# General framework for two-photon spontaneous emission near plasmonic nanostructures

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## Abstract

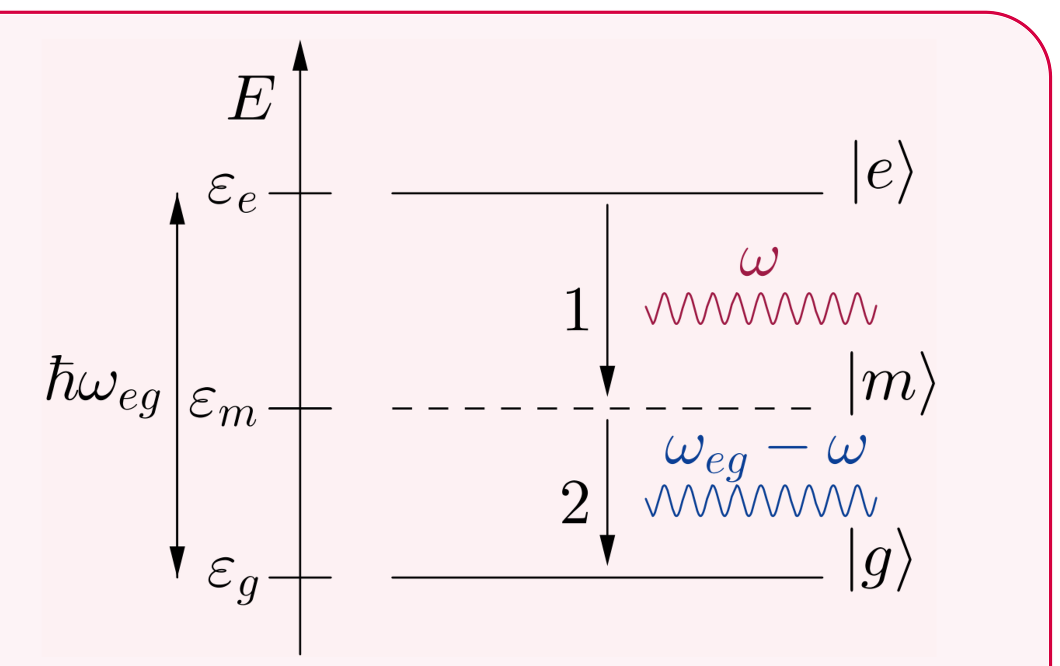
Our newly developed general framework efficiently computes the Two-Photon Spontaneous Emission rate of a quantum emitter positioned near any photonic structure beyond the dipolar approximation. This is particularly relevant for plasmonic nanocavities, which exhibit highly confined light. Interestingly, the influence of the environment is determined through the classical computation of Purcell factors via electromagnetic simulations, avoiding tedious analytic calculations. We show that placing a hydrogen-like emitter close to a silver nanodisk enhances the two-electric dipole and the two-electric quadrupole transition rates by 5 and 11 orders of magnitude, respectively. This discipline promises, for example, efficient entangled photon sources.

## Background

- **Two-Photon Spontaneous Emission (TPSE) processes:** second-order processes, 8 to 10 orders of magnitude slower than the competing single-photon spontaneous emission [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]
    - ✓ Breakdown of the electric dipole selection rule [3] → Forbidden transitions accessible [1]
  - ✗ Study of advanced nanostructures hampered by a lack of efficient numerical and theoretical methods

Need for an efficient and general framework which goes beyond the electric dipole approximation by considering higher-order multipolar contributions to second-order processes

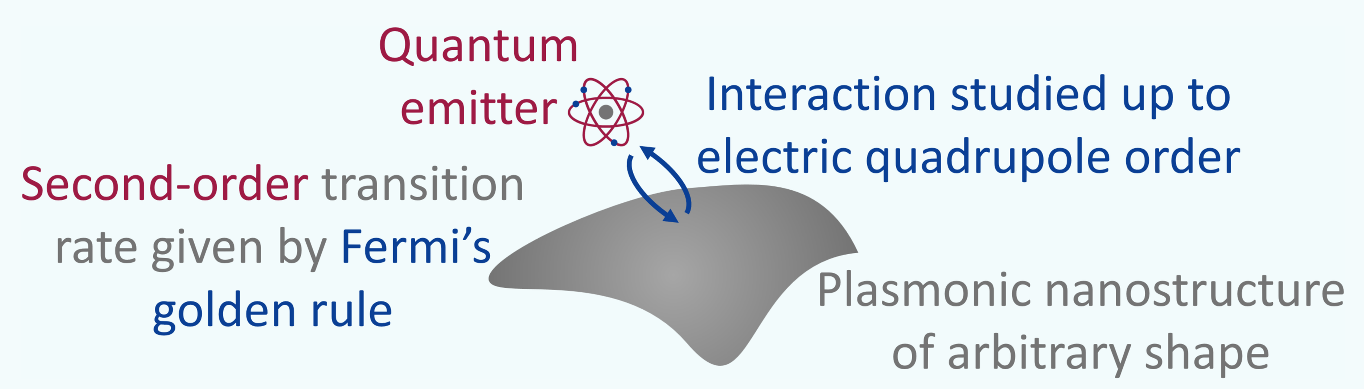
Electric Dipole (ED)
Magnetic Dipole (MD)
Electric Quadrupole (EQ)




