Optical Antenna Enhanced Spontaneous Emission from CVD-Grown Monolayer WS_2

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Abstract: We report on efficacy of light emission enhancement of CVD-grown WS_2 films under monomer and dimer plasmonic nanoantennas separated by a spacer layer from the WS_2 emitter.
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1. Introduction to the style guide, formatting of main text, and page layout

The direct band gap of monolayers of several chemically, structurally and electronically similar 2H phased semiconducting TMDs such as MoS_2, $MoSe_2$, WS_2, and WSe_2 has shown light emission in the visible and near-infrared spectrum. A drawback of TMD materials for optic and photonic application is the weak light-matter interaction originating from the atomic thickness of the monolayer material. This, however, can be enhanced via resonant (cavity, surface plasmon resonance) and non-resonant (waveguide dispersion, index tuning, metamaterials) systems [2-7]. Antennas act as an impedance transformer matching k-vectors of free-space with that of the atomic-system, and also provide local field enhancement utilized in the weak-coupling regime (Purcell effect) for emitters. As such, here we utilize optical antennas to increase the excitation rate and simultaneously enhance the local density of states (DOS) of the emission process, which modifies the spontaneous emission rate (Purcell effect). Hence, the nanoantenna-TMD system behaves as a dipole-coupled to an electromagnetic resonator that strongly modifies spontaneous emission of fluorescence in its spatial and spectral proximity. Such as system has shown to have short radiative lifetimes and can have a deep Subwavelength optical mode, thus opening the possibility of creating ultrafast, and efficient nanoscale emitters [12-17].

Nanoscale structures consisting of two metallic nanoparticles separated by a small gap (plasmonic dimer) support hybridized plasmon resonances because of the capacitive coupling between the plasmon modes of each nanoparticle. For a quantum emitter placed inside such gap, this coupling strongly localizes charges at the junction between the two nanoparticles, giving rise to large field enhancements at the center of the feed-gap of the dimer antenna. However, for emitters that are not of quantum dot dimensions, such as a TMD layer, the emitter layer must be in an offset from the mid gap, either below or above the plane of the antenna. For emitters with zero distance from the plasmonic antennas, the strong field gradients of the point dipole source can efficiently excite higher multipolar lossy modes of the antenna which are mostly dark or weakly coupled to the radiation field and therefore convert EM energy into heat. To avoid this emitter quenching effect and its coupling to lossy plasmonic surface waves, the emitter should be separated from the metallic nanoparticle by a distance previously reported to be about ~8-10 nm. In the case of TMDs, the dielectric spacer layer can be simultaneously used to control and maintain the material quality. Thus, for a layered material emitter that is positioned in an offset from the gap center, dimer antennas have higher filed enhancement only by a factor of about 2x compared to a monomer disk. That is, the cavity field enhancement is not originating from the gap between the metal particles, but from each monomer disc. In this work, we compare the TMD emission enhancement of monomer vs. dimer antennas relative to intrinsic PL emission (Figure 1a).

In previous TMD-plasmonic antenna cavity nanostructures the material system was based on exfoliated 2D material or colloidal cavities. However, little attention has been paid to a scalable approach for achieving the hybrid nanostructures. Thus, here we utilize large-area and high-quality WS_2 flakes prepared by chemical vapor deposition (CVD) instead of exfoliation. A CVD process is the preferred method for synthesizing TMD materials due to its reproduce-able nature of growing pristine monolayer materials. Moreover, fine control of optical antenna dimensions by electron beam lithography (EBL) enables 10's of nanometer precise control over the plasmonic cavity dimensions (inset Fig. 1b). Single- and Multi-layer Tungsten Disulfide (WS_2) was grown directly on SiO_2-on-

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Si substrate via ambient-pressure CVD utilizing a tube furnace. After deposition of WS_2 thin layers, standard EBL process was used to fabricate the plasmonic nanoantennas.

