Spontaneous emission beyond dipole approximation in nanoscopic environments

K. Słowik[*1], **M. Kosik**[1]

1. Institute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University, Grudziadzka 5, 87-100 Torun, Poland

The possibility to control the spontaneous emission lifetime of quantum emitters by tailoring their surroundings was first investigated in the pioneering work of Purcell [1]. An excited quantum emitter can decay to the photonic modes of its environment, whose local density can be modified by the surroundings. Such influence on emitters such as atoms, molecules and quantum dots has been experimentally verified in various types of cavities or band-gap environments, including semiconductor microstructures [2], photonic crystals [3], and plasmonic nanoparticles [4], where the emission rate was boosted by so-far the record 3 orders of magnitude.

Usually only the electric dipole contribution to the spontaneous emission of quantum emitters is considered, which is justified in the case of negligible spatial variations of the electric field over the size of the quantum emitter. However, nanoscopic environments are capable of localizing the electric field into nanometric spatial domains, providing high intensities and spatial modulations at the length scale of the emitter. Thus, higher order multipolar contributions to light-matter coupling may become relevant. Until now, the enhancement by a nanostructure of magnetic dipole emission of lanthanide ions, such as Eu^{3+} and Er^{3+}, was reported [5]. Large enhancement of quadrupole transitions was predicted [6]. Transitions driven with several multipolar mechanisms have been observed in semiconductor quantum dots [7].

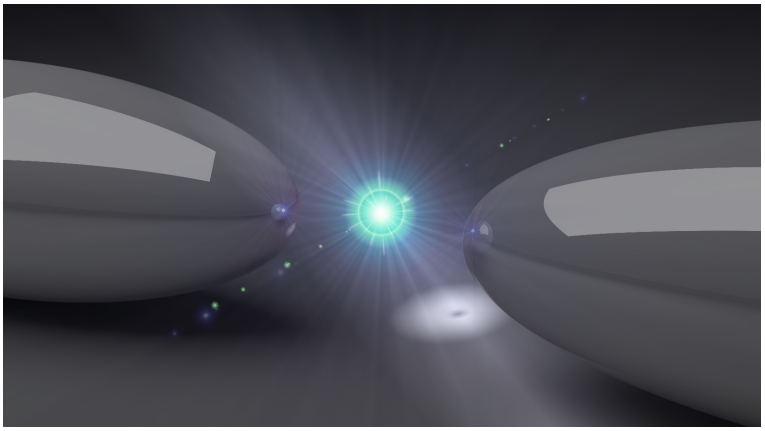

Fig. 1: Artistic impression of a pair of nanoellipsoids influencing radiative properties of a quantum emitter.

Here, we derive expressions for spontaneous emission rates beyond the electric dipole approximation, taking into account the magnetic dipole and electric quadrupole components [8]. Our approach generalizes the one introduced in Ref. [9], based on the Green's tensor formalism applied to quantize electromagnetic fields and account for resulting vacuum fluctuations. A Green's tensor naturally takes into account the structure of electromagnetic surroundings, and is suited to describe nanoparticles made of dispersive and lossy materials such as metals. Exemplary cases will be discussed where the terms beyond the electric dipole significantly influence the emission rate or even dominate it. Finally, we will investigate interference of different multipolar components, which may lead to an even stronger enhancement of the emission rate, or - contrary - to its suppression, leading to enhanced lifetimes of quantum emitters.

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[*]Corresponding author: karolina@fizyka.umk.pl