

# Plasmonically enhanced lasing by different size silver nanoparticles-silver film hybrid structure



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## ABSTRACT

We reported on the enhanced lasing in organic dyes based on plasmonic hybrid structure of Ag nanoparticles (NPs)-Ag film, the diameters of Ag NPs ranged from 20 nm to 100 nm. The lowest lasing threshold was achieved by the optimal size Ag NPs-Ag film hybrid structure, which was reduced by 5.2 times than that of the neat gain medium. Comparing to the separate Ag NPs or Ag film, the hybrid structure presented the more intense local electric field due to the plasmonics coupling between the localized surface plasmons of Ag NPs and the surface plasmon polariton of Ag film, and the stronger scattering due to the reinjection of the leaking photons by external feedback of Ag film. The effects of different sizes Ag NPs-Ag film hybrid structures on lasing were investigated. It found that when the Ag NPs in hybrid structure is small (diameter  $\leq 40$  nm), the enhanced localized electric field plays a major role on enhanced lasing; with the increase of Ag NPs size, the enhanced electric field and scattering have comparable contribution on enhancing lasing; for the larger size Ag NPs-Ag film (diameter  $\geq 80$  nm), the scattering effect is the dominant mechanism for random lasing. Then the lowest threshold was dominated by the balance of enhanced localized electric field and scattering effect. Our results could provide us a unique idea to effectively enhance the lasing of organic dyes, and realize the lower pumped threshold and stronger lasing.

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## 1. Introduction

Localized Surface Plasmons (LSPs) can be excited on metal nanoparticles (NPs) whereas delocalized surface plasmon polariton (SPP) can be excited on metallic film, which both can induce the field enhancement in the near-field region [1–4]. Various groups have already used metallic NPs to enhance lasing. For example, O. Popov et al. utilized gold NPs to enhance lasing from a polymer film doped with Rhodamine 6G [5,6]. X. Meng et al. achieved the enhanced emission of coherent random lasing in polymer films by introducing Ag NPs [7,8]. T. Zhai et al. demonstrated an enhanced random laser based on gold nano-island structures with a layer of dye-doped polymer [9]. E. Heydari et al. reported the emission enhancement for the gold NP-based waveguided random laser [10]. However, for the metallic film in planar waveguide structure, it

would increase the lasing threshold due to the quenching effect of metallic film on lasing of dyes, which is a major obstacle to obtain stimulated emission under electrical pumping [11,12]. Substantial efforts have been made to restore and optimize the optically pumped lasers in the presence of metallic film, such as polymer film deposited on distributed feedback metallic structure [13,14], adopting in low loss metal cladding [15–17], or introducing the spacer between polymer film and metallic film [18]. Despite all these attempts, it is still desired to further reduce the lasing threshold of optically pumped laser in the presence of metallic film.

Here, a particularly interesting plasmonic system which has received somewhat less attention, metallic NPs-metallic film hybrid structure was explored. We systematically investigated the effects of different sizes Ag NPs-Ag film hybrid structures on the lasing of gain medium, the diameters of Ag NPs ranged from 20 to 100 nm. The lowest lasing threshold of the gain medium deposited on 60 nm Ag NPs-Ag film hybrid structure was found, which greatly reduced 5.2 times than that of the gain medium deposited on glass. For comparing, the devices that gain medium deposited on Ag film,

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silver island film (SIF) were also prepared. It found that the Ag NPs-Ag film coupled system could better enhance lasing, which is because that the Ag NPs-Ag film hybrid structure presents the more intense local electric field due to the plasmonics interaction between LSPs of the Ag NPs and the SPP of Ag film, and the stronger scattering effect due to the re-injection of emitted light into the organic gain media of planar waveguide structure by the external feedback of Ag film.

At meanwhile, the effects of different sizes Ag NPs-Ag film hybrid structures on lasing properties were studied. It was noting that the enhanced localized electric field and scattering effect of Ag NPs-Ag film all work on the reduction of lasing threshold. However, according to the experiment and theoretical analysis, it was found that when the Ag NPs in hybrid structure is small (diameter  $\leq 40$  nm), the enhanced localized electric field plays a more significant role on the enhanced lasing of organic dyes; with the increase of Ag NPs size for hybrid structure, the enhanced localized electric field and scattering play the comparable role on the lasing; when the Ag NPs size is larger (diameter  $\geq 80$  nm), the scattering effect of Ag NPs-Ag film mainly contributes to the enhanced lasing. Then we concluded that the optimal size Ag NPs-Ag film could provide a balance between enhanced local electric field and scattering to realize maximum lasing. At meanwhile, the negative effect of metallic film is avoided to realize the lower pumped threshold than that of the metal-free device.

