

Figure S1 ( a ) Zoom-in structure diagram of how Rox was labeled on the tag of DNA chain; ( b ) and ( c ) are the absorption and fluorescence spectra of the Rox labeled DNA sequence ( $P_1$ ) respectively.

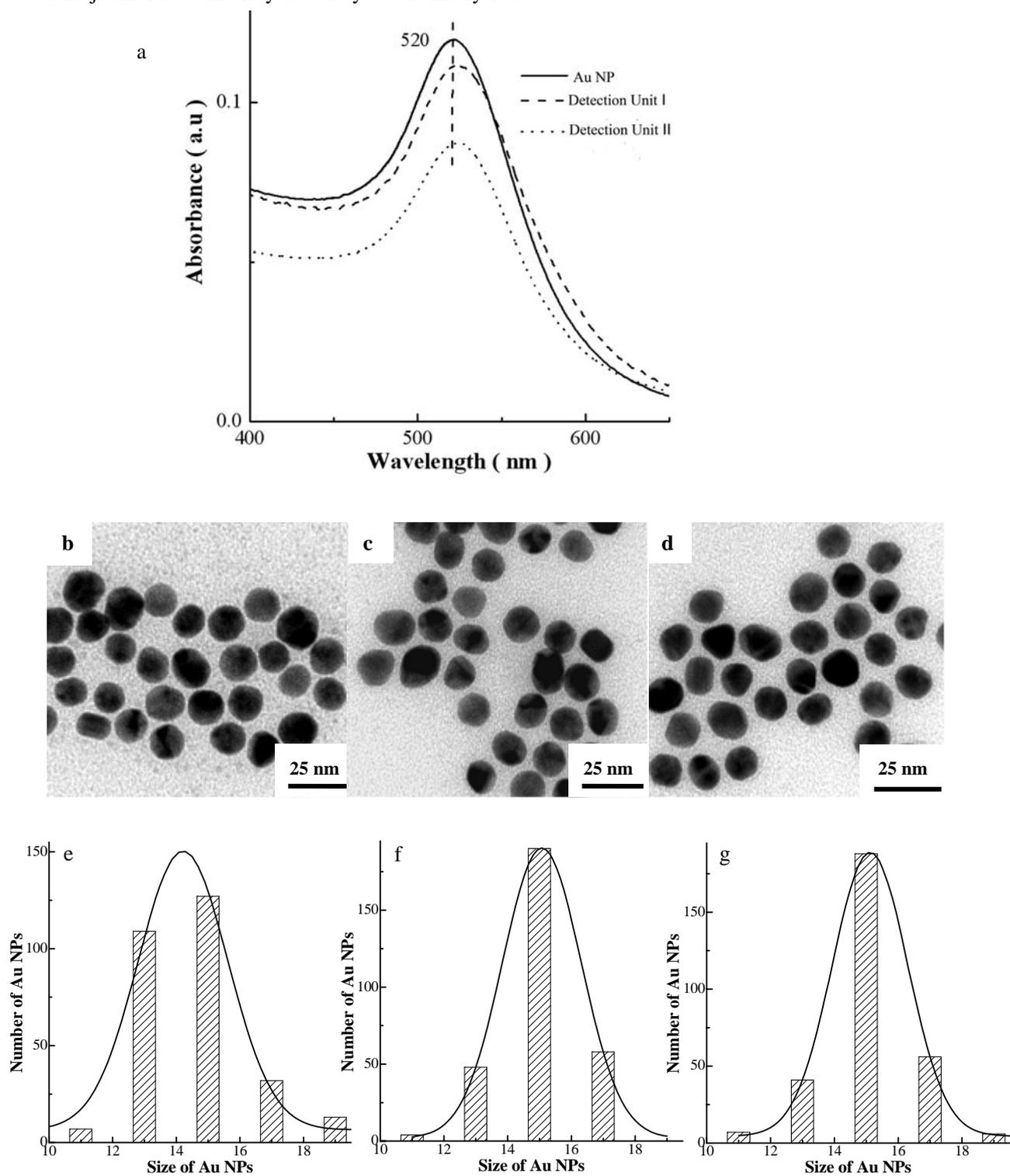
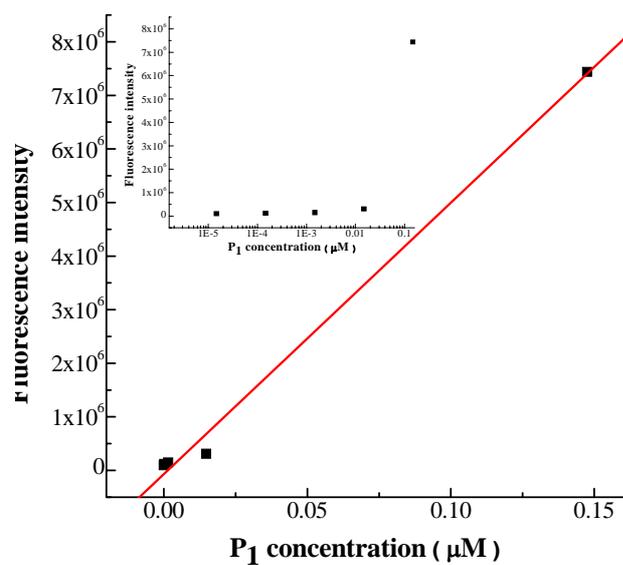
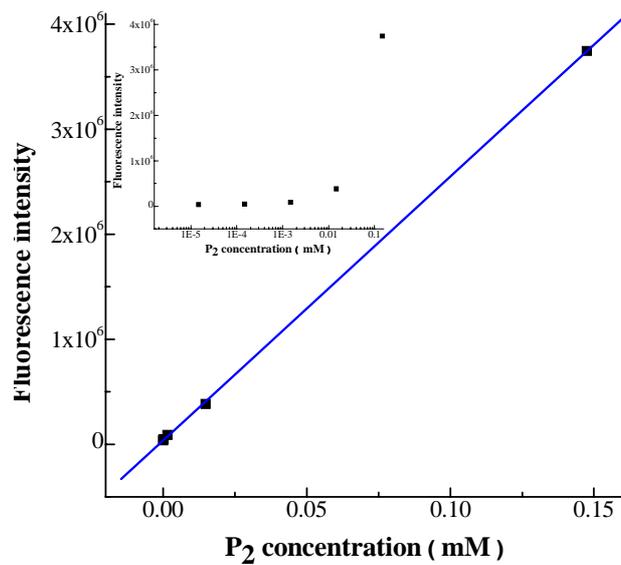


