Enhanced laser emission in opposite handedness using a cholesteric polymer film stack

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Abstract: We demonstrate an enhanced circularly polarized laser emission whose handedness is opposite to the cholesteric helix by the stacked cholesteric polymeric films. The dye-doped right-handed cholesteric polymer film is sandwiched between a mirror and a cholesteric polymer reflector. Due to the stimulated amplification and light recycling effects, the original laser emission from the middle active layer is not only dramatically enhanced but also converted to a left-handed circularly polarized emission with high purity. Moreover, a single 15 μm dye-doped cholesteric film is found to lase more efficiently when the top side faces the pump source than when the bottom side does. This phenomenon is attributed to the band gap broadening of the bottom side.

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

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1. Introduction

A cholesteric liquid crystal (CLC) layer can selectively reflect the circularly polarized light in the same handedness as its helix within a certain bandwidth from n_oP to n_eP, where n_o and n_e represent the ordinary and extraordinary refractive index of the LC and P is the intrinsic pitch length of the cholesteric phase [1]. Due to the periodic structure, CLC is regarded as a self-organized one-dimensional photonic crystal. Compared to the dielectric photonic crystals with a strong periodic index modulation, CLCs can be easily fabricated with a wide reflection band. As proposed by Dowling et al [2], at the edge of the reflection band the photons are strongly localized and density of states is significantly enhanced. Therefore, CLC is a good candidate to serve as a resonator based on distributed feedback (DFB). In the past few years, low threshold lasers, micro-cavity lasers, and sensors have been demonstrated using cholesteric medium [3-6]. These devices are mirrorless, small, and tunable.

Recently, the tendency towards miniaturization and portability in the emerging CLC-based mirrorless lasers brings forward new research interests in cholesteric polymer lasers. Many efforts have been focused on the lasing action in dye-doped cholesteric free standing polymer films [7, 8], spatially tunable lasing films [9], and cholesteric elastomer films [10]. Meanwhile, CLC polymer films have another important application as the end reflecting layers in the laser cavity [11, 12] or to create the extra defect laser modes [13]. Currently, there are several factors that limit the output power from the CLC polymer lasers, such as dye-bleaching during the UV polymerization process and deformation of the ordered cholesteric structure due to insufficient surface anchoring energy. In order to increase the output power and, in the meantime, lower the threshold, different approaches have been proposed, such as adding external reflectors [11-12, 14-16] or stacking multiple active polymer films [17]. Adding a cholesteric passive reflector helps to enhance the laser output based on a further coherent amplification [15] while the enhancement from a metallic mirror essentially results from the second pump of the active layer [16].

In this paper, we combine these two effects together for enhancing the output from a CLC polymer film laser by sandwiching the dye-doped CLC polymer film between a mirror and a CLC reflecting film. Unlike the single reflecting layer enhancement approaches, such a structure creates an incomplete resonant cavity because the metal layer and the cholesteric reflecting layer select a different polarization for reflection. Hence the output power is dramatically improved. More importantly, the output beam is circularly polarized, but opposite to the handedness of the cholesteric helix. Based on light recycling effect, the original impure circularly polarized laser (from a single active CLC polymer film) can be recycled and maximally purified by the two reflective layers. The responsible physical mechanisms are discussed in detail. In addition, the different lasing performance from different sides of the dye-doped CLC polymer film is investigated and the underlying mechanism is analyzed. This work cannot be realized using CLC cell stack because of indirect contact between each layer. Such an approach not only improves the output power but also opens a way for polarization manipulation and control in CLC polymer- based laser devices.

2. Experiment setup and sample preparation

Figure 1 shows the experimental setup at 20^o oblique incidence. A frequency-doubled, Q-switched Nd:YAG pulsed laser was used as the pumping source. The laser was operated at λ=532 nm, 4 ns pulse width, and 1 Hz repetition rate in order to reduce the accumulated thermal effect in the sample. The pump beam was split into two paths: one was monitored by an energy meter (LaserStar, Ophir) and the other linearly polarized pump beam was converted

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into a left-handed circularly polarized beam in order to reduce the band gap reflection from the cholesteric polymer film at the pumping wavelength. The pump beam was then focused by a lens (with a focal length 15 cm) onto the lasing sample with a spot diameter of ~160 μm. The generated laser emission was collected by another lens into a fiber-based spectrometer (HR4000, Ocean Optics, resolution = 0.8 nm).