## 2. Experiment

### 2.1. Synthesis of different size Ag NPs

Different sizes Ag NPs were synthesized according to the seed-mediated growth method by citrate reduction of silver nitrate ( $\text{AgNO}_3$ ) with  $\text{NaBH}_4$  as strong reducing agent [19]. Firstly, small Ag NPs were synthesized under chemically reducing  $\text{AgNO}_3$  in aqueous solution by a rapid nucleation-growth-ripening principle, the resulting Ag NPs were used as starter seeds. Then, slowly adding proper portions of Ag salt and citrate reducer into the starter

seeds solution obtained in the first step. In this way, we prepared Ag NPs with diameters about: 20, 40, 60, 80, 100 nm.

### 2.2. The devices based on different sizes Ag NPs-Ag film hybrid structure

In order to systematically study the properties of different sizes Ag NPs-Ag film hybrid structures, we developed the devices that gain media deposited on different sizes Ag NPs-Ag film, the diameters of Ag NPs were about 20, 40, 60, 80, and 100 nm, respectively. The device structures were Glass/Ag film (50 nm)/ $\text{SiO}_2$  (10 nm)/Ag NPs/LiF (5 nm)/PS:BMT-TPD (shown in Fig. 1). For comparison, the gain medium deposited on glass was also prepared as reference. The preparation process of devices was following manner:

Firstly, a continuous Ag film of thickness 50 nm was evaporated onto a glass substrate. The 10 nm- $\text{SiO}_2$  layer was radio frequency sputtered over the silver surface, which could serve to protect the metal surface from chemical attack, and to provide a means to chemically attach silver colloids. SIFs were deposited according to the procedure described elsewhere [20]. The Ag film coated with SIF was prepared. Fig. 2 shows the atomic force microscopy (AFM) images of the Ag NPs with different sizes.

Then the planar waveguide structures were fabricated. N,N'-bis(3-methylphenyl)-N,N'-diphenyl-[1,1':4,1''-terphenyl]-4,4''-diamine (BMT-TPD) was used as gain medium [21]. Polystyrene (PS) and BMT-TPD were dissolved in chloroform solution (PS: BMT-TPD = 4:1, wt%). The gain medium films were obtained by spin coating the solutions onto LiF layers at speed 4000 rpm, and were annealed at 110 °C for 10 min to remove the solvent. The 5 nm-LiF layer was deposited over the SIF surface to prevent quenching caused by direct contact between Ag NPs and gain medium. The thickness of gain medium layer is approximately 250 nm.

### 2.3. Characterization

The absorption and photoluminescence (PL) spectra were

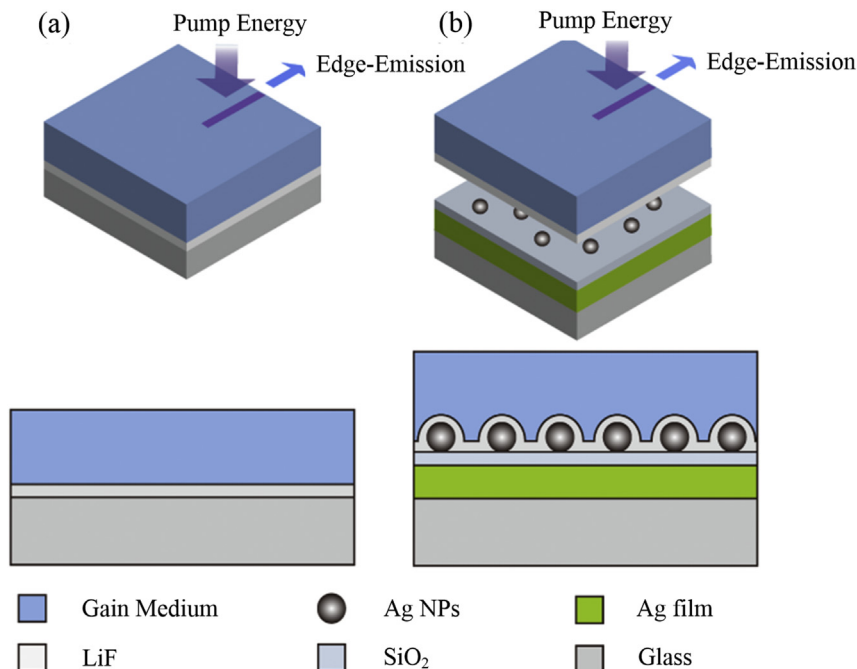


Fig. 1. The schematic illustrations of different devices. (a) Glass/LiF (5 nm)/PS:BMT-TPD (reference device); (b) Glass/Ag film (50 nm)/ $\text{SiO}_2$  (10 nm)/Ag NPs/LiF (5 nm)/PS:BMT-TPD.

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