

Supporting information

A planar plasmonic nano-gap and its array for enhancing light–matter interactions at nanoscale

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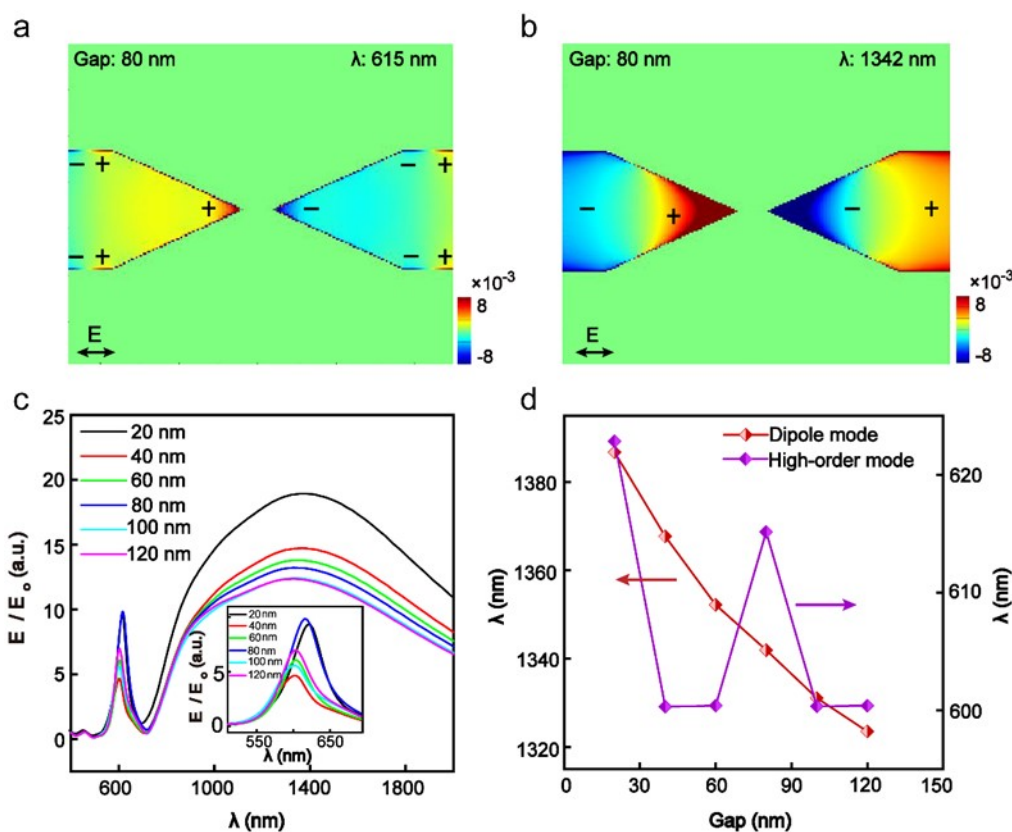


Figure S1. (a, b) Calculated charge distribution contours of a nano-gap with 80-nm gap size. The contours shown in upper panels are obtained on the cross section parallel to the planar nano-gap and passing through its longitudinal axis. The contours are obtained at the wavelength of 615 nm (a) and 1342 nm (b). (c) Simulated electric

field amplitude enhancement as a function of incident light wavelength in nano-gap structures with different gap sizes. Inset: enlarged image of the high order modes. (d) Evolutions of the resonance wavelength for dipole modes (red) and high order modes (purple) against different gap sizes. The data points are extracted from the spectra shown in (c).

