

# Supporting Information

## Engraving High-Density Nanogaps in Gold Thin Films via Sequential Anodization and Reduction for Surface-Enhanced Raman Spectroscopy Applications

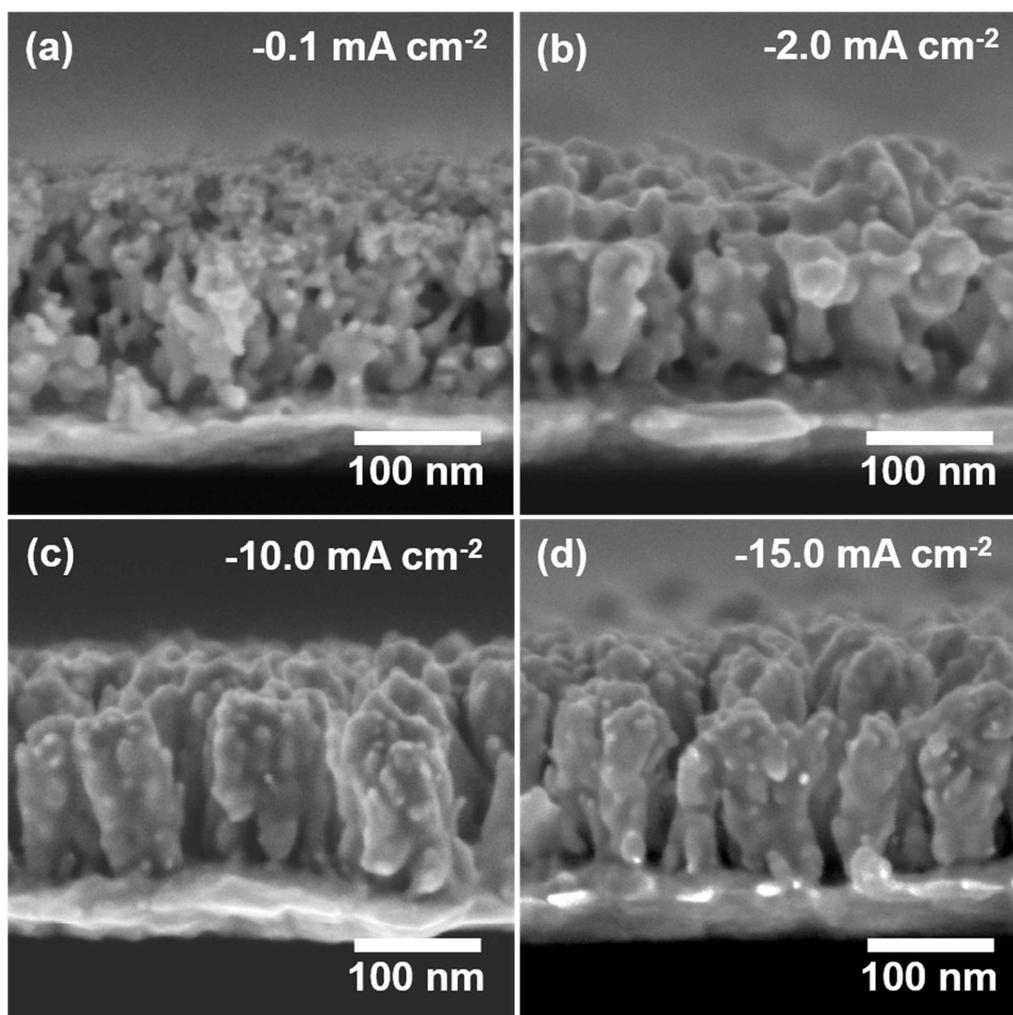
