Two-wavelength volume holographic recording in thick PQ-doped PMMA photopolymer

June Hua Lin [a], Ken Y. Hsu *[a], Shiuan Huei Lin [b]
[a] Department of Photonics, National Chiao Tung University, Hsinchu 30010, Taiwan; [b] Department of Electrophysics, National Chiao Tung University, Hsinchu 30010, Taiwan

ABSTRACT

We report holographic recording in thick phenanthrenequinone-doped poly(methyl methacrylate) (PQ/PMMA) photopolymer material via the two-wavelength technique. By using gating light at 325 nm and writing light at 647 nm two-wavelength holographic recording is achieved. Non-volatile readout of a holographic image under 24 hours continuous reconstruction is demonstrated. A four-level modeling for the photochemical procedures of the two-wavelength holographic recording is proposed, and dynamic behaviors of the holograms are illustrated. A planar integrated optical correlator system is constructed by selective writing of holographic optical elements via two-wavelength holographic recording on a photopolymer disk.

Keywords: photopolymer materials, real-time holography, non-volatile volume holograms, two-wavelength recording, spatial multiplexing holograms, temporal multiplexing holograms

1. INTRODUCTION

Volume holograms have been intensely investigated because there are many potential applications such as data storage, optical interconnects, interferometry and optical metrology, 4-D microscope, holographic optical elements, narrow-band spectral filters, image processing and recognition, and displays[1, 2]. The key to a success in these applications relies on suitable materials for recording holograms that match the needs of particular systems. For example, volume holographic data storage requires recording materials with large dynamic range and with negligible photo-induced shrinkage, while real-time interferometers and displays require recording materials with quick refreshing ability. In this investigation we are particularly interested in real-time volume holograms for applications of information storage and processing.

Popular materials for recording real-time volume holograms can be catalogued into inorganic[3, 4] and organic types [5, 6]. We are interested in developing organic materials for holographic data storage. Phenanthrenequinone-doped poly(methyl methacrylate) (PQ/PMMA) photopolymer has been our major interest, because the compositions of the material are easy to adjust and the fabrication does not require dedicated equipments [7-12].

We are particularly interested in developing materials for holographic storage of write-once and read-many (WORM) type. Basically, photopolymers including PQ/PMMA are dynamic or real-time materials. That is, holograms got enhanced upon optical exposure during the writing stage, and they got erased by the reconstructing beam during the reading stage. This kind of destructive reading should be avoided if we are interested in developing archival data storage. Thus, it is necessary to find a method for hologram fixing in these materials. A typical technique for hologram fixing in photopolymers is UV curing, which requires post processing after the holograms have been recorded. Here we investigate another technique for achieving non-volatile holograms in real time named two-wavelength recording. We demonstrate that during the writing stage, holograms can be recorded on PQ/PMMA under simultaneous illumination of 325 nm and 647 nm light beams. During the reading stage, holograms can be reconstructed by the 647 nm light beam without erasure. Of the two wavelengths 325 nm is a uniform beam called the gating light, because it controls the time duration that allows the material to be sensitive to 647 nm. The 647 nm light is called the writing light, because it is responsible for producing the interference fringes for holograms writing. We describe fabrication and testing of PQ/PMMA for two-wavelength holographic recording. A four-level system model of the material will be proposed, and dynamic behaviors of the holograms are analyzed. Non-volatile readout of a holographic image under 24 hours continuous reconstruction is demonstrated. We also demonstrate that a planar-integrated optical correlator for five-

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*ken@cc.nctu.edu.tw phone +886 3 5712121 56360; fax +886 3 5725601;

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channel image recognition can be constructed by selective writing of holographic optical elements on a PQ/PMMA disk via two-wavelength holographic recording.

2. TWO-WAVELENGTH HOLOGRAPHIC RECORDING IN PQ/PMMA

2.1 Sample preparation

The idea of two-wavelength holographic recording in photopolymers has been reported previously [13, 14]. They discovered that photosensitive molecules with cascaded-excited meta-stable intermediate levels offer the capability of two-wavelength recording. According to their results, molecules with α-diketone structure possess these characteristics of energy levels. Since PQ molecules are with α-diketone structure, hence it is interesting to investigate the possibility of using PQ/PMMA for two-wavelength hologram recording.

