Field enhanced harmonic generation in nanostructures

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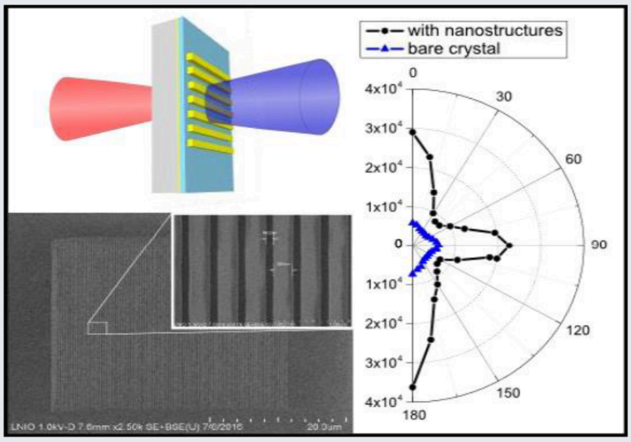
Non-linear optical processes such as high harmonic generation (HHG) rely on strong light-matter interaction. This can be achieved by squeezing light in space, compressing light in time, or by both. Traditionally HHG occurs in rare-gas atoms at an intensity of about 1014W/cm^2 achieved by means of mJ-level amplified lasers. This clearly prevents the development of hand-held systems. In 2011, Stanford and Ohio State Universities put in evidence for the first time HHG from a bulk semiconductor crystal. This experiment started a vast effort by numerous laboratories towards the understanding of the phenomenon, proposing different models and pushing experimental setups. We have recently found numerically a new effect so-called "near field phase matching" for the control of attosecond pulses using plasmonic field enhancement [1]. Following this, recent experimental findings at CEA [2,3] report on the third harmonic generation (THG) from two original nanostructures in the sub-terawatt regime. We first investigate a bi-layer structure composed of a thin ZnO crystal and an out-coupling gold layer with a 2D array of nanoholes showing extraordinary transmission. The THG signal is strongly amplified and shows polarization independent field amplification. To increase the light coupling and polarization selectivity we then introduce a novel hybrid Metal-Dielectric-Metal (MDM) metasurface that induces a boosted amplification of the THG compared to the bi-layer structure (see Fig. 1). Indeed, the Fabry-Perot plasmonic resonator allows a high confinement and amplification of the laser electric field over a large volume of the SiO2 dielectric film. THG signal is amplified by one order of magnitude. The THG polarization dependence with respect to the fundamental laser allows an ON/OFF switch of the enhancement and confirms the plasmonic origin of the amplification (see polarisation dependence in Fig 1.). However these experiments also suggest damage threshold and lifetime limitations of plasmonic metallic nano-structures at IR wavelength. To go beyond, we have designed an all-semiconductor field nano-amplifier based on an array of ZnO nanocones with resonance in the mid-infrared [4]. The laser intensity is enhanced locally by more than one order of magnitude as supported by FDTD simulation. Strong amplification of the 7th harmonic from our all 3D waveguide are reported [4]. Lifetime of such structure exceeds a week which opens applications of this novel nano-source of hot photons.



Example of a MIM structure for cHHG

[1] T. Shaaran, R. Nicolas, B. Iwan, M. Kovacev, H. Merdji, *Nano-plasmonic near field phase matching of attosecond pulses*, Scientific Reports 7, 6356 (2017)

[2] L. Shi, B. Iwan, R. Nicolas, *Self-optimization of plasmonic nanoantennas in strong femtosecond fields*, Optica 4, 1038 (2017)

[3] R. Nicolas, L. Shi, D. Franz, Q. Ripault, B. Iwan, W. Boutu, M. Kovačev, H. Merdji, *Plasmon-Amplified Third Harmonic Generation in Metal-Dielectric-Metal Resonators and Nanoholes Arrays*, Submitted to ACS Nano (Oct. 2017)

[4] D. Franz, R. Nicolas, W. Boutu, L. Shi, Q. Ripault, M. Kholodtsova, B. Iwan, U. Elu Etxano, M. Kovacev, J. Biegert, H. Merdji, *Amplification of high harmonics in 3D semiconductor waveguides*, Submitted to Light Science and Applications (Sept. 2017)

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