Research article

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Waveguided nematic liquid crystal random lasers

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Abstract: In waveguided nematic liquid crystal random lasers (NLCRLs), we realize polarized random laser (RL) emission and discover that the waveguide effect reduces the transmission loss of the RL whose polarization is parallel to the liquid crystal molecules (LCMs). Compared with the traditional liquid crystal random lasers, the waveguide NLCRLs can achieve the regulation of RLs strength, polarization, and wavelength in the same structure. The electric field can drive the rotation of LCMs to control the RL polarization and intensity. The drop of horizontal polarization laser and the increase of vertical polarization laser prove the role of the waveguide effect. In addition, the disorder of the waveguided NLCRLs is highly sensitive to temperature, which makes it easy to control the wavelength and intensity of the RL. As the temperature rises, the

waveguide effect is weakened, resulting in a weakening of the restriction along liquid crystal (LC) cell normal direction. The reduced laser intensity verifies the role of the waveguide effect.

Keywords: disorder; nematicliquid crystal; polarization directions; random laser; waveguide effect.

Random lasers (RLs), which result from the multiple scattering of light inside a disordered optical gain medium [1], have received wide attention since it was theoretically predicted [2] and experimentally demonstrated [3]. Compared with traditional lasers, RLs have unique optical characteristics, including multiple emission wavelengths, large-angle output, and easy production. Because of these characteristics, RLs have been used in sensors, lighting, and medical diagnosis [4–6]. Liquid crystal random lasers (LCRLs) are based on using liquid crystal (LC) as a multiple scattering medium. The random lasing wavelength and intensity can be controlled by liquid crystal molecules (LCMs) arrangement and LC phase through applying external conditions, e.g., temperature, low-frequency electric field, and magnetic field [7–9]. Various geometric structures have been applied in LCRLs including wedge cells [8] capillary [10] and photonic crystal fibers [11]. Sensitive responses to external conditions and a variety of geometric structures provide a large range of application prospects for LCRLs.

Nematic liquid crystals (NLCs) are fluid dielectrics with the long axis of the molecules in the same direction. The average direction (n) (the long axis orientation of rod-shaped molecules) is regarded as the LC optical axis [12]. In the uniaxial crystal NLCs, an electromagnetic plane wave can be described as the superposition of ordinarily (o-) and extraordinarily (e-) polarized wave components [13]. For these two types of polarized waves, LCs have different refractive indices. Therefore, the LCs can provide strong scattering. In 2004, Stefano Gottardo et al. [14] observed the emission from dye-doped polymer-dispersed liquid crystals (PDLCs) is anisotropic and extraordinarily polarized, which is controlled via an external electric field. Shumin Xiao et al. fabricated a PDLC film. A 9.2 V/ μm external electric field was applied to control the linewidth, intensity, and polarization of the RLs [1]. Their common

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tunable feature is to change the effective refractive index by controlling the LC molecular orientation. In 2007, Shtykov et al. [15] obtained amplifying lasing in a planarly aligned LC layer doped with a lasing dye. This kind of planarly aligned LC layer can be pumped by a continuous-wave beam to form optical spatial solitons to assist RL emission [16–19]. In traditional LCRLs, the waveguide effect of the LCs was ignored. Waveguided nematic liquid crystal random lasers (NLCRLs) provide a convenient structure to analyze the role of the waveguide effect in RLs transmission. And the RLs characteristics can be controlled in waveguided NLCRLs through temperature and electric field.

In this paper, we fabricated waveguided NLCRLs with uniform planar orientation. Adjusting the polarization of the pump, it was found that the o-wave pumping has a lower laser threshold than the e-wave pumping. The laser generated under arbitrary polarization pumping is horizontally polarized due to the anisotropic absorption of the laser dye and the waveguide effect. Under fixed o-wave pumping, the main plane of the LC rotates under voltage, which causes the observed laser to be changed from horizontal polarization to vertical polarization. In addition, during the process of heating the sample from room temperature to the clearing point (CP), the laser intensity first increased, and then rapidly decreased, until it disappeared completely at 42 °C. The corresponding laser peak first blue-shifted 0.49 nm from 572.86 nm (26 °C) and then continued to red-shifted to 577.24 nm (40 °C). The total red-shift of the laser peak is 4.87 nm in the range of 12 °C.

