Emergence of quantum phenomena in the coupling of plasmons with molecules and quantum dots

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Metallic nanostructures are often called plasmonic nanoantennas due to their capability to exploit the collective resonances of the free electrons, called plasmons, to manipulate light at the nanoscale. Notably, these structures can confine light in extremely low volumes, leading to a very large local density of states and to large coupling strengths with nearby emitters (Figure 1), such as quantum dots or molecules.

The behavior of these systems can typically be understood from classical electromagnetic calculations, but the constant improvement of fabrication techniques makes it possible to reach situations where purely quantum effects[1] could appear. In this talk, two situations of interest are considered. First, the regime of strong coupling provides a bridge between the classical and quantum worlds, where key phenomena such as Rabi splitting or anti-crossing can be understood classically but other effects such as the Jaynes-Cummings ladder are of purely quantum origin. In the talk, we first briefly discuss the possibility to reach strong coupling at the single emitter level with phononic antennas[2] –a close relative of the plasmonic ones- and then focus on how the characteristic anti-crossing of the strong coupling regime affects the photoluminescence signal[3].

On the other hand, in Surface Enhanced Raman Spectroscopy (SERS) plasmons strongly enhance the Raman signal from nearby molecules. State-of-the-art experimental results[4,5] may already operate in situations beyond conventional classical treatments. A new quantum model has been recently presented that describes the SERS process in a framework formally identical to that used to describe quantum optomechanical systems [6,7]. We apply a rigorous quantum treatment of this optomechanical framework and discuss several effects of potential interest to experiments, such as unexpected nonlinearities and large photon correlations. An improved understanding of the emitter-plasmon coupling at the quantum level in complex scenarios opens possibilities for new phenomena to emerge for fundamental studies and novel applications.

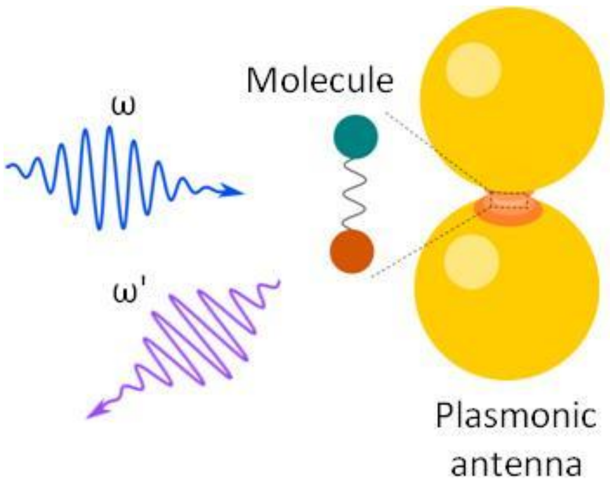

Figure 1. A plasmonic antenna, for example two spheres separated by a narrow gap, can interact strongly with nearby molecules, up to the point where strong coupling can be reached. The molecule-emitter interaction allows to manipulate light at the nanoscale, and can lead to emission at frequencies ω' different than the incoming light at ω, for example due to Raman processes or photoluminescence. In this talk, we apply quantum descriptions to gain new understanding on the coupling and on the emitted light.

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