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Huanting Bian[a], Fengfeng Yao[a], Hai Liu[c], Feng Huang[a], Yanbo Pei[ab], Chunfeng Hou[ab] & Xiudong Sun[ab]

[a] Department of Physics, Harbin Institute of Technology, Harbin, China

[b] Key Laboratory of Micro-Optics and Photonic Technology of Heilongjiang Province, Harbin, China

[c] Key Laboratory of Optical System Advanced Manufacturing Technology, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun, China

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Optically controlled random lasing based on photothermal effect in dye-doped nematic liquid crystals

Huanting Bian[a], Fengfeng Yao[a]*, Hai Liu[c], Feng Huang[a], Yanbo Pei[a,b], Chunfeng Hou[a,b] and Xiudong Sun[a,b]

[a]*Department of Physics, Harbin Institute of Technology, Harbin, China;* [b]*Key Laboratory of Micro-Optics and Photonic Technology of Heilongjiang Province, Harbin, China;* [c]*Key Laboratory of Optical System Advanced Manufacturing Technology, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun, China*

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We presented a way to control the output of the nematic liquid crystal (NLC) random laser with a response time of hundreds of milliseconds by a continuous-wave laser beam with the power of several milliwatts. The lasing intensity can be tuned continuously by variation of the control power in the range from 0 to 5 mW or the lasing output can be switched on and off by turning off and on the control beam when the control power reaches 6 mW. The control power-dependent lasing polarisation and the self-phase modulation of the control beam indicate that the photothermal effect, which leads to the change of the order of the NLC and subsequently the change of the long-range fluctuation of the dielectric tensor, is mainly responsible for the controllability of the random lasing.

Keywords: nematic liquid crystals; random lasers; photothermal effect; multiple scattering

1. Introduction

Random lasers (RLs) are disordered media with gain that do not possess a light-trapping cavity beyond the confinement provided by multiple scattering from the disorder itself.[1] For random lasing in dye-doped nematic liquid crystals (NLCs), the interference effect in recurrent multiple scattering by the long-range dielectric tensor fluctuation of the optically anisotropic NLCs provides the required optical feedback for lasing.[2] Because the orientation of NLCs and thus the dielectric properties can be externally controlled, NLC RLs can be developed into lasers with controllable features,[3,4] which is the peculiar merit of NLC RLs over all other types of observed RLs mainly including semiconductor powers,[5] piconjugated polymers,[6] scattering suspension in dyes,[7] random microcavities,[8] and random fibre lasers.[9] Flexible controllability is anticipated for the application of RLs in photonics and optically controlled RLs are especially luring. There are a variety of mechanisms through which the dielectric properties of NLCs can be manipulated optically,[10] which may be utilised to control random lasing in NLCs. However, the research concentrated on this aspect of NLC RLs is scarce. In the report by Lee et al.,[4] photoisomerisable dye (azo dye) was added into the dye-doped NLC, and this optically reversible controllability of the NLC RL is attributed to the UV-beam-induced isothermal nematic→isotropic (N→I) and green-beam-induced isothermal I→N phase transitions of the NLC by trans→cis and cis→trans back isomerisations of the azo dye, respectively, subsequently decreasing and increasing the spatial non-uniformity of the NLC orientational order, respectively. Thus, the NLC RL can be controlled with a two-step exposure of one UV and then one green beam, and the response time is in the order of tens of seconds.

In this article, we present a way to control random lasing in NLCs by a weak He-Ne laser beam at the power level of several milliwatts. With the variation of the intensity of the control light, the continuous reversible tuning of the output RL intensity was realised. The complete on–off state transition of lasing also operated by simply turning off and on the control light with certain intensity. The dependence of the lasing polarisation on the control beam and the dependence of the control effect on the polarisation of the control beam inferred that the optical control over the NLC RL was realised through controlling the NLC order and thus controlling the spatial fluctuation of the dielectric tensor $[\delta\varepsilon = \varepsilon(\boldsymbol{r} + \delta\boldsymbol{r}) - \varepsilon(\boldsymbol{r})]$ based on the photothermal effect. And this explanation was supported by the analysis of the self-phase modulation (SPM) of the control beam that coexisted with the controlling process. The optically controlling method based on the photothermal effect has several advantages than that based on the photoisomerisable process. Firstly, it has a response time less than 1 second while the response time based on the photoisomerisable process takes several tens of seconds; secondly, doping photoisomerisable dye is avoided; third, the

