

## Supporting Information

### Improved CdS Photocatalytic H<sub>2</sub> Evolution using Au-Ag Nanoparticles with Tunable Plasmon-Enhanced Resonance Energy Transfer

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## EXPERIMENTAL SECTION

**2.1 Chemicals.** Silver nitrate ( $\text{AgNO}_3$ , 99.99%), Cadmiumnitrate hexahydrate ( $\text{Cd}(\text{NO}_3)_2$  99.99%), sodium borohydride ( $\text{NaBH}_4$ , 98%) were purchased from Sigma-Aldrich. Sodium citrate ( $\text{C}_6\text{H}_5\text{Na}_3\text{O}_7$ , 98%), hydrogen tetrachloroaurate hydrate ( $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ ), ammonia (AR) and thiourea were purchased from Shanghai Aladdin Reagent. All chemicals were used as received without further treatment.

### 2.2 Synthesis of Au-Ag HNPs

The synthesis of Au-Ag HNPs is referred to previous work<sup>[1]</sup>. Briefly, 45nm silver nanoparticles were prepared by a two-step growth method. To prepare Au-Ag hollow nanoparticles (HNPs), add a certain amount of  $\text{HAuCl}_4$ (0.01M) to the water, and quickly inject sodium citrate after boiling. Subsequently, the synthesized Ag NPs are taken and injected into the reaction solution. The reaction solution was quickly taken out under different reaction times, and subjected to an ice bath to terminate the reaction. The resulting Au-Ag HNPs were centrifuged at 10000 rpm for 10 min, then disperse into 2mL of solution. Different reaction time can obtain Au-Ag HNPs with different plasmon absorption peak positions.

### 2.3 Synthesis of Au NPs and Au@Ag<sub>487</sub> core-shell NPs

Au colloids were prepared by a sodium citrate reduction method. Typically, an aqueous solution of  $\text{HAuCl}_4$  (0.25 mM, 40mL) was heated to boiling, followed by the rapid addition of sodium citrate solution (0.5 M, 200  $\mu\text{L}$ ). The solution was kept boiling for another 15 min, producing a stable and deep-red dispersion of Au nanoparticles. The citrate-protected Au-colloids suspension (denoted as Au-Cit) was then cooled to room temperature for next use. (Au@Ag) core-shell NPs were obtained by suspension of Au NPs heated to reflux and then silver nitrate (99.99%, sigma) and trisodium citrate were simultaneously added. After refluxing for 35 min, (Au@Ag) NPs were obtained. The position of the LSPR peak can be adjusted to 487 nm by controlling the amount of  $\text{AgNO}_3$  added.

### 2.4 Synthesis of Au-Ag HNPs@CdS<sub>x</sub> photocatalyst

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For prepared Au-Ag HNPs@CdS<sub>90</sub> composites, 2mL of Au-Ag HNPs solution in 50 mL of H<sub>2</sub>O. Then 3.6 mL of 0.1 M sodium citrate solution and 6 mL of 0.05 M cadmium nitrate solution were then added and stirred for 10 min, after which 1.6 mL of 28% ammonia was added into the suspension followed by the addition of 2.8 mL of 0.2 M thiourea solution. The solution was stored at 60 °C for 3 h. The final product was washed three times by centrifugation at 8000 rpm and dried under vacuum at 60°C for 12h. Au-Ag HNPs@CdS<sub>110</sub> and Au-Ag HNPs@CdS<sub>70</sub> were obtained by keeping the addition amount of Au-Ag constant, and enlarging the other composition by 1.5 times and reducing by 1 times respectively.

For prepared Au-Ag<sub>x</sub>@CdS<sub>90</sub>, Au@CdS<sub>90</sub>, (Au@Ag<sub>487</sub>)@CdS<sub>90</sub> composites, repeat the above experimental steps using Au-Ag<sub>x</sub>, Au and Au@Ag<sub>487</sub> as templates

## 2.5 Sample characterization

Scanning electron microscope (SEM, Hitachi SU8010), analysis of sample morphology. Transmission electron microscopy (TEM), high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and TEM energy-dispersive X-ray spectra (TEM-EDX) images were recorded on Hitachi HT-7700. Surface compositions were measured by X-ray photoelectron spectroscopy (XPS) using AMICUS ESCA3200. The concentration of the solution is measured by the absorption of the absorbance using UV-vis absorption (MAPADA UV-3200 spectrophotometer). PL spectra was performed on an FT300 (PicoQuant).

The photocatalytic activities of these photocatalysts were used Perfect light, Beijing Lab Solar IIIAG system. Equipped with 300W Xe lamp and  $\lambda > 420$  nm cut-off filter. Briefly, 1 mg of catalyst was added into 50 mL solution containing 0.35M Na<sub>2</sub>S<sub>9</sub> H<sub>2</sub>O and 0.25M Na<sub>2</sub>SO<sub>3</sub>. The mixture solution was stirred under a vacuum for 30 min in the dark. Hydrogen evolution was measured every half an hour using a gas chromatograph (GC7900, Tian Mei, Shanghai, TCD, nitrogen as carrier gas).

