Electronic Supplementary Information for

Photothermal Structural Modification of Porous Gold Nanoshells *via* Pulsed-Laser Irradiation: Effects of Laser Wavelengths and Surface Conditions

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Supplementary Note: Temperature rise calculation under irradiation with pulses

Calculation for the temperature of colloid by irradiation with laser pulses rise has been conducted using

$$\Delta T = E \cdot N \cdot (1 - 10^{-Ext})/c_{col} \cdot m_{col} \quad (1)$$

where E is the pulse energy, N is the number of irradiating pulses, *Ext* is the extinction of colloid at a wavelength, c_{col} and m_{col} are the specific heat capacity and the mass of the overall colloid, respectively. First of all, the extinction ($Ext = c_{Au} \cdot \varepsilon \cdot l$; c_{Au}, concentration of gold nanostructures; ε, extinction coefficient of irradiated colloid; l, optical path length) values at 355 nm and 532 nm can be obtained from Figure S1d (Ext_{355} = 0.280 and Ext_{532} = 0.239). While the path length was 1.0 cm in the UV-vis extinction measurement, it was 2.4 cm in the laser irradiation experiments (Figure S2). Thus, for the calculation, modified extinction values should be Ext'_{355} = 0.672 and Ext'_{532} = 0.574. The pulse energy, E, could be obtained by multiplying the laser fluence with the beam spot area. The diameter of the spot was d = 0.40 cm and the area was A = 0.126 cm^2. Thus, the pulse energy was E = 0.378 mJ at 3.0 mJ/cm^2 and E = 0.630 mJ at 5.0 mJ/cm^2. The number of irradiating pulses, N, could be deduced simply by multiplying the repetition rate of the laser (10 Hz) and the irrdiation time (600 s): therefore N = 6,000. The specific heat capacity and the mass of the colloid can be approximated to the respective values of water, c_{water} = 4.184 J/g·℃ and m_{water} = 2.4 g. Using the values given above, we have calculated the temperature rise values under irradiation with laser pulses.

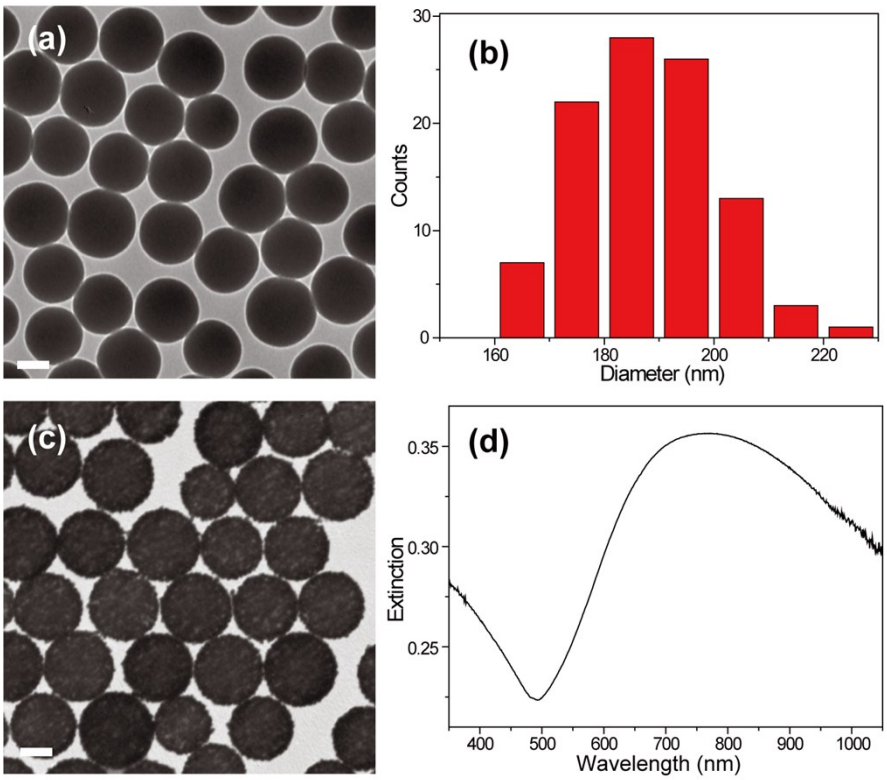

Figure S1 Preparation of porous gold nanoshells. (a) TEM image and (b) size distribution of core SiO_2 nanoparticles. (c) TEM image and (d) UV-vis extinction spectrum of porous gold nanoshells (scale bars: 100 nm).

