Tailoring two-photon spontaneous emission: framework and nanoantenna design for interference and directionality

Steve Smeets, Bjorn Maes, Gilles Rosolen

Micro- and Nanophotonic Materials Group, University of Mons, Belgium

Steve.Smeets@umons.ac.be

UMONS

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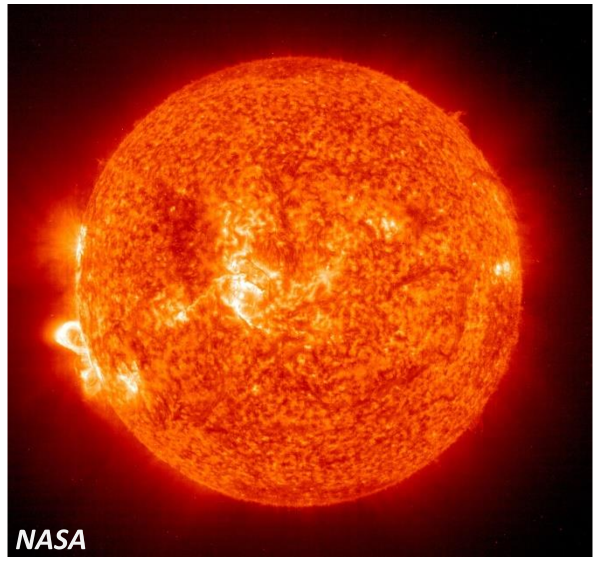
fnrs FREEDOM TO RESEARCH

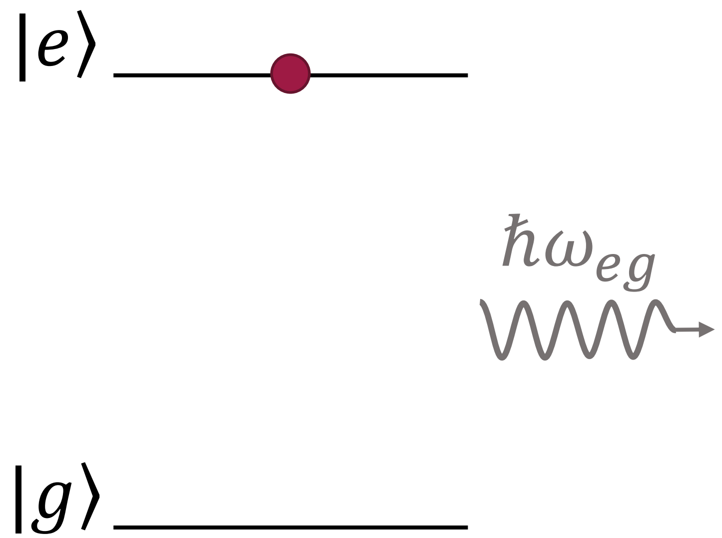
Spontaneous emission

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

  → Responsible for most of the light we see around us



One-photon spontaneous emission

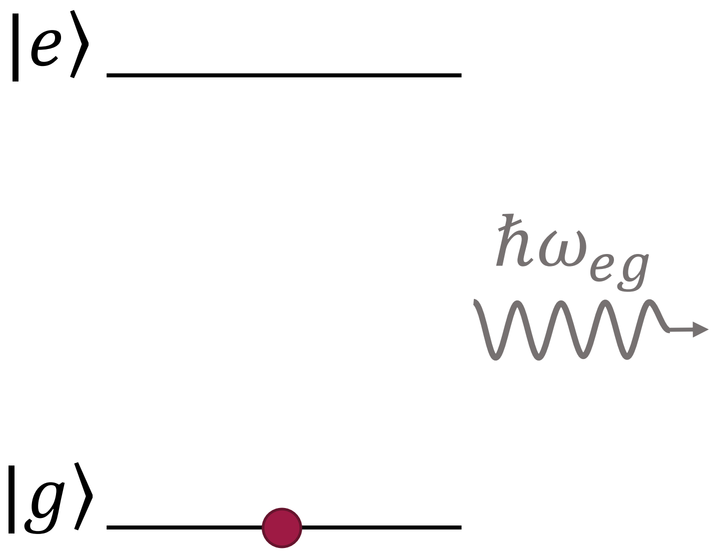


Spontaneous emission

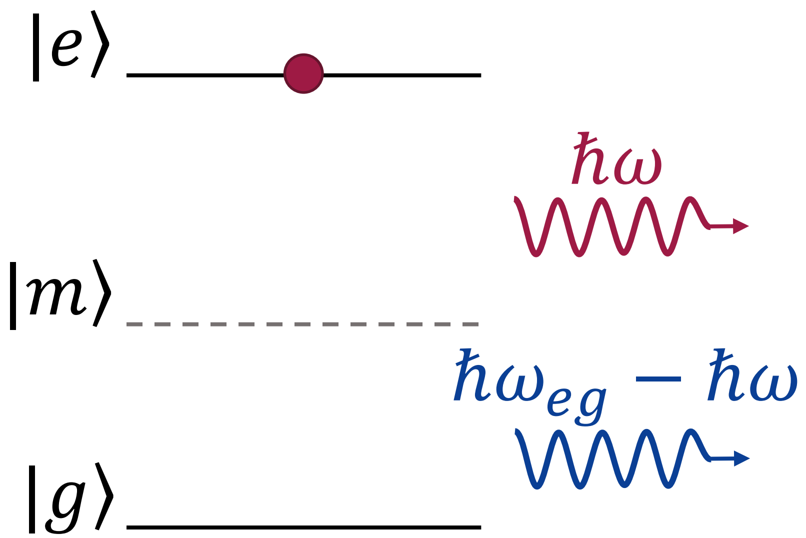

NASA, ESA and the Hubble Heritage (STScI/AURA)-ESA/Hubble Collaboration

- **Two-Photon Spontaneous Emission (TPSE):** second-order process
  - → 8 to 10 orders of magnitude slower than the emission of a single photon [1,2]
  - → Responsible of the 2s state lifetime
    - → Continuous spectrum coming from planetary nebulae
  - → Promising alternative to SPDC for entangled photon sources [3]
    - → 3 orders of magnitude more efficient for equal pump levels, more flexible

One-photon spontaneous emission



Two-photon spontaneous emission



[1] Rivera et al. *Science* 353, 263-269 (2016)
[2] Rivera et al. *PNAS* 114(52), 13607-12 (2017)
[3] A. Hayat et al. *PRB* 76, 035339 (2007)

Photonic environment

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

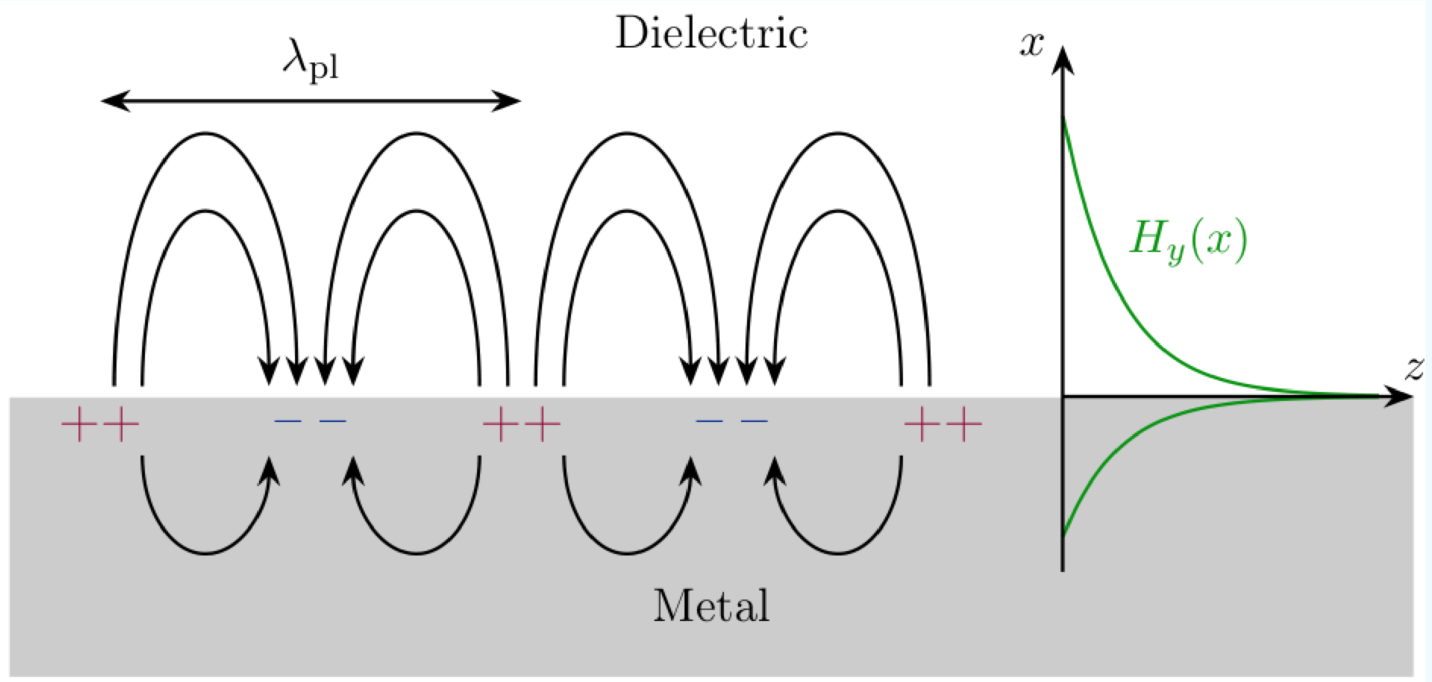
$$P = \frac{\Gamma^{(1)}}{\Gamma_0^{(1)}}$$

