High efficiency cholesteric liquid crystal lasers with an external stable resonator

Hamidreza Shirvani-Mahdavi,[1,2] Shima Fardad,[2] Ezeddin Mohajerani,[1] and Shin-Tson Wu[2*]

[1]*Laser and Plasma Research Institute, Shahid Beheshti University, G.C., Evin St., Tehran, Iran*
[2]*College of Optics and Photonics, University of Central Florida, Orlando, Florida 32816, USA*
**swu@mail.ucf.edu*

Abstract: An amplified cholesteric liquid crystal (CLC) laser performance is demonstrated by utilizing a binary-dye mixture (with 62 wt% DCM and 38 wt% PM597) as the active medium and an external stable resonator. The measured results show that the laser efficiency is enhanced as compared to the highest efficiency of each individual dye. Furthermore, using such an active CLC in an external stable resonator leads to a ~92X improved efficiency over the single CLC laser. In this instance, the binary-dye doped CLC simultaneously functions as laser oscillator and amplifier.

OCIS codes: (230.3720) Liquid-crystal devices; (160. 3710) Materials

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1. Introduction

One-dimensional photonic crystals are optical nanostructures with periodic multi-layer dielectric stacks establishing a distributed feedback along the material and defining allowed and forbidden photonic energy bands. Cholesteric liquid crystal (CLC) materials are special types of quarter-wave stack photonic crystals in which refractive index, in a periodic helical structure with a pitch length (p), contrary to common photonic crystals, continuously varies from the extraordinary refractive index (n_e) to the ordinary one (n_o) of the liquid crystal. Therefore, CLCs show a selective photonic band gap (PBG) into which the circularly polarized incident light with the same handedness as the cholesteric helix is reflected while the opposite handedness is transmitted. The photonic band edges (PBEs) occur at $\lambda_s = n_o\, p$ and $\lambda_l = n_e\, p$, where s and l specify the short and long wavelengths of the PBG, respectively. Since the density of photon states at the PBEs, against within the band, is very large, the group velocity approaches zero [1] and the possibility of lasing in CLCs in the presence of a proper laser dye as an active material, without any external reflectors, is considerable [2].

From the first demonstration of photonic band-edge lasing in CLCs [2] to date, enhancing the efficiency and decreasing the threshold energy of CLC lasers have become the most important scientific challenges. A high orientational order parameter for the liquid crystal as host in CLC lasers gives rise to a pronounced birefringence [3,4] establishing a low threshold energy and a high efficiency [5,6]. Laser dyes with a high order parameter of the transition dipole moment lead to an optimal performance for CLC lasers [7,8]. In addition to an extensive structural study on liquid crystals and laser dyes, a wide variety of optical techniques have been utilized to minimize the losses in CLC cavities and amplify the laser emission resulting in a low threshold and a high efficiency [9–15].

In this paper, we demonstrate two methods for significantly improving the optical efficiency of a CLC laser. Firstly, a binary dye mixture consisting of 62 wt% DCM and 38 wt% PM597 as the active medium is elicited an enhancement of the laser emission in the wavelengths between 595 nm and 613 nm, as compared to the highest efficiency of each individual dye. Secondly, using such an active CLC in an external stable resonator leads to a ~92X improved efficiency over the single CLC laser, in which the binary-dye doped CLC functions as laser oscillator and amplifier simultaneously.

