Supporting Information for

Polydopamine-Enabled Approach toward Tailored Plasmonic Nanogapped Nanoparticles: From Nanogap Engineering to Multifunctionality

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Materials and Characterization

Dopamine, sodium citrate, potassium gold(III) chloride ($KAuCl_4$), bicine, hydroxylamine hydrochloride ($NH_2OH{\cdot}HCl$), iron(III) chloride hexahydrate ($FeCl_3{\cdot}6H_2O$) iron(II) chloride ($FeCl_2{\cdot}4H_2O$), ammonium hydroxide, oleic acid, sodium dodecyl sulfate (SDS), styrene, tetradecane, potassium persulfate (KPS), zirconium(IV) chloride ($ZrCl_4$), terephthalic acid (H_2BDC), acetic acid, hexadecyltrimethylammonium bromide (CTAB), silver nitrate ($AgNO_3$), sodium borohydride ($NaBH_4$), 4-nitrothiophenol (NTP), and bovine serum albumin were purchased from Sigma Aldrich. Methanol (MeOH) and N,N-dimethylmethanamide (DMF) were obtained from Fisher Chemical. Hydrogen tetrachloroaurate(III) trihydrate ($HAuCl_4{\cdot}3H_2O$) was from Alfa Aesar. Ultrapure water (18.2 MΩ·cm) was purified using a Sartorius AG arium system and used in all experiments. Methoxy-poly(ethylene glycol)-thiol (PEG-SH, 5 kDa) and carboxymethyl-poly(ethylene glycol)-thiol (HOOC-PEG-SH, 3.4 kDa) were purchased from Laysan Bio, Inc. Lissamine rhodamine B ethylenediamine (RhB-NH_2) was purchased from Life Technologies. LIVE/DEAD® BacLight™ Bacterial Viability Kits was purchased from Thermo Fisher Scientific. The pair of detection/capturing monoclonal antibodies (8B1-C2-B1 and 10C5-H3-B6) was obtained as a gift from Dr. Weihua Lai's group in Nanchang University.

Transmission electron microscopy (TEM) observations were conducted on a Jeol JEM 2010 electron microscope at an acceleration voltage of 300 kV. UV–vis spectra were recorded using a Shimadzu UV1800 spectrophotometer. Fluorescence spectra were collected on a Fluoromax-3 spectrometer (Horiba Scientific). Temperatures of solutions were obtained using FLIR T420 thermal imaging infrared camera. A RENISHAW Raman microscope with WIRE 2.0 software and 632.8 nm (maximum energy: 50 mW) emission line of an air-cooled He−Ne laser was used for SERS measurements. The laser beam with a laser spot size of 2−5 μm was focused by a 50× objective. A single scan with an integration time of 15 s was performed. The bacterial cells were imaged using laser scanning confocal microscopy (ZEISS LSM 800 with Airyscan).

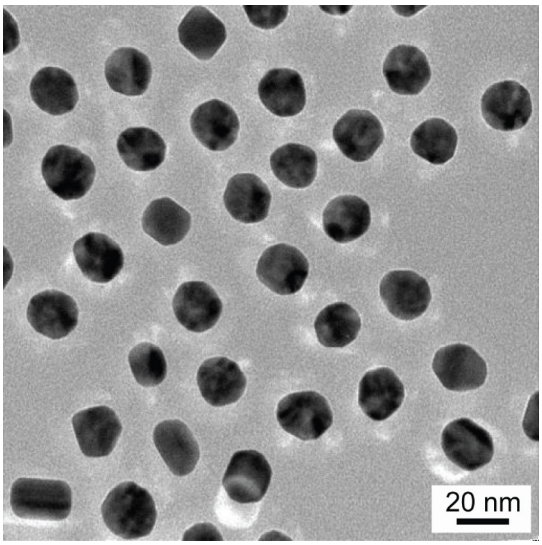

Figure S1. TEM image of 20 nm Au nanoparticles.

