Supporting Information

Single Molecule Level Plasmonic Catalysis - Dilution Study of p-Nitrothiophenol on Gold Dimers

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Experimental details and 3 Figures.

Experimental details

Sample preparation. SERS active gold nanoparticle dimers were prepared using a wet chemistry method (J Appl Phys, 2013, 113, 033102.). 200 μ l of GD colloid solutions were added to 200 μ l of pNTP in aqueous solution (c = 5 × 10⁻⁷ – 10⁻⁹ M) for 2 hours. Then, 20 μ l of the mixture were spin-coated onto cleaned glass slides marked with position indicators, then dried in vacuum.

Measurements. SERS measurements were performed on an inverted microscope with a $\times 100$ objective (NA=0.9, Olympus, Japan), using 632.8 nm laser excitation with a laser spot size of ~1 µm. All spectra were obtained from a single nanoparticle dimer placed in the center of the laser spot. A notch filter was placed in front of the entrance of the spectrometer (SP2750i, Princeton Instruments, USA), and Raman spectra were detected using a CCD detector (PIXIS400, Princeton Instruments, USA) with an acquisition time of 10 s. All spectra shown are raw and untreated data, only a linear baseline correction was applied for presentation.



Figure S1. Time dependent SERS spectra of pNTP (c= 10^{-9} M). The dominating band at 1332 cm⁻¹ (v_{NO2}) decreased and disappeared within 10 min, simultaneously, a new peak at 1017 cm⁻¹ was detected after 5 min.



Figure S2. Representation of number of molecules absorbed on gold dimers. About 1000, 100, and 10 molecules absorbed on a gold dimer for the concentrations of 5×10^{-7} , 10^{-8} and 10^{-9} M, respectively.



Figure S3. Comparison of low wavenumber SERS spectra of pNTP and TP with spectra recorded during the experiment on a single gold dimer. (a) time dependent SERS spectra of reacted pNTP ($c = 10^{-9}$ M) at 3 mW laser at 30 and 60 min. Normal SERS spectra and corresponding molecular structures of TP and pNTP are shown in (b) and (c), respectively.