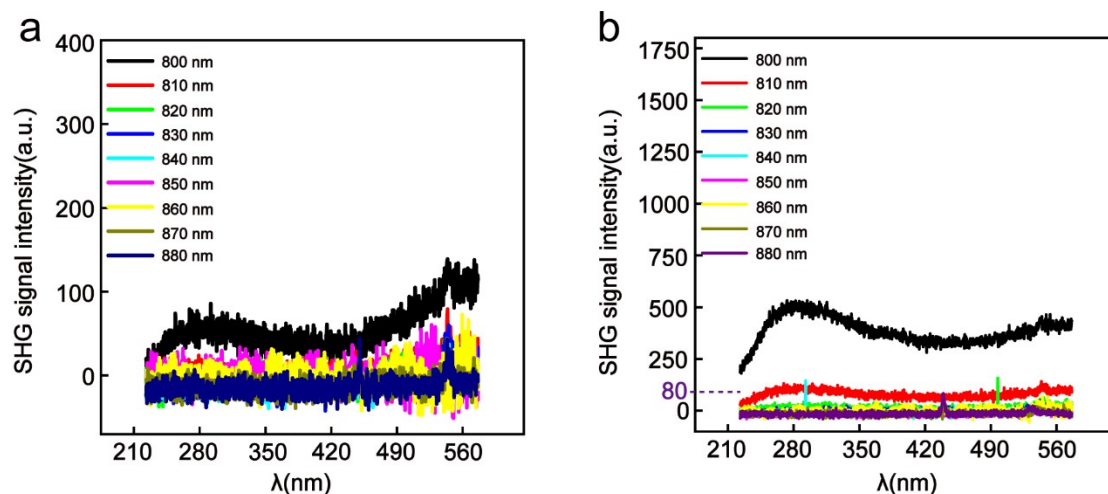


Figure S2. Second harmonic generation spectra of the blank substrate (SiO_2/Si) (a) and planar gold film (b). The spectra were collected at different excitation wavelengths, and the laser intensities were all kept at $8.5 \times 10^5 \text{ W/cm}^2$.

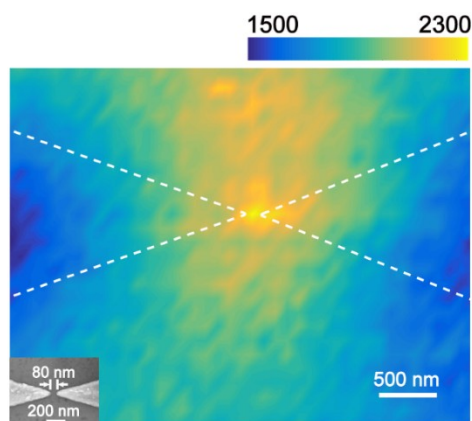


Figure S3. Raman intensity mapping of the nano-gap and adjacent regions. A typical nano-gap with a gap size of 80 nm was investigated. The intensity was monitored at 1187 cm^{-1} . A R6G molecule concentration of 10^{-2} M was employed to prepare the sample. The excitation wavelength is 785 nm. The power density is $4.36 \times 10^4 \text{ W/cm}^2$.

The integration time for each point is 2 s.

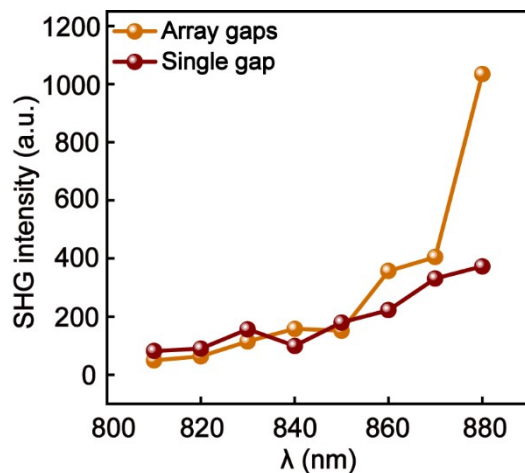


Figure S4. Dependence of SHG spectra on the excitation wavelength collected from an individual nano-gap and its array structure. The gap sizes are all kept at 80 nm. The power density of the excitation laser is kept at 8.5×10^5 W/cm² for all wavelengths.

