## **Electronic Supplementary Information**



**Fig. S1.** STEM-EDX measurement of single Ag/Au nanoshells. The Ag/Au nanoshell monomer consists of 73% Ag and 27% Au. The shell thickness is around 5-6 nm and the particle diameter about 65 nm.



Fig. S2. SEM and TEM images of Au nanostars.



Fig. S3. SEM and TEM images of Au/Au superstructures.



**Fig. S4.** SP experiments with Au nanostars and 4-NTB-MEG-OH as Raman marker. To emphasize SERS activity of a single Au nanostar, another type of Raman reporter is used. The Raman band of the nitro group appears significantly at 1340 cm<sup>-1</sup>. Single Au nanoparticle and single Au/Ag nanoshells with 4-NTB-MEG-OH Raman marker show no SERS activity at all. Raman mapping was performed with a 100x DF objective (220  $\mu$ W, 0.5 s, 632.8 nm laser excitation wavelength).



**Fig. S5.** Further additional DF scattering/LSPR spectra of individual AuNP, Au/Ag nanoshells, Au nanostars and Au/Au supraparticles.

Tab. T1 FEM computer simulations based on the parameters listed in the table.

	Au NP	Ag/Au nanoshell	Au nanostar	Au/Au supraparticle
Diameter	55 nm	d <sub>in</sub> = 57 nm d <sub>out</sub> = 67 nm	ca. 80-100 nm	ca. 110 nm
Surface area with Ra	7654 nm²	14103 nm²	26503 nm²	18151 nm²
N <sub>tips/satellites</sub>	-	-	26	30



Fig. S6. Correlative SP SERS/SEM experiments of Au nanospheres.



Fig. S7. Correlative SP SERS/SEM experiments of Au/Ag nanoshells.



Fig. S8. Correlative SP SERS/SEM experiments of Au nanostars.



Fig. S9. Correlative SP SERS/SEM experiments of Au/Au core/satellite nanoparticles.



Fig. S10. Raman spectrum of 4-MBA in solution.