An LC cell with a thickness of 125 μm was prepared. The indium tin oxide layer with high reflectivity ($r \geq 86\%$) is covered with polyimide (PI) film about 2–3 μm thick. The orientation of the LC molecules n (LC optical axis, in the sample plane and form an angle of 45° with the Z-axis) is

formed by rubbing PI film. The main plane of the LC is the horizontal plane where the LC optical axis is located. The NLC host E7 ($n_o = 1.517$, $n_e = 1.741$) mixed with 0.3 wt% pyrromethene 597 (PM597) guest dye. The LC bulk phase sequence is Crystal – (–40 °C) Nematic – (59 °C) Isotropic. Figure 1(a) shows the LC cell configuration and orientation.

In our experiment, the pumping source is a Q-switched Nd:YAG laser which outputs a wavelength of 532 nm with a round spot (pulse duration 10 ns, repetition rate 10 Hz, spot diameter 30 μm). The pump pulse energy and polarization are controlled by a Glan Prism group. To form a waveguide channel for the propagation of RLs in the cell plane, RLs were excited by a laser beam which was focused in the LC layer by a 5 $\times$ objective lens. The numerical aperture of the microscope objective lens we used in the experiment is 0.13, which is a weak focus system. The polarization transverse component can be ignored [20]. Figure 1(b) shows the light-emitting area captured by the charge coupled device (CCD) camera. The emitted light is converged by a focusing lens and collected by a fiber spectrometer (QE65PRO, ocean optics, resolution ~ 0.4 nm, integration time 100 ms). The temperature is controlled by the heating platform (HCS302-01, INSTEC, USA, resolution 0.01 °C). The waveguide structure can only limit the random walk in the x-direction (depends on the angle of total internal reflection [TIR]), and the divergence in the y-direction cannot be limited. In the experiment, we converged the light through the focusing lens group at the back end of the sample, which alleviated the leakage problem along the Y direction. The overall experimental device is shown in Figure 1(c).

In waveguided NLCRLs, TIR exists between the LC layer and the PI film layer. According to the conditions of TIR, the angle of TIR (θ_c):

Figure 1: (a) Cell configuration and LCMs orientation; (b) Beam transmission path image; (c) Experimental setup of waveguided NLCRLs laser experiment, microscope objectives (MO), attenuator (A), beam splitters (BS), energy meter (EM), polarizer (P), and spectrometer (SM).

Figure 2: The integrated intensity of the RLs under o-wave pumping (red dot) and e-wave pumping (black dot) as a function of pump energy (a) and polarization direction (b, θ is the angle between polarization direction and the horizontal plane), and random lasers (RLs) emission spectra at different pump energy for o-wave pumping (c) and e-wave pumping (d).

$$\theta_c = \arcsin \frac{n_2}{n_{\text{eff}}} \quad (1)$$

where n_{eff} is the effective refractive index of LC, $n_2 = 1.508$ is the refractive index of the PI film. When the angle between the direction of light propagation and the normal direction of the glass substrate is greater than θ_c , the total reflection will occur, otherwise, it will leak from the X direction. Therefore, the strength of the waveguide effect is positively correlated with Δn (the refractive index difference between the effective refractive index of the LC and the refractive index of the PI film). The LC effective refractive index n_{eff} can be described by the following formula [1]:

$$\frac{1}{n_{\text{eff}}^2} = \frac{\cos^2 \theta}{n_e^2} + \frac{\sin^2 \theta}{n_o^2} \quad (2)$$

where θ is the angle between the main plane of the LC (YZ -plane) and the polarization direction of the pump.

Figure 2(a) shows the integration intensity of RLs under the different pumping orientations. The random lasing threshold produced by o-wave pumping (polarization along the X -axis) is significantly lower than that of e-wave pumping (polarization along the Y -axis). The thresholds are 4.06 and 5.43 μJ , respectively. The effect is mainly due to polarization dependence of the diffusion constant D for the emitted photons, and indeed a larger diffusion constant for the e-wave compared to the o-wave is reported [8]. As the diffusion constant decreases, thereby increasing scattering strength, and, in turn, random lasing can be enhanced. As a result, under the same pump energy, the intensity of the RLs generated by the o-wave pumping is

stronger than that of the e-wave pumping. This is consistent with previous results [16, 17].

