Enhancing the laser power by stacking multiple dye-doped chiral polymer films

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Abstract: We demonstrate a method for enhancing the laser efficiency by stacking multiple dye-doped chiral polymer films. No laser emission was observed from a single 8 μm film. By stacking two films together, the laser efficiency is dramatically enhanced. Further increasing the number of stacked films, the output laser power is further increased. It is also observed that the output laser power in the forward direction is almost the same as that in the backward for the two- and three-layered films. However, in the six-layered film the output laser power is much stronger in the backward direction than the forward one. This is due to the absorption of the laser dyes and the distributed feedback in the chiral polymer films.

OCIS codes: (230.3720) liquid-crystal devices, (140, 0140) Lasers and laser optics

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1. Introduction

Mirrorless lasers in dye-doped cholesteric liquid crystals (CLCs) have been extensively studied in recent years due to its photonic crystal properties and simple fabrication process. [1-12]. A CLC cell is typically prepared by doping some chiral agents into a nematic LC host. Due to the twisting power of the chiral dopants, the LC molecules form a self-organized periodic helical structure. Since the host LC is a highly birefringent medium, the periodic helical structure gives a periodic modulation of the refractive index. Consequently, a one-dimensional (1D) photonic band gap (PBG) is established whose central wavelength is at $\lambda = np$, where p is the helical pitch and n the average refractive index. The abrupt change of the optical density of state at the band edges provides the possibility of generating laser emission when laser dyes are doped into the CLCs.

So far, however, the output laser power is still relatively low. For most of the applications, high output laser power is required. Several methods have been developed for enhancing the laser efficiency. For instances, the laser efficiency can be improved by choosing laser dyes and liquid crystals with higher order parameters, [13] adding a mirror reflector or passive CLC reflector to form a laser cavity, [14] and sandwiching the active CLC layer between a Fabry-Perot cavity. [15]

In this paper, we demonstrate the laser power enhancement by stacking multiple dye-doped chiral polymer films. It is known that a cavity laser with a longer optical gain medium can produce higher output laser power because more photons are available. While in a mirrorless laser, the optical gain medium, such as dye-doped chiral polymer film, provides not only photons but also feedback amplification of the optical gain. Therefore, increasing the film thickness within a certain range can enhance the output laser power. However, because of the limited anchoring energy induced by the rubbing layer on the glass substrates, it is very difficult to sustain uniform planar cholesteric structure for a LC cell thicker than 15 μm. The imperfect planar cholesteric structure would introduce an appreciable amount of light scattering and, consequently, dramatically decreases the lasing efficiency. In order to increase the effective film thickness with perfect planar cholesteric structure, we stack multiple 8 μm chiral polymer films together. The output laser power is significantly enhanced with the increase of the stacked films. No laser emission can be observed from a single 8 μm film. By stacking two films together, the laser efficiency is dramatically enhanced. Further increasing the stacked films, the laser power is further increased. Moreover, it is found that the output laser power in the backward direction is much stronger than that in the forward for the sample stacked with six 8-μm films.

2. Sample preparation

The chiral monomer host mixture was prepared by mixing the reactive mesogen monomer RMM154, reactive monomer RM82, and chiral CB15 (all from Merck) together. Afterwards, we doped 1 wt% of a highly fluorescent laser dye 4-(Dicyanomethylene)-2-methyl-6-(4-dimethlyaminostyryl)-4H-pran (DCM, from Exciton) to the chiral monomer mixture. The whole mixture was thoroughly mixed before it was capillary-filled into the empty LC cell (8 μm cell gap) in an isotropic state. 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^{o} pretilt angle. The sample was slowly cooled down to 55 °C to reduce the defect formation. Then a UV light was used to illuminate the sample for ~1 hr while keeping the temperature at 55 °C to turn the chiral monomer into chiral polymer film. Then, we separated the two glass substrates of the LC cell and peeled off the chiral polymer film from one of the substrates. The film was cut into several pieces and stacked together one by one very carefully to avoid the generation of the surface corrugation.

