Controlled Exciton-Plasmon Coupling in Hybrid Nanostructures

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Excitons and plasmons are the fundamental optical excitations in semiconductors and metals, respectively. Understanding optical properties of hybrid nanostructures consisting these two components necessarily involves exciton-plasmon coupling and its consequence. The challenges often arise from metallic nanostructures with ill-controlled shape, size, and morphology. I will discuss three experiments that aim to investigate well controlled exciton-plasmon coupling in hybrid photonic nanostructures. In the first example, we discuss Fano resonance in the scattering spectrum from the simplest hybrid molecule consisting of a gold nanoparticle and a semiconductor quantum dot. The Fano resonance is mediated by single photon absorption and scattering events. In the second experiment, we show that semiconductor quantum dot lifetime near an atomically smooth Ag film exhibits a narrower distribution compared to that near a thermally deposited film. In the third experiment, we demonstrate that cascaded exciton energy transfer in a monolayer semiconductor lateral heterostructure (MoS_2/WS_2) is extended to tens of microns in a planar metal-oxide-semiconductor structure. This energy transfer is facilitated by an exciton-surface plasmon polariton-exciton conversion process.

Theoretical Spectroscopy at Hybrid Interfaces

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Organic π-conjugated molecules are intriguing building blocks for opto-electronic applications, offering a playground for achieving tailored properties and functions through molecular size, functionalization, dimensionality, and molecule-substrate interaction. Owing to the weak inter-molecular binding, the single-molecule features naturally pre-determine the properties of condensed phases. Nevertheless, molecular assemblies, thin films, molecular crystals, and organic/(in)organic interfaces exhibit excitation spectra that substantially deviate from those of the gas phase.

From a theory point of view, the ab initio description of the interface electronic structure and corresponding optical excitations is an exciting though challenging issue as many-body effects play a dominant role. Only having proper theoretical concepts and numerical tools in hand which consistently capture the features of molecular materials, from single molecules to hybrid interfaces, allows for getting insight into the leading excitation processes.

Selected examples, including pyridine and poly(para-phenylene) on ZnO and others, will show how we explore, control, and predict level alignment and light-matter interaction at hybrid interfaces.