Supporting information for

Bimetallic Core-Shell Nanostars with Tunable Surface Plasmon Resonance for Surface-Enhanced Raman Scattering

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Calculation of enhancement factor

Herein all the SERS spectra were measured on the nanostars assembled on a Si substrate. Each measurement included numerous NPs within the laser spot. Assume all the BPE molecules adsorbed on the nanostars contributed to the overall SERS signal intensity. Therefore the average SERS enhancement factor (EF) from collective nanostars can be estimated by:

$$\mathrm{EF}=\frac{I_{\mathrm{SERS}} / N_{\mathrm{SERS}}}{I_{\mathrm{RS}} / N_{\mathrm{RS}}} \tag{S1}$$

where I_{SERS} and I_{RS} are the intensities of the selected scattering bands in the Raman spectra with and without SERS effect. N_{SERS} and N_{RS} are the numbers of BPE probed in the SERS and the Raman measurements. For normal Raman signal measurements, a total of 10 μL BPE (1 mM) ethanol solution was dispersed on a Si substrate (3×3 mm^2) and dried to yield a thin film of BPE, which can be visible under optical microscopy. For SERS signal measurements, 10 μL BPE (1 nM) solution was cast on the Si substrate loaded with Au@Au and Ag@Au nanostars. We supposed that all the BPE molecules distributed on the substrate. All Raman spectra and SERS spectra were measured under identical conditions (785 nm excitation line, laser power was 0.75 mW, 50×objective lens, integration time was 1s). The averaged relative intensity of the strong band observed at 1643 cm^{-1} shift in the SERS spectra was chosen as a reference to evaluate EF values. As presented in **Figure S6**, the Raman intensities were I_{SERS} = 109.7 and 534.5 (original intensity subtracted by that SERS substrate without BPE) for BPE of $N_{\mathrm{SERS}} = 1 \times 10^{-8}$ mol on the Au and Ag@Au nanostars-adhered substrate, and I_{RS} = 35.4 for BPE of $N_{\mathrm{RS}} = 1 \times 10^{-2}$ mol without nanostars, respectively. According to the above equation, the average EF was calculated to be 3.1×10^6 for the Au nanostars and 1.5×10^7 for the Ag@Au core-shell nanostars.

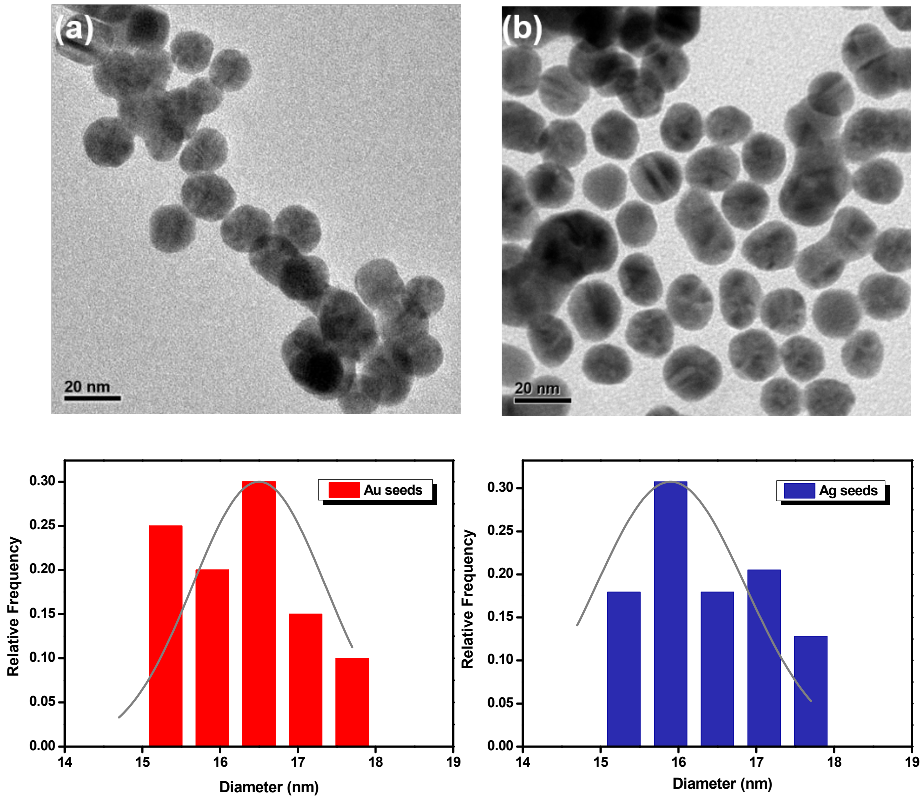

Figure S1. TEM images of (a) Au seeds and (b) Ag seeds, and the corresponding particle size histogram distribution.

