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Highly enhanced spontaneous emission with nanoshell-based metallodielectric hybrid antennas

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ABSTRACT

The metallodielectric hybrid nanoantenna integrating plasmonic nanostructures with dielectric planar substrate can improve the spontaneous emission greatly. We demonstrated that the performances of the hybrid antenna can be substantially optimized with specific plasmonic nanostructures by employing finite-difference time-domain method. The hybrid antenna with core-shell nanostructure can enhance spontaneous emission greatly rather than the individual spherical nanoparticle. Moreover, the performances of the hybrid antenna can be boosted further through using asymmetrical nanoshell. The mechanism of the high enhancement effect is due to the hybrid structure being able to couple efficiently with the electric field by a larger dipolar moment. And the emission directivity of the hybrid antenna is able to be modified by adjusting the geometry of the plasmonic nanostructures. The results should be beneficial for various fundamental and applied research fields, including single molecule fluorescence and surface enhance Raman spectroscopy, etc.

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1. Introduction

Metallic nanostructures exhibit extraordinary surface plasmon resonance that interacts strongly with optical fields. The plasmonic nanostructures can modify the spontaneous emission of an emitter greatly. Currently, plasmonic surface-enhanced spectroscopy is an exciting field of study attracting much attention, and metallic nanostructures are now commonly referred to as optical antennas. The behaviors of plasmonic nanostructures in surface enhanced fluorescence (SEF) have received great attention. The research topic has progressed extensively during the last decades from theoretical understandings to various effective practice configurations [1–7]. The general principle of SEF effect has been pointed out in the pioneering work of Purcell, i.e. the spontaneous emission of an emitter can be influenced and even modified by engineering its electromagnetic environment [8]. Enhancement of spontaneous emission is in demand in fundamental interests and various applied research fields, such as near-infrared fluorescence imaging [9], fluorescent labels [10], and detecting single miRNA [11], etc. To modify spontaneous emission, researchers have controlled the local electromagnetic environment of an emitter to highly enhance its Purcell factor, thus enhancing its emission rate. For instance, it was demonstrated that a conical metallic nanoantenna, gold nanorods, nanospheres, periodical metal structures and other designed nanostructures can enhance its spontaneous emission rate [12–16].

However, the electromagnetic enhancement of single plasmonic nanostructure is limited due to intrinsic loss of metal materials and quantum tunneling effect. That limits the ability of enhancement of spontaneous emission. Interestingly, it was found that hybrid structures can provide higher enhancement effect. The hybrid antenna, consisting of plasmonic and photonic elements, has received many attentions because of its high performance to engineer the spontaneous emission [17]. For instance, Chen et al. proposed to combine single metallic nanoparticles antenna (MNA) with a dielectric planar antenna (DPA) structures, and they exploited that this designing strategies of plasmonic nanoantennas can highly enhanced the spontaneous emission rate [18]. The concepts from cavity quantum electrodynamics was implemented to understand the maximum of the local density of optical states (LDOS) and the suppressed the non-radiative losses incurred by the metallic constituents.

Although the simple gold spherical nanoparticle provides a convenient and powerful model system as optical antennas, it does not result in very large enhancement effects. Compared to gold nanorod and nanoshell, gold spherical nanoparticle is an antenna with low enhancement effect [19, 20]. For instance, the core-shell nanoantenna (CSNA) structure (i.e. gold shell with a silica core) possesses several advantages rather than the spherical

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nanoparticle. There are at least two parameters for the CSNA, i.e. core radius and shell radius, to control the plasmon resonant frequency spreading from visible to infrared, and to concentrate the near field distribution. So the plasmonic enhancement effect is higher, which promises much larger modifications of several orders of magnitudes for the spontaneous emission enhancement. The hybrid antenna that consists of a single gold CSNA combined with a dielectric planar antenna would enhance spontaneous emission greatly rather than spherical gold nanoparticle.

In this study, we numerically investigated a hybrid antenna consisted of CSNA nanostructure and DPA by employing finite-difference time-domain (FDTD) method. The hybrid antenna can highly enhance the spontaneous emission than the spherical nanoparticle, which presents larger antenna efficiency and larger Purcell factor. To understand the mechanism of the high enhancement effect, the electric polarization and surface charge distribution were calculated for comparison. It was found that the hybrid structure is able to couple efficiently with the emitter and behaves a dipole-like oscillation with a large moment. The far-field radiation directivity under the control of the hybrid antennas were also calculated and compared, and it was found that most of the radiative flux is directed to the dielectric substrate leading to a high collection efficiency.

