

Temperature effects on the conversion coupling efficiency in dye-based plasmonic random laser gain media

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Original Research

Received:

24 October 2024

Revised:

29 November 2024

Accepted:

4 December 2024

Published online:

10 February 2025

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Abstract:

The impact of temperature on the conversion coupling efficiency between Rhodamine 6G (Rh6G) dye and hybrid nanoparticles, composed of gold (Au) and copper (Cu), and its influence on the performance of random lasers is investigated. The study focused on the interaction between the photophysical properties of Rh6G dye molecules and the plasmonic and thermal effects of Au/Cu nanoparticles (NPs) at varying temperatures. We analyzed the interaction between the dye molecules and nanoparticles as a function of pumping energy and temperature focusing on laser parameters laser threshold, full width at half maximum (FWHM), and peak intensity. Our results show that increasing pumping energy and temperature significantly affects the FWHM's narrowing, and peak intensity enhancement. We found that with increasing pumping energy, the FWHM narrowed to about 8 nm for Au and Cu nanoparticles, and the peak intensity was enhanced to about 40,000 a.u. for Au NPs and 28,000 a.u. for Cu NPs. While, we found that with increasing temperature, the FWHM decreased to about 0.6 nm for Au NPs and 0.8 nm for Cu NPs, and the peak intensity increased to about 5400 a.u. for Au NPs and 9400 a.u. for Cu NPs. This study provides insight into optimizing random laser performance through temperature control, potentially advancing the development of tunable photonic devices.

Keywords: Random laser; Rhodamine 6G; Copper nanoparticles; Gold nanoparticles; Temperature; Coupling efficiency; magneto-plasmonic

1. Introduction

Random lasers represent a unique class of light-emitting devices that differ fundamentally from conventional lasers by lacking a well-defined resonator [1, 2]. Instead, their feedback mechanism is provided by multiple scattering events within a disordered gain medium, leading to coherent or incoherent light emission [3]. The inherent randomness of these systems offers potential advantages in terms of simplicity, cost-effectiveness, and the generation of complex, multimode emission spectra by using nanostructures or nanoparticles as scatter points.

Using metallic nanoparticles as scattering centers in random lasers has become an area of intense research, offering unique opportunities to manipulate and control laser emission. Among these, metal nanoparticles, particularly nano gold (Au) [4, 5] and nano copper (Cu) [6] have garnered significant attention due to their exceptional optical and thermal properties, driven by surface plasmonic phenom-

ena (SPR) [5]. SPR is an optical technique used to detect molecular interactions [7], biological species [8], virus [9], DNA [10] and drugs detection [11, 12] and so on.

Metallic nanoparticles, (Au NPs) and (Cu NPs), have emerged as promising candidates due to their strong scattering properties and the ability to induce localized surface plasmon resonances (LSPRs) [13, 14]. These NPs are effective scatters and exhibit notable photothermal properties [15], which can be leveraged to modulate the laser output through external stimuli such as temperature [16]. Due to LSPRs in these NPs, the local electromagnetic field enhances around the nanoparticles [17] thus leading to increased scattering efficiency and, consequently, light amplification within the gain medium. They exhibit strong SPR effects, though their resonance frequencies differ due to variations in their dielectric functions and the size of particles and also efficient effect by thermoplasmonic impact in them. Since the scattering centers are plasmonic nanoparticles, they can absorb some of the pump energy

and convert it into heat through the thermoplasmonic effect. These differences enable tunable optical properties, allowing for the customization of random lasers across a wide spectral range [18–20].

This work investigates thermoplasmonic effect of the random lasing behavior using Rhodamine 6G (R6G) as the active medium contains laser-generated Au NPs and Cu NPs. The choice of R6G is motivated by its well-known high quantum yield and compatibility with visible light excitation, making it an excellent gain medium for visible-range lasing [21, 22]. The first type is Plasmonic nanoparticles such as Au NPs and the second type Cu NPs to achieve a thermoplasmonic phenomenon [23] within the active medium. The results showed that the thermo effect appears in the active media with Cu NPs, while the plasmonic effect in Au NPs was better than in the active media with Cu NPs. The impact of these two phenomena was clear on the emission spectra of random laser in terms of laser threshold, FWHM, and peak intensity [24].

