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Gold nano-urchins for plasmonic enhancement of random lasing in a dye-doped polymer

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Abstract

We report our results on a plasmonic random laser with three-dimensional (3D) gold nano-urchins as scatterers distributed in Rhodamine 6G dye doped polymer film. The performance of anisotropic urchin scatterers is first studied using electromagnetic simulations for absorption/scattering cross-section and local field enhancement. This is compared to gold nanoparticles of similar size. The simulation results indicate a two-fold local field enhancement, a higher scattering cross-section, and a low absorption cross-section in the 400 nm-570 nm spectral region of interest for nano-urchins. The effective scattering mean free path for urchins is calculated to be 90 μm less than nanoparticles. This suggests nano-urchins to be efficient scatterers over conventional nanospheres for random lasing. Random laser is then experimentally demonstrated using gold nano-urchin scatterers. Incoherent random lasing is observed for very low urchin number density of order $\sim 10^8 \text{ cm}^{-3}$. A three-fold increase in scatterer concentration is shown to reduce the threshold energy from 0.8 mJ to 0.28 mJ per pulse. This is accompanied by a linewidth decrease from the rhodamine 6G emission bandwidth of 58 nm to up to 3 nm. The random feedback mechanism has been validated using a different spot pump scheme and angular measurement of emission. With low gold nano-urchin concentration, being incoherent and in film form, this plasmonic random laser could be an economical solution for speckle-free imaging applications.

Introduction

Random lasing occurs as a result of stimulated emission through optical multiple scattering and amplification in tandem. Ever since the first observation of random lasing in 1966 [1], there have been many advancements in this field using dielectric and semiconductor particles as scatterers [2,3], leading even to strongly confined coherent random lasing [4]. Compared to dielectric particles, metallic scatterers form a distinct class due to the plasmonic resonance effect, specifically localized surface plasmon resonance (LSPR) effect for smaller nanoparticles [5]. The striking feature of these scatterers is that the LSPR wavelength varies significantly with the particle size or shape. Moreover, localized field enhancement and improved scattering cross-sections gives an upper hand for these metallic particles to show lower lasing threshold and smaller linewidths as compared to dielectric counterparts. Dice et al. used these concepts for the first time to show improved random lasing using silver nanoparticles [6]. Popov et al. later showed that gold nanoparticles in a polymer film can induce much better photon scattering due to surface plasmons [7]. Coherent random lasers using silver nanoparticles have been shown to have linewidths as low as 0.5 nm [8]. Various morphologies of gold and silver particles have been later proposed for random lasing. However, the fundamental description and experimental validation of amplification of surface plasmon

polaritons was given by Noginov et al. by studying the effect of having gain molecules near metallic particles at the plasmon resonance wavelength. First, stimulated emission of surface plasmon polaritons was demonstrated using polymeric gain medium over a thin silver film [9]. However, the feedback mechanism was not yet clear. This was accomplished by using specially designed gold core and dye doped silica shell scatterers which showed a clear lasing threshold and linewidth narrowing at the plasmon resonance due to coupling of plasmonic modes to photonic modes [10]. Varying volume fractions of metamaterial glancing silver nanorods are shown to produce high quality factor coherent random lasing [11]. Interestingly, gold nanorods induce up to 1000-fold fluorescence enhancement from weak emitters owing to plasmon resonance [12]. The tuning of scattering cross-section and hence lasing threshold, using dielectric TiO₂-silica core-shell particles has been demonstrated by Ernesto et al. wherein the thickness of silica shell layer is used as the parameter [13,14]. Moreover, plasmonic particles provide a scope for tuning emission wavelength based on LSPR resonance effect [15]. Plasmonic core-shell particles are demonstrated in this regard [16,17]. Owing to the ambient extreme sensitivity and field enhancement, plasmonic structures have been used effectively for sucrose sensing [18] and light trapping [19,20]. Anisotropic metallic structures like gold nanostars and nanowires/rods have a relatively broad SPR spectrum over which the scattering properties could be efficiently used [21]. Broadband random lasing using gold nanostars has been demonstrated in this context [22]. Plasmon resonance coupling of silver particles has been shown to reduce electrically pumped threshold of Zn/MgO structures by three times [23]. Recently, plasmon nano-gaps have been effectively used to vary optical scattering and achieve linewidth tunable random lasers [24]. Silver nanowires incorporated in a stretchable film have been shown to induce lasing spectral shift of up to 7 nm and a polarization change upon stretching [25]. In general, particles with tapered or sharp edges concentrate electric field and also help in increasing the scattering. The ease of fabrication of small silver nanoparticles and to incorporate them at an optical fiber tip for RGB laser is noteworthy here to realize ultra-compact plasmonic optical devices [26]. Three-dimensional Urchin-like structures of ZnO [27], GaAs [28] and TiO₂ [29] are promising as efficient scatterers for random lasing to reduce the threshold. Silver nanoflowers have been recently used for coherent ultra-low threshold and broadband random lasing opening avenues for three-dimensional metallic anisotropic scatterers [30]. However, there are no reports till date on metallic nano-urchins for random lasing. Compared to two-dimensional nanostars or nanorods, the metallic 3D urchins are expected to provide higher field and scattering enhancement owing to increased area.

