Research article

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The controllable intensity and polarization degree of random laser from sheared dye-doped polymer-dispersed liquid crystal

Abstract: The random laser from sheared dye-doped polymer-dispersed liquid crystal (DDPDLC) is investigated. As the emission intensity weakens, the threshold of random laser from DDPDLC increases from 2.0 mJ/pulse to 4.0 mJ/pulse, and the degree of polarization (DOP) increases from 0.1 to 0.78, obviously when the shear distance increases from 0 mm to 4 mm. As the liquid crystal droplets are gradually oriented in the shear direction caused by alignment direction of polymer chain and anisotropy of droplet shape, the scattering intensity perpendicular to the shear direction gradually decreases and that parallel to the shear direction gradually increases. The anisotropic absorption of the laser dye also plays a certain role as the shear distance is 0 mm. The controllable intensity and polarization degree of random laser have a huge potential for sensing applications.

Keywords: polarization degree; intensity; shear; controllable; random laser.

1 Introduction

Random laser is a stimulated emission phenomenon observed in disordered media, such as nanoparticles [1, 2], biomaterials [3], quantum cascade gain medium [4], semiconductors [5, 6], and liquid crystals [7–9]. The feedback of random laser is realized by multiple scattering of light. The advantages of technological simplicity and low fabrication cost [10] make the random laser have important research significance and application value in photonics, bio-medicine, sensing, and other fields. The optical anisotropy of the liquid crystal makes it unique in terms of laser [9, 11–13], and there are some ways to regulate the liquid crystal, such as applying an electric field [14, 15] or incorporating colloidal mixed silica nanoparticles [16].

Polarized light is a light beam in which the vibration direction of the light vector does not change or has a specific regular change, with important application value in the field of display application, polarization imaging, and medical treatment. The linear polarization of random laser has been realized in liquid-crystal-based media [17] and in suspensions of scattering nanoparticles source [18]. The polarized character of random lasing in dye-doped nematic liquid crystals has been addressed [18, 19]. The DOP can be adjusted to some extent by varying the material composition of the laser [20] or by adjusting the pump light [21]. In 2015, Zhai et al. [22] controlled the DOP of random laser by stretching silver nanowires embedded in a flexible substrate, providing the idea that random laser can be controlled by applying external force. The DOP of liquid-crystal-based laser has a wide adjustment range due to the huge optical anisotropy of liquid crystals. It has important research significance but has been rarely reported.

In this paper, the intensity and polarization of random laser emitted from sheared DDPDLC have been investigated. The shear force is applied on the DDPDLCs to form different shear distances. As the shear distance increases, the emission intensity and DOP of random laser change because of the orientation of liquid crystals caused by shear force.

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2 Sample preparation and experimental setup

2.1 Sample preparation

The materials used to fabricate the dye-doped PDLC consisted of 44.41 wt% of monomer (trimethylolpropane triacrylate) (Aladdin Industrial Corporation, Shanghai, China), 7.64 wt% of cross-linking monomer(N-vinylpyrrollidone) (Aladdin Industrial Corporation, Shanghai, China), 0.54 wt% of photo-initiator(rose bengal) (Aladdin Industrial Corporation, Shanghai, China), 1.02 wt% of coinitiator(N-phenylglycine) (Aladdin Industrial Corporation, Shanghai, China), 8.44 wt% of surfactant(octanoic acid) (Aladdin Industrial Corporation, Shanghai, China), and 1.36 wt% of lasing dye (PM597) (Moriizumi Optoelectronics Technology Co. Ltd., Shanghai, China) and 35.3 wt% of nematic liquid crystal (E7, $n_e = 1.7462$ and $n_o = 1.5216$) (Chengzhi Yonghua Display Company, Shijiazhuang, Hebei, China). All the materials were stirred in water bath heating at 65°C (the clearing point of the E7 is about 60°C) for 2 h to form a homogeneous mixture in the dark [23]. Then the mixture was injected into the empty cells by capillary action to form the liquid crystal cells. During this process, the cells were placed on a 65°C platform. The empty cell was formed by two pieces of ITO-coated glasses and had a thickness of 25 µm controlled by Mylar spacers. The ITO glass had no surface treatment. To polymerize the cells, a two-step UV curing process was adopted. In the first step, the LC cell was illuminated to a UV light ($\lambda \sim 365$ nm, I = 50 mW/cm²) (Spectronics, NY, USA) for 30 s at T = 90°C, which is higher than the clearing temperature of E7. The shear force was applied by a specific device to make the shear distances 0 mm, 1 mm, 2 mm, 3 mm, and 4 mm, respectively. In the second step, the cell was cured at 35°C for 5 min.

