

Organic polariton condensates: Modeling complex open quantum systems and structured environments

Jonathan Keeling



University of
St Andrews

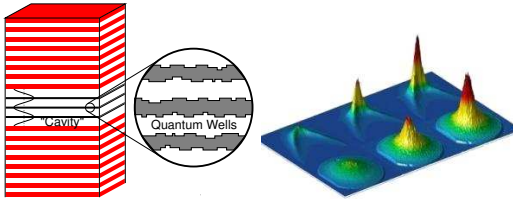
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Flatiron Institute, July 2019

Motivation: polariton condensates

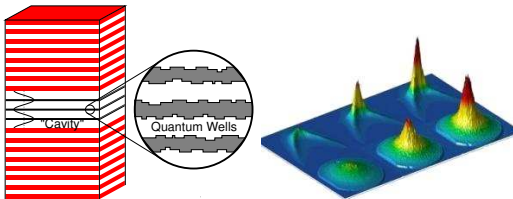
- CdTe Polariton Condensate $T \sim 20\text{K}$.



[Kasprzak *et al.* Nature, '06]

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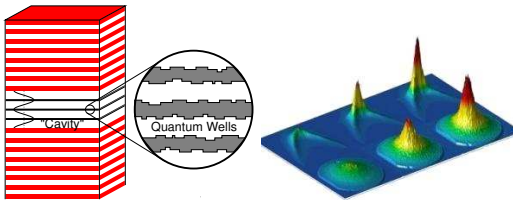
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Q2. Energetics vs dynamics (esp. spin state).

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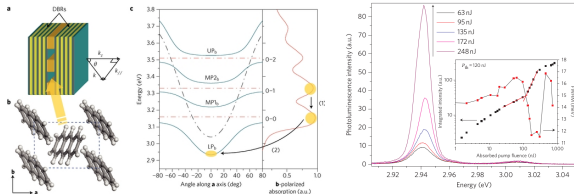


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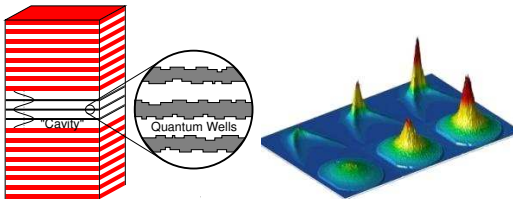
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[Kena Cohen and Forrest, Nat. Photon '10]

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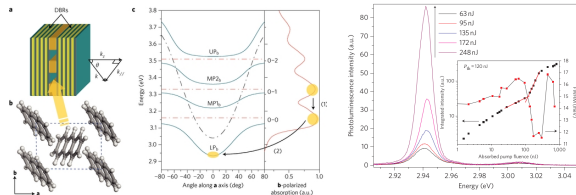


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Q1. Vibrational replicas?

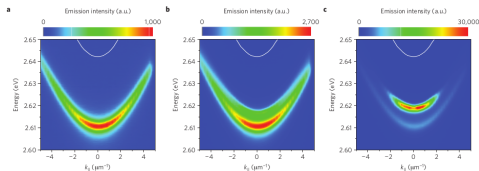
Q2. Relevance of disorder?

Q3. Lasing vs condensation?

[Kena Cohen and Forrest, Nat. Photon '10]

Motivation: polariton and photon condensates

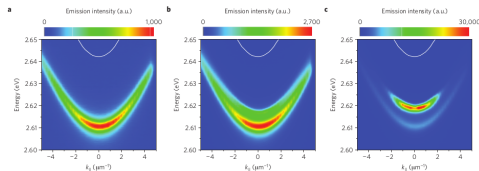
- Other organic materials, e.g. polymers:



[Plumhoff *et al.* ; Daskalakis *et al.* Nat. Mater. '14]

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Q1. Frenkel to Wannier crossover?

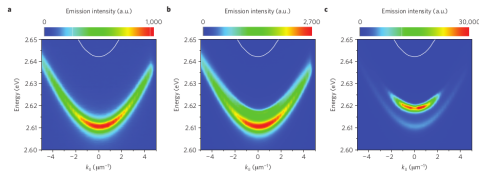
Q2. Optimal vibrational properties?

Q3. Nonlinearities?

