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Citation: Applied Physics Letters **96**, 111105 (2010); doi: 10.1063/1.3364942
View online: http://dx.doi.org/10.1063/1.3364942
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/96/11?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS **96**, 111105 (2010)

Optically band-tunable color cone lasing emission in a dye-doped cholesteric liquid crystal with a photoisomerizable chiral dopant

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(Received 31 December 2009; accepted 24 February 2010; published online 16 March 2010)

This work demonstrates the feasibility of an optically band-tunable color cone lasing emission (CCLE) based on a dye-doped cholesteric liquid crystal with a photoisomerizable chiral dopant. Experimental results indicate that the lasing band of the formed CCLE can be tuned optically among various color regions by adjusting the UV irradiated fluence. The optical band tunability of the laser is attributed to the presence of two chiral agents with twisting powers of opposite signs in the cell and the UV-irradiation-induced decrease of the right-handed twisting power of the photoisomerizable chiral dopant via *trans*→*cis* isomerization, subsequently inducing the other chiral agent to reduce the structural pitch of the cell. Total tunable wavelength range of the laser exceeds 100 nm. Moreover, the band-tunable laser exhibits a high spectral stability under illumination of a visible light or thermal treatment. © *2010 American Institute of Physics.* [doi:10.1063/1.3364942]

Dye-doped cholesteric liquid crystal (DDCLC) lasers have been thoroughly studied owing to their ease of fabrication, their interesting distributed feedback (DFB) lasing mechanism associated with the unique photonic crystal-like band structure of the cholesteric liquid crystal (CLC), and related potential applications.[1–6,8] As the fluorescence dyes doped in the CLC are excited, the spontaneously emitted fluorescence within the stop bands is inhibited to propagate but enhanced at the band edges. The fluorescence at band edges can propagate via a multireflection,[7] subsequently producing an extremely small group velocity and an extremely high density of photonic state. Based on the DFB effect of the active DDCLC multilayer in the multireflection process, the rates of spontaneous and stimulated emissions at band edges can both be amplified, enabling a high gain to exceed the loss for a low-threshold lasing emission.[1]

Our recent study demonstrated that a "single pitched" DDCLC laser can simultaneously emit a wide-band lasing emission with an angular dependence on the wavelength.[8] This emission is called color cone lasing emission (CCLE), which differs from the conventional scenario that, at most, two lasing peaks at the band edges can simultaneously occur along the helical axis in a DDCLC.[1–6] To implement such a CCLE in practice, this study elucidates the optical band tunability of a color cone laser based on a DDCLC cell with a photoisomerizable chiral dopant. Experimental results indicate that the lasing band of the DDCLC laser can be tuned optically among various color regions by varying the UV irradiated fluence. The basic mechanism responsible for inducing the band tunability of the laser is attributed to the presence of two chiral agents with twisting powers of opposite signs in the cell and the decay of the right-handed twisting power of the photoisomerizable chiral dopant via *trans* →*cis* isomerization, subsequently causing the other chiral agent to reduce the structural pitch of the cell. The overall tunable wavelength range for the tunable laser exceeds 100 nm. Furthermore, the band-tunable laser displays a high spectral stability under illumination of a visible light or thermal treatment.

The nematic liquid crystal (NLC), left-handed chiral agent, two laser dyes, and photoisomerizable chiral dopant (right-handed) used herein are ZLI2293 (from Merck), S811 (from Merck), 4-Dicyanmethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) and Pyrromethene 567 (P567) (from Exciton), and Isosorbide 2,5-bis(4-methoxy-cinnamate) (IBM), respectively. The homogeneous mixture, with a mixing ratio of ZLI2293:S811:IBM:DCM:P567 of 66.29:31.42:1.47:0.57:0.25 wt %, is prepared. The helical twisting power (HTP) values of S811 and IBM in ZLI2293 are -10.358 and 39.8 μm^{-1}, respectively. The empty cell is prefabricated by integrating two indium-tin-oxide-coated glass slides separated with two 23 μm thick plastic spacers. Both glass slides in the empty cell are precoated with a polyvinyl alcohol (PVA) film, and prerubbed in a parallel direction. The above mixture is then injected into the empty cell in order to form a DDCLC cell via capillary effect. Next, the DDCLC cell is placed in a clean and opaque specimen box at room temperature for about 7 days, enabling CLC in the cell to self-organize slowly to a perfect planar structure. The determined absorption and fluorescence emission spectra (not shown herein) of the DDCLC cell in the isotropic phase show that the maxima of the absorption and fluorescence emission of the cell are located at about 526 and 574 nm, respectively. When the wavelength exceeds 575 (690) nm, the absorption (the fluorescence emission) is extremely weak and can be neglected.

