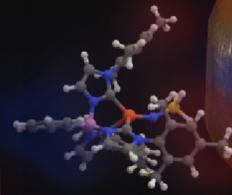


Mapping between Quantum & Nano-Photonics

Johannes Feist

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Universidad Autónoma de Madrid



Quick advertisement

<https://mmuscles.eu/tools>

Emitter properties

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

Cavity properties

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

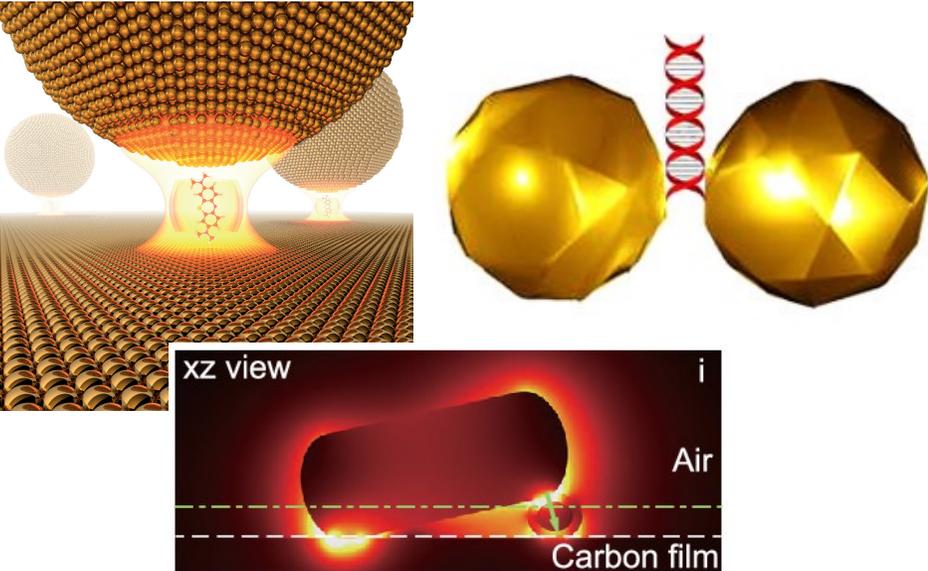
Emitter-cavity coupling

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

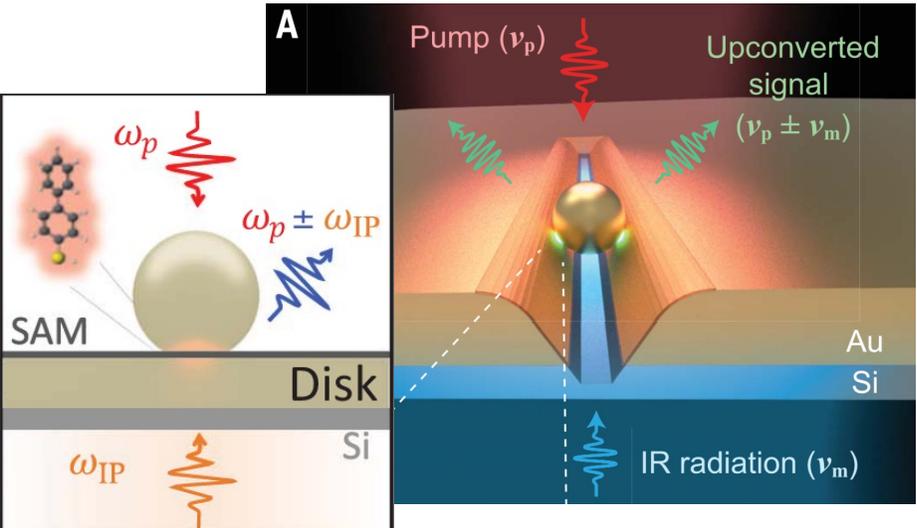
Plasmonic and hybrid nanocavities provide strong light-matter interactions

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.)?

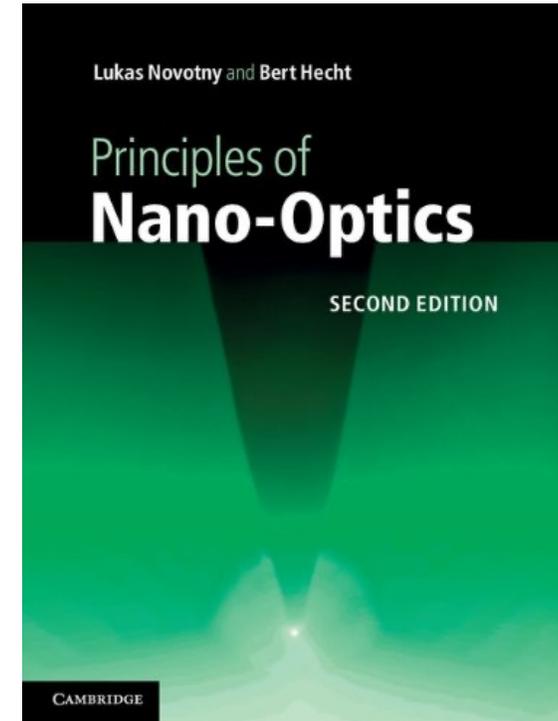
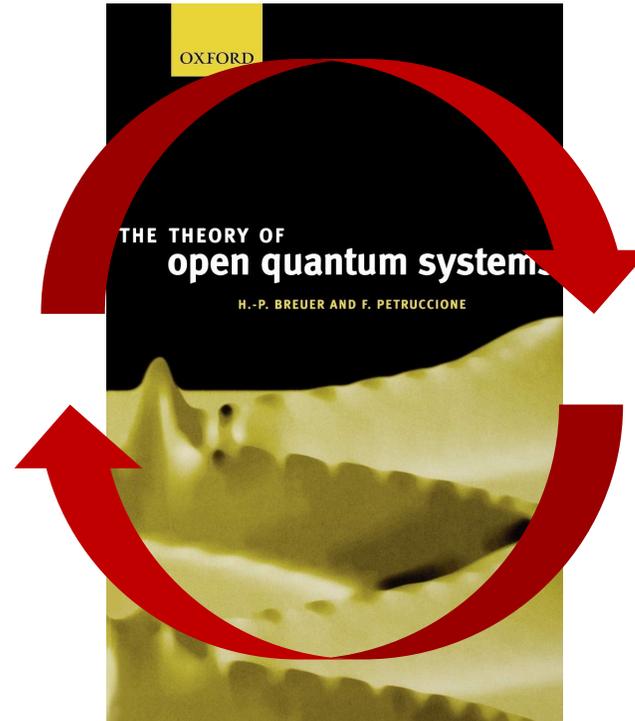
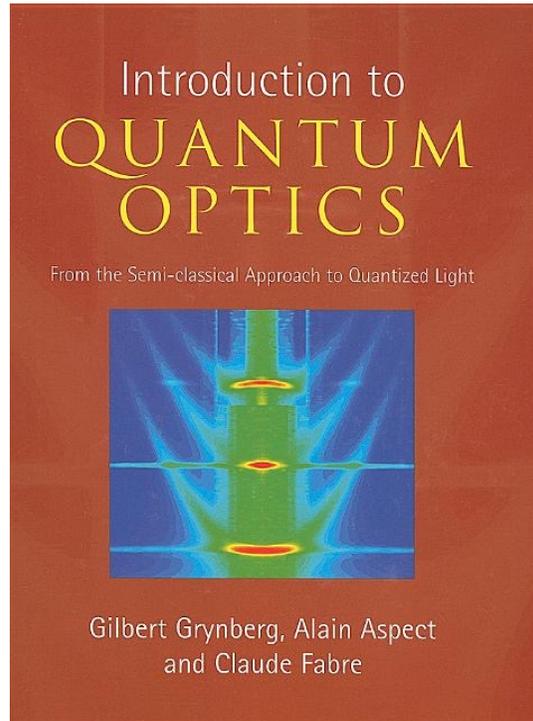
Descriptions of electromagnetic fields

“Traditional” Quantum Optics

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

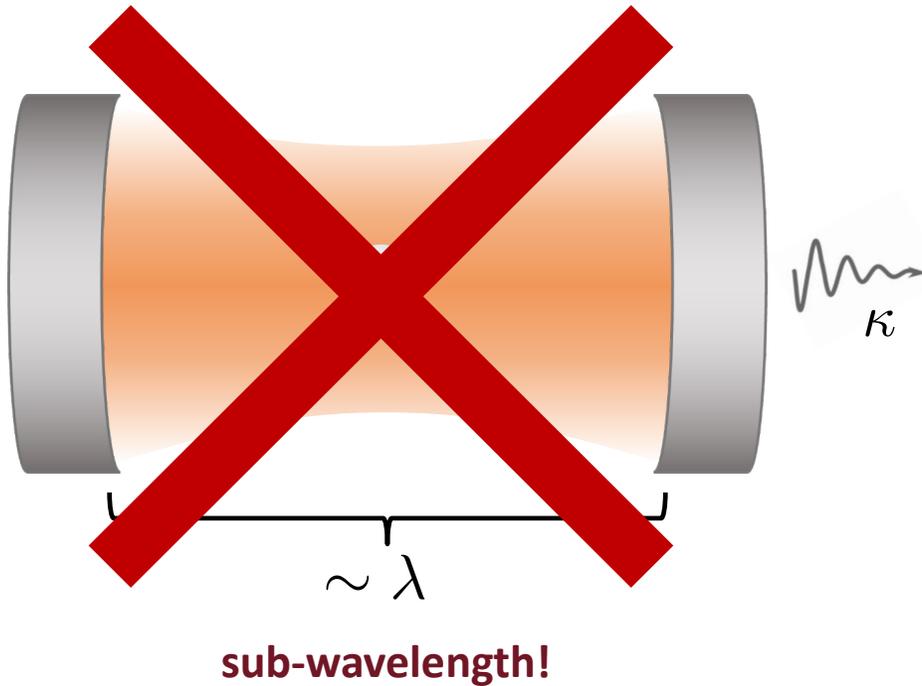
“Traditional” Nanophotonics

- 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”?