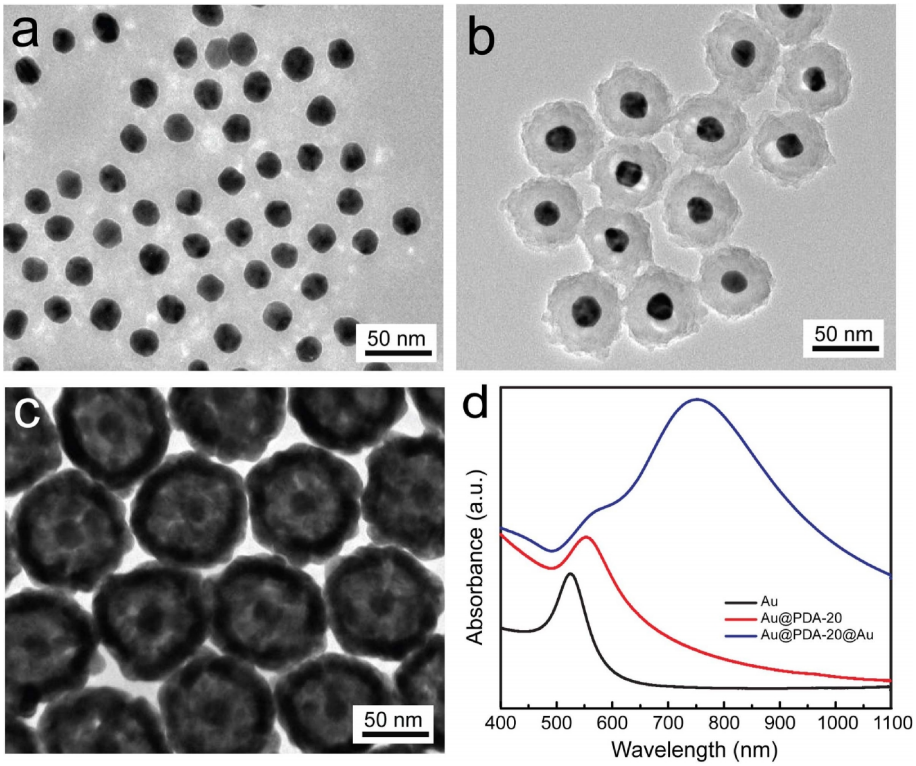

Figure S2. TEM images of 20 nm AuNP (a), Au@PDA-20 (b), and Au NNPs with a 20 nm nanogap. (c). UV−vis spectra of AuNP (black line), Au@PDA-20 (red line), and Au NNPs with a 20 nm nanogap (blue line).

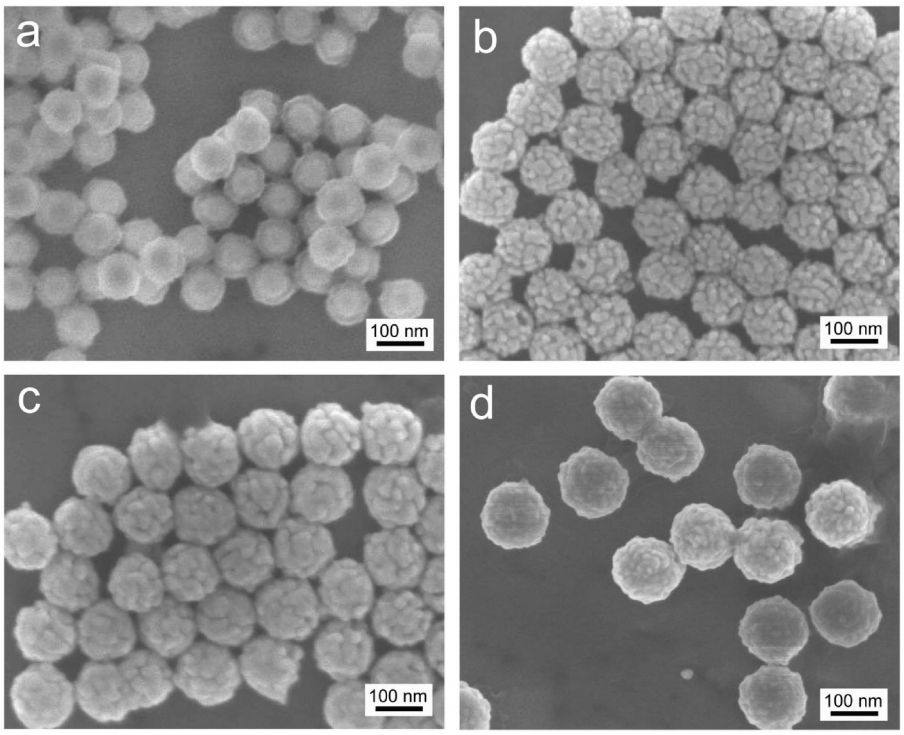

Figure S3. SEM images of the nanogapped nanoparticle at different amount of Au precursor: (a) 0 uL, (b) 25 uL, (b) 50 uL, (c) 80 uL.

