Volume polarization holographic recording in thick phenanthrenequinone-doped poly(methyl methacrylate) photopolymer

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Received May 27, 2011; revised June 28, 2011; accepted June 29, 2011;
posted July 5, 2011 (Doc. ID 148338); published August 5, 2011

Volume polarization holographic recording in phenanthrenequinone-doped poly (methyl methacrylate) photopolymer is obtained. Photoinduced birefringence in a 2 mm thick sample is measured by a phase-modulated ellipsometry. The birefringence induced in this material by linearly polarized beam at 514 nm reaches 1.2×10^{-5}. In addition, ability for recording volume polarization grating using two different polarization configurations is demonstrated and compared. The experimental results show that the diffraction efficiency of the hologram reaches to ~40% by using two orthogonal circularly polarized beams.

OCIS codes: 090.0090, 090.2900, 090.7330, 160.5470.

Polarization-selective holographic optical elements require recording material to be sensitive with the periodic change of the polarization state of interference pattern. Unlike the classical phase or amplitude hologram, the photoinduced anisotropy in those materials leads to the polarization-dependent diffraction. Those devices are useful for many applications, such as optical interconnection, holographic data storage and imaging processing, etc [1–3]. So far, azobenzene-containing polymers with *trans-cis-trans* photoisomerization property are the most popular recording media [4]. Those materials show full reconfigurable dynamic holographic recording by the intrinsic reversibility of the photoinduced processes. Thin transmission holographic recording configuration is often applied due to heavy absorption resulting from a high concentration of azobenzene dye in the polymer [5,6].

In recent years, thick phenanthrenequinone-doped poly (methyl methacrylate) (PQ/PMMA) photopolymer for volume holographic applications has attracted intense research interest because experiments demonstrated that thick PQ/PMMA photopolymers possess not only high optical quality, but also negligible shrinkage effect under light exposure [7,8]. Additionally, Veniaminov *et al.* have reported the observation of weak photoinduced birefringence in the PQ/PMMA film during recording of phase holograms [9]. Later on, photoanisotropy in PQ/PMMA thin layers was characterized by Trofimova *et al.* [10]. Photoinduced birefringence in the material resulted from the anisotropic polarizability of disubstituted phenanthrene group on photoproduct. A polarization holographic recording was successfully demonstrated in 1.8 mm thick PQ/PMMA by using two orthogonal linearly polarized beams [3]. The diffraction efficiency of the recorded grating was ~7%. These results suggest that PQ/PMMA is an attractive candidate for permanent volume polarization holographic applications.

In this Letter, we present experimental studies of photoinduced birefringence and polarization holographic recording in 2 mm thick PQ/PMMA photopolymers. We show that linearly polarized light is more efficient to induce anisotropy. Diffraction efficiency (~40%) of a hologram can be significantly improved by using two orthogonal circularly polarized recording beams. To the best of our knowledge, it is the first demonstration to obtain such high-diffraction efficiency of a polarization hologram in a PQ/PMMA photopolymer. The Bragg selectivity curve of a 2 mm thick volume hologram is demonstrated, supporting negligible shrinkage effect during holographic recording, which is one of the most important issues for developing a volume holographic photopolymer.

Thick PQ/PMMA samples studied in this work were prepared by a two-step thermo-polymerization method [7]. A saturated concentration (0.7% by weight) of PQ and 1 wt.% of thermal initiator, azobisisobutyronitrile, were dissolved into the purified MMA solution. The resulting solution was placed in an ultrasonic water bath at 30 °C for 1 h, until all the components were dissolved completely, and then was stirred (using a magneto stirrer) in an open-end glass bottle at 30 °C for 24 h. After the stirring, the viscous solution was poured into a glass mold with a 2 mm thick spacer. The sample was then baked at 45 °C for 72 h, until most of the monomers were polymerized. We then obtained a self-sustained solid block which can be removed from the mold for testing without postprocessing such as cutting and polishing. Our PQ/PMMA contains residual MMA molecules dispersed in the polymer for holographic recording [7].

