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Low threshold polymerised cholesteric liquid crystal film lasers with red, green and blue colour

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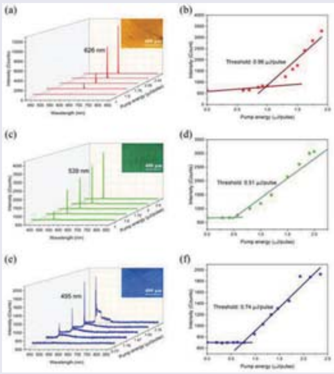
ABSTRACT
We demonstrate dye-doped low threshold polymerised cholesteric liquid crystal (PCLC) film lasers with red, green and blue (RGB) colour. The polymer network structure of lasers, which were fabricated by washing-out/refilling method, enhanced the stability of the laser thus avoiding the external interference like temperature and electromagnetic field and prevented the efficiency of laser dye from decreasing during polymerisation. The proposed RGB PCLC film lasers have application potential in such as coherent light source, multi-wavelength biomedical light source, white laser and other photonic applications.



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Introduction

Liquid crystal (LC) lasers have attracted significant attention due to their tunability, compact and easy fabrication process [1,2]. This technology is mainly based on cholesteric liquid crystals (CLC), which exists in a helical structure where the directors of LC molecules rotate regularly perpendicular to the helical axis [3,4]. The CLC structure corresponds to photonic crystal, which is a material with periodic microstructure composed of different media with different dielectric constant, which can form the photonic band-gap (PBG) due to its periodic orientation of the index ellipsoid. The PBG can block light at a certain range of wavelengths [5]. At the edges of the PBG, the group velocity of photons tends to be zero, and photon density of state diverges, photons will reflect in the structure and form a strong gain. This PBG microstructure gives rise to so called distributed feedback microresonator, which combine the emissive gain media and optical resonator in one structure [6,7].

When the light propagates within CLC, it will be amplified by stimulated emission of radiation, and certain wavelength of light will be reflected due to the Bragg's reflection effect. These repeated activities make the energy stimulates and ejects from the laser. Generally, by selecting different laser dye that absorbs the short laser pulse, the output lasing wavelength can cover a wide range, from ultraviolet (UV) light, visible light to infrared light [8,9]. Also, the influence of different kinds of excitation beams on optical properties of dye-doped CLC structures and lasing characteristics has been studied as well [10,11]. Since LC has good wavelength tuning ability especially under the external stimulus like electric field, temperature and pressure, the laser device can achieve wide tunability [12–14]. And being an all-organic and mirrorless device with low threshold and narrow linewidth, the self-assembly LC laser can form periodic structures without complicated preparation technology compared to other types

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of artificial fabricated photonic crystal [5], providing it with great potential.

Based on those remarkable characteristics of dye-doped CLC lasers, the polymerised CLC (PCLC) film has great potential in the related areas. It is realised through the polymerisation of the mixture of CLC and monomers under the UV light. A dye-doped flexible free-standing PCLC film has been demonstrated to act as laser [15]. It can operate without aligning or fixing, and it has great mechanical flexibility and thermal stability. Moreover, in order to solve the problems about the instability and deformability of a 3D CLC Bragg optical microcavity [16], one solution has been reported to PCLC with reactive mesogens (RMs) [17,18]. This stable film can avoid the interference of external environment and the distortions of LC molecules, so the pitch and wavelength can be better controlled. In addition, a novel method called washing-out /refilling technology has been reported by Guo et al. [19] to fabricate a single-layer CLC film. This kind of film contains double-handed circularly polarised light reflection band, thus provides a higher reflectance and a better lasing condition comparing with conventional CLC film. However, this polymerisation approach that uses UV light may damage the doped laser dye in the mixture. Lin et al. [20,21] demonstrated a wide-band tunable PBG device by refilling LC with dye in the gap of the PCLC gap. A spatial position adjustable and ultra-wide wavelength emissive laser can be achieved with the refilled PCLC film. It can not only increase the efficiencies of the laser dyes and the lasing properties, but also enhance the system stability by avoiding the thermally induced light scattering [22].