2. Sample preparation

In experiments, we first measured the order parameter of two commercial dyes DCM [(4-(dicyanomethylene)-2-methyl-6-(4-dimethlyaminostryl)-4H-pyran)] and PM597 [1,3,5,7,8-pentamethyl-2,6-di-t-butylpyrromethene-difluoroborate complex] (both from Exciton), and a binary mixture consisting of 62 wt% DCM and 38 wt% PM597. We doped 1.5 wt% of each dye in a nematic liquid crystal host: BL009 (Δn = 0.281, n_e = 1.810 from Merck). In addition, to measure the laser emission efficiency at various wavelengths, nine active CLC mixtures comprising BL009, 1.5 wt% binary-dye mixture, and the left-handed chiral dopant MLC-6247 (from Merck) with different concentrations (from 25.70 wt% to 29.28 wt%) corresponding to various wavelengths were prepared. The whole mixtures were thoroughly mixed before they were capillary-filled into the homogeneous LC cells in an isotropic state (at 105°C). The thickness of all the employed LC cells was 10 μm. The inner surfaces of the glass substrates were first coated with a thin transparent conductive indium-tin-oxide (ITO) electrode and then overcoated with a thin polyimide layer. The substrates were subsequently rubbed in antiparallel directions to produce ~2-3° pretilt angle. After a slow cooling process (0.3°C/min) for active CLCs, a defect-free single-domain cholesteric planar structure was formed.

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3. Results and discussion

A crucial parameter effective in the efficiency of dye-doped CLC lasers is the order parameter of the transition dipole moment (TDM) of the dye (S_{td}) with respect to the local director ($\hat{n}$) of liquid crystal. In practice, S_{td} can be calculated by the following equation [16]:

$$S_{td} = \frac{n_o I_{//} - n_e I_{\perp}}{n_o I_{//} + n_e I_{\perp}}, \tag{1}$$

where $I_{//}$ and $I_{\perp}$ are the fluorescence intensities emitted from a dye-doped nematic film polarized parallel and perpendicular to $\hat{n}$. The maximum possible value $S_{td} = 1$ (for $I_{\perp} = 0$) corresponds to the case of perfect alignment of the TDM parallel to $\hat{n}$, while $S_{td} = 0$ implies an isotropic orientational distribution. On the other hand, the values $S_{td} < 0$ correspond to a preferred orientation perpendicular to $\hat{n}$.

Figure 1 depicts the dependence of laser emission energy on the wavelength for the binary-dye doped CLC laser. For comparison purpose, the laser energies for DCM-doped and PM597-doped CLC lasers at the maximum fluorescent wavelength of each dye are also included. In all the experimental results reported here, the active cell was pumped by a frequency-doubled, Q-switched, Nd:YAG pulsed laser (λ = 532 nm, from Continuum) with pulse duration of 4 ns, and all the measurements were performed at 1 Hz laser repetition rate in order to reduce the accumulated thermal effect originating from dye absorption. The optical efficiency of the PM597-doped CLC laser is ~1.5X higher than that of DCM. As Fig. 1 shows, the laser emission energy for the binary-dye-doped CLC between λ = 595 nm and 613 nm is even higher than that of PM597-CLC; at λ~605 nm the improvement is over 20%. The higher laser efficiency of the binary-dye doped CLC is believed to originate from better alignment of the transition dipole moment of both PM597 and DCM with the director arising from an effective mutual interaction between these two dyes in the presence of the liquid crystal host. To confirm this, the order parameter of DCM, PM597, and the binary-dye mixture through measuring the fluorescence intensities ($I_{//}$ and $I_{\perp}$) were calculated to be 0.33, 0.45, and 0.55, respectively, which, on average, implies to a better alignment of the TDMs in the binary-dye-doped CLC mixture.

