Supplementary Information for "Plasmonic Nanosponges Filled by Silicon for Enhanced White Light Emission."

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1. FABRICATION METHOD

A gold film and a layer of amorphous silicon were places on a glass substrate in two stages (see Fig.S2). Initially, a layer of amorphous silicon thick was deposited on the surface of the glass substrate by the plasma-enhanced chemical vapor deposition method (see Fig.S2a). Sequentially a gold layer was deposited behind it by magnetic sputtering (see Fig.S2b). Final film thicknesses in bi-layer structure were 15 nm and 60 nm for the gold and silicon layers, respectively.

Si/Au NPs were fabricated using the femtosecond laser ablation method in the air at the room temperature (294 K) (see Fig.S1). Bilayer films (15nm/60nm Au/Si) on a glass substrate were employed as targets for the ablation process. As a source of the ultrashort laser pulses we used a commercial system TEMA-150 (Avesta project) operating at the wavelength of 1050 nm, spectral width of 7 nm, pulse duration of 150 fs and the pulse repetition rate of 1 kHz after a pulse picker system. Laser radiation with approximately 50 nJ per a pulse was focused using the Mitutoyo Plan Apo NIR Infinity Corrected Objective (10×, NA = 0.26) (see Fig.S1b). In the process of the laser printing of the NPs, the Au/Si bilayer target was placed on the distance of 40 μm from a receiving surface (glass or TEM grid) (see Fig.S1c). An example of a direct image of particles in scattered light on a glass substrate is shown in the Fig.S1.

