

# Optically switchable biphotonic gratings based on dye-doped cholesteric liquid crystal films

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(Received 2 March 2007; accepted 3 June 2007; published online 25 June 2007)

This study elucidates optically switchable gratings (BGs) based on biphotonic effect in dye-doped cholesteric liquid crystal films. When one circularly polarized green beam is switched on (off), the gratings can be turned on (off) by illumination with an interference field generated by two linearly polarized red beams. The biphotonic gratings are formed by two mechanisms—green-beam-induced dye reorientation through *trans-cis* isomerization and red-beam-induced suppression of dye reorientation by *cis-trans* back isomerization. These mechanisms result in a spatially periodic distribution with homeotropiclike and planarlike structures, respectively, in dark and bright regions of the interference field, generating the BGs. © 2007 American Institute of Physics.

[DOI: 10.1063/1.2752110]

Dye material-doped liquid crystals (LCs) have received considerable interest in the development of all optically controllable devices due to their large birefringence and ability to be flexibly controlled through interaction with photoexcited dyes.<sup>1–4</sup> In particular, biphotonic holographic gratings based on LCs combined with azo dyes have recently attracted increasing attention because their controllable elements have potential applications in integrated optics.<sup>5,6</sup> However, no investigation based on cholesteric liquid crystal (CLC) systems has been conducted. This work addresses the development of optically switchable biphotonic gratings (BGs) based on dye-doped CLC (DDCLC) films. The gratings can be switched on (off) by turning on (off) one green beam under irradiation with a red interference pattern. The mechanisms by which BGs are produced are green-beam-induced dye reorientation in the direction of the wave vector by *trans-cis* isomerization and red-beam-induced suppression of dye reorientation by *cis-trans* back isomerization. These mechanisms yield a spatially periodic distribution of homeotropiclike and planarlike textures, respectively, in the dark and bright regions of the red interference field, which creates the BGs. The diffraction efficiency of BGs depends markedly on the relative intensity of the green and red beams.

The nematic LC, chiral dopant, and azo dye employed herein are BL009 ( $n_0=1.5266$ ,  $\Delta n=0.2915$ ), CB15, and D2 (all purchased from Aldrich), respectively. The mixing ratio of BL009:CB15:D2 is 90.5:9.0:0.5 wt %. Two indium tin oxide glass slides separated by two 38- $\mu\text{m}$ -thick plastic spacers are used to fabricate an empty cell. Both glass slides are coated with polyvinyl alcohol alignment film and rubbed in the same direction. The homogeneously mixed compound is then injected into the empty cell to form a DDCLC cell; cell pitch is confirmed to be 1.5  $\mu\text{m}$  using a Fourier-transform infrared ray spectrometer.

Figure 1 displays the experimental setup for developing and investigating optically switchable BGs based on DDCLC cells. Two *s*-polarized (along *x* axis) red beams,  $E_{R1}$  and  $E_{R2}$ , from a He–Ne pump laser ( $\lambda_R$ : 633 nm,  $\leq 35$  mW), are focused and intersect at the DDCLC cell at a small angle of  $\theta=2.8^\circ$ . The beams have an equal intensity of  $I_{R1}=I_{R2}=I_{R1,2}=340\text{--}909$  mW/cm<sup>2</sup>. Since the red beams are coherent, an intensity-modulated interference field ( $E_R$ ) is generated in the crossing region, where the sample is placed. One right-circularly polarized green beam ( $E_G$ ) with an intensity of  $I_G=14\text{--}306$  mW/cm<sup>2</sup>, derived from an Ar<sup>+</sup> laser ( $\lambda_G$ : 514.5 nm) and passed through a quarter-wave plate, impinges simultaneously on the interference region. One *s*-polarized probe beam ( $E_{\text{pro}}$ ,  $\lambda_{\text{pro}}$ : 633 nm) with a weak intensity of 1 mW/cm<sup>2</sup>, derived from another He–Ne laser and passed through polarizer  $P_2$  with a transmission axis in the *x* direction, is almost normally incident on the sample. A detection system, consisting of a photodiode and an oscilloscope, is placed behind the DDCLC samples to detect and record first-order diffraction intensity of the probe beam when BGs form.

