Fabrication of the two-dimensional structure on dye-doped polymer film by multiple holographic recording

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ABSTRACT

The two-dimensional surface relief gratings (2D-SRGs) on azo-dye doped polymer film were fabricated by the two writing stages of holographic technique. The grooves structure of 2D-SRGs was depend on the rotational angle of two writing stages and the depth of grooves could be enhanced about 2~3 times by using the interface of nematic liquid crystals. The modulations of grooves depending on the polarization of writing beams were also disccussed. As the rotational angle between two writing stages was increased, the depth of grooves was gradually decreased for the S-polarization of writing beams; while it was increased for the P-polarization of writing beams.

Keywords: surface relief grating, grooves structure, rotational angle, dye-doped polymer film, nematic liquid crystals

1 INTRODUCTION

Azo polymer materials have revieved much attention in the recent years because of their potential applications in optical elements[1], optical data storage[2], optical switching devices[3,4] and photoalignment. Todorov's group demonstrated that the surface relief gratings were formed on azobenzene polymer films by an interference pattern of linearly polarized lights[5]. The trans-cis photoisomerization of azobenzene groups introduces the alignment of molecular long axis in the direction perpendicular to the polarization of the incident beam. The reorientation of azobenzene groups induces the large scale molecular motion and results in the expansion of irradiated azopolymer thin film. The formation of surface relief grating produces a preferred orientation to the overlying liquid crystal molecules. The alignment along the groove is due to the minimization of elastic strain energy[6].

The two-dimensional surface relief gratings can be fabricated by using multiple holographic recording[7]. The structure is highly symmetric and identical. It could be used as a template for fabricating well-ordered TiO_2 nanostructures[8]. S. Choi's group created the 2D-SRGs on fiber surface end which could be employed as beam splitter, beam deflector, diffractive lens, etc[9]. In the pervious studies, it took longer time to fabricate the SRGs by using the CW laser[10~12]. However, the SRGs could be quickly fabricated by using the pulse laser operated at single pulse[13].

In this study, we report the fabrications of 2D-SRGs on azo-dye doped polymer film by two-steps writing process of holographic technique. The 2D-SRGs based on the azo-dye doped polymer film with the interface of nematic liquid crystals were discussed. The dependence of surface modulation on the rotational angle between two writing stages were under investigation. The atomic force microscope(AFM) was employed to observe the surface modulations of 2D-SRGs. The study of two-dimensional surface relief gratings can lead to the potential applications in bistable alignment.

2 EXPERIMENT

The polymer solution was prepared by dissolving azo dye (DR1) of 4% weight concentration and polymethyl methacrylate (PMMA) of 96% weight concentration into toluene solvent. The prepared polymer solution was then spin-coated onto a glass substrate. The thickness of the resultant film was measured about 4 μm. In this experiment,

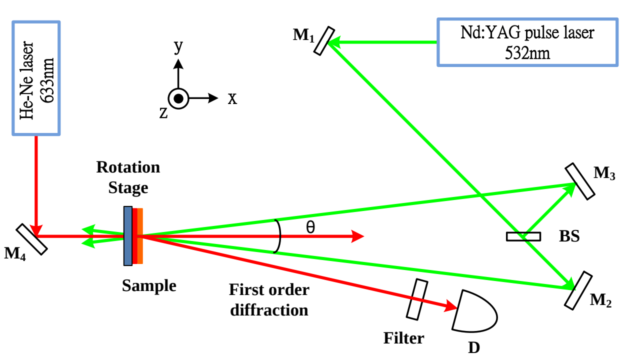

Figure 1：The experimental setup used to fabricate 2D-SRGs. M：mirrors, BS：50-50 beam splitter, D：detector, θ：interference angle.

