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 (P₁) respectively.
Figure S2 (a) UV-Vis absorption of Au NPs stabilized by citrate and further modified by P₁ or P₂; (b-d) TEM images of Au NPs before modified, P₁-modified and P₂-modified Au NPs (Unit I and Unit II), respectively; (e-g) are the corresponding statistic size distributions of 300 nanoparticles, respectively.
Figure S3 (a, b) Concentration titrations of fluorescence intensity at 601 nm spectra of free $P_1$ and $P_2$, respectively, and a log scaled titration was shown as the insert; (c) fluorescence spectra of $P_1$ (red) and $P_2$ (blue) after being modified on Au NPs surface.
Figure S4 Normalized histogram of SERS peak intensity at 1497 cm⁻¹ with the concentration of both \( P_0 \) and MHT fell down from \( 10^{-6} \) M to \( 10^{-9} \) 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 10⁻¹³ M.
Figure S9 AFM image of the smooth gold surface after being modified with four-(Unit I + Unit II) layers in a 10 μm × 10 μm scale. The dashed squares display the uniform distribution of Au aggregates at the 2 μm × 2 μm region.