

Supplementary Information

Photothermal antenna effects derived from one-to-one coupling nanohybrids of Au plasmonics and MoS₂ semiconductors

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Materials. Molybdenum disulfide (MoS₂ crystalline powder, < 2 μm, 99%) was obtained from Sigma-Aldrich Co. (USA). Sodium cholate (98%), chloroauric acid hydrated (HAuCl₄·4H₂O) were obtained from Aladdin Chemistry Co. Ltd. (Shanghai, China). All oligonucleotide sequences were ordered from Shanghai Sangon Biotech. Co. (Shanghai, China). Bovine serum albumin (BSA) and adenosine 5'-triphosphate (ATP) were purchased from BBI Life Science (Shanghai, China). Other chemicals were of analytical reagent grade and used without further purification. Ultrapure water (18.2 MΩ cm) was utilized throughout the whole experiment.

Experimental instrumentations. A KQ-400B ultrasonic bath (400 W, Kunshan Ultrasonic Instruments Co., Ltd., China) was adopted for the liquid-phase exfoliation of MoS₂ crystals. The optical absorption spectra were recorded on a UV-2450 UV-vis spectrophotometer (Shimadzu, Japan). X-ray photoelectron spectra (XPS) results were analyzed using an ESCALAB 250Xi X-ray photoelectron spectroscope (Thermo Electron, USA). Centrifugation treatments were conducted using a TGL-16M high-

speed refrigerated centrifuge (Xiangyi, China). Morphological characterization of nanostructures and energy dispersive X-ray spectroscopy (EDS) element mapping were conducted using a TecnaiG2 T20 UTWIN (FEI, USA) transmission electron microscope (TEM). Fluorescence measurements were conducted on F-7100 fluorescence spectrometer (Hitachi, Japan). Inductively coupled plasma-atomic emission spectra (ICP-AES) were employed for the quantitative analysis of Mo species concentrations (Agilent 7800ce, USA).

Liquid-exfoliated strategies. Briefly, a portion of 100.0 mL mixed aqueous dispersion, containing 5.0 mg/mL MoS₂ powders and 1.5 mg/mL sodium cholate, was treated with sonication (25 °C, 20 h). The resultant dispersion was centrifuged (3000 rpm, 30 min) and yellow-green supernatant was collected to remove un-exfoliated bulk crystals. In order to achieve size fractionation, the separated supernatant was centrifuged at 10000 rpm for further 30 min, and subsequently, the sediments of liquid-phase exfoliated MoS₂ samples (LE-MoS₂) and the supernatant were isolated.

Reactions between the LE-MoS₂ and chloroauric acid. Firstly, a portion of 9.0 mL chloroauric acid (0.5 mM) solution was pre-heated in a water bath (70 °C) and kept stirred (300 rpm) for 30 min. Subsequently, a portion of the MoS₂ or BSA/MoS₂ mixing aqueous dispersion (1.0 mL, BSA: 2 mg/mL; MoS₂: 200 µg/mL) was rapidly added to the treated chloroauric acid solution, whereas the reaction occurred immediately and terminated after 30 min. The resultant solution was taken out from the water bath for naturally cooling. As-obtained colorful dispersions were treated with centrifugation (10 000 rpm, 20 min), and sediments were further re-dispersed in a certain volume of ultrapure water for collecting the purified hybrids. Lastly, the products of BSA-covered Au/MoS₂ nanoarchitectures stocking solutions were stored at 4 °C for further experiments.

The calculation of Au and MoS₂ composition in hybrids. The products of Au/MoS₂ nanostructures were collected for ICP-AES detections. The ratios of MoS₂ were calculated through the concentrations of Mo species.

Preparation of gold nanoparticles. Gold nanoparticles (AuNPs) were prepared based on the well-known Frens method. Herein, citrate molecules were used as both reductants and stabilizers. Briefly, aqueous solutions of 50 mL of 0.5 mM HAuCl₄ were heated to 95 °C with vigorous magnetic stirring. Subsequently, the 1% (w/v) sodium citrate aqueous solutions (1.50 mL) were added all at once to generate AuNPs with average diameters of 21 nm. The yellow solutions of HAuCl₄ turned clear, dark blue and finally red, indicating the formation of AuNPs. After reaction for 15 min, the heated process was terminated and the resulting solutions were naturally cooled to room temperature. The final production was stored at 4 °C for further experiments.

