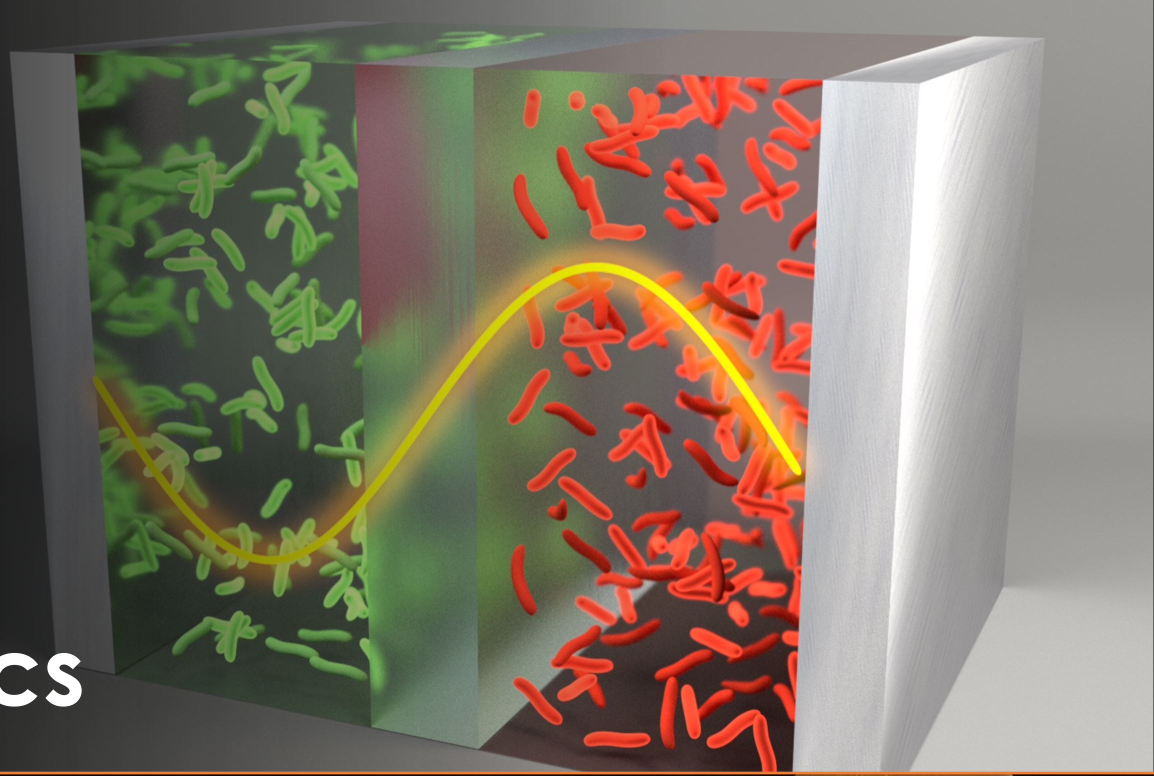
Universidad Autónoma de Madrid

IfiMAC Condensed Matter Physics Center



Mapping between
Quantum & Nano-Photonics

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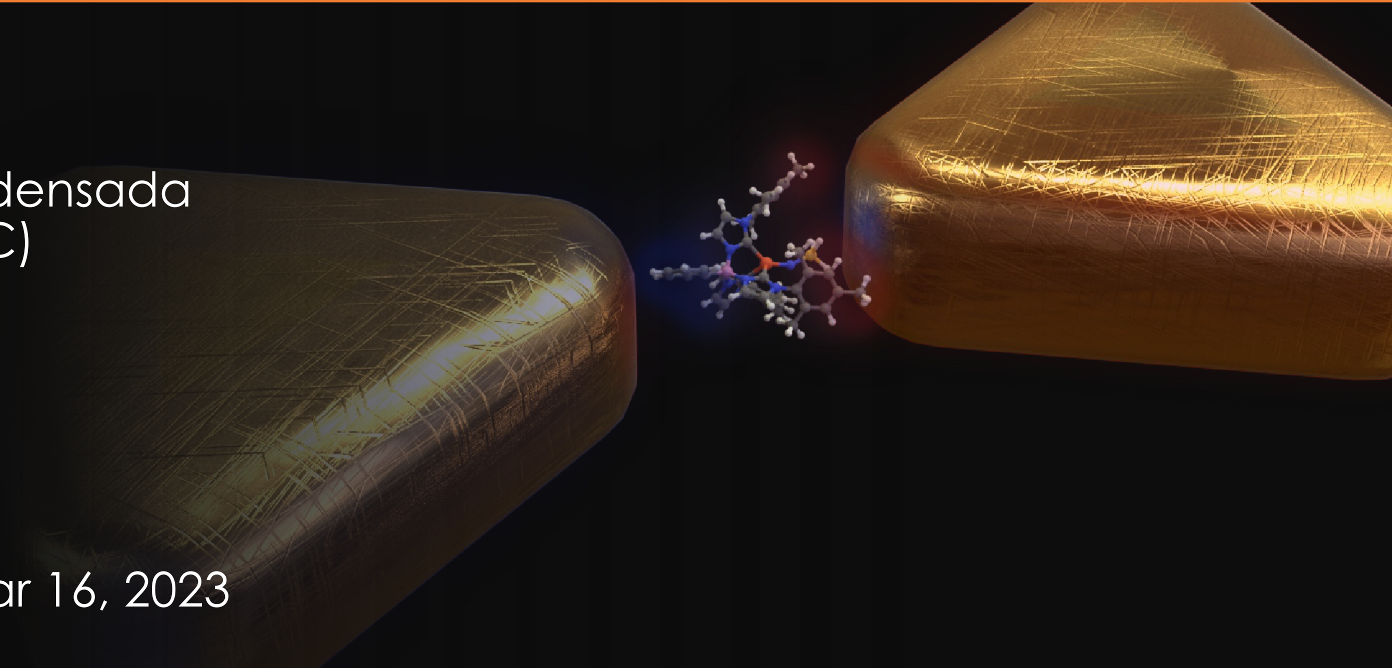
Universidad Autónoma de Madrid



Quantum Nanophotonics, Benasque, Mar 16, 2023

Quick advertisement

https://mmuscles.eu/tools

Emitter properties

$\hbar\omega$	1 eV	f	241.799 THz	λ	1.23984 um	k	5.06773 um^-1	ν	0.806554 um^-1
μ	1 D	$\hbar\gamma$	108.31 peV	γ	164.552 kHz	τ	6.07711 us	f_{osc}	0.00379222

Cavity properties

$\hbar\omega$	1 eV	f	241.799 THz	λ	1.23984 um	k	5.06773 um^-1	ν	0.806554 um^-1
$\hbar\gamma$	50 meV	γ	75.9634 THz	τ	13.1642 fs	Q	20		
V_{eff}	1 um^3	E_{1ph}	95118.7 V / m	λ_c	1.3646e-6				

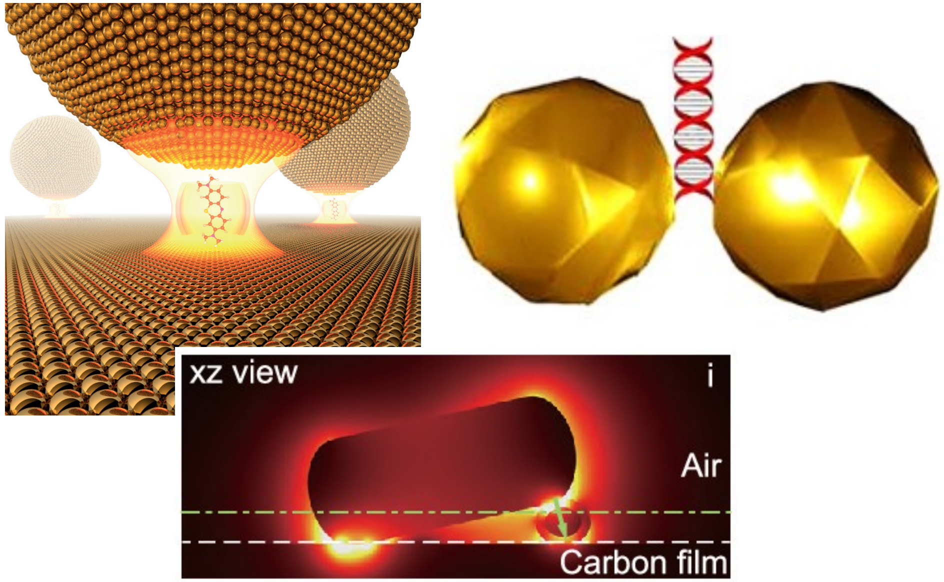
Emitter-cavity coupling

$\hbar\Omega_R$	3.96063 ueV	η	2.89661	C_1	1.58425e-4	C_2	1.12024e-4

Motivation

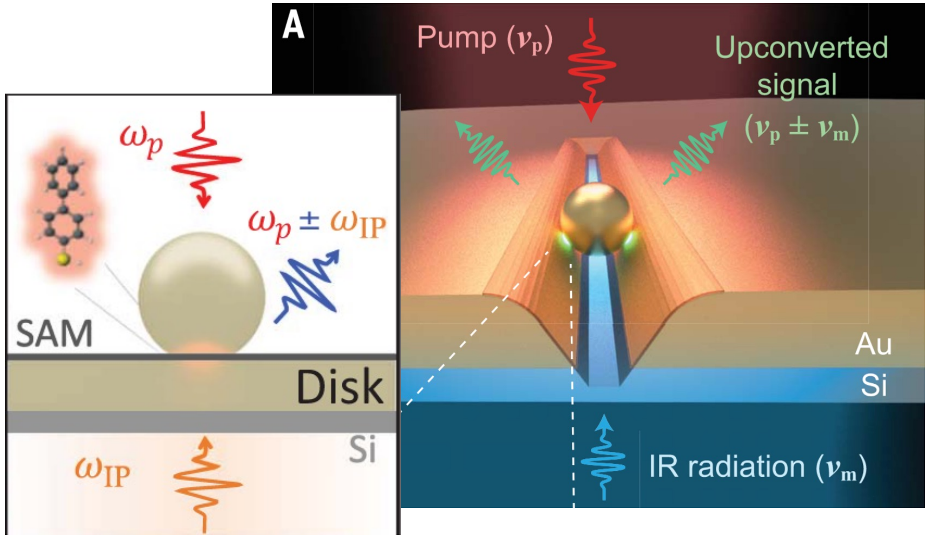
Plasmonic and hybrid nanocavities provide strong light-matter interactions

Few-emitter strong coupling



Chikkaraddy et al., Nature **535**, 127 (2016)
Heintz et al., ACS Nano **15**, 14732 (2021)
Lie et al., Nano Lett. **22**, 4686 (2022)

Frequency upconversion



Chen et al., Science **374**, 1264 (2021)
Xomalis et al., Science **374**, 1268 (2021)

→ **How can we understand quantum light-matter interactions in such (multi-mode) systems?**
→ **How can we use them for (ultrafast) quantum technologies (single-photon sources, nonlinear elements, etc.)?**