In this paper, we demonstrate dye-doped low threshold PCLC film lasers with red, green and blue (RGB) colours. The polymer network structure of lasers, which were fabricated by washing-out/refilling method, enhanced the stability of the laser thus avoiding the external interference like temperature and electromagnetic field and prevented the efficiency of laser dye from decreasing during polymerisation. The proposed RGB PCLC film lasers have application potential such as coherent light source, multi-wavelength biomedical light source, white laser and other photonic applications.

Experiment

Materials preparation

The materials in our experiment include nematic liquid crystals (NLC) E7 (from HCCH, Zhenjiang, China), chiral dopant R5011 (from HCCH), RMs, photoinitiator Darocur1173 (from Sigma-Aldrich, Darmstadt, Germany), laser dyes DCM (from Sigma-Aldrich) and C540A (from Exciton, Lockbourne, OH, USA). The RM material was mixed by RM257, RM82, RM006, RM021 and RM010 (all from Shijiazhuang Sdyano, Shijiazhuang, China), with 30:20:20:20:10 weight ratio [17,18]. Figure 1 shows the chemical structures of RMs, which belong to LC polymers [23]. Here, the RM257 and RM82 are macromolecules that form polymer network. The others are small molecules that are used to stabilise the structure. The photoluminescence (PL) of DCM and C540A excited under UV light is in range of 616–678 and 516–590 nm, respectively.

The central wavelength λ_c of reflection band of CLC is determined by the equation: $\lambda_c = n_{av}p$, where n_{av} is the average refractive index and p is the pitch of helical structure. The bandwidth $\Delta\lambda$ of reflection is given by $\Delta\lambda = (n_e - n_o) \times p$, where n_e and n_o is the ordinary and extraordinary refractive index of NLC, respectively. Three mixtures with different concentrations of NLC

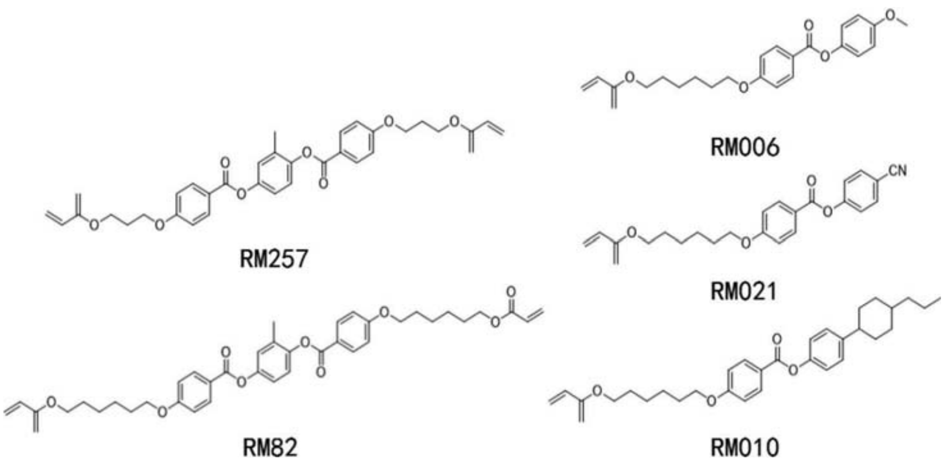

Figure 1. Chemical structures of RMs.

Table 1. The compositions of samples.

					Refilled materials		
					Laser dye		
NLC	Chiral dopant	RMs	Photo-initiator	PCLC film	DCM	C550A	NLC
71.79 wt%	2.21 wt%	25 wt%	1 wt%	A	1 wt%	–	99 wt%
71.48 wt%	2.52 wt%			B	–	4 wt%	96 wt%
71.00 wt%	3.00 wt%			C	–		

and chiral dopant that correspond to different reflection band for RGB lasers generation are shown in Table 1. The NLC and chiral dopant were mixed with RMs (25 wt%) and photo-initiator Darocur1173 (1 wt%) thoroughly to achieve PCLC film. During mixing process, the mixtures were heated up to 70°C in isotropic phase of NLC and stirred at about 1000 rpm with the help of the magnetic stirrer for 10 min. The refilled materials include laser dye of DCM or C540A and NLC.

