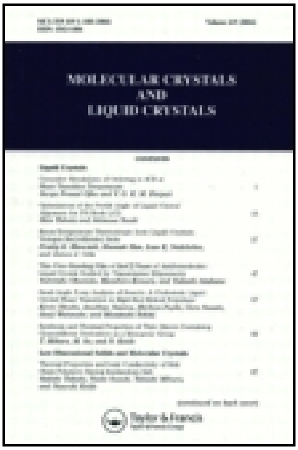

Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

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Version of record first published: 05 Oct 2009

Mol. Cryst. Liq. Cryst., Vol. 511, pp. 1/[1471]–10/[1480], 2009

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400903048453

Inscribing Holographic Patterns into a Polymer Liquid Crystals

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Two-beam interference holographic patterns have been inscribed into a diacrylate mesogen in a liquid crystalline phase. The recorded holographic pattern can be permanently stored by polymerization of the diacrylate mesogen. In the polymer storage medium, the local orientational ordering is determined by the interfering characteristics of the writing holographic pattern. The mesogenic groups in the regions exposed to constructive interference filed are found to orient in a unique direction, whereas a well aligned polymeric plate is not obtained in those regions exposed to destructive interference field.

Keywords: diacrylate mesogen; holographic recording; optical storage; polymer storage medium; two-beam interference

INTRODUCTION

Polymeric recording media capable of higher recording capability for optical storage have attracted massive attention because of their potentially vast availability, high resolution, self-development properties, ease of fabrication, and low cost. Azobenzene based polymers have been found to be useful materials for holographic recording. In these polymers, the orientation of azo chromophores can be induced by illumination with linearly polarized light leading to the generation of anisotropic effects such as birefringence and dichroism [1]. Azobenzene polymers have been intensively studied since the possibility of the holographic recoding of a surface relief grating (SRG) on an

The authors acknowledge the financial support from NSC of Taiwan through project NSC962221E110042 and the "Aim for Top Universities Plan" funded by NEC of Taiwan.
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azobenzene polymer was reported in 1995 [2,3]. Technically, SRGs can be produced by exposing an azobensene polymer to a holographic gratings pattern generated by two coherent beams. The microscopic mechanisms of the formation of the SRGs are not well understood. It is thought that the photoinduced directional mass transport is responsible for the SRG formation [4]. Due to their easy and reversible formation abilities, the SRGs are thought to have a wide range of applications in information storage [5–7], optical switching [8], nonlinear optics [9], the fabrication of optical elements [10–13] and photonic devices [14–16]. They have also been considered to be used as alignment layers for liquid crystal displays [17–20].

Photopolymerizable acrylic materials seem to be another catalogue of candidates for optical storage due to their real-time capabilities and easy handling [21–23]. In this article, we demonstrate that holographic patterns can be inscribed in to a thin layer of photocrosslinkable mesogenic diacrylate polymer. The use of mesogenic polymers in optical storage may introduce a new dimension of consideration, as in a liquid crystalline phase, the mesogenic diacrylate groups can be aligned by an external field because of their anisotropic nature in electric and magnetic characteristics. The alignment state of the recording materials can be frozen up by photopolymerization of the diacrylate monomers.

EXPERIMENTAL

The mesogenic monomer used in this study was a commercial diacrylate compound RM257 (Merck). In its monomer form, the diacrylate compound exhibits the nematic phase in a quite wide temperature range. The chemical structure and the phase sequence of the compound are shown in Figure 1. Samples were prepared by injecting RM257 into cells made using ultrasonically cleaned glass substrates by capillary action at an elevated temperature where the monomer was in isotropic phase. The thickness of the material layer was 10 μm. The temperature of sample was controlled by a Linkam TMS93 temperature system (Linkam Scientific Instrument).