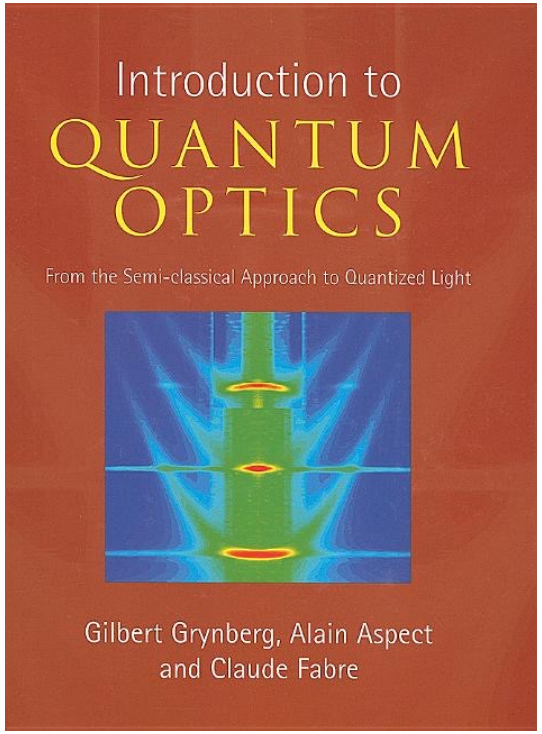
Descriptions of electromagnetic fields

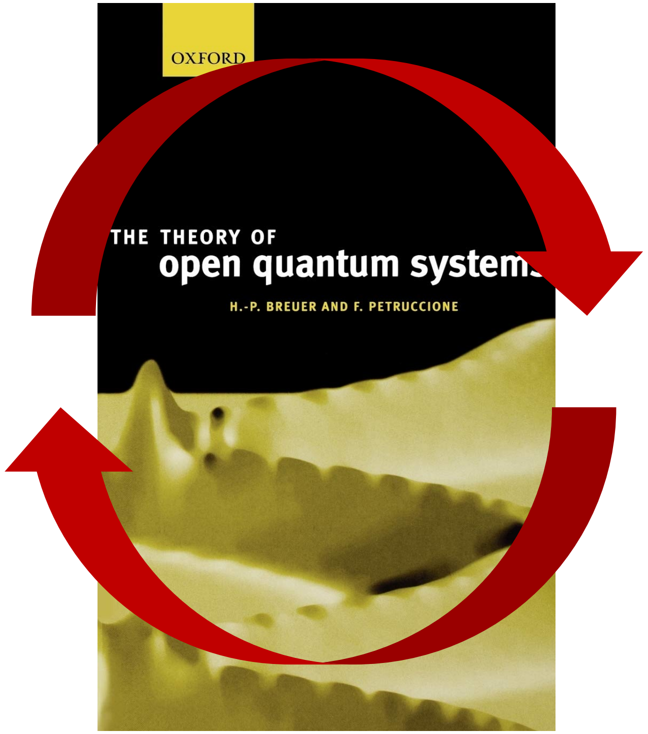
“Traditional” Quantum Optics

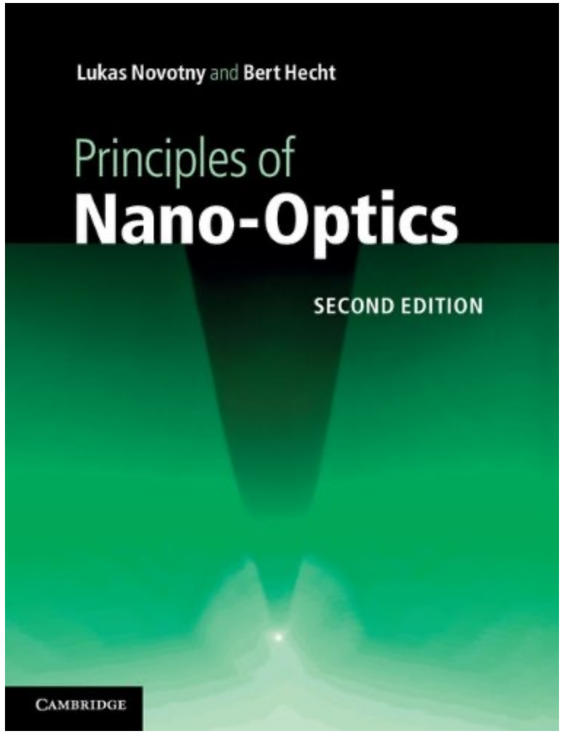
- Discrete modes (often only one)
- Losses described as Lindblad terms

“Traditional” Nanophotonics

- Continuous modes (any frequency and direction)
- Green’s function (tensor) determines “everything”

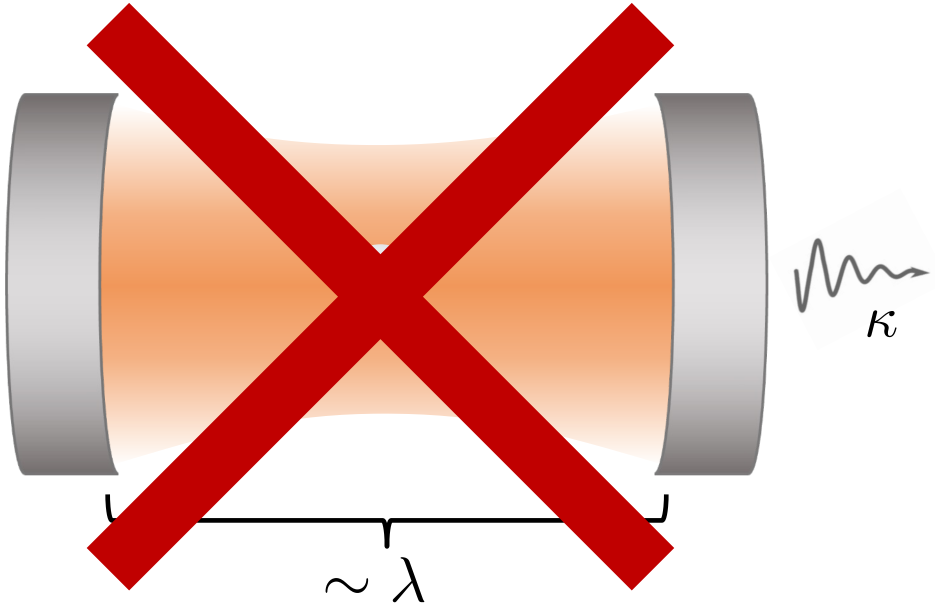






Subwavelength cavity QED

We want/need a quantum description of light-matter interactions in nanophotonic structures.
But what do we really mean with **"light"** and **"matter"**?



sub-wavelength!

"Traditional" quantum optics / cavity QED

1) Cavity mode is a propagating EM field with boundary conditions → discrete modes
2) Losses are small perturbation on top

Subwavelength cavity QED

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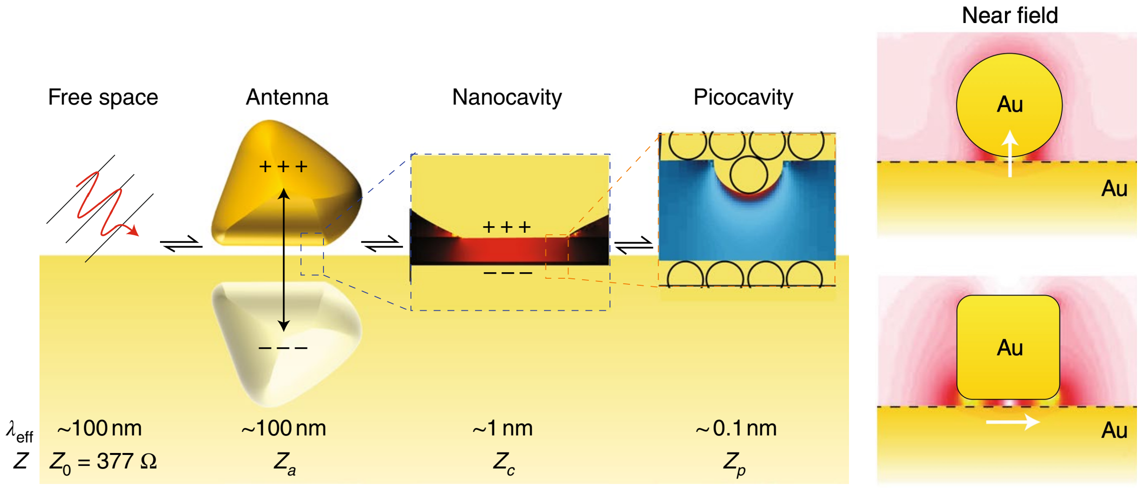


J. J. Baumberg et al., Nat. Mater. **18**, 668 (2019)

"Traditional" quantum optics / cavity QED

1) Cavity mode is a propagating EM field with boundary conditions → discrete modes
2) Losses are small perturbation on top

Nanophotonic (subwavelength) quantum optics

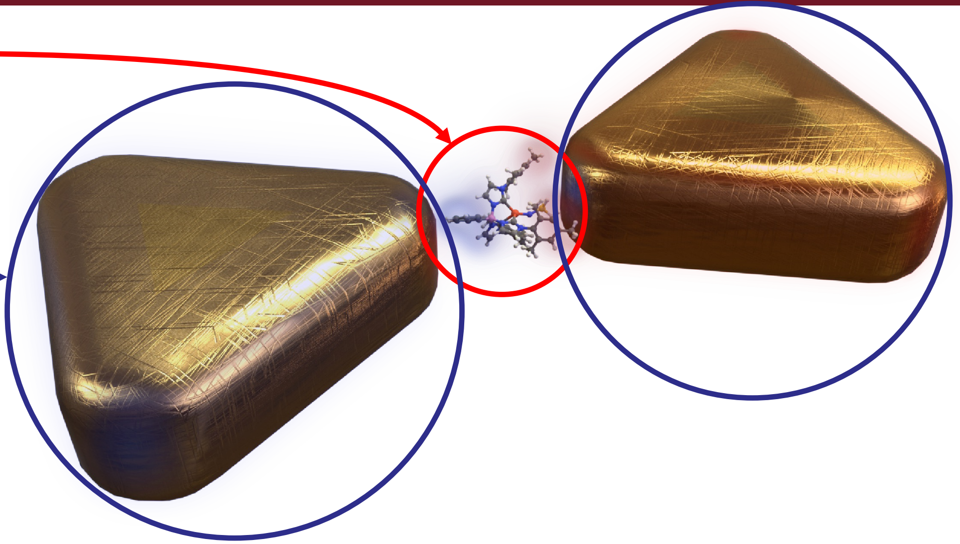
1) Material structure is integral part of "light" modes
2) "Photons" are actually **mixed light-matter excitations** (e.g., surface plasmon **polaritons**)
3) Fast material and radiation losses integral to description (lifetimes on the order of femtoseconds)

Side comment for theorists:
Dominant interaction is with charges in the material (Coulomb force / longitudinal fields), not with propagating (transverse) fields → fundamental interaction is $\vec{\mu} \cdot \vec{E}$, not $\vec{p} \cdot \vec{A}$ and $\vec{A}^2$
("an electron by any other name is still an electron")

Quantization strategy: Macroscopic QED

1) Separate:
   a) **Emitter(s)** (collections of charged particles)
      Described through **quantum chemistry** or similar

   b) **“Cavity”** (arbitrary material structure with **linear** response)
      Described through **macroscopic electromagnetism (Maxwell)**
      Local response: permittivity $\epsilon(\vec{r},\omega)$ and permeability $\mu(\vec{r},\omega)$



C. Climent et al., Angew. Chemie Int. Ed. **58**, 8698 (2019)

2) Treat **“cavity” material** + **free-space EM modes**:
   a) Find system of coupled harmonic oscillators reproducing macroscopic Maxwell equations.
      Dissipation: coupling to “internal” harmonic oscillators.
   b) Diagonalize (formally → solutions expressed through Green’s functions)
   c) Quantize harmonic oscillators (promote variables to operators)
      → **quantized medium-assisted EM field:** infinite collection of bosonic modes (quantum harmonic oscillators)

$$H_F = \sum_\lambda \int_0^\infty \mathrm{d}\omega \int \mathrm{d}^3\mathbf{r}\, \hbar\omega\, \hat{\mathbf{f}}_\lambda^\dagger(\mathbf{r},\omega)\hat{\mathbf{f}}_\lambda(\mathbf{r},\omega)$$

$$\hat{\mathbf{E}}(\mathbf{r}) = \sum_\lambda \int_0^\infty \mathrm{d}\omega \int \mathrm{d}^3\mathbf{r}'\, \mathbf{G}_\lambda(\mathbf{r},\mathbf{r}',\omega)\cdot\hat{\mathbf{f}}_\lambda(\mathbf{r}',\omega) + \mathrm{H.c.}$$

