UNIVERSIDADE FEDERAL DO RIO DE JANEIRO

Conselho Nacional de Desenvolvimento Científico e Tecnológico

CAPES

4TH SYMPOSIUM ON THE CASIMIR EFFECT

TWO-PHOTON SPONTANEOUS EMISSION IN A PHOTONIC CAVITY

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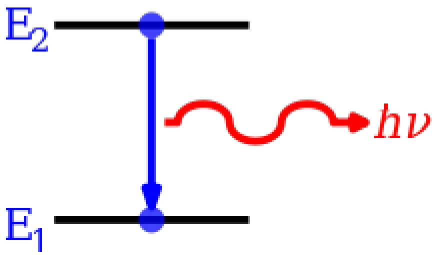
Spontaneous emission (SE)

- An **excited atom**, even when isolated, **decays** to its fundamental state.

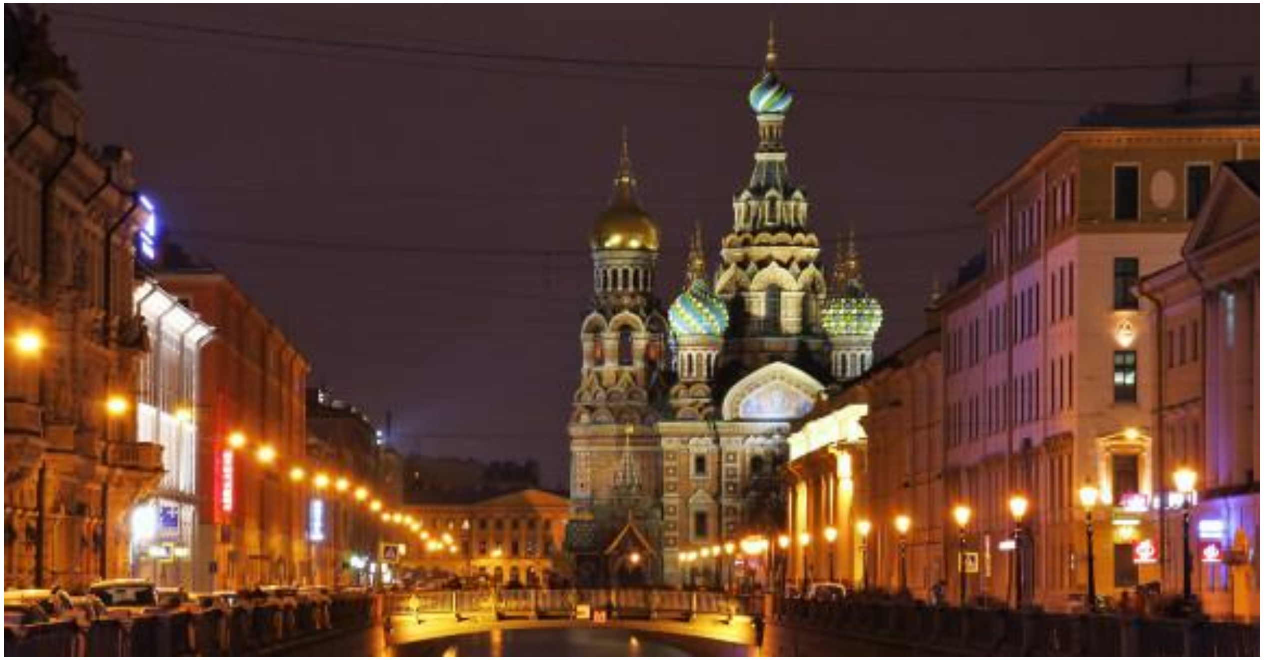


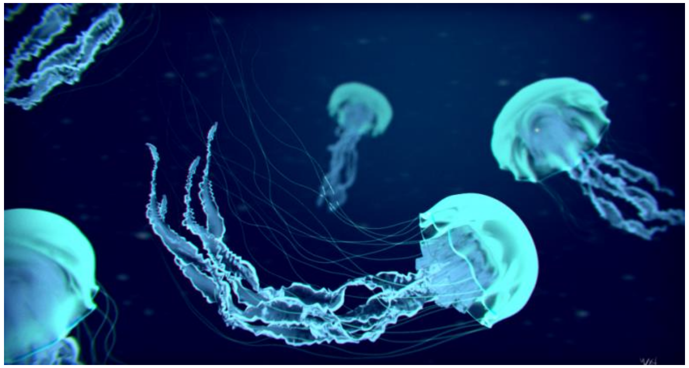
- Phenomenon induced by **quantum vacuum fluctuations**.

- Quantum electrodynamics (QED): excited atom + zero photons is not a stationary state of the atom-field system.

