Transmission and reflection phase gratings formed in azo-dye-doped chiral nematic liquid crystals

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Transmission and reflection gratings were simultaneously formed in azo-dye-doped chiral nematic liquid crystals (N*LCs) with planar alignment. The formation process is based on a phototuning of the Bragg reflection band of the N*LC. The helical pitch of the photoreactive N*LC was spatially controlled with intensity variation of interference light. The resultant periodic structure showed both transmissive and reflective diffractions due to the spatially modulated light intensities. The observed dependence of diffraction efficiencies on the polarization states of the probe beam was well explained by considering a spatial modulation of the helical pitch. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3072363]

It is known that chiral nematic liquid crystals (N*LCs), which have helical arrangement of mesogenic molecules with self-assembly, exhibit unique and interesting optical properties.[1] The spatially periodic variation in the dielectric tensor in the helical structure induces the Bragg reflection of circularly polarized light. The Bragg reflection arises when the wavelength of incident light is in the range of $n_oP<\lambda<n_eP$, where P is the pitch of the N*LC structure and n_o and n_e are the principal refractive indices of the LC medium. The helical pitch can be controlled with electric fields, magnetic fields, and temperature. In addition, Sackmann[2] reported photochemical control of the helical pitch. These of N*LCs provide many useful applications such as light modulators,[3,4] displays,[5–8] laser devices,[9–12] and diffraction gratings.[13–17] Chanishvili *et al.*[11] reported experimental investigation of phototunable lasing in dye-doped N*LCs. Hrozhyk *et al.*[18] reported optical tuning of the reflection of azobenzene containing N*LCs and demonstrated the ability to write information into the N*LCs. According to the above examples of studies for photonics in the N*LCs, it is found that strict periodic patterns can be self-organized in the N*LCs and photoreactive N*LCs are suitable materials for realizing phototunable photonic devices.

We have studied polarization holographic recordings in an azo-dye-doped nematic LC.[19,20] Since photoisomerization reactions of the azo-dye affect the alignment of the LC molecules, spatial distributions of the molecular reorientation are induced with irradiation of polarized interference light. In our previous studies, it was shown that the reconstructing properties of polarization holograms that are recorded in the azo-dye-doped nematic LC are strongly dependent on the initial alignment of the LC.[19,20]

In the present letter, holographic gratings were formed in azo-dye-doped N*LCs to provide insight into the relationship between the periodic helical structure and the optical property. The gratings showed both transmissive and reflective diffractions controlled with spatially modulated helical pitch. The diffraction intensities depended on the polarization state of incident light and the helical pitch of the N*LC. The diffraction properties were discussed using a model of helical pitch modulation.

In the present study, three types of azo-dye-doped N*LCs with different compositions were prepared by mixing a low-molar-mass nematic mixture E7 (Merck, Japan), a right-hand chiral dopant CB15 (Merck, Japan), and an azo-dye DR1 (Aldrich). The weight ratios of the mixtures (E7:CB15:DR1) were prepared to be 62.9:36.5:0.6 (sample 1), 62.1:37.3:0.6 (sample 2), and 61.3:38.1:0.6 (sample 3). The mixtures were sandwiched between rubbed poly(vinyl alcohol)-coated glass plates with 10 μm thick spacers, which gave planar alignment cells. The spectral characterization was measured using a spectrometer (Ocean Optics USB4000) at room temperature. All samples showed strong absorption of green light. The center wavelengths of the selective reflection bands of samples 1, 2, and 3 were observed at 642, 617, and 601 nm, respectively. The width of the reflection bands was about 50 nm.

Figure 1 shows transmission spectra of all samples around the reflection band when the samples were irradiated by a frequency-doubled Nd doped yttrium aluminum garnet laser light with an operating wavelength of 532 nm (pump beam). The intensity of linearly polarized pump beam, which was set perpendicular to the rubbing direction, was selected at 0, 0.57, and 1.1 W/cm^2. The exposure time was 60 s. The reflection band corresponded to the helical pitch showed blueshift with the pump beam intensity.

