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Low-threshold and high efficiency lasing upon band-edge excitation in a cholesteric liquid crystal

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A low threshold and high efficiency laser based on dye-doped cholesteric liquid crystals (CLCs) is demonstrated using an input excitation with the same handedness of circular polarization as the helical structure of the sample at the shorter wavelength band edge of the reflection band. The responsible mechanism originates from the dramatic increase of the optical density of state (DOS) at the band edges. The calculated DOS of the CLC system confirms the authors' experimental results. © 2007 American Institute of Physics. [DOI: 10.1063/1.2710777]

Liquid crystals containing chiral molecules such as cholesteric liquid crystals (CLCs), ferroelectric liquid crystals, cholesteric blue phases, and cholesteric glasses have recently attracted great attention as self-organized photonic crystals. Different from other photonic crystals, the photonic band gap of the CLC is polarization dependent. A circularly polarized light in the opposite handedness to the liquid crystal helix transmits through the CLC, while for the one in the same handedness a photon density of state (DOS) vanishes in the photonic band gap.^{1,2} Since the photon group velocity approaches zero and DOS is enhanced at the edge of the photonic band gap, low-threshold lasers have been theoretically predicted³ and experimentally demonstrated.^{4–16} Moreover, an anomalous strong absorption occurs at the band edge of an absorbing CLC. A low-threshold CLC laser upon such phenomena has been theoretically predicted.¹⁷ So far, however, no experimental demonstration has been carried out.

In this letter, we experimentally demonstrate the low-threshold laser action upon the band-edge effect due to the anomalous absorption in a dye-doped CLC. To enhance the band-edge effect, we match the pump laser wavelength to the higher energy band edge of the photonic band gap of the dye-doped CLC by controlling the operating temperature. We also perform theoretical calculation about the DOS, which shows good agreement with the experimental result.

The CLC host was prepared by mixing 72.5% nematic liquid crystal BL006 with 27.5% of a right-handed chiral dopant MLC6248 (both are from Merck). Afterwards, a 0.1 wt % laser dye PM597 (exciton) was doped into the mixture. The CLC was filled into a 12- μm -thick LC cell by capillary action. The inner surfaces of the glass substrates were coated with a thin polyimide alignment layer and rubbed in antiparallel directions. The pretilt angle is $\sim 3^\circ$. The helical axis of the CLC was perpendicular to the glass substrates. The temperature of the sample was controlled by a temperature controller.

To investigate the laser emission properties, a second harmonic laser from a Q-switched neodymium-doped yttrium aluminum garnet pulsed laser (MiniliteII, Continuum) with vertical linear polarization was used for the excitation. The wavelength, pulse width, and pulse repetition frequency are $\lambda = 532$ nm, 4 ns, and 1 Hz, respectively. The linear polarized light beam was converted into right-handed circularly polarized (RCP) or left-handed circularly polarized (LCP) light by a quarter-wave plate and was focused by a lens onto the sample. The spot diameter on the sample was measured to be about 160 μm . The emitted laser light from the sample was collected into a fiber-based spectrometer (Ocean Optics, HR4000).

The dashed lines in Fig. 1(a) show the transmission

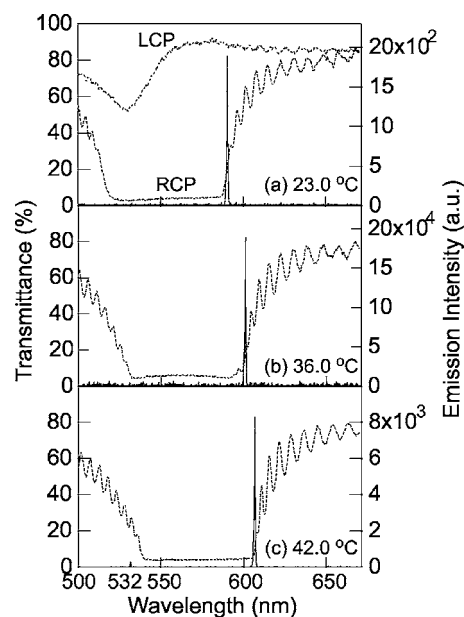


FIG. 1. (a) Transmission spectra of the CLC system for the RCP and LCP incident lights and the emission spectrum at 23.0 °C. [(b) and (c)] Transmission spectra for the RCP incident light and emission spectra at 36.0 and 42.0 °C.

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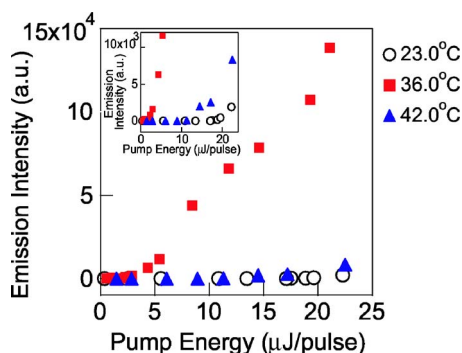


FIG. 2. Emission intensity as a function of pumping energy at 23.0, 36.0, and 42.0 °C.

spectra of the dye-doped CLC for the LCP and RCP incident lasers at 23.0 °C. For the LCP incident light, the absorption of the laser dye was observed at $\lambda \sim 530$ nm. On the other hand, for the RCP incident light, the suppressed transmittance in the spectral range from 519 to 590 nm is due to the stop band of the CLC. The stop band covers the wavelength of the excitation beam ($\lambda = 532$ nm) at room temperature. The dashed lines in Figs. 1(b) and 1(c) show the transmission spectra for the RCP incident light at 36.0 and 42.0 °C, respectively. The stop band shifts toward the longer wavelength side as the temperature increases, which is attributed to the temperature dependence of the helical pitch length of the CLC. At 36.0 °C, the high-energy band edge is located at the wavelength of the excitation beam, as Fig. 1(b) shows. When the temperature is increased to 42.0 °C, the stop band is shifted to the 540–607 nm spectral range. In this case, the pumping laser wavelength is outside the stop band, as shown in Fig. 1(c).