Photoelectrochemical analyses were performed using (CHI760E Shanghai Chenhua) a standard three-electrode system. A Pt sheet was used as the counter electrode and a saturated Ag/AgCl (0.1976 V versus RHE) was employed as the reference electrode. To prepare the work electrode,

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1 mg of photocatalyst was uniformly dispersed in a solution containing deionized water (375  $\mu\text{L}$ ), ethanol (125  $\mu\text{L}$ ), and Nafion (30  $\mu\text{L}$ ), and ultrasonicated for 20 min. The resulting colloid was dropped onto a FTO substrate with an area of 1  $\text{cm}^2$ , and then dried at 60°C for 1 h. The photocurrent density-time (i-t) were collected in the dark or under a 300 W Xe lamp (Institute for Electric Light Sources, Beijing). The electrolyte was 0.5 M  $\text{Na}_2\text{SO}_4$  solution.

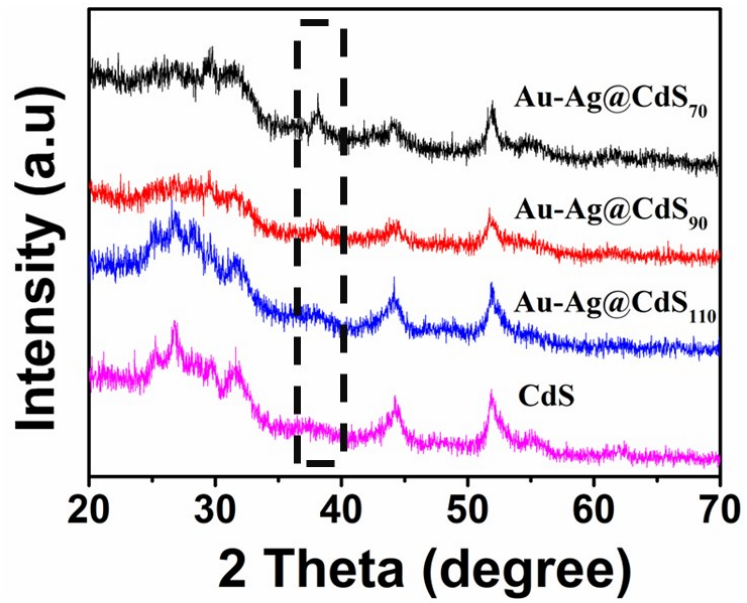


Figure S1. XRD patterns of CdS and Au-Ag@CdS<sub>x</sub> composites.

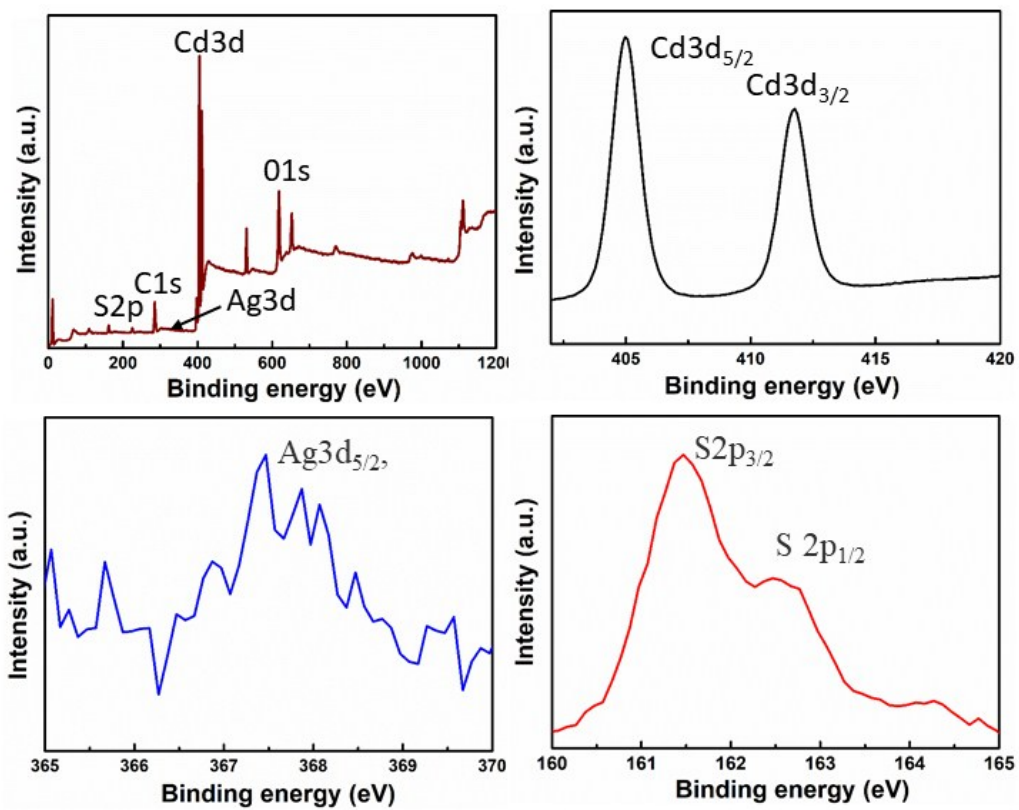


Figure S2. Full (a) and individual XPS spectra of Au-Ag<sub>487</sub>@CdS<sub>0.2</sub>.