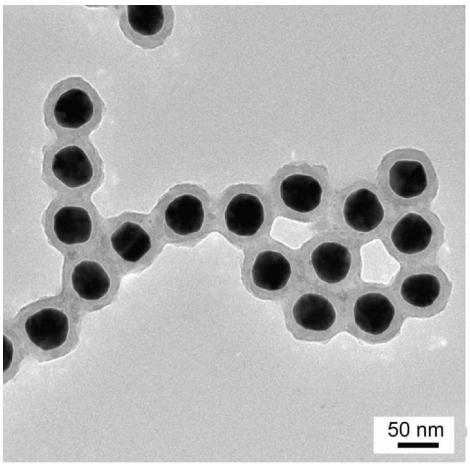

Figure S4. TEM image of Au(50nm)@PDA.

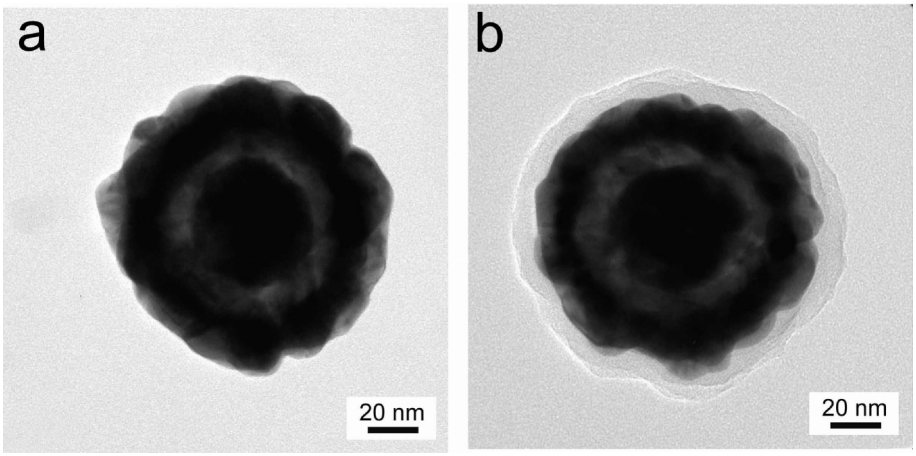

Figure S5. TEM images of Au(50nm)@PDA@Au and Au(50nm)@PDA@Au@PDA.

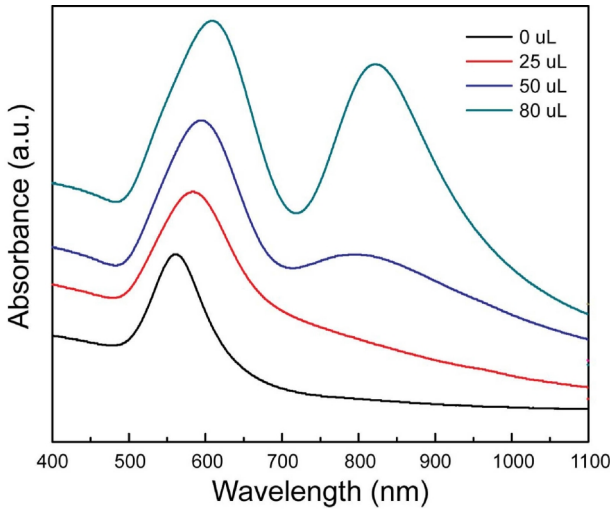

Figure S6. UV−vis spectra of Au nanogapped nanoparticle at different stage of growth.

