Beaming of WS$_2$ Photoluminescence from a film coupled microsphere antenna

Shailendra K. Chaubey*, Sunny Tiwari, Gokul M. A., Diptabrata Paul, Atikur Rahman, G.V. Pavan Kumar*

Department of Physics, Indian Institute of Science Education and Research, Pune-411008, India

*E-mail: shailendrakumar.chaubey@students.iiserpune.ac.in

*E-mail: pavan@iiserpune.ac.in

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Abstract: Engineering optical emission from two dimensional, transition metal dichalcogenides (TMDs) materials such as Tungsten disulphide (WS$_2$) has implications in creating and understanding nanophotonic sources. One of the challenges in controlling the optical emission from 2D materials is to achieve narrow angular spread using a simple photonic geometry. In this paper, we study how the photoluminescence of a monolayer WS$_2$ can be controlled when coupled to film coupled microsphere dielectric antenna. Specifically, by employing Fourier plane microscopy and spectroscopic techniques, we quantify the wavevector distribution in the momentum space. As a result, we show beaming of the WS$_2$ photoluminescence with angular divergence of $\theta_{1/2} = 4.6^\circ$. Furthermore, the experimental measurements have been supported by three-dimensional numerical simulations. We envisage that the discussed results can be generalized to a variety of nanophotonic 2D materials, and can be harnessed in nonlinear and quantum technology.
1. Introduction:

2D transition metal dichalcogenides (TMDs) have attracted major attention in recent years because of their unique optical and electronic properties [1] [2] [3]. High exciton binding energy and direct bandgap makes them suitable candidates for optoelectronics device applications [3] [4] [5]. TMDs show other unique properties such as valley polarization which make it suitable for valleytronics and spin-orbit interaction studies [6, 7].

Influencing and controlling the emission properties of the TMDs is key to improving the efficiency of optoelectronic devices. Recent advancements in the field of plasmonic and dielectric nanoantenna have enabled the control over the fundamental emission properties such as wavevector distribution and Purcell enhancement [8-11]. Furthermore, plasmonic nanostructure coupled to TMDs has been studied for strong coupling [12-14], PL enhancement [15-18], surface enhanced Raman scattering [19, 20], spectrum tailoring [21, 22], trion enhancement [23] and directional emission [24]. Although plasmonic nanostructure provide great control over the emission properties, they suffer large Ohmic losses and have broad resonances [25]. On the other hand dielectric structures such as microdisk, cylinder and microsphere show low absorption, and sharp resonances which make them suitable candidate for controlling the emission properties without much losses.

To this end, studying the optical transition characteristics of TMDs coupled whispering gallery mode of the dielectric microsphere and disk has gained relevance. A variety of prospects like PL enhancement [26], lasing [26-28], directional emission [29] and out of plane dipole excitation [30] has been achieved using such geometries. Recently hybrid metallo-dielectric cavity has been used to enhance light matter interaction [31-33], build hybrid metasurfaces [34], tailor the quality factor [35], and in designing directional sources [36-38]. Especially, dielectric particle on the mirror is one such geometry which is widely used for hybridizing surface plasmon and WGM [34], studying
wavevector and polarization state of emission from fluorescent dye \textsuperscript{39} and probing magnetic resonance \textsuperscript{40}. Although, basic emission properties of TMDs such as, Purcell enhancement, polarization of emission has been studied in great detail but the wavevector distribution is relatively less explored using geometries involving complicated fabrication and etching techniques \textsuperscript{41}.

In this context, tunability of directionality of the optical emission from nanostructures coupled to TMDs remains a challenging problem and highly directional emission from TMDs is yet to be achieved. Motivated by this, we study the optical emission properties of a monolayer WS\textsubscript{2} coupled to a metallo-dielectric antenna. In this paper we have reported the photoluminescence beaming of monolayer WS\textsubscript{2} coupled to this antenna. Specifically we study the wavevector distribution of monolayer WS\textsubscript{2} coupled to a metallo-dielectric antenna. Using the Fourier plane imaging we have characterized the beaming and observe that the divergence angle is as narrow as $\theta_{1/2} = 4.6^\circ$ which correspond to the NA of 0.06. We numerically study the near-field and far-field properties of our system by placing an in-plane and out of plane dipole using finite element method based simulations.

