Tailoring two-photon spontaneous emission: framework and nanoantenna design for interference and directionality

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Abstract. We develop a framework that computes two-photon spontaneous emission (TPSE) spectra of a quantum emitter near an arbitrarily shaped nanostructure. The model considers the interaction up to the electric quadrupolar order, which is relevant for nanophotonic structures sustaining strongly confined fields that are used to enhance and to tailor spontaneous emission processes. Moreover, we consider interference effects between multipolar two-photon emission channels, for the first time to our knowledge. First, we show for a $s \rightarrow s$ transition of a hydrogen atom placed under a silver plasmonic nanodisk a substantial enhancement in the photon-pair emission rates by 5 and 11 orders of magnitude for the two-electric dipole (2ED) and two-electric quadrupole (2EQ) transitions, respectively. Then for the same emitter under a plasmonic graphene nanotriangle, we demonstrate a breakdown of the electric dipole approximation in the TPSE process where the interference between the 2ED and 2EQ transitions is important, as it increases the total rate by 63 %. Third, we explore platforms where entangled photons of different energy are emitted in the far-field in different directions. In the end, our framework is a complete tool to design emitters and nanostructures for the TPSE process, leading to a rich assortment of functional nanoantennas.

Two-photon spontaneous emission (TPSE) is a broadband second-order process in the field of light-matter interaction that involves the simultaneous emission of two photons from a quantum emitter. Enhancing and tailoring this process is of great interest, as it promises several applications, especially for quantum applications [1]. TPSE is an alternative to conventional entangled photon pairs sources using the parametric down-conversion process in nonlinear crystals.

Despite the interest in controlling the TPSE process, it typically occurs 8 to 10 orders of magnitude slower than the competing spontaneous emission of a single photon [2]. However, electromagnetic interactions with the surrounding environment are known to modify spontaneous emission rates of quantum emitters: the Purcell effect. Specifically, in nanophotonic structures sustaining strongly confined fields such as plasmonic nanostructures, higher-order transitions can be enhanced and even outperform the electric-dipole single photon transitions, such as multiquanta and multipolar transitions [2, 3].

First, we develop a general framework that calculates the two-photon Purcell effect of a quantum emitter near an arbitrarily shaped nanostructure by considering the interaction up to the electric quadrupolar order [4, 5]. It is based on the classical computation of Purcell factors that can be computed by modeling point emitters in conventional electromagnetic simulations, thus allowing the consideration of complex geometries without available analytical models. The formulation in terms of Purcell factors allows the separate computation of the radiative and non-radiative emissions channels, thus enabling computation of the quantum efficiency.

Specifically, the framework is used to calculate the two-photon Purcell effect for an $s \rightarrow s$ transition (wavelength of 470 nm) of a hydrogen atom positioned 10 nm under a plasmonic silver nanodisk. Figure 1 demonstrates a substantial enhancement in the photon-pair emission rates by 5 and 11 orders of magnitude for the two-electric dipole (2ED, upper graph) and two-electric quadrupole (2EQ, lower graph) transitions, respectively [5].

Then, we incorporate in our framework the interference between the 2ED and 2EQ decay channels [6]. Indeed, between two s states these decay channels occur simultaneously, and interference effects must be considered if the difference between the transition rates of both channels is less than 3 orders of magnitude because the interference can lead to a modification greater than 10 % of the total transition rate. Furthermore, in our model interferences between multipolar two-photon transitions are calculated through the calculation of classical interferences between multipolar sources superposed at the position of the emitter.

