Second Harmonic Generation from Symmetric and Asymmetric Gold Nanoantennas

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Abstract

We measured second harmonic generation (SHG) from a series of symmetric and asymmetric gold nanoantennas. By systematically varying the arm lengths, we find that the resonance of antenna plasmonic modes with laser excitation results in SHG enhancement in both symmetric and asymmetric nanoantennas. But for asymmetric antennas, large enhancement can occur even without such resonance.

How to Perform SHG Measurement

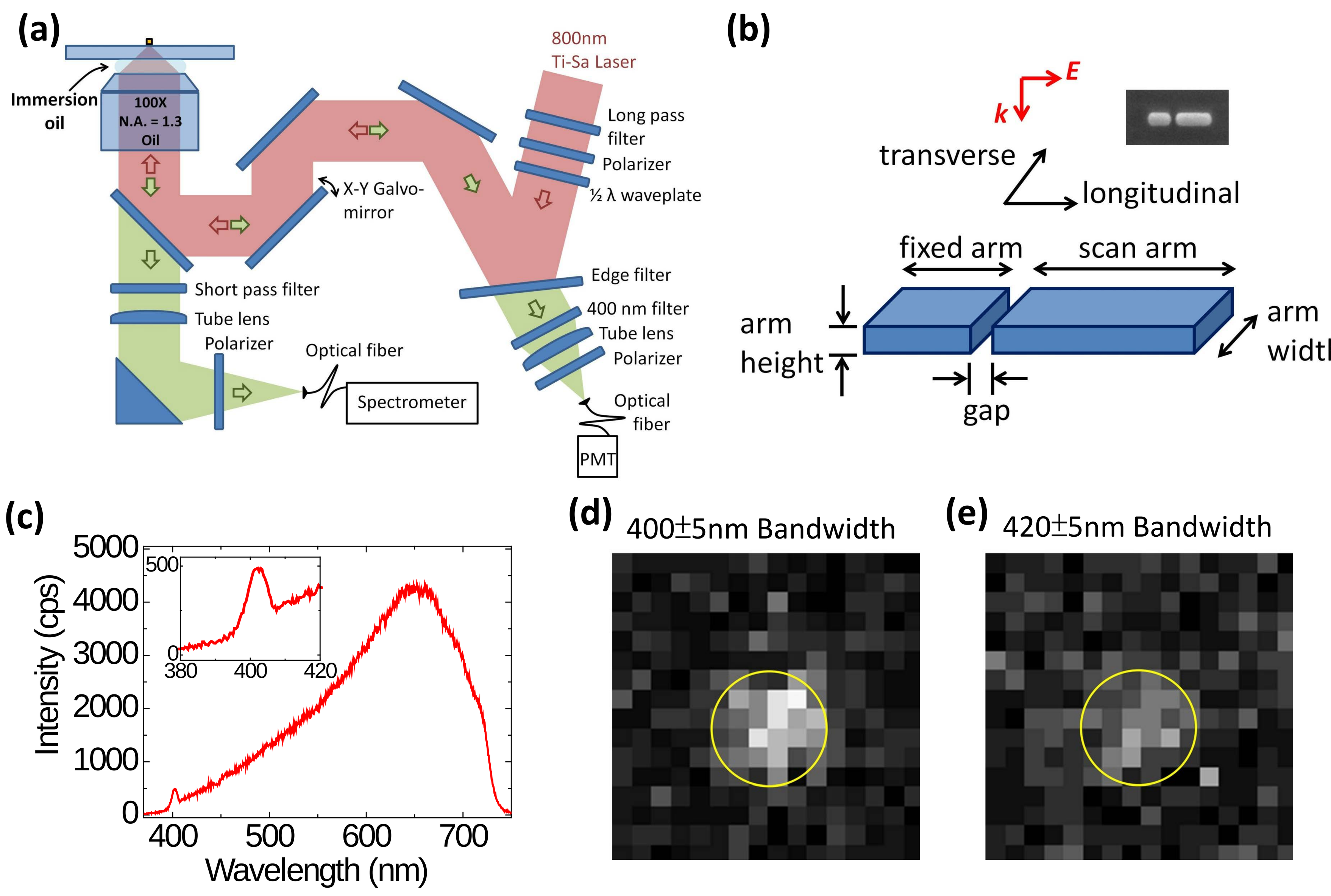

Fig. 1 (a) Diagram of experimental setup. (b) Antenna structure showing the orientation of the excitation polarization is along the longitudinal direction. Inset shows the SEM image of a typical antenna. (c) Emission spectra showing both SHG and TPPL signal. SHG intensity can be determined by subtracting the TPPL background from the spectra. (d) Scan image at 400 nm with 10 nm bandwidth, and (e) Scan image at 420 nm with 10 nm bandwidth. Note that SHG is determined by subtracting (d) from (e) to remove the TPPL background.