*Corresponding author. Email: fengfyao@hit.edu.cn

optical set-up is simplified since it does not need two wavelengths beams to excite trans→cis and cis→trans isomerisations of the azo dye.

2. Materials and optical set-up

In our experiment, the NLC used was E7 (provided by SLICHEM, China) and the dye was Pyrromethene 650 (Exciton), which was doped into the NLC E7 with a concentration of 0.3% by weight. The cell was fabricated with two indium tin oxide (ITO)-coated glass slides separated by two 100 μm-thick mylar spacers, in which inner surfaces of ITO glass substrates were covered with rubbed polyvinyl alcohol. The cells were filled up with the dye-doped NLC through the capillarity effect to form homogeneous alignment of the NLC molecules with the nematic director parallel to the rubbing direction in the plane of the cell.

Figure 1(a) shows the experimental set-up. A Q-switched Nd:YAG second-harmonic generation pulse laser (532 nm, 6 ns, 1 Hz) was focused onto the sample using a lens (L, f = 8.89 cm). The pump pulse was set to be p-polarised, and the pump pulse energy was set to a fixed value to induce a random lasing emission. The control beam, which is the output of a linearly polarised, continuous-wave (CW) He-Ne laser (λ = 632.8 nm), was also focused by a lens with a focal length of 3.5 cm, and then used to irradiate the sample in an oblique incident direction (at an incident angle of ~30°). Any intensity and polarisation direction of the He-Ne laser beam can be achieved by using an attenuator (A), a polariser (P) and a half-wave plate (λ/2), to control the RL emission. The two incident laser spots were superposed on the sample. The output intensity and polarisation of the RL emission were measured by two high-speed photodetectors D_1 and D_2 (DET 10 A/M, from Thorlabs), which detected the reflected intensity and transmitted intensity through an analyser (P) of the RL after a non-polarised beam splitter (NBS). Two lenses were used to collect the RL beam to the detectors and a bandpass filter F (central wavelength: 630 nm, bandwidth: 30 nm) was used to block the scattering light of the pump beam for the RL measurements.

3. Results and discussions

Firstly, the emission spectra of the NLC RL were measured by an Ocean Optics USB2000 + spectrometer (range: 350–1100 nm, resolution: 1.5 nm) with the