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

## Framework [4]

### System → Perturbative approach




### Relation between the TPSE rates and Purcell factors

→ Muniz: 2ED transition, only for symmetric structures [2]

→ Our work: 2ED, 2MD and 2EQ transitions, also for asymmetric structures

$$\frac{\gamma^{(2)}_{\mathrm{2ED}}(\omega;\boldsymbol{R})}{\gamma^{(2)}_{\mathrm{2ED},0}(\omega)} = \sum_{i,j,a,b=1}^{3} \hat{\mathcal{D}}^{eg}_{iajb}(\omega,\omega_{eg}-\omega)\, F^{\mathrm{ED}}_{ij}(\omega;\boldsymbol{R})\, F^{\mathrm{ED}}_{ab}(\omega_{eg}-\omega;\boldsymbol{R})$$

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

Vacuum

Emitter's position

Transition rate tailoring

- Normalized tensors: multipolar second-order transition moments
- Depend only on the electronic structure of the emitter
- Calculated analytically for a specific transition of the emitter

- Tensors expressed as a function of Purcell factors of the two emitted quanta of complementary energy
- Depend only on the photonic environment
- Computed classically with COMSOL Multiphysics® (FEM)

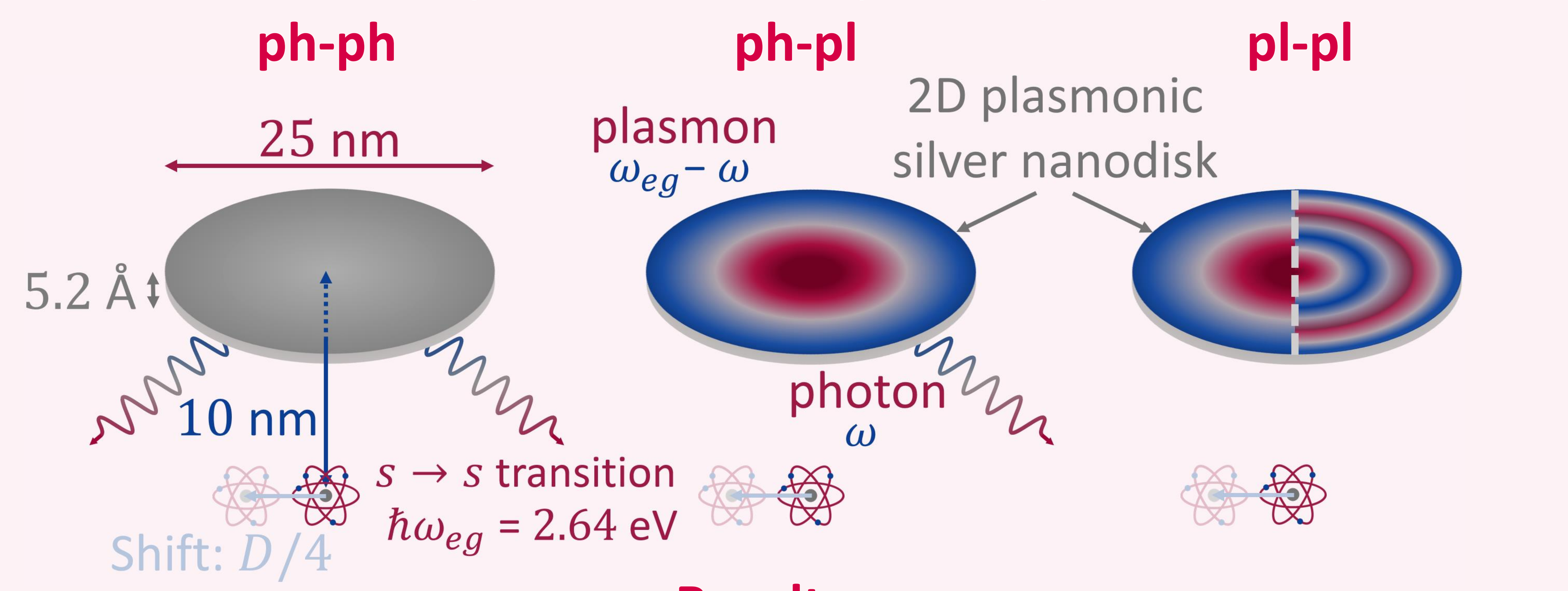
$$\frac{W_\varphi}{W_0} = P_\varphi = \frac{\Gamma^{(1)}_\varphi}{\Gamma^{(1)}_0}$$