Cell fabrication

The mixture was then capillary-filled into an empty LC cell with gap of 10 μm. The LC cell consists of two indium-tin-oxides (ITO) coated glass substrates at the inner surface and is anti-parallel rubbed using polyimide (PI). After the CLC/RM mixture was filled in the cell totally, the sample was cooled down to reduce their defects.

The sample was then exposed under UV light (UVEC-4II, LOTS, Shenzhen, China) with intensity of 15 mW/cm^2 for 15 min. The RMs would be photo-polymerised and form a stable three-dimensional polymer network because of the radicals or cations formed by the photo-initiator [23–25]. In the next step, LC cell was immersed in toluene for more than 1 day to dissolve E7 and unpolymerised RM monomers completely. The polymer film was washed repeatedly and fetched out from the cell. Next, NLC E7 and laser dye DCM or C540A as refiller were refilled into polymer scaffold film with weight ratio shown in Table 1. For PCLC film A, mixture of 1 wt% DCM and 99 wt% E7 was refilled. For films B and C, mixture of 4 wt% C540A and 96 wt% E7 was refilled. The polymer scaffold with refilled material re-exhibited helical structure of CLC. The fabrication process is demonstrated in Figure 2.

Optical setup

Figure 3 depicts our experimental configuration to test the laser performance of the three samples. A Q-switched neodymium-doped yttrium aluminum garnet (Nd:YAG) laser with wavelength of 355 nm, repetition rate of 10 Hz, pulse duration of 5 ns and pulse energy of 80 mJ was used to pump the samples. The beam from the pump source was collimated and pulled up by two reflection mirrors M1 and M2.

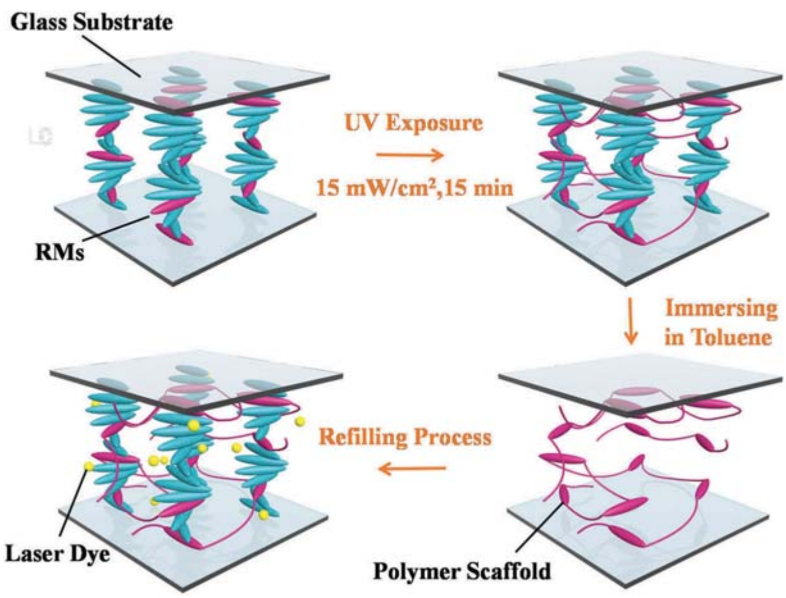

Figure 2. (Colour online) Schematic of fabrication process of PCLC film.

