A general framework for two-photon spontaneous emission near plasmonic nanostructures

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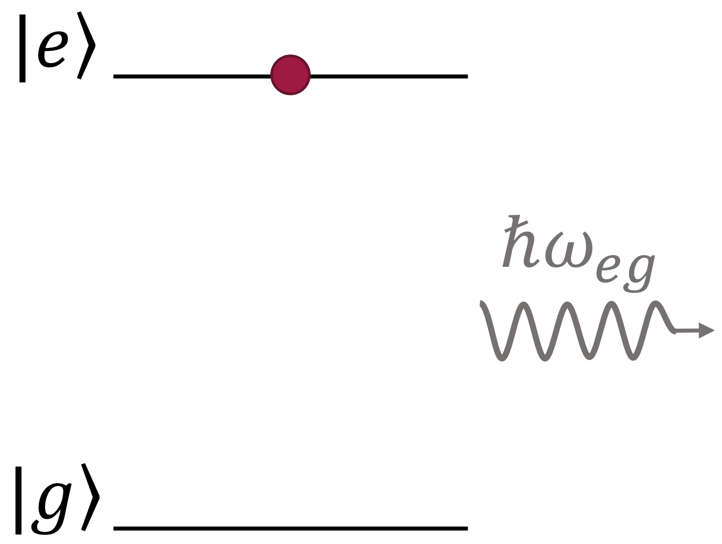
Spontaneous emission

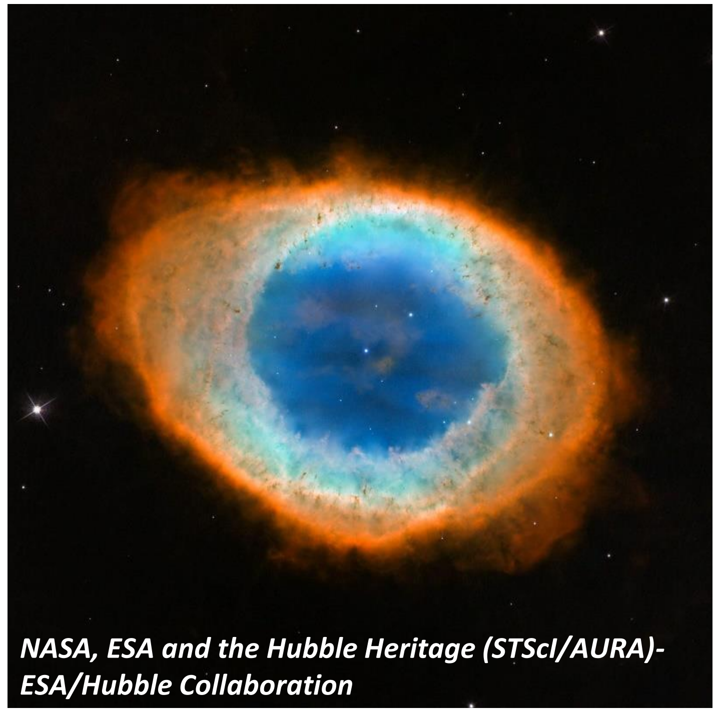
- **Fundamental process** in the field of light-matter interaction

