

Supplementary Information

Dual-Band Moiré Metasurface Patches for Multifunctional Biomedical Applications

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Fabrication of AMMP

The Au thin film (100 nm in thickness) and MgF₂ thin film (300 nm in thickness) were deposited sequentially onto a premium glass substrate using electron beam deposition. A Ti adhesion layer (5 nm in thickness) was used between the Au film and the glass substrate. MNSL was then applied to create Au moiré metasurface on the MgF₂ surface. Specifically, polystyrene (PS) spheres with a diameter of 800 nm (Thermo Scientific Inc.) were used in the MNSL.³⁰ Oxygen plasma etching (March Plasma CS170IF) was applied to tune the size of the PS spheres in bilayer. The Au moiré metasurface was formed by depositing an Au layer (20 nm in thickness) onto the PS-patterned MgF₂ followed by selective removal of PS via ultrasonication of the substrate in toluene for 5 minutes.

Deposition of PMMA and BSA on AMMP

All the AMMP were cleaned for 10 minutes using a UV ozone cleaner (BioForce Nanosciences, Inc.) before the deposition. The PMMA (Mw 46,000) with a thickness of ~20 nm was spin-coated onto the AMMP. A sub-monolayer of BSA proteins was formed on the AMMP by

incubating the substrates in the BSA aqueous solution (1 mM, BAH64, Equitech-Bio, Inc.) for 24 hours followed by rinse with DI water.

Structural and optical characterizations

Scanning electron micrographs of samples were obtained using FEI Quanta 650. MIR spectra were recorded with a Thermo Scientific Nicolet Continuum FTIR microscope coupled with a Nicolet 6700 FTIR spectrometer. NIR spectra were obtained with a Bruker Hyperion 2000 IR microscope coupled with a Bruker Vertex v80 FTIR spectrometer. Spectrometers and microscopes were continuously purged with ultrahigh-purity N₂ gas to suppress atmospheric spectral lines (i.e., CO₂ and water vapor). The measurement areas on the samples were ~60 μm X 60 μm, as defined by an aperture in the optical microscope. The number of quasiperiodic units was ~900 within the measurement areas. The spectra were normalized using an Au mirror as a reference.

Laser heating

A diode laser with a wavelength of 1310 nm (Thorlabs, Inc.) was used for the experiments focused on the plasmon-enhanced photothermal effects. The laser beam was focused onto the sample surfaces through a 40X objective in an inverted optical microscope (TiE, Nikon Inc.).

Optical simulations

We applied FDTD methods to simulate the optical spectra and electric-field distributions of various plasmonic nanostructures. We used the commercially available software package (FDTD Solutions, Lumerical). The optical properties of Au were adapted from Johnson and Christy (for wavelengths from 0.7 μm to 2 μm)⁵⁵ and Palik (for wavelengths from 2 μm to 6 μm)⁵⁶. The refractive index of MgF_2 was set as 1.38. Perfectly matched layers were applied as boundary conditions. The simulated area was 18 μm X 18 μm to include \sim 80 quasiperiodic units. The mesh size was set as 8 nm.