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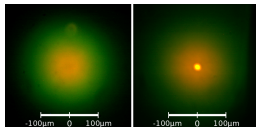
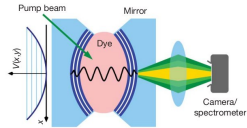
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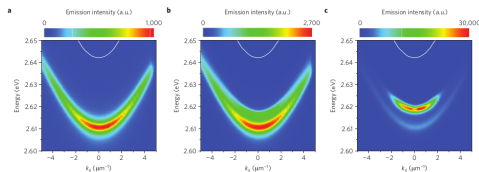
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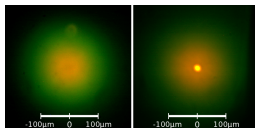
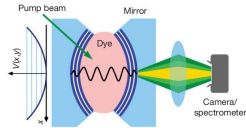
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Q1. Relation to dye laser?

Q2. Relation to polaritons?

Q3. Thermalisation breakdown?

[Klaers *et al.* Nature, '10]

What kinds of modelling

- Bottom up

- ▶ DFT (or quantum chemistry)
→ electronic structure
- ▶ Time-dependent DFT /MD
→ vibrational spectra
- ▶ FDTD/transfer-matrix
→ cavity modes

- Tractable microscopic toy models

- Top-down

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Illustration by Dick Cador.

[Auerbach, Interacting Electrons (Springer, 1998)]

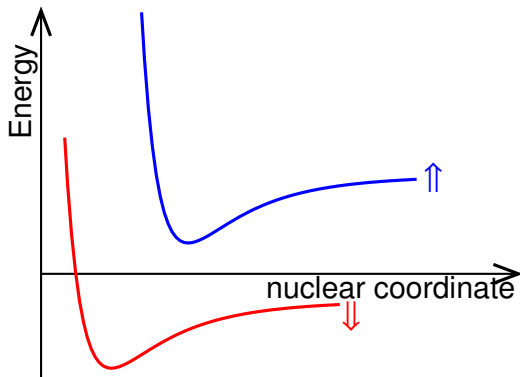
[From Auerbach, Interacting electrons and quantum magnetism]

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Toy model: Dicke-Holstein

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 - ▶ Two-level system, HOMO/LUMO
 - ▶ Single DoF Potential Energy Surface

● Simplified archetypal model: Dicke-Holstein

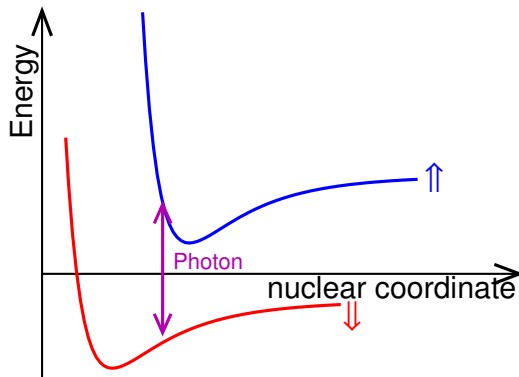


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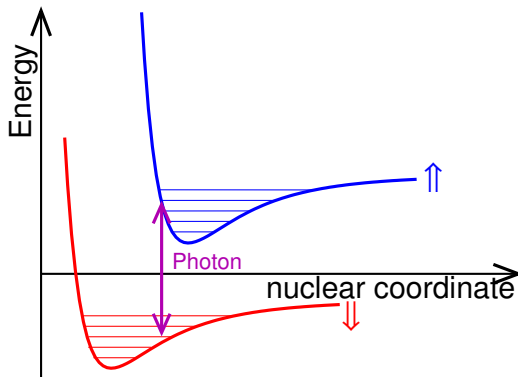


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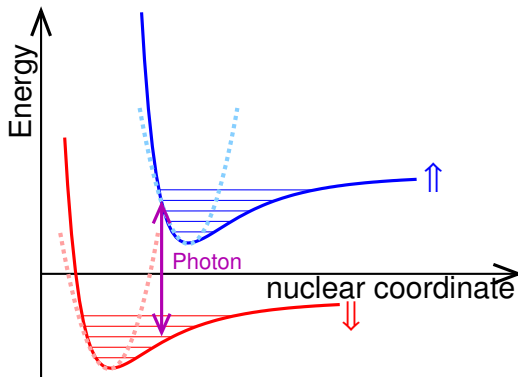
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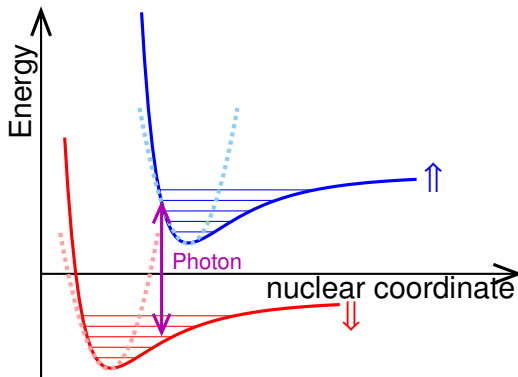
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 - Dicke Holstein model
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- 3 Lasing from organic molecules
- 4 TEMPO: methods for non-Markovian models
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One excitation subspace, questions

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- $\hat{x}_i = (\hat{b}_i + \hat{b}_i^\dagger) / \sqrt{2}$

- Restrict, $\hat{a}^\dagger \hat{a} + \sum_i \sigma_i^+ \sigma_i^- = 1$.