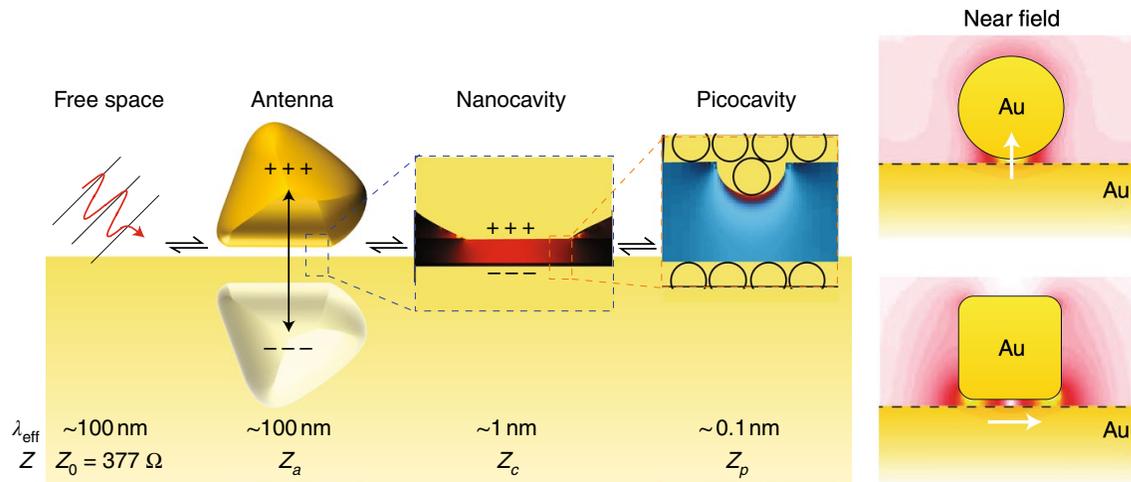


“Traditional” quantum optics / cavity QED

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

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”?



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:

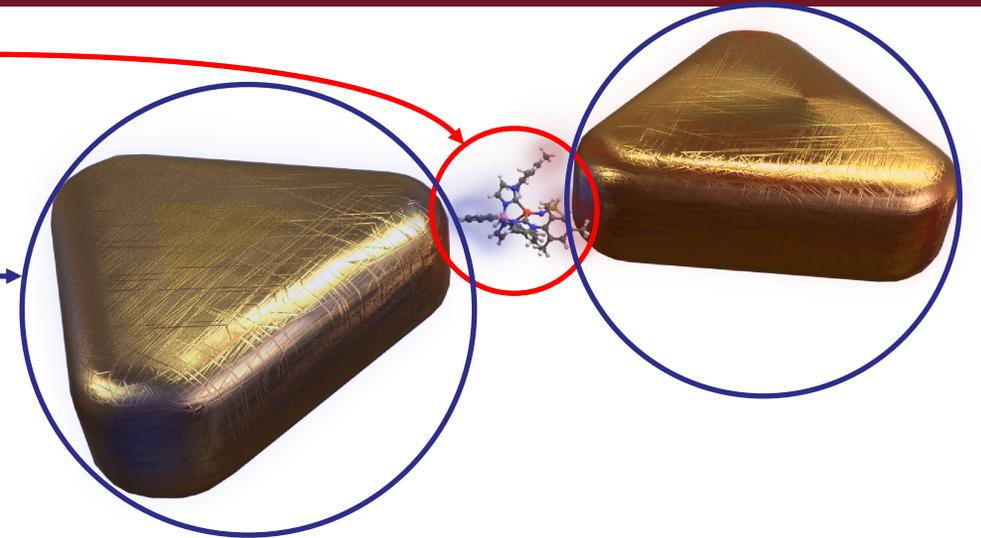
Dominant interaction is with charges in the material (Coulomb force / longitudinal fields), not with propagating (transverse) fields → fundamental interaction is $\vec{\mu} \cdot \vec{E}$, not $\vec{p} \cdot \vec{A}$ and \vec{A}^2 (“an electron by any other name is still an electron”)

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 \rightarrow solutions expressed through Green’s functions)

c) Quantize harmonic oscillators (promote variables to operators)

\rightarrow **quantized medium-assisted EM field**: infinite collection of bosonic modes (quantum harmonic oscillators)

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

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

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)**

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

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

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

$J(\omega)$ fully characterizes the “cavity”

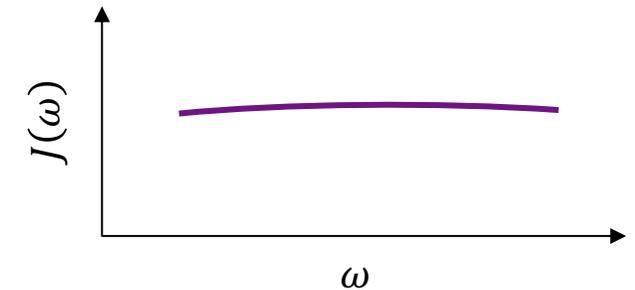
→ two systems with the same spectral density are indistinguishable for the emitter

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

Markovian $J(\omega)$

Spectrally flat

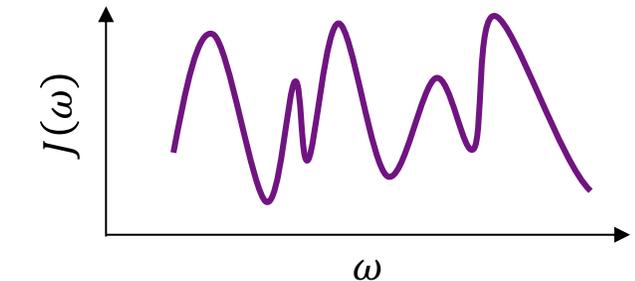
“Memoryless” environment



Non-Markovian $J(\omega)$

Arbitrary densities

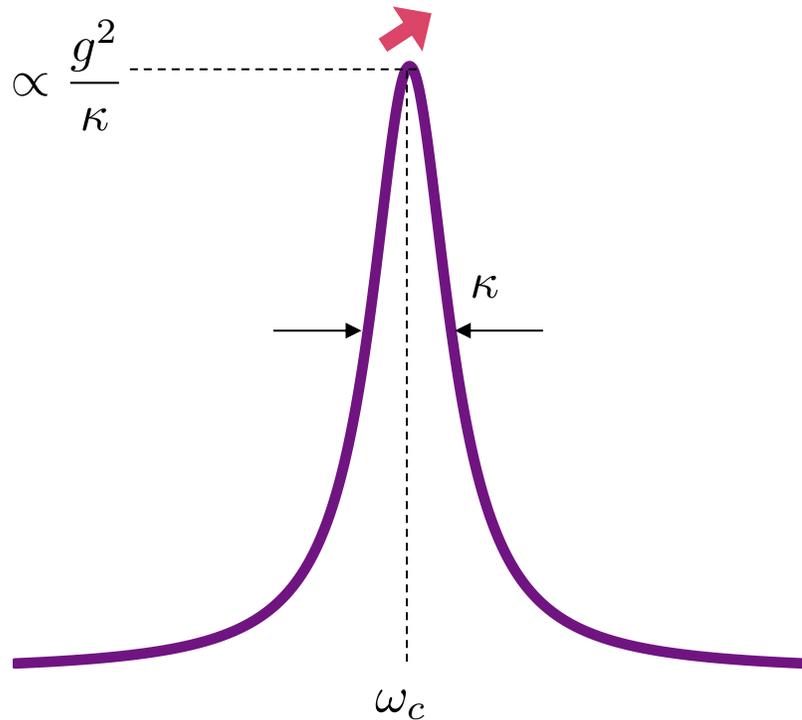
Dynamics depend on previous times



Formulation used here: “Emitter-centered modes”

Feist, Fernández-Domínguez, García-Vidal, Nanophotonics 10, 477 (2020)

Lorentzian spectral density



Lorentzian spectral density (“resonance”):

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

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

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

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

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

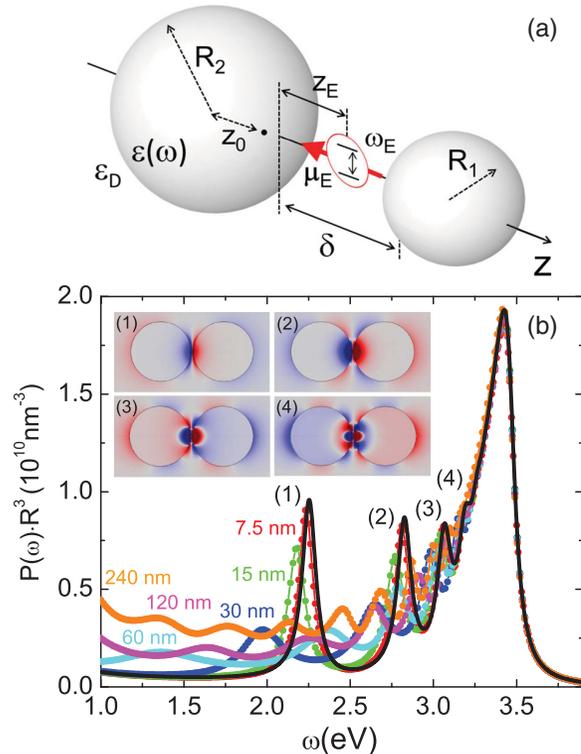
Multiple lossy modes: **sum** of Lorentzians

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

Nanophotonic spectral densities

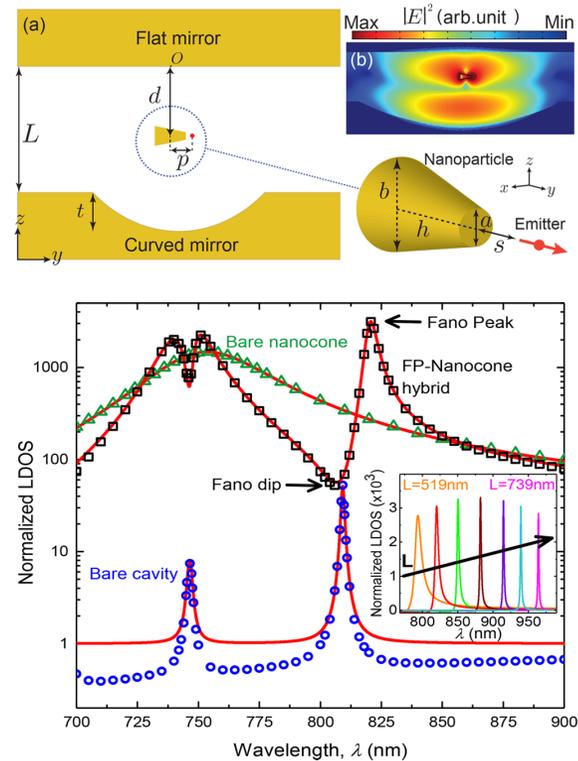
Spectral densities of some example systems

