Supporting Information for

Nanoparticle-enhanced Plasma Discharge Using Nanosecond High Voltage Pulses

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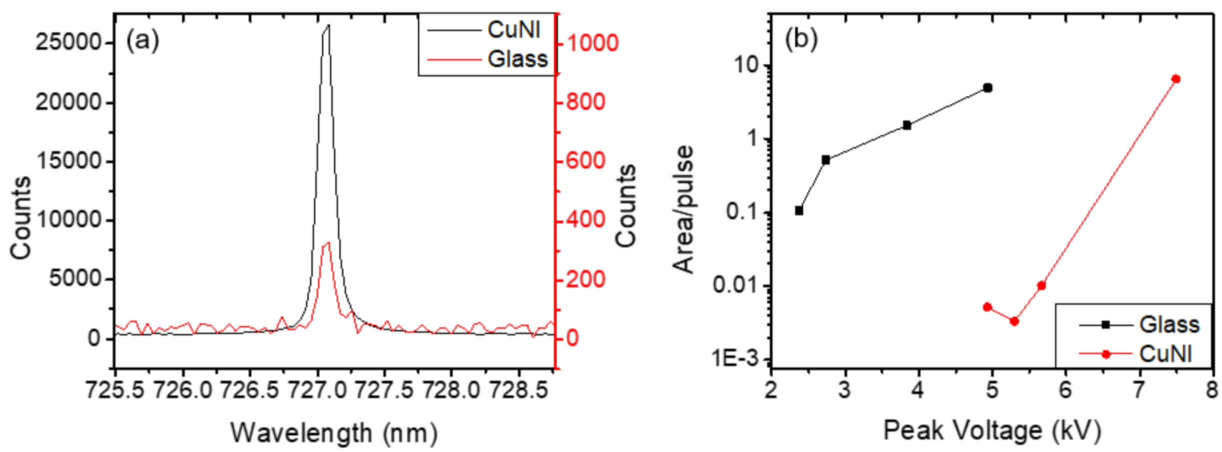

Figure S1. Enhancement of Ar plasma emission with Cu nanoparticles. (a) emission of Ar on different substrate with 5 kV peak voltage (b) emission intensity changing with peak voltage on different substrate. A 1000x enhancement can be observed near 5 kV.

Figure S2 shows a photograph of our experimental setup in which the high voltage cable from our nanosecond high voltage pulse generator is attached to a glass slide-based reactor, which consists of two parallel copper electrodes separated by an approximately 5mm gap. The gas flow cell is constructed by sandwiching four glass slides together with 3D-printed manifold as shown in Figure S2b. This flow cell has a low profile and fits easily underneath our high numerical aperture microscope objective lens, enabling plasma emission spectra in the visible wavelength range to be obtained with high collection efficiency, as illustrated in Figure S2c.

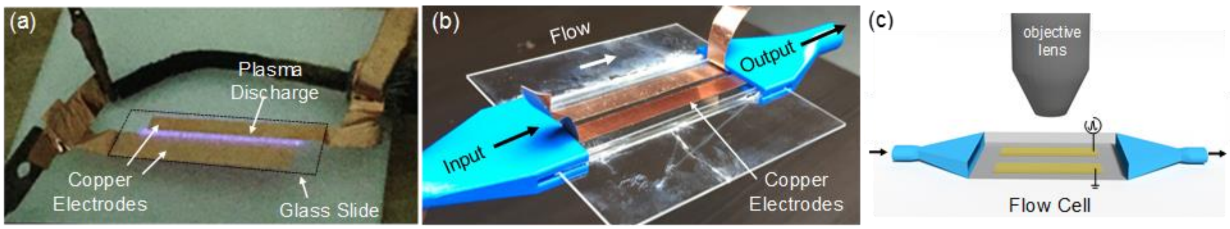

Figure S2. Photographs of (a) the plasma discharge across a 5mm gap on a glass slide and (b,c) glass-slide flow cell.