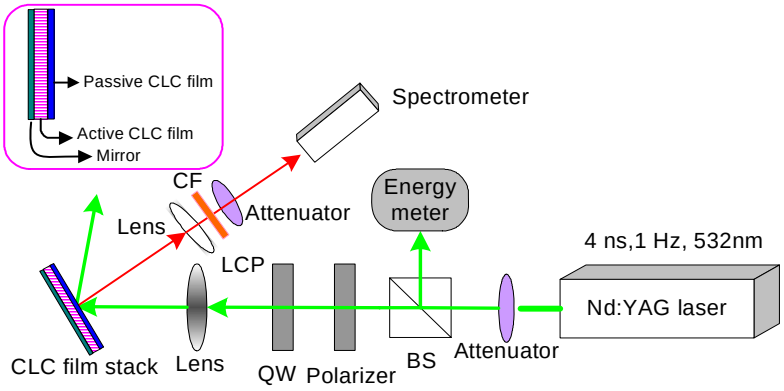

Fig. 1. Experimental setup: BS: Beam splitter; QW: Quarter-wave plate; CF: Color filter; LCP: left-handed circularly polarized light.

The mixture was prepared by mixing 44% of a chiral reactive mesogen RMM154, 44% of a reactive mesogen RM82 and 12% of a right-handed chiral agent CB15 (all from Merck). To make the active CLC lasing film, a laser dye DCM (4-(Dicyanomethylene)-2-methyl-6-(4-dimethlyaminostyryl)-4H-pran) (from Exciton) was doped into the mixture at 1.7 wt% concentration. For the passive reflective film the CB15 concentration was slightly lowered in order to shift the reflection band to cover the lasing wavelength. After the mixture was thoroughly stirred for several hours it was filled into an empty cell in an isotropic state through capillary action. The inner surfaces of the cell were first coated with indium-tin-oxide (ITO) electrodes and then polyimide alignment layers. In reality, these ITO layers were not needed because no voltage was applied to the cell. Both substrates were rubbed in anti-parallel directions to produce a small (~3^{o}) pretilt angle. The sample was slowly cooled down to ~55 ^{o}C so that less defect was formed in its cholesteric planar structure. Subsequently the sample was exposed under a UV light for ~45 min while keeping the temperature at 55 ^{o}C. The upper surface of the polymer film, which is facing the UV lamp during exposure, is called *top side* in this paper. In contrast the lower surface of the polymer film, which is further away from the UV lamp but closer to the heating stage, is called *bottom side*. We differentiate these two sides because they exhibit a dramatic difference in the lasing performance when different sides face the pumping laser.

The thickness of the active CLC polymer film is 15 μm and the passive CLC polymer film is 8 μm. To fabricate the stacked CLC polymer laser, we first peeled off the 15 μm dye-doped CLC film from the substrates and put it in optical contact on a substrate which was coated with a thin aluminum layer. Afterwards the 8 μm CLC film was peeled off from the substrates and attached in optical contact with the 15 μm film. Any air gap between the polymer films will deteriorate the lasing power. In order to achieve a stronger laser output, when assembling the film stack, the top side of the active CLC film should face the passive CLC film and the pump beam because of the different lasing capabilities between the top side and the bottom side. The device structure is illustrated in the upper-left corner of Fig. 1.

3. Experimental results and discussions

First, we characterized the reflection band of both active and passive films. We measured the reflection band using a white light source (DH-2000, Instec, UV-VIS-NIR) and a spectrometer (the same one shown in Fig. 1) at normal incidence. Results are shown in Fig. 2,

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where the top side reflection bands of the 15 μm active film and the 8 μm passive film are shown in red and blue curves, respectively. Lasing spectrum centered at λ=613 nm with 53 μJ/pulse pumping energy is also included in the figure as the green curve shows. From Fig. 2, laser action takes place at the long wavelength band edge of the active film and, in the meantime, within the reflection band of the passive film. The thicker film (15 μm) has a lower reflectivity because multi-domains and defect lines are formed during fabrication due to insufficient surface anchoring energy and lower order parameter of the LC polymer material.

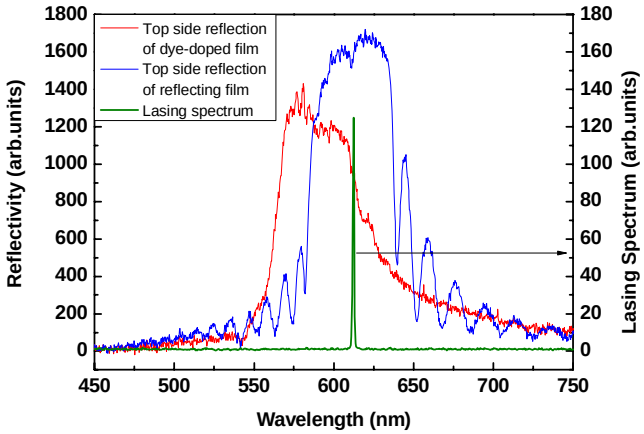

Fig. 2. Top side reflection bands of the 15 μm active film (red) and the 8 μm passive film (blue), and the lasing spectrum at 53 μJ/pulse pump (green).

