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Random lasing from cholesteric liquid crystal microspheres dispersed in glycerol

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We demonstrate random lasing from a scattering system formed by a cholesteric liquid crystal dispersed in glycerol. Strong scattering of light is produced from the interference between the cholesteric liquid crystal microsphere and glycerol and leads to random lasing. The optical properties of random lasing, such as intensity, threshold, and the temperature effect on lasing emission are demonstrated. The random laser is distinguished from the band-edge laser generated within the cholesteric liquid crystal microspheres by analyzing the positions of the photonic band-edge of the cholesteric liquid crystal and the photoluminescence of the doped laser dye. The random laser from cholesteric liquid crystal microspheres in glycerol possesses a simple fabrication process, small volume, and low threshold, which enable it to be used in speckle-free imaging, target identification, biomedicine, document coding, and other photonic devices.

OCIS codes: (160.3710) Liquid crystals; (290.4210) Multiple scattering; (140.2050) Dye lasers.

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1. INTRODUCTION

Random lasers were first reported by Letokhov in 1968 [1]. Studies carried out in the literature have shown that random laser devices are characterized by features such as low spatial coherence, omnidirectional emission, multiple lasing wavelengths, and tiny dimensions [2,3]. Consequently, these compact random laser devices are of significant interest for a wide range of applications including speckle-free imaging, target identification, biomedicine, document coding, and photonic devices [4–6]. To generate a random laser, it is crucial to produce recurrent multiple scattering in the diffusive materials [7]. Many materials, such as ZnO powders [8,9], polymers [10], porous glass [11], and biological structures [12], have been used to fabricate random lasers.

Liquid crystal (LC) is a good scattering material for random lasers due to the large optical birefringence. In comparison with conventional diffusive materials, the LC is stimulus-responsive material under stimuli such as electric field, magnetic field, optical irradiation, or environmental temperature [13–17]. LC random lasers have been investigated extensively during the past decades [5,10,16–23]. Liu *et al.* reported gain narrowing and random lasing from dye-doped polymer dispersed liquid crystal (PDLC) with nanoscale liquid crystal droplets [10]. Ferjani *et al.* [18] reported statistical analysis of random lasing emission properties in nematic liquid crystals. The molecules of cholesteric liquid crystal (CLC) are arranged as a helical structure where the director is twisted uniformly in space as a function of position along the helical axis, perpendicular to the director. He *et al.* have reported random lasing in a dye-doped CLC polymer solution, where a mixture of (E-CE)C/AA solution was used as the scatter material, and the effects of concentration of (E-CE)C/AA solution and the thickness of the sample on the random lasing were investigated [22]. Humar and Musevic first reported 3D microlasers from self-assembled CLC microdroplets, and the structure of the CLC microdroplet within glycerol was analyzed [23]. Recently, Zhu *et al.* investigated the random laser emission in a sphere-phase liquid crystal [24]. In the aforementioned works, strong scattering of light has been generated between microspheres or liquid crystal molecules, formed by LC and polymer, sphere-phase LC, or polymer-dispersed LC. However, the reports on random laser based on CLC are rare.

Herein we report on a random laser from dye-doped cholesteric liquid crystal microspheres dispersed in glycerol. To the best of our knowledge, this is the first study of this kind. A scattering system that consists of cholesteric liquid crystal microspheres and glycerol can diffuse light strongly and generate random lasing due to the interference associated with multiple scattering. The random laser from CLC microspheres in glycerol possesses a simple fabrication process, small volume, and low threshold, which enable it to be used in speckle-free

imaging, target identification, biomedicine, document coding, and other photonic devices.

2. EXPERIMENT

First, the cholesteric liquid crystal was prepared by adding 3.18 wt. % of high-twisting-power chiral dopant R5011 (HCCH) and 1.8 wt. % of laser dye pyrromethene 597 (Exciton) into 95.02 wt. % of nematic liquid crystal E7 (HCCH) with $n_o = 1.52$ and $n_e = 1.71$, where the n_o and n_e are the ordinary and extraordinary refractive indexes, respectively. Then the CLC was dispersed in glycerol by a magnetic stirrer with rotation speed of 1000 rpm for 10 min, at weight ratio of 4:96. Finally, the mixture was filled into a LC cell formed by two indium-tin-oxide (ITO)-coated glass substrates without surface treatment. The cell gap is 150 μm.