Here, we report on the incoherent random lasing from 3D gold nano-urchin scatterers in dye doped polymer films. First, the absorption effect, scattering cross-section, and local field enhancement of the gold nano-urchins is compared to conventional spherical gold particles at pump laser and dye emission wavelengths through simulations. The simulations show that the urchins provide a higher scattering cross-section and superior local field enhancement, and at the same time have minimal absorption which improves overall optical gain in the system. The optical scattering mean free path calculations point out to an enhanced scattering by urchins. This should lead to a significantly lower lasing threshold. Next, we experimentally demonstrate random lasing with low scatterer concentration of order $\sim 10^8 \text{ cm}^{-3}$ of gold nano-urchins. We also study the effect of a small variation in concentration on the lasing threshold and linewidth. The results indicate an incoherent feedback random lasing with linewidth up to 3 nm.

Methods

The local electric field intensity and absorption/scattering cross-section of the nano-urchins and nanoparticles are simulated using finite-difference time-domain (FDTD) 3D Electromagnetic Simulator Lumerical FDTD [31]. Nano-urchins are modelled according to the practical transmission electron microscopy (TEM) image to keep the data in close relation to the experiment. A typical simulation setup is shown in figure 1. Two scenarios are chosen, one is for single particle (urchin and sphere) and another is for a group of 12 particles to validate the results for a random medium case. A random distribution of 12 particles is chosen based on computational optimization, and more number can be simulated in general. Mesh size of 5 nm is chosen for all the simulations. Illumination source is Gaussian and local field monitor is placed across the center of the particle. Similarly, absorption and scattering cross-sections are measured using a total field scattered field (TFSF) source. The 3D analysis group for this simulation calculates the absorption cross-section by measuring net power flow into TFSF source and the scattering cross-section by net power flow out of the TFSF source.

Figure 1: FDTD simulation setup to study scattering and absorption cross-sections, and local field enhancement showing the source, field monitor and the simulation region for a group of 12 particles to emulate a random medium. Shown image is a typical setup for a group of urchins. Similar setup is used for single particles of urchins and spheres.

The random laser is fabricated using rhodamine 6G as the gain medium, gold nano-urchins as the scatterers, and poly-vinyl alcohol (PVA) polymer for the film. Two samples are fabricated with same gain but different scatterer concentrations of gold nano-urchins. 2 mM rhodamine 6G in dimethyl sulfoxide (DMSO) and 174 mM PVA in DMSO (15 mg/mL) solutions are prepared. Gold nano-urchins sample (Sigma-Aldrich) with particle concentration of $3.8 \times 10^9 \text{ cm}^{-3}$ is used. 25 μL of PVA solution, 25 μL of rhodamine 6G solution and 50 μL of gold nano-urchins are mixed, drop-casted on a glass substrate and heated at 70° C. for 3 hours. This is named sample S1. In a similar way, 150 μL of gold nano-urchins is used for preparing another sample S2 with same PVA and gain concentrations. The films are dried thoroughly to remove solvents. Sample S2 ($5.4 \times 10^8 \text{ cm}^{-3}$) has thrice the scatterer concentration as S1 ($1.8 \times 10^8 \text{ cm}^{-3}$) with all other parameters fixed. We intend to study the effect of this small variation in

concentration, a factor of three, on the lasing performance. Optical characterization of the films is done through absorption (Shimadzu UV-1800 spectrophotometer) and photoluminescence (Edinburgh Instruments FS5) spectroscopy. Nano-urchin morphology is visualized through TEM (Jeol 2100F). Average film thickness is measured to be 160 μm (Keyence VK-X1100).