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- Competition of $g\sqrt{N}$ vs ω_v , $\omega_v \lambda_0^2$
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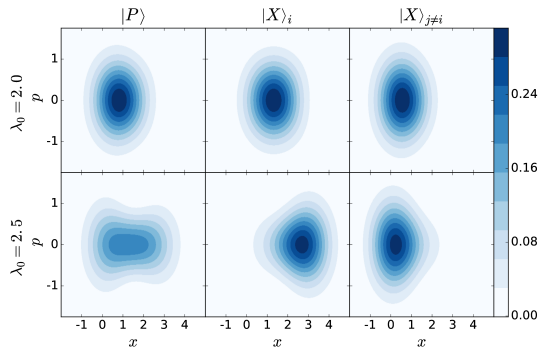
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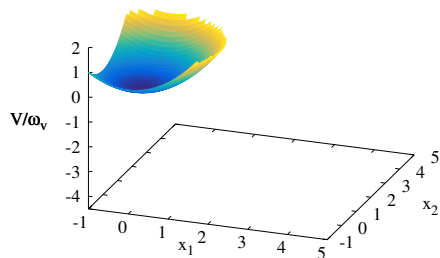
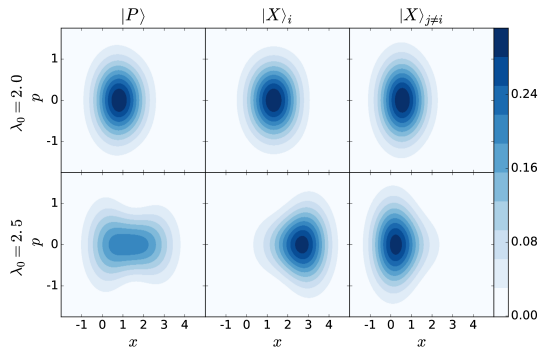


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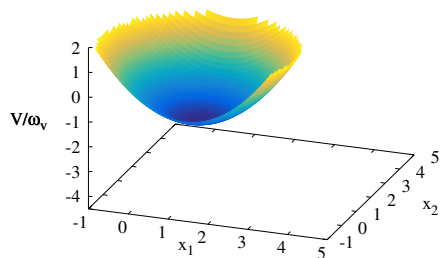
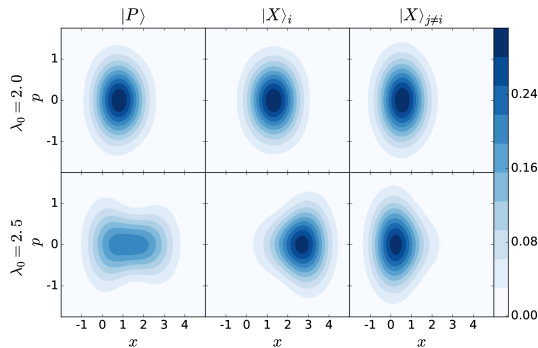


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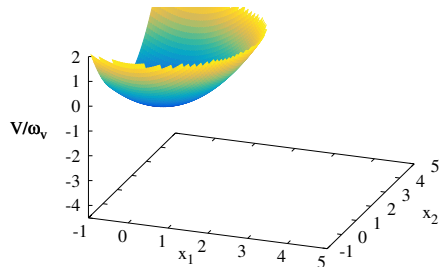
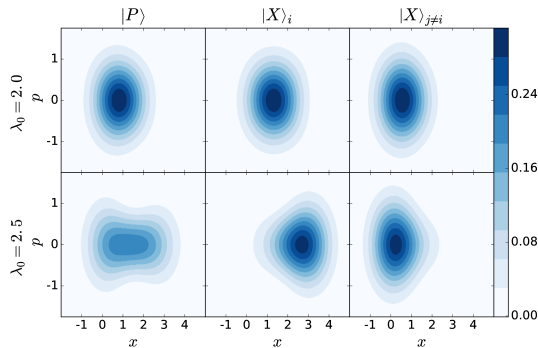


