

## Electronic Supplementary Information

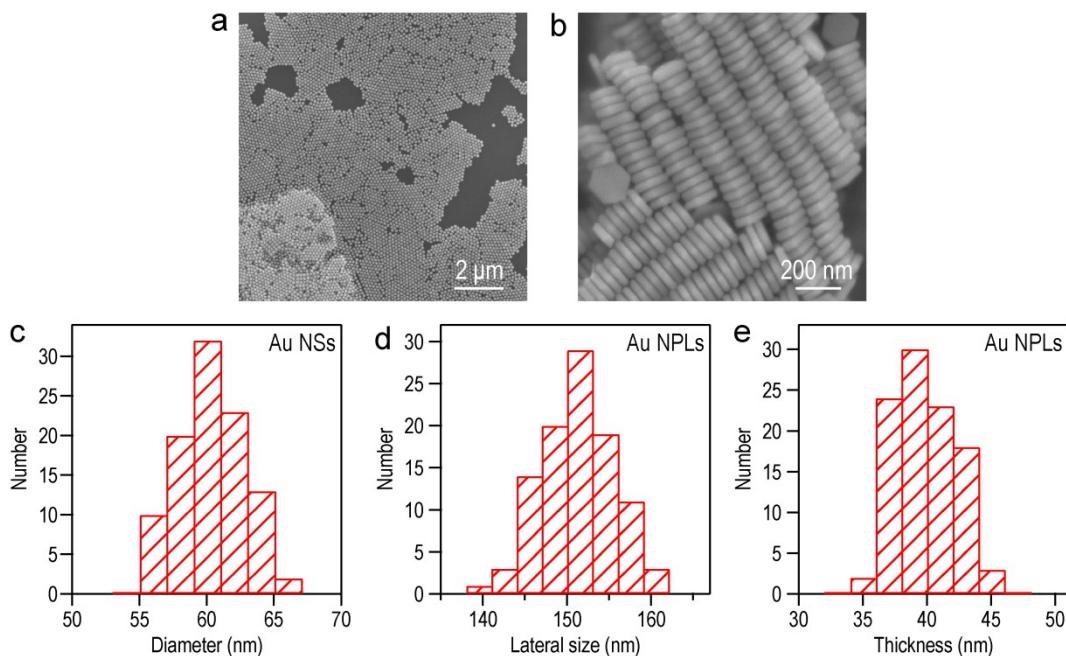
### Switching plasmonic Fano resonance in gold nanosphere–nanoplate heterodimers

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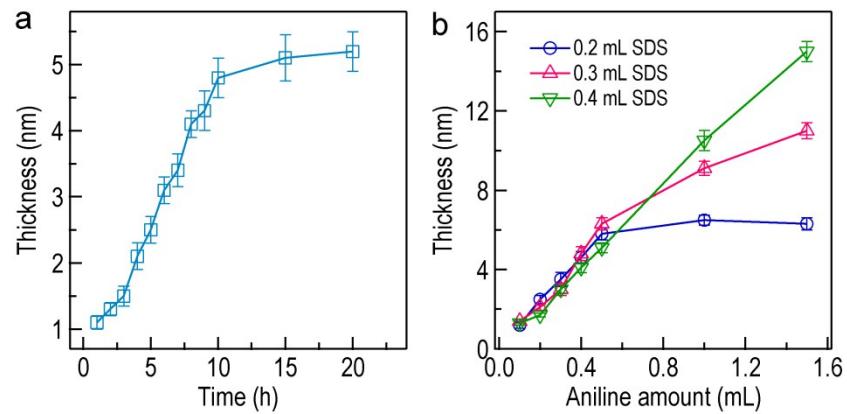
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### Supplementary figures

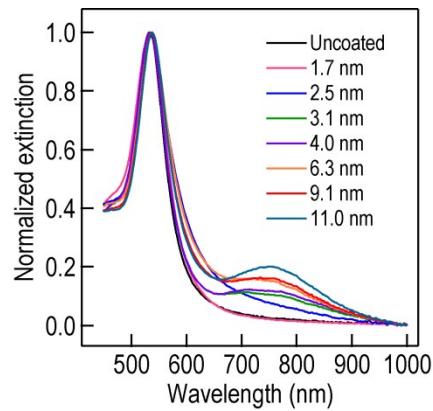


**Fig. S1** Au nanosphere (NS) and nanoplate (NPL) samples. (a) SEM image of the Au NSs. (b) SEM image of the Au NPLs. Most of the NPLs are stacked together and aligned vertically.