Due to the guest-host effect [21–23], most dye molecules are aligned along the optical axis of the LC but there are some exceptions. Because of the anisotropic absorption of the laser dye, RLs with multiple polarization directions are present in the sample. For horizontally polarized RLs ($n_{\text{eff}} = n_e$), Δn is 0.233 ($n_e - n_2$), $\theta_c \approx 60^\circ$. But for vertically polarized RLs ($n_{\text{eff}} = n_o$), Δn is 0.009 ($n_o - n_2$), $\theta_c \approx 83.6^\circ$. A sufficiently long transmission path allows the waveguide to select the polarization RL that satisfies TIR conditions. Finally, a horizontally polarized RL (the degree of polarization is 1) is received at the back of the sample (Figure S1 in the Supplementary material). As shown in Figure 2(b), the random lasing emission is horizontal polarization with whether an o-wave or an e-wave pump. Figure 2(c) and (d) show the emission spectra of RLs under the o-wave pump and e-wave pump, respectively. It can be seen that e-wave produces fewer laser modes than o-wave pumps. It is because the lower intensity modes for e-wave pumping are lost during transmission.

In order to explore the influence of the waveguide effect on the RL polarization and intensity, the sinusoidal AC voltage (20 kHz, 24 V) is applied along the X -axis. Other voltage conditions are shown in Figures S2–S5 (see in the Supplementary material). Because o-wave pumping has a lower pumping threshold, the subsequent experiments are carried out under o-wave pumping. After a period of response time, the LCMs turn to be perpendicular to the horizontal plane. Figure 3(a) shows the change of RLs after

applying voltage under o-wave pumping. As the LCMs rotated under-voltage drive, due to the guest-host effect, the dye molecules are arranged along the optical axis of the LC. Based on *Fermi's Golden Rule* the molecular transitions and the rate of emission strongly depend on the coupling of the pump electric field E and the transition dipole moment d of the dye molecules [8]. After the voltage is applied, the coupling of the optical field with the dye molecules becomes stronger, resulting in an increase in the total RL intensity (Figure S6 in the Supplementary material).

The vertical polarization (Figure 3b) and horizontal polarization (Figure 3c) RLs emission are obtained at a voltage of 0 and 24 V. With the increase of the voltage, the vertical polarization RL enhance more than five times, and the horizontal polarization RL is reduced by 97%. The guest-host effect is one of the influencing factors. However, due to the high anchoring energy of friction orientation, a small amount of horizontal polarization can still be generated. At this time, the refractive index of the LC to the vertical polarization RLs is n_e . The $\Delta n = n_e - n_2$ is increased to 0.233, the waveguide effect is increased and the loss is reduced. And the refractive index of the LC to the horizontal polarization RLs is n_o . The $\Delta n = n_o - n_2$ is reduced to 0.009, the waveguide effect is reduced and the loss is increased. Until the vertical polarization dominates. The waveguide effect helps to increase the degree of polarization.

In the waveguided NLCRLs, the waveguide effect is affected by temperature. Adjusting the sample temperature from room temperature (26 °C) to the nematic–isotropic (N–I) transition point. Some interesting phenomena of RLs

have been observed. The expressions of n_e and n_o varying with temperature are as follows [24]:

$$n_e(T) \approx A - BT + \frac{2(\Delta n)_o}{3} \left(1 - \frac{T}{T_c}\right)^\beta \quad (3)$$

$$n_o(T) \approx A - BT - \frac{(\Delta n)_o}{3} \left(1 - \frac{T}{T_c}\right)^\beta \quad (4)$$

where A and B are constants greater than 0, $(\Delta n)_o$ is the birefringence when Kelvin temperature $T = 0$ K, β is the material constant and T_c is the CP temperature of the LC. And n_e decreases as the temperature increases, and n_o is the opposite. The refractive index of PI decreases in temperature at a rate of $10^{-4}/^\circ\text{C}$, which is one order of magnitude larger than that of inorganic glasses [25, 26]. When the temperature is lower than the CP of the LC, the refractive index of PI film is regarded as a constant n_2 . Under o-wave pumping, the $\Delta n(T) = n_o(T) - n_2$ is a temperature-dependent function.