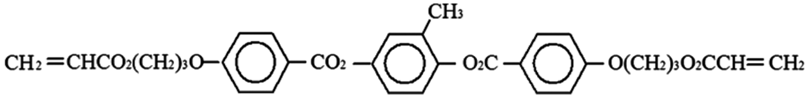

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FIGURE 1 Chemical structure of RM257, and phase sequence of the diacrylate mesogen.

Figure 2 shows the experimental setup used for the holographic recording. The light source used was an Ar^+ laser (2060Beamlok, Spectra-Physics). A s-polarized high power Ar^+ laser beam, operating at $\lambda = 488$ nm, was split into two equal amplitude coherent beams using a 50/50 beam splitter (BS), and then the two beams were allowed to recombine with an angle of α and interfere to form a holographic gratings in the space. In this arrangement, the gratings are parallel to the polarization of the two coherent beams, i.e., perpendicular to the incident plane. The spatial period of the gratings Λ, expressed in terms of the wavelength λ of the beam and the angle α between the two interfering beams, is

$$\Lambda = \frac{\lambda}{2\sin(\alpha/2)}. \tag{1}$$

A sample was installed in the interference region and exposed to the holographic pattern. A desired spacing of the interference pattern for a

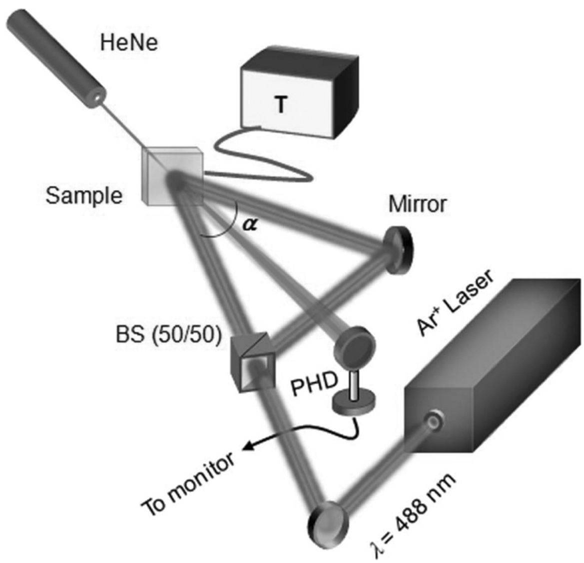

FIGURE 2 Schematically showing the optical setup for holographic recording. A 488 nm wavelength laser beam, produced by a Ar^+ laser, is split into two equal intensity p-polarized coherent beams by a 50/50 beam split. The two coherent beams obliquely incident at an angle $\alpha/2$, respectively, recombine and interfere at the sample. The time evolution of the diffractive gratings formed in the sample is monitored using a HeNe laser readout beam. T is the temperature system used for controlling sample temperature.

particular wavelength can be obtained by properly adjusting α. The effect of the holographic recording of the polymer was detected in the transmission mode. A 633 nm wavelength beam from a 5 mW HeNe laser was used as the read-out beam. The wavelength of 633 nm was selected for the read-out beam to avoid introducing extra photoreaction, which would interfere the holographic recording, in the diacrylate medium. The diffraction pattern was displayed on a black board and was monitored using a digital camera (PowerShot A620, Canon).

The phase behavior and the orientational states of the diacrylate mesogen in the sample were evaluated by analyzing optical textures of the samples using polarizing microscope (Axioscop40, Zeiss). Photomicrographs of the optical textures of the samples were taken by the Canon digital camera, which was installed in the polarizing microscope, for further analysis. An electrooptic measurement rig based on the polarizing microscope was set up to assist us in the evaluation of orientational states of the samples.

