Direct imaging of optical field enhancement, propagation and antenna effects for molecular plasmonics

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The exploitation of plasmon resonances to promote the interaction between conjugated molecules and optical fields motivates intensive researches. Their objectives are to understand the mechanisms of plasmon-mediated interactions, and to realize molecularly- or atomically-precise metal nanostructures combining field-enhancements and optical antenna effects. In this presentation, we present examples of plasmonic-field mappings based on multiphoton photoemission PEEM, [1] or scanning tunneling microscopy(STM)-induced light emission [2], two techniques among those which offer today's best spatial resolutions for plasmon microscopy.[3]

By imaging the photoemitted electrons, using well-established electron optics, two-dimensional intensity maps reflecting the actual distribution of the optical near-field are obtained. The imaging technique involves no physical probe altering the measure. This approach provides full field spectroscopic images with a routine spatial resolution of the order of 20 nm (down to 2 nm with recent aberration corrected instruments). We have analyzed the optical response of various metal nano-objects and imaged for the first time the so-called short-range propagative modes in nanorods [4] as well as polarization and spectral selection of hot-spots at the tips of gold nano-triangles [5] or nano-stars [6].

A particularly interesting geometry is that of a nano-scale gap between two metallic objects, as present between the tip and the sample in a STM. Such geometry is ideally suited to exhibit simultaneously field enhancement and antenna effects. Hence, an unfamiliar property of the junction of a STM is its ability to behave as a highly localized source of light. This phenomenon can be exploited to probe opto-electronic properties, in particular plasmonic fields, with ultimate subnanometer spatial resolution. Moreover, it permits to insert molecular systems in the junction, in particular through self-assembly. The analysis of current- or field induced photonic processes in the junction of a STM operating in an organic environment offers new insights into elementary mechanisms underlying photonic responses. We have studied the response of self-assembled metal particles and conjugated organic systems.[7] Time-resolved or nonlinear optical processes may be exploited as well.[8]

As a conclusion, we will report an application of plasmonics in two-photon fluorescence (TPF) enhancements through nanoantenna or Purcell effects in hybrid systems coupling gold nanoparticles to fluorophores. First results indicate the existence of an optimal distance between fluorophore and metal surface, allowing TPF enhancement.[9]

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