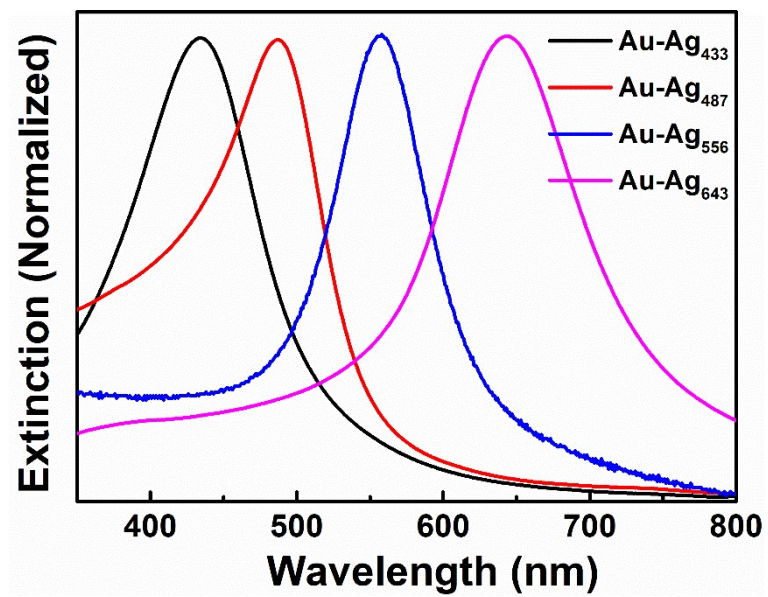


Fig S3. UV-Vis absorption spectra of Au-Ag<sub>433</sub>, Au-Ag<sub>487</sub>, Au-Ag<sub>556</sub>, Au-Ag<sub>643</sub>.

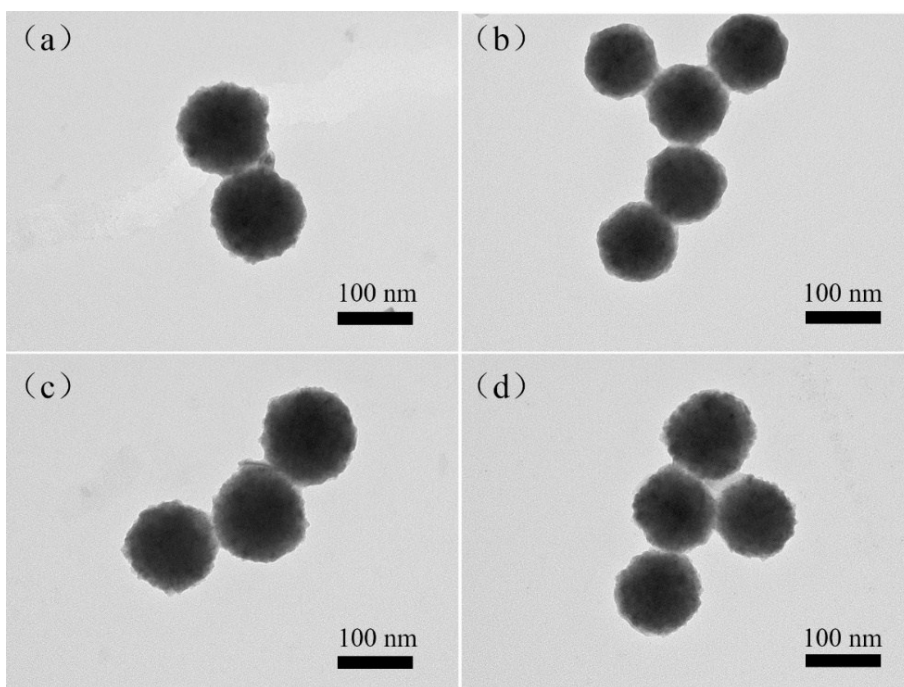


Figure S4. TEM images of Au-Ag<sub>643</sub>@CdS<sub>90</sub>, Au-Ag<sub>556</sub>@CdS<sub>90</sub>, Au-Ag<sub>487</sub>@CdS<sub>90</sub>, Au-Ag<sub>433</sub>@CdS<sub>90</sub>.



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[1] X. Yue, J. Hou, H. Zhao, P. Wu, Y. Guo, Q. Shi, L. Chen, S. Peng, Z. Liu, G. Cao *Journal of Energy Chemistry*. **2020**, 49, 1-7.