Photonic modeling of two-photon spontaneous emission processes beyond the electric dipole approximation

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Abstract

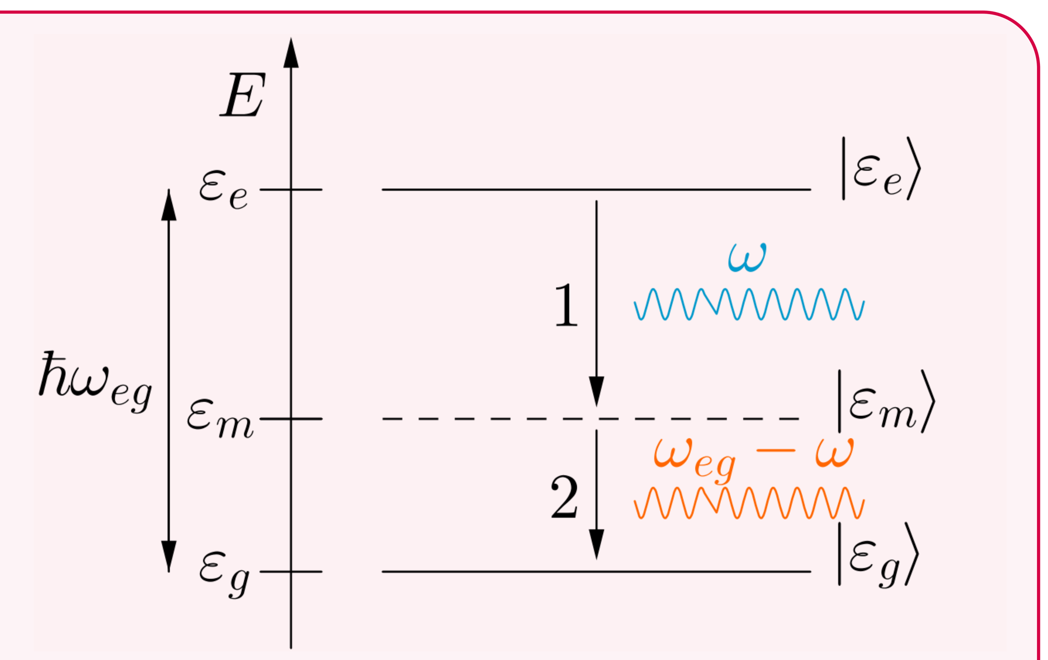
We present a framework that can be used to study Two-Photon Spontaneous Emission (TPSE) processes of a quantum emitter placed near a nanostructure beyond the standard electric dipole approximation. This is relevant for current nanocavities, used for tailoring and enhancing transition rates of spontaneous emission processes. This discipline promises, for example, efficient entangled photon sources in quantum computing.

The developed framework relies on the classical computation of Purcell factors of the one-photon spontaneous emission. 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 [3]
  - → Light confinement at the atomic scale
    - ✓ Light emission enhancement via the Purcell effect by several orders of magnitude [1, 2]
    - ✗ Standard electric dipole approximation no longer appropriate [2]
  - ✗ Study of advanced nanostructures hampered by a lack of efficient numerical and theoretical methods

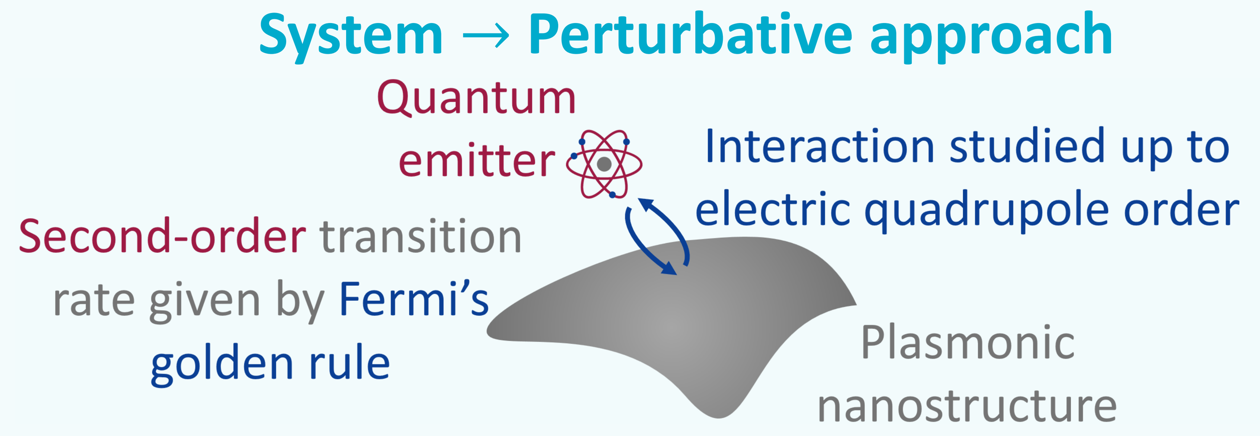
Need for an efficient framework which goes beyond the electric dipole approximation by considering higher-order multipolar contributions to high-order processes → Electric dipole (E1), Magnetic dipole (M1), Electric quadrupole (E2)



