

Multi-wavelength laser from dye-doped cholesteric polymer films

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Abstract: A multi-wavelength laser is demonstrated using a dye-doped cholesteric polymer film whose reflection bandwidth is broadened with several oscillations. Due to the abrupt change of the density of state between oscillation peak and valley, each oscillation functions as a photonic band gap for generating a laser wavelength under the excitation of a pumping laser. As a result, a multiple wavelength laser is generated. Results indicate that the dye-doped cholesteric liquid crystal polymer film is a good candidate for fabricating broadband lasers such as white light lasers. Potential applications include experimental testing of laser materials, identification markers, information displays, and inertial confinement laser fusion.

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1. Introduction

Multi-wavelength lasers have attracted great attention [1–3] because of their wide range of applications, including telecommunications [4], high-precision broadband sensing and spectroscopy [5], metrology, molecular fingerprinting [6], optical clocks [7], and even attosecond physics. For example, a multi-wavelength laser is ideal for characterizing optical amplifiers where the gain profile must be saturated across its whole spectrum for reliable measurements, or polarization mode dispersion and polarization-dependent loss measurements that are usually carried out with tunable sources. Multi-wavelength lasers have been demonstrated using a number of techniques including spectrum slicing in LEDs [8], amplified spontaneous emission from erbium-doped amplifiers (EDFAs) [9], supercontinuum generation in fiber [10], semiconductor optical amplifier (SOA) cavities [11–13], femtosecond pulses [14] as well as in erbium-doped fiber (EDF) oscillators that use a fiber Lyot [15].

In this paper, we demonstrated a multi-wavelength laser from a dye-doped cholesteric liquid crystal polymer film. A cholesteric liquid crystal (CLC) exhibits a self-organized periodic helical structure [16–19]. Since liquid crystals are highly birefringent, the periodic helical structure gives rise to a periodic modulation of the refractive index and consequently offers a one-dimensional photonic band gap with the central wavelength at $\lambda_o = np$, where p is the helical pitch and n the average refractive index. The electromagnetic modes are suppressed within the bandgap but enhanced at the band edge because the group velocity tends to vanish near the band edge so that the density of states (DOS) and spontaneous emission rate are enhanced accordingly. The sharp increase of DOS at photonic band edge allows the possibility for generating a low threshold and high efficiency laser. Therefore, under the excitation of a pump laser, a single wavelength laser with narrow linewidth usually occurs at either edge of the bandgap [17–19]. Recently, multi-wavelength lasers based on cholesteric liquid crystal (CLC) have also been demonstrated. For example, Lee et al. and Hands et al. have obtained one- and two-dimensional spatially distributed multi-wavelength lasers in CLCs [20,21]. In these two examples, however, there is actually only one laser wavelength at a spot. Lee et al. observed a very interesting color cone laser [22], which gives a different laser wavelength at a different angle. But at a specific angle, there is only one laser wavelength. Lin et al. reported a multi-wavelength laser in a dye-doped CLC with oversaturated chiral dopant, which is actually based on the scattering mechanism [23]. Multi-wavelength has also been observed in CLC polymer network [24], which is due to the defects inside the sample. No matter it is caused by scattering or defects, however, the lasing efficiency is reduced dramatically due to large optical loss. In this paper, we obtained a multi-wavelength laser from a dye-doped cholesteric polymer

film (CPL) based on multiple Bragg reflections [16,17]. The fabrication process of CPL is very simple. Moreover, it provides a possibility for obtaining high efficiency multi-wavelength lasers. Thus, the dye-doped CPL is a potential candidate for fabricating broadband lasers.

2. Sample preparation and experimental setup

To fabricate the CPL, we first mixed the reactive mesogen monomer RMM154 (31 wt%), reactive monomer RM82 (31 wt%), and chiral CB15 (38 wt%) (all from Merck) together. Afterwards, we doped 1 wt% of a highly fluorescent laser dye 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminoethyl)-4H-pran (DCM, from Exciton) to the chiral monomer mixture. The whole mixture was thoroughly mixed before it was capillary-filled into an empty LC cell (15 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 $\sim 2\text{-}3^\circ$ pretilt angle. The sample was slowly cooled down to 55°C to reduce the defect formation. We prepared two samples: sample-1 and sample-2, but only sample-2 was illuminated by a UV light for ~ 3 hr while keeping the temperature at 55°C in order to turn the chiral monomer into cholesteric polymer film. If the sample was cured at below 55°C , phase separation would occur.

