Supporting Information

Enhancing Quantum Yield in Strained MoS$_2$ Bilayers by Morphology-controlled Plasmonic Nanostructures towards Superior Photodetectors

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a) CVR-grown bilayer MoS$_2$ and chemically synthesized Au nanostructures:

Figure S1a shows the Raman spectrum of bilayer MoS$_2$ with characteristic peaks of the in-plane Raman mode ($E^{1}_{2g}$) and the out-of-plane Raman mode ($A_{1g}$) at the frequencies of 384.2 cm$^{-1}$ and 406.4 cm$^{-1}$, respectively with 22.2 cm$^{-1}$ distance between two modes confirming the growth of bilayer MoS$_2$. Figure S1b presents the UV-visible absorption spectrum of Au nanoparticles with various morphologies overlapping with the incident excitation wavelength. We tailored the size of the nanoparticles to employ the spectral overlap between localized surface plasmon resonances (LSPRs) of Au nanoparticles and the incident excitation wavelength (520 nm).

![Figure S1](image_url)

**Figure S1.** a) Raman spectrum of bilayer MoS$_2$ on a sapphire substrate; b) UV-vis absorption spectra of various nanostructures (the dashed vertical line corresponds to the laser excitation wavelength).
b) Size distribution histograms of Au nanostructures:

The size distribution maps of Au nanoparticles of various shapes are shown in Figure S2. The average size of nanocubes ~ 83 nm; octahedra ~ 74 nm; Rhombic dodecahedra ~ 64 nm; Nanorods ~ length - 65 nm; width - 23 nm. The histogram plot demonstrates the uniform sizes of chemically synthesized Au nanoparticles.

Figure S2. Size distribution histograms of Au nanostructures. (a) Nanocubes; (b) Octahedra; (c) Rhombic dodecahedra; (d) Nanorods.
c) XY-field distributions of bilayer MoS$_2$ interfaces with various Au nanostructures:

Au nanocube being symmetric structures, the longitudinal and transverse mode of Au nanocube substantially degenerate, resulting in a ring-like plasmonic mode. While the nanorod with high-aspect-ratio, breaks this degeneracy and creates longitudinal & transverse mode. For complex nanostructures such as OD and RD electric field are concentrated at the vertices. Au sphere has small interface area with MoS$_2$ resulting in limited e-field confined region.

![Figure S3](image-url)

**Figure S3.** FDTD-simulated XY-field distributions of bilayer MoS$_2$ interfaces with various Au nanostructures.
d) Prolonged time-resolved photoresponse performance of the hybrid device:

we performed time-resolved evolution of our bilayer MoS$_2$, which was integrated with the plasmonic structure with laser on/off modes for a prolonged time for $V_{ds} = 10$ V and $P = 318.471$ mW/cm$^2$, as shown in Figure S4. We observe a gentle increase in the photocurrent under laser illumination and a sharp drop due to pacific relaxation when the laser is switched off. The rising time of our device is estimated to be 0.26 s and the decay time is 0.45 s. The rising and decay times of our device show significant improvement compared to the Pt-strip-enabled MoS$_2$ bilayer$^2$.

\[ \text{Figure S4: Prolonged time-resolved photoresponse performance of the hybrid MoS2-Au nanocube device at } V_{ds} = 10 \text{ V; laser power density } = 318.471 \text{ mW/cm}^2. \]
e) The photoresponsivity measurement of hybrid MoS$_2$ bilayer with other Au-OD & RD:

The photoresponsivity measurement of hybrid MoS$_2$ bilayer with other Au nanostructures such as Au-OD and RD has been carried out, and the results are displayed in Figures S5. The plot of photocurrent versus time of bilayer MoS$_2$ that is decorated with Au OD & RD at various bias voltages ($V_{ds} = 1, 3, 5, 7, 10$ V) demonstrates the reproducibility and consistent behavior of our hybrid as-grown bilayer MoS$_2$ that is decorated with Au nanostructures.