The photoinduced birefringence was measured by a photoelastic modulated polarimetry (shown in Fig. 1). During experiments, the samples were excited by either (a) a linearly polarized Ar^+ laser beam at 514 nm with azimuth angle 30° with respect to the incident plane, or (b) a circularly polarized Ar^+ laser beam. We tried to keep both beams operating at the same intensity of 19 mW/cm^2. The beam was incident oblique with an angle of 10°. During exposure, the birefringence of the sample was probed by a weak HeNe laser beam ($\lambda =$ 632.8 nm), which was sinusoidally phase-modulated in a pair of cross polarizers. The azimuth angle of the induced optical axis α and phase retardation Δ_Q can be

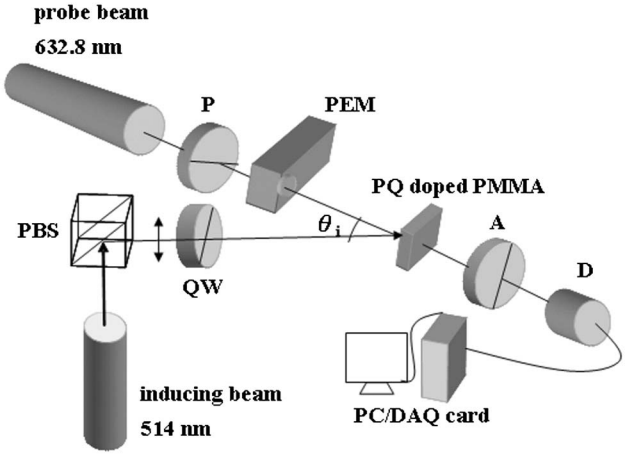

Fig. 1. Experimental setup of photoelastic modulator polarimetry: the photoinduced birefringence is monitored in real time. Linearly or circularly polarized light at 514 nm is used for excitation.

obtained by calculating harmonic components of the Fourier spectrum of intensity of output light. The details of the measurement have been described in [15].

The temporal responses of the azimuth angle α during exposure are shown in Figs. 2(a) and 2(d). The temporal responses of phase retardation Δ_Q are shown in Figs. 2(b) and 2(e). The absolute value of the induced birefringence Δn can be calculated from the formula $|\Delta n| = \lambda\Delta_Q/(2\pi nd)$, where λ is wavelength of the probing beam, d is the thickness of and n is the refractive index of polymer (1.48). The results are shown in Figs. 2(c) and 2(f). Obviously, under the excitation of a linear polarized beam, one can also observe the azimuth angle of the induced optical axis changed from 45° to the saturation value of ~32°, which indicates the material changed from an isotropic to a birefringence medium with its optical axis parallel to the azimuth angle of the inducing beam. In contrast, there is no significant change of angle α in the case of excitation with circularly polarized beam. Small angle Δ_Q (3°) detected in the beginning of exposure is attributed to the intrinsic birefringence of the PMMA polymer. Moreover, under excitation, the change of the birefringence value Δn induced by a linearly polarized beam is ~16 times larger than that induced by a circularly polarized beam. From these measurements, we can conclude that the linearly polarized light is more efficient to induce linear birefringence in a PQ/PMMA photopolymer. It is useful for us to design the polarization configuration of recording polarization hologram.

