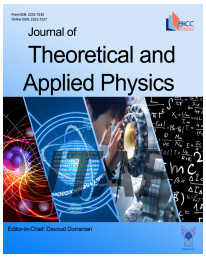

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Robust mouse tissue imaging by plasmonic random laser

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Original Research

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Abstract:

Given the major applications of optical random laser in the next generation devices, tissue imaging is proposed in this article by the aid of plasmonic random laser media. For this purpose, we use Rhodamine 6 G as the main gain medium and fill it by Gold nanoparticles, graphene and the mixture of them as random laser generator under the Nd:YAG's second harmonic and use them in the imaging of mouse tissue. For this purpose, Nd:YAG laser with the first harmonic select to produce nanoparticles for 4 minutes' exposure times and the second harmonic of the laser practice as the pump light to collect the random lasing. In the 45-degree arm, the mouse tissue puts as the object and the transmitted random lasing after the tissue collect by spectrometer. Our results show good random laser emission at the maximum of 3.69 mJ pumping power and thus resolution in the imaging recording from the tissues. This low cost laser medium can suggest to next generation of imaging systems based on the plasmonic random lasers.

Keywords: Laser ablation in liquid; Plasmonic random laser; Nanoparticles; Mouse tissue

1. Introduction

Random lasers (RL) as new type of coherent light generator uses disordered scattering medium like as metallic [1], magnetic [2], dielectric [3], core shells [4] nanoparticles (NPs), semiconductor powders [5], semiconductor polycrystalline thin films [6], covered optical fibers [7], and polymers [8, 9]. These lasers have a number of potential advantages over traditional ones, including lower cost and simpler construction with potential applications in areas such as imaging, sensing, telecommunications, biomedical and so on [10]. Among these new kind of lasers, use of plasmonic NPs as scattered points to tune the spectral response generate significant interest due to their improved performance [11]. This owes to the greater scattering strength and enhanced local field offered by the plasmonic scatterers. However, absorption losses and fluorescence quenching are the major setbacks in plasmonic random lasers, until now, the design of plasmonic random lasers with low threshold and enhanced emission intensity continues to be a challenging area of research [1]. Furthermore, control the threshold power and also emission intensity are two main challenges in RL design and construction which go ahead by using different materials and structure like as plasmonic core shell NPs in the dye host medium [12], polymeric film doped with laser dye [13], light emitting polymers [14], silver nanowires in multilayer construction [1], gold nanoparticle array coated with a gain medium [15, 16] and so on.

Based on low cost and efficient properties of above mentioned RLs, researchers noted that the plasmonic random laser could be useful for applications such as sensing, on chip telecommunications and also imaging [17, 18]. Their applications in imaging fields have been studied extensively in recent years for fluorescence imaging of biological samples by the aid of dye-doped polymer film as the scattering medium [19] or non-invasive detection of skin cancer by TiO_2 NPs [20] and so on. In this new field, the broadband emission spectrum of RLs allows for the simultaneous excitation of multiple fluorophores, enabling multiplexed imaging. This is particularly useful in biological imaging, where multiple biomarkers or targets need to be visualized simultaneously. Additionally, the random scattering medium in a RL can enhance the imaging depth and resolution [21]. The multiple scattering events within the medium help to diffuse and spread the emitted light, reducing the effects of scattering and improving imaging depth [22]. This can

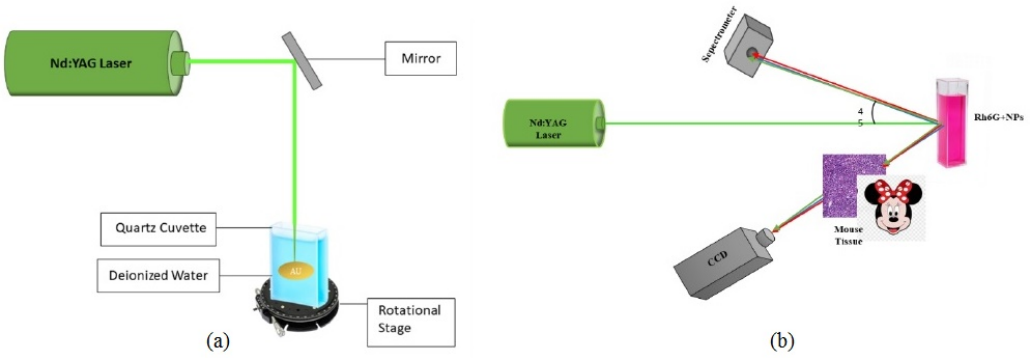

Figure 1. The schematic diagram of (a) the laser ablation in liquids process and (b) imaging by RL process.

be advantageous for imaging deep tissues or samples with high scattering properties with lowest damage and cost effectiveness which drifts us to use different scattering points to record imaging from the mouse tissue by RL [23, 24].

