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Formation mechanisms of multiple holographic gratings in spirooxazine-doped polymer films

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ABSTRACT

Multiple holographic gratings were stored in the same location of spirooxazine doped polymer films using peristrophic multiplexing techniques. Accompanied by UV irradiation, three sets of holographic gratings with different directions were recorded and readout by the He–Ne laser (632.8 nm) and the Nd:YAG laser (532 nm), respectively. Diffraction efficiency of each grating was controlled almost uniform by adjusting recording time. It was found that the growth rate of the holographic grating recorded later was lower than that of the earlier one, resulting from the weakened anisotropic distribution and the decreased population of photomerocyanine molecules. A kinetics description of isomerization gratings, competing with orientation gratings, agrees well with experimental results. Due to the thermal stability of the isomerization grating, multiple interference fringes in the photochromic film were reserved and observed by Confocal Laser Scanning Microscope.

1. Introduction

In this rapidly evolving age of information, there has arisen the need for high-density and rapid access memory system [1]. Holographic data storage allows a great number of bits to be stored in the recording media with high sensitivity [2]. Recently, how to further improve the density and read-out (write-in) rate of holographic storage has been a subject of intense current studies [3–7]. To a great extent, the lack of suitable recording materials has long been a binding constraint to the development of holographic storage. In fact, holographic recording media must satisfy stringent criteria, including high dynamic range, dimensional stability, optical clarity and flatness, millimeter thickness, rapid response time and non-destructive readout [8]. Recently, polymer films containing photochromic materials have been investigated in the applications of holographic recording, molecular switching, optical modulation, and nonlinear optics [9]. A photochromic compound is characterized by its ability to alternate between two different chemical forms having different absorption spectra, in response to irradiation by light of appropriate wavelengths. As a well-known photochromic dye, spirooxazines (SOs) have the reaction of a reversible photochemical cleavage of the C—O bond in the SO rings upon UV-light excitation. The subsequent isomerization, photomerocyanines (PMCs), can revert to the spiro forms thermally or photochemically [10]. To date, some investigations on photochromic spirooxazines have been carried out in synthesis, reaction mechanism, optical switch and memory etc. [11–14]. However, little attention has been given to the formation mechanisms of multiple holographic gratings in spirooxazine doped polymer films, which may be helpful to the application of high-density holographic storage.

In this paper, we report the multiple holographic gratings recorded with a He–Ne laser (632.8 nm) by peristrophic multiplexing with axis of rotation perpendicular to the recording media. The relationship between the growth rate and overlapping times for the holographic gratings was studied. The corresponding physical process was also discussed. A kinetics description for isomerization gratings, competing with orientation gratings, agrees well with experimental results. Multiple interference fringes were observed by Confocal Laser Scanning Microscope.

2. Materials and methods

2.1. Materials and film preparation

6′-Piperidino-1,3,3-trimethylspiro[indolino-2,3′-[3H]naphtha-[2,1-b][1,4]oxazine (SO) was synthesized with microwave

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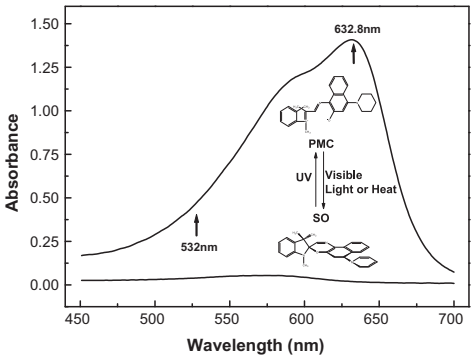

Fig. 1. Absorbance spectra for SO (PMC) doped PMMA films, and the molecule transformation between SO and PMC (insert).

irradiation. Commercially available poly(methyl methacrylate) (PMMA) was used without further purification. Both SO and PMMA were dissolved in $CHCl_3$ and then cast on a clean glass substrate. After the solvent evaporated, the composite film was obtained and used for holographic recording. The pre-set dye concentration was 15.0 wt% and typical film thickness was 10 ± 1 μm measured with a Precision Ellipsometer.

2.2. Absorbance spectra

Fig. 1 shows absorbance spectra for SO/PMMA film in visible wavelength. After UV irradiation, SO molecules are transformed to photomerocymine (PMC) molecules, resulting in an abroad absorbance band in visible wavelength. For the maximal absorbance at around 633 nm and a much lower absorbance at around 532 nm, holographic recording measurements are made possible by the interferential laser beams generated by a linearly polarized He–Ne laser (632.8 nm) as writing beams and a linearly polarized double-frequency Nd:YAG laser (532 nm) as a probe beam to monitor the holographic grating dynamics.