2. Experimental setup

In this work, we prepared Rh6G dye solution by dissolving the dye in ethanol solvent at a suitable concentration to achieve optimal fluorescence (The concentration of Rh6G was adjusted to 5×10^{-5} mol/L to optimize the fluorescence output while minimizing reabsorption effects). Separate solutions of gold and copper nanoparticles were prepared using laser ablation technique (first harmonic of Q-switched Nd:YAG) by dispersing the nanoparticles in distilled water supplemented with Polyvinylpyrrolidone (PVP) polymer [17] (ratio 1:100 mL) to enhance the stability and homogeneity and reduce the oxidation process of the nanomaterials. The concentration of nanoparticles was controlled by setting a fixed pulse energy (500 mJ for Au NPs & 600 mJ for Cu NPs) and frequency (10 HZ) for a specific period (4

min for Au NPs & 8 min for Cu NPs). To build a random laser system, we mixed the prepared nanoparticle solutions with the Rh6G dye solution and then the mixture was placed into a transparent quartz cuvette for optical measurements. We used a pulsed Nd:YAG laser operating at 532 nm as the pumping source to excite the mixture (Fig. 1).

We equipped the sample cell with a temperature control system (Peltier element with water bath) to control the sample temperature [25]. This system allowed us to systematically change the temperature of the sample, where the temperature was monitored using a temperature sensor (thermal camera). The sample temperature was controlled using the mentioned heating system, with a temperature range of 30 °C to 50 °C. A thermal camera (FLIR C2 thermal imager) was used to monitor the temperature in real-time, with an accuracy of ± 2 °C and thermal sensitivity < 0.10 °C. The emission spectrum of the random laser was collected using an optical fiber coupled to a high-resolution spectrometer.

3. Results and discussion

We initially performed the measurements at room temperature and then took regular temperature changes on the same sample. We recorded the laser output for each temperature setting to monitor changes in peak intensity, FWHM, and threshold behavior. We collected data at 5 °C intervals to evaluate the temperature dependence of the random laser output. The absorption spectra of Rh6G with concentrations of (5×10^{-5} mol) dissolved in ethanol are recorded as shown in figure 2 (a).

Figure 2 (b) shows the absorption spectra of the R6G dye and the Au & Cu nanomaterials separately. We noticed that the highest absorbance of the dye is at wavelength 531 nm, the highest absorbance of the Au NPs is at wavelength 535 nm, and the highest absorbance of the Cu Nps is at wavelength 298 nm. In addition, figure 2 (c) shows the

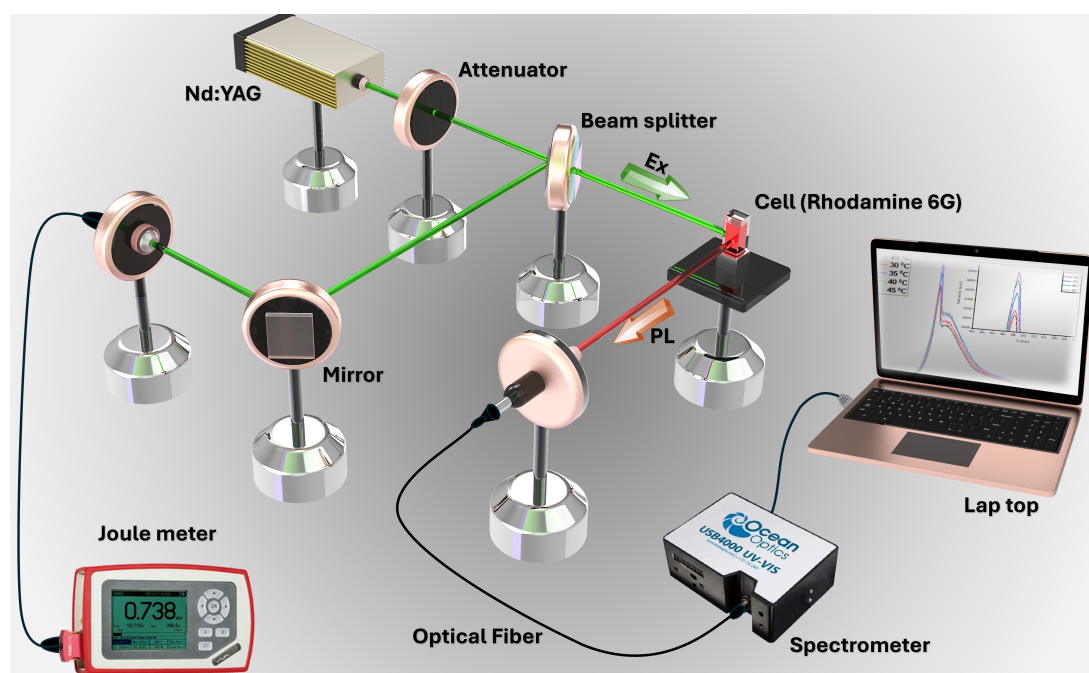


Figure 1. The Schematic diagram of the experimental setup to produce NPs and record random lasing.

