Supplementary Materials

Direct electron-beam patterning of transferrable plasmonic gold nanoparticles using $HAuCl_4$/PVP composite resist

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Figure S1 to S6.

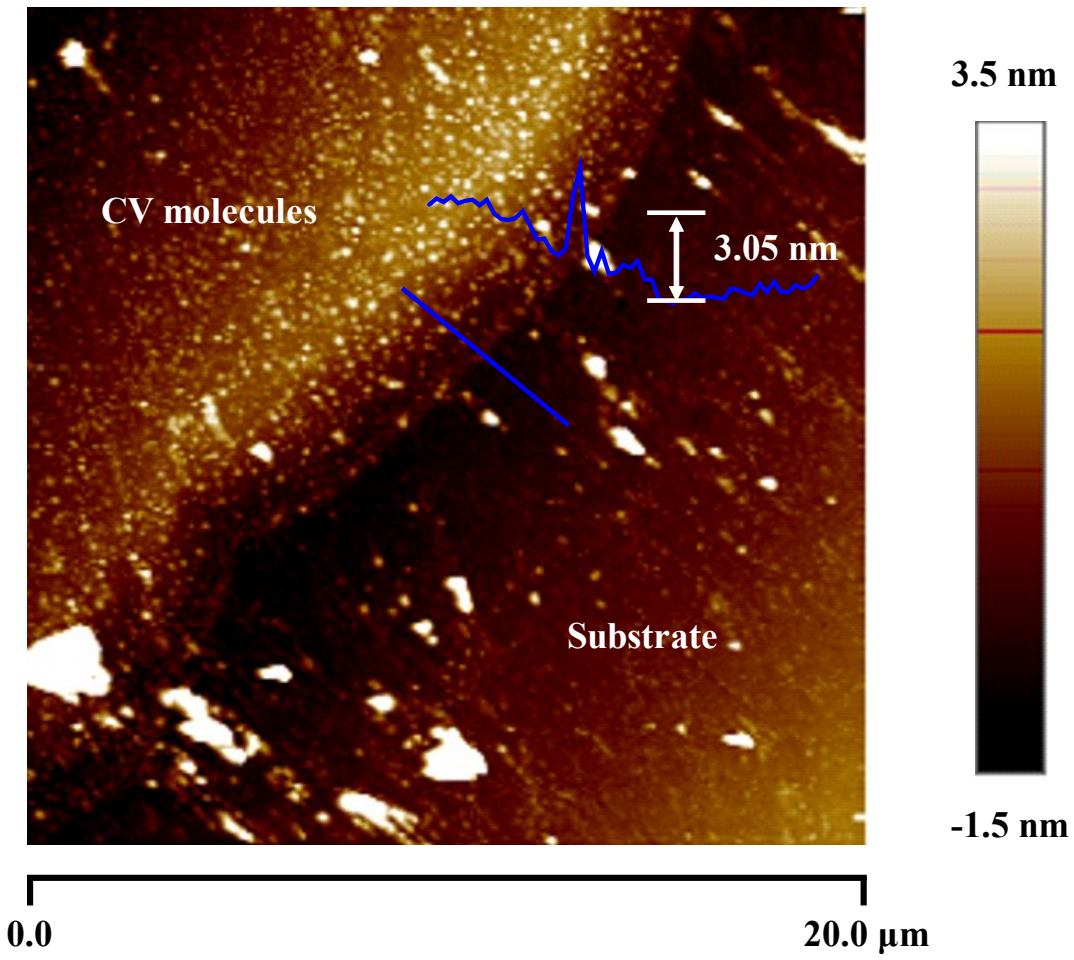

Fig. S1 (a) Atomic force microscopy test of sample in boundary to confirm the thickness of modified molecule.

In order to confirm a monolayer of CV molecules absorbed on the Au film, we just keep half of the Au substrate in CV solution at the required concentration for 2h. Then the sample was rinsed several times with deionized water and blow-dried using the N_2 stream. After that, we use the line scan over the boundary to measure the values of height variations. The ~ 3 nm thickness CV molecules was adsorbed on the surface of Au substrate in comparison to pure Au substrate (as shown in Fig. S1), which means that a monolayer CV molecular was obtained on the surface of 100-nm Au film substrate.

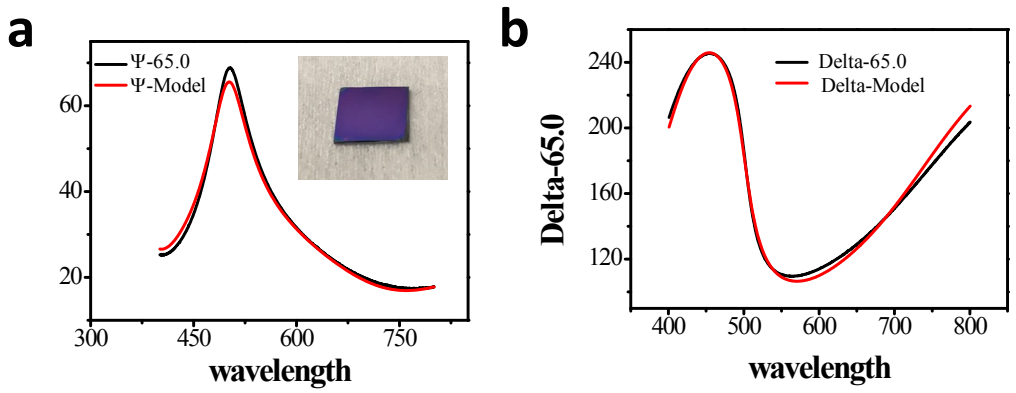

Fig. S2 (a) ψ and (b) Δ delta are ellipsometry test for a $HAuCl_4$/PVP composite resist film, in which black line is experimental data and red line is model data.

