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. This discipline promises for example improved nanoscale imaging, better detectors, and efficient entangled photon sources for quantum computers. 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, with an eye towards new physical effects and applications.

Context

Spontaneous emission (SE): fundamental process in the field of light-matter interaction

→ **Motivation:** most radiative decay channels of SE are too slow to be accessible **(forbidden transitions)**

→ **For example:** **two-photon spontaneous emission (TPSE)** is 5 orders of magnitude slower than the emission of a single photon (Figure A)

Purcell effect: SE of quantum emitters can be modified by their optical environment

→ Modification usually studied under the **electric dipole approximation**

→ **Solution:** consider coupling with **surface plasmons** ➔ forbidden transitions are strongly enhanced, which make them accessible (Figures B and C)

→ **Therefore:** the **electric dipole approximation is no longer appropriate** for highly confined light [2]

➔ We need to **study high-order transitions** by considering **higher-order multipolar channels**

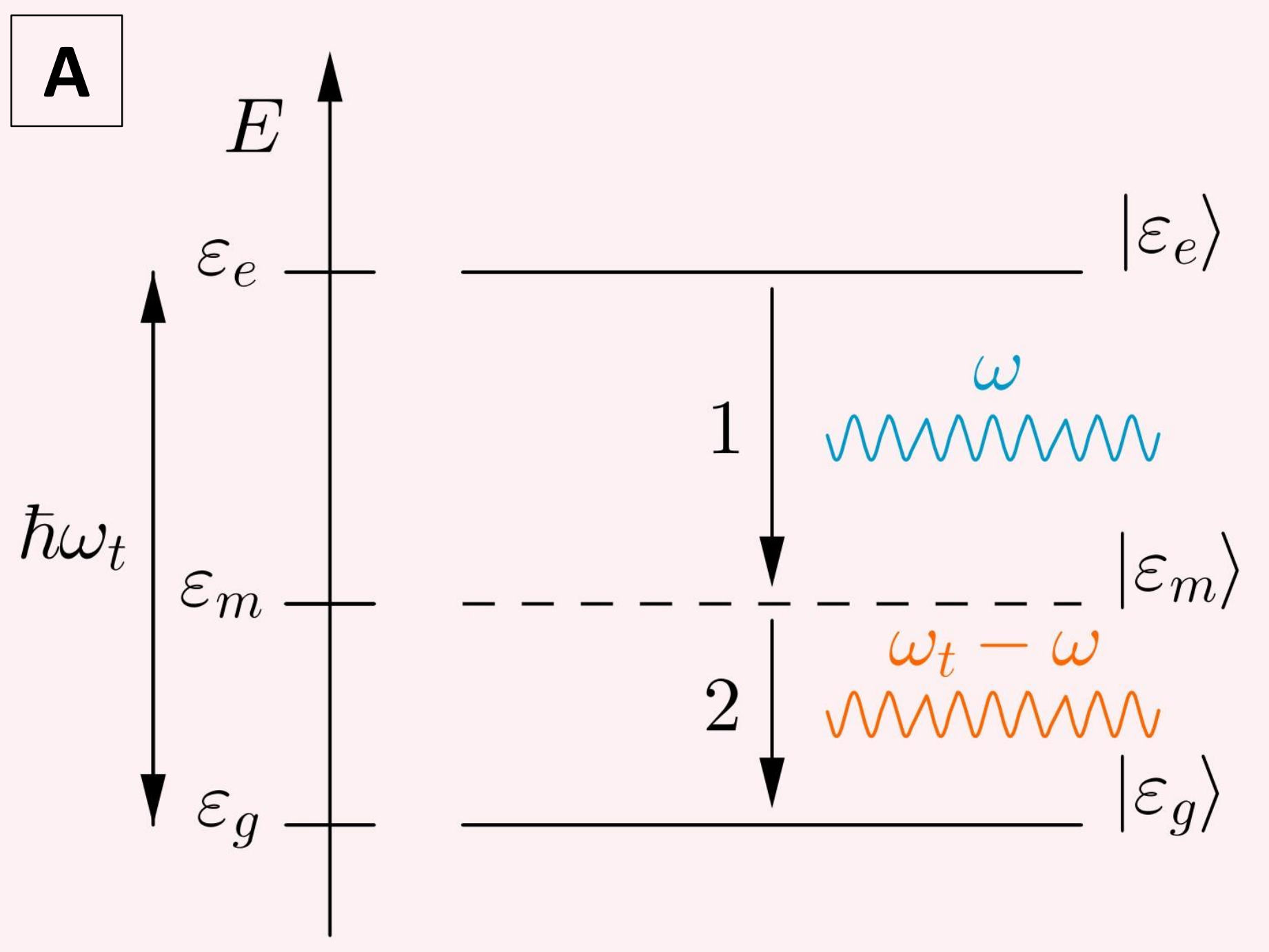

Figure A - Second-order transition: an excited emitter emits a first photon $\hbar\omega$. Then, it emits a second photon $\hbar(\omega_t-\omega)$ from a virtual intermediate state.

