Circularly polarized high-efficiency cholesteric liquid crystal lasers with a tunable nematic phase retarder

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Abstract: A high efficiency dye-doped cholesteric liquid crystal (CLC) is demonstrated by optimizing the dye concentration and using an electrically tunable nematic liquid crystal (NLC) phase retarder. The state of polarization of laser emission in CLC lasers, contrary to our expectations, due to the refractive index mismatch at the boundaries is not exactly circular. A double-cell structure including a CLC laser and an adjustable NLC phase retarder with a mirror reflector on one of the inner surfaces not only purifies the polarization state of the laser output but also improves the laser efficiency by 6.7X, over the single-direction dye-doped CLC laser.

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

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1. Introduction

Cholesteric liquid crystal (CLC) materials are special types of quarter-wave stack reflectors in which refractive index, in a periodic helical structure with a pitch length (p), continuously varies from n_e to n_o which are extraordinary and ordinary refractive indices of the liquid crystal, respectively. As a result of this periodic structure, CLCs show a selective reflection band. Within the band, 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 $\lambda_l = n_e\, p$. The unique properties of this one-dimensional self-organized photonic band gap structure as distributed Bragg reflector has stimulated a wide variety of applications [1–4]. The first unambiguous demonstrations of lasing in dye-doped CLCs were carried out by Kopp and Genack et al. [5]. In this case, the density of photon states at the PBEs, against within the band, is very large so that the group velocity approaches zero, and the possibility for lasing is considerable.

To make CLC lasers practically useful, the laser threshold energy (E_{th}) should be further reduced and the slope efficiency (η) enhanced. To achieve this objective, extensive researches have been conducted [6–12]. The improved LC structure towards enhancing the orientational order parameter establishes a significant birefringence, and consequently, a reduction in E_{th} and an increase in η [6]. A laser dye with a high order parameter of the transition dipole moment leads to an optimal performance for CLC lasers [7,8]. Moreover, a polymerized CLC restricts thermal and rotational motion of the dye molecules, thus reducing non-radiative emissions within the system and decreasing the laser threshold energy [9,10]. In addition to abovementioned studies, a wide range of useful optical designs for minimizing the losses in CLC cavities have been performed resulting in more efficient PBE CLC lasers [11,12].

In this paper, we demonstrate a high efficiency CLC laser by controlling the polarization state of laser emission. Due to the refractive index mismatch at the boundaries of the CLC and substrate interfaces, and thus, the Fresnel reflection as well as reflection band effect, the state of polarization (SOP) of laser emission is no longer circular. By using an adjustable retarder whose retardation and angle of the fast axis with respect to the lab frame of reference can be regulated, we can improve the SOP of the laser emission which in turn leads to a 6.7X increased slope efficiency over the single-direction CLC laser.

2. Sample preparation

In order to determine the optimum dye concentration, we made a CLC mixture having a long wavelength edge identical to the maximum fluorescence of the laser dye DCM (4-(dicyanomethylene)-2-methyl-6-(4-dimethlyaminostryl)-4H-pran). The mixture is composed of high birefringence nematic liquid crystal BL009 (Δn = 0.281, n_e = 1.810 from Merck) and 26.6 wt% of right-handed chiral agent MLC-6248 (helical twisting power = 11.3 $(\mu\text{m wt\%})^{-1}$ from Merck). This mixture was divided into nine parts, and then, the laser dye DCM (from Exciton) was doped into them with the concentrations: 0.1, 0.2, 0.5, 1.0, 1.5, 2.1, 2.7, 3.0, and 3.3 wt%. The whole mixtures were thoroughly mixed before they were capillary-filled into the homogeneous LC cells in an isotropic state (at 105°C). The cell gap was controlled at 10 μm. This is because in a short-pitch CLC layer, the strong chiral turn competes with the surface anchoring force so that multi-domain cholesteric defects cannot be completely eliminated, especially for the cell gap larger than 10 μm. Hence, light scattering is introduced which increases the loss in the laser cavity. 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), a defect-free single-domain cholesteric planar structure was formed. On the other hand, a retarder cell (10-μm thick) was capillary-filled with BL009 (Merck); one of the inner surfaces was deposited with a thin aluminum reflector, instead of ITO.

3. Results and discussion

To achieve a high efficiency CLC laser, two parameters were carefully controlled: dye concentration and refractive index mismatch at the boundaries. Figure 1 depicts the measured

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results of dye concentration effect on laser threshold and slope efficiency for single-direction CLC lasers.

