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Spontaneous Emission Control of 2D Semiconductors Using Nanospheres on Gold Mirror Films

Songyan Hou[1,2], Landobasa Yosef Mario A L Tobing[2], Xingli Wang[1,2], Zhenwei Xie[1,2], Jin Zhou[2], Tay Beng Kang[1,2], Daohua Zhang[2], Muhammad Danang Birowosuto[1*], Edwin Hang Tong Teo[1,2*], Hong Wang[1,2*]

[1] CNRS International NTU THALES Research Alliance
50 Nanyang Drive, Border X Block, Level 6
Singapore 637553, Singapore
[2] School of Electrical and Electronic Engineering, Nanyang Technological University
50 Nanyang Avenue
Singapore 639798, Singapore
[*]Corresponding authors email: MBIROWOSUTO@ntu.edu.sg, HTTEO@ntu.edu.sg, EWANGHONG@ntu.edu.sg

Abstract

The plasmonic resonance can be robustly tuned by adjusting the thickness of dielectric spacer layer between gold Nanospheres and a Gold Mirror Films (NGMF). Additionally, great modification of spontaneous emission was observed in MoS_2 when coupled into NGMF nanostructures.

1. Introduction

Two-dimensional (2D) materials have opened up plenty of possibilities for photonics and electronics during past few decades[1]. Among 2D materials, monolayer molybdenum disulfide (MoS_2) is a direct bandgap two-dimensional (2D) material with a visible wavelength emission, opening great perspectives for applications in nanoscale optoelectronic photonic device. The excitons in monolayer MoS_2 remains stable even at room temperature because they are tightly bound with large Coulomb interactions within thin film[2]. Various complex excitons reported in monolayer MoS_2 include neutral excitons, tritons, bound excitons and biexcitons. These remarkable optical properties demonstrate that monolayer MoS_2 is a promising material for nanophotonics and nanoelectronics. However, compared to other low dimensional semiconductors, weak spontaneous emissions and low intrinsic quantum yield (QY) in monolayer MoS_2 become a hindrance for further applications in high speed photonic devices. Thus, manipulating spontaneous emission in MoS_2 needs further exploration. Spontaneous emission can be strongly controlled by engineering the surrounding photonic environment of emitter and photonic density of states of free space. Plasmonic nanocavities have attracted great interest due to their strong local electric field enhancement at room temperature, thus offering various approaches to increase spontaneous emission rate of semiconductor emitters[3-7]. Among all the plasmonic cavities, nanoparticle on gold mirror films (NGMF) system provides an intriguing plasmonic structures which can adopt scalable fabrication technique.

In this work, we characterize NGMF with tunable plasmonic resonances from 620 nm to 690 nm, which exhibit 211-fold emission enhancement when integrated with MoS_2 monolayer. We also show excellent agreement between emission and lifetime enhancements.

2. Methods and Results

A 130 nm gold thin film was deposited onto a glass slide via electron beam evaporation with 5 nm titanium adhesive layer. The dielectric spacer layer of Al_2O_3 layer was then deposited using atomic layer deposition (ALD) with various thicknesses ranging from 1 nm to 25 nm. Monolayer MoS_2 was grown on SiO_2/Si substrate via chemical vapor deposition (CVD) system, which was then transferred onto Al_2O_3-deposited gold thin film. A 2 uL droplet containing 100 diluted gold

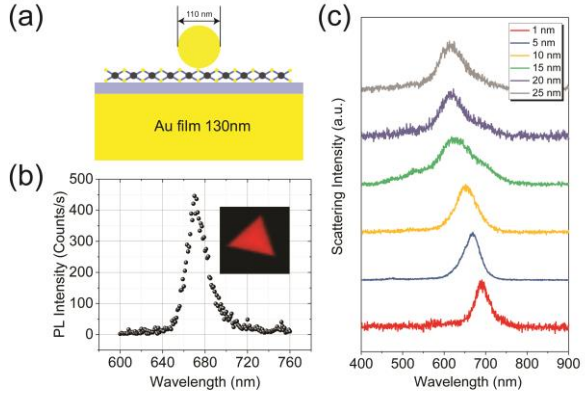

Figure 1. NGMF nanostructure schematic and characterization. (a) Illustration of MoS_2 monolayer situated into a NGMF system with various gap thickness. (b) Typical PL spectrum of MoS_2 monolayer on glass substrate. Inset: PL intensity mapping a MoS_2 monolayer sample. (c) Dark field scattering spectrum of NGMF systems with various gap thicknesses.

