Rapid communication

Z. ZHENG^{1,2} F. GUO^{1,2} Y. LIU¹ L. XUAN^{1, \bowtie}

Low threshold and high contrast polymer dispersed liquid crystal grating based on twisted nematic polarization modulator

- State Key Laboratory of Applied Optics, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Science, Changchun 130033, P.R. China
- ² Graduate School of Chinese Academy of Science, Beijing 100039, P.R. China

Received: 13 November 2007/Revised version: 27 January 2008 Published online: 27 February 2008 • © Springer-Verlag 2008

ABSTRACT An electrically tunable mode is proposed to overcome the bottleneck problem of a conventional polymer dispersed liquid crystal (PDLC) grating and realize its low threshold and high contrast through an alignment-controlled PDLC. The results indicate that the threshold voltage $V_{\rm PP}$ of the grating is as low as 0.75–0.8 V, the saturation voltage $V_{\rm PP}$ is 7 V and the contrast ratio of the grating reaches 245, which is about three times larger than the conventional PDLC grating. Stability testing indicates a stable state of contrast during a long time.

PACS 42.40.Eq; 42.70.Df; 42.79.Kr

1 Introduction

The diffractive transmission grating based on polymer dispersed liquid crystal (PDLC) is an attractive new electro-optical device. Due to its electrically tunable characteristic, such a grating exhibits a bright prospect in applications of integrated optics, optical communication, laser techniques, optical data storage, etc. [1–4]. The conventional PDLC grating is always prepared with a mixture of liquid crystals and photosensitive monomers. However, because of monomer mixing, the threshold voltage of such a grating is much higher than the other liquid crystal devices, which limits its application. In addition, because of the high threshold (usually $10 \text{ V/}\mu\text{m}$ [5, 8, 9]) and poor tunable performance, the contrast ratio of a PDLC grating is very low. The value of the contrast ratio always lies in the range of 10-30 [6-16]. To improve the shortcomings mentioned above, some researchers adopted kinds of surfactant monomers, such as octanoic acid [17], high-dielectric HRL-410 [18] or fluorine monomers [19], to

decrease the anchoring energy between the liquid crystal layer and the polymer layer. The threshold voltage decreases with the decreasing of the anchoring energy accompanied by a different-level improvement of electro-optical (E-O) properties. These works are helpful for materials development in the PDLC grating field. However, it is difficult to realize the improvement in practical application by modifying materials properties only. Therefore, there is an urgent need to develop a low threshold voltage PDLC grating with high contrast ratio from other aspects, such as device structure, drive mode or external conditions.

In this work, we design an electrically tunable mode of PDLC grating based on a twisted nematic (TN) cell (provided by North Liquid Crystal Display Research and Development Center, Changchun, China. The tested cell gap is $6.67~\mu m$) positioned in front of an alignment-controlled polymer dispersed liquid crystal (ACPDLC) grating, and we realize a much lower threshold voltage and a higher contrast compared with conventional PDLC gratings.

2 Details of the design and discussion

The principle diagram is shown in Fig. 1. A polarizer is inserted between the He-Ne laser and the TN cell to produce a polarized incident beam, and its polarization direction turns through 90 degrees when the beam is transmitted through the TN cell, because of the optical rotation property of the TN cell without an applied voltage. In the case that a voltage over the threshold is applied to the TN cell, the twisting effect disappears. Therefore, the polarization direction of the incident beam is unaltered. That is to say, the TN cell is a polarization-direction modulator. Because of the optical anisotropy of liquid crystal molecules contained in the PDLC grating, the refractive index of the LC layer is changeable with the polarization direction of the incident beam. Thus, driving the TN modulator with a very low voltage changes the liquid crystal's refractive index, and consequently changes the refractive-index modulation (Δn) of the PDLC grating, and, finally, a much better electrooptical property could be realized. The polarization directions of the incident beam in the cases of no applied voltage and a drive voltage on the TN modulator are labeled in Fig. 1; the black arrow is the case of the former and the red arrow is the latter.