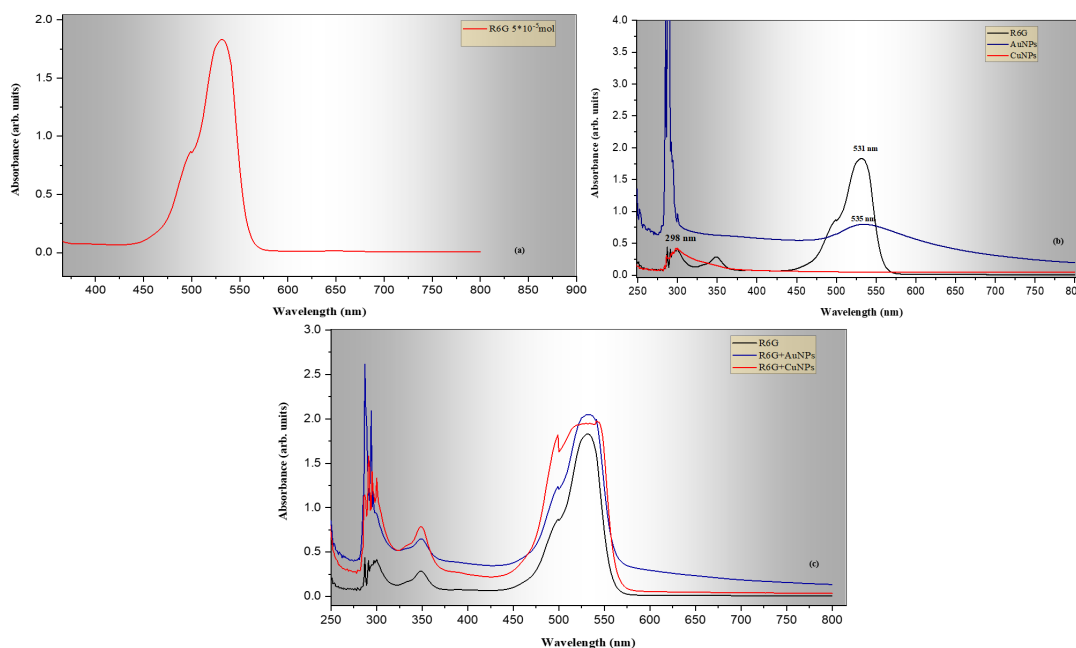


Figure 2. Absorption spectra of (a) Rh6G dye dissolved in ethanol with concentration (5×10^{-5} mol), (b) Rh6G, Au and Cu NPs, and (c) Rh6G with Au and Cu NPs as a mixture.

UV–visible absorption spectra of Rh dye only and the absorption spectrum of Rh dye with nanogold once and with nanocopper another time [26]. It is observed that the intensity increased when adding Au and Cu NPs in (each one individually) Rh6G dye, noting that the intensity of adding gold nanoparticles is greater than in the case of adding copper nanoparticles. This can be attributed to the plasmonic enhancement caused by the Au NPs. The difference in intensity between the addition of Au and Cu nanoparticles is due to their distinct plasmonic properties and interaction efficiency with the dye molecules [27]. The strong LSPR of Au NPs leads to more efficient coupling with the dye molecules, resulting in less quenching and more effective fluorescence enhancement of Rh6G. We observed that Au NPs achieved an absorption efficiency of more than 80%, while Cu NPs peaked at approximately 40% for the same concentration. We also notice that after mixing the nanomaterials with the dye, the absorption efficiency becomes

somewhat similar.