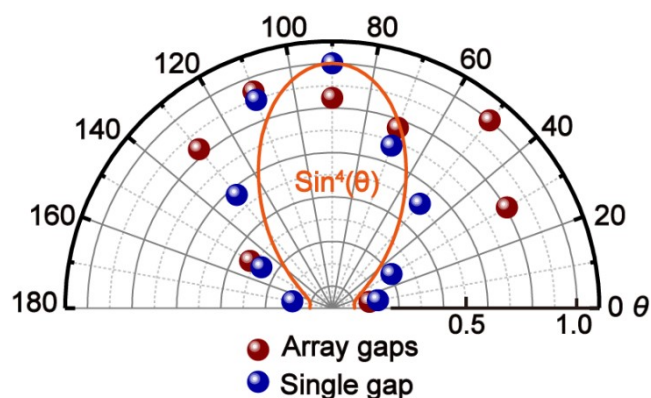


Figure S5. Normalized polarization polar plots of second harmonic generation intensities of a single nano-gap (blue spheres) and nano-gap array (red spheres). Angle θ is defined as the angle between the longitudinal axis of the nano-gap and polarization direction of the incidence light. The second harmonic generation intensities in are obtained 450 nm with the excitation wavelength at 900 nm. The nano-gap has a gap size of 80 nm. The power density of the excitation laser is kept at 8.5×10^5 W/cm² for all polarization directions.

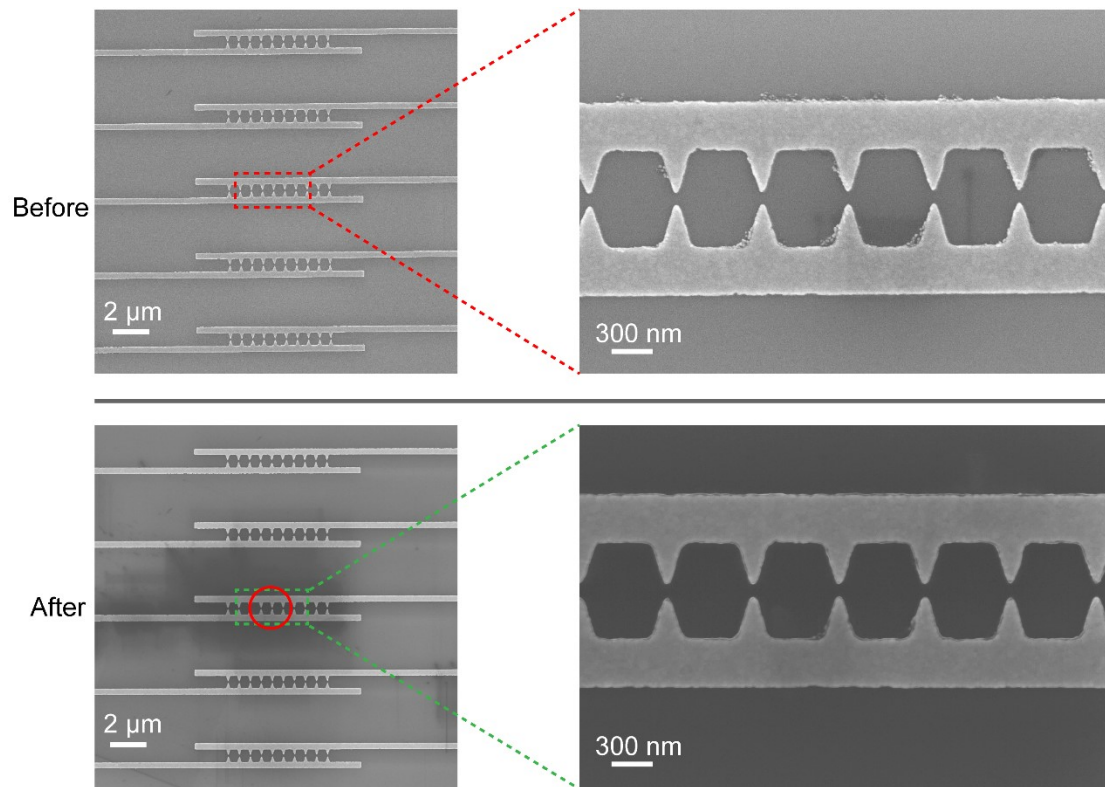


Figure S6. SEM images showing the same nano-gap nanostructures before (upper panel) and after (lower panel) femtosecond laser irradiation. The right images show the enlarged part marked by red and green dashed rectangles for the two samples shown in the left images. The light intensity was 8.5×10^5 W/cm². The red circle indicates the focusing spot.