Photonic modeling of two-photon spontaneous emission processes

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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-quanta emission processes [2]

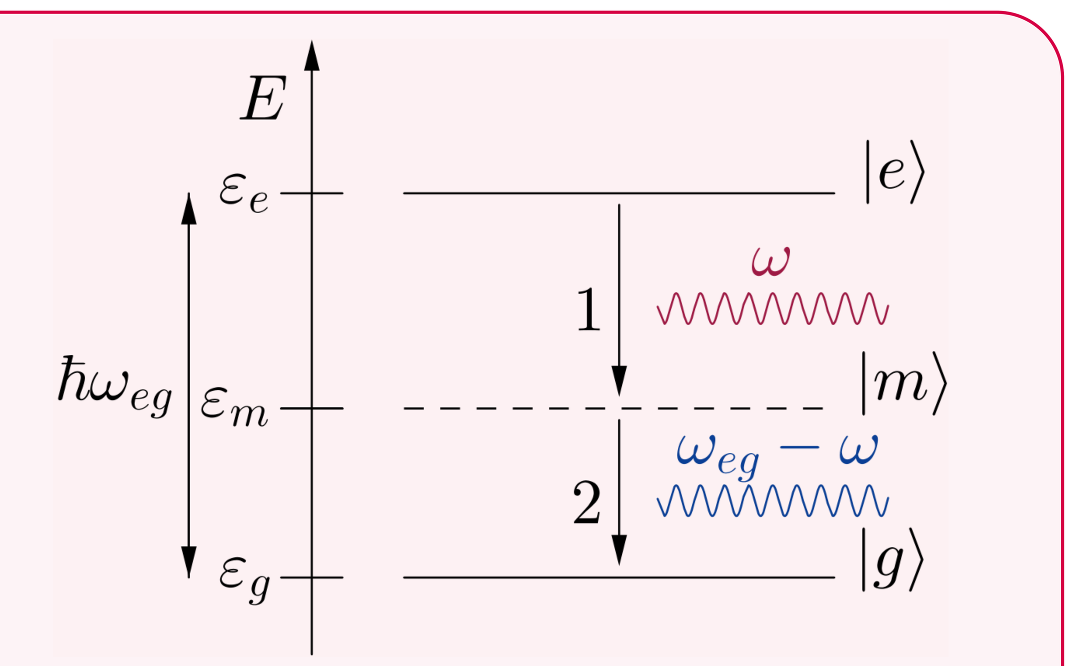
→ Light confinement at the atomic scale

✓ 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

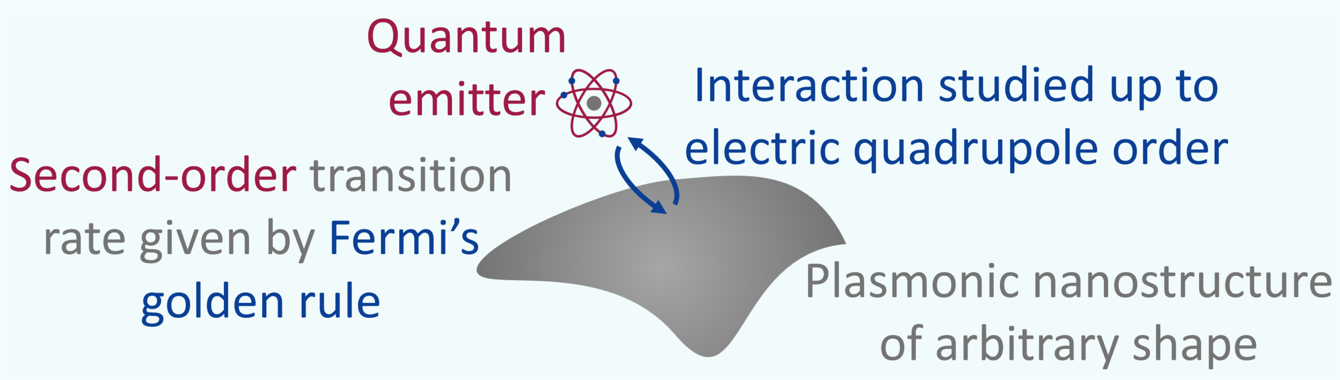
Need for an efficient framework which goes beyond the electric dipole approximation by considering higher-order multipolar contributions to high-order processes → Electric Dipole (ED), Magnetic Dipole (MD), Electric Quadrupole (EQ)



Second-order transition: an excited emitter emits a first quantum ω then emits a second quantum $\omega_{eg}-\omega$ from a virtual intermediate state