In order to confirm the thickness of initial $HAuCl_4$/PVP composite resist film, ellipsometer was used to measurement the values of ψ and delta of light reflected from the sample surface. The thickness of initial film is acquired by further fitting of curves (as shown in Fig. S2a and Fig. S2b). The thickness of initial film is ~110 nm.

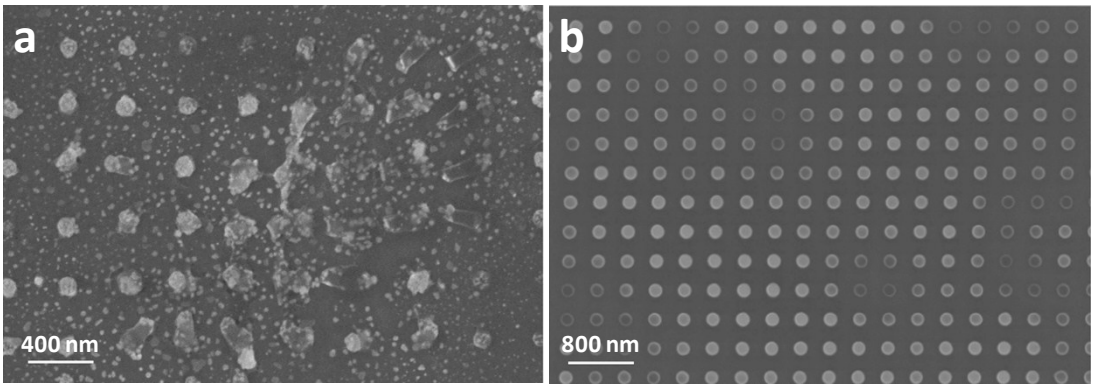

Fig. S3 SEM images of $HAuCl_4$/PVP composite with different concentration ratio. (a) Mixed 100 mM $HAuCl_4$ solution and PVP solution by 1:1 volume ratio. (b) Mixed 10 mM $AuCl_4$ solution and PVP solution by 1:1 volume ratio.

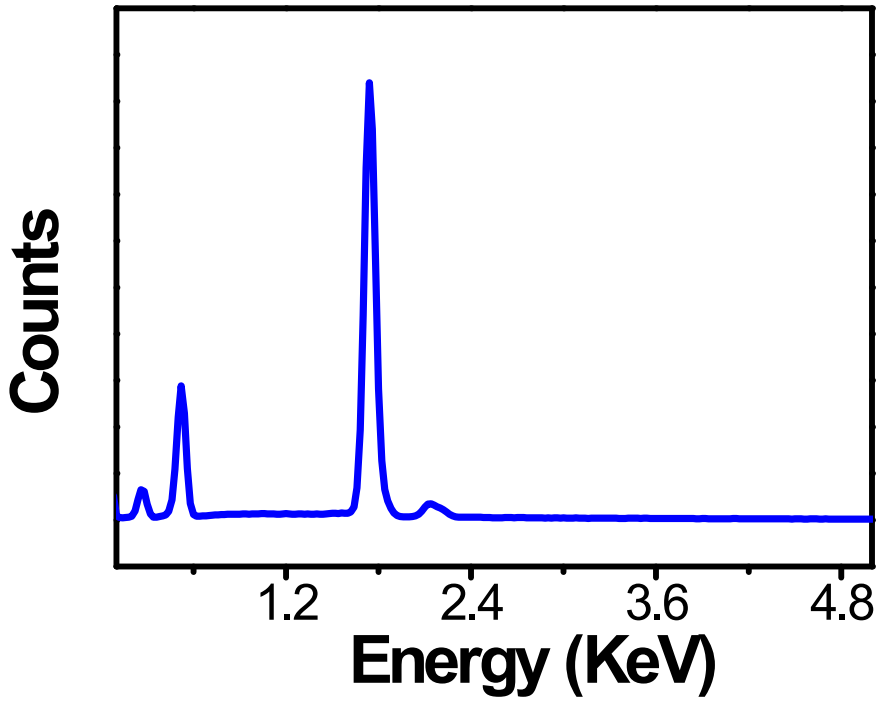

Fig. S4 EDX spectrum of Au particle after EBL and annealing processes.