SE

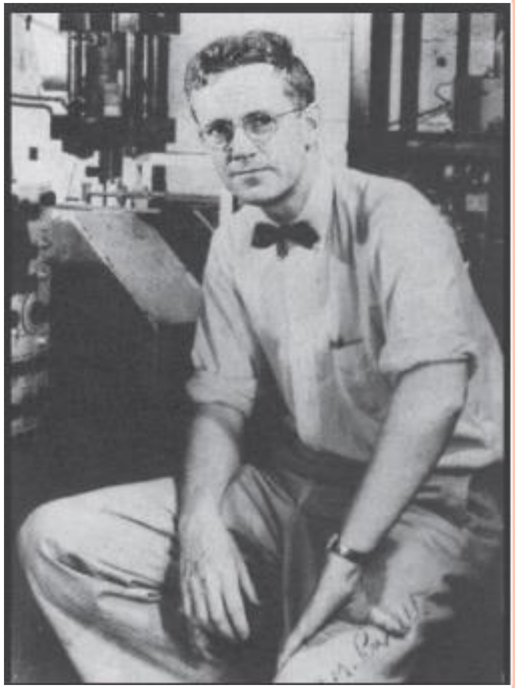
- Most of the light we see is from SE.





PURCELL EFFECT

- E.M. **Purcell (1946)**: bodies in the vicinities of an emitter change its SE rate.



- Reason: the presence of the bodies affects the **boundary conditions** (BC) on the electromagnetic field.

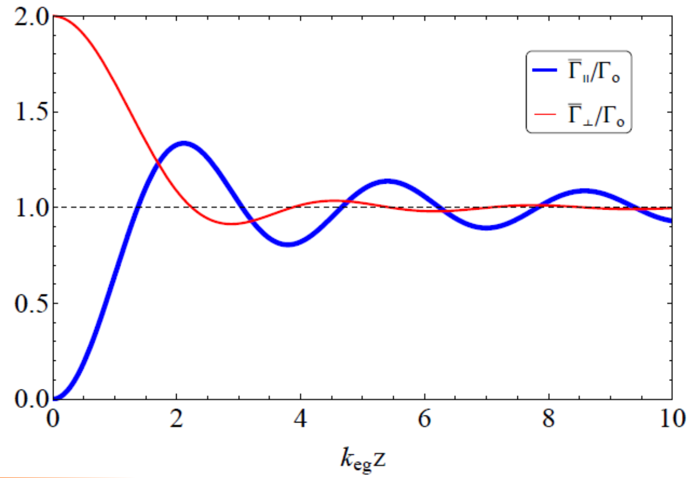
$$\Gamma(\mathbf{R}) = \frac{\pi}{\epsilon_o \hbar} \sum_{\mathbf{k}p} \omega_k |\mathbf{d}_{eg} \cdot \mathbf{A}_{\mathbf{k}p}(\mathbf{R})|^2 \delta(\omega_k - \omega_{eg}).$$

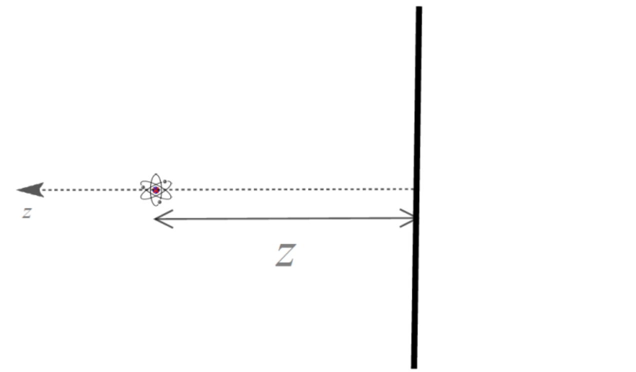
$$\frac{\Gamma}{\Gamma_o} = \frac{6\pi c}{\omega_{eg}} \hat{\mathbf{n}}_{eg}^* \cdot [\mathrm{Im}\mathbb{G}(\mathbf{R}, \mathbf{R}, \omega_{eg})] \cdot \hat{\mathbf{n}}_{eg},$$

$$\nabla \times \nabla \times \mathbb{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{\omega^2}{c^2} \mathbb{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbb{I}\delta(\mathbf{r} - \mathbf{r}').$$