Figure 4(a) shows the integration intensity of the RLs spectrum at different temperatures. Each point is the averaged integrated intensity of 50 spectra in the range of 500–600 nm. Initially, when the temperature increases from 26 to 28 °C, the intensity of the RL increases. Within the initial range of 2 °C, the small change in refractive index has little effect on the waveguide effect. However, the thermal motion of the molecules enhances disorder and scattering. After 28 °C, the intensity of RLs gradually decreases with the increase of temperature and disappears at 42 °C. There are two reasons for this result. On the one hand, the RLs in the experiment are horizontal

Figure 3: (a) Schematic diagram of LC molecular arrangement; Vertically (b) and horizontally (c) polarized random lasers (RLs) spectra at voltages of 0 V (black line) and 24 V (red line).

Figure 4: RLs integrated intensity (a) and peak (b) at different temperatures.

Figure 5: (a) Random lasing spectra at 26 and 30 $^{\circ}\text{C}$; (b) Fourier transform of the left spectra.

polarization, as shown in Figure 2(b). At this time, $\Delta n(T) = n_e(T) - n_o$, and the waveguide effect decreases with increasing temperature which results in a weakening of the restriction along the X-axis. On the other hand, the anisotropy of LC decreases with increasing temperature, resulting in the disorder and scattering decreases, and the laser threshold increases. RLs cannot be generated after the N-I phase transition point (59 $^{\circ}\text{C}$). This result is different from previous studies [27, 28].

Figure 4(b) shows the peak position of the RLs at 26–40 $^{\circ}\text{C}$. Each point is the averaged peak position of 50 spectra at each temperature. Within 2 $^{\circ}\text{C}$ of the increase from room temperature, the peak position of the RLs blue-shifts from 572.86 to 572.37 nm. However, from 28 to 40 $^{\circ}\text{C}$, the red-shifts continually until 577.24 nm. Within the temperature range of 12 $^{\circ}\text{C}$, the wavelength shifts by 4.87 nm.

Through Fourier transform, we analyzed two spectra with a central wavelength of 571.80 nm at 26 $^{\circ}\text{C}$ and 573.40 nm at 30 $^{\circ}\text{C}$, as shown in Figure 5(a). The power Fourier transform of the emission spectrum from a well-defined laser cavity yields peaks at Fourier components [29]:

$$p_m = \frac{mnl_c}{\pi} \quad (5)$$

where m is the Fourier resonance ordinal number, n is the effective refractive index of the LC, and l_c is the cavity path

length [30]. Because the RLs are horizontal polarization, $n = n_e$. As shown in Figure 5(b), the laser cavity yields peaks at Fourier components $p_{m1} = 18.68 \mu\text{m}$ and $p_{m2} = 33.96 \mu\text{m}$, respectively. Take $m = 1$, and get $l_{c1} = 33.69 \mu\text{m}$, $l_{c2} = 61.25 \mu\text{m}$. It is obvious that the cavity path length increases ($l_{c1} < l_{c2}$) with the increase of temperature, this shows that the disorder of the sample is reduced.

In the scattering system, the scattering mean free path (l_s) is affected by the disorder. l_s increases ($l_{s1} < l_{s2}$) with the decrease of disorder. Larger l_s will reduce the number of light scattering in the LC, and reduce the path of photon walking between LC molecules. Moreover, the scattering mean free path of long wavelength is larger than that of short wavelength, and long wavelength produces greater gain than the short wavelength, resulting in the red-shift of emission wavelength. At 28 $^{\circ}\text{C}$, with the abnormal increase of laser intensity, the blue-shift of the peak position is observed. This is because when the sample is just heated, the thermal movement of molecules leads to the enhancement of the internal disorder of the sample, which increases the scattering intensity. As a result, the scattering mean free path decreases and the gain of short wavelength increases, resulting in a blue-shift of emission wavelength.

Conclusion: To summarize, we have reported the polarization-dependent and external controllable characteristics of waveguide-assisted multi-variable controllable NLCRLs, and obtained RLs with high polarization. With the

increase of the voltage, the horizontal polarization RLs is gradually converted into vertical polarization. This result provides a new way for RL polarization control. In addition, the disorder of the NLCRLs is highly sensitive to temperature, which makes it easy to control the wavelength and intensity of the RL. We envision that the waveguided NLCRLs may open a window in sensors, wavelength tunability, and polarization control.

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