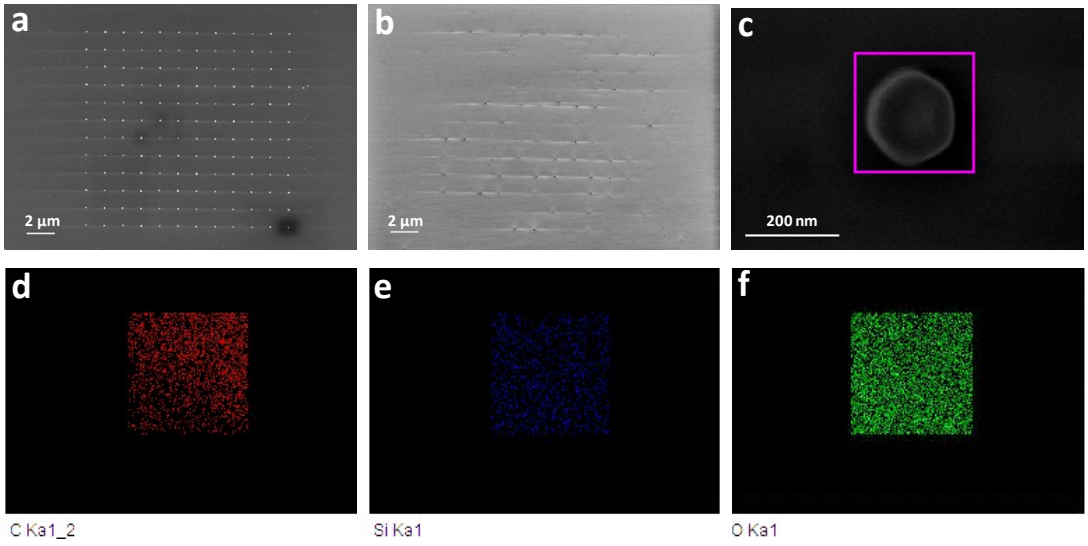

Fig. S5 SEM images and EDX mapping of sample before and after peeling off Au nanoparticles from SiO_2 substrate. (a) Au nanoparticle array on SiO_2 substrate. (b) The SiO_2 substrate morphology after peeling off Au nanoparticles form the substrate. (c-f) The EDX mapping of residual substance on substrate, which indicates that just few organic carbon element is existed

and all Au particle are peeled off.

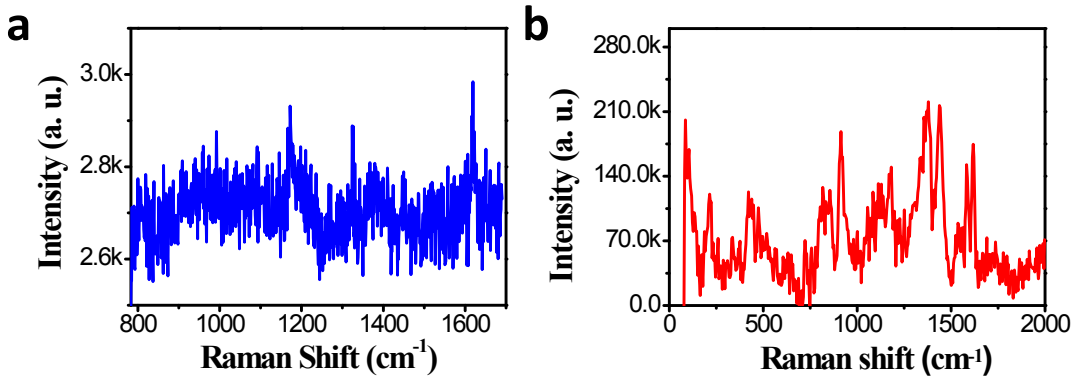

Fig. S6 (a,b) stand for the Raman intensities of CV molecules from capillary sample and SERS substrate, respectively.

In our work, the SERS EF was calculated according to the following formula:

$$EF = \frac{I_{SERS} N_{bulk}}{I_{bulk} N_{SERS}}$$

Where I_{SERS} and I_{bulk} are the Raman intensities of crystal violet (CV) molecules obtained from SERS substrate and capillary sample in the same laser and integration conditions (as shown in Fig. S6), respectively.

N_{SERS} and N_{bulk} corresponding to the number of detected molecules on SERS substrate and reference sample. Here, we choose the Raman peak at 1172 cm^{-1} for calculating Raman enhanced factor. N_{SERS} can be acquired based on following formula :

$$N_{SERS} = \frac{\pi r^2}{\sigma}$$

where "r" and "σ" is the radius of laser spot (~ 1 μm) and single CV molecular (~4 nm^2) (*e. g. A. Kudelski et al. Chem. Phys. Lett., 2005, 414, 271*).

N_{bulk} can be acquired based on formula :

$$N_{bulk} = \pi r^2 h c N_A$$

where "r" is still the radius of laser spot, "h" is the depth of laser illuminate capillary sample (~ 11 μm) (e. g. W. Lin et al. Appl. Phys. A: Mater. Sci. Process. 2010, 101, 185.), "c" is the concentration of sample solution (10^{-2} M), "N_A" is Avogadro's constant.

So we calculated the Raman enhanced factor is about 9.8×10^5.