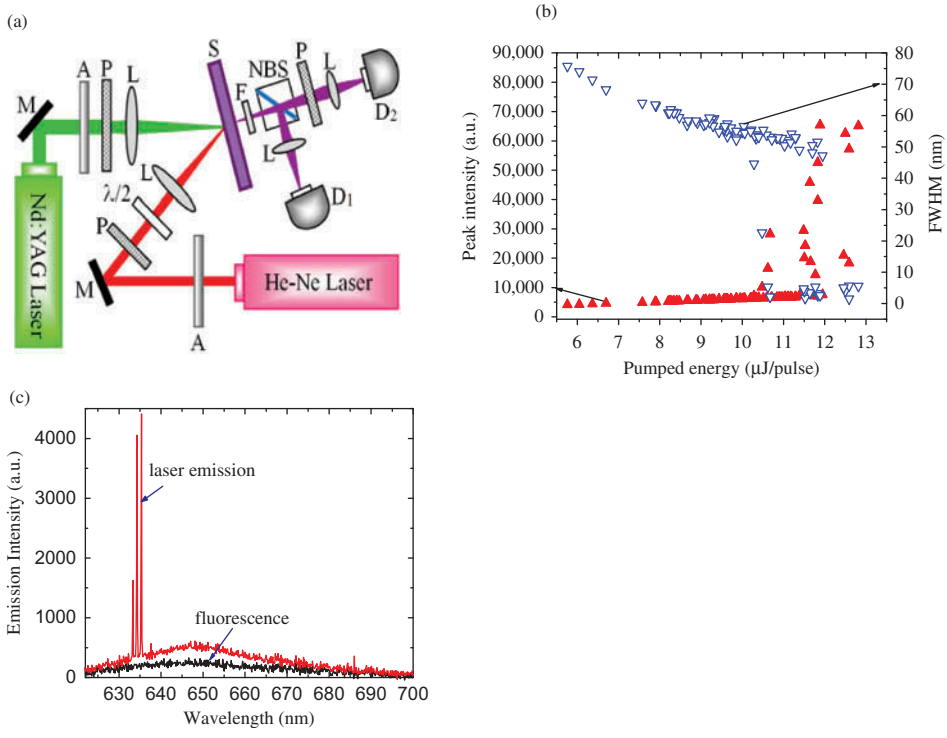

Figure 1. (colour online) (a) Experimental set-up. A, P, M, λ/2, L, F, NBS and D were the attenuator, polariser, mirror, half-wave plate, lens, bandpass filter centred at 630 nm, non-polarised beam splitter and high-speed photodetector, respectively. S was the dye-doped NLC cell. (b) Variations of the peak intensity of fluorescence output and corresponding full widths at half-maximum with the incident pump energy. (c) Fluorescence and lasing spectra emitted from the dye-doped NLC cell.

control beam blocked from irradiating the sample. With the increase of the pump energy, the peak intensity of the fluorescence output and the corresponding full widths at half-maximum varied non-linearly as shown in Figure 1(b), and a pump energy threshold value of about 12 μJ/pulse was obtained. Figure 1(c) shows the typical fluorescence spectra emitted from the dye-doped NLC. It shows a spontaneous emission curve from the Pyrromethene dye at low pump energy and discrete sharp peaks near 633 nm emerge when increasing the pump energy above the threshold value. For a given pump area, lasing modes always appear at the same wavelengths, but the relative weights of the different modes fluctuate randomly from shot to shot. The emission peaks are not evenly spaced, and the number of the modes depends strongly on the pump area. When a different area is excited and thus a different disorder configuration probed by moving the sample, a completely different set of lasing modes emerges. These above-mentioned characteristics are consistent with those reported for RLs in organic thin films.[11,12]

Secondly, the all-optically reversible controllability of the NLC RL with a fixed pump energy of around 20 μJ was observed. Fixing the control beam *p*-polarised and at an input power of about 6 mW, we found the random lasing emission disappeared and reappeared when turning on and off the control beam, as shown in Figure 2(a). And after hundreds of such switching cycles were performed, no obvious descent of the lasing intensity occurred. For the input power of the *p*-polarised control beam below 5 mW, tuning of the random lasing emission intensity by changing the input power of the control beam was observed. Figure 2(b) and (c) shows the typical results when turning on and off the *p*-polarised control beam at input powers of 2.4 and 0.8 mW, respectively. Similarly, during the control process with hundreds of switching cycles, the lasing intensity could be recovered without evident decline after turning off the control beam, and the output lasing intensity in the presence of the control beam basically maintained invariable. During the above controlling process, SPM of the control beam after propagating through the NLC film was also observed. The diffraction patterns on the projection screen in the far field were shown in the right-hand side of Figure 2. At low control powers (<2.0 mW), a steady-state broadening of the laser spot is observed, and at the same time, a sharp decline of the random lasing intensity takes place. The lasing intensity drops to 14% of the value obtained without the control beam when the power of the control beam increases to ~2.0 mW at which a main ring diffraction pattern appears. On further increasing the power of the control beam (>2.0 mW), the size and the number of the main