2. Experimental part

To get efficient RL and use of this emission as the main light in tissue imaging, we fabricate three different gain medium based on Rhodamine 6G as a host medium and gold NPs, graphene NPs and also the mixture of them [25]. These NPs were fabricated by laser ablation in liquids method by the aid of first harmonic generation of Nd:YAG laser (as shown schematically in Fig. 1 in the diluted water solution [26] and then mix with 10^{-4} and 10^{-5} M of the host dye.

After that, Rhodamine dye R6G was dissolved in an ethanol solvent to form a solution with different concentrations $(1 \times 10^{-4}, 1 \times 10^{-5})$ mol/m^3, then the gold nanoparticles were mixed with the dye solution to prepare the main gain medium in a ratio of (1:3), that is, once a volume of 3 mL of dye with a concentration of 1×10^{-4} mol/m^3 with 1 mL of gold nanoparticles and again a volume of 3 mL of dye with a concentration of 1×10^{-5} mol/m^3 with a volume of 1 mL of nanoparticles.

In the second step, we use second harmonic of Nd:YAG laser as the pump light due to the main absorption region of gold NPs in RL process onto the Rh6G dye plus gold, graphene and the mixture of them. The pulsed laser is focused on the gain medium and the detector is positioned at a 45° angle in the direction of the pump beam and RL occurrences and emissions were recorded and analyzed by the spectrophotometer software on the computer as shown schematically in Fig. 1(b). In the parallel way, we use the 100 and 200 micrometers mouse tissue which were cuted and fixed onto the glass substrate and put it in front of output