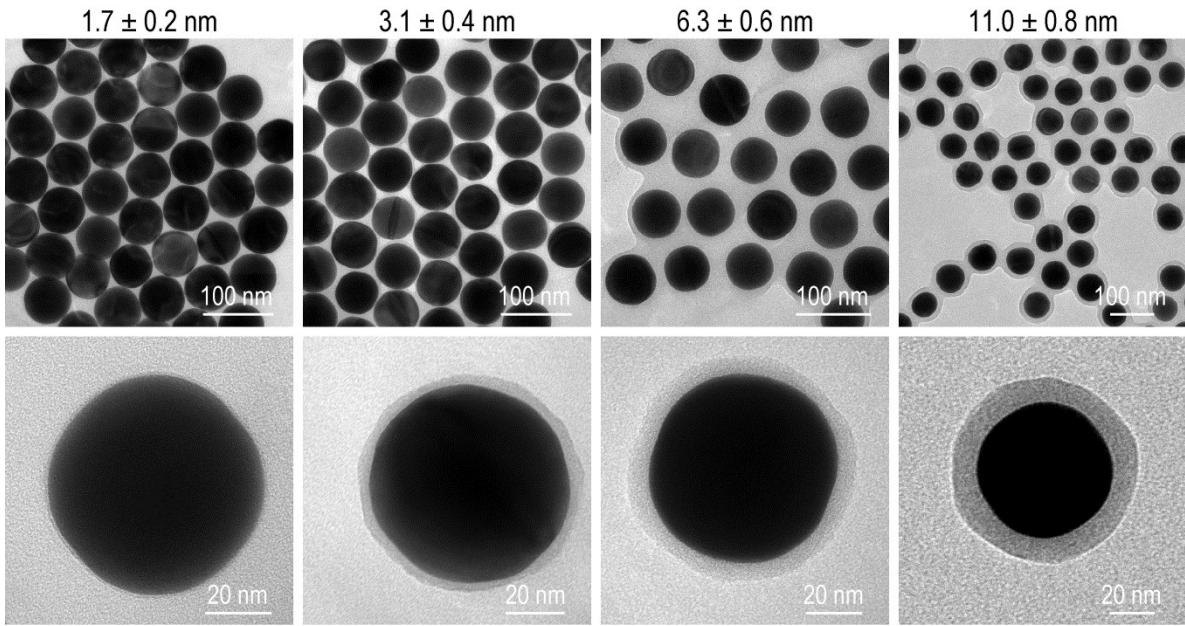
(c–e) Histograms of the diameters of the Au NSs, the lateral sizes and the thicknesses of the Au NPLs, respectively.