We fabricate the samples using a two-step thermal polymerization method. The composition of the material is 0.7 wt.% photosensitizer of PQ, 1 wt.% thermal initiator of AIBN (2,2-azo-bis-isobutyrolnitrile), and the rest of the component is monomer of MMA (methyl methacrylate). In the first step, the three components are mixed and stirred completely at 30 ºC for 24 hours. In the second step, the well mixed liquid is poured into a container of the desired geometry and put into an oven at 45 ºC for 72 hours until the sample becomes a solid bulk. The solid bulk can be removed from the container to be used for optical experiments. Fig. 1 shows the procedures for fabrication and photos of the samples. This kind of PQ/PMMA has been successfully used in holographic data storage experiments at 514 nm wavelength. Now we investigate the possibility for two-wavelength recording in this material.

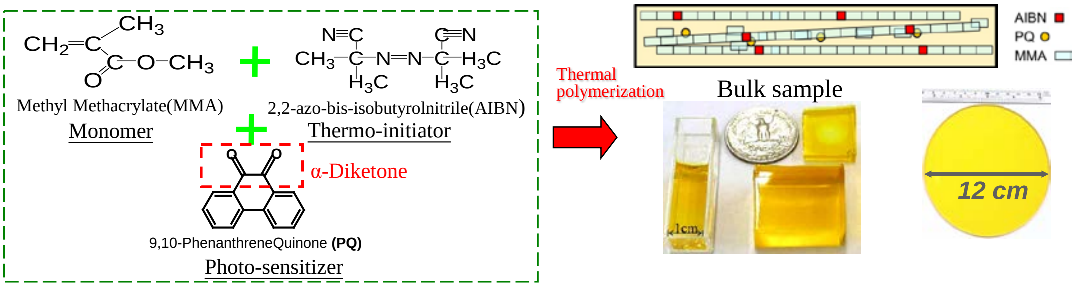
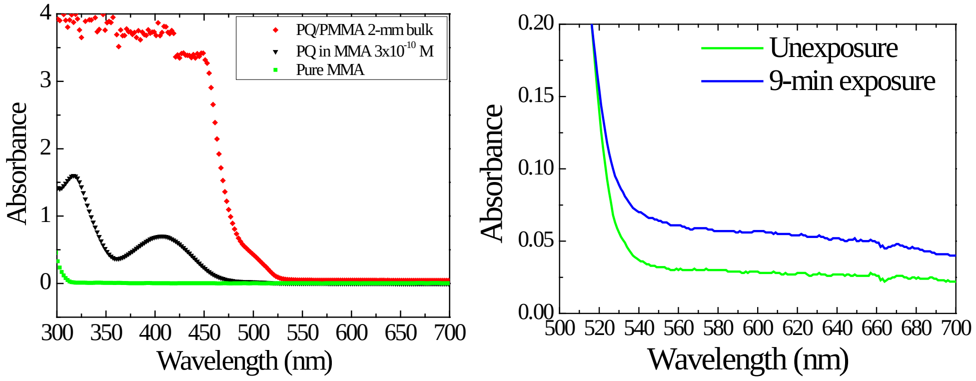


Figure 1. Fabrication and photos of PQ/PMMA.

2.2 Real-time holography of two-wavelength recording



(a)

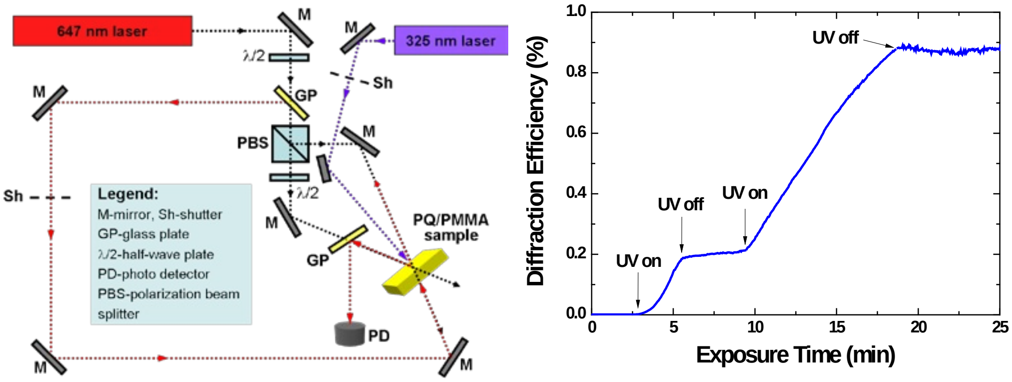
(b)