  → Responsible for most of the light we see around us

One-photon spontaneous emission

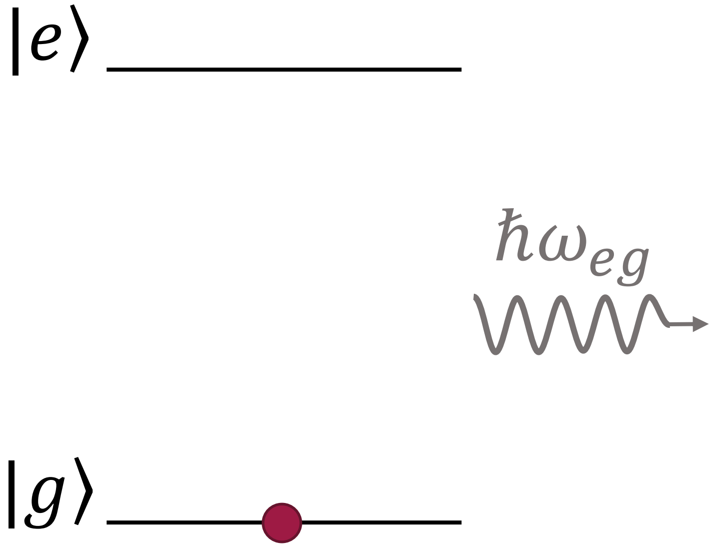


Spontaneous emission

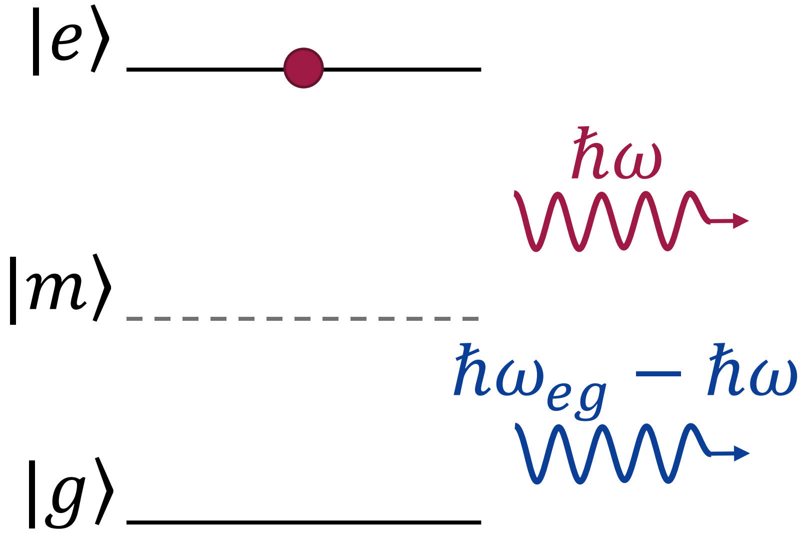

NASA, ESA and the Hubble Heritage (STScI/AURA)-ESA/Hubble Collaboration

- **Fundamental process** in the field of light-matter interaction
  - → Responsible for most of the light we see around us
- **Two-Photon Spontaneous Emission (TPSE):** second-order process
  - → 8 to 10 orders of magnitude slower than the emission of a single photon [1]
  - → Continuous spectrum
  - → Responsible of the lifetime of the 2s state → Explanation of the continuous spectrum coming from planetary nebulae

One-photon spontaneous emission



Two-photon spontaneous emission



[1] Rivera et al. *Science* 353, 263-269 (2016)