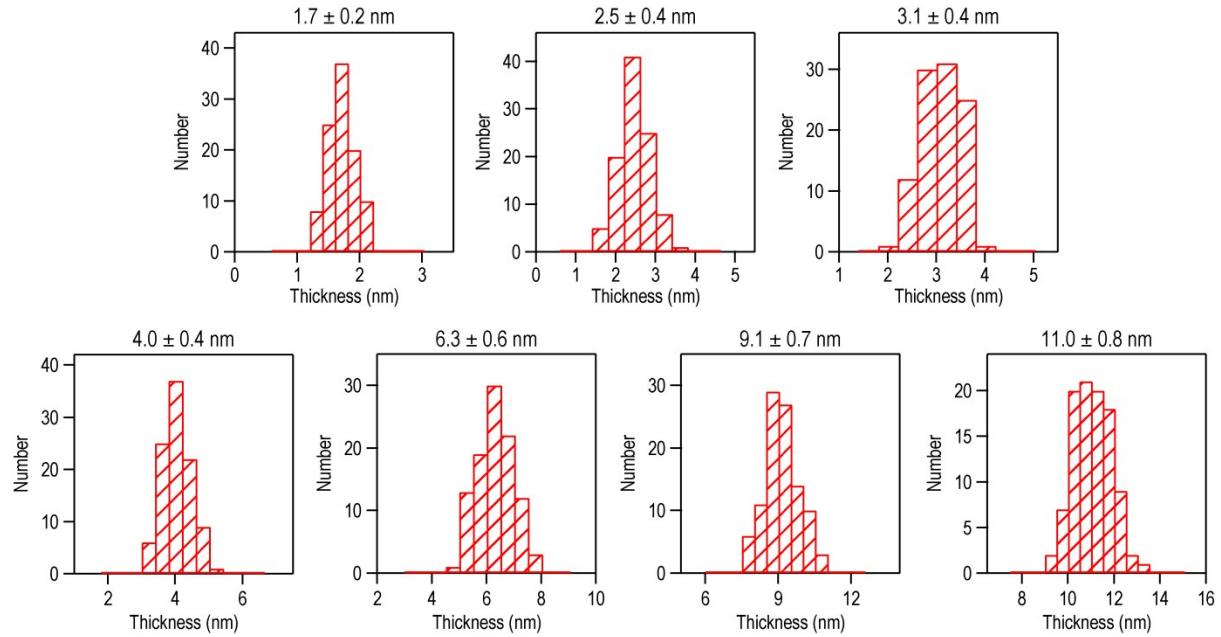
**Fig. S2** PANI coating on the Au NSs. (a) Variation of the thickness of the PANI shell on the Au NSs with the reaction time. (b) Variations of the thickness of the PANI shell with the aniline amount when SDS was added in the coating solution at different amounts.



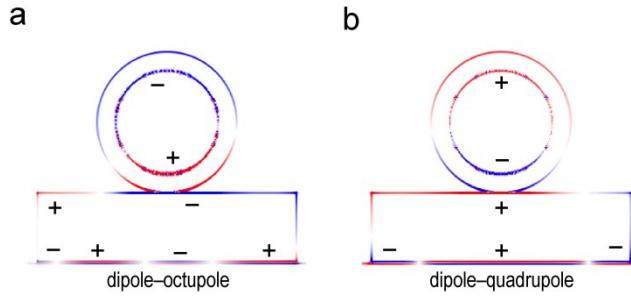
**Fig. S3** Normalized extinction spectra of the PANI-coated Au NSs with the PANI shell of different thicknesses.