Plasmonic nanogap antenna



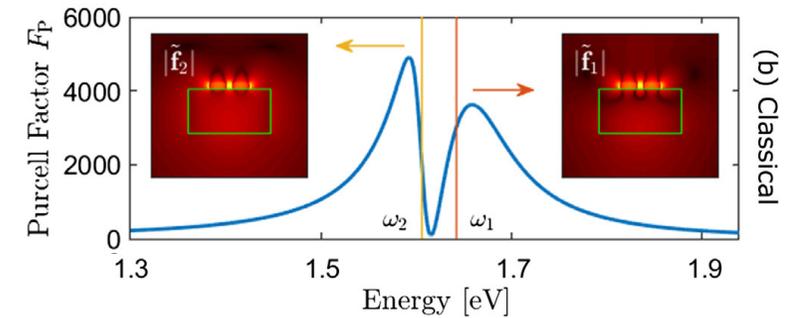
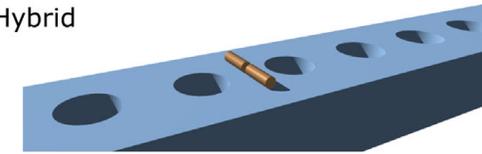
R.-Q. Li et al., Phys. Rev. Lett. **117**, 107401 (2016)

Hybrid plasmonic – dielectric cavities



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

(a) Hybrid



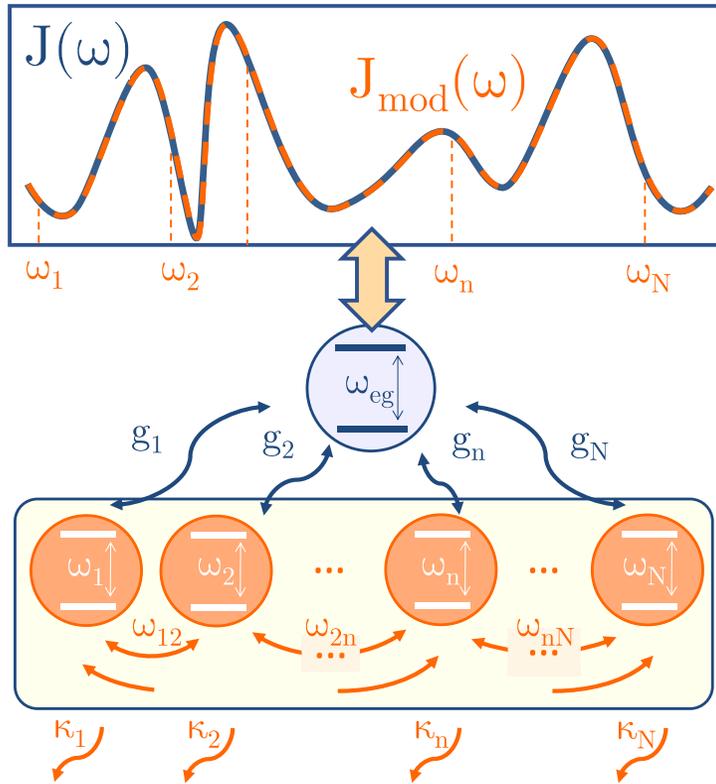
S. Franke et al., Phys. Rev. Lett. **122**, 213901 (2019)

“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:

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

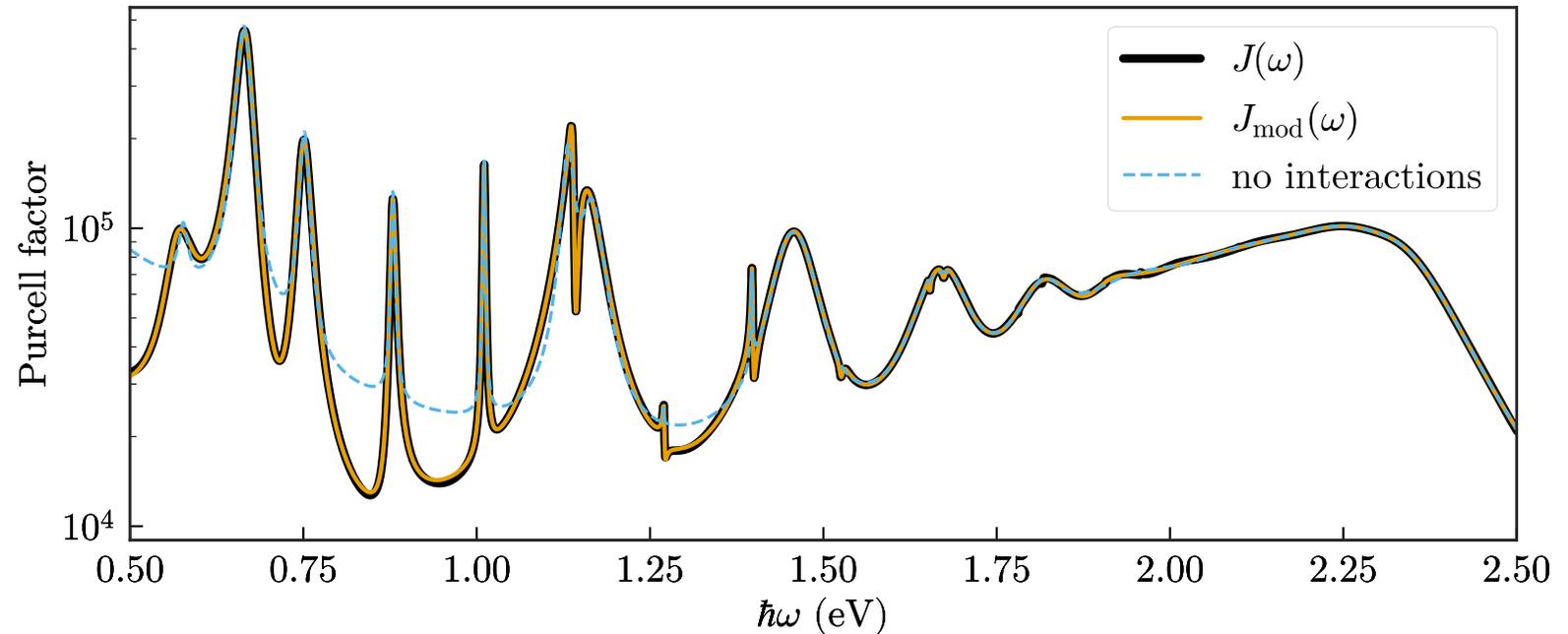
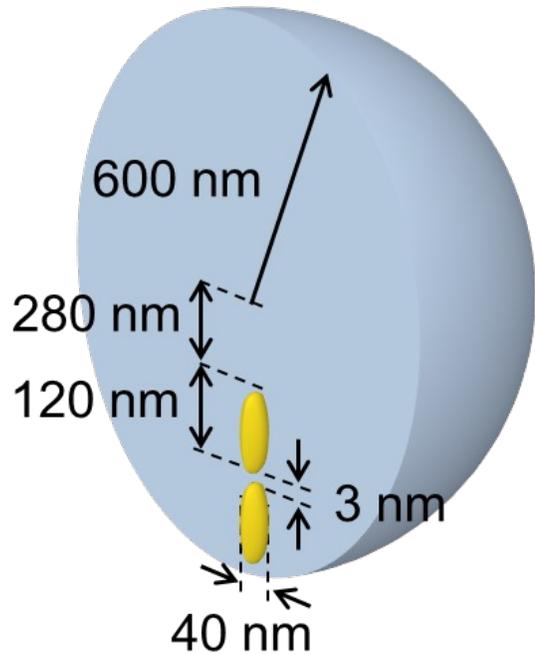
Interactions give much more flexibility for describing spectral density!

“Just” need to find model system that reproduces nanophotonic $J(\omega)$

→ fitting to obtain parameters – only approximation in the method!

Few-mode quantization of arbitrary spectral densities

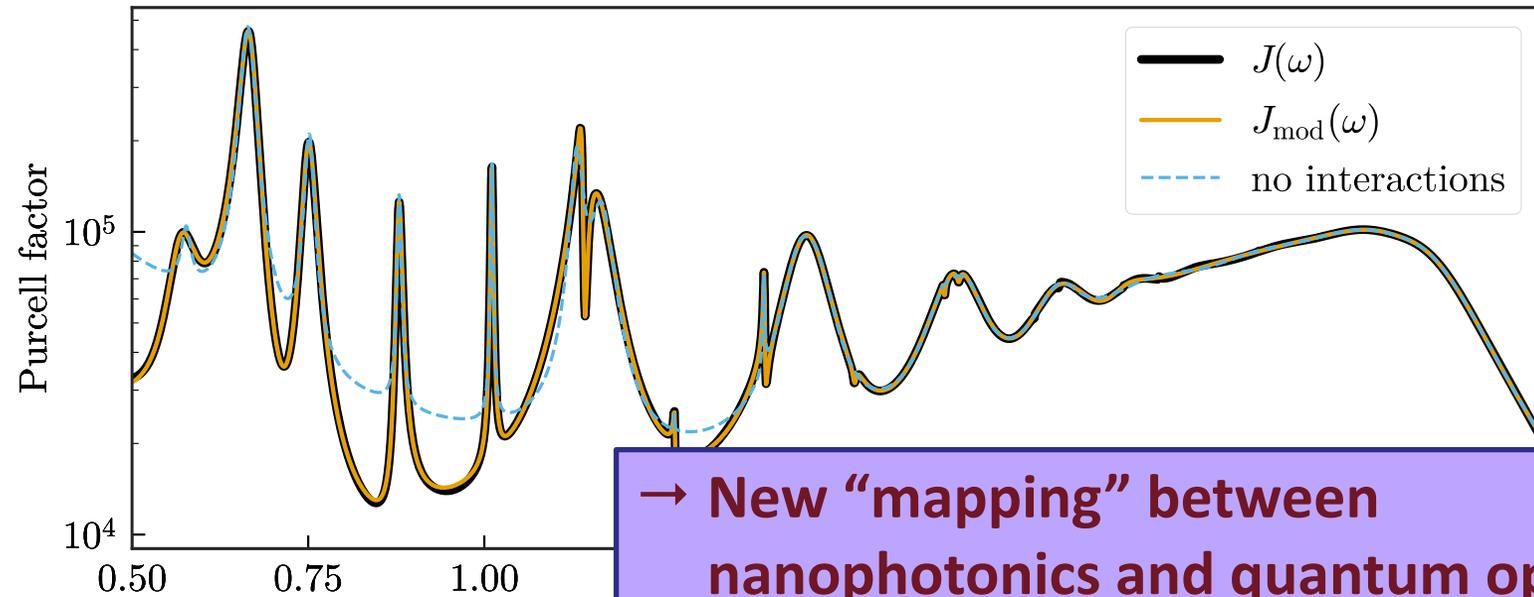
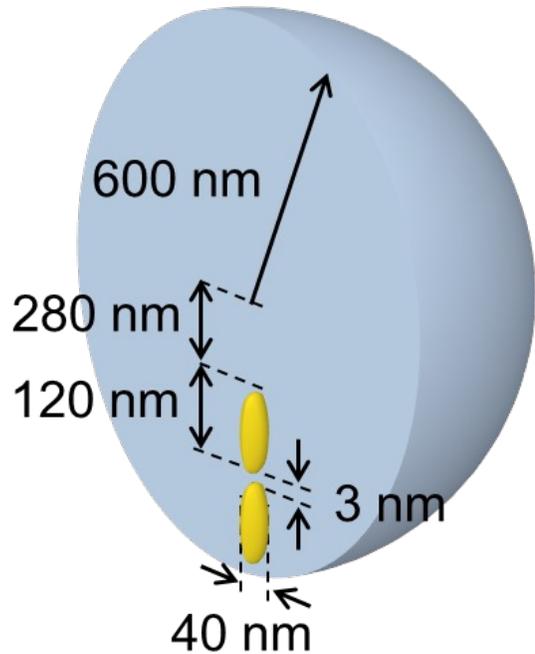
Hybrid test system: high-dielectric microsphere with ellipsoidal plasmonic nanoantenna
Complex spectral density with several interference (Fano-like) features.