- Decomposition into radiative and non-radiative parts

- Power emitted by a classical radiating point source
- To calculate for different source orientations

## 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 11 orders of magnitude for the 2ED and 2EQ transitions at $\omega = \omega_{eg}/2$
- ✓ Emitter off-axis → Additional peaks

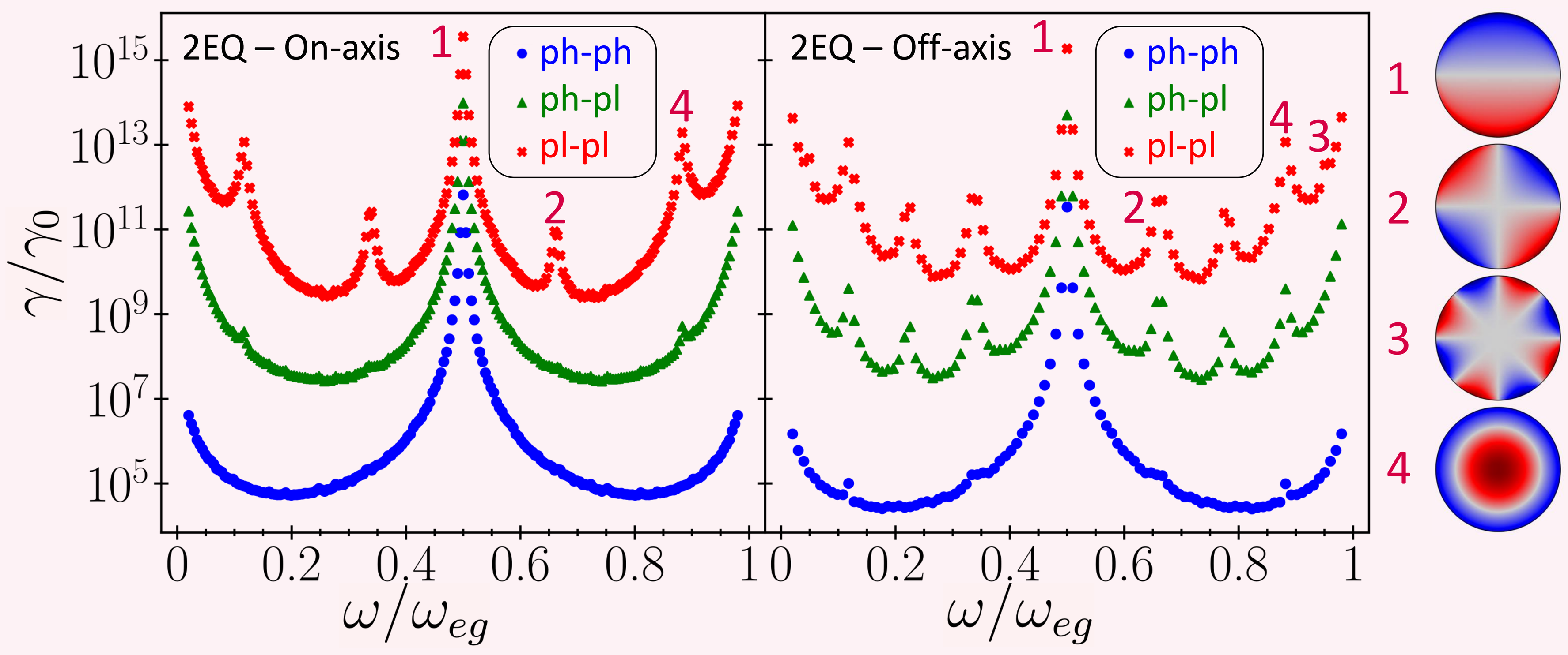



**Figure –** Relaxation channels of the 2EQ contribution to the total spectral TPSE rate for an emitter placed on-axis and off-axis under the nanodisk

## Conclusion

### Framework

→ **Efficiently computes TPSE rate** of a quantum emitter near an **arbitrary shaped nanostructure** and **beyond the electric dipole approximation**

→ **Based on the computation of Purcell factors via classical simulations**
- ✓ Avoids tedious analytic calculations
- ✓ Allows the study of complex geometries and design optimization
- ✓ Allows the separate calculation of the radiative and non-radiative channels

**Application** to a hydrogen-like emitter close to a silver nanodisk → Enhancement

**Perspective:** study interference effects between multipolar TPSE 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. *PRL*, *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).
[4] Smeets et al. General framework for two-photon spontaneous emission near plasmonic nanostructures. Submitted (2023)

### Acknowledgments

We acknowledge support from the FRS-FNRS (Research project T.0166.20) and from Action de Recherche Concertée (project ARC-21/25 UMONS2)

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