the polymer sample was assembled by two glass substrates. One glass substrate was coated with dye doped polymer film (DDPF) and the other was coated with polyimide (PI) layer, which was rubbed for homogeneous alignment. The cell gap was controlled by two teflon spacers with the thickness of 12 μm. For the purpose of comparison, another cell was fabricated without the injection of the liquid crystals. The experimental setup for the fabrication of 2D-SRGs on the DDPF is shown in Fig. 1. The surface relief gratings were formed by two equal intensity of coherent writing beams from the pulsed Nd:YAG laser operated at 532 nm with the pulsed width of 6 ns, and the energy density of 0.2J/cm^2 for each beam. The interference pattern was produced by the writing beams with S-polarization (parallel to the *z*-direction). The grating pitch Λ corresponding to the interfringe spacing of the interference pattern is given by

$$\Lambda = \frac{\lambda}{2\sin\frac{\theta}{2}} \tag{1}$$

where λ is the wavelength of the Nd:YAG pulsed laser and θ is the incidence angle of the writing beams onto the sample. The angle between two writing beams is 6^o and the periodicity of the grating is about 5μm from equation (1). The He-Ne laser (633nm) with unpolarization was introduced straight to the sample to serve as the probe beam.

In order to fabricate the 2D-SRGs, the sample was fixed in a rotation stage, which could be rotated around the *x*-axis. After the first recording, the rotation stage was rotated an angle φ then the second grating was recorded again. The two-dimensional grating structues were formed by superimpoising one grating onto another grating[14,15]. The structure and the depth of two dimensional surface relief gratings were measured by using the atomic force microscope (AFM).

3 RESULTS

3.1 Dye-doped polymer film (DDPF)

The 2D-SRGs were fabricated with the various angle φ of 30^o, 45^o, 60^o, and 90^o between two-steps writing on the DDPF sample and their images were measured by AFM, as shown in Fig. 2(a)~2(d), respectively. Two gratings were overlapped to form a network like structure in Fig. 2(a)~2(c) and an egg-crate like structure in Fig. 2(d). In order to realize the orientation dependence of surface modulation under the second writing, the surface modulation of the first grating was chosen about 425nm. The surface modulations of the second recording at various angle of 30^o, 45^o, 60^o, and 90^o were measured about 368, 378, 377, and 380nm, respectively. The first-order diffraction efficiencies of various structure corresponding to 2(a)~2(d) were measured about 0.78, 1.0, 0.92, and 0.88% .

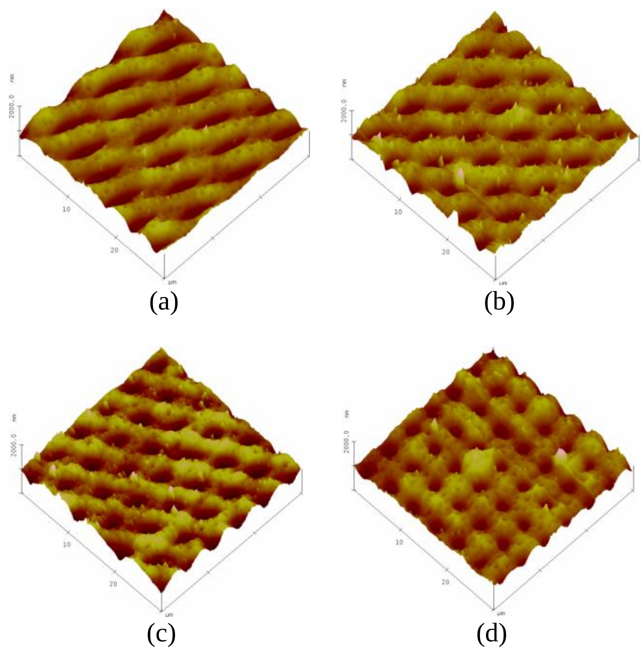

Figure 2 : The AFM images of 2D structure on DDPF for the various angle. (a) 30^o、(b) 45^o、(c) 60^o and (d) 90^o.

The surface modulation for the first recording was about 1.14 times than that for second recording.The surface modulation depends on the quantities of azo dye molecules from trans to cis state. At the first recording, some azo dye molecules transfer from trans to cis state and the residual trans molecules become less. At the second recording, the depth of surface modulation was smaller than that of the first recording due to less photoisomerization. The first-order diffraction efficiency and the surface modulation of SRGs under the second writing were dependent on the rotation angle between two steps writing for DDPF, as shown in Fig. 3. The surface modulations of SRGs under