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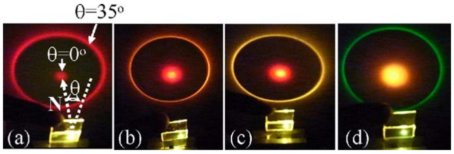

FIG. 1. (Color online) Four obtained CCLE patterns distributed from long- to short-wavelength region based on the DDCLC cell under preillumination of one UV light with $I_{UV}=3\ mW/cm^2$ and $t_{UV}=$(a) 0 s, (b) 20 s, (c) 80 s, and (d) 160 s, respectively.

Because the wavelength (532 nm) of the pumped pulses is near 526 nm, the cell can be excited efficiently.

This work employs two experimental setups for detecting the reflection and lasing spectra of the DDCLC cell, respectively, at each oblique angle θ (relative to the cell normal **N**). Our previous study includes the same setups and the same method and parameters in the measurements of reflection and lasing spectra of the cell and, therefore, are not repeated here for brevity.[8] The optically band-tunable lasing experiments are performed using one UV light source (<400 nm) with a changeable intensity (I_{UV}) to preirradiate or not the cell for a variable time (t_{UV}) before the reflection and lasing spectra measurements.

Figures 1(a)–1(d) display four obtained CCLE patterns on the screen (behind the cell) by the excitation of the pumped pulses ($E=15\ \mu J/pulse$), if the DDCLC cell with the photoisomerizable IBM is preirradiated by one uniform UV beam with a fixed irradiated intensity of $I_{UV}=3\ mW/cm^2$ and a variable irradiated time of $t_{UV}=0$ s, 20 s, 80 s, and 160 s, respectively. Figures 2(a)–2(d) further display the detailed lasing spectra of the four obtained CCLEs in Figs. 1(a)–1(d), respectively, as well as the corresponding reflection spectra at various oblique angles ($\theta=0°-50°$). Obviously, the wavelength of the lasing signal in each CCLE decreases with an increasing oblique angle and is located close to the long wavelength edge (LWE) of the CLC reflection band (CLCRB) at each oblique angle. Our previous study includes both the explanation of the mechanism for the formation of the CCLE and its related features and, therefore, are not repeated here for brevity.[8] In particular, the red, orange, yellow, and green lasing rings at nearly 35° [as displayed in Figs. 1(a)–1(d), respectively] are similar in that their wavelength at 35° is close to the value at which the LWE of CLCRB, measured at nearly 35°, overlaps the short wavelength edge of CLCRB, measured at 0°; briefly, $\lambda_{LWE}(35°)=\lambda_{SWE}(0°)$ [as evident in the reflection spectra measured at 35° and 0° (green and black, respectively) in Figs. 2(a)–2(d)]. As is widely assumed, beams of fluorescence with the same wavelength $[\lambda_{LWE}(35°)=\lambda_{SWE}(0°)]$ and propagating at nearly 35° and 0° may reinforce indirectly each other owing to the enhancements of the related respective rates of spontaneous emission.[8] Efforts are currently underway to thoroughly elucidate the formation of the unique lasing ring at nearly 35° in each CCLE. Instead, this study investigates the optical band-tunable feature of such a DDCLC color cone laser.

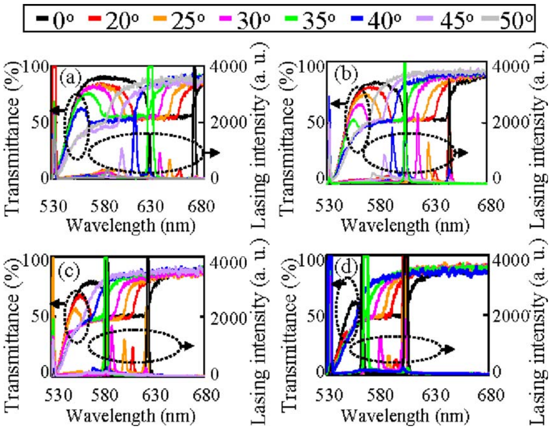

FIG. 2. (Color online) Detailed lasing spectra of the four obtained CCLE patterns and corresponding reflection spectra measured at $\theta=0-50°$ based on the DDCLC cell under preillumination of one UV light with $I_{UV}=3\ mW\ cm^{-2}$ and $t_{UV}=$(a) 0 s, (b) 20 s, (c) 80 s, and (d) 160 s, respectively.