Photonic environment

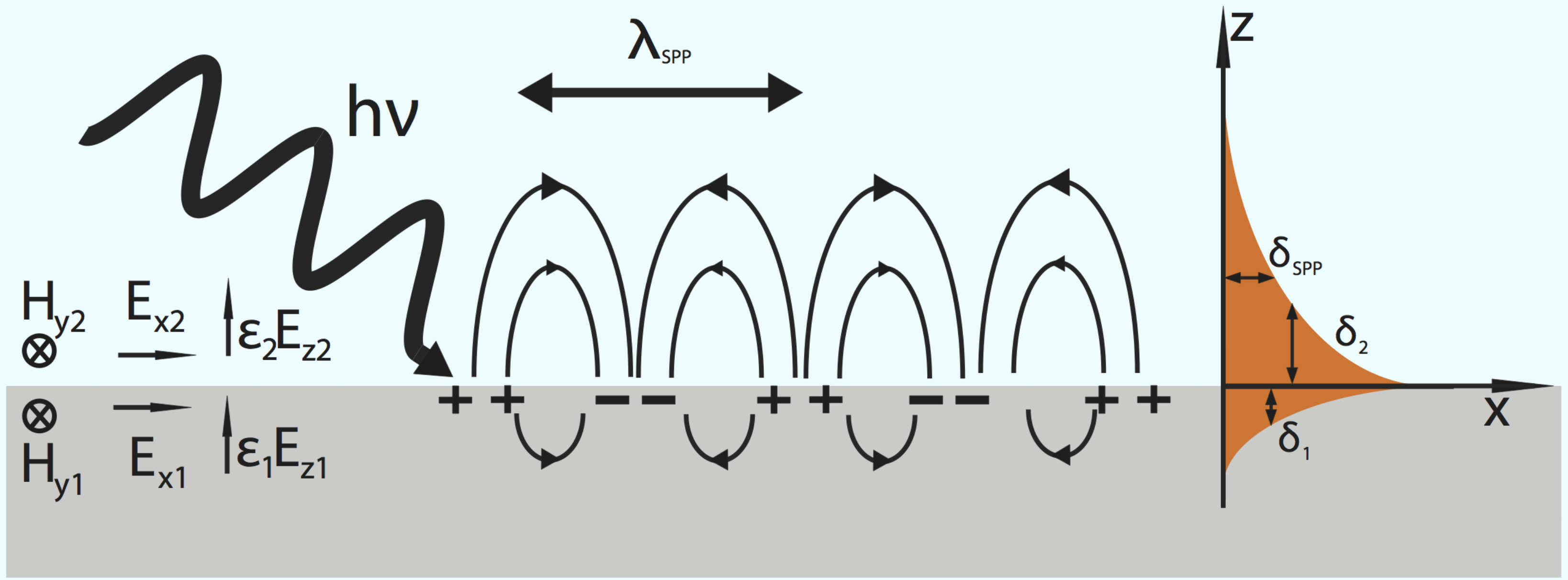
- **Purcell effect (1946):** the spontaneous emission rate of an emitter depends on its environment

$$P = \frac{\Gamma^{(1)}}{\Gamma_0^{(1)}}$$

- **2D plasmonic nanostructures:** ideal to harness two-quanta emission processes [2]

Surface plasmons

→ Collective oscillation of electrons at the interface between a metal and a dielectric
→ The wavelength of the light can be squeezed by two orders of magnitude



Anil Thilsted, via Wikimedia Commons

[2] Muniz et al. *Physical Review Letters* 125(3), 033601 (2020)

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→ Light confinement at the nanoscale

✓ 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], TPSE can dominate [4]

✗ 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

→

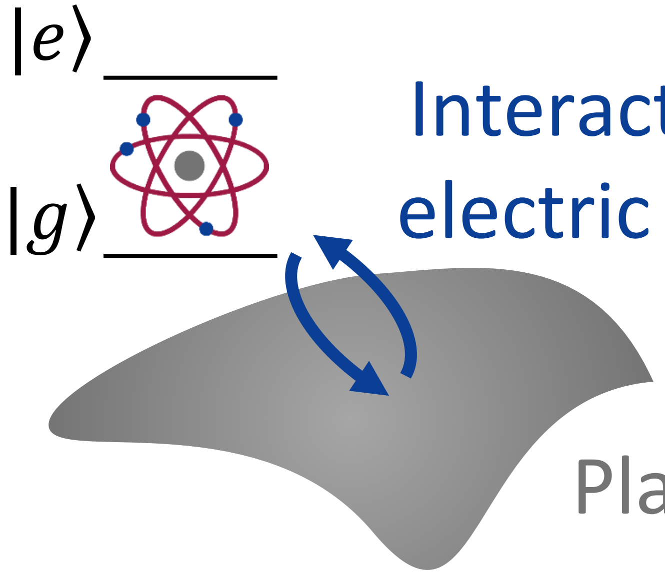
Electric Dipole (ED)
Magnetic Dipole (MD)
Electric Quadrupole (EQ)