**Result and Discussion:**

Monolayer WS\textsubscript{2} has been grown using chemical vapor deposition (CVD) \textsuperscript{42,43} characterized using Raman and PL spectra (See supplementary Information S1). WS\textsubscript{2} flakes were transferred to Al\textsubscript{2}O\textsubscript{3} gold mirror using wet chemical method. 3 nm Al\textsubscript{2}O\textsubscript{3} layer is used between WS\textsubscript{2} and mirror to avoid the charge screening which might lead to PL quenching. Microsphere has been dropcasted on the transferred monolayer WS\textsubscript{2}. Monolayer WS\textsubscript{2} placed between dielectric microsphere and gold mirror is excited with 532 nm laser as shown in the figure 1. 100 X, 0.95 numerical aperture (NA) objective lens was used in backscattered configuration for both excitation and collection.
Backscattered signal was collected using the same objective lens and projected into EMCCD and spectrometer for imaging and spectroscopy. This microsphere gold mirror acts a metallo-dielectric antenna and direct the WS$_2$ signal to a narrow range of wavevector. Directional beaming of PL was probed using Fourier plane as shown in the inset of the figure 1.

**Figure 1.** Schematic of the experiment. WS$_2$ monolayer was placed over a gold film separated by a 3 nm Al$_2$O$_3$ spacer layer. Polystyrene microsphere of diameter 7 µm is placed over the monolayer WS$_2$ flake. Microsphere is excited with 532 nm CW laser. The emitted light was collected using the same objective lens and projected to spectrometer and EMCCD for spectroscopy and Fourier plane imaging, respectively. Inset represents the Fourier plane image showing the beaming of the WS$_2$ photoluminescence.

To project the Fourier plane from the back aperture of the objective lens to the EMCCD, 4f configuration was used \cite{44, 45}. Combination of edge and notch filters was used in output path to
efficiently reject the elastically scattered light. It was clear from the PL spectrum that the elastically scattered light was effectively rejected (See supplementary information S1). For detailed experimental setup see supplementary information S2.

**Figure 2.** Beaming of WS<sub>2</sub> photoluminescence. (a) Bright-field image of a microsphere over WS<sub>2</sub>, which is placed over a gold mirror with 3 nm spacer layer of Al<sub>2</sub>O<sub>3</sub>. Boundaries of WS<sub>2</sub> monolayer are shown with red dotted lines. Microsphere was excited with 532 nm CW laser (b) PL spectrum of WS<sub>2</sub> monolayer coupled to mettalo-dielectric antenna, collected after rejecting the elastically scattered light. (c) Fourier space intensity distribution of the PL emission from the WS<sub>2</sub> coupled to antenna. (d) Measured energy-momentum spectra of PL emission form the same across the white dotted line.
Figure 2a is the bright-field image of a 7µm polystyrene microsphere placed on a WS$_2$ monolayer over gold mirror. Red dotted line shows the boundaries of the WS$_2$ monolayer. Microsphere was excited using 532 nm tightly focused laser which excites the PL of the WS$_2$ monolayer. Because of the near-field excitation, it couples to the whispering gallery modes (WGMs) of the microsphere. Figure 2 (b) shows the PL spectra for the WS$_2$ coupled to metallo-dielectric antenna. WGMs of microsphere can be seen as sharp peaks riding over a broad PL spectrum. To study the wavevector distribution of emission, Fourier plane microscopy and spectroscopy has been performed. Fourier plane image map the wavevectors of the light in terms of θ and Φ coordinates. Radial coordinate in Fourier plane image is numerical aperture ($NA = n \sin \theta$), and Φ is azimuthal coordinate varies from 0 to $2\pi$. Figure 2 (c) shows the Fourier plane image of the PL emission from this system. It can be observed from the figure 2 (c) that the θ spread is very small and most of the emitted wavevectors are beaming at the center of the Fourier plane image. The metallo-dielectric antenna has utilized the photonic nanojet effect of the microsphere which focused the emitted light into small area $^{[46, 47]}$. Photonic nanojet is narrow high intensity propagating electromagnetic wave when a dielectric microsphere is illuminated with plane wave. It is not a resonant phenomenon and completely dependent on the wavelength and relative size of the microsphere so it can be used for broad wavelength range. When microsphere is illuminated with light beam, it act as a micro lens and focusses the light to a small area by photonic nanojet effect. By similar effect the emitted PL from the WS$_2$ monolayer is focused into a small areas toward the other side of the microsphere $^{[48]}$. This leads to a narrow range of wavevector distribution for the PL emission and WS$_2$ PL beaming is observed.