Figure 2(a) presents the optical switching process of the BG- $E_R$  with  $I_{R1,2}=909$  mW/cm<sup>2</sup> is turned on at  $t=14$  s and  $E_G$  with  $I_G=204$  mW/cm<sup>2</sup> is turned on and off at  $t=19$  and

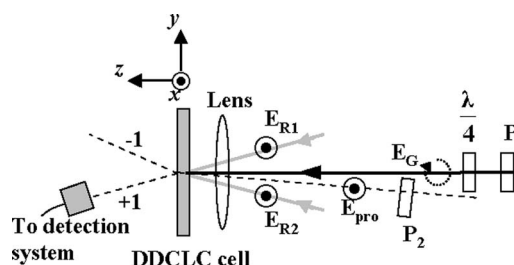


FIG. 1. Schematic of the experimental setup for investigating optically switchable biphotonic gratings (BGs) in dye-doped cholesteric liquid crystal (DDCLC) films;  $E_G$ : green beam;  $E_{R1,2}$ : red pump beams;  $E_{\text{pro}}$ : probe beam;  $P_1$ ,  $P_2$ : polarizers;  $\lambda/4$ : quarter waveplate for 514.5 nm.

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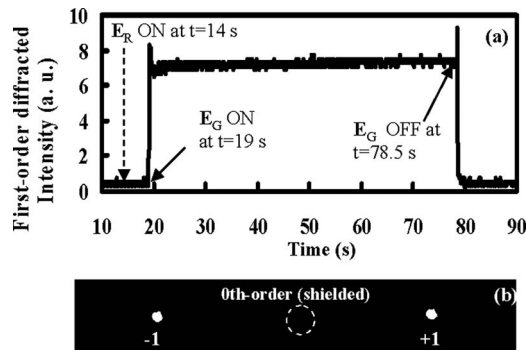


FIG. 2. (a) Optical switching feature of the first-order diffraction beam from the BG and (b) the diffraction pattern of the probed red beam from a stable BG based on a DDCLC cell.

78.5 s, respectively. No diffraction occurs when the red pump beams interfere with each other at the sample during  $t=14$ –19 s. Once  $E_G$  is turned on at  $t=19$  s, the BG is rapidly generated and diffraction intensity of the probe beam increases markedly, reaching a constant after  $t=20$  s. Thereafter, the BG is switched off and probe beam diffraction rapidly declines to zero when  $E_G$  is turned off at  $t=78.5$  s. Response times required to optically switch on and off the BG are 1 and 0.2 s, respectively. Figure 2(b) depicts the stable diffraction pattern of the probe beam during BG formation—the zeroth order is too strong and must be shielded.

A separate experiment is performed to clarify the mechanism of formation of the BG based on the DDCLC cell. One of the pumped red beams  $E_{R2}$  (Fig. 1) is blocked, and an analyzer A is placed behind the sample. Polarization of the incident  $s$ -polarized probe beam through the DDCLC sample is almost linear at  $\delta=-75^\circ$  with respect to the  $x$  axis ( $0^\circ$  in the  $x$ - $y$  plane in Fig. 1) before  $E_G$  and  $E_{R1}$  are turned on. The DDCLC cell can exhibit optical activity, which causes the polarization of the outgoing probe beam to rotate  $75^\circ$  counterclockwise (observed in the wave vector direction). As is widely known, the CLC medium with a planar structure can be optically active in the regime  $\lambda > \Delta n P$  with normal incidence (excluding the case of exact Bragg reflection  $\lambda=P$ ), where  $\lambda$  is incident beam wavelength in a vacuum,  $P$  is CLC cell pitch, and  $\Delta n$  is the difference between ordinary and extraordinary refractive indices of the LC material.<sup>7</sup> As incident, linearly polarized beam passes through the CLC cell, it can be decomposed into orthogonal right- and left-circularly polarized eigenmodes of waves propagating along the  $z$  axis with indices of  $n_R$  and  $n_L$ , respectively. The corresponding outgoing wave is also linear but rotated through an angle, such that

$$|\delta| = \pi d |n_R - n_L| / \lambda. \quad (1)$$

The CLC cell used in this work can be simply confirmed in the regime of optical activity, by substituting  $\lambda=0.633 \mu\text{m}$ ,  $P=1.5 \mu\text{m}$ , and  $\Delta n=0.2915$  into the inequality of  $\lambda > \Delta n P$ . Accordingly, the value of  $|n_R - n_L|$  in the present unilluminated DDCLC cell can be estimated at  $\sim 6.9 \times 10^{-3}$  by substituting  $|\delta|=75^\circ$ ,  $\lambda=0.633 \mu\text{m}$ , and  $d=38 \mu\text{m}$  into Eq. (1).

