

Electronic Supporting Information for:

## **Enhanced Control of Plasmonic Properties of Silver-Gold Hollow Nanoparticles Via a Reduction-Assisted Galvanic Replacement Approach**

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Table S1. Experimental details for the synthesis of spherical Ag cores (masses are given per 100 mL).

Ag Size (nm)	Tannic Acid (mg)	Citrate (mg)	Centrifuge Ag		Centrifuge AgAu	
			RCF	min	RCF	min
55-60	50	72	7500	12	7500	5
70-75	30	15	6000	10	6000	5
85-90	38	10	6000	8	5000	5
110-120	60	9	4500	9	4500	5

Table S2. FWHM for AgAu hollow nanoparticles as a function of Au concentration. The bulk measurements are related to Fig. 1A, while single particle measurements are related to Fig. 1B-F as noted in the LSPR (nm) column.

Au éq.	Bulk FWHM (nm)	SP FWHM (nm)
Ag	135	73 ± 7
0.25Au:Ag	215	110 ± 17
0.50Au:Ag	200	108 ± 10
0.75Au:Ag	195	122 ± 15
Au:Ag	237	145 ± 20

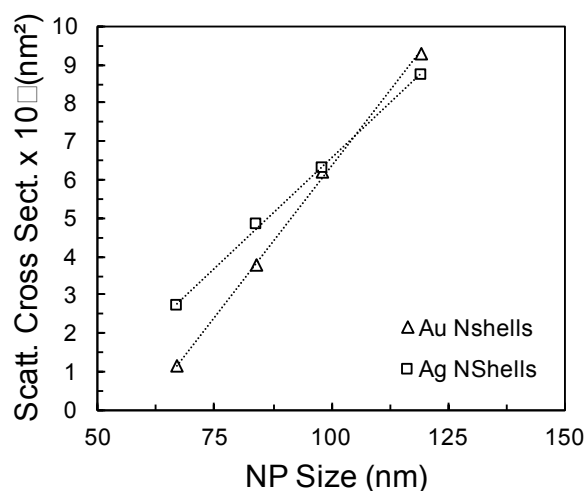


Fig. S1 (A) Mie theory simulation of the scattering cross section of Au (Δ) and Ag (□) hollow nanoparticles as function of size with a 11-nm shell thickness.

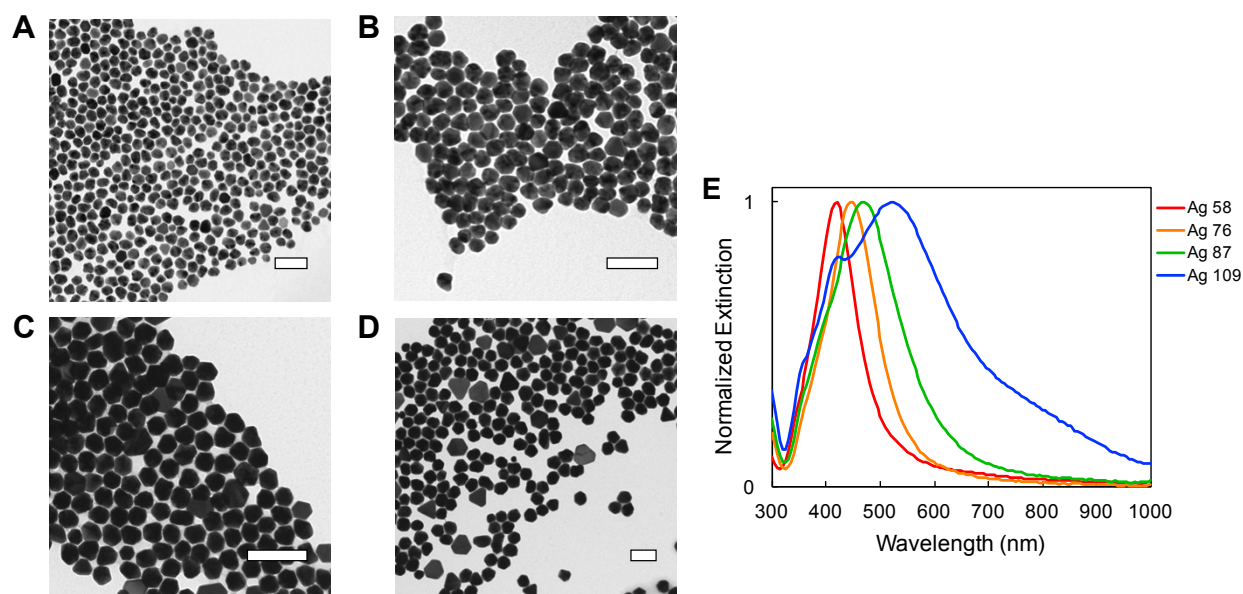


Fig. S2 Ag NPs before formaldehyde-assisted galvanic replacement: (A)  $58 \pm 8$  nm diameter,  $\lambda_{\max} = 430$  nm; (B)  $76 \pm 8$  nm,  $\lambda_{\max} = 450$  nm; (C);  $87 \pm 9$  nm,  $\lambda_{\max} = 475$  nm; (D)  $109 \pm 12$  nm,  $\lambda_{\max} = 520$  nm. Scale bars, 200 nm. (E) Normalized extinction of Ag NPs of sizes shown in A-D.