To get the wavelength information associated with the wavevectors, a thin slice of the Fourier plane image has been filtered using the slit of spectrometer at $k_x/k_0 = 0$ (see white dotted line in
figure 3 (c)) and dispersed using spectrometer to obtain the energy-momentum (E-k) spectrum. It can be seen in the E-k spectrum, emission is concentrated at \( k_x/k_0 = 0 \) for entire PL wavelengths. This further rejects the possibility of elastically scattered light at the center of the Fourier plane image. Since all the emission is concentrated near \( k_x/k_0 = 0 \) it rejects the role of WGM in the directionality. To quantify the spread of the emission wavevectors, intensity cross-cut profile of the Fourier plane image across the white dotted line has been plotted in figure 3 (d). These cross-cut profile has been fitted with Lorentzian function to calculate the full width at half maxima (FWHM). We found that at the FWHM of the intensity, the divergence angle \( \theta_{1/2} = 4.6^\circ \). We have performed the multiple set of experiment and average angular divergence over 4 sample is found \( \theta_{1/2} = 5^\circ \) with standard deviation of 0.5° (See supplementary information S3)
Figure 3. Comparison of wavevector distribution in different geometry. Fourier space intensity distribution (a) showing photoluminescence beaming from a microsphere on Si/SiO$_2$ substrate mirror, (b) microsphere on Si/SiO$_2$ substrate (c) WS$_2$ on gold mirror. (d), (e) and (f) are the intensity cross cut along the white dotted line. A ring shaped emission was observed in the Fourier plane image as shown by red arrow in 3 (d).

To highlight the role of our unique system, similar experiments has been performed in different configurations. Figure 3 (a) shows the wavevector distribution of WS$_2$ PL without coupling to any antenna directly on Si/SiO$_2$ substrate. It is clear from the figure 3 (a) that PL emission from WS$_2$ is isotropic in nature. Figure 3 (b) shows the wavevector distribution when a microsphere is placed directly over the WS$_2$. It can be seen from the figure 3(b) that the emission is directional in nature. Figure 3 (c) shows the wavevector distribution of the WS$_2$ PL in case of the WS$_2$ coupled to metallo-dielectric antenna. In case of metallo-dielectric antenna wavevector distribution is further narrowed down and beaming of the emission is observed as discussed. Intensity cross-cuts along the white dotted line is plotted in the figure 3 (d), (e) and (f). To quantify the angular spread, cross-cut curve in figure 3 (d) and (e) was fitted using Lorentzian function (See supplementary info S4). FWHM of the fit is calculated for both the intensity profile shown in figure 3 (e) and 3 (f). At FWHM intensity, angular diverge for the microsphere on Si/SiO$_2$ substrate and metallo-dielectric antenna are found to be 11.1° and 4.6° respectively. Angular divergence in case of the microsphere on Si/SiO$_2$ substrate is reduce because of photonic nanojet effect of the microsphere [48] [49]. In case of the metallo-dielectric antenna this divergence is further reduced. Since photonic nanojet effect depend on the ratio of the microsphere diameter to wavelength (D/λ) we have performed the microsphere size dependent experiment. We have performed the experiment with metallo-dielectric antenna made with 3 μm and 5 μm SiO$_2$ microspheres on the gold mirror. It is observed
that the angular spread is narrowed down for the bigger microsphere which further confirm that beaming is caused because of the photonic nanojet effect as increasing the microsphere size increase the ratio $D/\lambda$. (See supplementary information S5). This confirms that the beaming of the emission is caused by photonic nanojet effect.