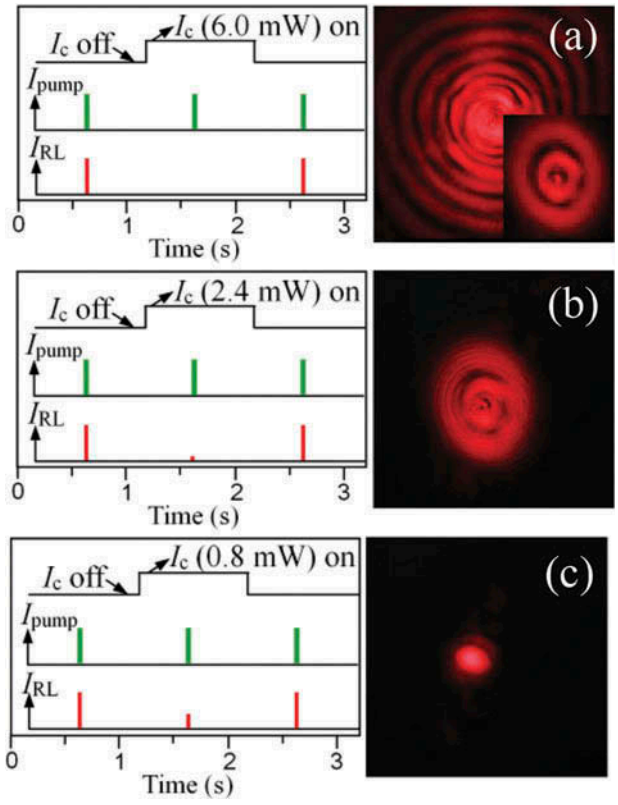

Figure 2. (colour online) Typical manipulation of the random lasing (I_{RL}) by turning off and on a *p*-polarised control beam (I_c) with an input power of 6.0 mW ((a) left), 2.4 mW ((b) left) and 0.8 mW ((c) left) at 633 nm in the presence of the pump pulse (I_{pump}). The concurred SPM ring patterns of the control beam were shown on the right of the corresponding figures of the controlling processes. At the control power of 6 mW, the lasing could be switched off and on completely by turning on and off the control beam. The inset of (a) is the SPM pattern of the *s*-polarisation control beam at 6.0 mW.

diffraction rings increase, in the process of which the RL is further weakened until the disappearance of the random lasing emission.

Figure 3 shows the dependence of the output intensity of the NLC RL on the power of the control beam with the polarisation direction at 0°, 30°, 60° and 90° to the initial *p*-polarised direction. From the figure, we can see that the lasing intensity decreases when the power of the control beam rises from 0 to 5 mW for all the considered polarisations. For the control beam with the zero-degree polarisation (i.e., *p*-polarised), the RL intensity drops to a half, 30% and ~3% of that without the control beam when the control power reaches 0.5 , 1 and 5 mW, respectively. And for the same input power of the control beam, the polarisation direction with smaller angle to the nematic director leads to sharper fall of the lasing intensity. For the control beam with an input power

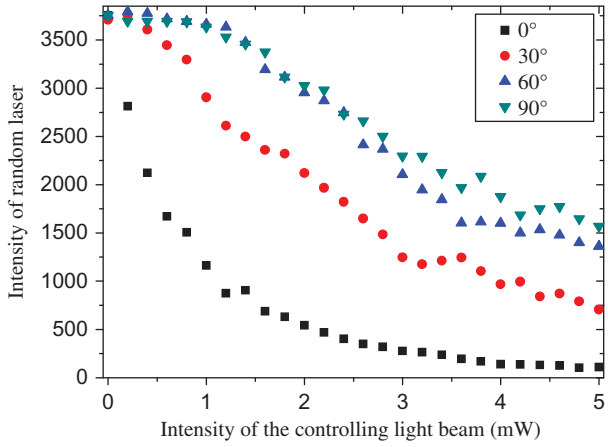

Figure 3. (colour online) Intensity of the RL output as a function of the power of the control beam from 0 to 5 mW. The data plotted in square, circle, up-pointing triangle and down-pointing triangle were measured with the polarisation direction of the control beam at 0° (along the *p*-polarised direction), 30°, 60°, and 90°, respectively.

of 2 mW, the RL intensity drops to 14%, 57%, 79% and 81% at 0°, 30°, 60°, and 90° polarisation directions, respectively. This indicates a continuous tuning of the output intensity of the RL emission by changing the power of the control beam, and the decline rate of such a tuning process can be adjusted by the polarisation direction of the control beam. And as mentioned earlier, above a threshold power (~6 mW for the *p*-polarised He-Ne CW laser in our experiment condition), the lasing output disappears and this optical tuning behaviour changes into the light switching behaviour.

