Cholesteric liquid crystal laser with wide tuning capability

Tsung-Hsien Lin, Yi-Jan Chen, Chun-Hui Wu, and Andy Y.-G. Fuh\textsuperscript{a)}

Institute of Electro-optical Science and Engineering and Department of Physics, National Cheng Kung University, Tainan, Taiwan 701, Republic of China

J.-H. Liu and P.-C. Yang

Department of Chemical Engineering, National Cheng Kung University, Tainan, Taiwan 701, Republic of China

(Received 13 October 2004; accepted 16 February 2005; published online 15 April 2005)

This letter examines a planar cholesteric cell (CLC) doped with two collocated laser dyes as a one-dimensional photonic crystal. Adding phototunable chiral material (AzoB) allows the CLC photonic crystal to be lased at the band edges of the photonic band gap with a tuning range of over 100 nm. Tuning is performed by irradiating the chiral AzoB material with UV light, causing the material to undergo trans-cis isomerization in the CLC film. The tuning range is the visible region from 563 to 667 nm. Moreover, the tuning is reversible. © 2005 American Institute of Physics. [DOI: 10.1063/1.1897439]

The use of liquid crystals as photonic crystals (PCs) was investigated. PCs, which have a periodic dielectric structure with a periodicity in the range of optical wavelengths, have attracted much interest, because they have potential fundamental uses and practical applications. Photonic crystals have band gaps that are analogous to electronic band gaps in semiconductors. The group velocity of the photonic band edge is real and tends towards zero, so the gain is much higher at the edge of the photonic band. Planar cholesteric liquid crystals (CLCs) with a pitch that is comparable to the optical wavelength can be regarded as a one-dimensional photonic band gap material. When linearly polarized light propagates into a right-handed CLC cell along the helical axis, right-handed circularly polarized light is reflected if the wavelength of the light, $\lambda$, equals $np$ ($\lambda = np$), where $n$ is the mean refractive index of a liquid crystal and $p$ is the pitch of the helix.

Lasing action is expected at the edges of the band gap in the one-dimensional photonic crystal (1D PC), in which the density of states of light exhibits a narrow singularity. Equivalently, the group velocity of light approaches zero at the band edges. Dye-doped CLC, which is a 1D PC, has been observed to exhibit laser activity at its band edges. The primary role of the CLC in these laser systems is as a distributed cavity host for active materials, such as the DCM laser dyes used in the experiment performed herein. Some works have investigated control of the laser action in the dye-doped CLC system, using the external stimuli of mechanical stress, temperature, an electric field, and an optical field.

The authors recently reported lasing in chiral photonic liquid crystals. Adding a tunable chiral monomer (TCM) enables the lasing frequency to be tuned over a range of 30 nm. However, the process is irreversible. This letter reports a CLC laser with a tuning range of over 100 nm, and reversible tuning.

A left-hand CLC sample was prepared by mixing a chiral material (S811, Merck) with a nematic liquid crystal (ZLI2293, Merck) in a suitable ratio. Notably, the Bragg reflection edges of the dye-doped CLC sample had to be within the emission spectrum of the laser dye to induce lasing. After homogeneous mixing, 0.5% of two laser dyes, 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostryl)-4H-pyran (DCM, Exciton) and Pyrromethene 580 (P580, Exciton) were dissolved in the cholesteric host. Finally, left-hand chiral molecular AzoB was added. Figure 1 presents the chemical structure of AzoB. It does not have a liquid crystalline phase. The trans-form and cis-form AzoB influence the twisting power of the cholesteric liquid crystal cells to different degrees. The final compound was injected into an empty cell that was made from two glass plates coated with indium tin oxide and separated by a 25-μm-thick spacer. The surfaces of the glass plates were coated with a polyimide and rubbed to form a homogeneously aligned cell. The sample thus formed was a planar CLC whose helical axis was perpendicular to the surface of the cell.

The pumping source of the dye-doped CLC cells was a single pulse of the second-harmonic generation (SHG) laser ($\lambda = 532$ nm) from a Q-switched Nd:yttrium–aluminum–garnet (YAG) laser. The duration of the Q-switched pulse was about 8 ns. The pumping laser was focused on to the sample at an angle of incidence of 45° to the normal of the surface, using a lens with a focal length of $f = 10$ cm. The CLC cells were lased in the direction of the surface normal. A detector connected to a spectrometer recorded the lasing intensity from the cell.

