Quantum theory of room temperature single quantum-dot strong-coupling with plasmonic nanoresonators

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Plasmonic nanomaterials and nanophotonics have the unique ability to confine light in extremely sub-wavelength volumes and massively enhance electromagnetic fields. Fundamentally, such high-field enhancement can alter the local density of states of a photoactive molecule to unprecedented degrees and control its exchange of energy with light. For high enough field enhancement, one enters the strong-coupling regime, where the energy exchange between the excited states of molecules/materials and plasmons is faster than the de-coherence processes of the system. As a result, the excitonic state of the molecule becomes entangled with the photonic mode, forming hybrid excitonic-photonic states. These hybrid-states are part light, part matter and allow for the characteristic Rabi oscillations of the atomic excitations to be observed. Until recently, the conditions for achieving strong-coupling were most commonly met at low temperatures, where de-coherence processes are suppressed. As a major step forward, we have recently demonstrated room-temperature strong coupling of single molecules in a plasmonic nano-cavity [1] achieved using a host-guest chemistry technique, controlling matter at the molecular level.

Here, linking nano-spectroscopy of quantum emitters with strong coupling allows to lithographically realise a strong-coupling set-up that couples dark plasmonic modes and quantum dots. A quantum nanophotonic model which incorporates a non-degenerated multi-level emitter strongly coupled with a broadband resonator explains and quantifies this boost of coupling strength due to collective coupling to band-edge states. Our combined theoretical and experimental findings [2] pave the road for a wide range of ultrafast quantum optics experiments and quantum technologies at ambient conditions. Moreover, the pronounced position-dependent spectral changes may lead to new types of quantum sensors and near-field quantum imaging modalities.

[1] R. Chikkaraddy, B. de Nijs, F. Benz, S. J. Barrow, O. A. Scherman, E. Rosta, A. Demetriadou, P. Fox, O. Hess and J. J. Baumberg, Nature 535 (2016), 127.

[2] H. Gross, J. M. Hamm, T. Tuffarelli, O. Hess and B. Hecht, (2017), submitted.