Figure 2 summarizes the transmittance of the 632.8 nm He–Ne laser light for the three samples with varying pump beam intensity. When the probe beam was right-handed circularly polarized (RCP), the transmittance of sample 3 changed drastically. The reason for the significant change in transmittance of sample 3 can be explained by considering the blueshift of the reflection band with the irradiation as shown in Fig. 1. In addition, the transmittance of sample 2 changed slightly in the high intensity region. On the other hand, the transmittance for the left-handed circularly polarized (LCP) probe beam had very little dependence on the pump beam intensity since all samples were right-hand helical structure. As a result, it is confirmed that the polarization-selective transmission and the reflection gratings coexisting

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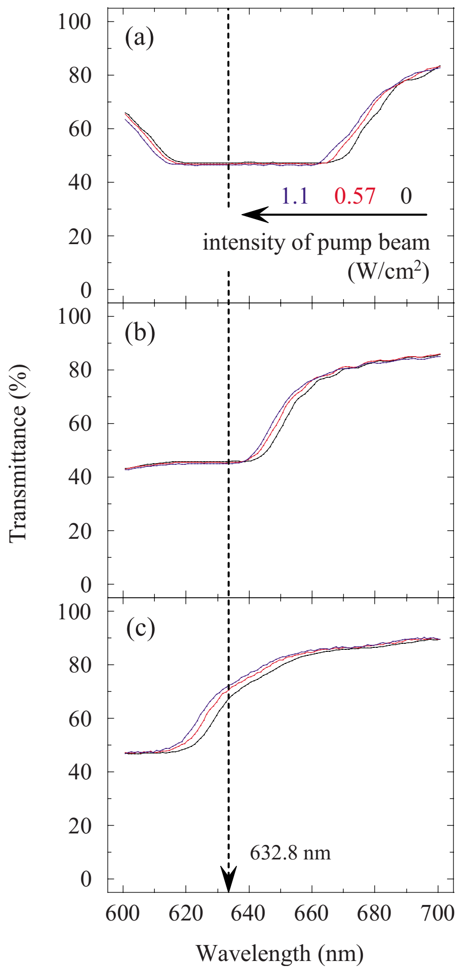

FIG. 1. (Color online) Transmission spectra for varying intensities of the pump beam for samples (a) 1, (b) 2, and (c) 3.

in this material can be realized with spatial intensity modulation induced by interference.

Holographic gratings formed in the three samples, and the diffraction properties were investigated. The experimental setup is shown in Fig. 3(a). The intensities of the s-polarized two pump beams were 0.56 W/cm^2. The incident angles of the pump beams were $\pm 1.5°$, resulting in a grating pitch of approximately 10 μm. The diffraction properties of the gratings were evaluated using the He–Ne laser. The diffraction beams appeared for both transmissive and reflective directions because both the transmittance and reflectance were spatially modulated according to the helical pitch modulation, as shown in Fig. 3(b). The measured positive first-order diffraction efficiencies are shown in Fig. 4. The polarization state of the probe beam was varied by rotating a quarter-wave plate. The strongest diffraction was observed for sample 3 because the transmittance and reflectance were modulated most deeply at the probe wavelength. This reason can be also explained by considering the blueshift of the reflection band. In addition, the diffraction efficiencies obviously depended on the polarization state of the probe beam. To explain this dependence, the diffraction properties were calculated using a model of the helical pitch modulation as shown in Fig. 3(b). We assumed that the variation in the helical pitch is proportional to the pump intensity. By denoting the maximum value of the variation in the helical pitch as ΔP, the helical pitch modulation is given by $P(x)=P_0-\Delta P\cos^2(\pi x/\Lambda)$, where P_0 is the helical pitch in the initial state and Λ is the grating pitch. The angle between the x-axis and the local director is given by $\theta(x,z)=2\pi z/P$. The electric field distributions of the transmitted and reflected light in the x-direction were calculated by employing Berreman's 4×4 matrix method.[21] Antireflection layers were installed at the air-N*LC boundaries to reduce the effect of multiple interference in the calculation.[22] By Fourier transforming the electric fields of the transmitted and reflected light, the diffraction efficiencies for the transmissive and reflective directions were calculated. The calculation was performed for sample 3, and the result is shown in Fig. 4. The parameters used in the calculation were a cell thickness of