Subsequently the underlying mechanism of the above optically controlled random lasing was considered. It was believed that the spontaneous fluctuations of the nematic director leading to the fluctuation of the dielectric tensor of the NLC resulted in recurrent multiple scattering to provide the optical feedback for random lasing,[2] which could be manipulated by controlling the director orientation or the order of the NLC. The mechanisms responsible for the photo-induced reorientation or the order change of the NLC by single focused beam include optical-field-induced reorientation, photothermal induced order and density change, and photoisomerisation.[10,13–19] For the optical-field-induced reorientation, the optical electric field reorients the nematic director to the polarisation direction, leading to a local refractive index change.[10,13,14] During this process, the optical field will also align the NLC molecules into better uniformity and diminish the fluctuation of the nematic director, resulting in the descent of the lasing intensity. And for the photothermal effect the NLC is heated by light locally, and then the order of the NLC will decrease with the temperature,[10,16–18] leading to the enhancement of the fluctuation of the nematic director and the dielectric tensor, and the change of the lasing behaviour.[20] Photoisomerisation effect [10,13,19] could be excluded because there was no photoisomerisable dye doping into our NLC sample. To distinguish which mechanism accounts for the optically controlled RL, the influence of the control beam on the lasing output polarisation was investigated. Figure 4 shows the normalised transmitted intensity of RLs through an analyser at different powers of the 30° polarised control beam (there is similar dependence on the control power at other polarisation directions). The RL output at a control power of zero was linearly polarised at the direction in proximity to the bisection between the nematic director and the polarisation direction at the maximal scattering (approximately 10° from the nematic director for the NLC sample used here). This may be because the dye molecules with anisotropic absorption and emission are aligned with the nematic director, which together with the anisotropic scattering of the NLC determines the lasing polarisation direction.[21] A negligible change of the polarisation direction of the dominant polarised component of RLs was observed under the application of the control beam with different incident powers. However, the polarisation degree reduced from 0.98 for the control power of zero to 0.97 and 0.93 for those of 1.5 and 3 mW, respectively. This result indicates that the optical-field-induced director reorientation does not play a predominant role

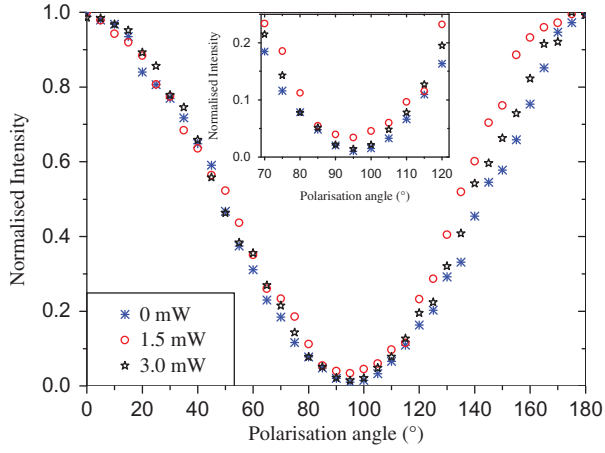

Figure 4. (colour online) Normalised transmitted intensity of the RL emission after an analyser for the 30° linearly polarised control beam with different input powers.

