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Electronic Supplementary Information

Facile fabrication of homogeneous and gradient plasmonic arrays with tunable optical properties *via* thermally regulated surface charge density

Shunsheng Ye,[a] Hongyu Wang,[a] Hongyang Su,[a] Lingxia Chang,[a] Shuli Wang,[a] Xuemin Zhang,[b] Junhu Zhang,*[a] and Bai Yang[a]

[a] *State Key Lab of Supramolecular Structure and Materials, College of Chemistry, Jilin University, Changchun 130012, P. R. China. *E-mail: zjh@jlu.edu.cn*

[b] *Department of Chemistry, College of Sciences, Northeastern University, Shenyang 110004, P. R. China*

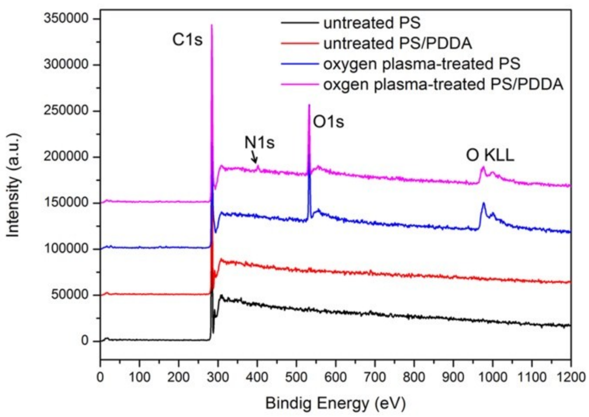

Fig. S1 XPS survey scans of untreated PS before (black line) and after (red line) PDDA adsorption. XPS survey scans of oxygen plasma-treated PS before (blue line) and after (magenta line) PDDA adsorption.

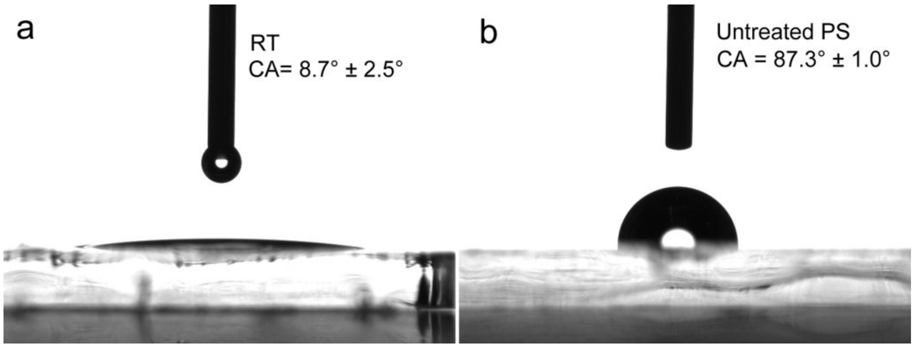

Fig. S2 Water contact angle measurements on (a) an oxygen plasma-modified PS film without thermal annealing and (b) an untreated PS film.

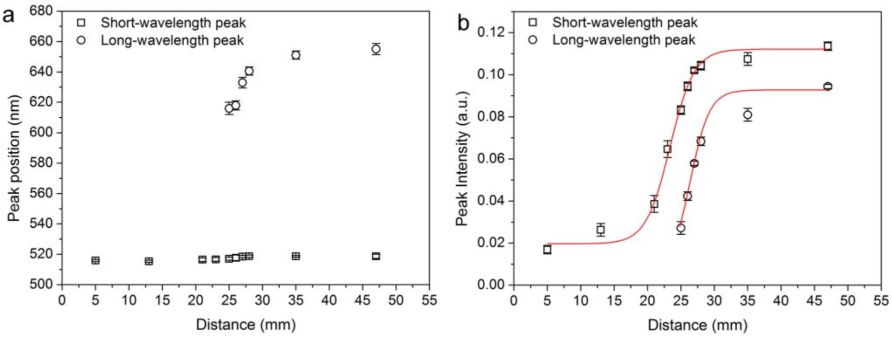

Fig. S3 (a) Dependence of wavelengths of the short- and long-wavelength resonance bands on distance. (b) Intensities of the two resonance bands plotted against distance. The squares are fitted with a sigmoidal curve and the circles are fitted with part of a sigmoidal curve.

