Supporting Information

Direct Assembly of Large Area Nanoparticle Arrays

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Structural Characterization of Gold NSs and Gold NRs

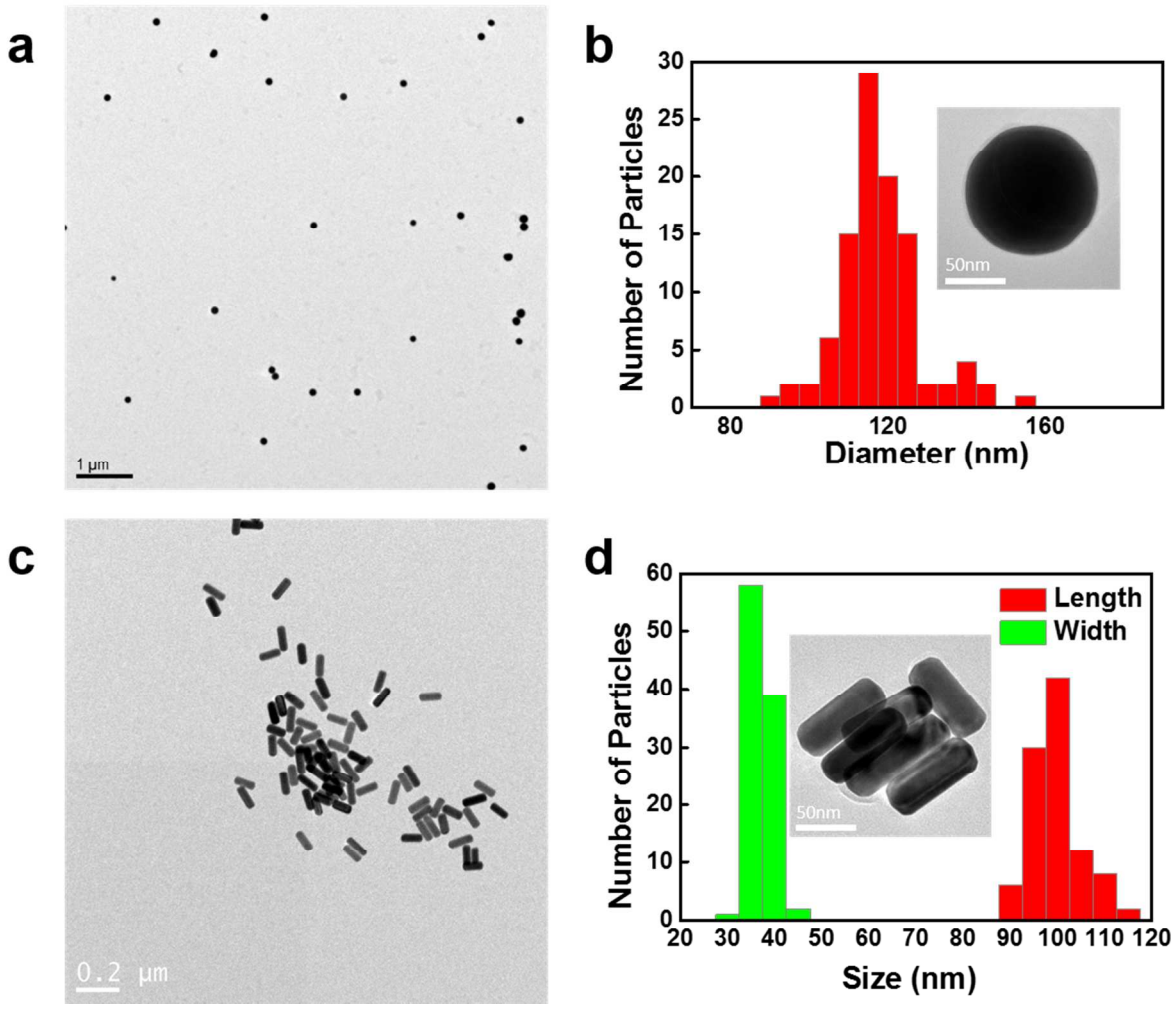

Figure S1. The transmission electron microscope images for (a) gold NSs with (b) 110 nm diameter distribution and (c) gold NRs with (d) 100 nm in length and 40 nm in width distribution.