Figure 2 shows the fluorescence spectra of the two laser dyes that were dissolved in a nematic liquid crystal (ZLI2293) and pumped by a single pulse from the SHG laser ($\lambda = 532$ nm) Nd:YAG laser. The dotted line is a fluorescence spectrum of Pyrromethene 580 with a peak at $\sim 570$ nm; the solid line is the DCM fluorescence spectrum with an emis-

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\textsuperscript{a)}Author to whom correspondence should be addressed; electronic mail: andyfuh@mail.ncku.edu.tw

![FIG. 1. Chemical structure of AzoB.](image-url)
sion peak at ~610 nm. The emission spectrum became broad when the two dyes are added, so the lasing could be tuned over a wide range.

Figure 3 depicts the shifts of the reflection band of a CLC cell doped with dyes, chiral dopants of S811 and AzoB irradiated with UV light at 350 nm for various durations at room temperature. The dopant AzoB undergoes a transformation from the trans- to the cis-form when irradiated at UV wavelengths. Azo derivatives usually exist in the relatively stable trans-form. Figure 3 presents the blueshift of the reflection band under UV irradiation. This result is consistent with that reported by Lee et al. According to Ref. 16, such a blueshift is caused by the photoisomerization of the azobenzene derivatives from the trans- to cis-configuration. The rod-like trans-azobenzene molecule promotes the stabilization of the cholesteric phase. However, the bent cis-azobenzene molecule tends to disorganize the molecular orientations of the host liquid crystal phase, changing the geometrical structure rather than the chirality of the AzoB derivatives.

Experimentally, the pitch of a CLC cell was modified by adding S811 to produce a reflection band initially with a long-wavelength edge at ~520 nm. Adding left-hand AzoB TCM changes the pitch to shift the reflection band with the long-wavelength edge to 670 nm. Essentially, an intrinsic shift occurs to a shorter wavelength when AzoB is introduced to the left-handed host. However, the twisting power of AzoB is much smaller than S811 and can be neglected. And the shift to longer wavelength caused by the conformation of the azobenzene unit compensates for this shift. Here, the shift induced by the conformation of the AzoB is believed to be larger than that caused by its molecular chirality, so that a total shift of λ to longer wavelength was observed in the left-handed host. Similar results were observed in Ref. 17. The mixing ratio of the final compound was ZLI 2293 (62.2 wt %), S811 (31.1 wt %), and TCM (AzoB) (6.7 wt %). In this case, under UV irradiation, the period of exposure governed the wavelength of the long-wavelength reflection band edge from 670 to 560 nm. The range of the shift was within the width of the combined fluorescence spectra of the two laser dyes, and so was expected to generate laser light from red to green. When the cells are kept in the dark at room temperature, the cis-states return gradually to their initial stable trans-states by the thermally induced cis to trans-backisomerization of azobenzene.

Figure 4 plots the variation of the lasing wavelength of the sample with the period of exposure to UV, when the sample is pumped using a Q-switched Nd:YAG laser (λ = 532 nm, 8 ns, and 1 μJ/pulse). A comparison of Fig. 3 with Fig. 4 reveals that lasing always occurs at the long-wavelength edge of the CLC reflection band, and can be tuned by changing the UV exposure time. Initially, the wavelength of lasing was 667 nm. It was tuned to 563 nm by irradiating the sample for ~20 min. At this time, the AzoB molecules were isomerized completely to the cis state. The cis-AzoB molecules were inversely isomerized by heating,
and returned to the transform at room temperature, so the lasing wavelength was expected to relax to the initial wavelength of ~667 nm. Figure 5 plots the measurements. The molecules took approximately a day to relax. The lasing wavelength changed insignificantly in an hour.

Stable lasing at the two edges of the CLC reflection band was observed as the pumping energy was increased to ~3 μJ, as shown in Fig. 6. In order to verify the cause of the two lasing wavelengths, a separate experiment with the sample doped only DCM dye was performed. Two lasing wavelengths similar to Fig. 6 were observed as the pumping energy was increased to ~3 μJ. It indicates that direct dye excitation rather than the transform mechanism (Foster or Dexter) between the DCM and P580 dyes is the cause. Increasing the pumping energy enabled the long and short wavelength stop band edges (LWSE and SWSE) to obtain sufficient gain to lase. However, it is noted in Fig. 6 that the intensity of the SWSE lasing is smaller than that of LWSE one. This is due the lasing threshold is considerably higher at the SWSE than at the LWSE.19

In conclusion, this work demonstrates the feasibility of a CLC laser with a tuning range of over 100 nm. Doping with two laser dyes broadens the emission spectrum of the CLC sample. The addition of a TCM (AzoB) supported the wide-range tuning of lasing. The lasing is stable, and can be tuned over 100 nm. Additionally, the tuning is reversible. All these characteristics enable the device to be used in various applications.

The authors would like to thank the National Science Council (NSC) of the Republic of China (Taiwan) for financially supporting this research under Contract No. NSC 93-2112-M-06-016.