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Supplementary Information

Novel and simple route to fabricate fully biocompatible plasmonic mushroom arrays adhered on silk biopolymer

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1. Angle-dependence of the Silk-Au MA

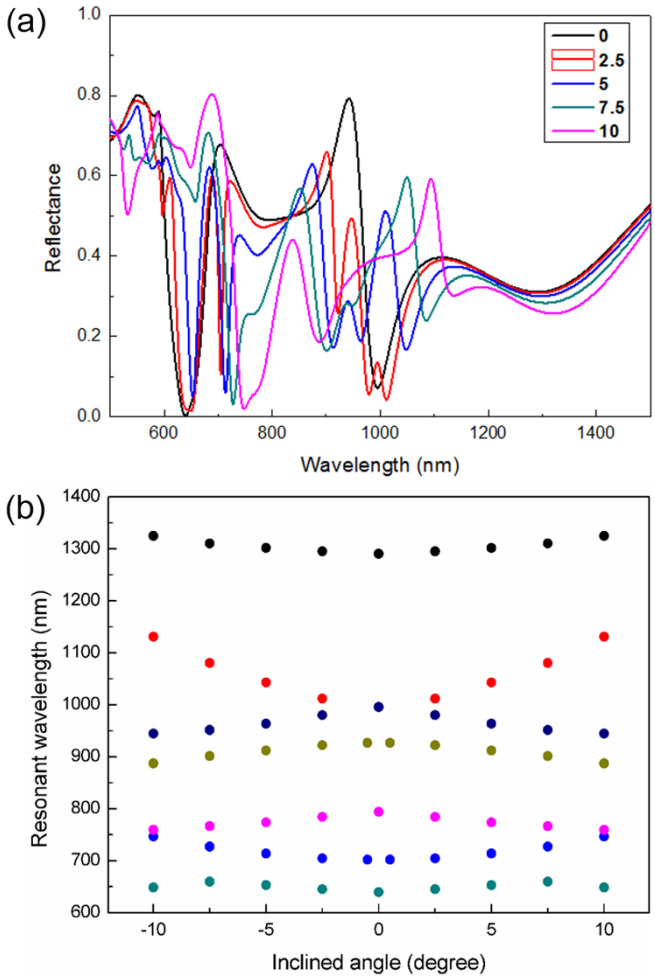

Fig. S1 (a) Effect of the angle of incidence on the reflection for the Silk-Au MA structure in simulation. (b) Plot of wavelength of resonance modes as a function of incident angle.

To investigate the angle-dependence of plasmonic resonances shown in the Silk-Au MA, numerical simulations were performed by employing a commercial FDTD software (FDTD solutions, Lumerical Solutions) to generate the reflectance spectra. The Bloch boundary condition including a single Au mushroom in the *x* and *y* directions (lateral) was used; this includes perfect matching layers (PMLs) on the top and bottom boundaries of the simulation unit-cell. The complex dielectric constants of Au were taken from the Palik handbooks. The refractive index of silk used herein was 1.5. For structure excitation, a plane wave source, normally launched to the top Au cap, was used. The plane wave covers a wavelength range of 500–1500 nm with 5000 frequency points. Reflected light was recorded on the top PML.

As shown in Fig. S1, since surface plasmons in a periodic metal structure are coupled to the Bragg resonance, the Silk-Au MA exhibits the angular dispersion of plasmonic modes. We also confirmed that there was a degenerated Bragg resonance around $\lambda \sim 1000$ nm due to the symmetry of the periodic

lattice for the normal incidence. This resonance could be split when the symmetry was broken by the oblique incident.

2. Atomic force microscopy image of the Au nanoparticle array on a silk film

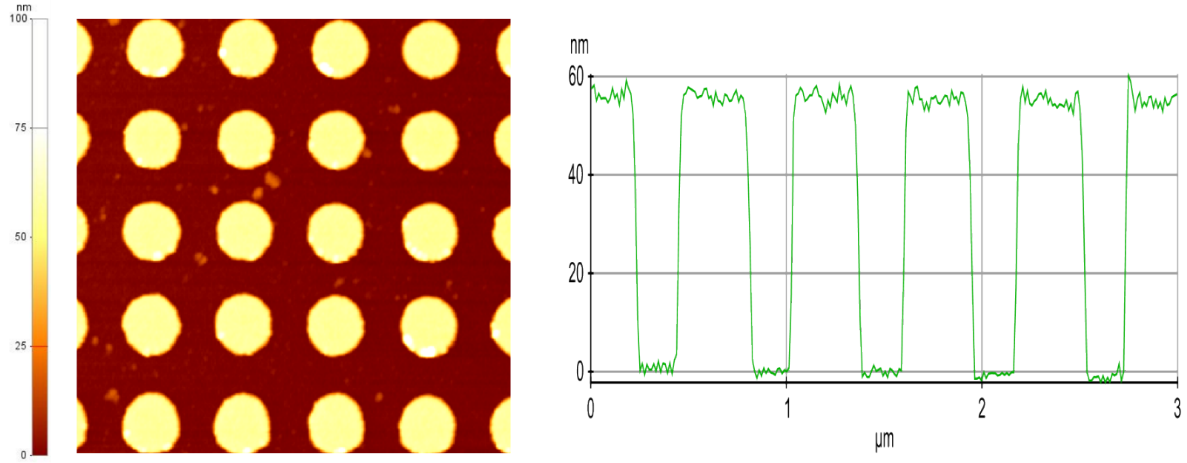

Fig. S2 Atomic force microscopy image of the nanoparticle array on the silk film after 1 hour ultra-sonication.