Random lasing measurements are done using the optical setup shown in figure 2(a), consisting of Nd:YAG pump laser (Opotek Vibrant- 532 nm, 10 Hz, 5 ns), focussed using a 5 cm focal length cylindrical lens to form a line pump pattern of area 6.2 mm^2 at focus (measured with Edmund Optics 4M beam profiler camera) on the sample. The input pulse energy of the laser is varied by changing the Q-switch delay time. A neutral density (ND) filter of optical density (OD) 1 is used before the lens for all the measurements. The emission from the sample is collected at the edge using a fiber-optic spectrometer (Ocean Optics USB4000). The light collection in the fiber is optimized based on different angular measurements. Integration time of 100 ms is used to make sure they are single shot spectra. The mechanism of light propagation, scattering and edge lasing is depicted in the figure 2(b). The input light is effectively scattered, confined and amplified in the film plane.

Figure 2: (a) Experimental schematic of the random lasing setup with edge detection of the output light from the sample, (b) Mode of light scattering and propagation in the film sample.

Results and Discussion

Normalized optical absorption spectra of gold nano-urchins and gold nanoparticles of both same average diameter of 100 nm are shown in figure 3(a). The absorption spectrum is co-plotted with normalized rhodamine emission spectrum to understand the photon absorption effect. Interestingly, gold nano-urchins being anisotropic in structure, have a very broad absorption spectrum from 500 nm to 800 nm, which is not the case for spherical nanoparticles which have a relatively narrower absorption bandwidth. The absorption peak wavelength for nano-urchins is 670 nm whereas for gold spherical nanoparticles it is 570 nm which overlaps fully with the rhodamine 6G emission. The absorption overlap of gold nano-urchins with rhodamine 6G PL is significantly less. This property of urchins gives provision for laser dye like rhodamine 6G to be used efficiently without inducing any absorptive losses in the random laser system. TEM image of gold nano-urchins is given in figure 3(b) where an average diameter of 100 nm can be realized. The urchin spikes of 20 nm length are seen in the particle. The plasmonic local field is expected to be high at these tips of nano-urchins.

Figure 3: (a) Normalized absorption of gold nano-urchins and nanoparticles, and PL spectrum of rhodamine 6G in DMSO, and (b) TEM image of gold nano-urchins with average diameter of 100 nm.

To compare the lasing performance of gold nano-urchins with isotropic gold nanospheres, FDTD simulations are conducted as described earlier. To evaluate lasing performance using plasmons, we look at three aspects: absorption cross-section, scattering cross-section, and the local field enhancement. Figure 4 shows the results for local field enhancement for urchins and spheres. It is worth noting that a Gaussian input field has been used for all the simulations shown here. This is to keep the simulations in close relation to experimental condition where a laser has been used as the excitation source. Figures 4(a) and 4(b) show field enhancement at a single particle of same size of sphere and urchin respectively. It is observed that the field intensity magnitude for urchin is 2.1 times that of a sphere. Since we are studying random particle distribution experimentally, the simulation is done for a typical ensemble of 12 particles to validate the field enhancement factor. Results are given in figures 4(c) and 4(d). An enhancement factor of 1.9 is obtained for urchins as compared to spheres for this ensemble. The enhancement is thus consistent at around a factor of 2 for urchins. This plasmonic field enhancement aids the input pump light to be coupled efficiently to the scattering particle and the gain medium for overall better optical gain. Thus urchins couple the input light efficiently, increasing the scattering and gain performance.

Figure 4: Electric field data at rhodamine emission peak wavelength of 570 nm from FDTD simulations-(a) for single gold nanoparticle, (b) for single gold nano-urchin, (c) for 12 gold nanoparticles, and (d) for 12 gold nano-urchins.

The absorption and scattering cross-section simulation for gold nano-urchins and spheres of same size is done as explained earlier with a single particle setup. Scattering cross-section comparison is given in figure 5(a). The pump and emission peak of rhodamine 6G are marked. At pump wavelength of 532 nm, urchins have higher scattering cross-section than spheres. Again, it has to be noted that these results are for Gaussian input wave and not a plane wave. At the rhodamine 6G emission peak of 570 nm, both particles have similar scattering cross-sections. Thus the pump photons are scattered more efficiently by urchins. We initially employed coherent backscattering (CBS) experiment for scattering mean free path estimation. However, due to the transparent nature of the sample, we could not get the CBS profile. Thus we estimated it using the theoretical method. Using the scattering cross-section data, the scattering mean free path (l_s) of photons for urchins and spheres can be calculated. This is done using the equation $l_s = (p\sigma)^{-1}$ where p is the particle concentration and σ is the scattering cross-section. The sample film has a total volume of 8 mm³. The particle concentration for sample S2 in this confined volume is $\sim 7 \times 10^{10}$ cm⁻³. The scattering cross-section of urchins and nanoparticles at 532 nm pump and 570 nm rhodamine 6G emission peak is considered. The calculation gives l_s of 270 μ m and 360 μ m for urchin and sphere respectively at pump laser wavelength. Similarly, l_s is 304 μ m and 340 μ m for urchin and sphere respectively at rhodamine PL peak wavelength. At pump wavelength, l_s is 90 μ m less for urchins. At optical length scales this value is significant and suggests that the effective scattering distance for urchin is less, which contributes to an enhanced optical feedback and amplification. Using the Ioffe-Regel relation for mean free path l_s and wavelength λ of 532 nm we see that in present case, $l_s \gg \lambda$ which signifies a weak scattering phenomenon which gives rise to incoherent feedback lasing. Next crucial aspect is the photon absorption by scatterers which has to be low for efficient random lasing. Figure 5(b) shows the absorption cross-section for urchin and sphere at pump and gain emission wavelengths. At both the wavelengths, urchins have lower absorption cross-sections. This indicates that the pump photons and fluoresced photons are absorbed less by urchins, leading to low absorptive loss in the system.