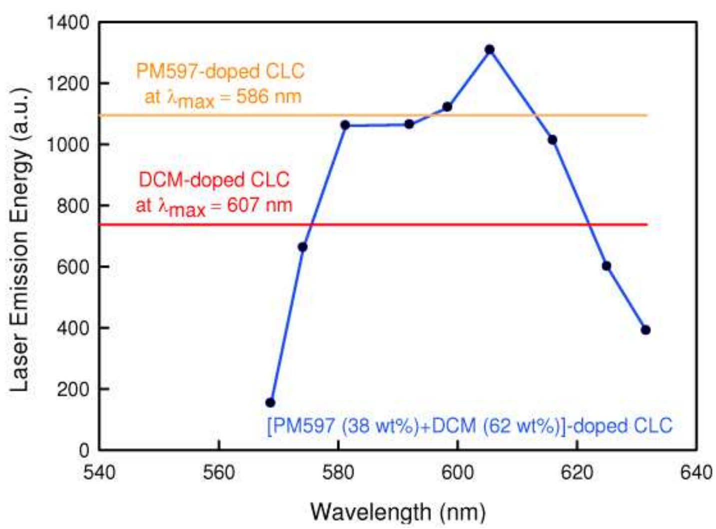

Fig. 1. The laser emission energy of binary-dye doped CLC vs. wavelength (blue line), PM597-CLC at λ = 586 nm (orange line), and DCM-CLC at λ = 607 nm (red line). Pump energy = 25 μJ/pulse for all the measurements.

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The reasons we mixed the laser dye PM597 with DCM (as a commonly employed laser dye in CLC lasers) are threefold: 1) the pump wavelength (532 nm), according to our measurement, is approximately on the absorption peak of PM597-BL009 mixture. 2) The solubility of PM597 in the liquid crystal host is very good. 3) A negligible overlap between the fluorescence spectrum of PM597-BL009 and the absorption spectrum of DCM-BL009 prevents the fluorescence resonance energy transfer, and thus emits a considerable fluorescence which will enhance the output efficiency. The binary-dye mixture consisting of 62 wt% DCM and 38 wt% PM597 was empirically obtained to be optimal for the BL009 host. Of course, this ratio could vary depending on the LC host material because of the detailed guest-host molecular interactions.

Owing to the symmetry of the helical CLC structure, laser emission from the active CLC occurs equally in both forward and backward directions parallel to the helix with a left-handed circular polarization (LCP). To achieve a defect-free planar texture in the active CLC which may die away affected by insufficient surface anchoring force in the bulk area, the cell gap should not exceed 10 μm. Moreover, for such a short cavity length the lasing efficiency is significantly restricted which gives rise to a noticeable diffraction and consequently a highly divergent laser beam [9]. So as to resolve this drawback, an external stable resonator, as schematically shown in Fig. 2, is utilized. Two similar plano-convex lenses (whose plane surfaces facing the CLC laser), in both sides of the active cell, may cause the stability of resonator. Without the lenses, there will be a large walk-off loss from the resonator, so that only waves that are accurately aligned with the resonator axis will remain within the cavity and be able to oscillate. To prevent successive reflections, all the lenses were with anti-reflection coatings. Due to the symmetry of CLC laser emission, the intervals between the lenses and active CLC cell were selected to be identical, i.e., $d_l = d_r = d$.

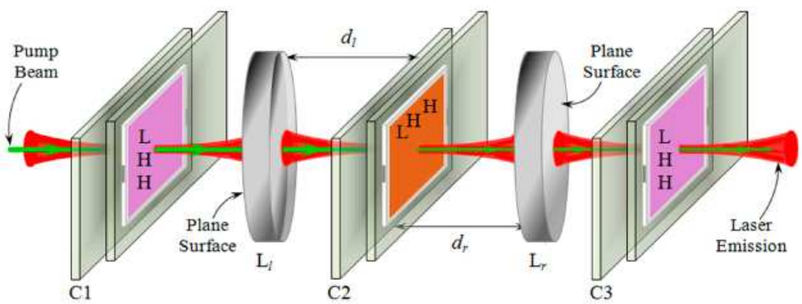

Fig. 2. A schematic diagram of the CLC laser including an external stable resonator. C1 and C3: CLC reflectors; L_l and L_r: plano-convex lenses; C2: the active CLC cell. C1 and C3 are, in practice, fixed on the plane surfaces of L_l and L_r, respectively.