Figure 1(a) illustrates the experimental setup of a random laser generated from CLC microspheres dispersed in glycerol. A Q-switched Neodymium-doped Yttrium Aluminum Garnet (Nd:YAG) laser (Q-smart 450, Quantel) with a wavelength of 532 nm was used to pump the sample. The pulse width, repetition rate of the Nd:YAG laser was 10 ns, and 10 Hz, respectively. An attenuator (A) was used to adjust the intensity of the pump laser. In our experiment, the laser beam was separated into two beams by a nonpolarized beam splitter (NPBS, BS016-50:50, Thorlabs). One beam was monitored by an energy meter, and another beam was focused to the sample through an objective (LMH-20X-532, Thorlabs). The radius of the focused laser beam on the sample was ~30 μm (from center to edge of the laser spot). The laser emission was collected by a spectrometer with a resolution of 0.05 nm (USB2000+, Ocean Optics).

A simple illustration of random lasing from CLC microspheres dispersed in glycerol is demonstrated in Fig. 1(b). The green and red rays represent the pump light and output laser, respectively. The laser dye and CLC microsphere are

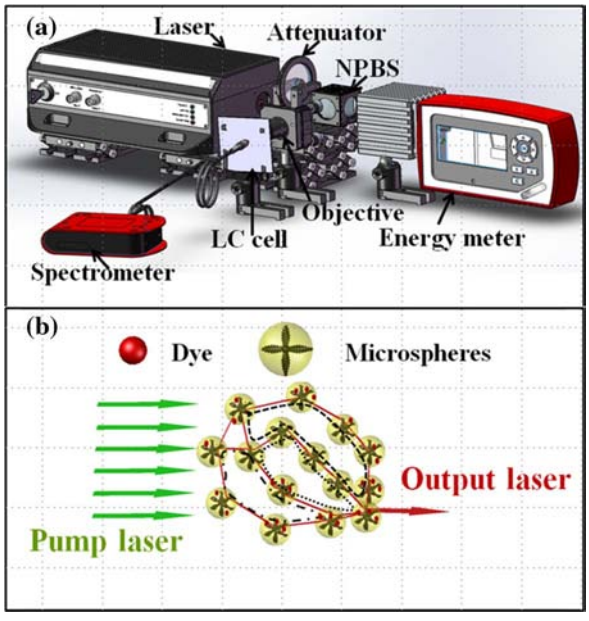

Fig. 1. (a) Schematic of optical setup for random lasing generation. NPBS, non-polarizing beam splitter; (b) schematic of closed loop formed in CLC microspheres/glycerol scattering system.

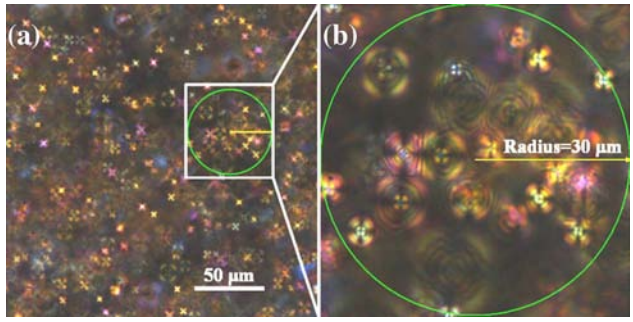

Fig. 2. (a) Optical image of CLC microspheres observed under polarized optical microscopy. (b) A magnified image of selected region. The scale bar is 50 μm.

represented by red and yellow balls. In glycerol, the CLC microspheres form naturally due to the surface tension between the CLC and glycerol, where the surface tension intends to reduce the surface area for a given volume. In CLC, the helical structure of LC that originates from the center of the microsphere gives rise to concentric shells of the constant refractive index [23]. When a laser is pumped on the CLC microspheres, the photoluminescence of laser dye PM597 will scatter between different microspheres. The scattering is complex, yet completely coherent, meaning that the phase of each optical wavelet undergoes a random path. When some of the rays return to their starting positions under constructive interference, a closed-loop path (dashed line) will be formed and enable coherent light. If the amplification along such a loop patch exceeds the loss, laser emission will occur [11,24,25]. The random lasing obtained here was unpolarized.

Figures 2(a) and 2(b) show optical image and magnified image of CLC microspheres observed under polarized optical microscopy (Nikon Ti, POM). The petal-like images indicate the periodic modulation of the refractive index in the CLC microspheres. We can see that the CLC microspheres are distributed in the three-dimensional space and the size of the microspheres varies from 0.5 to 10 μm. The sizes of microspheres were measured and marked by software on POM from Nikon.

In our experiment, the microspheres of pure LC are clustered in glycerol quickly, thus difficult to stabilize within glycerol. The optical images of LC for 0, 5, and 15 min are shown in Figs. 3(a)–3(c). It is clear to see that there is a significant change of LC microspheres in glycerol. In contrast, the CLC microspheres are more stable than the LC microspheres. Figure 3(d) shows the optical images of the CLC microspheres after two months, where many microspheres are still stable inside the glycerol.

