Enhancing Fluorescence of Diamond Color Centers Near Gold Nanorods Via Geometry Optimization

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Abstract

Detecting light emitted by fluorescent molecules with resolution down to single photon is an important problem in various fields of sciences and applications, such as solid-state physics, quantum information processing and medicine [1-3]. The detection probability can be improved via enhancement of excitation and emission. Enhancement both of these processes can be reached by localized surface plasmon resonance (LSPR) of metal nanoparticles. LSPR phenomenon strongly confines the in-coupled electromagnetic field and increase the excitation via the strong intensity enhancement at the position of the emitter [4,5]. In addition, LSPR can promote effectively the scattering of light emitted by a fluorescent molecule into the far-field [6,7]. However, the properties of LSPRs is strongly material and shape dependent [8]. To obtain the optimal geometric parameters of the plasmonic structure - emitter coupled system numerical calculations needed.

Our purpose was to determine the optimal configuration of the gold nanorod - emitter coupled system to enhance excitation or emission. We have modeled this nanophotonic problem in COMSOL Multiphysics® using the RF Module. The optical properties of inspected configurations were determined by integrating the power outflow through closed surfaces. The emitter was modeled as an electric point-like dipole with frequency corresponding to the 532 nm excitation wavelength of nitrogen (NV) and silicon (SiV) vacancy centers, as well as 650 nm and 738 nm corresponding to the emission wavelength of NV and SiV centers, respectively. The wavelength dependent properties were determined by sweeping the dipole oscillation frequency. The nanorod geometry, dipole position and orientation were varied during optimization (Figure 1). The optimization was realized by implementing GLOBAL algorithm using LiveLink™ for MATLAB® [9], and two different optimization approaches were compared. Namely, maximization of the total decay rate enhancement with respect to homogeneous environment i.e., the Purcell factor and the quantum efficiency QE was realized by setting a minimal criterion regarding the quantum efficiency (QE_crit) and Purcell factor (P_crit), respectively (Figure 1).

The wavelength dependent decay rate enhancements and the apparent QE qualify the excitation and emission enhancement achievable in proximity of a plasmonic nano object. Therefore, the enhancement of the radiative- (γ^rad), non-radiative decay rate (γ^non-rad) with respect to homogeneous environment and the apparent QE of the optimal

configurations obtained via QE_crit and P_crit optimization approaches are presented in Figure 2. The local and global maxima appearing on the spectra of optical responses are related to transversal and longitudinal LSPRs of the nanorod with different orders.

At 738 nm and 650 nm emission wavelengths both optimization approaches have found an optimal configuration, in which the nanorod geometry is tuned into the LSPR bands at the wavelengths of interest. Accordingly, the optimal configurations result in global maxima in radiative decay rate enhancements and apparent QE. However, at 532 nm none of the found configurations presents a maximum in the optical responses caused by inherent losses in gold. Although, the QE_crit approach resulted in configuration with higher radiative decay rate and so in a better excitation or emission enhancement, P_crit approach is more straightforward in applications.

Reference

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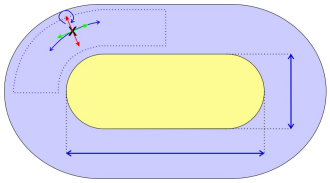
Figures used in the abstract



$$Purcell \quad factor = \frac{\gamma^{rad} + \gamma^{non-rad}}{\gamma_0^{rad} + \gamma_0^{non-rad}}$$

$$QE = \frac{\gamma^{rad}}{\gamma^{rad} + \gamma^{non-rad}}$$

Figure 1: Schematic of the coupled system used for optimization and to determine the decay rate enhancements (Purcell factor) and quantum efficiency (QE) via power flow integration. Yellow: gold nanorod, grey: diamond coating, green/red arrow: dipole, blue arrows: dipole position, orientation and nanorod geometry tuning.

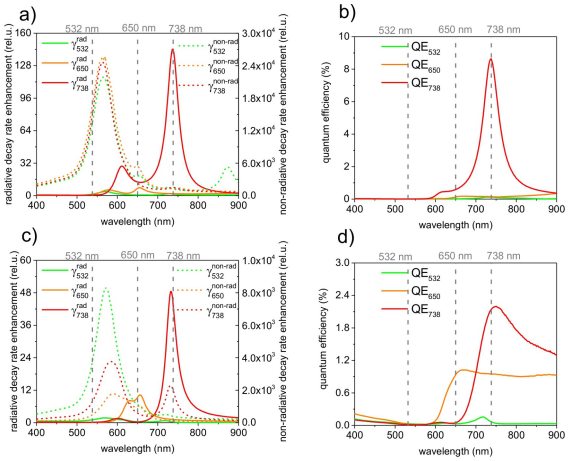


Figure 2: Wavelength dependent (a, c) radiative and non-radiative decay rate enhancements and (b, d) quantum efficiency of optimal configurations obtained via (a, b) QE_crit method, and (c, d) P_crit method.