At initial state, the liquid crystal droplets in DDPDLC were randomly distributed, as shown in Figure 1B, then the shear force was applied by a specific device to make the liquid crystal droplets oriented in the shear direction,
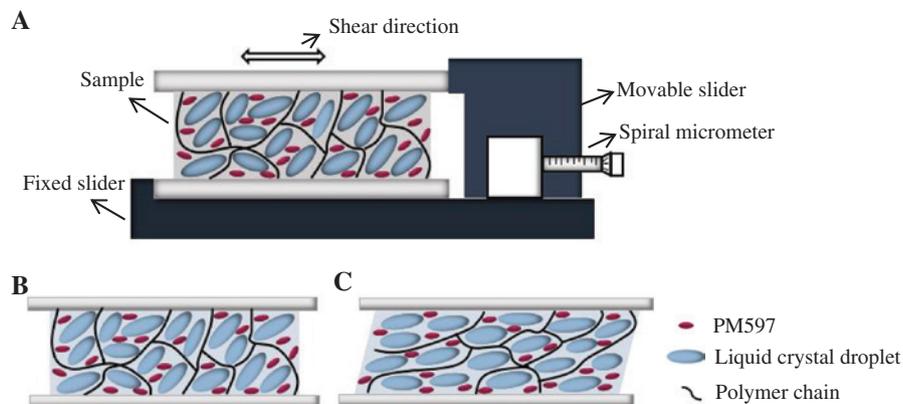
Figure 1: The schematic diagrams of the shearing device and internal structure of the sample.
(A) The device for applying shear force to DDPDLC. (B) The internal structure of the sample before being sheared. (C) The internal structure of the sample after being sheared.
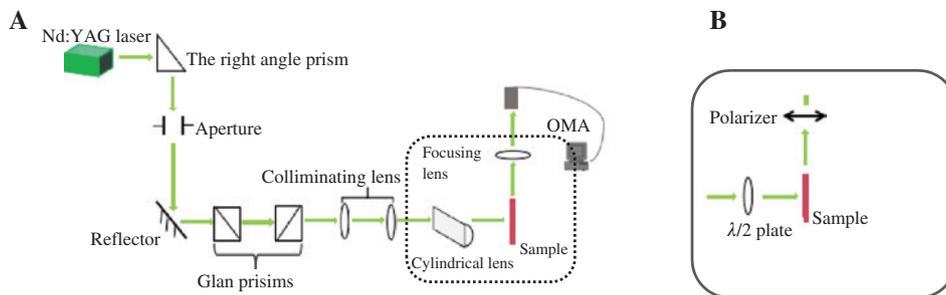
Figure 2: The sketch of the experimental setup.
(A) The group of Glan prisms is used to adjust the pump energy. The two lens are used to collimate the pump beam. (B) The half wave plate is used to make sure the polarization direction of the pump is parallel to the shear direction. The polarizer is used to detect the random laser polarization direction.

as shown in Figure 1C. The structure of specific device is shown in Figure 1A. The sheared direction is parallel to the long side of the DDPDLC and is set to be parallel (0°) unless otherwise stated.

2.2 Experimental setup

The experimental setups for the random laser are shown in Figure 2. The samples were optically pumped by a frequency-doubled Nd:YAG laser system (532 nm, 10-Hz repetition rate, 8-ns pulse duration, PowerLite Precision II 8010) (Continuum, Boston, MA, USA). The pump energy could be adjusted through a group of Glan prisms. The pump beam was collimated by two lens with different focal lengths (7.5 cm and 5 cm) and was focused onto the sample by cylindrical lens, which were aligned to shape a laser pump stripe, as shown in Figure 2A. The length and width of pump strips are 8 mm and 0.2 mm, respectively. In the part of the study on the influence of shear force on the polarization of random laser, the component surrounded by the dotted lines in Figure 2A would be replaced by Figure 2B. A half wave plate was placed in front of the cylindrical lens, used to make sure the polarization direction of the pump is parallel to the shear direction. A polarizer was placed behind the focusing lens, used to detect the random laser polarization direction. The emitting signals were recorded by Optical Multichannel Analyzer (OMA) with spectral resolution of 0.1 nm (Acton Research Corporation Company, CA, USA).

3 Results and discussion

The emission intensities of the random laser from DDPDLCs with different shear distances were investigated.