**Figure S5:** Time-dependent photocurrent measurements of bilayer MoS$_2$ that is decorated with a) Au OD; b) RD nanostructures and studied at various bias voltages ($V_{ds} = 1, 3, 5, 7, 10$ V) with a constant laser power density of 318.471 mW/cm$^2$. 
f) **Photodetection of MoS$_2$ hybridized with Au nanorod:**

The plots of the photocurrent versus the illumination power ($V_{ds}$ = 3 V) for MoS$_2$ that is hybridized with Au nanorods and nanocubes are shown in Figure S6a. In addition, the photoresponsivity measurements of hybrid MoS$_2$ bilayer that is decorated with Au nanorods and excited at 520 nm and 634 nm with various bias voltages ($V_{ds}$ = 1, 3, 5, 7, 10 V) are shown in Figures S6 b & c, which again demonstrate the reproducibility of our device.

![Figure S6: a) Plot of photocurrent versus laser power intensity at $V_{ds}$ = 3 V for bilayer MoS$_2$ that is decorated with Au nanocubes and nanorods; b) and c) Time-dependent photocurrent measurements of bilayer MoS$_2$ that is decorated with Au nanorods and studied at various bias voltages ($V_{ds}$ = 1, 3, 5, 7, and 10 V) with illumination by a 520 nm and 634 nm laser with a power density of 318.471 mW/cm$^2$.](image)

![Figure S7: a) Plot of photocurrent versus laser power intensity at $V_{ds}$ = 3 V for bilayer MoS$_2$ that is decorated with Au nanocubes and nanorods; b) and c) Time-dependent photocurrent measurements of bilayer MoS$_2$ that is decorated with Au nanorods and studied at various bias voltages ($V_{ds}$ = 1, 3, 5, 7, and 10 V) with illumination by a 520 nm and 634 nm laser with a power density of 318.471 mW/cm$^2$.](image)

**Figure S6:** a) Plot of photocurrent versus laser power intensity at $V_{ds}$ = 3 V for bilayer MoS$_2$ that is decorated with Au nanocubes and nanorods; b) and c) Time-dependent photocurrent measurements of bilayer MoS$_2$ that is decorated with Au nanorods and studied at various bias voltages ($V_{ds}$ = 1, 3, 5, 7, and 10 V) with illumination by a 520 nm and 634 nm laser with a power density of 318.471 mW/cm$^2$.

![Figure S7: a) Plot of photocurrent versus laser power intensity at $V_{ds}$ = 3 V for bilayer MoS$_2$ that is decorated with Au nanocubes and nanorods; b) and c) Time-dependent photocurrent measurements of bilayer MoS$_2$ that is decorated with Au nanorods and studied at various bias voltages ($V_{ds}$ = 1, 3, 5, 7, and 10 V) with illumination by a 520 nm and 634 nm laser with a power density of 318.471 mW/cm$^2$.](image)

**g) Band structure modification and charge transfer of MoS$_2$ by hybridizing with Au.**

Figure S7a shows the Fermi level shift of MoS$_2$ hybridized with Au toward the conduction band with a new isolated energy level that is formed in the bandgap, thereby reducing the interfacial Schottky barrier between Au and MoS$_2$\(^3\). In order to gain more insight into the exciton and plasmon coupling, time-resolved photoluminescence (TR-PL) measurements have been carried out for MoS$_2$ and MoS$_2$ decorated with Au nanoparticles as shown in Figure S7b. The PL decay time of as-grown MoS$_2$ is
4.483 ns, but it shortens to 3.183 ns when MoS$_2$ decorated with Au nanoparticles. This shorten decay time results from the charge transfer between Au and MoS$_2$\textsuperscript{4}.

**Figure S7:** a) Partial density of states (PDOS) of MoS$_2$ and Au-MoS$_2$ hybrid by first-principle calculations; b) Time resolved photoluminescence spectra of MoS$_2$ and Au-MoS$_2$. 

\[ \text{Lifetime:} \quad \text{MoS}_2 = 4.483 \pm 0.5 \quad \text{Au+MoS}_2 = 3.183 \pm 0.4 \]
h) X-ray photoelectron spectroscopy (XPS) study for MoS$_2$ hybridized with Au.

In order to understand hybridization nature of bilayer MoS$_2$ with Au plasmonic nanostructures, we performed the X-ray photoelectron spectroscopy (XPS) as discussed in Figure S8. The binding energies of Mo 3d and S 2p are shifted by ca. 0.23 and 0.7 eV compared to pristine bilayer MoS$_2$.