**Figure 4.** Effect of focusing Fourier plane image of the emission for focusing in different plane of the microsphere, (a), (b) (c) and (d) are schematic of different focusing scheme and (e) (f) (g) and (h) are corresponding Fourier plane images.

Since in photonic nanojet effect microsphere act as a micro-lens and emission wavevectors depend at which plane microsphere focuses the light, focusing dependent experiment has been performed.
Input light has been focused in the different plane of the microsphere and wavevector distribution of PL emission is studied. Figure 4 (a) (b) (c) and (d) are the different focusing scheme in and (e) (f) (g) and (h) are corresponding Fourier plane images. It is observed that the divergence is smallest in case when input lased is focused slightly below the sample plane. When the objective lens focusses the light slightly below the sample plane microsphere act as a micro-lens and focused the light at the sample. When objective lens focused the light in sample plane, the light that reaches the sample plane is defocused because of the second focusing caused by microsphere lens.

**Figure 5.** Calculated Fourier plane imaging of emission from dipoles placed between microsphere and mirror. (a) Experimentally measured Fourier plane image of emission from a TMD monolayer sandwiched between a 3 µm SiO₂ microsphere and 160 nm gold mirror. Calculated Fourier plane
image of emission from a dipole oscillating in-plane (x axis) (b) and out-of-plane (z axis) (c). Calculated near-field electric field of a 3 μm SiO₂ microsphere placed on a gold mirror excited by placing an oscillating dipole at 645 nm wavelength with in-plane (x axis) (d) and out-of-plane (z axis) dipole in the gap between microsphere and mirror (e).

To corroborate the experimental findings and to understand the directional emission of TMD coupled WGMs emission from the mettalo-dielectric antenna, we performed finite element method based numerical calculations in COMSOL Multiphysics. We performed near-field calculations and calculated Fourier plane images by using reciprocity arguments. We calculated the near-field profile of a 3 μm diameter SiO₂ microsphere placed on a 160 nm thick gold mirror. The gap between the microsphere and gold mirror was set as 5 nm for accounting the Al₂O₃. We used a 3 μm microsphere because of the computational constraints. Figure 4(a) shows the experimentally measured Fourier plane image of the emission from a WS₂ monolayer coupled to metallo-dielectric antenna formed by a 3 μm SiO₂ microsphere and gold mirror. To understand the wavevector distribution we calculated the near-field electric field of a 3 μm SiO₂ placed on a gold mirror and excited by placing in-plane and out-of-plane dipoles. We placed an oscillating dipole at 645 nm oriented along the in-plane (x axis) (figure 4(d)) and out-of-plane (z axis) (figure 4(e)). We chose the wavelength of oscillation to be 645 nm as the PL emission has a peak around this wavelength. Since the contribution of in plane dipole dominates the emission in 2D materials, the experimental Fourier plane image Figure 5 (a) matches well with the calculated Fourier plane image for the in plane dipole Figure 5 (b) In case of the gold mirror, microsphere on gold mirror form a cavity which localized the electric field near the contact point of mirror and the microsphere. To understand this effect, microsphere on glass and microsphere on mirror system is excited with Gaussian beam numerically (See supplementary information S6). As can be seen that in case of
microsphere on gold mirror near-field electric field is more localized. We have already seen in focusing dependent experiment that localization of the electric field leads to narrowing of wavevector distribution. To corroborate this effect numerically, we have performed the calculation by placing 3 dipoles (1) 240 nm apart (2) 80 nm apart. (See supplementary Information S7). In case of 80 nm separation emission is more directional which explains the narrowing down of the emission wavevector on gold mirror.