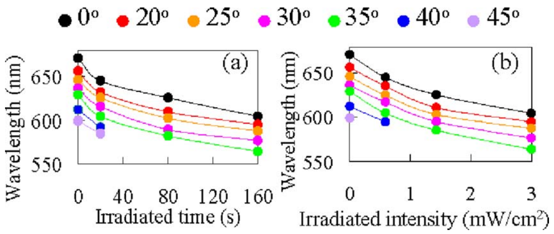

FIG. 3. (Color online) Variations in the wavelength of the lasing signals at the LWE of CLCRB in the obtained CCLE patterns at $\theta=0°-45°$ with (a) the irradiated time (0–160 s) and (b) the irradiated intensity ($0-3\ mW/cm^2$) of the UV light based on the DDCLC cell.

Next, rather than the irradiated time, the irradiated intensity of the UV light on the cell is changed to examine the optical band-tunability of the CCLE in the DDCLC. Four CCLE patterns and their detailed lasing spectra and the corresponding reflection spectra of the cell measured at $\theta=0°-50°$ (not shown herein), which resemble those lasing patterns displayed in Figs. 1(a)–1(d) and those spectra in Figs. 2(a)–2(d), respectively, can also be obtained if the cell is preilluminated by the UV light with a fixed $t_{UV}=160$ s and a variable $I_{UV}=0\ mW/cm^2$, $0.59\ mW/cm^2$, $1.43\ mW/cm^2$, and $3\ mW/cm^2$, respectively. Figures 3(a) and 3(b) plot similar experimental results with respect to the detailed lasing bands within 0°–45° in the tunable CCLE by varying t_{UV} and I_{UV}, respectively. Such a similarity between the variations of the lasing wavelength of the CCLE with t_{UV} and I_{UV} implies that the UV irradiated fluence (i.e., the product of t_{UV} and I_{UV}) is responsible for the band-tunable effect of the color cone laser. Therefore, experimental results in Fig. 3 clearly indicate that the lasing band of the laser can be tuned optically from the long-wavelength region of 599.35–671.11 nm (orange-reddish) to the short-wavelength region of 564.27–604.42 nm (greenish-orange) by increasing the UV irradiated fluence from 0 to $480\ mJ/cm^2$. This optical band tunability of the DDCLC color cone laser differs from the conventional scenario in which normally only a wavelength of edge lasing signal alone emitted at 0° is optically tunable in common DDCLC lasers described elsewhere.[2–5]

Because a separate experiment (data not shown) has confirmed the lack of such a band-tunable feature of the CCLE based on a similar DDCLC cell without IBM chiral agent, the optical band tunability of the CCLE illustrated above (Figs. 1–3) is certainly attributed to the modification of the structural pitch of the cell via the UV-irradiation-induced *trans*→*cis* isomerization of the isomerizable IBM. Details are as follows. The chiral dopant, IBM (synthesized and offered by our colleagues, J.-H. Liu *et al.*), is a right-handed

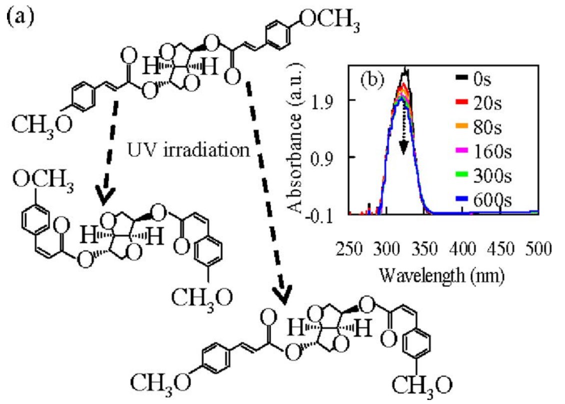

FIG. 4. (Color online) (a) Chemical structures of the photoisomerizable chiral dopant IBM at rodlike *trans*- and bent *cis*-states. (b) Variation in the measured absorption spectra (250–500 nm) of the IBM in ZLI2293 (without doping laser dyes) in the isotropic state with the UV irradiated time (0–600 s) at a fixed intensity of 3 mW/cm^2.

