LETTER

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Random lasing characteristics in dye-doped semiconductor CdS nanoparticles*

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Abstract

A new strategy to obtain random lasing from semiconductor CdS nanoparticles is presented. A device is prepared with dye (DCM) solution containing CdS NPs. Good lasing behavior is reached and the multiple light scattering process is responsible for lasing generation. We also manage to show that the lasing modes and output intensity exhibit angular dependence. The result indicates that CdS nanoparticles can be used as ideal microlaser material in future semiconductor laser devices.

Keywords: random laser, laser threshold, CdS nanoparticles

(Some figures may appear in colour only in the online journal)

Introduction

The realization of random lasers aims to combine the flexibility of gain media with the technological advantages of disordered media light emitters, having the ultimate goal of electrical pumping. To date, various types of random lasers have been exploited in versatile dye-doped device lasers to enhance light emission, including ZnO random lasers, TiO_2 nanoparticles random lasers, LCs random lasers and LCs with Ag nanoparticles [1–8]. Random lasers can be easily achieved in a dye-filled fluidic with dispersed Ag nanoparticles, despite the absence of an optical cavity, in which the optical feedback is provided by multiple scattering between nanoparticles. Apart from similarities with semiconductor random lasers in the physics mechanism, integrating the lasing source onto miniaturize particles provides significant benefits such as low costs, easy fabrication, portability, and high degrees of functionality. Furthermore, random lasers may also perform as a potential light source for retrieval of contaminated information [9]. Recently, the LCs and nanoparticle structures offer a new route for on-LCs random lasing source integration [10–13, 15]. Lee *et al* demonstrated an LCs random laser based on the inherent fluctuations of the local dielectric tensor due to the main effect responsible for the recurrent multiple scattering events as a light wave is propagating through the LC medium which largely reduces the demands of high standard fabrication techniques. These researches opened the door for exploration in random lasers, and thereafter, much attention has been paid to on-particle random lasers investigation due to its optical character with easy fabrication processes. Typically used as scatterers are such things as, dielectric particles, noble metal particles, liquid crystals, semiconductors and so on. In this letter, we present for the first time a strategy to obtain random lasing from CdS NPs. The novelty of this system relies on the random laser fabrication process by getting rid of any surface treatment procedures. We have used a cell, filled with dye solution containing the CdS NPs that act as scatterers. The objective of this paper is to study the influence of the excited emission from the semiconductor CdS NPs with DCM solutions on the lasing properties. Our approach is based on the lasing characteristic measurement and associated discussion describing the characterization of the lasing modes in CdS nanoparticles. By optimizing the new material, namely CdS NPs, the basic spectroscopic properties and the output characteristics of the DDCdS NPs lasers at different angles with respect to the incident pump beam had been obtained.

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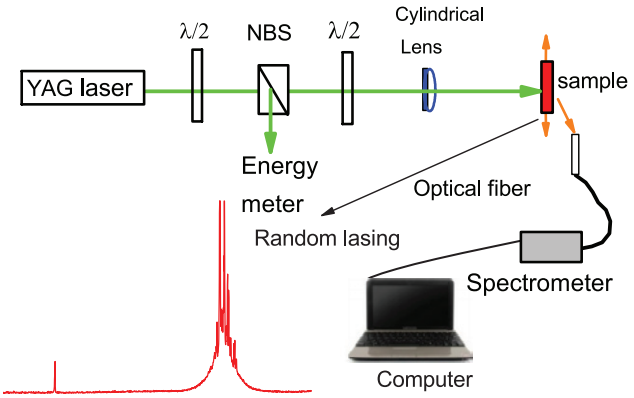
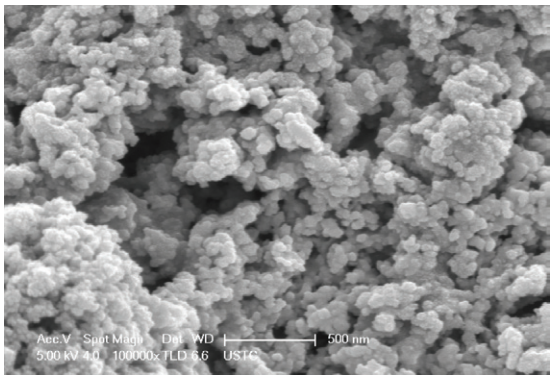


Figure 1. Probing laser emission of the optical setup used to investigate lasing action.



SEM

Figure 2. SEM image shows the photograph of CdS nanoparticles.

