Strong Coupling in Plasmonic systems and their Interaction with Molecules

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Abstract

We study the optical properties of molecules deposited metallic nanostructures with respect to the free molecules. We show theoretically and experimentally that molecular excited states can be strongly coupled to plasmonic modes. Upon coupling new hybrid states are form, the lower and the higher polariton. These modes have the characteristic of both molecular and plasmonic states. As the coupling strength grows, a new mode emerges, which is attributed to long-range molecular interactions mediated by the plasmonic field. The new, molecular-like mode repels the polariton states, and leads to an opening of energy gaps. By tuning the plasmonic modes to be on/off resonance with respect to molecular system excited state, one can shift these hybrid modes and by that modify the photo-physical and even the chemical properties of these molecules, and form a new kind of tunable hybrid materials.

In the same aspect, we study and demonstrate the strong coupling between plasmonic modes of metallic nanocavities (holes). The geometric parameters of the cavity, the distance between them and density of electrons participating in the modes are all determine the nature of hybridization. We study by cathodeluminescence together with linear and nonlinear optical measurements the nature of coupling in nanocavities milled either in aluminum or in silver and discuss their application for hybrid materials and catalysis. We further use such strong coupling to strongly enhance the nonlinear responses of the metallic surface and to tune actively the electromagnetic field at the sub-micron scale.

References

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Figures:

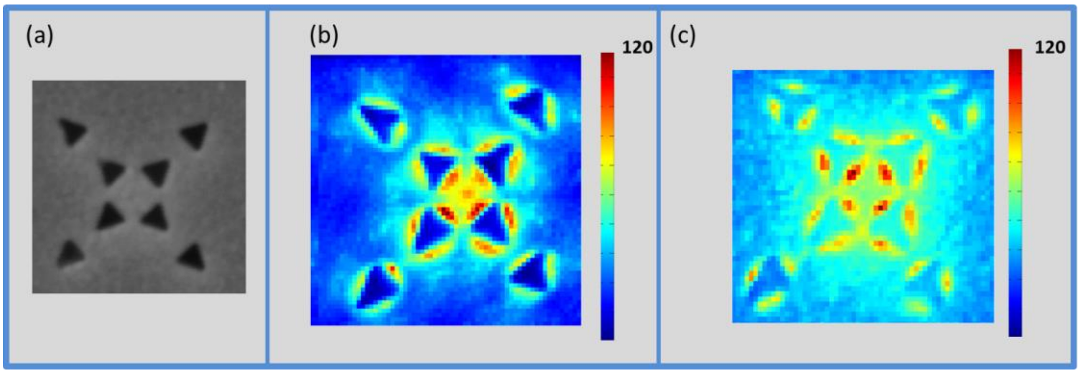

Figure 1: Images of Cathtodoluminescence of metallic nano cavities at 400nm±20nm. (a) SEM image of the studied plasmonic structure, the triangular hole side length is about 200nm and the distance between them is about 400nm. The CL revealed the nature of coupling and the difference between (b) silver and aluminum (c).

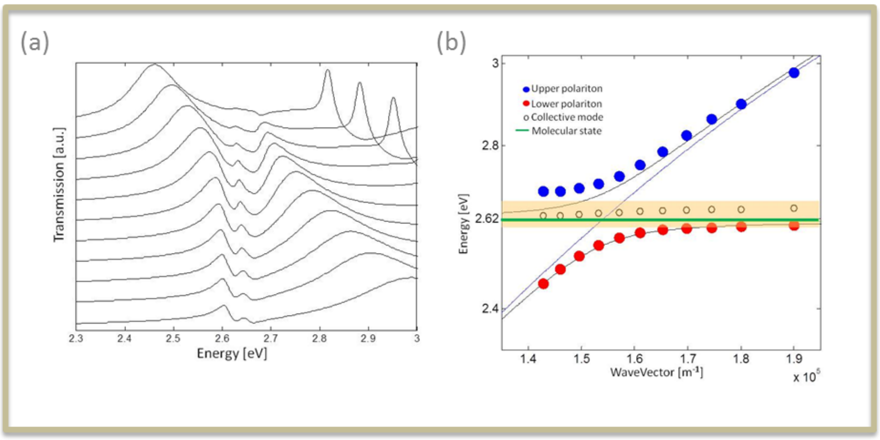

Figure 2: Simulations of a similar system. (a) Transmission spectra for the series of Ag slit arrays covered by a 10 nm thin film of molecular layer with a density of 3x 10^{25} m^3. An additional mode is clearly seen at about 2.64 eV. (b) Anti-crossing behavior of the hybrid system with RS value of 0.15 eV. The peak position of the additional mode barely changes with detuning of SPP mode. (ref[5])