Because the passive CLCs reflect circularly polarized light with the same handedness as the cholesteric helix while preserving the polarization state of light upon reflection, the two passive CLC cells (C1 and C3) with left-handed helix serve as reflectors of the external resonator. In this case, the reflection bands of both passive cells should cover the long wavelength edge of the active cell's photonic band gap where lasing takes place (Fig. 3).

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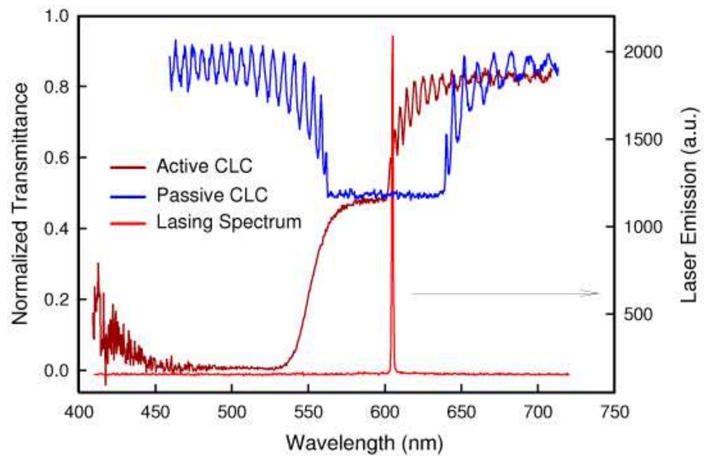

Fig. 3. The normalized transmittance of active CLC (brown line) and passive CLC (blue line). The red line shows the lasing spectrum of the active CLC cell at 10 μJ/pulse pump.

As Fig. 3 shows, the short wavelength edge of the active cell is obscured by the absorption of the binary dye. The lasing wavelength is located at λ~605 nm. The reflectivity of the passive CLC cells C1 and C3 (10-μm thick) for the LCP state light in the reflection band region was measured to be ~96.9%. Therefore, the laser emission emerges from both sides of the resonator symmetrically. In practice, in order to establish an optimal oscillation the cells C1 and C3 were fixed on the plane surface of the lenses L_l and L_r, respectively, and each of these two sets was attached to a five-axis lens positioning apparatus (Newport Co.) providing five degrees of precision positioning. Thus, the interval between each cell (C1 and C3) and its respective lens is about 1 mm (the glass substrate thickness of the cells), which may be negligible in comparison with d.

To realize the performance of external resonator, we should note that some energy is always died away from the intra-resonator laser field because of scattering, absorption, and imperfect mirror reflectivities as transmissive output coupling through both reflectors. Even if we completely neglect these loss mechanisms, there is still loss resulting from the resonator instability. A laser with an unstable resonator will have a large loss associated with the escape of radiation passing the reflectors and consequently need active media with higher gain to sustain laser oscillation. Accordingly, to stabilize the external resonator as depicted in Fig. 1, we need to specify the stability condition with the aid of *ABCD* law [17]. Since the configuration of the active CLC cell and external resonator has been pondered to be symmetric, by using the paraxial approximation and the common ray tracing method, the following condition is obtained:

$$0 \le \frac{8d^2}{f^2} - \frac{8d}{f} + \frac{3}{2} \le 1, \tag{2}$$

where f is the focal length of the lenses. Equation (2) leads to a stable oscillation provided that $d \le (2-\sqrt{3}) f/4$ or $d \ge (2+\sqrt{3}) f/4$. In our experiment, to reduce aberration we chose the lenses with $f = 30$ cm and diameter $D = 50$ mm which implies to $d \le 2$ cm or $d \ge 28$ cm. On the other hand, to fulfill the paraxial approximation, the separation of the lenses and the active cell should be as short as possible. Therefore, by considering experimental limitations we selected d ~1.6 cm.