Huttner and Barnett, Phys. Rev. A 46, 4306 (1992); Dung, Knöll, Welsch, Phys. Rev. A 57, 3931 (1998)
Scheel, Knöll, Welsch, Phys. Rev. A 58, 700 (1998); Philbin, New J. Phys. 12, 123008 (2010)
Scheel and Buhmann, Acta Phys. Slov. 58, 675 (2008); Buhmann, Dispersion Forces I & II (2012)

Quantization strategy: Macroscopic QED

3) Reintroduce emitter & do unitary transformation of EM modes to simplify Hamiltonian:
Final result: **Emitter – EM mode interaction fully characterized by "spectral density" (~ local density of states)**

$$H = H_e + \int_0^\infty \left[\omega a^\dagger(\omega) a(\omega) + \hat{\mu}_e \sqrt{J(\omega)} \left(a(\omega) + a^\dagger(\omega)\right)\right] \mathrm{d}\omega$$

$$J(\omega) = \frac{\omega^2}{\pi\epsilon_0 c^2} \boldsymbol{\mu}_E \cdot \operatorname{Im} \mathbf{G}(\mathbf{r}_E, \mathbf{r}_E, \omega) \cdot \boldsymbol{\mu}_E$$

Purcell Factor: $P(\omega) = \dfrac{J(\omega)}{J_0(\omega)}$

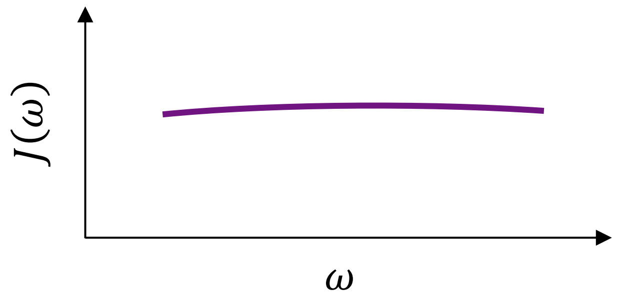
$J(\omega)$ fully characterizes the "cavity"
→ two systems with the same spectral density are indistinguishable for the emitter

Spectral density is the "central" quantity in open quantum systems theory.

Markovian $J(\omega)$

Spectrally flat

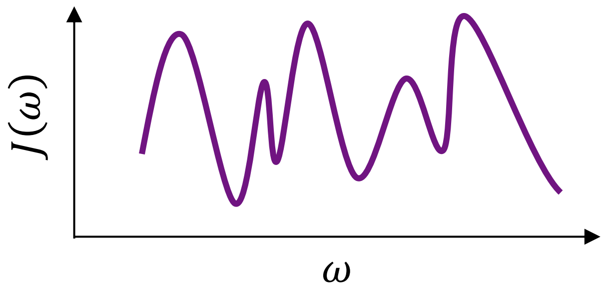
"Memoryless" environment



Non-Markovian $J(\omega)$

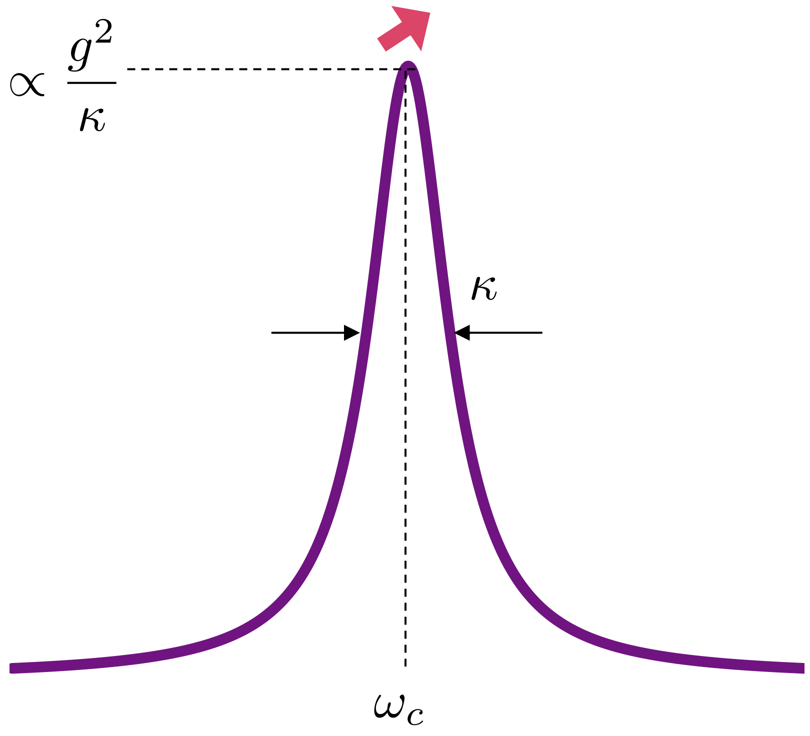
Arbitrary densities

Dynamics depend on previous times



Formulation used here: "Emitter-centered modes"
Feist, Fernández-Domínguez, García-Vidal, Nanophotonics 10, 477 (2020)

Lorentzian spectral density



Lorentzian spectral density ("resonance"):

$$J(\omega) = \frac{g^2}{\pi} \frac{\kappa/2}{(\omega-\omega_c)^2 + (\kappa/2)^2} = \frac{g^2}{\pi} \mathrm{Im}\left[\frac{1}{\omega_c - i\kappa/2 - \omega}\right]$$

$$H = H_e + \int_0^\infty \left[\omega a^\dagger(\omega) a(\omega) + \hat{\mu}_e \sqrt{J(\omega)}\left(a(\omega) + a^\dagger(\omega)\right)\right] \mathrm{d}\omega$$

Exactly equivalent to a Lindblad master equation with a lossy mode!

$$\partial_t \rho = -i[H_R, \rho] + \kappa \mathcal{L}_a[\rho]$$

$$H_R = H_e + \omega_c a^\dagger a + g\hat{\mu}_e(a + a^\dagger) \qquad \mathcal{L}_a[\rho] = a\rho a^\dagger - \frac{1}{2}\{a^\dagger a, \rho\}$$

Multiple lossy modes: **sum** of Lorentzians

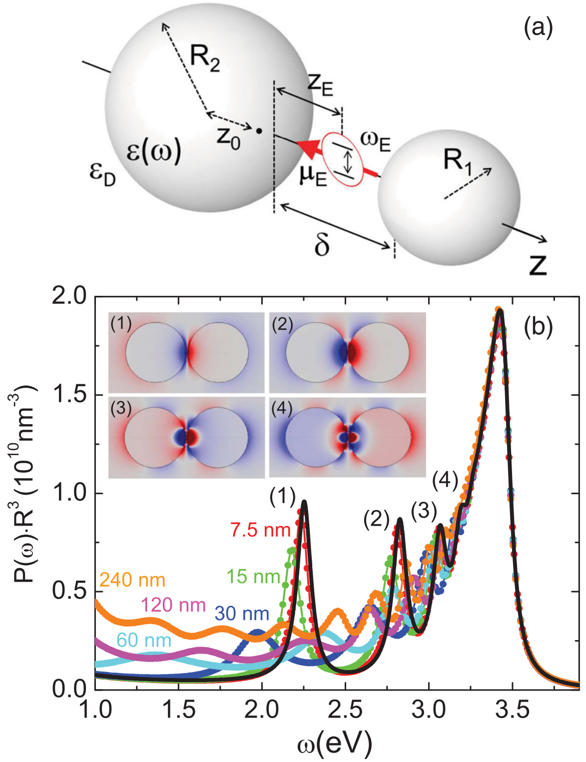
For clean systems with relatively sharp isolated resonances, this is "enough". **But does it always work?**

Imamoğlu, Phys. Rev. A 50, 3650 (1994)
Tamascelli, Smirne, Huelga, Plenio, Phys. Rev. Lett. 120, 030402 (2018)

Nanophotonic spectral densities

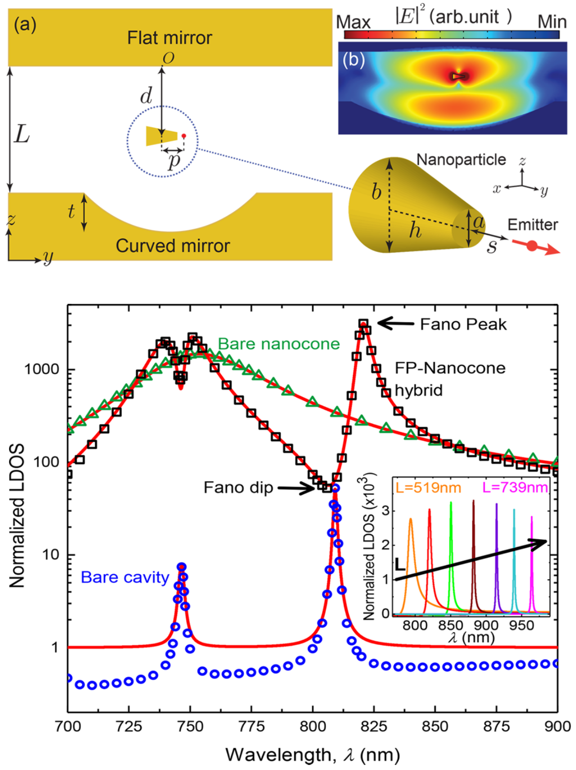
Spectral densities of some example systems

Plasmonic nanogap antenna

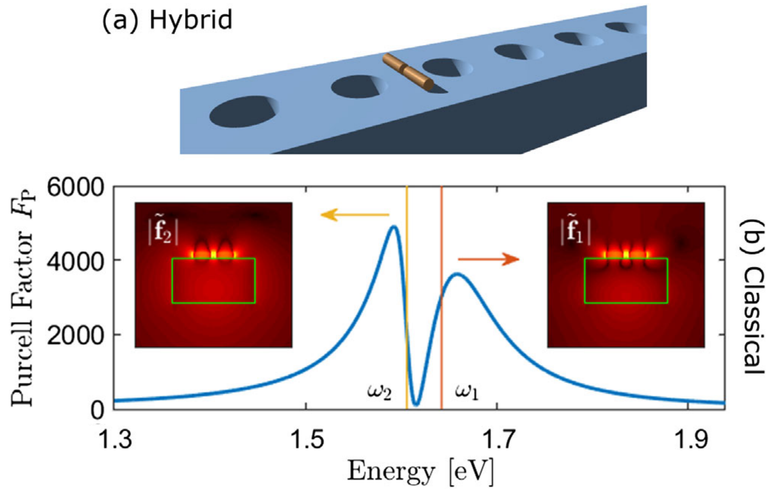


R.-Q. Li et al., Phys. Rev. Lett. **117**, 107401 (2016)

Hybrid plasmonic – dielectric cavities



B. Gurlek et al., ACS Photonics **5**, 456 (2018)



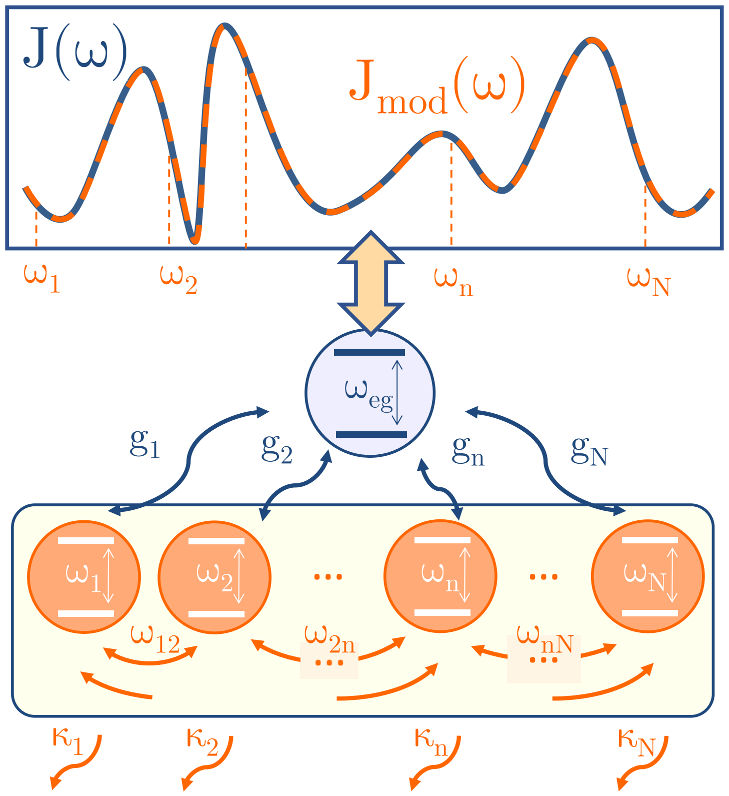
S. Franke et al., Phys. Rev. Lett. **122**, 213901 (2019)

“Resonances” are not necessarily Lorentzian, with interference effects (Fano), nonsymmetric peaks, etc.

