Photonic modeling of two-photon spontaneous emission processes

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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 and is relevant for current plasmonic nanocavities that are ideal for tailoring and enhancing transition rates of spontaneous emission processes.

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.

References

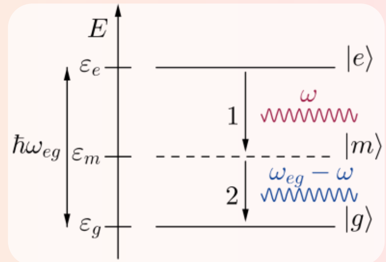
[1] Muniz *et al,* Physical Review Letters 125(3), 033601 (2020)
[2] Rusak *et al,* Nat Commun 10, 5775 (2019)

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
- **2D plasmonic nanostructures:** ideal to harness two-quanta emission processes [1]
  - → Light confinement at the atomic scale
    - ✓ Light emission enhancement via the Purcell effect by several orders of magnitude [2]
    - ✗ Standard electric dipole approximation no longer appropriate [2]
  - ✗ Study of advanced nanostructures hampered by a lack of efficient numerical and theoretical methods

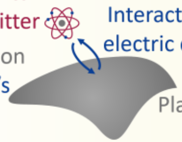
Need for an efficient framework which goes beyond the electric dipole approximation by considering higher-order multipolar contributions to high-order processes

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



Framework

System:
perturbative approach



Relations established for 2ED, 2MD and 2EQ contributions to the total TPSE rate (general)

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

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

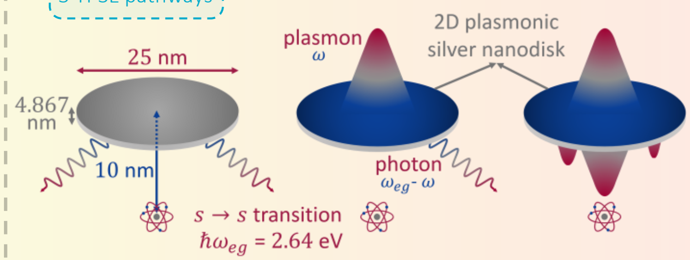
Transition rate tailoring — Vacuum — Emitter's position

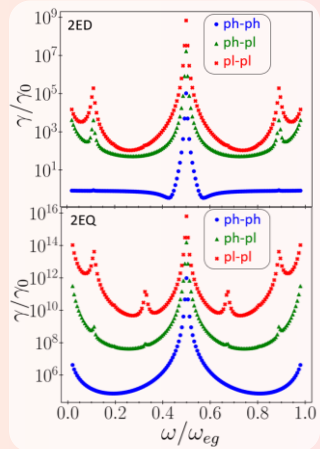
- Normalized tensors: second-order matrix elements of multipolar moment operators (MO)
- Depend only on the emitter's electronic structure
- Calculated analytically for a specific transition of the emitter

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

Application and Conclusion

3 TPSE pathways





Results

✓ Agreement with the analytical results for the 2ED transition [1]
✓ Photon-pair emission rate enhanced by, respectively, 5 and 12 orders of magnitude for the 2ED and 2EQ transitions at $\omega = \omega_{eg}/2$

Perspective: study interference effects between TPSE multipolar channels

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