The atomic force microscopy images for box and rod templates.

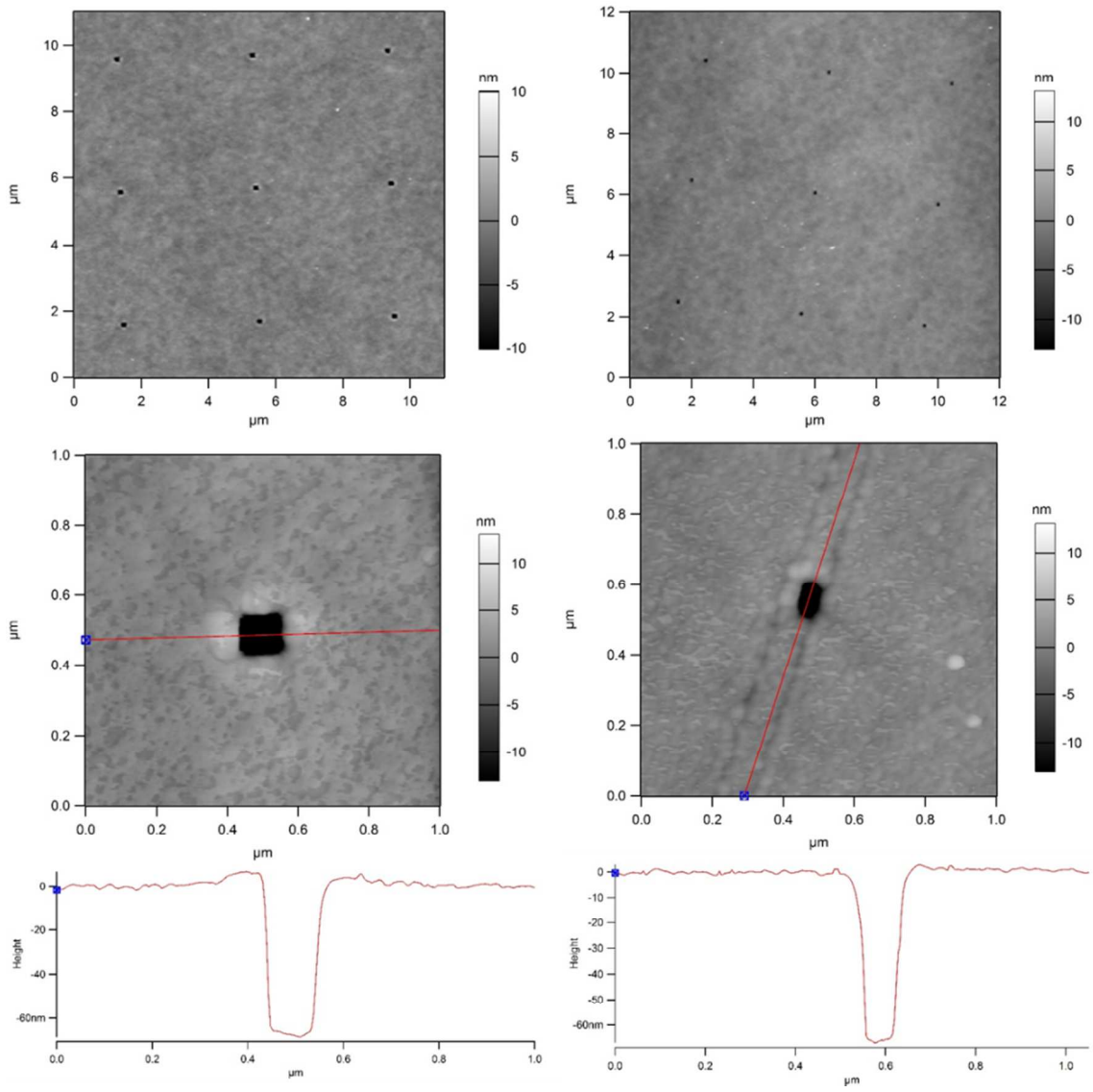

Figure S2. The atomic force microscopy images for box array (Left) and rod array (Right) and the height profile which shows 60 nm PMMA thickness.