In addition, the contrast ratio, the other important parameter which reflects the tunable performance of the PDLC grating, is worth being considered carefully. The contrast is defined as the first order diffraction efficiency ratio under the conditions of no applied voltage and a drive voltage on the TN cell.

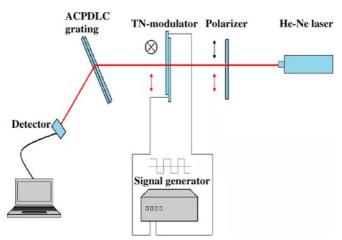


FIGURE 1 Electrically tunable setup to realize low threshold and high contrast with a TN modulator and an ACPDLC. Arrows represent polarization direction of the incident beam in the cases of no applied voltage (colored black) and a drive voltage (colored red). 'S' represents the direction perpendicular to the surface

The diffraction efficiency of the grating is dependent directly on Δn ; therefore, the larger the variation range of Δn , the better the tunable property is obtained: then, the higher contrast could be reached. In order to extend the variation range of Δn , two points are noted by us. First, the index matching between the LC layer and the polymer layer in the grating, which means that the ordinary refractive index of LC (n_0) is approximately equal to the polymer refractive index (n_p) . Second, the uniform alignment of liquid crystals in the grating is a vital problem. The most perfect case is a uniform alignment of LC molecules along the grating strips, as schemed in Fig. 2. When the polarization direction of the incident beam is parallel to the direction of the strips, the refractive index of the LC layer is n_e , and Δn reaches a maximal value; contrarily, when it is perpendicular to the direction, $\Delta n \approx 0$ because of index matching.

For the purpose of index matching between materials in our experiments, two kinds of acrylates with better photo-reactive activity and solubility are selected. One is penta-functional dipentaerythritol hydroxyl pentaacrylate (DPHPA) (supported by Aldrich, $n_{\rm D} = 1.49$ at 20 °C); the other is difunctional phthalic diglycol diacrylate (PDDA) (supported by Eastern Acrylic Chem. Tech. Co., Ltd., Beijing, $n_D =$ 1.5458 at 20 °C). They are mixed with the weight ratio of 1:1. To obtain a good morphology of the PDLC grating, a small amount of polymerized fluoridizer, Actyflon-G04 (supported by

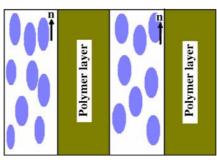


FIGURE 2 Sketch map of alignment-controlled PDLC grating. The directors (*n*) of LC molecules are parallel to each other and along the direction of polymer grooves

XEOGIA Fluorine-Silicon Chem. Co., Ltd., $n_D = 1.5492$ at 20 °C) is added as surfactant frequently. In addition, some rose bengal (0.5 wt. \%, supported by Aldrich) and N-phenylglycine (2 wt. %, supported by Aldrich) are mixed together as photoinitiator and coinitiator. The mixture is stirred at 40 °C for 24 h; then, the nematic liquid crystal TEB30A (supported by Slichem Co., Ltd., Shi Jia Chuang, China, $\Delta n = 0.1703$, $n_e =$ 1.6925 at 20 °C) is mixed into it with the monomer/LC ratio of 7:3 and restirred at 40 °C for 12 h. The refractive index of the polymer, estimated as the average value of monomers contained in the mixture, is 1.520, which is very close to the ordinary refractive index of LC $n_0 = 1.522$. The index matches well between the polymer and the LC. The mixture is injected into the LC cell with 10-µm cell gap and exposed in a two-beam interference field for about 20 min. The intensity of a single beam is 3.6 mW/cm² and the wavelength is 532 nm. Finally, the cell is irradiated

under UV for about 3 min for structure fixing.