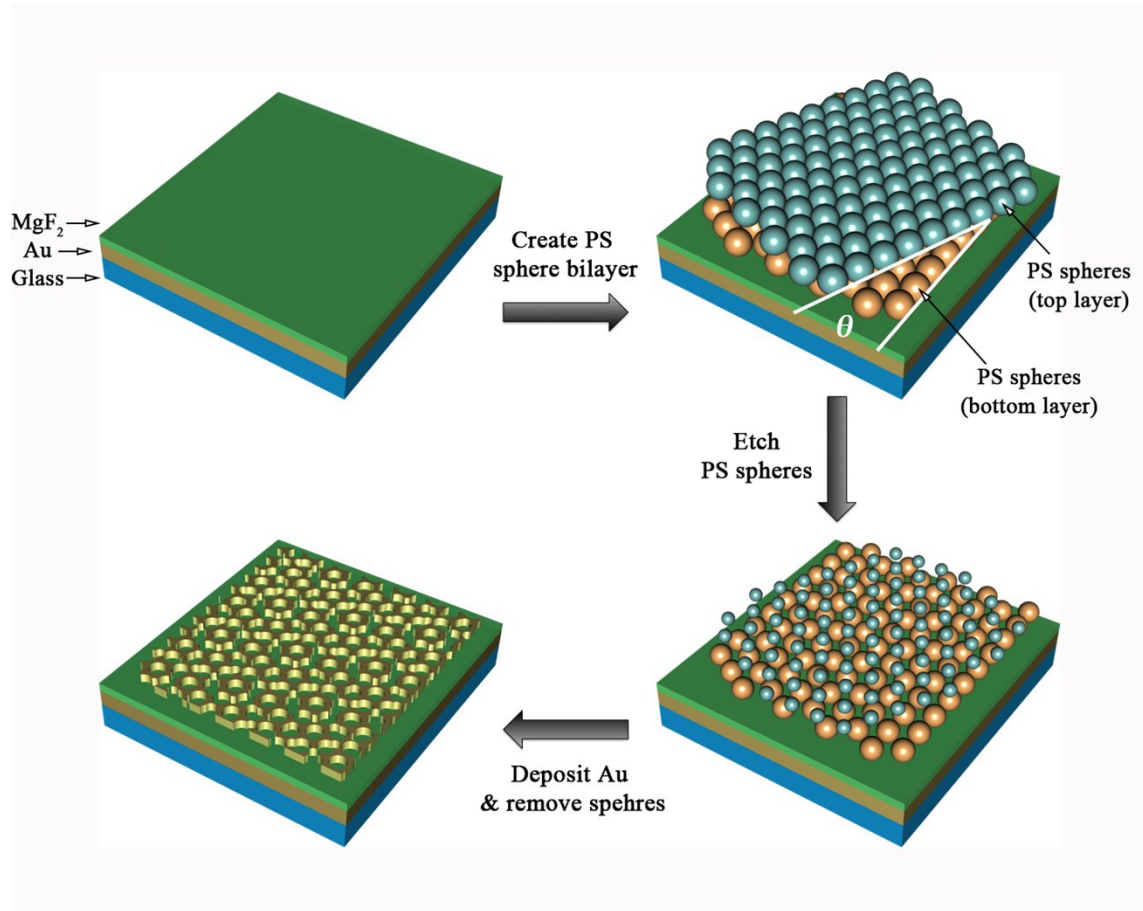


Figure S1. Fabrication process of AMMP. Firstly, an Au thin film and a MgF_2 thin film are deposited on a glass substrate. Secondly, two monolayers of polystyrene (PS) nanospheres with a relative in-plane rotation are formed on the surface of MgF_2 via directed assembly. Thirdly, oxygen plasma etching is applied to reduce the size of PS nanospheres. Finally, Au is deposited onto the substrate and PS spheres are selectively removed to leave Au moiré metasurface on the substrate. The relative in-plane rotation angle (θ) can be controlled to tune the moiré pattern.

