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Thermally tunable random laser in dye-doped liquid crystals

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A thermally tunable random laser in dye-doped liquid crystals (DDLCs) is reported. The gain medium of PM597 dye-doped E7 nematic LC is injected into a glass cell. The experimental results show that the random lasing is still obtained when the cell temperature becomes higher, even above the nematic–isotropic transition, and that its polarization changes at the same time. Temperature has little effect on the full width at half maxiumum of the random lasing. The center wavelength of the random lasing shifts from 575.69 nm to 593.43 nm when the temperature increases from 25.5°C to 148°C. Meanwhile, a random laser based on a solution of laser dye is first reported in this article. The reasons are possibly that nanoparticles consisting of dye molecules provide a new scattering mechanism in both solution and isotropic phase.

Keywords: random laser; thermal effects; multiple scattering; liquid crystal

1. Introduction

Random lasers originate from active disordered media, in which multiple scattering provides feedback for radiation light. Diffusive lasing has a very peculiar behavior, as randomness of laser emission is observed in time, space, and frequency [1]. To create a random laser, light diffusion must be combined with light amplification. The threshold at which a random laser starts to manifest is determined by the careful balance between gain and loss [2]. The loss for a random laser is due to light that escapes through the sample surface; the overall gain depends on the excitation energy and on how strongly the material scatters light (expressed in terms of a photon-diffusion constant). The more the material scatters light (the smaller the diffusion constant), the longer the light is kept inside and the larger the overall gain can grow [3].

The tunable laser is an important component in optical communications and other applications. By use of liquid crystals as disordered media, random laser action can be controlled by the external environment (such as temperature and electric field) [3,4]. Because the phase or molecular orientation of a liquid crystal depends on the external environment, its anisotropy distribution changes at the same time. The tunable random laser explores a new way for practical application of random lasers and has potential applications in photonics, temperature-sensitive display, and remote temperature sensing [3].

Some researchers have studied the thermal behavior of random lasing in dye-doped liquid crystals (DDLCs). However, the study evidenced that the random lasing disappeared when the temperature is higher than the clear point of the liquid crystals [3–7] or 70°C [8], indicating that it cannot be used at high temperature. The structure of a wavelength-tunable random laser in DDLCs is complex [9]. In this paper, we report a thermally tunable random laser in DDLCs for a cell that can be used at high temperature (above 148°C). Both the intensity and center wavelength can be tuned via the temperature. In addition, the polarization of random lasing changes at the same time.

2. Experimental setup

The PM597 dye (Pyrromethene 597 provided by Exciton, with 0.4% by weight) was doped into a nematic liquid crystal, NLC E7. The LC bulk phase sequence is crystal–(10°C)–nematic–(61°C)–isotropic, while the principal refractive indices of E7 are n_o = 1.521 and n_e = 1.746. The mixture consisting of NLC E7 and PM597 was injected into a wedge-shaped glass cell. The cell was then filled with the mixture through the capillary effect to form homogeneously aligned DDLCs cells. The cell was formed by two glass plates separated by Mylar spacers, having a variable thickness between 25 and 200 μm. The plates were covered with rubbed polyimide

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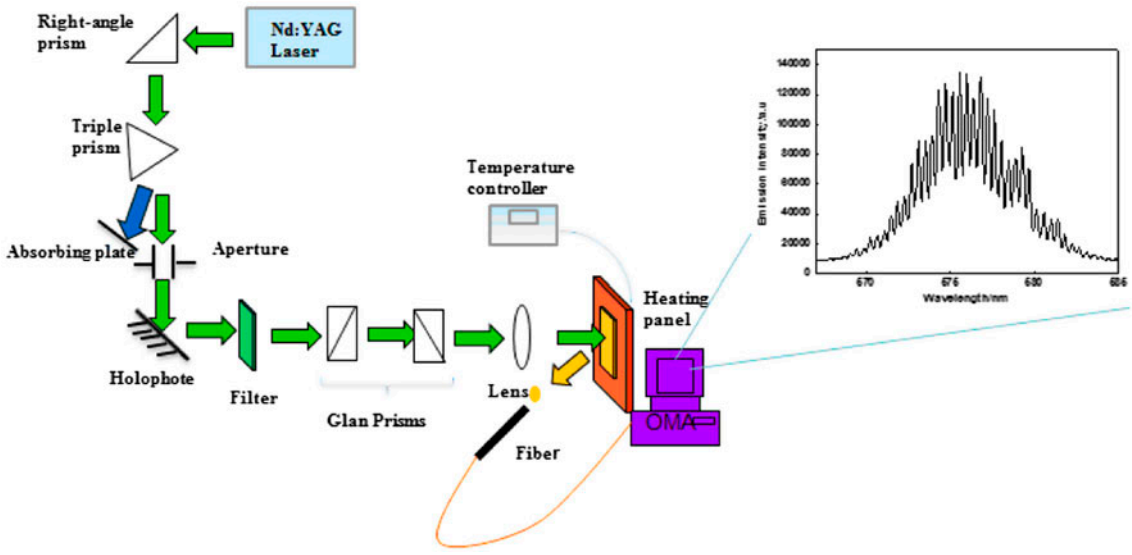

