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Temperature and orientation dependence of surface relief gratings based on dye-doped polymer film with the interface of nematic liquid crystals

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ABSTRACT

The formation of surface relief grating on dye-doped polymer film with the interface of nematic liquid crystals has been investigated by means of the holographic technique. The first-order diffraction efficiency of surface relief grating depends on the temperature and the orientation of molecular director in the interface of nematic liquid crystals. The diffraction efficiency is roughly independent of thermal fluctuations of molecular director in the most part of nematic temperature range and apparently drops near the transition temperature. The morphology of surface relief grating demonstrates that the surface modulation is larger for molecular director parallel to the groove direction. The experimental result also shows that the first-order diffraction efficiency is dependent on the surface modulation of surface relief grating.

1. Introduction

Thin films of polymers containing azobenzene chromophores have received much attention in the last decade because of their potential applications in optical data storage, optical switching, and electro-optical devices. Todorov's group demonstrated that the birefringence gratings could be fabricated on the azobenzene-containing polymers with the interference pattern of linearly polarized light [1]. The optical printing has also been accomplished by the use of various polymer matrices including guest-host [1], liquid crystalline [2–5], and amorphous polymers [6–8]. The photoisomerization of azobenzene groups under illumination of polarized laser introduces the alignment of molecular axis along the direction perpendicular to the polarization of incident light. The reorientations of chromophore generate the massive motion of the polymer material and the corresponding free volume requirements [9]. This geometrical transformation results in the expansion of irradiated azopolymer thin films.

The fabrications of surface relief gratings on thin azobenzene polymer films were reported by using the light-induced mass transport process [10–14]. The driving force of mass transport is due to the pressure gradients originated from the interfering light and unequal isomerization. During the spatially periodic photoisomerization, the increase in pressure gradients induces the polymer flow and generates the spatial modulation on the polymer film. 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 but also the controlled surface modification of the polymer film [10–12]. The formation of in-plane anisotropy in the polymer produces a preferred orientation to the overlying liquid crystal molecules [15]. Due to the minimization of the elastic strain energy, the liquid crystal molecules are reoriented along the direction of grating grooves [16–19].

The feasibility of increasing the first-order diffraction efficiency of surface relief grating has been demonstrated by using the holographic grating technique with the interface of nematic liquid crystals [20]. In this work, we report the dependence of the first-order diffraction efficiency on the molecular director and the temperature in the nematic phase. The surface modulation of relief gratings measured by the atomic force microscopy will be compared with the first-order diffraction efficiency between parallel and perpendicular orientation of molecular director relative to the grooves of surface relief grating.

2. Experimental

The polymer solution was prepared by dissolving azo dye of 4% weight concentration (DR1, purchased from Sigma Aldrich) and polymethyl methacrylate (PMMA, $M_w = 40{,}000$) of 96% weight concentration into toluene solvent. The absorption spectrum with maximum absorption around ~500 nm and the chemical structure of DR1 are shown in Fig. 1. The prepared polymer solution was then spin-coated onto a glass substrate. The polymer film was dried up in the vacuum oven at temperature of 80 °C for 48 hours in order to

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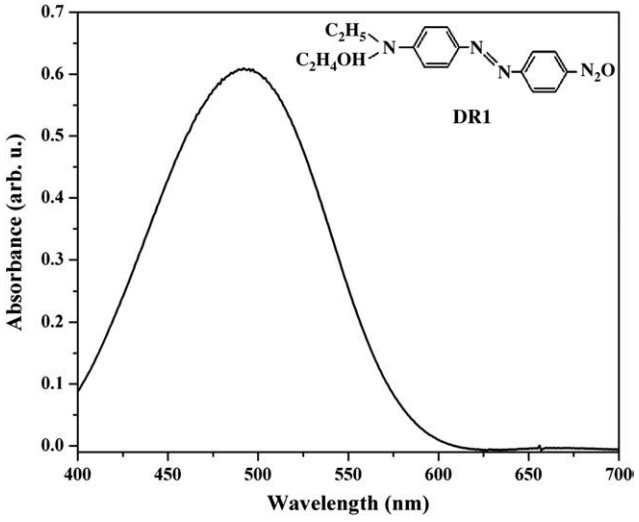

Fig. 1. The chemical structure and the absorption spectrum of DR1.