Uniform alignment of LCs in a PDLC grating is a decisive point for a better E-O property and it is studied in the experiments. What is interesting is that there is a self-alignment phenomenon in the LC layer of the PDLC grating. It could be expressed clearly according to Berreman's groove model, which indicates that LC molecules could be anchored by the grooves formed by the polymer [20]. Therefore, the LCs could self-align without any other controlling. The anchoring energy could be calculated as the following equation:

$$W_{\rm B} = 0.25 K A^2 q^3, \tag{1}$$

where A is the depth of the groove formed by the polymer after exposure, $q=2\pi/\Lambda$ is the wavenumber of the grating, Λ is the grating pitch and K is the Frank elastic constant. By assuming a simple sinusoidal surface relief structure with A=100 nm, $\Lambda=1$ μ m and $K\sim10^{-12}$ N. Thus, $W_{\rm B}\sim10^{-3}$ mN·m⁻¹, which is about three orders of magnitude smaller than rubbing induced anchoring energy [21]. Although this energy is small, its alignment effect on the LCs could not be omitted.

To explain the uniformity of LC alignment, the angular transmittance intensity of a PDLC grating is tested through polarized optical microscopy (using an Olympus BX-51 microscope) under crossed polarizers. In testing, the polarization direction of one polarizer is parallel to the grating grooves and the other is perpendicular to them; the two polarizers are fixed on the microscope, the grating is inserted between the two polarizers and then the sample is turned. Finally, the rotation angletransmittance dependence is obtained (Fig. 3). It is indicated by the symbols ' Δ ' in Fig. 3; a light-to-dark change could be found in the transmittanceangle dependence. The transmittance ratio of light and dark states is about 170, which shows the self-alignment of the LCs in grating. The E-O property is tested through the tunable mode of Fig. 1 and the first order diffraction intensity decreases rapidly when the TN cell is driven by the applied voltage. The contrast ratio tested is 125 as shown by the symbols '\Delta' in Fig. 5. To improve the E-O tunable performance of the grating further, a more uniform alignment of LCs is a main aspect. Normally, the simplest way to obtain a uniform alignment of LCs is rubbing; however, the morphology or the phase separation of the grating might be affected by a strong rubbing. It was noted that the self-alignment of LCs is induced by grooves, thus, the uniformity of alignment could be enhanced through a weak rubbing of substrates

before exposure, which does not affect the formation of the grating, as well as improves the E-O performance or contrast. Subsequently, the alignment-controlled PDLC (ACPDLC) grating is prepared; its transmittance—angle dependence is shown by the symbols 'O' in Fig. 3. There arises an evident light-to-dark transformation and the transmittance ratio in that case could reach 280, which increases notably compared with the self-alignment sample. In order to evaluate the alignment ef-

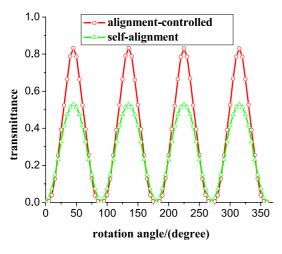


FIGURE 3 Angular transmittance intensity of self-alignment PDLC grating (shown with symbols 'Δ') and alignment-controlled PDLC grating (shown with symbols '○') under crossed polarizers

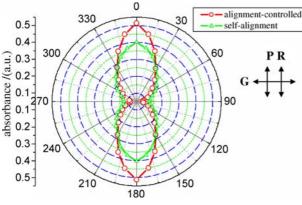


FIGURE 4 Polarized IR absorbance at 2227 cm⁻¹ of ACPDLC grating and self-alignment grating, as a function of rotating angles of two samples. The double arrow **P** is the polarization direction. The other two double arrows are the grating vector **G** and the rubbing direction **R** of the ACPDLC before it is rotated

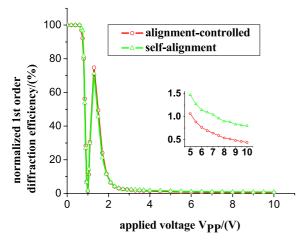


FIGURE 5 Electro-optical property of the self-alignment PDLC grating (shown by symbols 'Δ') and alignment-controlled PDLC grating (shown by symbols ')'). The *inset* shows the magnified scale for comparing the state of the applied voltage

