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Emission rate studying through nanodiamonds with embedded nitrogen vacancy centers

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Abstract. High-refractive index nanoparticles, including diamond nanoparticles, can exhibit strong optical resonances, whose spectral position depends on particle size and shape. In this work we demonstrate that these resonances can be used to control the emission of active defects within the particles. We present the theoretical concept of an active dielectric nanoantenna, and study the dependence of the radiation properties on the position of the defect within the particle, as well as on particle size.

1. Introduction

Dielectric nanoparticles are actively studied within many research directions of nanophotonics since resonant behavior of small particles can substantially enhance light-mater interaction at the nanoscale. One of the potential applications areas of this enhancement is the emission control of single photon emitters through excitation of Mie resonances [1]. Efficient coupling of resonances of quantum sources with localized modes of high-index dielectric materials offers new opportunities for emission enhancement. In previous works, it was shown how to couple resonances of quantum emitters with resonances of dielectric and plasmonic nanoparticles [2,3], photonic crystals [4,5], and optical cavities [5,6]. By placing emitters inside the high-index dielectric nanoparticle we obtain the smallest possible structure with enhanced emission control, and we refer to it as an active nanoantenna [6,7].

The simplest structure that can serve as an active nanoantenna is a diamond nanosphere with an embedded quantum emitter – nitrogen vacancy center (NV-center). Diamonds have excellent optical properties, including high refractive index (~2.4), low losses in visible and high stability.

In this work we present the theoretical concept of active dielectric nanoantenna. We analyze the effect of high-order optical modes of relatively large diamond particles on the emission rate as compared to the small nanoparticles that do not support Mie resonances.

2. Concept of active dielectric nanoantenna

The optical properties of nanoantennas, and, as a result, the emission properties of the NV-Centres will depend on size, shape and material of nanoparticles. Larger refractive index leads to larger enhancement of the intensity of the optical field inside the particle. As the size of the particle increases, the number of supported resonances increases and high-order resonances appear.

To analyze multiple resonances and to reveal the relation between sizes and emission rate enhancement, it is common to use the Purcell factor, and here we calculate it for diamond nanoparticles with embedded NV-centers. Utilizing the theory developed in Refs. [8,9], we study the Purcell Factor dependence on dipole position inside the sphere (see Fig. 1). Purcell factor is calculated as a ratio of emission rate of the dipole inside a diamond sphere ($n = 2.4$) to that without the sphere. Due to the fact that the positions of NV centers in realistic nanodiamonds are not known, we assume that they can be randomly located inside a diamond. Moreover, larger nanodiamonds will contain multiple defects, and the observed emission enhancement will be proportional to the average Purcell factor:

$$\bar{F}_p = \frac{\int_{v_{min}}^{v_{max}} \iiint_V F_p(r,v) dV P_0(v) dv}{V \int_{v_{min}}^{v_{max}} P_0(v) dv},$$

(1)

where V – volume of the diamond particle, v is frequency, and $P_0(v)$ – typical luminescence intensity spectrum of the NV-center.
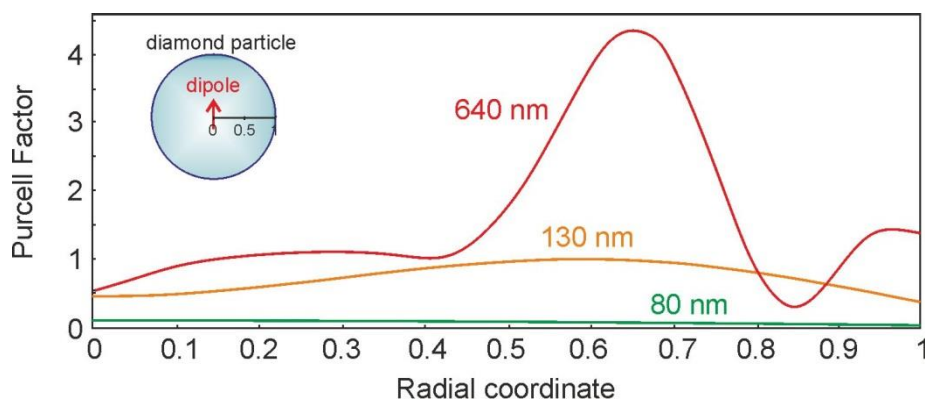

Figure 1. Purcell factor dependence on the normalized radial position of the particle within a spherical nanodiamond for threeThe diamond particle sizes and dipole radial coordinate dependence on Purcell factor particle sizes.

From the results presented in Fig. 1 it is clear that in the small nanoparticles (less than 100 nm), the emission rate is suppressed due to low local density of optical states [10]. Higher order resonances in large diamonds produce large Purcell factors that should lead to significant emission rate enhancement. We see that the Purcell factor in the diamonds of the diameter of 130 nm reaches the values of 1, while for the sphere of 640 nm it is 4.5. Importantly, as Fig. 1 shows, the values of Purcell factor change dramatically as the position of the defect varies along the radius. There is also an optimal position for the NV-center inside the nanodiamond that will produce the strongest emission enhancement. This position depends on the size of the nanodiamond, and in case of a spherical particle of the radius of 640 nm, the normalized optimal position is 0.66, while for the 130 nm sphere it is 0.6. In real experiments, however, various effects including roughness, irregular shape, and a large number of emitters of different orientations in different locations will reduce the observable emission rate. By further calculating the effects of randomness of the emitter orientation and location, and comparing the results to the deeply subwavelength nanodiamonds, we reveal that the Purcell factor for large resonant nanodiamonds is 4.5 times larger than in subwavelength nanoparticles.

While such technology is not available at the moment, we envisage that in future, by correctly choosing the size and shape of dielectric nanodiamonds and controllably placing the NV-center at a desired depth, we will be able to efficiently overlap the optical Mie-type resonances of particles with the photoluminescence spectrum and achieve manifold emission rate enhancement.

3. Conclusion

We have studied the Purcell effect in spherical nanodiamonds with multiple embedded NV-centers and found that it is possible to obtain the emission rate increase of 4.5. We predict that by controlled positioning of the NV-centers inside the nanoparticle one can achieve much larger values of the Purcell factor.

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