Figure 1. Experimental setup.

alignment layers to induce the homogeneous alignment of the NLC molecules at the interface. Laser dyes (PM597 or DCM, 0.01 mol/L) were dissolved in C_4H_8O. The laser dye solutions were then confined in five different geometries: glass cell (185 μm), glass bottle, plastic test tube, cuvette, and free droplet.

Figure 1 shows the experimental setup for examining the thermally tunable random laser in DDLCs. The pump light originates from a Nd:YAG laser with a pulse duration of 8 ns and repetition rate of 10 Hz. The wavelengths of output light were 355 nm, 532 nm, and 1064 nm. The direction of the mixing light is changed by a right-angled prism and the mixing light is separated by a triple prism. An aperture is used to obtain a uniform part of the green light spot (532 nm) and the other wavelengths of light (532 nm and 1064 nm) are absorbed by an absorbing plate. Then the green light passes a reflection mirror, green filter and Glan prism. It is finally focused by a lens (focal length is 21 cm) on the cell with an incident angle of 30°. Pump light is linearly polarized light and its polarization angle is 0° (vertical relative to horizontal plane). A high resolution optical multi-channel analyzer (OMA) was used to measure the random lasing signal, having a resolution of about 0.1 nm. The Glan prisms were placed in front of the lens to vary the incident pulse energy. The sample was placed in a purposely built heating panel, as shown in Figure 1.

If the pump energy is above the threshold energy, the random lasing is obtained immediately. Meanwhile, yellow spots were observed in both forward and backward directions. The spectrum was detected by the OMA, as shown by the illustration of Figure 1.

3. Results and discussion

The thermal behavior and characteristics of random laser action in a dye-doped nematic liquid crystalline system were studied by varying the temperature in the range from 23.6°C (room temperature) to 148°C (isotropic phase, Figure 2).

Figure 3(*a*) illustrates the temperature dependence of the intensity (black curve) and full width at half maximum (FWHM, blue curve) of the random lasing in DDLCs (the curve breaks because the random lasing disappears in the temperature range 43.2°C–44°C). It shows that the intensity of random lasing gradually reduces when the temperature increases from 23.6°C to 44°C,

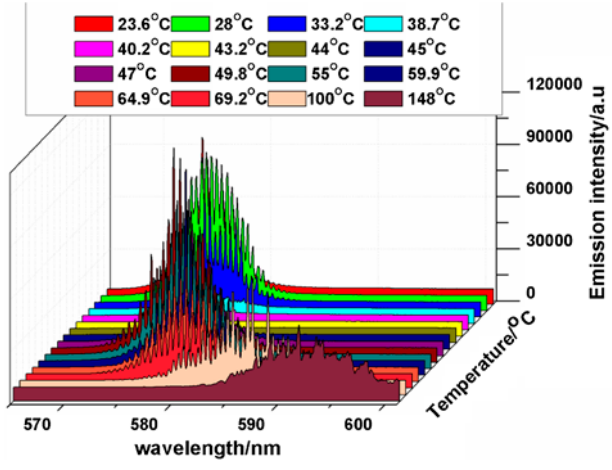

Figure 2. Temperature dependence of the random lasing in DDLCs.

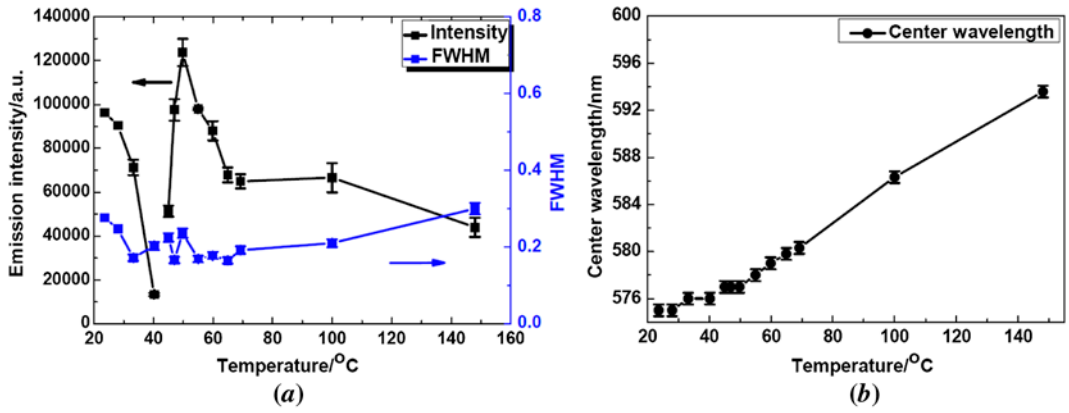

Figure 3. (*a*) Temperature dependence of the emission intensity (black curve) and FWHM (blue curve) of the random lasing in DDLCs. (*b*) Temperature dependence of the center wavelength of the random lasing in DDLCs. (The color version of this figure is included in the online version of the journal.)