For the antenna mediated pumping under a 532-nm wavelength Gaussian beam with a micron radius, the 75-nm radius dimer cavity enhanced photoluminescence emission of monolayer WS_2 flakes exhibits a 3.2-fold (2.7-fold) enhancement in peak intensity (in integrated PL count) relative to emission from the control sample at the same excitation power density (Figure 1b). Whereas, the 75-nm radius monomer cavity enhances the PL emission by 1.9-fold (1.8-fold). This is because the absorption and emission processes of the WS_2 monolayer is coupled to the local fields of an optical antenna. The coupled system has a larger absorption cross-section compared with that of the isolated emitter (Figure 1c). Also, the cavity resonance is matched with emission wavelength of the monolayer WS_2, leading to higher Q factor and consequently Purcell effect up to 30, as evident in band narrowing of the spectral response of the emitter.

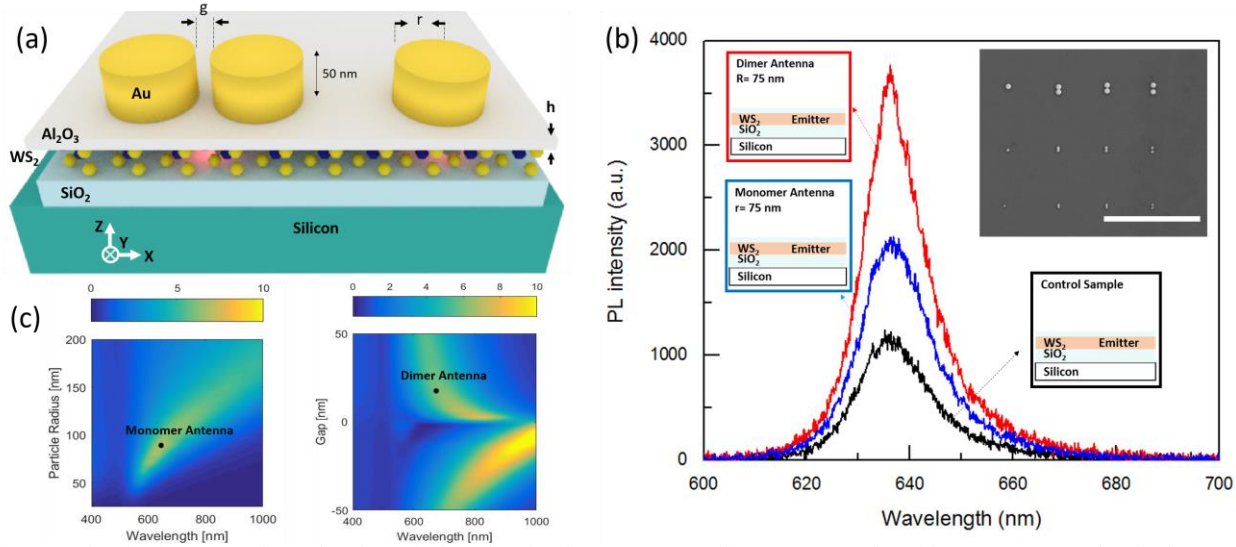

Figure 1: Photoluminescence of WS2 is enhanced when placed under a monomer or dimer antenna cavity with a resonance wavelength close to emission. a) Schematic of the purposed optical antenna types for PL enhancement of monolayer TMDs. b) PL intensity of CVD-grown monolayer WS2 before and after fabrication of 2 different optical antennas. Insets is an SEM image of a set of various fabricated antenna cavities on monolayer WS2; the scale bar is 5 microns. (c) Far field scattering efficiency (Q_{scat}) of monomer nanodisc antennas for radius range of 50 nm to 200 nm and for dimer nanoantennas of single 75 nm radius and in a gap sweeping range from -50 nm (overlapping charge transfer mode) to 50 nm (gap plasmon mode). Denoted points correspond to two cavity enhanced PL spectrum presented.

We have demonstrated that optical nanoantennas can be used to control the emitting properties of monolayer TMDs. These emission dynamics point toward visible light sources based on lithographically fabricated nanoantennas. Both monomer and dimer nanoantenna architecture are scalable to emission resonances of other members of the TMD family such as $MoTe_2$, at telecommunications wavelengths in the near infrared. Finally, the resultant nano-cavities may efficiently enhance light-matter-interaction for optical and photonic components based on 2D materials, such as LEDs, lasers, optical modulators and solar cells [7-11].

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