fect of the ACPDLC grating further, the order parameter of LCs in this kind of grating is tested through polarized Fourier transform infrared (FTIR) spectroscopy (using a Bio-Red 3000) and compared with the order parameter of the self-alignment one. In testing, the absorbance of liquid crystals at 2227 cm⁻¹ is detected when rotating the sample for a period. The absorbancerotation angle dependence is shown in Fig. 4. Symbols '()' represent the ACPDLC and symbols ' Δ ' are the selfalignment condition. It is evident in the figure that the maximum absorbance is obtained when the rotation angle is 0° or 180° (i.e. the polarization direction is parallel to the grating grooves) and the minimum is obtained at 90° or 270°. So, it could be concluded that the average alignment of LCs is parallel to the grating grooves no matter what types the gratings are. It could also be found that, for the ACPDLC type, $A_{\text{max}} = A_{0/180} =$ 0.512, $A_{\min} = A_{90/270} = 0.087$; for the self-alignment type, $A_{\text{max}} = A_{0/180} =$ 0.401, $A_{\min} = A_{90/270} = 0.126$. According to the equation given by Xuan et al. [22], $S = (A_{\text{max}} - A_{\text{min}})/(A_{\text{max}} +$ $2A_{\min}$), the LC order parameter (S) of the ACPDLC grating is calculated as 0.62, which is 0.2 larger than the selfalignment grating, 0.42. Thus, it is certain that the alignment uniformity of the ACPDLC grating is better than the self-alignment grating. From the E-O property shown by the symbols '()' in Fig. 5, it is found that the contrast ratio reaches 245, two times that of the self-alignment sample. The threshold voltage V_{90} is about 0.75–0.8 V (V_{PP}) and the saturation voltage is 7 V (V_{pp}) , which are much lower than those of a conventional PDLC grating. The photographs of a first order diffraction spot under the conditions of no applied voltage and a 7 V square wave signal (V_{pp}) are given in Fig. 6 a and b. It is significant that the intensity of the first order diffraction beam is much more blurry (Fig. 6b) when there is an applied voltage on the TN cell for the reason of index matching of the LC and the polymer. However, the diffraction spot is bright in the case of no applied voltage (Fig. 6a), because the refractive index of the LC is approximatively equal to $n_{\rm e}$ in the ACPDLC grating, so that the refractive-index modulation Δn reaches a maximum; subsequently, the diffrac-

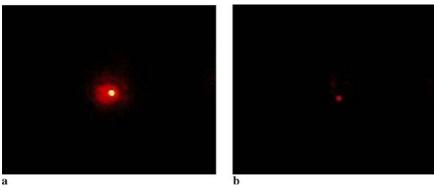


FIGURE 6 First order diffraction spot of ACPDLC grating switched by TN modulator, no applied voltage on the TN modulator (\mathbf{a}) and an applied square wave signal with the peak-to-peak value of $7 \text{ V}(\mathbf{b})$

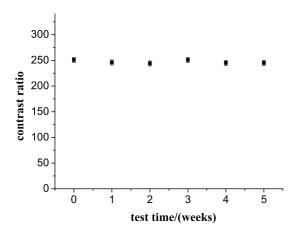


FIGURE 7 Stability testing for contrast of ACPDLC grating during five weeks

tion efficiency of the grating in that case is largest.

Finally, the stability of the ACPDLC grating is tested. The prepared grating is placed in a dry and dark box. The E-O performance and its contrast ratio are tested and recorded once a week. Results during five weeks are shown in Fig. 7. There is no evident change. Some little fluctuation might be ascribed to the error caused by testing. Such results indicate that the LC alignment in the ACPDLC grating is stable, and the anchoring energy for alignment is sufficient.

3 Conclusions

We have prepared an ACPDLC grating with a uniform LC

alignment and index matching, which could be electrically switched easily by an excellent tunable mode with a TN modulator under a much lower applied voltage. The high contrast ratio is obtained through that tunable mode. The experimental data indicates that the threshold voltage and saturation voltage are $0.75-0.8 \text{ V } (V_{PP})$ and $7 \text{ V } (V_{pp})$, respectively. The contrast ratio of the ACPDLC grating is 245, which is three times higher than that of a conventional PDLC grating. Moreover, stability testing shows that the alignment of the LCs in the ACPDLC is fine and stable, which ensures the perfect E-O property and high contrast of the device. Such a kind of tunable mode contained in the ACPDLC and TN cell overcomes two main problems (high threshold voltage and low contrast), and realizes its practicability.