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3. Results and discussion

To test the laser performance, we used a frequency-doubled, Q-switched, linearly polarized Nd:YAG pulsed laser (λ=532 nm, from Continuum) which produces a single 4-ns-long pulse as pumping light source. 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 convert the linear polarization into left-handed (LH) circular polarization to avoid the reflection from the reflection band of the cholesteric polymer film. A lens with 15 cm focal length focused the incident beam to a small spot of ~160 μm diameter at the sample. The output laser emission from the sample was collected by a lens to a fiber-optics based universal serial bus (USB) spectrometer (0.4 nm resolution; USB HR2000, Ocean Optics). The setup is illustrated in Fig. 1.

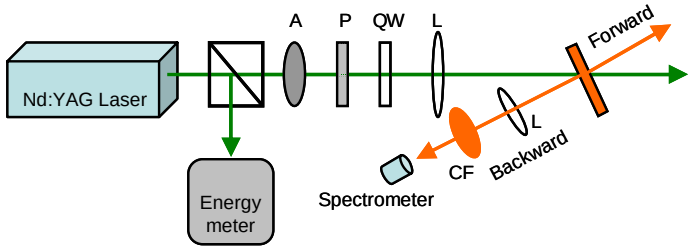

Fig. 1. Experimental setup. A: attenuator; P: polarizer; QW: quarter wave plate; L: lens, CF: color filter.

First, we measured the emission intensity from the backward direction (the propagation direction is opposite to that of the pump laser) of the sample, as shown in Fig. 1. For a single 8 μm dye-doped chiral polymer film, only fluorescence emission was observed even when the pumping energy was increased to 140 μJ. However, by stacking two films together we detected laser emission when the pumping energy was around 11 μJ, as Fig. 2 shows.

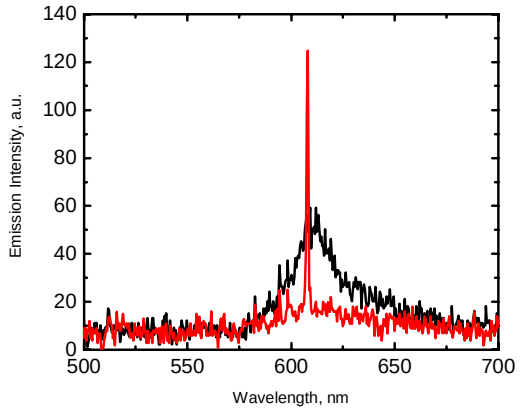

Fig. 2. Emission spectra from a single 8 μm film (black line) at 140 μJ pump energy and double 8 μm films (red line) at 14 μJ.

Further stacking one more film on top of the two combined films, the threshold is decreased to 3.9 μJ and the lasing efficiency is doubled. By stacking two 3-layered films together, the threshold pumping energy is further reduced to 2.5 μJ while the output laser power is tripled as compared to that of the 3-layered films, as shown in Fig. 3. In Fig. 3, the laser emission was measured from the backward direction (as shown in Fig. 1) of the samples.

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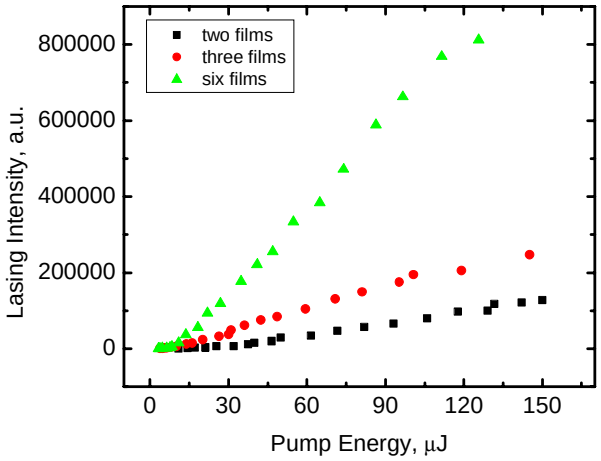

Fig. 3. The output laser power as a function of pumping energy from two- (squares), three- (circles), and six- (triangles) stacked films.

