

Cholesteric liquid crystal laser in a dielectric mirror cavity upon band-edge excitation

Yuko Matsuhisa, Yuhua Huang, Ying Zhou, and Shin-Tson Wu

College of Optics and Photonics, University of Central Florida, Orlando, Florida 32816

Yuuki Takao, Akihiko Fujii, and Masanori Ozaki

*Department of Electrical, Electronic and Information Engineering, Graduate School of Engineering,
Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871 Japan*
swu@mail.ucf.edu

Abstract: Low threshold laser action of dye-doped cholesteric liquid crystals (CLCs) is demonstrated using an input circularly polarized light whose handedness is the same as the cholesteric helix of the sample at the high-energy band edge of the reflection band. The mechanism originates from the dramatic increase of the photon density of state at the band edges. We also demonstrate an enhanced laser action of a CLC in a dielectric multilayer cavity. In such a device configuration, the band-edge excitation at high-energy band edge improves the lasing performance not only for the same handedness circularly polarized pump beam as the cholesteric helix but also for the opposite one. It stems from the polarization independence of the dielectric multilayers.

©2007 Optical Society of America

OCIS codes: (230.3720) liquid-crystal devices, (140, 0140) Lasers and laser optics

References and links

1. E. Yablonovitch, "Inhibited spontaneous emission in solid-state physics and electronics," *Phys. Rev. Lett.* **58**, 2059-2062 (1987).
2. S. John, "Strong localization of photons in certain disordered dielectric superlattices," *Phys. Rev. Lett.* **58**, 2486-2489 (1987).
3. D. Roundy, E. Lidorikis, and J. D. Joannopoulos, "Polarization-selective waveguide bends in a photonic crystal structure with layered square symmetry," *J. Appl. Phys.* **96**, 7750-7752 (2004).
4. H. Kosaka, T. Kawashima, A. Tomita, M. Notomi, T. Tamamura, T. Sato, and S. Kawakami, "Superprism phenomena in photonic crystals," *Phys. Rev. B* **58**, R10096 (1998).
5. H. Kajii, Y. Kawagishi, H. Take, K. Yoshino, A. A. Zakhidov, and R. H. Baughman "Optical and electrical properties of opal carbon replica and effect of pyrolysis," *J. Appl. Phys.* **88**, 758-763 (2000).
6. S. T. Wu and D. K. Yang, *Reflective Liquid Crystal Displays* (Wiley, New York, 2001).
7. V. I. Kopp and A. Z. Genack "Apparatus and method for mode selection in a photonic band edge laser," US patent 6,411,635, B1 (June 25, 2002).
8. V. I. Kopp, Z. Zhang and A. Z. Genack, "Lasing in chiral photonic structures," *Prog. Quantum Electron.* **27**, 369-416 (2003).
9. Y. Huang, Y. Zhou, C. Doyle, and S. T. Wu, "Tuning the photonic band gap in cholesteric liquid crystals by temperature-dependent dopant solubility," *Opt. Express* **14**, 1236-1242 (2006).
10. T. Matsui, R. Ozaki, K. Funamoto, M. Ozaki, and K. Yoshino, "Flexible mirrorless laser based on a freestanding film of photo polymerized cholesteric liquid crystal," *Appl. Phys. Lett.* **81**, 3741-3743 (2002).
11. I. P. Il'chishin, E. A. Tikhonov, V. G. Tishchenko and M. T. Shpak, "Generation of tunable radiation by impurity cholesteric liquid crystals," *Sov. JETP Lett.* **32**, 24-27 (1980).
12. T. H. Lin, Y. J. Chen, C. H. Wu, A. Y. G. Fuh, J. H. Liu, and P. C. Yang, "Cholesteric liquid crystal laser with wide tuning capability," *Appl. Phys. Lett.* **86**, 161120 (2005).
13. A. Chanishvili, G. Chilaya, G. Petriashvili, R. Barberi, R. Bartolino, G. Cipparrone, A. Mazzulla, R. Gimenez, L. Oriol, and M. Pinol, "Widely tunable ultraviolet-visible liquid crystal laser," *Appl. Phys. Lett.* **86**, 051107 (2005).
14. G. Chilaya, "Light-controlled change in the helical pitch and broadband tunable cholesteric liquid-crystal lasers," *Crystallography Reports* **51**, Suppl. 1, S108-S118 (2006).

