Driving Nanophotonics to the Atomic Scale

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Plasmonic nanocavities formed at the junction of two metallic interfaces provide a great platform to explore atomic-scale morphologies and complex photochemical processes by optically monitoring the excitation of their intense surface plasmonic modes. In recent years, optical spectroscopy of these cavities has proven to be extremely sensitive to atomic-scale features that determine the chemistry and the optoelectronics in the gaps. In this regime, classical theories fail to address the fine details of the optical response, and more sophisticated quantum theories are needed [1].

We develop a quantum atomistic description of the total energy and the optical response of metallic nanojunctions within Time-dependent Density Functional Theory (TDDFT) that allows to properly address atomic-scale features of the field enhancement and localization of light at the gap [2] (see Figure below), providing new keys to understand the ultraresolution obtained in molecular vibrational spectroscopy [3]. The small effective volumes associated to these new type of optical 'pico-cavities' are of particular interest in quantum nanooptics, as they provide large values of the coupling strength between the photons in the cavity and excitons of an emitter or mechanical vibrations of a molecule. We exploit this opportunity of large coupling in molecular optomechanics of single molecules [4,5].

The atomic scale is a challenging regime in plasmonics, progressively achieved experimentally. New theoretical tools, as those pointed out here, become necessary to understand this regime, and implement new concepts in optoelectronics, quantum nanooptics, and field-enhanced spectroscopy.

References

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Figure

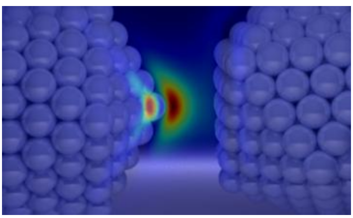

Figure: Time-Dependent Density Functional Theory (TDDFT) calculation of the electromagnetic field localized around atomic-scale features [2] in a metallic 'pico-cavity' to be exploited in molecular spectroscopy, nanoscale optomechanics, and optoelectronic transport. The blue spheres represent sodium atoms building the plasmonic cavity. Bright colours from red to blue express the intensity of the electromagnetic field. (in collaboration with D. Sánchez-Portal's group).