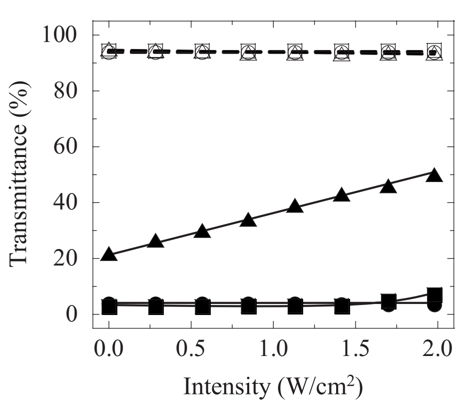

FIG. 2. Dependences of the transmittance on the intensity of the pump beam. Filled circles, filled squares, and filled triangles represent measurements by the RCP probe beam for samples 1, 2, and 3, respectively. Open circles, open squares, and open triangles represent measurements by the LCP probe beam for samples 1, 2, and 3, respectively.

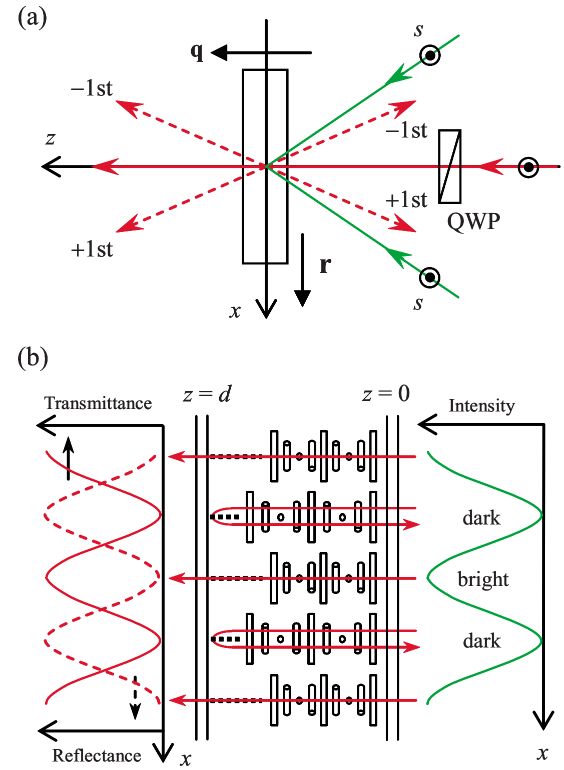

FIG. 3. (Color online) (a) Experimental setup for holographic recording and reconstruction. **q** is the wave vector of the helical structure and **r** is the rubbing direction. (b) Schematic illustration of the helical pitch modulation. d is the cell thickness.

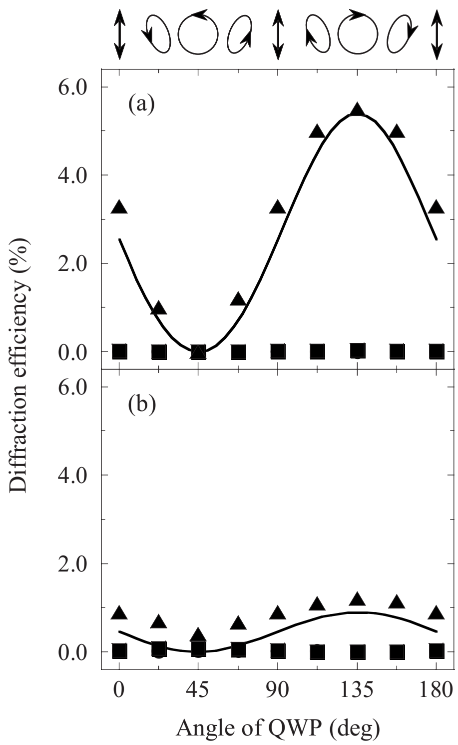

FIG. 4. Dependences of the positive first-order diffraction efficiencies on the polarization state of the probe beam for (a) the reflective direction and (b) the transmissive direction. The polarization states of the probe beam are schematically shown in the top of the figure. Filled circles, filled squares, and filled triangles represent the measured data for samples 1, 2, and 3, respectively. Solid curves represent the calculation results for sample 3.