2. Methods

The finite-difference time-domain (FDTD) method, a powerful technique for metallic nanostructures with arbitrary geometries, is employed to calculate near-field electromagnetic properties [21]. The FDTD method has been used widely to calculate the electromagnetic optical responses of various metal nanostructures, like as absorption and scattering cross sections, near-field distribution and enhancement, and decay rates etc in specific position near metallic structure. And near-field to far-field transformation method is implemented to obtain the far-field directivities modified by the nanostructures. Specifically, to understand the interactions between the nanoantenna and fluorophore, a configuration comprising a single emitter and a single nanoantenna was studied systematically. An emitter was located at a short distance from antenna, which is implemented in the FDTD method by having a classical point current source placed in the proximity of the nanostructure. The dipole orientation is determined by the components of the current source. The emitter's radiative decay rate Γ^0_{rad} in vacuum changes to $\Gamma_{tot} = \Gamma_{rad} + \Gamma_{nrad}$. Here, Γ_{rad} is the energy that reaches the far field (the radiative decay rate) and Γ_{nrad} is the radiated energy absorbed by the whole structure. We define the ratio $\eta_a = \Gamma_{rad}/\Gamma_{tot}$ as the antenna efficiency of the optical nanoantenna and the ratio $\eta_0 = \Gamma^0_{rad}/\Gamma^0_{tot}$ as the quantum efficiency of the emitter in vacuum. All the quantities with “0” above represent the quantities in vacuum. The far field decay rate was obtained indirectly by integrating the Poynting vector over closed surfaces that contain the nanoantenna and dipolar source, while the total decay rate was obtained over closed surfaces only containing the dipolar source [22]. In addition, Purcell factor is defined as $F = \Gamma_{rad}/\Gamma^0_{rad}$. When the single emitter is coupled to the nanoantenna, its quantum efficiency can be written as

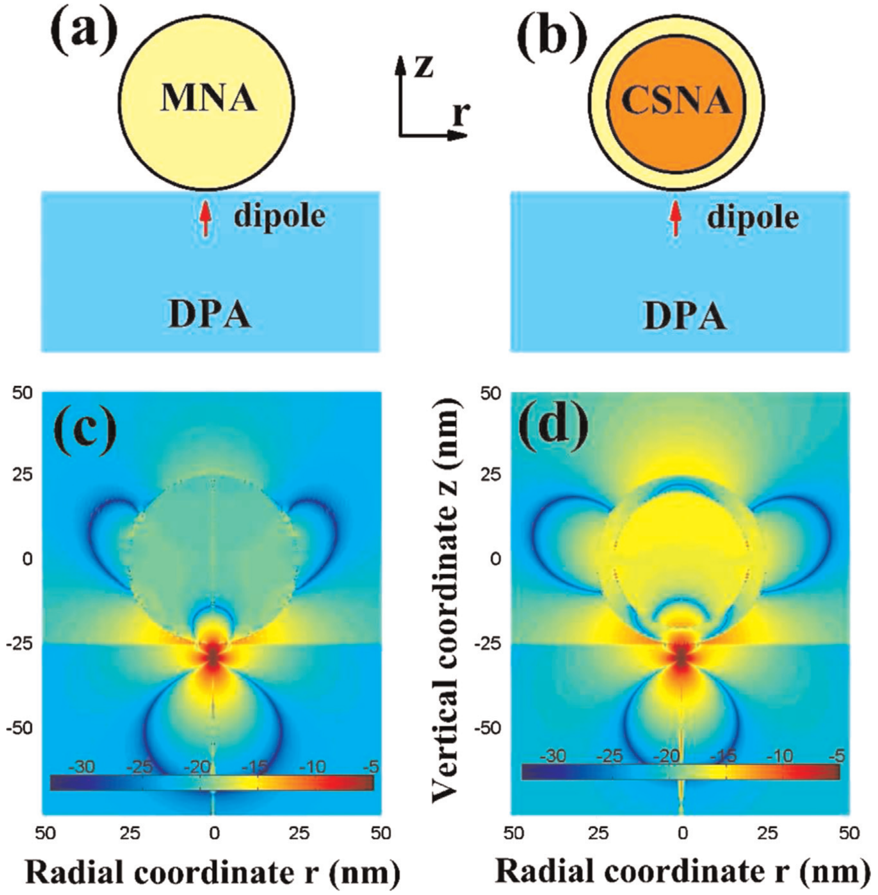

Fig. 1. Configuration of a MNA (gold-sphere) (a) and a CSNA (b) with a DPA of refractive index n respectively, where the emitter is polarized along the z direction. The z component of intensities of the electric field distribution in $\rho - z$ plane around the MNA (c) and CSNA (d), where the radius of MNA is 25 nm, radius of SiO_2 core is 20 nm, and thickness of gold shell is 5 nm. The color bars are in log scale. The refractive index of the substrate is 2.0 in both cases. The interface-emitter distance is fixed at 4 nm. The emission wavelength is 549 nm for MNA-DPA and 670 nm for CSNA-DPA corresponding to the resonant peak of Purcell factor for MNA case and CSNA case, respectively.

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