- **2D plasmonic nanostructures:** ideal to harness two-quanta emission processes [4]

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



[4] Muniz et al. *PRL* 125(3), 033601 (2020)

Photonic environment

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- **2D plasmonic nanostructures:** ideal to harness two-quanta emission processes [4]

→ Light confinement at the nanoscale

✓ Light emission enhancement via the Purcell effect by several orders of magnitude [1]

✓ Breakdown of the electric dipole approximation [1] → Forbidden transitions accessible [1], TPSE can dominate [2]

✗ Study of advanced nanostructures hampered by a lack of efficient numerical and theoretical methods

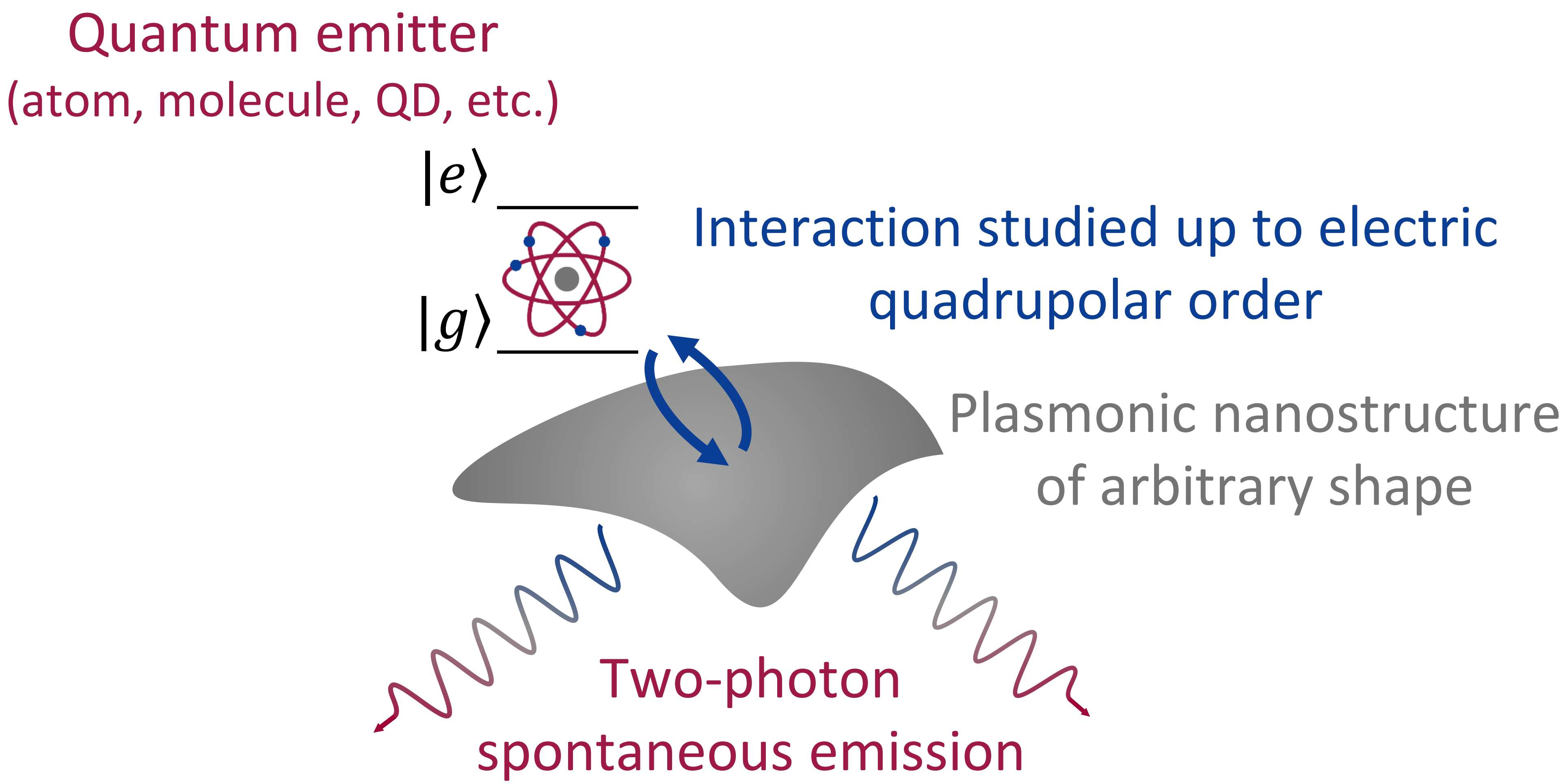
Need for an efficient and general framework which goes beyond the electric dipole approximation by considering higher-order multipolar contributions to second-order processes

→

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

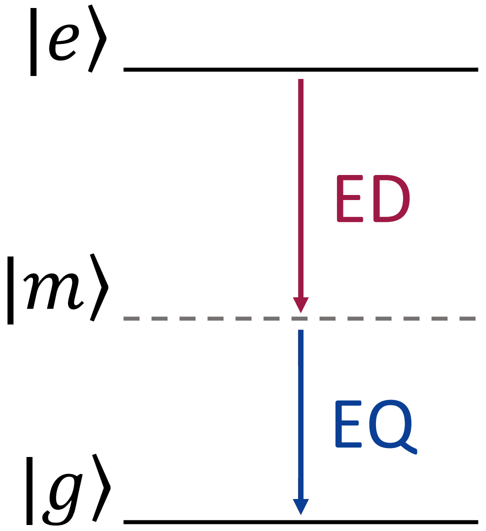
[1] Rivera et al. *Science* 353, 263-269 (2016)
[2] Rivera et al. *PNAS* 114(52), 13607-12 (2017)
[4] Muniz et al. *PRL* 125(3), 033601 (2020)

Fermi's golden rule approach



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

$$\Gamma^{(2)}_{\mathrm{tot}}(\mathbf{R}) = \Gamma^{(2)}_{\mathrm{2ED}} + \Gamma^{(2)}_{\mathrm{2MD}} + \Gamma^{(2)}_{\mathrm{2EQ}} \left(+\Gamma^{(2)}_{\mathrm{mixed}}\right) + \Gamma^{(2)}_{\mathrm{int}}$$



[6] Smeets et al. *PRA 107, 063516* (2023)

TPSE rate as a function of Purcell factors

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

Vacuum

Two-photon Purcell effect

Emitter's position

Emitter contribution

- 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

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)

[6] Smeets et al. *PRA 107, 063516* (2023)

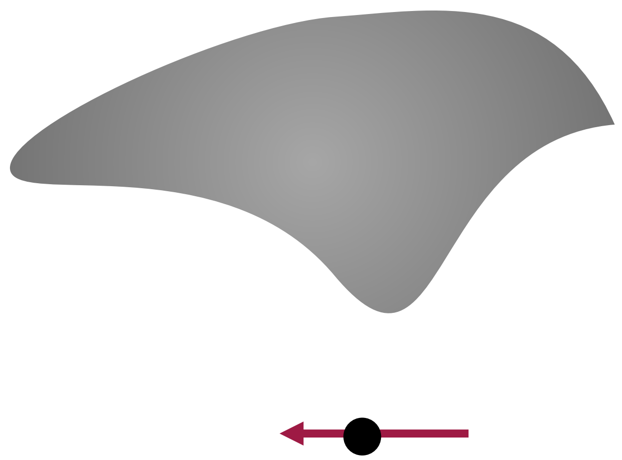
TPSE rate as a function of Purcell factors