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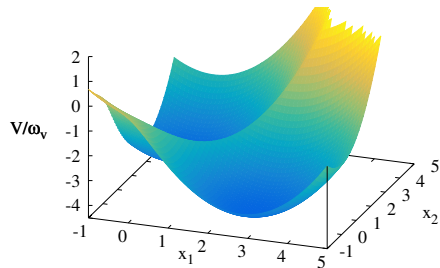
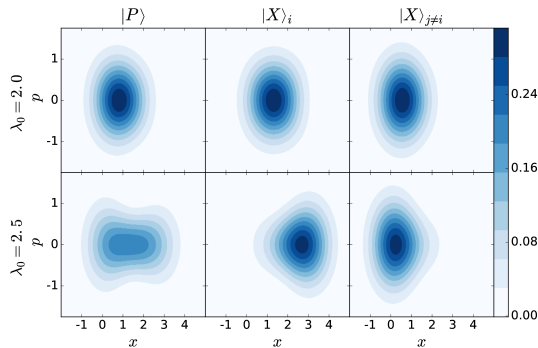


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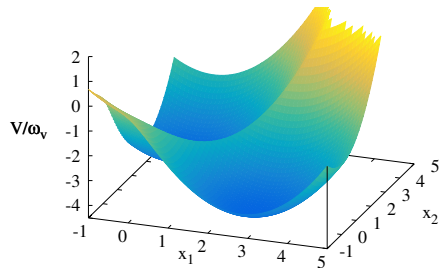
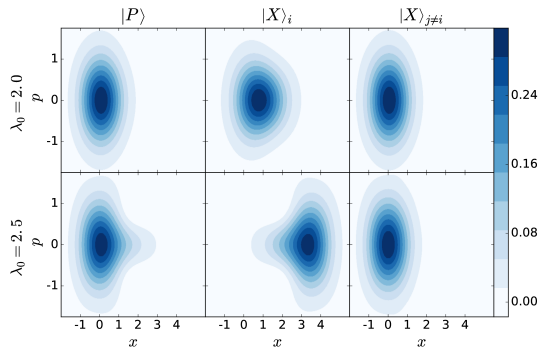


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Use permutation symmetry

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Extending to arbitrary N , polariton-polaron ansatz

- Polaron ansatz $\mathcal{D}_i(\lambda) = \exp\left(\lambda(\hat{b}_i^\dagger - \hat{b}_i)\right)$ [Wu *et al.* PRB '16, Zeb, Kirton, JK ACS Phot. '17]

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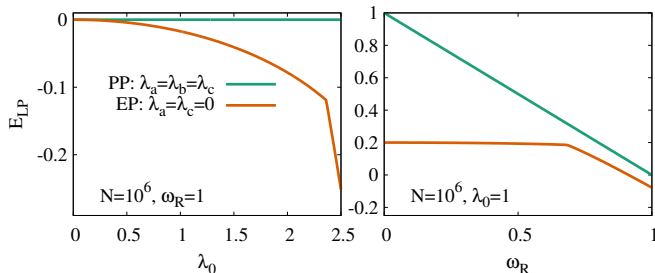
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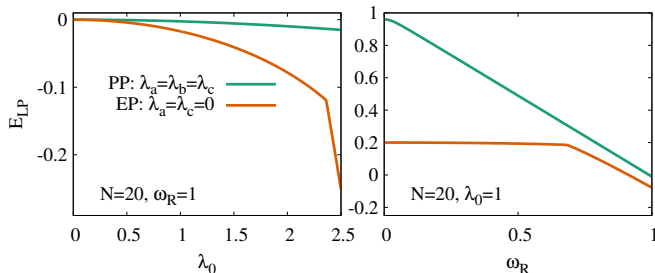
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$$|\Psi\rangle = \left[\alpha |P\rangle \prod_j \mathcal{D}_j(\lambda_a) + \frac{\beta}{\sqrt{N}} \sum_i |X\rangle_i \mathcal{D}_i(\lambda_b) \prod_{j \neq i} \mathcal{D}_j(\lambda_c) \right] |0\rangle_V$$

- Closed form $E[\lambda_a, \lambda_b, \lambda_c]$, minimise:

- ▶ At $N \rightarrow \infty$ suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$
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[Herrera and Spano PRL 2016]