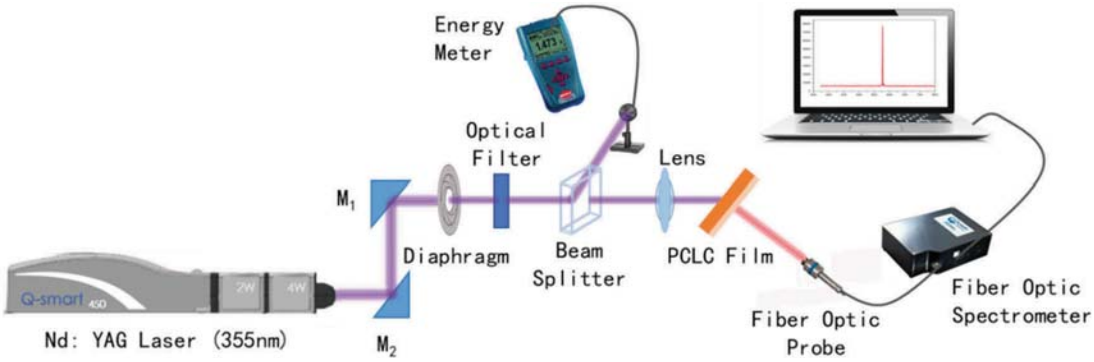

Figure 3. (Colour online) Schematic optical setup of lasing generation in PCLC film.

A diaphragm was used to adjust the beam diameter, and a beam splitter was applied to divide laser beam into two beams, where one beam was captured by an energy metre for power measurement and another beam was used to pump the cholesteric film sample. The pump beam impinged on sample through a convex lens with incident angle 45° to the normal direction of sample surface. A fibre optic spectrometer (HR2000+, from Ocean Optics, Shanghai, China) with a fibre probe was used to measure the output emission spectrum along the normal direction of sample surface.

Results and discussion

The optical setup of transmission measurement consists of white-light source, fibre optic spectrometer, optic fibre and sample stage. The transmission spectrum that is measured through unpolarised light and

