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Biphotonic recording effect of polarization gratings based on dye-doped liquid crystal films

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This study investigates a holographic recording effect of biphotonic polarization gratings (BPGs) written on dye-doped liquid crystal (DDLC) films. One linearly polarized green light (λ_G=514.5 nm) and a polarization-modulated interference pattern formed by two mutually coherent orthogonal ($\pm 45^\circ$ with respect to the polarization of the green light) polarized red lights simultaneously excite the DDLC films to generate a BPG. The formation of BPGs depends primarily on the dichroism of the dye molecules and a sequence of mechanisms: photoisomerization, anisotropic adsorption and inhibition of dye adsorption. The *cis*-isomer absorbance-modulated distribution associated with the red polarization-modulation pattern induces the dye adsorption-modulated pattern, in turn, yielding the permanent BPG which generates a modulated twisted nematic (TN) structure pattern in the sample. It is found that each BPG is verified to be electrically switchable and thermally erasable. The switching time is in the order of milliseconds. Additionally, the recording time to form a BPG decreases with increasing intensity of the green pump beam.

1. Introduction

Holographic gratings (HGs), including intensity gratings (IGs) and polarization gratings (PGs), have been intensively examined over the last decade because of their participation in interesting physical mechanisms and their potential applications in optics. Various optical materials, e.g. azo materials and polymers, have been studied in this field [1–5]. Photoinduced anisotropic adsorption is important in the formation of these HGs [6–12]. Voloschenko *et al.* first addressed the adsorption effect in homogeneously aligned dye-doped liquid crystal (DDLC) films [6]. Simoni and Francescangeli reported on this effect in similar systems [8]. Slussarenko *et al.* presented a PG in a DDLC cell [2]. Fuh *et al.* recently developed IGs and PGs in not only DDLC films but also dye-doped PDLC films [4, 5, 11].

Although a few biphotonic IGs and PGs have been recently been developed for use in azo/polymer systems, most of the researchers cited above have demonstrated and exploited the photoinduced molecular reorientation effect based on the HGs in DDLC films using a single-colour (generally, green light) pump beam [13–18]. Particularly, Lee *et al.* exploited the photoinduced anisotropic adsorption effect of the biphotonic IGs using green and red writing beams in DDLC films [19]. This report, however, presents biphotonic polarization, rather than intensity, gratings in homogeneously aligned methyl red (MR)-doped LC films. A red polarization-modulated interference pattern generated by the superposition of two coherent red lights linearly polarized at $\pm 45^\circ$ to the polarization of the green light, and one linearly polarized green light, simultaneously pumps and modulates the DDLC cells to yield a BPG. The formations of the BPGs are attributed to the dye dichroism, the polarization of light and a series of processes photoisomerization, anisotropic adsorption and inhibition of dye adsorption. The *cis*-isomer absorbance-modulated distribution produces the dye adsorption-modulated pattern, yielding the twisted nematic (TN) structure-modulated BPGs. The mechanism of the BPG formation is also verified experimentally using one red and one green light with different polarizations. The recording time of the BPG formation is found to decrease with the increase of the green pump-beam intensity. Additional experiments reveal that each formed BPG is electrically switchable and thermally erasable, and the switching time is in the order of milliseconds.

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2. Experiment

The materials employed in this experiment are a nematic liquid crystal, E7 (from Merck), and an azo dye, MR (from Aldrich). The dichroic ratio, D, defined as $A_{//}/A_{\perp}$, of MR is around six for visible light, where $A_{//}$ and $A_{\perp}$ are the dye absorbance when the pump-polarization is parallel and perpendicular, respectively, to the optical axis of the LCs in a homogeneously aligned MR-doped E7 cell. The mixing ratio of these two materials is 99:1 wt% of E7:MR. Each empty cell comprises two indium-tin-oxide (ITO) coated glass slides, which are separated by two 12 μm thick plastic spacers. One of these two ITO glass slides is coated with an alignment film, polyimide (PI, from Nissan), and rubbed in the direction **R**; the other is not. The mixed compound is then injected into the empty cell to yield a DDLC film. The homogeneous alignment of the DDLC cell is verified by conoscopy. The surface with (without) a rubbed PI film is called the reference (command) surface, denoted S_R (S_C). According to continuum elastic theory, the PI film with a strong anchoring force can align LCs parallel to **R** from S_R to S_C, even if S_C is not rubbed.

