

Dynamics of Non-Linear Hybrid ITO-Plasmonic Metasurfaces

Navarro-Cia, Miguel; Aouani, Heykel; Rahmani, Mohsen; Maier, Stefan A.

License:

None: All rights reserved

Document Version

Peer reviewed version

Citation for published version (Harvard):

Navarro-Cia, M, Aouani, H, Rahmani, M & Maier, SA 2016, Dynamics of Non-Linear Hybrid ITO-Plasmonic Metasurfaces. in *META'16 in Malaga: The 7th International Conference on Metamaterials, Photonic Crystals and Plasmonics Proceedings*. META Proceedings: META'16 in Malaga, META Conferences, Spain, pp. 487-488, META'16, Malaga, Spain, 25/07/16.

[Link to publication on Research at Birmingham portal](#)

Publisher Rights Statement:

In addition, authors are encouraged to post and share their work online (e.g., in institutional repositories or on their website) at any point before and after the conference. <http://metaconferences.org/ocs/index.php/META16/index/about/submissions#copyrightNotice>

General rights

Unless a licence is specified above, all rights (including copyright and moral rights) in this document are retained by the authors and/or the copyright holders. The express permission of the copyright holder must be obtained for any use of this material other than for purposes permitted by law.

- Users may freely distribute the URL that is used to identify this publication.
- Users may download and/or print one copy of the publication from the University of Birmingham research portal for the purpose of private study or non-commercial research.
- User may use extracts from the document in line with the concept of 'fair dealing' under the Copyright, Designs and Patents Act 1988 (?)
- Users may not further distribute the material nor use it for the purposes of commercial gain.

Where a licence is displayed above, please note the terms and conditions of the licence govern your use of this document.

When citing, please reference the published version.

Take down policy

While the University of Birmingham exercises care and attention in making items available there are rare occasions when an item has been uploaded in error or has been deemed to be commercially or otherwise sensitive.

If you believe that this is the case for this document, please contact UBIRA@lists.bham.ac.uk providing details and we will remove access to the work immediately and investigate.

Dynamics of Non-Linear Hybrid ITO-Plasmonic Metasurfaces

M. Navarro-Cía^{1*}, H. Aouani², M. Rahmani^{2,3}, and S. A. Maier²

¹School of Physics and Astronomy, University of Birmingham, Birmingham B15 2TT, UK

²The Blackett Laboratory, Department of Physics, Imperial College London, London SW7 2AZ, UK

³Nonlinear Physics Centre, Research School of Physics and Engineering, The Australian National University, Canberra, ACT 0200, Australia

*corresponding author: m.navarro-cia@bham.ac.uk

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-off 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.

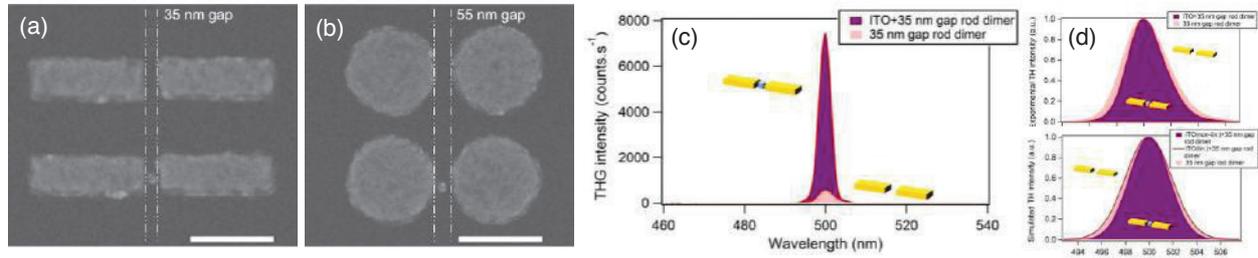


Figure 1. (a) SEM image of a 35 nm gap nanorod dimer without (top) / with (bottom) a ~ 25 nm ITO nanoparticle at its gap; scale bar: 200 nm. (b) SEM image of a 55 nm gap nanocylinder dimer without (top) / with (bottom) a ~ 25 nm ITO nanoparticle at its gap; scale bar: 200 nm. (c) Experimental THG spectra 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.

Acknowledgements. This work was funded by the Engineering and Physical Sciences Research Council (EPSRC) through the Active Plasmonics programme, the Leverhulme Trust, the US Army International Technology Centre Atlantic (USAITC-A) and the Office of Naval Research (ONR and ONR Global). M.N.-C. is supported by a Birmingham Fellowship.

REFERENCES

1. Zhang, Y., N. K. Grady, C. Ayala-Orozco, and N. J. Halas, “Three-Dimensional Nanostructures as Highly Efficient Generators of Second Harmonic Light,” *Nano Lett.*, Vol. 11, No. 12, 5519-5523, 2011.
2. Schumacher, T., K. Kratzer, D. Molnar, M. Hentschel, H. Giessen, and M. Lippitz, “Nanoantenna-enhanced ultrafast nonlinear spectroscopy of a single gold nanoparticle,” *Nature Comm.*, Vol. 2, 333, 2011.
3. Navarro-Cía, M. and S.A. Maier, “Broad-Band Near-Infrared Plasmonic Nanoantennas for Higher Harmonic Generation,” *ACS Nano*, Vol. 6, No. 4, 3537–3544, 2012.
4. Aouani, H., M. Navarro-Cia, M. Rahmani, T. P. H. Sidiropoulos, M. Hong, R. F. Oulton, and S. A. Maier, “Multiresonant Broadband Optical Antennas As Efficient Tunable Nanosources of Second Harmonic Light,” *Nano Lett.*, Vol. 12, No. 9, 4997-5002, 2012.
5. Harutyunyan, H., G. Volpe, R. Quidant, and L. Novotny, “Enhancing the Nonlinear Optical Response Using Multifrequency Gold-Nanowire Antennas,” *Phys. Rev. Lett.*, Vol. 108, No. 21, 217403, 2012.
6. Aouani, H., M. Rahmani, M. Navarro-Cía, and S. A. Maier, “Third-harmonic-upconversion enhancement from a single semiconductor nanoparticle coupled to a plasmonic antenna,” *Nature Nanotech.*, Vol. 9, 290-294, 2014.
7. Metzger, B., M. Hentschel, T. Schumacher, M. Lippitz, X. Ye, C. B. Murray, B. Knabe, K. Buse, and H. Giessen, “Doubling the Efficiency of Third Harmonic Generation by Positioning ITO Nanocrystals into the Hot-Spot of Plasmonic Gap-Antennas,” *Nano Lett.*, Vol. 14, NO. 5, 2867-2872, 2014.
8. Aouani, H., M. Navarro-Cía, M. Rahmani, and S. A. Maier, “Unveiling the Origin of Third Harmonic Generation in Hybrid ITO–Plasmonic Crystals,” *Adv. Opt. Mat.*, Vol. 3, 1059-1065, 2015.