Dynamics of strongly coupled surface plasmons and molecular excitations

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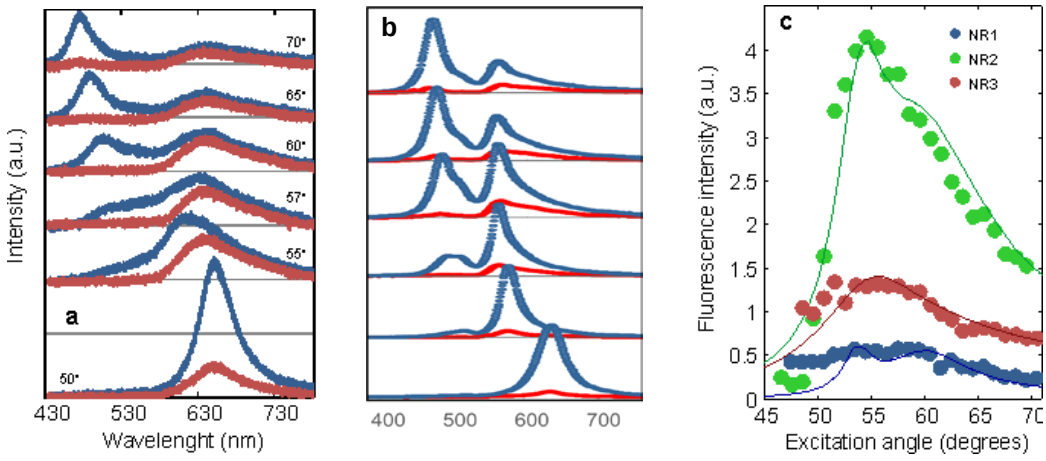
During the last decades surface plasmons polaritons (SPPs) have attracted vast interest because of their strongly enhanced light-matter interactions, which opens routes towards optical devices beyond the diffraction limit. Of particular interest is strong coupling between SPPs and molecular excitations (ME), which manifests itself through the formation of new hybrid modes exhibiting Rabi splittings even up to several hundreds of meV depending on the molecular species and concentration [1,2]. The obtained new hybrid ME-SPP polaritons, differ a lot from both MEs and SPPs, and could provide the means to enable interactions between photons/plasmons, needed for active all optical components. In addition, they have effect on properties of the molecules.

We have studied strong coupling between SPPs and various molecules [2,3], and shown that in all of them the fluorescence of the MEs reveals the coexistence of strongly coupled polaritons and non-coupled MEs. As the molecular fluorescence is directly related to the ME occupation, it provides an ideal tool for studying the polariton dynamics. We have utilized dye molecules with an large Stokes shift, Nile Red (NR), to demonstrate the absence of scatterings among the polaritons and to show that the modes decay via dephasing and internal relaxation of the molecules to a fluorescing state of the dye [3]. These result provides essential information on the dynamics of the strongly coupled modes. Studies with other molecules with different Stokes shifts and dynamics are on their way [4].



a: *Scattering spectra measured from the NR2-sample at different angles and polarizations: s (red) & p (blue). The s spectra match well with the fluorescence of NR, while p consists of fluorescence and the scattered ME-SPP polaritons.* ***b:*** *Scattering spectra measured from a R6G-sample at different angles and polarizations. Both spectra contain fluorescence as well as scattered ME-SPP polaritons.* ***c:*** *Measured total fluorescence (circles) for three NR-samples together with the quantum mechanical three state model (solid lines).*

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