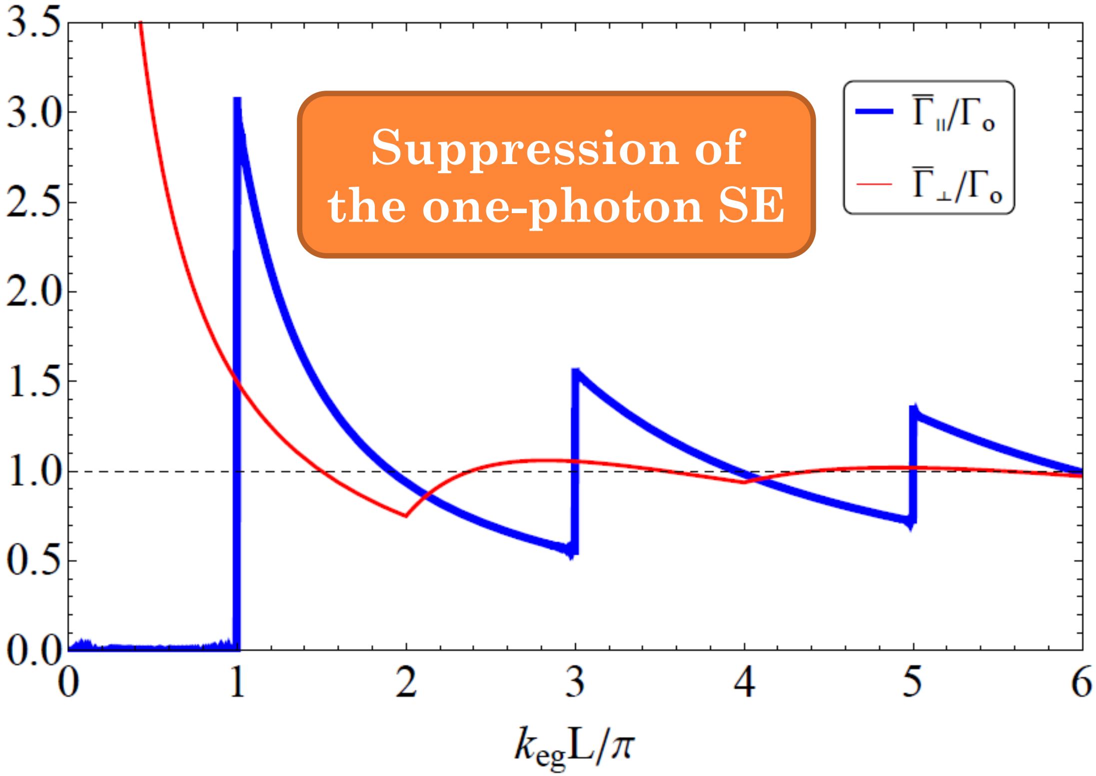
L. Novotny and **B. Hecht**, *Principles of nano-optics*. Cambridge university press, 2012.

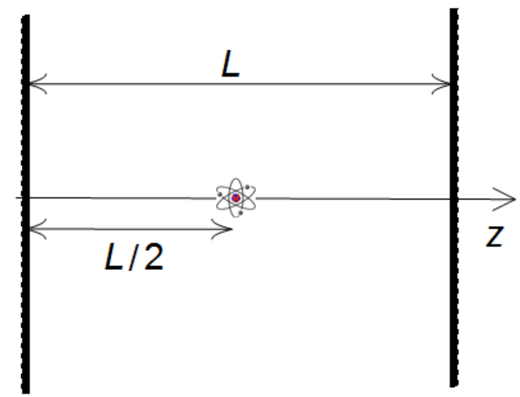
PURCELL EFFECT ON THE ONE-PHOTON SE





Morawitz, Phys. Rev (**1969**)

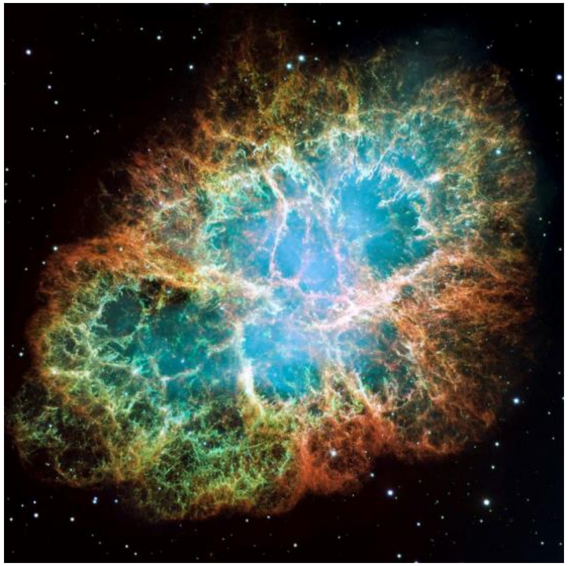




Hulet et al., PRL (**1985**)

Two-photon spontaneous emission (TPSE)

- **Second order process** in perturbation theory.
- Relevant process when the one-photon SE is forbidden, as for intance, due to **selection rules**.
- Ex: **2s – 1s** transition in **H**.
- **Broadband** spectrum of emission.



- Explains the emission spectrum of planetary nebulae.

L. Spitzer and J. L. Greenstein, The Astrophysical Journal, vol. **114**, p. **407 (1951)**.

Purcell effect on the TPSE

- **Not widely discussed** in the literature.

- The progress in **near-field optics, plasmonics,** and **materials science** in general has improved our **control** over **radiation-matter interactions**.

- In some situations the TPSE can even dominate conventionally fast transitions!