3D printed cells for EPD and its assembly

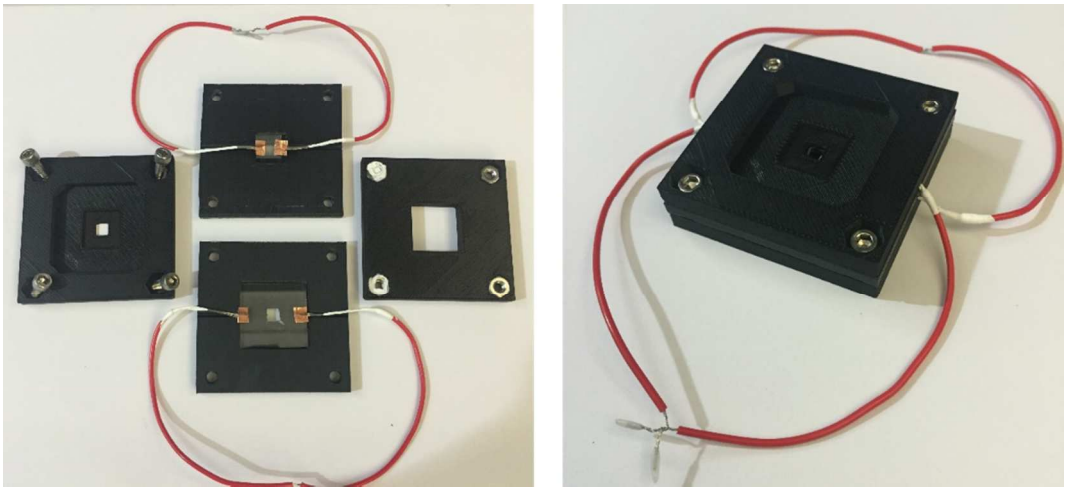

Figure S3. Digital images of assembly method and 3D printed, small-scale EPD cell with 10 mm (L) × 10 mm (W) × 2 mm (H) tank. Before and after assembly.