Figure 2. The spectra of PQ/PMMA, PQ diluted in MMA, and pure MMA;(b) UV-induced spectra of PQ/PMMA

In order to achieve non-volatile reading and two-wavelength holographic recording, two properties of the materials should be met. First, the material should not be sensitive to the longer wavelength λ_1, so that hologram reconstruction with λ_1 will not cause erasure during the reading stage. Second, the material should be sensitive to λ_1 under simultaneous illumination of an uniform beam at a shorter wavelengths λ_2, so that the interference fringes produced by wavelength λ_1 can be recorded during the writing stage. In order to find suitable wavelengths of λ_1 and λ_2 for PQ/PMMA, we performed a absorption spectroscopy measurement of the material. The results are shown in Fig. 2a. In order to see the behavior of PQ molecules more clearly, the absorption spectra of pure MMA and that of PQ in dilute solution of MMA are also shown in the same figure. It can be seen that there are two absorption peaks in the range from 300 nm to 500 nm. We chose a He-Cd laser at wavelength 325 nm as the light source for gating light.

According to the absorption spectrum, PQ/PMMA is transparent for wavelengths longer than 550 nm. Further, as shown in Fig.2b, the material demonstrated light induced absorption in this range when it is under simultaneous illumination of 325 nm. Thus, any wavelength longer than 550 nm would be suitable to serve as $\lambda_{1.}$ We chose 647 nm from a krypton laser as the writing light in our experiments.

We performed two-wavelength holographic recording experiment in PQ/PMMA. A bulk PQ/PMMA of 2 mm thick was placed in a typical holographic recording system, as shown in Fig. 3a. A uniform beam of He-Cd laser at 325 nm and with intensity of 90 mW/cm^2 was used as the gating beam. The writing beam was derived from a krypton laser at 647 nm. It was split into two beams and interfered on the sample with an intersection angle of 28º. An electronic shutter was used to control the ON and OFF of the gating beam. The strength of the hologram was real-time monitored by a probe beam at 647 nm, which was directed along the backward-propagation direction of one of the writing beams. Experimental results are shown in Fig. 3b. It is seen that the hologram starts to build up when the UV gating beam is ON, and it remains constant when the UV gating beam is OFF whiles the sample was kept illuminated by the 647 nm light. This is exactly what is required for the two-wavelength recording: the 325 nm light controls the gating period during which the PQ/PMMA is sensitive to 647 nm light for the hologram writing, otherwise, the 647 nm light cannot produce any photo-chemical reaction in the material. Thus, two-wavelength hologram writing and non-volatile reading have been achieved.



(a) (b)

Figure 3.(a) The experimental setup for holographic recording and other experiments;(b)the demonstration of switchable recording by using the gating light.

Above experimental result shows that illumination of gating light at 325 nm is necessary for PQ/PMMA to be able to record holograms with writing light at 647 nm. However, uniform illumination of the gating light tends to reduce the modulation index in the material and thus to reduce the holographic grating strength during the writing stage. Further experiments show that too strong intensity on gating light indeed reduces the maximum diffraction efficiency, though it takes shorter time to reach the maximum value. Therefore, a trade-off between the maximum diffraction efficiency and speed to reach peak diffraction efficiency should be made by choosing suitable power levels for two-wavelength holographic recording. In order to determine proper power levels of the lasers, it is necessary to develop a system modeling for the photo-chemical reaction of two-wavelength holographic recording in PQ/PMMA.

2.3 Four-level modeling of the two-wavelength holographic recording in PQ/PMMA

According to our previous investigations, holographic recording in PQ/PMMA can be described as a photo-chemical process of molecular attachments between PQ radicals and MMA molecules. When the material is illuminated with a light beam at suitable wavelength PQ molecules absorb the photon energy to become radicals, so that they react with MMA molecules to form a photoproduct based on the photoreaction of one molecule to one molecule attachment. Since the refractive index of these photoproducts is different from the rest of the material, thus the intensity of the optical interference fringes is recorded as a corresponding distribution of the refractive index. This photochemical process can be described by a four level system.

