Strong enhancement of second harmonic emission by plasmonic resonances at the second harmonic wavelength

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Abstract

We perform second harmonic (SH) spectroscopy of aluminum nanoantenna arrays that exhibit plasmonic resonances at the second harmonic wavelength. We find that a plasmonic resonance at the second harmonic boosts the overall nonlinear process by more than an order of magnitude. Experimental results can be well understood from a simple model that accounts for the resonant emission process at the second harmonic and the almost off-resonant absorption at the fundamental wavelength.

1. Motivation

- Localized surface plasmon resonances at the fundamental wavelength can significantly boost the optical nonlinearity of metallic nanoparticles due to the strongly confined and enhanced electromagnetic near-field at the fundamental wavelength.
- There has been no quantitative analysis of the exact role of the plasmonic resonances at the harmonic wavelength concerning competing mechanisms of increased radiation efficiency [1,2] and increased absorption [3] at the harmonic wavelength.

2. Experimental setup

Fig. 1. (a) Exemplary measured laser spectrum (red) and its corresponding SH spectrum (blue) with respect to the reflectance spectrum of an Al nanoantenna array (black).

(b) Setup for measuring polarization-resolved SH spectra. FL: fused silica lens, S: sample, Q: quartz crystal, A: analyzer, KG: KG filters.

3. Analytical model

Linear response:

\[ u(\omega) = \frac{A}{\omega^2 - \omega_0^2 + 2i\omega\gamma} \]

P-polarized second-order nonlinear polarization:

\[ P^{(2)}(2\omega) = \chi^{(2)}(\omega) E^2(\omega) \]

Nonlinear response [4,5]:

\[ P^{(2)}(2\omega) = \chi^{(2)}(\omega) E^2(\omega) \]

SH emission enhancement

Miller’s rule:

\[ \chi^{(2)}(2\omega, w, w) = \chi^{(2)}(\omega, \omega, \omega) + 2\chi^{(2)}(\omega, w, w) \]

4. Measurement

Second harmonic spectroscopy

Fig. 2. Measured (a) and modeled (b) polarization-resolved SH spectra together with the corresponding reflectance spectra of four Al nanoantenna arrays with different antenna lengths L. Scale bar: 200 nm.

Quantitative analysis of the blue-shift

Fig. 3. Measured blue-shift of the peak of the p-polarized SH conversion efficiencies with respect to the blue-shift extracted from the model, for the four nanoantenna arrays depicted in Fig. 2.

SH emission enhancement

Fig. 4. (a) Measured p-polarized SH intensity divided by the s-polarized SH intensity for the nanoantenna array with an antenna length of 130 nm. (b) Absolute value squared of the modeled field enhancement factor \(|L_2(\omega)|^2\) of the same nanoantenna array. Linear reflectance spectra are shown in black.

References


5. Conclusion and outlook

We experimentally demonstrate that plasmonic resonances at the SH wavelength can boost the SH signal by more than one order of magnitude, which paves the way to our further research of the following more complex plasmonic nanostructures:

- (a) Chiral nanostructures; (b) Hybrid dielectric nanoantennas; (c) Magneto-plasmonic nanostructures; (d) Metasurfaces.