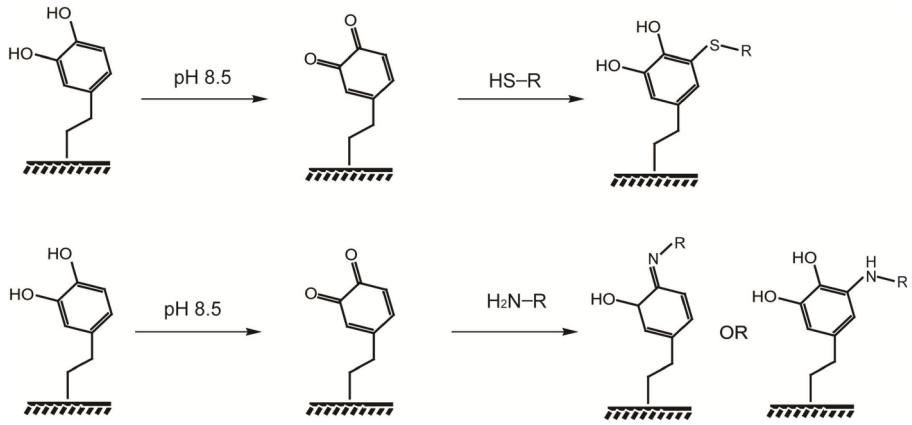

Figure S7. The PDA-coated nanoparticles conjugated with thiol and amine groups *via* Michael addition and/or Schiff base reaction.

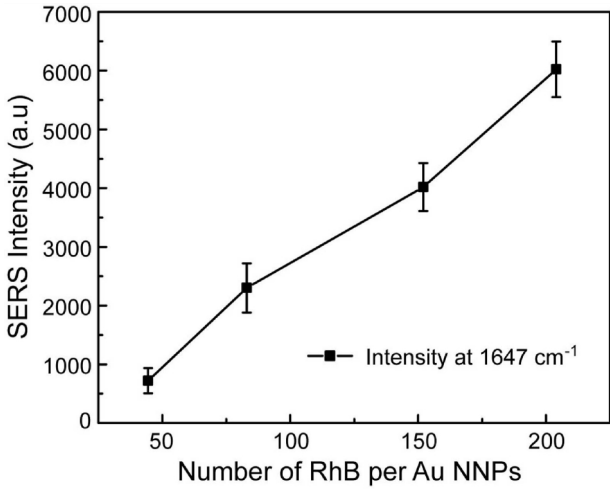

Figure S8. SERS intensity at 1647 cm^{-1} as a function of the average number of RhB molecules loaded in each Au NNP.

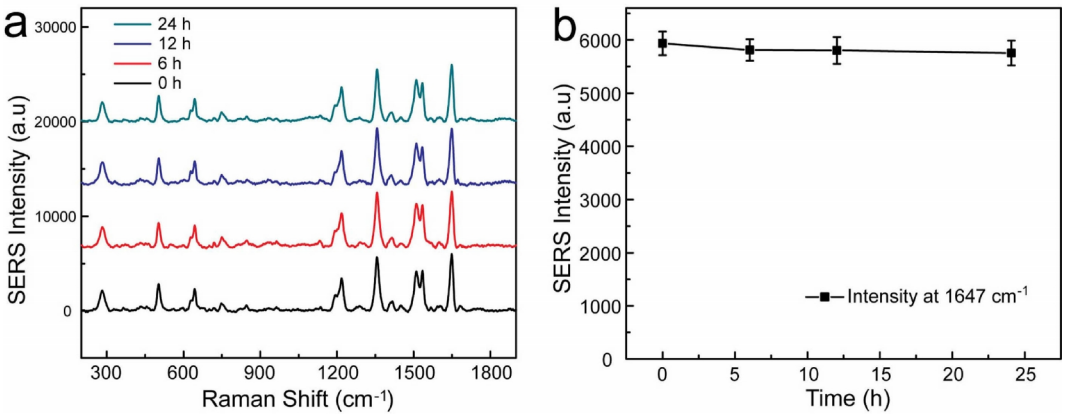

Figure S9. (a) SERS spectra of the as-prepared nanogapped nanoparticle dispersed in water at 0, 6, 12, and 24 h. (b) The time-dependent SERS intensity at 1647 cm^{-1} during 24 h.