One option: Quantization of “quasinormal modes” (Franke et al., PRL 2019). But complex to implement and limited when many modes contribute to a single peak.

Can we find a simpler and more general approach?

Coupled modes spectral density



What if we use multiple modes, and allow them to interact?

Lindblad master equation of **coupled modes** with losses:

$$\partial_t \rho = -i[H_R, \rho] + \sum_i \kappa_i \mathcal{L}_{a_i}[\rho]$$

$$H_R = H_e + \sum_{ij} \omega_{ij} a_i^\dagger a_j + \sum_i g_i \hat{\mu}_e (a_i + a_i^\dagger)$$

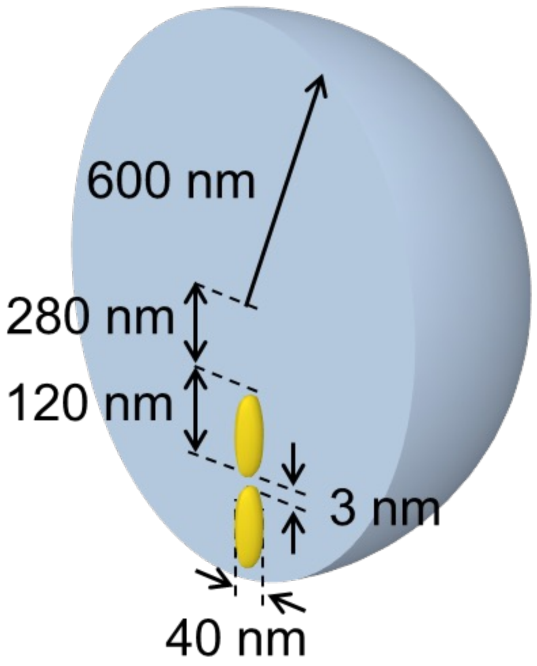
Exactly equivalent to a spectral density:

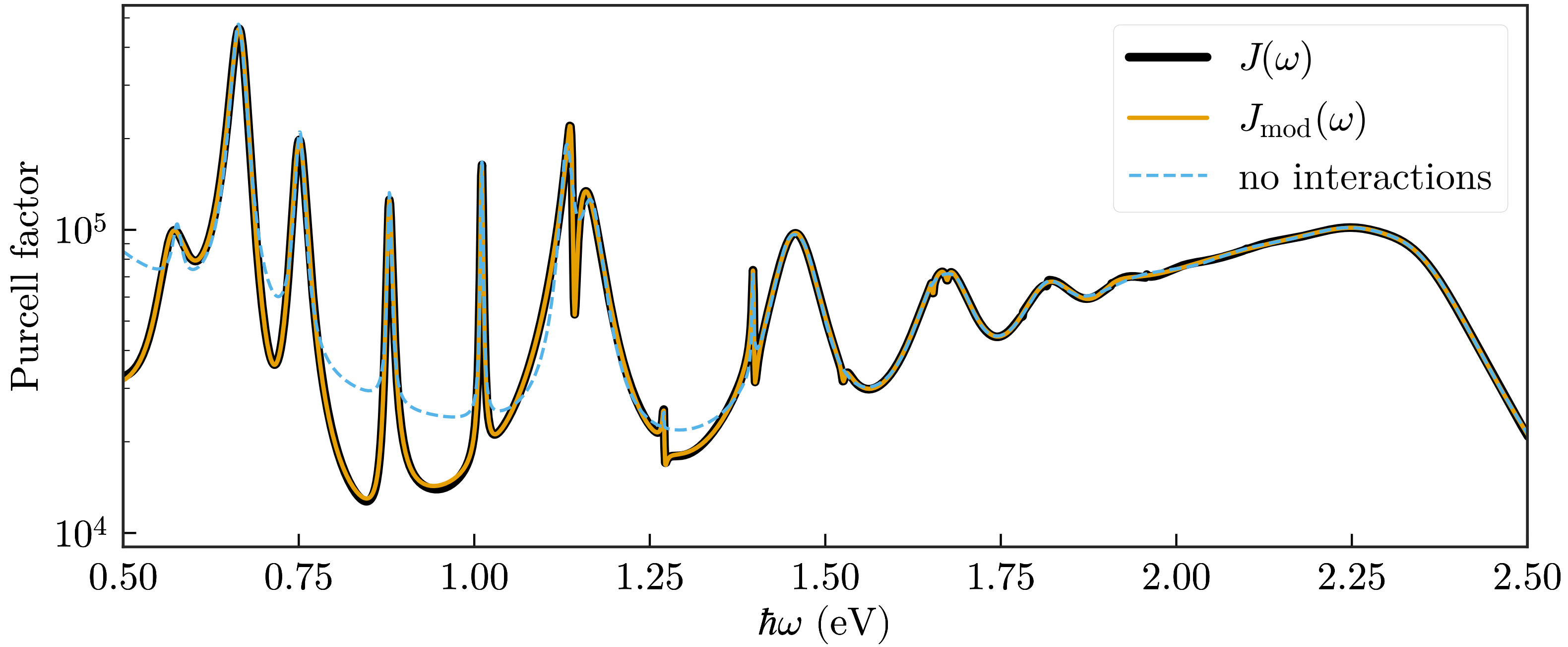
$$J_{\mathrm{mod}}(\omega) = \frac{1}{\pi} \vec{g} \cdot \mathrm{Im}\left[\frac{1}{\tilde{\mathbf{H}} - \omega}\right] \cdot \vec{g} \qquad \tilde{\mathbf{H}} = \begin{pmatrix} \omega_{11} - \frac{i}{2}\kappa_1 & \omega_{12} & \cdots \\ \omega_{21} & \omega_{22} - \frac{i}{2}\kappa_2 & \cdots \\ \vdots & \vdots & \ddots \end{pmatrix}$$

Interactions give much more flexibility for describing spectral density!
"Just" need to find model system that reproduces nanophotonic J(ω)
→ fitting to obtain parameters – only approximation in the method!

Few-mode quantization of arbitrary spectral densities

Hybrid test system: high-dielectric microsphere with ellipsoidal plasmonic nanoantenna
Complex spectral density with several interference (Fano-like) features.



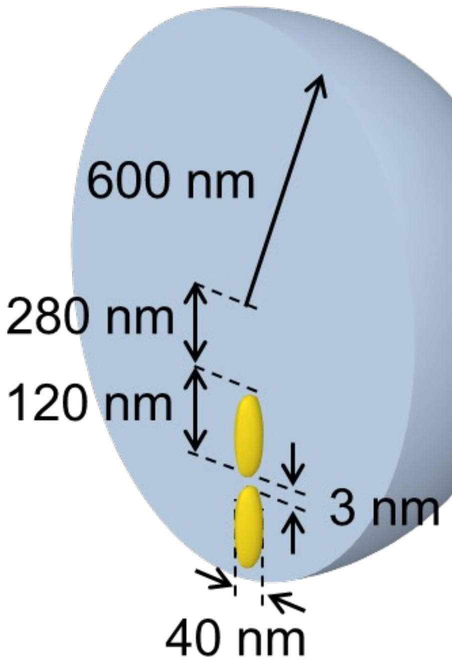


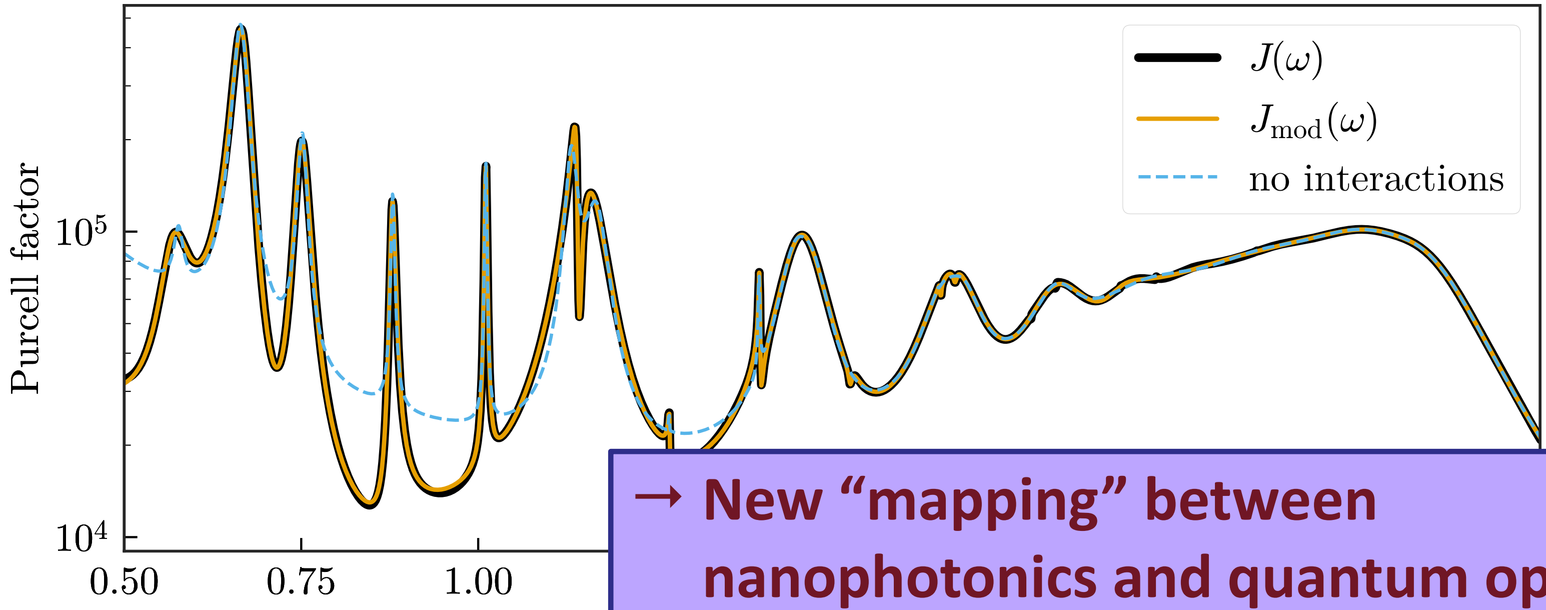
- **Almost perfect fit of full spectrum using 20 modes!**
  (Quasi-normal modes: orders of magnitude more in the spectral region)
- Fit with **non-interacting** modes (sum of Lorentzians) **cannot reproduce** complex interference structure and overestimates density in several regions.