N. Rivera *et al*, "*Making two-photon processes dominate one-photon processes using mid-ir phonon polaritons*", PNAS, p. **201713538 (2017)**

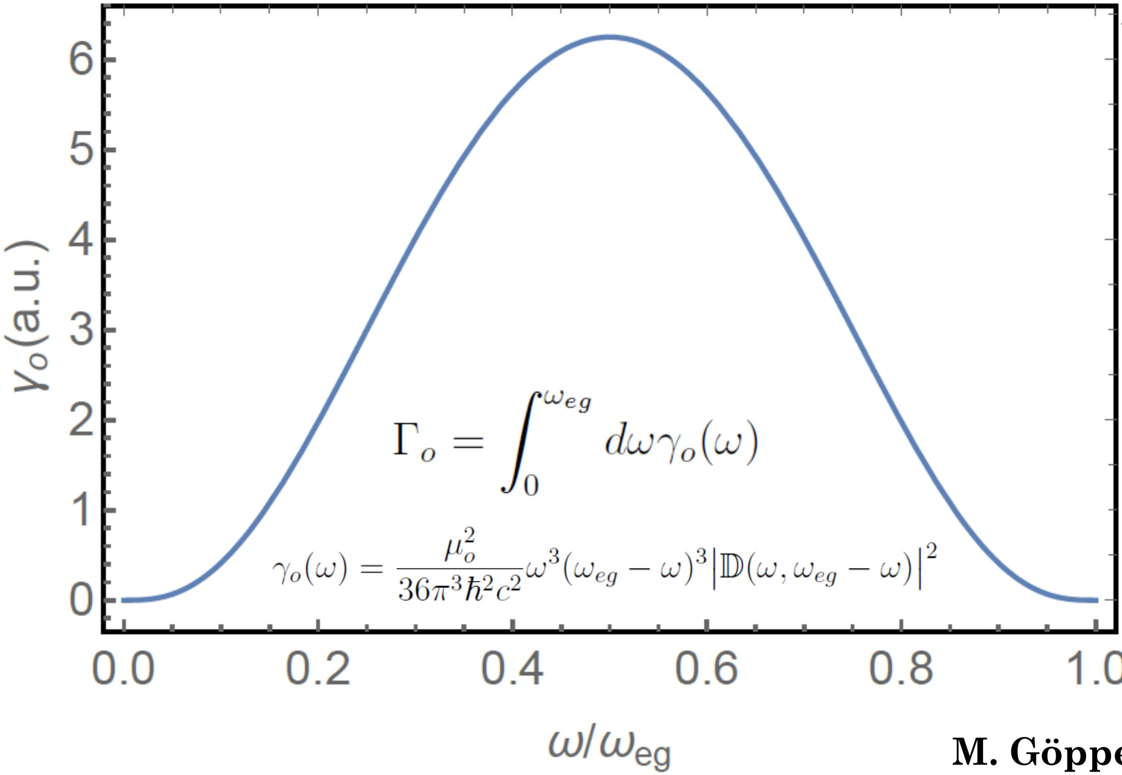
- TPSE is a rich phenomenon, with very much to be explored yet.

TPSE rate: field modes approach

- **Second order Fermi's golden rule** gives

$$\Gamma(\mathbf{R}) = \frac{\pi}{4\epsilon_o^2\hbar^2} \sum_{\mathbf{k}p,\mathbf{k}'p'} \omega_k\omega_{k'} \left|\mathbf{A}_{\mathbf{k}p}(\mathbf{R}) \cdot \mathbb{D}(\omega_k, \omega_{k'}) \cdot \mathbf{A}_{\mathbf{k}'p'}(\mathbf{R})\right|^2 \delta(\omega_k + \omega_{k'} - \omega_{eg}),$$

$$\mathbb{D}(\omega_k, \omega_{k'}) := \lim_{\eta\to 0^+} \sum_m \left[\frac{\mathbf{d}_{em}\mathbf{d}_{mg}}{\omega_{em} - \omega_k + i\eta} + \frac{\mathbf{d}_{mg}\mathbf{d}_{em}}{\omega_{em} - \omega_{k'} + i\eta}\right].$$