3. Results and discussion

Figure 1a presents the experimental setup of a BPG. One linearly polarized (in y-direction) green light, $\mathbf{E}_G$, derived from an Ar^+ laser (λ_G=514.5 nm) is a green pump beam. Two red pump beams, $\mathbf{E}_{R1}$ and $\mathbf{E}_{R2}$, linearly polarized at $\pm 45^\circ$ from **R** (along y-axis), are output by a high power He–Ne laser (λ_R=632.8 nm). The green light and the two red lights simultaneously pump a single region of the cell from the S_C and S_R sides, respectively. The angle of intersection of the two red lights is $\theta \sim 1^\circ$ and their incident bisector is normal to the surface of the DDLC film. The angle of incidence of the green light is 0°. The red lights have an equal intensity of $I_{R1,2}$=180 mW cm^{-2} and the green light has intensity I_G=2 mW cm^{-2}. None is focused. The two red writing beams are coherent and have orthogonal polarizations, so they produce an interference field ($\mathbf{E}_R$) that has a polarization-modulated pattern with 0° linear (with respect to x-axis), circular and 90° linear (with respect to x-axis) polarization of constant intensity in the intersecting region of the DDLC cell, as shown in figure 1b. After the DDLC cell is pumped for 2400 seconds, a BPG is formed, as confirmed using a polarizing optical microscope (POM). The first order diffraction efficiency of a BPG recorded using the conditions described above, probed by a linearly polarized ($\mathbf{E}_P$, along y-axis) He–Ne laser beam is about 0.7%. The diffraction efficiency is defined as the ratio of the intensity of the diffracted beam to that of the incident beam. Further, the polarization of the first-order diffracted beam is found to be perpendicular to that of the incident probe beam. It is reasonable since the formed BPG is a TN grating with the TN structure under the Mauguin condition. In addition, simulation using Jones matrix and Fourier optics is made. The simulated result verifies the experimental finding.

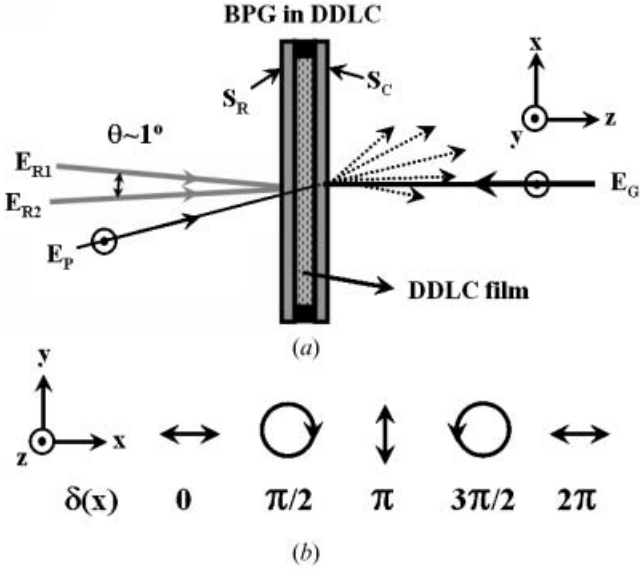

Figure 1. (a) Schematic diagram of the experimental setup for studying the BPGs in DDLC films; S_C: command surface, S_R: reference surface. The polarization of the green light, $\mathbf{E}_G$, is the y-direction. A weak red beam, $\mathbf{E}_p$, is utilized to probe the formed BPGs. (b) Configuration of the spatially modulated polarization states of the interference field created by two mutually orthogonal polarized He–Ne laser beams, $\mathbf{E_{R1}}$ and $\mathbf{E_{R2}}$. δ is the phase shift between $\mathbf{E_{R1}}$ and $\mathbf{E_{R2}}$ at z=0.

Figures 2a and 2b show the grating patterns observed under a POM with crossed and parallel polarizers, respectively. The transmission axis of the polarizer is set parallel to the grating stripes. The grating spacing (Λ) is measured to be around 32 μm, which value agrees with the theoretical grating spacing determined by the equation $\delta=(4\pi/\lambda)y\sin(\theta/2)=2\pi$. The grating patterns in figures 2a and 2b are complementary and appear to be organized by a TN structure-modulated pattern. Figure 2c presents the self-diffraction pattern of the red pump beams diffracted from the formed grating. The annotations on the figure refer to the nth-order diffraction signal of a BPG.