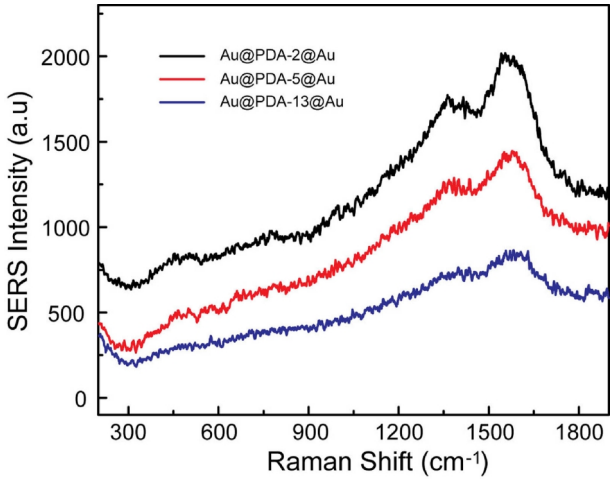

Figure S10. SERS spectra of different Au NNPs without Raman dyes.

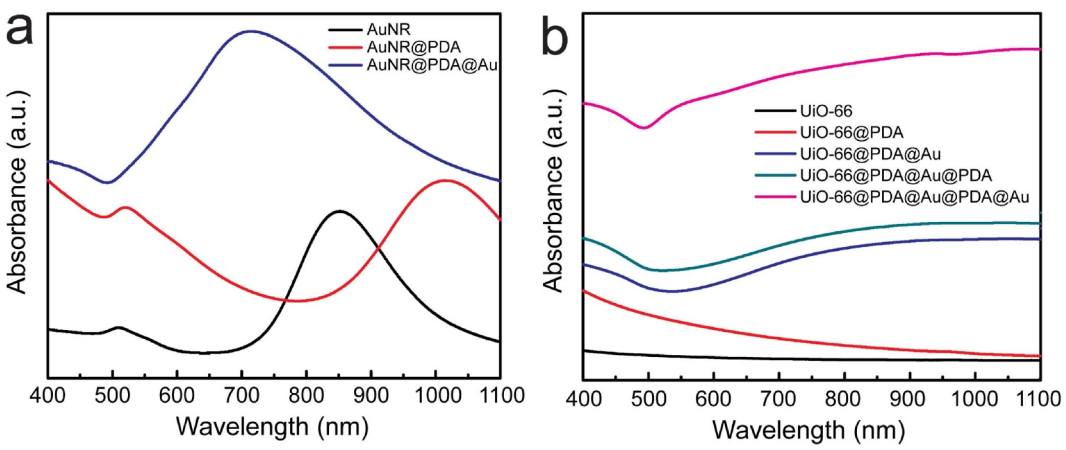

Figure S11. (a) UV–vis spectra of plasmonic AuNR NNPs at different stages. (b) UV–vis spectra of plasmonic UiO-66-cored NNPs at different stages.

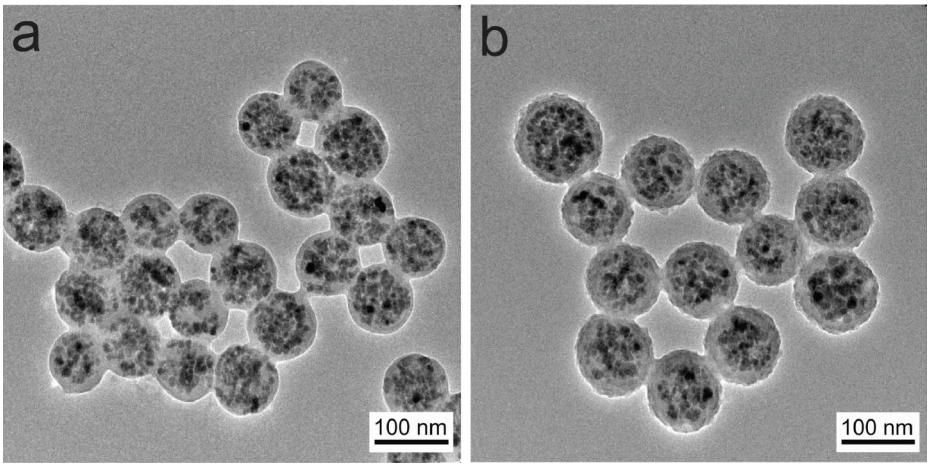

Figure S12. TEM images of MagNP (a) and MagNP@PDA (b).