Figure 3 shows the effect of pumping energy on the RL emission spectrum at room temperature for gold and copper nanoparticles. Emission intensity is found to linearly increase with the laser pumping energy for both nanomaterials but with a different spectral narrowing. The enhanced localized electromagnetic field (EM) in the vicinity of metal nanostructures may enhance the density of pump light available for the gain media and consequently may increase the probability of the R6G dye molecules that are to be excited simultaneously to the higher energy levels. Au NPs can affect the radiative rate of fluorophores [27, 28] higher than Cu NPs. The peak intensity of the random laser spectrum is evident at high pumping energies. When metallic NPs are excited resonantly, they scatter the energy of emitters with the greater scattering cross sections, and then easily lead to the occurrence of spectral narrowing random lasing [29].

Figure 4 shows the intensity of the emission spectrum of a

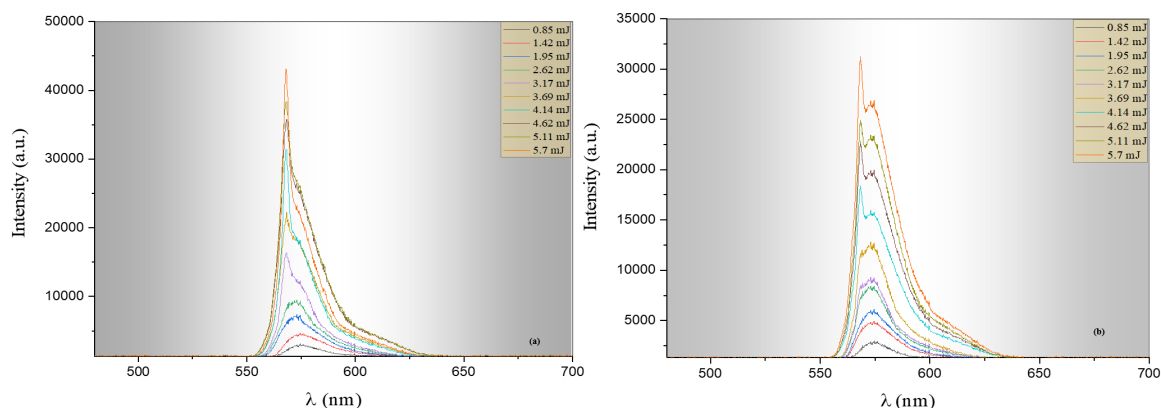


Figure 3. Effect of pumping energy on the emission spectrum of RL at constant temperature (room temperature) for two different nanomaterials (a) Au NPs and (b) Cu NPs.

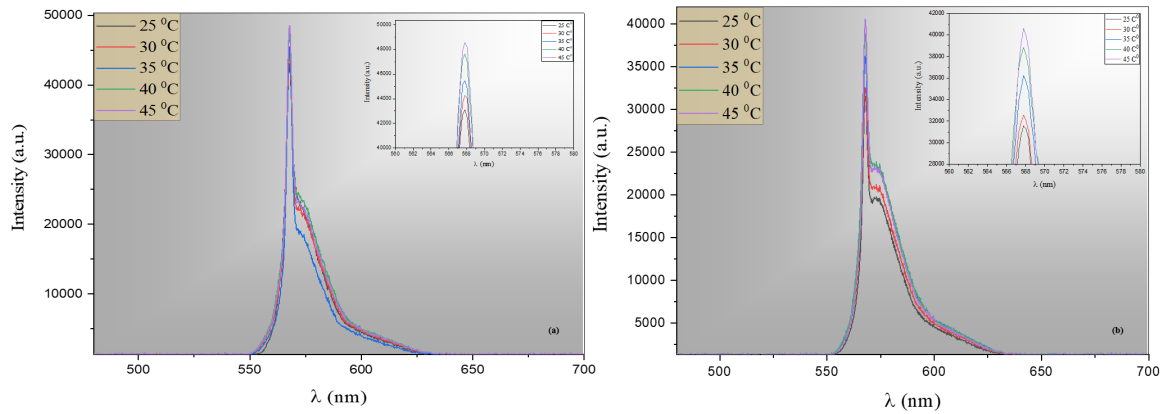


Figure 4. Effect of temperature on the emission spectrum of RL for two different nanomaterials (a) Au NPs and (b) Cu NPs.

random laser as a function of wavelength after measuring it for several changes in the temperature of the solution (active medium). As the temperature of the solution increases, the refractive index of water will decrease, which may lead to a change in the SPR of the gold nanoparticles (Au NPs). This change in the plasmon resonance can lead to a high coupling [148] between the energy levels of Rh6G and Au NPs. With this enhanced coupling, the energy transfer from the excited dye molecules to Au NPs can become more efficient, decreasing the non-radiative relaxation pathways and increasing the radiative recombination of the dye molecules. As a result, the intensity of the emission spectrum of Rh6G in the presence of Au NPs increases with increasing temperature.