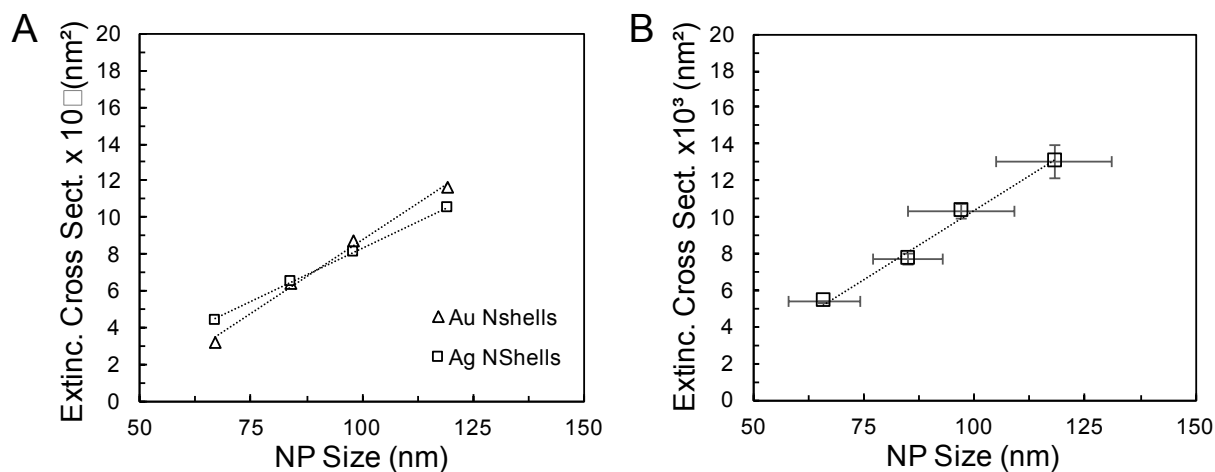


Fig. S3 (A) Mie theory simulation of extinction cross sections for Au ( $\Delta$ ) and Ag ( $\square$ ) hollow nanoparticles as function of size with a 11-nm shell thickness. (B) Experimental extinction cross sections ( $\sigma = \ln(10) D/\rho L$ ), where  $D$  is the optical density,  $\rho$  the number of density in particles/cm<sup>3</sup> and  $L$  the sample length in cm) for the hollow nanoparticles shown in Figure 3.

Table S3. NP size, refractive index sensitivity (RIS), full width at half maximum (FWHM), and sensing figure of merit (FOM) for several hollow NP sizes with 11 nm of shell thickness

NP Size (nm)	$r_1/(r_2-r_1)$	RIS (nm/RIU)	FWHM (nm)	FOM
$66 \pm 8$	2.1	237	150	1.6
$85 \pm 8$	2.7	291	190	1.5
$94 \pm 9$	3.1	310	200	1.5
$98 \pm 12$	3.5	345	245	1.4
$118 \pm 13$	4.6	444	440	1.0

Note: Overall performance of the AgAu hollow nanoparticle were compared using figure of merit (FOM) values. FOMs were calculated by dividing the RIS (in nm/RIU) with the FWHM (in nm) of the extinction peak observed in water.

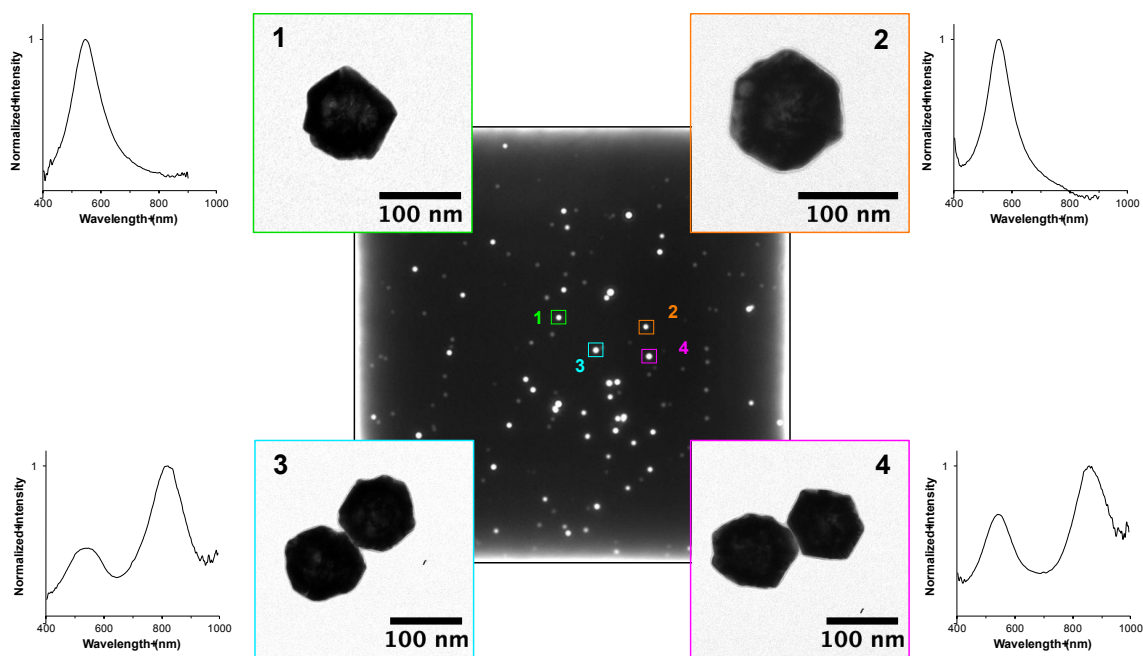


Figure S4. Darkfield optical microscopy image (center) and correlated single NP spectra and TEM images corresponding to the regions indicated in the optical image. This is an example of the single nanoparticle LSPR/TEM correlation performed to validate our data collection technique. Dimers and aggregates were eliminated from data based on the spectrum shape, as assessed by these correlated measurements.