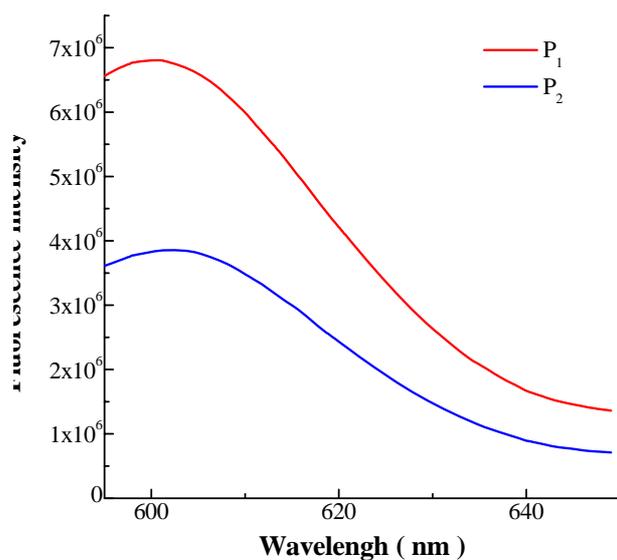
Figure S2 ( a ) UV-Vis absorption of Au NPs stabilized by citrate and further modified by P<sub>1</sub> or P<sub>2</sub>; ( b-d ) TEM images of Au NPs before modified, P<sub>1</sub>-modified and P<sub>2</sub>-modified Au NPs (Unit I and Unit II), respectively; ( e-g ) are the corresponding statistic size distributions of 300 nanoparticles, respectively.



