Synthesis of Au@Ag Core-Shell Nanocubes with Finely Tuned Shell Thicknesses for Surface-Enhanced Raman Spectroscopic Detection

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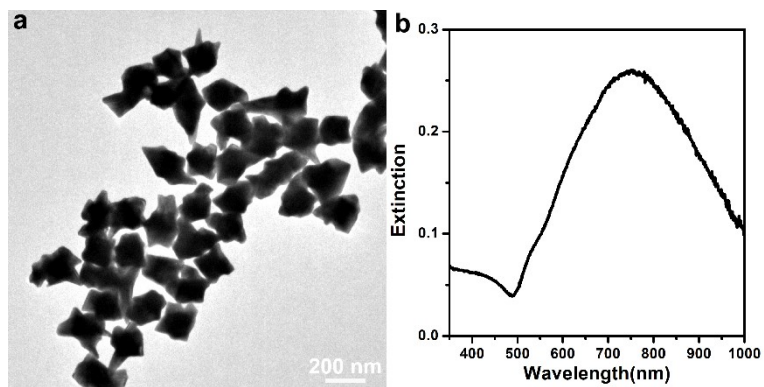
Figure S1. TEM images (a) and UV-vis spectra (b) of the Au nanoparticles produced at the seed volume of 1 μL. The concentrations of CTAB, HAuCl₄, and AA in the reaction mixture are 15 mM, 0.19 mM, and 8.83 mM, respectively.
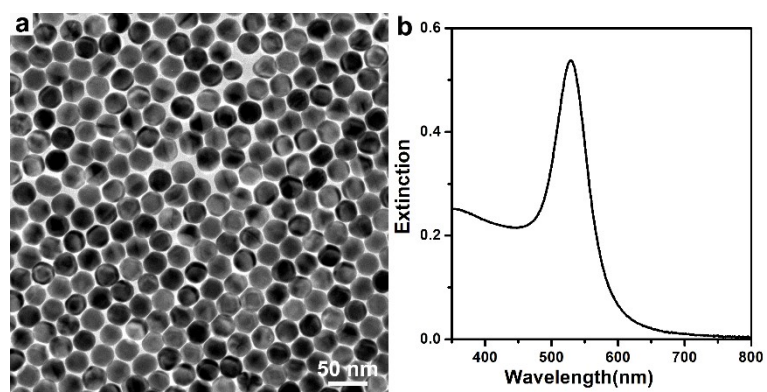
Figure S2. TEM images (a) and UV-vis spectra (b) of the Au nanoparticles produced at the seed volume of 150 μL. The concentrations of CTAB, HAuCl$_4$, and AA in the reaction mixture are 15 mM, 0.19 mM, and 8.83 mM, respectively.
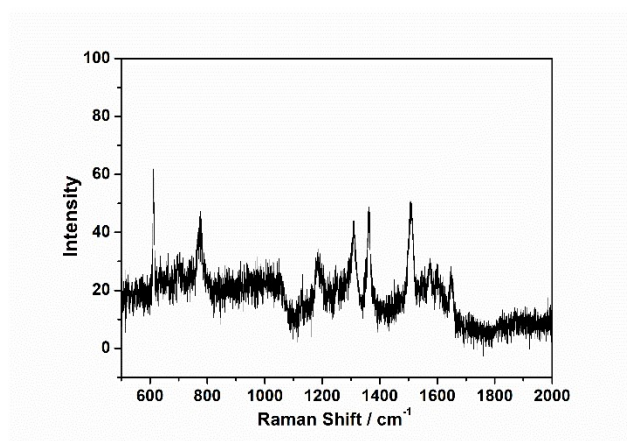
Figure S3. The Raman spectra of 10^{-2} M R6G molecules on the blank glass substrate.