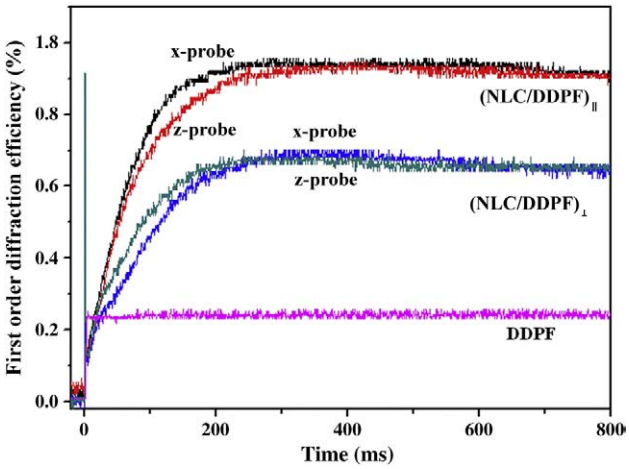

Fig. 3. Time evolutions of the first-order diffraction efficiency for DDPF and NLC/DDPF in either parallel or perpendicular configuration at room temperature.

remove residual solvent completely. The thickness of the resultant film was measured about 4 μm. The glass transition temperature (Tg) of DR1-PMMA is around 90 °C [21]. In our experiment, the polymer sample was assembled by a pair of glass substrates, as shown in Fig. 2. One glass substrate was coated with DR1-PMMA film (DDPF) and the other was coated with polyimide (PI) layer, which was rubbed unidirectionally for homogeneous alignment of liquid crystal molecules. The cell gap was controlled by two teflon spacers with the thickness of 25 μm. The nematic liquid crystals pentyl-cyanobiphenyl (5CB, clearing point ~35 °C) in the isotropic phase was then capillarily filled into the empty cell. For the purpose of comparison, another cell was fabricated without the injection of the liquid crystals.

The experimental setup for the fabrication of surface relief grating on the DR1-PMMA film is illustrated in Fig. 2. The DR1-PMMA cell was placed inside a temperature-controlled chamber operated in the range from 27 to 35 °C. The surface relief gratings were performed 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, the repetition rate of 10 Hz, and the energy density of 4.5 mJ/cm^2. The polarizations of writing beams are fixed in the *x*-direction. The angle between two writing beams is 3° and the resultant grating spacing is approximately 10 μm. The cw He-Ne laser with polarization either in the *x*- or *z*-direction was introduced normal to the sample to serve as the probe beam. The surface modulations of DR1-PMMA were achieved with a single shot of a pair of writing beams at a fixed temperature. The first-order of the diffracted probe beam was isolated from the writing beams with the glass filter and pinhole system. The diffraction efficiency was detected with a photodiode and recorded as a function of time with a digitizing storage oscilloscope.

In order to investigate the dependence of diffraction efficiency on the orientation of nematic liquid crystals, the measurement of the first-order diffraction efficiency has been performed in two geometric configurations of dye-doped polymer film in contact with nematic liquid crystals (NLC/DDPF). In the parallel geometrical configuration, the director of nematic liquid crystals was arranged in the direction of *x*-axis and, correspondingly, parallel to the polarization of writing beams. In the perpendicular geometrical configuration, the director of nematic liquid crystals was arranged in the direction of *z*-axis and, correspondingly, perpendicular to the polarization of writing beams.

3. Results and discussion

The dynamics of formation of surface relief gratings on the film of DDPF with and without the interface of liquid crystals was studied through the real-time probing by using the holographic technique. The first-order diffraction efficiencies of surface relief gratings were recorded as a function of time for DDPF and NLC/DDPF in either parallel or perpendicular configuration at room temperature, as shown in Fig. 3. The surface relief gratings of DDPF and NLC/DDPF were generated immediately with the single pulse of writing beams. The DDPF grating showed a stable diffraction efficiency of ~0.2%. The saturated diffraction efficiencies of NLC/DDPF gratings were around 0.94% for parallel configuration and 0.64% for perpendicular configuration. Both NLC/DDPF gratings showed the higher diffraction efficiency than the DDPF grating. However, the diffraction efficiencies of NLC/DDPF gratings exhibited the dramatic dip in a short period (t = ~180 ms) as soon as the formation of surface relief gratings was completed. The ordering state of nematic liquid crystals is very sensitive to the variation of temperature in the nematic phase. The temporarily heterogeneous orientation of nematic liquid crystals could be easily induced by the photothermal effect in the pumping

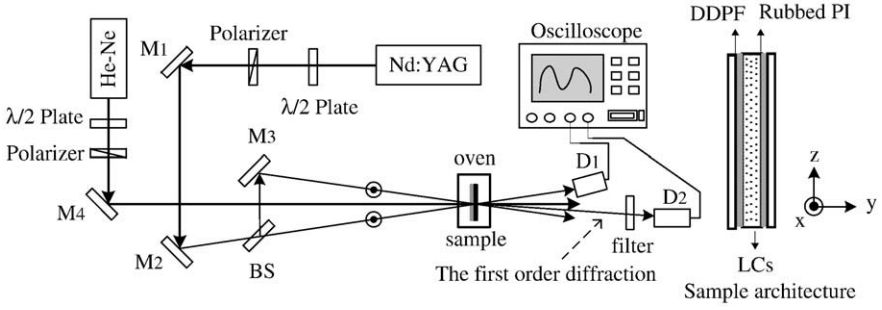

Fig. 2. The experimental setup and the configuration of sample cell.