Hydrogen's enhanced spectrum in the vicinity of nanophotonic structures

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Abstract

Particular nanophotonic structures support strongly confined fields that can enhance an emitter's higher order transitions to even surpass dipolar ones. It is therefore possible to break conventional optical selection rules, which can result in various novel physical and chemical effects. To model such systems, we used a conventional quantum chemistry software (Gaussian) to compute the transition rate of the Hydrogen atom close to a graphene nanoflake modeled using COMSOL. We get a good agreement with an analytical method. Our combined framework is now ready to be used for more complex systems.

Systems of interest: Those that are subjected to weak light-matter coupling[1].

Perturbation theory

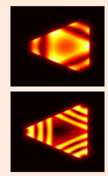
Fermi's golden rule

$\Gamma_{eg} \propto |\langle\psi_g|\hat{O}|\psi_e\rangle|^2$

Structure-dependent operator

?

GENOME: Generalized normal mode expansion of the electromagnetic Green Tensor.



Operator's properties

- All Multipolar terms are included.
- Emitter's extension is considered – no point-dipole approximation.

$$G(r,r') = G_0(r,r') + \sum_m \gamma_m E_m(r) \otimes E_m(r')$$

Eigenpermittivity modes

More details in [2]

Material property

m[th] Electric field eigenpermittivity-mode

$$\Gamma = \Gamma_0 + \frac{2\pi}{k^2}\frac{e^2\hbar^3}{\pi\varepsilon_0 m_e^2 c^2}\sum_m \mathrm{Im}\left[\frac{\varepsilon_i-\varepsilon_b}{(\varepsilon_m-\varepsilon_i)(\varepsilon_m-\varepsilon_b)}\left(\int \psi_e(\vec{r})\boldsymbol{E}_m(\vec{r})\vec{\nabla}\psi_g(\vec{r})d\vec{r}\right)^2\right]$$

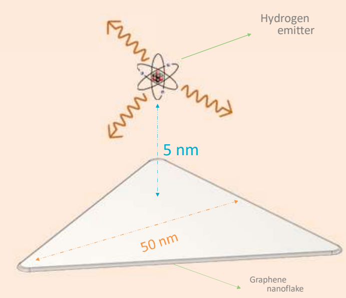
Total rate

Rate in vacuum

Wavefunction of the excited state

Wavefunction of the ground state

System in study



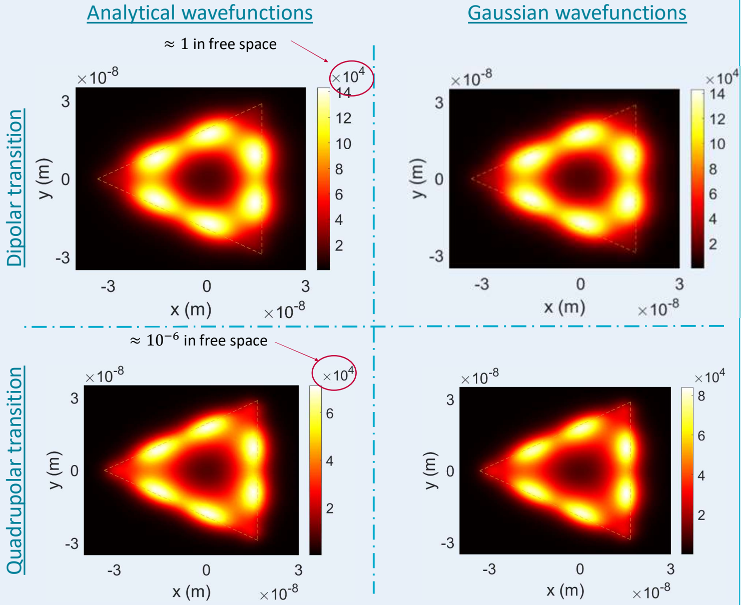
COMSOL Multiphysics

Gaussian (Quantum chemistry software)

Transition rate maps

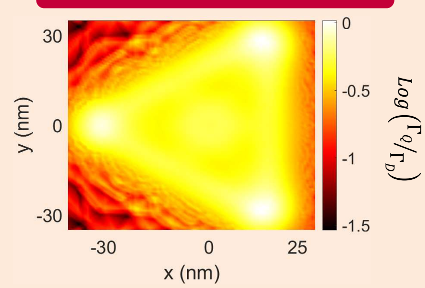
Higher-order transitions normalized with the dipolar transition rate in free-space ($\Gamma_{D0} = 4.5 \times 10^5\ s^{-1}$).

→ **Strong enhancement**



The transition series $6s \rightarrow 4p, d$ of a H emitter with a transition wavelength of $\lambda = 2.63\ \mu m$. The emitter is placed 5 nm above the nanoflake. Left-hand side is calculated using Hydrogen's analytical wavefunctions, the right-hand side is calculated with the use of Gaussian. The maps are generated by calculating the rate for the various positions scanned by the emitter while being moved horizontally in the z=5nm plane.

Quadrupolar/dipolar – ratio map



Near the nanoflake, the quadrupolar and the dipolar transition rates have the same order of magnitude.

Conclusion

I. We have shown the validity of combining Gaussian and COMSOL to simulate emitters in photonic environments.

II. We have also shown that in such environments, quadrupolar transition rate can become on equal footing with the dipolar.

III. This method can now be applied to study more complex emitters (dye molecules, CNT…) in various structures (Tips, nanoantennas, nanodimers…).

Acknowledgments

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