SHG Resonance with Antenna Longitudinal Modes

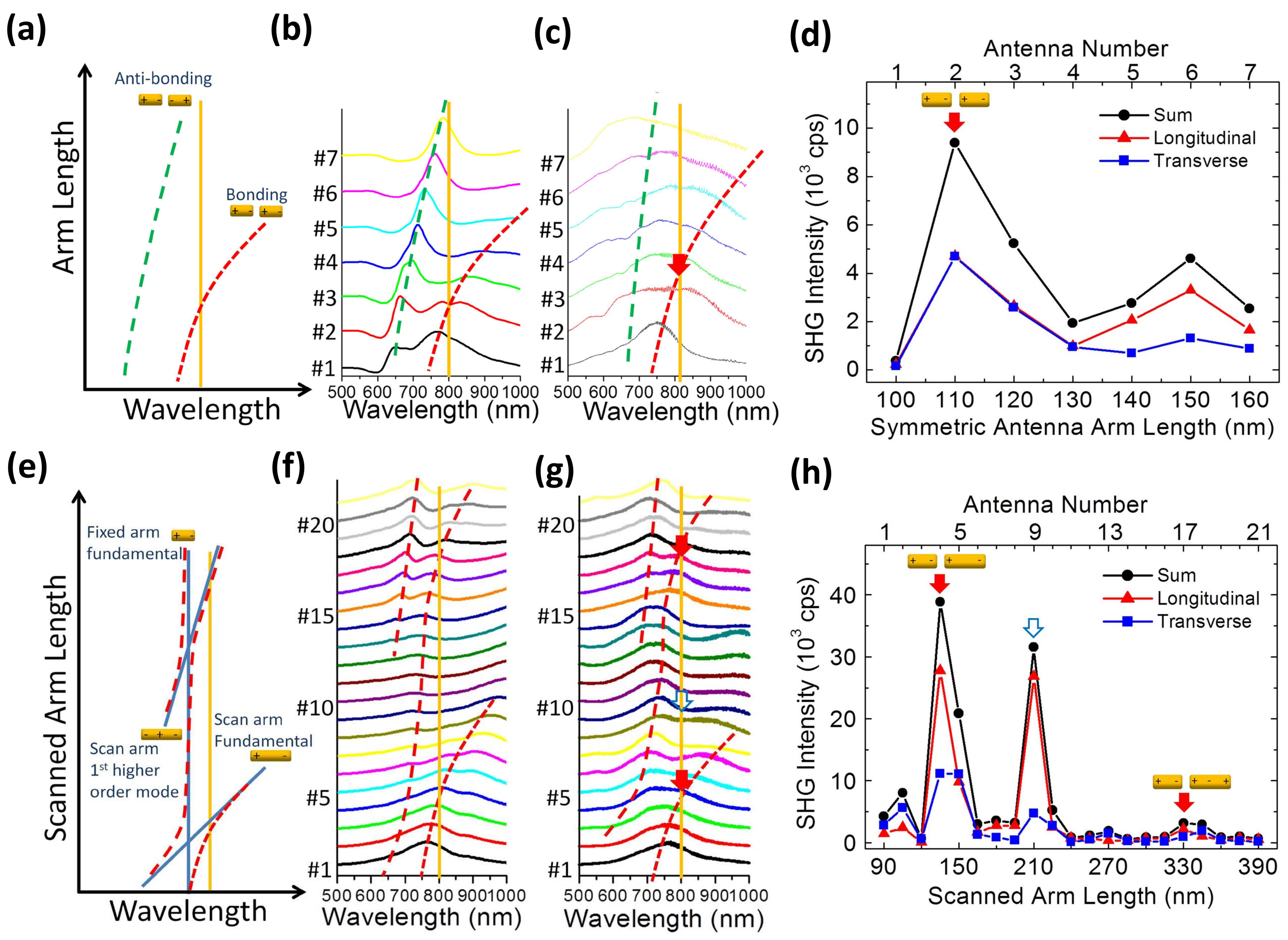

Fig. 2 . (a) Diagram showing the wavelength of the longitudinal bonding (red dash curve) and anti-bonding (green dash curve) resonance for symmetric antenna Array A. Longitudinal spectra for the seven symmetric antennas based on (b) simulated near-field intensity and (c) dark field scattering experiment. The orange line denotes the laser excitation wavelength. (d) SHG intensity as a function of antenna number and scanned arm length. (e) – (h) are the corresponding figures for the asymmetric antenna Array B. The solid red arrows shows the position where the laser excitation overlaps with the longitudinal plasmonic resonance modes. The blue arrow marks the antenna which is not in resonance with the antenna plasmonic modes but show relatively large SHG.

Nanoantenna Sample List

Symmetric gold nanoantennas were fabricated by focused-ion beam milling into single-crystalline gold platelets drop-casted on top of indium tin oxide (ITO) layer coated on a microscope cover glass [1]. Dimension of the three arrays of nanoantennas are shown below:

Array	Antenna Type	Width (nm)	Height (nm)	Gap (nm)	Antenna arm length (nm)	No. of Antennas
A	Symmetric	100	50	20	100 – 160 (step 10)	7
B	Asymmetric	100	50	20	Fixed arm = 120 90 – 390 (step 15)	21
C	Asymmetric	70	50	20	Fixed arm = 120 90 – 400 (step 10)	32