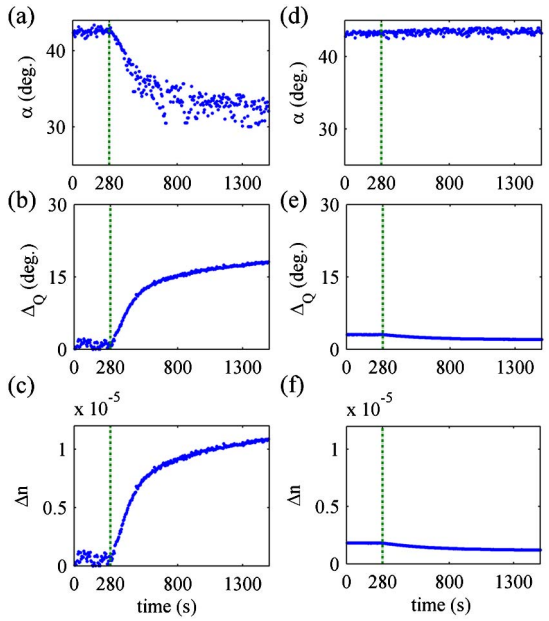

Fig. 2. (Color online) Temporal response of azimuth angle of induced optical axis α, phase retardation Δ_Q and induced birefringence Δn. The dashed line in each figure indicates the starting point of excitation. (a), (b), (c) are excited by linearly polarized light, while (d), (e), (f) are excited by circularly polarized light.

For polarization holographic experiments, a 514 nm Ar^+ laser beam was used to write gratings in a two beam interference setup with transmission geometry. The intersection angle of the two beams was 30° (outside sample). Two polarization configurations have been investigated, which were generated by the interference of two recording plane waves with orthogonal linearly or circularly polarized states, respectively. For linear polarization configuration, the polarization states of the two recording beams were set as s-polarized and p-polarized, with respect to the incident plane. For circular polarization configuration, right-hand and left-hand circularly polarized beams were used. During recording, the diffraction efficiency, defined as the ratio of the intensity of diffracted beam to that of the incident beam, is monitored by blocking one beam periodically and measuring the intensity of the diffracted beam. Additionally, after recording, the Bragg selectivity curve of the hologram was monitored by rotating the sample mounted on a rotational stage.

Dynamics of the diffraction efficiency, which is retrieved by the p-polarized beam during recording, with different intensity of each writing beam for linear polarization configuration is shown in Fig. 3. It is seen that the recording of the polarization hologram in PQ/PMMA is strongly intensity-dependent. The sample is nearly inactive until the beam intensity is larger than 10 mW/cm^2. While the intensity is too large (>30 mW/cm^2), the diffraction efficiency also decreases. An optimized intensity in our case is close to 26 mW/cm^2 and the maximal diffraction efficiency reaches ~7%. As proposed by

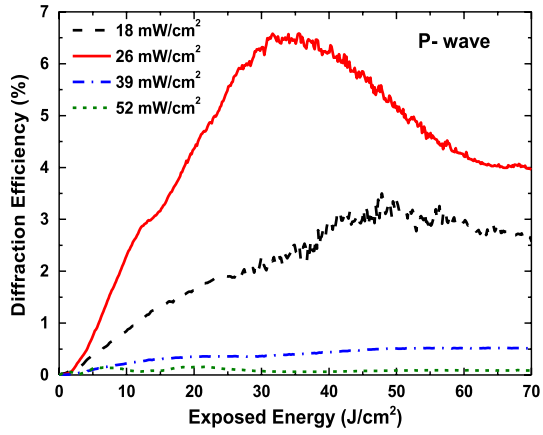

Fig. 3. (Color online) Dynamics of the recording processes for the case of linear polarization configuration with different intensity of writing beam.

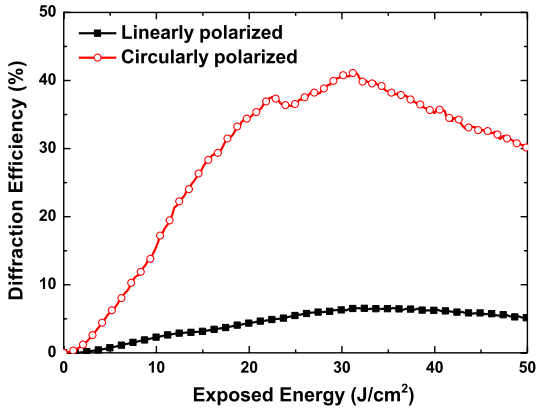

Fig. 4. (Color online) Dynamics of the recording processes for both cases of circular and linear polarization configurations.