[1] Rivera et al. *Science* 353, 263-269 (2016)
[2] Muniz et al. *Physical Review Letters* 125(3), 033601 (2020)
[3] Rusak et al. *Nat Commun* 10, 5775 (2019)
[4] Rivera et al. *Proceedings of the National Academy of Sciences 114(52)*, 13607-12 (2017)

Fermi’s golden rule approach

Quantum emitter

Atom, molecule, QD

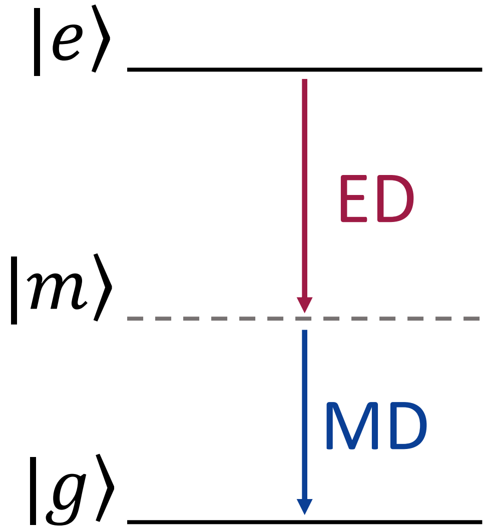


Interaction studied up to electric quadrupole order

Plasmonic nanostructure of arbitrary shape

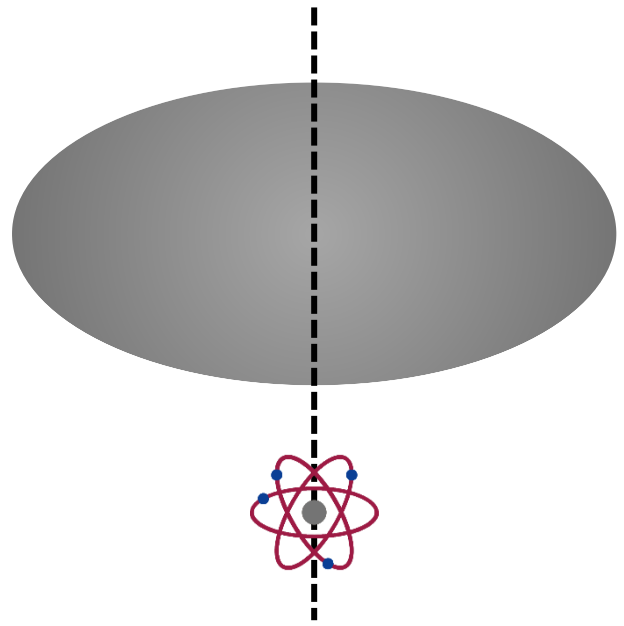
- **Second-order transition rate** given by Fermi’s golden rule

$$\Gamma^{(2)}_{\mathrm{tot}}(\boldsymbol{R}) = \Gamma^{(2)}_{\mathrm{2ED}}(\boldsymbol{R}) + \Gamma^{(2)}_{\mathrm{2MD}}(\boldsymbol{R}) + \Gamma^{(2)}_{\mathrm{2EQ}}(\boldsymbol{R}) + \Gamma^{(2)}_{\mathrm{mixed}}(\boldsymbol{R})$$

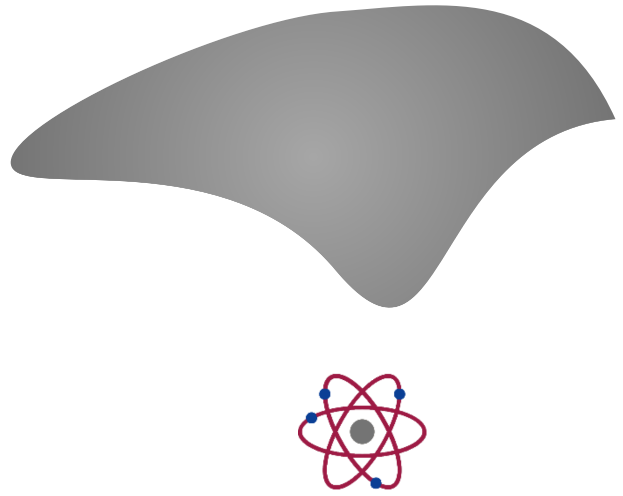


Derivations of TPSE rates

- **Former derivation** [2,5]
  - → Only for the 2ED contribution
  - → Can be applied only for symmetric structures with the emitter at specific positions



