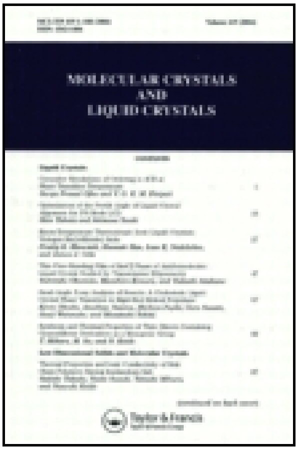

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The First Order Diffraction of Dye-Doped Polymer Film Assisted with Nematic Liquid Crystals

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The First Order Diffraction of Dye-Doped Polymer Film Assisted with Nematic Liquid Crystals

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The dynamic behavior of surface relief grating of dye-doped polymer film has been investigated. The dependence of the first-order diffraction efficiency on the concentration of dye doped polymer film is studied. The first-order diffraction efficiency of surface relief grating of dye-doped polymer film can be enhanced through the use of nematic liquid crystals as the interface. The surface modulations of relief gratings measured by the atomic force microscopy were compared for both dye-doped polymer films in contact with or without liquid crystals.

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INTRODUCTION

In the last decade, thin films of polymers containing azobenzene chromophores have become attractive optical recording materials for a variety of applications, including optical data storage, optical switching, and electro-optical devices. Birefringence of these thin films can be optically induced by the polarized light through photoisomerization of the azobenzene group [1]. The process involves *trans-cis* photoisomerization of azobenzene groups and thermal *cis-trans* relaxation, and introduces the alignment of azobenzene groups in the direction perpendicular to the polarization of the incident light. Thus, the reorientation

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of azobenzene groups induces the large-scale molecular motion and the free volume requirements [2]. This geometrical transformation results in the expansion of irradiated azopolymer thin films [3].

The fabrications of surface relief gratings on thin azobenzene polymer films were reported by using the light-induced mass transport process [4–8]. The irradiation of these films with an interference pattern of coherent light can induce not only the alignment of the azobenzene chromophores throughout the volume of polymer but also the controlled surface modification of the polymer film [4–6]. The formation of in-plane anisotropy in the polymer produces a preferred orientation to the overlying liquid crystal molecules [9]. The alignment along the groove direction is due to the minimization of the elastic strain energy of liquid crystals [10]. The aligning properties of liquid crystal molecules on the surface relief gratings (SRGs) have been also studied [11–13].

In this work, the surface relief grating of dye-doped polymer film is reported. The dependence of the first-order diffraction efficiency of the surface relief grating on the concentration of dye doped in the polymer thin films is studied. The diffraction efficiency of the surface relief grating can be enhanced through the use of nematic liquid crystals as the interface. The morphologies of inscribed surface relief on dye-doped polymer films were observed by atomic force microscopy.

EXPERIMENTAL

The sample structure with optical pump and probe system is schematically shown in Figure 1. The azo-dye DR-1 doped PMMA (polymethyl

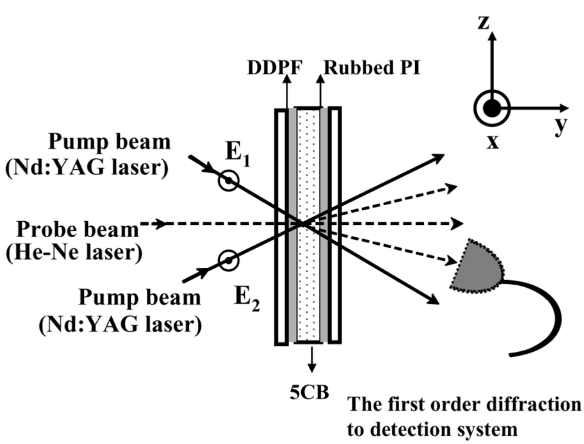


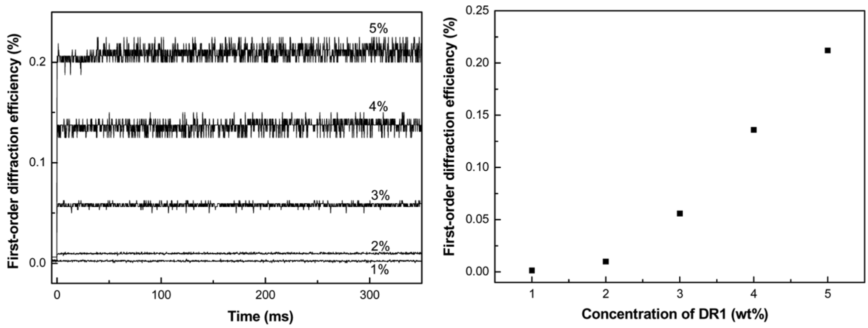
FIGURE 1 Experimental setup and the configuration of sample cell.