Extending to arbitrary N , polariton-polaron ansatz

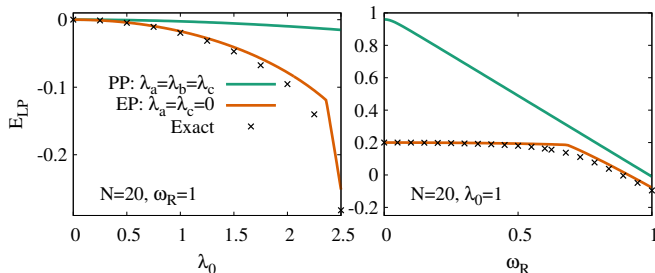
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Lasing from organic molecules

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 - Dicke Holstein model
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- 3 Lasing from organic molecules
- 4 TEMPO: methods for non-Markovian models
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Dicke-Holstein model: Strong coupling lasing

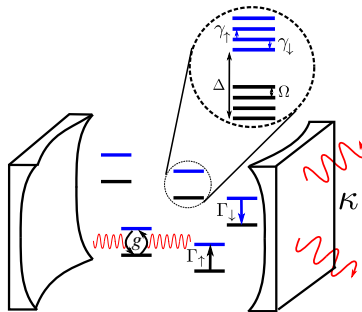
- Hamiltonian without RWA, with A^2 :

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^{N_m} \left[\omega_0 \sigma_i^+ \sigma_i^- + g \sigma_i^x (a + a^\dagger) + \omega_v \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) + \frac{g^2}{\varepsilon} (a^\dagger + a)^2 \right]$$

- Incoherent processes:

$$\partial_t \rho = -i[H, \rho] + \sum_j \gamma_j \mathcal{L}[\rho]$$

- Photon loss $\propto \mathcal{L}[a]$
- Molecular pump, decay $\Gamma_1 \mathcal{L}[\sigma^+], \Gamma_2 \mathcal{L}[\sigma^-]$
- Dephasing $\Gamma_x \mathcal{L}[\sigma^x]$
- Vibrational relaxation: $\gamma_1 \mathcal{L}[\hat{b}^\dagger - \lambda_0 \sigma^+ \sigma^-], \gamma_2 \mathcal{L}[\hat{b} - \lambda_0 \sigma^+ \sigma^-]$
(Thermal: $\gamma_1 = \gamma_2[\bar{n}_v + 1], \gamma_2 = \gamma_1 \bar{n}_v$)



[Strashko, Kirton & JK, PRL '18]

Dicke-Holstein model: Strong coupling lasing

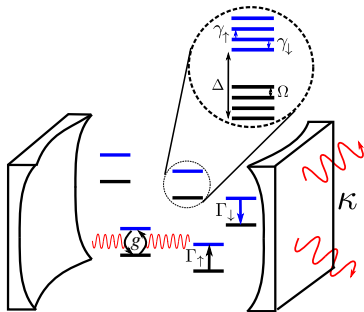
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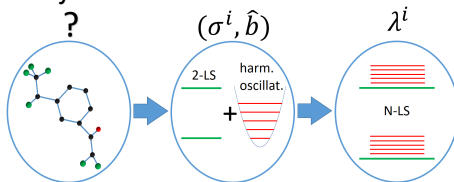
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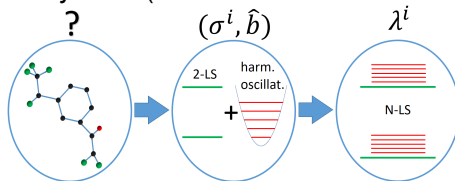
Mean-field theory for Holstein-Dicke

- Exciton-vibron $\rightarrow$ N -level system



Mean-field theory for Holstein-Dicke

- Exciton-vibron $\rightarrow$ N -level system (Generalized Gell-Mann matrix)

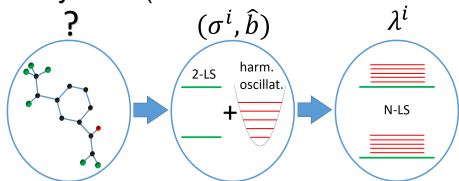


λ^i : $\lambda = \lambda^\dagger$, $\text{Tr}(\lambda) = 0$, so now

$$H_0 = \omega_c a^\dagger a + \frac{g^2 N_m}{\varepsilon} (a^\dagger + a)^2 + \sum_n \left[A_i + B_i (a^\dagger + a) \right] \lambda_i^{(n)}$$