15. S. M. Morris, A. D. Ford, M. N. Pivnenko, and H. J. Coles, "Enhanced emission from liquid-crystal lasers," *J. Appl. Phys.* **97**, 023103 (2005).
 16. Y. Zhou, Y. Huang, and S. T. Wu, "Enhancing cholesteric liquid crystal laser performance using a cholesteric reflector," *Opt. Express* **14**, 3906-3916 (2006).
 17. Y. Zhou, Y. Huang, A. Rapaport, M. Bass, and S. T. Wu, "Doubling the optical efficiency of a chiral liquid crystal laser using a reflector," *Appl. Phys. Lett.* **87**, 231107 (2005).
 18. K. Amemiya, M. H. Song, Y. Takanishi, K. Ishikawa, S. Nishimura, T. Toyooka, and H. Takezoe "Lowering the lasing threshold by introducing cholesteric liquid crystal films to dye-doped cholesteric liquid crystal cell surfaces," *Jpn. J. Appl. Phys.* **44**, 7966-7971 (2005).
 19. Y. Matsuhisa and R. Ozaki, M. Ozaki, and K. Yoshino, "Single-mode lasing in a one-dimensional periodic structure containing helical structure as a defect," *Jpn. J. Appl. Phys.*, **44**, 629-632 (2005).
 20. Y. Matsuhisa and R. Ozaki, K. Yoshino, and M. Ozaki, "High Q defect mode and laser action in one-dimensional hybrid photonic crystal containing cholesteric liquid crystal," *Appl. Phys. Lett.* **89**, 101109 (2006).
-

1. Introduction

Photonic crystals, which exhibit an ordered structure with periodic dielectric constant in an optical wavelength range, have attracted much attention from both fundamental and practical points of view [1-5]. In a photonic crystal, the photon's density of state is suppressed in a certain energy range called photonic band gap or stop band, but enhanced at the edges of the band gap because there the photon's group velocity approaches zero [1].

Cholesteric liquid crystals (CLCs) spontaneously form a one-dimensional (1D) periodic structure and their molecules have a high birefringence. Consequently, CLCs give a periodic modulation for the refractive index and can be regarded as a 1D photonic crystal. The central wavelength of the CLC photonic band gap is determined by $\lambda = np$, where n is the average refractive index and p is the helical pitch length [6]. Different from other photonic crystals, the photonic band gap of the CLCs is polarization dependent. When the incident light is circularly polarized with the same sense as the cholesteric helix, the light is reflected by the stop band. In contrast, the light in the opposite handedness is transmitted through the CLC medium.

Dye-doped CLC lasers have been extensively studied by utilizing the suppression of the photon group velocity and the enhancement of the photon density of state at the band edges because of their advantages such as simple fabrication process, wide tuning range, and mirrorless structure [7-14]. So far, however, the band edge effect due to the enhancement of the photon density of state has only been realized for an output lasing emission. The band edge effect has not been used for an input excitation.

In this paper, we demonstrate, for the first time, the band edge effect not only for output lasing emission but also for input excitation. To investigate the band-edge effect for input excitation, we match the pump laser wavelength to the higher energy band-edge of the CLC by controlling the sample temperature. Consequently, we observe a significant reduction in lasing threshold. Moreover, to achieve higher lasing performance, the CLC and dye materials and resonator structures all need to be taken into consideration. For instance, the laser efficiency can be improved by choosing laser dyes and liquid crystals with higher order parameters [15]. Another approach is to incorporate reflectors to the CLC medium, which decreases the lasing threshold and enhances the lasing efficiency [16-20]. In this paper, we also demonstrate the laser action upon band-edge excitation in the CLC using dielectric mirrors as the reflectors.

2. Sample preparation and experimental setup

To compare the lasing performance, we fabricated two CLC cells with and without dielectric mirrors. The CLC host mixture was prepared by mixing nematic liquid crystal BL006 with 27.5 wt% right-handed chiral agent MLC-6248 (both are from Merck). Afterwards, we doped 0.1 wt% laser dye PM597 (Exciton) to the CLC mixture. In general, the dye concentration in the CLC lasers is ~1-2 wt% and the pump beam is heavily absorbed by the dye molecules.

However, in the present study the incident laser beam is reflected inside the CLC medium several times due to the band edge effect. Thus, in order to prevent absorption saturation we reduce the dye concentration to ~0.1 wt%. For the CLC cell without dielectric mirrors, the CLC mixture was capillary-filled into the cell in an isotropic state. The inner surfaces of the glass substrates were coated with a thin polyimide alignment layer and rubbed in anti-parallel directions to produce ~3° pretilt angle. The cell gap is controlled at 12 μm by the spacer balls. On the other hand, for the CLC cell with dielectric mirrors as shown in Fig. 1, the dielectric multilayer has five pairs of alternately stacked SiO₂ and TiO₂ layers. The refractive indices of the SiO₂ and TiO₂ are 1.46 and 2.35, and their thicknesses are 111 and 69 nm, respectively. The surface of the dielectric mirror was coated with a polyimide alignment layer and gently rubbed. For the convenience of discussion, we abbreviate the CLC cell with dielectric mirrors as DM-CLC. Both cells have same cell gap and same LC materials. The temperature of the sample was monitored by a temperature controller.

