Supporting information: Dynamic plasmonic nano-traps for single molecule surface-enhanced Raman scattering

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Excitation of the SPP field

When the radial polarized (RP) 532-nm laser beam is focused by a high-numerical-aperture objective lens into a Kretschmann coupling-type glass plate coated with a thin Ag film, surface plasmon polaritons (SPPs) are excited at the surface plasmon resonance (SPR) angle. Figure S1a shows the reflected light at the back focal plane of the objective lens, the excited SPP results in a near zero reflection at the excitation positions, as indicated by the red arrow. Similarly, the result for the RP 1070-nm laser beam diverging into the Kretschmann structure, and Fig. S1b is the corresponding divergent ring-shaped SPP field. Since the excited position of the SPP is controlled by setting the position of the Ag film and divergence angle of the incident beam, a combined dual plasmonic field with inner focused and outer divergent SPPs is produced. Moreover, the intensity of the excited SPP field is regulated independently by the exciting laser beam.

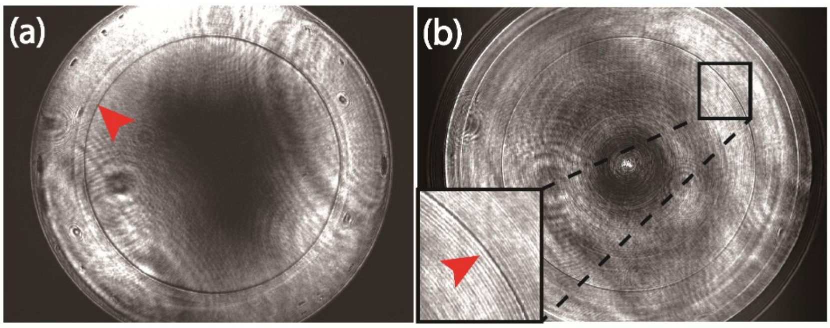

Fig. S1. Images of the excited SPP fields produced by the focused and divergent laser beams. (A) and (B) are reflected images on the back focal plane of the objective lens showing the distribution of the corresponding SPP fields. The dark ring corresponds to low reflectance, resulting from SPP excitation by the laser beam.

Distribution of the electric field for particles with different numbers and diameters

The energy is mostly localized in the narrow nano-gap between the particle and the film, which is important in furthering SERS research. Because of limitations in experimental conditions, only particles of radius 100 nm were studied in experiments. However, relatively smaller particles will provide better performance based on calculated EF of particles with different diameters (Fig. S2). The full width at half maximum of the plasmonic virtual probe and the electronic field compressed in the plasmonic nano-gap of single-particle–film structure, with NPs of various diameters, were compared (Fig. S3). The results indicate that the energy is highly focused in the nanosized plasmonic gap, and better results may be achieved using particles of optimized size.