Figure 3A depicts the dependence of emission integrated intensity on pump energy for the DDPDLCs whose shear distances are 0 mm, 1 mm, 2 mm, 3 mm, and 4 mm, respectively. As the shear distance increases, the intensity of the random laser decreases and the threshold increases from 2.0 mJ/pulse to 4.0 mJ/pulse, obviously. Such behavior is associated with orientation of the liquid crystal molecules caused by alignment direction of polymer chain and anisotropy of droplet shape in the sheared DDPDLC [24].

The multiple scattering in the DDPDLC can provide optical feedback for random laser [25], which mainly depends on the refractive index difference between the liquid crystal and the polymer. The scattering intensity is proportional to the refractive index difference. In the case of the DDPDLC, the refractive index $<n>$ of the liquid crystal droplets is expressed as Equation (1) [23]:

$$\frac{1}{<n>} = \left[\frac{\cos^2 \theta}{n_e^2} + \frac{\sin^2 \theta}{n_o^2} \right]^{\frac{1}{2}} \tag{1}$$

The variable θ is the angle between the liquid crystal director direction and the pump light polarization direction. As the orientations of the liquid crystal droplets are randomly distributed, the refractive index difference $\Delta n = <n> - n_p$ is a random value between $0.03(n_o - n_p)$ and $0.26(n_e - n_p)$; n_p is the refractive index of polymer (~1.50). As the shear distance increases, alignment of liquid crystal droplets is gradually parallel to the sheared direction [26]. The Δn value decreases to nearly $0.03(n_o - n_p)$ in the perpendicular direction and increases to nearly $0.26(n_e - n_p)$ in the parallel direction. The magnitude of the decrease is greater than the increase, resulting in the reduction of the total scatting intensity, so as the emission intensity. In order to confirm that the total scattering intensity is
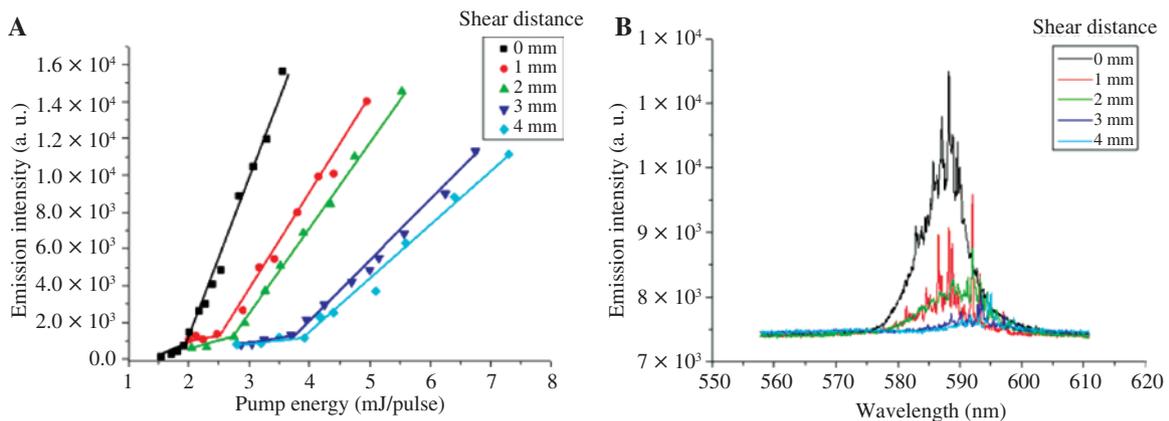
Figure 3: Emission spectra with different shear distances.
(A) The dependence of emission integrated intensity on pump energy for the DDPDLCs with different shear distance. (B) Evolution of emission spectra as a function of shear distance.

indeed weakened, the spectrum is analyzed. Transmission mean free path L_t is expressed as [27]:

$$L_t = \frac{\lambda^2}{2n\Delta\lambda} \qquad (2)$$

The variable λ is wavelength of the main peak of emission spectra, n is the refractive index of DDPDLC, and $\Delta\lambda$ is the peak separation. Figure 3B shows the emission spectra of DDPDLCs with different shear distances under the same pump energy. Obviously, the random laser emitted from DDPDLCs is coherent, and the peak separations are almost unchanged. Therefore, DDPDLCs with different shear distances are considered to have the same n and $\Delta\lambda$. According to Equation (2), L_t is proportional to λ. As shown in Figure 3B, the wavelength of the main peak is red-shifted, as the shear distance increases, which means L_t increases. As the scattering intensity is inversely proportional to L_t [28], the scattering intensity is reduced, resulting in the decrease of emission intensity as the distance increases.