I. Medina et al., Phys. Rev. Lett. 126, 093601 (2021)

Few-mode quantization of arbitrary spectral densities

Hybrid test system: high-dielectric microsphere with ellipsoidal plasmonic nanoantenna
Complex spectral density with several interference (Fano-like) features.





→ **New "mapping" between nanophotonics and quantum optics**
→ **Couplings and losses do not commute (otherwise: sum of Lorentzians)**
→ **Mode interactions are an intrinsic feature of nanophotonic systems**

- **Almost perfect fit of full spectrum using 20 modes!**
  (Quasi-normal modes: orders of magnitude more in the spe
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I. Medina et al., Phys. Rev. Lett. 126, 093601 (2021)

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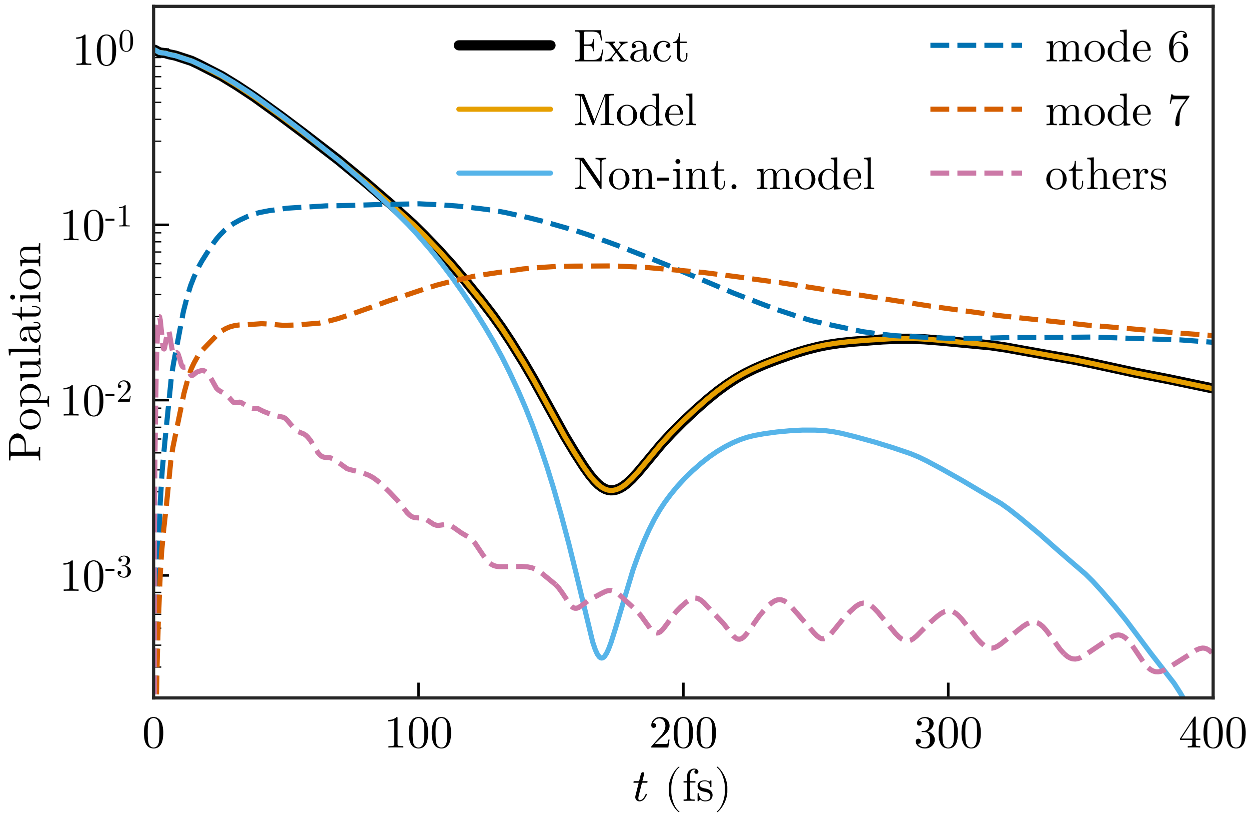
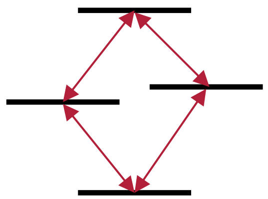
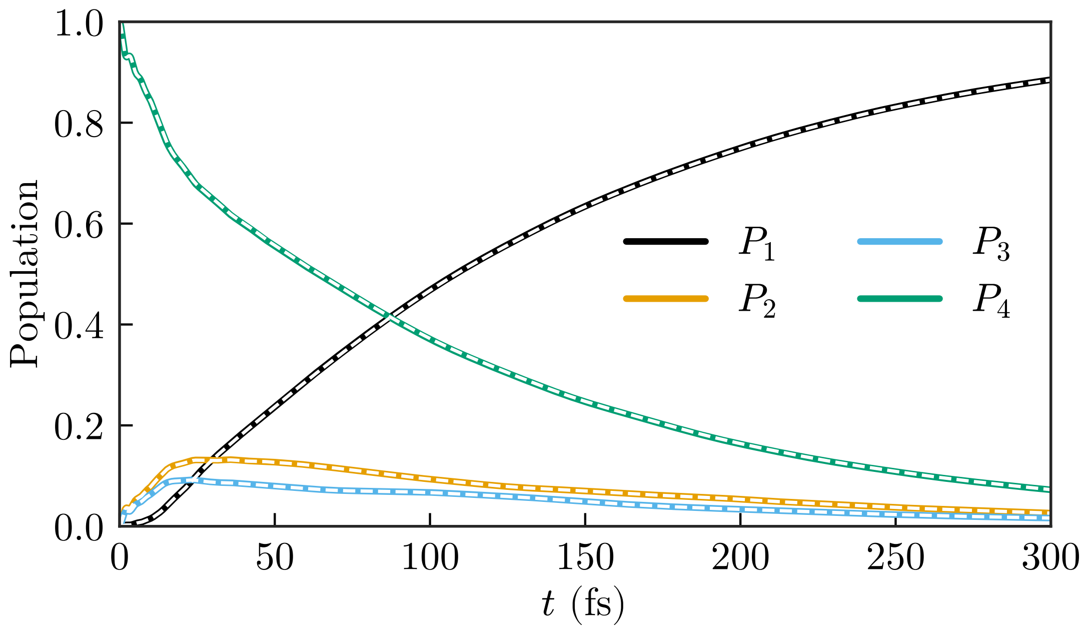


Spontaneous emission dynamics confirm correctness of model.

Works for any coupling regime (weak or strong).

Emitter can have any internal structure.





I. Medina et al., Phys. Rev. Lett. 126, 093601 (2021)

Few-mode quantization of arbitrary spectral densities

Have access to spatially resolved EM field

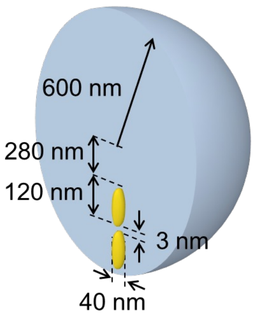
Spontaneous emission: no coherent field!
→ Need quantum calculation

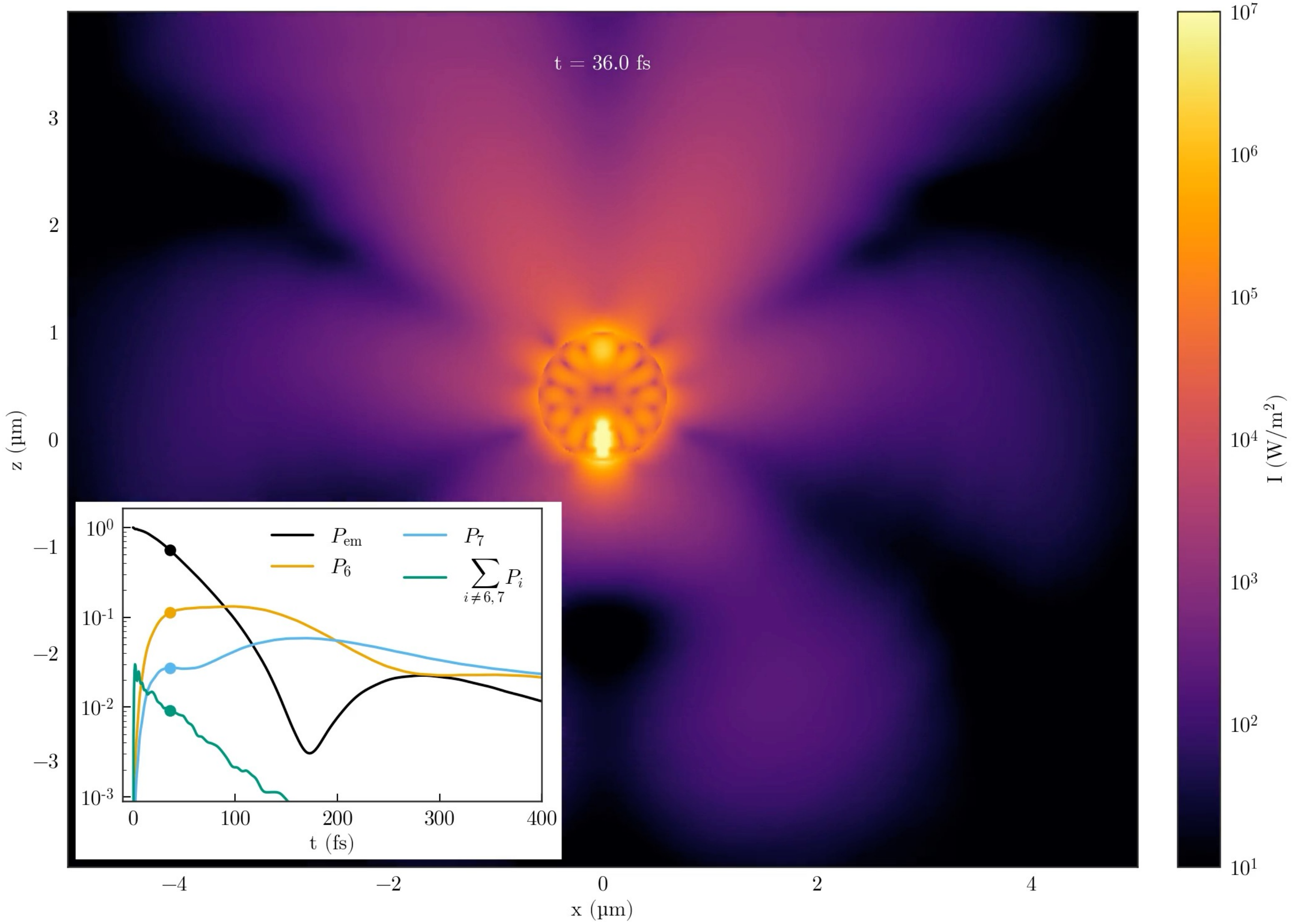
$\langle \vec{E}(\vec{r}) \rangle = 0$

Can be transformed to “chain” form with next-nearest neighbor coupling

M. Sánchez-Barquilla, JF, Nanomaterials 11, 2104 (2021)

Up to now: **single** emitter and **single** polarization direction
Can we go beyond that?