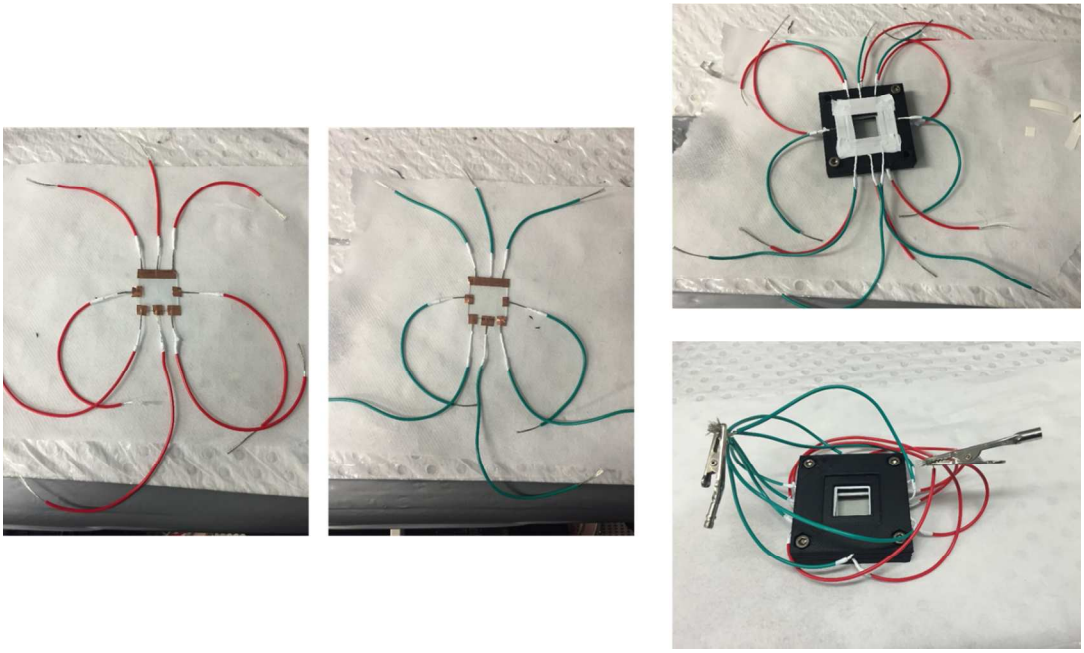

Figure S4. Digital images of assemble method and 3D printed large scale EPD cell with 20 mm (L) × 20 mm (W) × 2 mm (H) tank.

Scattering spectrum of single gold nanoparticles in arrays

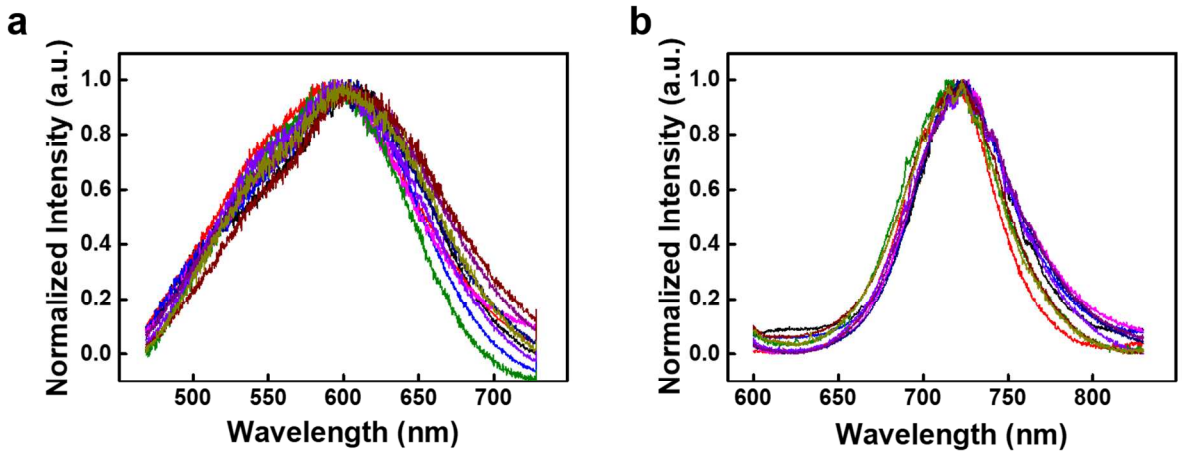

Figure S5. Single particle scattering spectra of gold NSs and gold NRs assembled in arrays via EPD. (a) Scattering spectrum shows the consistency for randomly ten gold NSs in a line of array. (b) Scattering spectrum shows the consistency for randomly ten gold NRs in a line of array

Change of Zeta potential and double layer thickness as a function of salt concentration