Preparation of BSA-covered MoS₂ and AuNPs. The aqueous dispersions of nanomaterials (LE-MoS₂ and AuNPs) were incubated with 2 mg/mL BSA for 2 hours. Subsequently, the samples were centrifuged at 10000 rpm for 20 min, and the sediments were collected. The sediments were then re-dispersed into the ultrapure water under the treatment of sonication for the preparation of BSA-MoS₂ and BSA-AuNPs aqueous dispersions.

IR treatments and fluorescence measurements. The DNA-cy3 was added in OtO-Au/MoS₂ dispersion, and the fluorescence intensities (F_0) were detected at the excitation wavelength of 550 nm. Subsequently, the mixture was treatment with IR (808 nm, 1.0 W/cm²) for 10 min, and the temperature of solution was increased. After the solution was naturally cooled to room temperature, the fluorescence (F) was re-measured. The quenching ratio is estimated via an equation: $(F_0-F)/F_0$. In order to calculate sensing performance of the quenching system, ATP is added in the mixture solution and the incubation was conducted for 30 min. After that, the fluorescence of

system in the presence of ATP was detected.

Table S1. The DNA name corresponding to the sequence in the content.

Name	Sequence
Poly(T)-Cy3	5'-TTTTT TTTTT TTTTT TTTTT-3' -Cy3
Poly(A) -Cy3	5'-AAAAA AAAAA AAAAA AAAAA-3' -Cy3
Poly(C) -Cy3	5'-CCCCC CCCCC CCCCC CCCCC-3' -Cy3
Poly(G) -Cy3	5'-GGGGG GGGGG GGGGG GGGGG-3' -Cy3
ATP Aptamer-Cy3	5'-ACCTG GGGGA GTATT GCGGA GGAAG GT-3'-Cy3
cDNA	5'-ACCTT CCTCC GCAAT ACTCC CCCAG GT-3'

Figure S1.

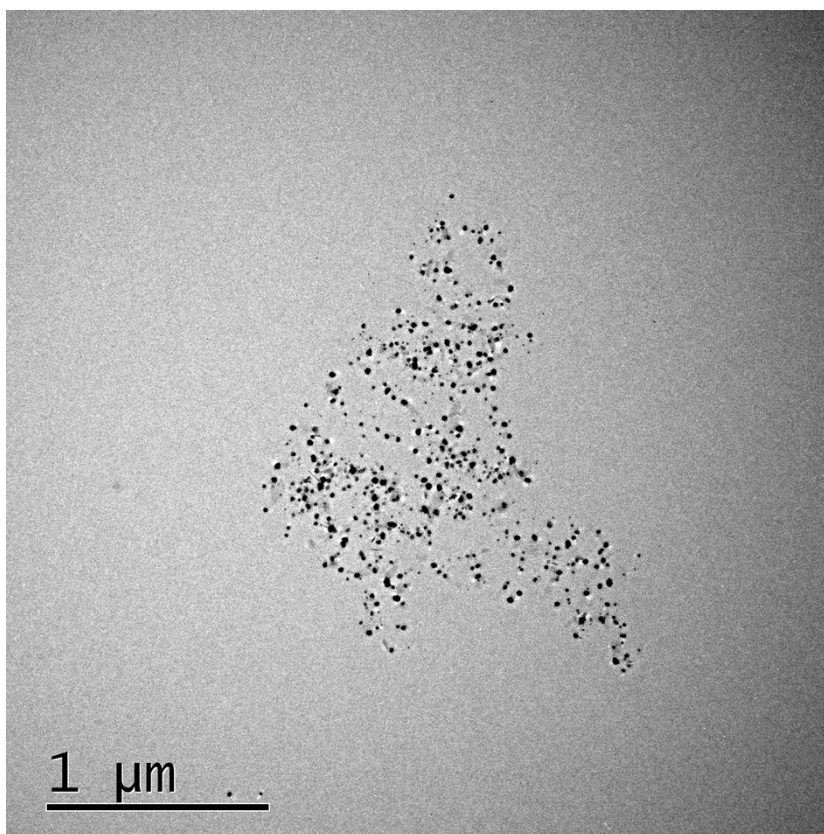


Fig. S1. The TEM image of as-prepared Au/MoS₂ nanostructures indicating the consistent distribution of Au and MoS₂ structures.