RESULTS AND DISCUSSIONS

RM257 is a photocrosslinkable diacrylate mesogen which exhibits the nematic phase within the temperature range of $70 \sim 120°C$. When the compound is in the monomeric form, the mesophases of the compound can be easily recognized and distinguished by observing its optical textures. At room temperature, a thin layer of monomeric RM257 sandwiched between glass substrates is in solid state and exhibits a fan shape texture in the polarizing microscope indicating a random orientation order of the mesogenic groups of the compound. The polymerization of the compound was carried out by exposing the compound to a UV radiation. After the polymerization, the optical texture of the RM257 film will not change with temperature. Experimental evidences for samples polymerized in a particular mesophase also revealed that the optical texture of a RM257 thin layer in the polymeric form is identical to that of RM257 in the mesophase in which the cross-link of diacrylate units was initiated, and is independent on temperature, i.e., the molecular ordering of the mesogen in a particular mesophase is frozen up by polymerizing the compound in the mesophase.

A sample at room temperature was exposed to a holographic gratings generated by two beams interference for 30 min. The spatial period of the interference pattern Λ was 30 μm. Throughout the present study, the power of each of two coherent laser beams was fixed at 1.0 W/cm^2. After processing, the sample was examined. No ablation

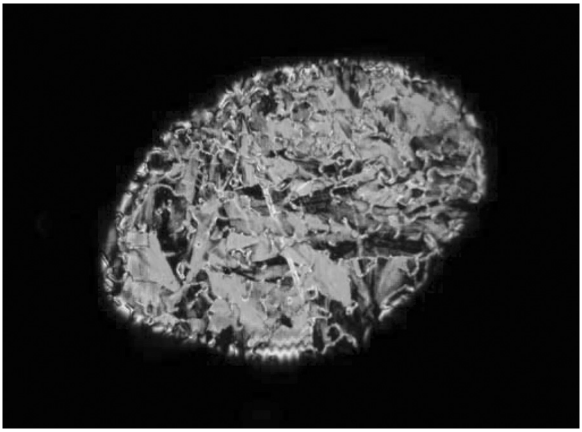

FIGURE 3 Photomicrograph of a RM257 polymer film after it was exposed to a two-beam interference pattern at room temperature. The photograph was taken when the sample was at 160°C. The optical texture of the region exposed to the interference pattern is unchanged. The black appearance of the other region in the picture is due to the diacrylate mesogen in the region is in isotropic state.

type of damage has been found. Figure 3 shows a photomicrograph of the sample after photoprocessing. The photograph was taken when the sample temperature was 160°C, which is well above the clear temperature of RM257. At the elevated temperature, the RM257 is in isotropic state and appears black in the polarizing microscope. The optical texture of the region of the sample exposed to the interference pattern, as illustrated in Figure 3, is unchanged indicating that RM257 in this region has been polymerized after exposure. In the present study, we have not provided any photo initiator for the polymerization of the monomers. The free radicals required for initiating the crosslink of the monomers were produced directly in the diacrylate compound when it was exposed to the laser beam.

Then the view field of the microscope was focused on the region exposed to the holographic interference pattern. In the polarizing microscope with the axis of the analyzer being set perpendicular to that of the polarizer, only a faint trace of a striped pattern could be seen. The stripes were found to be perpendicular to the incident plane defined by the two coherent laser beams, i.e., parallel to the polarization of the s-polarzed laser beams. The clarity of the appearance of the striped pattern increased when the analyzer of the microscope was rotated. When the analyzer was rotated 30°, the pattern