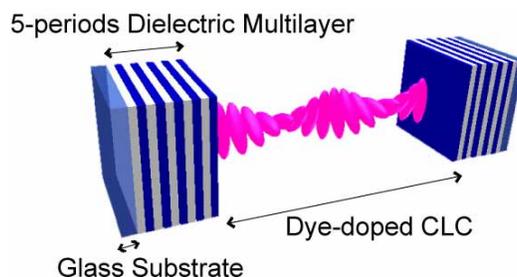


Fig. 1. The CLC cell with dielectric multilayers.

To investigate the laser performance, we used a frequency-doubled, Q-switched, linearly polarized neodymium-doped yttrium aluminum garnet pulsed laser (Minilite II, Continuum) for the excitation. The wavelength, pulse width, and pulse repetition frequency are $\lambda=532$ nm, 4 ns, and 1 Hz, respectively. The pump beam was separated into two paths by a beam splitter: one was sent to an energy meter (Laserstar, Ophir) for monitoring the input energy, and the other passed through a quarter-wave plate and was focused onto the CLC cell. The quarter-wave plate converts the linearly polarized pump beam into right-handed circularly polarized (RCP) or left-handed circularly polarized (LCP) light. The diameter of the irradiation spot on the sample was approximately 160 μm. The emitted laser light from the sample was collected by a lens to a fiber-based spectrometer (Ocean Optics, HR4000, resolution=0.8 nm).

3. Result and discussion

3.1 CLC without dielectric mirrors

We first studied the laser action of the CLC cell without dielectric mirrors. Figure 2(a) shows the measured transmission spectra for the RCP incident light at the indicated temperatures.

At room temperature, the stop band covers the wavelength of the pumping beam ($\lambda=532$ nm) as shown by the dashed lines in Fig. 2. As the temperature increases, the stop band shifts toward the longer wavelength side due to the temperature dependent CLC helical pitch length. At 36.0 °C, the high-energy band edge coincides with the pumping laser wavelength. As the cell is heated to 42.0 °C, the stop band shifts toward a longer wavelength so that the pumping beam wavelength falls outside the stop band. Figure 2(b) shows the normalized lasing spectra for the RCP pump beam as a function of temperature at the pump energy of ~22 μJ/pulse.

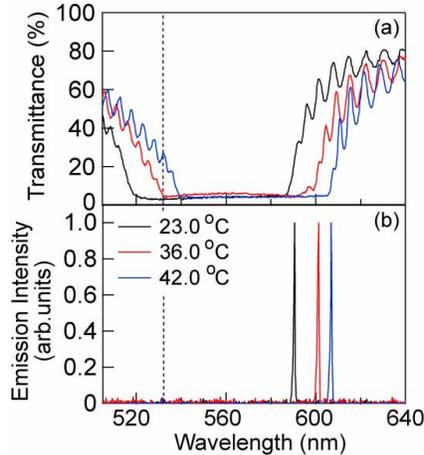


Fig. 2. Transmission and emission spectra of CLC as a function of temperature.

Next, we investigated lasing threshold for LCP and RCP pump beams as a function of high-energy band edge wavelength by changing the sample temperature. Results are shown in Fig. 3. For the LCP pump beam, the lasing threshold gradually increases with the red-shift of the band-edge. This stems from the fluorescence spectrum of the doped dye. The dye used in this experiment exhibits its maximum fluorescence at $\lambda=580$ nm. Therefore, the lasing threshold reaches the lowest when the laser action occurs at 580 nm. Under such a circumstance, the high-energy band edge is located at $\lambda=510$ nm because the stop-band width is ~ 70 nm in this experiment. As a result, the lasing threshold gradually increases when the high-energy band edge departs from $\lambda=510$ nm. On the other hand, for the RCP pump beam the lasing threshold changes drastically with the band-edge wavelength. The lasing threshold for the RCP pump beam is much higher than that for the LCP one when the high-energy band edge is below the pumping laser wavelength 532 nm. This is because the stop band covers the wavelength of the pump beam as shown by the black line in Fig. 2(a). The RCP incident beam is reflected by the stop band. By contrast, the LCP incident beam does not feel the reflection band so that all the excitation energy is able to pump the dye molecules. As a result, the LCP light has a lower lasing threshold than the RCP light at $\lambda < 532$ nm.