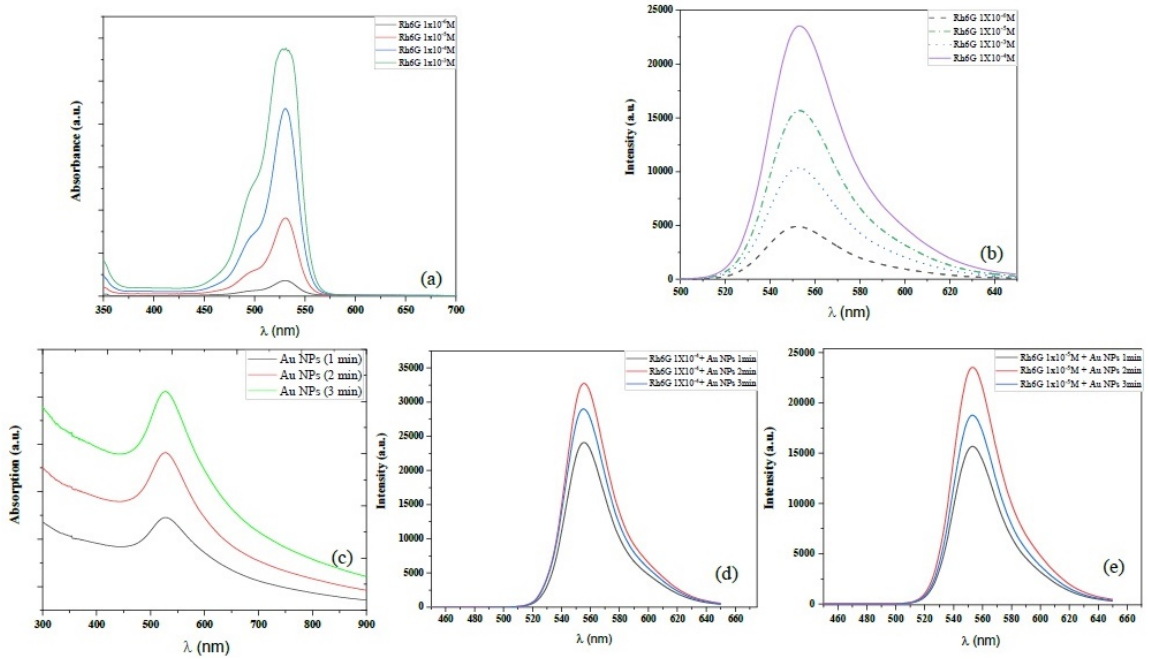

Figure 2. (a) Absorption spectra of Rh6G dissolved in ethanol with different concentration, (b) Rd6G fluorescence emission spectra at different concentrations, (c) Au Nps absorption spectrum at different time periods, (d) R6G fluorescence spectrum of dye concentration $(1 \times 10^{-4}$ M) mixed with different concentrations of nanoparticles and (e) R6G fluorescence spectrum of dye concentration $(1 \times 10^{-5}$ M) mixed with different concentrations of nanoparticles.

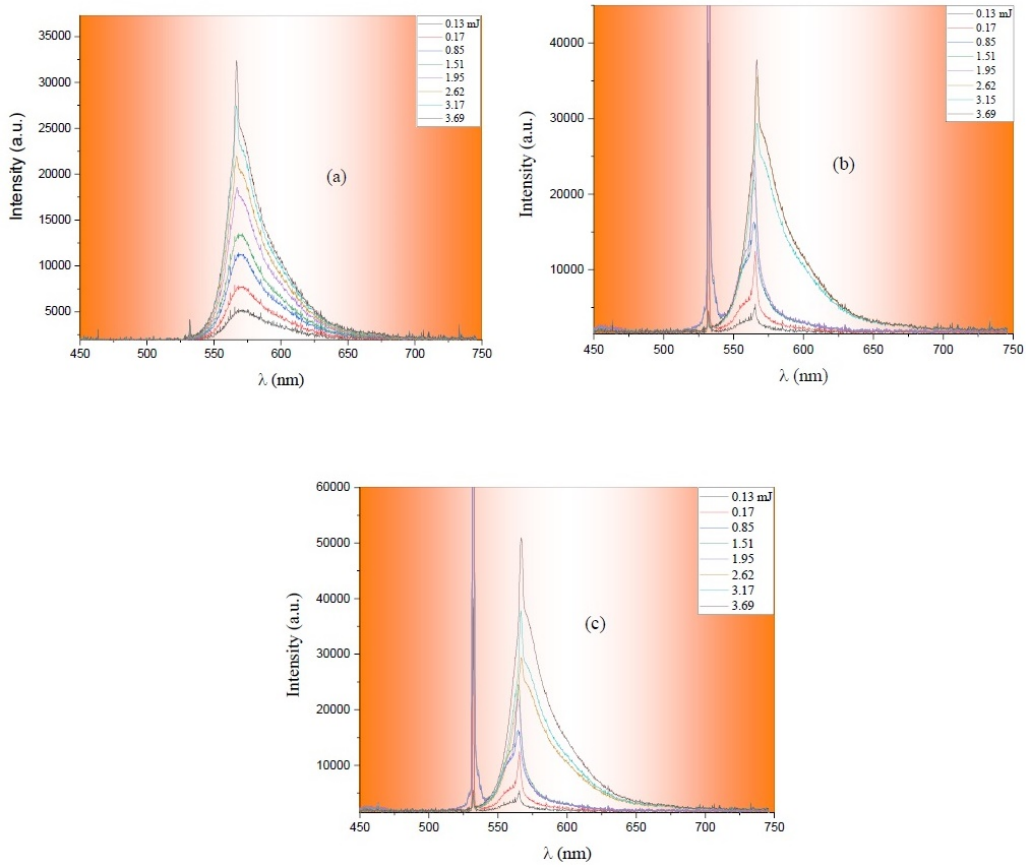

Figure 3. Random laser spectra of Rh6G dye at a concentration of 1×10^{-4} M with (a) G, (b) Au, (c) Au-G NPs