- **Our derivation** [6]
  - → 2ED, 2MD, and 2EQ contributions
  - → Can be applied for arbitrary shaped nanostructures with the emitter at any position



[2] Muniz et al. *Physical Review Letters* 125(3), 033601 (2020)
[5] Muniz et al. *Phys. Rev. A* 100, 023818 (2019)
[6] Smeets et al. *Phys. Rev. A,* Submitted (2023)

TPSE rate as a function of one-photon Purcell factors

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

Vacuum

Transition rate tailoring

Emitter's position

Emitter contribution

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

Photonic environment contribution

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

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

- ➢ W_φ: Power emitted by a classical radiating point source
- ➢ To calculate for different source orientations (6 for ED/MD, 15 for EQ)

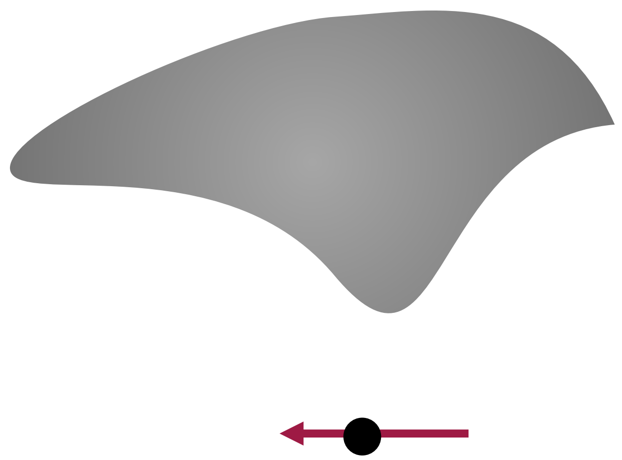
TPSE rate as a function of one-photon Purcell factors