Whereas binding energies for Au 4f$_{7/2}$ and Au 4f$_{5/2}$ are 83.47 eV and 89.8 eV, respectively. This shows that there is no chemical interaction between pristine MoS$_2$ and Au/ MoS$_2$. The peak shift confirms the synergetic effect and charge transfer between MoS$_2$ and Au.

<table>
<thead>
<tr>
<th>Plasmonic integrated MoS$_2$</th>
<th>Thickness/ No. of MoS$_2$ layers</th>
<th>FET</th>
<th>Wavelength (nm)</th>
<th>Photocurrent</th>
<th>Responsivity</th>
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<tbody>
<tr>
<td>Exfoliated MoS$_2$ with Au electrode$^5$</td>
<td>7nm-10 nm</td>
<td>Yes</td>
<td>500-1550</td>
<td>~ 60 nA @ 532 nm</td>
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<td>Exfoliated MoS$_2$- Au resonating and non-resonating nanostructures$^6$</td>
<td>Bilayer</td>
<td>-</td>
<td>532, 1070</td>
<td>~ 5 pA @ 1080 nm</td>
<td>5.2 A W$^{-1}$ @ 1070 nm</td>
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<td>1.1$\times 10^5$ A W$^{-1}$ @ 532 nm</td>
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<tr>
<td>Exfoliated MoS$_2$- Au nanostructures$^7$</td>
<td>4.5nm</td>
<td>Yes</td>
<td>477-720</td>
<td>4.6 µA @ 532 nm</td>
<td>-</td>
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<tr>
<td>Exfoliated MoS$_2$- Au nanospheres$^8$</td>
<td>4-5 layers</td>
<td>Yes</td>
<td>400-800</td>
<td>3 µA @ 514 nm</td>
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<tr>
<td>CVD grown MoS$_2$- Au core-shell</td>
<td>Monolayer</td>
<td>Yes</td>
<td>600-800</td>
<td>2.5 fold increase in photocurrent</td>
<td>~ 0.9 nA µW$^{-1}$ @ 630 nm</td>
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Figure S8: a) XPS spectra of MoS$_2$ and Au–MoS$_2$ in the (a) Mo 3d; (b) S 2p; and (c) Au 4f region
Table 1 Performance metrics of plasmon enhanced MoS$_2$ photodetectors

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<tr>
<th>Method</th>
<th>Type</th>
<th>Yes/No</th>
<th>$\lambda$</th>
<th>Current @ $\lambda$</th>
<th>Power at $\lambda$</th>
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<tr>
<td>Exfoliated MoS$_2$-Pt nanostrps$^9$</td>
<td>Bilayer</td>
<td>Yes</td>
<td>325, 532</td>
<td>~30 nA @ 325 nm</td>
<td>14 A W$^{-1}$ @ 325 nm</td>
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<tr>
<td>CVD grown MoS$_2$-Ag nanowire network$^{10}$</td>
<td>Monolayer</td>
<td>-</td>
<td>532</td>
<td>~0.8 µA @ 532 nm</td>
<td>0.15 A W$^{-1}$ @ 532 nm</td>
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<tr>
<td>CVR grown MoS$_2$-Different Au nanostructures (Cube, OD, RD, Sphere, Rod)- <em>Our work</em></td>
<td>Bilayer</td>
<td>-</td>
<td>520, 634</td>
<td>MoS$_2$ ~ 0.1 nA @ 520 nm</td>
<td>MoS$_2$-cube Au NPs - 790 µA W$^{-1}$ @ 532 nm; 0.159 mW cm$^{-2}$</td>
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<td>MoS$_2$/Au Cube ~ 1.4 nA @ 520 nm</td>
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<td>MoS$_2$/Au OD ~ 1 nA @ 520 nm</td>
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<td>MoS$_2$/Au RD ~ 0.8 nA @ 520 nm</td>
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<td>MoS$_2$/Au Sphere ~ 0.4 nA @ 520 nm</td>
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<td>MoS$_2$/Au Rod ~ 1.7 nA @ 520 nm</td>
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<td>@ 318.471 mW cm$^{-2}$</td>
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References:


