Supporting Information

Structural and Optical Control of DNA-mediated Janus Plasmonic Nanostructures

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The etching processes under nitrogen atmosphere and in air.

The 5 μ l 2 M NaCl solution was added to the 0.2 nM Au-Ag JNs in 100 μ l deionized water under nitrogen atmosphere and in air, respectively. Incubate and let the reaction last for half an hour at room temperature, then wash the mixture with deionized water and prepare for UV/Vis characterizations. As can be seen that the solution color in air turns into green and the third peak shows an obvious blue-shift while the sample under nitrogen atmosphere shows little change. This demonstrates the reduction of Ag NPs is due to the etching effect induced by Cl⁻/O₂ pairs.



Figure S1. UV-vis extinction spectra of Au-Ag JNs under nitrogen atmosphere and in air.



Figure S2. The EDS spectra of (a) the as-grown Au-Ag JNs and (b) the sample with Ag NPs being fully etched away.

Compare the transverse and longitudinal plasmonic modes of the Au-Ag JN, Au-Au dimer and Ag-Ag dimer

For transverse polarization, the calculated extinction spectra of the Ag-Ag and Au-Au dimers display only one peak due to the LSPR of Ag and Au NPs, while that of the Au-Ag JN shows two peaks at positions close to the characteristic LSPR peaks of Au and Ag NPs. For longitudinal polarization, the CTP peak of the Au-Ag JN is located between those of the Au-Au and Ag-Ag dimers. The peak at the shorter wavelength of the Au-Ag JN originates mainly from the longitudinal LSPR of the Ag NP, while those of the Ag-Ag and Au-Au dimers are corresponding to the bonding dipolar dimer plasmon (BDP) mode.



Figure S3. FDTD simulated spectra for the incident light polarized along the transverse (a) and longitudinal (b) axes of the Au-Ag JN, Au-Au dimer and Ag-Ag dimer. For transverse polarization, the electric field distributions 1 and 3 are recorded for Au-Au and Ag-Ag dimers at peak positions 397 and 523 nm, those of 2 and 4 are taken for Au-Ag JN at the peak positions 401 and 530 nm, respectively. For longitudinal polarization, the electric field distributions 1 and 4 recorded for the Ag-Ag dimer at peak positions 399 and 676 nm, those of 3 and 6 are taken for the Au-Au dimer at positions 526 nm and 728 nm, those of 2 and 5 are taken for the Au-Ag JN at 393 and 701 nm, respectively.



Figure S4. UV/Vis extinction spectra of the Au-Ag JN solutions treated with different amount of NaCl. The right column shows the color of the solutions, from bottom to top corresponding to 0, 10, 30, 40, 50, 100 to 200 mM of NaCl, respectively. A dashed curve is added to guide the eye through the evolution of the CTP mode with varying salt concentration.



Figure S5. TEM images of samples with 0.1 M NaCl at the different solution temperature. From left to right, they are corresponding to (a) 25 °C, (b) 60 °C, (c) 80 °C, and (d) 95 °C. All the scale bars are 50 nm.



Figure S6. The TEM images (a-c) for the composite nanostructures in Figure 5(b-c), respectively. All the scale bars are 50 nm. The synthetic yields for the nanostructures in a, b and c are estimated to be 50%, 20% and 5%, respectively.