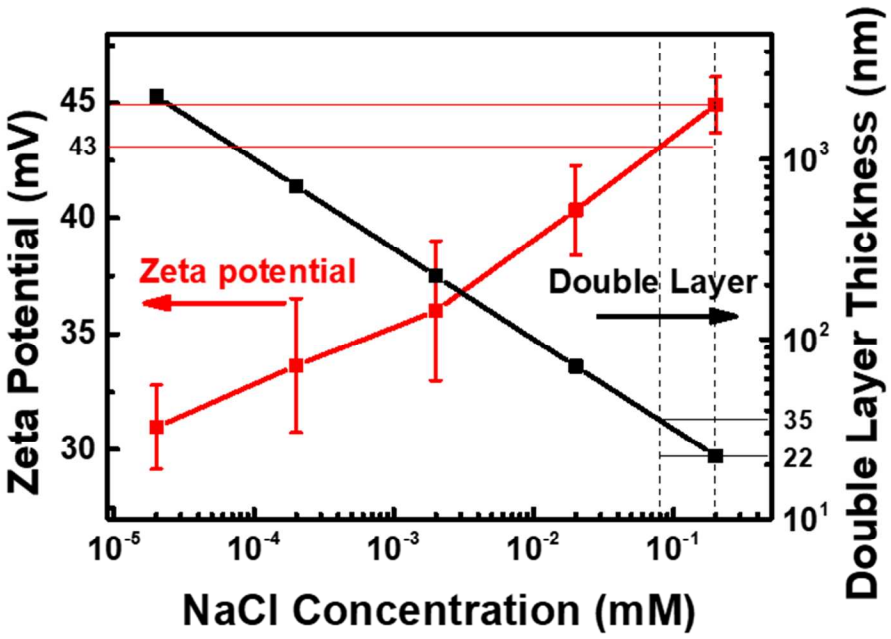

Figure S6: The plot of zeta potential (left axis) and double layer thickness (right axis) as a function of NaCl concentration in 110 nm gold NSs solution. (room temperature 293K, pH = 7, p-DADMAC coated particles) Black dash line indicate the applied NaCl concentration region in experiment.

The screen effect on the surface of the PMMA-ITO template.

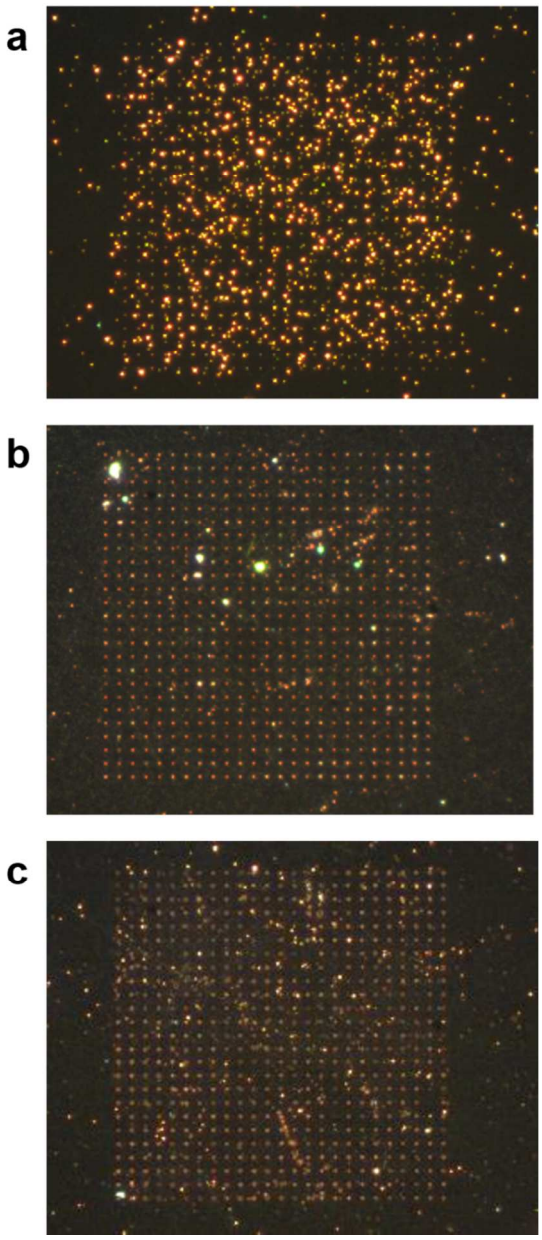

Figure S7: Gold nanorod EPD assembly under different electrolyte concentration and applied potential to demonstrate the influence of electric field overlapping near the electrode surface. EPD condition (Potential, NaCl concentration) (**a**): 3V, 0 mM. (**b**): 3V, 0.2 mM. (**c**):4V, 0.2 mM.

The schematic of reflectance dark-field microscope attached with polarizer.

