Nanohybrid architectures for strong light-matter interaction

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Light-matter interaction is at the root of several phenomena of everyday life and typically involves an energy exchange between a quantum emitter (dye molecules, low-dimensional semiconductors, etc.) with an external electromagnetic field. The possibility of controlling the coupling of photons to material quasiparticles and/or collective excitations (e.g. excitons, phonons, and plasmons) can drastically reshape the quantum emitter properties [1]. In particular, by properly engineering the coupled system, an exchange rate of energy faster than any other competing relaxation process can be promoted: the system enters in the so called "strong-coupling" regime, and new hybrid light/matter states are formed [2,3]. Novel and distinctive physico-chemical properties can thus emerge in the hybrid architecture, with significant implications in various fields, spanning from quantum information and biophysics to non-linear photonics and polariton chemistry [4,5].

Here, we focus on hybrid platforms composed of nanopatterned metallic surfaces and quantum emitters featuring strong coupling between fundamentally distinct excitations. Both steady-state and ultrafast pump-probe spectroscopies were exploited to investigate the radiative properties of the hybrid heterostructures and to assess the presence of the typical anti-crossing behavior between polariton branches of plexcitonic states. Experiments of time-resolved spectroscopies can indeed enrich the knowledge on the intrinsic photophysics of these heterostructures, shedding light on the decay dynamics of the hybrid states. Quantitative results in terms of Rabi splitting energy, *i.e.* the energy separation between the lower and the upper polariton branches, will be reported and data on polariton relaxation dynamics will be shown. In particular, the presence of two distinctive bleaching signals in the transient absorption spectra confirms the existence of coherent exciton-plasmon states. Among the various architectures, particular attention will be devoted to long-range ordered plasmonic arrays of subwavelength holes milled in a metallic film. Indeed, the latter combine the required field confinement along with an open architecture, thus facilitating both the placement of the material and the excitation and probing of polaritons [6,7]. Periodic and quasi-periodic (Penrose-type lattice) arrays will be considered [8,9], and their extraordinary optical transmission properties investigated. The roles of both the symmetry and the nanohole packing on the interaction strength with different nanomaterials will be discussed. This work is stimulating toward a further exploration on the symmetry-dependent properties of long-range ordered 2D arrays, with a particular focus on quasi-periodic structures which show rotational symmetry-related unique properties in terms of per-hole transmission and near-field distribution. More in general, the presented results can set the base for investigations on novel hybrid architectures with interesting perspectives in numerous fields where strong light-matter interactions can be fruitfully exploited, such as the realization of devices optimized for photocatalysis.

Acknowledgement

This work is part of the REPLY project that received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement No 101002422)

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