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

Vacuum

Transition rate tailoring

Emitter's position



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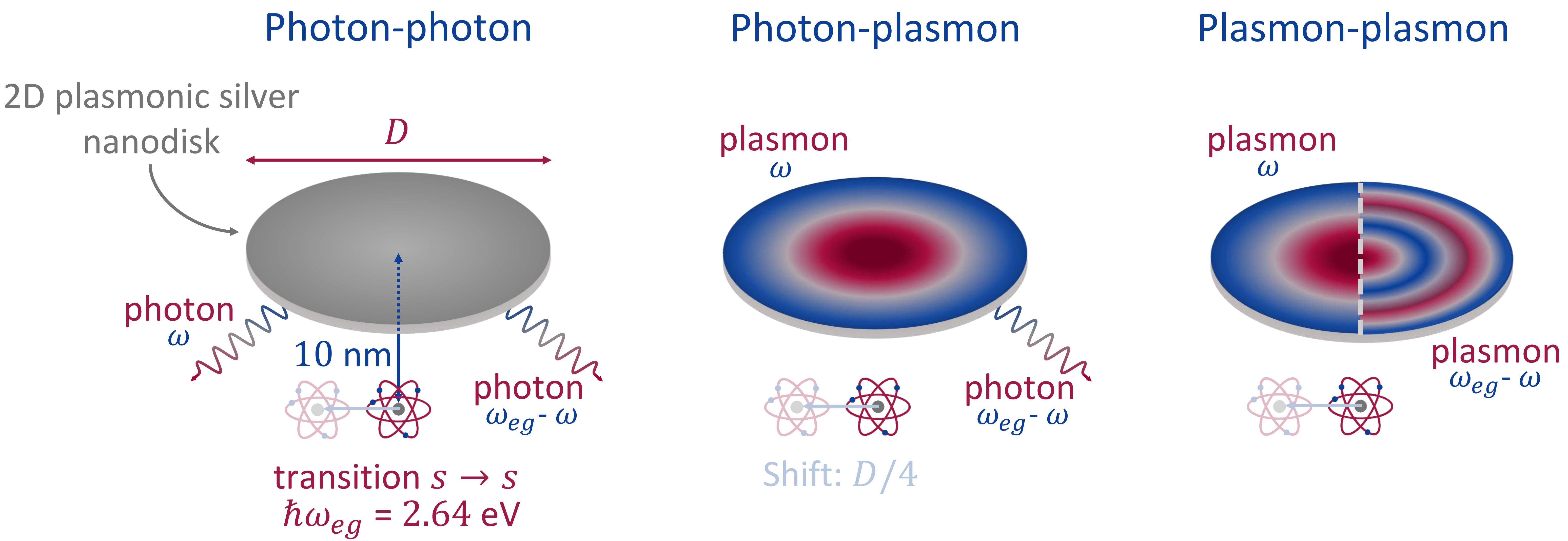
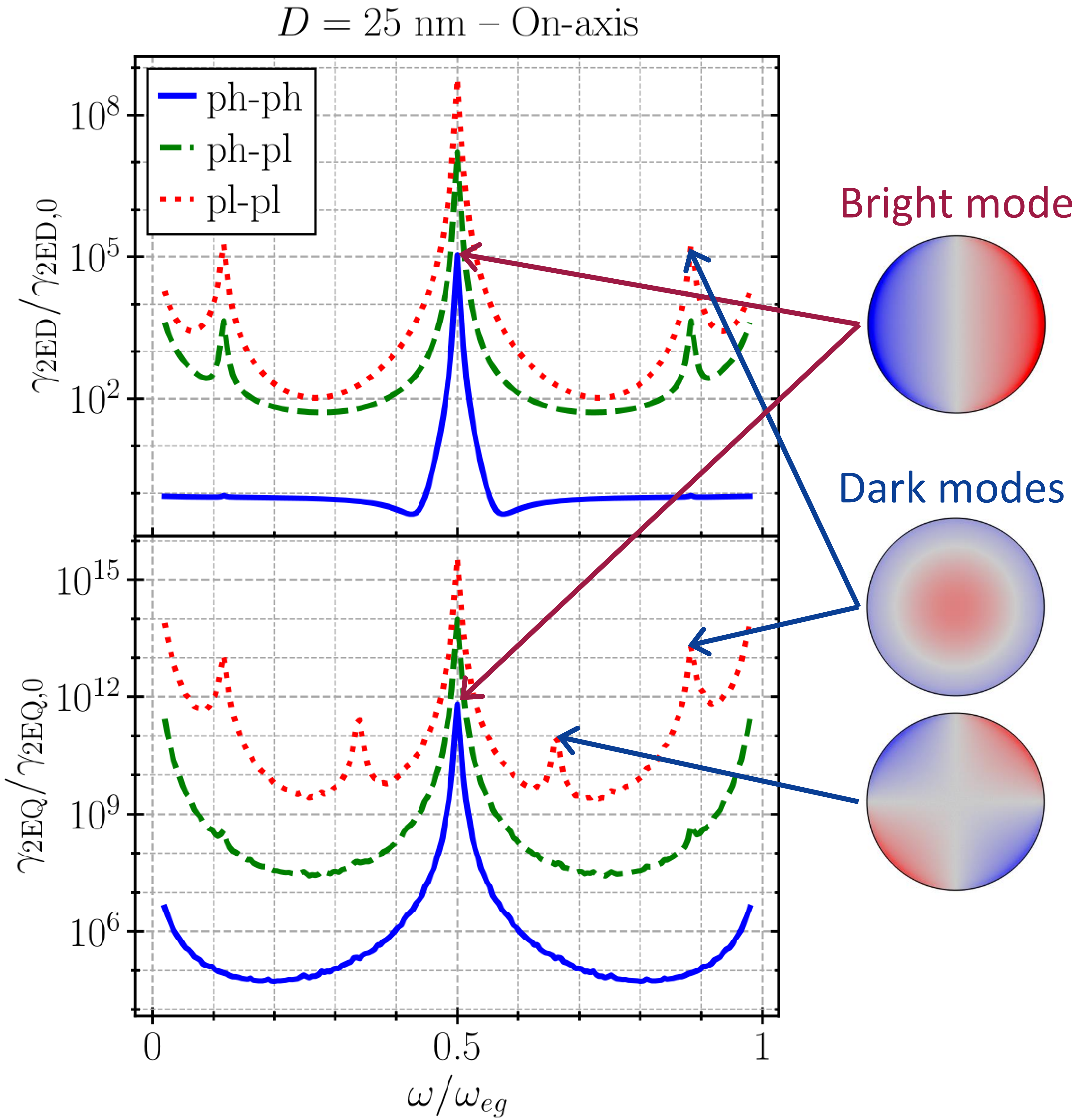
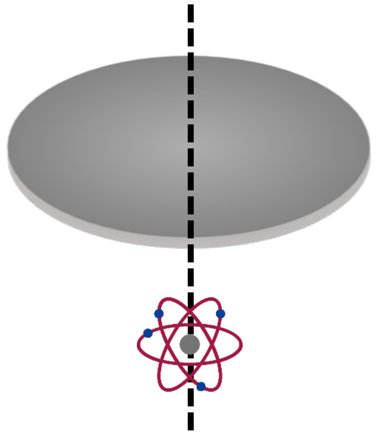
System