In the part of the study on the influence of shear force on the polarization of random laser, the polarization direction of the pump light is always parallel to the shear direction of the cells. The polarizer is rotated to change the angle (φ) between the polarization direction of the emitted random laser and the shearing direction. The relationship between the intensity and the angle (φ) is presented in Figure 4A. The random laser intensity emitted from the cells decreases, as the φ increases from 0° to 90° and increases as the φ increases from 90° to 180°.

The linear polarization degree of random laser can be calculated according to the expression [21]:

Table 1: The DOP of random laser from DDPDLCs with different shear distances.
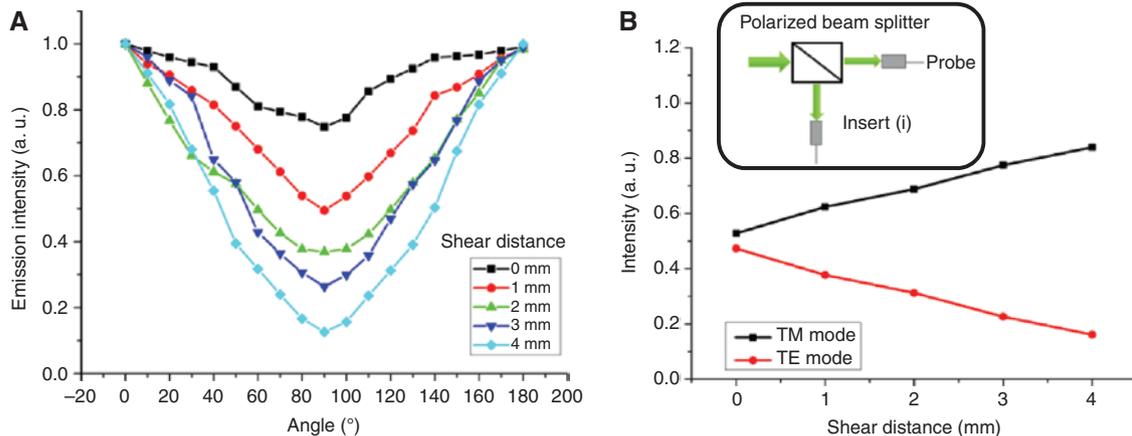
Shear distance	0	1	2	3	4
DOP	0.1	0.34	0.47	0.58	0.78

$$DOP = \frac{I_\parallel - I_\perp}{I_\parallel + I_\perp} \qquad (3)$$

where $I_\parallel$ and $I_\perp$ represent the parallel polarization component and perpendicular polarization component of the lasing emission, respectively. According to formula (3), it is obvious that the DOP increases gradually as the shear distance increases, as shown in Table 1. It illustrates that the DOP of the random laser from DDPDLCs can be controlled by applying a shearing force.

On the one hand, as the liquid crystal droplets are gradually oriented in the shear direction, the scattering intensity of ordinary light perpendicular to the shear direction is gradually reduced, and the scattering intensity of extraordinary light parallel to the shear direction is gradually enhanced. In other words, as the shear distance increases, the proportion of TE modes in the emitted random laser becomes smaller and smaller, and the proportion of TM mode becomes larger and larger, as shown in Figure 4B. The insert (i) shows the sketch for measuring TM and TE mode beams, and the emitted random laser is divided by a polarized beam splitter.

On the other hand, as the PM597 dye is a rod-like dye, and there is a host-guest effect between the nematic liquid crystal molecules and the dye molecules, the dye molecules tend to be oriented along the shear direction as the shear

Figure 4: The relationship between emission intensity and shear distance. (A) The normalized Intensity of random laser depends on the rotation angle φ. (B) The dependence of proportion of TM mode and TE mode on shear distance. Insert (i). The sketch for measuring TM and TE mode beams.

distance increases. As the absorption efficiency of the dye for pump light with polarization direction parallel to the transition dipole moment of dye is greater than pump light with other polarization directions, more random laser is emitted whose polarization direction is parallel to the shear direction. When the shear distance is 0 mm, the DOP is 0.1. The direction of dye's transition dipole moments is randomly distributed, thus only a small fraction of the overall emission could be polarimetrically evaluated [18].

The two points above explain why the DOP of the emitted random laser can be controlled by applying shear force.

4 Conclusion

In conclusion, the controllable intensity and polarization degree of random laser from DDPDLC were investigated. We find that the emission intensity of emitted random laser from DDPDLC weakens and the DOP increases with the increase of shear distance. As the liquid crystal droplets are gradually oriented in the shear direction, the scattering intensity is gradually reduced in perpendicular direction. The controllable intensity and polarization degree of random laser by shearing the DDPDLC have prospects of wide range of applications, such as optical sensors as a laser light source [29] and liquid crystal display [18].

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