ACKNOWLEDGEMENTS The authors would like to thank the support from the Natural Science Foundation (Grant Nos. 60578035 and 50473040) and the Science Foundation of Jilin Province, China (Grant Nos. 20050520 and 20050321-2).

REFERENCES

- 1 R.L. Sutherland, L.V. Natarajan, V.P. Tondiglia, Chem. Mater. **5**, 1533 (1993)
- 2 X.H. Sun, X.M. Tao, T.J. Ye, P. Xue, Y.S. Szeto, Appl. Phys. B 87, 65 (2007)
- 3 D.J. Pikas, S.M. Kirkpatrick, D.W. Tomlin, L.V. Natarajan, V.P. Tondiglia, T.J. Bunning, Appl. Phys. A 74, 767 (2002)
- 4 R.L. Sutherland, L.V. Natarajan, V.P. Tondiglia, T.J. Bunning, W.W. Adams, Appl. Phys. Lett. **64**, 1074 (1994)
- 5 A.F. Senyurt, G. Warren, J.B. Whitehead, C.E. Holyer, Polymer **47**, 2741 (2006)
- 6 H. Xianyu, J. Qi, R.F. Cohn, G.P. Crawford, Opt. Lett. 28, 792 (2003)
- 7 H.L. Zhang, H. Xianyu, J. Liang, Y. Betremieux, G.P. Crawford, J. Noto, R. Kerr, Appl. Opt. 46, 161 (2007)
- 8 T.J. Bunning, L.V. Natarajan, V.P. Tondiglia, R.L. Sutherland, Annu. Rev. Mater. Sci. **30**, 83 (2000)
- 9 M. Mucha, Prog. Polym. Sci. 28, 837 (2003)
- 10 Y.J. Liu, B. Zhang, Y. Jia, K.S. Xu, Opt. Commun. 218, 27 (2003)
- 11 Y.J. Liu, X.W. Sun, H.T. Dai, J.H. Liu, K.S. Xu, Opt. Mater. 27, 1451 (2005)
- 12 E.Y. Shin, J.A. Jung, E.H. Kim, B.K. Kim, Polym. Int. **54**, 922 (2005)
- 13 R. Caputo, L.D. Sio, A. Veltri, C.P. Umeton, A.V. Sukhov, J. Display Technol. 2, 38 (2006)
- 14 J.M. Wofford, L.V. Natarajan, V.P. Tondiglia, R.L. Sutherland, P.F. Lloyd, S.A. Siwecki, T.J. Bunning, in *Liquid Crystals X* (Proc. SPIE **6332**) (SPIE, Bellingham, WA, 2006), p. 63 320Q-1
- 15 K. Beev, L. Criante, D.E. Lucchetta, F. Simoni, S. Sainov, Opt. Commun. 260, 192 (2006)
- 16 K. Beev, L. Criante, D.E. Lucchetta, F. Simoni, S. Sainov, J. Opt. A Pure Appl. Opt. 8, 205 (2006)
- 17 J. Klosterman, L.V. Natarajan, V.P. Tondiglia, R.L. Sutherland, T.J. White, C.A. Guymon, T.J. Bunning, Polymer 45, 7213 (2004)
- 18 S.T. Wu, T.S. Mo, Y.G. Fuh, S. Wu, L.C. Chien, J. Appl. Phys. 40, 6441 (2001)
- 19 M.D. Sarkar, J. Qi, G.P. Crawford, Polymer 43, 7335 (2002)
- 20 D.W. Berreman, Phys. Rev. Lett. **28**, 1683 (1972)
- 21 H. Yokoyama, Mol. Cryst. Liq. Cryst. **165**, 265 (1988)
- 22 L. Xuan, T. Tohyama, T. Miyashita, T. Uchida, J. Appl. Phys. 96, 1953 (2004)