10 μm, a wavelength of the probe beam of 633 nm, $P_0 = 370$ nm, $n_o = 1.56$, $n_e = 1.71$, and $\Delta P = 4.1$ nm. Here, ΔP was obtained by fitting the experimental data, and the other parameters were determined from the experimental condition and the observed spectrum. The calculated diffraction efficiencies for the polarization state of the probe beam were in good agreement with the experimental result.

In summary, we demonstrated that N*LC with modulated helical pitch can be used as transmission and reflection phase gratings. The formation mechanism is based on the photoinduced shift in the Bragg reflection band of the N*LC. The observed polarization dependence of the diffraction efficiencies is well explained by considering the model of the helical pitch modulation. The rapid change in the reflectance around the reflection band enables highly efficient gratings with low light intensities.

[1] P. Yeh and G. Gu, *Optics of Liquid Crystal Displays* (Wiley, New York, 1999).
[2] E. Sackmann, J. Am. Chem. Soc. **93**, 7088 (1971).
[3] J. S. Patel and R. B. Meyer, Phys. Rev. Lett. **58**, 1538 (1987).
[4] D.-K. Yang, L.-C. Chien, and J. W. Doane, Appl. Phys. Lett. **60**, 3102 (1992).
[5] D. L. White and G. N. Taylor, J. Appl. Phys. **45**, 4718 (1974).
[6] D. W. Berreman and W. R. Heffner, Appl. Phys. Lett. **37**, 109 (1980).
[7] D. K. Yang, J. W. Doane, Z. Yaniv, and J. Glasser, Appl. Phys. Lett. **64**, 1905 (1994).
[8] D.-K. Yang, J. L. West, L.-C. Chien, and J. W. Doane, J. Appl. Phys. **76**, 1331 (1994).
[9] V. I. Kopp, B. Fan, and H. K. M. Vithana, Opt. Lett. **23**, 1707 (1998).
[10] T. Matsui, R. Ozaki, K. Funamoto, M. Ozaki, and K. Yoshino, Appl. Phys. Lett. **81**, 3741 (2002).
[11] A. Chanishvili, G. Chilaya, G. Petriashvili, R. Barberi, R. Bartolino, G. Cipparrone, A. Mazzulla, and L. Oriol, Appl. Phys. Lett. **83**, 5353 (2003).
[12] S. Furumi, S. Yokoyama, A. Otomo, and S. Mashiko, Appl. Phys. Lett. **84**, 2491 (2004).
[13] D. Subacius, P. J. Bos, and O. D. Lavrentovich, Appl. Phys. Lett. **71**, 1350 (1997).
[14] A. Y.-G. Fuh, C.-H. Lin, M.-F. Hsien, and C.-Y. Huang, Jpn. J. Appl. Phys., Part 1 **40**, 1334 (2001).
[15] B. I. Senyuk, I. I. Smalyukh, and O. D. Lavrentovich, Opt. Lett. **30**, 349 (2005).
[16] H.-C. Yeh, G.-H. Chen, C.-R. Lee, and T.-S. Mo, Appl. Phys. Lett. **90**, 261103 (2007).
[17] H.-C. Yeh, G.-H. Chen, C.-R. Lee, and T.-S. Mo, J. Chem. Phys. **127**, 141105 (2007).
[18] U. A. Hrozhyk, S. V. Serak, N. V. Tabiryan, and T. J. Bunning, Adv. Mater. (Weinheim, Ger.) **17**, 1735 (2007).
[19] T. Sasaki, H. Ono, and N. Kawatsuki, Appl. Opt. **47**, 2192 (2008).
[20] T. Sasaki, H. Ono, and N. Kawatsuki, J. Appl. Phys. **104**, 043524 (2008).
[21] D. W. Berreman, J. Opt. Soc. Am. **62**, 502 (1972).
[22] W. H. Southwell, Opt. Lett. **8**, 584 (1983).