Figure 5: (a) and (b) are the scattering and absorption cross-section data respectively for a single gold nano-urchin and nanoparticle with the pump laser and dye emission wavelengths marked. Gaussian input field has been used for the simulations.

Through simulations, we have shown that gold nano-urchins provide local field enhancement by a factor of two, a higher scattering cross-section at pump wavelength, and a low absorption cross-section at both pump and gain emission wavelengths. This proves that nano-urchins are better scatterer candidates than isotropic spheres for improved random lasing performance.

Random lasing experiments are performed using the setup shown in figure 2(a) earlier. The spectral results are analysed to obtain the threshold and linewidth plots for the two samples S1 and S2. The pump pulse energy dependent emission is shown in figure 6(a) and 6(b) for sample S1 and S2 respectively. Specifically, the emission below threshold, near threshold and above threshold is shown to illustrate the effect of particle concentration on random lasing. It is observed that the sample with thrice the particle concentration shows a distinct single peak lasing with smaller linewidth above threshold energy. The lasing mechanism is observed to be due to incoherent feedback with a single emission profile unlike a coherent feedback type, where strong particle confinement leads to sub-nm sharp lasing spikes due to constructive interferences in the system. Moreover, the threshold energy for sample S2 is three-fold less compared to S1. This is observed by plotting pump pulse energy dependent maximum PL intensity and linewidth. The plots are given in figures 6(c) and 6(d). For sample S1, a sharp variation in PL intensity is observed at 0.8 mJ pump pulse energy. This translates to a fluence of 0.13 mJ/mm^2 . Around this threshold energy, a linewidth reduction from 58 nm to 20 nm has been observed. Pumping above the threshold energy, the linewidth remains constant at around 12 nm to 10 nm. Higher pulse energies did not show any change in linewidth characteristics. Higher input energy also photo-bleaches the gain medium, leading to reduced PL, and hence this was avoided. The photobleaching effect can be seen in figure 6(c) where the last point at 1.1 mJ is saturated. We attribute this to the fact that the scattering mean free path is slightly larger than the transverse thickness of the sample, due to which more absorption loss and hence dye degradation happens along the increased optical path in the plane of the sample. Similarly, for sample S2, the threshold and linewidth profile is given in figure 6(d). Interestingly, the threshold for this sample is reached at 0.28 mJ pulse energy. This translates to a pump fluence of 0.05 mJ/mm^2 . This fluence is nearly three times less than that for sample S1. Also at this threshold energy, the linewidth approaches a value of 5 nm which is four times less than the sample S1. Above the threshold energy, the linewidth remains almost constant at 3 nm. Also, above this

particle concentration for sample S2, we did not observe any further change in the PL and linewidth with the same gain concentration. Hence we chose two typical samples with one having thrice the scatterer concentration as the other. The experimental results indicate that a small variation in particle concentration leads to a significant enhancement in lasing performance.

Figure 6: Random lasing results showing pump laser energy dependent emission for (a) sample S1 and (b) sample S2. Threshold and linewidth profiles for (c) sample S1 and (d) sample S2. The intensity and linewidth values for a given pump pulse energy are taken for the maximum PL intensity point in the spectrum.

Feedback mechanism in the random laser

The feedback mechanism in the random laser is attributed to two aspects, scattering in the plane of the sample and scattering in the thickness of the sample. These are shown in figure 7 (a) and (b) respectively.

Figure 7: Depiction of two random feedback mechanisms occurring in the random laser, (a) in the plane of the sample and (b) across the thickness of the sample. Green spot is the pump stripe. (c) The imaged random laser emission and its intensity map at the edge of sample.