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

Vacuum

Emitter's position

Two-photon Purcell effect



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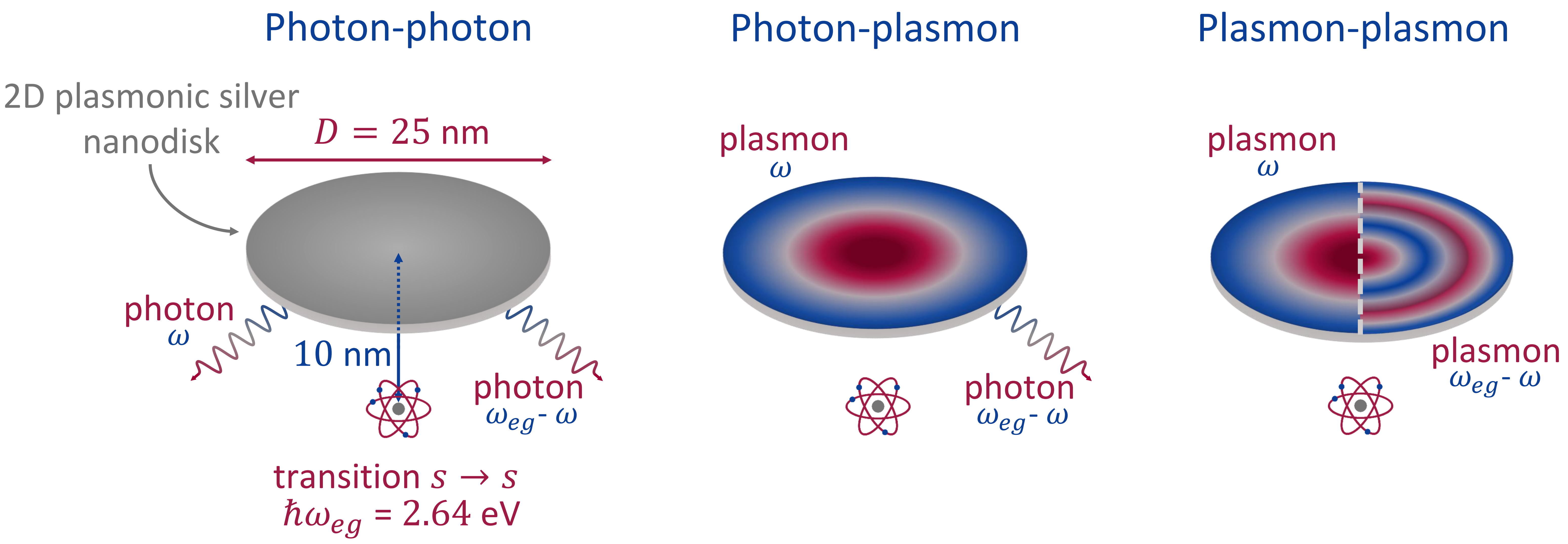
$$\frac{W_\varphi}{W_0} = P_\varphi = \frac{\Gamma^{(1)}_\varphi}{\Gamma^{(1)}_0}$$

  - W_φ: Power emitted by a classical radiating point source
  - To calculate for different source orientations (6 for ED/MD, 15 for EQ)
  - For interference: calculation of interference between classical multipolar sources

[6] Smeets et al. *PRA 107, 063516* (2023)

1) Silver nanodisk

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



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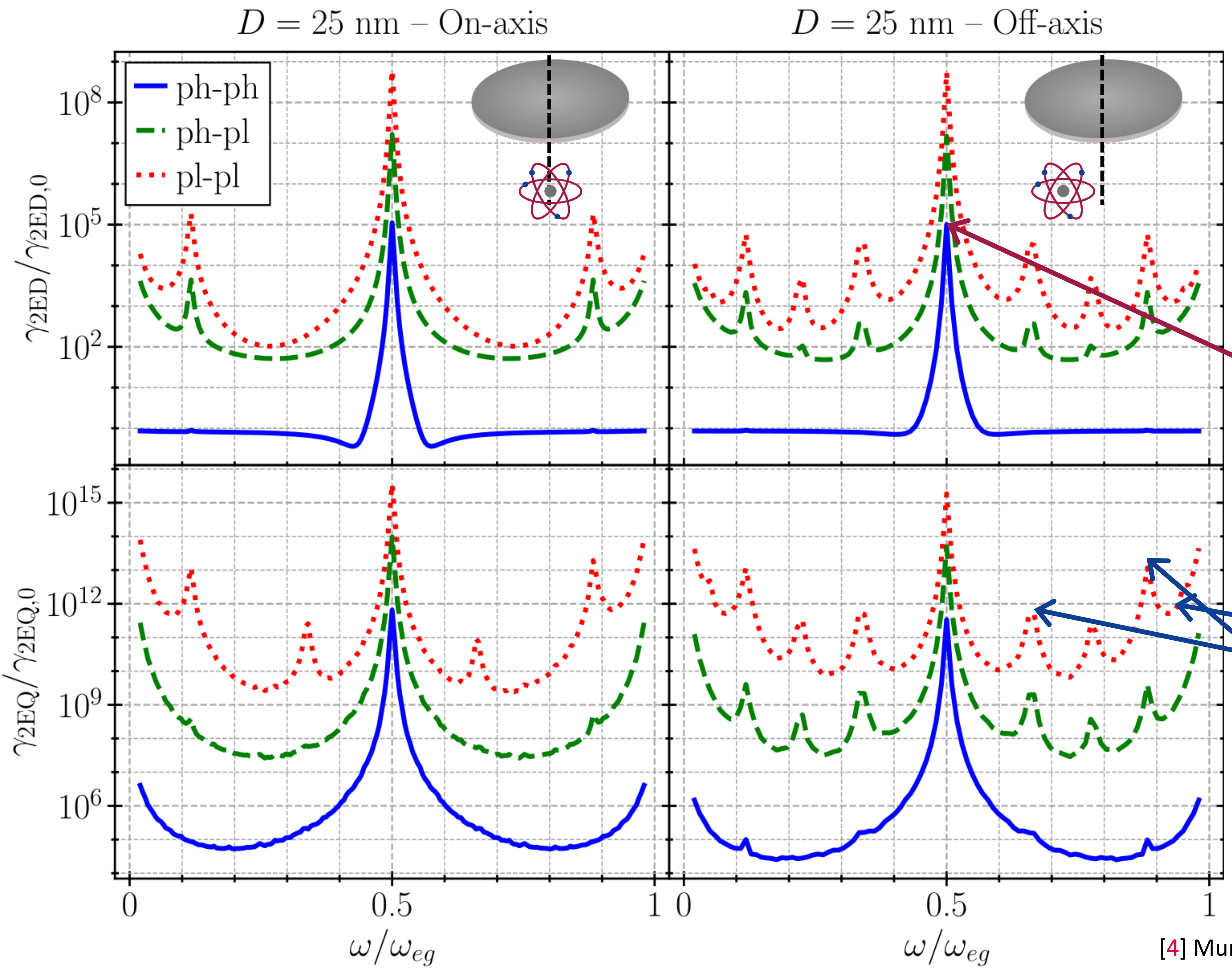
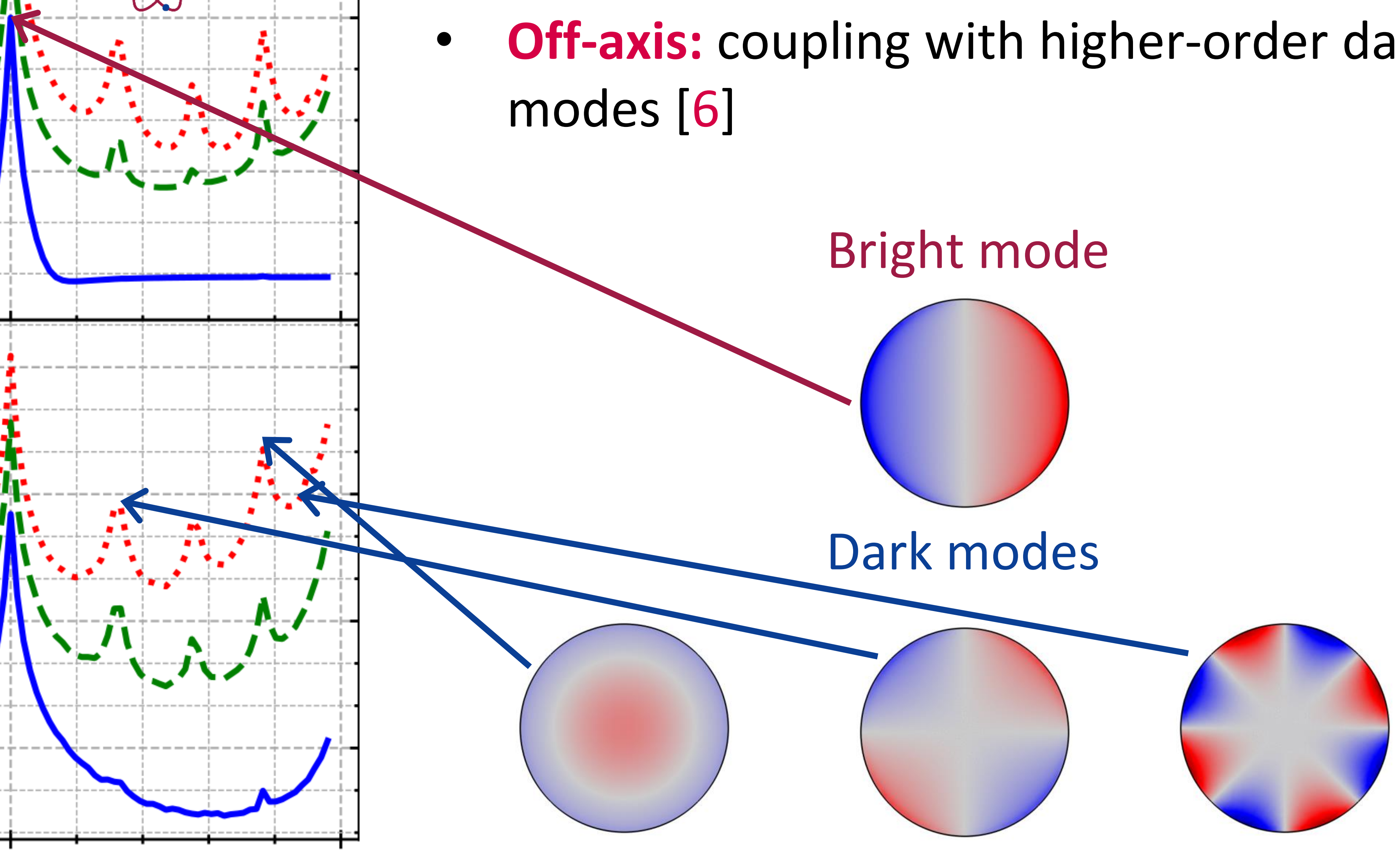
1) Silver nanodisk