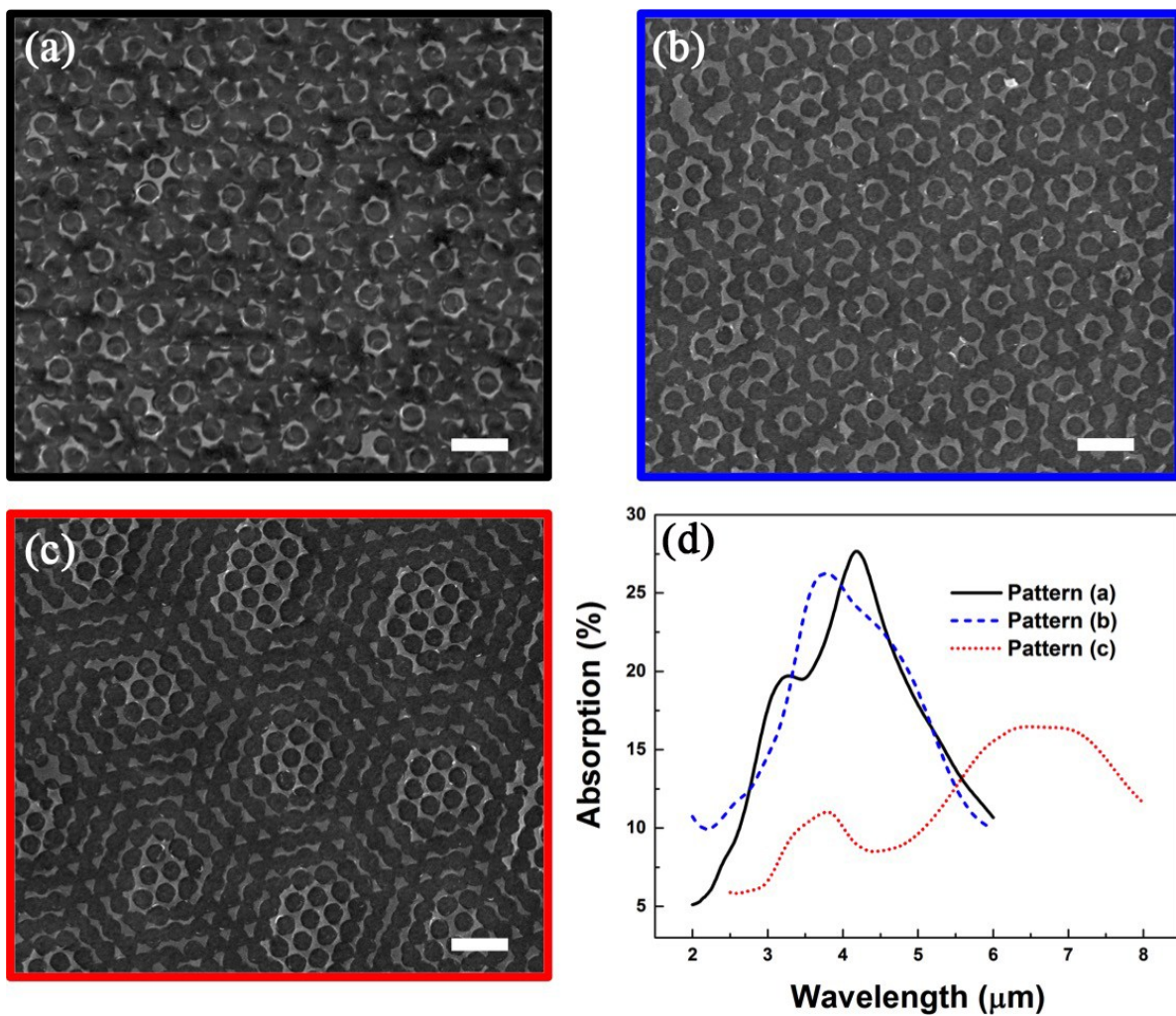


Figure S2. (a) - (c) Scanning electron micrographs of three different types of Au moiré metasurfaces based on the different relative in-plane rotation angles of the bilayers of PS nanospheres in MNSL. The scale bars are 2 μm. (d) Simulated absorption spectra of the Au moiré metasurfaces in (a) - (c).

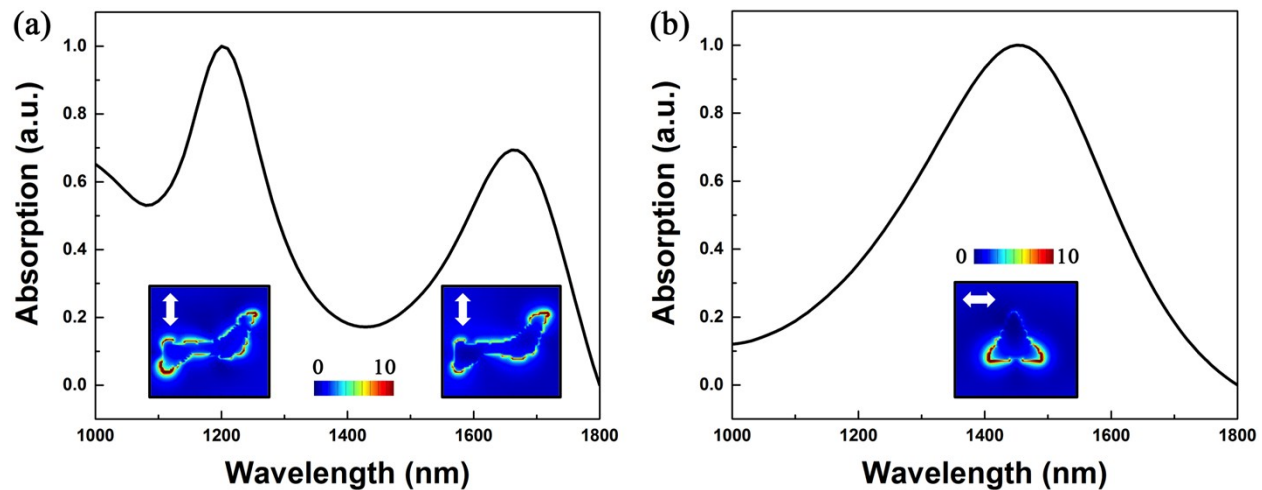


Figure S3. Simulated absorption spectra of representative (a) z-shaped nanostructure and (b) triangular nanostructure, respectively. The insets show the electric field ($|E|$) distributions at the corresponding resonance peaks. The white arrows depict the polarizations of incident light.