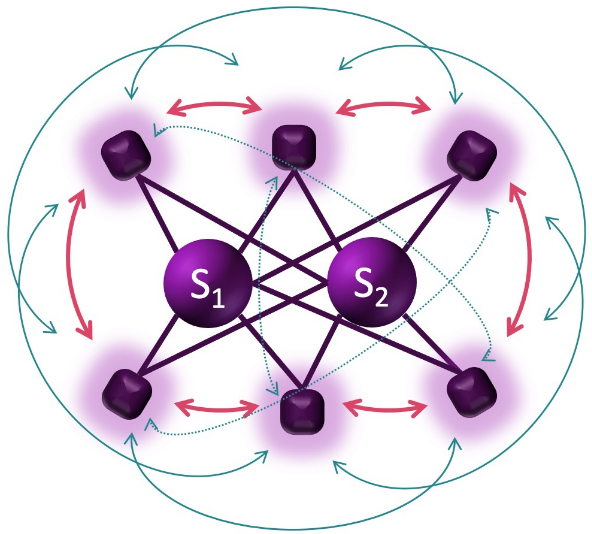
I. Medina et al., Phys. Rev. Lett. 126, 093601 (2021)

Extension to multiple emitters

- **Macroscopic QED:**
  Light-matter interaction for multiple emitters is **fully characterized** by a **generalized spectral density** describing EM-mediated interaction between emitters n and m:

$$\mathcal{J}_{nm}(\omega) = \frac{\omega^2}{\pi \epsilon_0 c^2} \mathbf{n}_n \cdot \operatorname{Im} \mathbf{G}(\mathbf{r}_n, \mathbf{r}_m, \omega) \cdot \mathbf{n}_m$$

- **Few-mode quantization model easily extended to that case**
  → coupling vector becomes a matrix

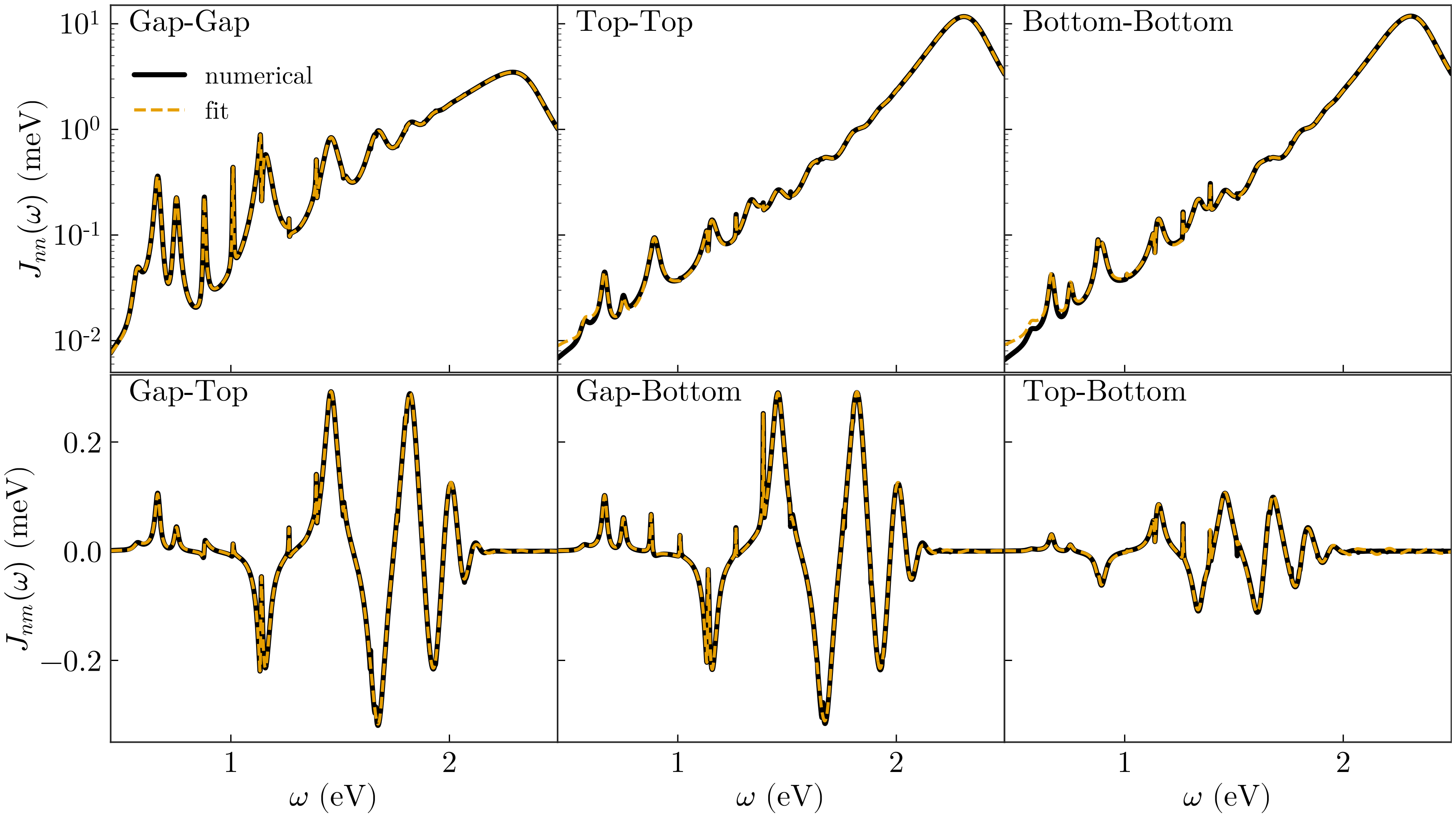


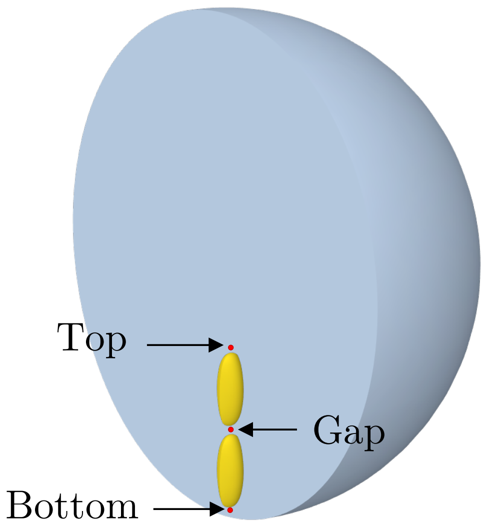
$$H_R = \sum_n H_{e,n} + \sum_{ij} \omega_{ij} a_i^\dagger a_j + \sum_i g_{ni} \hat{\mu}_{e,n} (a_i + a_i^\dagger)$$

$$\mathcal{J}_{\text{mod}}(\omega) = \frac{1}{\pi} \mathbf{g} \operatorname{Im} \left[\frac{1}{\tilde{\mathbf{H}} - \omega} \right] \mathbf{g}^T$$

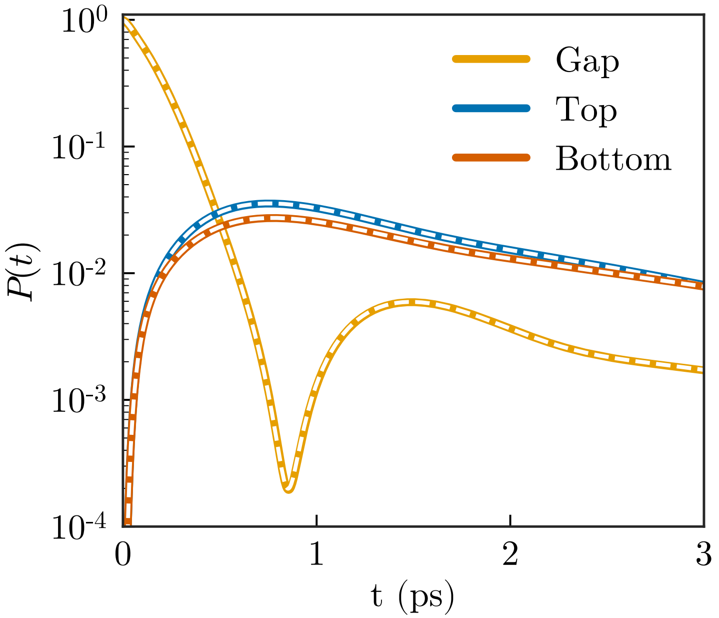
J. Feist et al., Nanophotonics **10**, 477 (2020)
M. Sánchez-Barquilla et al., Nanophotonics **11**, 4363 (2022)

Few-mode quantization for multiple emitters

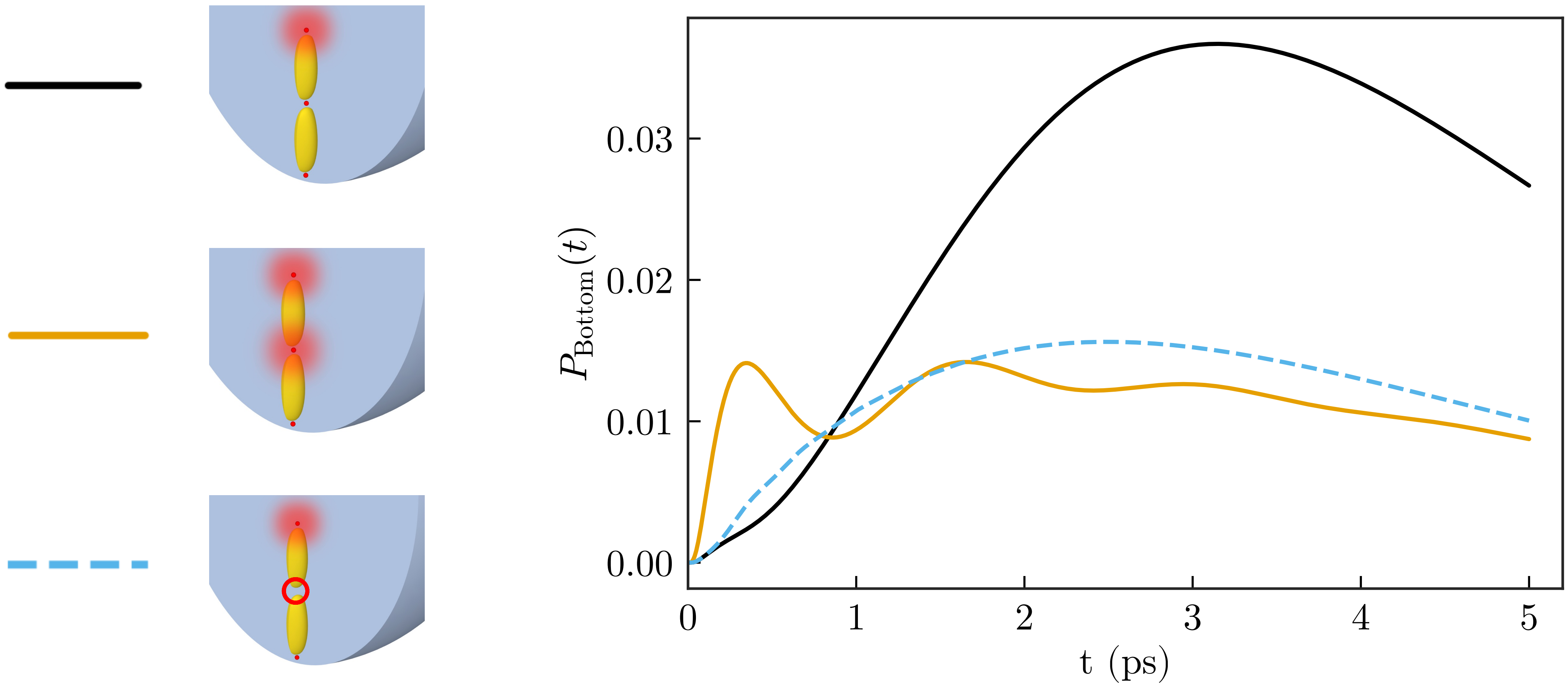




Fitting again gives a few-mode model that reproduces dynamics fully:



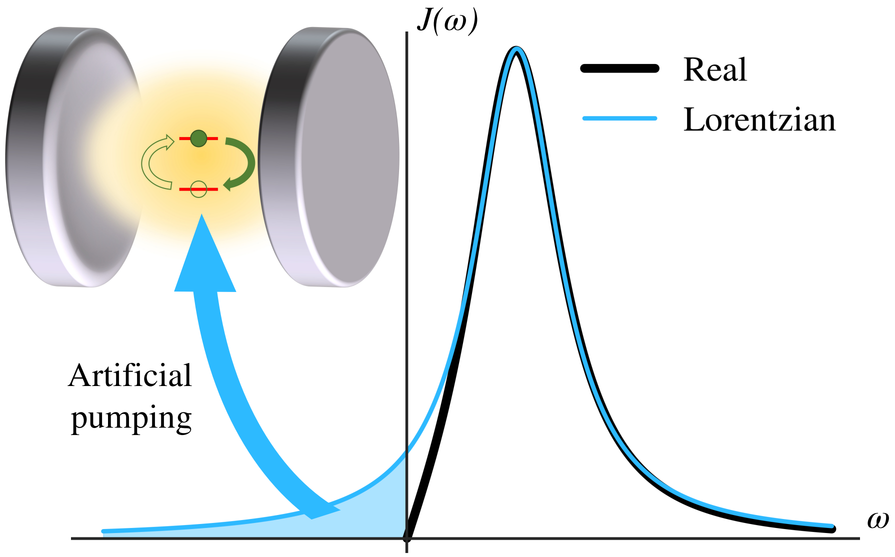
Control of energy transfer



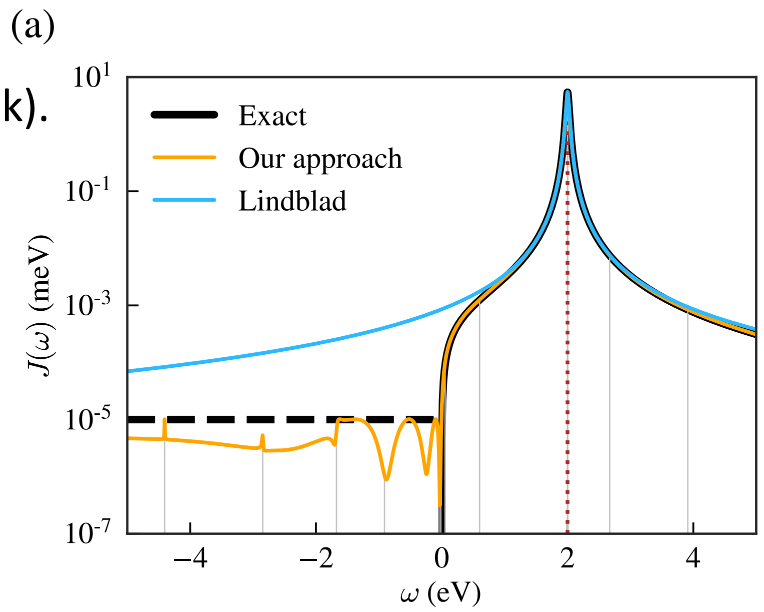
Quantum state of Gap emitter controls energy transfer from Top to Bottom emitter.

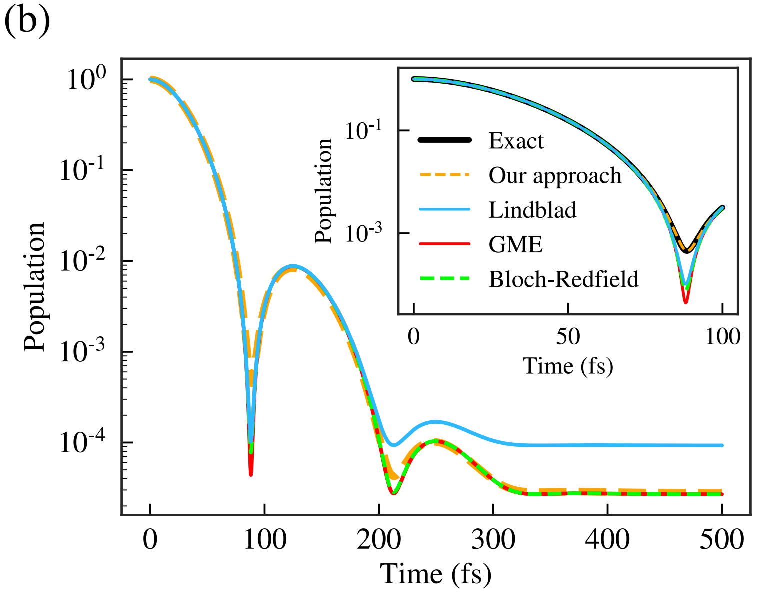
New perspective on ultrastrong coupling

Well-known in ultrastrongly coupled systems: Cannot use “normal” Lindblad term because it induces artificial pumping, e.g., from the ground state (e.g., Mikołaj’s talk). Can the mapping provide a new perspective on this?



Use **interference** of coupled modes to **suppress negative frequencies** → “standard” Lindblad master equation in the ultrastrong coupling regime!





Yes! Cavity mode + Lindblad gives a Lorentzian spectral density, spanning the whole real axis.
“Real” spectral densities have only positive frequencies!
→ **negative frequency components introduce artificial pumping**
(emission of negative-frequency photons to the bath
= absorption of photons from the bath)

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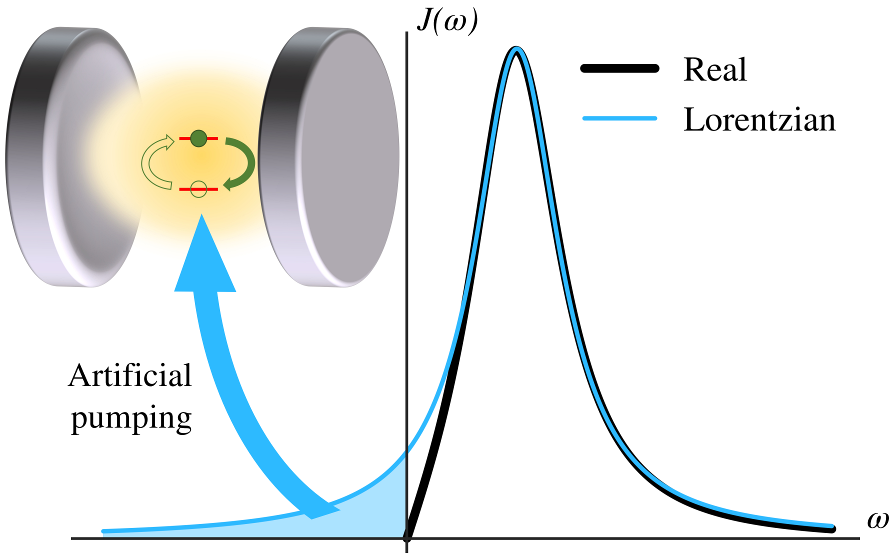

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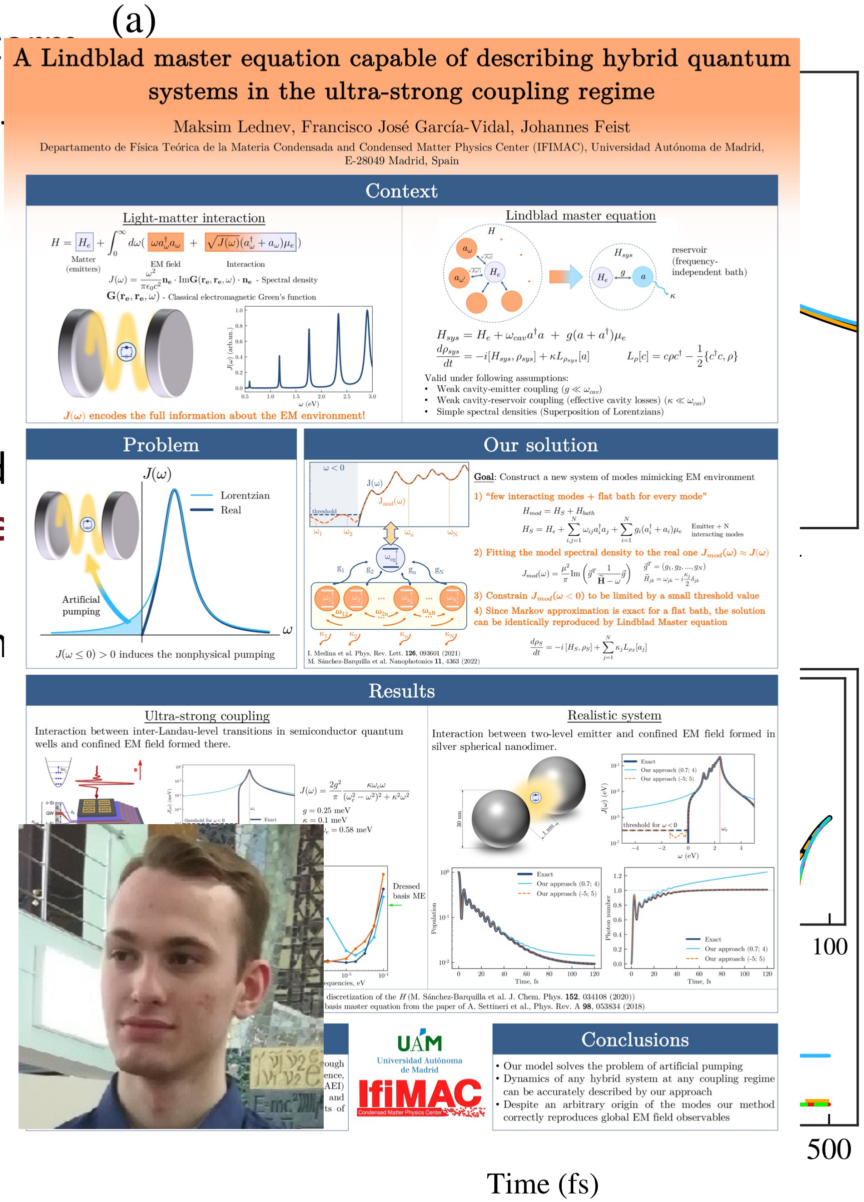