Next, we measured the pump energy dependent lasing output power from our stacked CLC polymer laser, which consists of a mirror, an active CLC film (15 μm) and a passive CLC film (8 μm). Results are plotted in Fig. 3.

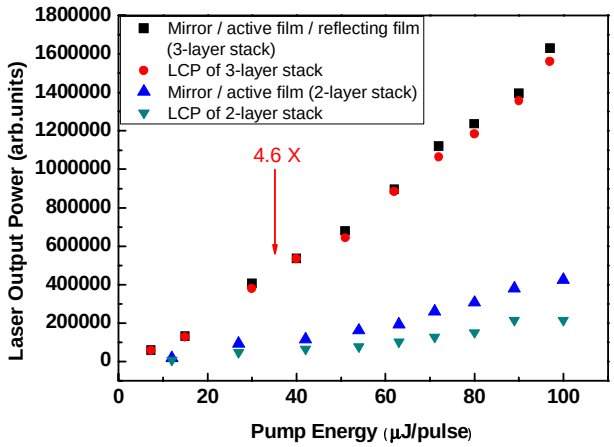

Fig. 3. The pump energy dependent laser output power of the total emission and LCP component from a {mirror/active CLC film/reflecting CLC film} laser and a {mirror/active CLC film} laser.

In Fig. 3, the total emission and the corresponding left-handed circularly polarized (LCP) component of this 3-layer CLC laser (mirror/active CLC film/passive CLC film) are shown in

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black and red, respectively. The LCP component occupies ~96% of the total emission. We then removed the passive CLC film from this film stack and measured the laser emission again at the same pumping spot. For this 2-layer CLC laser (mirror/active CLC film), the total emission and its LCP component are shown in blue and green, respectively. The LCP component occupies only ~52% of the total emission. Moreover, after removing the passive film the laser emission is ~4.6X weaker than that of the 3-layer CLC laser. The lasing threshold was measured to be 0.46 and 3.0 μJ/pulse for the 3-layer stacked CLC polymer laser and the 2-layer mirror reflected CLC polymer laser, respectively.

The results shown in Fig. 3 indicate that when the active layer is sandwiched between a mirror and a passive CLC reflector, we can obtain a dramatically enhanced LCP output whose polarization is opposite to the handedness of the cholesteric helix. This phenomenon is attributed to the further stimulated amplification of the reflected laser beam and light recycling effect. Intrinsically the single right-handed CLC film generates circularly polarized laser light in the same handedness as its helix. Hence for our 15 μm dye-doped CLC film, the laser emission is mostly right-handed circularly polarized (RCP). Here the lasing wavelength is designed to be within the reflection band of the passive CLC reflector. Therefore, when the laser light is reflected back by the passive CLC reflector, its original polarization state maintains, i.e. RCP in our case. Such RCP emission at the band edge wavelength will get further stimulated amplification when passing through the active CLC layer due to its polarization-dependent DFB. On the other hand, the mirror reflection converts RCP into LCP which is highly transmitted by the passive CLC film and, in turn, contributes to the final enhanced LCP output. As a consequence, the originally generated laser light successively experiences the amplification process each time the RCP part propagates through the active layer, and then gets converted to LCP by the mirror. As this process continues, the output laser light is gradually enhanced and purified into an LCP.

Taking the surface reflection between the air and polymer films (estimated to be ~4%) into consideration, the measured 96% LCP output represents a highly purified polarized emission, almost reaching its ideal case of polarization recycling. The CLC polymer laser with mirror reflection only generates a linearly polarized emission so that ~52% of LCP component can be detected within the total output, as a result of coherent superposition of the original RCP and the reflected LCP.