methacrylate) film was prepared by the spin-coating method using a toluene solvent at a weight concentration of $1 \sim 5\%$. The solution was filtered through a 0.2 μm Teflon filter and cast onto a glass substrate. The resultant film was dried in the vacuum oven at 80°C for 48 hours in order to remove residual solvent completely. The thin layer of polyimide (PI) was spin-coated onto another glass substrate and rubbed with a velvet cloth unidirectionally (parallel to x-axis initially). The sample cell was assembled from a pair of glass substrates coated with DR-1 doped PMMA and PI films, respectively, and spaced by a pair of 25-μm-thick Teflon sheets. Nematic liquid crystals (5CB) were capillarily filled into the sample cell. The experimental setup has been described in details elsewhere [14]. Briefly, a Q-switched Nd:YAG laser with a BBO frequency-doubling crystal was employed for the pulsed excitation beam at 532 nm. Two S-polarized pump beams split from the excitation beam with a beam splitter were introduced and crossed inside the sample cell. The He-Ne laser with S-polarization was served as a probe beam and incident normally into the sample. The surface relief grating was achieved by a pair of pump beams operated with a single shot at room temperature. The first-order diffracted probe beam was isolated from the pump beams with the glass filter and pinhole system. The diffraction efficiency was measured by a photodiode and recorded as a function of time with a digitizing storage oscilloscope.

RESULTS AND DISCUSSION

The holographic gratings of dye-doped PMMA film (DDPF) were performed with various concentrations of azobenzene dye of $1 \sim 5\%$ in the absence of liquid crystals inside the sample cell. The first-order diffraction efficiencies of gratings for DDPF were recorded as a function of time, as shown in Figure 2(a). Initially, the diffraction efficiencies rise sharply for all curves of various concentrations of dye on irradiation of the single pump pulse, and subsequently the diffraction efficiencies almost maintain at the constant levels. In general, the photoabsorption of dye increases with the concentration and results in the stronger photoinduced reorientational effect. Therefore, the difference of refractive index Δn between the bright and dark field of interference increases with concentration of dye, and the saturation value of the first-order diffraction efficiency also increases with the concentration of dye, as shown in Figure 2(b).

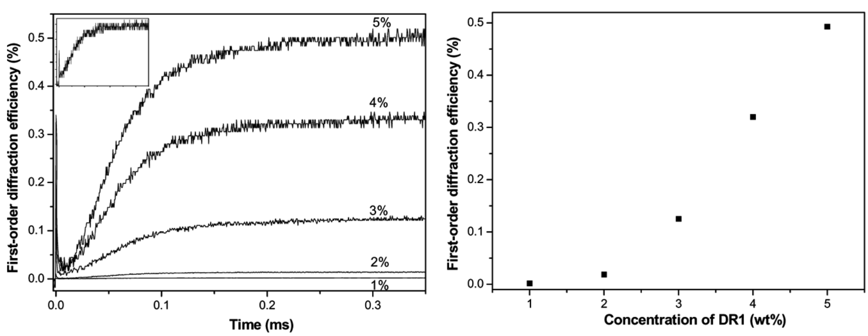
The holographic gratings of dye-doped polymer film in contact with liquid crystals (LC/DDPF) were investigated using the same experimental conditions as DDPF at room temperature. The first-order



(a) (b)

FIGURE 2 (a) The first-order diffraction efficiencies of DDPF as a function of time at various concentrations of dye, and (b) The first-order diffraction efficiencies of DDPF as a function of dye concentrations.

diffraction efficiencies of gratings for LC/DDPF were recorded as a function of time, as shown in Figure 3(a). The temporal profiles of diffraction efficiency for DDPF and LC/DDPF are obviously different. The diffraction efficiency of LC/DDPF exhibits the pattern of rise-decay-rise to a stable level for various concentrations of dye. The temporal profile of diffraction efficiency also exhibits the same pattern even at the low concentration of 1%, as shown in the inset of Figure 3(a). The possible cause of the decay of diffraction efficiency is due to the photothermal effect in the pumping region where the



(a) (b)

FIGURE 3 (a) The first-order diffraction efficiencies of LC/DDPF as a function of time at various concentrations of dye. The insert is the enlarged diffraction efficiency at concentration of 1%, and (b) The first-order of diffraction efficiencies of LC/DDPF as a function of dye concentrations.

inhomogeneous orientation of liquid crystals is temporarily induced by the photothermal fluctuation [14]. In the nematic phase, the ordering state of liquid crystals is very sensitive to the variation of temperature. The uniform orientation state of molecular director might be distorted due to photothermal fluctuation.