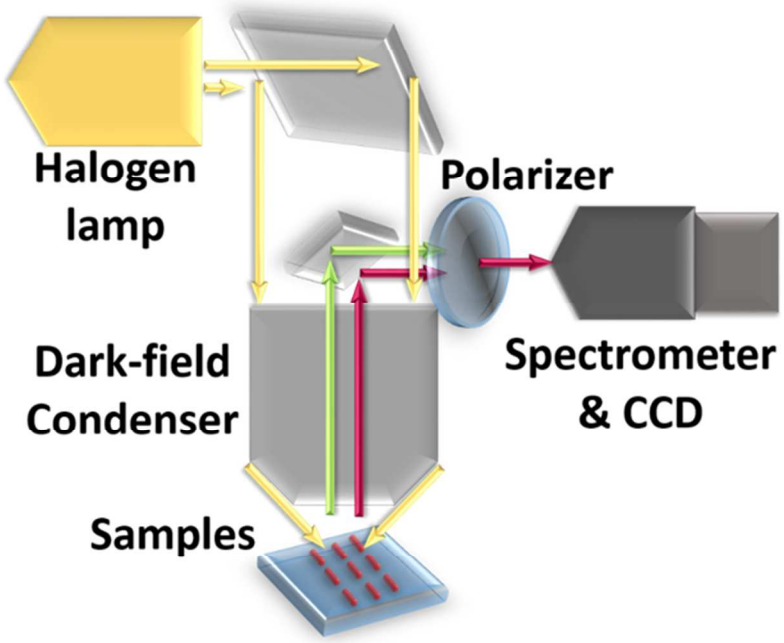

Figure S8: Schematic of reflectance dark-field microscope with polarizer.

Structural Characterization of letter “Au” and corresponding rectangular array

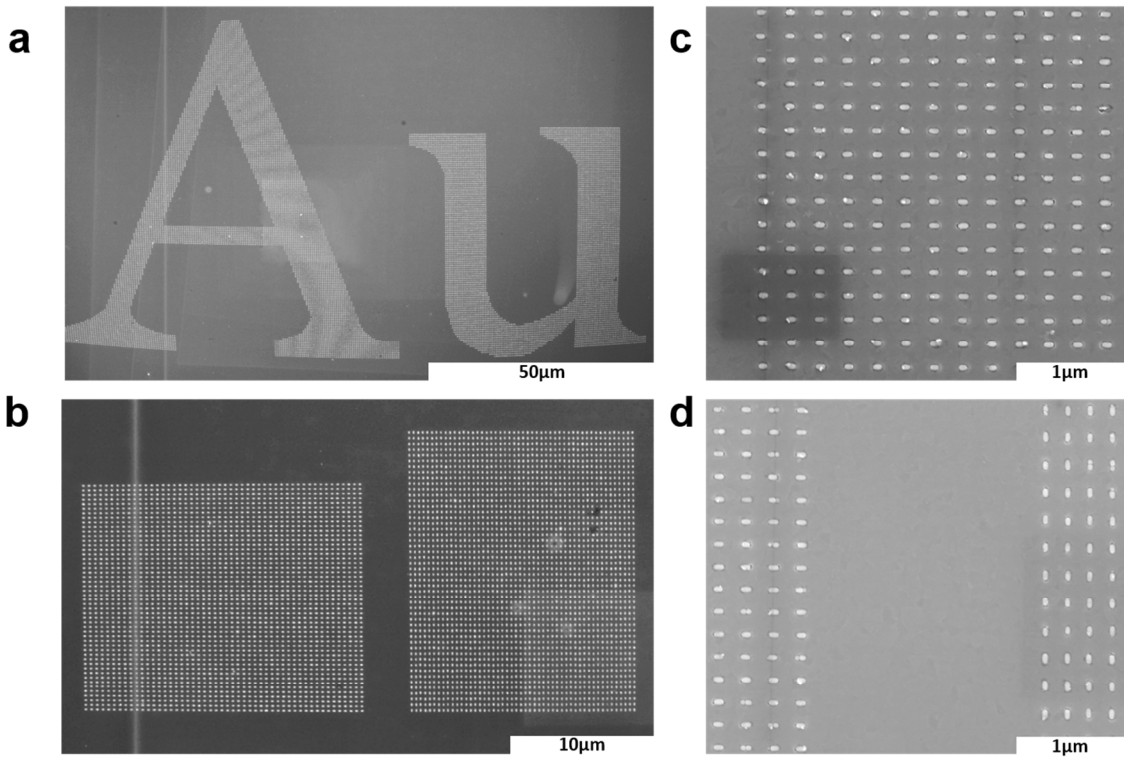

Figure S9: The scanning electron microscope image of (a) Letter “Au” assembled with gold NRs with perpendicular orientation and (b) corresponding rectangular array with same array structure. High magnification scanning electron microscope image of (c) the corner and (d) between rectangular arrays.