Temperature changes can change the refractive index of the surrounding medium, modify the electron density in nanoparticles, and affect thermal diffusion within the laser medium [29]. Figure 5 shows the intensity of the emission spectrum of a random laser as a function of wavelength after measuring it for several changes in the temperature of the solution (active medium). Increasing the temperature of the solution leads to the thermal expansion of the copper nanoparticles (Cu NPs) and this changes their shape and size which affects their scattering properties. This change leads to an increase in non-radiative decay processes which results in a decrease in fluorescence emission. This can directly affect the gain of the random laser since fluores-

cence is essential for the stimulated emission process. As the fluorescence intensity decreases with temperature, the output intensity of the random laser decreases accordingly due to a decrease in the optical gain. These effects can lead to changes in the intensity and FWHM of the laser output. FWHM decreases from 14 to 6 nm when the pumping source energy increases from 0.85 to 5.7 mJ for Au NPs as shown in Fig. 5 (a). FWHM decreases from 16 to 8 nm when the pumping source energy increases from 0.85 to 5.7 mJ for Cu NPs as shown in Fig. 5 (b). The same goes for the peak intensity, as it was found that increasing the pumping source energy leads to an increase in the peak intensity from 3222 to 43000 (a.u.) when the energy increases from 0.85 to 5.7 mJ for Au NPs as shown in Fig. 5 (a). Also when increasing the pumping source energy, the peak intensity increases from 2575 to 31000 (a.u.) when the energy increases from 0.85 to 5.7 mJ for Cu NPs as shown in Fig. 5 (b).

We observe the behavior of random laser properties under the influence of temperature once when we use Au NPs with the dye and another time when we use Cu NPs with the dye, where in both cases the peak intensity decreases and FWHM increases with increasing temperature. It was found that increasing the temperatures leads to a decrease in FWHM from 6 to 5.4 nm when the temperatures increase from 25 to 45 °C for Au NPs as shown in Fig. 6 (a). Also, when increasing the temperatures, the FWHM decreases

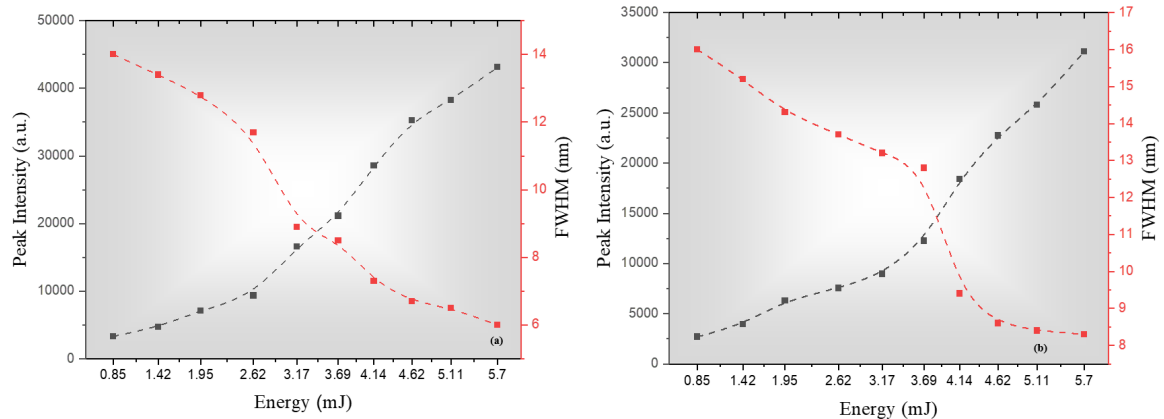


Figure 5. Represents the peak intensity and FWHM as a function of different pumping source energy (a) R6G + Au Nps and (b) R6G + Cu NPs.