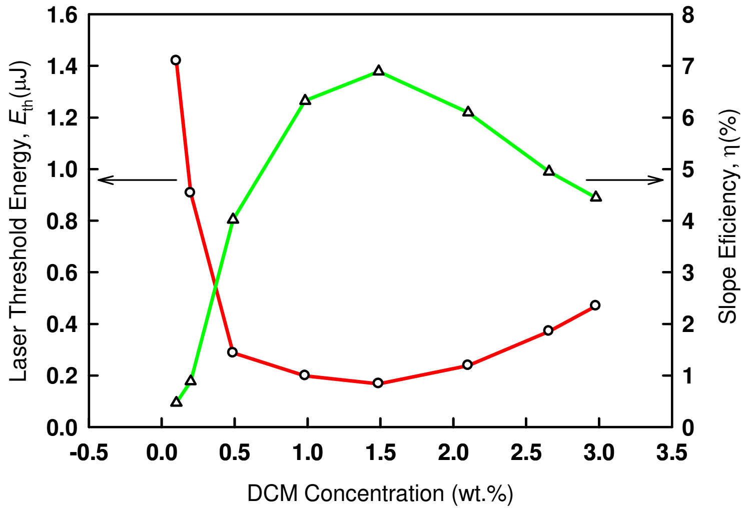
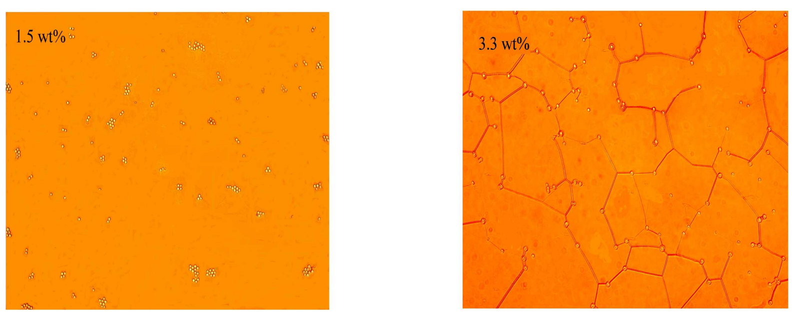


Fig. 1. The laser threshold energy (red) and the slope efficiency (green) as functions of the dye concentration.

From Fig. 1, as the DCM concentration increases the laser threshold decreases sharply, saturates, and then gradually climbs up. The optimal dye concentration is ~1.5 wt%. A low dye concentration is equivalent to a small absorption cross-section (σ_{abs}), and thus, low excitation. Consequently, more pumping energy is needed in order to exceed lasing threshold. On the other hand, if the dye concentration is too high, molecular aggregation occurs, which results in increased light scattering, higher cavity loss, and finally higher threshold energy. At 3.3 wt% dye concentration, no lasing effect was observed.



(a) (b)

Fig. 2. The planar textures of dye-doped CLC with (a) 1.5 wt%, and (b) 3.3 wt% concentrations.

Figure 2 shows a comparison between the planar textures of dye-doped CLC with (a) 1.5 wt% and (b) 3.3 wt% concentrations observed from a polarized optical microscope (Olympus BX51). The defect lines in the sample with 3.3 wt% dye clearly indicate high aggregation of dye molecules in the CLC host. On the other hand, the slope efficiency is entirely opposite to the laser threshold energy; at minimum E_{th}, η reaches maximum. This is in good agreement with laser theory because a low E_{th} corresponds to a large σ_{abs}, and thus, a large stimulated-emission cross section. As a result, η is enhanced. The slope efficiency, from the rate equations, can be shown to be proportional to $1/E_{th}$.

The existence of boundaries (substrates) at two sides of the CLC layer establishes a refractive index mismatch which causes the Fresnel reflection as well as the CLC reflection due to the stop band effect. These two types of reflections are fundamentally different for two reasons. First, the reflectance of Fresnel reflection is $R = (n_1 - n_2)^2/(n_1 + n_2)^2$ at normal

