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Enhanced Raman and Fluorescence by Metal Nanostructures

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Abstract – Enhanced Raman and fluorescence from molecules near complex materials consisting of metal nanostructures is theoretically and experimentally investigated for sensing applications. Plasmon resonances supported by metal nanopaticles are shown to be responsible for the electromagnetic enhancement needed in such processes. A variety of shapes and configurations are studied. In particular, nanoflower-shaped metal particles are good candidates for SERS substrates without the need of aggregation; dimer nanoantennas are experimentally shown to enhance fluorescence decay rates, and can yield large SERS enhancement factors; and nanotrimers are proposed for twofold enhancement of fluorescence requiered when excitation and emission bands do not overlap.

Complex metal nanostructures play a crucial role in various electromagnetic processes stemming from molecular (spontaneous) emission occurring nearby. 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 and optoelectronic systems, due to the strong impact on the local (near-field) EM density of states of surface-plasmon resonances in dimer nanoantennas. Experimentally (Fig. 1), resonant enhancement of the radiative and nonradiative decay rates of a fluorescent dye is observed for dimers with optically coupled arms with narrow (~20 nm) gaps [1], in agreement with our electrodynamic model calculations [2,3]. On the other hand, metallic nanowire trimers have been theoretically investigated, with associated multiple plasmon resonances that can be exploited to doubly enhanced inelastic and/or nonlinear optical processes [4]; this has straightforward implications for low efficiency emitters in, e.g., (bio)molecular sensing or optoelectronic devices.

In this regard, it is crucial to characterize the surface-plasmon resonances for a variety of metal nanoparticles. We have indeed calculated the scattering cross sections for nanowires of various shapes (circle, triangles, rectangles, and stars), either isolated or interacting, including near-field intensity maps (with corresponding enhancement factors) and surface charge distributions [5]. Good agreement with experimental results for the dimer nanoantennas used in enhanced fluorescence is in turn achieved [6]. In the case of metal nanoparticles resembling a star/flower, which have also been obtained experimentally, large enhancement factors are shown to occur, making them specially suitable as SERS substrates [7].

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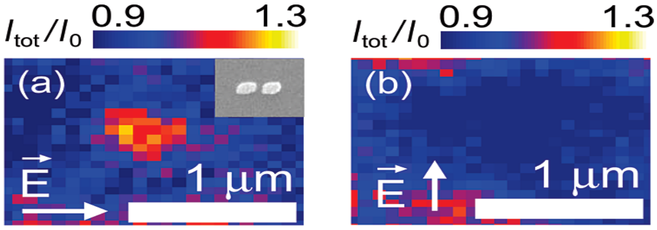

Figure 1: (a) Fluorescence intensity around a single resonant gold nanoantenna [on-scale SEM image in inset of (a)], for polarization of the detected light parallel, and (b) perpendicular to the antenna axis [1].

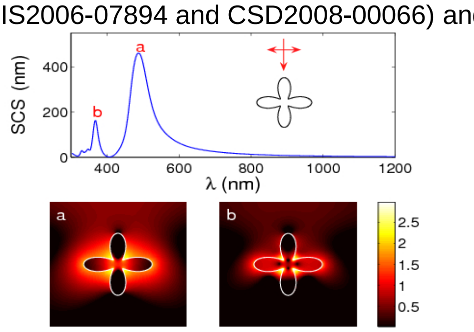

Figure 2: Scattering Cross Section for a Ag four-petal nanoflower with average radius of 30 nm and oscillation amplitude of 20 nm (blue curve). (a,b) Electric-near-field intensity distribution in log scale at both plasmon resonances [7].

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