Efficient Third Harmonic Generation from Metal-Dielectric Hybrid Nanoantennas

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1. Fabrication of the hybrid nanoantenna

Arrays of hybrid nanoantennas composed by a Au nanoring and Si nanodisk were fabricated on a sapphire substrate using electron beam lithography (EBL) and reactive ion etching (RIE). A commercially available crystalline silicon thin layer fabricated by a hetero-epitaxial process on a sapphire substrate was first etched down by RIE process to obtain the desirable thickness. Then the substrate was spin-coated with positive tone resist, PMMA (poly(methyl methacrylate)), and baked at 180 °C for 5 minutes. The substrate was exposed with an electron beam followed by a development procedure in a MIBK (methyl isobutyl ketone) : IPA (isopropanol) = 1 : 3 solvent. The nanostructured PMMA was covered with a 40 nm Cr layer deposited by thermal evaporation, and then removed by a lift-off process in acetone. The remaining Cr mask pattern was transferred to the silicon layer via the second RIE, and then removed by commercial etchant, providing the arrays of the Si nanodisk. After the fabrication of the Si nanodisk, the second PMMA layer was coated on the substrate and treated by the same EBL procedures. The position of the second PMMA mask was aligned to the Si nanodisk using cross-shaped markers. A Au layer was then thermally evaporated onto the substrate, and the following lift-off process provided the final sample (Figure S1).

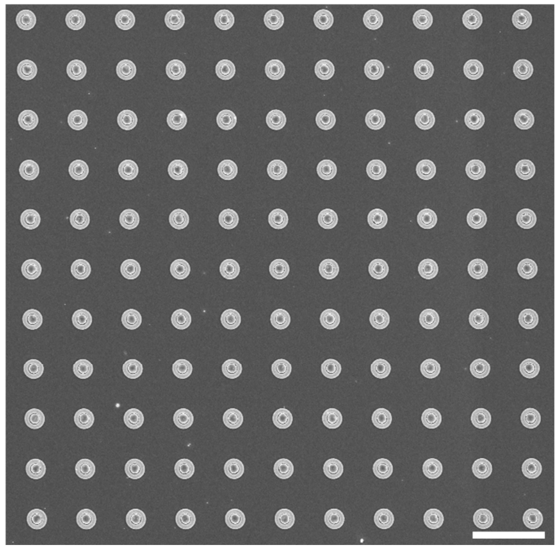

Figure S1. SEM image of the fabricated array of the hybrid nanoantennas. Scale bar corresponds to 5 μm.

2. Extinction spectra of the isolated Si nanodisk and Au nanoring

Simulated extinction spectra of the isolated Si nanodisk and Au nanoring are shown in Figure S2 as reference.

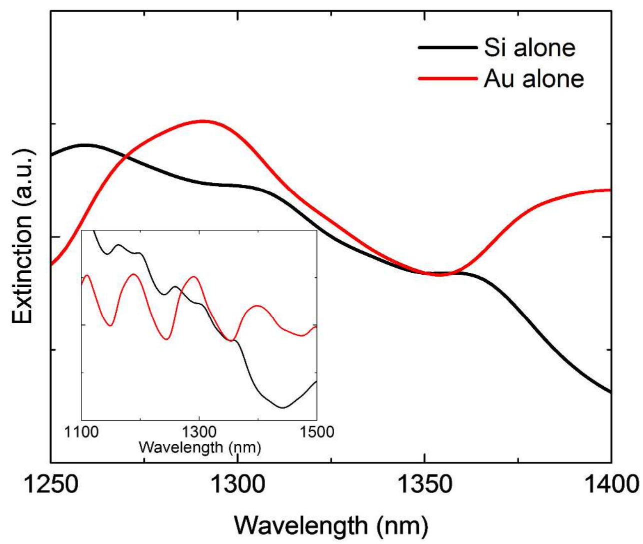

Figure S2. Simulated extinction spectra of the isolated Si disk and Au ring. The inset shows the same spectra for a broader wavelength range.

