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Quantum emitter coupled to plasmonic nanotriangle: Spatially dependent emission and thermal mapping

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ABSTRACT

Herein we report on our studies of radiative and non-radiative interaction between an individual quantum emitter and an anisotropic plasmonic nanostructure: a gold nanotriangle. Our theoretical and three-dimensional electromagnetic simulation studies highlight an interesting connection between: dipole-orientation of the quantum emitter, anisotropy of the plasmonic nanostructure and, radiative and non-radiative energy transfer processes between the emitter and the plasmonic geometry. For the out of plane orientation of quantum emitter, the total decay rate and non-radiative decay rate was found to be maximum, showing radiation extraction efficiency of 0.678. Also the radiative decay rate was greater for the same orientation, and showed a pronounced spatial dependence with respect to the nanotriangle. Our study has direct implication on two aspects: designing nanoparticle optical antennas to control emission from individual atoms and molecules and geometrical control of quenching of emission into plasmonic decay channels.

1. Introduction

Understanding interaction between individual quantum emitters such as atoms and molecules with plasmonic nanostructure has emerged as an important area of research in the context of quantum nanophotonics. By tailoring the geometry of the plasmonic nanostructures, especially by introducing anisotropy in the geometry, one can systematically tune the local density of optical states (LDOS) [1,2], which can further effect the radiative and non-radiative processes of a quantum emitter in its vicinity [3–6]. In the context of quantum emitter interacting with a single anisotropic plasmonic nanostructure, there are two important questions that have emerged in recent times. Firstly, how does the spatial location of the quantum emitter with respect to individual anisotropic plasmonic nanostructure affect its radiative and non-radiative processes? This question is of direct relevance in designing optical antennas to control atomic and molecular emission [7–14]. With the emergence of nanofabrication methods, accurate placement of single quantum emitters have been achieved [15–17], which may open new avenues in quantum optics and quantum information processing [18,19]. The second question is how anisotropic plasmonic geometries can be harnessed to effectively control non-radiative energy transfer between a single quantum emitter and a single anisotropic plasmonic nanostructure. This question has direct implication on the process of quenching of emission by plasmonic channels, and also has connection in designing thermoplasmonic nanoprobes [20–25] that are nowadays extensively utilized for applications such as photothermal therapy [26–28], near field sensors [29] etc.

Motivated by the above-mentioned questions, herein we report on our numerical and theoretical studies of radiative and non-radiative energy transfer processes between an individual quantum emitter and an anisotropy plasmonic geometry – gold nanotriangle. The rationale behind the choice of this geometry was that gold nanotriangle can be nanofabricated by both bottom-up and top-down approaches with excellent control over the geometrical parameters [30–34].

In this study, we address the issue of dipole-orientation dependent radiative and non-radiative energy transfer process, and evaluate its spectral and spatial dependence with respect to the plasmonic geometry. We found the out-of-plane dipole (z-polarized) emitter to exhibit greater total decay rates compared to in-plane emitters. Interestingly, the radiative decay rate of dipole emitters show a strong spatial dependence with respect to the geometry.

2. Theory

2.1. Decay rate of a quantum emitter

Under the dipole approximation of particle-field Hamiltonian,

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one can represent the atom as an oscillating dipole for all practical calculations [1,35]. The decay rate of a molecular or an atomic emitter can be expressed using Fermi's golden rule, considering weighted sum of all possible decay channels according to which, the total decay rate of an excited emitter is proportional to transition dipole moment $p_{trans} = \langle i|\hat{p}|f\rangle$ where $|i\rangle$ and $|f\rangle$ denotes initial and final states of the emitter respectively and $\hat{p}$ represents the dipole operator. When a dipolar emitter is placed in the vicinity of a metallic nanostructure, the emitter will now have two channels to decay. One of those is to directly couple to the far field, γ_{fs}, and other is to couple to the near field of the structure, γ_{couple}. Thus coupled power, P_{couple}, can either get scattered away by the structure to far field or can get absorbed by the structure. Scattered power together with γ_{fs} forms the total radiative decay rate. That part of emitted power which ends up in getting absorbed by the structure is a measure of non-radiative decay rate of the emitter. So, in presence of nanostructure we can write the total decay rate of the emitter, γ_{tot}, as

$$\gamma_{tot} = \gamma_{couple} + \gamma_{fs} \tag{1}$$

where, γ_{couple}, γ_{fs} are decay rate due to near field coupling with the nanostructure, free space radiative decay rate in the presence of nanostructure respectively. Now, γ_{couple} can further be expressed as a sum of absorptive decay rate, γ_{abs}, (which forms the non-radiative part) and scattered decay rate, γ_{scatt}, (which forms the radiative part) respectively. The scattered electric field and electric field of the emitter combine to give total radiated power in the far field. Depending upon the phase of the scattered field with respect to electric field of the dipolar emitter, one can have a destructive interference and hence excite optically dark modes which show complete radiation quench in the far field.