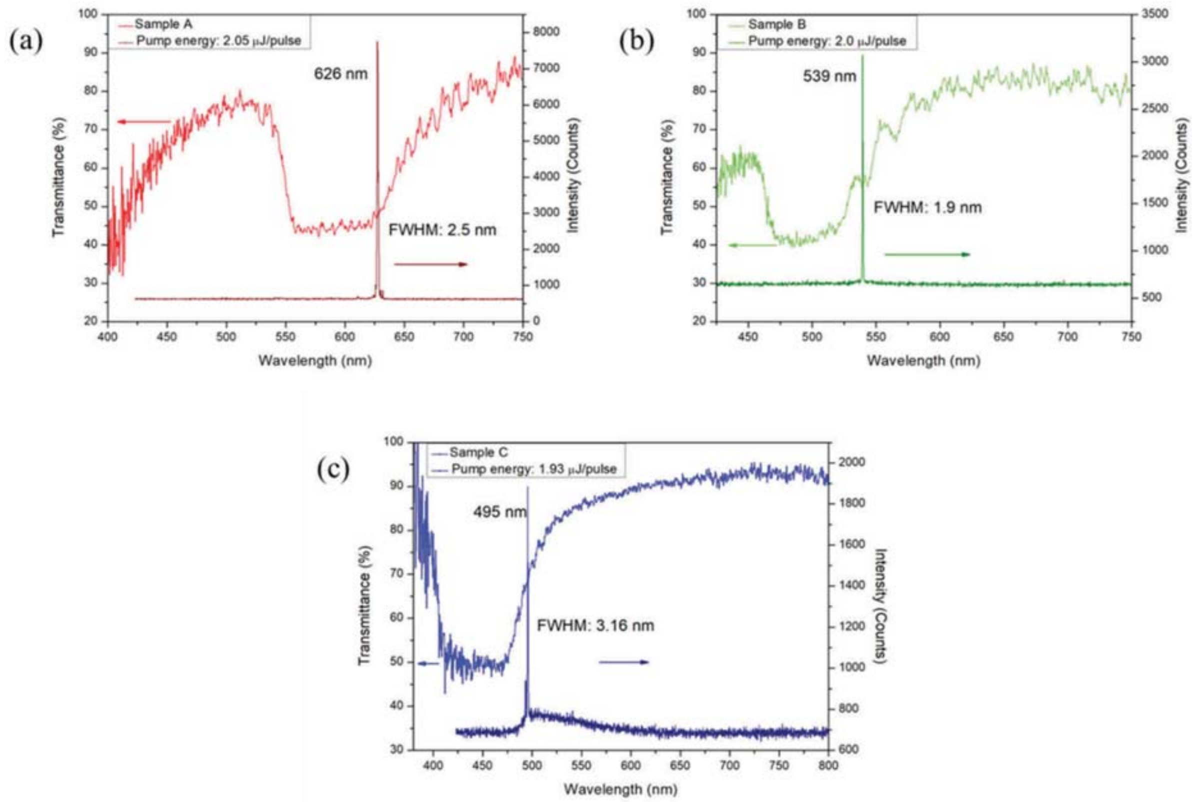

Figure 4. (Colour online) Transmission spectrum and the laser emission spectrum of PCLC film (a) A, (b) B and (c) C, with the pump energy of 2.05, 2.0 and 1.93 μJ/pulse, respectively.

the laser emission spectrum of the fabricated PCLC films A, B and C are shown in Figure 4. As shown in Figure 4(a), the PBG of PCLC film A is around 548–647 nm. Under the excitation of the Nd:YAG laser with pump energy of 2.05 μJ/pulse, the peak lasing occurs at 626 nm with full-width at half maximum (FWHM) of 2.5 nm. In Figure 4(b), the PBG of PCLC film B is about 463–550 nm. When the pump energy is 2.0 μJ/pulse, the position of lasing peak wavelength is 539 nm and the FWHM is 1.9 nm. For PCLC film C as shown in Figure 4(c), the PBG is around 402–498 nm, and the generated laser peak is at 495 nm with FWHM of 3.16 nm. It can be clearly seen that the lasing peaks of the three PCLC films all appear at the long edge of PBG, indicating band-edge lasing. Notably, with the increase of the concentration of chiral dopant: 2.21 wt% of sample A, 2.52 wt% of sample B and 3.00 wt% of sample C, the PBG and lasing wavelength both blue-shift. PCLC films A, B and C act as the optical micro-cavity to generate RGB lasers, respectively.

Figure 5(a,c,e) depicts the laser spectrum of samples A, B and C under different pump energies.