3. RESULTS

Figure 4(a) depicts the photoluminescence (PL) spectrum of laser dye PM597 (blue line) in CLC microspheres and random lasing (black line). When the sample was excited by a pulse pump laser with wavelength of 532 nm and energy of 70.1 μJ/pulse, several discrete lasing peaks appeared at 564 nm, 570 nm, 573 nm, 576 nm, 578 nm, 581 nm, 584 nm, 587 nm, and 592 nm, respectively, with corresponding full width at half maximums (FWHM) of 0.4 nm, 0.9 nm, 0.5 nm, 0.5 nm, 0.8 nm, 0.4 nm, 0.6 nm, 0.5 nm, and 0.3 nm,

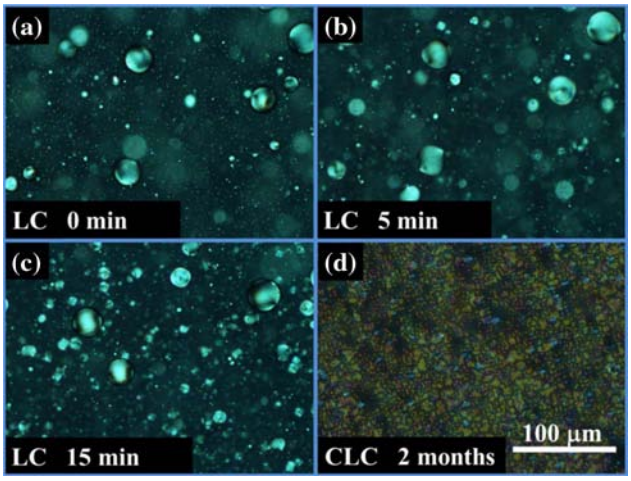

Fig. 3. Optical images of (a) LC for 0 min; (b) LC for 5 min; (c) LC 15 min; and (d) CLC for two months. The scale bar is 100 μm.

respectively. The PL spectrum of PM597 (centered at 598 nm) was overlapped with an emission laser. Here, the transport mean free path (L) of the fluorescence photons in the scattering of the glycerol/CLC system was measured to identify the formation mechanism of the random lasing. The L is calculated by $L \approx \lambda/(\pi * \theta)$, where λ is wavelength and θ is the scattering cone [17]. In our experiment, we have $\theta = 279$ mrad and $\lambda = 633$ nm, thus $L \approx 0.36$ μm. Given the condition: $k * L = 2\pi * L/\lambda_0 = 3.9 > 1$ (where k is the magnitude of the local wave vector and λ_0 is wavelength of output laser), the random lasing in the present study should result from the diffusion of the fluorescence photons via the multiscattering from the glycerol/CLC system [17,26].

In glycerol, the CLC microspheres were 3D microcavities with photonic band-gap (PBG) structure, which would prefer to generate a band-edge type laser [23]. To identify the lasing obtained in our experiment from the band-edge type lasing, we measured the transmittance (black color) and reflectance (blue color) of the CLC microspheres, as shown in Fig. 4(b). The yellow region represents the PBG of a CLC microsphere that ranges from 429 to 497 nm. If a band-edge type laser were generated, the obtained lasing should be located around the photonic band-edge, either in range of 429 ~ 434 nm (short

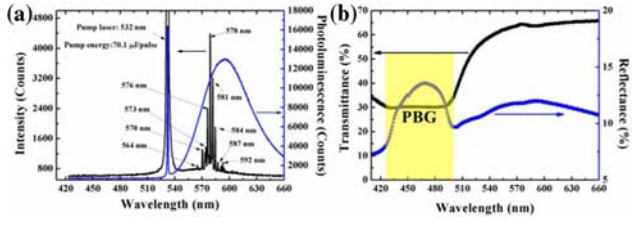

Fig. 4. (a) PL spectrum of PM597 (black color) in CLC microspheres and the spectrum of random laser (blue color). The output laser peaks are at 564, 570, 573, 576, 578, 581, 584, 587, and 592 nm. The pump energy is 70.1 μJ/pulse. (b) The transmittance (black color) and reflectance (blue color) of CLC microspheres. The yellow region represented the PBG of CLC microspheres in range of 429 ~ 497 nm. The wavelengths of output lasing are far from the photonic band-edge, indicating the lasing is photonic band-edge type lasing.