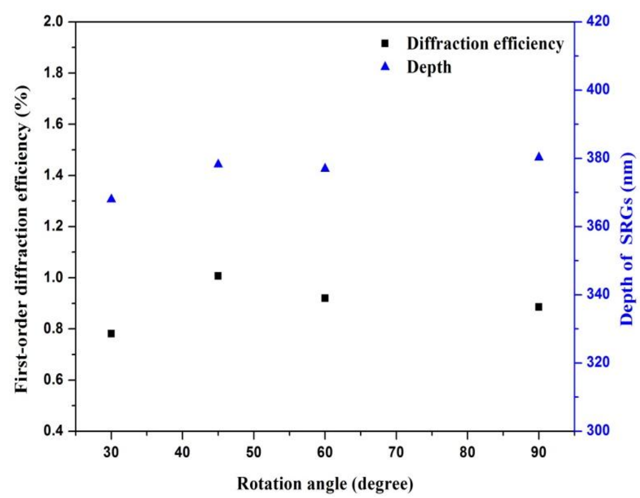

Figure 3 : The dependence of the first-order diffraction efficiency and the depth of surface relief on the rotation angle for DDPF samples in the second writing.

the second writing fluctuated between 365nm and 380nm when the rotation angle was increased. It can be concluded that the first-order diffraction efficiency and the depth of surface modulation under the second writing were independent of the rotational angle.

3.2 DDPF with the interface of nematic liquid crystals (NLC/DDPF)

The AFM images of 2D-SRGs on the NLC/DDPF at the various rotation angle φ of 15^o, 30^o, 45^o, 60^o, 75^o, and 90^o were shown in Fig. 4(a)~4(f). With the interface of liquid crystals, the surface of relief structures were more smooth comparing with that of DDPF. The depth of surface relief structure was fixed about 1.35μm for the first writing, which was around three times larger than that of DDPF. The surface modulations at the various rotation angle φ of 15^o, 30^o, 45^o, 60^o, 75^o, and 90^o were about 1.24, 1.13, 1.07, 0.95, 0.89, and 0.75μm, respectively, and the first-order diffraction efficiencies were about 3.16, 2.76, 2.65, 2.38, 2.06, and 1.79% , respectively, for the second writing. The result also shows that the depth of surface relief structure for the first writing is larger than that of the second writing.

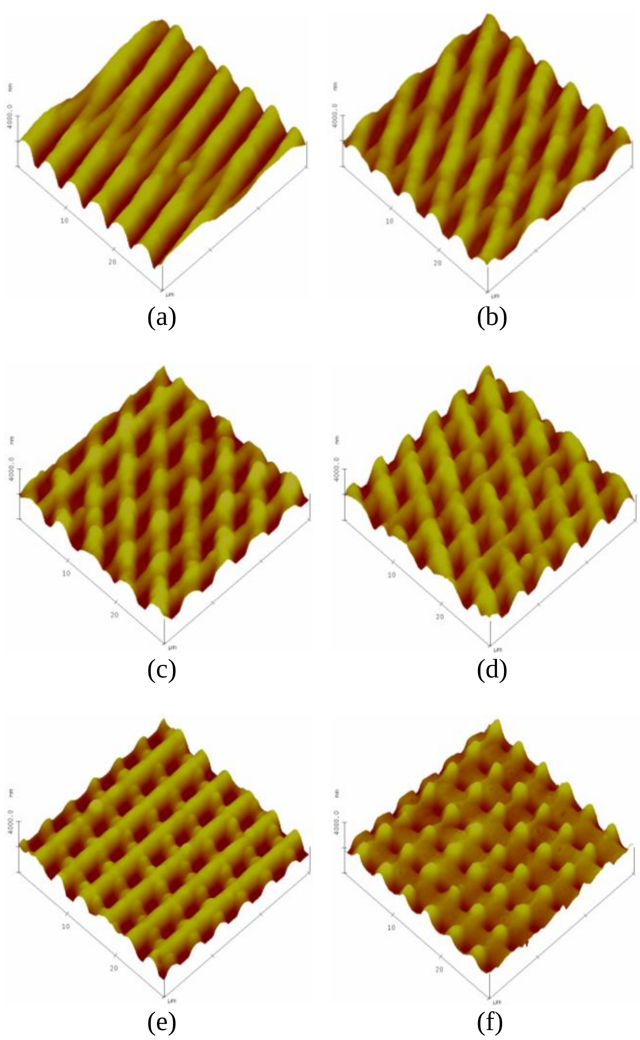

Figure 4 : The AFM images of 2D structure on NLC/DDPF for the various angle. (a) 15^o、(b) 30^o、(c) 45^o、(d) 60^o、(e)75^o and (f)90^o