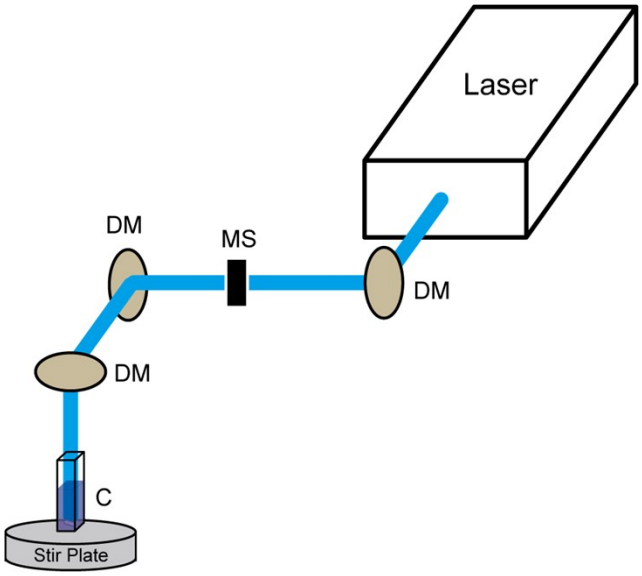

Figure S2 Schematic of the laser irradiation setup for the structural modification of porous gold nanoshells (DM, dichroic mirror; MS, mechanical shutter; C, cell). Laser pulses of 355 nm or 532 nm were irradiated for 10 min.

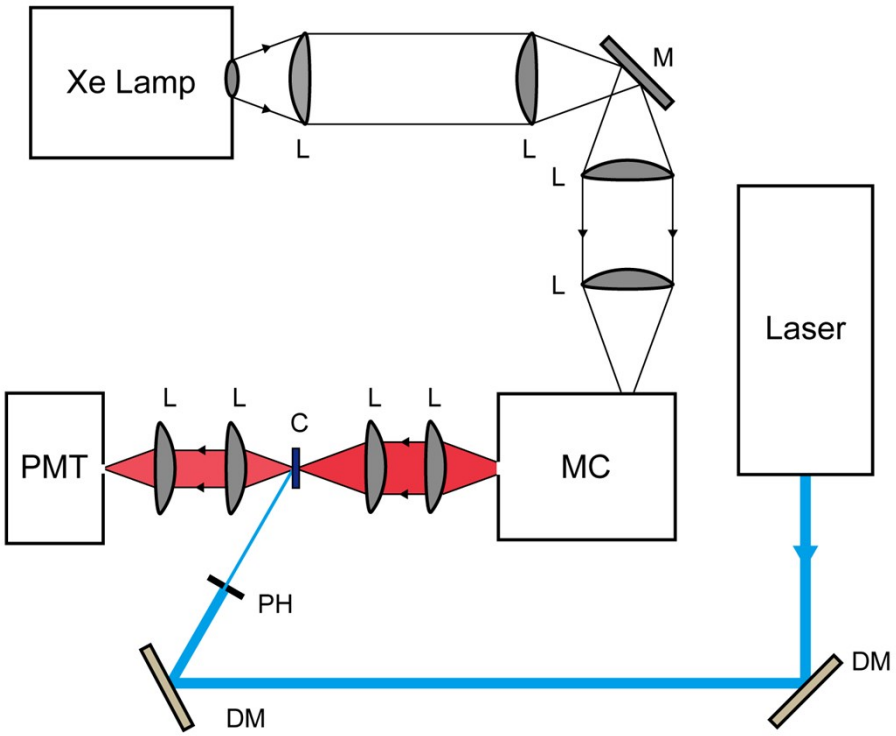

Figure S3 Schematic illustration for the experimental setup of the transient-absorption measurement (L, lense; M, mirror; MC, monochromator; DM, dichroic mirror; PH, pinhole; C, cell; PMT, photomultiplier tube).