nanospheres was then drop cast on the samples, forming the final NGMF nanostructures (Figure 1a). Figure 1b is a typical photoluminescence (PL) spectrum of MoS_2 monolayer on glass sample with a dominant A exciton peak at 675 nm. The homogeneous PL emission from a triangle MoS_2 flake indicates a high quality of MoS_2 monolayer. The dark field spectrum in Figure 1c show that the plasmon resonance of NGMF structures can be precisely tuned from 690 nm to 620 nm via varying the spacer thickness from 1 nm to 25 nm as the spacer thickness is varied from 1 nm to 25 nm.

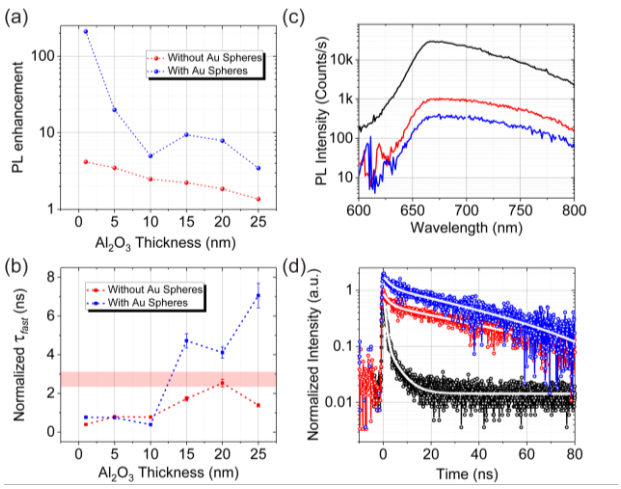

Figure 2. Spontaneous emission modification of MoS_2 monolayer coupled to NGMF. (a-b): MoS_2 monolayer PL intensity enhancement (a) and fast decay (b) as a function of spacer thicknesses. (c-d): Typical PL spectra (c) and decay curves (d) of MoS_2 monolayer coupled to NGMF. The black, red, and blue curves correspond to the 1, 15, and 25 nm spacer, respectively.

Upon 532 nm pulse laser excitation, NGMF nanostructure with 1 nm spacer thickness exhibits 211-fold PL enhancement at 676 nm, followed by a significant decrease as the spacer thickness increases to 5 nm, and remains almost the same with some fluctuation when the spacer is thicker than 10 nm, as shown in Figure 2a, c and d, respectively. It should be noted that monolayer MoS_2 is a direct bandgap material with strong in-plane PL emission, while NGMF exhibits strong field enhancement at out-of-plane direction. Such orthogonal properties have led to weaker with MoS_2 as compared to NGMF based on nanocubes[3]. Samples of MoS_2 monolayer onto Al_2O_3 coated gold film without Au nanospheres show similar trend with much smaller field enhancement from gold film only. The enhancement in spontaneous emission rate can be determined by decay time which can be measured by time resolved fluorescence measurements. Figure 2b illustrates spontaneous emission decay modification of MoS_2 monolayer with/without gold nanospheres. When the spacer is thinner than 10 nm, the fast decays are almost the same (below 1 ns which is the limited to our photodetector resolution) for the samples with and without gold spheres. However, MoS_2 monolayer coupled into NGMF structures with more than 10 nm spacer exhibits longer lifetime than MoS_2 monolayer on SiO_2/Si. At large spacer thickness, the electric field confinement shifts from the spacer layer into the gold nanospheres, causing a shorter decay due to the quenching resulting from direct contact of MoS_2 monolayer with the gold nanosphere. For samples without spheres, the effect of gold mirror weakens as spacer increases, therefore, the decay becomes comparable with monolayer on SiO_2/Si substrate.

3. Conclusions

We have successfully demonstrated that the plasmonic resonance can be greatly tuned by varying spacer thickness in NGMF structures. Strong modification of spontaneous emission from MoS_2 monolayer has been observed in NGMF plasmonic structures. These NGMF could potentially be used as bright light emitting diodes when electrically pumped, with estimated 3GHz operational speed.

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