Supporting Information for

Gate-Tunable Plasmon-Enhanced Photodetection in a Monolayer MoS_2 Phototransistor with Ultrahigh Photoresponsivity

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Section S1. Gold-mediated exfoliation

Monolayer MoS_2 was prepared by a gold-mediated exfoliation method[1]. This advanced method starts with gold (Au) evaporation (~ 100 nm) on bulk MoS_2. The interaction between Au and S atoms on the topmost layer is much stronger than that between the same layer and the bottom layer, thus enabling peel-off of the topmost layer by using thermal tape. The thermal tape is then released on a hot plate (=120℃) and cleaned by O_2 plasma treatment. The top Au film is etched by potassium iodide and iodine (KI/I_2) wet etching, followed by a 10 min acetone and isopropyl alcohol (IPA) rinse to remove the etchant and the residues.

Section S2. Raman and PL spectra of the monolayer MoS_2

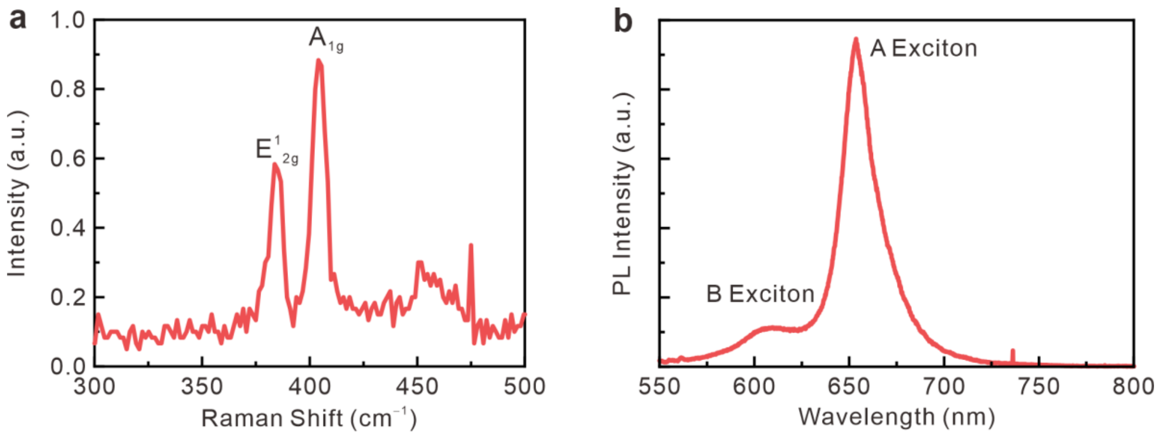

Figure S1. Material properties of monolayer MoS_2 obtained by gold-mediated exfoliation method. (a) Raman spectra and (b) photoluminescence (PL) spectra of monolayer MoS_2.

Section S3. The measured optical constants of noble metal (Au, Ag, Al) films

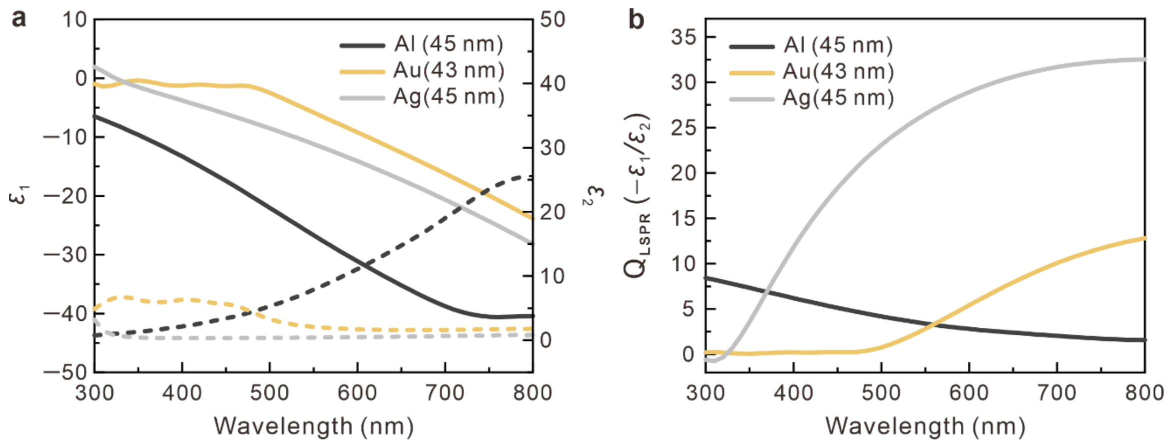

Figure S2. The measured optical constants of noble metal (Au, Ag, Al) films growth by thermal evaporation. The base pressure of the vacuum chamber is approximately 3×10^{-6} torr, and the deposition rate is approximately 1 Å s^{-1}. (a) The optical constants of Al, Au, and Ag were measured by spectroscopic ellipsometry (VASE, J.A. Woollam Co.). The real (solid line) and imaginary (dashed line) parts of the dielectric function are used for the FDTD simulation. (b) Calculated quality factors of the localized surface plasmon resonance in spherical structures (Q_{LSPR}). The results suggest that Ag is the best plasmonic material in the visible.