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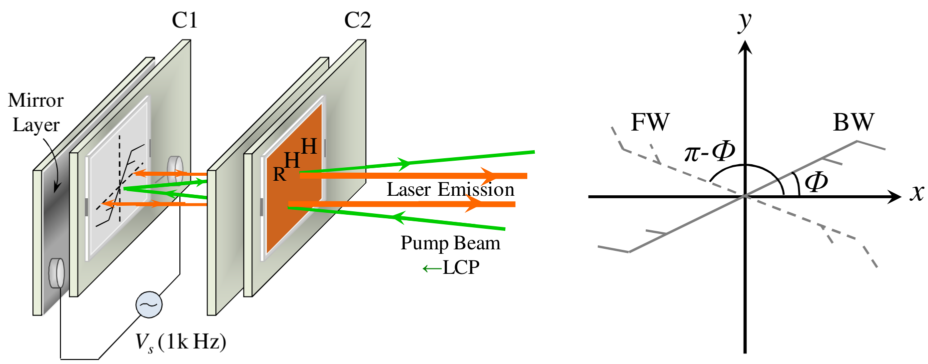
incidence, where n_1 and n_2 are the refractive indices at the boundary, whereas the reflectance of CLC reflection equals to one in the bandgap for one normal mode, due to Bragg effect caused by the interference between successive reflected beams in the photonic structure's interfaces of the cholesteric liquid crystal [13]. Another difference is upon CLC reflection, the handedness of circularly polarized light does not change, while it changes to opposite handedness of rotation upon Fresnel reflection, if the incidence is from low-to-high index medium. This is why the polarization state of CLC lasers, by contrary, is not exactly circular. Although, the effect of cell boundaries can theoretically be removed by matching the refractive index of the substrates to $\bar{n} = \sqrt{(n_e^2 + n_o^2)/2}$ of the CLC, because of a wide variety of CLCs with different ordinary and extraordinary refractive indices, it is not practically feasible.



(a) (b)

Fig. 3. (a) Schematic illustration of the performance of the NLC retarder (C1) along with the CLC laser (C2), and (b) the coordinates system of the reference frame of lab and the state of fast axis of the retarder for the forward (FW) and backward (BW) beams.

To resolve this problem and also enhance the efficiency of the CLC laser, we used an electrically tunable NLC retarder. Figure 3 depicts a CLC laser with right-handed helix along with an NLC retarder whose fast axis is rotated by Φ from the *x*-axis of *x*-*y* coordinates and one of the inner surfaces acts as a mirror reflector. If the electric field of the optical wave is defined as $\vec{E} = \vec{E}_0 \exp[i(kz - \omega t)]$, the Jones vector for a wave with an elliptical polarization is $\frac{1}{2}\begin{bmatrix} 1 \\ e_0 \exp(i\Delta) \end{bmatrix}$ where e_0 and Δ are the ellipticity and the phase difference between the components of $\vec{E}$ in the directions of x and y (Fig. 3(b)), respectively. The $e_0 = 1$ for $\Delta = \pm\pi/2$ corresponds to a left-handed (+ π) or right-handed (-π) circular polarization. The effect of the retarder on the polarization of a beam propagating in the direction of *z*-axis can be obtained by using the following Jones matrix [14]:

$$Q(\Phi) = \begin{bmatrix} \cos\left(\frac{\delta}{2}\right) - i\sin\left(\frac{\delta}{2}\right)\cos(2\Phi) & -i\sin\left(\frac{\delta}{2}\right)\sin(2\Phi) \\ -i\sin\left(\frac{\delta}{2}\right)\sin(2\Phi) & \cos\left(\frac{\delta}{2}\right) + i\sin\left(\frac{\delta}{2}\right)\cos(2\Phi) \end{bmatrix}, \quad (1)$$

where δ is the phase retardation of the NLC retarder. For a positive dielectric anisotropy NLC, the phase retardation can be expressed as [15]:

$$\delta = \frac{2\pi d}{\lambda} \Delta n(V_s) \quad (2)$$

In Eq. (2), $\Delta n(V_s)$ is the voltage-dependent birefringence, λ is the wavelength, *d* is the cell gap of the retarder, and V_s is the applied voltage to the retarder. To adjust δ, the voltage V_s

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must be higher than the Freedericksz transition threshold. Consequently, the effective Jones matrix for the laser emission and also the pump beam propagating in the direction –z, reflecting in the mirror, and then coming back in the direction + z, can be expressed as:

$$Q_{eff.}(\Phi,V_s) = Q(\Phi,V_s)\begin{pmatrix}-1 & 0\\ 0 & 1\end{pmatrix}Q(\pi-\Phi,V_s), \quad (3)$$

where the Jones matrix $\begin{pmatrix}-1 & 0\\ 0 & 1\end{pmatrix}$, $Q(\pi-\Phi,V_s)$, and $Q(\Phi,V_s)$ show the effects of the reflector on the input beam and the retarder on the forward (-z) and backward (+ z) beams, respectively (Fig. 3(b)). If the polarization of laser emission is a pure RCP, upon reflection from the mirror it will be converted to LCP. The retarder could compensate this phase change by the voltage alone. But for our complicated CLC system, both δ and Φ should be adjusted for two reasons. Firstly, as mentioned above, the CLC laser polarization is not completely RCP. And secondly, this system is modifying not only the CLC laser polarization but also the pump beam polarization so that the laser output could be maximized.