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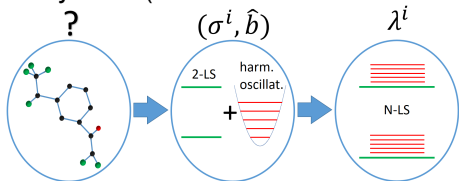
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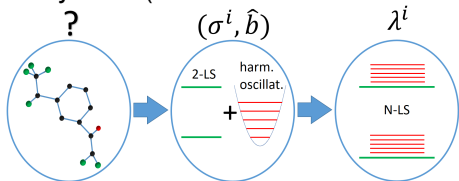
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$\text{Re}[\xi^{(k)}]$ — growth/decay rate
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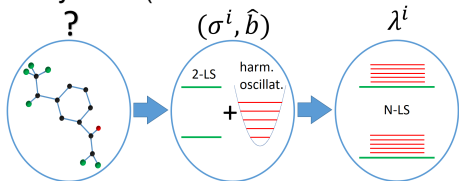
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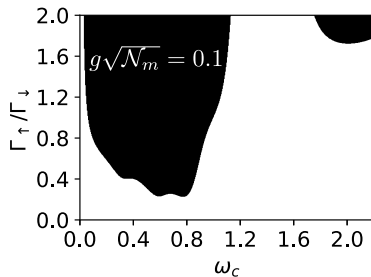


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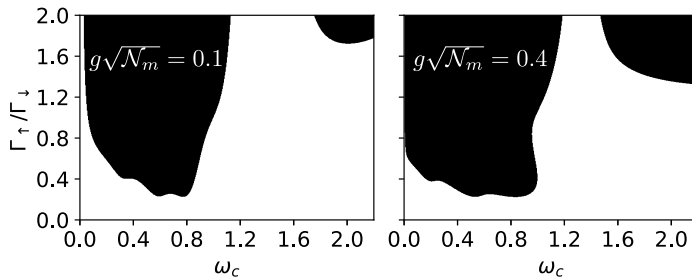
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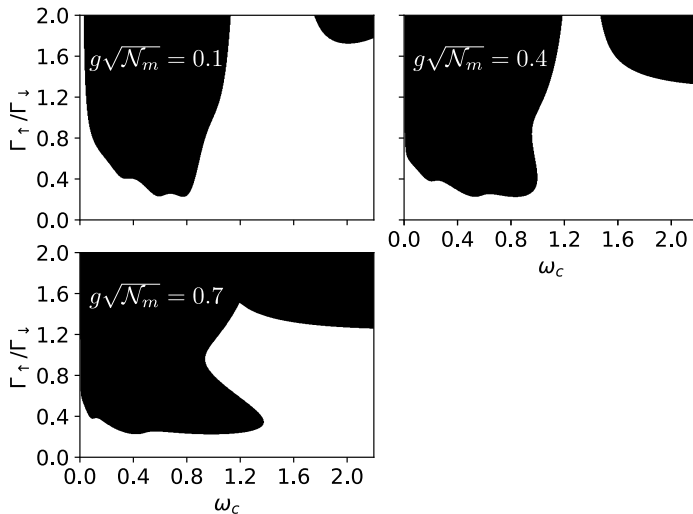
Evolution of phase diagram vs coupling



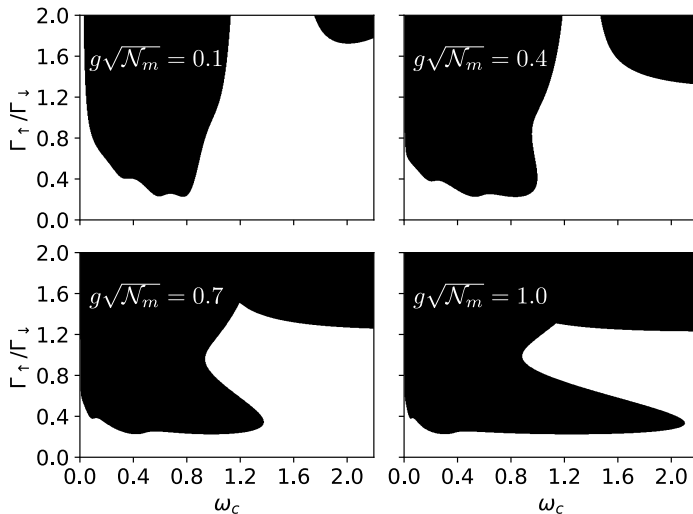
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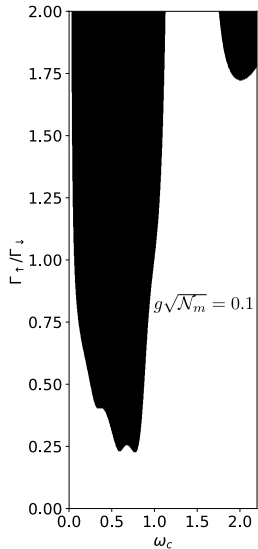
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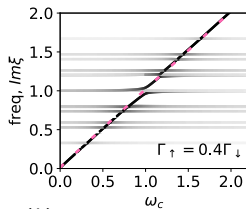
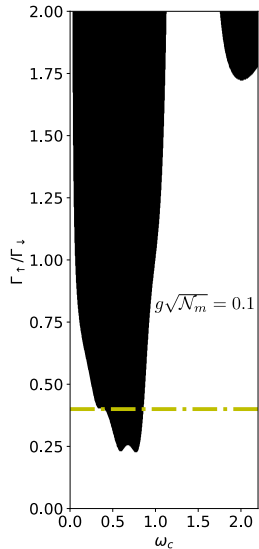