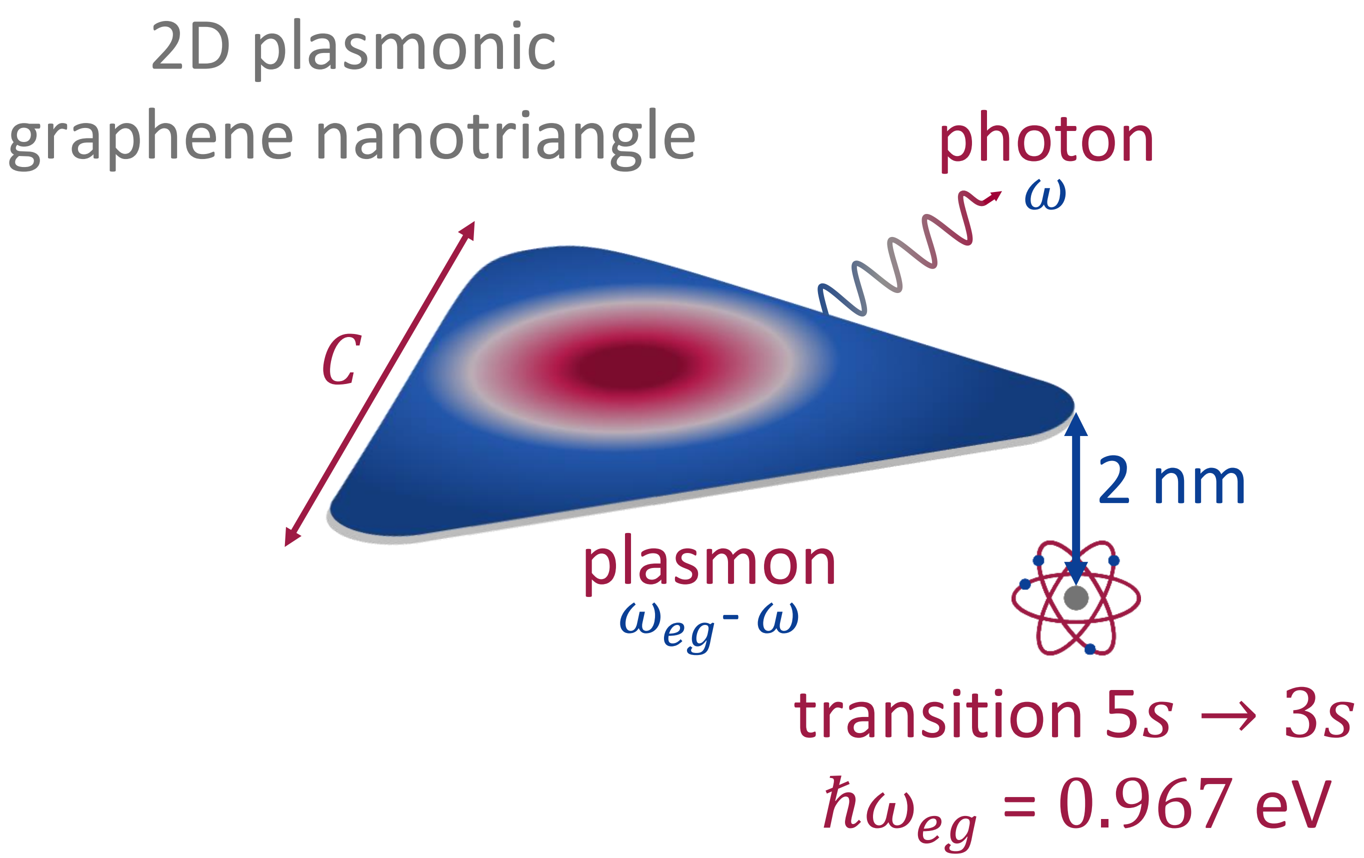


- **2ED:** agreement with analytical results [4]
- **2EQ:** new results [6]
- **Off-axis:** coupling with higher-order dark modes [6]