- **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.

Few-mode quantization of arbitrary spectral densities

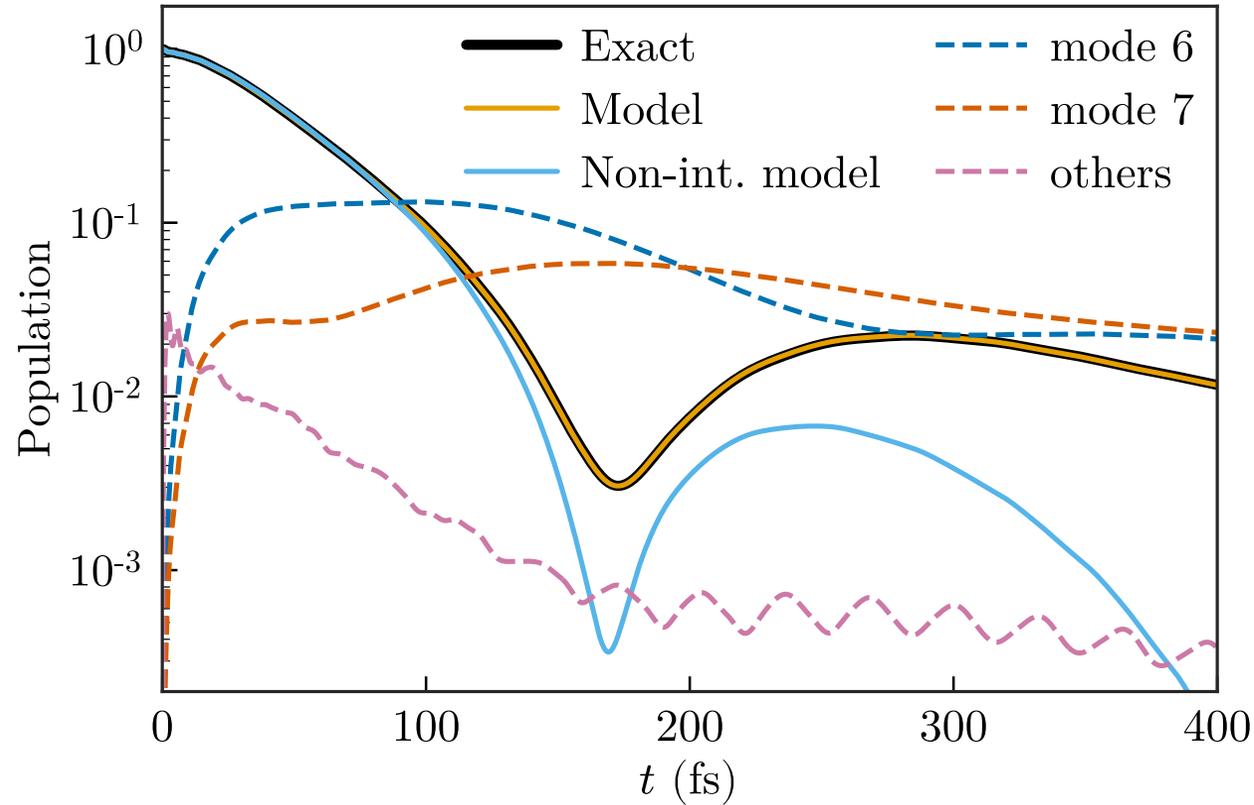
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- **Almost perfect fit of full spectrum using 20 modes!**
(Quasi-normal modes: orders of magnitude more in the spectrum)
- Fit with **non-interacting** modes (sum of Lorentzians) **cannot** capture the complex interference structure and overestimates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

→ 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

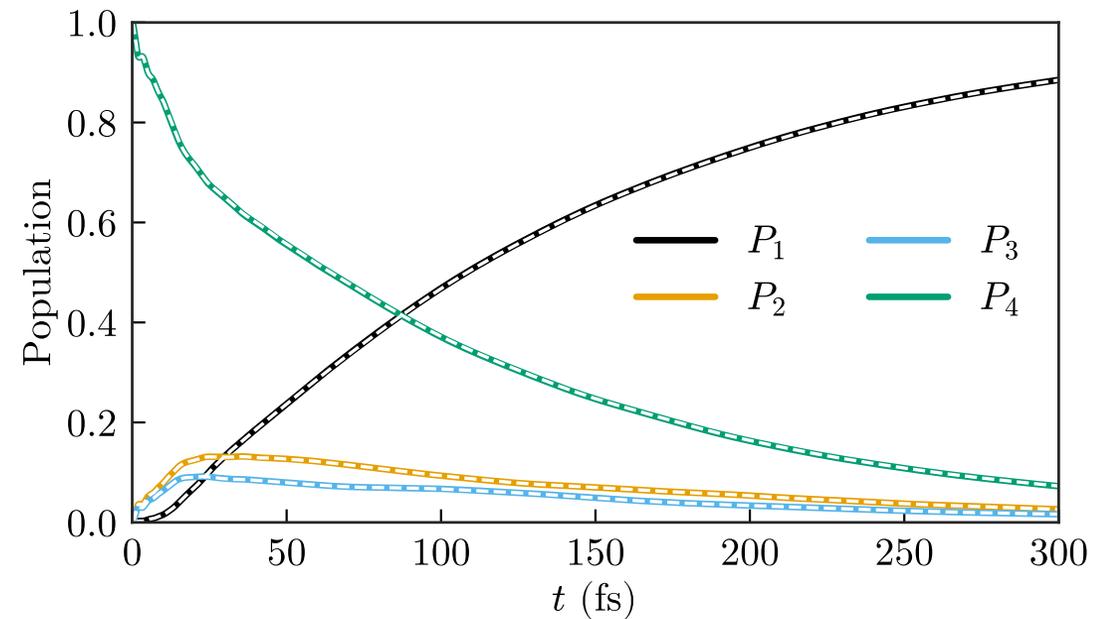
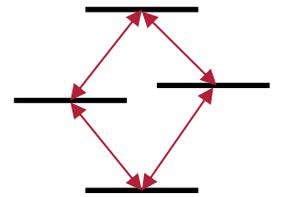
Few-mode quantization of arbitrary spectral densities



Spontaneous emission dynamics confirm correctness of model.

Works for any coupling regime (weak or strong).

Emitter can have any internal structure.



Few-mode quantization of arbitrary spectral densities

Have access to spatially resolved EM field

Spontaneous emission: no coherent field!

→ Need quantum calculation

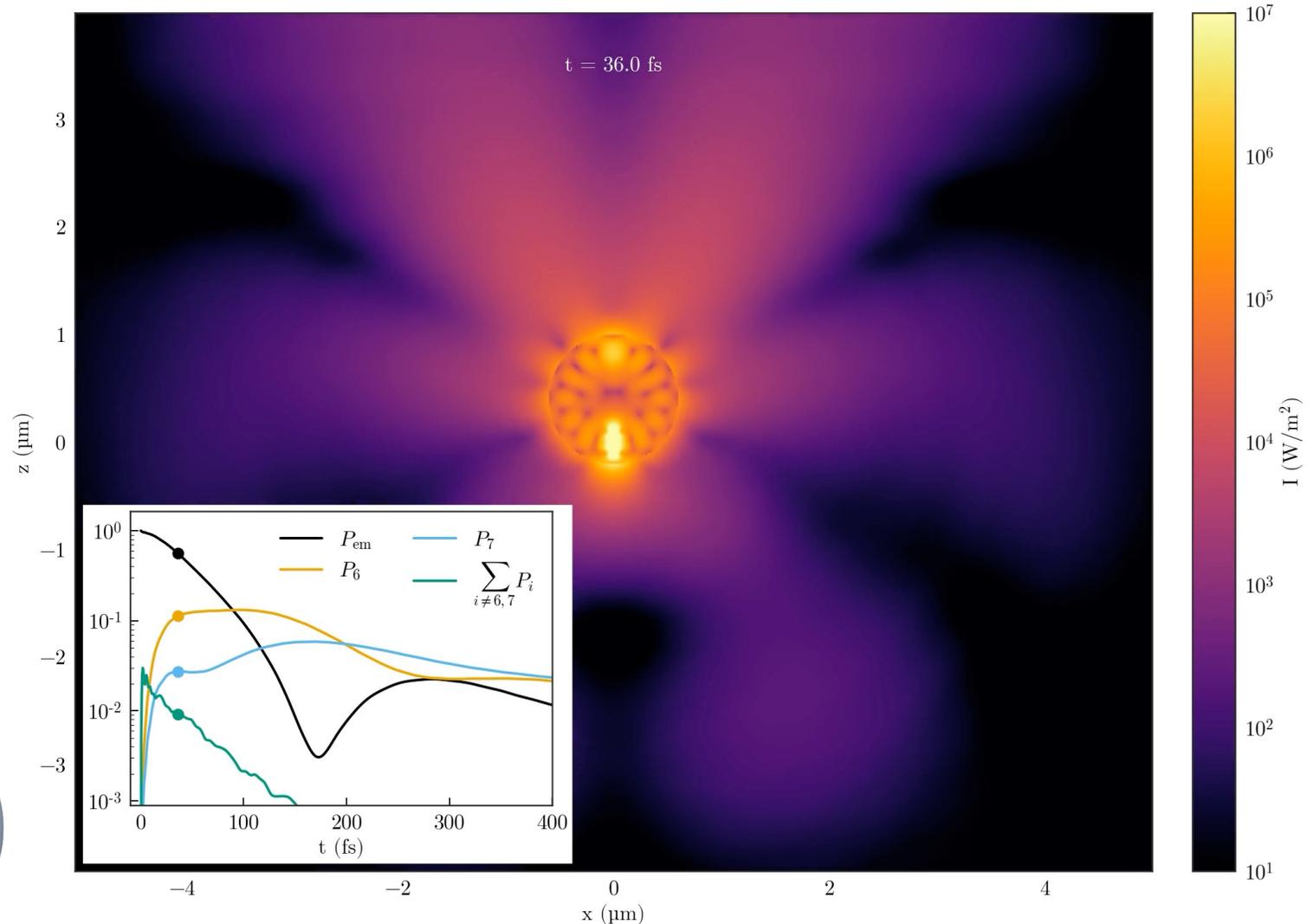
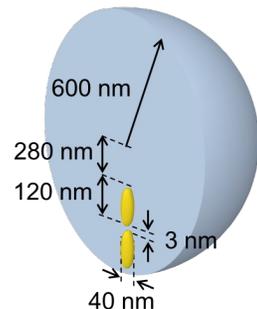
$$\langle \vec{E}(\vec{r}) \rangle = 0$$