Table 1. The influence of factors on the vanishing temperature of random lasing in DDLCs.

Pump energy (μJ/pulse)	Vanishing temperature (°C)	Sample thickness (μm)	Vanishing temperature (°C)	Dye concentration (wt.%)	Vanishing temperature (°C)
11	44	45.86	44–45	0.2	46
20	44	107.61	44–45	0.3	45
30	43	136.30	44–45	0.6	45

and it fades to zero at the temperature of 44°C. This is because increasing sample temperature will diminish the nematic order parameter and result in a higher lasing threshold, approaching the behavior of a totally disordered system [6]. The study shows that pump energy, sample thicknesses, dye concentration, and rubbing direction have little influence on the vanishing temperature of random lasing in DDLCs (Table 1). However, it was found to reappear at a temperature approaching the nematic–isotropic (N-I) transition (45°C). Ferjani et al. explained that the unexpected reoccurrence of random lasing at higher temperature, in proximity to the N-I transition, was found to be related to a different scattering mechanism, which was micro-droplet nucleation and critical opalescence [7]. Theoretically, the emission intensity completely vanishes when the temperature of the cell is higher than the N-I transition, because the system is in an isotropic phase. But the random lasing was still obtained when the temperature increased up to 148°C, indicating that a different scattering mechanism occurs. This will be discussed later. Temperature has little effect on the FWHM of the random lasing (blue curve in Figure 3(*a*)), but the center wavelength shifts from 575 nm

Table 2. The influence of temperature on the polarization of the random lasing. β is the polarization angle of the random lasing in DDLCs, *T* is the temperature of the sample, and θ is the angle between the rubbing direction and the polarization direction of pump light.

	θ (°)			
β (°) *T* (°C)	**0**	**30**	**60**	90
25	**4**	**22**	**60**	82
30	**4**	**22**	**60**	82
35	**4**	**22**	**60**	82
40	**4**	**22**	**60**	82
50	**6**	**6**	**6**	6
60	**6**	**6**	**6**	6
70	6	6	6	6

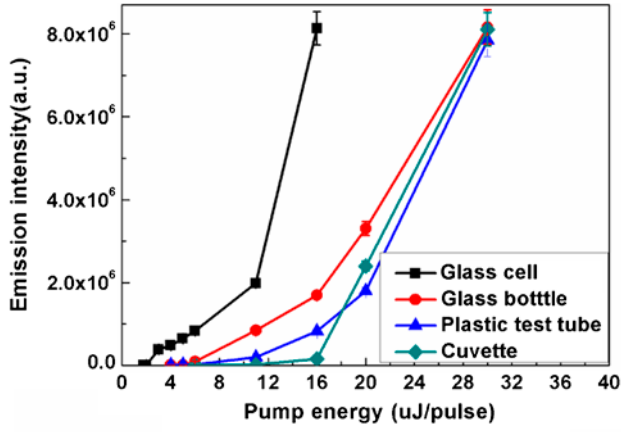

Figure 4. Input–output characteristics of different structures. (The color version of this figure is included in the online version of the journal.)

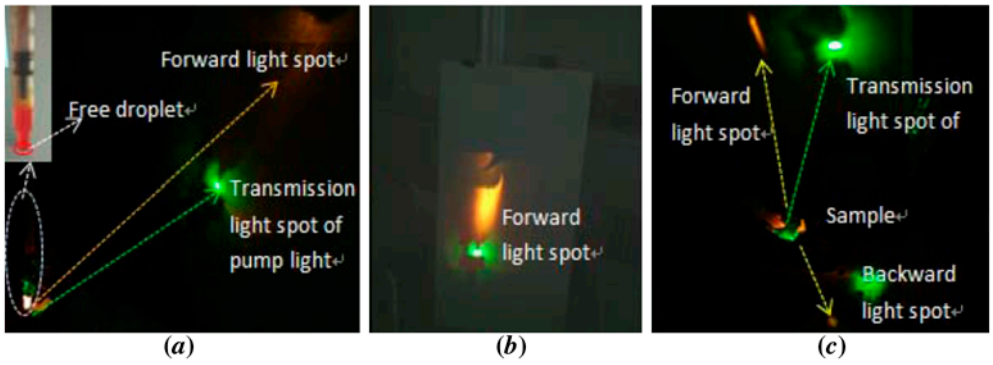

Figure 5. Time dependence of light spots in free droplet of DDLCs: (*a*) 0.5 min; (*b*) 1 min; (*c*) 1.5 min. (The color version of this figure is included in the online version of the journal.)

to 593.6 nm when the temperature increases from 23.6°C to 148°C (Figure 3(*b*)), which is the native characteristic of the PM597 dye. The experiment was reversible.

