Supplementary Information for

“Chip-scale Plasmonic Sum Frequency Generation”

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1. Fabrication of the MIM nanostructure

The MIM nanostructure is fabricated on the 1-mm-thick glass substrate. A 100-nm-thick gold slab and a 14-nm-thick alumina film are sequentially deposited on the glass substrate by the e-beam evaporation. The top nanostructure is then defined by the e-beam lithography and developed in 1:3 methyl isobutyl ketone/isopropyl alcohol solution. Finally, a 5-nm-thick chromium and a 30-nm-thick gold film are deposited by the e-beam evaporation and the MIM nanostructure is obtained after the liftoff.

2. SFG experiment

The experimental setup for SFG is shown in Fig. S1. An 800 nm ultrafast amplified Ti:sapphire laser (Coherent Inc.) provides the sources for the two pump light. 5% of its output is used as one pump beam while 95% is used to pump an optical parametric oscillator whose output wavelength can be tuned to 1500 nm as the other pump beam. The pulse duration of the pump beams is 110 fs and the repetition rate is 76 MHz. Both the 800-nm and the 1500-nm pump beams from the lasers are x-polarized. A quarter wave plate and a polarizer are adopted to change the 800-nm pump beam to y-polarized. An objective lens with a numerical aperture of 0.4 focuses the two pump beams onto the sample. A delay line is used to make sure the two pump pulses overlap in the time domain when they arrive at the sample. The SFG signal is collected by the same objective lens and detected by a high-resolution Raman spectrometer (Princeton Instrument). The integration time of the spectrometer is set as 300 s. Filters are introduced before the spectrometer to remove the two pump light. A linear polarizer can also be inserted before the filters to allow for the selective collection of the SFG in different polarizations.

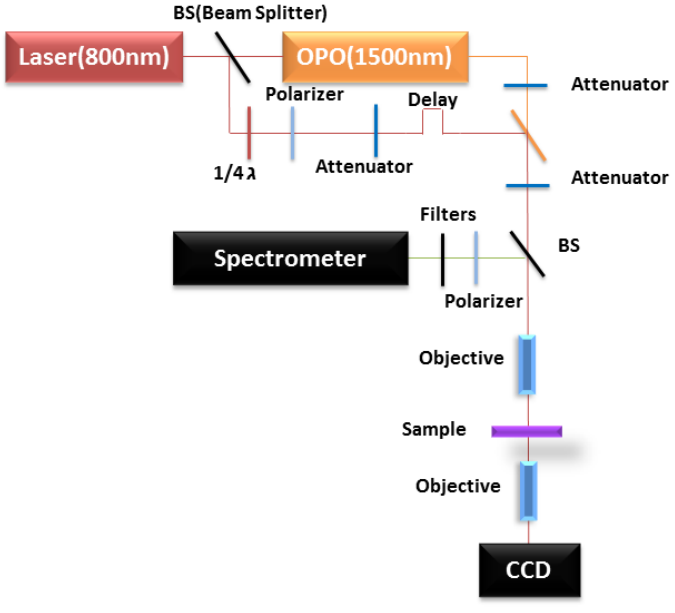

Fig. S1. Experimental setup. BS means the Beam Splitter. 1/4 λ means the Quarter Wave Plate

3. Resonance peak changed independently

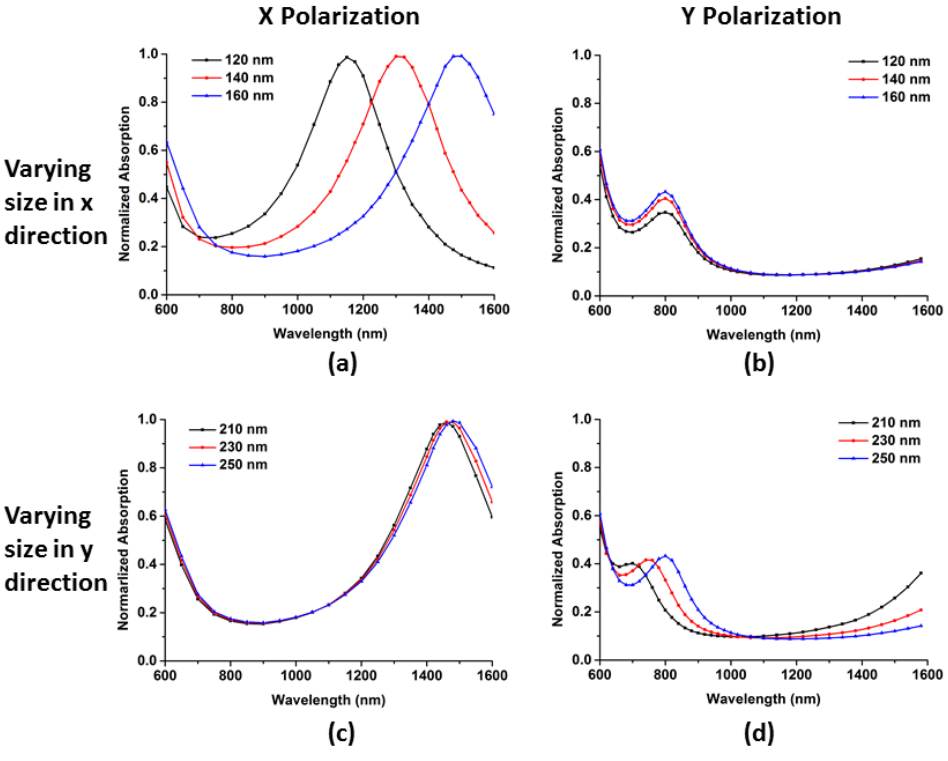

Fig. S2. Simulated absorption is displayed when the geometric sizes of the top nanostructures in x- and y- directions vary for different polarized pump. (a, c) and (b, d) are for X- and Y- polarizations, respectively. (a, b) are the absorption with increasing size in x direction from 120 nm to 160 nm. (b, d) are the absorption with increasing size in y direction from 210 nm to 250 nm

The simulated absorption when the geometric sizes of the top nanostructures in x- and y- directions vary independently is provided in Fig. S2. With increasing size in x direction, the resonance for x polarization shifts from 1100 nm to 1500 nm (Fig S2(a)), while the resonance for y polarization stays at 800 nm (Fig S2(b)). With increasing size in y direction, the resonance for y polarization shifts from 700 nm to 800 nm (Fig S2(c)), while the resonance for x polarization stays at around 1500 nm (Fig S2(d)). This proves that the resonance peak along orthogonal polarization directions can be tuned independently.