The SERS enhancement factor (EF) can be calculated by the equation[1,2]: EF = (I_{SERS} × N_{Raman}) / (I_{Raman} × N_{SERS}), where I_{SERS} and I_{Raman} represent the peak intensities in the SERS spectrum with 1 × 10^{-6} M R6G and the normal Raman spectrum with a 1 × 10^{-2} M R6G solution, respectively. N_{Raman} is the total number of probe molecules in the excitation volume for the normal Raman measurements, which can be calculated from the laser spot size, the depth of penetration of the focused beam, the density of R6G, and its molecular mass. N_{SERS} is the total number of adsorbed molecules on the NPs in the SERS effective area and can be calculated by the following equation: $N_{SERS} = N_d A_{laser} A_N \rho_b$. N_d is the number density of the nanoparticles, A_{laser} is the area of the laser irradiated on the substrate, A_N is the projected area of the nanoparticles, and ρ_b is the binding density of the R6G molecules. Figure S3 show the Raman spectra of 10^{-2} M R6G from the as-prepared substrates. In this experiment, we select the intensity of the predominant peak of R6G at 613 cm^{-1} for the EF calculations.
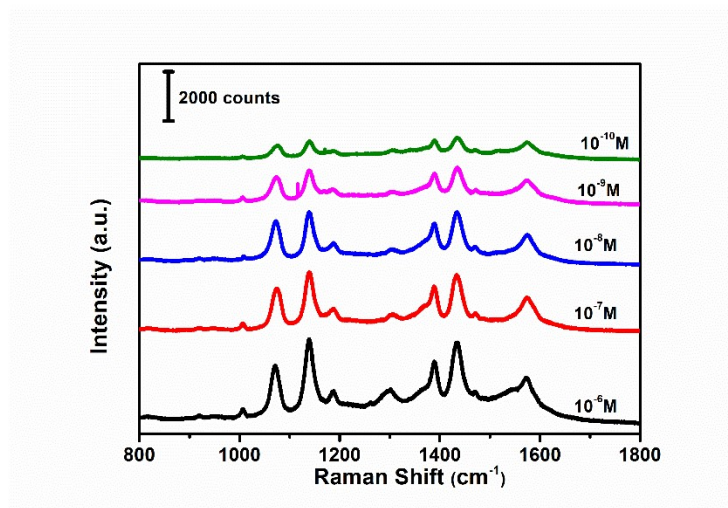
Figure S4. SERS spectra of 4-ATP molecules at different concentrations (from 10^{-6} to 10^{-10} M) on Au@Ag$_{8.4}$ nanocubes substrates. The excitation laser wavelength for Raman measurements is 633 nm, the laser power is 0.5 mW, and the acquisition time is 2 s.

Table S1. Details of the size of Au nanoparticles obtained at different amount of seeds and corresponding LSPR band positions.

Sample	CTAB (mM)	HAuCl$_4$ (mM)	AA (mM)	seed (uL)	size (nm)	LSPR band position (nm)
Figure 1a	15	0.19	8.8	3	102 ± 7	625
Figure 1b	15	0.19	8.8	5	90 ± 5	598
Figure 1c	15	0.19	8.8	10	55 ± 3	549
Figure 1d	15	0.19	8.8	30	42 ± 3	537
Figure 1e	15	0.19	8.8	50	38 ± 3	530
Figure 1f	15	0.19	8.8	80	35 ± 3	529

Table S2. Details of the Ag shell thicknesses of Au@Ag core-shell nanocubes obtained under different AgNO$_3$ concentrations and corresponding LSPR band position of Au cores and Ag shells.

Sample	AgNO$_3$ (uM)	Ag shell thicknesses (nm)	LSPR band position of Au cores (nm)	LSPR band position of Ag shells (nm)
Figure 2a	15	3.6	532	408
Figure 2b	22	5.0	523	410
Figure 2c	29	6.2	519	412
Figure 2d	36	7.3	513	416
Figure 2e	44	8.4	509	417
Figure 2f	51	10.0	508	419

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