We calculate the interference between the 2ED and 2EQ decay channels for a $5s \rightarrow 3s$ transition (wavelength of 1.28 μm) of a hydrogen atom positioned 2 nm under a plasmonic graphene nanotriangle. Figure 2 shows a breakdown of the electric dipole approximation in the TPSE process where interference effects have a

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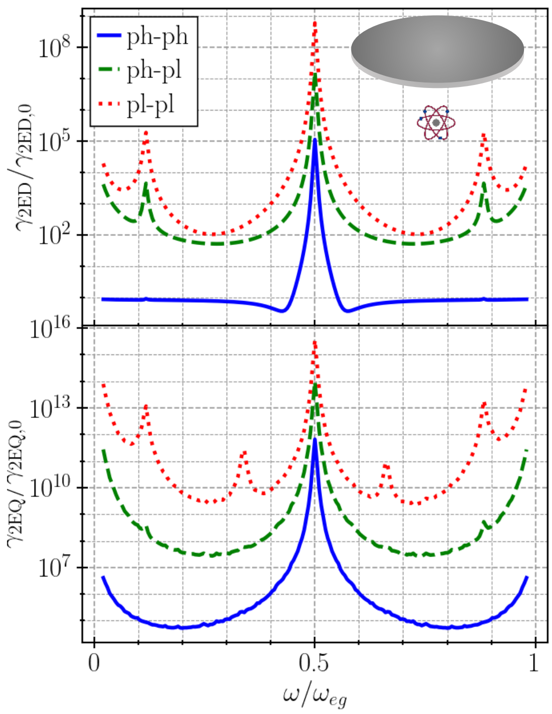

Fig. 1. Two-photon Purcell effect of the 2ED and 2EQ decay channels for the emission of two photons (ph-ph, solid), one photon and one plasmon (ph-pl, dashed), and two plasmons (pl-pl, dotted), with $\hbar\omega_{eg}$ the transition energy. We consider the s-s transition (470 nm wavelength) of a hydrogen atom that is 10 nm below a 25 nm diameter silver nanodisk, on its axis of symmetry.

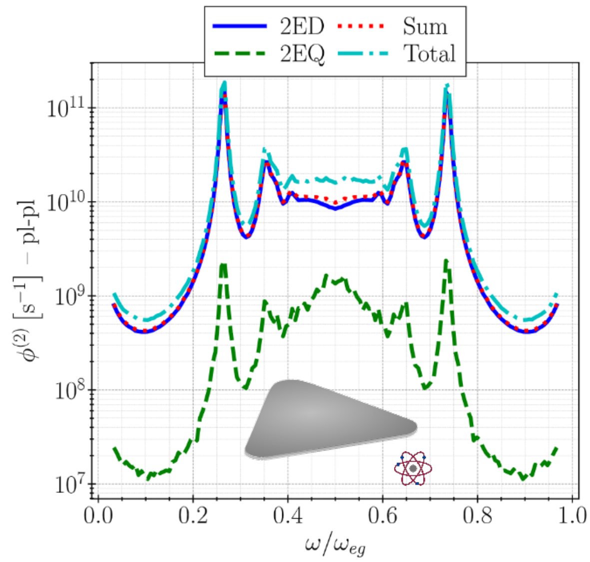

Fig. 2. Interference between the 2ED (blue, solid) and the 2EQ (green, dashed) decay channels for the excitation of two plasmons. At $\omega/\omega_{eg} = 0.5$, the total rate that includes the interference contribution (cyan, dash-dotted) is 63 % larger than the rate neglecting it (red, dotted). We consider the 5s-3s transition (1.28 µm wavelength) of a hydrogen atom that is 2 nm below a corner of a 23 nm-side length equilateral triangle of graphene with a doping of 0.31 eV.

significant contribution to the transition rate. For two quanta emitted at the same frequency ($\omega = \omega_{eg}/2$, with $\hbar\omega_{eg}$ the transition frequency), the 2EQ transition is 6 times smaller than the 2ED transition, but the interference increases the total rate by 63 %.

Finally, we will show that the framework is a powerful tool to design TPSE nanoantennas, like the rich assortment of nanoantennas that exists to tailor the single-photon emission process [7]. Currently, we analyze results for two perpendicular silver nanodisks in which entangled photons of different energy are emitted in the far-field in different directions, which can be interesting for applications.

In conclusion, we developed a powerful framework based on the classical computation of Purcell factors to design nanoantennas for the two-photon spontaneous emission process. It can be used for arbitrarily shaped nanostructures in order to optimize the efficiency and directionality, amongst other applications.

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