In our electrostatic simulations, we extracted the nanoparticle geometries (i.e., size and shape) from an HRTEM image (Figure 2a) and imported these into the COMSOL Multiphysics simulation package. The nanoparticle film had an area of 550nm x 550nm and thickness of 5nm. A built-in material model for Au was used for the nanoparticles. Two metal electrodes are placed in-plane with the nanoparticle film with a separation of 725nm. The total simulation volume was set to be 1250 x 1000 x 800nm with air surrounding the particle film and electrodes. A schematic diagram illustrating the simulation cross section is given in Figure S3. We performed a quasi-static simulation to determine the electric field profile using the electric current physics AC/DC module in COMSOL. Here, a 100MHz sinusoidal signal of unit amplitude was applied across the nanoparticles, and the electric field was monitored. In the low-frequency region, ranging from radiofrequency waves to infrared wavelengths, the amount of metallic losses is very low, and metals thus tend to behave like perfect reflectors[1]. As a result, no field penetration was assumed inside the nanoparticles. Floating potential boundaries were used at the nanoparticle surfaces and the electric field was calculated in the surrounding air medium. The field enhancement at the surface of the nanoparticle is normalized to the electric field at the same position without the nanoparticle film.

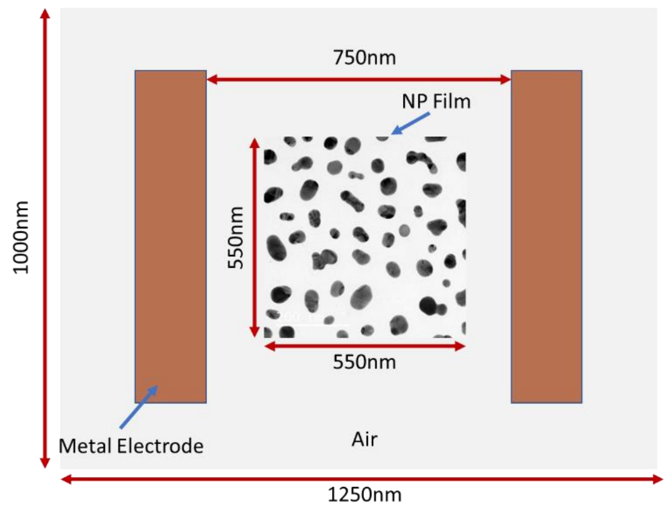

Figure S3. A schematic diagram of the structure of our simulated sample.

Figure S4 shows the nanoparticle size distributions of Au and Pt nanoisland films measured by atomic force microscopy (AFM).

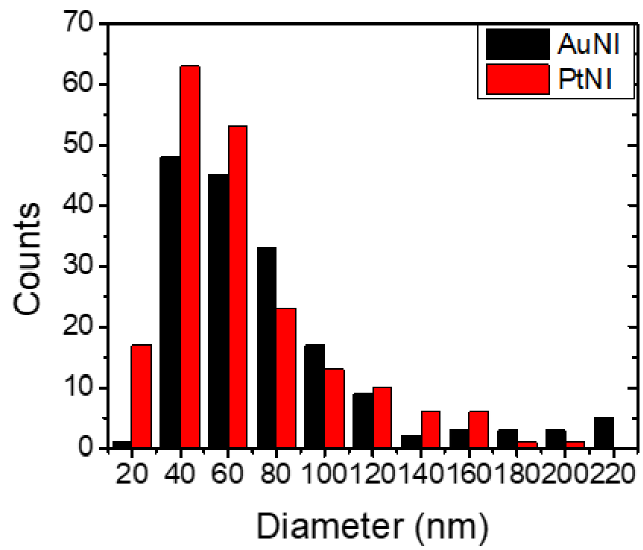

Figure S4. Distribution of particle sizes

For COMSOL simulations, we use a non-uniform tetrahedral mesh with a minimum element size of 0.25nm is used. The mesh grid for the particles is shown in the figure below.

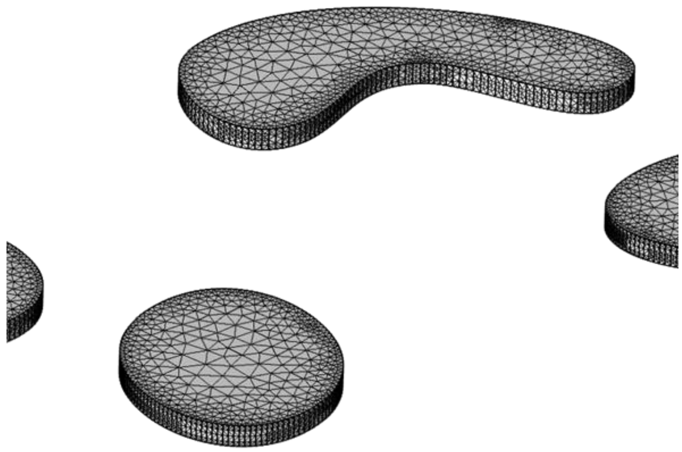

Figure S5. Non-uniform tetrahedral mesh use in our COMSOL simulations.