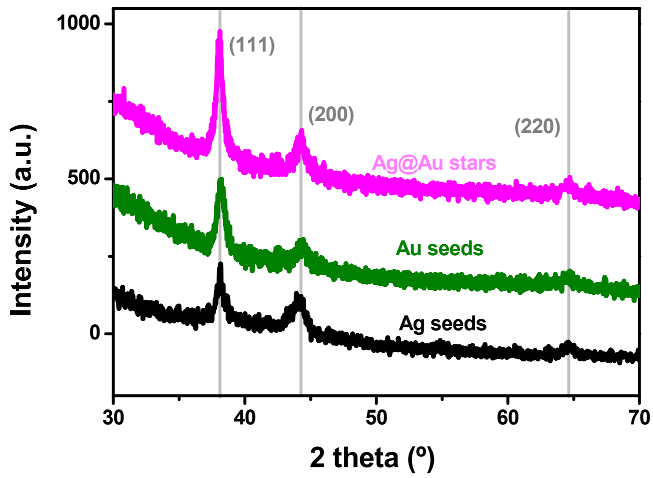

Figure S2. XRD patterns for Ag@Au NSs towards Ag and Au seeds

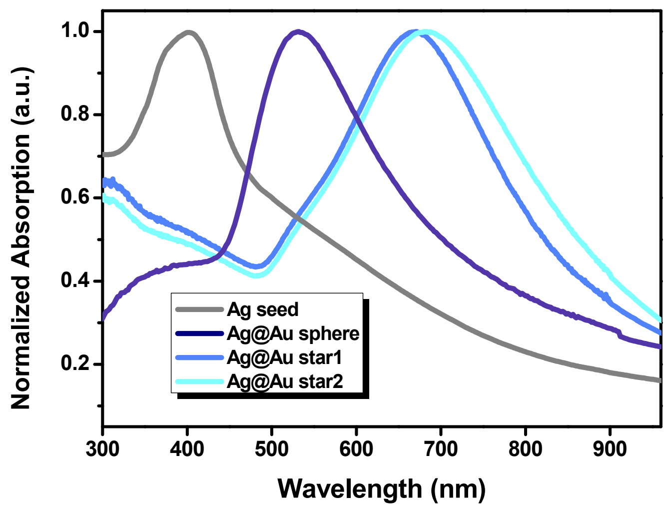

Figure S3. The extinction spectra of Ag seeds and corresponding Ag@Au nanoparticles with and without branched.

Figure S4. Schematics of the models for theoretical calculations. The core with a diameter of 20 nm surrounding five 30 nm length tips. Since the size of gold and silver seeds all are distributed between 15 and 20 nm, and the final diameters reach 50-60nm. Consequently, the nanostars were assumed into two parts: perfect sphere core part with a diameter of 20 nm, and five branches parts with 30 nm length and not sharp tip with a diameter of 10 nm evenly distributed on one plane, which was evaluated by TEM results(**Figure 3l**, **Figure 4l**).

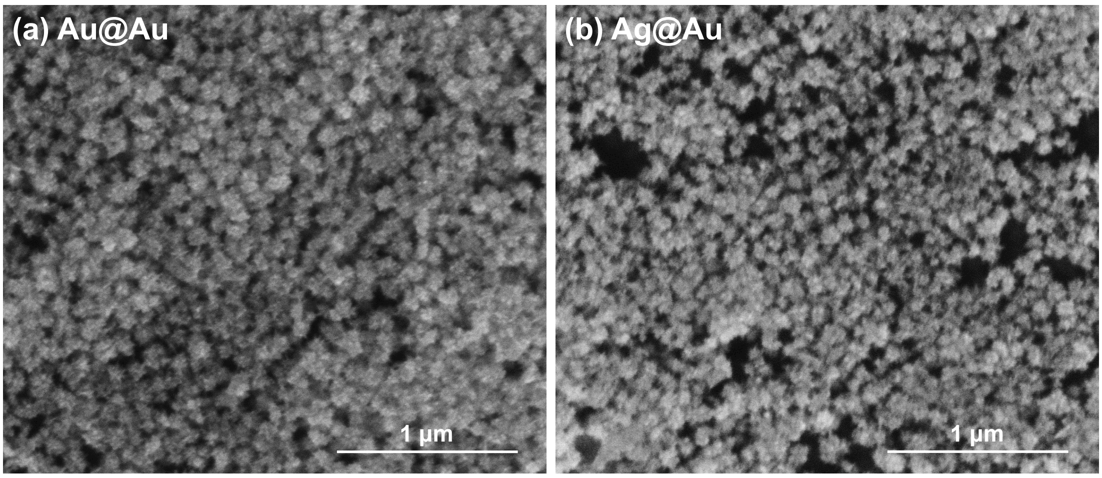

Figure S5. The SEM images of the (a) Au@Au and (b) Ag@Au NSs adhering on Si substrate.

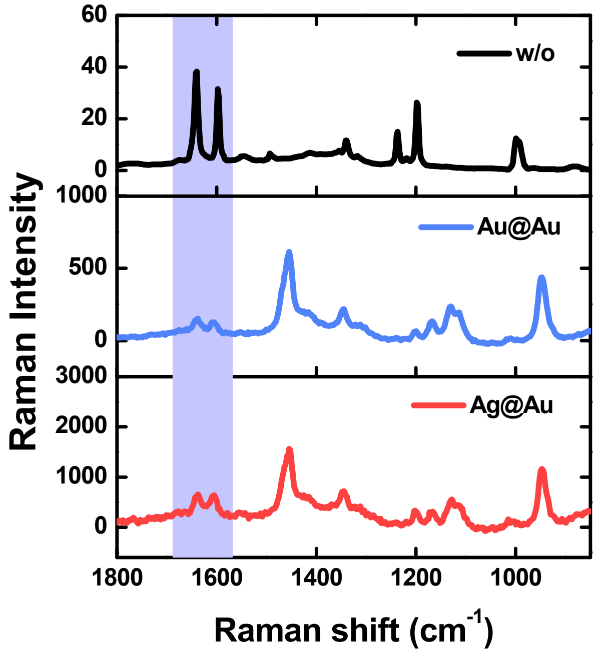

Figure S6. The Raman spectra of 10 µL BPE (1 mM) dispersed on bare Si substrate and 10 µL BPE (1 nM) solution cast on the Si substrate with Au@Au and Ag@Au nanostars