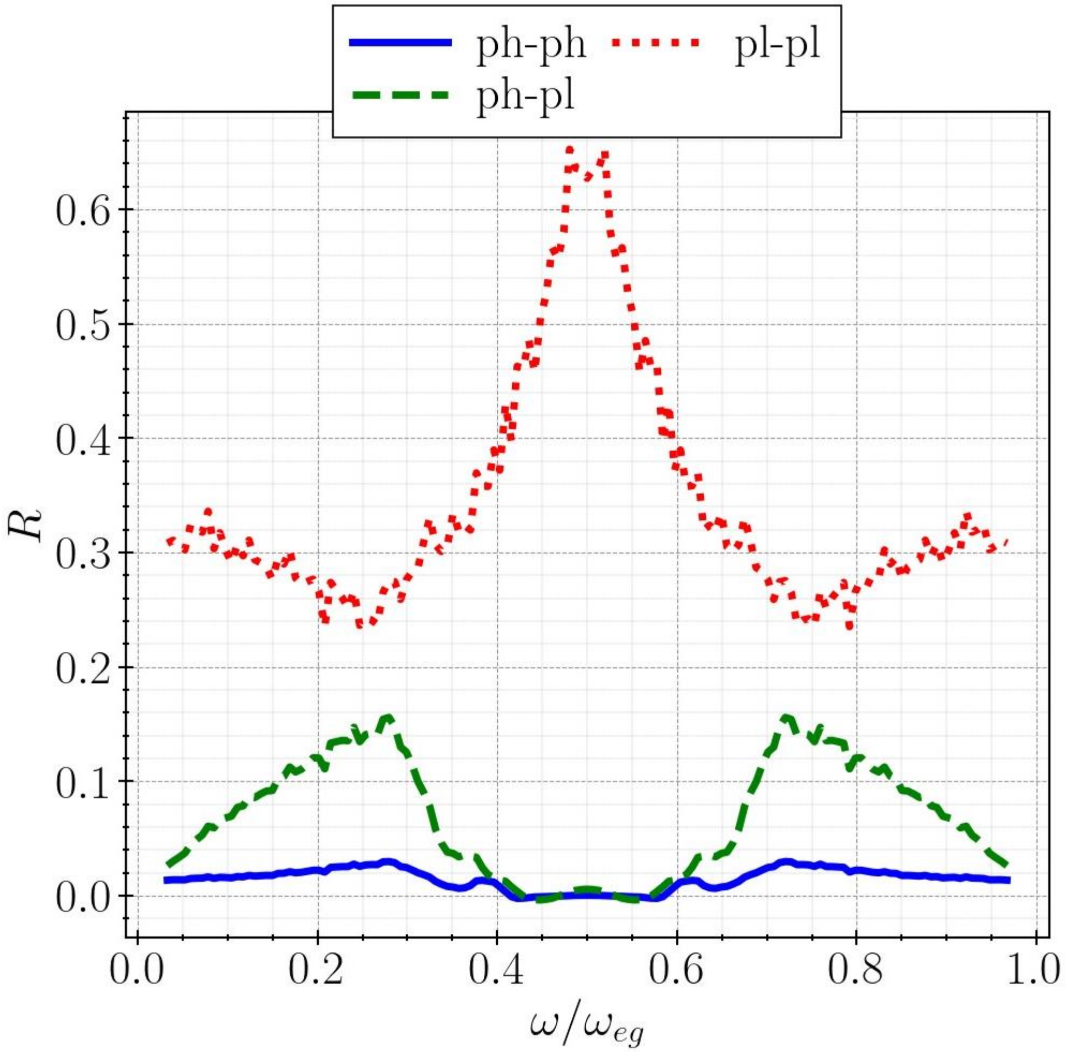


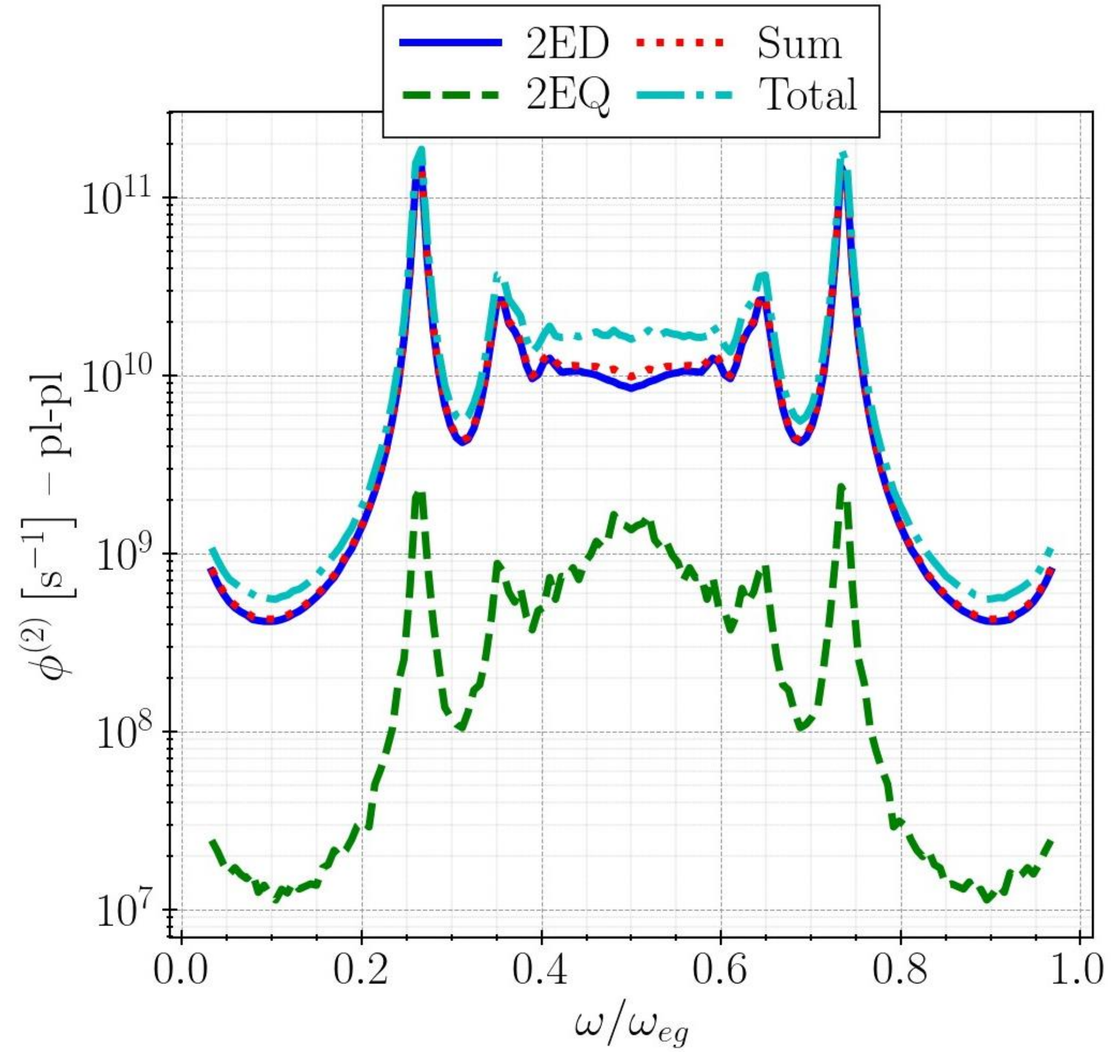
[4] Muniz et al. *PRL* 125(3), 033601 (2020) [6] Smeets et al. *PRA 107, 063516* (2023)

2) Interference near graphene nanotriangle



$E_F = 0.31$ eV, $C = 23$ nm





- Metric $\in [-1,1]$

$$R(\omega;\mathbf{R}) := \frac{\gamma^{(2)}_{\mathrm{2ED}\cap\mathrm{2EQ}}(\omega;\mathbf{R})}{\gamma^{(2)}_{\mathrm{2ED}}(\omega;\mathbf{R}) + \gamma^{(2)}_{\mathrm{2EQ}}(\omega;\mathbf{R})}$$

Important interference effect

$$\phi^{(2)}_{\mathrm{2EQ}}(\omega_{eg}/2) \approx 0.17\,\phi^{(2)}_{\mathrm{2ED}}(\omega_{eg}/2)$$

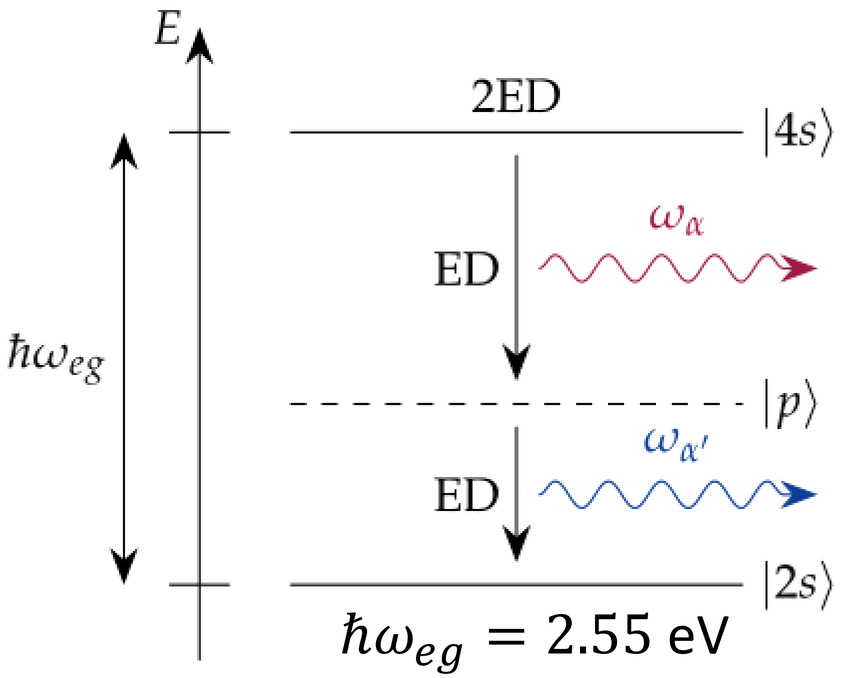
$$R(\omega_{eg}/2) = 63\,\%$$

[7] Smeets et al. *submitted* (2024)

3) Nanoantenna for directional emission

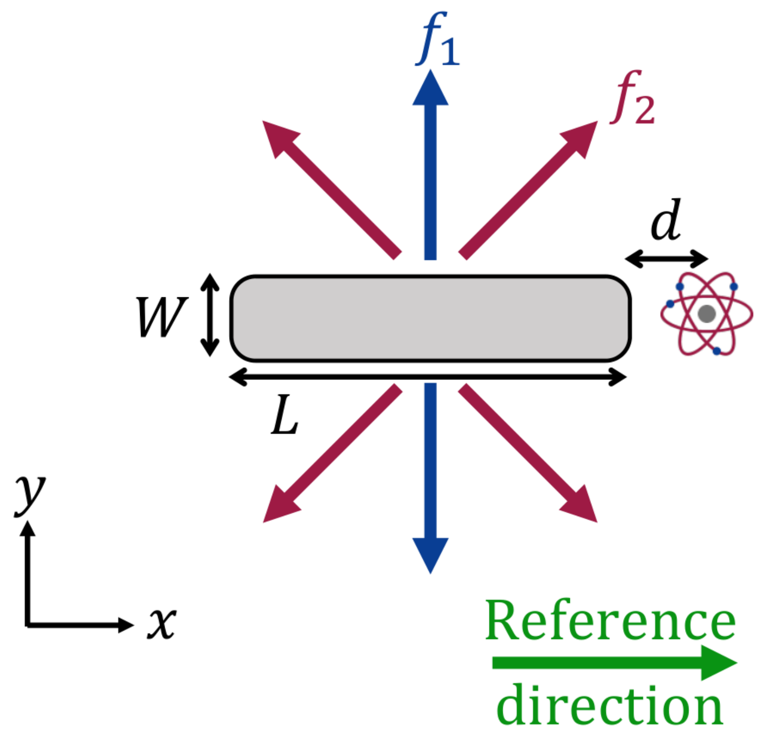
- **Goal:** emit the two photons from the TPSE in different directions at different frequencies