Now, the total radiation extraction efficiency, (modified quantum efficiency of the emitter in the presence of nanostructure) can be written in terms of γ_{abs} and γ_{rad} as

$$\eta = \frac{\frac{\gamma_{rad}}{\gamma_{rad}^{0}}}{\frac{\gamma_{abs}}{\gamma_{rad}^{0}} + \frac{\gamma_{rad}}{\gamma_{rad}^{0}}} \tag{2}$$

where γ_{rad}^{0} is the radiative decay rate of emitter in absence of nanostructure.

2.1.1. Radiative and non-radiative decay rates

Radiative decay rate for an emitter coupled to nanostructure consists of two parts (i) Direct out-coupling of power by the emitter to the far field (ii) Scattered component of the coupled power by the scatterer. If the dimensions of the scatterer is less than emission wavelength, the scattered power from the structure will be majorly dependent on the induced dipole moment, ($\vec{p}_{induced}$), in the structure. Using this approximation, normalized radiative decay rate can be represented as [36],

$$\frac{\gamma_{rad}}{\gamma_{rad}^{0}} = \frac{|\vec{p} + \vec{p}_{induced}|^2}{|\vec{p}|^2} = \frac{P_{rad}}{P_{rad}^{0}} \tag{3}$$

where $\vec{p} = [p_x, p_y, p_z]$ is dipole moment of the emitter and P_{rad}^{0} is power emitted by dipolar in the absence of nanostructure. Exact expression for the radiative decay rate will depend on morphology and polarizability of the structure.

Non-radiative decay rate from emitter is quantification of power absorbed by the nanostructure out of power coupled by emitter to its near field. For an emitter with emission frequency ω and wavenumber $k=\omega/c$, to nanostructure with wavelength dependent dielectric permittivity $\epsilon(\omega)$ it can be expressed as [36],

$$\frac{\gamma_{abs}}{\gamma_{rad}^{0}} = \frac{3}{16}\,\mathrm{Im}\frac{\epsilon(\omega)-1}{\epsilon(\omega)+1}\frac{1}{k^3 z^3}\frac{(p_x^2 + p_y^2 + 2p_z^2)}{|\vec{p}|^2} = \frac{P_{abs}}{P_{rad}^{0}} \tag{4}$$

Neglecting edge and curvature effects Eq. (4) implies that non-radiant energy transfer is dependent only on the distance, z, between emitter and the structure and relative dielectric permittivity $\epsilon(\omega)$ of the structure.

2.2. Mapping non-radiative transfer of energy

Energy absorbed by the nanostructure will dissipate mostly as heat. In metallic nanostructures, major source of dissipation is joule heating. So non-radiatively transferred energy can considered, in case of metallic nanostructures, to get dissipated entirely as heat due to joule heating. For far-field illumination, heat power density created will be proportional to absorption cross section of the structure. But, in this case of local excitation, heat power generated will depend on photonic density of states rather than on absorption cross section of the structure. Since the dissipation methodology is the same (joule heating), heat dynamics in the system will be governed by heat diffusion equation with source of heat being electromagnetic power dissipation density in structure due to power emitted by dipole source as,

$$\rho C_p\left(\frac{\partial T}{\partial t} + \mathbf{u}_{trans}.\,\nabla T\right) + \nabla.\,(\mathbf{q} + \mathbf{q}_r) = -\,\alpha T\!:\frac{dS}{dt} + Q \tag{5}$$

where ρ is density of material, C_p is specific heat capacity at constant pressure, T is absolute temperature, $\mathbf{u}_{trans}$ is velocity vector of translational motion, $\mathbf{q}$ and $\mathbf{q}_r$ are heat flux by convection and radiation respectively, α is coefficient of thermal expansion, S is the kirchhoff stress tensor and Q is the electromagnetic dissipation power density. For steady state calculations, the time derivatives will vanish from Eq. (5).

With this hindsight, we go on to calculate decay rates of a quantum emitter in the vicinity of an anisotropic nanostructure and quantify the non-radiative energy transfer from the emitter.

3. Methods

3.1. FDTD calculations

We used Finite Difference Time Domain (FDTD) method to calculate extinction spectrum, near field electric field and normalized decay rates using commercially available solver by Lumerical solutions Inc. The structure under study is a gold nanotriangle of edge length 160 nm and thickness of 30 nm placed over glass substrate (see Fig. 1(a)). Triangle was modeled to have rounded corners with corner radius of 20 nm to avoid field singularity at corners and also to mimic experimentally realizable object. The area near the triangle was discretized by a non-uniform conformal variant mesh with meshing size of 0.6 nm and rest of the simulation area with size of 1 nm. Simulation area was terminated by Perfectly Matched Layers (PMLs) to avoid spurious reflections from boundaries. Wavelength dependent dielectric permittivity of gold was taken from experimental details provided by Johnson and Christy [37] and that of glass (SiO2) from Palik [38]. For extinction spectrum calculation, a broadband Total Field Scattered Field (TFSF) source[1] (illumination wavelength, 400–1200 nm) was used. Absorption and scattering cross sections were calculated using an in-built analysis group in Lumerical FDTD solver and extinction spectrum was calculated in the post

[1] A TFSF source uses a plane wave illumination and divides simulation area into total and scattering fields.

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