In addition, TMDs and plasmon monolayer sandwiched second-harmonic generation, [17] a nanodisk array, [20,28] and trion [19] have been either fabricated over TMDs - [11] orbit interaction studies. This utility of AgNW both as a plasmonic cavity and nanoparticle, [18] have been achieved using such con - [12] optical and electronic properties. (TMDs) have attracted signi - [13] cant attention because of their novel optical and electronic properties. [1–4] Direct bandgap in their monolayer makes them a suitable candidate for electronics applications. [3–5] Very high oscillator strength leads to a narrow peak with a pronounced optical transition. [5–7] In addition, TMDs exhibit valley polarization effects, making them suitable candidate for spin–orbit interaction studies. [8–10] Coupling 2D materials such as a tungsten disulfide (WS2) monolayer to plasmonic nanocavities can enhance and influence light–matter coupling. In this article, we report on our experimental studies of directional photoluminescence (PL) from WS2 monolayer sandwiched in a unique kind of cavity: silver nanowire (AgNW) on a gold mirror.

1. Introduction

In the last few years, 2D transition metal dichalcogenides (TMDs) have attracted significant attention because of their novel optical and electronic properties. [1–4] Direct bandgap in their monolayer makes them a suitable candidate for electronics applications. [3–5] Very high oscillator strength leads to a narrow peak with a pronounced optical transition. [5–7] In addition, TMDs exhibit valley polarization effects, making them suitable candidate for spin–orbit interaction studies. [8–10] Coupling 2D materials such as a tungsten disulfide (WS2) monolayer to plasmonic nanocavities can enhance and influence light–matter coupling. In this article, we report on our experimental studies of directional photoluminescence (PL) from WS2 monolayer sandwiched in a unique kind of cavity: silver nanowire (AgNW) on a gold mirror.

Plasmonic nanocavities are important architectures to study because of the very high electric field inside the cavity caused by hybridization of gap plasmon. [11] Ultrasmall volume inside the cavity provides a large Purcell enhancement factor. [12,13] To this end, nanocavity formed by a nanoparticle on a gold film has been utilized for strong coupling at the single-molecule level, [14] single-molecule surface-enhanced Raman scattering (SERS), [15,16] controlled reflectance properties, [17] and enhanced spontaneous emission. [13,18]

Of relevance to this study is the AgNW on a gold mirror cavity. Nanocavity formed by a AgNW on a gold film, apart from the abovementioned advantages, can also work as an optoplasmic waveguide of cavity’s emission, as shown recently by our group. [19] Such nanocavities are also studied in the context of Purcell enhancement, [18] remote excitation of the molecules, [19] spontaneous emission enhancement, [18] wavevector distribution, [19] and trion enhancement. [20]

Coupling surface plasmon polaritons to TMDs can facilitate interesting optical properties. To this end, studying the optical transition characteristics of TMDs in vicinity of a plasmonic nanostructure near resonance has gained relevance. [21–24] A variety of prospects, such as PL and Raman enhancement, [25,26] enhanced spin–orbit interaction, [20,27] remote excitation of SERS, [28] spectrum tailoring, [29] strong coupling, [30] and trion enhancement, [20] have been achieved using such configuration. For these purposes, many plasmonic structures, such as bowtie antenna, [31] nanodisk array, [32] nanocube, [29] nanoparticle, [30] and NW, [20,28] have been either fabricated over TMDs [13] or TMDs is transferred onto the structures. [14] Specifically, in the context of 2D materials, TMDs have been coupled to a single AgNW for studying remote SERS, [27] second-harmonic generation, [35–38] logic operation, [39] Rabi splitting, [40] and plasmon–exciton interconversion. [20] Silver film–AgNW cavity has been recently utilized for the trion enhancement and enhancing the spin–orbit coupling. [20]

Apart from these effects, a plasmonic AgNW can act as a subwavelength waveguide as well as a nanoscale antenna, which can be harnessed to route an excited signal and emit the signal at the distal end. [41] This utility of AgNW both as a plasmonic cavity and as a waveguide for 2D materials has not been explored, which we do in this article.

