

the spontaneous emission rate and introduces an additional non-radiative decay channel with rate γ^{nr} due to absorption in the metal.^{8,36} The new radiative decay rate γ^{r} and the quantum efficiency η can be related to the initial values by the following expression³⁷

$$\eta = \frac{\eta_o}{(1 - \eta_o)/F + \eta_o/\eta_a}, \quad (1)$$

where $F = \gamma^{\text{r}}/\gamma_o^{\text{r}}$ is the Purcell factor and $\eta_a = \gamma^{\text{r}}/(\gamma^{\text{r}} + \gamma^{\text{nr}})$ is the antenna efficiency. Under weak excitation, the fluorescence signal of the isolated emitter is $S_o = \xi_o \eta_o |\mathbf{d} \cdot \mathbf{E}_o|^2$. Here, ξ_o represents the collection efficiency, \mathbf{d} is the transition electric dipole moment, and \mathbf{E}_o is the electric field at the emitter position. With an optical antenna, the local electric field and the collection efficiency get also modified and the signal becomes $S = \xi \eta |\mathbf{d} \cdot \mathbf{E}|^2$.^{16,18} Assuming that the signal is collected over all angles, such that $\xi = \xi_o = 1$, the fluorescence enhancement reads

$$\frac{S}{S_o} = \frac{\eta}{\eta_o} \frac{|\mathbf{d} \cdot \mathbf{E}|^2}{|\mathbf{d} \cdot \mathbf{E}_o|^2}. \quad (2)$$

Furthermore, if the metal nanostructure and the emitter are arranged in a configuration that almost preserves the dipolar radiation pattern of the isolated emitter,^{18,37} reciprocity implies that S/S_o can be well approximated by replacing the electric field enhancement $|\mathbf{d} \cdot \mathbf{E}|^2/|\mathbf{d} \cdot \mathbf{E}_o|^2$ with the Purcell factor F ⁵⁸. We thus write

$$\frac{S}{S_o} \simeq \frac{F}{(1 - \eta_o)/F + \eta_o/\eta_a}. \quad (3)$$

From Eqs. (2) and (3) one immediately notes that the maximum fluorescence and the maximum field enhancements do not correspond because of η_o and η_a .

The calculation of F and η_a is performed here using the body-of-revolution (BOR) finite-difference time-domain (FDTD) method, which exploits the cylindrical symmetry of the nanocones to significantly reduce the computational burden in comparison to standard 3D-FDTD methods⁵⁹. The computational details can be found in Ref. 37 and references therein. We focus our attention on gold³³ optical antennas made of one or two finite nanocones. The emitter is always at 10 nm from the sharp end of the nanocone, unless otherwise specified, and it is positioned and oriented along the nanocone axis. This distance is chosen in order to ignore effects due to nonlocality in the optical constants of the metal interface⁶⁰ and convergence issues in the FDTD method.⁶¹ The mesh pitch is 1 nm or 0.5 nm, depending on the nanocone geometry. For the case of an optical antenna made of two nanocones, the gap between them is fixed to 20 nm. The nanocone dimensions are chosen such that the LSPP