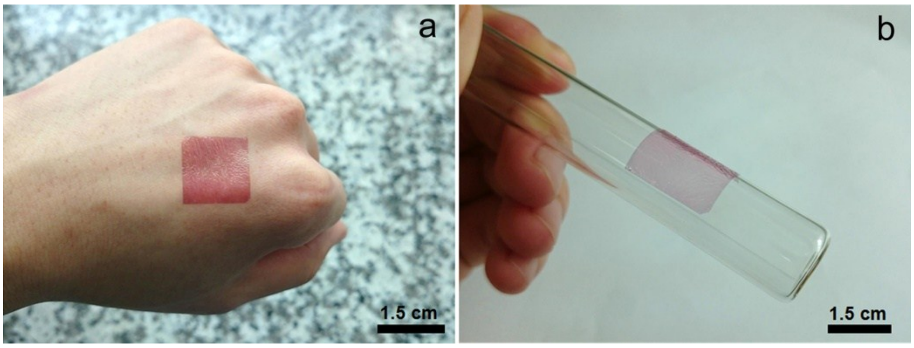

Fig. S4 Au nanoparticle-modified PS films transferred onto (a) skin and (b) a curved surface.

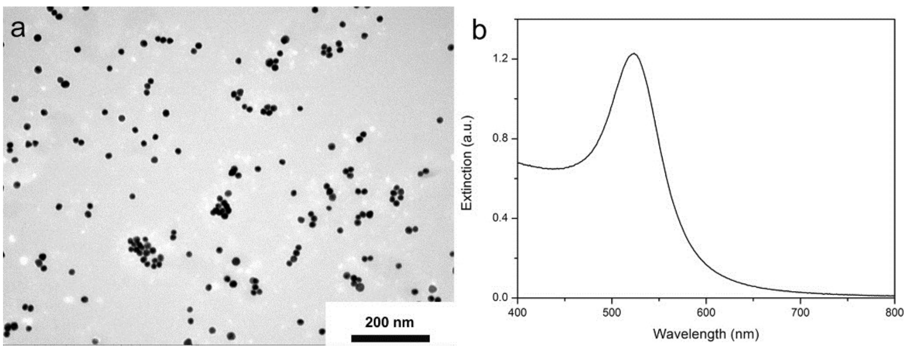

Fig. S5 (a) TEM image of the Au nanoparticles synthesized *via* the seeded growth method. The mean size is about 18 nm. (b) UV−vis extinction spectrum of the Au nanoparticle sol.

Calculation of Enhancement Factor

The enhancement factor (EF) was calculated based on the following equation:

$$EF = \frac{I_{SERS}}{I_{bulk}} \times \frac{N_{bulk}}{N_{SERS}} \quad (1)$$

where I_{SERS} and I_{bulk} are the intensity of characteristic bands in SERS and normal Raman spectra(see Figure S6), respectively; N_{bulk} and N_{SERS} are the number of detected molecules in the laser spot volume in normal and SERS measurements, respectively. N_{bulk} can be estimated as follows:

$$N_{bulk} = \frac{\rho A h}{M} N_A \times 90\% \quad (2)$$

where ρ is the density of MB powder (1.14 g/cm^3), A is the area of laser spot (ca. 1μm^2), h is the penetration depth of the laser (ca. 1 μm), M is the molar mass of MB (319.86 g/mol), and N_A is the Avogadro constant. N_{SERS} is calculated from the following equation:

$$N_{SERS} = \frac{N_d A A_N \gamma}{\sigma} \quad (3)$$

where N_d is the number density of our 2D Au nanoparticle arrays, A_N is the area of a single Au nanoparticle (calculated as ca. 452 nm^2), γ is the coverage of a MB monolayer on one Au nanoparticle (assumed to be 30%), and σ is the area occupied by one MB molecule (0.8 nm^2)[1], respectively. By substituting equation 2 and 3 into equation 1, we get:

$$EF = \frac{I_{SERS}}{I_{bulk}} \times \frac{3\rho h N_A \sigma}{M N_d A_N} = \frac{I_{SERS}}{I_{bulk} N_d} \times 1.14 \times 10^7\ \mu m^{-2}$$

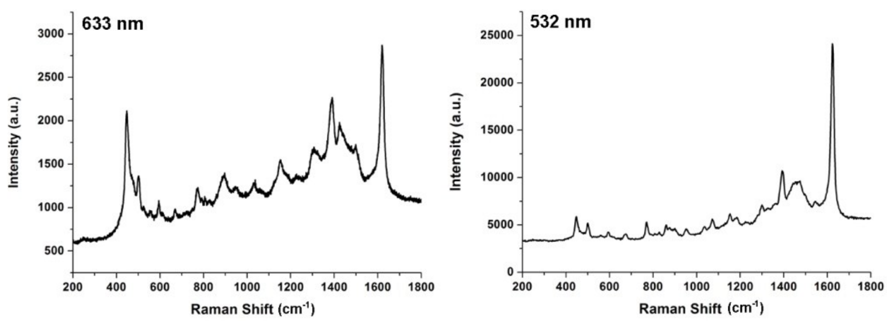

Fig. S6 Normal Raman spectra of MB excited by 633 nm and 532 nm laser lines.

References

1 G. Laurent, N. Félidj, J. Aubard, G. Lévi, *Phys. Rev., B,* 2005, **71**, 045430.