Framework

System → Perturbative approach



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

$$\frac{\gamma^{(2)}_{2\mathrm{ED}}(\omega;\boldsymbol{R})}{\gamma^{(2)}_{2\mathrm{ED},0}(\omega)} = \sum_{i,j,k,l=1}^{3} \bar{\mathcal{D}}_{ij}(\omega,\omega_{eg}-\omega)\bar{\mathcal{D}}^*_{kl}(\omega,\omega_{eg}-\omega) F^{\mathrm{ED}}_{ik}(\omega;\boldsymbol{R})F^{\mathrm{ED}}_{jl}(\omega_{eg}-\omega;\boldsymbol{R})$$

$$\frac{\gamma^{(2)}_{2\mathrm{EQ}}(\omega;\boldsymbol{R})}{\gamma^{(2)}_{2\mathrm{EQ},0}(\omega)} = \sum_{\substack{\mu,\nu,\alpha,\beta=1\\ \nu\geq\mu,\,\beta\geq\alpha}}^{5} \bar{\mathcal{Q}}_{\mu\nu}(\omega,\omega_{eg}-\omega)\bar{\mathcal{Q}}^*_{\alpha\beta}(\omega,\omega_{eg}-\omega) F^{\mathrm{EQ}}_{\mu\alpha}(\omega;\boldsymbol{R})F^{\mathrm{EQ}}_{\nu\beta}(\omega_{eg}-\omega;\boldsymbol{R})$$

Vacuum

Emitter's position

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

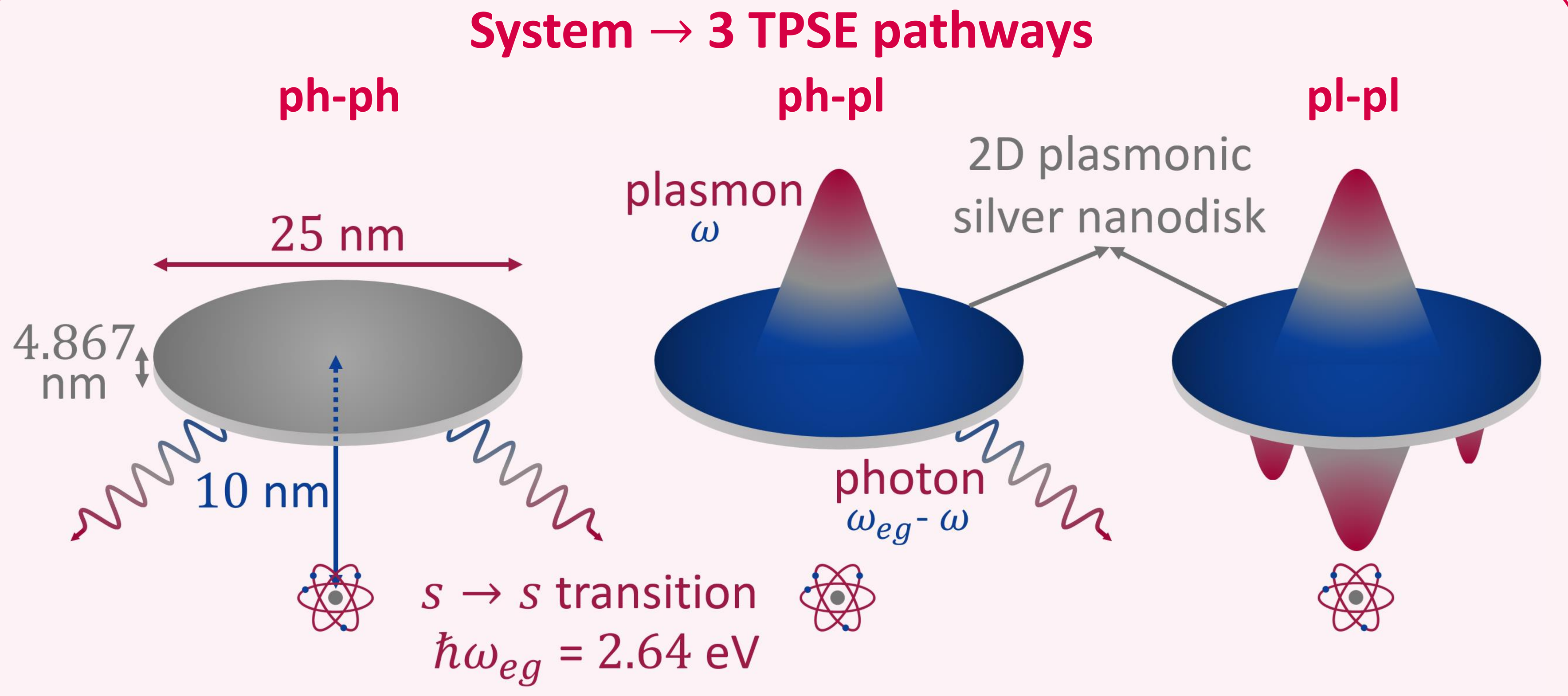
- Functions expressed in terms of Purcell factors of the two emitted quanta of complementary energy
- Depend only on the photonic environment
- Computed classically with COMSOL Multiphysics® software (finite element method)