Figure 1 shows the studied geometry. It contains a WS2 monolayer sandwiched between a plasmonic AgNW and a gold mirror.
AgNW–WS₂–Au cavity). A thin, Al₂O₃ layer is deposited on gold mirror, which acts as a buffer layer. In such waveguide-cavity systems, a wavevector analysis becomes an important aspect of study to quantify the emission process. Motivated by this, our study focuses on the wavevector distribution of the PL emission in the AgNW–WS₂–Au cavity. Specifically, we excite one end of the cavity with a focused 532 nm laser and study the PL spectrum from the other end of the cavity as a function of angle and polarization. To achieve this, we use polarization-resolved Fourier-plane optical microscopy to study the emission direction of the AgNW–WS₂–Au cavity and quantify the in-plane angular distribution. By studying the angular distribution and polarization-resolved spectra, we show that the AgNW–WS₂–Au cavity can modify the spectral feature of the WS₂ monolayer by interconversion between exciton and trion.

2. Result and Discussion

2.1. Directional PL from AgNW–WS₂–Au Cavity

Figure 2a is the bright-field image of the AgNW placed on a gold mirror AgNW–WS₂–Au cavity. Excitation of NW end, with a tightly focused 532 nm laser, excites the propagating plasmons along the NW. In addition, the excitation of hybridized gap plasmons between the AgNW and the metal film creates high local electric field, which enhances the PL emission from WS₂ monolayer. Because of the near field interaction, the PL emission gets coupled to the NW surface plasmon polaritons (SPPs) travelling along the length of the NW. Because of the spatial discontinuity at the NW end, these SPPs are out-coupled as free space photons. In Figure 2b, we observe strong PL emission from the AgNW–WS₂–Au cavity not only from the excitation and distal ends, but also throughout the NW. This is because of the high electric field in the NW on a metal film cavity, which acts as a hotline along the length of the cavity. Figure 2c shows the emission spectrum collected from the distal end of the AgNW–WS₂–Au cavity. To
study the wavevector of emission from the cavity, we performed Fourier-plane imaging, which maps the emission wavevectors in terms of $\theta$ and $\varphi$ spreading. Radial coordinate in Fourier plane is $NA = n \sin \theta$, and $\varphi$ is the tangential coordinate that varies from 0 to $2\pi$. Fourier-plane image in Figure 2d shows that the emission from the NW end is directed toward higher $k_y/k_0$ and covers only a small range of radial and azimuthal angles, indicating highly directional emission. Multiple measurements for the momentum space image are shown in Figure S4, Supporting Information.

A relevant question to ask is how does the presence of gold mirror influence the performance of the cavity on and off the absorption band of WS$_2$ monolayer? To explore this, we performed experiments similar to Figure 2, but placing the WS$_2$ on a glass coverslip and dropcasting the AgNW over it and exciting it with 532 and 633 nm lasers. In the case of 532 nm excitation, we do not observe any plasmon-assisted propagation from the distal end of the NW because of high absorption of incoming laser by WS$_2$ and NW, whereas for 633 nm excitation, we do observe plasmon propagation (see Figure S3, Supporting Information). Although we observe a plasmon propagation with 633 nm, the PL of WS$_2$ cannot be excited at this wavelength.

2.2. Polarization-Resolved Angular Emission from AgNW–WS$_2$–Au Cavity

The emission at the distal end of NW is mediated through the SPPs of the NW, which generally shows rich polarization signature. In addition to this, emission from the WS$_2$, which is confined in the AgNW–WS$_2$–Au cavity, is enhanced and out-couples through the NW end, after propagating through the cavity. To study the effect of NW and metal film cavity, we performed polarization-resolved Fourier-plane imaging on the PL emission from the distal end of NW. Figure 3a represents the PL image of the AgNW–WS$_2$–Au cavity after rejecting the elastically scattered light captured using electron multiplying charged coupled device (EMCCD). Figure 3b is the momentum space image of the PL emission from the distal end of the NW. Figure 3c is the momentum space image analyzed for the polarization along the long axis of the NW, and Figure 3d represents the momentum space image analyzed for the polarization transverse to NW. For the output polarization along the NW, we observe that light is directional, and most of the emission is centered around $\varphi = 0$. While in case of the output polarization transverse to the axis of the NW, the $\varphi$ spread is more. This means that photons, which are polarized along the wire, are more directional in comparison with the photon whose polarization is transverse to the NW. This is attributed

![Figure 3](https://www.advancedsciencenews.com/advancedphotonicsres/2021/2/2100002/2100002-3.png)