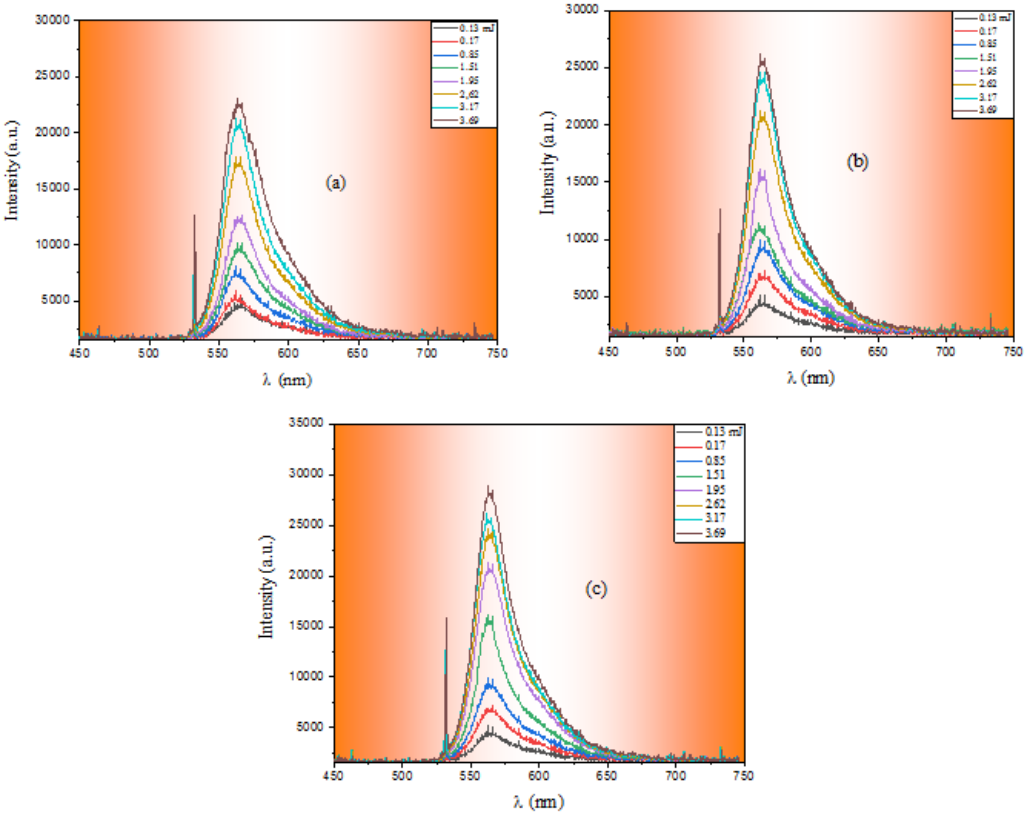

Figure 4. Random laser spectra of Rh6G dye at a concentration of 1×10^{-5} M with (a) G, (b) Au, (c) Au-G NPs.