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 rate and Purcell factors:

→ Established for the 2E1 [3], 2M1, and 2E2 transitions

→ Examples: spectral distribution contributions of the transitions 2E1 and 2E2 to the total two-quanta transition rate

$$\frac{\gamma^{(2)}_{2\mathrm{E1}}(\omega;\boldsymbol{R})}{\gamma^{(2)}_{2\mathrm{E1},0}(\omega)} = \sum_{i,j=1}^{3} \left|\bar{\mathcal{D}}_{ij}(\omega,\omega_{eg}-\omega)\right|^2 P_i^{\mathrm{E1}}(\omega;\boldsymbol{R})\, P_j^{\mathrm{E1}}(\omega_{eg}-\omega;\boldsymbol{R})$$

$$\frac{\gamma^{(2)}_{2\mathrm{E2}}(\omega;\boldsymbol{R})}{\gamma^{(2)}_{2\mathrm{E2},0}(\omega)} = \sum_{\mu,\nu=1}^{6} C_{\mu\nu} \left|\bar{\mathcal{Q}}_{\mu\nu}(\omega,\omega_{eg}-\omega)\right|^2 P_\mu^{\mathrm{E2}}(\omega;\boldsymbol{R})\, P_\nu^{\mathrm{E2}}(\omega_{eg}-\omega;\boldsymbol{R})$$

Emitter's position ($\boldsymbol{R}$)

Vacuum ($\gamma^{(2)}_{2\mathrm{E1},0}(\omega)$, $\gamma^{(2)}_{2\mathrm{E2},0}(\omega)$)

Transition rate tailoring

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

- Voigt notation for the second-order matrix elements of Q

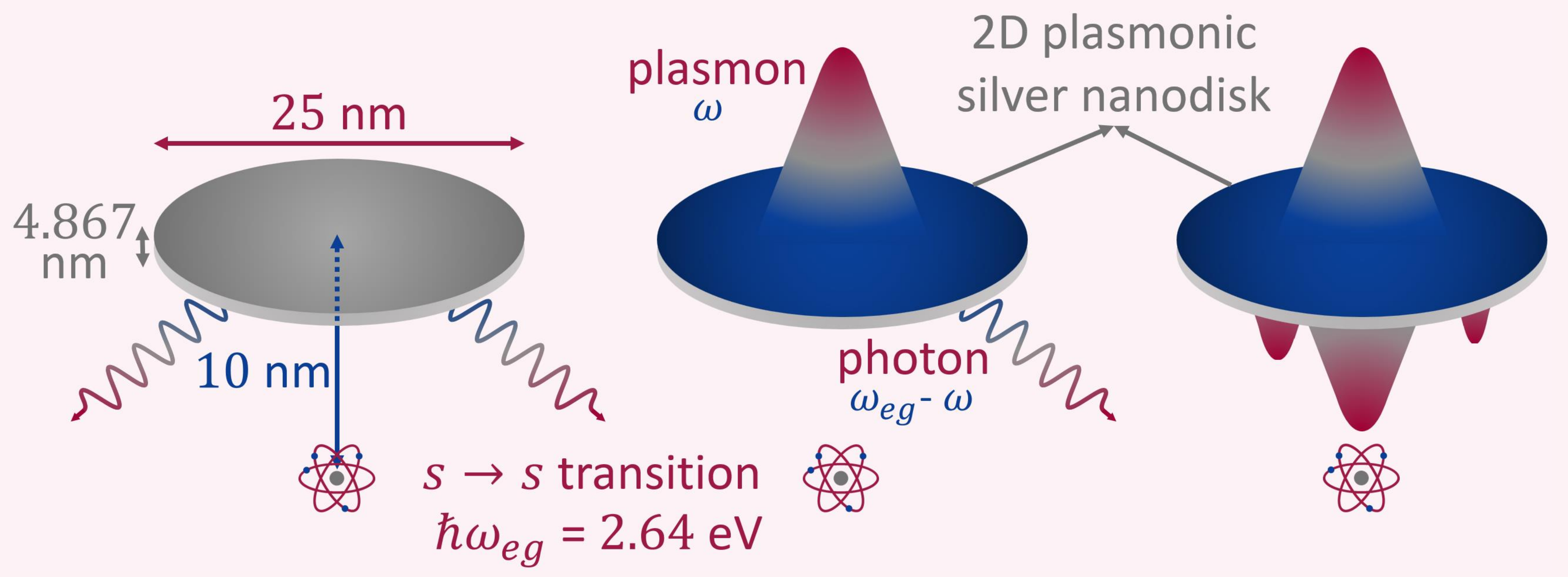
- Purcell factors of the two emitted quanta of complementary energy
- Depend only on the photonic environment
- Computed classically with COMSOL Multiphysics® software (finite element method)