Figure S2.

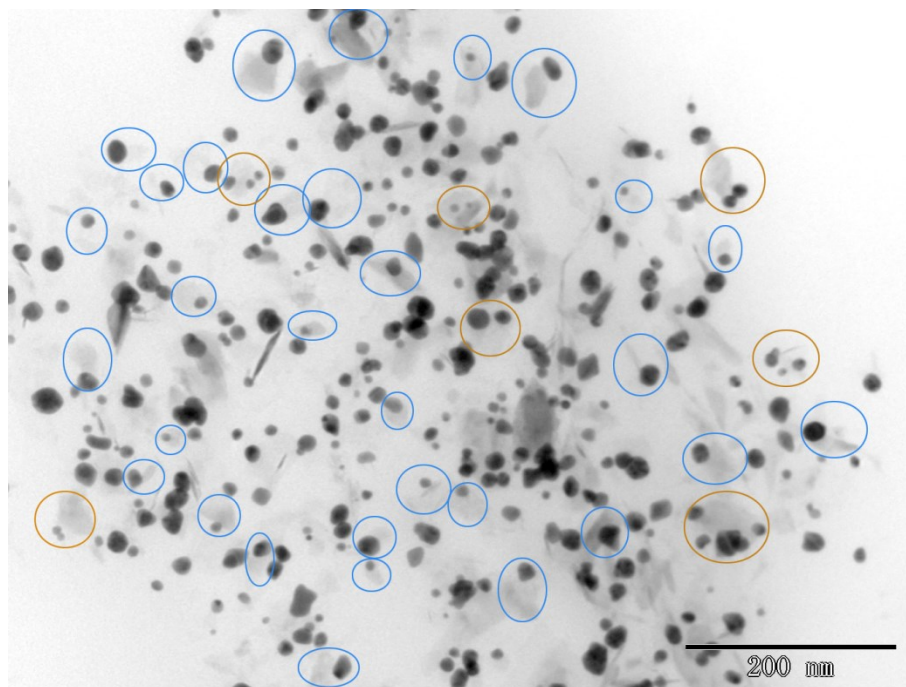


Fig. 2. The TEM image of as-prepared Au/MoS₂ nanostructures indicating the one-to-one (blue circle) and multiple-to-one (yellow circle) coupling formats.

Figure S3

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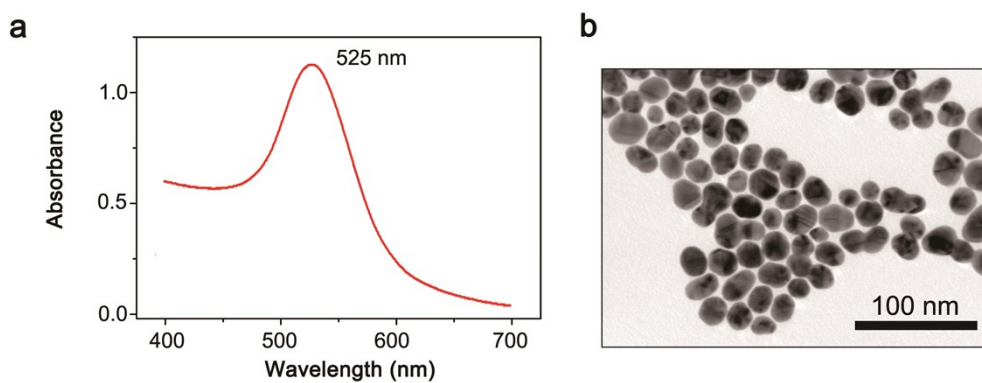


Fig. S3. The Absorption spectrum (a) and TEM image (b) of as-prepared AuNPs with the average size of 21 nm.