In two-wavelength holographic recording in PQ/PMMA, the gating light is a UV at 327 nm with higher photon energy, and the writing light is at red wavelength of 647 nm with lower photon energy. Under illumination of gating light, PQ molecules are first pumped from ground level S_0 to an excited level S_1 of singlet states. Then, the molecules undergo a fast interstate relaxation to reach ground level T_1 of triplet states. Since T_1 is a meta-stable level, hence the number of molecules on this level will be accumulating under the pumping of gating light. From there, the molecules can again be pumped to one of excited levels T_n of triplet states to become radicals that are capable of performing molecular attachment with MMA molecules.

Note that, energy gap between S_1 and S_0 is larger than photon energy of 647 nm and fits that of 327 nm. Thus, without gating light there will be no molecule accumulation on the meta-stable level $T_{1,}$ and holographic writing or erasure will not occur for single wavelength illumination at red wavelength. Also note that, there is an absorption band for molecules on T_1 level, which includes UV wavelength of the gating light as well as red wavelength of the writing beam. Therefore, UV-induced-absorption of red light can occur and thus holographic writing can be performed under simultaneous illumination of gating and writing lights. The red light induces a refractive index distribution change that is proportional to the interference fringes of the writing beams, yet the UV light produces a uniform change of the refractive index and that will cause a reduction in the modulation index of the holographic grating. Thus, the two lights compete with each other for molecules on the meta-stable level, and that will affect obtainable diffraction efficiency of the holograms. In order to analyze dynamic behaviors of holographic grating we can write down rate equations of the four-level system and obtain numerical solution on it.

In fact, dynamic behavior of the holographic grating can be estimated simply by measuring the light-induced absorption of the two wavelengths. Light absorption results in decreasing of photon flux, and that will induce a corresponding rate change of population density at each level. By combining these relationships, the photo-induced absorbance can be derived as a function of photon flux of the UV and red lights, population density and pumping constant at each level.

Since the dynamics of light-induced absorption can be measured experimentally, hence the corresponding absorbance can be calculated thereafter. By curve fitting of the calculated values for the light-induced absorbance with that of the derived formulas the pumping constants can thus be found. These pumping constants then can be used to calculate the temporal behavior of diffraction efficiency of the holographic grating. The maximum diffraction efficiency can be plotted as a function of the intensity ratio between the UV and red light, thus suitable range of the intensity ratio between the two lights can be found. Fig. 4 shows dynamic behavior of a hologram with intensity ratio of 0.3. It can be seen that simulation result fits well with that of the experimental measurement.

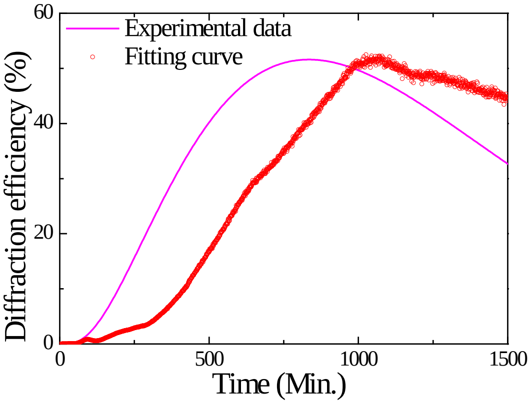
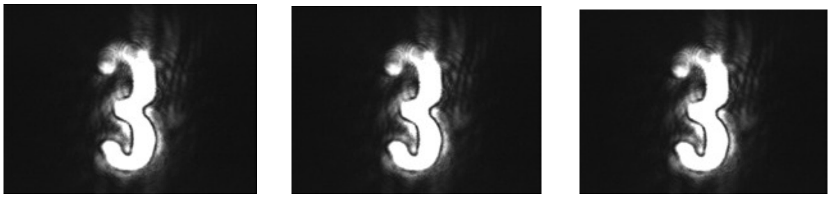


Figure 4. Experimental data and simulation curve of the diffraction efficiency

2.4 Non-volatile image readout from the two-wavelength holographic recording in PQ/PMMA

We performed holographic image recording and reconstruction by two-wavelength recording in PQ/PMMA. The holographic recording system was the same as that shown in Fig. 3(a), except that an image of digit 3 was put into the object beam. Gating light was turned OFF when the hologram diffraction efficiency reached about 5%. Then, the hologram was reconstructed continuously by red light for 24 hours. The reconstructed images at the beginning and that after reading 3 minutes, and that after 24 hours are shown in Fig. 5(a), (b), and (c), respectively. It is seen that brightness of the readout images remains almost unchanged during this period. Therefore, a non-volatile holographic recording and reconstruction has been demonstrated.