Explaining phase diagram: strong coupling



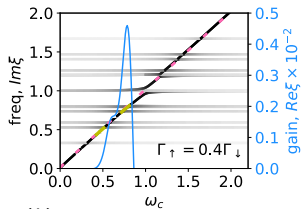
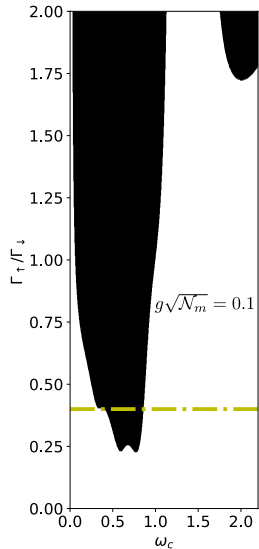
• Frequencies: $\text{Im}[\zeta^{(k)}]$,

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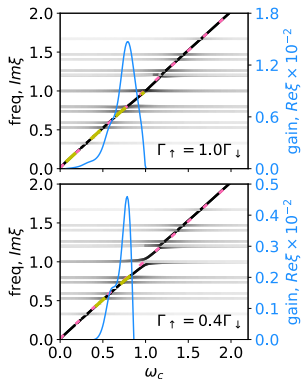
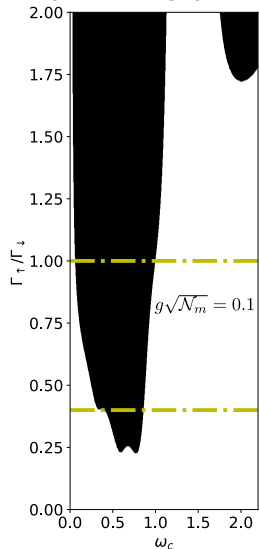
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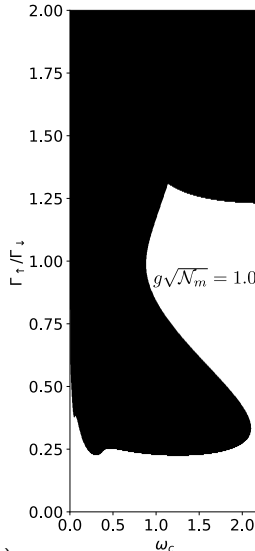
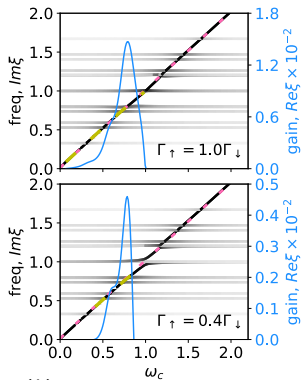
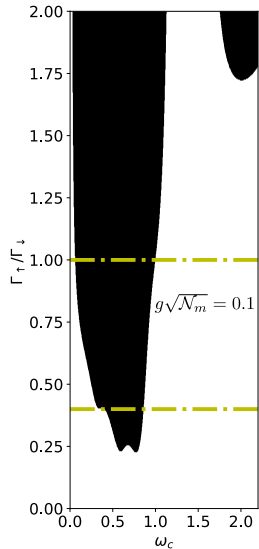
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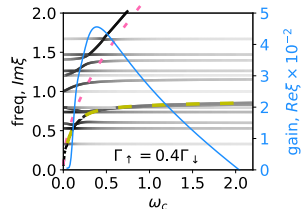
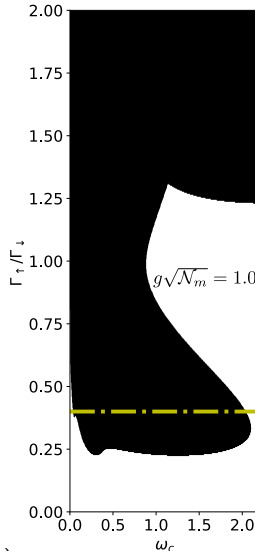
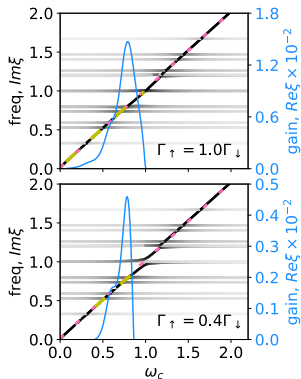