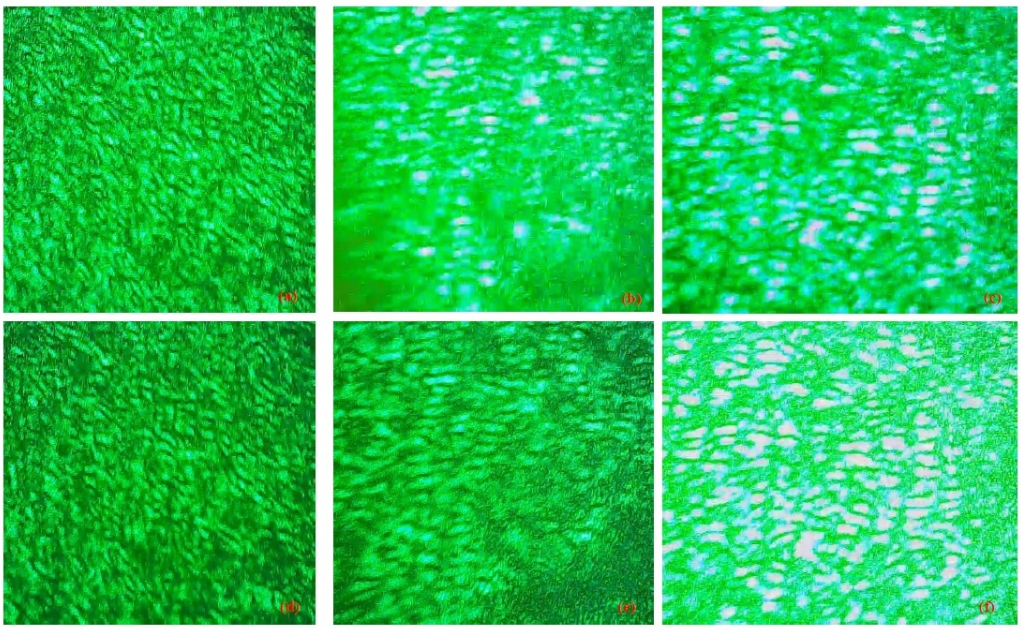

Figure 5. CCD image of mouse tissues; (a), (b) and (c) for tissue 1 and (d), (e) and (f) for tissue 2 by random laser from Rh6G dye with Au NPs at a concentration of 1×10^{-4} M and for different energies (low, medium, high), respectively.

RL from the solution. Main three extracted colors from each images which recorded by CCD camera was used as the main comparison tools in imaging process in accordance with the RL which recorded in different incidence power.

3. Results and discussion

To characterize produced NPs, at the first step, the absorption spectra of Rh6G dye with different concentrations (1×10^{-6}, 1×10^{-5}, 1×10^{-4}, 1×10^{-3} M) which dissolved in ethanol have been studied, as shown in Figure 2.

Based on this figure, the dye concentration 1×10^{-4} M showed the best fluorescence spectrum and thus reinforced our selection of it as the best suitable concentration for random laser action on the basis of its absorption spectrum as shown in Fig. 2(b). Moreover, increasing the dye concentration shifts the fluorescence spectrum towards longer wavelengths. In addition, absorption spectra of Au samples were produced in three different ablation times measured using a UV-Vis spectrophotometer and their behavior is shown in Fig. 2(c).

To find the best concentration of dye and Au NPs, two concentrations of dye R6G (1×10^{-4}, 1×10^{-5} M) mixed with three classes of gold NPs and then the concentrations were taken to measure the fluorescence of the sample under continuous pumping, 532 nm. The emission spectrum of different concentrations of Au NPs mixed with a solution of Rh6G dyes and at different concentrations (1×10^{-4}, 1×10^{-5} M), shows the best emission spectrum of R6G with as-prepared Au NPs with the time period 2 min because they have the highest emission intensity, which means it can be used as a gain medium in random laser beams as shown in Fig. 3(d,e). Now after selection the best concentrations, we record the RL from the main gain medium by incidence energy, from 0.13 to 4.14 mJ, for sample with graphene NPs, gold NPs, graphene plus gold NPs as respectively shown in Figs. 3(a) to (c) which indicate the appearance of random and incoherent laser beams, where the upper part of the spectrum represents closed paths, a state (steady wave) occurring within the random medium.

Because the main pumping pulsed laser in RL recording fit to the gold NPs plasmonic absorption spectrum, we can see very fine and nice RL peak in the main fluorescence spectrum of the host dye medium. In addition, it is evident that we have lower full width at half maximum (FWHM) when we use the mix of gold and dye Rh6G NPs (Fig. 3(b)) by this fact that the threshold power in lasing decreases in this sample also. Furthermore, in the sample with mix NPs, we have more emission intensity with the same incidence energy in the comparison with other ones. In the same manner, we record the RL from above mentioned three gain medium in s 10^{-5} M concentration of the dye shown in Fig. 4.

When comparing the first and second concentrations, we note that the first concentration (10^{-4} M) has a higher intensity and a lower threshold than the second concentration (10^{-5} M) for the same conditions.

Now after RL recording and also in the same time, we record the images of mouse tissue by the CCD camera for two different tissues with 100 and 200 micrometer thicknesses and 10^{-4} M in the low (0.13 mJ), middle (1.51 mJ) and high (3.69 mJ) pumping energy respectively as shown in Figure 5.

Figures 5 (a) to (c) shows the CCD image of mouse tissue with 200 micrometer thickness for low, middle and high pumping intensity and the three others for the tissue with 100 micrometers. It is obvious that by enhanced in the pumping power and thus appearance more small peaks as random lasers in the main fluorescence spectrum, we have more hot spot and resolution in the mouse tissue imaging. Furthermore, the imaging of two tissues were recorded in

