Metal-Dielectric Nanodimers with Hybridized Resonances Probed by Second-Harmonic Polarization

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Abstract: We fabricate hybrid nanodimers made of gold and barium titanate nanoparticles by a pick-and-place technique. By overlapping their resonances, we achieve 100-times enhancement of the second-harmonic signal at the hybridized mode while reshaping its polarization.

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1. Introduction

Most studies of nonlinear effects at the nanoscale focus on top down fabricated metallic structures (Au, Ag, etc.) [1]. More recently, semiconductors (such as Si, Ge, III-V) nanostructures, also called all-dielectrics, showed low losses and multiple resonances due to Mie scattering that can be exploited for linear and nonlinear nanophotonics [2,3].

Here, we aim at combining the high field localization of plasmonics with the optical properties of all-dielectric systems, in particular the bulk nonlinearities of the broadly transparent non-centrosymmetric material barium titanate ($BaTiO_3$ or BTO). To obtain such a multifunctional structure, we assemble nanoparticles of different nature by a bottom-up pick-and-place technique combining the advantages of metals and of dielectrics at the same time. We demonstrate hybrid nanodimers made of gold and barium titanate nanoparticles, where the electric dipole mode of the gold nanoparticle overlap with the magnetic dipole mode of the $BaTiO_3$ nanoparticle. We take advantage of this interplay between the resonances to enhance the second-harmonic generation (SHG) and reshape the polarization dependence of the second-harmonic signal. We achieve two orders of magnitude enhancement of the SHG efficiency at the hybridized mode compared to single $BaTiO_3$ nanoparticles of similar sizes.

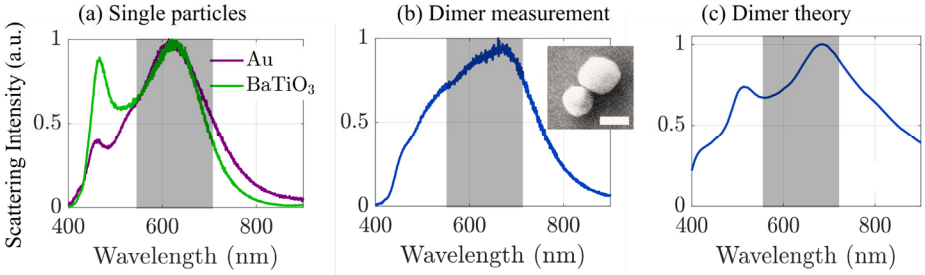

Fig. 1. (a) Experimentally measured forward linear scattering of a single 210 nm size gold (purple) and a single 280 nm size barium titanate (green) nanoparticles. The highlighted region of the spectra is showing the spectral range of the hybridized mode. (b) Measured forward linear scattering of one nanodimer with sizes of Au/BTO particles of 213 nm / 314 nm. Inset: scanning electron micrograph (SEM) the nanodimer, scale bars are 200 nm. (c) Modeled forward linear scattering of a nanodimer with Au (212 nm) and BTO (280 nm) nanoparticles.

2. Fabrication and Linear Characterization

We first measure the linear scattering from the individual nanoparticle. The typical features of a single $BaTiO_3$ nanoparticle are a wide peak centered around 620 nm [the magnetic dipole (MD) resonance], which perfectly overlap with the electric dipole (ED) of the gold [see Fig. 1(a)]. Another sharp peak is visible around 460 nm on the measured linear scattering curve and belongs to the magnetic quadrupole (MQ) resonance of $BaTiO_3$, see Fig. 1(a).

Then, we fabricated the hybrid nanodimers by the pick-and-place technique from gold and barium titanate nanoparticles, see the inset in Fig. 1(b) [4,5]. The single nanoparticles are electrostatically manipulated under the beam of an electron microscope with a sharp metallic tip and assembled on an indium tin oxide covered glass

substrate into hybrid nanodimers. In the linear scattering measurements of the hybrid dimer, we can differentiate the main peak at 640°nm [see Fig.1 (b)], which correspond to the modeled spectrum [Fig. 1(c)].

3. Reshaping of the nonlinear polarization response

Then, we perform second-harmonic generation (SHG) measurements by impinging a femtosecond near infrared laser onto the same dimer. First, we measure wavelength dependent second-order responses [the SHG signal on Fig. 2(a)] over a wide range (800 to 1400 nm) using a Ti: Sapphire oscillator (range with full red circles) and an optical parametric oscillator (range with empty circles), with corresponding simulations. Then, we measure the polarization dependence [Fig. 2(b)] of the second-order responses for an isolated dimer, for the same wavelength range. We show a strong reshaping of the SHG polar responses of the hybrid dimer at the hybridized mode of the plasmon resonance of Au and the Mie resonance of the $BaTiO_3$. Indeed, a clear polar orientation changes is observed from 550 nm onward (highlighted in red), which corresponds to the linear scattering measurements [see Fig. 1(b)].

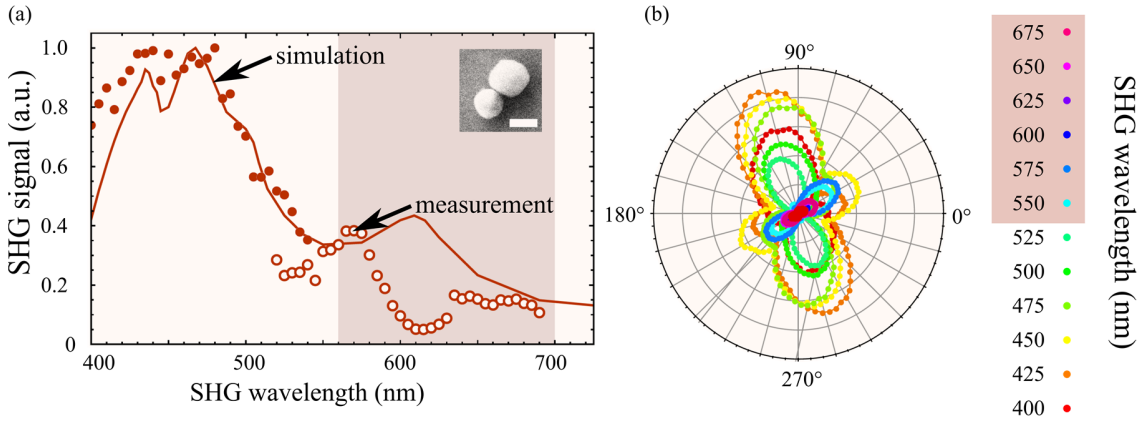

Fig. 2. (a) Normalized SHG signal of the nanodimer (inset) as a function of the second-harmonic wavelength. The solid red line shows the modeled spectrum and the red dots are the measurements points (filled laser, empty OPO range). SEM micrograph of the nanodimer in the inset, scale bar 200 nm. (b) Measured polar dependence of the normalized SHG signal from the same nanodimer. Reshaping range is highlighted in red.

We also estimate the SHG enhancement at the different wavelength by comparing the signal of the hybrid dimer to an isolated $BaTiO_3$ nanoparticle. We find that for the region below 550 nm, we have a one order of magnitude enhancement and above 550 nm, in the hybridized mode range, up to two orders of magnitude.

4. Conclusion

We have shown strong nonlinear effects in a well-controlled bottom-up fabrication scheme adapted to nanoparticles independently of their composition or shapes. Such a flexible fabrication approach can have impact on nonlinear optics and dielectric meta-optics, especially for the polarization control and switching at different wavelengths.

5. References

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