Electron dynamics and nonlinearities in optical nanoantennas

Javier Aizpurua

Center for Materials Physics (CSIC-UPV/EHU) and Donostia International Physics Center (DIPC), E-20018 San Sebastian

A nanoscale gap between two metallic nanoparticles is an ideal platform to exploit the interplay between electron currents and photonic excitations. The capability of a metallic gap to enhance the amplitude of the induced plasmonic field produces a variety of non-linear effects [1,2] which can be exploited in different applications in optoelectronics, such as optical rectification, light emission driven by DC currents, or high-harmonic generation, among others. Furthermore, in ultranarrow gaps, tunneling of electrons at optical frequencies has been found to screen the plasmonic bonding gap resonance, and activate a new distribution of optical modes characterized by optical charge transfer [3].

Here we address the complex ultrafast dynamics of photoelectrons driven by single-cycle optical pulses in nanoscale gaps. By solving Schrödinger equation within the framework of Time-Dependent Density Functional Theory (TDDFT), the currents of the electrons photoemitted across the gap can be monitored, identifying ultrafast electron bursts where electron quiver occurs when the amplitude of the induced field at the plasmonic gap is reversed within the optical cycle. The properties of the amplitude and carrier-envelope phase (CEP) of the incident pulse, together with the gap length determine the complex electron dynamics [4,5,6] (see Fig. 1).

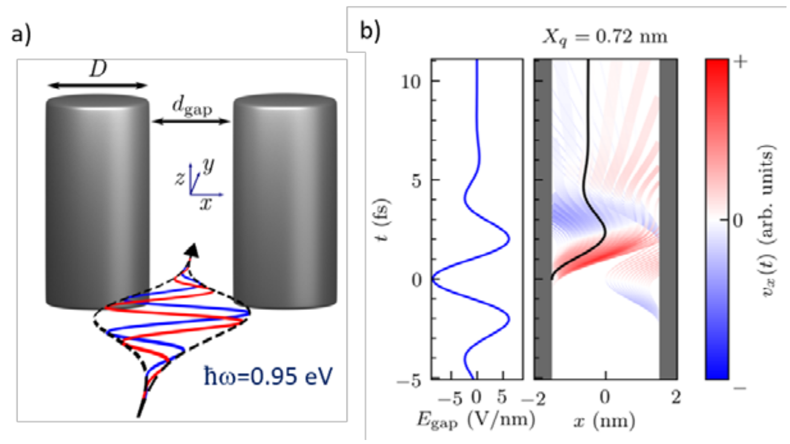

Fig. 1: a) *Schematics of a gap nanoantenna constituted by two metallic nanowires, excited by a single-optical cycle pulse of energy* $\hbar\omega$=0.95 eV. b) Evolution in time of the electromagnetic field, E_{gap}, at the nanoantenna gap (left), and of the emitted photoelectrons velocity (right panel).

Experimental measurements of the current autocorrelations for pairs of such pulses with controlled relative delay between them, confirms the ultrafast dynamics of the photoelectrons in the gap and its complexity.

References:

[1] D.C. Marinica et al., *Nano Lett*. **12**, 1333 (2012).
[2] A. Babaze et al., *ACS Photonics* **7**, 701-713 (2020) .
[3] K.J. Savage et al., *Nature* **491**, 574 (2012).
[4] G. Aguirregabiria et al., *Faraday Discussions* **214**, 147-157 (2019).
[5] M. Ludwig et al., *Nature Physics* **16**, 341–345 (2020) .
[6] M. Ludwig et al., *Phys. Rev. B* **101**, 241412(R) (2020).