Azo dyes are known to be typically in the *trans*-state in the dark. According to Lee *et al.* [19], *trans*-MR dyes can absorb much more green light than red light. The green light-excited dyes can photoisomerize from the *trans* form to the *cis* form, diffuse and then be adsorbed by the inner surface of the DDLC cell. The anisotropically adsorbed dyes then reorient the LCs. However,

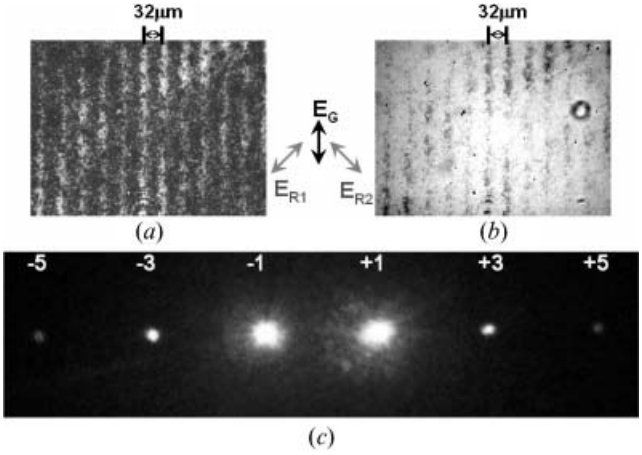

Figure 2. Formed BPGs with a twisted nematic (TN) structure-modulated pattern are observed under an optical polarizing microscope with (a) crossed and (b) parallel polarizers. The spacing of the grating is ~32 μm. The stripes of a BPG are parallel to the polarization of the green light. (c) Self-diffraction pattern of the red light from a BPG.

when simultaneously pumped with the red light, the *cis*-state dyes can immediately reverse to the *trans*-state, and adsorb less. Hence, the red light resists adsorption of the green light-excited dyes via *cis–trans* inverse isomerization. Therefore, in this work, the green light initially excites the *trans*-MR molecules to become *cis*-isomers uniformly throughout the irradiated region. The red interference pattern, with a polarization-modulated distribution with linear (0° with respect to the *x*-axis), circular and linear (90° with respect to the *x*-axis) polarization of constant intensity, then excites the *cis*-isomers. Theoretically, the *cis*-isomers have a minimum and maximum absorbance (α_{min} and α_{max}) at 0° and 90° (with respect to the *x*-axis) linear polarization, respectively, corresponding to the situations in which the long axes of the dyes are perpendicular and parallel to the polarization of the red exciting field. The mean absorbance α_{avg} in the regions of left- or right-circular polarization is between α_{min} and α_{max}. This is verified experimentally, and the results are shown in figure 3. The higher absorbance of *cis*-isomers in the red band corresponds to the stronger inhibition of dye adsorption. Therefore, the *cis*-isomer absorbance-modulated distribution induces the dye adsorption-modulated pattern, generating a TN structure-modulated grating.

Figure 3 shows the images observed under a polarizing optical microscope of regions in DDLC cells pumped in different conditions. In this part of experiment, one linearly polarized (in the *y*-direction) green light, $\mathbf{E}_G$, with an intensity ~2 mW cm^{-2} (Ar^+ laser, λ_G=514.5 nm) and one linearly or circularly polarized red light, $\mathbf{E}_R$, (He–Ne laser, λ_R=632.8 nm) with an