The solid lines in Fig. 1 show the emission spectra as a function of temperature when the pump energy is ~ 22 $\mu\text{J/pulse}$. Above a threshold, the laser action was observed at the low-energy band edge. Continue to increasing the temperature, the lasing wavelength shifts to a longer wavelength, which is in accordance with the redshift of the band edge. Figure 2 shows the emission intensities as a function of pumping energy. The lasing efficiency strongly depends on the temperature. As shown in the inset of Fig. 2, the lasing efficiency at 23.0 °C is approximately 4.4 times less than that at 42.0 °C. From Fig. 1, the stop band overlaps the excitation wavelength and a portion of the pump energy is reflected at 23.0 °C. While at 42.0 °C, since the excitation wavelength is outside the reflection band, all the excitation energy is able to pump the dye molecules. As a result, higher optical gain is achieved in the sample at 42.0 °C than that at 23.0 °C. Consequently, higher laser efficiency is obtained when the sample temperature is controlled at 42.0 °C.

The most interesting discovery is that the lasing efficiency at 36.0 °C is further enhanced by ~ 17 times as compared to that at 42.0 °C. This is contributed by the band-edge effect. At the edges of the photonic band gap, the DOS and photon dwell time are significantly enhanced because of the multiple internal reflections of the CLC. Therefore, when the band edge is within the absorption spectrum of the laser dye, the excitation of the dye molecules is dramatically enhanced, which means much more dye molecules are elevated to the excited state by anomalous absorption of the pumping photons. As a result, the laser efficiency is dramatically enhanced.

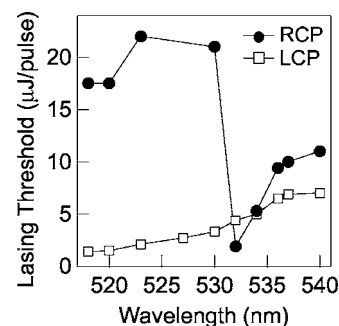


FIG. 3. Lasing threshold as a function of wavelength of high-energy band edge for the LCP and RCP excitation beams.

We also investigated the lasing threshold of the dye-doped CLC sample for the RCP and LCP pumping beams as a function of the wavelength of the high-energy band edge by changing the temperature. The results are shown in Fig. 3. For the LCP excitation beam, the lasing threshold gradually increases with the redshift of the band edge. This stems from the spontaneous emission spectrum of the doped dye, which determines the optical gain. The dye used in this experiment exhibits its maximum spontaneous emission at $\lambda = 580$ nm. Therefore, the lasing threshold reaches the lowest when the lasing wavelength occurs at 580 nm and then gradually increases when the lasing wavelength departs from $\lambda = 580$ nm.

On the other hand, for the RCP excitation beam the lasing threshold is high when the shorter band edge of the band gap is below the pumping wavelength $\lambda = 532$ nm. This phenomenon can be explained as follows. At a short wavelength, the stop band covers the wavelength of the excitation beam, as Fig. 1(a) shows. As a result, the incident beam is reflected by the stop band, which causes the lasing threshold to increase.¹⁸ Note that the lasing threshold is significantly decreased from 21 to 1.9 $\mu\text{J/pulse}$ when the shorter band edge is located at the pumping wavelength of 532 nm. Moreover, the lasing threshold for the RCP excitation beam is ~ 2.3 times smaller than that for the LCP one. This is contributed by the above-mentioned band-edge effects. When the band-edge wavelength gets longer than 532 nm, the lasing threshold increases again and becomes higher than the threshold for the LCP excitation beam, which is attributed to the reflection induced by interference. From the result, the band-edge excitation is remarkably effective to increase the lasing efficiency and lower the lasing threshold.

To confirm the experimental results, we calculated the DOS of the CLC system^{19,20} using the 4×4 matrix method.²¹ In our calculations, the thickness and the extraordinary and ordinary refractive indices of the CLC are 12.0 μm and 1.700 and 1.506, respectively, according to our experimental conditions. The absorption of the CLC medium is neglected. We investigated the DOS as a function of wavelength of the high-energy band edge for LCP or RCP incident light by changing the helical pitch. As plotted in Fig. 4, the DOS for the LCP incident light is insensitive to the wavelength of the band edge. This is because a circularly polarized light in the opposite handedness to the CLC helix could not interact with the CLC medium. On the other hand, for the RCP incident light, the DOS at a shorter wavelength is smaller than that of the LCP light due to the overlapping with the stop band. The low DOS reduces the optical gain. However, in the vicinities of $\lambda = 532$ nm the DOS is drastically

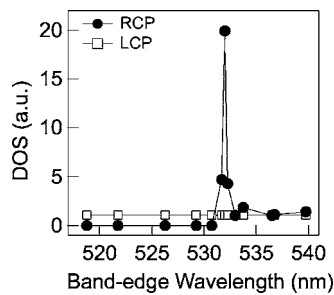


FIG. 4. DOS as a function of wavelength of high-energy band edge for the LCP or RCP incident light.

increased to 20 because the band edge coincides with the pumping wavelength. This high DOS greatly enhances the lasing efficiency and decreases the lasing threshold at 532 nm, as shown in Fig. 3. The calculated results agree well with our experimental observations.

In conclusion, we have demonstrated a low-threshold laser action in dye-doped CLC based on band-edge excitation. The lasing efficiency is dramatically enhanced and the lasing threshold significantly decreased by the band-edge excitation. Our theoretical calculations on the DOS in the CLC confirm our experimental results.

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