( a )



( b )



( c )

Figure S3 ( a , b ) Concentration titrations of fluorescence intensity at 601 nm spectra of free P<sub>1</sub> and P<sub>2</sub>, respectively, and a log scaled titration was shown as the insert; (c) fluorescence spectra of P<sub>1</sub> (red) and P<sub>2</sub>(blue) after being modified on Au NPs surface

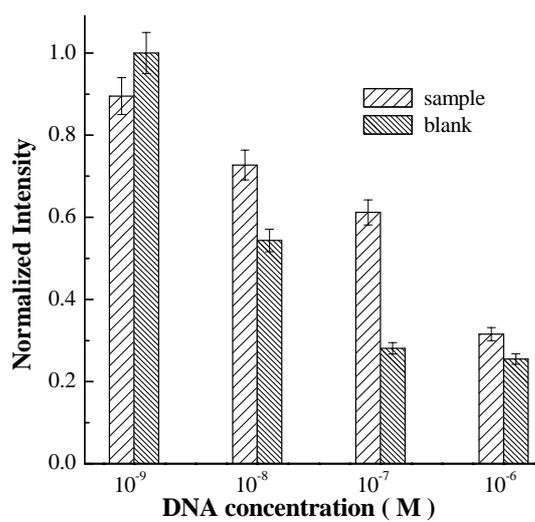


Figure S4 Normalized histogram of SERS peak intensity at 1497 cm<sup>-1</sup> with the concentration of both P<sub>0</sub> and MHT fell down from 10<sup>-6</sup> M to 10<sup>-9</sup> M.

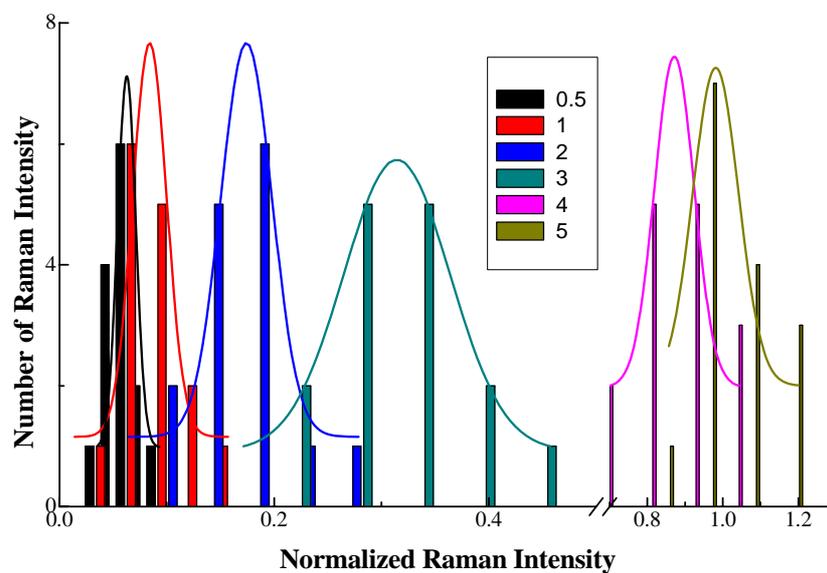


Figure S5. Histogram of the normalized difference of SERS intensity with increasing the layers of the modified Unit I and Unit II with the probe target concentration at  $10^{-14}$  M.