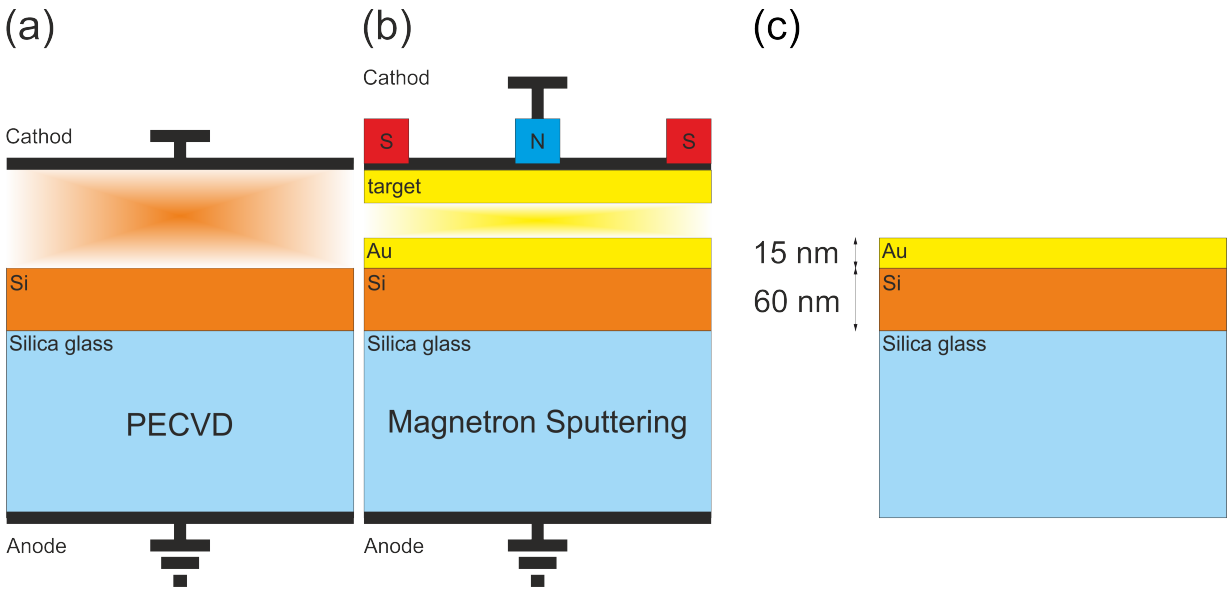


Figure S1. **Step by step pictures of the Au/Si film fabrication process.**

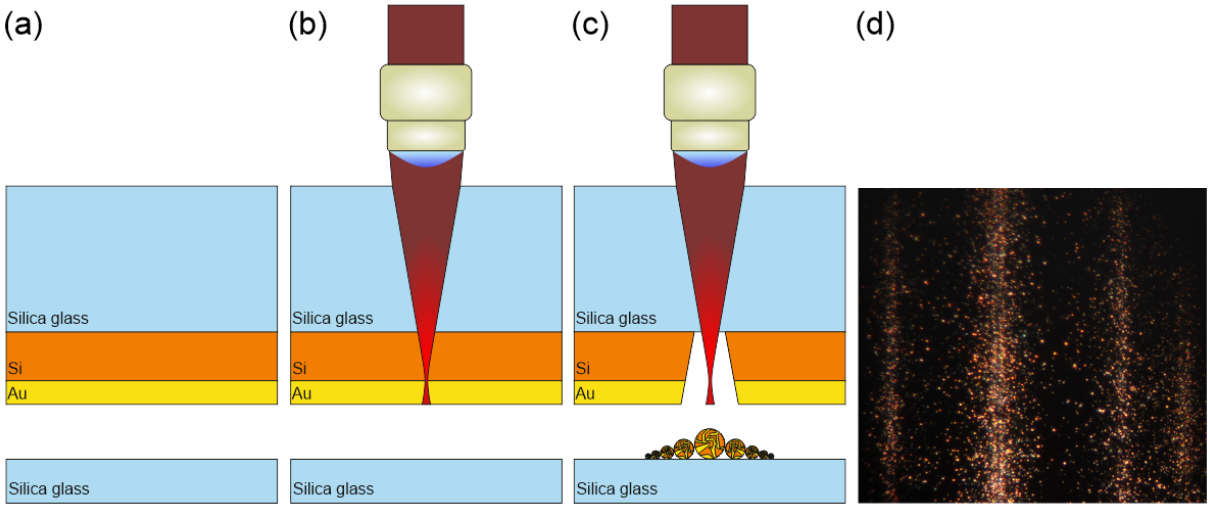


Figure S2. **Step by step pictures of the Au/Si nanoparticles fabrication process.**

2. CRYSTALLOGRAPHIC CHARACTERIZATION.

Si/Au NPs were further characterized by transmission electron microscopy (TEM) using a JEOL - ARM 200F Cold FEG TEM/STEM operating at 200 kV and equipped with a spherical aberration (Cs) probe corrector with a 0.19 nm point resolution in TEM mode and 0.078 nm in Scanning mode (STEM).

The EDS analysis has shown an Au concentration within the whole particle in the range of 30 70 at% Au, with however, noticeable concentration inhomogeneity. Si/Au is a simple eutectic system with very low solubility Therefore, the phase diagram suggests that NPs concentration leads to a biphase alloy composed of a quasi-pure Si phase and a quasi-pure Au phase. The buoyancy of the Au network within Silicon makes it difficult to interpret local EDS analysis or Fast Fourier Transform (FFT). Indeed, several grains of different nature may be superimposed and therefore lead to a convoluted signal. Probing a smaller area increases the probability to probe a single grain but at the cost of the resolution. To tackle this issue, microdiffraction technique has been used, intersecting the probed area with the edge of the NPs (Fig. S3a) in order to minimize the number of crystals within the probed area.

Electron Diffraction pattern has been obtained on the area shown in Fig. S3a. The experimental pattern matches with the one of a pure Gold crystal in [112] zone axis. This is further confirmed by the measurement of interplanar distances (Tab. I).