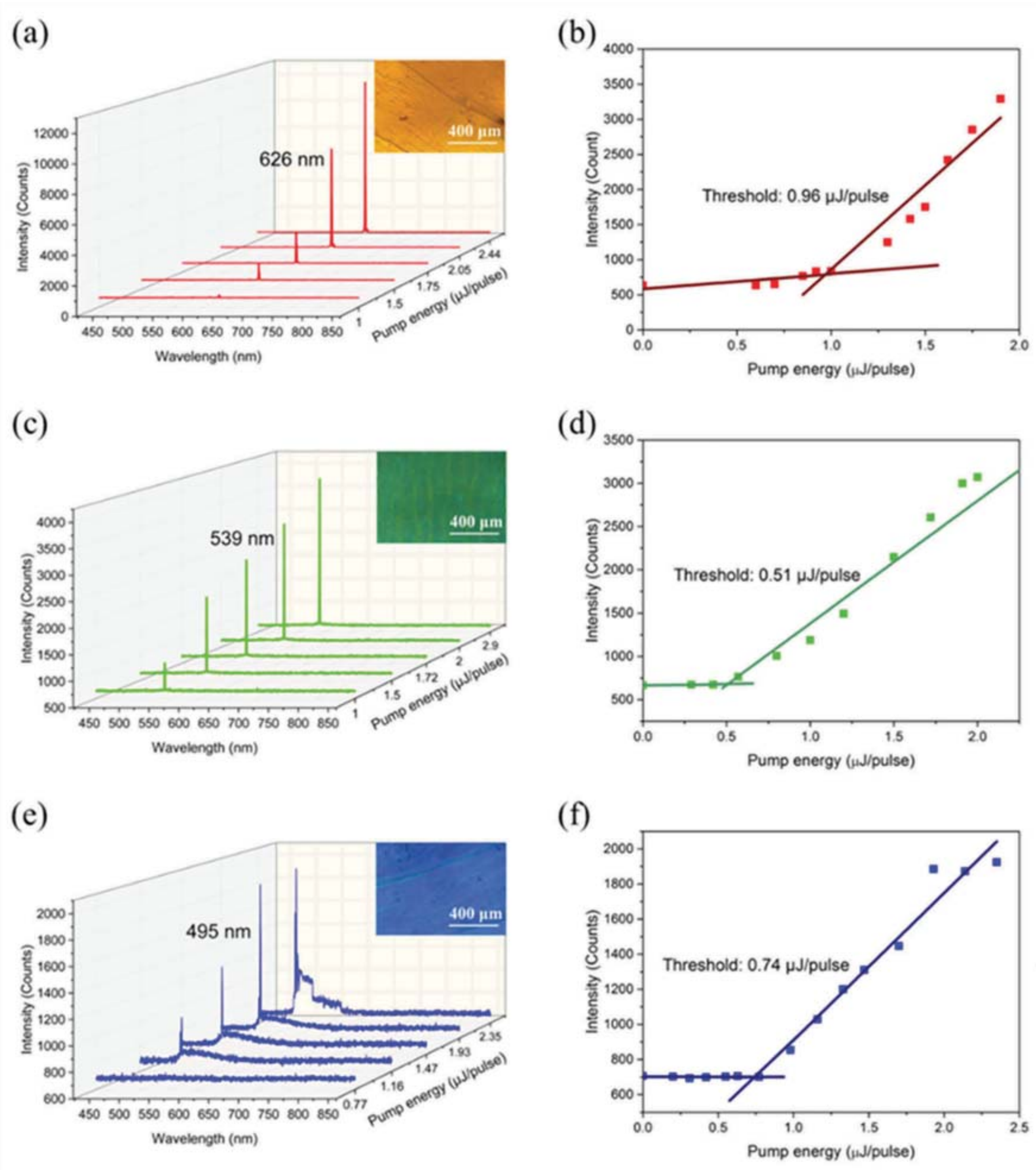

Figure 5. (Colour online) Laser spectrum under different pump energy of (a) sample A, (c) sample B and (e) sample C and the laser intensity versus pump energy of (b) sample A, (d) sample B and (f) sample C.

As the pump energy increases, the peak lasing intensity increases as well. The optical images of samples A, B and C are also shown in the insets, which are captured by polarisation optical microscopy (POM). The laser intensity as a function of pump energy of the three samples is shown in Figure 5(b,d,f), respectively. The thresholds of RGB lasers are 0.96, 0.51 and 0.74 μJ/pulse, respectively, which is much lower than the wide range tunable dye-doped CLC laser with threshold of 18 μJ/pulse [26]. The obtained thresholds are also lower than CLC film lasers reported by Lin et al. [21], where the minimal threshold is 2.32 μJ/pulse. The low-viscosity coefficient of the refilled NLCs (E7, with viscosity coefficient of 90 cP) may lead to stronger capillary action and infiltration rate, thus increase the ratio of NLC refilling volume and the dye molecules can absorb light more efficiently [20]. Besides, the sample preparation process guarantees a low concentration of impurities and imperfections in the bulk, which also leads to the reduction of threshold energy. Therefore, the probability of laser absorption and scattering becomes lower, resulting in easier lasing generation.

When the pump energy begins to increase from zero, there is no laser emission initially, after the pump energy reaches the threshold, the output lasing intensity increases sharply with the increase of pump energy. However, the output lasing will saturate or even decrease beyond a certain range of pump energy, since the higher energy will damage the polymer structure and influence the intensity of output lasing. And when the pump light intensity exceeds a certain limit, the energy transfer process in the dye reaches saturation, which cannot provide further increase of the fluorescence intensity.

Conclusion

In summary, dye-doped low threshold PCLC film lasers with RGB colours have been demonstrated. The laser emissions are generated at the long edge of the PBG with narrow FWHM. High stability and machining property of laser have been achieved due to the polymer network structure. The lower viscosity coefficient of refilled NLC might be useful for reduction of threshold. The proposed RGB PCLC film lasers have application potential such as coherent light source, multi-wavelength biomedical light source, white laser and other photonic applications.

Disclosure statement

No potential conflict of interest was reported by the authors.

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