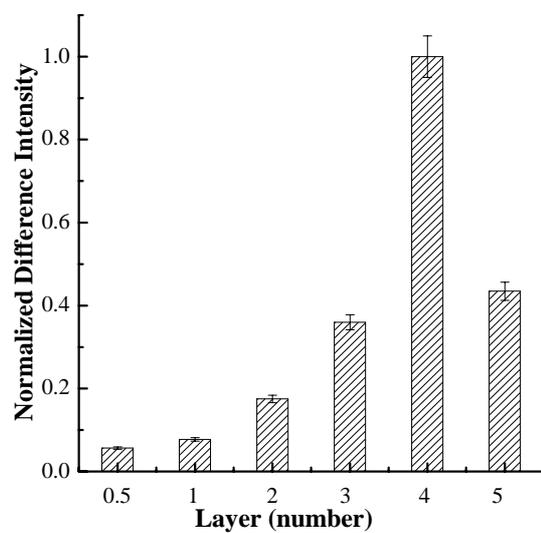


Figure S6. Histogram of the normalized difference of SERS intensity between signal and blank background with increasing the layers of the modified Unit I and Unit II with the probe target concentration at  $10^{-14}$  M.

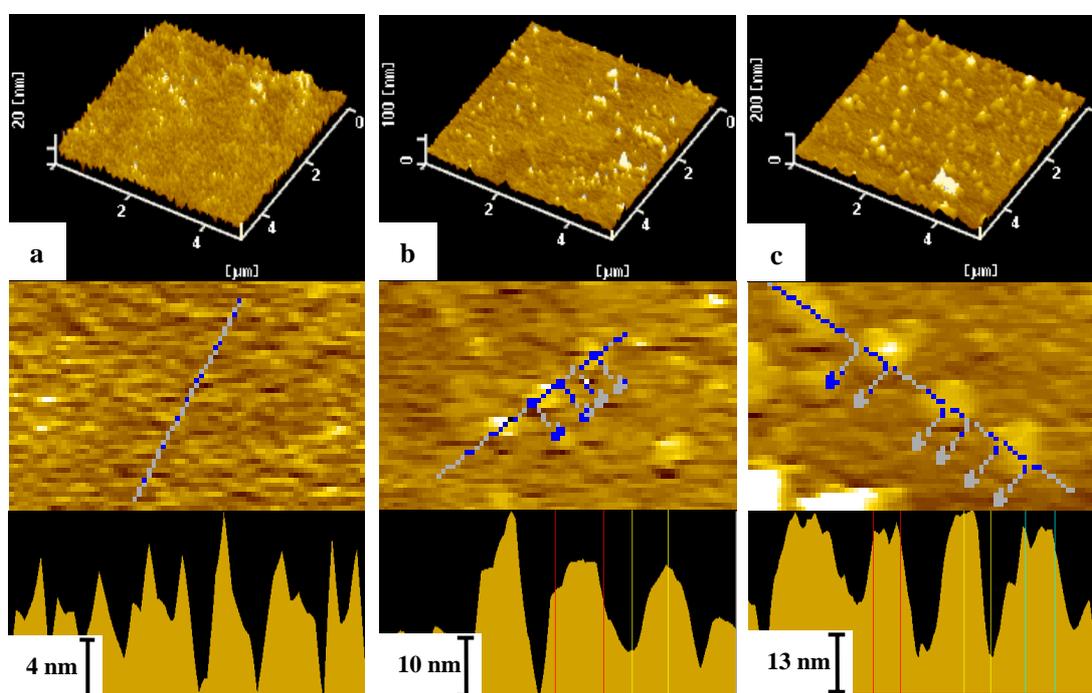


Figure S7 3D images and height of AFM images of the smooth gold surface (a); after being modified with one (Unit I + Unit II) layer; (b) and further four-layer one (c), respectively. The concentration of the target DNA strand is at  $10^{-14}$  M.