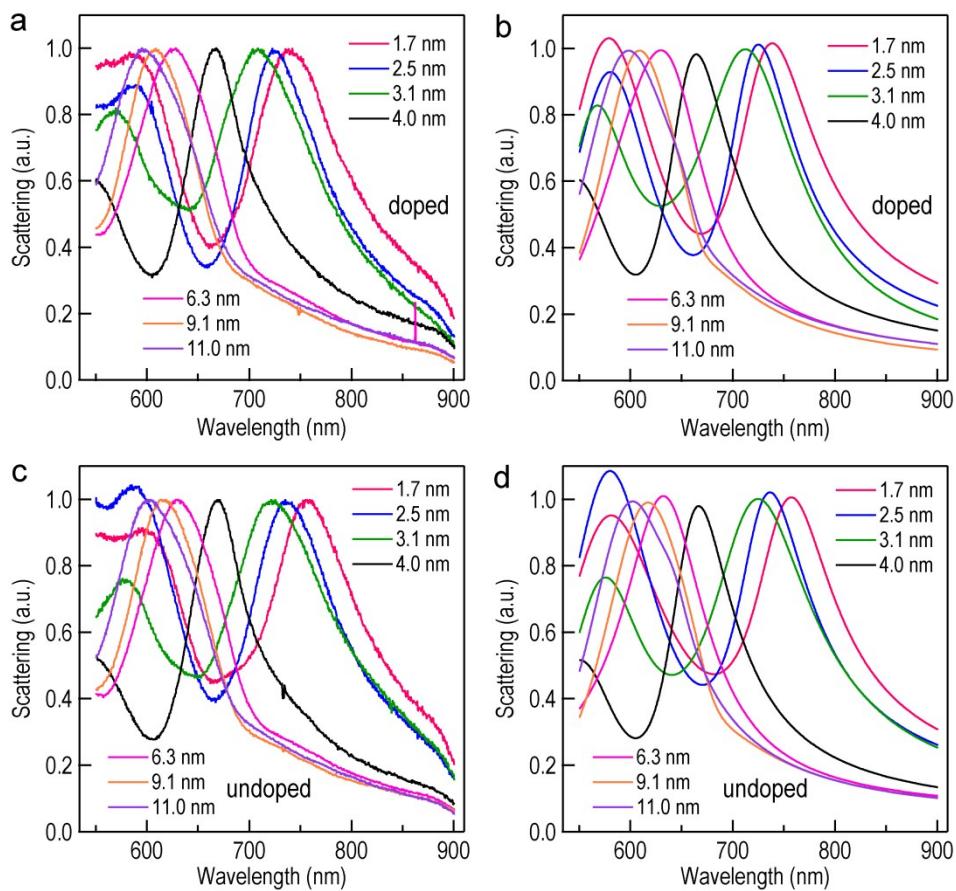
**Fig. S4** Low- (top row) and high-magnification (bottom row) TEM images of four representative PANI-coated Au NS samples. The thicknesses of the PANI layers are  $1.7 \pm 0.2$  nm,  $3.1 \pm 0.4$  nm,  $6.3 \pm 0.6$  nm and  $11.0 \pm 0.8$  nm, respectively.



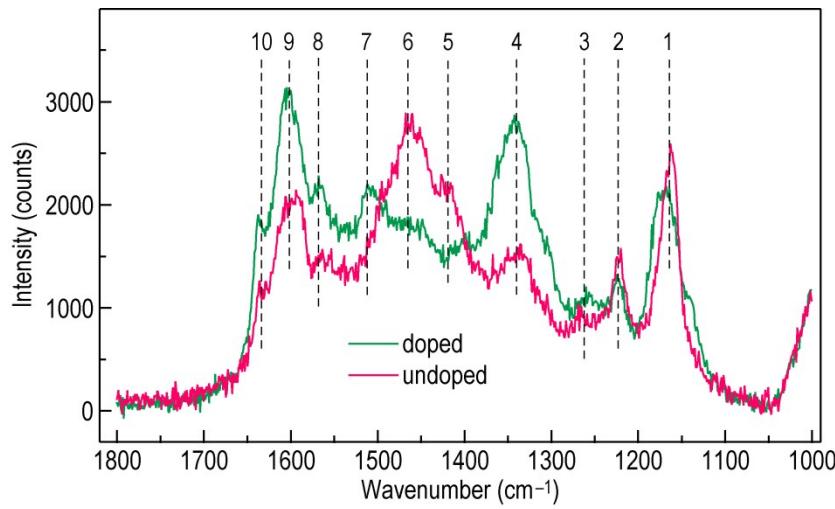
**Fig. S5** Histograms of the thicknesses of the PANI layers of the seven PANI-coated Au NS samples.



**Fig. S6** Simulated charge distribution contours of the Au NS–NPL heterodimer with a gap distance of 11 nm. (a) Charge distribution of the main peak at 604 nm. (b) Charge distribution of the bump at 725 nm.