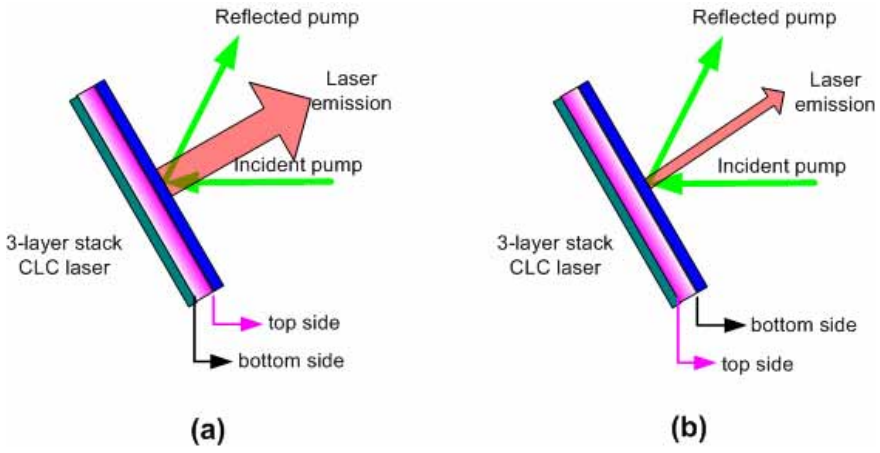

Fig. 4. Sample layout in the experimental setup: (a) top side of the active film is facing the pump beam and (b) bottom side of the active film is facing the pump beam. The 3-layer stack CLC laser: mirror (dark green) / active CLC film (magenta) / passive CLC film (blue).

Figures 4(a) and 4(b) show the sample layout in the experiment, where the top side (dark magenta) and the bottom side (light magenta) of the active film faces towards the pumping beam, respectively. During the assembly of the 3-layer CLC polymer laser, we found that if the top side of the 15-μm active film was directly attached to the passive film (i.e., the top side

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is facing the pump source), the laser output was much stronger than when the bottom side was directly attached to the passive film. This is caused by the drastically different lasing performance of a single 15-μm active film between the top and bottom sides facing the pump.

In a similar layout as shown in Figs. 4(a) and (b) except without the mirror and the CLC reflecting film, the laser output was measured and results are shown in Fig. 5. The units on the ordinate scale are intentionally kept the same as that used in Fig. 3 to allow an easy comparison.

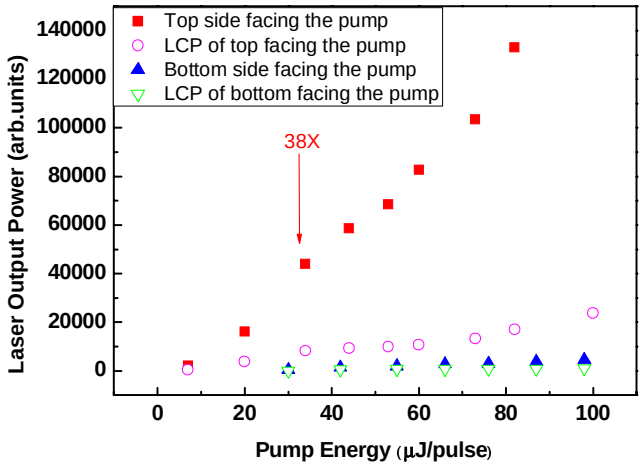

Fig. 5. The pump energy dependent laser output power from a single 15 μm active CLC film with the top side facing the pump source and the bottom side facing the pump source.

When the top side is facing the pump source (Fig. 4(a) layout with the active film only), the total emission and LCP component are shown in red and pink, respectively. Correspondingly, when the bottom side is facing the pump (Fig. 4(b) layout with the active film only), the total emission and LCP component are shown in blue and green, respectively. Therefore the emission in the case of the top side facing the pump is, on average, ~38X stronger than that of the bottom side facing the pump. In addition, no matter whether the top or the bottom side is placed towards the pump, laser light is generated from both directions at almost equal power. The corresponding lasing thresholds for these two cases are 5.65 and 20 μJ/pulse. By measuring the LCP component, we found that there was ~13% LCP with the top side facing the pump and ~24% LCP with the bottom side facing the pump. The minority LCP component originates from the defects in the planar structure, which not only scatter the light but also deteriorate the output power and the purity of the polarizations.

Comparing the output power when the top side facing the pump (red squares) in Fig. 5 and that from a 2-layer mirror-reflected CLC polymer laser (blue triangles) in Fig. 3, we notice that the former one is ~2.3X stronger than the latter one. Because of the direct contact of the aluminum mirror and the active CLC film, the mirror will reflect back both the laser emission generated towards it and the pump beam. Under this circumstance, the active layer can be pumped once again however the bottom side of the active film generates a very weak laser emission when being pumped. As the result, the enhancement contribution due to re-pump of the active layer is unnoticeable.

In terms of the LCP component percentage, we can see that the 3-layer CLC polymer laser device performs a dramatic improvement in purifying the output polarization state from 87% of RCP output (13% of LCP as measured in Fig. 5) to 96% of LCP output, which is the maximum it can reach.

