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Influence of nonlocality on fluorescence from a dipolar emitter coupled to a metallic nanoshell

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It is has long been known that emission from classical or quantum emitters depends not only on their intrinsic properties but also on their environment [1]. Experiments on fluorescent molecules or quantum dots in the vicinity of planar metallic surfaces [2] or photonic crystal cavities [3] have shown that, under certain conditions, large enhancement of fluorescence and spontaneous emission rates can be achieved. A particularly promising template for engineering the emitter environment is provided by metallic nanoparticles (NPs). The optical response of these NPs is dominated by localized surface plasmon (LSP) modes, which modify drastically the local density of electromagnetic (EM) states and focus EM fields in regions below the diffraction limit, opening new possibilities to control light-matter interactions.

In the past both fluorescence enhancement and quenching were observed experimentally for emitters close to plasmonic NPs. It is now understood that the final result depends on the interplay between emitter excitation, which is enhanced due to the plasmonic environment, and emission into two decay channels: radiation in the environment and ohmic losses in the metal [4]. An optimum emitter-NP distance, for which the two mechanisms lead to a maximum fluorescence enhancement, can then be obtained. This is shown Fig. 1(a) for an emitter, described here as a classical dipo-le of dipole moment **p** parallel to the external electric field **E**, placed at a distance d from the surface of a spherical gold NP with radius R = 40 nm. The dipole is excited at wavelength λ = 532 nm and emits at λ = 560 nm, values which are appropriate for Rhodamine 6G, and R is selected so that the plasmon resonance lies close to these wavelengths (516 nm). It can be seen that fluorescence, η, is enhanced by about 2.5 times if the dipole is 20 nm away from the NP surface. This value could be significantly enhanced if absorptive losses in the metal were reduced, by choosing for example a different NP material. Silver is a good plasmonic material characterized by low absorptive losses. However, for a silver sphere of similar size, the main LSP peak is at much shorter wavelengths (about 360 nm). In order to bring the LSP reso-nance within the wavelength region of interest, we take advantage of the tunability provided by hybridization of the plasmonic modes excited at the inner and outer sur-faces of a metallic nanoshell [5]. To this end, in Fig. 1(b) we consider a complex NP consisting of a SiO_2 core of radius R_1 = 32.7 nm, covered by a silver shell of thick-ness 7.3 nm,

parameters which were chosen so that the total radius R and LSP wavel-ength remain the same as in Fig. 1(a). Reduction of ohmic losses in the metallic mate-rial leads then to much stronger fluorescence enhancement, of more than an order of magnitude, and this holds for a much larger range of emitter-NP distances [6].

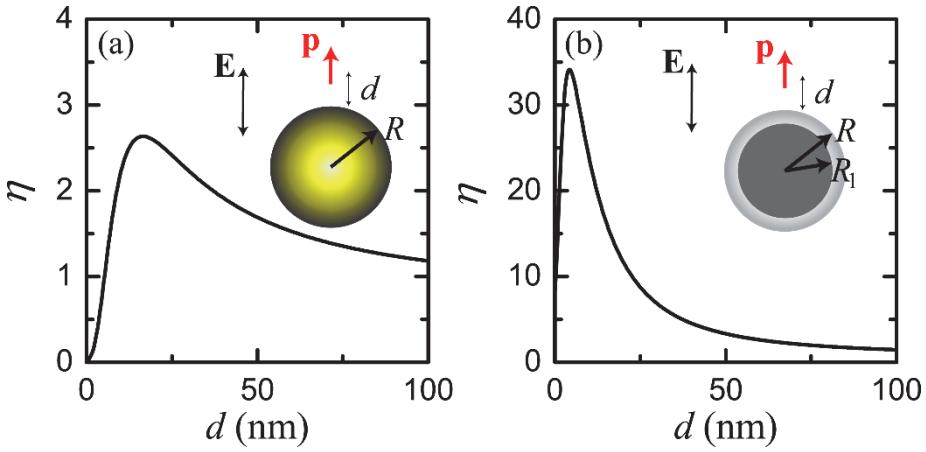

Figure 6: (a) Dependence of fluorescence enhancement, η, for an emitter with dipole moment **p**, polari-zed parallel to the external electric field **E**, on distance d from a gold nanosphere of radius R = 40 nm. (b) Same as (a), for a silver nanoshell consisting of a SiO_2 core of radius R_1 = 32.7 nm and a silver shell of thickness 7.3 nm, so that the total NP radius is R = 40 nm.

An important issue that arises when such emitters are placed close to plasmonic NPs, or when the metallic shell of the NP is very thin, is that the nonlocal character of the dielectric function of the metal becomes relevant. For example, it was recently shown, through theoretical studies with a simple hydrodynamic model, that emission rates for a classical dipole in the vicinity of a metallic NP can be significantly redu-ced because of nonlocality [7]. Within the same model, it has also been shown that strong shifts of the plasmonic modes are also expected [8]. Here we will explore the-se issues in a systematic way, and study the effect of nonlocality for different NP si-zes, shell thicknesses, and emitter distances, by use of both the hydrodynamic model and the recently proposed Generalized Nonlocal Optical Response theory [9]. Our re-sults are expected to shed light on the importance of a description of the optical response of plasmon-emitter hybrids that goes beyond classical electrodynamics and facilitate the design of novel architectures for controlling light-matter interactions.

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