in the optically controlled process. Otherwise at the same time, the nematic director also reoriented towards the optical electric field direction (namely the polarisation direction of the control beam), resulting in the rotation of the polarisation direction of the dominant polarised component of RLs towards the optical electric field direction.[21] This change of the lasing polarisation was not observed. So, we believe that the photothermal effect plays a crucial role in the optically controlled random lasing behaviour observed here. For the RL system consisting of homogeneously aligned NLC E7, the degree of anisotropy was about 0.2 as reported in the work by Strangi et al.[2] Heating the NLC sample used here to the isotropic phase, an absorption coefficient of 30 cm^{-1} was obtained. Comparing with the reports of the photothermal-effect-based SPM in the same NLC E7 system, [17,18] NLCs in our experiment absorbed the similar optical power in the unit volume leading to the similar temperature increase. The resulted temperature increase due to the photothermal effect consequently results in the decrease of the NLC order and the increase of the director fluctuation. The increase of the director fluctuation influences RLs from two aspects. One is that the increase of the director fluctuation enhances the loss of spontaneous emission photons and lasing resonance and thus decreases the lasing output intensity. The other is it will improve the optical feedback for RLs, thus increasing the lasing intensity. The reported experimental results show that the former takes the predominant role so that the laser intensity decreases with the temperature.[20] And in the meanwhile, the decrease of the NLC order leads to the deterioration of the lasing polarisation. In addition, because of the anisotropic adsorption, the controlling effect is the most significant when the polarisation direction of the control beam is parallel to the nematic director and decreases when the two directions deviate from each other, as shown in Figure 3.

The proposed mechanism can be substantiated further by the concurred SPM of the control beam. When adjusting polarisation of the control beam from *p*-polarised to *s*-polarised, the SPM effect became weaker, but did not disappear at *s*-polarisation, as shown in the inset of Figure 2(a). From Figure 2, we can also see that the random lasing intensity decreases with the increase of the change of the refractive index locally leading to SPM (i.e., with the increase of the number and the diameter of the diffraction rings). This is because when the polarisation of the control light is parallel to the nematic director of NLCs, the light is absorbed the most leading to the most significant SPM and controlling effect. And with the increase of the angle between the polarisation of the control light and the nematic director, the absorbed light decreases, leading to the decrease of the SPM and controlling effect. As for the SPM effect arising from the optical-field-induced reorientation cannot be produced by the incident beam of *p*-polarised direction, and the optical-field-induced refractive index change rises with the increase of the angle between the polarisation direction and the nematic director, [10,13–15] which is inconsistent with our experiment results.

Besides, the pump pulse could destroy the SPM pattern, which would restore within half a second as shown in Figure 5. It is believed that this effect was also due to the local temperature increase through the photothermal effect leading to the decline of the NLC order, because it existed for all polarisation direction and was strongest at the *p*-polarised direction. Because of the high instantaneous power of the pulse, it is inferred that the temperature of NLCs on the focus increases a lot and the intensity-dependent orientation distribution of NLC micro-domains induced by the control beam is obscured, leading to the disappearance of the SPM pattern, as shown in t = 0.05 s of Figure 5. Within half a second, the pattern restored completely, indicating that the thermal disturbance of the pump pulse vanished. This indicates the restoring time from the light-induced enhancement of the fluctuation of the dielectric tensor was in the order of hundreds of milliseconds. It was observed if we increased the repetition frequency more than 2 Hz, the RL intensity decreased with the time due to the interferences between adjacent pulses. So, we can infer that the response time of the

Figure 5. (colour online) Dynamic evolution of the SPM diffraction pattern when a pump pulse was impinged on the dye-doped NLC film at time zero.

controlling over the RL by the control beam is also several hundreds of milliseconds.

4. Conclusions

In conclusion, a weak He-Ne laser with the power of several milliwatts was used to control the RL in NLCs. The lasing intensity can be tuned continuously with the variation of the control power and the lasing output can be switched on and off by turning off and on the control beam when the control power is above a certain value. The response time of the control process is several hundreds of milliseconds and there is no obvious decline of the output intensity after hundreds of control cycles. The influence of the control beam on the polarisation of the random lasing and the SPM of the control beam indicate that the photothermal effect takes the predominant role in the controllability of the RL in NLCs.

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