Multiple scattering happening in the plane of the sample experiences more pump photons at the input side owing to the stripe pump as compared to a circular spot pump. Hence the overall fluoresced photon density would be more which further leads to amplification. Similarly, in the thickness direction, the feedback also occurs at the interface of the film where internal reflections happen other than the general scattering. The internally reflected light again scatters back leading to overall higher emission. The stripe pumping plays a significant role of pumping more photons to the dye molecules at the same time in the sample, which is not possible in the case of a circular spot. Hence we see the random laser emission, signified by higher intensity and linewidth narrowing, at the edge of the sample. The random laser emission and its corresponding intensity map is shown in figure 7(c).

Waveguide effect and detection angle dependence

The waveguide effect in random laser is schematically depicted in figures 7(a) and 7(b) based on the optical scattering and internal reflection feedback. However, to better understand that the random laser emission indeed occurs as a consequence of waveguide effect in our sample, we carried out two experiments: circular spot pumping of the sample, and angular measurements of the emission profile. For circular spot pumping we used a plano-convex lens with 5 cm focal length (same as that of the cylindrical lens). The emission is detected in two directions, one along edge of the sample and other perpendicular to the film plane. Figures 8(a) and 8(b) show the spectral data for edge collection and perpendicular-to-plane collection respectively. It is observed in both the cases that the required feedback for random lasing is not achieved. With stripe pumping, the threshold energy is 0.8 mJ, but with circular spot pumping, we observed general spontaneous emission from the sample. The spectral data signify that in

film samples where transverse dimensions are less (thickness), the effective optical feedback should occur in the plane of the film where scatterer density is more. Technically, a stripe pump spot should accomplish this as more region is pumped with photons. In the case of circular spot pumping in a film sample, a very small fraction of scatterers see the pumped photons at the interaction area thereby forming no multiple scattering required for random lasing. Thus, in our case here, we attribute the effective optical feedback and lasing to waveguide effect due to stripe pumping.

Figure 8: Spectral data for circular spot pumping of the sample showing no signature of random lasing for (a) edge collection and (b) perpendicular-to-plane collection. Angular measurements of the random laser emission: (c) linewidth and intensity variation, and (d) the corresponding emission spectra at different angles in the quadrant..

To study the nature of the lasing mode, angular emission data has been taken in a quadrant from 0° to 90° , where 0° corresponds to the edge and 90° to perpendicular to sample plane, in steps of 15° . Stripe pumping as mentioned for random lasing experiment has been used. The data is shown in figures 8(c) and 8(d). As expected, the lasing mode intensity is maximum at the edge detection angle (where the waveguide effect is maximum) and gradually reduces as we move perpendicular to the sample plane. Thus the waveguided nature of lasing mode is clearly understood here. Another point to note is the non-existence of multimodes in the emission spectrum. The fundamental fluorescence spectrum of rhodamine 6G narrows to give diffuse random lasing. The emission of such diffusive random lasers has been theoretically analysed earlier [32,33]. In random lasers governed by weak localization, as in our case here,

mode mixing and mode overlapping are governed only by the atomic transitions of the optical gain medium. In our system, it is rhodamine 6G dye's fluorescence. Owing to regime far from the Anderson localization, many modes are enveloped within the fluorescence curve. This can be visualized as an existence of many wide-linewidth modes under the envelope. Hence we do not see any sub nanometer spikes which are characteristic of coherent random lasers.

Conclusion

We have proposed and successfully demonstrated an incoherent random laser with low particle density of 3D gold nano-urchins of order $\sim 10^8 \text{ cm}^{-3}$. Through electromagnetic simulations, we observed a local field enhancement factor of two, a higher scattering cross-section at pump wavelength, and a low absorption cross-section for nano-urchins in comparison to nanospheres. The scattering mean free path, calculated from the cross-section data, was 90 μm less for urchins suggesting an enhanced effective scattering and optical feedback required for lasing. Thereafter, a random laser with gold nano-urchin scatterers was experimentally demonstrated. A three-times increase in scatterer concentration led to a threshold fluence reduction from 0.13 mJ/mm^2 to 0.05 mJ/mm^2 . This was accompanied by a linewidth as low as 3 nm from 58 nm emission bandwidth of rhodamine 6G. The lasing was observed to be incoherent type with no sub-nm coherent feedback spikes. The waveguiding mechanism in the film sample has been validated along with angular measurements giving information on mode properties. Our results indicate the potential of gold nano-urchin 3D as efficient scatterers for random lasing. Gold nanoparticles being biocompatible, we envisage that this polymer film incoherent random laser with low particle density could be an economical solution for light source for applications in bio-sensing and -imaging.

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