Physica B 407 (2012) 4826–4830

Contents lists available at SciVerse ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Random lasers in dye-doped polymer-dispersed liquid crystals containing silver nanoparticles

LongWu Li *, LuoGen Deng

School of Physics, Beijing Institute of Technology, Beijing 100081, China

ARTICLE INFO

Article history:
Received 23 May 2012
Received in revised form
1 September 2012
Accepted 3 September 2012
Available online 18 September 2012

Keywords:
Random laser
Polymer-dispersed liquid crystals
Nano-sized liquid crystal droplets
Silver nanoparticles

ABSTRACT

We report on the observation of random lasers for the first time in dye-doped polymer-dispersed liquid crystals (PDLCs) containing nano-sized liquid crystal droplets and silver nanoparticles. The dye-doped polymer-dispersed liquid crystal containing silver nanoparticles film is exposed by a collimated 532 nm Nd: YAG (yttrium aluminum garnet) laser beam, so that it is quickly cured. Under the excitation of a frequency-doubled Nd: YAG (yttrium aluminum garnet) laser operating at a wavelength of 532 nm, random lasing from dye-doped PDLCs containing Ag nanoparticles is observed as a result of cooperative effect due to light scattering of nano-sized liquid crystal droplets and the local field enhancement capabilities around silver nanoparticles. We show that the threshold of the random lasing is about 0.95 μJ/pulse which is lower than the lasing threshold of dye-doped polymer-dispersed liquid crystals containing nano-sized liquid crystal droplets. The linewidth of the lasing peaks is shown to be 0.2 nm. We also propose a possible mechanism to explain the random lasing from dye-doped polymer-dispersed liquid crystals containing silver nanoparticles.

1. Introduction

Random lasers have attracted considerable attention over the last decade due to their interesting physics and potential applications in photonics and bio-medicine [1–5]. Many of the passive or active disordered materials, such as ZnO powders [2], polymers [6], dye-doped liquid crystals (DDLCs) [7–12], and dye-doped polymer-dispersed LCs (DDPDLCs) [13,14], can be employed to generate random lasing. In recent years, metal nanostructures have been realized to significantly enhance the spontaneous emission rate of semiconductor quantum wells [15], rare-earth ions [16], and laser dyes [17] because of the interaction between emission centers and surface plasmons. Utilizing these advantages, some researchers reported random lasers mediated by metal nanoparticles with incoherent emissions [18,19]. They observed narrowing of an emission spectrum and increasing in emission intensity, which are typical characteristics of incoherent random lasers. However, metal nanoparticles have received little attention as scatters in dye-doped liquid crystals or polymer-dispersed liquid crystals random laser. And scarcely have people conducted experiments on amplifying random media have been made by combining three kinds of materials (polymer-dispersed liquid crystals, metal nanoparticles and dye) which give rise to multiple scattering and optical gain. To address the above situation, we experimentally and theoretically discuss on random lasing with coherent feedback effect in dye-doped polymer-dispersed liquid crystals containing silver nanoparticles. In this paper, we have made an amplifying random medium containing Ag nanoparticles with a size (~5 nm in diameter) and nano-sized liquid crystal droplets and demonstrated that coherent random laser emissions around 625 nm can be attained in dye-doped polymer-dispersed liquid crystals with embedded nano-sized liquid crystal droplets and silver nanoparticles.

A single sharp emission peak, with full width at half maximum (FWHM) less than 0.2 nm, appears at a threshold pump energy, while further increase in the pump energy brings about multiple laser spikes. With the purpose of comparison, we have studied stimulated emission in a random laser based on dye-doped polymer-dispersed liquid crystal containing nano-sized liquid crystal droplets, similar to those of Ag nanoparticles-dispersed dye-doped polymer-dispersed liquid crystals containing nano-sized liquid crystal droplets, and found that the Ag nanoparticles-dispersed one has a lasing threshold lower than those of the nano-sized liquid crystal droplets-dispersed ones. We have clarified the importance of the presence of Ag nanoparticles for the observation of distinctive laser spikes from the contribution localized surface-plasmon resonance (LSPR) and multiple scattering of light. The comparison indicates that Ag nanoparticles are better scattering elements toward the design of random lasers with high intensity and low threshold, thereby expanding the study of random lasers.

* Corresponding author.
E-mail address: llwnn2012@126.com (L.W. Li).

0921-4526/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.physb.2012.09.023