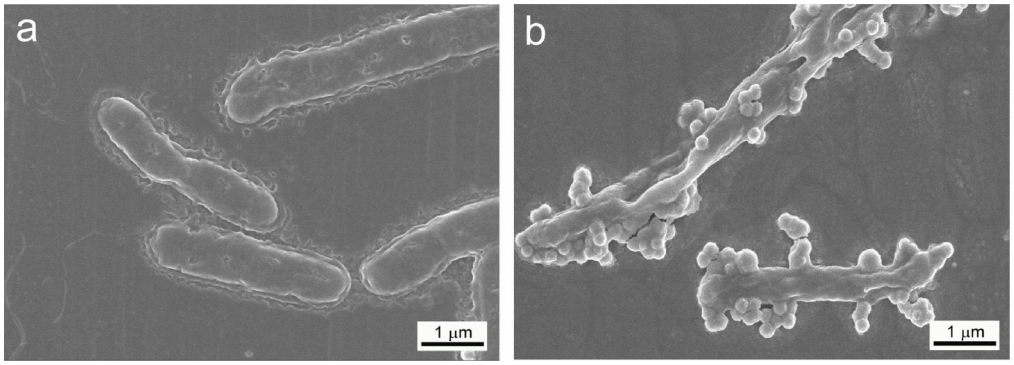

Figure S13. SEM images of *E. coli* O157:H7 before (a) and after (b) captured by magnetic NNPs.

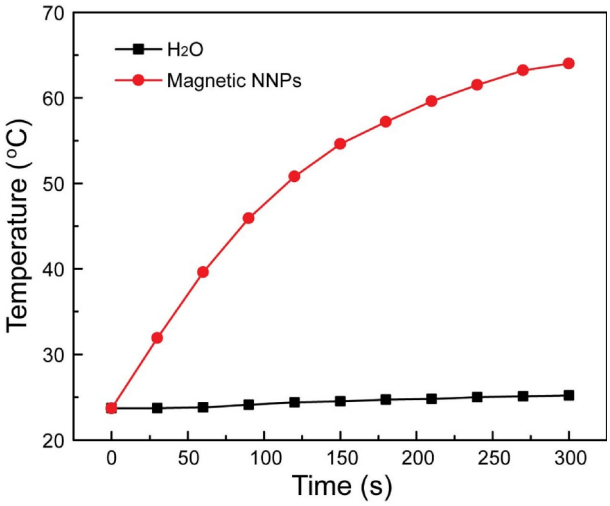

Figure S14. Photothermal conversion of magnetic NNPs exposed to an 808 nm laser (1 W/cm^2) at $OD_{808\ nm}$=1.5.

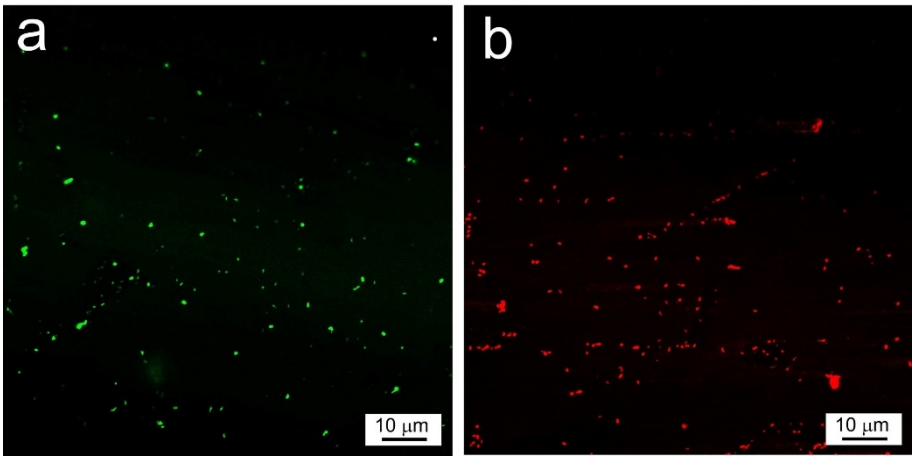

Figure S15. Fluorescence images of the captured bacteria before (a) and after (b) photothermal treatment by exposure to an 808 nm laser (1 W/cm^2) for 15 min.