Section S4. Full-Color palettes with varied size and period of plasmonic Ag nanodisk arrays

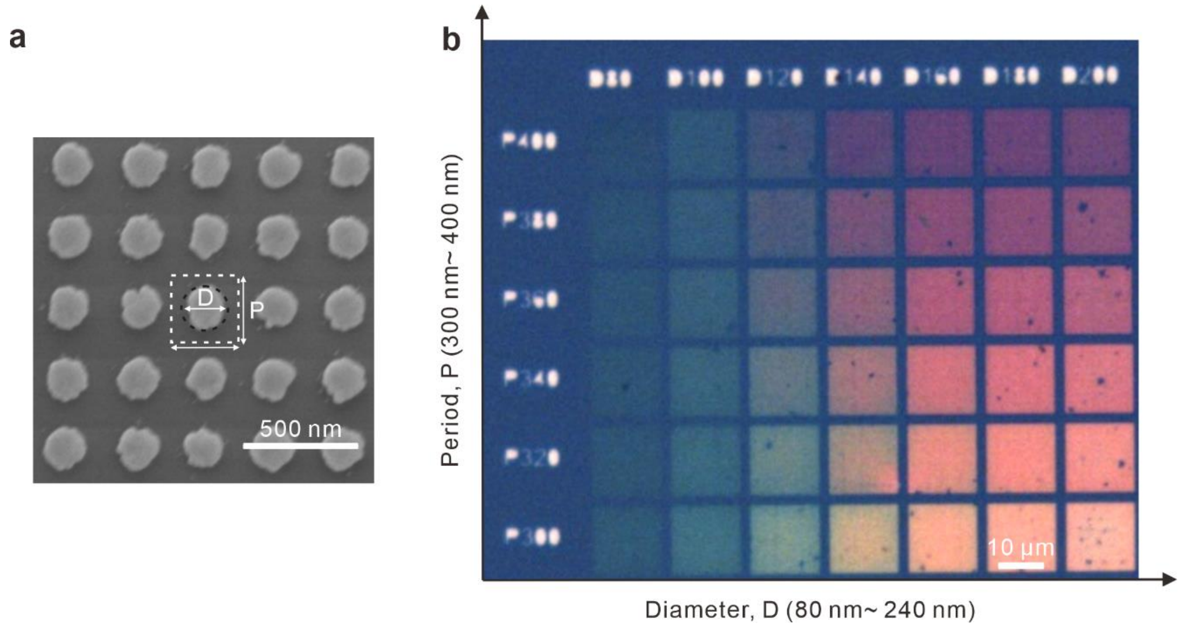

Figure S3. Full-color palettes with varied size and period of plasmonic Ag nanodisk (ND) arrays on SiO_2/Si substrate. (a) SEM images of a fabricated AgND array with a diameter of 140 nm and a periodicity of 260 nm. (b) Optical image of the reflective colors from different AgND arrays with the designed disk size D= 80– 240 nm) and the designed period P= 220– 400 nm. The real size of the fabricated AgNDs is slightly larger than the designed size. The image was taken using a 20× objective lens in microscope reflection mode under a white light illumination.

Section S5. Reflection spectra of different Ag nanodisk arrays with varied disk size and period