In order to verify that our mesh size is sufficient to accurately predict the underlying physics associated with these metal nanoparticles, simulations have been performed on a 5nm radius disk with 5nm thickness by varying the minimum element size in the non-uniform tetrahedral mesh from 0.1nm to 5nm to verify the convergence below 1nm.

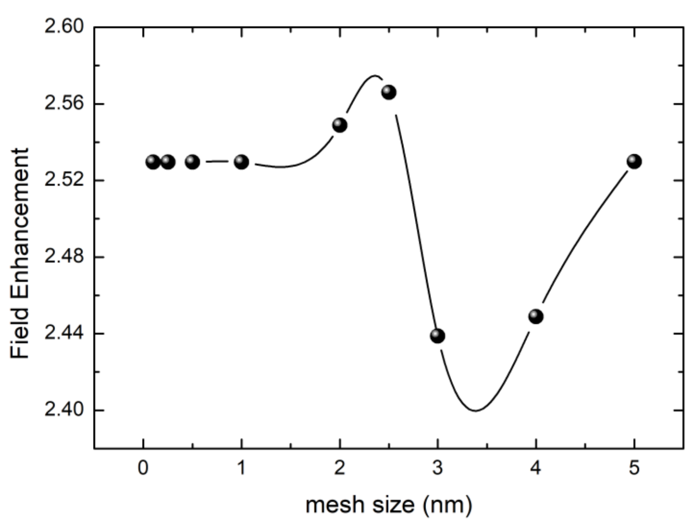

Figure S6. Mesh convergence test for 5nm thick circular discs of radius 5nm.

We define electric field enhancement as the ratio of the absolute values of simulated local electric field with nanoislands to the electric field without nanoislands (on bare glass slide).

For COMSOL simulations,

$$E_{enhancement}(x,y) = \frac{|E|_{with\ particle}(x,y)}{|E|_{without\ particle}(x,y)}$$

For FDTD simulations,

$$E_{enhancement}(x,y) = \frac{|E|^2}{|E_0|^2}$$

where Eo is the electric field of incident wave and E is the electric field with the nanoislands.

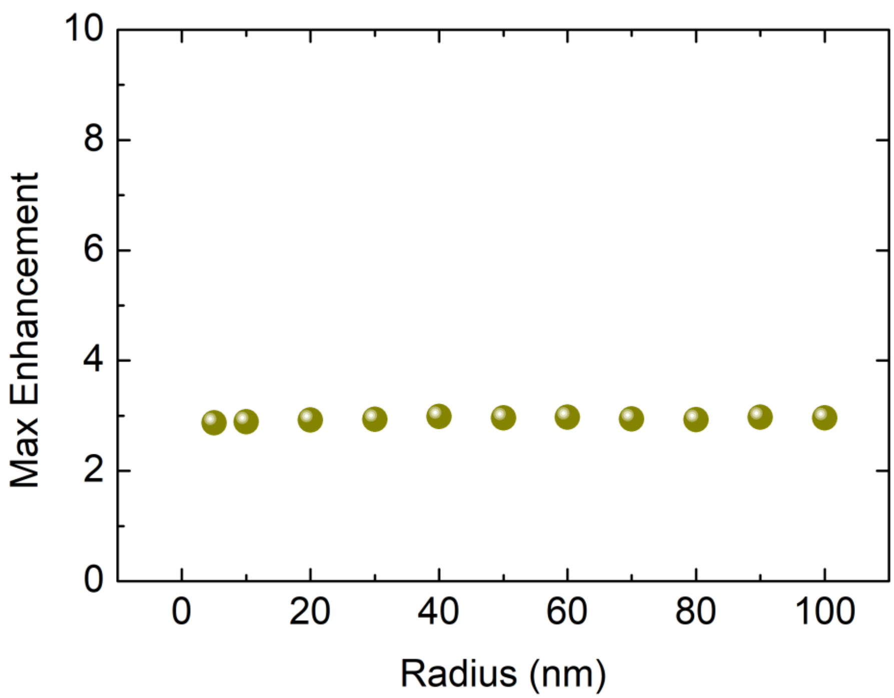

Figure S7. Maximum electric field enhancement plotted as a function of particle size.

1. Lourtioz, J.-M., Benisty, H., Berger, V., Gerard, J.-M., Maystre, D., Tchelnokov, A., *Photonic Crystals Towards Nanoscale Photonic Devices*. 2008: Springer.