**Fig. S7** Fitting of the plasmonic Fano resonance according to the two-oscillator model. (a, b) Measured and fitted scattering spectra of the heterodimers with different gap distances at the doped state of PANI, respectively. (c, d) Measured and fitted scattering spectra of the heterodimers at the undoped state of PANI, respectively.



**Fig. S8** Raman spectra of PANI at the doped and undoped states. The Raman spectra were measured on the individual Au NS–NPL heterodimer with a gap distance of 6.3 nm under the excitation of a 633-nm laser. The Raman scattering peaks are labeled from 1 to 10. The assignments of these peaks are listed in Table S2.

## Supplementary tables

**Table S1** Fitting parameters for the measured scattering spectra in the doped state

Gap distance (nm)	$\omega_s$ (eV)	$W_s$ (eV)	$\omega_a$ (eV)	$W_a$ (eV)	$a$	$b$	$q$
1.7	1.70	0.11	2.13	0.22	3.15	0.30	1.39
2.5	1.72	0.10	2.12	0.20	3.01	0.28	1.39
3.1	1.73	0.14	2.19	0.16	2.46	0.25	1.33
4.0	1.89	0.11	2.24	0.21	1.49	0.27	1.02
6.3	2.00	0.27	1.90	0.13	0.76	0.79	0.28
9.1	2.03	0.18	1.86	0.06	0.98	0.82	0.07
11.0	2.08	0.214	1.85	0.05	0.97	0.99	0.09

**Table S2** Fitting parameters for the measured scattering spectra in the undoped state

Gap distance (nm)	$\omega_s$ (eV)	$W_s$ (eV)	$\omega_a$ (eV)	$W_a$ (eV)	$a$	$b$	$q$
1.7	1.65	0.10	2.12	0.25	3.46	0.29	1.44
2.5	1.70	0.10	2.12	0.21	3.27	0.26	1.50
3.1	1.72	0.14	2.15	0.19	2.23	0.24	1.33
4.0	1.88	0.11	2.22	0.20	1.82	0.34	1.01
6.3	1.94	0.15	2.23	0.58	0.42	0.68	0.11
9.1	2.01	0.19	1.83	0.07	0.96	0.84	0.19
11.0	2.06	0.20	1.92	0.05	0.99	1.17	0.00

**Table S3** Fitting parameters for the simulated scattering spectra shown in Fig. 5a

Gap distance (nm)	$\omega_s$ (eV)	$W_s$ (eV)	$\omega_a$ (eV)	$W_a$ (eV)	$a$	$b$	$q$
1	1.66	0.11	2.03	0.24	1.16	1.06	1.02
2	1.77	0.10	2.15	0.33	1.03	0.64	0.89
3	1.91	0.09	2.19	0.40	0.99	0.48	0.74
4	1.97	0.08	2.46	0.46	0.96	0.46	0.71
6	1.99	0.15	1.69	0.24	1.38	0.80	0.52
9	2.02	0.18	1.67	0.22	1.22	1.10	0.47
11	2.03	0.19	1.67	0.21	1.20	1.08	0.42

**Table S4** Assignments of the Raman peaks of PANI

No.	Wavenumber ( $\text{cm}^{-1}$ )	Assignment <sup>a</sup>
1	1164	$\delta(\text{C}-\text{H})_{\text{Q}}$
2	1223	$\nu(\text{C}-\text{N})_{\text{Q}}$
3	1262	$\nu(\text{C}-\text{N})_{\text{B}}$
4	1340	$\nu(\text{C}\sim\text{N}^{+})$
5	1419	Phz
6	1465	$\nu(\text{C}=\text{N})_{\text{Q}}$
7	1512	N–H bending
8	1568	$\nu(\text{C}-\text{C})_{\text{Q}}$
9	1602	$\nu(\text{C}=\text{C})_{\text{Q}}$
10	1634	Phz

<sup>a</sup>  $\delta$ , in-plane bending;  $\nu$ , stretching; B, benzenoid ring; Q, quinonoid ring;  $\sim$ , bond intermediate between single and double bonds; Phz, phenazine-like segment.