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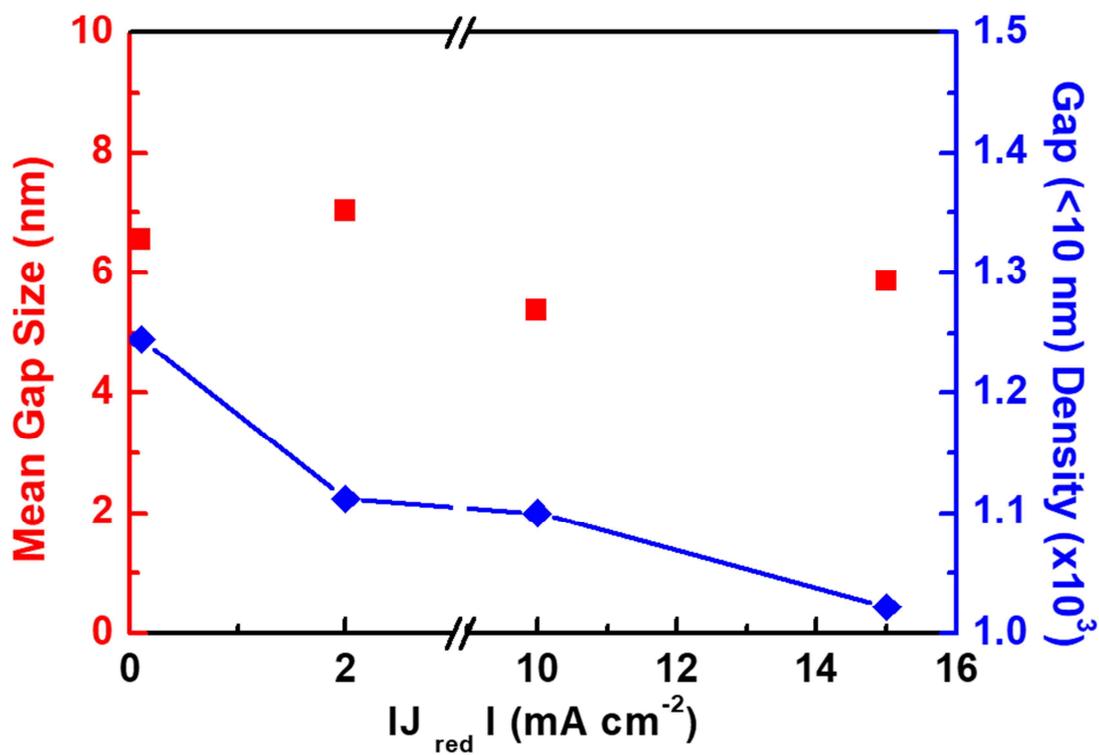
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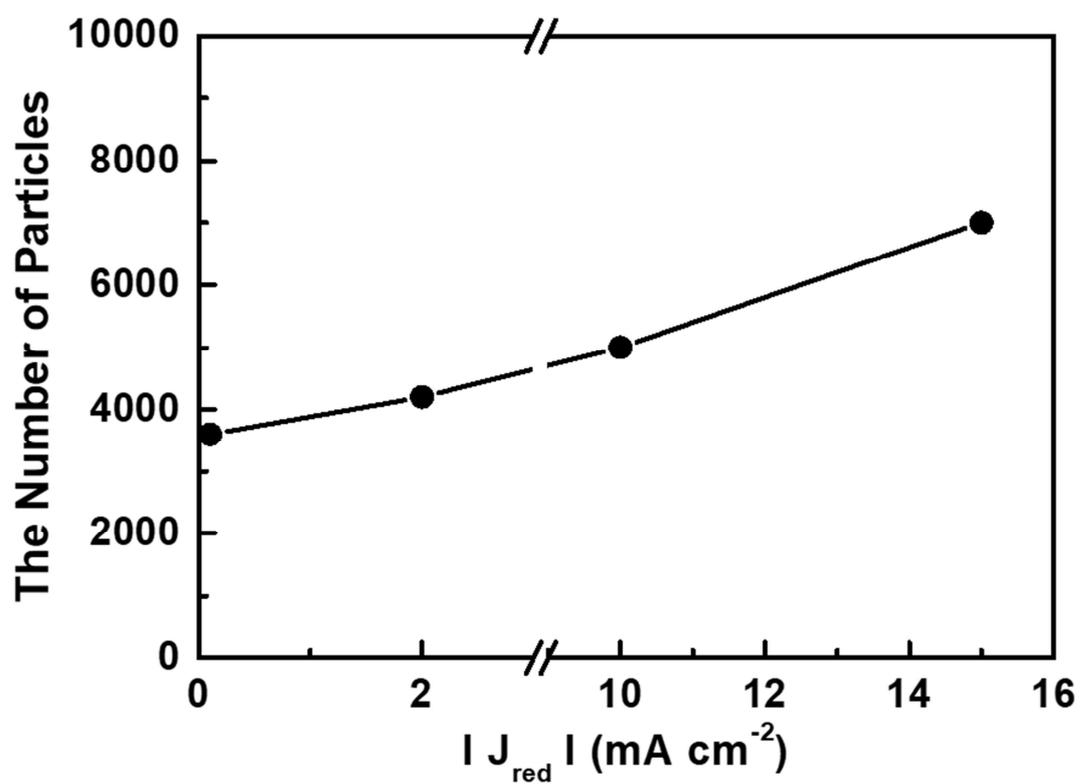
<sup>‡</sup>K.M.B. and J.K. contributed equally to this work