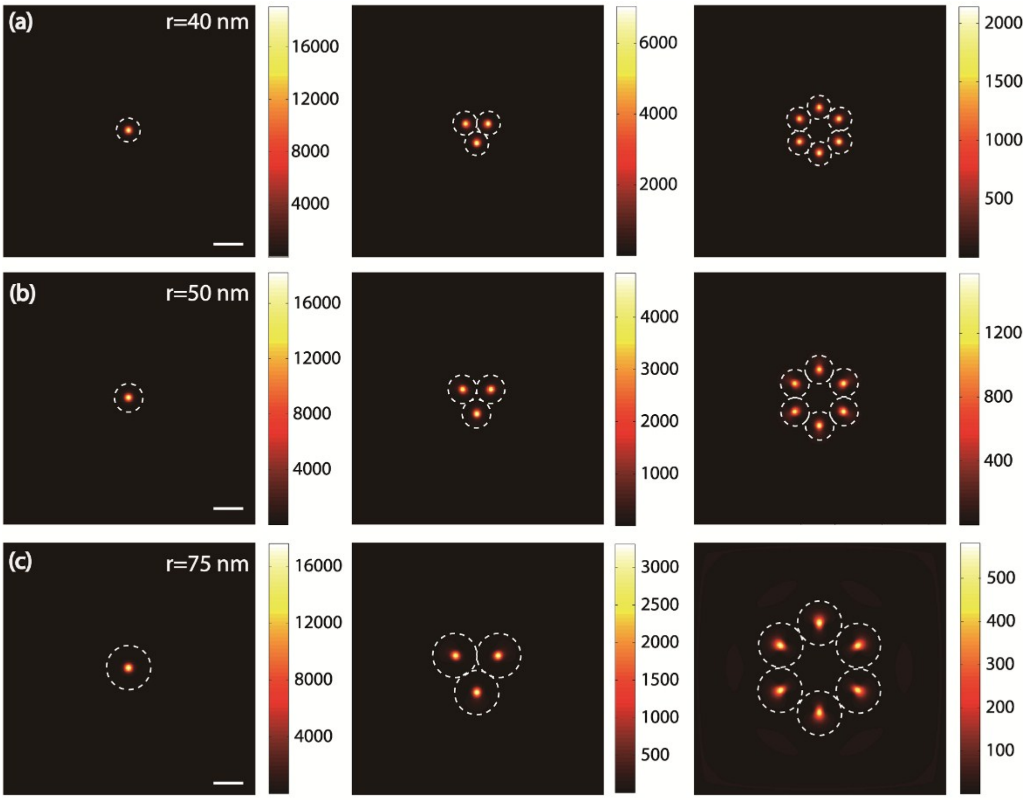

Fig. S2. Dependence of intensity of the electronic field on the radius of Au nanoparticles. (A) *r*=40 nm, (B) *r*=50 nm, (C) *r*=75 nm. The relatively smaller particles exhibit better performance, i.e. *r*=40 nm in these calculations. Other parameters are in accordance with experimental conditions. The scale bar is 100 nm.

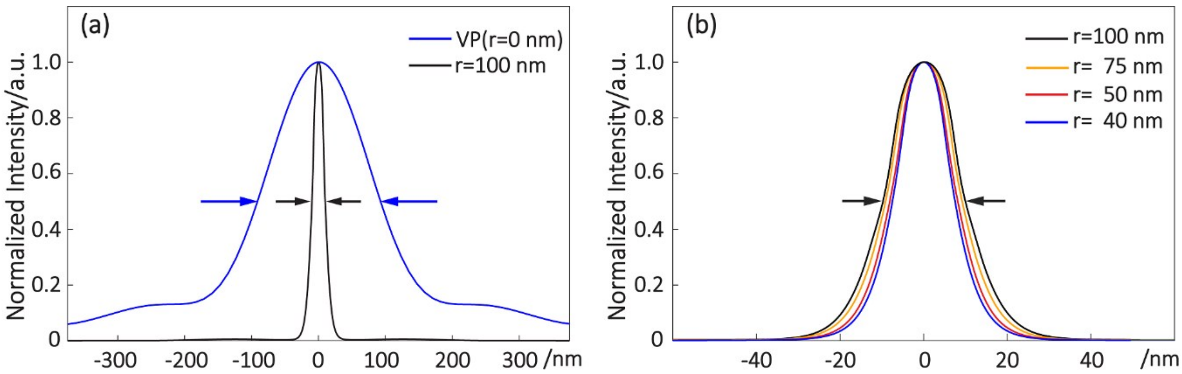

Fig. S3. Dependence of full width at half maximum (FWHM) of the electronic field on radius of Au nanoparticles. (A) FWHM of the compressed electronic field in the nano-gap is much smaller than the plasmonic virtual probe. (B) FWHM of the electronic field in the nano-gaps with particles of various sizes particle trapped on the film. All other parameters are in accordance with the experimental conditions. The results indicate that the energy is highly focused in the nanosized plasmonic gap. The suggestive super-resolution can be optimized by choosing appropriate nanoparticles.