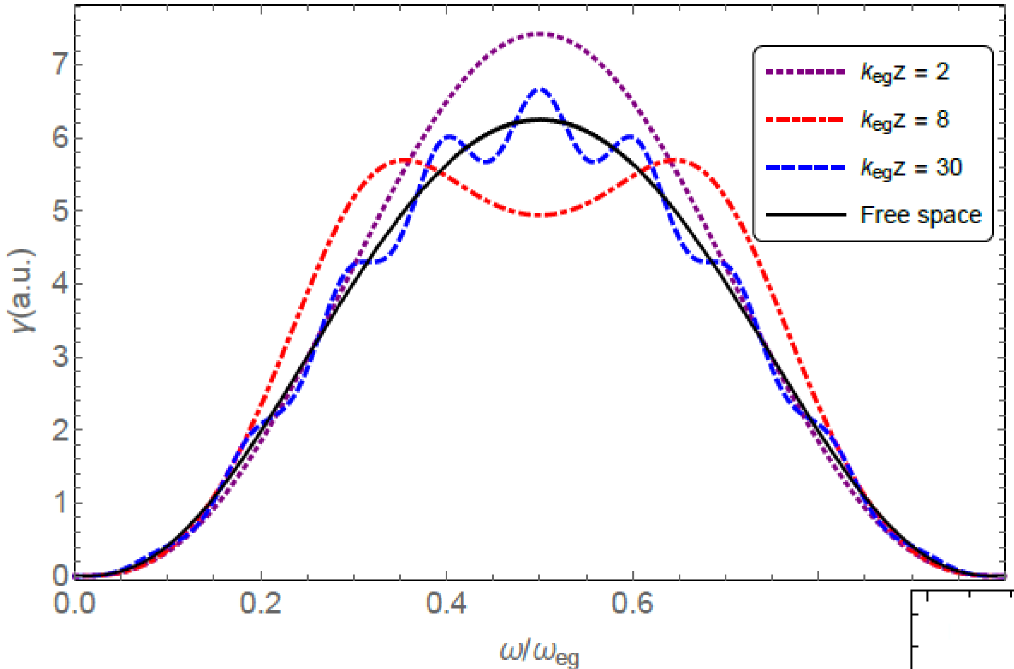


Y.M. *et al*, **arXiv:1906.10716 (2019)**

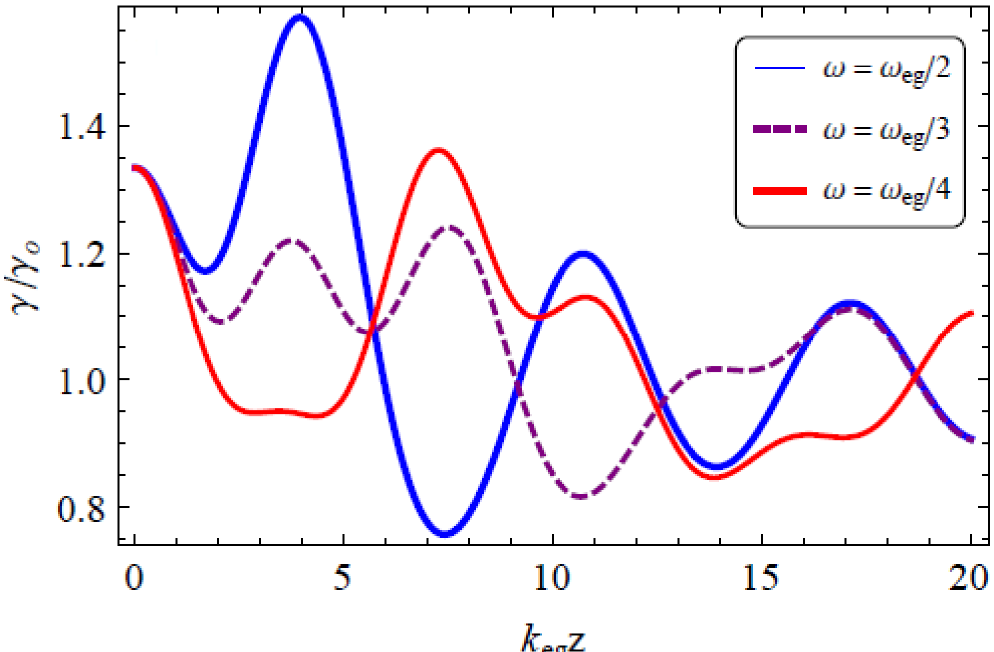
Free space TPSE spectral distribution

M. Göppert-Mayer, Annalen der Physik (**1931**)

AN EMITTER NEAR A PERFECT MIRROR (S → S)



The TPSE rate and the spectral distribution change by the presence of a body.



Anharmonic oscillations for frequencies different from half the transition frequency.

Green's function method

- The imaginary part of the Green's function can be written in terms of the field modes as

$$\mathrm{Im}\mathbb{G}(\mathbf{r},\mathbf{r}',\omega) = \frac{\pi c^2}{2\omega}\sum_{\mathbf{k}p}\mathbf{A}^*_{\mathbf{k}p}(\mathbf{r}')\mathbf{A}_{\mathbf{k}p}(\mathbf{r})\delta(\omega-\omega_k).$$

- Using the previous identity, we recover the well known expression for the TPSE rate, namely

$$\Gamma = \frac{\mu_0^2}{\pi\hbar^2}\int_0^{\omega_{eg}} d\omega\,\omega^2(\omega_{eg}-\omega)^2\mathrm{Im}\mathbb{G}_{il}(\omega)\mathrm{Im}\mathbb{G}_{jn}(\omega_{eg}-\omega)\mathbb{D}_{ij}(\omega,\omega_{eg}-\omega)\mathbb{D}^*_{ln}(\omega,\omega_{eg}-\omega).$$

N. Rivera et al., Science, vol. **353**, no. **6296**, pp. **263–269** (**2016**).

- This constitutes an **alternative demonstration** of this formula!