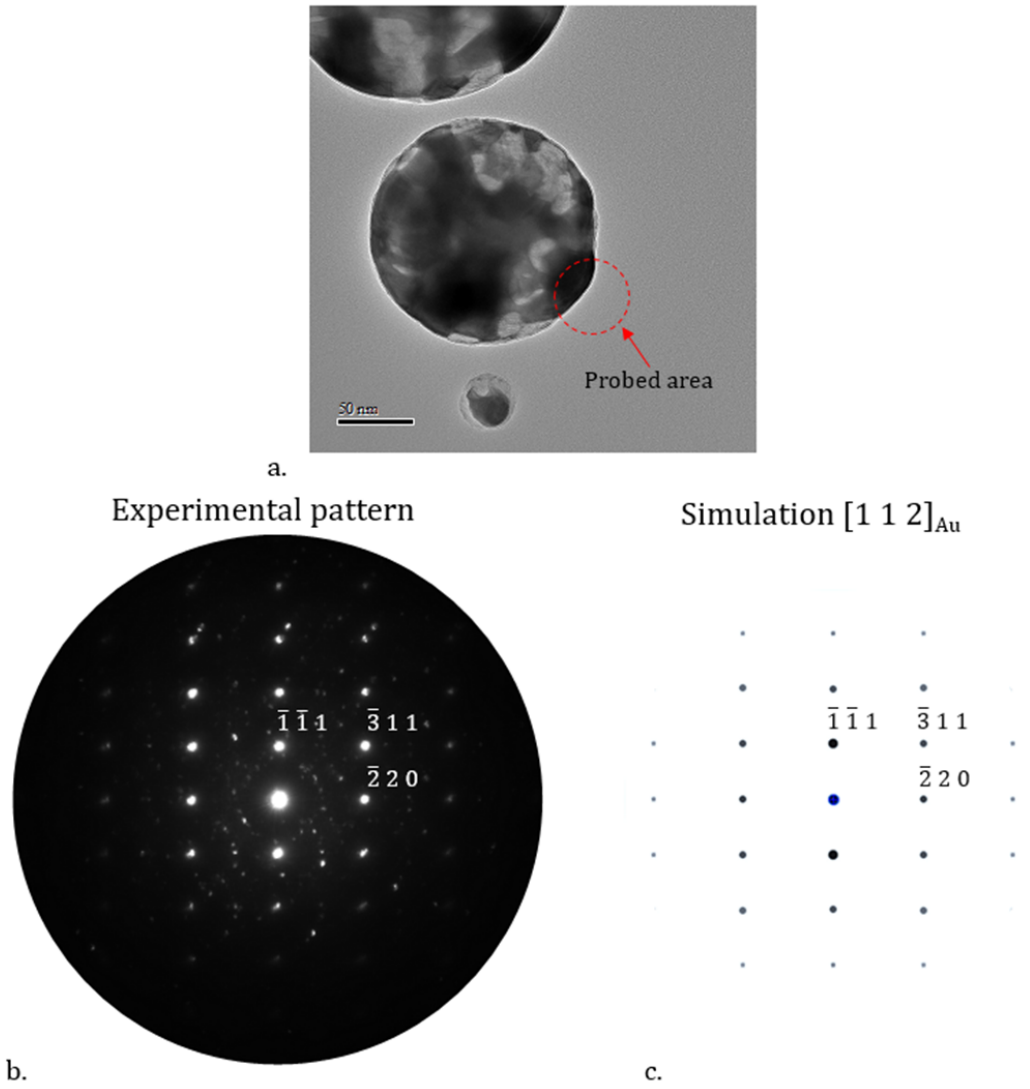
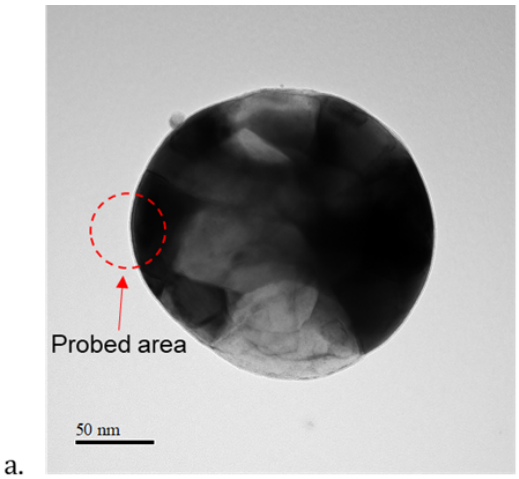


Figure S3. **Bright-field micrograph** (a) with the experimental (b) and simulated diffraction pattern of pure Gold in a [1 1 2] Zone Axis.

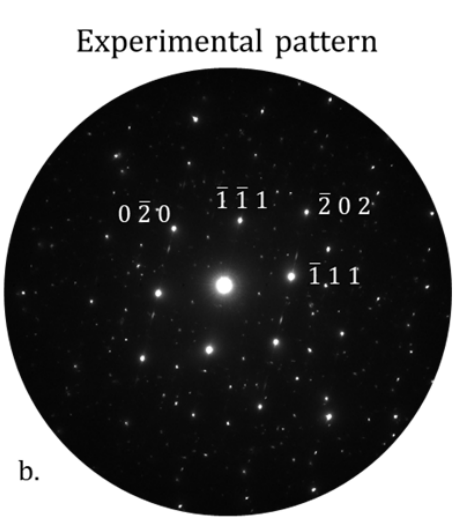
The procedure has been repeated for the investigation on the Si-rich phase (Fig. S4). A satisfying matching of the

Table I.

