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The enhanced random lasing from dye-doped polymer films with different-sized silver nanoparticles

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ABSTRACT

We reported on the enhanced random lasing from organic dyes doped with silver nanoparticles (Ag NPs), the sizes of Ag NPs ranged from 8 nm to 250 nm. The effects of different sizes of Ag NPs on the lasing properties were studied. We found a strong dependence of the random lasing properties on the size of the Ag NPs, and the lowest threshold was achieved by the introduction of Ag NPs with the diameter of 150 nm. By studying the enhanced localized electromagnetic (EM) field due to localized surface-plasmon resonance and the scattering effect of Ag NPs in experiment and Mie theory, we found that the enhanced localized EM field plays a major role on enhanced lasing of organic dyes for the small Ag NPs (diameter < 50 nm); and the scattering effect is the dominant underlying mechanism for random lasing for the large Ag NPs (diameter ≥ 100 nm), which also suggest that the lowest threshold and strongest lasing are dominated by the photon scattering.

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1. Introduction

Recently, metal nanoparticles (NPs) have been widely investigated for the applications in nanolasers [1–5] and optoelectronic devices [6] to biosensors [7]. It had been demonstrated that the coherent and incoherent random lasing of gain media were induced by the golden or silver NPs. Generally speaking, metal NPs enhance the lasing efficiency via two different mechanisms [8]: (i) enhancement of localized electromagnetic (EM) field in the vicinity of metal NP due to localized surface-plasmon resonance (LSPR) [9–11] and (ii) enhancement of the scattering strength, resulted from large scattering cross section of metal NP. As for the gold NPs, Popov et al. studied the enhancement effect of different sizes of gold NPs on the lasing characteristics in a polymer film doped with Rhodamine 6G (Rh6G) [8,12]. T. Zhai et al. showed a threshold reduction for waveguide-plasmonic scheme constructed by coating the gain medium onto gold nanoisland structures [13]. E. Heydari reported the threshold reduction in the gold NP-based random laser by tuning the coupling between the gain material and the LSPR of Au-NPs [14]. In these works, the LSPR spectra of Au NPs overlap sufficiently with emission spectra of the dyes, and photon scattering is expected to be dominant mechanism to enhance lasing properties for Au NPs. As for the Ag NPs, Dice et al. firstly demonstrated an incoherent random laser emission by introducing a suspension of 55 nm-Ag NPs in the solution of Rh6G in 2005 [15]. And then, Meng et al. reported the enhanced emission of coherent random lasing in polymer films by introducing Ag NPs with the diameter of 2 nm and 50 nm respectively [16,17]. And they also discussed the coupling between LSPR of core–shell Ag NPs and organic dyes [18]. In these works, the enhanced localized EM field is considered to be the dominant mechanism, especially for the sizes of Ag NPs below 50 nm. However, Rh6G was used as laser dyes which emission spectrum doesn't overlap with the LSPR spectra of Ag NPs. The deviation between emission spectrum of Rh6G and LSPR spectra of Ag NPs is not optimal for the lasing properties. According to the above works about the Ag NPs, we can know that the present researches are all limited to used several small size Ag NPs (2 nm, 50 nm) to enhanced lasing properties, and the research about the large size Ag NPs (≥100 nm) on lasing properties is absent. Therefore, systematic investigation on the lasing properties affected by Ag NPs of different size with different LSPR is desirable.

Here, we report on the effects of Ag NPs with different sizes on

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the lasing properties of organic gain medium. The diameters of Ag NPs ranged from 8 to 250 nm with different LSPR peaks. To this end, different Ag NPs were doped into gain medium, and kept constant the mass concentrations to ensure the same gain volume and enable a more critical examination, and the lowest lasing threshold was achieved by introducing Ag NPs with a diameter of 150 nm. In addition, it was also found that the enhanced localized EM field plays a major role on the enhanced lasing of organic dyes with small Ag NPs (<50 nm); the scattering effect mainly contributes to the enhanced lasing for the large Ag NPs (≥100 nm).

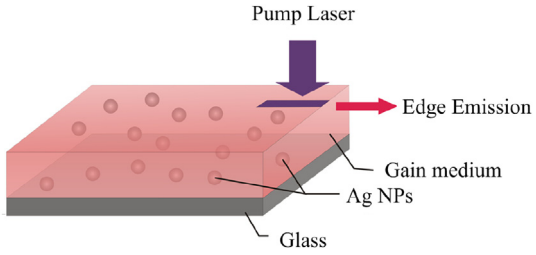

Fig. 2. Schematic illustration of glass/PS: Alq_3: DCJTB doped with Ag NPs.

2. Experiment

Ag NPs having a well-controlled size were synthesized by a seed-mediated growth method [19]. Different sizes of Ag NPs were prepared in two steps by citrate reduction of silver nitrate ($AgNO_3$) with $NaBH_4$ as strong reducing agent in water. First, small Ag NPs were synthesized under chemically reducing $AgNO_3$ in aqueous solution by a rapid nucleation-growth-ripening principle; the resulting Ag NPs were used as starter seeds. And then, slowly adding the proper portions of Ag salt and citrate reducer into the starter seeds solution as obtained in the first step. The slow process ensures aggregation of the released Ag atoms on the Ag nucleation centers without the formation of new centers. In this way, the final sizes of the NPs were tuned, and we prepared NPs with the diameters as: 8, 20, 50, 100, 150, 180, 200 and 250 nm. Fig. 1 shows the Atomic Force Microscopy (AFM) images of Ag NPs with different sizes.