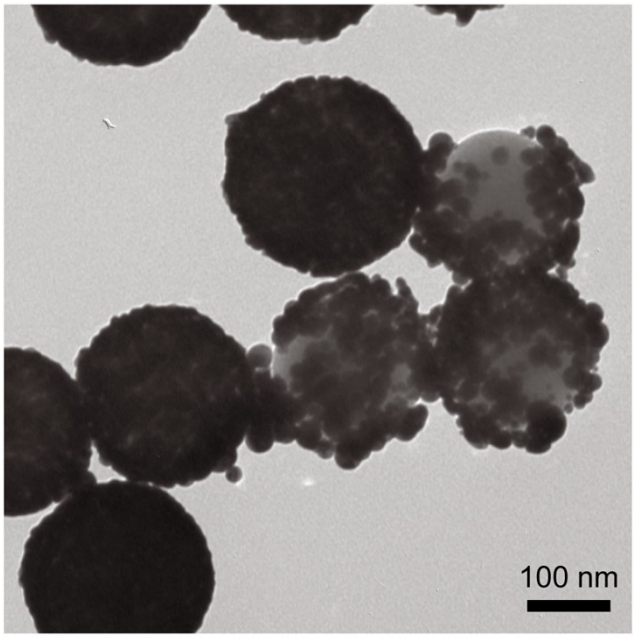

Figure S4 TEM image of porous gold nanoshells irraidated with 532 nm pulses at a fluence of 5 mJ/cm^2.

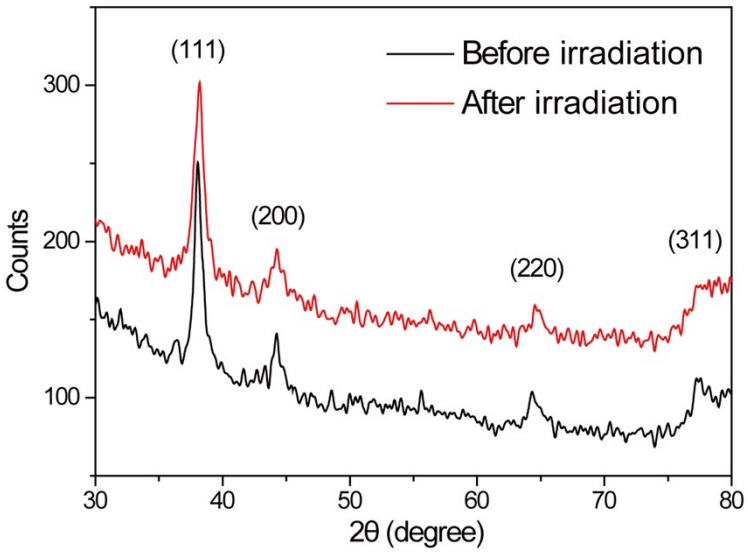

Figure S5 XRD patterns of porous gold nanoshells before and after laser irradiation (355 nm, 6 mJ/cm^2).

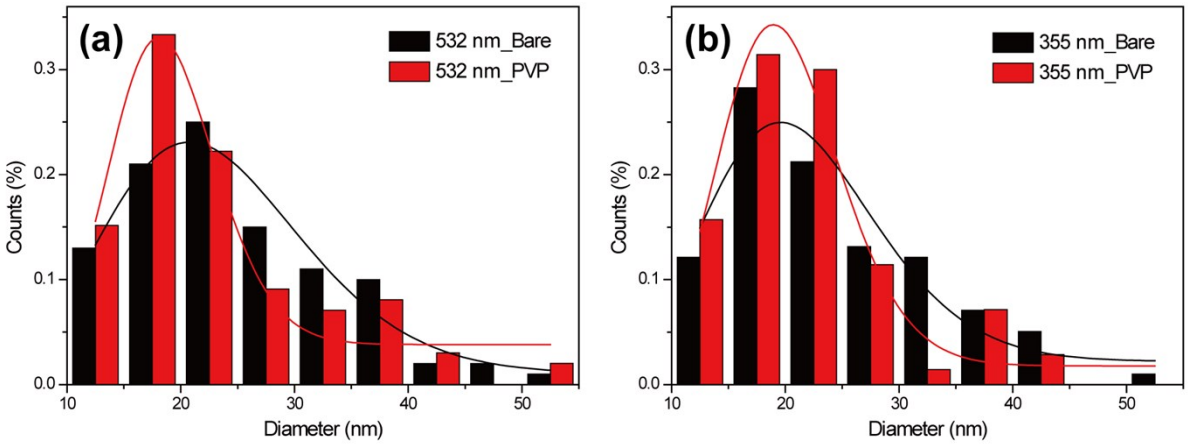

Figure S6 Size distribution of gold nanospheres, which were generated by 6 mJ/cm^2 pulses under the indicated conditions.