  → Two designs

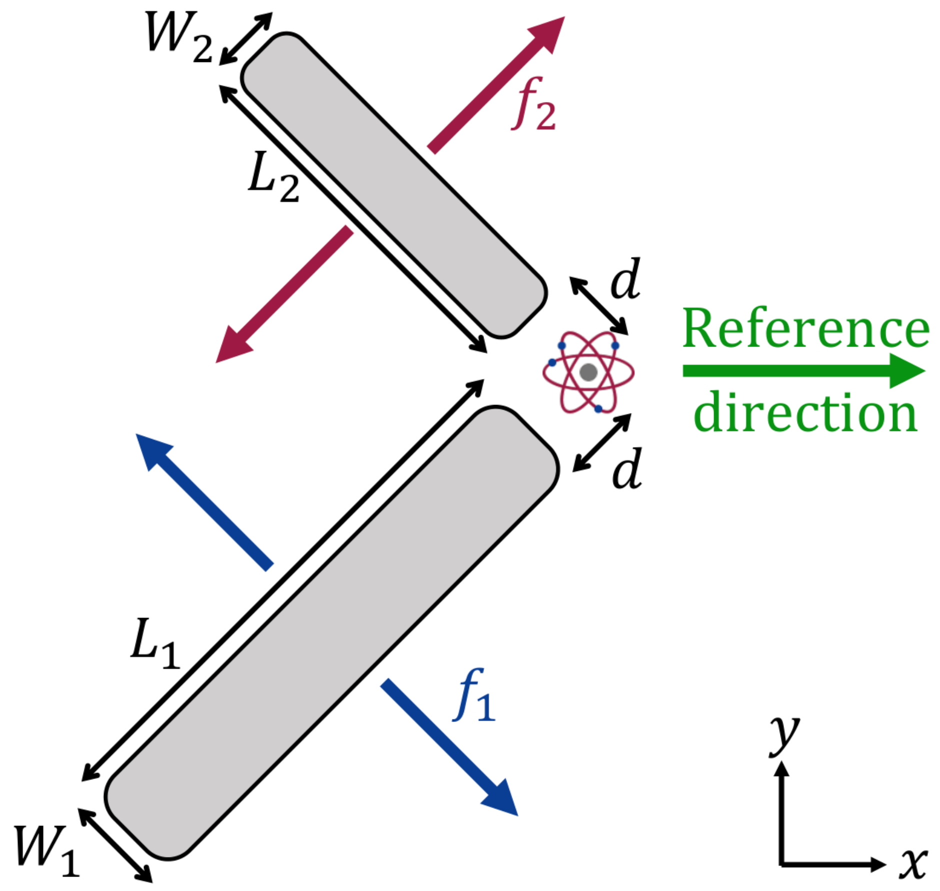


$\hbar\omega_{eg} = 2.55$ eV
($\lambda = 486$ nm)

1) Exploitation of a dipolar and a quadrupolar mode on a single silver nanorod

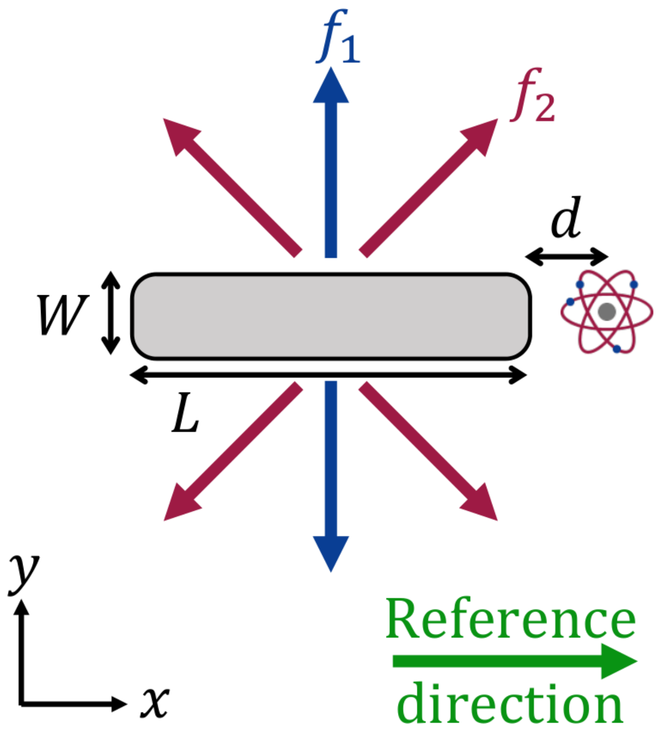


2) Exploitation of a dipolar mode on two perpendicular silver nanorods of different sizes

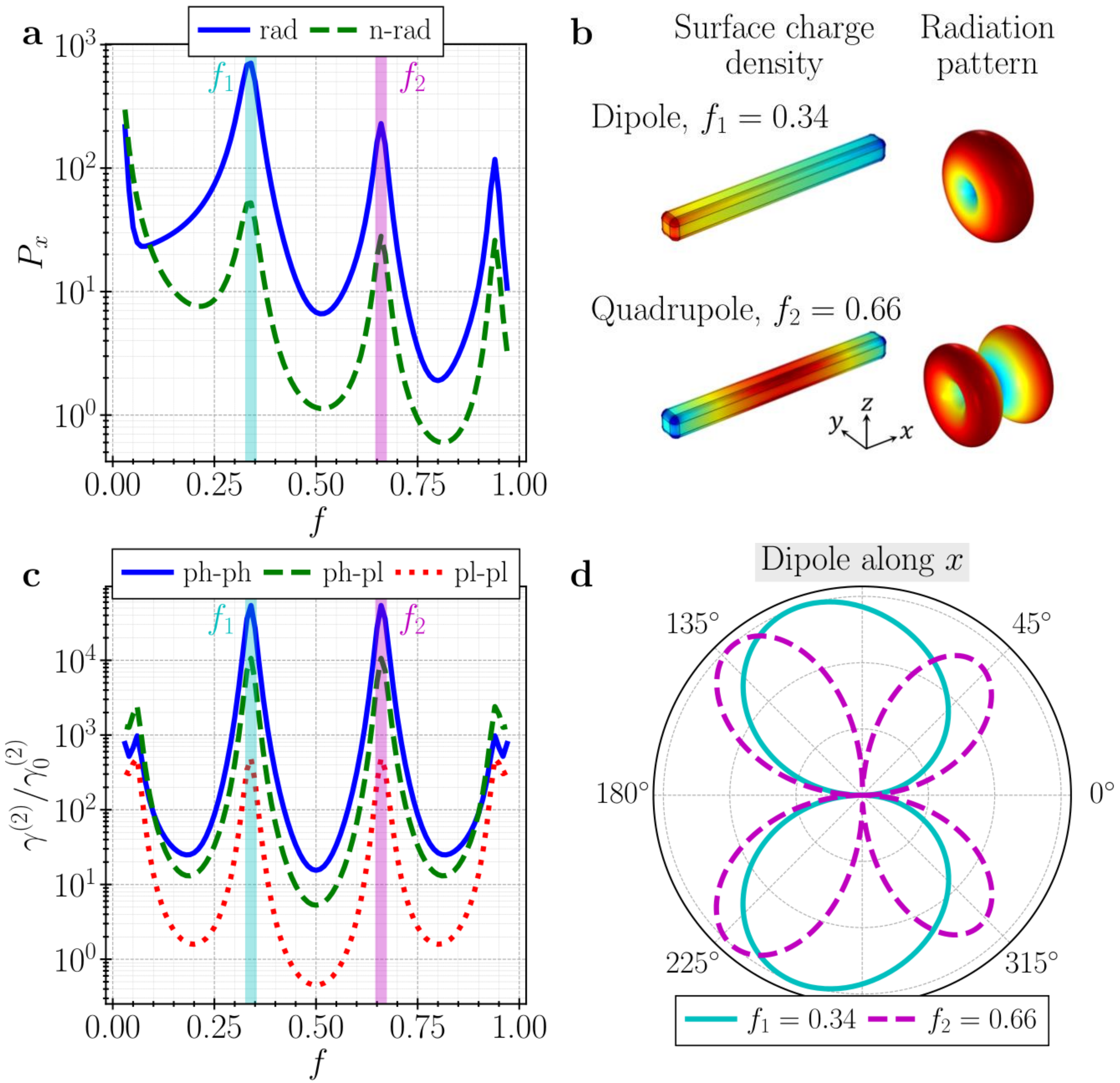


[8] Smeets et al. *submitted* (2024)