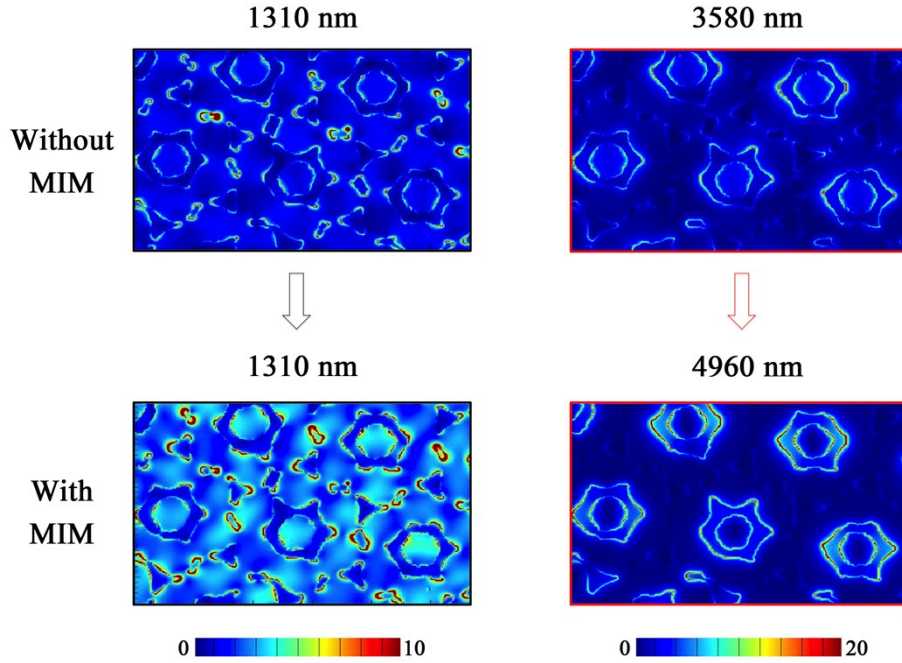


Figure S4. Comparisons of electric field ($|E|$) distributions of the Au moiré metasurface with and without MIM configuration in NIR and MIR regimes. The numbers indicate the resonance wavelengths at which the electric field distributions were simulated. The field distributions are at the bottom surfaces of the moiré metasurfaces.

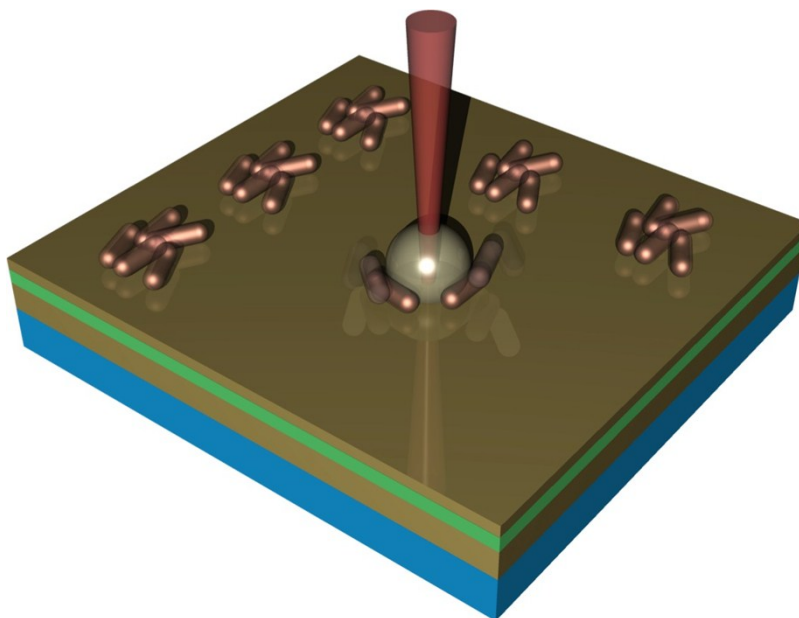


Figure S5. Schematic illustration of patterning bacteria clusters into an array on the AMMP via steering the laser beam that is used to generate the microbubbles.