Figure 1 depicts the experimental setup for testing the laser performance of the two samples. A frequency-doubled, Q-switched, linearly polarized Nd:YAG pulsed laser ($\lambda = 532$ nm, from Continuum) which produces a single 6-ns pulse is used as the pumping laser. All the measurements were performed at 1 Hz laser repetition rate to reduce the accumulated thermal effect because of the 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 in the forward direction of 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).

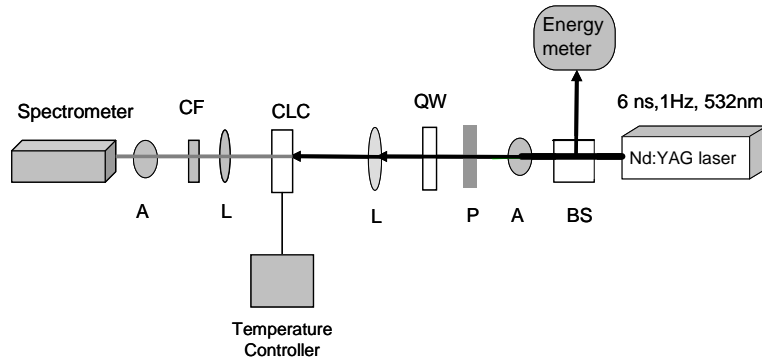


Fig. 1. Experimental setup for characterizing the laser performances. BS: beam splitter; A: aperture; P: polarizer; QW: quarter wave plate; L: lens; CF: color filter.

3. Results and discussion

When the pump energy is below ~ 3 $\mu\text{J}/\text{pulse}$ (threshold energy, ~ 2.7 $\mu\text{J}/\text{pulse}$ and ~ 3 $\mu\text{J}/\text{pulse}$ for samples 1 and 2, respectively), only the spontaneous fluorescence takes place and the emission intensity is very low, as shown by the black lines in Figs. 2(a) and 2(b). As the pump energy exceeds the threshold, the emitted laser intensity is significantly increased and the linewidth of a single lasing wavelength is dramatically decreased to ~ 1.1 nm and ~ 0.7 nm for samples 1 and 2, respectively, as shown by the red lines in Fig. 2(a) and (b). From Fig. 2, only a

single laser wavelength was generated from sample 1 (without UV illumination), but multiple laser wavelengths were generated from sample 2 (with UV illumination).

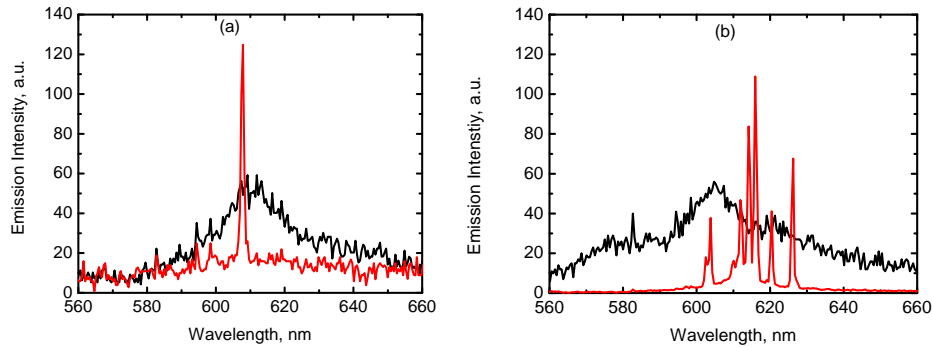


Fig. 2. Fluorescence (black) and laser (red) emission spectra from (a) sample-1, and (b) sample-2.

