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Stable random lasing in CdSSe micropowders

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Abstract. **The random lasing in CdS micropowder in the green region (near 520 nm) at room temperature under optical pumping by pulsed Nd^{3+}: YAG and N_2 lasers at wavelengths of 355 and 337 nm, respectively, is studied. The influence of fluctuations of pump pulse output energy on the random lasing action in CdS micropowder is determined. It is shown that the use of a pump laser with highly stable output power and high spatial beam uniformity (in particular, an N_2 laser) provides stable random lasing in CdSSe micropowders. The lasing is characterised by the presence of a clear structure of narrow lines in the spectrum and stability of their position during pulsed operation at a repetition rate of 700 Hz at wavelengths of 525, 570, 600, and 650 nm with thresholds in the range of 1100–1600 kW cm^{-2}.**

Keywords: *semiconductor lasers, optical pumping, random lasing, micropowders, semiconductors.*

1. Introduction

An important area in the field of the development of new-type lasers is the obtaining and study of random lasing in various scattering active media. This effect is caused by the random formation of numerous gain loops under multiple scattering of radiation in an active disordered medium [1, 2]. Random lasing has been obtained in different regions of UV, visible, and IR spectral ranges in active media based on nano- and microstructures of II–VI semiconductors, crystalline matrices with rare-earth activators and laser dyes with intentionally introduced scatterers [1–8]. Random lasing in the visible spectral range was achived at a limited set of wavelengths in microstructured active scattering media based on some II–VI semiconductors, in particular, CdZnO [9], ZnSe [10, 11], CdS [12], and CdSe [13]. Random white lasing in a mixture of semiconductor micropowders with the general formula ZnCdSSe was recently reported in [14]. Random lasers are promising for image visualisation, data transfer, labelling of materials, friend-or-foe recognition as biosensors in medicine and illumination technologies [1, 3, 4].

The position of narrow lines in random lasing spectrum often chaotically changes from one pump pulse to another, as was repeatedly observed previously [1, 14, 15]. It was also established that, under fixed experimental conditions with a specified above-threshold pump level, random lasing may either occur or be absent (with only intense photoluminescence instead of it). The purpose of the present work is to clarify the reasons for instability of random lasing in active scattering media and possible ways to eliminate it.

2. Experimental

The objects of study were microcrystalline powders of CdS compound and CdSSe solid solutions with submicron size of crystallites. Thin (~500 μm) layers of micropowders were deposited on polished planes of quartz plates. Optical pumping of random lasing in micropowders was performed by third-harmonic pulses of a nanosecond Nd^{3+}: YAG laser at a wavelength $\lambda = 355$ nm with a pulse repetition rate (PRR) of 10 Hz and by N_2-laser pulses at $\lambda = 337$ nm with an PRR of 700 Hz. The pump radiation was focused to a round spot on a sample of micropowder active medium. The radiation from micropowders was coupled to the monochromator input through an optical fibre. The micropowder emission spectra after each pump pulse were recorded by a CCD line array at the monochromator output. The monochromator spectral resolution in the measured wavelength range was 0.14 nm. Measurements were performed at 300 K.

3. Results and discussion

The most widespread pump sources for random lasers are solid-state Nd^{3+}: YAG lasers with pico- and nanosecond radiation at wavelengths of 266, 355, and 532 nm with a PRR ~10 Hz [1, 2]. Previously we reported some results on implementing random lasing in CdS micropowder upon excitation by single third-harmonic pulses of an Nd^{3+}: YAG-laser at $\lambda = 355$ nm [14].

