Abstract

We present a framework that computes the Two-Photon Spontaneous Emission (TPSE) transition rates of a quantum emitter placed near a nanostructure of arbitrary shape and beyond the electric dipole approximation. Interestingly, it relies on the classical computation of one-photon Purcell factors. The developed framework is relevant for current plasmonic nanocavities that are ideal for tailoring and enhancing transition rates of spontaneous emission processes. This discipline promises, for example, efficient entangled photon sources in quantum computing. Finally, we show that placing an emitter close to a silver nanodisk enhances the TPSE transition rate of its electric dipole and quadrupole transitions by 5 and 12 orders of magnitude, respectively.

Background

- **Two-Photon Spontaneous Emission (TPSE) processes**: second-order processes, 8 to 10 orders of magnitude slower than the competing spontaneous emission of a single photon [1]
- **2D plasmonic nanostructures**: ideal to harness two-quantum emission processes [2]
  - Light confinement at the atomic scale
  - Light emission enhancement via the Purcell effect by several orders of magnitude [1, 3]
  - Standard electric dipole approximation no longer appropriate [3]
- Study of advanced nanostructures hampered by a lack of efficient numerical and theoretical methods

Framework

**System → Perturbative approach**

Quantum emitter

Interaction studied up to electric quadrupole order

Plasmonic nanostructure of arbitrary shape

Relation between the TPSE rates and Purcell factors

- Established by Muniz: 2ED transition, not for arbitrary shape [2]
- Established by us: 2ED, 2MD and 2EQ transitions, for arbitrary shape
- Examples: 2ED and 2EQ contributions to the total TPSE transition rate

\[
\begin{align*}
\gamma_{2ED} (\omega, R) &= \sum_{j, k=1}^{3} D_{j,k} (\omega, \omega_{eg} - \omega) D_{k,j} (\omega, \omega_{eg} - \omega) F_{L_{j}} (\omega, R) F_{E_{k}} (\omega_{eg} - \omega, R) \\
\gamma_{2MD} (\omega, R) &= \sum_{j, k=1}^{3} Q_{j,k} (\omega, \omega_{eg} - \omega) Q_{k,j} (\omega, \omega_{eg} - \omega) F_{L_{j}} (\omega, R) F_{E_{k}} (\omega_{eg} - \omega, R) \\
\gamma_{2EQ} (\omega, R) &= \sum_{j, k=1}^{3} Q_{j,k} (\omega, \omega_{eg} - \omega) Q_{k,j} (\omega, \omega_{eg} - \omega) F_{L_{j}} (\omega, R) F_{E_{k}} (\omega_{eg} - \omega, R)
\end{align*}
\]

Transition rate tailoring

- Normalized tensors: second-order matrix elements of multipolar moment operators (MO)
- Depend solely on the electronic structure of the emitter
- Calculated analytically for a specific transition of the emitter

Function expressed in terms of Purcell factors of the two emitted quanta of complementary energy

- Depend only on the photonic environment
- Computed classically with COMSOL Multiphysics® software (finite element method)

Applicability: to an s → s transition of an emitter close to a silver nanodisk

Application

- Enhancement of 5 and 12 orders of magnitude for the two-photon electric dipole and electric quadrupole transitions, respectively

Perspective: study interference effects between TPSE multipolar channels [3]

Conclusion

- Computes TPSE transition rates of a quantum emitter near a plasmonic nanostructure of arbitrary shape and beyond the electric dipole approximation
- Based on the computation of one-photon Purcell factors

Application

- to an s → s transition of an emitter close to a silver nanodisk

References


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