Undo approximations:
Modeling quantum emitters in proximity of nanophotonic structures

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Abstract

Several methods have been developed to calculate the spontaneous emission rate for extended emitters in the vicinity of a nanophotonic structure. While taking the emitter's spatial extension, these methods have taken approximations regarding the full interaction, either by coupling to one single plasmonic mode or adding higher order rates individually thus neglecting interference terms.

Here we apply a new framework [1] to extended emitters, by which, all these interactions can be considered, and we show both under, and over-estimation of the Purcell Factor when using a point-dipole.

System under study

Systems of interest: Extended emitters near plasmonic structures[2]

In the weak-coupling limit, the Fermi Golden rule still holds: $\Gamma = \frac{2\pi}{\hbar^2}\frac{e^2\hbar}{\pi\epsilon_0 m_e^2 c^2}\int\int d\mathbf{r}d\mathbf{r}'\ \psi_e^*(\mathbf{r})\psi_e(\mathbf{r}')\,(\mathrm{Im}\,G_{ij}(\mathbf{r},\mathbf{r}',\omega_0))\,(p_i\psi_g(\mathbf{r}))(p_j^*\psi_g^*(\mathbf{r}'))$ [2]

$G(r,r') = G(|r-r'|) + \frac{1}{k^2}\sum_m \frac{\epsilon_i-\epsilon_b}{(\epsilon_m-\epsilon_i)(\epsilon_m-\epsilon_b)}E_m(r)\otimes E_m^{\mathrm{t}}(r')$ [3]

[1]

$$\Gamma = \frac{2\pi}{k^2}\frac{e^2\hbar}{\pi\varepsilon_0 m_e^2 c^2}\sum_m \mathrm{Im}\left[\gamma_m\left(\int\sum_{ia} C_{ia}\phi_a(r)E_m(r)\nabla\phi_i(r)dr\right)^2\right]$$

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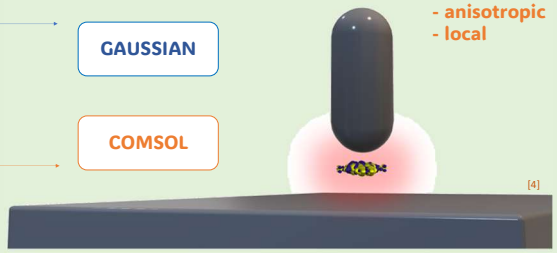
Where:

- $\phi(r)$ are KS-orbitals and C_{ia} their respective contribution to an excited state.
- $E_m(r)$ are eigen-permittivity modes and γ_m their respective weights [3].

Type of medium:

- Inhomogeneous
- lossy
- dispersive
- anisotropic
- local



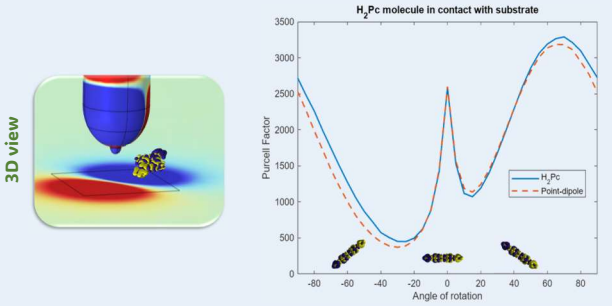
[4]

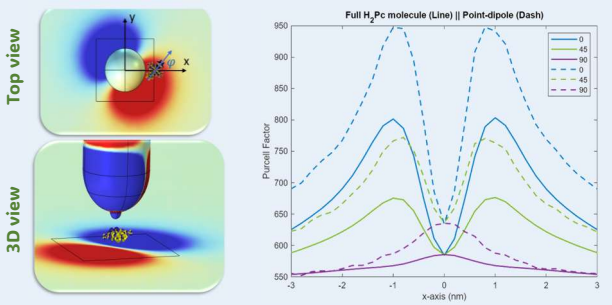
Preliminary results

Full molecule – Point dipole comparison within a gap of 2 nm

Wavelength = 652 nm | Tip height = 36 nm, Tip diameter = 6 nm | Tip material = Silver, Substrate material = Silver

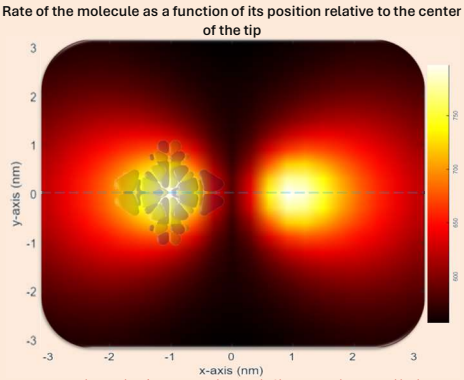
The molecule shows a slightly higher enhancement in comparison to the dipole in the case of it being laterally exposed to the tip as shown in the side image [Negative angle range]





The point-dipole's rate enhancement follows closely the full molecular predicted rate, showing the same pattern overall, though overestimating the enhancement.

Rate map



H_2Pc molecule is translated flat and parallel to the substrate within the gap of the Tip-mirror setup

Conclusion

I. Modeling a molecule as a point-dipole can sometimes overestimate the Purcell-Factor – especially in hot spots.

II. The slightly higher molecular enhancement over its dipolar counterpart can be attributed to either higher transition modes or the molecule's spatial extension enabling it to feel hot spots close to the nanoscatterer.

III. Further investigating the exact reason for any difference in rate enhancement for the current method is under way.

Acknowledgments

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