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?



Extension to multiple emitters

- **Macroscopic QED:**

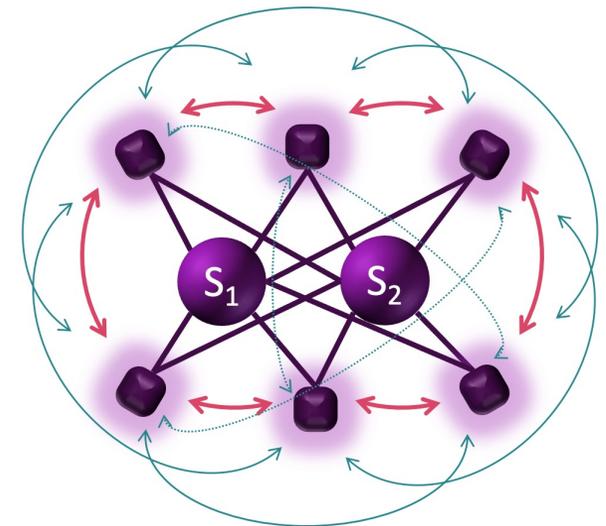
Light-matter interaction for multiple emitters is **fully characterized** by a **generalized spectral density** describing EM-mediated interaction between emitters n and m :

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

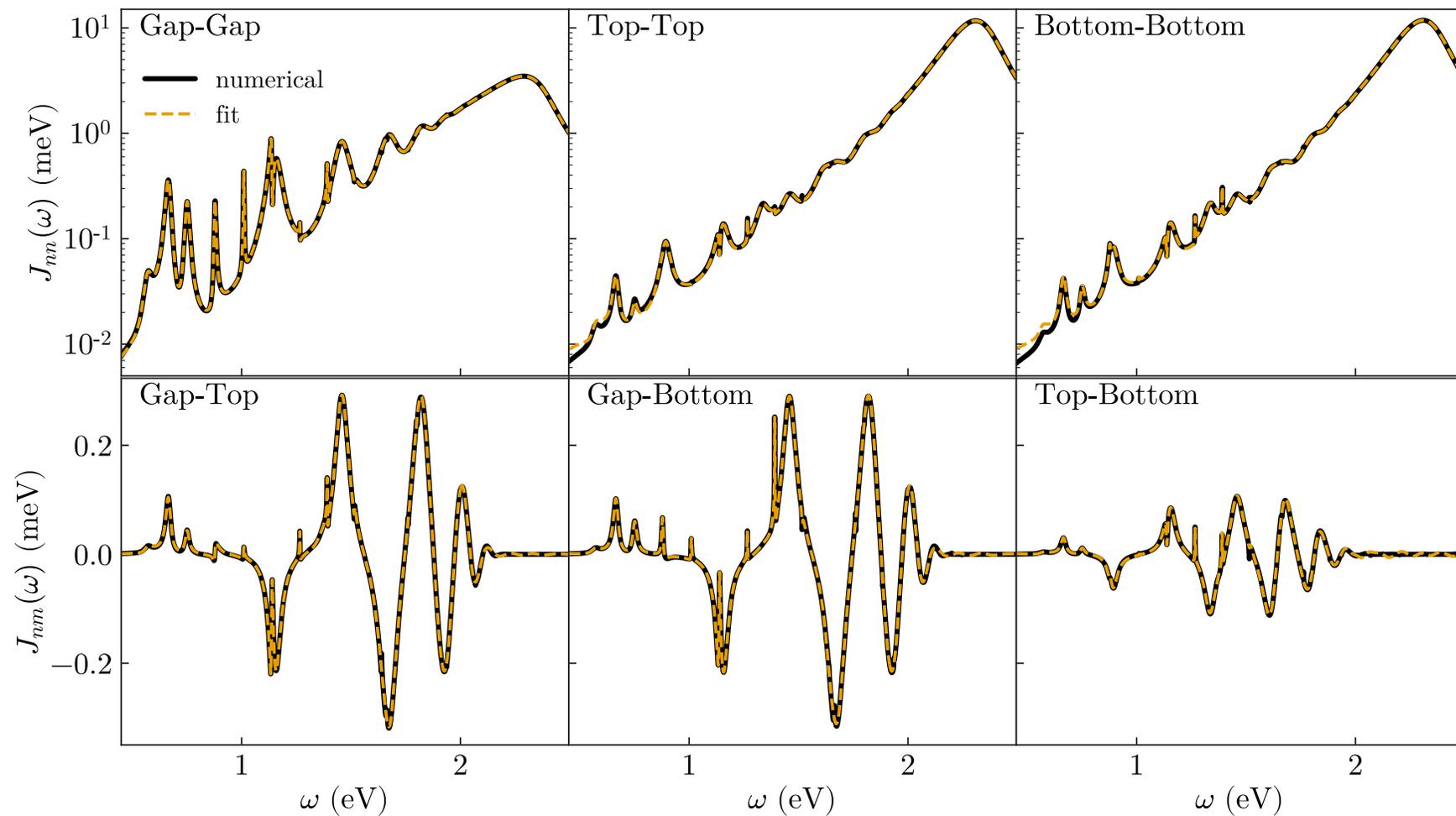
- **Few-mode quantization model easily extended to that case**
→ **coupling vector becomes a matrix**

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

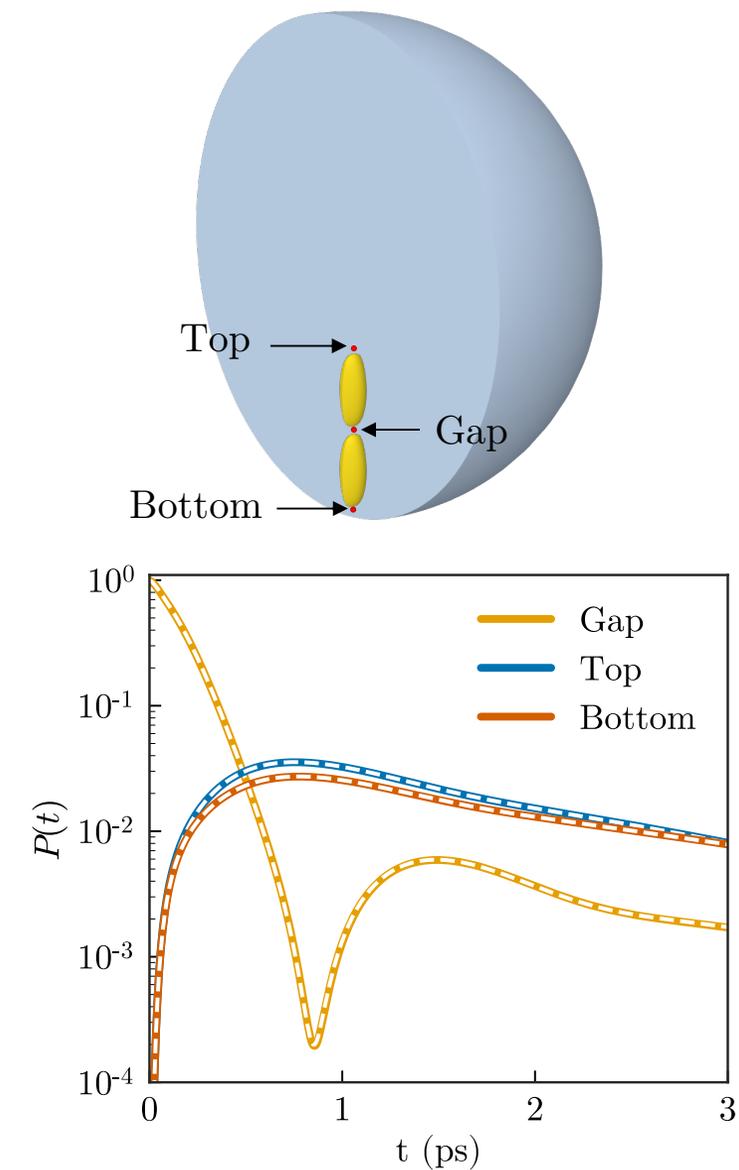
$$\mathcal{J}_{\text{mod}}(\omega) = \frac{1}{\pi} \mathbf{g} \text{Im} \left[\frac{1}{\tilde{\mathbf{H}} - \omega} \right] \mathbf{g}^T$$