Samplepreparation and experimental setup

We have purchased from Exciton, devices that are very simple, low cost, and provide a reliable method of fabricating dye-doped CdS NPs. The DDCdS NPs device was prepared using DCM dye (0.5 wt%, $4-Dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran$) ethanol solution as an active dopant and CdS nanoparticles as scatterers. The ingredients were thoroughly mixed in a small vessel. Then, the mixture was absorbed in this vessel to make it infill the tube through capillary action and was then injected into the empty cells forming a dye-doped CdS cell (10 μm), which was placed between two glass slides. As shown in figure 1, the set up was used to measure the lasing spectra response pump intensity. The cells were optically pumped by a pulsed (8 ns)-Q-switched, frequency double ($\lambda = 532$ nm) Nd-YAG laser. The pump beam through NBS separated into two paths: one path was monitored by an energy meter, and the other path was used as the excitation beam. The excitation beam was focused on the sample through a cylindrical lens to form an excited stripe (0.3 mm wide and 8 mm long). The fiber tip was placed on the laterals or edges of the sample to collect the lasing signal. The output light was coupled from a fibre into an ocean optics spectrometer with 0.1 nm resolution.

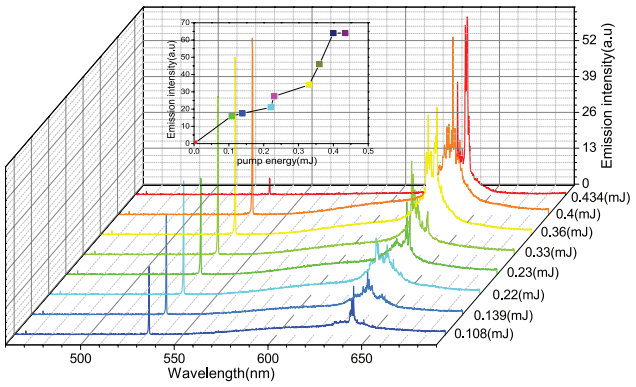


Figure 3. Typical emission spectra of random lasing at various pump pulse energy.

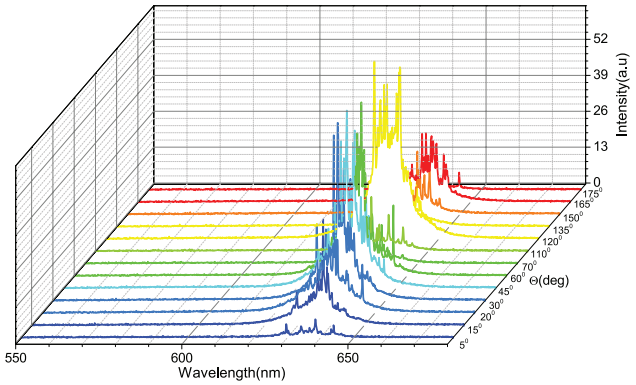


Figure 4. Emission spectra as a function of the angle.

Resultsand discussion

The size of CdS nanoparticles can be measured by scanning electron microscopy (SEM). Figure 2 shows the image of the CdS nanoparticles. We found that the size of CdS nanoparticles was 5 nm. The nanoparticles dispersed in dye solution was then dropped into cells. As a result, dye-doped CdS nanoparticle samples were obtained. These samples were pumped using a light pulse of 532 nm from a Q-switched laser with a pulse duration of 8 ns. The laser was incident normal to the sample surface. Emission spectra were also detected using a single fiber spectrometer equipped with a computer. All measurements were performed at room temperature. We examined the emission characteristics of the sample under pump energy excitation through pulsed laser. Figure 3 shows the random lasing spectra of CdS NPs under different excitation intensity at room temperature.

When the excitation intensity exceeds a threshold of 0.108 mJ, sharp peaks with linewidths as narrow as 0.1 nm emerge from the broad spontaneous emission spectrum, indicating the appearance of lasing action. As the excitation intensity increases, multiple spikes between 620 nm and 650 nm, can be detected. The spectra from the different angles at the same excited area for the DDCdS NPs are shown in figure 4.

To examine the directional emission property of the sample device, we measured the angular dependence of the

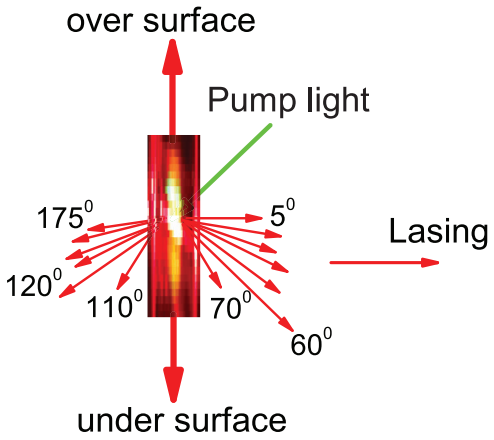


Figure 5. The measurement degrees are given.

