Extreme Nanocavity-Enhanced Molecular Spectroscopy

Javier Aizpurua

Center for Materials Physics (CSIC-UPV/EHU) and DIPC, Paseo Manuel lardizabal 5, Donostia-San Sebastián, Spain
aizpurua@ehu.eus

A plasmonic nanogap is a superb configuration to explore the interplay between light and matter. Light scattered off, or emitted from a nanogap carries the information of the surrounding electromagnetic environment with it. This situation becomes even more appealing when a single molecule is located in such a plasmonic cavity or in its proximity, with the molecule playing an active role either in the electromagnetic coupling with the nanocavity, or even participating in processes of charge injection and transfer, as revealed through cutting-edge molecular spectroscopy. In this talk, the process of interaction between a molecular emitter and a nanocavity will be addressed by means of different theoretical frameworks which involve aspects of condensed matter physics [1], quantum chemistry [2], and cavity-quantum electrodynamics [3]. A battery of methodologies to address the dynamics of electrons photo-emitted from nanogaps, ultra resolution in atomic-scale photoluminescence, or non-linear regimes in molecular optomechanics will be described, and many of the theoretical insights obtained will be interpreted in the context of state-of-the-art experimental results in nanocavity-enhanced molecular spectroscopy.

References

[1] A. Babaze, R. Esteban, A. G. Borisov, and J. Aizpurua, Nano Lett., 21 (2021) 8466-8473.
[2] A. Roslawska, T. Neuman, B. Doppagne, A. G. Borisov, M. Romeo, F. Scheurer, J. Aizpurua, and G. Schull, Phys. Rev., 12 (2022) 011012.
[3] R. Esteban, J. J. Baumberg, and J. Aizpurua, Acc. Chem. Res., 55 (2022) 1889-1899.

Figures

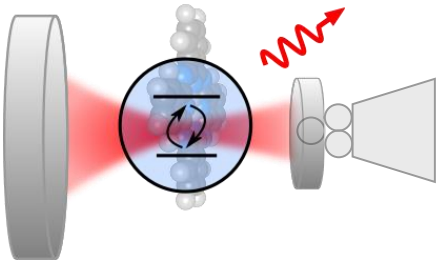

Figure 1: Molecular electronic excitation constituted by a two-level system, coupled to an optical picocavity constituted by an atomic protrusion at the tip of a scanning tunneling microscope over a surface. Light emission (depicted with a red arrow) is produced in such a picocavity, which allows for identifying atomically-resolved intramolecular intensity of light emission, Purcell effect, and Lamb shift.