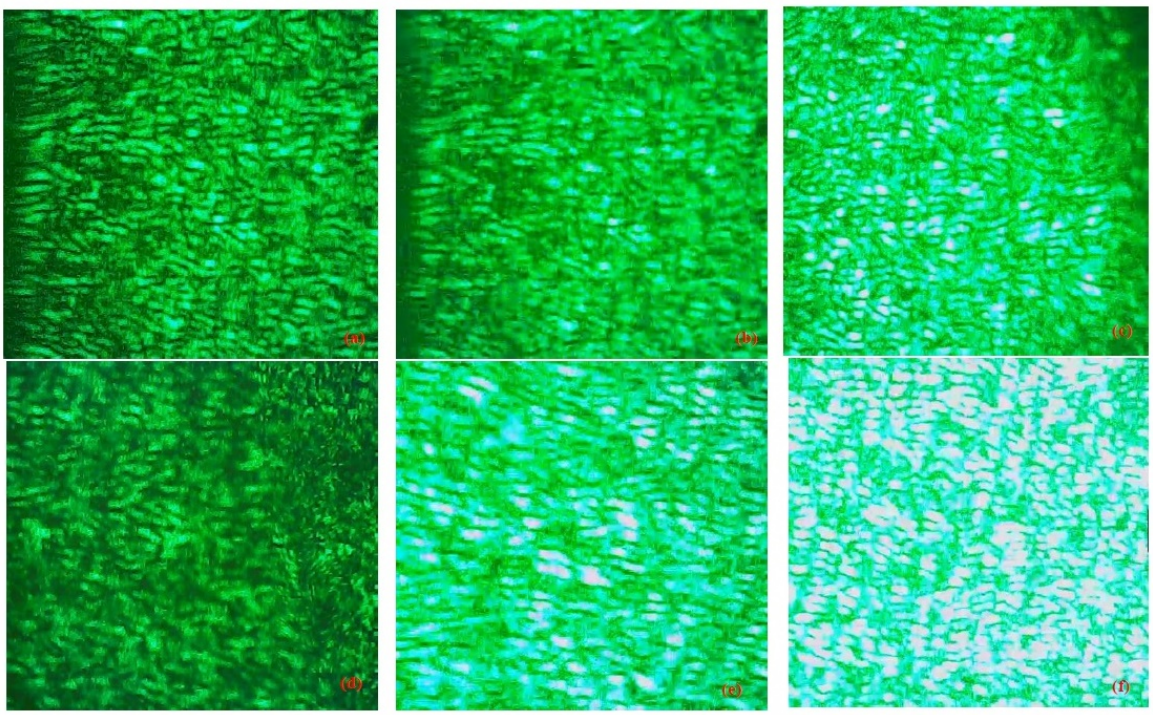

Figure 6. CCD image of mouse tissues; (a), (b) and (c) for tissue 1 and (d), (e) and (f) for tissue 2 by random laser from Rh6G dye with Au NPs at a concentration of 1×10^{-5} M and for different energies (low, medium, high), respectively.