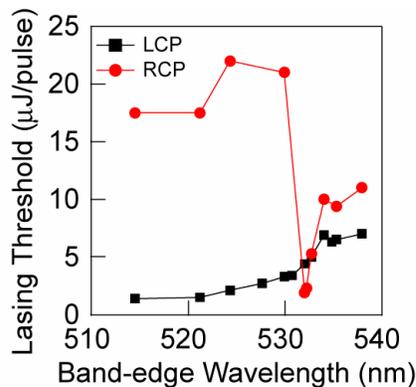


Fig. 3. Band-edge wavelength dependence of lasing threshold for the LCP and RCP pump beams. The band edge wavelength is controlled by the temperature.

Note that the lasing threshold is decreased from 21 $\mu\text{J/pulse}$ to 1.9 $\mu\text{J/pulse}$ and output laser power increased significantly when the band edge coincides with the pumping laser wavelength ($\lambda=532$ nm). Moreover, the lasing threshold for the RCP excitation beam is $\sim 2.3\times$

lower than that for the LCP one because of the band edge effects. At the band edges of the photonic band gap, the density of state and photon dwell time are enhanced due to the multiple internal reflections of the CLC. Therefore, the excitation of the dye molecules is dramatically enhanced, which means much more dye molecules are elevated to the excited state by absorbing the pumping photons. When the band-edge is located at a longer wavelength than 532 nm, the lasing threshold increases again and becomes higher than the threshold for the LCP pump beam due to the reflection induced by interference. From the results, the band-edge excitation is remarkably effective to lower the lasing threshold for the circularly polarized light having the same handedness as the cholesteric helix.

3.2 CLC with dielectric mirrors (DM-CLC)

Figure 4 shows the transmission spectra of the 5-period dielectric multilayer and CLC. From Fig. 4, we find that the pump beam wavelength ($\lambda=532$ nm) is located outside the band gap of the dielectric mirror, and the stop band of the dielectric mirror covers the low-energy band edge of the CLC. We investigated the laser performance of the DM-CLC cell. The red line represents the laser spectrum at 2.1 $\mu\text{J}/\text{pulse}$ of pumping energy. Above a threshold, the emission intensity is drastically increased as the pump energy increases and laser action is observed at the low-energy band edge of the CLC. The full width at half-maximum of the lasing peak is less than 0.8 nm which is the spectral limitation of our spectrometer.

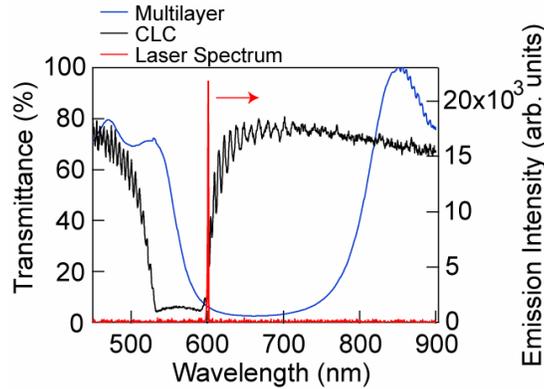


Fig. 4. Transmission spectra of the CLC cell, dielectric mirror, and laser spectrum of DM-CLC cell.

We also investigated the temperature dependence of laser action of the DM-CLC cell. The lasing wavelength shifts to longer wavelength as the temperature increases, which corresponds to the wavelength of the low-energy band edge of the CLC as shown in Fig. 2. Figure 5 shows the lasing threshold and output laser power at 20.0 $\mu\text{J}/\text{pulse}$ of the DM-CLC for the RCP and LCP pumping beams as a function of the wavelength of the high-energy band edge by changing the temperature. We found that the lasing threshold of the DM-CLC sample [as shown in Fig.5 (a)] at any laser wavelengths is much (6.3 \rightarrow 15.5X) lower than that of the CLC without dielectric mirrors (as shown in Fig. 2) for both RCP and LCP pumping beams. This stems from the external feedback of the emission provided by the dielectric mirrors.^{16, 17} Since the stop band of the dielectric mirrors is polarization independent, the feedback induced by the dielectric mirrors is also polarization independent.