SHG by 800 nm Excitation

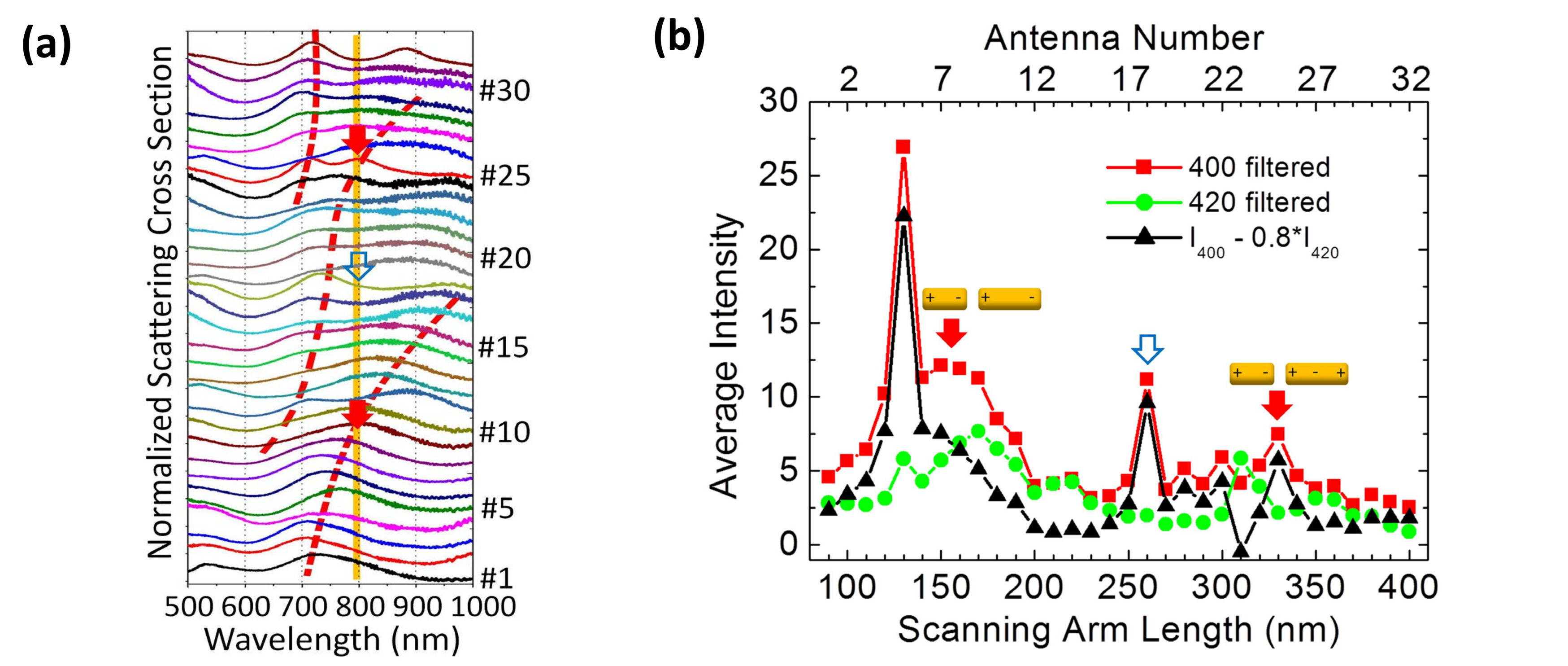

Fig. 3 (a) Longitudinal dark field scattering spectra for the 32 asymmetric antennas of Array C. The orange line denotes the laser excitation wavelength. (b) Longitudinal SHG intensity as a function of antenna number and scanned arm length. As in Array B, the solid red arrows shows the position where the laser excitation overlaps with the longitudinal plasmonic resonance modes. The blue arrow marks antenna #18 which is not in resonance with the antenna plasmonic modes but shows a relatively large SHG.

TPPL by 1064 nm Excitation

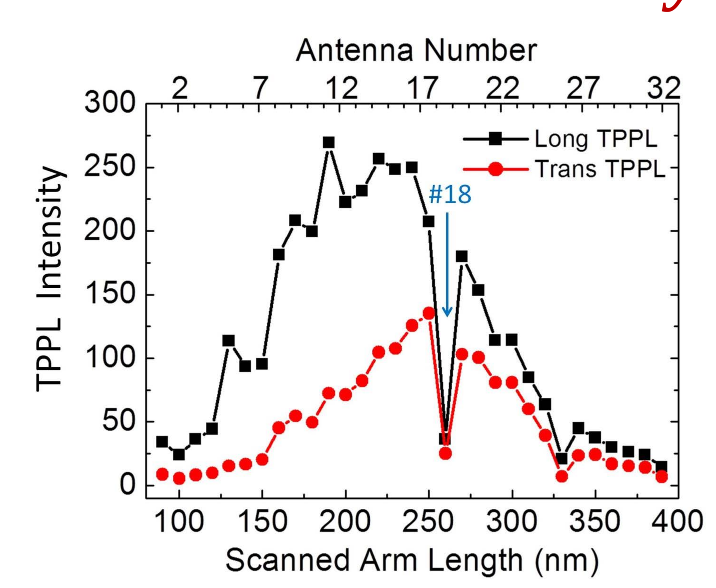

Fig. 4 Longitudinal and transverse TPPL of asymmetric antenna Array C using 1064 nm excitation. Antenna #18 which is not in resonance with 800 nm excitation shows relatively large SHG. Its fundamental mode is close to be in resonance with 1064nm, yet it shows a dip in 1064nm TPPL .

Conclusion

Using two-photon laser excitation in longitudinal configuration, we measured longitudinal and transverse SHG intensity from symmetric and asymmetric gold nanoantennas of varying arm lengths. Comparing antennas where the laser is in resonance with the first order bonding plasmonic mode, the asymmetric antenna produces greater SHG enhancement by a factor of ~4. Furthermore, asymmetric antenna of specific dimensions can result in large SHG enhancement peaks without resonance of laser excitation with any of the antenna's plasmonic modes. The origin of this enhancement is still under investigation.

Acknowledgement

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Reference

[1] J. S. Huang, et al. "Atomically flat single-crystalline gold nanostructures for plasmonic nanocircuitry," Nat Commun. **1**, (2010).