2. Sample preparation and experimental setup

The dye-doped PDLC films were prepared by mixing a pre-polymer and 0.1 ml ethanol solution (1.35×10^{-4} g/ml) containing 10 times diluted silver nanoparticles (with a diameter 5 nm) and photo-polymerized by an Q-switched, and frequency doubled 532 nm Nd: YAG (yttrium aluminum garnet) pulsed laser operating at 532 nm. The prepolymer consists of 44.41 wt% of monomer, trimethylolpropane triacrylate, 7.64 wt% of cross-linking monomer, N-vinylpyrrollidone, 0.54 wt% of photo-initiator, rose bengal, 1.02 wt% of coinitiator, N-phenylglycine, 8.44 wt% of surfactant, octanoic acid, and 35.3 wt% of liquid crystal (E7), all from State Key Laboratory of Polymer Material Engineering, College of Polymer Science and Engineering, SiChuan University. And 1.36 wt% of lasing dye, 4-dicyanomethylene-2-methyl-6-4-dimethylami-nostyryl-4H-pyran (DCM) from Exciton. The liquid crystal used has an ordinary refractive index of $n_o = 1.5216$ and an extraordinary refractive index of $n_e = 1.7462$, and a birefringence of $\Delta n = 0.2246$. The mixture of the dye-doped polymer-dispersed liquid crystals containing nano-sized liquid crystal droplets and Ag nanoparticles was injected into the empty cell, forming a dye-doped PDLC cell, which was made up of two pieces of indium-tin-oxide (ITO) coated glass slides, and then subjected to a uniform exposure of a Q-Switched and frequency doubled 532 nm Nd: YAG (yttrium aluminum garnet) pulsed laser. The exposure intensity on the samples was about 20 mW/cm^2 and the exposure time was about 20 min. The laser uniform exposure induced rapid polymerization, and resulted in the nanoscale LC droplets and the silver nanoparticles which were fixed in the polymer matrix. The thickness of the samples was 10 μm. The pump laser used was a linearly polarized, Q-switched, and frequency doubled 532 nm Nd: YAG (yttrium aluminum garnet) pulsed laser (LABest Optronics Co.; Ltd) with a pulse duration of 8 ns and repetition rate of 10 Hz. The schematic setup of the lasing measurement is shown in Fig. 1.

In experiments of random lasers, the sample was optically pumped by the second harmonics of an Q-switched Nd: YAG (yttrium aluminum garnet) pulsed laser (532 nm, 10 Hz repetition rate, 8 ns pulse duration). A half-wave plate ($\lambda/2$ for 532 nm) was placed in front of the polarizer and polarizing beam splitter (PBS) for adjusting incident pulse energy. The excited stripe width could be varied by an adjustable slit. One end of the excited stripe was close to the edge of the sample. The laser emission from the edge of the film was measured. The pump beam was separated into two paths: one was monitored by an energy meter and the other was used as the pump beam. The detector was placed on the edge of the sample to collect the lasing signal and connected to a spectrometer. The laser beam was focused on the samples through adjustable slit to form an excited stripe with a length 9 mm and a width that could be varied from 1 mm to 0.04 mm. The emission was collected along the edge of the sample and connected to a spectrometer that was monitored real time by a computer. In the other experiment of coherent of random lasing from dye-doped polymer dispersed liquid crystals, the sample cell, materials, material parameters, weight percent concentration, sample preparation, experimental apparatus and methods of measurement we used were the same with those used in this experiment mentioned above. The only difference was that there were no silver nanoparticles.

3. Results and discussion

For scanning electron microscopy (SEM) measurement, one side ITO glass of the test samples was removed so that the random distribution of LC droplets and Ag nanoparticles can be gauged by scanning electron microscopy (SEM). Fig. 2 shows the SEM image of the dye-doped PDLC film containing nano-sized liquid crystals and Ag nanoparticles. Fig. 3 depicts the evolution of emission spectra from the dye-doped PDLC containing nano-sized liquid crystal droplets without Ag nanoparticles with various pumping energies of (a) 1.97 μJ, (b) 2.2 μJ, (c) 2.8 μJ, and (d) 3.2 μJ /pulse at excited areas 1.35 mm^2. It can be seen from Fig. 3 that, a broad band is observed below the lasing threshold, and the corresponding linewidth (FWHM) is about 20 nm. Many narrow random lasing peaks with ≤ 1 nm (FWHM) appear at the top of the emission spectra when the incident pumping energy exceeds 1.97 μJ. The linewidth of the lasing peaks is more than 20 times narrower than the amplified spontaneous emission (ASE) linewidth below the threshold. At small pumping intensity, only

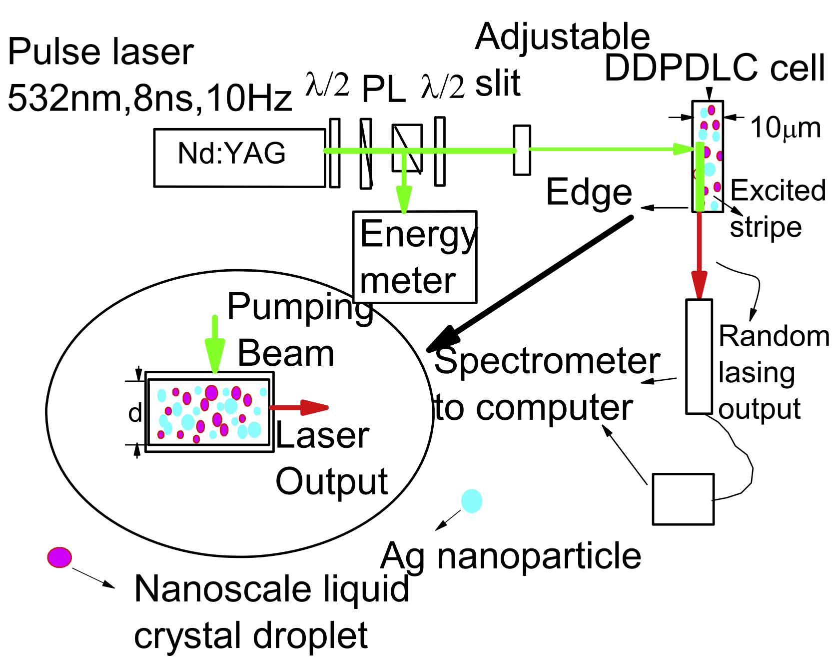

Fig. 1. Schematic setup of lasing measurement.