarization dependence in an HPDLC grating along the direction of the shear. Small shear will cause an increase in the notch depth for light linearly polarized in the shear direction and a decrease in the notch depth for light with the opposite polarization. Figure 3 shows the case when the shear was large enough to completely eliminate the notch with the perpendicular to the shear. In general, for light polarized perpendicular to the shear direction, the notch blue-shifts, narrows, and decreases in depth or even vanishes. While the notch, red-shifts for light polarized in the shear direction. When a field is applied across a sheared sample the usual DE dependence is observed (i.e., DE drops with increasing field strength). However, the field for complete switching increases by a factor of about three for light polarized in the shear direction and drops for light polarized in perpendicular to the shear. Switching speeds,

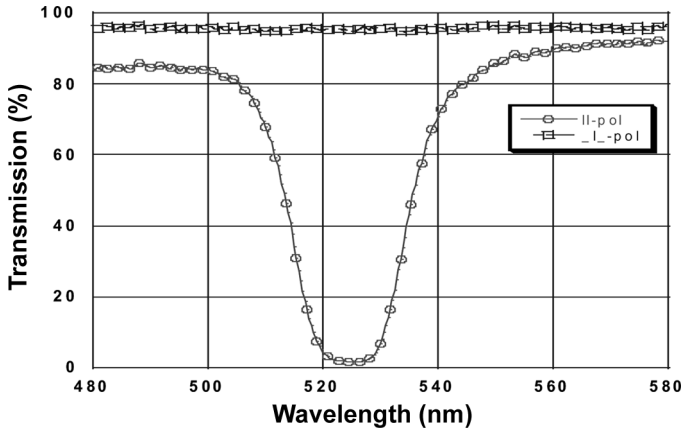


FIGURE 3 Sample with 30% LC and window displacement of ~0.76 mm after a holographic exposure time of 0.7 second. The transmission notch in the polarization perpendicular to the shear vanishes while the transmission notch in the other polarization deepens and broadens.

for light polarized in the shear direction decreases by about the same factor of three.

Structural integrity of the holographic grating region depends on the percentage of monomer polymerized. Some structural integrity is needed before a memory of the shear can be successfully retained. However, if too much polymerization occurs a larger force is required to induce the shear and the system returns to an un-sheared state when the force is removed. Transmission spectra of sheared cells for small holographic exposure times are shown in Figure 4. A short holographic exposure (**A**) produces a broadening of the notch, an increased DE, and a decrease in out-of-band transmission (i.e., increased scatter). As the holographic exposure time increases the bandwidth decreases slightly while out-of-band transmission and DE both increase until a best holographic exposure time is found for the other set conditions (laser intensity, cell thickness, syrup variations, etc.). Further increase of the holographic exposure time results in a decrease in optical performance.

Another factor in the final performance of a sheared cell is the magnitude of the shear distance (**B**). Variations in shear distance are shown in Figure 5 for a sample cell with 30% LC by weight, a gap thickness of 8 microns and holographic exposure of one second

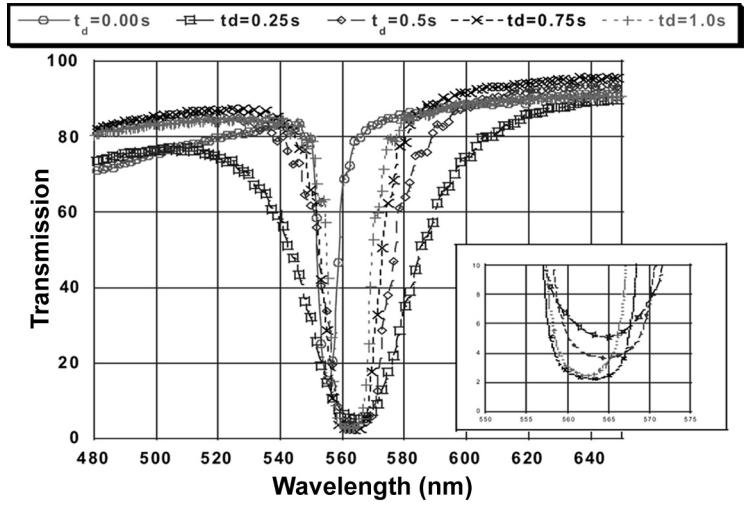


FIGURE 4 The effect of changing the holographic exposure time from 0 to 1 second. The initial narrow notch ($t = 0$) red-shifts and broadens as the exposure time begins to increase from zero. The notch then narrows and deepens, reaching its deepest point with $t \sim 0.75$ second.