- **Parameters** (square section):
  - → $d = 15$ nm
  - → $L = 412$ nm, $W = 39$ nm
  - → $f_1 = 0.34$ ($\lambda = 1.43$ µm)
  - → $f_2 = 0.66$ ($\lambda = 736$ nm)



3) Nanoantenna for directional emission



- P_x dominates
- The two modes contribute to the same TPSE peak (symmetric spectrum)
  - ✓ $\frac{\gamma^{(2)}_{\text{ph-ph}}}{\gamma^{(2)}_0} = 5.4\ 10^4$
  - ✓ High quantum efficiency

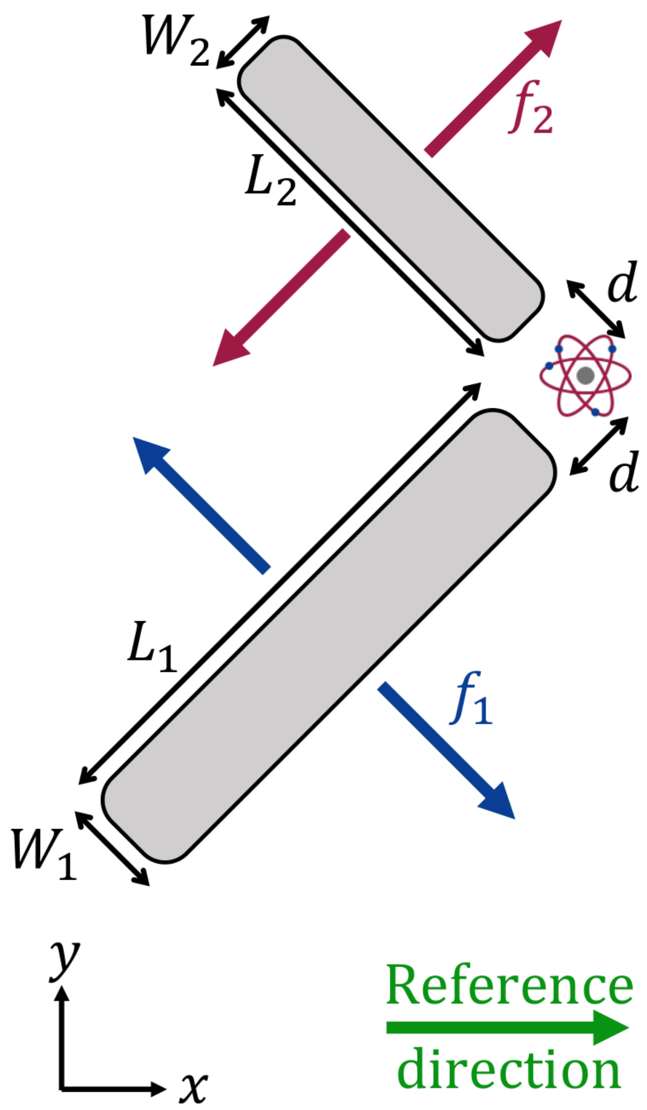
$$\eta^{(2)} := \frac{\gamma^{(2)}_{\text{ph-ph}}}{\gamma^{(2)}} = 83\ \%$$

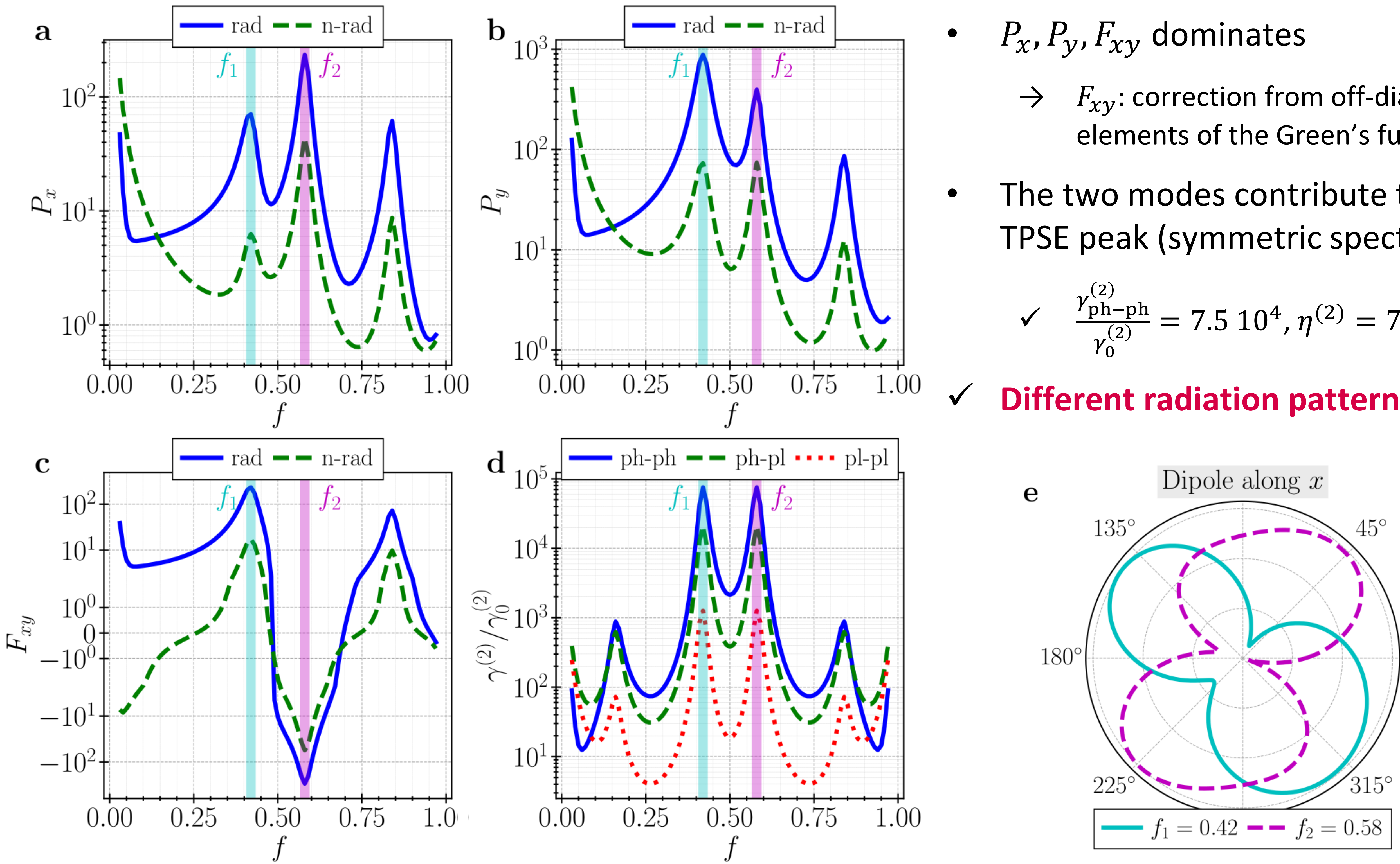
✓ **Different radiation patterns**

[8] Smeets et al. *submitted* (2024)

3) Nanoantenna for directional emission

- **Parameters** (square section):
  - → $d = 15$ nm
  - → $L_1 = 289$ nm, $W_1 = 36$ nm
  - → $L_2 = 146$ nm, $W_2 = 21$ nm
  - → $f_1 = 0.42$ ($\lambda = 1.16$ µm)
  - → $f_2 = 0.58$ ($\lambda = 838$ nm)





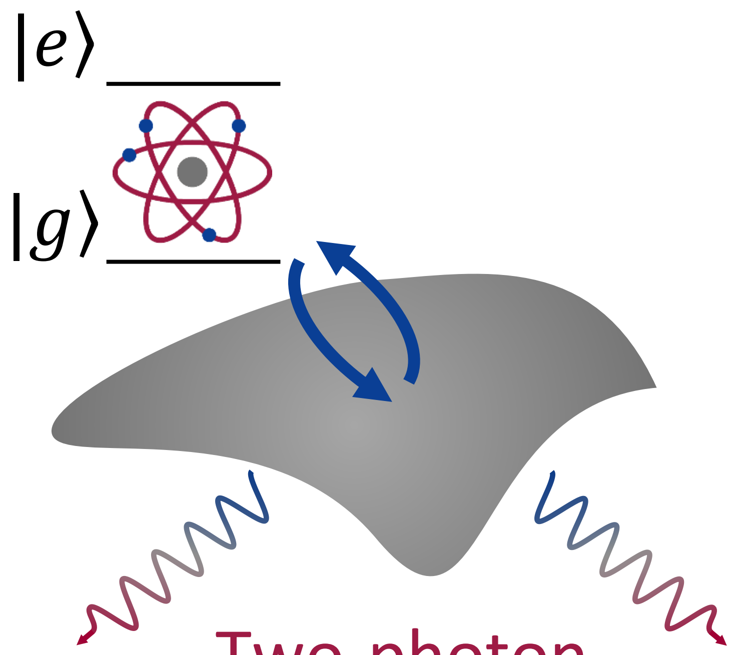
- P_x, P_y, F_{xy} dominates
  - → F_{xy}: correction from off-diagonal elements of the Green's function
- The two modes contribute to the same TPSE peak (symmetric spectrum)
  - ✓ $\frac{\gamma^{(2)}_{\text{ph-ph}}}{\gamma^{(2)}_0} = 7.5\ 10^4$, $\eta^{(2)} = 77$ %

