DOI: 10.3969/j.issn.1003-501X.2017.02.004

Strong coupling and ultrafast dynamics in organic semiconductor/metal hybrid nanostructures

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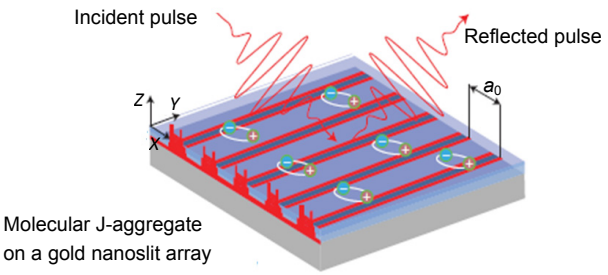

Schematic of coherent ultrafast spectroscopy on a hybrid nanostructure consisting of a 50-nm-thick film of J-aggregate molecules in a polymer matrix coated onto a gold nanogroove array with period a_0. Strongly localized SPP fields (in red) exist in and near the grooves. Incident and reflected laser pulses and J-aggregate excitons are shown schematically.

Abstract: Metallic nanostructures can support the strongly confined interface waves: surface plasmon polaritons (SPPs). SPPs have recently been used in a variety of applications due to their abilities to guide light in the scale of nanometer. Whereas, intrinsic weak optical nonlinearities and short propagation lengths of SPPs hinder their applications in novel active plasmonic devices.

One promising solution is to couple SPPs to nonlinear optical resonances, such as excitons (Xs) in molecular or semiconducting nanostructures. Consequently, hybrid nanostructures containing J-aggregate molecules and metallic nanostructures have attracted considerable interest. In these systems, vacuum field fluctuations lead to a coherent exchange of energy between ensembles of excitons and plasmons and the formation of new hybrid polariton states. Strong coupling between Xs and SPPs enables an efficient transfer of the strong optical nonlinearities of the excitonic emitters to the passive plasmonic nanostructures on the ultrashort time scale of femtosecond.

Here, we give a brief review of our studies in the area of active plasmonics. We focus on hybrid J-aggregate/metal nanostructures consisting of J-aggregate excitons and surface plasmon polaritons supported by metallic nanogroove arrays. We first introduce two experimental methods used in our study: chrip-compensated spectral interferometry and nonlinear pump-probe spectroscopy. The strong coupling between J-aggregate excitons and SPPs is studied in detail by probing both the static optical properties and ultrafast dynamics of the strongly coupled X-SPP systems. We show that two different energy transfer channels: a coherent resonant dipole-dipole interaction and an incoherent exchange of photons, are coexisting in the hybrid system. The interplay between both pathways results in a pronounced modification of the radiative damping due to the formation of super- and subradiant polariton states.

We also investigate the coherent energy exchange, Rabi oscillations between the excitonic and the SPP systems in real time. Using nonlinear pump-probe spectroscopy, coherent polariton dynamics of the hybrid X-SPP systems is studied. It is found that the optical response of the individual resonances is drastically altered by the optical dipole coupling between excitons and SPPs. Coherent X-SPP population transfer induces transient oscillations in exciton density, leading to a periodic modulation of the normal mode splitting and thus optical nonlinearity in a 10 fs timescale.

Keywords: surface plasmon polaritons; hybrid nanostructures; strong coupling; ultrafast spectroscopy

Citation: Wang Wei, Zhang Hong. Strong coupling and ultrafast dynamics in Organic semiconductor/metal hybrid nanostructures[J]. *Opto-Electronic Engineering*, 2017, **44**(2): 161-171.

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