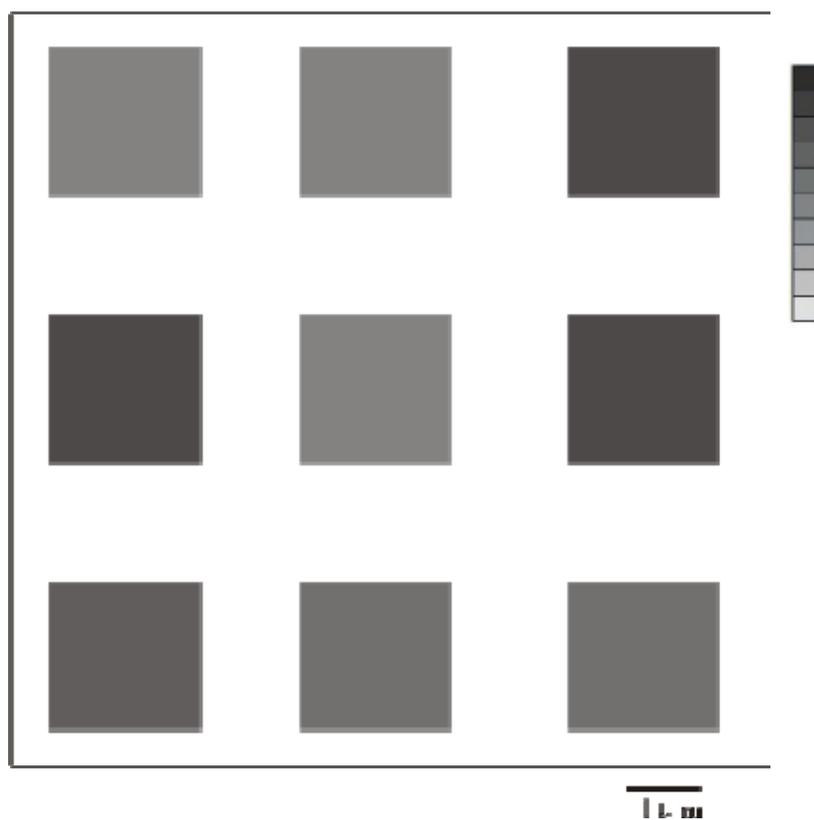


Figure S8 A 10 μm × 10 μm Mapping image from one gold surface being modified with two-(Unit I + Unit II) layer. The color reflects the variation of Raman intensities obtained from each square (2 μm × 2 μm). The concentration of the target DNA strand is at 10<sup>-13</sup> M.

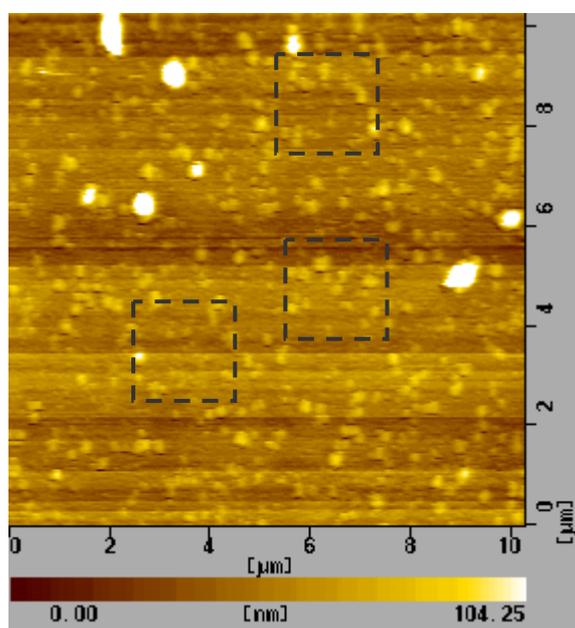


Figure S9 AFM image of the smooth gold surface after being modified with four-(Unit I + Unit II) layers in a  $10\ \mu\text{m} \times 10\ \mu\text{m}$  scale. The dashed squares display the uniform distribution of Au aggregates at the  $2\ \mu\text{m} \times 2\ \mu\text{m}$  region.