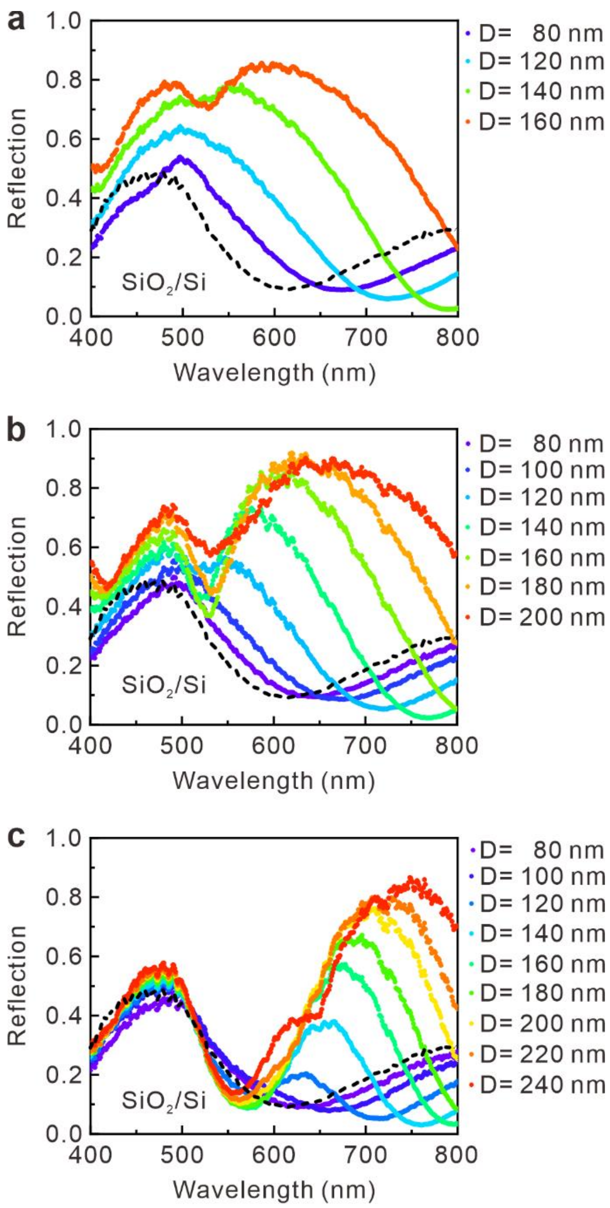

Figure S4. Measured reflection spectra of different Ag nanodisk (AgND) arrays with varied disk size and period on the SiO_2/Si substrate. (a) Experimentally measured reflection spectra from AgND arrays with a varied diameter of the AgND at a fixed period of P= 260 nm. (b) P= 300 nm. (c) P= 400 nm. The black line shows the reflection spectra from the SiO_2/Si substrate. A clear redshift in the resonance peak with increasing diameter of AgND is strongly related to the localized surface plasmon resonance in the AgND arrays.

Section S6. Calculated absorption spectra specifically for the monolayer MoS_2 layer

The simulation results of the monolayer MoS_2 absorbance were done with the commercial Lumerical FDTD software. In the simulation setup, a 300 nm silica layer was located on the silicon substrate. On top of the silica, the monolayer MoS_2 with a thickness of 0.7 nm was then defined. The permittivity of the monolayer MoS_2 was taken from the literature[2]. In the case of plasmonic enhancement, the silver nanodisk (diameter = 160 nm, height = 40 nm) was built on the monolayer MoS_2. To construct the nanodisk arrays, periodic boundary conditions (period = 260 nm) were set in the x and y directions, while the PML boundaries were applied in the z direction, with a planewave source propagating in the same direction. The mesh for each simulation was set to be 1 nm in the x and y directions and 0.2 nm in the z direction. To obtain the absorbance of the monolayer MoS_2, a field monitor and an index monitor were added to cover the entire region of the MoS_2 layer. The absorbance was then calculated by applying equation (1) and integrating through the volume of the MoS_2 monolayer.

$$Absobance = \frac{1}{2} \times \varepsilon_0 \times \omega \times |E|^2 \times \varepsilon_{img} \qquad (1)$$

where ε_0, ω, E, and ε_{img} represent the permittivity of free space, the angular frequency, the normalized electric field (by the source), and the imaginary part of epsilon of the monolayer MoS_2, respectively.

Section S7. Metrics of the reported TMD-based plasmonic photodetectors

Devices	Material	Plasmonic Structure	Responsivity ($A\ W^{-1}$)	Detectivity ($cm\ Hz^{1/2}\ W^{-1}$)	Decay Time (s)	Enhanced Ratio
PhotoFET	FL MoS_2	Au nanoparticles (SHINs)[3]	NA	NA	NA	2-3
	1L MoS_2	Au nanoshells[4]	~0.87 mA W^{-1} @630 nm	NA	NA	1.5-3.5
	FL MoS_2	Au nanoparticles[5]	~ 2-3 mA W^{-1}@632 nm	NA	NA	2
	FL MoS_2	Au nanoarrays[5]	~ 2-3 mA W^{-1} @632 nm	NA	NA	3
	FL MoS_2	Au nanoarrays[6]	20 A W^{-1} @ 466 nm	6×10^{10}	0.58 s	8.2
	1L MoS_2	**Ag nanoarrays (This work)**	**~27000 A/W @ 620 nm**	$\mathbf{1.3 \times 10^{12}}$	**0.4 s**	**7.2**
M-S-M PD	1L MoS_2	Ag nanowires[7]	59.6 A/W @532 nm	4.51×10^{10}	> 10 s	250
	1L MoS_2	Ag nanoparticles (SHINs) [8]	287.5 A/W @570 nm	NA	15 s	8.8
	1L MoS_2	Ag nanocubes[9]	7940 A/W @520 nm	NA	1.6 s	38
	FL MoS_2	Au nanoparticles[10]	64 mA/W @980 nm	NA	2.6 ms	14

Table S1. Device performance of the reported plasmonic-enhanced monolayer or multi-layer MoS_2 photodetectors.

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