To calculate the far-field profiles we used the reciprocity arguments [50]. For in-plane x oriented dipole the calculated Fourier plane image matches very well with the experimentally obtained Fourier plane image. Majority of the light is coming out in a beaming manner and is confined to a very narrow range of wavevectors. The calculated Fourier plane image for emission from out-of-plane dipole shows that the emission is coming out in a doughnut shaped pattern and is also directed towards the center of the Fourier plane image. In the Figure 2 (c) the emission also contains a doughnut shaped pattern but it is very weak and the simulations show that it is because of the z-dipoles. The results shows that the contribution of in-plane dipoles is more pronounced in case of metallo-dielectric antenna.

4. **Conclusion:**

To summarize, we have experimentally studied wavevector distribution of PL coupled it to a metallo-dielectric antenna. By performing the experiment in the different configuration we have shown that the PL emission of WS₂ sandwiched between microsphere and gold mirror out-couples in a beaming manner. Microsphere on the mirror geometry acts as an excellent metallo-dielectric antenna to direct the emission from TMDs in a very small range of wavevectors. In Fourier plane at the FWHM intensity, the angular divergence $\theta_{1/2} = 4.6^\circ$ was achieved. We have shown that increasing the size of the microsphere further reduces the angular spread of the PL emission. The
experimental findings were corroborated with three dimensional finite element method based numerical simulations. This hybrid directional antenna will find relevance in 2D material based nanophotonic chips and nano-lasers.

4. Experimental Methods:

**WS₂ synthesis:** The substrates was first cleaned by acetone and then with isopropanol (IPA) followed by O₂ plasma cleaning at 60 W for 10 minutes. An alumina boat containing 500 mg of WO₃ is loaded with substrates was placed inside a quartz container of 3.5 cm internal diameter, inside the furnace. Another boat containing 500 mg of Sulfur was kept inside the tube, 15 cm away from the WO₃ boat. The Sulfur boat was put outside the furnace and was heated independently utilizing another heater. At first, the tube was flushed with 500 standard cubic centimeters each moment (SCCM) of argon for 10 min, and afterwards, the stream was diminished to 30 SCCM. The furnace was increased to 850°C slowly at a constant rate. As the furnace reached 850°C, the Sulfur was evaporated using a second heater. These temperatures were kept up for 10 min for the growth of the WS₂ monolayer. Once growth is completed, the system was left to cool down to room temperature.

**Sample Preparation:** Microspheres were purchased from Sigma-Aldrich. Gold mirror was prepared by thermal vapor deposition (TVD) by depositing 160 nm gold film on a glass cover-slip. Monolayer WS₂ flakes were transferred onto the gold mirror using wet chemical method. Briefly, polystyrene solution is spin coated on the WS₂ sample followed by baking at 100°C. Sample was place in water and WS₂ was fished on desired substrate with polystyrene support film when it started floating in water. Polystyrene was removed using toluene followed by cleaning the sample using acetone and IPA. Al₂O₃ spacer layer was deposited using atomic layer deposition. After
transferring the WS$_2$ monolayer on the Al$_2$O$_3$ coated gold mirror, MS were dropcasted on it and sample was dried in vacuum desiccator.

**Experimental Set-up:** Sample was excited using a 532 nm CW laser. 100 X, 0.95 numerical aperture (NA) objective lens was used in backscattered configuration for both excitation and collection. Backscattered signal was collected using the same objective lens and projected into EMCCD and spectrometer for imaging and spectroscopy.

**Supporting Information**

Supporting information file is available.

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Supplementary Information

Beaming of WS₂ Photoluminescence from a film coupled microsphere antenna

Shailendra K. Chaubey*, Sunny Tiwari, Gokul M. A., Diptabrata Paul, Atikur Rahman, G.V. Pavan Kumar*

1Department of Physics, Indian Institute of Science Education and Research, Pune-411008, India

*E-mail: shailendrakumar.chaubey@students.iiserpune.ac.in
*E-mail: pavan@iiserpune.ac.in