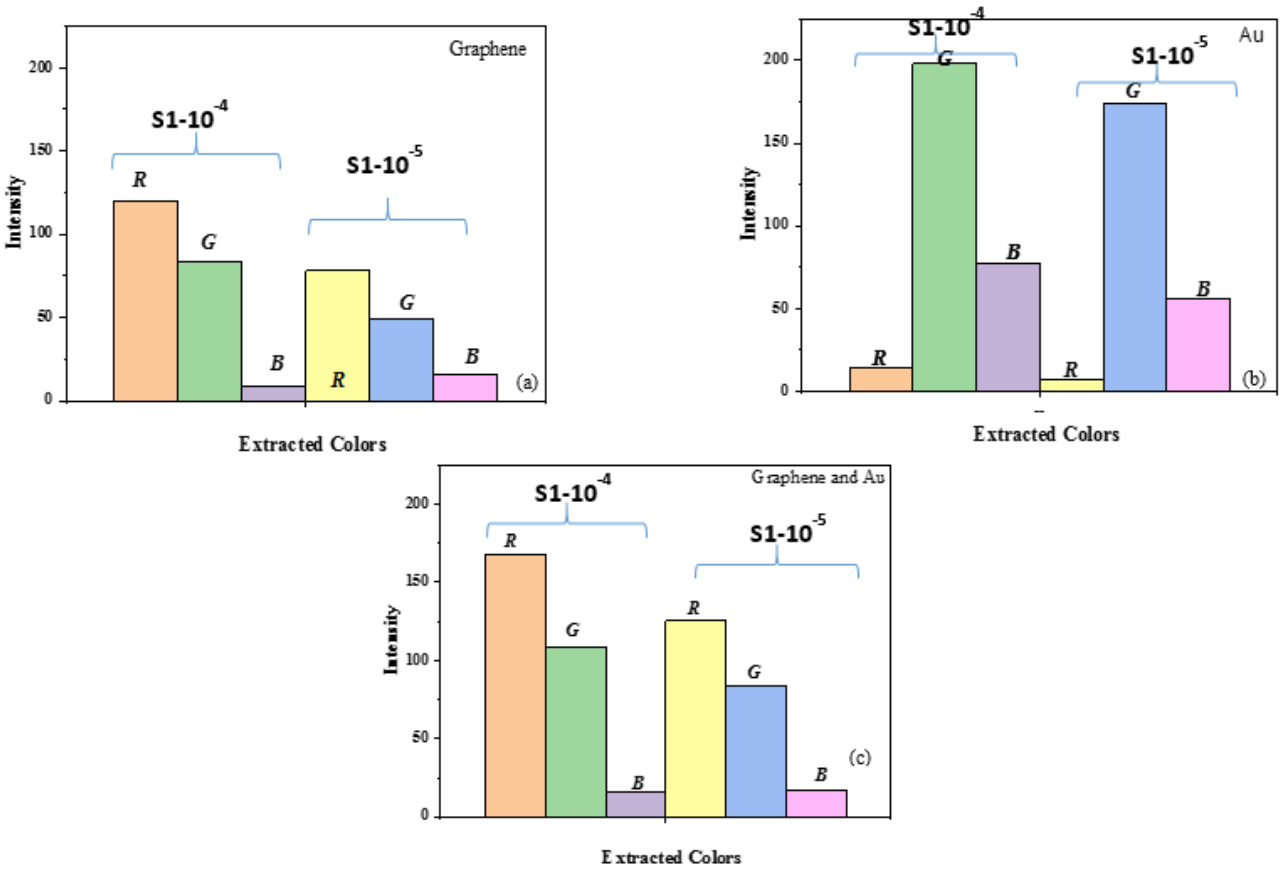

Figure 7. Three main colors extraction from three different concentrations as (a) graphene, (b) gold and (c) Mixture of graphene and gold NPs in two different dye's concentrations.

three main pumping energy region by the main gain medium at 10^{-5} M as shown in Figure 6 in the same arrangement of the sample's images which recorded previously by 10^{-4} M. The main red hotspots which indicated the RL peaks in the fluorescence shoulder of the main gain medium helps us to record higher resolution images which is sharper in the sample with gain medium by the concentration of 10^{-4} M. To get more sense about the RL effect in the resolution of our imaging systems by the aid of RL, we extract the main three colors' share as red (R), green (G) and blue (B) as shown in Fig. 7. This extraction was done for three different samples as graphene, gold and mixture of them in the main host dye medium with two different concentrations. In addition, this extraction was done for high energy pumping and thus the main RL was happens in the gain medium.
As shown in this figure, the red color share in the mixture of graphene and gold NPs is the maximum in 10^{-4} M Rh6G dye as the host medium as confirmed before in the RL result also. It means that we have the best output from the CCD camera in the sample with the mixture of gold and graphene NPs in the dye host medium with concentration of 10^{-4} M.

4. Conclusion

In sum, we fabricate the gold NPs, graphene NPs and the mixture of them as scatterer points in the Rh6G dye medium in two different concentrations. The samples with different concentrations were examined by continuous wave and also pulsed green light to record the fluorescence and random laser which indicate the best gain medium achieve with 10^{-4} M dye and the mixture of gold and graphene NPs. In addition, the main sample for higher resolution tissue imaging approved by CCD image of the samples and also the main three colors, RGB, extraction from the samples.

Ethical approval
All experimental procedures were performed in accordance with guidelines and regulations approved by a regional Institutional Animal Care and Use Ethics Committee of the "Ethical committee of Vice president of research of Shahid Beheshti university/IR.SBU.REC.1405".

Authors Contributions
All the authors have participated sufficiently in the intellectual content, conception and design of this work or the analysis and interpretation of the data (when applicable), as well as the writing of the manuscript.

Availability of data and materials
Data presented in the manuscript are available via 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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