Figure 3 shows the dynamics of probe beam transmitted intensity in the separate experiment when analyzer A is crossed with polarizer  $P_2$  during the excitation of  $E_G$  with  $I_G=62, 162, 255$ , and  $270 \text{ mW/cm}^2$  and  $E_{R1}$  with  $I_{R1}=909 \text{ mW/cm}^2$ . During the first stage ( $t=19$ –99 s), trans-

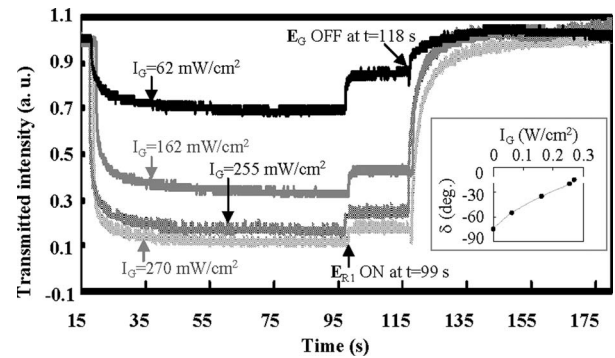


FIG. 3. Dynamics of transmitted intensity of a probe beam when an analyzer A (crossed with  $P_2$ ) is inserted behind the cell during excitation of green beam with four intensities and a pumped red beam with an intensity of  $909 \text{ mW/cm}^2$ . The inset figure plots the variation of angle of rotation of the polarization of the probe beam due to the optical activity of the DDCLC cell with the green-beam intensity.

mitted intensity of the probe beam through crossed A abruptly declines when  $E_G$  is turned on at  $t=19$  s, and the stronger the green beam becomes, the deeper the transmitted intensity drops. This experimental result suggests that the circularly polarized green beam stimulates the azo dyes by *trans-cis* isomerization<sup>5,8,9</sup> to reorient parallel to the wave vector ( $\mathbf{k}_G$ ) to minimize the probability of being photoexcited again. The homeotropically aligned dyes then induce the LC molecules to tilt toward the  $z$  axis, in turn forming a distorted planar structure.<sup>9</sup> As  $I_G$  increases, more and more LC molecules tilt toward the  $z$  axis, producing a homeotropic-like structure, in which capacity for optical activity of the CLC cell decreases. The variation of  $\delta$  with  $I_G$  (inset in Fig. 3) confirms this inference, in which the angle  $\delta$  of rotation of the linear polarization of the probe beam caused by the cell declines as  $I_G$  increases, such that the transmitted intensity of the probe beam via A decreases in Fig. 3. The experimental result for the first stage (Fig. 3) is therefore reasonable. The reduction in optical activity of the CLC cell corresponds to the probe beam experiencing a drop in the difference between indices  $|n_R - n_L|$ . During the second stage ( $t=99$ –118 s), the pumped red beam  $E_{R1}$  is also turned on at  $t=99$  s, and then the transmitted intensity of the probe beam increases immediately but only to specific extent because of the inherent limitation on He-Ne pump laser power. Additionally, the increase in transmitted intensity increases as  $I_G$  decreases. This experimental result is explained as follows. The absorption spectrum (Fig. 4) indicates that an absorptive peak of *trans*-D2 doping in the CLC cell is in the green-yellow band, and no absorption can occur when the wavelength exceeds  $620 \text{ nm}$  in dark (black line). Once the

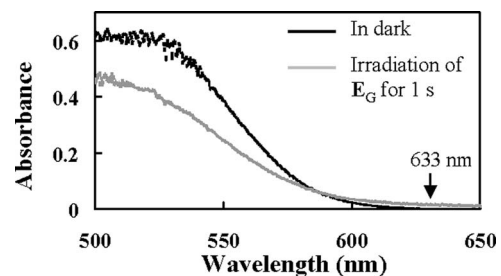


FIG. 4. Absorption spectrum of a D2-doped CLC cell in the dark (black line) and in the irradiation of  $E_G$  with intensity of  $300 \text{ mW/cm}^2$  for 1 s (gray line).