The results show that the threshold energy of sample 2 is slightly higher than sample 1. The reason is some of the laser dyes in sample 2 are bleached by the UV light, causing the quantum emission efficiency in sample 2 is smaller than that in sample 1. As a result, sample 2 needs slightly higher threshold energy to generate laser emission. To understand the physical mechanism of the multi-wavelength laser generated from the dye-doped CPL and the laser linewidth from sample 2 slightly narrower than that from sample 1, we measured the reflection spectra of the two samples. Results for sample-1 and sample-2 are shown in Figs. 3(a) and 3(b), respectively. We can see that the reflection band of sample-1 is quite flat on the top and the band edge is very sharp. While for sample-2, the top of the reflection band is not flat and, the band edge is not as sharp as that of sample-1. Instead, the reflection band is broadened and there exhibit a few oscillations within the reflection band. According to the electromagnetic theory, in sample 1, the electromagnetic modes are suppressed within the reflection band and enhanced at the band edge. Consequently, the density of states (DOS) and spontaneous emission rate are suppressed within the reflection band and enhanced at the oscillation valley. The abrupt change of DOS at the band edge leading to a laser emission at the band edge under the excitation of a pumping laser. In sample 2, each oscillation functions as a photonic band gap. The electromagnetic modes are suppressed at the oscillation peak and enhanced at the oscillation valley. The abrupt change of DOS between each oscillation peak and valley provides the possibility of generating a laser emission under the excitation of

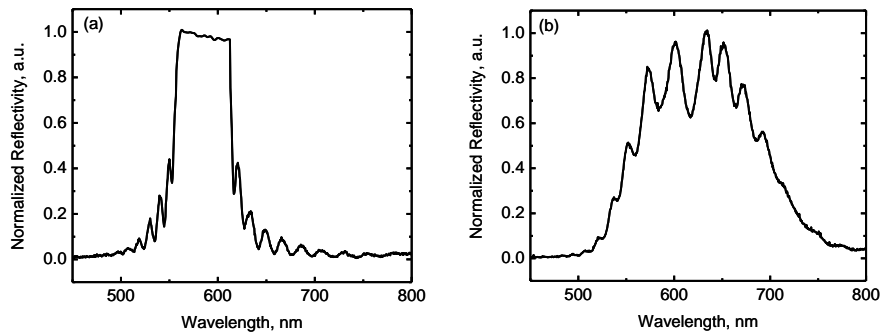


Fig. 3. Measured reflection spectra of (a) sample-1, and (b) sample-2.

a pumping laser. Since there are several oscillations in the sample, multiple wavelengths can be produced simultaneously under the excitation of a pumping laser. As for the slightly narrower laser linewidth in sample 2 than in sample 1, we believe it is due to the slightly higher reflectivity provided by sample 2 than sample 1. According to laser principle, narrower laser linewidth can be generated by the feedback cavity with higher reflectivity. In sample 1, the reflectivity at the band edge change sharply. While in sample 2, the oscillation peak and valley are within the reflection band. It is very possible that the reflection at the lasing position in sample 2 is higher than in sample 1. Thus the laser linewidth from sample 2 is slightly narrower than in sample 1.

To understand why the reflection band of the UV cured sample (#2) is not as flat as the one without UV curing (sample-1) but exhibits many oscillations within the band gap, we checked the structure of the two samples under a polarizing optical microscope. The photographs are depicted in Figs. 4(a) and 4(b) for sample-1 and sample-2, respectively. From Fig. 4(a), we see that the surface of sample-1 is quite uniform except there are some defect lines between the spacers, indicating that sample-1 has perfect planar cholesteric structure inside. In contrast, the surface of sample-2 is uneven, as in Fig. 4(b). This implies that the cholesteric structure inside sample-2 is not perfectly planar and instead, some domains are formed inside after UV illumination, in which the helical structure is directed to different directions. As a result, the reflection band is broadened and there exhibits oscillations within the reflection band.

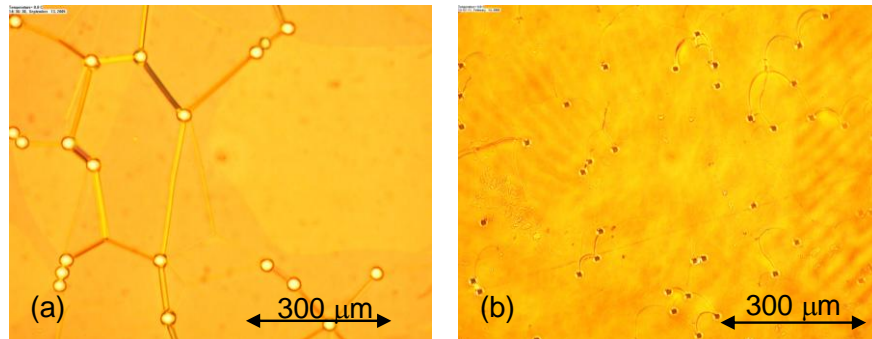


Fig. 4. Photographs of (a) sample-1, and (b) sample-2.