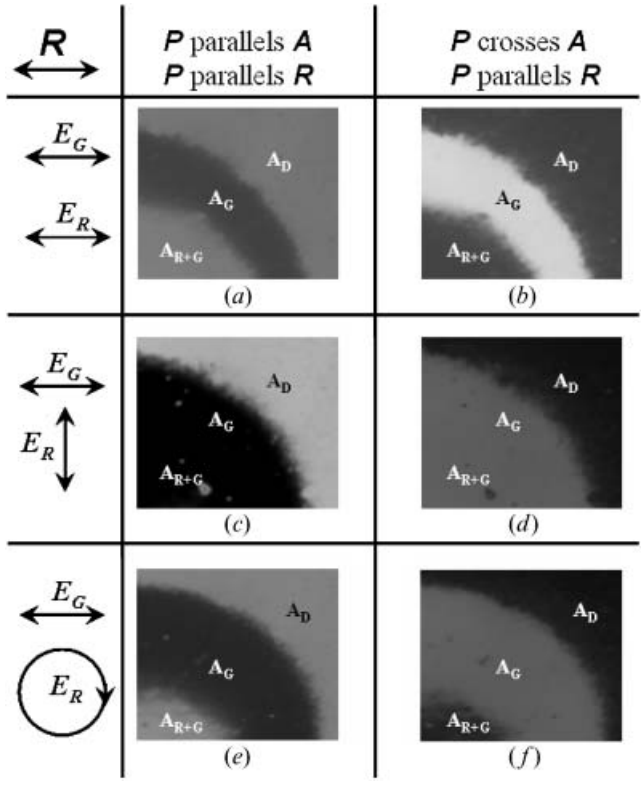

Figure 3. The images observed under a polarizing optical microscope of a region A_{R+G} (A_G) in a DDLC cell pumped by one linearly polarized ($\mathbf{E}_G$) green light (Ar^+ laser, λ_G=514.5 nm, I_G~2 mW cm^{-2}) with (without) simultaneous irradiation of one red light (He–Ne laser, λ_R=632.8 nm, I_R~180 mW cm^{-2}) with various polarizations ($\mathbf{E}_R$) for 2400 seconds; (a) $\mathbf{E}_G$//$\mathbf{E}_R$, and P//A, (b) $\mathbf{E}_G$//$\mathbf{E}_R$, and P⊥A, (c) $\mathbf{E}_G$⊥$\mathbf{E}_R$, and P//A, (d) $\mathbf{E}_G$⊥$\mathbf{E}_R$, and P⊥A, (e) $\mathbf{E}_R$ is a circular polarization, and P//A, (f) $\mathbf{E}_R$ is a circular polarization, and P⊥A. A_D represents the dark area, without being pumped. P and A are the polarizer and analyzer. **R** denotes the rubbing direction on S_R.

intensity ~180 mW cm^{-2} simultaneously illuminate the cell from the S_C and S_R sides, respectively, for 2400 seconds. A_{R+G} (A_G) is the region in a DDLC cell pumped by one linearly polarized green light ($\mathbf{E}_G$) with (without) simultaneous irradiation of one red light with various polarizations ($\mathbf{E}_R$). A_D represents the dark region, without being pumped. Figures 3a and 3b (figures 3c and 3d) [figures 3e and 3f] show the POM images of the pumped spot with $\mathbf{E}_G$//$\mathbf{E}_R$ ($\mathbf{E}_G$⊥$\mathbf{E}_R$) [$\mathbf{E}_R$ is a circular polarization] under parallel and crossed polarizers, respectively. It is clear to see that the region of A_{R+G} in figure 3d is a bright region (which is a TN structure), but that in figure 3b is a dark region. It means that the *cis*-isomer has a lower (higher) absorbance at 0° (90°) linear polarization condition, since the higher the absorbance of *cis*-isomer in the red band, the less the dye molecules adsorption on S_C.

Compare the region of A_{R+G} in figure 3b and in figure 3d with that in figure 3f; it clearly shows that the area of A_{R+G} in figure 3b (3d) is larger (smaller) than that in figure 3f. Thus, the mean absorbance α_{avg} of the *cis*-isomer with circularly polarized red light is higher (lower) than that with 0° (90°) condition.

The experimental data presented in figures 4 and 5 reveal that the BPGs formed are electrically switchable and thermally erasable. Figure 4 show that, when a 1 kHz electric field is applied, the diffraction signal from the BPGs decreases as the applied voltage increases, because the LCs are all aligned parallel to the applied field, and the TN structure-modulated grating is eliminated. After the applied voltage is switched off, the LCs return to their original state so the diffraction signal reappears, as shown in figure 4a. The switching times (10~90%) are measured to be τ_{rise}~1 ms and τ_{decay}~250 ms with the sample applied with an ac applied voltage V=60 V (1 kHz). The decay time is much longer than the rise time. It is due to the fact that as the ac applied voltage is switched off, the orientations of the LCs and MR molecules return to their initial state without any electric potential. In addition, a TN cell experiences a backflow effect after the applied high voltage has been switched off [20, 21]. The backflow gives an echo effect in the relaxation process, and increases the decay time.

Figure 5a reveals that the diffraction signal decreases as the temperature increases, and disappears when the temperature exceeds the clear point (about 65°C) of the DDLC materials. However, the diffraction cannot recover its initial value when the temperature returns to 25°C, indicating that the grating can be partially erased thermally. This work ascribes the thermally erasable BPGs to the thermal disturbance that causes desorption of dyes from S_C. Figures 5b and 5c depict the morphologies of a BPG before and after thermal treatment, respectively.