The saturation value of the first-order diffraction efficiency for LC/DDPF also increases with the concentration of dye, as shown in Figure 3(b). Although, the unavoidable photothermal fluctuation is occurred for LC/DDPF and results in the decay of diffraction efficiency due to the light scattering, the first-order diffraction efficiency of LC/DDPF is larger than that of DDPF. Comparing with the dye-doped polymer films only, the first-order diffraction efficiency of dye-doped polymer film in contact with nematic liquid crystals is almost the same as DDPF at the concentration of 1% and is enhanced about 50% at the concentration of 2%. Above the concentration of 3%, the first-order diffraction efficiency of LC/DDPF is around 2.3 times as large as that of DDPF. With the assistance of nematic liquid crystals as the interface, the enhancement factor of the first-order diffraction efficiency of dye-doped polymer film increases with the concentration of dye at low value of concentration and then reaches the constant level at certain value of concentration. Thus, the first-order diffraction efficiency for dye doped polymer film can be enhanced through the use of nematic liquid crystals as the interface with the appropriate amount of dye concentration.

The morphologies of inscribed surface relief for dye-doped polymer film (dye concentration of 5%) in contact with or without liquid crystals were observed by using the atomic force microscopy. The relief grating of DDPF was recorded with a spatial period of 13 μm and

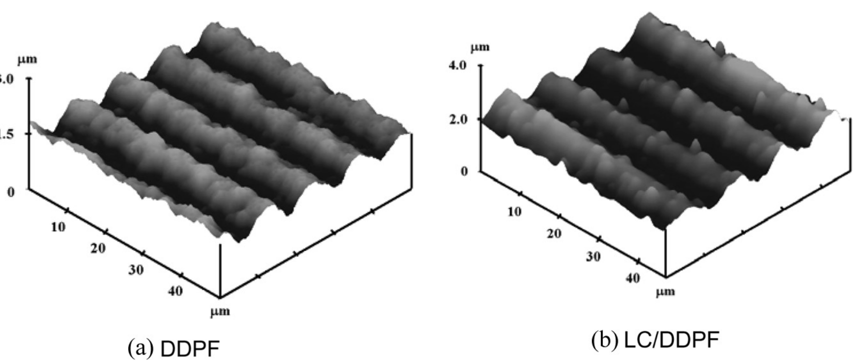


FIGURE 4 (a) The morphology of inscribed surface relief grating of DDPF, and (b) The morphology of inscribed surface relief grating of LC/DDPF.

the surface modulation of 622 nm, as shown in Figure 4 (a). However, the relief grating of LC/DDPF was recorded with the surface modulation of 1311 nm, as shown in Figure 4 (b). The surface modulation of LC/DDPF is about two times as large as that of DDPF. The first-order diffraction of DDPF and LC/DDPF are 0.212 and 0.493%, respectively. The ratio of surface modulation is almost consistent with the ratio of the first-order diffraction efficiency between DDPF and LC/DDPF. The photoisomerization of azo-dye molecules in polymer film induces the change of surface properties. The light-induced reaction can be amplified and stabilized by incorporating with the nematic liquid crystal layer. Therefore, the depth of surface modulation for LC/DDPF depends on the concentration of azo-dye.

CONCLUSIONS

In conclusions, the dynamic behavior of surface relief grating for LC/DDPF has been investigated by means of the holographic method associated with the pump-probe technique. The measurements of the first order diffraction efficiency of dye-doped PMMA film in contact with or without nematic liquid crystals were performed with various concentrations of dye. Comparing with DDPF, the first-order diffraction efficiency of LC/DDPF can be enhanced as large as around 1.3 times with an appropriate amount of dye concentration and shows the temporal pattern of rise-decay-rise to a stable level.

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