According to the above experimental results, we think that the helical structure oriented at different directions is due to the nonuniform UV illumination. Since the laser dye has some absorption at 365 nm, which is the light source for curing the sample, the UV light intensity inside the CLC sample is different along the z-direction (thickness of the sample). As a result, the polymerization speed is different along the z-direction. The interaction between molecules may cause the local movement of the chiral and monomer molecules. Therefore, a gradient distributed pitch length is induced which in turn broadens the reflection band [25]. In the meantime, the interaction between molecules may also cause the change of the direction of the helical structure. It is also known that blue shift occurs for a CLC film as the incident angle increases. If the direction distribution is not uniform, some oscillations on the broadened reflection band will occur. This explains why our measured reflection band is broadened with oscillations.

Figures 5(a) and 5(b) show the beam image and profile captured with a digital camera and a CCD, respectively. It is seen that the laser emission from our dye-doped cholesteric polymer film exhibits a Gaussian-like beam shape similar to that from a conventional dye-doped CLC [26]. This indicates that the emission is indeed a stimulated laser.

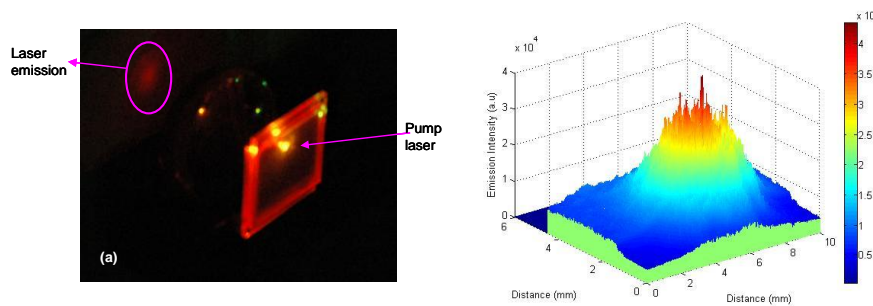


Fig. 5. (a) Lasing beam image and (b) beam profile.

The laser emission from sample-2 as a function of the pump energy is plotted in Fig. 6. As the pump energy exceeds a threshold, the laser emission is almost linearly proportional to the pump energy. The laser efficiency is around 1%, which is not high as compared to the CLC laser. The reason is because the Δn of the reactive monomer used in our experiment is relatively small (only ~ 0.15). To enhance the laser efficiency, a high birefringence monomer should be employed.

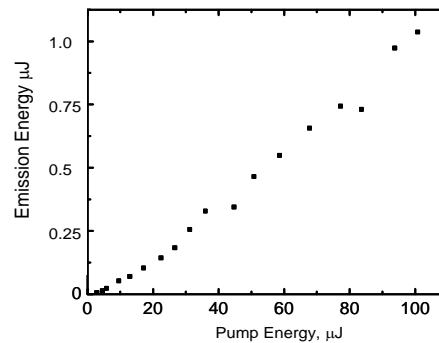


Fig. 6. Laser emission as a function of the pump energy.

5. Conclusion

A multi-wavelength laser is obtained from a dye-doped cholesteric polymer film whose reflection band is broadened allowing for several oscillations. Due to the abrupt change of the density of states between the oscillation peak and valley, each oscillation functions as a photonic band gap, which provides the possibility of generating a laser wavelength under the excitation of a pumping laser. As a result, a multiple wavelength laser is generated. The results indicate that the dye-doped cholesteric liquid crystal polymer film could be a good candidate for fabricating broadband lasers such as white light lasers, which have many applications in experimental testing of laser materials, identification markers, displays, information technology, and inertial confinement laser fusion.

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