The Purcell factors relation

- Choosing the basis which diagonalizes the Green's function we have

$$\gamma(\omega) = \frac{\mu_0^2}{\pi\hbar^2}\omega^2(\omega_{eg}-\omega)^2\sum_{i,j}\mathrm{Im}\mathbb{G}_{ii}(\omega)\mathrm{Im}\mathbb{G}_{jj}(\omega_{eg}-\omega)|\mathbb{D}_{ij}(\omega,\omega_{eg}-\omega)|^2.$$

- We define the **Purcell factors** as

$$P_i(\mathbf{R},\omega) := \frac{6\pi c}{\omega}\mathrm{Im}\mathbb{G}_{ii}(\mathbf{R},\mathbf{R},\omega).$$

- In this way, we can write

$$\frac{\gamma(\omega)}{\gamma_o(\omega)} = \sum_{i,j}\frac{|\mathbb{D}_{ij}(\omega,\omega_{eg}-\omega)|^2}{|\mathbb{D}(\omega,\omega_{eg}-\omega)|^2}P_i(\omega)P_j(\omega_{eg}-\omega).$$

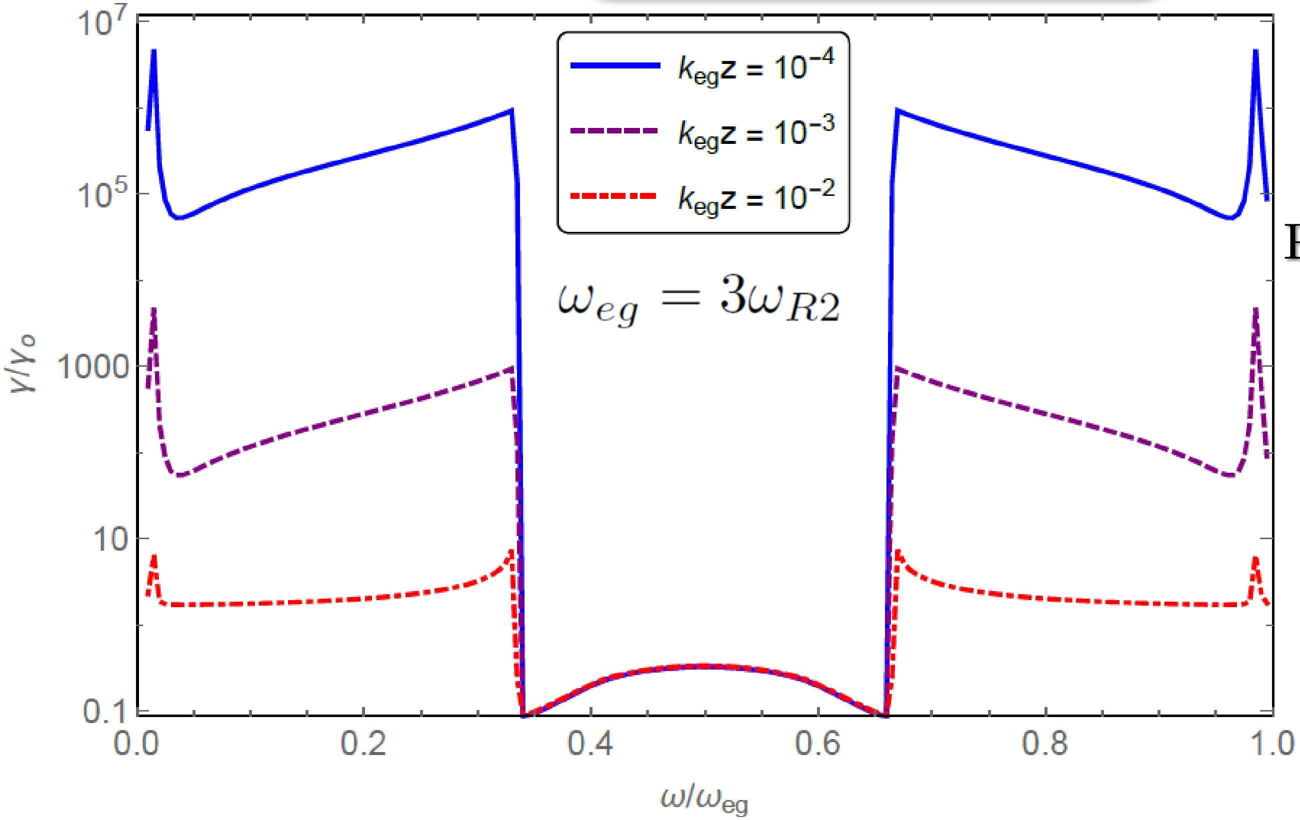
- The **TPSE** rate **dependence** on the **local density of states** (LDOS) was made explicit!