✓ **Different radiation patterns**

[8] Smeets et al. *submitted* (2024)

Conclusion

- **Framework** [6,7]
  - → Efficiently computes TPSE rate of a quantum emitter near an arbitrary shaped nanostructure beyond the electric dipole approximation, including interferences
  - → 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 system optimization (emitter and environment)
  - → Quantum applications
- **Interference** [7] between 2ED and 2EQ transitions increase total TPSE rate by 63 % near a graphene nanotriangle
- **Tailoring directionality** [8]
  - → Two nanoantenna designs to emit TPSE photons in different directions
  - → Directivity can be improved with hybrid metal-dielectric nanostructures



Two-photon spontaneous emission

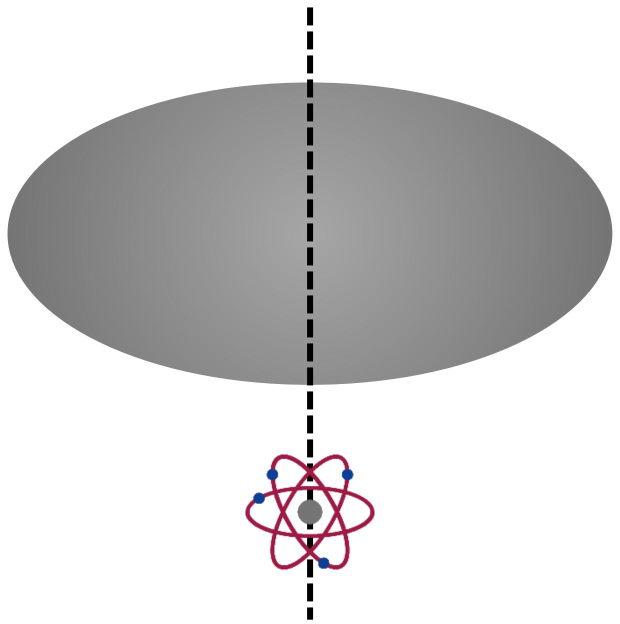
[6] Smeets et al. *PRA 107, 063516* (2023)
[7] Smeets et al. *Submitted* (2024)
[8] Smeets et al. *Submitted* (2024)

Bibliography

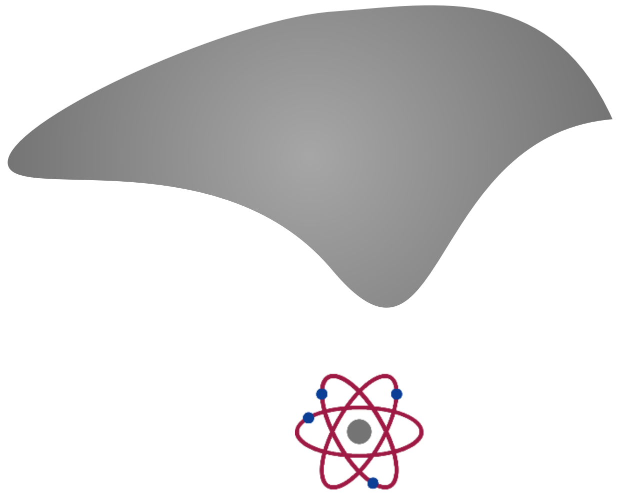
- [1] Rivera et al. Shrinking light to allow forbidden transitions on the atomic scale. *Science* 353, 263-269 (2016)
- [2] Rivera et al. Making two-photon processes dominate one-photon processes using mid-IR phonon polaritons. *Proceedings of the National Academy of Sciences* 114(52), 13607-12 (2017)
- [3] A. Hayat et al. High-rate entanglement source via two-photon emission from semiconductor quantum wells. *Phys. Rev. B* 76, 035339 (2007)
- [4] Muniz et al. Two-photon spontaneous emission in atomically thin plasmonic nanostructures. *Physical Review Letters* 125(3), 033601 (2020)
- [5] Muniz et al. Quantum two-photon emission in a photonic cavity. *Phys. Rev. A* 100, 023818 (2019)
- [6] Smeets et al. General framework for two-photon spontaneous emission near plasmonic nanostructures. *Phys. Rev. A* 107, 063516 (2023)
- [7] Smeets et al. Interference between multipolar two-photon transitions in quantum emitters near plasmonic nanostructures. *Submitted* (2024)
- [8] Smeets et al. Tailoring directivity of two-photon spontaneous emission using plasmonic nanoantennas. *Submitted* (2024)

TPSE rate derivations

- **Former derivation** [4,5]
  - → Only for the 2ED contribution
  - → Can be applied only for symmetric structures with the emitter at specific positions



- **Our derivation** [6,7]
  - → 2ED, 2MD, 2EQ contributions + interferences
  - → Can be applied for arbitrary shaped nanostructures with the emitter at any position



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[6] Smeets et al. *PRA 107, 063516* (2023)
[7] Smeets et al. *Submitted* (2024)

Framework extension

- **Including interferences in the total TPSE rate**

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

Interference between multipolar TPSE channels

- **Interferences must be considered** when the ratio between two multipolar pathways is greater than $2.5\ 10^{-3}$ since they can lead to a modification greater than $10\ \%$ of the total transition rate

- When magnetic transitions are negligible and mixed transitions are forbidden: **3 contributions**

$$\Gamma^{(2)}_{\mathrm{tot}}(\boldsymbol{R}) \approx \Gamma^{(2)}_{\mathrm{2ED}}(\boldsymbol{R}) + \Gamma^{(2)}_{\mathrm{2EQ}}(\boldsymbol{R}) + \Gamma^{(2)}_{\mathrm{2ED}\cap\mathrm{2EQ}}(\boldsymbol{R})$$

Ideally of the same order of magnitude for greater interference effects

Interference between the 2ED and the 2EQ channels

- **Need to calculate TPSE rates in vacuum** → Analytical calculation

Interference as a function of Purcell factors

$$\frac{\gamma^{(2)}_{\text{2ED}\cap\text{2EQ}}(\omega;\mathbf{R})}{\sqrt{\gamma^{(2)}_{\text{2ED},0}(\omega)\gamma^{(2)}_{\text{2EQ},0}(\omega)}} = 2\sum_{i,j=1}^{3}\sum_{\mu,\nu=1}^{5} \hat{\mathcal{D}}^{eg}_{ij}(\omega,\omega_{eg}-\omega)\left(\hat{\mathcal{Q}}^{eg}_{\mu\nu}(\omega,\omega_{eg}-\omega)\right)^{*} F^{\text{ED}\cap\text{EQ}}_{i\mu}(\omega;\mathbf{R})\,F^{\text{ED}\cap\text{EQ}}_{j\nu}(\omega_{eg}-\omega;\mathbf{R})$$

Vacuum — Emitter contribution — Environment contribution

- Tensors F expressed as a function of **one-photon Purcell factors**, present for the two emitted quanta with complementary energies

$$\left(\gamma^{(2)}_{\text{2ED},0}\,\gamma^{(2)}_{\text{2EQ},0}\right)^{1/4} F^{\text{ED}\cap\text{EQ}}_{i\mu} = \frac{1}{\sqrt{2}}\left[\left(\sqrt{\gamma^{(2)}_{\text{2ED},0}}+\sqrt{\gamma^{(2)}_{\text{2EQ},0}}\right)P^{\text{ED+EQ}}_{i\mu} - \sqrt{\gamma^{(2)}_{\text{2ED},0}}\,P^{\text{ED}}_{i} - \sqrt{\gamma^{(2)}_{\text{2EQ},0}}\,P^{\text{EQ}}_{\mu}\right]$$

Purcell relative to the superposition of an ED along $\vec{e}_i$ + an EQ along $\vec{e}_\mu$ (15 combinations)

Purcell relative to an ED along $\vec{e}_i$ (3)

Purcell relative to an EQ along $\vec{e}_\mu$ (5)

→ Can be positive or negative ⇒ **Increase or decrease of the total TPSE rate**

→ **Quantum interference computed via interferences between classical multipolar sources**

2. Interference