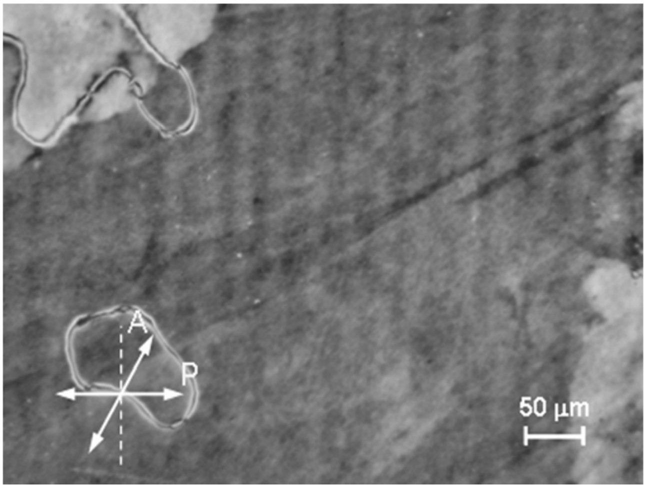

FIGURE 4 Photomicrograph of a holographic gratings pattern recorded in a RM257 polymer plate. The holographic recording was carried out while the RM257 was at room temperature. The white arrowed bars represent the polarization axes of the polarizer and the analyzer of the microscope, respectively.

had the best contrast, and can be, as illustrated in Figure 4, clearly seen. The spacing of the stripes was measured and found to be exactly the same as that of the interference pattern. This indicates that the holographic interference pattern has been inscribed into the RM257 layer.

Those regions in the sample exposed to the destruction field of the interference pattern exhibits a milky white appearance, which is almost unchanged when the sample was rotated in the polarizing microscope. The milky white appearance is due to the light leakage resulted from a disorder distribution in the director of the diacrylate mesogen in these regions. The appearance of the regions in the sample exposed to the construction field was found to vary between black and white alternately when the sample was rotated in the polarizing microscope. This indicates that the mesogenic groups in these regions are aligned to orient in a unique direction. The optimum contrast between the stripes corresponding to the constructive and the destructive interference of the holographic pattern, respectively, was found to appear when the axis of the analyzer of the microscope was adjusted to make an angle of 60° against the axis of the polarizer of the microscope, with the stripes being aligned perpendicular to the axis of the polarizer. The mechanisms for the molecular alignment of the polymer layer are still under study. It is thought that the mesogenic groups in the polymer are aligned by the holographic pattern generated by two polarized coherent laser beams. However, the orientation of the mesogen is not parallel to the polarization of the interference pattern. We examined the diffractive effect of the sample using the experimental rig shown in Figure 2, however, a recognizable diffraction pattern was not found.

For inscribing a holographic pattern in to an isotropic medium, a sample was heated to 145°C. This temperature was set to ensure the mesogen was in the isotropic state, and besides with a consideration for preventing the thermal-set of diacrylate monomers in the sample. The sample temperature was kept during the exposure which lasted for 30 min. The spatial period of the interference pattern generated by the 488 nm laser beam was 30 μm. After the exposure, the mesogen was polymerized, and did not exhibit any texture corresponding to a defective mesogenic layer. Figure 5 shows photomicrographs of the holographic pattern recorded in the RM257 layer while it was in isotropic state. A gratings structure, with white stripes inlaid into a black background, is clearly illustrated. The stripes are parallel to the polarization of the two coherent laser beams. The spatial period of the recorded grating is exactly the same as that of the writing pattern. When the sample was rotated, the milky white appearance of those stripes corresponding to the areas exposed to the destructive interference of the writing pattern never became completed black, whereas those areas exposed to constructive interference of the writing pattern appeared black and white alternately. This again confirms that in the regions exposed to destructive interference field the mesogenic groups are not aligned, whereas the mesogenic groups in those regions exposed to the constructive interference field are well aligned to form the one director region. The optical characteristics of these oriented regions also suggest that the diacrylate polymer in these regions becomes uniaxial optical media and the optical axes for all these striped regions have been aligned to orient in the same direction. The milky white appearance of those regions exposed to the destructive interference pattern is due to light leakage resulted from disordered orientation of mesogenic groups in these regions. It was also found that although an absolute darkness could not be achieved, the brightness of the appearance of these regions varied sinusoidally as the sample was rotated (c.f. Fig. 5) indicating a certain degree of alignment of the mesogenic groups in these regions. To understand the causes that lead to the orientational disorder of the mesogenic groups in the regions exposed to destructive interference of the holographic pattern requires an insight into the dynamics of photopolymerization

