Photonic modeling of high-order light-matter interactions

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Abstract

Controlling and understanding the behavior of a quantum emitter close to a nanostructure is under extensive research. However, the study of advanced nanostructures is hampered by a lack of efficient numerical and theoretical methods.

Therefore, the main objective is to implement novel modeling methods for high-order transitions, beyond the standard dipolar approach, which is relevant for the current nanocavities with highly confined light. Then, the developed framework will be applied for innovative structures.

References:

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

Context

Motivation: Two-Photon Spontaneous Emission (TPSE) is around 5 to 8 orders of magnitude slower than the emission of a single photon → **How to make it accessible ?**

Solution: coupling with **surface plasmons** ⇒ Emission rate enhanced by the **Purcell effect**

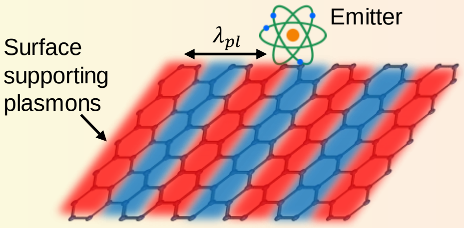

Figure A – Emitter coupled to a surface plasmon supported by a 2D material. The **light confinement** makes the wavelength approaches the emitter size.

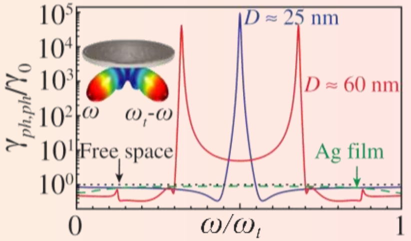

Figure B - Photon-pair production rate $\lambda_{ph,ph}(\omega)$ **enhancement** for an electric dipole transition of an emitter placed 10 nm above a bilayer Ag nanodisk [1].

Problem: the electric dipole approximation is not appropriate for highly confined light [1]
⇒ We need to develop a framework which **studies TPSE beyond the standard electric dipole approximation**

Framework in progress

- Let's consider a system composed of a quantum emitter located by $\boldsymbol{r_0}$ close to a surface of arbitrary shape. The **interaction Hamiltonian** $\hat{V}$ is **studied up to the quadrupolar order**

$$\hat{V} = -\,\hat{\boldsymbol{d}}.\hat{\boldsymbol{E}}(\boldsymbol{r_0}) - \hat{\boldsymbol{m}}.\hat{\boldsymbol{B}}(\boldsymbol{r_0}) - \left[\hat{\boldsymbol{Q}}\nabla\right].\hat{\boldsymbol{E}}(\boldsymbol{r_0})$$

- The approach is based on **Fermi's golden rule**. The n-order transition rate from an initial state $|i\rangle$ to a final state $|f\rangle$ is then given by

intermediate state

$$\Gamma_{i\to f}^{(n)} = \frac{2\pi}{\hbar}\left|M_{fi}^{(n)}\right|^2 \delta\left(E_i - E_f\right) \qquad M_{fi}^{(1)} = \langle f|\hat{V}|i\rangle \qquad M_{fi}^{(2)} = \sum_{l} \frac{\langle f|\hat{V}|l\rangle\,\langle l|\hat{V}|i\rangle}{E_i - E_f}$$

- **First step:** express $\Gamma_{i\to f}^{(n)}$ depending on **Purcell factors.** For example, for a second-order electric dipole transition the spectral TPSE rate is

$$\gamma_{\mathrm{ph,ph}}(\omega) = \gamma_0(\omega) \sum_{a,b}^{3} t_{ab}(\omega)\, P_{a,r}(\omega) P_{b,r}(\omega_t - \omega)$$

  where $t_{ab}(\omega)$ is a tensor that depends on the electronic structure of the emitter and $P_{a,r}(\omega)$ is the radiative Purcell factor for a transition electric dipole moment oriented along $\boldsymbol{e}_a$ [1]

- **Second step:** compute classically the Purcell factors with the COMSOL Multiphysics® software (finite element method)

Results and Conclusions

- **Characteristics of the framework**
  - Concerns the TPSE of a quantum emitter coupled with plasmonic nanostructures
  - Based on the Fermi's golden rule
  - Based on **numerical calculations** of the Purcell factors
    ⇒ Study and optimize nanostructures
  - Goes **beyond the standard electric dipole approximation** by considering magnetic dipole and electric quadrupole transitions
- Figure B has been reproduced with the framework ⇒ **COMSOL can be used**
- **Planning**
  - Finish the framework: express $\Gamma_{i\to f}^{(2)}$ depending on Purcell factors for a magnetic dipole transition and an electric quadrupole transition → estimation
  - Study interference effects between multipolar transition channels of TPSE

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