Light Emission Enhancement of 2D Materials in Monomer vs. Dimer Nanoantennae

Mohammad H. Tahersima[1], M. Danang Birowosuto[3], Zhizhen Ma[1], William C. Coley[2], Michael Valentin[2], Sahar Naghibi Alvillar[2], Cesare Soci[3], Ludwig Bartels[2], Volker J. Sorger[1]

1 Department of Electrical and Computer Engineering, George Washington University, 800 22nd Street NW, Washington, DC 20052, USA
2 Chemistry, Materials Science & Engineering and Electrical Engineering, University of California, Riverside, California 92521, USA
3 Division of Physics and Applied Physics, Nanyang Technological University, 21 Nanyang Link, Singapore 637371, Singapore

Abstract: We show that the emission enhancement from a TMD emitter- monomer antenna cavity system rivals that of dimers at much reduced lithographic effort.
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The atomic thickness of these two-dimensional (2D) materials results in low photo absorption limiting the achievable photo luminescence intensity. Such emission can, in principle, be enhanced via nanoscale antennae resulting in; a) an increased absorption cross-section enhancing pump efficiency, b) an acceleration of the internal emission rate via the Purcell factor mainly by reducing the antenna's optical mode volume beyond the diffraction limit, and c) improved impedance matching of the emitter dipole to the free-space wavelength [2-6]. Plasmonic dimer antennae show orders of magnitude hot-spot field enhancements when an emitter is positioned exactly at the mid-gap. However, a 2D material cannot be grown, or easily transferred, to reside in mid-gap of the metallic dimer cavity. Hence, it is not plausible to simply take the peak intensity as the emission process enhancing value. In addition, a spacer layer between the cavity and the emissive material is required to avoid non-radiative recombination channels. Using both computational and experimental methods, in this work we show that the emission enhancement from a 2D emitter- monomer antenna cavity device rivals that of dimers at much reduced lithographic effort. We observe a representative 3-fold higher emission for the dimer cavities as compared to intrinsic emission of chemical vapor deposition synthesized WS_2 flakes (Figure 1.c).

The emission intensity profile of an emitter in an optical-cavity-antenna is governed by:

$$I_{out} = \left[I_0 . \left(\frac{\int \frac{|E|^2}{|E_0|^2} ds}{C_{geo}} \right) \right]_{\lambda - Pump} [QE . F_p]_{\lambda - Emit} \tag{1}$$

Regarding the internal photon generation enhancement process of the TMD-cavity system, we focus on the near electric field enhancement and Purcell product in the Eqn. 1. The overall fluorescence enhancement of monolayer WS_2 by the plasmonic optical antenna can be expressed as the product of excitation rate enhancement, the spontaneous emission probability enhancement (Purcell effect), and outgoing portion of the spontaneous emission.

Since the emitter is excited optically, the excitation rate enhancement is then proportional to ratio of the squared electric field of the emitter with the optical cavity and without the cavity. Here, care must be taken to describe the physical observable accurately. It is tempting to consider the peak field enhancement of a dimer. This is however not an accurate interpretation of the actual experiment often as well as conducted here. Because the spot size of our pump laser beam is significantly larger than the physical area of the antennae, the excitation and hence the photon generation is not a local, hot-spot like effect, but rather originates from an average across the pump beam dimensions. To obtain a) an accurate field enhancement originating from a ~0.8 micrometer large pump diameter, and b) a complete picture of the nature of electric field enhancement distribution due to presence of either monomer or dimer antennae, we calculate the spatially resolved electromagnetic field profile both at the location of the monolayer WS_2 as well as at the cross section of the optical antenna for both the excitation and emission wavelengths (Figure 1.b, d). The photon generation rate, here defined as the product between the quantum efficiency and the Purcell factor, is equal to the quantitative radiative decay rate enhancement. Also, the *QE* of the fluorescence process is estimated by the ratio of the radiated power measured in the far-filed to the total power injected by the emitter.