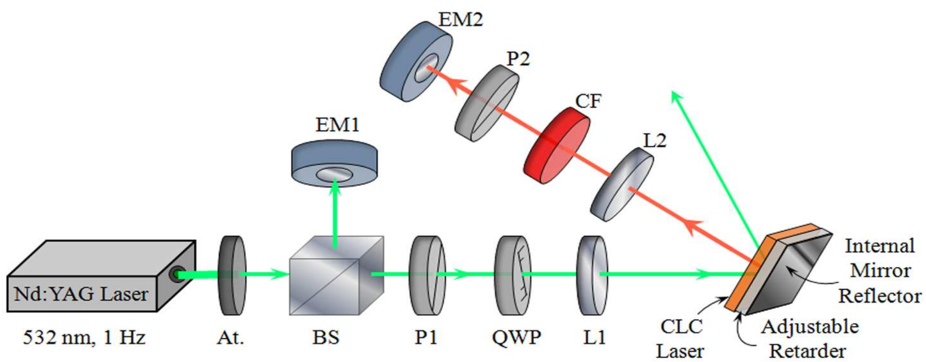

Fig. 4. Experimental setup: At: attenuator; BS: beam splitter; P1: polarizer; QWP: quarter-wave plate; L1 and L2: lenses; CF: color filter; P2: rotatable polarizing for measuring the ellipticity; EM1 and EM2: energy meters.

At first, by rotating a polarizer, the maximum (E_{Max}) and minimum (E_{min}) energy of the single-direction CLC laser (with 1.5 wt% DCM) were measured and the ellipticity of the polarization state was calculated as $e_0 = (E_{min} / E_{Max})^{1/2} = 0.93$. This measurement showed that SOP of the CLC laser is not exactly circular. In order to examine the effect of the adjustable retarder on the CLC laser performance, we used a reflective configuration at 30° oblique incidence according to Fig. 4. The pump source is a frequency-doubled, Q-switched, Nd:YAG pulsed laser (λ = 532 nm, from Continuum) with pulse duration of 4 ns. All the measurements were performed at 1 Hz laser repetition rate in order to reduce the accumulated thermal effect originating from dye absorption. A linear polarizer and a quarter-wave plate were used to create a left-handed circular polarization (LCP) to avoid the reflection from the stop band of the CLC film. The slope efficiency and threshold energy are both dependent on the spot size of pump beam [16], so that by decreasing the spot size the former increases and the latter decreases. But, based on our experiment, the smaller the pump beam spot size is, the faster the laser energy is saturated, which is in good agreement with laser theory. Therefore, in the experiment related to Fig. 5 in order for more exact comparison between the slope efficiency of the three kinds of laser in greater range of the excitation energy (50 µJ), we chose the pump spot size to be 170 µm, while to measure E_{th} and η versus the dye concentration as depicted in Fig. 1, this restriction is not determinative. In other words, by changing the spot size the amount of optimal dye concentration will not change; and consequently, so as to increase the measurement accuracy the spot size was chosen to be 65 µm. The output laser emission from the sample was collected by a lens to an energy meter (PD10, Ophir). The rotatable polarizer P2 is only applied when measuring the ellipticity of the output laser.

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To adjust the retarder, it was set on a rotatable mount with sensitivity 1° and completely connected to one of the faces of the CLC laser. Then by regulating Φ and V_s (at 1 kHz frequency), maximum laser emission was obtained. It should be mentioned that the selections of Φ and V_s are not unique. In this experiment, we set V_s = 1.1 V_{rms} (the threshold voltage of the NLC retarder is ~1V_{rms}) and Φ = 34°. To demonstrate how the adjustable retarder improves the lasing efficiency, as shown in Fig. 5, we measured the laser emission energy as a function of pump energy for the single-direction CLC laser, the CLC laser with an external mirror reflector, and the CLC laser with the adjustable retarder having a reflector on one of the inner surfaces as illustrated in Fig. 3(a). Moreover, the threshold energy was measured to be 5.6, 3.5, and 1.3 μJ/pulse, respectively. The laser wavelength (λ = 604 nm) was measured by an Ocean Optics spectrometer (HR2000, resolution = 0.4 nm).