- Purcell factors → Decomposition into radiative (photons) and non-radiative (plasmons) parts → 3 TPSE pathways



3. Application





- **Results – 2ED**
  - ✓ Agreement with analytical results [2]
  - ✓ Photon-pair emission rate enhanced by 5 orders of magnitude
  - ✓ Plasmon-pair emission rate enhanced by 8 orders of magnitude
- **Results – 2EQ**
  - ✓ New results
  - ✓ Photon-pair emission rate enhanced by 11 orders of magnitude
  - ✓ Plasmon-pair emission rate enhanced by 15 orders of magnitude

[2] Muniz et al. *Physical Review Letters* 125(3), 033601 (2020)

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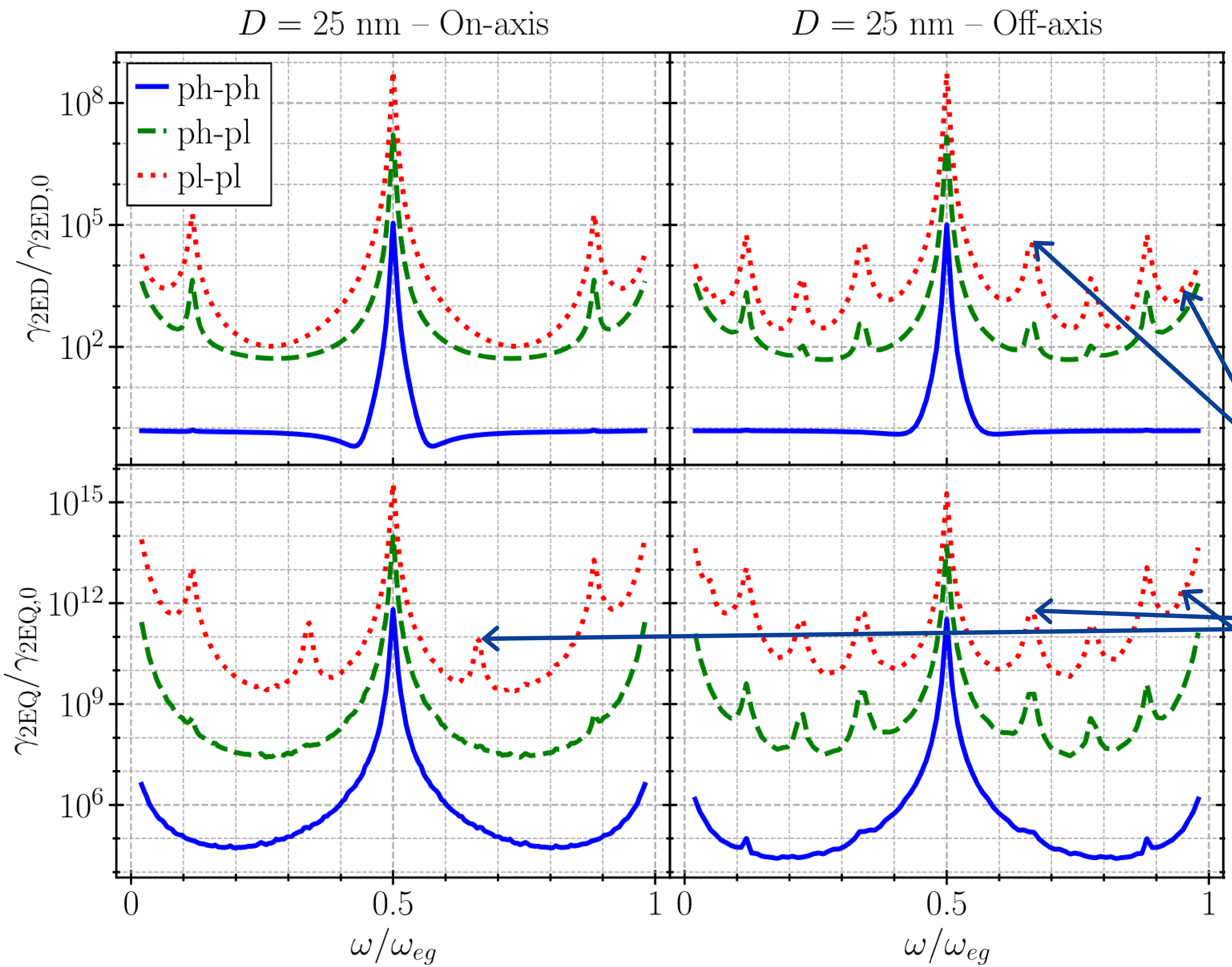
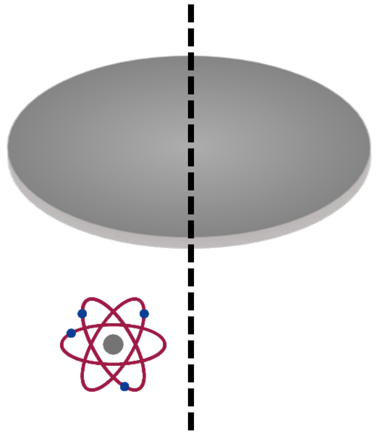
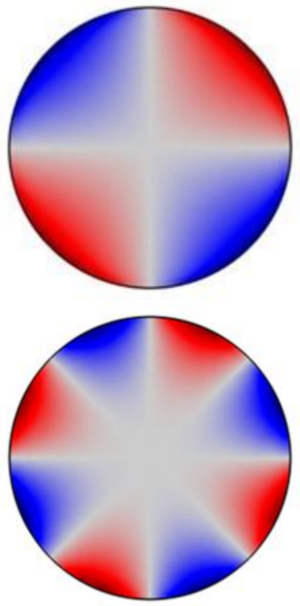




- **Emitter off-axis**
  - → New results: the framework can be applied with emitter at any position
  - → New peaks

Dark modes



Conclusion

- **Our 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
    - ✓ Allows the study of complex geometries
    - ✓ Allows the separate calculation of the radiative and non-radiative channels
  - → Efficient and useful tool for design optimization
  - → Applications: spectroscopy, quantum applications
  - → Paper: Smeets et al. General framework for two-photon spontaneous emission near plasmonic nanostructures. *Phys. Rev. A,* Submitted (2023)