When we measured the laser emission intensity from the forward direction of the samples as a function of pump energy, we found that the output laser power from the forward direction is not always equal to that from the backward direction. In our experiment, the output laser power from the forward of the two-layered and three-layered films is almost the same as that from the backward. However, for the six-layered film, the output laser power from the forward direction is much lower than that from the backward, as Fig. 4 shows. The physical mechanism is discussed as follows.

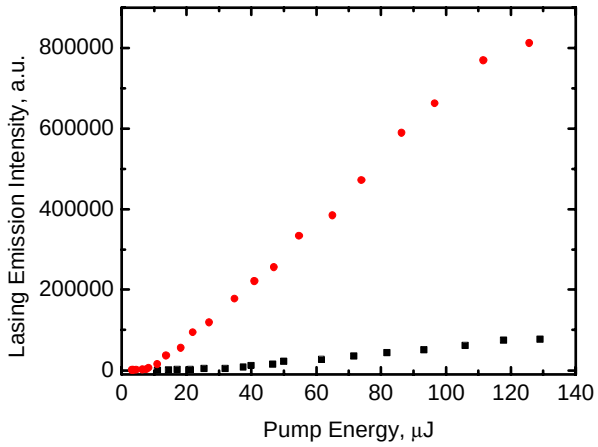

Fig. 4. The output laser power as a function of pump energy from the forward (squares) and backward (circles) directions of the six-stacked films.

It is known that the laser emission can be generated through the feedback amplification only when the optical gain inside the medium is lager than the optical losses. For chiral polymer films, the feedback amplification is provided by the internal distributed feedback of the photonic band gap. The feedback efficiency is determined by the bandwidth of the band gap and the sample thickness. A thicker sample with a wider bandwidth can more efficiently

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amplify the optical gain traveling in the sample to overcome the losses in the medium. In our experiment, all the samples exhibit the same photonic bandwidth of about 70 nm. In a single 8 μm film, the optical gain amplification provided by the feedback is not sufficient to overcome the optical losses. Therefore, we cannot observe laser emission from a single 8 μm film. By stacking two 8 μm films together, the thickness is doubled. The feedback efficiency is dramatically enhanced due to the doubled internal distributed feedback, which can provide sufficient optical gain amplification to overcome the associated optical losses. Therefore, laser emission occurs.

By stacking more films together, the optical gain can be more efficiently amplified due to the longer internal distributed feedback. Therefore, the output laser power is increased. If the sample has no absorption at the pumping laser wavelength, then the output laser power from both sides of the sample should be equal. However, the laser dye employed in our experiments exhibits a relatively large absorption at λ= 532 nm. The pump light intensity will be gradually decreased by the absorption of the laser dyes when the pump light propagates through the sample. If the sample is thin and the pump light intensity is high enough, the attenuation of the pump energy by the absorption of the dye can be ignored and does not affect the laser output noticeably. Therefore, the output laser powers from both sides of the sample are almost the same. But when the pump laser passes through a thicker sample, most of the pump energy could be absorbed by the doped dyes. As a result, the pump intensity at the rear surface of the sample, where the pump laser leaves the sample, is much weaker than that at the front surface, where the pump laser enters. Consequently, much fewer dye molecules could be pumped to the excited state at the rear surface than at the front surface to produce photons. Since the internal distributed feedback can feedback the photons locally, the area with more photons can certainly get higher amplified optical gain and consequently higher output laser power. Thus, the laser power from the backward direction is much stronger than that from the forward direction.

5. Conclusion

We have demonstrated a method to enhance laser efficiency by stacking multiple dye-doped chiral polymer films together. No laser emission was observed from a single 8 μm film. By stacking two films together, the laser efficiency is dramatically enhanced. Further increasing the stacked films, the output laser power can be further increased. It is also observed that the output laser power from the forward direction is almost the same as that from the backward for the two and three stacked films. In the six-layered film, however, the laser power from the backward is much stronger than that from the forward, which is attributed by the absorption of the imbedded laser dyes and the internal distributed feedback of the chiral polymer films.

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