Spectral and energy characteristics of resonatorless lasing in solutions of Rhodamine 6G with nanoparticles of various nature with femtosecond pumping

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Abstract. The results of measurements of the spectral and energy characteristics of cavity-free lasing in Rhodamine 6G dye solutions with Al nanoparticles irradiated with femtosecond laser pulses are presented. A comparison is made of the threshold generation values in a pure dye solution and in a dye solution with nanoparticles upon irradiation with femtosecond pulses.

1. Introduction

The development of laser emitters in recent years is associated with their significant miniaturization and the creation of highly efficient active media. One of these lasers is considered to be a stochastic laser, known in foreign literature as a random laser. The active medium of this laser is a composite of laser-active molecules of nanoparticles of various materials. According to the type of stimulated emission, a random laser can be divided into two classes [1]. The first of which is a random laser based on solutions of laser dyes with embedded nanoparticles of various substances. In such structures, the effect of multiple scattering by small inhomogeneities increases the residence time of stimulated emission photons in an excited active medium. The second class includes stochastic lasers, the principle of generation of which is based on the appearance of stimulated emission in the so-called microresonators, the mirrors of which are well-reflecting faces of microcrystals. Closed resonators are formed randomly during the action of the pump pulse. The first theoretical justifications for the possibility of obtaining laser radiation in randomly inhomogeneous active media were presented by Letokhov [2]. In this work, a possible mechanism for the formation of feedback in a medium with amplification due to strong scattering was proposed. In 1986, V. M. Markushev, this effect was experimentally demonstrated on samples of dielectric powders activated by neodymium ions. [3]. In 1994, Lawandy N. M. in [1] demonstrated isotropic laser emission in Rhodamine 640 (R640) solutions with scattering TiO₂ nanoparticles [1]. In the experiment [1], when a certain threshold pump energy is reached, the width of the spectrum of the secondary radiation sharply narrows from 70 nm to 4 nm.

Random lasers can be used in many fields of science and technology, such as laser lighting, the creation of sensors and displays, information technology, spectroscopy[4]. Among the advantages of a random laser, it is customary to consider the angular dependence, the possibility of tuning the wavelength in a wide spectral range, and the different aggregate state of the active medium (liquids, crystals, powders, gases). Among the disadvantages in comparison with other lasers is the lack of directivity and low spatial coherence, however, these properties give an advantage in some areas.
Solid-state lasers are most often used as pumps for random lasers, but the use of lamps and electric discharges is also known. Earlier in our experiments, we used a nanosecond solid-state laser as a pump. Our main objectives are to increase the efficiency of a random laser, namely, lowering the generation thresholds and increasing the output radiation power, in order to expand the scope of its application. One of the ways to achieve these goals, in our opinion, is to use femtosecond laser pulses.

Femtosecond radiation has a number of unique properties: high intensity, wide optical spectrum, which served as a strong incentive for the introduction of femtosecond radiation as a tool in scientific research, production, technology and medicine.

The works devoted to the study of luminescence processes of organic molecules in spherical particles under femtosecond excitation began, mainly, at the beginning of the XXI century. The possibility of stimulated emission in droplets with the Rhodamine 6G dye (concentration 10⁻³ mol / L) upon two-photon absorption of femtosecond pulses at a wavelength of 0.82 μm was studied in [5]. It was shown that at intensities of femtosecond pulses up to 10 W / sm² spontaneous luminescence is observed with a characteristic feature - the quadratic dependence of the luminescence energy on the pump energy.

It was also shown in [6] using the biological medium of the DsRed2 fluorescent protein during nano- and femtosecond pumping that femtosecond pumping of the medium reduces the threshold average pump intensity by more than two orders of magnitude compared to nanosecond pumping.

The use of gold nanoparticles in solutions with dyes during femtosecond pumping in the range of wavelengths 850–900 nm was demonstrated in [7]. The results of the work show that a change in the diameter of gold nanoparticles leads to a shift in the generation peak due to the plasmon resonance effect. A decrease in the diameter of the nanoparticles leads to a shift of the peak to the short-wavelength region.

At the moment, there are not many experimental works in the literature using solutions of fluorescent dyes with nanoparticles when irradiated with femtosecond laser pulses. This suggests that this area is still poorly understood. Based on this, the goal of this work is an experimental study of the fluorescence of a dye with nanoparticles upon irradiation with femtosecond laser pulses.

2. Experimental setup
The experiment was conducted on the installation, the scheme of which is shown in Figure 1.

![Figure 1. The scheme of the experimental setup: 1 – Ti: Sa laser, 2 – diaphragm, 3 – power meter, 4 – drop with an active medium, 5 – fiber, 6 – spectrometer, 7 – PC](image-url)

Femtosecond radiation (λ≈0.8 μm, t≈50 fs) passed through aperture 2 (d = 8 mm) and was directed to an Ophir 3 power meter, then the power meter was removed and the radiation fell into a droplet with an active medium (Rhodamine dye 6G in dibutyl phthalate + nanoparticles) 4. Secondary
radiation from the droplet was transferred to the AvaSpec 6 spectrometer using a fiber 5 and using the software, secondary radiation spectra were output to computer 7.

3. Results

Figure 2 shows the luminescence spectrum of a dye solution at a laser pulse energy of 5 mJ.

![Figure 2. The luminescence spectrum of the dye solution at a laser pulse energy of 5 mJ](image)

Based on the obtained spectra, we plotted the dependence of the luminescence intensity of the solution on the laser pulse energy in relative units (Figure 3).

![Figure 3. Dependence of the luminescence intensity of pure dye solution (a) and dye solution with nanoparticles (b) on the laser pulse energy in relative units (1), quadratic dependence (2)](image)

Figure 3a shows that, up to a pump energy of 6 mJ (0.45 rel. units), the curve has the form of a typical quadratic dependence, which indicates spontaneous luminescence at two-photon absorption, then at energies above 6 mJ (0.45 rel. units) stimulated luminescence arises during two-photon absorption, which is explained by the transition of the energy dependence from a quadratic character to a dependence with increasing growth rates [8]. After a pump pulse energy of 10 mJ (0.7 rel. units) and more, the destruction of the solution begins.

From previous experiments [9] with nanosecond pump pulses, we know that when nanoparticles are added to a dye solution (Rhodamine 6G), the stimulated emission thresholds decrease. In this work, we also performed an experiment with a solution of the Rhodamine 6G dye with Al nanoparticles with a concentration of 0.625% by volume (Figure 3b).
4. Summary
From Figure 3b and from the conclusions made on the basis of the analysis of experimental data obtained in solutions with a pure dye, it can be seen that, at the used pump pulse energies, spontaneous luminescence is not observed, and stimulated emission immediately begins. Currently, work in this area continues in the direction of decreasing the energy of the laser pump pulse.

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