

Optoelectronics in plasmonic nanogaps

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Abstract

A plasmonic nanogap is an ideal platform to explore and test quantum effects in the optical response of nanoscale structures. As the separation between interfaces in a nanogap becomes below nanometric distances, the optical response of the system enters a strong nonlocal regime where the quantum nature inherent to the coherent oscillation of interacting electrons becomes apparent (see schematics of a typical plasmonic gap in the figure). We have developed full quantum mechanical calculations within time-dependent density functional theory (TDDFT) to address nonlocal effects in plasmonic gaps [1]. By doing so, we have identified a tunneling regime for separation distances of the interfaces below 0.5 nm, which totally modifies the spectral fingerprints of the cavity [2]. Quantum tunneling screens plasmonic modes localized at the cavity and establishes charge transfer across the gap producing lower energy modes of the optical response, as recently demonstrated experimentally [3]. By applying both a full quantum mechanical framework as well as a semiclassical approach, we explore the interactions between photons and electrons in these subnanometric gaps in a variety of situations of practical interest in plasmonics and in optoelectronics. Among other topics of interest, we consider the presence of an emitter in the nanogap under the strong coupling regime where hybrid plexcitonic modes are produced, identifying the situation where resonant electron transfer (RET) can be established. Moreover, we explore subnanometric gaps produced by novel materials such as rigid organic molecules producing controlled aggregates for field-enhanced spectroscopy [4], or 2D materials such as graphene, MoS₂ or CdSe, where a distinctive optical response is obtained within the nanogap [5]. The results presented here stress the importance of the plasmonic gap as a canonical structure in nanophotonics, giving rise to the emergence of new optoelectronic processes that can be tailored on demand.

References

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Figure

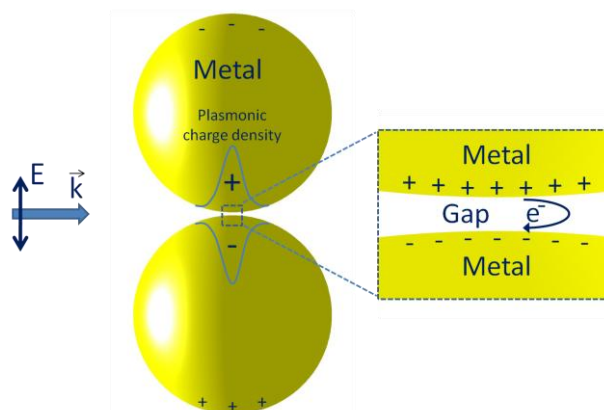


Figure: Nanooptics in a plasmonic gap. Sketch of the surface charge density associated to the surface plasmon mode at the nanogap formed by a metallic dimer, excited by a planewave linearly polarized along its axis (E) that propagates with k vector. The interaction between the two nanoparticles localizes and enhances the plasmonic field at the nanogap. For nanometric and subnanometric separation distances (see zoom-in), an interplay between the plasmon charge densities induced by light and electronic states e^- shows a rich and complex variety of optoelectronic processes with potential for technological application.