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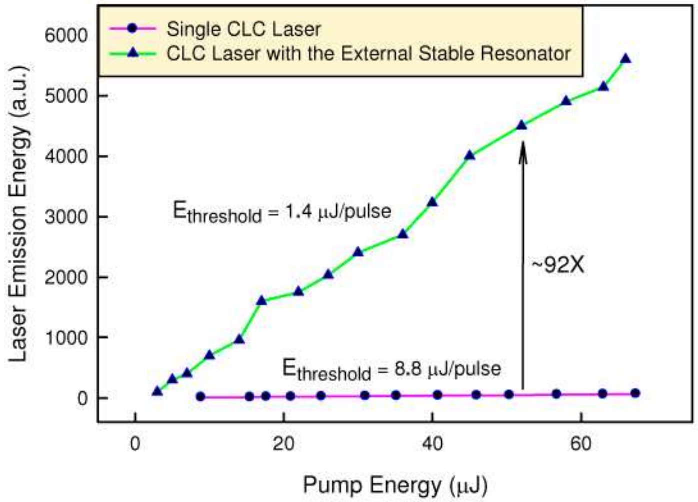

Fig. 4. The laser emission energy dependence on the pump energy of a single CLC laser (pink line) and a CLC laser with an external stable resonator (green line). In this experiment, because the laser output with an external stable resonator is significantly higher than that of the single CLC laser, in order to show this contrast an arbitrary scale which is different from Fig. 1 is used.

To study the impact of external stable resonator on the lasing efficiency, we measured the laser emission energy as a function of pump energy for a 10-μm active CLC at λ = 605 nm, with and without the resonator. Figure 4 shows the measured results. The single CLC laser creates a quite weak laser emission due to a thin emissive layer and consequently a short distributed feedback length. By inserting the active CLC into the external stable resonator, the laser emission originated from the CLC laser, under the influence of reflectors, is bounced back and forth repeatedly through the active medium (C2). As a result, since the circular polarization state of the reflected beams is preserved by the CLC reflectors, the field of each beam may coherently interact with the active CLC and thus be significantly amplified. Accordingly, while the active CLC operates as a laser oscillator individually, it functions as a laser amplifier in conjunction with the external resonator. In this case, the laser output is 92X stronger than that of the single CLC laser alone. Regarding the polarization of output, it should be noted that the existence of boundaries (substrates) at two sides of the active CLC layer establishes a refractive index mismatch which causes the Fresnel reflection as well as the CLC reflection due to the stop band effect. This gives rise to the laser output polarization not completely circular [18]. In the case of external resonator there are some extra boundaries of the passive CLCs which are expected to intensify this effect. But our measurements reflect that the ellipticity of laser's polarization for the single CLC laser (~0.93) and the active CLC with the external stable resonator (~0.91) are markedly identical, and thus the output polarization state is still (to a large extent) left-handed circular, the same as its original polarization state. Moreover, the threshold energy is decreased from 8.8 μJ/pulse for a single CLC laser to 1.4 μJ/pulse. The spot size of the pump beam (w) was regulated at ~185 μm. Our experiment indicates that the amplification factor is noticeably sensitive to the spot size of the pump beam. If the spot size is smaller than ~75 μm, no amplification was observed.

4. Conclusion

We have demonstrated an approach to significantly improve the lasing efficiency of a CLC band-edge laser. This is attained through exploiting a particular binary dye as the active material and an external stable resonator. A binary-dye mixture consisting of 62 wt% DCM and 38 wt% PM597 improves the lasing efficiency up to 20% (at λ~605 nm) for the

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wavelengths between 595 nm and 613 nm, as compared to the highest efficiency of each individual dye. With an external stable resonator the laser efficiency is further improved by ~92X over the single CLC laser. In this instance, the active CLC behaves as laser oscillator and laser amplifier simultaneously. As a result, the threshold energy is reduced from 8.8 μJ/pulse for the single CLC laser to 1.4 μJ/pulse. In addition, the experiment indicates that the amplification factor is noticeably sensitive to the spot size of the pump beam, so that if the spot size is smaller than ~75 μm no amplification would occur.

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