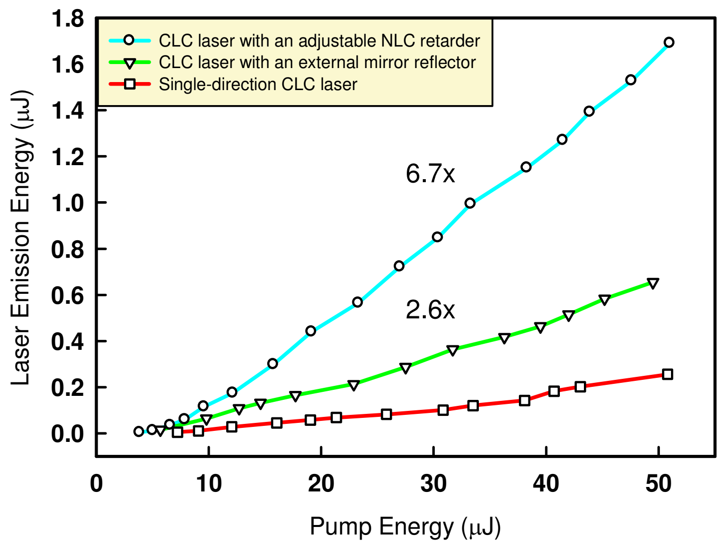

Fig. 5. Laser emission energy as a function of pump energy for the single-direction CLC laser (red line), the CLC laser with an external mirror reflector (green line), and the CLC laser with the adjustable retarder (blue line).

From Fig. 5, the slope efficiency of the double-cell structure including the CLC laser and the adjustable retarder shows a considerable enhancement in the laser emission, on average, 6.7X higher than that of single-direction CLC laser. To understand the reason, first we consider a CLC laser with an external mirror reflector. Owing to the symmetry of the helical CLC structure, laser emission (without considering the reflector) occurs equally in both forward and backward directions parallel to the helix with a nearly right-handed circular polarization (RCP). The polarization of the backward emission, after reflecting in the mirror reflector due to π phase change is converted to a state close to LCP. Because of the existence of glass substrate ($t \approx 1.1\,\text{mm}$ thick) as an interface, there is no coherent relationship between these laser emissions. On the other hand, the pump beam polarization (LCP), due to the effect of mirror, is converted to RCP, so the backward pump beam no longer pumps the active CLC at 30° angle [17]. Accordingly, in this instance, it is expected the output laser energy is twice of the single-direction laser, but the experiment shows ~2.6X (Fig. 5). Of course this difference may be caused by the effect of multiple surface reflections from the glass substrates. Regarding the CLC laser with the adjusted retarder, we believe two effects are established: double pumping of the active CLC and the coherent interaction of the backward laser emission with the excited dye molecules in the active CLC.

As seen from Fig. 3(a), the pump beam at 30° relative to the cell normal crosses the active materials at two points by distance $l \approx 2.5$ mm ($l = 4t\tan(\pi/6)$). Since the pump spot size ($D = \pi\omega_o$) was considered to be 170 μm, the Rayleigh range of the beam ($Z_R = \pi\omega_o^2/\lambda$) is about 17 mm (where ω_o and λ are the beam waist and pumping beam wavelength, respectively). This means that the pump spot size is approximately constant in the crossing points with the active

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material. The incoming LCP pump beam can excite the active material (at the input point), while the outgoing beam may only stimulate it to some extent, because after passing the retarder and upon reflection from the mirror its polarization will change. By adjusting the retarder at a proper retardation and Φ the polarization of the outgoing beam may be preserved resulting in maximal excitation in the output point of the active material. These two stimulated points of active material emit laser lights in two directions normal to the active cell's surface, and the backward laser emissions' polarization (with RCP states) under the influence of the adjusted retarder may be quite well preserved. Consequently, the adjusted retarder acts as a polarization modifier for both pump beam and backward laser emissions, simultaneously. In other words, the optimal retardation and Φ of the adjusted retarder are the amounts in which the SOP for both the backward pump beam and laser emissions, to a large extent, preserves. As a result of double pumping of active CLC irrespective of the other effects, a fourfold laser emission is expected. But, a backward laser emission with the same polarization as created in lasing can coherently interact with the active CLC and cause an enhanced gain and consequently an amplified output laser. This is why the slope efficiency is increased by 6.7X. In this case the ellipticity was measured to be 0.96 showing that an improved polarization state in addition to high efficiency emission.

4. Conclusion

We have presented a method to establish a circularly polarized high-efficiency laser emission from a cholesteric liquid crystal band-edge laser. This is achieved by selecting the optimal dye concentration and using an electrically adjustable NLC retarder which can rotate with respect to the CLC laser. By adjusting the phase retardation and angle of the fast axis of the retarder, the polarization state of the backward pump beam and laser emission improves. This, in turn, causes a double CLC pumping as well as an increased gain in the active CLC resulting in a high efficiency laser output. The measurement indicates that the slope efficiency is increased by 6.7X as much of single-direction laser. Moreover, the ellipticity of the laser emission is also improved from 0.93 to 0.96.

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