Optoelectronic response of quantum emitters 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. 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. Furthermore, we consider the presence of an emitter in the nanogap, as depicted in the figure below, under the strong coupling regime where hybrid plexcitonic modes are produced. Once the plexcitonic response obtained within classical and quantum approaches are proven to be consistent, we adopt the classical approach to incorporate resonant electron transfer (RET) into the optoelectronic response of the cavity-emitter system by including the electron transfer rates as an extra broadening into the description of the emitter. As observed in the spectra of the figure, the spectral fingerprint of the emitter is lost when it is located at short distances from the interfaces (below 1.2 nm). At very close distances from the metal interfaces, RET from the excited state of the emitter into the continuum of metallic states occurs (see scheme of states in the figure), quenching the plexcitonic fingerprint [3]. This is an effect intrinsically different to the classical quenching of emission by classical interaction with surface plasmons. The results presented here emphasize the importance of quantum effects in the coupling between single emitters and plasmonic antennas.

References

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Figure

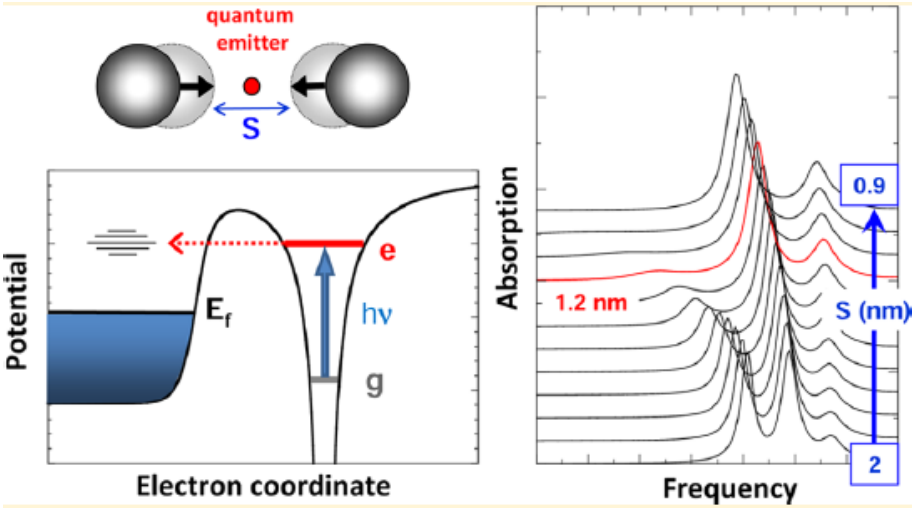

Figure: Top left: Scheme of a quantum emitter located at the gap of a plasmonic dimer with separation distance S. Bottom left: Process of resonant electron transfer (RET) depicted as a red arrow between the excited (e) state of the emitter and the continuum of states of the metal, related to the Fermi Energy E_f. Light of energy hν drives the emitter from the ground state (g) to the excited. Right: Absorption spectra of the coupled emitter-gap system for different separations of the gap (0.9nm-2nm). For small separation distances, the emitter's fingerprint is quenched.