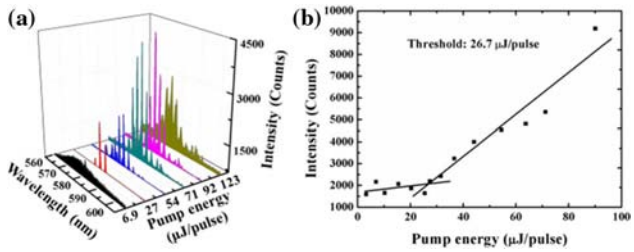

Fig. 5. (a) Lasing spectra under different pumping energies. (b) Dependence of lasing intensity as function of pump energy. The threshold is 26.7 μJ/pulse at 578 nm.

edge) or 494 ~ 497 nm (long edge). However, the lasing peaks were in range of 578 ~ 596 nm in our experiment, which were far from the photonic band-edge. Therefore, we can confirm that the emission lasing obtained here was random lasing, but not the band-edge type.

Figure 5 demonstrates spectrum and input–output characteristics of random lasing. The peaks and intensity of the lasing were variable under different pump energies, which were the typical behaviors of random lasing [2]. This phenomenon further confirmed that the obtained lasing was a kind of random lasing generated from the CLC microsphere/glycerol scattering system. In Fig. 5(a), we can see that the lasing intensity increases with the increase of pump energy up to 71 μJ/pulse. The intensity of lasing reaches it maximum when the pump energy increases to 123 μJ/pulse, above which the intensity reduces with the increase of pump energy. When the pump energy was larger than the critical value of 123 μJ/pulse, the local temperature of the CLC microsphere might be increased above the clear temperature of liquid crystal and then triggered the transition of the CLC from the liquid crystal phase to the isotropic phase. As the effective refractive index of the liquid crystal decreased in the process of phase transition with increased temperature [27], it would destroy the closed-loop path previously formed in the CLC microspheres/glycerol scattering system. Therefore, a poorer random lasing was obtained at the fixed detection position. From the plot of the output light as a function of the pump energy [Fig. 5(b)], the sample exhibits a lasing threshold of 26.7 μJ/pulse at wavelength of 578 nm, above which the lasing intensity increases rapidly with the increase of pump energy.

To investigate the temperature effect on random lasing, the evolution of the lasing spectrum at different temperatures is shown in Fig. 6(a). When the temperature increased from 24°C to 54°C, and the sample was excited by a pump laser with an energy of 50 μJ/pulse; in this case, both the number and intensity of lasing peaks were reduced. There were six lasing peaks (566, 568, 571, 572, 575, and 577 nm) at 24°C, four lasing peaks (569, 570, 573, and 577 nm) at 40°C, and only two lasing peaks at 52°C, indicating a suppressed lasing oscillation. The possible reason is explained by the following. The effective index of CLC is 1.63 (24°C) and 1.57 (54°C), according to data provided by producer. In contrast, the measured indexes of glycerol are 1.467 (24°C) and 1.460 (54°C). Therefore, the index mismatch between the CLC droplet and the glycerol decreases from 0.170 (24°C) to 0.103

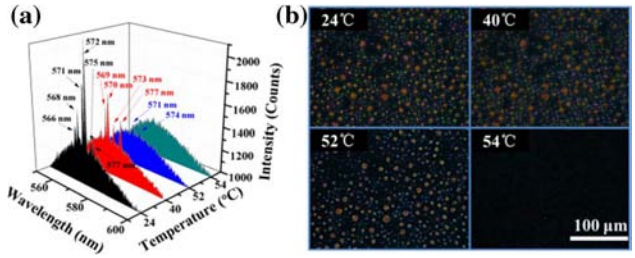

Fig. 6. (a) Temperature effect on random lasing. (b) Images of the CLC microspheres observed under polarizing optical microscope at different temperatures.

(54°C) when temperature increases, which might lead to weaker diffusion between the CLC microspheres and the glycerol, and finally poorer random laser oscillations. However, the real underlying mechanism is still under investigation.

Figure 6(b) demonstrates the images of the CLC microspheres observed under a polarizing optical microscope at different temperatures. The number and morphology of CLC microspheres changes a lot with the increase of temperature from 24°C to 52°C. At 54°C, the CLC is in isotropic phase, which leads to a black image.

4. CONCLUSION

In summary, we demonstrate random lasing from dye-doped CLC microspheres dispersed in glycerol. A scattering system that consists of CLC microspheres and glycerol can diffuse light strongly, and thus generate random lasing due to the interference associated with multiple scattering. The lasing properties, including intensity and threshold, have been studied. The random laser is distinguished from the photonic band-edge laser generated within CLC microspheres by analyzing the positions of the photonic band-edge of CLC. The temperature effect on laser emission is also investigated. The lasing intensity of the random laser decreases as the temperature increases. The underlying mechanism might be the change of the effective refractive index of CLC and glycerol, which would change or destroy the closed-loop path previously formed in the CLC microspheres/glycerol scattering system. The random laser from CLC microspheres in glycerol possesses simple a fabrication process, small volume, and low threshold, which enable it to be used in speckle-free imaging, target identification, biomedicine, document coding, and other photonic devices.

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