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Behaviors of random laser in dye-doped nematic liquid crystals

Fengfeng Yao [a,*], Huanting Bian [a], Yanbo Pei [a,b], Chunfeng Hou [a,b], Xiudong Sun [a,b]

[a] Institute of Modern Optics, Department of Physics, Harbin Institute of Technology, Harbin 150001, China
[b] Key Laboratory of Micro-optics and Photonic Technology of Heilongjiang Province, Department of Physics, Harbin Institute of Technology, Harbin 150001, China

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ABSTRACT

Random lasing in the nematic liquid crystals (NLCs) with a high doping concentration of the laser dye was observed and characterized. With increasing the pump energy after the occurrence of the random laser (RL), the RL intensity first increases gradually to a maximum, then drops sharply to zero, accompanied by the gradual enhancement of scattering manifested by the growth of far-field diffraction rings of the transmitted pump beam in number. The threshold energy per unit pump area, slope efficiency, and maximal output intensity of the NLC RL depend heavily and nonmonotonically on the pump angle. A model involving the pump pulse induced molecular reorientation in NLCs leading to the pump angle dependent enhancement of scattering is proposed to explain the pump angle dependent properties of RLs.

1. Introduction

Random laser (RL) is a laser whose feedback is not provided by an external resonator, but by scatterers randomly distributed in an active medium, and the interference of the scattered light gives rise to resonant modes at particular frequencies [1,2]. RLs have been realized across different systems, such as semiconductor powders [3–5], piconjugated polymers [6], scattering suspension in dyes [7], and liquid-crystal-based media [8–12]. For the applications of RLs, controlling RL emission is one of the research focuses of this field [13,14]. Thanks to the merits of the nematic liquid crystal (NLC) that its optical properties are susceptible to the weak external perturbation of a small external electric field, magnetic field, or light, NLC RLs can be developed into lasers with flexibly controllable features [9,10,12]. Because the pump light used to excite RLs is generally of high intensity, it is very possible that the pump light is able to induce the change of the properties of NLCs, leading to the change of random lasing properties. Studying this process is important not only for getting full knowledge into the lasing properties of NLC RLs, but also for developing new tools to control and tune the NLC RLs. However less research working on this effect was reported.

In this paper, dye-doped NLC RLs were observed and characterized under the coexistence of the pump pulse induced change of optical properties of NLCs manifested by the far-field diffraction rings for both the RL and pump pulse itself. The experimental data show that the RL intensity initially increases with the pump energy to a maximum, then drops sharply to zero. And the threshold energy per unit pump area, slope efficiency, optimal pump energy for the maximal output of random lasing and maximal output of random lasing depend nonmonotonically on the incident angle of the pump pulse. The following mechanism is suggested to explain the above observed phenomena and data. The optical scattering of NLCs changes due to the pump pulse induced reorientation or photothermal effect, leading to the change of the lasing properties. At lower pump energy, optical field induced molecular reorientation takes crucial role. At higher pump energy, the photothermal effect sets in and develops with the further increase of the pump energy into the predominant mechanism. The above pump pulse induced nonlinear effect predicts the lasing features consistent with those observed in the experiments.

2. Materials and optical setup

The empty cell was fabricated with two pre-rubbed polyvinyl-alcohol (PVA) coated glass slides separated by two 100 μm-thick Mylar spacers. The cells were filled up with NLC E7 (from SLICHEM, China) doped with 0.4 wt% Pyrromethene 650 dye (from Exciton) through the capillarity effect to form homogeneous alignment with the nematic director parallel to the rubbing direction in the plane of the cells. The samples were optically pumped with the pulse derived from a Q-switched Nd:YAG laser (532 nm, 6 ns, 1 Hz) as shown in Fig. 1(a). The pump pulse was focused onto the sample using a lens (L, $f=100$ mm), yielding a beam waist of about 200 μm at the focus position. The linearly

* Corresponding author.
E-mail address: fengfyao@hit.edu.cn (F. Yao).

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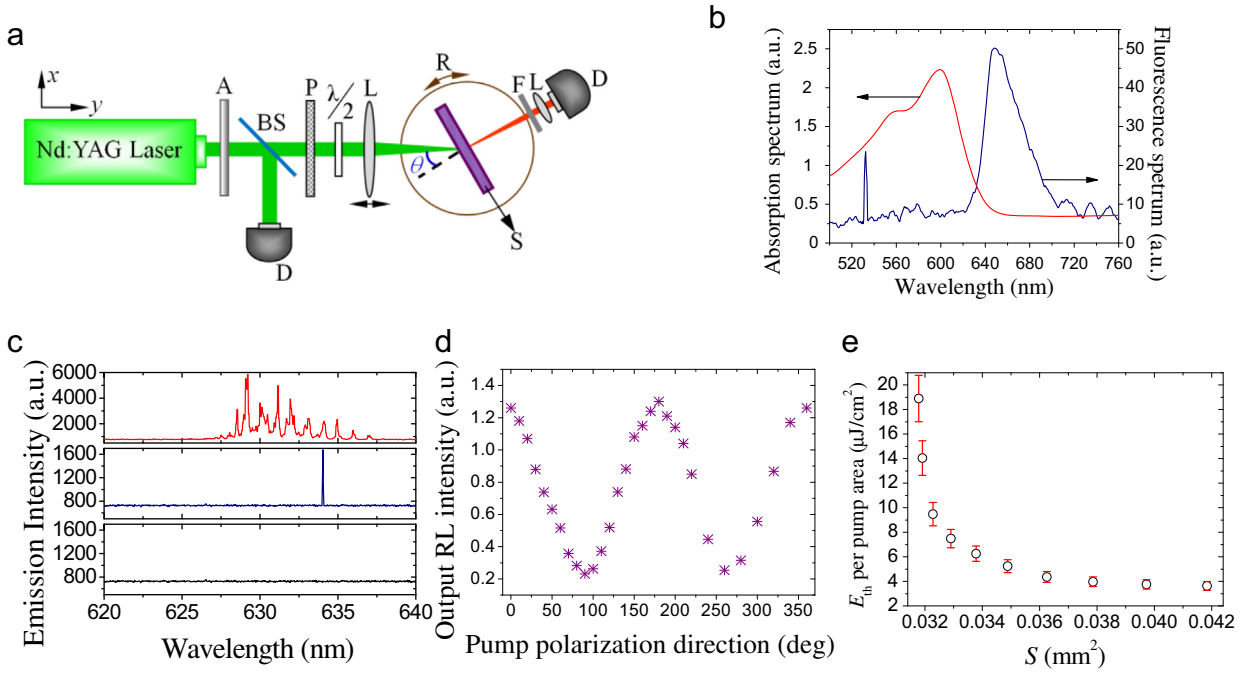