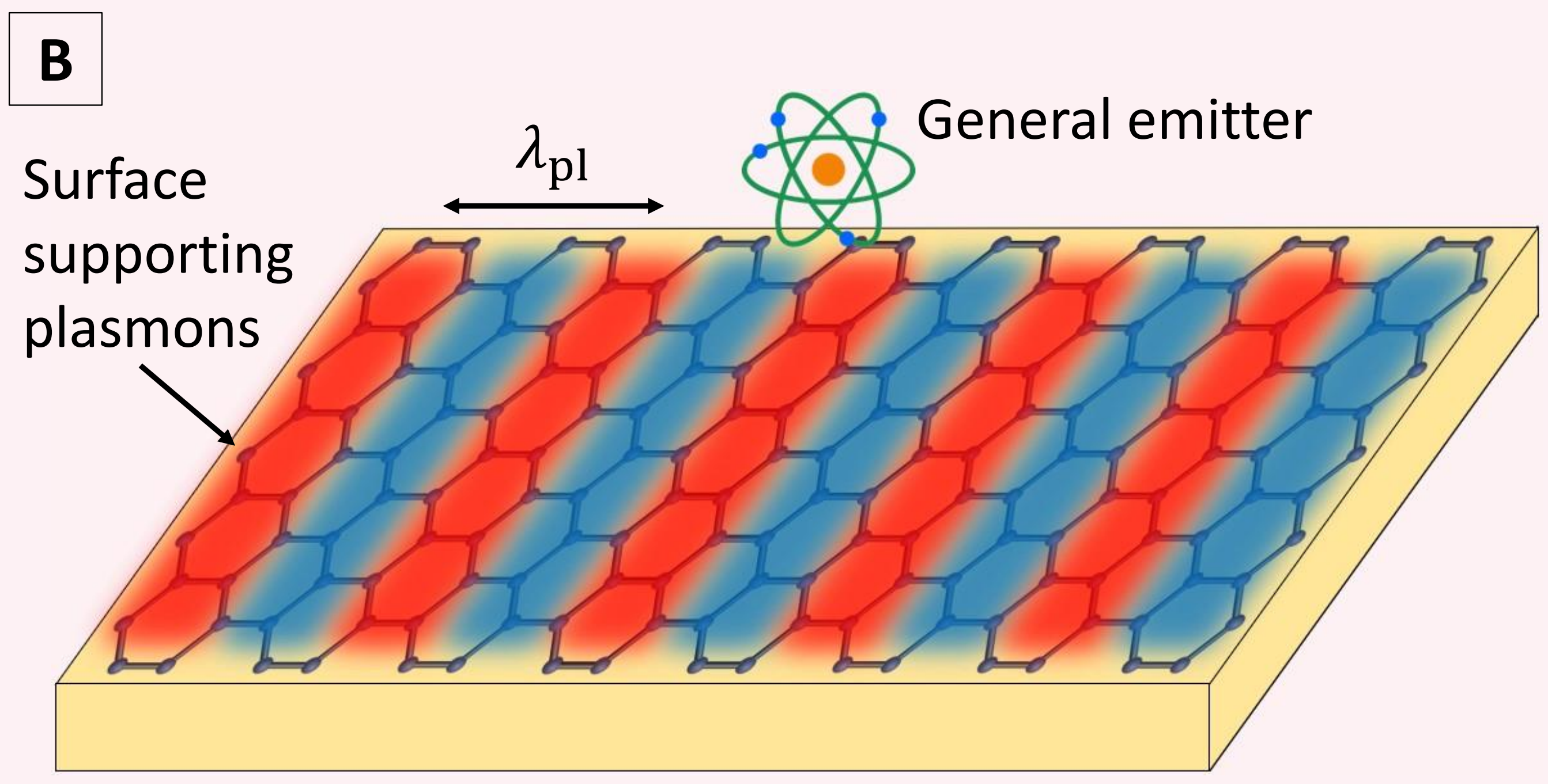

Figure B – Representation of an emitter coupled to a surface plasmon supported by a 2D material. The plasmon's wavelength λ_{pl} is reduced by two orders of magnitude with respect to the photon wavelength **(light confinement)** and approaches the emitter size. Figure adapted from [1].