The polarization of the random lasing at different temperatures was determined by varying the angle between the rubbing direction and the polarization direction of pump light (named as θ). Table 2 shows that the random lasing is linearly polarized light and its polarization depends on the rubbing direction when the temperature is below 40°C. But the polarization angle of the random lasing is fixed (remaining at 6°) and is almost parallel to the polarization direction of pump light when the temperature is over 50°C, no matter if the value of θ is different. This means that the polarization of the random lasing depends on the rubbing direction and the polarization of pump light in nematic phase and in isotropic phase (or proximity of N-I transition), respectively.

In order to explain the random laser action in the isotropic phase, the laser dye solution consisting of PM597 and C_4H_8O was confined in four different geometries: glass cell (185 μm), glass bottle, plastic test tube, and cuvette. After pumping by laser (wavelength is 532 nm), yellow light spots can be found in forward or backward directions and the spectrum has obvious splitting peaks. Figure 4 shows the input–output characteristics of the four systems. The threshold energies were 1.8 μJ/pulse, 4 μJ/pulse, 4 μJ/pulse, and 8 μJ/pulse, respectively.

Then the laser dye solution was put into an injector so that it could become a free droplet, as shown in the illustration of Figure 5(*a*). The OMA was able to detect the random lasing when the energy of the pump light was above 5 μJ/pulse. Light spots (Figure 5) and center wavelength (Figure 6) change as pumping time increased, because evaporation of the solvent (C_4H_8O) changed the size of the free droplets. This means that the random lasing occurs without the external structure. The micro-droplets or particles are absent in DDLCs (PM597 and E7) or laser dye solution, as observed by means of

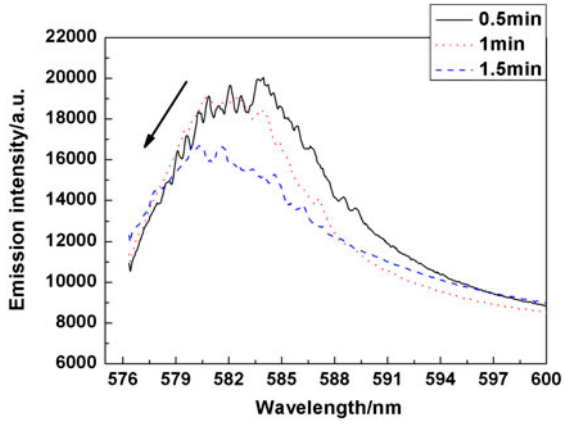

Figure 6. Time dependence of the lasing spectra in free droplet of DDLCs. (The color version of this figure is included in the online version of the journal.)

an optical microscope. However, there are some particles with sizes of about 610 nm in the the laser dye solution (PM597), as revealed in a density instrument (Marve). Thus, we deduce that it is nanoparticles consisting of dye molecules that provide a new scattering mechanism, and hence the random lasing is still obtained in both isotropic phase and laser dye solution. The random lasing was still obtained in different solvents, such as alcohol and acetone. A similar experimental result can be obtained in DCM:C_4H_8O solution.

4. Conclusion

This paper has reported the temperature dependence of the intensity, FWHM, center wavelength, and polarization of random lasing in DDLCs in a cell. The experimental result shows that the intensity of random lasing gradually reduces when the temperature increases from 23.6°C to 44°C, and it fades to zero at a temperature of 44°C. However, it reappears at the temperature approaches the nematic–isotropic (N-I) transition because

of micro-droplet nucleation and critical opalescence. Surprisingly, it is found that the random lasing is still obtained when the cell temperature becomes higher, even above the N-I transition (i.e. above 61°C). Temperature has little effect on the FWHM of the random lasing. The center wavelength of the random lasing shifts from 575.69 nm to 593.43 nm when the temperature increases from 25.5°C to 148°C. The study also shows that the polarization of the random lasing depends on the rubbing direction and the polarization of pump light in nematic phase and in isotropic phase (or proximity to N-I transition), respectively. Furthermore, it was found that the random lasing occurs in a laser dye solution consisting of PM597 and C_4H_8O in five different geometries: glass cell (185 μm), glass bottle, plastic test tube, cuvette, and free droplet. The reasons are possibly that nanoparticles consisting of dye molecules provide a new scattering mechanism in both solution and isotropic phases.

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