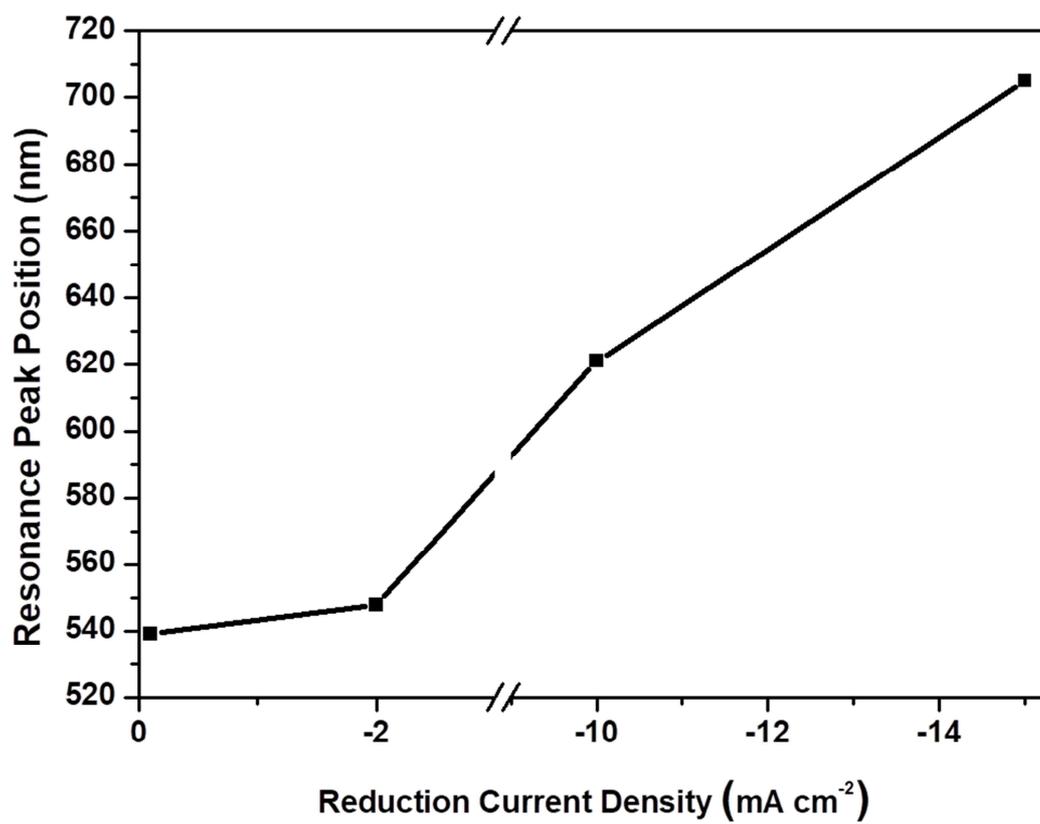
**Figure S1.** Cross-sectional SEM images of the Au nanostructures formed with the control of reduction current density. (a)  $-0.1 \text{ mA cm}^{-2}$ , (b)  $-2.0 \text{ mA cm}^{-2}$ , (c)  $-10.0 \text{ mA cm}^{-2}$ , and (d)  $-15.0 \text{ mA cm}^{-2}$ , respectively.



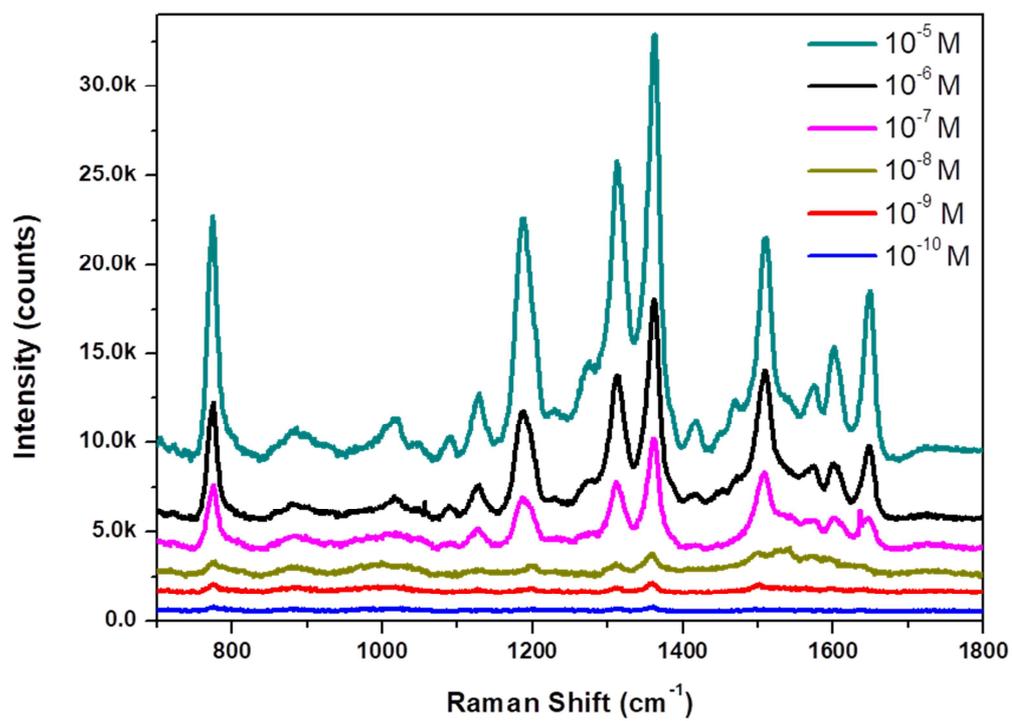
**Figure S2.** The mean gap size and the nanogap (< 10 nm) density per unit area of 1  $\mu\text{m}^2$  as a function of the reduction current density ( $J_{red}$ ).