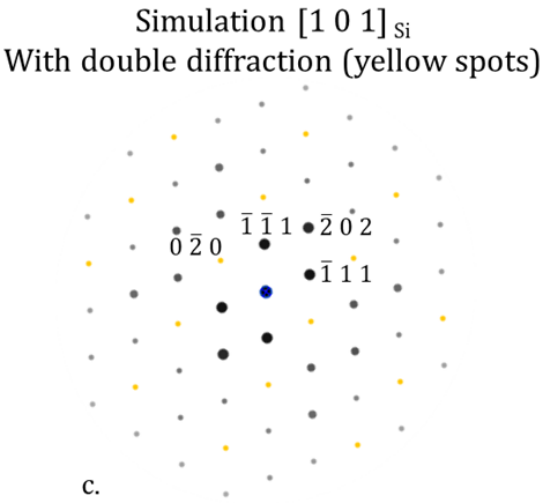
(h k l)	$d_{experimental}$ (nm)	$d_{theoretical}$ (nm)
($\bar{1}$ $\bar{1}$ 1)	0.319 ±0.005	0.314
($\bar{2}$ 0 2)	0.196 ±0.005	0.192
(0 $\bar{2}$ 0)	0.280 ±0.005	0.272



a.



b.



c.

Figure S4. **Bright-field micrograph** (a) with the experimental (b) and simulated diffraction pattern of pure Silicon in a [1 0 1] Zone Axis.

experimental diffraction pattern has been obtained for the zone axis [1 0 1] of pure Silicon (Fig. S4b,c and Tab. II). Supplementary spots have been identified as double diffraction spots.

Table II.

(h k l)	$d_{experimental}$ (nm)	$d_{theoretical}$ (nm)
($\overline{1}$ $\overline{1}$ 1)	0.236 ±0.005	0.237
($\overline{3}$ 1 1)	0.147 ±0.005	0.145
($\overline{2}$ 2 0)	0.125 ±0.005	0.124