Finally, figure 6 gives the variation of the measured recording time of the BPGs with the intensity of the green pump beam. The recording time is defined as the time required for the first-order diffraction efficiency to reach 0.1% after the green beam is on. It is clear to see from figure 6 that the recording time decreases with

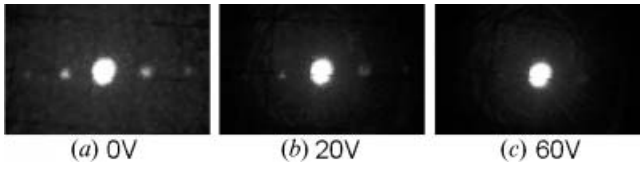

Figure 4. Diffraction patterns from a BPG probed by a He–Ne laser beam with an ac applied voltage (1 kHz) of (a) 0 V, (b) 20 V and (c) 60 V.

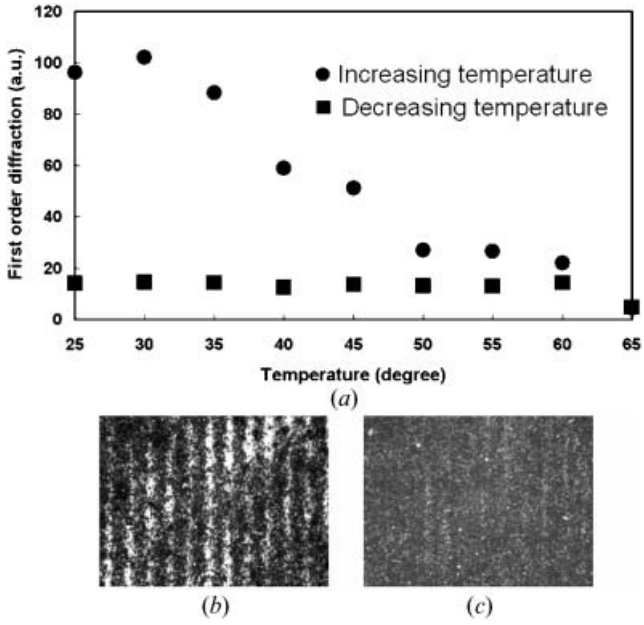

Figure 5. (a) Partially erasable characteristic of a BPG presented in a heating–cooling cycle; (b) and (c) are the morphologies of a BPG before and after thermal treatment, respectively.

increasing green pump-beam intensity. The reason is that a higher green pump-beam intensity induces a faster adsorption rate of the dyes. The adsorbed dyes align the LCs, and then generate a TN BPG.

4. Conclusions

In conclusion, this investigation proposes and studies a biphotonic recording effect of polarization gratings called BPGs that are based on DDLC films. The BPGs are written by the simultaneous irradiation of one linearly-polarized green light and a polarization-modulated red interference field, formed from two coherent and mutually orthogonal linearly-polarized (±45° with

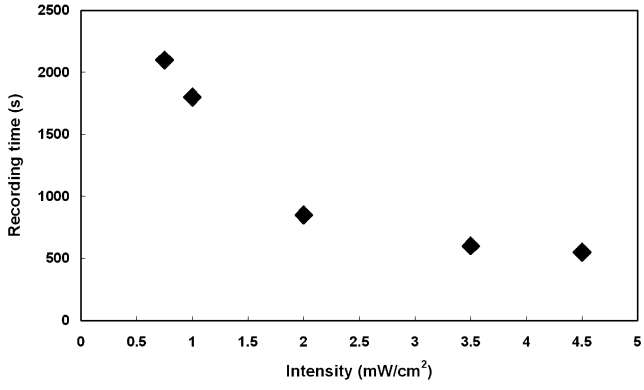

Figure 6. Variation of the recording time of BPGs with the intensity of the green pump beam.

respect to the polarization of the green light) red light. The BPGs are formed by the *cis*-isomer absorbance-modulated pattern, which suppresses the dye adsorption-modulated pattern, via the polarization-modulated interference field. The pattern generates the TN structure-modulated BPGs. The other experimental data also reveal that the BPG is electrically switchable and thermally erasable. The rise- and decay-time of a BPG are in the order of milliseconds. Moreover, it is also found that the recording time to form a BPG is dependent on the intensity of the green pump beam. The recording time is decreased with an increase of the green pump-beam intensity.

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