chiral compound with two cinnamoyl (C═C) photochromatic group.[9] Figure 4(a) shows the chemical structures for the *trans*- and *cis*-isomers of IBM. Generally, this isomerizable chiral agent remains stable at a rodlike *trans*-state in the dark. Once irradiated by the UV light, the *trans*-IBM converts into the *cis*-state with two possible bent structures [Fig. 4(a)] via *trans*→*cis* isomerization. Figure 4(b) shows the decrease in the absorption of IBM in ZLI2293 (without doping laser dyes) in the isotropic state within the spectrum region of 300–340 nm when increasing the irradiated time of the UV light from 0 to 600 s at a fixed intensity of 3 mW/cm^2. Notably, a similar finding (data not shown) is also obtained if the UV irradiated intensity is increased at a fixed irradiated time. Experimental results in Fig. 4(b) are because the concentration of *cis*-isomer via the continued *trans*→*cis* isomerization can increase when increasing the UV irradiated time/intensity. The *cis*-isomers in the DDCLC cell may disorganize the local order of the NLC molecules due to their bent structures, resulting in the subsequent decay of the right-handed twisting power of IBM in NLC. Owing to the presence of the two chiral agents (S811 and IBM) with twisting powers of opposite signs in the cell and the UV-irradiation-induced decay of the right-handed twisting power of IBM in the cell via *trans*→*cis* isomerization, the power of the other chiral agent (S811) to twist NLC can rise to decrease the structural pitch of the cell. Accordingly, as the UV irradiated time or intensity gradually increases, the pitch can gradually contract in the cell. This explains why CLCRB and the band edges, and thus, the lasing band of the obtained CCLE can gradually blueshift with increasing the UV irradiated fluence (Fig. 3). However, as the LWE of the CLCRB for large oblique angles (>45°) enters the absorption region of the laser dyes (≤ 575 nm), the strong reabsorption effect of the fluorescence significantly diminishes the likelihood of lasing emission at those large angles. Consequently, the angle range and thus the band of lasing emission decrease with the increase of the UV irradiated fluence (Fig. 3).

This study also examines the spectral stability for the tunable DDCLC color cone laser. Experimental results indicate that, under the post-illumination of a visible light or post-thermal treatment (i.e., several heating-cooling cycles of the cell), the lasing band of the tunable color cone laser remains unchanged in the spectrum. This finding suggests that the *trans*→*cis isomerization for IBM is irreversible, which is in contrast with the unstable features of usual azo-mate*rials, in which the cis-dyes can reverse to *trans*-state by slow thermal or rapid optical (under the illumination of a light in long-wavelength region) *cis-trans* back isomerization.[2,4,5] Efforts are currently underway in our laboratory to demonstrate that neither a CCLE effect nor an associated band-tunable feature are found in a pumped pulses-induced unstable and, thus, an imperfect CLC structure based on a DDCLC with a reversibly photoisomerizable azo-material. Consequently, the irreversibility of the IBM significantly increases the spectral stability of such a tunable color cone laser described in this study.

In summary, this work develops an optically band-tunable color cone laser based on a DFB DDCLC cell with a photoisomerizable chiral dopant. Experimental results indicate that the lasing band of the formed CCLE based on the DDCLC cell can be optically tuned among various color regions by varying the UV irradiated fluence. The total optically tunable range of the lasing band for the DDCLC laser exceeds 100 nm. Such a laser with a wide and stable optically band tunability presented herein has unique advantages beyond those unstable single-wavelength-tunable lasers based on azo-associated DDCLC cells reported previously.

The authors would like to thank the National Science Council of the Republic of China, Taiwan (Contract No. NSC 97-2112-M-006-013-MY3) and the Advanced Opto-electronic Technology Center, National Cheng Kung University, under projects from the Ministry of Education for financially supporting this research. Ted Knoy is appreciated for his editorial assistance.

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