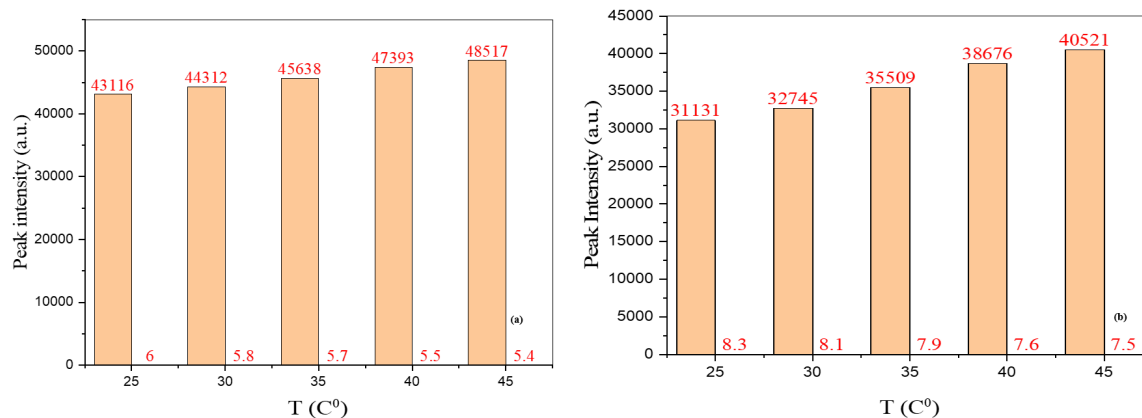


Figure 6. Peak intensity as a function of FWHM at different temperatures. (a) Au NPs and (b) Cu NPs.

from 8.3 to 7.5 nm when the temperatures increase from 25 to 45 °C for Cu NPs as shown in Fig. 6 (b). The same goes for the peak intensity, as it was found that increasing the temperature leads to an increase in the peak intensity from 43116 to 48517 (a.u.) for Au NPs and from 31131 to 40521 (a.u.) for Cu NPs when the temperature increases from 25 to 45 °C as shown in Fig. 6 (a, b). These changes come from enhanced in velocity and kinetic energy of NPs with temperature enhancement which yield more scattering and thus better laser actions in the main medium.

In addition, because Au NPs fabricate uniformly in the comparison with Cu NPs with laser ablation in liquids by Nd:YAG laser, we have more efficiently RL in the gain media with Au NPs from intensity and FWHM view point. Mainly as shown in Fig. 3, the SPR of Au NPs appear in the green region which is the main pumping wavelength in RL gathering process.

4. Conclusion

In this study, the effect of temperature and pumping energy on the conversion coupling efficiency between Rhodamine 6G dye in the presence of Au/Cu nanoparticles and the performance of random lasers was investigated, focusing on key parameters such as the laser threshold, FWHM and peak intensity. The results showed that the thermal effect was observed in the active media with Cu NPs, while the plasmonic impact on Au NPs. As the temperature increased (from 25 to 45 °C), notable shifts in the laser threshold were observed, attributed to temperature-induced changes in the optical properties of the Au/Cu nanoparticles, such as thermal expansion and plasmonic resonance. These changes impacted the light scattering and absorption within the gain medium, which subsequently altered the feedback mechanism of the random laser system. Our study revealed a narrowing of the FWHM at higher temperatures (6 to 5.4 nm) for Au NPs and (8.3 to 7.5 nm) for Cu NPs, and increasing in the peak intensity from 43116 to 48517 (a.u.) for Au NPs and from 31131 to 40521 (a.u.) for Cu NPs. This indicates an enhancement in the coherence of the laser output, likely due to improved coupling efficiency between the nanoparticles and the dye molecules.

Similarly, variations in pumping energy (0.85 to 5.7 mJ) significantly influenced the random laser's output

characteristics. Higher pumping energies reduced the FWHM from 14 to 6 nm for Au NPs and 16 to 8 nm for Cu NPs. Also, the peak intensity was enhanced to about 40,000 a.u for Au NPs and 28,000 a.u for Cu NPs, indicating a more efficient energy transfer between the Rh6G and the nanoparticles. This suggests optimizing the pumping energy in conjunction with temperature control can improve random laser performance with enhanced emission intensity and reduced lasing thresholds.

Authors contributions

Mariam Kadhim Jawad do the experimental part and write first version of the manuscript, Saddam Flayeh Haddawi measure the RL, investigation and help in the manuscript writing process, Jassim Mohammed Jassim advised all of the parts and Seyedeh Mehri Hamidi supervised the work and edit final version of the manuscript.

Availability of data and materials

The data that support the findings of this study are available from the corresponding author, upon reasonable request.

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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