**Figure S3.** The number of nanoparticles protruded in the 3D NPOP Au nanostructures per unit volume ( $1 \mu\text{m} \times 1 \mu\text{m} \times 0.2 \mu\text{m}$  (i.e., the height of each structure)) as a function of  $J_{red}$ .



**Figure S4.** Plot of resonance peak position depending on reduction current density.



**Figure S5.** SERS spectra of R6G molecule for concentration from  $10^{-5}$  to  $10^{-10}$  M.

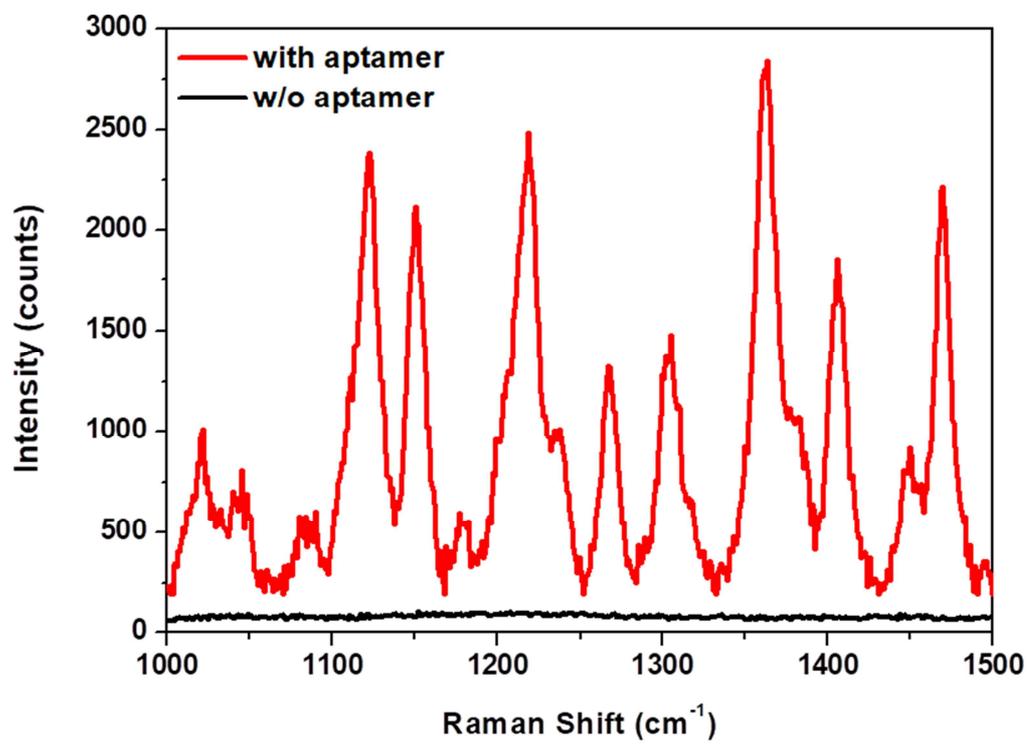