S1. Optical characterization of monolayer WS₂

Figure S1. Raman and Photoluminescence (PL) spectrum of the monolayer WS₂, (a) PL spectrum including excitation wavelength to make sure that we are efficiently rejecting the Raleigh scattered light. Excitation wavelength is shown with arrow has negligible intensity in comparison to PL intensity. (b) Raman spectrum of the monolayer WS₂ where peak E₂g¹ and A₁g are at 350 cm⁻¹ and 417 cm⁻¹. Since WS₂ was first grown on Si/SiO₂ we can see the silicon peak at 520 cm⁻¹. PL and
Raman spectrum is collected using 532 nm laser. We see very high PL count which shows that the WS$_2$ is monolayer. Further Raman measurement shows a large $E_{2g}^1$ at 350 cm$^{-1}$ and small $A_{1g}$ peak at 350 cm$^{-1}$. Intensity of $E_{2g}^1$ peak is an order of magnitude higher than of $A_{1g}$, which further confirms the sample is monolayer WS$_2$. Sample are grown on Si/SiO$_2$ substrate we can see silicon Raman at 520 cm$^{-1}$.

S2. Experimental Setup:

![Experimental Setup Diagram]

**Figure S2.** Experimental setup. The sample was excited with 0.95 NA, 100x objective lens. The backscattered light was collected using the same lens. The 532 nm laser light was expanded using a set of two lenses L1 and L2. M1 is a mirror. The polarization of the incoming laser was controlled by a $\lambda/2$ wave plate in the path. BS1 and BS2 are beam splitters to simultaneously excite the sample.
with laser and its visualization using white light. Lens L3 is used to loosely focus white light on 
the sample plane. F1, F2, and F3 are set of two edge filters and one notch filter to reject the 
elastically scattered light for Fourier plane and energy-momentum imaging. Lenses L4 and L5 are 
used to project the emission to the Fourier plane onto the spectrometer or EMCCD. M2 is a flip 
mirror, used to project the light on the spectrometer for spectroscopy and energy-momentum 
imaging. Lenses L6 and L7 are flip lenses used to switch from real plane to Fourier plane.

S3. PL beaming from multiple Sample:

![Figure S3](image_url)

**Figure S3.** Multiple Fourier plane image showing the PL beaming from showing the repeatability 
of result (a), (b) and (c) are the Fourier plane image showing the PL beaming from 3 different 
microsphere. (d), (e) and (f) are the cross cut along the white dotted line. We have calculated the 
FWHM of this cross cut graph and average spread in Fourier plane is $\theta = 10^\circ$. 


S4. Advantage of gold mirror as substrate:

Figure S4. Effect of the gold substrate, (a) and (b) are the cross cut of the Fourier plane images on gold and Si/SiO$_2$ substrate. Red curve is the Lorentzian fit for the black cross cut. FWHM for these two curve is found to be $\theta = \sin^{-1} (k) = 9.2^\circ$ and $22^\circ$ respectively for gold and Si/SiO$_2$ substrate.
S5. Size dependence of microsphere:

**Figure S5.** Microsphere size dependence: (a), (b) and (c) are the Fourier plane image of three different size of microsphere corresponding to 3, 5 and 7 µm microsphere. (d), (e) and (f) are the cross cut profile across the white dotted line. The FWHM corresponding to this three Fourier plane images are $\theta = \sin^{-1}(k_x) = 18^\circ$, $19^\circ$ and $10^\circ$ respectively.
Figure S5: Near field electric field distribution in x-y plane for a microsphere of size 2 micron when excited using a Gaussian beam of 532 nm wavelength. (a) When microsphere is placed on glass substrate (b) when microsphere is placed over gold mirror. We can see that in case of gold mirror near field electric field is more localized in comparison to glass substrate.
**Figure S7**: Near field and far field in Fourier plane image for a microsphere of size 2 µm: Near field electric field distribution when 3 dipole are placed at a distance (a) 240 nm apart and (b) 80 nm apart. (c) and (d) are the far field Fourier plane corresponding to (a) and (b)

As can be seen from the Figure S7 that when dipole are placed 80 nm apart directional which explains the high directionality from metallo-dielectric antenna.