In this study, the radiation of an Nd^{3+}: YAG pump laser (355 nm) was focused to a spot 400 μm in diameter on the surface of a CdS micropowder sample. A sharp increase in intensity, narrowing of the emission spectrum of CdS micropowder near $\lambda = 520$ nm and occurrence of a narrow-line structure in the spectral maximum were observed when the lasing threshold of 640 kW cm^{-2} was exceeded. The established signs of lasing occurrence in CdS micropowder without any deliberately introduced mirror cavities are typical indicators of the achievement of random lasing action in disordered active media with a high degree of scattering, which were revealed in a number of works [1, 2, 16]. Figure 1 shows a sequence of 15 emission spectra of CdS micropowder at a fixed pump

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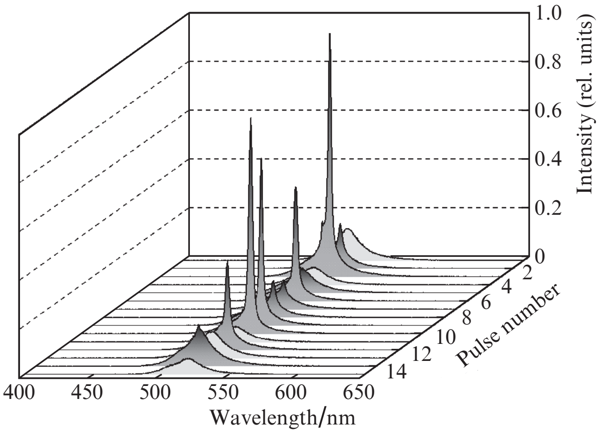

Figure 1. Photoluminescence (pulses 1, 4, 11, 13, and 15) and random lasing (pulses 2, 3, 5–10, 12, and 14) spectra for CdS micropowder upon excitation by 15 successive emission pulses of an Nd^{3+}:YAG laser ($\lambda = 355$ nm) with a power density of $\sim$1200 kW cm^{-2}.

power density of $\sim$1200 kW cm^{-2} and invariable experimental conditions. Random lasing in CdS micropowder was achieved for pump pulses 2, 3, 5–10, 12, and 14, as is evidenced by the presence of a narrow line with an FWHM of $\sim$7 nm in the emission spectrum. At the same time, only intense photoluminescence of micropowder was observed for the pump pulses 1, 4, 11, 13, and 15 with a power density of $\sim$1200 kW cm^{-2} (which is double as much as the lasing threshold).

The output pulse energy instability of the solid-state Nd^{3+}:YAG laser in the present work did not exceed 10%. Therefore, fluctuations of the excitation pulse energy could not reduce the pump level below the threshold (640 kW cm^{-2}). The observed random lasing instability is most likely caused by the energy redistribution within the excitation spot from one pump pulse to another, which leads to a change in the shape and number of randomly formed gain loops in the micropowder active medium. As a result, the number and length of gain loops may be insufficient to overcome the lasing threshold and, correspondingly, only intense photoluminescence occurs (Fig. 1, pulses 1, 4, 11, 13, and 15).

Thus, the random lasing action is extremely sensitivity to intensity fluctuations within the pump spot. Therefore, further studies were performed with a nitrogen pump laser, characterised by high stability of output radiation. Figure 2 shows the random lasing spectra of CdS micropowder pumped by an N_2 laser for two excitation spot diameters: 30 and 210 μm.

An evident difference between the lasing spectra of CdS micropowder in the range of 515–520 nm under excitation by a nitrogen laser and the lasing spectra obtained by pumping with a Nd^{3+}:YAG laser is the clear structure of narrow lasing lines with an FWHM of $\sim$1 nm (Fig. 2). The high stability of the N_2 laser output power and sufficiently uniform intensity distribution within the excitation spot provided a stable random lasing action in CdS micropowder at a PRR of 700 Hz: random lasing was achieved for all pump pulses exceeding the threshold, and the position of narrow spectral lines did not vary under fixed pumping conditions. A significant increase in the pump power density above the threshold led only to a slight intensity redistribution between the lines in the lasing spectrum of CdS micropowder (Fig. 2).