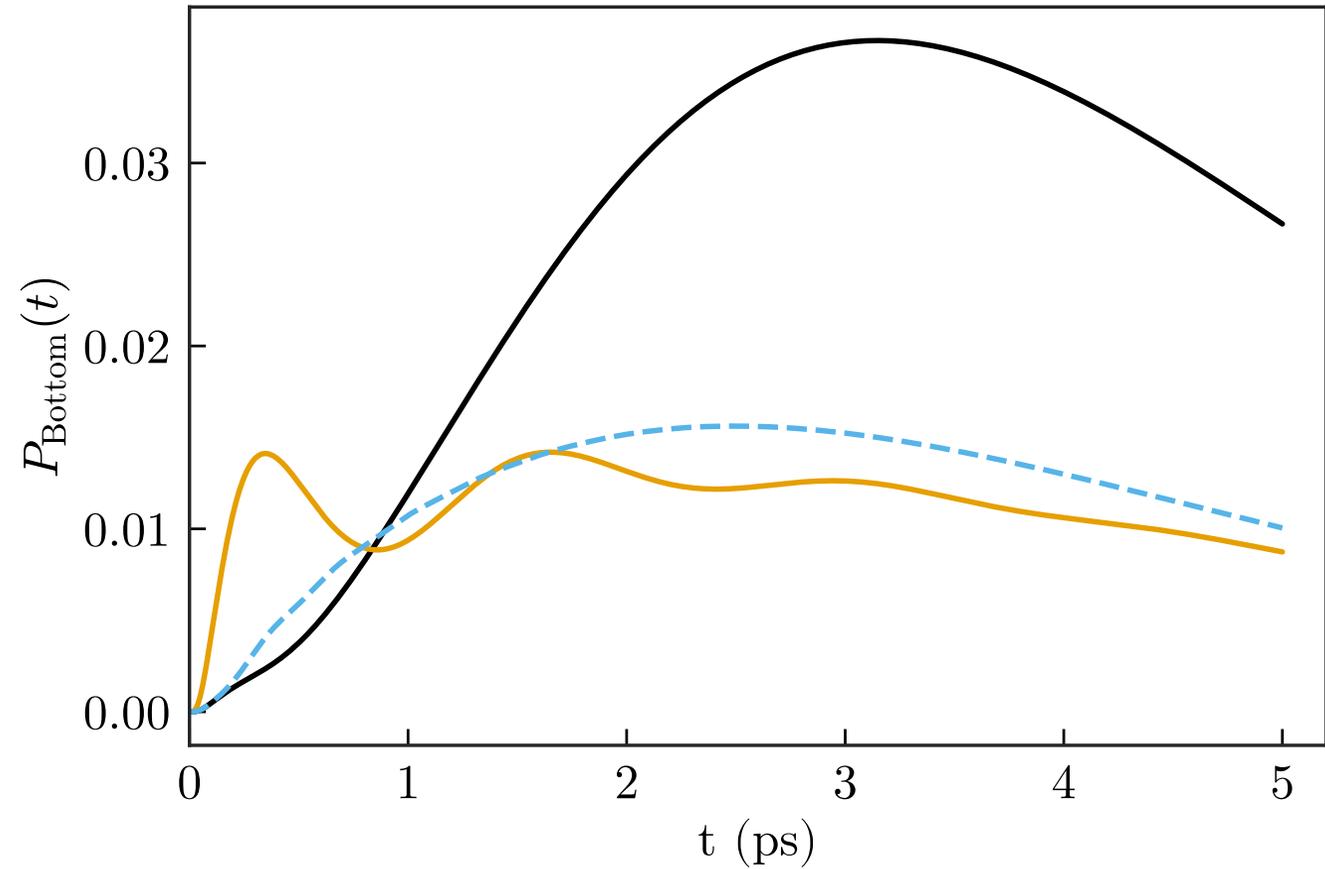
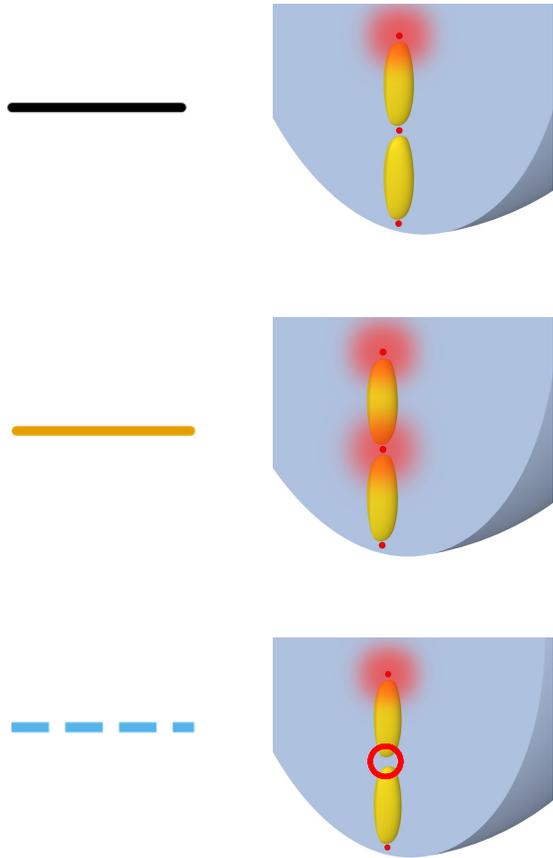
Few-mode quantization for multiple emitters



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



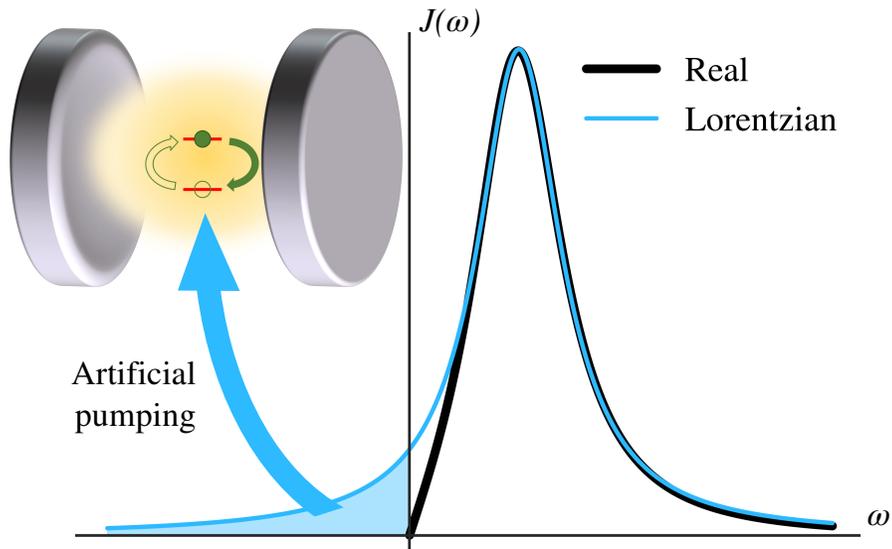
Control of energy transfer



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?

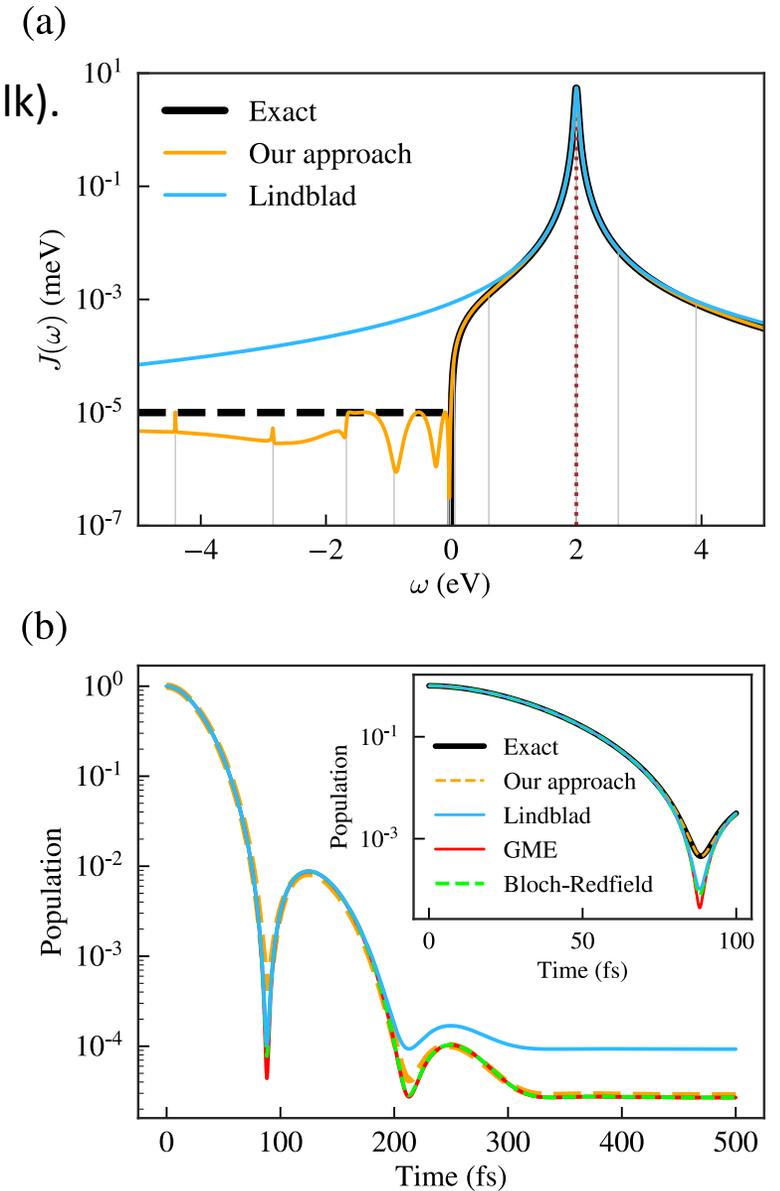


Use **interference** of coupled modes to **suppress negative frequencies** → “standard” Lindblad master equation in the ultrastrong coupling regime!

Yes! Cavity mode + Lindblad gives a Lorentzian spectral density, spanning the whole real axis.