2.3. Optical setup

Measurements of diffractive signals of transmission holograms are carried out using the system described in Fig. 2. The writing and reading beams irradiate at the center of a rotatable disk on which the sample is fixed. The diameter of the laser beams is ~0.3 cm. The power densities of writing beams are adjusted to 33 mW/cm^2, and that of the reading beam is set as 1 mW/cm^2 to reduce the possible transformation from PMC to SO molecules. The intersecting angle between the red beams is ~9.8°. A UV source is used to irradiate the sample (Hayashi LA-410, 25 mW/cm^2). The first order diffraction can be observed clearly. I_1^+ appears on a screen, patterns of which can be captured by a digital camera. Points A, B and C are the positions on which the diffractive signals will project. I_1^- is synchronously registered on a photodiode interfaced with a computer.

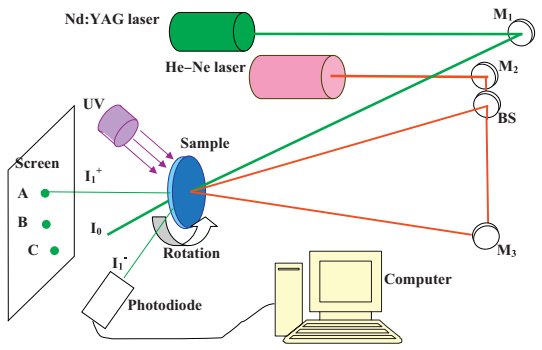

Fig. 2. Scheme of the holographic setup. M, mirror; BS, beam splitter.

3. Results and discussion

3.1. Multiple holographic gratings kinetics

The film was exposed to UV irradiation until maximal coloration was observed. Then, with the UV source still on, the He–Ne lasers were turned on for 5 s, and then the sample was rotated 30° rapidly, the writing beams irradiated the film for 10 s, then the sample was rotated another 30° immediately and the sample was irradiated for 15 s. Kinetics of the diffractive efficiency for each holographic grating were all recorded by the photodiode, as shown in Fig. 3(a). Meanwhile, the diffractive patterns of the multiple holographic gratings captured at 5 s, 15 s and 30 s were shown in Fig. 3(b).

The diffraction efficiency of the first grating increases sharply. At 5 s, the first rotation of the sample results in the remove of the first diffractive signal from Point A to Point B. Meanwhile, the photodiode begin to record the dynamics of the second grating. It is obvious that the growth rate and the maximal diffraction efficiency of the latter grating are both lower than that of the former. At 15 s, the second rotation is carried out. The diffractive signals at Points A and B remove to Points B and C, respectively. The growth rate and the maximal diffraction efficiency of the third grating become further lower. However, the diffractive signal intensity of the earlier recorded grating decreases during the recording of the later one. At 30 s, diffractive signal intensities for the three sets of the holographic gratings almost reach the same value.

To better understand the multiple holographic grating formation mechanisms, the single holographic grating dynamics was also recorded. As shown in Fig. 4(dotted), with UV irradiation, the

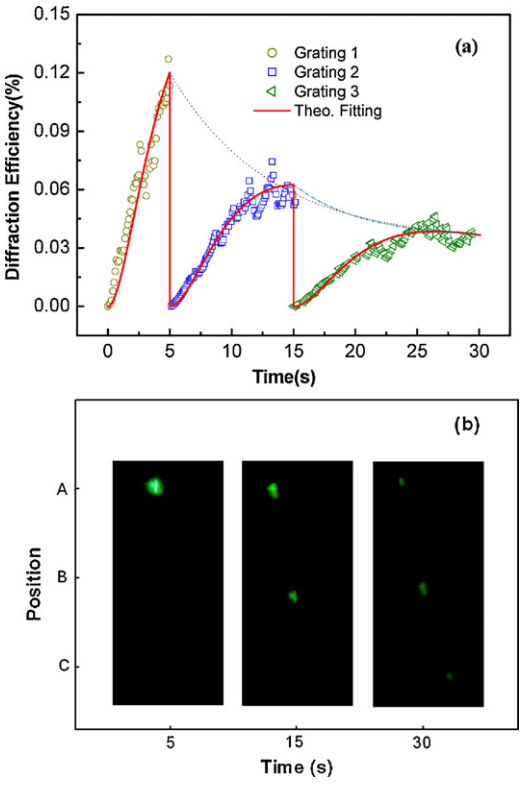

Fig. 3. (a) Diffractive signal kinetics of the multiple holographic gratings with the peristrophic multiplexing. The dots are the experimental measurements. The solid line is the theoretical fit. (b) Real-time diffractive signal patterns at 5, 15 s and 30 s.