The gain medium was fabricated as follows: Polystyrene (PS), tris(8-hydroxyquinolinato)aluminum (Alq_3) and 4-(dicyanomethylene)-2-tert-butyl-6(1,1,7,7 tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB) were fully dissolved in chloroform solution (PS: Alq_3: DCJTB = 200:100:3.5, wt%). Different sizes of Ag NPs were doped into the solution maintained a constant mass concentration of 1.28×10^{-3} g/cm^{-3}. The solution was then spin-coated with 1500 rpm spin speed atop of the glass substrate, the device structure is shown in Fig. 2. For comparison, the gain medium without Ag NPs on glass substrate was also fabricated. The thickness of gain medium film is about 420 nm.

In the experiment, the thicknesses of organic gain media layer were measured with Stylus Profiler (Dektak 6M, USA). The absorption and photoluminescence (PL) spectra were obtained by UV–Vis spectrophotometer (HITACHI U-3010, Japan) and Fluorescence Spectrometer (Fluoromax-4 spectrofluometer) respectively. The devices were pumped by the third harmonic of Nd: YAG laser (355 nm, 10 Hz repetition rate, and 25 ps pulse duration). Through a pinhole filter, a slit and a cylindrical lens, the laser beam was formed as a stripe with the size of 5 mm × 1 mm, and was perpendicular to the surface of the devices. Edge emission spectra were measured by Fiber Optic Spectrometer (Ocean Optics SpectraSuite, USB2000). The lasing threshold, peak intensity and the full width at half maximum (FWHM) were measured.

3. Results and discussion

Fig. 3 is the LSPR bands of Ag NPs with different sizes, together with the absorption and emission spectra of Alq_3 and DCJTB. The LSPR bands presented are the absorption spectra of Ag NPs. Alq_3: DCJTB is the gain media which is a typical donor-acceptor lasing system reported in previous published reports [20,21]. Alq_3 is used as donor and DCJTB is used as acceptor, which is the Förster resonance energy transfer system [22–25]. It can be clearly seen that the donor emission, centered around 518 nm, overlaps well with the acceptor absorption peak at 512 nm, enabling efficient resonant energy transfer between the donor and acceptor. In addition, the LSPR bands of different sizes of Ag NPs have difference overlaps with the absorption and emission spectra of the Alq_3 and DCJTB,

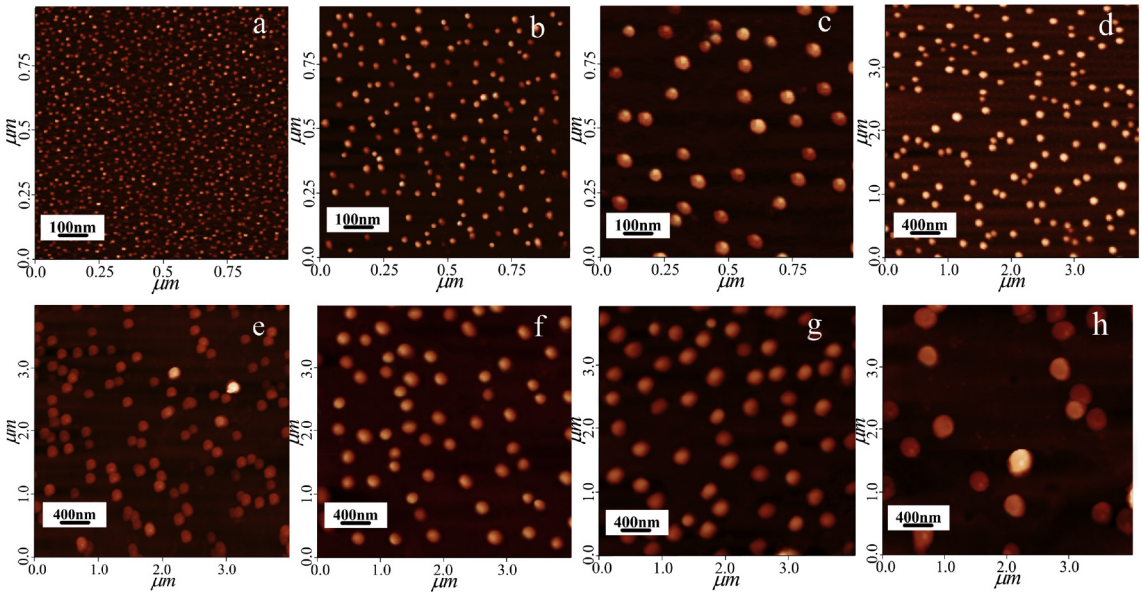

Fig. 1. AFM images of Ag NPs with different sizes. The average sizes of Ag NPs are (a) 8 nm, (b) 20 nm, (c) 50 nm, (d) 100 nm, (e) 150 nm, (f) 180 nm, (g) 200 and (h) 250 nm.

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