Fig. 1. (a) Experimental setup. (A) BS, P, $\lambda/2$, L, F, and D were the attenuator, beam splitter, polarizer, half-wave plate, lens, band pass filter centered at 630 nm, and high-speed photodetector or spectrometers, respectively. The lens was positioned on a one-dimension translation stage which was used to move the focus along the pump beam to change the pump area on the NLC film surface. S was the dye-doped NLC cell, and it was mounted on a rotator R. The sample could be rotated in the x–y plane to form any arbitrary incident angle of the pump beam θ. (b) Absorption and fluorescence spectra of the dye doped NLC sample measured by USB2000+. (c) Typical spectra emitted from the dye-doped NLC cell was obtained at $\theta=15°$ by the spectrometer HR4000. The incident pump energies for the spectrum curves from top to bottom were ~40, ~15, and ~12 μJ/pulse. (d) Dependence of the lasing intensity on the angle between the polarization direction of the pump light and the nematic director. (e) Dependence of the threshold energy per unit pump area on the pump area S on the NLC surface for $\theta=15°$.

polarized pump pulses were set to various intensities and any polarization direction by using an attenuator (A), a polarizer (P) and a half-wave plate ($\lambda/2$) before the lens. The focus of the pump pulse could be adjusted along the pump beam by positioning the focusing lens on a one-dimension translation stage. The external incident angle of the pump pulse θ could be varied arbitrarily by mounting the sample on a rotary stage, and the pump beam irradiated the same sample region while θ was varied. The area of the pump spot on the NLC film surface S could be varied by moving the one-dimension translation stage on which the lens L was mounted. After filtering the scattered light (by a bandpass filter F with a central wavelength of 630 nm and a bandwidth of 30 nm) and focusing it into a small spot, the RL output from the sample was measured by a high-speed photodetector D (DET 10 A/M, from Thorlabs) or Ocean Optics spectrometers USB2000+ (range: 350–1100 nm, resolution: 1.0 nm) and HR4000 (range: 550.0–666.2 nm, resolution: 0.065 nm). And the far-field diffraction patterns of the light beam were projected on the screen placed ~1 m away from the sample.

3. Basic characterizations

Firstly, the absorption spectrum and the emission fluorescence spectrum of the dye-doped NLC cell at low pump energy were measured by an Ocean Optics USB2000+ spectrometer, as shown in Fig. 1(b). The maximum of absorption lies at about 600 nm, and the pump light at 532 nm still has a high absorption. The fluorescence emission pumped by a 532 nm pulse laser mainly locates in the wavelength range of 620–710 nm, with the peak at about 650 nm. The RL emission spectra from the dye doped NLC cell were characterized by the spectrometers USB2000+ and HR4000 under the application of a p-polarized pump pulse for θ of ~15°. An incident pump energy threshold value of about 15 μJ/pulse was obtained at S~0.040 mm^2. With the increase of the pump energy around the threshold value, the peak intensity of the emission increases by several tens of times, and the full width at half-maximum (FWHM) of the emission spectra abruptly drops from ~40–50 nm to below 0.1 nm as shown in Fig. 1(c). Above the threshold energy, discrete sharp peaks near 633–634 nm emerge in the fluorescent spectrum. At $S<0.033$ mm^2, the spectral position of the peaks with randomly relative intensity weight varies randomly from pulse to pulse when the same part of the sample is excited repeatedly. When a different part of sample is excited by moving the sample and thus a different disorder configuration is probed, a completely different set of lasing modes emerges. These observations indicate that RL has formed in our NLC system.

For the homogeneously aligned dye-doped NLC system used here (dye molecules in NLCs tend to align along the nematic director due to the dipole–dipole interaction between dye molecules and NLC molecules), its anisotropic properties of scattering and absorption result in distinctive characteristics of the NLC RL. The lasing threshold is pump polarization dependent (as shown in Fig. 1(d), the lasing intensity undergoes a five-fold lowering when the polarization of the pump pulse is changed from the direction parallel to the director to the perpendicular, which is as reported in Ref. [8]). The emission is a linearly polarized RL as reported in our previous article [11], and the pump polarization has negligible influence on the polarization state of the RL. Resembling other RL systems [15], the threshold energy per unit pump area E_{th}/S depends strongly on S. Fig. 1(e) shows the relation between the threshold energy per unit pump area and S for our dye-doped NLC samples. The measured threshold energy per unit pump area drops with S with a gradual decrease in the rate of descent, and finally holds steady when S is above ~0.040 mm^2. This indicates that the threshold energy per unit pump area for the occurrence of the RL emission remains almost stable above a certain pump area $S=0.040$ mm^2.