An emitter near a half-space dielectric medium (s → s)

- By symmetry, the **Green's function** is **diagonal** in the **cartesian basis**.

$$P_1(\omega) = P_2(\omega) = P_{\|}(\omega) \;:=\; \frac{\overline{\Gamma}_{\|}}{\Gamma_o}$$

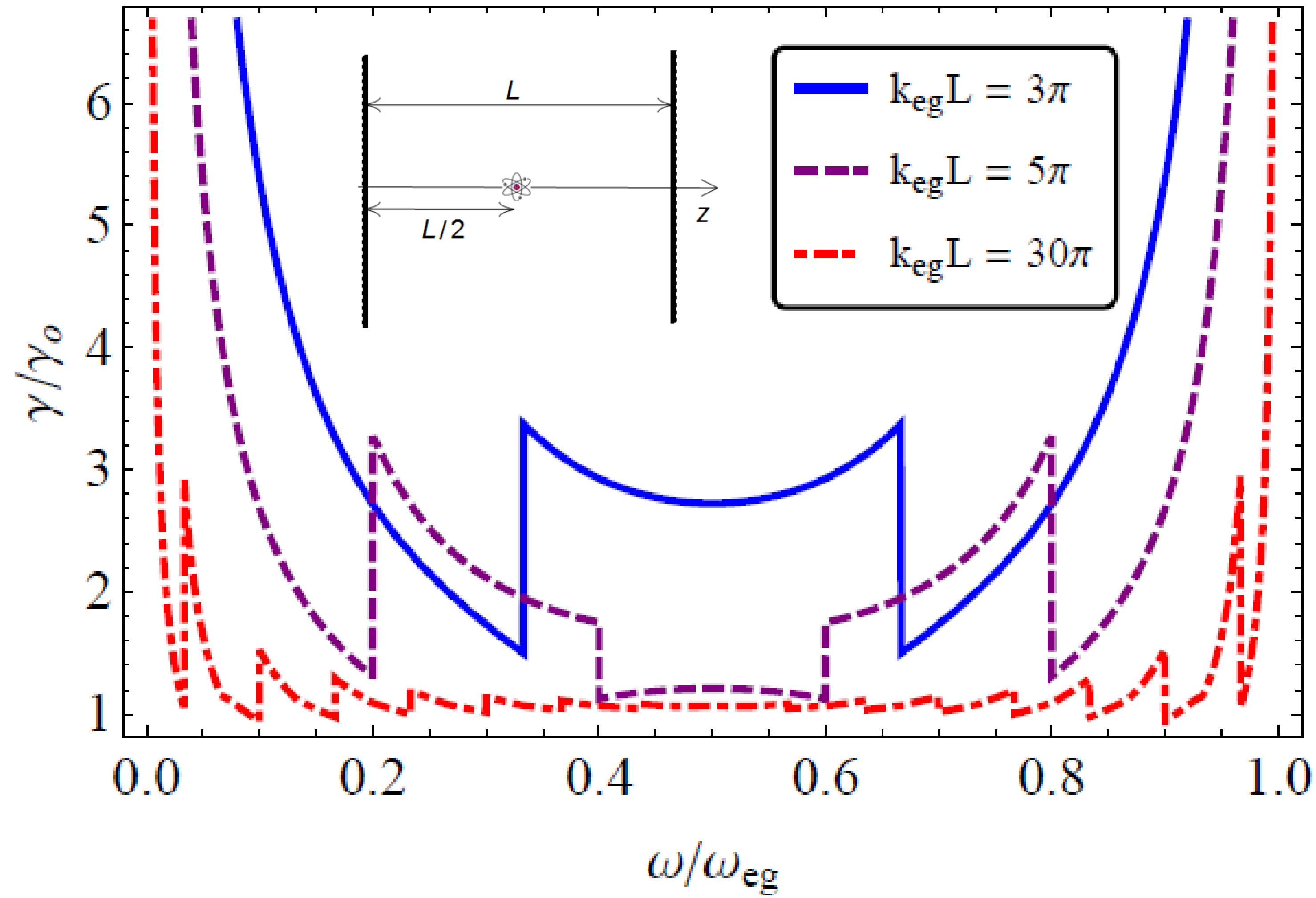
$$P_3(\omega) = P_{\perp}(\omega) \;:=\; \frac{\overline{\Gamma}_{\perp}}{\Gamma_o}$$