(a) (b) (c)

Figure 5.Retrieved images: (a) at the beginning;(b)after 3 minutes;(c) after 24 hours.

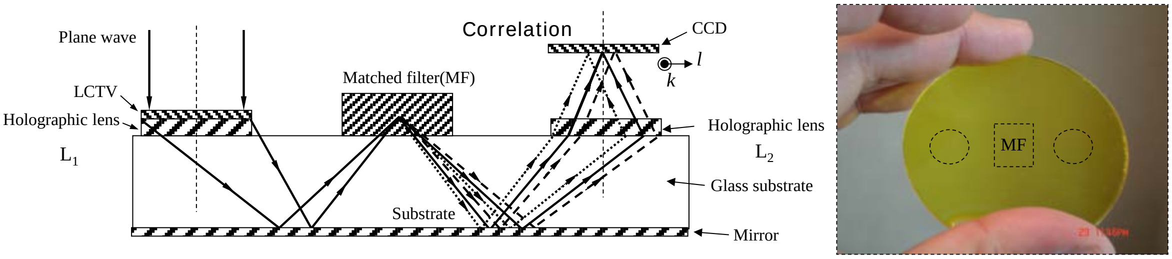
3. APPLICATION OF TWO-WAVELENGTH HOLOGRAPHIC RECORDING

3.1 Selective writing of holographic optical elements (HOE) for planar integration of optical correlation system

Fig. 6(a) shows the schematic diagram of the system. A PQ/PMMA disk of 2 mm thick was put on top of a glass substrate. The bottom of the glass substrate is a mirror of metallic coating. By using a gating light to select the positions for writing holographic optical elements one by one, an image correlation system can be fabricated on the polymer disk.

In the first step, appropriate writing lights (a plane wave plus a focused wave at 647 nm) were directed onto the position of holographic lens L_1. Under simultaneous illumination of the gating light at 325 nm onto this position, L_1 was written. Note that in this step only L_1 was written, because the rest of the photopolymer disk was not illuminated by gating light. In the second step, a liquid crystal TV (LCTV) was placed on top of L_1 lens. A target image was presented on LCTV and with illumination of 647 nm the Fourier spectrum of this image was displayed at the position of matched filter MF. At this time, a plane wave of 647 nm was also incident on the matched filter position at an appropriate angle. Thus, with simultaneous illumination of 325 nm onto this position, a matched filter for this target image was written.

Then, a point source of 647 nm originated at the desired position of the correlation peak (on CCD plane) was directed onto the position of holographic lens L_2, and this wave interfered with a plane wave that was counter-propagating with the reference wave. Under simultaneous illumination of 325 nm at the position of L_2, holographic lens L_2 was written. Thus, later on, when the first target image is presented into the system, the correlation peak will be directed into the designated position and detected by CCD camera. In a similar way, matched filters for other target images can be recorded one by one as volume holographic optical elements at the same matched filter position in the photopolymer bulk, with reference wave incident at different angles. Thus, the correlation system is integrated onto a planar bench, as shown in the photo of Fig. 6(b).



(a) (b)

Figure 6. (a)Compact correlation system;(b)Photo of the system.

Fig. 7 shows target images and a correlation result of a five-channel optical correlator. Experimental results showed that correlation peaks of different target images appeared at the designated positions of the CCD camera and thus images are recognized correctly.


(a) (b) (c)

Figure 7.(a)Target patterns;(b)Input pattern;(c)Correlation results.

4. CONCLUSIONS

In summary, we have presented two-wavelength holographic recording in thick PQ-doped PMMA photopolymer. Principles of two-wavelength recording and non-volatile readout are illustrated. Fabrication of the material has been described, and two-wavelength holographic recording has been demonstrated with gating light at 325 nm and writing at 647 nm. A four energy-level system modeling for the photochemical process of two-wavelength holographic recording in PQ/PMMA has been proposed. Dynamic behaviors of holographic gratings have been analyzed. A non-volatile holographic memory with 24 hours non-destructive readout has been demonstrated. Application of two-wavelength holographic recording for selective writing of holographic optical elements in a PQ/PMMA disk has been demonstrated.

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