In order to understand why the top side and bottom side exhibit such a dramatic difference

#77832 - $15.00 USD Received 5 December 2006; revised 25 February 2007; accepted 28 February 2007

in the laser generation when they are pumped, we further investigate the reflection band of a CLC polymer film from both sides. Results are shown in Figs. 6(a) and 6(b).

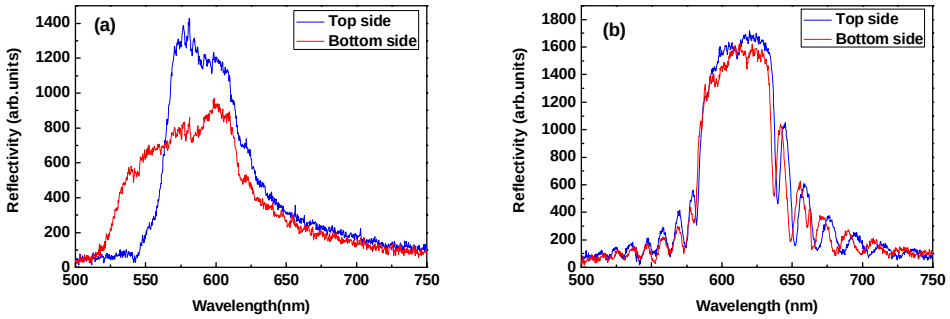

Fig. 6. The top side (blue) and bottom side (red) reflection bands of the CLC polymer film: (a) the 15 μm dye-doped CLC polymer film (b) the 8 μm passive CLC polymer film.

Figure 6(a) describes the top and bottom side reflection bands of the 15-μm dye-doped CLC polymer film and 6(b) describes those of the 8 μm passive CLC polymer film. For the 15 μm dye-doped CLC film, after curing process the top side reflection band still maintains a relatively good shape and high reflectivity with a bandwidth of ~50 nm. However the bottom side reflection band shows an obviously broadened bandwidth to ~100 nm. Currently we believe this bandwidth broadening could be attributed to the gradient pitch formed inside the cholesteric structure from the top surface to the bottom surface. Because the employed laser dye has a strong absorption for the UV light, during exposure an UV intensity gradient would be formed through the top surface to the bottom of the film. Near the top side the UV intensity is stronger and thus the polymerization reaction is faster. In the meantime, a diffusion of the chiral dopant CB15 is expected so that the top and bottom areas have different chiral concentrations and thus different pitches, leading to the observed bandwidth broadening. In addition, the ordered structures will be somewhat deteriorated during the monomer polymerization so that more defects are observed in the film after UV exposure. These defects will, in turn, lead to the decreased reflectivity within the reflection band. It is inferred that near the top side the cholesteric periodic structure is more ordered than that near the bottom side.

Under excitation, photons will be generated whereas the generation of laser light requires a suitable feedback with a reflectivity high enough to overcome the cavity loss. Accordingly, when the top side is pumped, the generated photons near the top surface experience more periodic structure and thus a better feedback. Moreover, at this time, the area near the top side is more strongly pumped than the bottom area due to the absorption of DCM. However, in the case of bottom side facing the pump, the photons generated in the stronger pump area cannot get sufficient distributed feedback. This gives rise to the difference in lasing output power between the two sides.

Unlike the broadened reflection band in dye-doped CLC polymer film, the 8 μm CLC polymer film shows almost the same band shape between the top and bottom sides. There is a very small blue shift and slight reflectivity decrease observed in the reflection band of the bottom side. This is due to the slightly different measurement positions on the film, where the reflection band is not perfectly uniform over the whole cell. Nevertheless the reflection band shape and reflectivity are not changed noticeably because no laser dye is present, and hence no obvious UV intensity gradient and pitch gradient are formed throughout the film thickness. As a polarization recycling layer and polarization dependent reflector, the passive film shows a higher reflectivity than the 15 μm dye-doped CLC film does, providing good polarization conversion efficiency.

#77832 - $15.00 USD Received 5 December 2006; revised 25 February 2007; accepted 28 February 2007

4. Conclusions

By sandwiching the active CLC polymer film between a mirror and a passive CLC reflector, we demonstrate a dramatically enhanced laser emission whose polarization state is circular, but opposite to the handedness of the intrinsic cholesteric. Such a polymer laser device shows significant purity improvement in the output polarization state. Moreover the drastically different lasing properties between the top and bottom sides of a dye-doped CLC polymer film under pump are investigated. This work will be valuable for improving the performance of highly portable and stable CLC-based polymer lasers and, meanwhile, provide new possibilities for operating the polarization states as needed. The underlying mechanism is also helpful in enhancing the other types of light emitting devices, such as LED, and in the applications of polarization manipulation and purification.

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