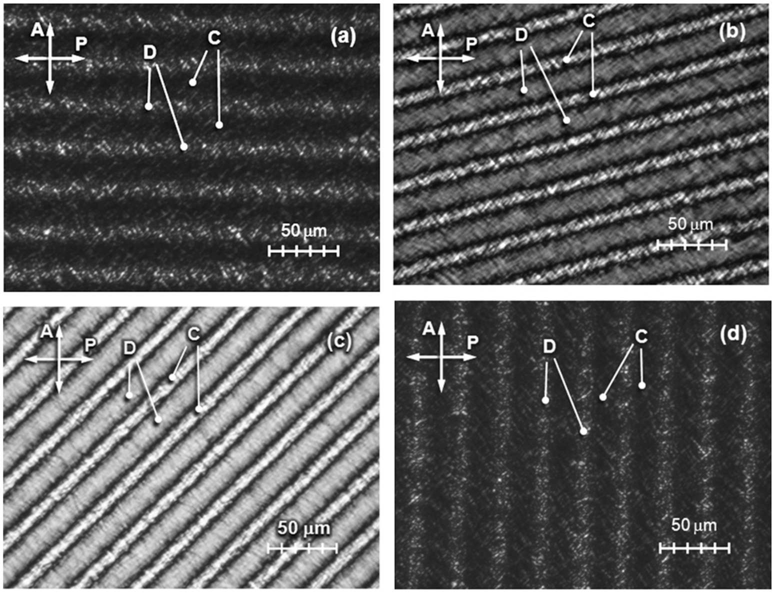

FIGURE 5 Photomicrographs of a holographic inscribed RM257 polymer plate which is rotated in a polarizing microscope. The two crossed white arrowed lines in each photograph represent the directions of the polarizing axes of the polarizer P and the analyzer A of the microscope, respectively. During inscribing the gratings the recording medium RM257 was kept at 145°C. In these pictures, the sample is in the positions where the stripes of the holographic gratings make an angle of (a) 0°, (b) 15°, (c) 45°, and (d) 90°, respectively, to the polarizer of the microscope. In these pictures, "C" and "D" represent constructive and destructive interference zones, respectively.

FIGURE 6 A diffractive pattern of a readout beam after it propagates across the holographic gratings illustrated in Figure 5. The readout beam, operating at 633 nm, is provided by a HeNe laser.

concerned linearly polarized light. Anyway an optical isotropic medium does not form in these regions.

As discussed above, the recorded pattern inscribed by the two-beam interference consists of two types of striped regions of different orientational ordering and thus different refractive indexes. The alternated arrangement of these two types of mesogenic stripes in the diacrylate polymer leads to the formation of diffractive gratings. Figure 6 shows the diffractive pattern of a p-polarized laser readout beam. The diffractive pattern shows that the holographic pattern generated by two-beam interference has been inscribed into the diacrylate polymer, and that the recorded optical information can be read out using a proper light beam. The experimental evidence also shows that better effects, in terms of holographic recording, can be achieved when the mesogenic diacrylate medium is in isotropic state.

CONCLUSIONS

Holographic interference pattern can be optically recorded into a thin layer of diacrylate polymer, and can be permanently stored in the media via the polymerization of the diacrylate monomer. Better result, in terms of holographic gratings recording, can be obtained when the diacrylate mesogen is in isotropic state during optical process. For the photopolymerizable diacrylate, the radicals required for initiating the cross-linking of the monomer are produced during the exposure to the holographic pattern. In the case of two-beam interference, a diffractive grating is inscribed in the polymer layer. The generation of the grating results from a periodic variation in optical refractive index of the polymer thin film due to a reorientation of mesogenic groups driven by the interfering light. In the regions exposed to constructive interference of the coherent laser beams, the mesogenic groups are driven to orient in a unique direction, whereas in those regions exposed to the destructive interference of the two beams the mesogenic groups are not well aligned.

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