The logo of the University of Melbourne filled by gold NRs *via* EPD assembly.

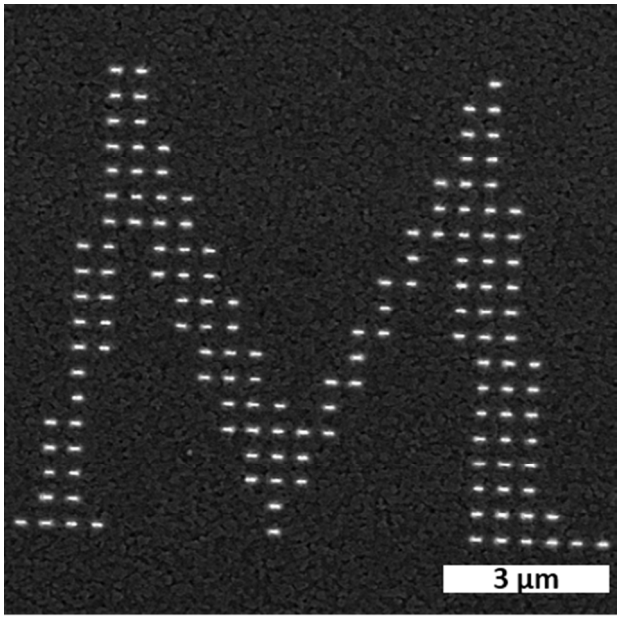

Figure S10. Scanning electron microscopy image of the letter "M" in the text "Melbourne".

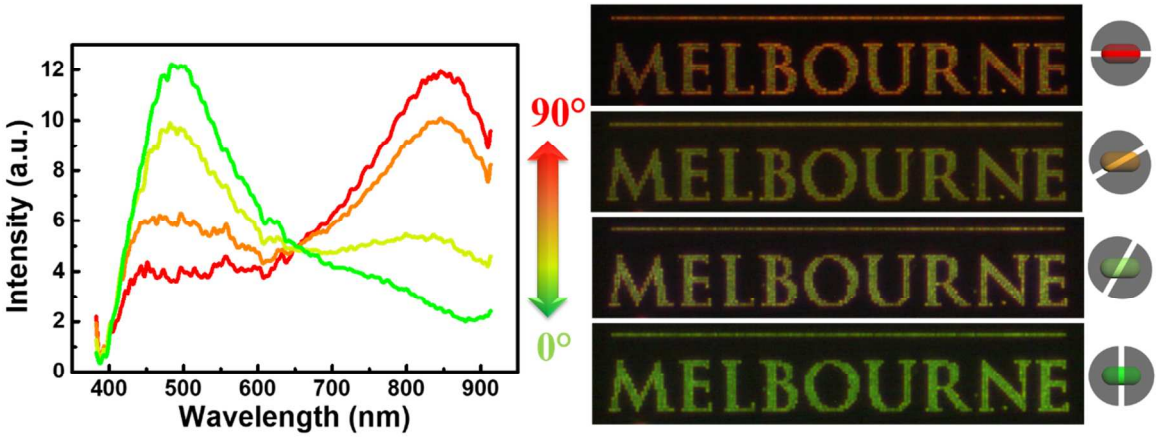

Figure S11. Dark-field scattering spectra of the logo of "Melbourne" under 0 (Green), 30 (Yellowish green), 60 (Orange) and 90 (Red) polarization angle and corresponding dark-field images.