**Figure 3.** Emission-polarization-resolved Fourier-plane imaging of distal end PL emission, which indicates the momentum-space distribution of the emitted light from a particular region in the geometry. a) Real-plane PL intensity distribution of the NW captured using EMCCD integrated over all polarizations (indicated by orthogonal arrows). b) Corresponding momentum space PL intensity distribution, collected from the distal end of the NW (shown by dotted box in (a)). Momentum space PL intensity distribution of the emission from the distal end of the NW when polarization is analyzed c) along the long axis of the NW and d) transverse to the long axis of the NW.
to the polarization maintaining properties of surface plasmon polaritons in an NW.\textsuperscript{[43]}

### 2.3. Polarization-Resolved PL Spectral Characteristics in AgNW–WS\textsubscript{2}–Au Cavity

In the previous section, we discussed about how polarized excitation and collection can influence the directional emission from the AgNW–WS\textsubscript{2}–Au cavity. This further motivated us to study the spectral characteristics as a function of excitation and collection polarization. First, we checked the excitation polarization dependence. The red and black curves in Figure 4a are the PL spectrum from the distal end of the NW AgNW–WS\textsubscript{2}–Au cavity when input excitation polarization is along and transverse to the long axis of the NW, respectively. It can be clearly seen that the output PL intensity is greater when excitation polarization is along the long axis of the NW. With this hindsight, we fixed the excitation polarization along the long axis of the wire and collected the polarization-resolved PL emission from distal end. In Figure 4b, the red and black curves represent the PL emission along and perpendicular to the long axis of the NW. It can be clearly seen that intensity in the case of output polarization along the long axis of the NW is more in comparison with the transverse output polarization. It is also observed that the PL peak in case of the output polarization along the long axis of the NW is red shifted in comparison with the output polarization transverse to the long axis of the NW. This red shift is found to be approximately $\Delta E = 23$ meV.

As the PL spectrum has the contribution of both the neutral exciton and the charged exciton (trion), such red shift can be attributed to the exciton to trion conversion.\textsuperscript{[44]} Trions are essentially formed when exciton is bound by an excess of electron or hole, also called charged exciton. Photoionization and doping are the two main ways to convert exciton into trion.\textsuperscript{[45,46]} In this case, WS\textsubscript{2} is slightly n-doped, so the trions in the system are negatively charged. Exciton to trion conversion is reported by coupling with SPP in metal–insulator–metal (MIM) cavity.\textsuperscript{[20]} In the AgNW–WS\textsubscript{2}–Au cavity, there are two types of plasmon involved. One is propagating along the wire, and the other is localized in the cavity.\textsuperscript{[47]} The intensity component along the NW is larger as compared with the transverse to the NW.

At higher irradiation power, it has been observed that exciton converts to trion, which leads to red shift in PL spectra.\textsuperscript{[46]} To further confirm this, we deconvoluted the PL spectra into two peaks by Voigt function double fit (see Figure S5, Supporting Information). The deconvoluted spectrum is consistent with the reported exciton and trion peak,\textsuperscript{[45]} and trion binding energy is found to be around 50 meV, which is consistent with the reported value.\textsuperscript{[20,48]} However, the propagating SPP may experience reabsorption by WS\textsubscript{2}, NW, or gold film during the propagation, which leads to the red shift for the component having polarization along the wire. Absorption coefficient is fairly constant above 550 nm for WS\textsubscript{2},\textsuperscript{[49]} so the contribution from the reabsorption of WS\textsubscript{2} is less probable, whereas reabsorption by AgNW or metal film may be a possible reason for the abovementioned red shift.\textsuperscript{[50]}

### 3. Conclusion

In summary, we have experimentally studied the polarization-resolved directional PL emission from the AgNW–WS\textsubscript{2}–Au cavity as a function of angular and spectral signatures. Through detailed experiments and analysis, we have shown the role of each component of the cavity, and how it influences the directional emission characteristics of the WS\textsubscript{2} monolayer. By performing polarization- and momentum-resolved PL spectroscopy from the distal end of the AgNW–WS\textsubscript{2}–Au cavity, we reveal the possibility of studying exciton to trion conversion at room temperature. It is to be noted that the NW-based cavities inherently facilitate waveguiding characteristics, which means the excitation and collection in the geometry are spatially offset. Such separation of excitation and collection of optical locations can be of relevance in 2D material-based nonlinear nanophotonic chips and open-cavity nanolasers. We envisage that the confined electric fields facilitated by the spatially extended cavity can be further harnessed by engineering various parameters, such as the cavity geometry and material properties.

