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Abstract- Optical nonlinearities are intrinsically weak, but can be strengthened in scenarios that provide field enhancement mechanisms. Hybrid structures combining the high inherent material nonlinearity of semiconductors along with plasmonic-induced strong fields are promising platforms for on-chip nonlinear optics. However, due to their complexity, understanding the mechanisms involved is sometimes challenging. Here, the spectral response of metal dimers loaded with an ITO nanoparticle at their gap is investigated numerically and experimentally to provide an understanding and design rules.

Nonlinear metasurfaces is a thriving research field, which creates transformative opportunities for manipulating light in a very compact way [1]-[4]. Since the length scales are short, nonlinear interactions are enhanced by exploiting materials with large nonlinearity in combination with plasmonic strong fields [3], [5]-[8]. Realizing experimentally this hybrid approach has been made possible thanks to advances in nanofabrication.

Unfortunately, the recently proposed hybrid structures are relatively complex and a full understanding of the mechanisms involve in the high nonlinear efficiency observed is yet to be provided. To address this, we investigate here different metal dimers loaded with an ITO nanoparticle at their gap (see Fig. 1(a,b)) with a systematic combined approach involving numerical and experimental spectral analysis. The methodology shown here allows us to trace the origin of the nonlinear signal to the semiconductor or metal.

All dimers (with and without ITO nanoparticle) investigated were fabricated by a combination of etch-down and lift-of methods. They were modelled with a full-wave finite-difference time-domain technique and their linear and nonlinear responses were studied by Fourier-transform infrared spectroscopy and a custom nonlinear inverted microscope, respectively.

Figure 1(c) shows the experimental third harmonic intensity for a nano-dipole with and without the ITO nanoparticle at its gap. Remarkably, the hybrid metasurface shows a six-fold intensity. The origin of this enhancement could be either a favorable change in the resonant condition of the nano-dipole or the boost of the ITO nonlinearity by the strong field expected at the gap of the nano-dipole. To resolve this, we compare the experimental normalized spectra to the numerically-computed ones (see Fig. 1(d)). The nonlinear simulations allow us to switch on and off the third order susceptibility of the metal and ITO independently, which proves to be a very powerful tool. Both measurements and simulation results show that the third harmonic linewidth of the bare plasmonic configuration is larger than that of the hybrid nanostructure when considering the ITO nanoparticle nonlinear. Meanwhile, the simulations reveal that the hybrid and bare nanostructures exhibit identical spectra when ITO is considered linear. This clearly indicates that the third harmonic signal is generated in our system from the ITO nanoparticle.

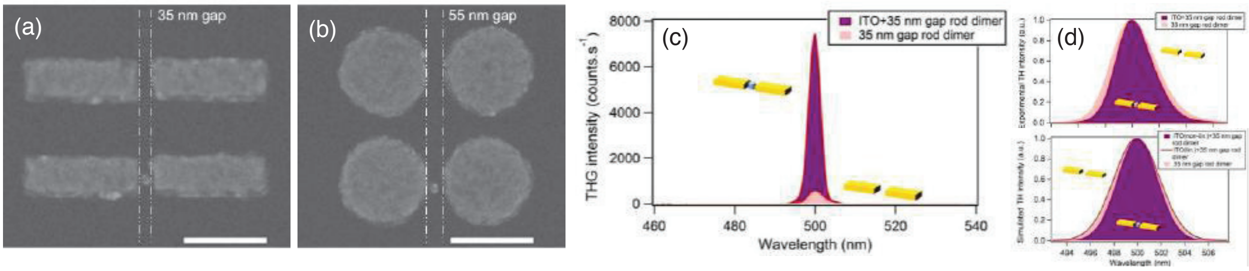

Figure 1. (a) SEM image of a 35 nm gap nanorod dimer without (top) / with (bottom) a ~25 nm ITO nanoparticle at its gap; scale bar: 200 nm. (b) SEM image of a 55 nm gap nanocylinder dimer without (top) / with (bottom) a ~25 nm ITO nanoparticle at its gap; scale bar: 200 nm. (c) Experimental THG spectra from the nano-dipoles (a,b) under parallel polarized excitation. (d) Experimental, top, and simulated, bottom, normalized THG spectra from a 35 nm gap nanorod dimer with and without the ITO nanoparticle at its gap under parallel polarized excitation.

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