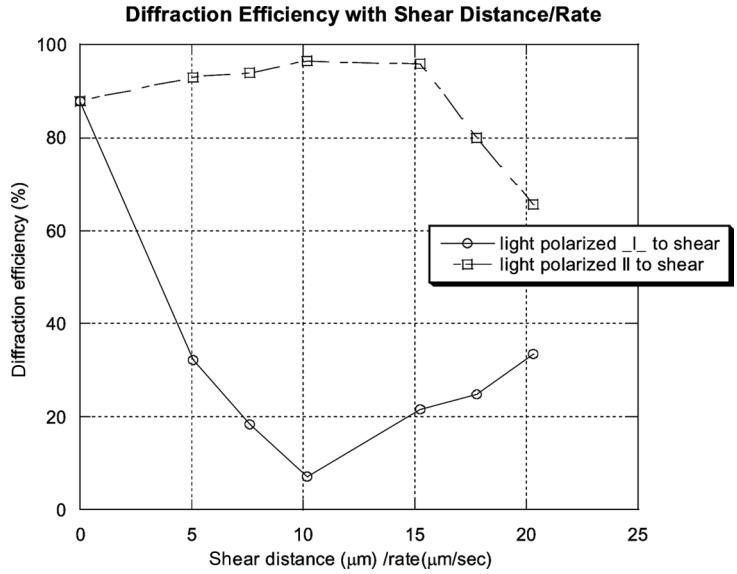


FIGURE 5 DE vs. shear rate shows the dramatic difference in behavior between the two polarizations.

at an intensity of $30\,mW/cm^2$. DE in the direction perpendicular to the shear drops very quickly while a corresponding increase of DE in the shear direction rises relatively slowly. When the shear becomes too large ($>10\,\mu m$) the sample begins to release from the substrates actually reducing the shear force. Further increasing of the shear causes delamination of the film, a large reduction in the shear transmitted to the film, and an increased scatter off the film glass interface. This interesting behavior of the polarization-dependent DE with shear distance could be because the LC droplets elongate out of the plane of the grating as seen in the TEM photographs (see Figure 6). The LC inside these droplets organizes in the direction along the major axis of the ellipsoid formed by the shear. For normal incident light, linearly polarized in the direction perpendicular to the shear, the **E** field will line up almost completely with the ordinary axis ($n_0 = 1.527$) of the LC. This closely matches the index of refraction of the polymer ($n_p = 1.524$) resulting in very little index modulation and almost no diffraction for this polarization. Light linearly polarized parallel to the shear encounters a mixture of ordinary and extra ordinary ($n_e = 1.809$) indices, the magnitude of which depends on the tilt angle, the angle of the ellipsoid with the grating vector (in this case also normal to the sample). This value would clearly be larger than that of the unsheared

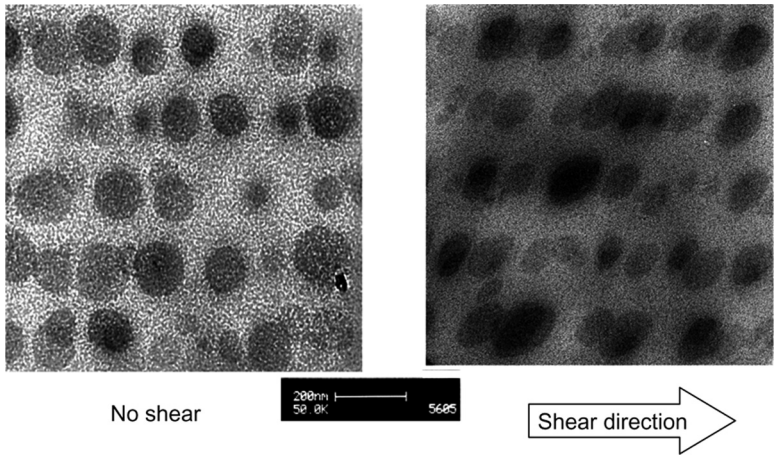


FIGURE 6 TEM comparison between an unsheared (left) and a sheared (right) HPDLC clearly shows the elongation and tilt of the LC droplets. (scale 200 nm)

sample, so the index modulation/DE would increases for this polarization. Only a small shear with a small tilt angle is needed to form the ellipsoid and the LC alignment, resulting in a quick drop in DE for light polarized perpendicular to the shear. The fact the droplets before shearing are already ellipsoids randomly oriented and shaped may actually be reason why DE in the perpendicular direction does not spontaneously drop to zero with the slightest shear. However, large tilt angles require significantly larger shear distances which accounts for the relatively slower growth of DE for light polarized parallel to the shear.