We see that the theoretical values agree with experimental results in Table 1. From Wheeler's limit, we expect to find higher quality factors for smaller antennae. Such narrowing of the spectral response due to smaller cavity radius is evident from our experimental results summarized in Figure 1.a, which suggests that the quality factor of nanoantenna increases with reducing dimensions of each disc.

In conclusion, we have demonstrated that optical nanoantenna can be used to control the emitting properties of monolayer TMDs. This control was achieved using two types of the metallic cavities (monomer vs dimer). These emission dynamics were also supported by numerical calculations. In particular, we have demonstrated that the

fluorescent enhancement of 2D materials unlike quantum dots is an areal average effect. Such optical 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 [7-15].
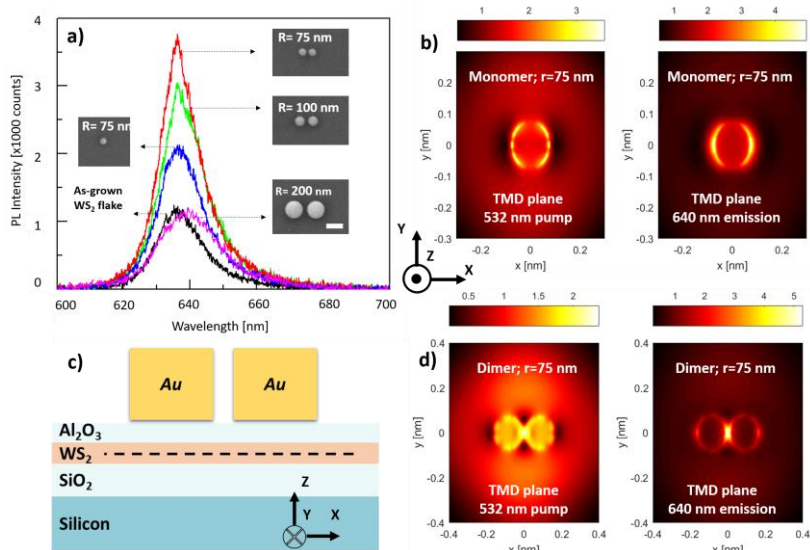

Figure 1: a) PL intensity of CVD-grown monolayer WS_2 before and after fabrication of 4 different optical antennae. Insets are SEM images of each type of optical cavity; the scale bar is 400 nm. Electric field distribution profile of the emitter at the proximity of monomer (b) and dimer nanodisc optical antenna (d). c) Schematic of the optical antenna on monolayer TMDs.

Table S1. Summary of simulation and experimental results for both monomer or a dimer antennae. Theoretical values are presented for three cases: spatial maxima of PL enhancement, averaged PL enhancement over the area of antennae, and over the area of excitation beam.

Monomer 75 nm radius	Formula	Peak	Antenna Average	Beam Average
Simulation Excitation rate enhancement	$\gamma_{exc}/\gamma_{exc}^0$	5.3	3.0	1.5
Simulation Radiative rate enhancement	$F_p \times QE$	52.2	10.0	1.7
Simulation PL enhancement	$(\gamma_{exc}/\gamma_{exc}^0) \times (F_p \times QE)$	276.7	30.0	2.1
Experimental PL enhancement	$< EF_{exp} > = \left(\frac{I_{cav}}{I_0}\right)/\left(\frac{S_{cav}}{S_0}\right)$	NA	$1.79 \times (400^2/75^2)$ = 50.9	1.8
Dimer 75 nm radius	Formula	Peak	Antenna Average	Beam Average
Simulation Excitation rate enhancement	$\gamma_{exc}/\gamma_{exc}^0$	6.8	2.3	1.4
Simulation Radiative rate enhancement	$F_p \times QE$	39.0	10.1	2.3
Simulation PL enhancement	$(\gamma_{exc}/\gamma_{exc}^0) \times (F_p \times QE)$	265.2	23.0	3.2
Experimental PL enhancement	$< EF_{exp} > = \left(\frac{I_{cav}}{I_0}\right)/\left(\frac{S_{cav}}{S_0}\right)$	NA	$2.74 \times (400^2/(2\times75^2))$ =39.7	2.7

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