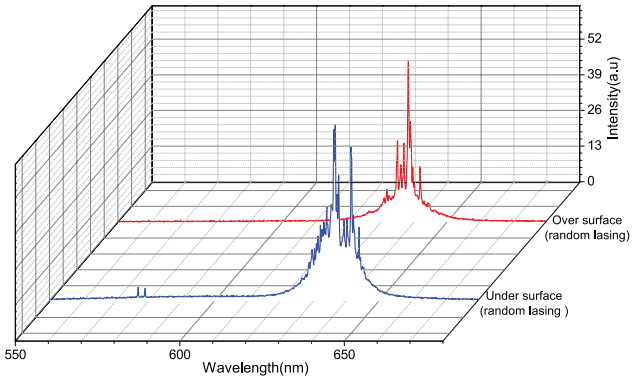


Figure 6. The spectra from a sample of over and under surfaces, respectively.

emission spectrum with a fixed pump laser energy of 0.434 mJ. Figure 4 shows the emission spectra at different collection angles of 5°, 15°, 20°, 30°, 45°, 60°, 70°, 110°, 120°, 135°, 150°, 165°, and 175°, at an excitation energy of 0.434 mJ. The peak positions and the intensity vary with the angles. It can be seen from the measured spectra that for lasing modes the lasing intensity from 30°–60° and 120°–165° is much stronger than those measured from other angles. This means that the laser emission does stem from the scattering of the disordered CdS NPs [1]. The model of Cao [14], which is based on optical scattering, also describes our results in figure 4 very well. Figure 5 shows the measurements were made at the angles of 0°–180°. Figure 6 shows the emission spectra at over and under surfaces, at an excitation energy of 0.434 mJ. Random lasing was found to vary along the measuring angles. It indicates that the angular distribution of the random lasing and the scattered light are different from each other.

This is evident from the above figures, in which gain length is reduced to the minimum length, the photon has to travel in-order to undergo a stimulated emission in a cavity and as a result the resonant lasing cavities that can provide stimulated emission can be formed easily, and hence the photons can have sufficient gain required for lasing by forming a relatively small loop. Due to the reduction in gain length, as the pumping energy is increased to E_{th}, more cavities are formed and there is an enhancement in the number of mode structures. It can also be observed that there is a wavelength shift for the entire emission spectrum due to emission and reabsorption process taking place within the dye solution. The plausible reason is that multiple scattering phenomena assures the localization of photons within the media containing gain medium and for random lasing to occur the formation of random cavities by the scatterers (CdS NPs) is essential, and can be considered a form of confinement of light within the sample. It is also shown that the addition of CdS NPs helps to stabilize the dye in the scattering mixture, and increases the multiple scattering repeatability of the photons. Random grains show a pronounced variation of mode spacing with excitation area, angle, and film location. Since these variations are observed, we conclude that work on CdS nanoparticles describes the formation of laser cavities by scattering from random grains. Evidently, the random lasing of DDCdS NPs is driven by optical scattering.

At the different angles, each dye molecule absorbs along the direction of its absorption transition dipoles, and emits a fluorescent photon according to its transition emission dipole, which is different. The spontaneous emission probability P can be expressed as a function of the dipole transition intensity S_{ed} as $P \propto S_{ed}$. From the viewpoint of the formation of the random lasing in dye-doped CdS NPs, lasing has two effects. One effect is that it improves the optical feedback for random lasers and thus enhances the transition of the dye molecules from upper level to lower levels and increases the stability lasing output intensity. The other effect is that the increase of the multiple scattering enhances the rates of spontaneous emission, decreases the threshold energy per unit pump area, and increases the lasing intensity. In our experiment, we have observed that the orientation of the dye molecules does contribute mainly to the angle dependent lasing properties. This explains why transition emission dipoles of dye molecules can emit fluorescence in specific directions, even if they are isotropically distributed. Considering the angle-dependent lasing emission, increases from 0–60 or from 180–120 degrees, resulting in gradual increases in transition emission dipoles for a given pump energy. The larger the transition emission rates of dipoles the more remarkable is the linear effect produced, accompanied by the scattering intensity around the emitted light becoming stronger and stronger. When the angle is from 60–120 degrees, the transition emission rates of dipoles gradually becomes small, the random lasing also decreases. Thus, at the different direction angles, the orientation of the pump-pulse-induced dye molecules contributes to the formation of random lasers, and enhances the slope efficiency greatly. It is manifested by the experiment results observed at $\Theta \leqslant 60°$ or $\Theta \geqslant 110°$, under the application of a pump energy of 0.434 mJ. Once the strength of the transition emission rates of dipoles attains a critical value approximately equal to that produced by the pump energy of 0.434 mJ, between 60° and 120°, the reorientation of the dye molecules is evident and the absorbing in the linear process becomes considerable, leading to the absorption transition dipoles playing a main role in the formation and amplification of random lasers. This will induce the increase of the threshold population inversion for the random lasing emission between 60° and 120° compared to that at $0° \leqslant \Theta \leqslant 60°$, or $120° \leqslant \Theta \leqslant 180°$.