Veniaminov and Chuang [9,11], there are two photoinduced effects, induced refractive index change and birefringence occurring simultaneously in the PQ/PMMA. The speed of former effect is slower, but the change is larger. As exposed with stronger light, the fraction of an oriented ensemble of photoproducts conducting the photoanisotropy may be smaller. Figure 4 shows dynamics of diffraction efficiency retrieved by a left-hand circularly polarized beam during recording for circular polarization configuration with intensity of 26 mW/cm^2. For comparison, the curve of linear polarization configuration is plotted on the same graph. Obviously, significant improvement is observed. With exposure energy of 30 J/cm^2, the maximal value reaches ~40%, which is almost six times larger. As noted above, linearly polarized light is more efficient to induce linear birefringence. Figure 5 shows the state of polarization of the resulting light field in one pitch of interference pattern for both polarization configurations. It is seen that with the linear polarization configuration, the polarization ellipse of the optical field changes periodically in the hologram plane. In contrast, with the circular polarization configuration, it is possible to achieve linear polarization of the total field. The rotation of polarization in the hologram plane generates the holographic grating. Thus, the induced birefringence is larger using the circular polarization configuration.

Figure 6 shows a typical Bragg selectivity curve for a strong hologram (~40% diffraction efficiency) recorded with a circularly polarization configuration. It can be seen clearly that the hologram has a sinc-squared selectivity curve as expected for a 2 mm thick hologram. The peak of diffraction efficiency is retrieved using a reference beam incident at almost the same angle during writing. With the resolution of our rotational stage of 2×10^{-4} degree, the induced shrinkage coefficient in our PQ/PMMA sample is calculated to be smaller than 10^{-4}, using the formulas in [12]. This result suggests the shrinkage is not a suspect because the high-bandwidth hologram can be reconstructed completely. This is one of the most desireable properties for developing a volume holographic photopolymer.

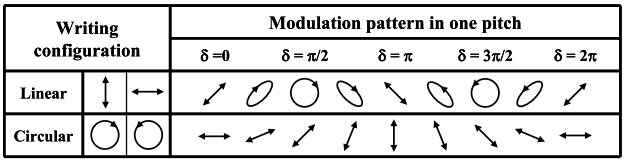

Fig. 5. The state of polization of the resulting light field of the two orthogonally polarized waves with phase difference δ.

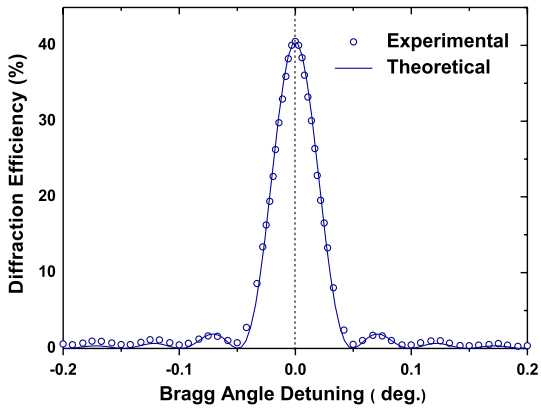

Fig. 6. (Color online) The Bragg selectivity curve for a 2 mm thick sample for a polarization hologram with diffraction efficiency ~40%. The symbols represent measured results and the curve is theoretically fitted by using the coupled wave equation [6] .

In summary, we have studied the photoinduced birefringence of a 2 mm thick PQ/PMMA photopolymer. The induced birefringence of this material can reach 1.2×10^{-5} by exposing it to a linearly polarized beam at 514 nm. We have successfully performed volume polarization holographic recording. The experimental results show that, with circular polarization configuration, the maximal diffraction efficiency of hologram can reach ~40%. A clear sinc-squared Bragg selectivity curve indicates that the shrinkage is neglible in our PQ/PMMA. These results suggest our PQ/PMMA can be an attractive candidate for permanent volume polarization holographic recording.

Financial support by National Science Council of Taiwan (NSCT), under contract #NSC 97-2628-E-009-034-MY3 are gratefully acknowledged.

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