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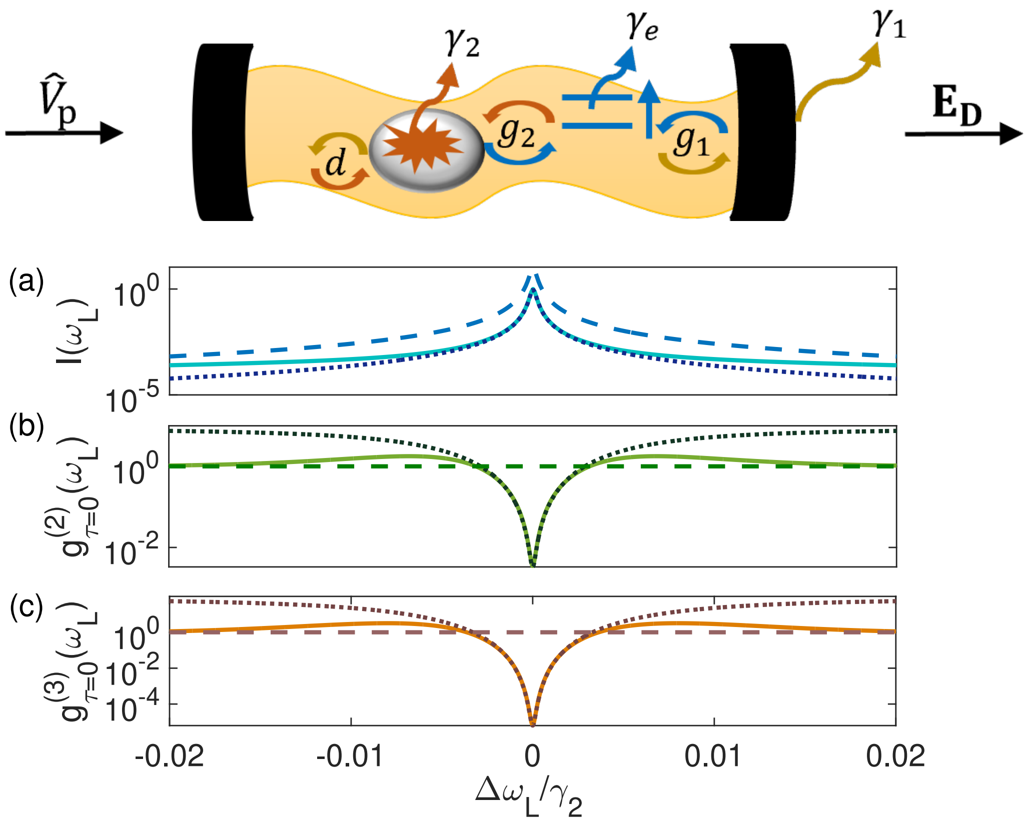
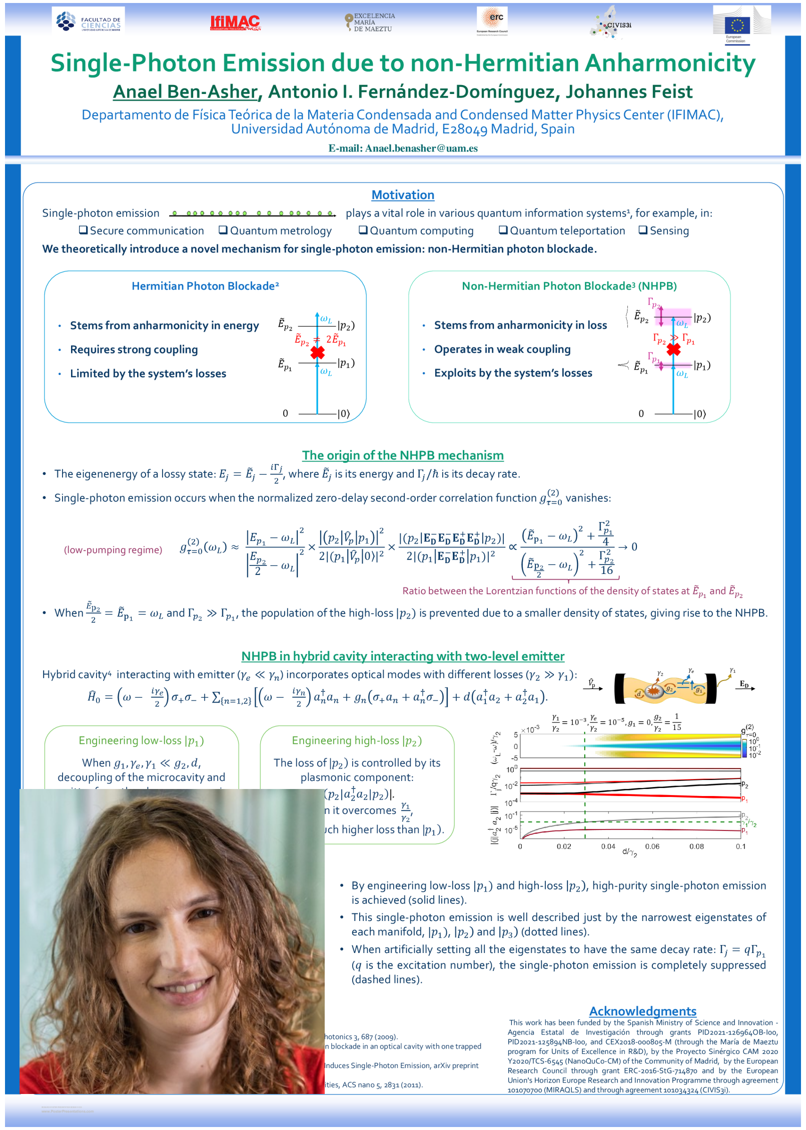
Engineering of non-Hermitian systems

Can engineer nonlinearity of the losses in a hybrid system to obtain efficient single-photon emission through non-Hermitian photon blockade.

Requires non-commutativity of losses & couplings!

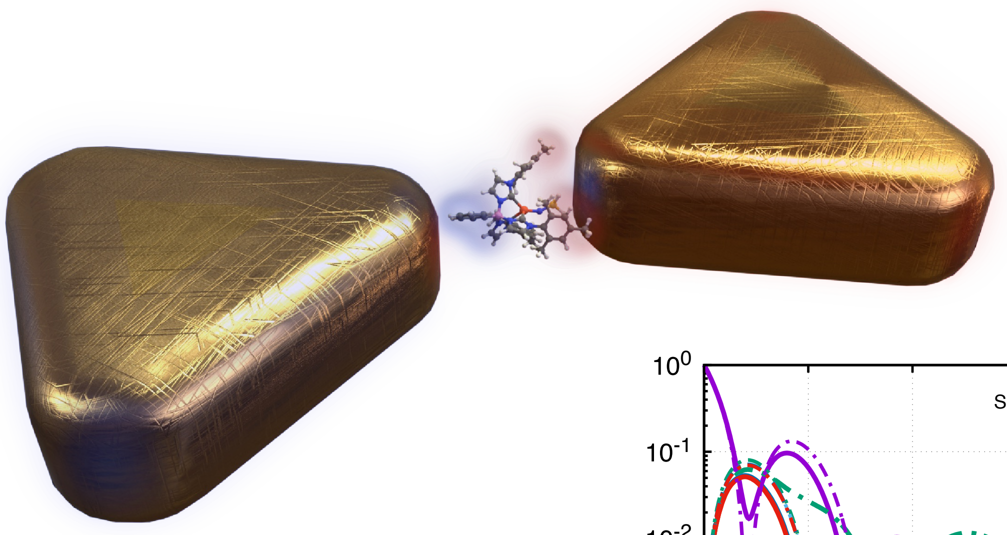
A. Ben-Asher et al., arXiv:2212.06307

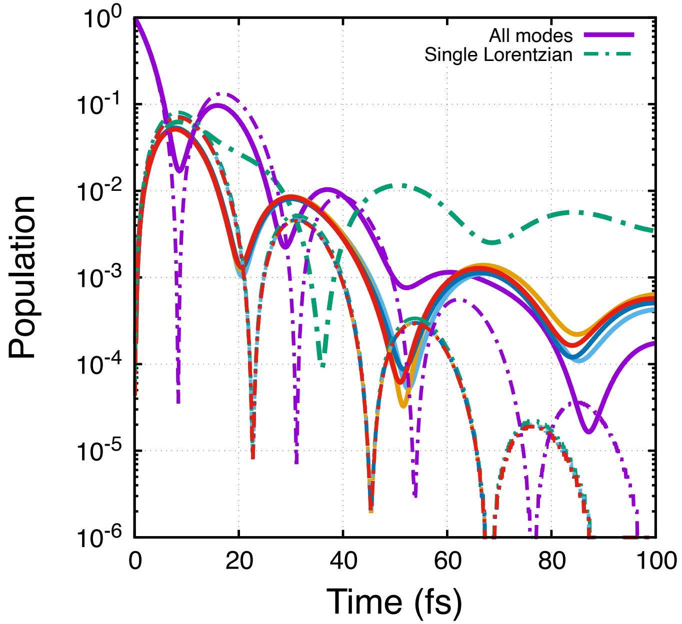


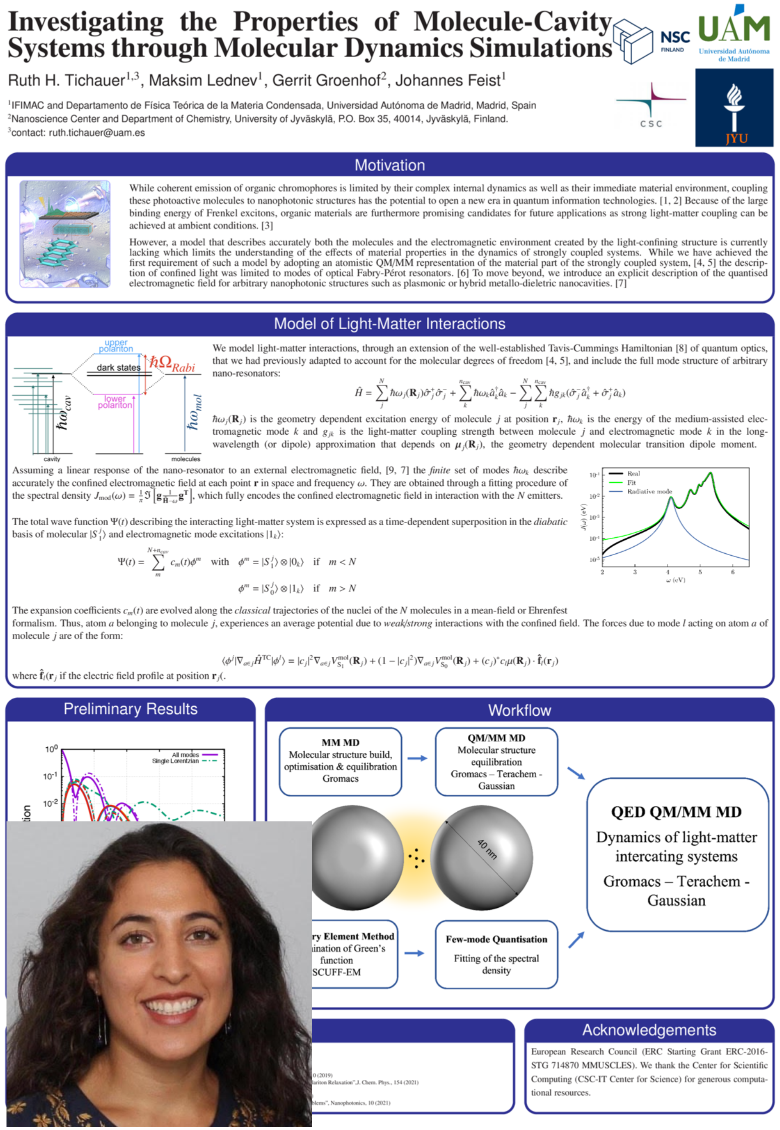


Combine few-mode quantization with molecular dynamics

We can now do molecular dynamics simulations with the full complexity of the molecules & the full complexity of the nanophotonic system in an efficient manner.







Summary

- **Novel few-mode quantization method for nanophotonic systems**
  - Efficient & simple mapping between nanophotonics and quantum optics
  - Mode interactions are an intrinsic feature of nanophotonic systems
  - Naturally non-Hermitian
  - Can deal with multiple emitters
  - Not restricted to any particular system or coupling regime
- **Example applications**
  - Ultrastrong coupling
  - Nonclassical light emission. R. Sáez-Blázquez et al., Nano Lett. 22, 2365 (2022), A. Ben-Asher et al., arXiv:2212.06307
  - Molecular dynamics in nanophotonic structures
- **Outlook:**
  - Exploit mapping to provide **new directions for quantum optics & nanophotonics**
  - **Further theory developments**
    - Input-output theory
    - Non-dipole interactions

Team & Acknowledgements

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Open-source codes:
https://github.com/jfeist