In addition, it turned out that the size of the excitation spot on the micropowder surface affects significantly the

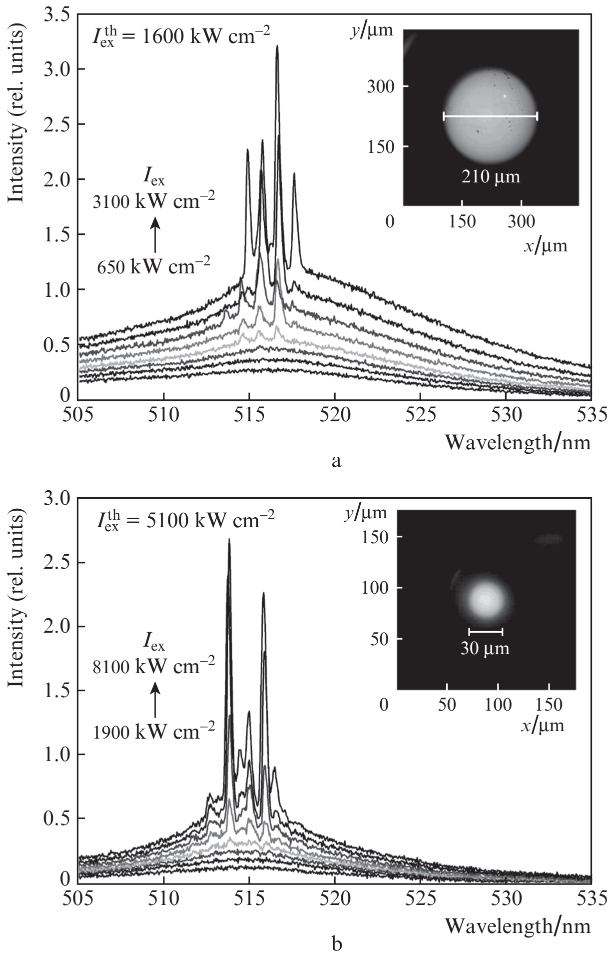

Figure 2. Emission spectra of CdS micropowder at a temperature of 300 K and different power densities I_{ex} of N_2 laser radiation ($\lambda = 337$ nm), focused to spots with diameters of (a) 210 and (b) 30 μm; I_{ex}^{th} is the threshold power density. The insets show the spatial power density distributions in excitation spots.

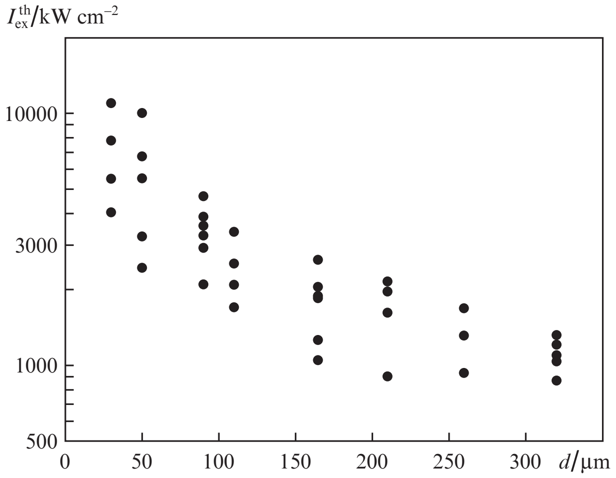

Figure 3. Dependence of the random lasing threshold in CdS micropowder on the excitation spot diameter under N_2 laser pumping (λ = 337 nm).

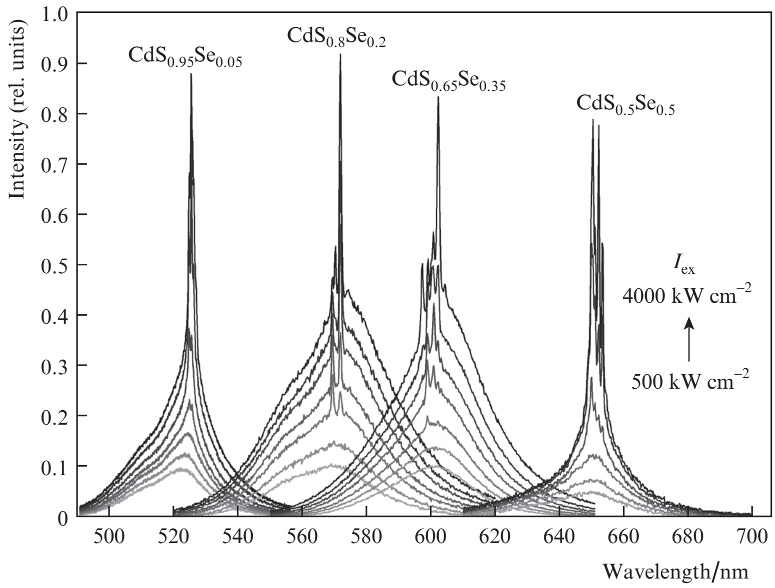

Figure 4. Emission spectra of CdSSe micropowders at a temperature of 300 K and different power densities I_{ex} of N_2 laser radiation (λ = 337 nm).