Py medium modeled by

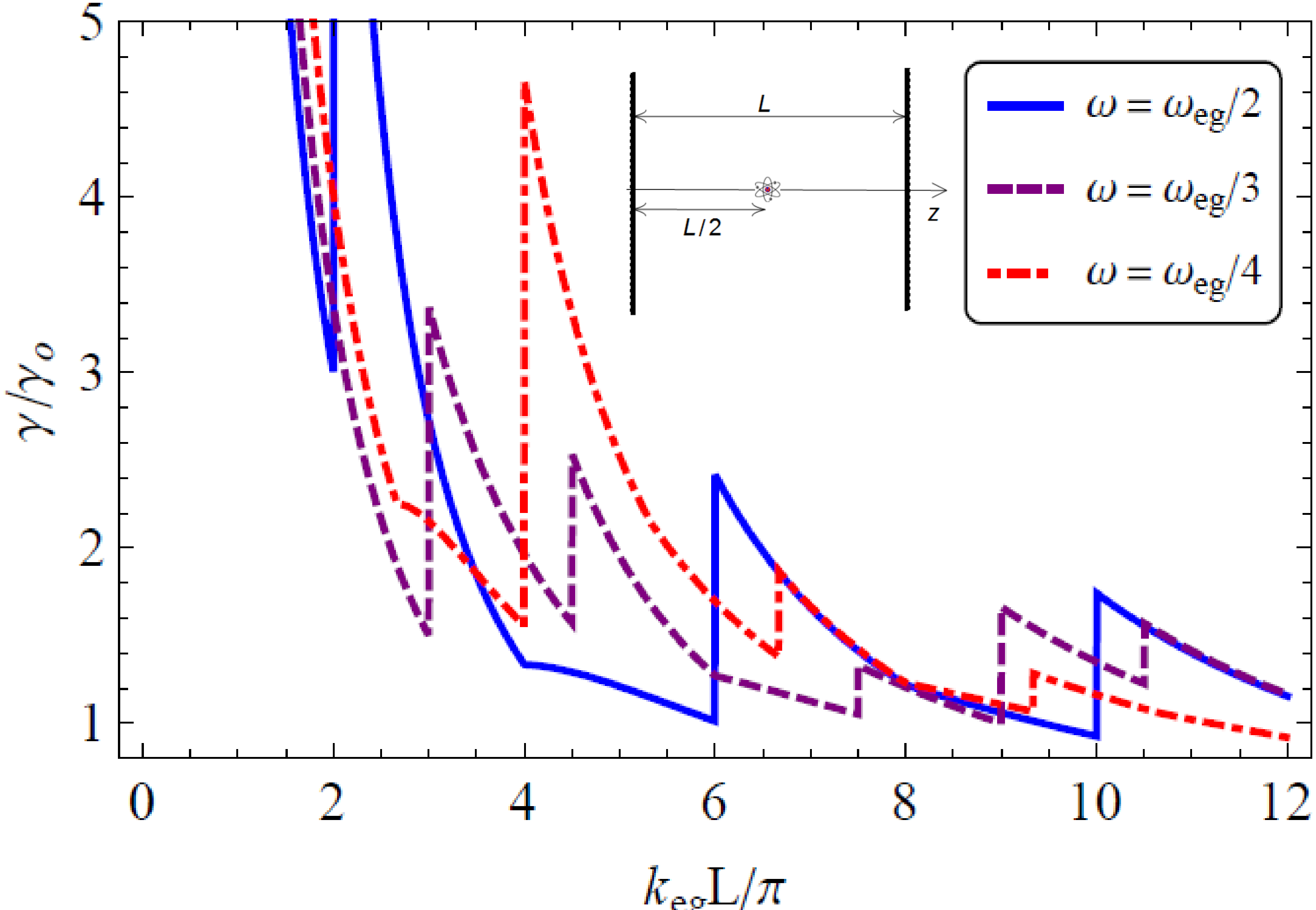
$$\omega_{R1} = 5.54 \times 10^{14} \text{ rad/s}$$
$$\omega_{R2} = 1.35 \times 10^{16} \text{ rad/s}$$
$$\Gamma = 1 \times 10^{11} \text{ rad/s}$$

An emitter between two perfect mirrors (s → s)



The TPSE is not suppressed, in contrast to what happens to the one-photon SE in this situation.

AN EMITTER BETWEEN TWO PERFECT MIRRORS (S → S)



Abrupt changes in the spectral density due to discontinuities in the LDOS.

CONCLUSIONS

- We obtained a new formula for computing the **TPSE rate** in terms of the **field modes**.
- We have shown its **equivalence** with the more usual **Green's function expression**.
- We derived a simple relation between the TPSE spectral density and the corresponding **Purcell factors**.
- For an emitter near a **dielectric**, the TPSE spectral density changes abruptly at the **resonance frequencies**.
- For an emitter placed between **two parallel mirrors**, complete supression never occurs for s→s transitions.

THANK YOU!