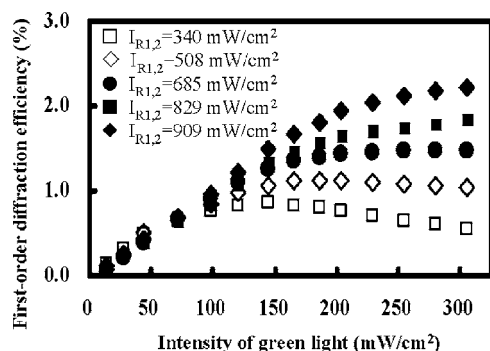


FIG. 5. Variation of the first-order diffraction efficiency of a probe beam with relative intensity of green and red beams.

DDCLC has been irradiated by  $E_G$  with an intensity of  $300 \text{ mW/cm}^2$  for 1 s, the *trans* isomers conform to the *cis* isomers, and their absorption in the green-yellow and red bands drops and rises, respectively (gray line).<sup>5</sup> Experimental results during the second stage (Fig. 3) reveal that restimulation by strong red beam can suppress additional dyes to reorient the LCs toward  $\mathbf{k}_G$  via *cis-trans* back isomerization, causing the cell to recover its planarlike structure to some extent. The capacity of the cell for optical activity then increases and transmitted intensity through the crossed A thus increases. During the third stage ( $t \geq 118 \text{ s}$ ), transmitted intensity increases when the green beam is shut off at  $t = 118 \text{ s}$ , and then recovers to its initial value. This experimental finding demonstrates that the isomers in the *cis* state all conform to the *trans* state due to the red-beam-induced and thermal *cis-trans* back isomerization, which restores a perfect planar structure.

As explained, BGs are formed via two mechanisms that operate separately in the dark and bright fringes of  $E_R$ . One is green-beam-induced reorientation of LCs parallel to  $\mathbf{k}_G$ , generated by the interaction of LCs with rotated dyes via *trans-cis* isomerization; the other mechanism is red-beam-induced suppression of dye rotation by *cis-trans* back isomerization. These two mechanisms are responsible for, respectively, a spatially periodic distribution of the homeotropiclike and planarlike structures, yielding the BGs.

Figure 5 plots the variation of the first-order diffraction efficiency  $\eta_1$  of the probe beam with the relative intensity of the green and red beams. Typically,  $\eta_1$  increases as  $I_{R1,2}$  increases for a fixed  $I_G$ . This experimental result is reasonable because, based on a previous discussion, as  $I_G$  ( $I_{R1,2}$ ) in-

creases, the texture in the dark (bright) regions of  $E_R$  typically becomes homeotropiclike (planarlike). Therefore, no matter  $I_G$  or  $I_{R1,1}$  increases, the difference between textures and, thus, the effective refractive indices  $\Delta n_{\text{eff}}$  "seen" by the probe beam in these two regions increases, thereby increasing  $\eta_1$ . However,  $\eta_1$  falls rather than increasing as  $I_G$  increases above  $144 \text{ mW/cm}^2$  when  $I_{R1,2}$  equals the weakest intensity of  $340 \text{ mW/cm}^2$ ; the red beams are too weak to suppress the strong green-beam-induced reorientation of the dyes and the LCs in the bright regions of  $E_R$ , such that  $\Delta n_{\text{eff}}$  and, therefore,  $\eta_1$  weaken.

In summary, this investigation develops optically switchable BGs and analyzes the mechanisms through which gratings form on DDCLC cells. The gratings can be switched on (off) by turning on (off) the circularly polarized green beam, illuminated in a red interfering field created from two coherent linearly polarized red beams. The mechanisms of BG formation are green-beam-induced dye reorientation in the direction of the wave vector via *trans-cis* isomerization and the red-beam-induced suppression of dye reorientation via *cis-trans* back isomerization in the dark and bright fringes of the red interfering field. These mechanisms result in a spatially periodic distribution with homeotropiclike and planarlike textures, which in turn cause the BGs. The diffracted performance of BGs depends significantly on the relative intensity of the green and red beams.

The authors would like to thank the National Science Council of the Republic of China, Taiwan, for financially supporting this research under Contract No. NSC 95-2112-M-006-020-MY2.

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