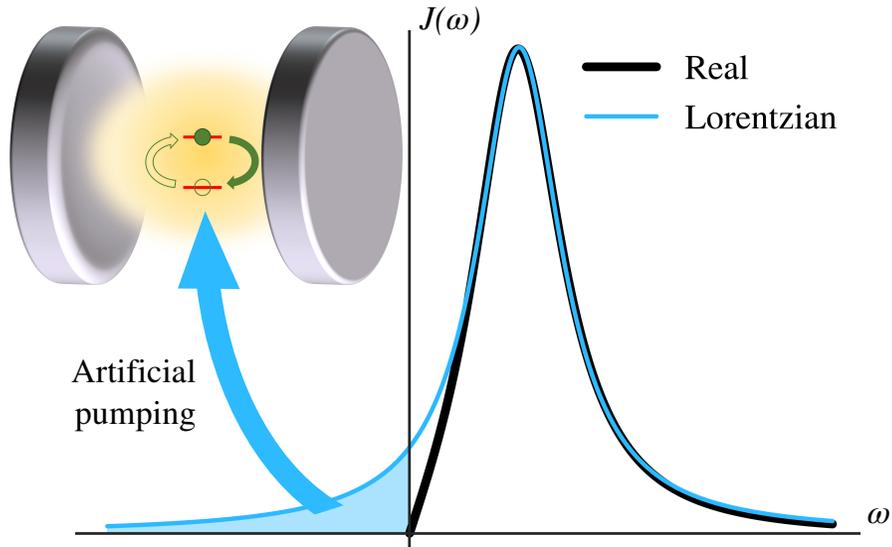
“Real” spectral densities have only positive frequencies!

→ **negative frequency components introduce artificial pumping** (emission of negative-frequency photons to the bath = absorption of photons from the bath)



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“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)

(a)

A Lindblad master equation capable of describing hybrid quantum systems in the ultra-strong coupling regime

Maksim Lednev, Francisco José García-Vidal, Johannes Feist

Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, E-28049 Madrid, Spain

Context

Light-matter interaction

Matrix EM field Interaction

$$H = H_c + \int d\omega \left(\omega a^\dagger a c_\omega + \sqrt{J(\omega)} (a^\dagger + a) \mu c_\omega \right)$$

$J(\omega) = \frac{1}{4\pi\epsilon_0} \text{Tr} \left[\mathbf{G}(\mathbf{r}_e, \mathbf{r}_e, \omega) \cdot \mathbf{D}_e \cdot \text{Spectral density} \right]$

$\mathbf{G}(\mathbf{r}_e, \mathbf{r}_e, \omega)$ - Classical electromagnetic Green's function

$J(\omega)$ encodes the full information about the EM environment!

Lindblad master equation

reservoir (frequency-independent bath)

$$H_{app} = H_c + \omega_{cav} a^\dagger a + g(a + a^\dagger) \mu c_e$$

$$\frac{d\rho_{app}}{dt} = -i[H_{app}, \rho_{app}] + \kappa L_{app}[\rho_{app}] \quad L_{app}[\rho] = c\rho c^\dagger - \frac{1}{2}\{c^\dagger c, \rho\}$$

Valid under following assumptions:

- Weak cavity-emitter coupling ($g \ll \omega_{cav}$)
- Weak cavity-reservoir coupling (effective cavity losses) ($\kappa \ll \omega_{cav}$)
- Simple spectral densities (Superposition of Lorentzians)

Problem

$J(\omega) = J_{Lorentzian} + J_{Real}$

$J(\omega \leq 0) > 0$ induces the nonphysical pumping

Our solution

Goal: Construct a new system of modes mimicking EM environment

- “few interacting modes + flat bath for every mode”
- Fitting the model spectral density to the real one $J_{mod}(\omega) = J(\omega)$
- Constrain $J_{mod}(\omega < 0)$ to be limited by a small threshold value
- Since Markov approximation is exact for a flat bath, the solution can be identically reproduced by Lindblad Master equation

$\frac{d\rho_{mod}}{dt} = -i[H_{mod}, \rho_{mod}] + \sum_{j=1}^N \kappa_j L_{mod}[\rho_{mod}]$

Results

Ultra-strong coupling

Interaction between inter-Landau-level transitions in semiconductor quantum wells and confined EM field formed there.

$J(\omega) = \frac{2g^2}{\pi} \frac{\text{Re}(\omega)}{(\omega^2 - \omega_{cav}^2)^2 + \kappa^2 \omega^2}$

$g = 0.25$ meV
 $\kappa = 0.18$ meV

Realistic system

Interaction between two-level emitter and confined EM field formed in silver spherical nanodimer.

discretization of the H (M. Sánchez-Barquilla et al. J. Chem. Phys. **152**, 034108 (2020))
Lindblad master equation from the paper of A. Settineri et al., Phys. Rev. A **98**, 053854 (2018)

Conclusions

- Our model solves the problem of artificial pumping
- Dynamics of any hybrid system at any coupling regime can be accurately described by our approach
- Despite an arbitrary origin of the modes our method correctly reproduces global EM field observables



Time (fs)



100

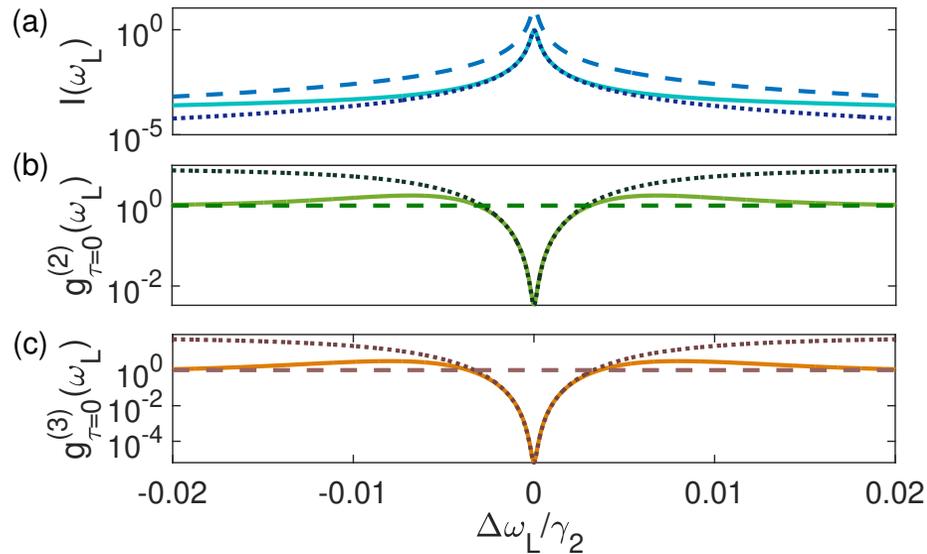
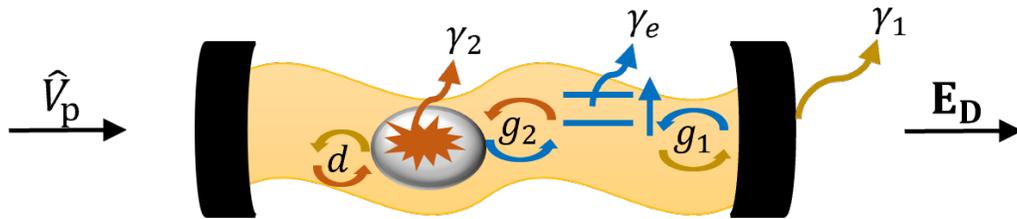
500

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



Single-Photon Emission due to non-Hermitian Anharmonicity
Anael Ben-Asher, Antonio I. Fernández-Domínguez, Johannes Feist
 Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC),
 Universidad Autónoma de Madrid, E28049 Madrid, Spain
 E-mail: Anael.benasher@uam.es

Motivation

Single-photon emission plays a vital role in various quantum information systems¹, for example, in:

- Secure communication
- Quantum metrology
- Quantum computing
- Quantum teleportation
- Sensing

We theoretically introduce a novel mechanism for single-photon emission: **non-Hermitian photon blockade**.

Hermitian Photon Blockade²

- Stems from anharmonicity in energy
- Requires strong coupling
- Limited by the system's losses

Non-Hermitian Photon Blockade³ (NHPB)

- Stems from anharmonicity in loss
- Operates in weak coupling
- Exploits by the system's losses

The origin of the NHPB mechanism

- The eigenenergy of a lossy state: $E_j = \tilde{E}_j - \frac{i\Gamma_j}{2}$, where \tilde{E}_j is its energy and Γ_j/\hbar is its decay rate.
- Single-photon emission occurs when the normalized zero-delay second-order correlation function $g_{\tau=0}^{(2)}$ vanishes:

$$\text{(low-pumping regime)} \quad g_{\tau=0}^{(2)}(\omega_L) \approx \frac{|E_{p_1} - \omega_L|^2}{|E_{p_2} - \omega_L|^2} \times \frac{|(p_2|V_p|p_1)|^2}{2|(p_1|V_p|0)|^2} \times \frac{|(p_2|E_0 E_0^\dagger E_0^\dagger E_0|p_2)|^2}{2|(p_1|E_0 E_0^\dagger E_0^\dagger E_0|p_1)|^2} \propto \frac{(E_{p_1} - \omega_L)^2 + \frac{\Gamma_{p_1}^2}{4}}{(E_{p_2} - \omega_L)^2 + \frac{\Gamma_{p_2}^2}{4}} \times \frac{\Gamma_{p_1}^2}{\Gamma_{p_2}^2}$$

Ratio between the Lorentzian functions of the density of states at \tilde{E}_{p_1} and \tilde{E}_{p_2} .

- When $\frac{\tilde{E}_{p_2}}{2} = E_{p_1} = \omega_L$ and $\Gamma_{p_2} \gg \Gamma_{p_1}$, the population of the high-loss $|p_2\rangle$ is prevented due to a smaller density of states, giving rise to the NHPB.

NHPB in hybrid cavity interacting with two-level emitter

Hybrid cavity⁴ interacting with emitter ($\gamma_e \ll \gamma_n$) incorporates optical modes with different losses ($\gamma_2 \gg \gamma_1$):

$$H_0 = \left(\omega - \frac{i\gamma_e}{2} \right) \sigma_x \sigma_z + \sum_{n=1,2} \left[\left(\omega - \frac{i\gamma_n}{2} \right) a_n^\dagger a_n + g_n (a_n \sigma_+ + a_n^\dagger \sigma_-) \right] + d (a_1^\dagger a_2 + a_2^\dagger a_1)$$

Engineering low-loss $|p_1\rangle$

When $g_1, \gamma_e, \gamma_1 \ll g_2, d$, decoupling of the microcavity and

Engineering high-loss $|p_2\rangle$

The loss of $|p_2\rangle$ is controlled by its plasmonic component: $|p_2\rangle \sim |a_2^\dagger a_2|p_2\rangle$. If it overcomes $\frac{\gamma_1}{\gamma_2}$, it has much higher loss than $|p_1\rangle$.