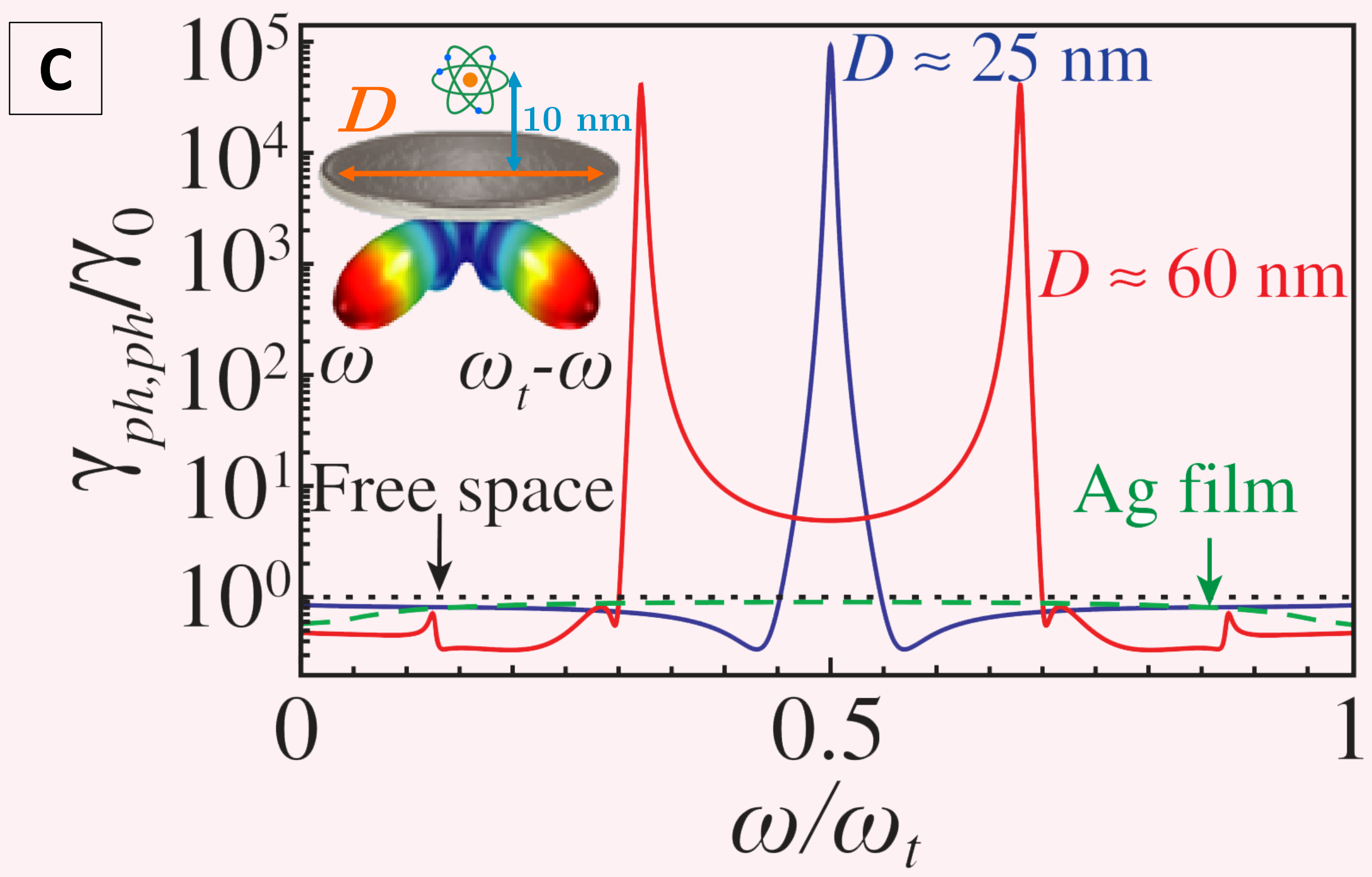

Figure C - Photon-pair production rate $\gamma_{\mathrm{ph,ph}}(\omega)$ for an electric dipole transition of an emitter placed 10 nm above a bilayer Ag nanodisk. They observe a **TPSE rate enhancement with 2D plasmonic nanostructures** [3].

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**

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

- 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

$$\Gamma_{i\to f}^{(n)} = \frac{2\pi}{\hbar}\left|M_{fi}^{(n)}\right|^2 \delta(E_i - E_f)$$

- For example, the first- and second-order $M_{fi}^{(n)}$ are

$$M_{fi}^{(1)} = \langle f|\hat{V}|i\rangle \qquad M_{fi}^{(2)} = \sum_l \frac{\langle f|\hat{V}|l\rangle\langle l|\hat{V}|i\rangle}{E_i - E_f}$$

where the summation runs over all intermediate states of the system

- The **first step** is to express $\Gamma_{i\to f}^{(n)}$ depending on **Purcell's 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 part of the Purcell's factor for a transition electric dipole moment oriented along the unit vector $\boldsymbol{e}_a$ [3]

- The **second step** is to compute, classically, the **Purcell's factors** with the COMSOL Multiphysics® software (finite element method)

Perspectives

The framework in progress will be used to study high-order transitions of quantum emitter coupled with 2D plasmonic nanostructures of arbitrary shapes. These structures are used for tailoring and enhancing transition rate of spontaneous emission processes.

→ Study of enhancement, suppression and interference effects between multipolar channels of high-order transitions

References

[1] Rivera et al. Shrinking light to allow forbidden transitions on the atomic scale. Science, 353(6296), 263-269 (2016).
[2] Rusak et al. Enhancement of and interference among higher order multipole transitions in molecules near a plasmonic nanoantenna. Nat Commun 10, 5775 (2019).
[3] Muniz et al. Two-photon spontaneous emission in atomically thin plasmonic nanostructures. *Physical Review Letters*, *125*(3), 033601 (2020).

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