random lasing threshold. It was found that a decrease in the spot diameter from 210 to 30 μm (Fig. 2) leads to a rise in the lasing threshold from 1600 to 5100 kW cm^{-2}. Figure 3 shows how the spread of random lasing thresholds for different points of the irradiated CdS micropowder surface varies with a change in the excitation spot diameter from 30 to 320 μm. One can also clearly see a significant decrease in the lasing threshold with an increase in the excitation spot diameter (i.e., the size of active medium). This behaviour can be explained as follows. Due to the scattering in a larger volume of active medium, the stimulated emission has a longer path before leaving the medium (i.e., a larger length of gain loops) and, as a result, acquires a larger gain [17, 18]. This leads to a decrease in the threshold with an increase in the volume of active medium. Thus, the increase in the lasing threshold with a decrease in the excitation spot size testifies in favour of the mechanism of feedback formation specifically due to the occurrence of random gain loops in the active scattering medium.

Stable lasing action was also achieved in micropowders of ternary solid solutions CdSSe, whose advantage over the II–IV-type binary compounds is the possibility of smooth shift of the emission spectrum within the visible spectral range. The room-temperature emission spectra of the solid solutions pumped by an N_2 laser at λ = 337 nm are presented in Fig. 4.

$CdS_{0.95}Se_{0.05}$, $CdS_{0.8}Se_{0.2}$, $CdS_{0.65}Se_{0.35}$ and $CdS_{0.5}Se_{0.5}$ solid solutions exhibit intense photoluminescence in the green, yellow, orange and red spectral regions, respectively, under excitation by an N_2 laser with a power density of about 700 kW cm^{-2}. An increase in the pump level caused a sharp rise in the radiation intensity, narrowing of spectral bands, and occurrence of a clear structure of narrow emission lines in their maxima. As a result, random lasing at wavelengths of 525, 570, 600, and 650 nm was achieved (Fig. 4). The threshold pump power densities were 1100, 1300, 1200 and 1600 kW cm^{-2} for $CdS_{0.95}Se_{0.05}$, $CdS_{0.8}Se_{0.2}$, $CdS_{0.65}Se_{0.35}$ and $CdS_{0.5}Se_{0.5}$, respectively.

The results of this study may be useful in the development of random lasers with radiation in the visible spectral range (including white light), image visualisation systems, illumination technologies, etc.

4. Conclusions

Room-temperature random lasing in CdS micropowder at a wavelength close at about λ = 520 nm under excitation by 10-ns pulses of Nd^{3+} : YAG and N_2 lasers was achieved. It was shown that a pulse-to-pulse change in the nonuniformity of intensity distribution in the excitation spot plane under pumping by Nd^{3+} : YAG-laser radiation leads to instability of random lasing in CdS micropowder under fixed experimental conditions. The pumping by N_2-laser radiation with its high spatial homogeneity made it possible to achieve stable lasing in CdS micropowder, which was characterised by a clear structure of narrow lines in the range of 515–520 nm and stability of their spectral position in the pulsed mode with a repetition rate of 700 Hz. In addition, stable lasing was obtained for the first time in micropowders of $Cd_{0.95}Se_{0.05}$, $CdS_{0.8}Se_{0.2}$, $CdS_{0.65}Se_{0.35}$, and $CdS_{0.5}Se_{0.5}$ solid solutions at wavelengths of 525, 570, 600, and 650 nm, respectively, with threshold intensities ranging from 1100 to 1600 kW cm^{-2}.

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