![Figure 4](image-url)

**Figure 4.** Dependence of PL spectral features from the distal end of the AgNW–WS\textsubscript{2}–Au cavity as a function of excitation and collection polarization. a) PL spectrum as a function of excitation polarization along (red curve) and perpendicular (black curve) to the NW axis. b) PL spectrum as a function of collection polarization along (red curve) and perpendicular (black curve) to the NW axis. Apart from the reduction in the intensity, the change in polarization also leads to red shift in the peaks. The PL spectrum collected from the distal end of the NW; red and black curves correspond to the input polarization along the long axis of the NW and transverse to the long axis of the NW, respectively.
as composition, shape, and size of the NW and the underlying gold mirror.

4. Experimental Section

WS$_2$ Synthesis: The WS$_2$ monolayers were grown using atmospheric pressure chemical vapor deposition (APCVD) on 300 nm SiO$_2$-coated silicon wafer following the procedure mentioned in the previous studies.[5,12] The substrates were sonicated in acetone and isopropyl alcohol for 10 min each and blow-dried. They were further cleaned using O$_2$ plasma at 60 W for 5 min. The substrates were loaded on to an alumina boat containing 500 mg of WO$_3$ with the smooth side facing the powder. The boat containing the WO$_3$ was placed inside a quartz tube of 3.5 cm inner diameter in the heating zone of the furnace. Another boat containing 500 mg of sulfur was placed upstream inside the tube, 15 cm away from the WO$_3$ boat. The sulfur boat was placed outside the furnace and was heated separately using a heater coil. Initially, the tube was flushed with 500 standard cubic centimeter per minute (SCCM) of argon for 10 min, and then, the flow was reduced to 30 SCCM and was maintained throughout the experiment. The furnace was ramped up to 850 °C at a rate of 5 °C/min[5]. As the furnace reached 850 °C, the heater coil was heated to 240 °C to evaporate sulfur. These temperatures were maintained for 10 min for growing WS$_2$ monolayer. After the growth, the system was allowed to cool naturally.

Optical Characterization: A chemically synthesized WS$_2$ monolayer was further characterized by PL and Raman spectroscopy. PL and Raman spectra of the same are shown in Figure S1, Supporting Information.

AgNW–WS$_2$–Au Cavity Preparation: AgNWs with an average diameter of 300 nm have been synthesized by a polyol process, as reported in the previous study.[5] For the preparation of the gold mirror, 160 nm of a gold film has been deposited on the glass coverslip using thermal vapor deposition. Monolayer WS$_2$ flakes have been transferred onto the gold mirror. Polystyrene is used as a support film for this transfer as reported here.[14] A 3 nm Al$_2$O$_3$ spacer layer was placed using atomic layer deposition in between gold mirror and WS$_2$ to prevent the charge screening with the aim to avoid PL quenching. To form a cavity, AgNWs were dropcasted on gold mirror with WS$_2$ sandwiched in between NW at which we have performed the experiment is 300 nm thick and ≈13 µm in length. For different set of measurements shown in the Supporting Information, the NWs of length 12–20 µm have been used. A field emission scanning electron microscope image showing the diameter of the wire is given in Figure S7, Supporting Information.

Experimental Setup: One end of the AgNW–WS$_2$–Au cavity has been excited using 532 nm laser with polarization along the cavity. A 100×, 0.95 numerical aperture (NA) objective lens was used in backscattered configuration for both excitation and collection. Signal from the distal end of the NW is collected by spatially filtering the region, and the real/Fourier plane is projected into EMCCD/spectrometer. To transfer the Fourier plane from the back aperture of the objective lens to the EMCCD, 4f configuration is used.[35] High NA excitation ensures efficient excitation of surface plasmon in the NW as well as high electric field in the cavity. Combination of edge and notch filter has been used to efficiently reject the elastically scattered light. It can be seen from the PL spectra that the contribution from the elastic scattering is negligible (Figure S1, Supporting Information). Polarizer and half wave plate are used in input path to control the input polarization. An analyzer is used in output path to analyze the output light for polarization-resolved measurements. See Figure S2, Supporting Information, and our previous reports[56] for detailed experimental setup of Fourier-plane optical microscopy.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

Keywords

2D materials, directional emission, excitons, Fourier-plane imaging, plasmonics, trions, tungsten disulfide

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