The effect of LC concentration over a small range from 27–33% by weight gave an optimum performance at $\sim$30% LC. For light polarized in the shear direction, higher concentrations resulted in an increased scattering of the baseline with no further improvement in notch depth but an increased bandwidth. Lower concentrations produced samples with less DE and bandwidth. A high degree of polarization-dependence is observed in all the samples indicating successful shearing. However, only a small amount of shear was required to increase the out-of-band baseline, independent of polarization.

CONCLUSIONS

Shearing in HPDLC's films creates tilted elliptical LC droplets which in turn align the LC more effectively. The tilt of the LC domains with

respect to the grating direction results in a larger index modulation for one polarization, leading to a polarization-dependent reflection. In addition, this ordering of the LC droplets directors by shear reduces the scatter when compared to random droplet directors. Shear during the early formation of a HPDLC in a reflection geometry leads to a significant increase in the achievable diffraction efficiency ($\sim$99%). The effect is permanent and does not require a holding force.

REFERENCES

[1] Ashmead, A. C., Popovich, M. M., Clark, J., Sagan, S. F., & Smith, R. T. (2000). *V*, Lewandowski, R. J., Haworth, L. A., & Girolamo, H. J. (Eds.), *Proc. SPIE*, *4021*, 180–186.
[2] Sutherland, R. L., Natarajan, L. V., Tondiglia, V. P., Siwecki, S. A., Chandra, S., & Bunning, T. J. (2001). *Liquid Crystals V*, Khoo, I.-C. (Ed.), Proc. *SPIE*, *4463*, 1–10.
[3] Qian, S., Colegrove, J., Liu, P.-Y., & Quan, X. (2001). *Organic Photonic Materials and Devices III*, Kippelen, B. E. & Bradley, D. D. C. (Eds.), *Proc. SPIE*, *4279*, 69–77.
[4] Tanaka, K., Kato, K., & Date, M. (1999). *Jpn. J. Appl. Phys.*, *38*, L277–L278.
[5] Bowley, C. C. & Crawford, G. P. (2000). *J. Opt. Technol.*, *67*, 717–722.
[6] Sagan, S. F., Smith, R. T. & Popovich, M. M. (2001). *Projection Displays VII*, Wu, M. H. (Ed.), *Proc. SPIE*, *4294*, 75–83.
[7] Qian, S., Colegrove, J., Liu, P.-Y., & Quan, X. (2001). *Organic Photonic Materials and Devices III*, Kippelen, B. E. & Bradley, D. D. C. (Eds.), *Proc. SPIE*, *4279*, 69–77.
[8] Yeralan, S., Gunther, J., Ritums, D. L., Cid, R., Storey, J., Ashmead, A. C., & Popovich, M. M. (2001). Sutherland, R. L., Prather, D. W., & Cindrich, I. (Eds.), *Proc. SPIE*, *4291*, 79–88.
[9] Caputo, R., De Sio, L., Veltri, A., Umeton, C., & Sukhov, A. V. (2004). *Opt. Lett.*, *29*, 1261–1263.
[10] Abbate, G., Vita, F., Marino, A., Tkachenko, V., Slussarenko, s., Sakho, O., & Stumpe, J. (2006). Molecular crystals. *Liquid Crystals*, *453*, 1.
[11] Sutherland, R. L., Natarajan, L. V., Tondiglia, V. P., Lloyd, P. F., & Bunning, T. J. (2006). *J. Appl. Phys.*, *99*, 123104.
[12] Natarajan, L. V., Shephard, C., Branndelik, D., Chandra, S., Sutherland, R. L., Bunning, T. J., Tondiglia, V. P., & Tomlin, T., (2003). *Chemistry of Materials*, *15*, 2477.
[13] De Rosa, M. E., Tondiglia, V. P., & Natarajan, L. V. (1998). *J. Appl. Pol. Sci.*, *68*, 523.
[14] Takeuchi, Y., Tanaka, K., & Kato, K. (1998). *IEICE TRANSACTIONS on Electronics*, vol. E81-C Núm. 11, 1685.