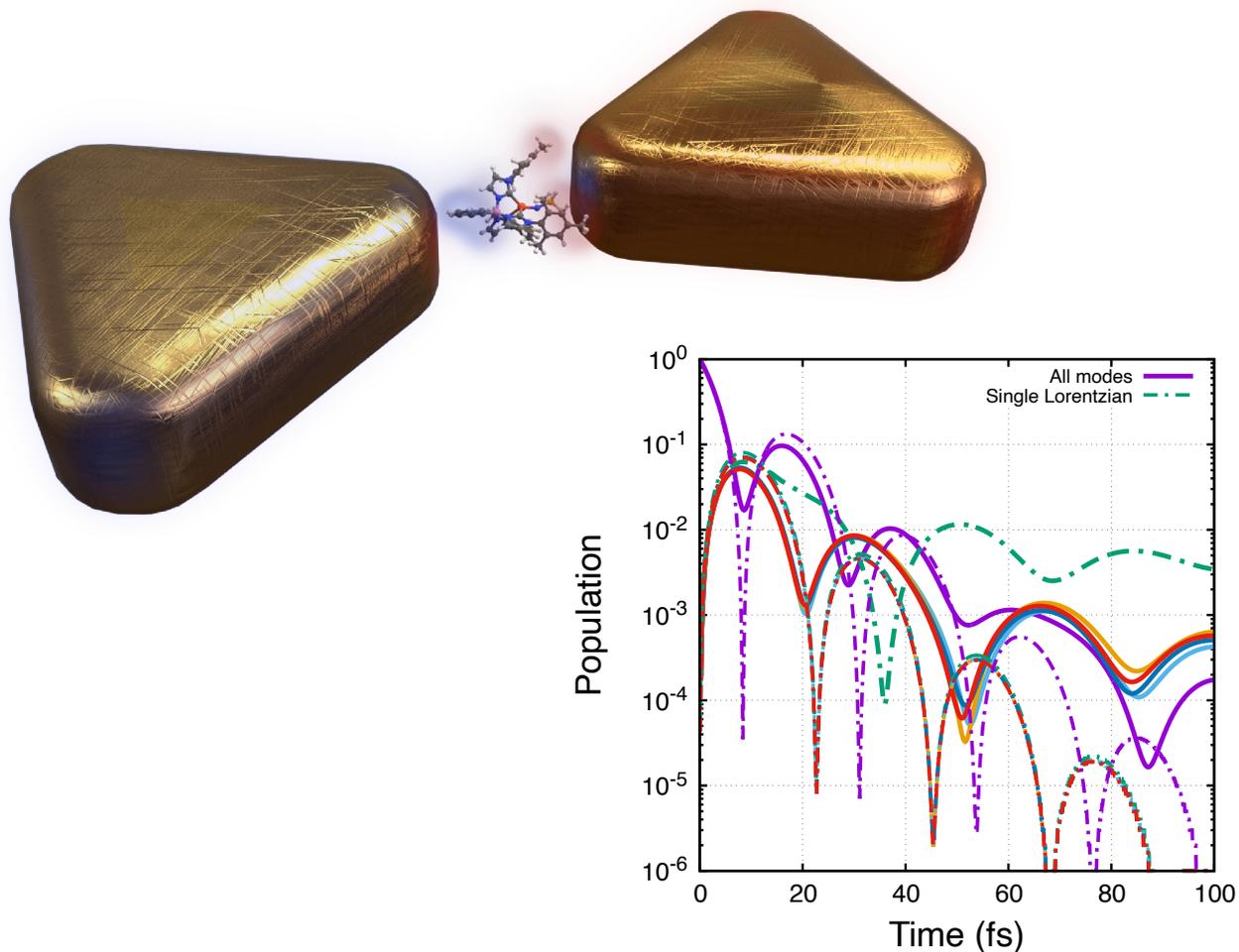
- By engineering low-loss $|p_1\rangle$ and high-loss $|p_2\rangle$, high-purity single-photon emission is achieved (solid lines).
- This single-photon emission is well described just by the narrowest eigenstates of each manifold, $|p_1\rangle$, $|p_2\rangle$ and $|p_2\rangle$ (dotted lines).
- When intentionally setting all the eigenstates to have the same decay rate: $\Gamma_j = q\Gamma_n$ (q is the excitation number), the single-photon emission is completely suppressed (dashed lines).

Acknowledgments

This work has been funded by the Spanish Ministry of Science and Innovation - Agencia Estatal de Investigación through grants PID2022-140140GB-I00, PID2021-140140GB-I00, and CEX2020-110105M through the María de Maeztu program for units of excellence in R&D, by the Proyecto Sinérgico CAM 2020 through grant S2020-TCS-EG6 (MHC/CAM of the Community of Madrid, by the European Research Council through grant ERC-2020-505-714879 and by the European Union's Horizon Europe Research and Innovation Programme through agreement 101019000 (MIRAGL5) and through agreement 101019000 (CVIS3).

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.



Investigating the Properties of Molecule-Cavity Systems through Molecular Dynamics Simulations

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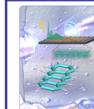
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Motivation



While coherent emission of organic chromophores is limited by their complex internal dynamics as well as their immediate material environment, coupling these photoactive molecules to nanophotonic structures has the potential to open a new era in quantum information technologies. [1, 2] Because of the large binding energy of Frenkel excitons, organic materials are furthermore promising candidates for future applications as strong light-matter coupling can be achieved at ambient conditions. [3]

However, a model that describes accurately both the molecules and the electromagnetic environment created by the light-confining structure is currently lacking which limits the understanding of the effects of material properties in the dynamics of strongly coupled systems. While we have achieved the first requirement of such a model by adopting an atomistic QM/MM representation of the material part of the strongly coupled system, [4, 5] the description of confined light was limited to modes of optical Fabry-Pérot resonators. [6] To move beyond, we introduce an explicit description of the quantised electromagnetic field for arbitrary nanophotonic structures such as plasmonic or hybrid metallo-dielectric nanocavities. [7]

Model of Light-Matter Interactions

We model light-matter interactions, through an extension of the well-established Tavis-Cummings Hamiltonian [8] of quantum optics, that we had previously adapted to account for the molecular degrees of freedom [4, 5], and include the full mode structure of arbitrary nano-resonators:

$$\hat{H} = \sum_k h\omega_k(\mathbf{R})\hat{a}_k^\dagger\hat{a}_k + \sum_j h\nu_j\hat{a}_j^\dagger\hat{a}_j - \sum_j \sum_k h g_{jk}(\hat{a}_k^\dagger + \hat{a}_k)\hat{a}_j$$

$h\nu_j(\mathbf{R}_j)$ is the geometry dependent excitation energy of molecule j at position \mathbf{r}_j , $h\omega_k$ is the energy of the medium-assisted electromagnetic mode k and g_{jk} is the light-matter coupling strength between molecule j and electromagnetic mode k in the long-wavelength (or dipole) approximation that depends on $\mu_j(\mathbf{R}_j)$, the geometry dependent molecular transition dipole moment.

Assuming a linear response of the nano-resonator to an external electromagnetic field, [9, 7] the *finite* set of modes $h\omega_k$ describe accurately the confined electromagnetic field at each point \mathbf{r} in space and frequency ω . They are obtained through a fitting procedure of the spectral density $J_{\text{mod}}(\omega) = \frac{1}{2} \Omega \left[\mathbf{E}_{\text{mod}} \cdot \mathbf{R} \right]^2$, which fully encodes the confined electromagnetic field in interaction with the N emitters.

The total wave function $\Psi(t)$ describing the interacting light-matter system is expressed as a time-dependent superposition in the *diabatic* basis of molecular $|S_j^i\rangle$ and electromagnetic mode excitations $|1_k\rangle$:

$$\Psi(t) = \sum_{j=1}^{N_{\text{mol}}} c_{j,0}(t) \phi_j^m \quad \text{with} \quad \phi_j^m = |S_j^i\rangle \otimes |0_k\rangle \quad \text{if} \quad m < N$$

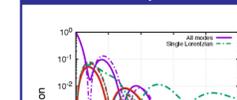
$$\phi_j^m = |S_j^i\rangle \otimes |1_k\rangle \quad \text{if} \quad m > N$$

The expansion coefficients $c_{j,0}(t)$ are evolved along the *classical* trajectories of the nuclei of the N molecules in a mean-field or Ehrenfest formalism. Thus, atom i belonging to molecule j , experiences an average potential due to *weak/strong* interactions with the confined field. The forces due to mode k acting on atom i of molecule j are of the form:

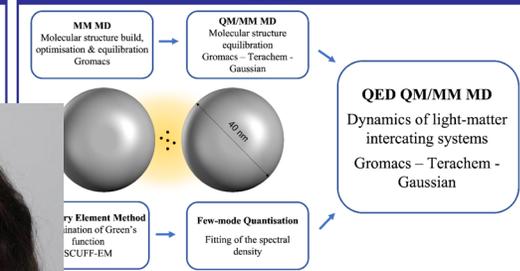
$$\langle \phi_j^m | \nabla_{\mathbf{r}_i} \hat{H}^{TC} | \phi_j^m \rangle = |\epsilon_j|^2 \nabla_{\mathbf{r}_i} V_{\text{em}}^{(m)}(\mathbf{R}_j) + (1 - |\epsilon_j|^2) \nabla_{\mathbf{r}_i} V_{\text{em}}^{(m)}(\mathbf{R}_j) + (\epsilon_j)^2 c_{j,0}(\mathbf{R}_j) \cdot \hat{\mathbf{E}}(\mathbf{r}_i)$$

where $\hat{\mathbf{E}}(\mathbf{r}_i)$ is the electric field profile at position \mathbf{r}_i .

Preliminary Results



Workflow



Acknowledgements

European Research Council (ERC Starting Grant ERC-2016-STG 714870 MMUSCLES). We thank the Center for Scientific Computing (CSC-IT Center for Science) for generous computational resources.

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 "Nanosciences" J. Chem. Phys., 154 (2021)
 "Nanosciences" J. Chem. Phys., 154 (2021)
 "Nanosciences" J. Chem. Phys., 154 (2021)

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



Universidad Autónoma
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European Research Council
Established by the European Commission

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Open-source codes:
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