

# Interplay between nonlinear effects at the nanoscale in engineered optical nanoantennas

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## Abstract

Nanostructures can be exploited to enhance nonlinear optical processes via the strong light confinement by their resonant modes. The sensitive dependence of these modes on the geometry and material composition offers ample opportunities for tailoring the optical response of the system. I report about sum-frequency generation (SFG) by individual dielectric and plasmonic nanocylinders, pumped by two pulsed beams with different photon energies. A rich size- and polarization-dependent behavior is observed, disclosing strategies to manipulate the nonlinear properties of nanoscale systems.

## 1. Introduction

Nanoscale optical integration is nowadays a strategic technological challenge and the ability of generating and manipulating nonlinear optical processes in sub-wavelength volumes is a key-enabling asset for the realization of efficient probes for sensing and photonic sources for the next-generation communication technologies. Yet, to date, confining nonlinear processes beyond the diffraction limit remains a challenging task because phase-matching conditions, which ensure efficient energy transfer from the fundamental to the nonlinear wave in the bulk, cannot be realized at the nanoscale.

Significant efforts have been recently devoted to the investigation of nonlinear optical processes in plasmonic nanoantennas [1,2]. The strong local field enhancement they exhibit makes them promising candidates to overcome the lack of phase-matching at the nanoscale. In this framework, we devised a plasmonic nanoantenna working in the near-infrared region of the electromagnetic spectrum, which allows boosting the second-harmonic generation (SHG) efficiency. This is achieved by optimizing the nanoantenna geometry to feature (i) a doubly resonant response at both the fundamental and emission wavelengths; (ii) a spatial overlap between the modes involved in the process; and (iii) a broken symmetry, to enable efficient SHG through

electric dipole emission. We found that this nanoantenna concept behaves like a strongly coherent nanoscale light source, featuring low photoluminescence in the visible range and a marked third-harmonic generation (THG) along with an intense SHG [3]. We recently applied this concept to an extended array of non-centrosymmetric nanoantennas demonstrating its potential for nonlinear sensing [4].

Due to the relatively low nonlinear conversion efficiencies in these nanoscale systems, a widely accepted approach consists in treating independently different order nonlinear processes such as, for instance, THG and SHG. We show that this paradigm can dramatically fail in these non-centrosymmetric plasmonic nanoantennas, highlighting a SHG-mediated cascaded pathway in THG [5]. This mechanism can be unveiled thanks to the commensurate emission yields of SHG and THG in this nanostructure. We have disentangled this cascading effect from bulk mediated THG based on the characteristic nonlinear emission fingerprint of these processes, which can be identified through the analysis of the overall emission polarization.

## 2. Results

In this context, I will also present the evidence of interference effects between THG and degenerate sum-frequency generation (SFG), produced by combining the original pump beam with a frequency-doubled beam, both on AlGaAs nanopillars and on asymmetric Au nanoantennas. We recently demonstrated that both these nanoscale systems can sustain intense SHG associated with the bulk second-order susceptibility of the material [6]. I will show that, in coincidence with the first order anapole configuration of the AlGaAs system [7], SFG can be controlled by the polarization of the impinging beams, obtaining signal yields higher than THG generated by the original pump beam alone. Conversely, the polarization state of the nonlinear radiation emitted by Au nanoantennas depends by the interplay between SFG and THG. For both systems, due to the degeneracy in the output wavelength, we are able to observe a marked interference between the two

processes, which is mediated by the resonances in these nanosystems.

These results demonstrate the possibility of tailoring the coupling efficiency between nonlinear processes in nanoscale systems nanoantennas and represent a crucial step towards the realization of nanoscale photon conversion and manipulation at room temperature, thereby opening new possibilities in quantum optics and in nonlinear optical sensing.

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