Supporting Information



Fig. S1 (A) (B) SEM image of Ag nanoparticles with HMD and (C) (D) without HMD. (B) and (D) are magnified images of the white squares shown in (A) and (C), respectively. The white arrows indicate the aggregated nanoparticles. (E) Histogram of the number of nanoparticles in the aggregates.



Fig. S2 (A) SERS intensity map at 1590 cm<sup>-1</sup> with different pH condition at same place. (B) Individual SERS spectra with different pH at allowed in (A).



Fig. S3 One of the 2D Gaussian fitting of the darkfield image of the nano assembly. Each graph corresponds to the line profile of the indicated position by the triangles and the result of the gaussian fitting. The spatial accuracy was calculated by the shown equation (from Ref. S-1) where N is the number of photons (~250), s is the full width of half-maximum of the spot (~340 nm), and a is the pixel size on the image (70 nm). The position accuracy is ~22 nm.

## Reference

S-1 X. Nan, P. A. Sims, P. Chen and X. S. Xie, J. Phys. Chem. B, 2005, 109, 24220–24224