With the difference of the angles at a fixed pump energy, the strength of the dipole transition effect will be different. For $0° \leqslant \Theta \leqslant 60°, 120° \leqslant \Theta \leqslant 180°$, the enhancement of transition dipoles by the reorientation of the pump-energy-induced dye molecules facilitates the formation and amplification of random lasers. However, for $60° < \Theta < 120°$, it does not benefit the amplification of random lasers. When the transition emission strength of the dipoles attains a critical value at which too much loss of spontaneous photons counterbalances the net gain owned by the original system with a negligible disturbance induced by the pump energy, the creation of random lasers gradually disappears. We propose a simple explanation based on re-absorption. Due to the overlap between absorption and stimulated emission cross sections of the transition dipole, unexcited molecules can absorb photons emitted from excited states, which decreases the gain:

$$g(\lambda_{em}) = \sigma_{em}^{dye}(\lambda_{em})N^* - \sigma_{abs}^{dye}(\lambda_{em})(N - N^*), \qquad (1)$$

where N stands for the total density of dye molecules and N^* for the density of excited dye molecules. At the change of the g value from the maximum to $g < g_{th}$, a nearly lasing strength was produced at different measured angles of Θ in devices by the reorientation of the pump-energy-induced dye molecules. Hence, the g values should be inferred as well in the lasing regime. In addition, the ratio of dye molecules $\gamma(\lambda_{em})$ that must be excited to reach the threshold also depends on re-absorption. We wrote the ratio of dye molecules $\gamma(\lambda_{em})$:

$$\gamma(\lambda_{em}) = \frac{N^*}{N} = \frac{\sigma_{abs}^{dye}N - \frac{lnR}{\ell\sigma_{em}^{dye}(\lambda_{em})(1+cf)}}{(\sigma_{em}^{dye}(\lambda_{em}) + \sigma_{abs}^{dye}(\lambda_{em}))N}, \qquad (2)$$

where ℓ is the dimension of the active laser medium in the cells, and R is the effective reflection coefficient representing the residual optical feedback caused by the NPs. Where we have introduced the volume fraction f of scatterers (the scatterers in the active medium was a variable), and the phenomenological constant c. In the regime, where losses due to absorption of the dye molecules is non-negligible, the gain g at the lasing wavelength is assumed to be limited by absorption losses caused by dye molecules and is therefore the volume fraction f of scatterers dependent. For lower f or lower g, $\gamma(\lambda_{em})$ takes on lower values according to equation (2), indicating a higher lasing threshold. Lower f or g values cause a shift in the laser wavelength to shorter wavelengths. Therefore, a decrease in gain g due to small volume fraction f leads to a higher laser threshold and a blue-shifted emission spectrum. It is obvious that each $\gamma(\lambda_{em})$ value corresponds to the lasing wavelength close to threshold. The random lasing wavelength is red (or blue)-shifted when the fraction f of the scatterers increases (or decreases), that is, when the loss of the system gain decreases (or increases). In fact, since the mode of the lasing spectra does depend on the sample position, film thickness, angle and excitation spot, the feedback is apparently provided by cavities which are formed by multiple scattering of the light [14]. In future work we will apply scanning near-field optical spectroscopy to try and resolve sample devices so that we may more precisely understand the nature of the laser microcavities.

Summary

In conclusion, we have fabricated a DCM solution filled with CdS NPs lasers with lasing threshold as low as 0.108 mJ and narrow laser line widths of 0.1 nm in the 640 nm wavelength region. By varying the measurement angle, different random laser action is obtained and explained by a dipoles transition model. The CdS NPs of optical quality promotes an enhancement of radiation and lasing properties, leads to higher emission stability of devices, and longevity of working time. Therefore, these results illustrate important progress in CdS NPs based on random lasing devices and ought to encourage renewed interest in CdS NPs as an emitting material.

Acknowledgments

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