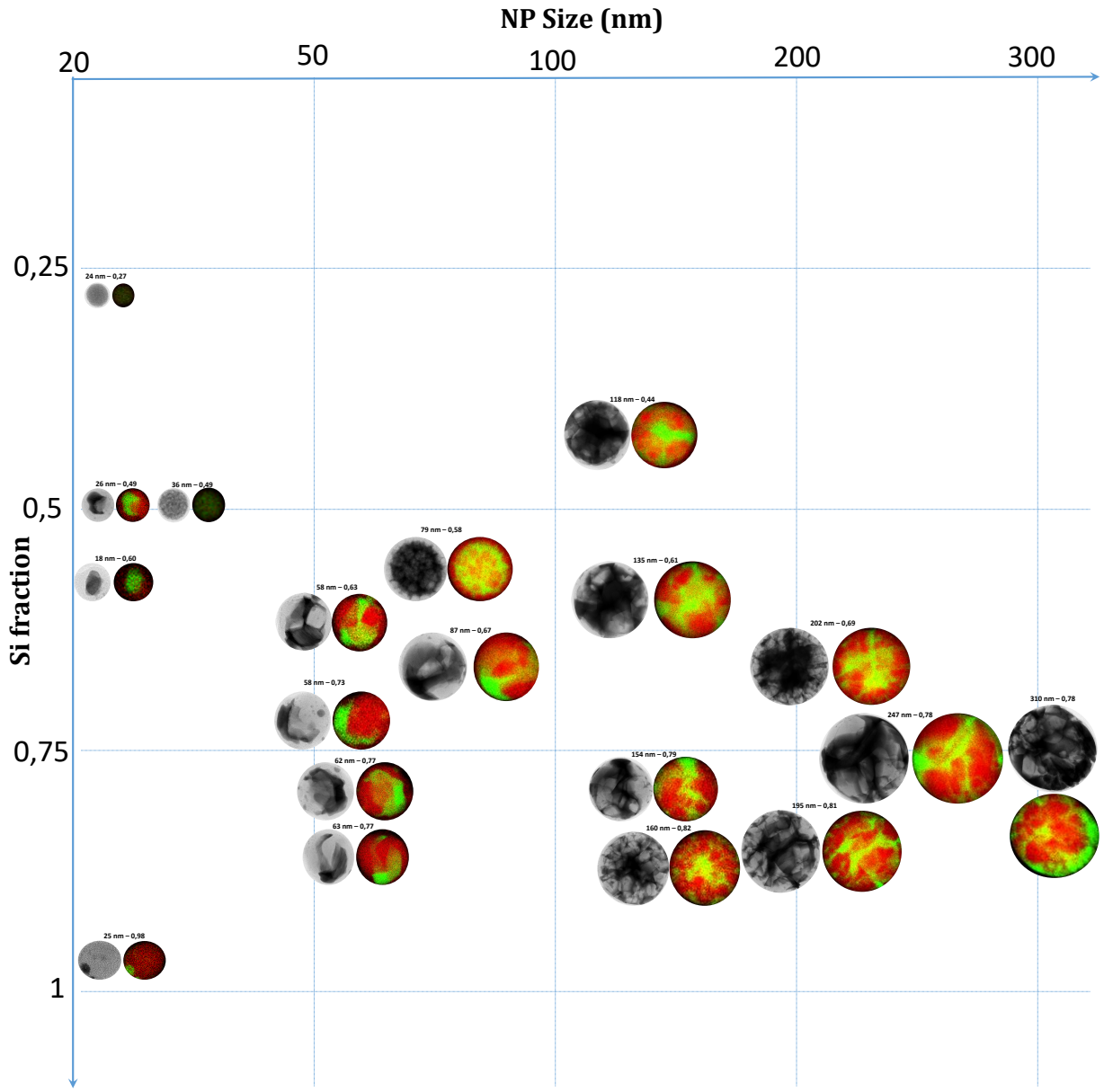


Figure S5. **Experimental map of Au-Si nanoparticales structure.**

3. CALCULATION OF MODE DECOMPOSITION FOR SI NANOSPHERES.

Scattering C_{sca} cross-section for spherical nanoparticles is expressed via the following expressions:

$$C_{sca} = \frac{W_{sca}}{I} = \frac{2\pi}{k^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2), \tag{S1}$$

where W_{sca} is the scattered power by a nanosphere, $k = 2\pi n_r/\lambda$, n_r is the refractive index, I is the incident intensity, a_n and b_n are the scattering coefficients that describe interaction of the nanoparticle with a plane wave. These assumptions lead to simplified form of the scattering coefficients:

$$a_n = \frac{m\psi_n(mx)\psi_n'(x) - \psi_n(x)\psi_n'(mx)}{m\psi_n(mx)\xi_n'(x) - \xi_n(x)\psi_n'(mx)}, \tag{S2}$$

$$b_n = \frac{\psi_n(mx)\psi_n'(x) - m\psi_n(x)\psi_n'(mx)}{\psi_n(mx)\xi_n'(x) - m\xi_n(x)\psi_n'(mx)}, \tag{S3}$$

with $x = ka$, and the Riccati - Bessel functions : $\psi_n(\rho) = \rho j_n(\rho)$, $\xi_n(\rho) = \rho h_n^{(1)}(\rho)$, where j_n and $h_n^{(1)}$ are spherical Bessel and Hankel functions, respectively. These scattering coefficients are shown in Fig. S6.

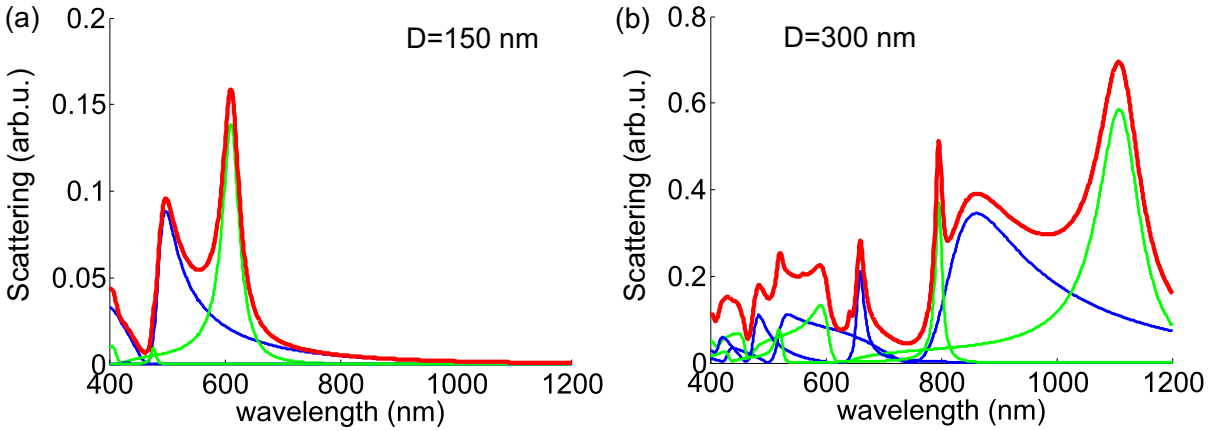


Figure S6. Calculated mode decomposition for Si nanospheres with diameter 150 nm (a) and 300 nm (b). Green and blue lines correspond to magnetic and electric modes, whereas red lines are total scattering cross-sections.