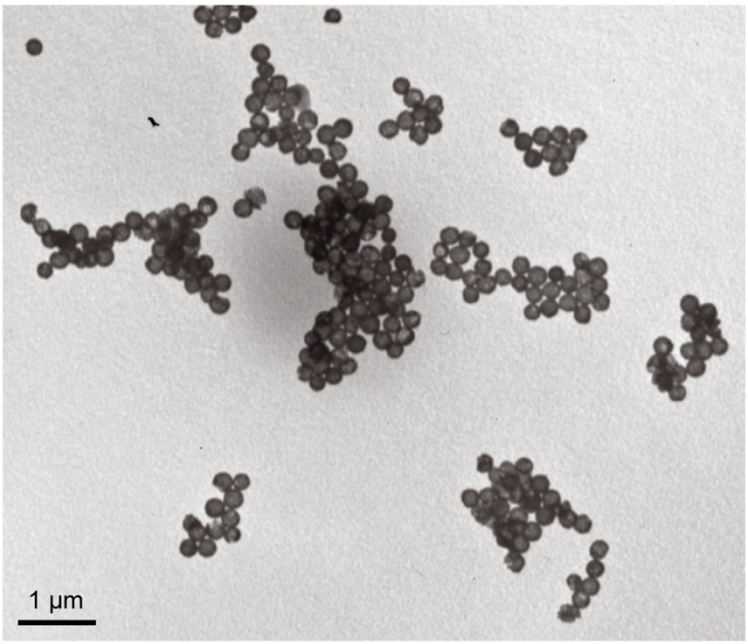

Figure S7 Low-resolution TEM image acquired after the core SiO_2 etching of irradiated porous gold nanoshells. The irradiation was conducted with 355 nm laser pulses at a fluence of 4 mJ/cm^2 after the addition of PVP.

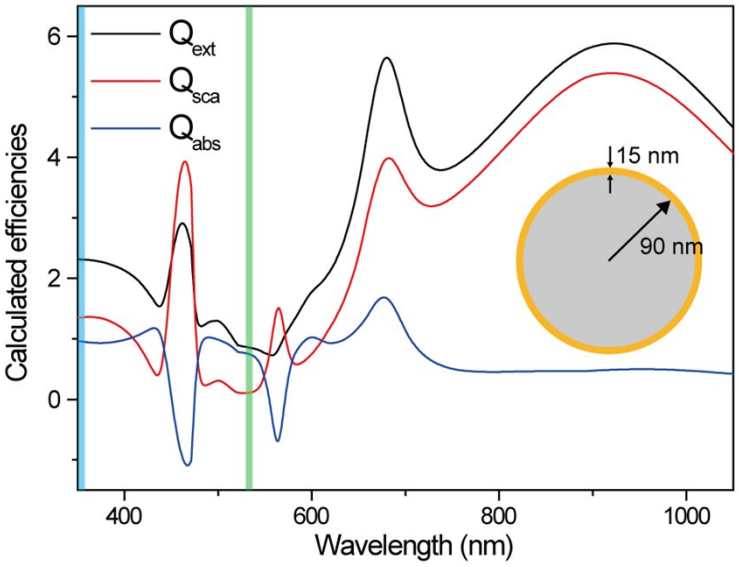

Figure S8 The extinction, absorption and scattering spectra of gold nanoshells supported on SiO_2 cores, simulated using the Mie calculation. The SiO_2 core radius and the gold shell thickness were set to be 90 nm and 15 nm, respectively.

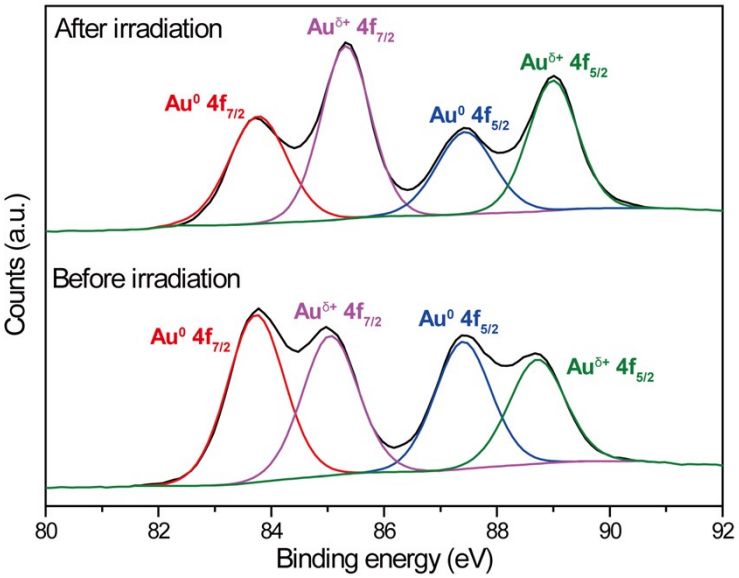

Figure S9 Au 4f XPS spectra of porous gold nanoshells before and after laser irradiation (355 nm, 6 mJ/cm^2). Each spectrum has been deconvoluted *via* the Gaussian fitting.