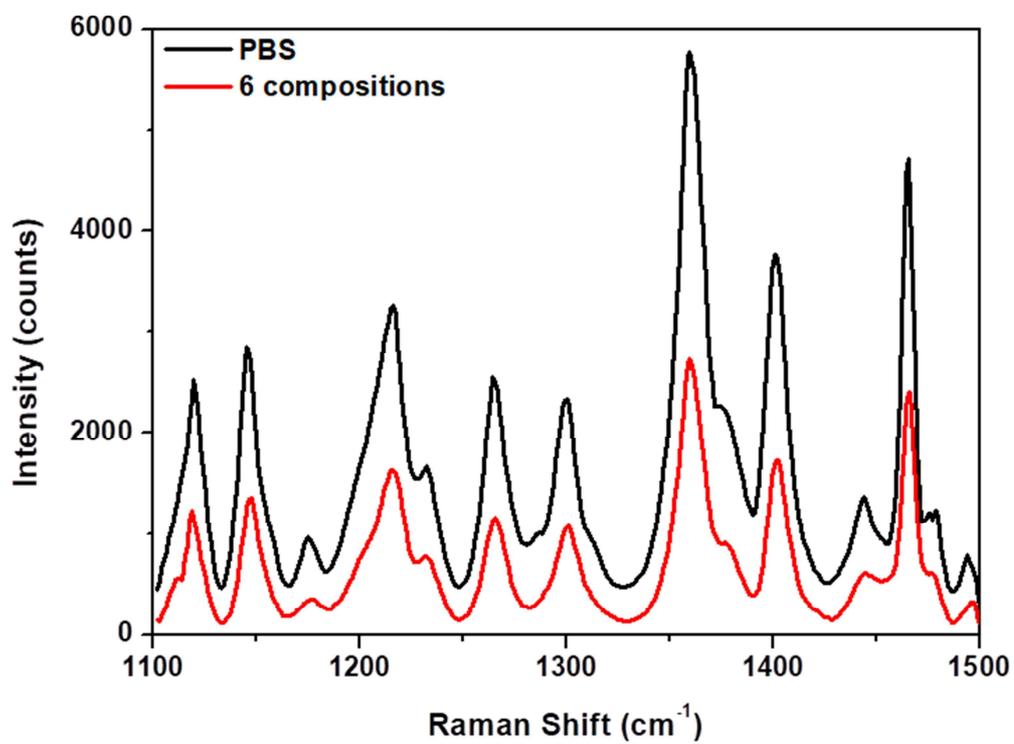
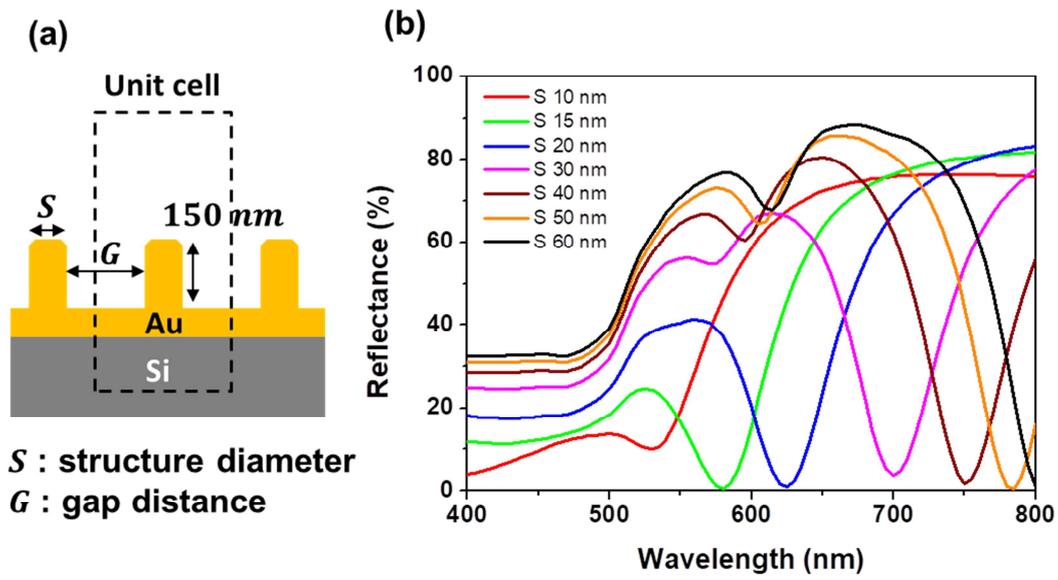


Figure S6. SERS spectra of immobilized Cy5 aptamer.



**Figure S7.** Selective detection of Hg<sup>2+</sup> ions from a mixture solution containing six different ion species.



**Figure S8.** Simulation model and reflectance spectra of the Au nanopillar samples (a) simulation model (b) reflectance spectra with changing the diameter ( $S$ ) of Au nanopillars