For the RCP pumping beam, the lasing threshold and the output laser power strongly depends on the band-edge wavelength. The lasing threshold is high and the output laser power is low at the short wavelengths due to the overlapping with the stop band. However, the lasing performance can be further improved when the higher energy band edge of the CLC matches with the pumping wavelength ($\lambda = 532$ nm). We can see that the lasing threshold for the RCP pumping beam is decreased from 1.4 $\mu\text{J}/\text{pulse}$ to 0.3 $\mu\text{J}/\text{pulse}$ when the higher energy band edge of the CLC overlaps with the pumping wavelength. Moreover, its output laser power is

enhanced more than 13.1X over that at the other band-edge wavelengths for the RCP pump beam. This is contributed by the coincidence of the CLC's band edge with the pumping wavelength.

While for the LCP pumping beam, different from the CLC without dielectric mirrors, the lasing threshold of the DM-CLC for the LCP pumping beam is slightly decreased when the high-energy band edge of the CLC is located at the pumping wavelength. The reason is when the LCP pumping beam passes through the CLC layer after the first dielectric mirror, the pumping beam transmits the CLC layer without any reflection because the handedness of the circular polarized pumping beam is opposite to the helical handedness of the CLC. After the CLC layer, however, a small portion of the LCP pumping beam will be reflected by the dielectric mirror, and in the meantime, the handedness of the circular polarization becomes RCP due to the π phase change at the interface of the dielectric mirror.¹⁴ The RCP pumping beam will be oscillated inside the CLC layer and the band edge effect occurs again. As a result, the lasing threshold of the DM-CLC for the LCP pumping beam is slightly reduced when the high energy band edge of the CLC layer is located at $\lambda = 532\text{nm}$. Accordingly, the output laser power is also dramatically enhanced for the RCP and slightly increased for the LCP pumping beam when the higher energy band edge of the CLC layer is located at the pumping wavelength, as illustrated in Fig. 5(b).

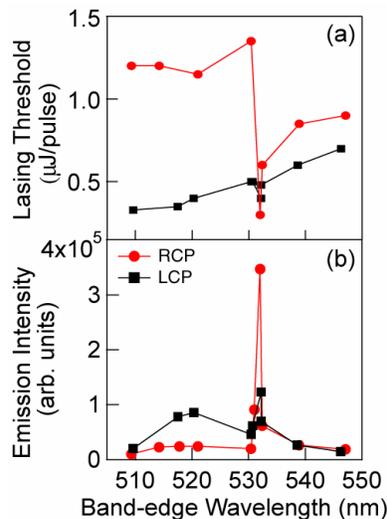


Fig. 5. (a). Lasing threshold and (b). emission intensity at 20 $\mu\text{J/pulse}$ as a function of CLC band-edge wavelength.

Finally, we compare the lasing characteristics of the CLC with and without dielectric mirrors upon band-edge excitation. Figure 6 shows the emission intensity as a function of pump energy of the CLC with and without dielectric mirrors for the RCP pump beam when the high-energy band edge coincides with the pumping beam wavelength. The lasing threshold of the CLC cell is 1.9 $\mu\text{J/pulse}$ (shown in Fig. 3) while the threshold of the DM-CLC is 0.3 $\mu\text{J/pulse}$ (shown in Fig. 5), which is reduced by $\sim 6.3\text{X}$. Moreover, by adding the dielectric mirrors the output lasing power is enhanced by $\sim 3.4\text{X}$.

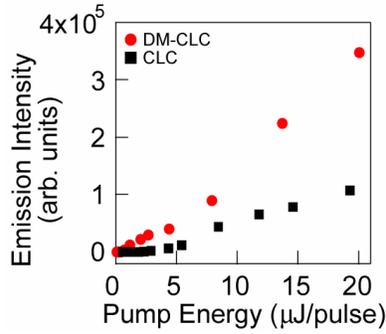


Fig. 6. Emission intensity as a function of pump energy of CLC with and without dielectric mirrors upon band-edge excitation.

4. Conclusion

We investigated the laser action of the dye-doped CLC with right handed helix and achieved a lower lasing threshold by the band-edge excitation for the RCP pump beam. We also demonstrated laser action of the CLC with the dielectric mirrors and found that its output lasing power was much higher and lasing threshold lower than that of the CLC without the dielectric mirrors. Furthermore, the band-edge excitation helps to improve the lasing performance of the CLC with the dielectric mirrors not only for RCP pump beam but also for the LCP one. This is because the employed dielectric mirror is polarization independent.

Acknowledgment

The authors would like to thank R. Ozaki from National Defense Academy for useful discussions. This work is partially supported by Grant-in-Aids for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan and Japan Society for the Promotion of Science. The UCF part is supported by AFOSR under contract FA9550-05-C-0057.