Large area “Au” logo

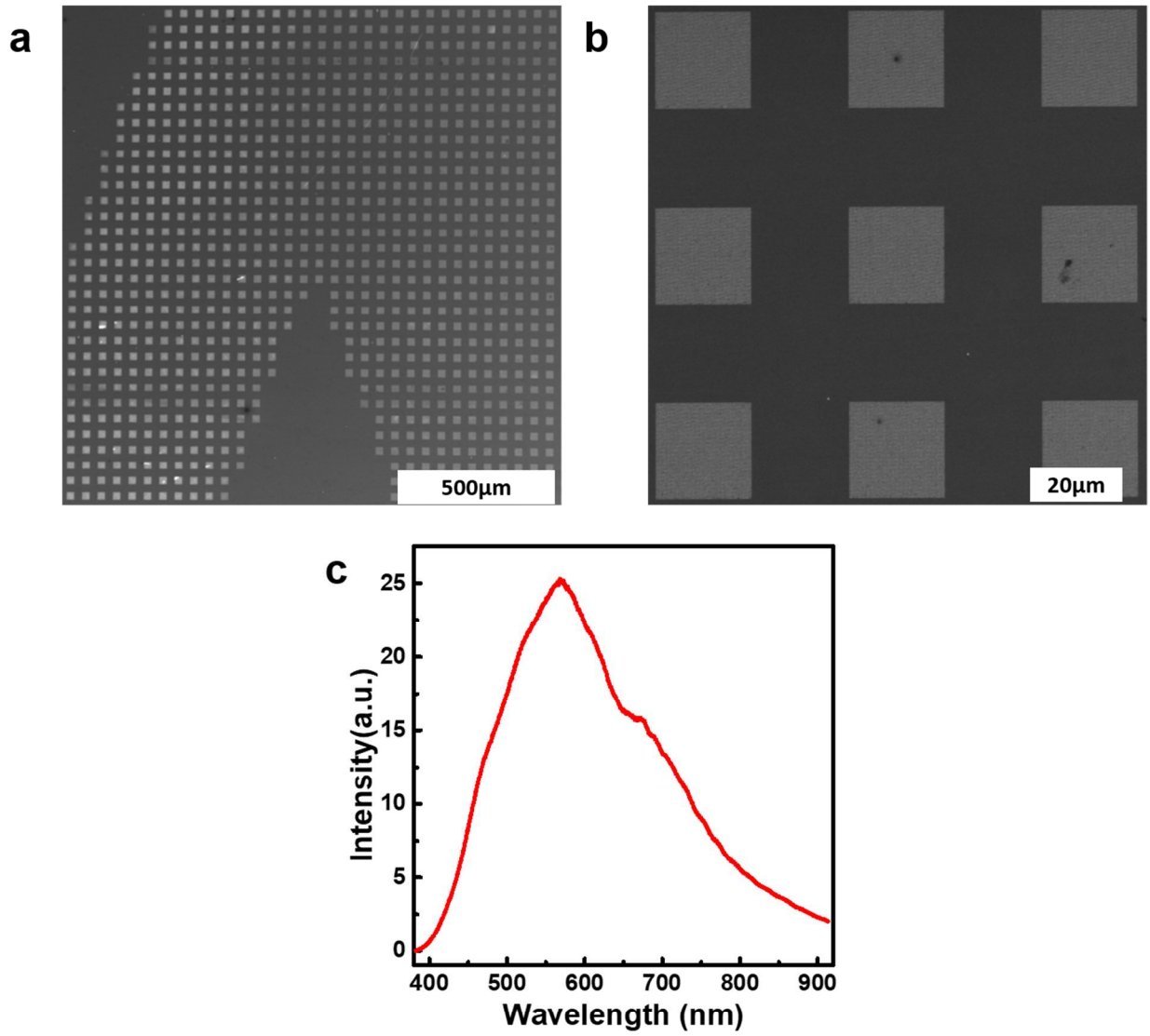

Figure S12 Scanning electron microscopy images of “Au” logo filled by gold NRs *via* EPD assembly. (a) Top part of the “Au” logo of the letter “A”. (b) 9 of the Pixels in “Au” logo filled by gold NRs. (c) Dark-field microscopy scattering spectrum of one pixel in “Au” logo.