Power emitted by a classical emitter modelled by a radiating point source

$$\frac{W_{\mathrm{MO},\varphi}(\boldsymbol{R})}{W_{\mathrm{MO},0}} = P^{\mathrm{MO}}_{\varphi}(\boldsymbol{R}) = \frac{\Gamma^{(1)}_{\mathrm{MO},\varphi}(\boldsymbol{R})}{\Gamma^{(1)}_{\mathrm{MO},0}}$$

Application

System → 3 TPSE pathways



Results

✓ Agreement with the analytical results for the 2ED transition [2]

✓ Photon-pair emission rate enhanced by, respectively, 5 and 12 orders of magnitude for the 2ED and 2EQ transitions at $\omega = \omega_{eg}/2$

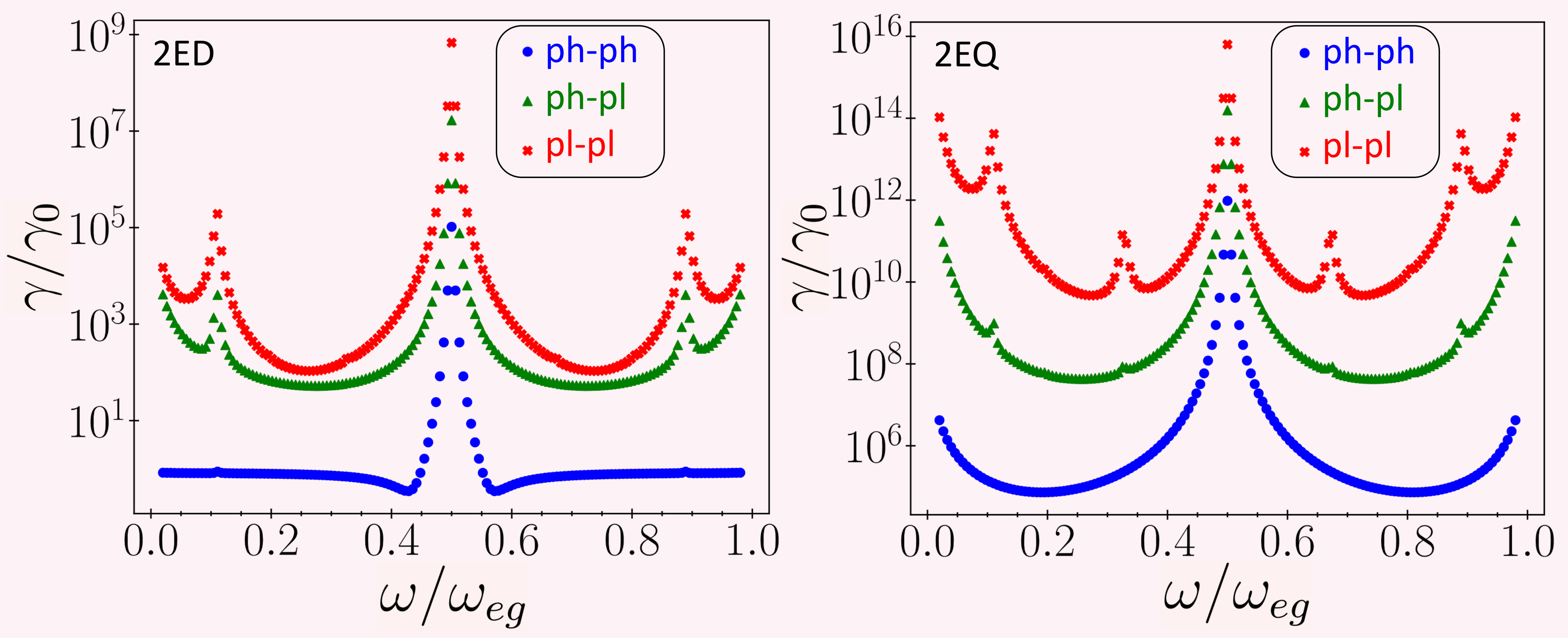


Figure – Relaxation channels of the 2ED and the 2EQ contributions to the total spectral TPSE rate between two symmetric states of an emitter

Conclusion

Framework

→ 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 $s \to s$ transition of an emitter close to a silver nanodisk

→ 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]

References

[1] Rivera et al. Shrinking light to allow forbidden transitions on the atomic scale. Science, 353(6296), 263-269 (2016).

[2] Muniz et al. Two-photon spontaneous emission in atomically thin plasmonic nanostructures. *Physical Review Letters*, *125*(3), 033601 (2020).

[3] Rusak et al. Enhancement of and interference among higher order multipole transitions in molecules near a plasmonic nanoantenna. Nat Commun 10, 5775 (2019).

Acknowledgments

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