3. Details of the third harmonic generation (THG) measurement

The THG signal from the fabricated nanoantennas was measured using the experimental setup illustrated in Figure S3. A pulsed Yb:KGW PHAROS laser system was used as the pump of a collinear optical parametric amplifier ORPHEUS with a LYRA wavelength extension option (Light Conversion Ltd., pulse duration of 180 fs, repetition rate of 100 kHz). The excitation beam was reflected by a short-pass dichroic mirror (Thorlabs DMSP805 for λ = 1300−1500 nm or DMSP1000 for λ = 1500−1800 nm) and focused onto the sample with a 100× (NA = 0.9) air objective from Nikon, giving rise to an excitation spot of #1400 nm FWTM (full width at tenth of maximum). The third harmonic emission was collected in a backscattering configuration via the same objective and detected with an avalanche photodiode (MPD PDM series by Picoquant) for imaging or by a spectrograph (PI Acton SP2300 by Princeton Instruments) for spectral measurements. The sample was fixed to an XYZ piezo-scanner stage (Nano-Drive, Mad City

Laboratories) to perform the scanning. The conversion efficiency was determined by directly measuring the collected TH emission power with a calibrated silicon photodetector (Newport), and the excitation power with a germanium photodetector (Thorlabs), and then computing the ratio between the both. For <1 pW TH powers, values were calibrated by using the measured TH spectra.

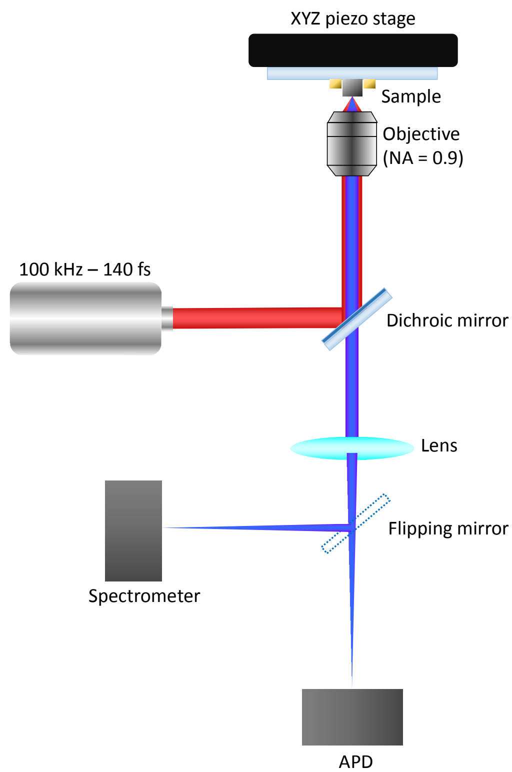

Figure S3. Schematic image of the experimental THG measurement setup.

4. Extinction spectra when the dimensions of the hybrid structures were varied

Figure S4 shows the extinction spectra of the hybrid structures in which the geometrical parameters were modified to expand the spectral range of operation of our nanosystem. The dimensions of the hybrid nanoantennas corresponding to the figures are as follows. (a) Si nanodisk: 840 nm diameter and 195 nm height. Au nanoring: 940 nm inner diameter, 1720 nm outer diameter and 145 nm height. (b) Si nanodsik: 960 nm diameter and 220 nm height. Au nanoring: 1060 nm inner diameter, 1960 nm outer diameter and 160 nm height.

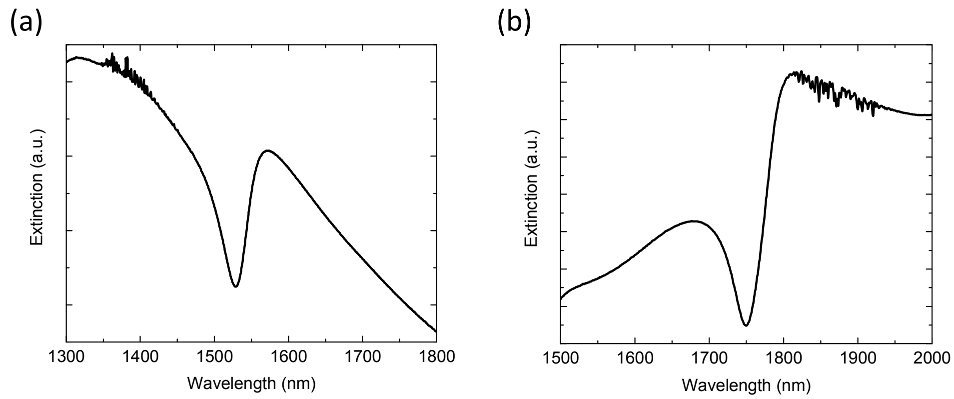

Figure S4. Experimental extinction spectra of the hybrid nanoantenna which generates TH at (a) $\lambda = 515$ nm and (b) $\lambda = 590$ nm.