Capillary-Force-Assisted Self-Assembly of Gold Nanoparticle into Highly Ordered Plasmonic Thin Films for Ultrasensitive SERS

Jun Dong,*[a] Haoran Wu,[a] Yi Cao,[c] Jiaxin Yuan,[c] Qingyan Han,[a] Wei Gao,[a] Chengyun Zhang,[a] Jianxia Qi,[b] and Mengtao Sun*[c]

[a.]School of Electronic Engineering, Xi'an University of Posts and Telecommunications, Xi'an, 710121, China

[b.]School of Science, Xi'an University of Posts and Telecommunications, Xi'an, 710121, China

[c.]School of Mathematics and Physics, Beijing Key Laboratory for Magneto-Photoelectrical Composite and Interface Science, University of Science and Technology Beijing, Beijing, 100083, China.

†E-mail: mengtaosun@ustb.edu.cn(M. Sun), and dongjun@xupt.edu.cn (J. Dong).

1. Simulation of electric field enhancement.

The local electromagnetic field distribution was simulated by Finite Element Method (FEM) and realized by COMSOL software. Based on the SEM results, the local electromagnetic field distribution of gold nanoparticles in the capillary excited by 532nm laser was calculated by finite element method. The diameter of AuNPs is 25 nm. **Fig.** S1(a) and (b) show the local electromagnetic field distribution of gold nanoparticles under different polarization directions (polarization along Y direction and polarization along X direction), respectively. Figure S1(c) shows the local electromagnetic field distribution of gold nanoparticles inside the capillary. It can be seen from the figure that, under the excitation of 532 nm laser, the local electromagnetic field intensity on the upper inner surface of the capillary is higher than that on the lower surface after the absorption and scattering of the upper surface and the air layer inside the capillary. The coupling effect between the nanoparticles on the inner wall of the upper and lower capillaries resulted in local enhancement of the electromagnetic field, and the "hot spot" was mainly concentrated in the gap between the nanoparticles. When target molecules are located in these "hot spots", their Raman scattering spectra intensity is greatly enhanced.

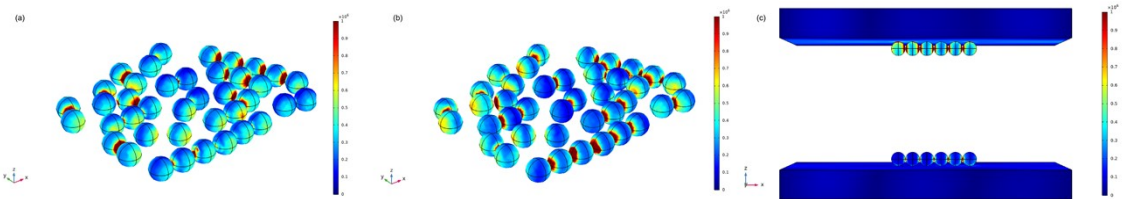

Figure S1. Under the excitation of a planar light wave with wavelength of 532 nm, the local electromagnetic field distribution of gold nanoparticles is obtained under different polarization directions (a) polarized in the Y direction and (b) polarized in the X direction. (c)The finite element method was used to simulate the local electric field distribution of gold nanoparticles in the capillary excited by 532nm laser.

2. Simple "drip self-priming" operation.

Apply two drops (10μL) of different concentrations of thiram to the orange peel in ethanol. The solvent ethanol was then completely evaporated at room temperature, mimicking the situation of a real sample. The analytes were then separated and extracted by adding another drop (10μL) of pure ethanol to the narrow skin surface of

the sample. Although the rough surface of the sample skin makes sampling or detection very difficult, due to the very small tip of the capillary used, it is easy to reach the pits in the skin, and due to the existence of capillary action, the target solution is easy to self-imbibe into the capillary, thus achieving the collection of the thiram sample of the rough fruit surface. Finally, after drying, the laser spot is focused on the inner surface of the capillary tube to collect the Raman signal of the thiram.