Enhanced single molecule fluorescence and SERS by metallic nanoantennas

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Electromagnetic processes stemming from molecular (spontaneous) emission close to complex metal nanostructures pumped at/near surface-plasmon resonances are investigated. Single molecule fluorescence close to metallic nanoantennas is thoroughly explored by calculating radiative and nonradiative decay rates (and quantum yields), addressing crucial issues as the modification and enhancement (or quenching) of spontaneous emission in (bio)molecular systems, due to the strong impact on the local (near-field) electromagnetic density of states of surface-plasmon resonances in dimer nanoantennas. In fact, a strong, 5-fold enhancement of the radiative decay rate from highly efficient fluorescent dye molecules is experimentally shown around resonant optical nanoantennas [1], resulting in an increase of the apparent quantum yield. This improvement is in agreement with electrodynamic model calculations [1]. In this regard, it is crucial to characterize theoretically and experimentally the surface-plasmon resonances for a variety of nanoparticles. like nanowires of various shapes, either isolated or interacting, including near-field intensity maps and surface charge distributions [2,3]. Indeed, near-field intensity enhancements straightforwardly yield the corresponding electromagnetic contribution to Raman enhancement factors, thus providing a tool to predict the performance of metal nanoantennas as SERS (surface-enhanced Raman spectroscopy) substrates [4].

In addition, metallic nanowire trimers have been theoretically investigated, exploring the rich phenomenology associated with multiple plasmon resonances. The enhanced local electromagnetic field associated with such surface-plasmon resonances can be exploited to achieve strong single molecule fluorescence enhancements [5], with straightforward implications for low efficiency emitters as in (bio)molecular sensing.

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