The dependence of the first-order diffraction efficiency and the depth of surface relief gratings on the rotation angle were shown in Fig. 5 for the second writing with S-polarization on NLC/DDPF. Both diffraction efficiency and depth of SRGs were gradually decreased as the rotation angle was increased. The possible reason is due to the change of molecular director of nematic liquid crystals. Two geometric configurations were defined to discuss the result. In the parallel geometrical configuration, the director of nematic liquid crystals was parallel to the polarization of writing beams, and the perpendicular geometrical configuration, the director of nematic liquid crystals was perpendicular to the polarization of writing beams. In the first writing, the SRGs was formed in the parallel geometrical configuration. The direction of grooves were parallel to the orientation of nematic liquid crystals when the formation of surface relief gratings was completed. The liquid crystal molecules on the SRGs were along the original direction. Therefore, in the second writing, the configuration of the polarization of writing beams and the direction of nematic liquid crystals were transformed from parallel to perpendicular when the rotational angle increases. In the case of parallel configurations, the strong interaction force between the azo dye and the liquid crystal molecules led to the higher diffraction efficiency and depth of SRGs.

The dependence of the first-order diffraction efficiency and the depth of surface relief gratings on the rotation angle in the second writing with P-polarization for NLC/DDP was shown in the inset of Fig. 5. As the rotation angle was increased, both the first-order diffraction efficiency and the

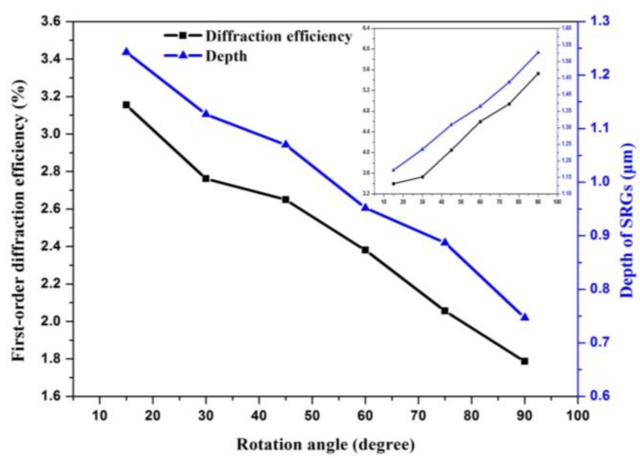

Figure 5 : The dependence of the first-order diffraction efficiency and the depth of surface relief gratings on the rotation angle in the second writing with S-polarization for NLC/DDPF. Inset is for the second writing with P-polarization.

depth of surface relief structure were gradually increased. In the first writing, the SRGs was formed under the parallel geometrical configuration and the direction of grooves were perpendicular to the orientation of nematic liquid crystals. In the first writing, the SRGs was formed under the parallel geometrical configuration and the direction of grooves were perpendicular to the orientation of nematic liquid crystals. Due to the surface anchoring energy, the liquid crystal molecules were reoriented along the direction of grooves. Therefore, when the rotational angle was increased, the geometrical configuration of the polarization of writing beams and the direction of nematic liquid crystals was transformed from perpendicular to parallel which led to the gradual increase of depth of SRGs in the second writing.

4 CONCLUSION

In conclusion, we demonstrate that the 2D-SRGs structure can be fabricated easier by using pulsed laser with two steps writing. The morphology and depth of 2D-SRGs become more smooth and deep by using the nematic liquid crystals as an interface. The first-order diffraction efficiency and the depth of SRGs under second writing gradually decay for the writing beams with S-polarization, and increase for the writing beams with P-polarization as the rotation angle increas, respectively. The results indicated that the polarization of writing beams and the directir orientation made a significant influence on the diffraction efficiency and the depth of SRGs. The depth of surface modulation was deep when the polarization of writing beams were parallel to the direction of nematic liquid crystals. These structures of 2D-SRGs could be utilized further in bistable device such as the substrate of bistable liquid crystal alignment.

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