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

Power emitted by a classical emitter, modelled by a radiating point source ($W_{\mathrm{MO},\varphi}(\boldsymbol{R})$)

Application

System → 3 TPSE pathways: ph-ph, ph-pl, pl-pl



Results

- ✓ Agreement with the analytical results for the 2E1 transition [3]
- ✓ Photon-pair emission rate enhanced by, respectively, 5 and 12 orders of magnitude for the 2E1 and 2E2 transitions at $\omega = \omega_{eg}/2$

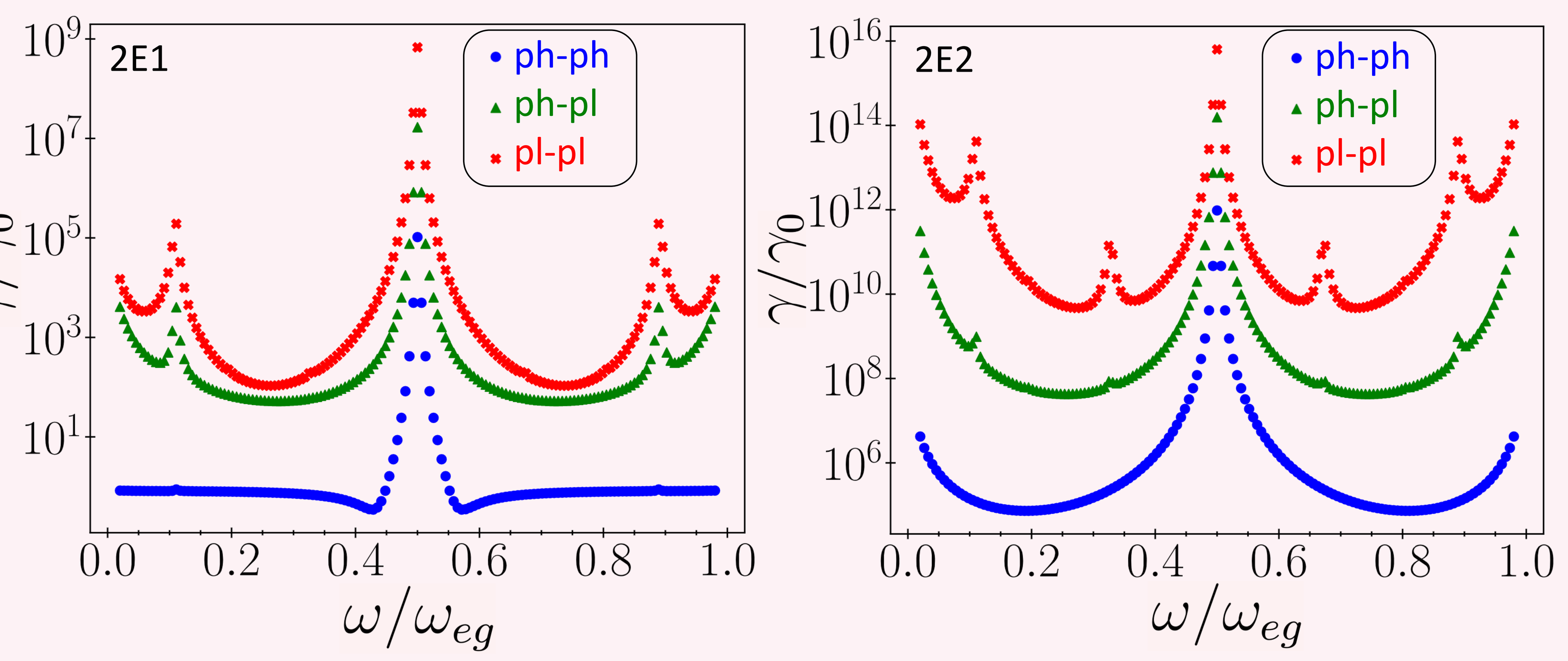


Figure – Relaxations channels of the 2E1 and the 2E2 contributions to the total spectral TPSE rate between two symmetric states of an emitter

Conclusion

Framework

→ Study two-photon spontaneous emission processes of a quantum emitter near a plasmonic nanostructure beyond the electric dipole approximation

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

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

Acknowledgments

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