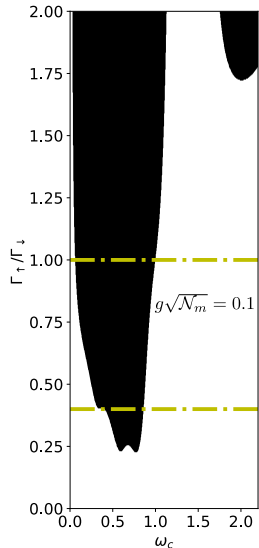
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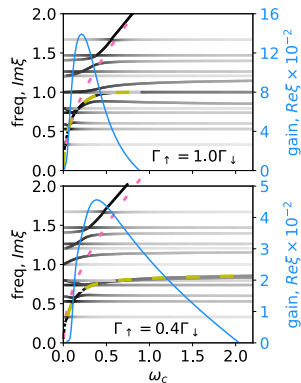
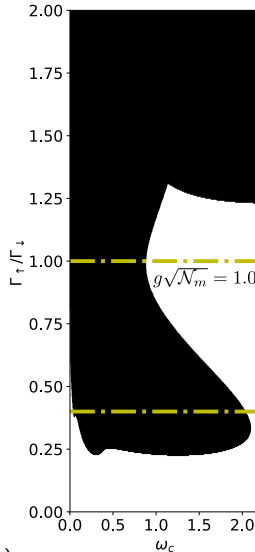
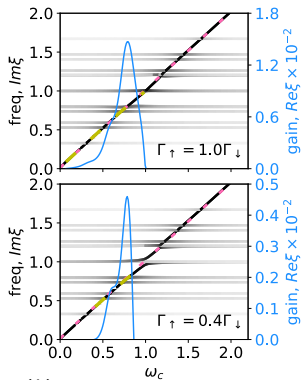
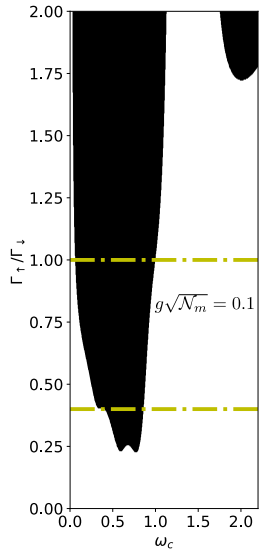
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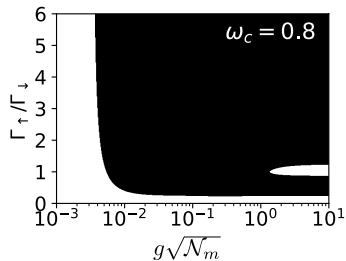


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Does strong coupling reduce threshold?

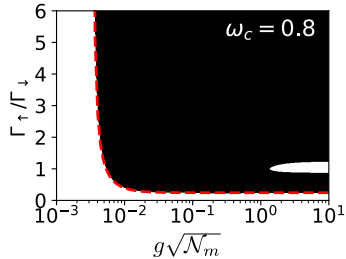
- Threshold vs g at optimal/sub-optimal ω_c

• Weak-coupling (photon BEC) prediction



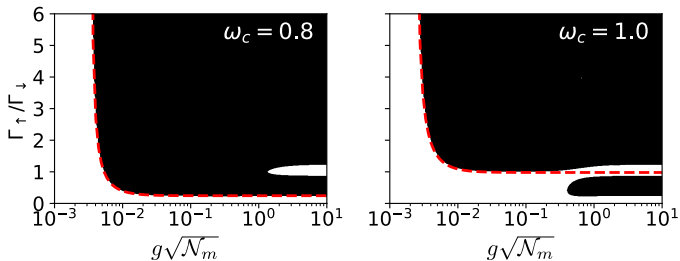
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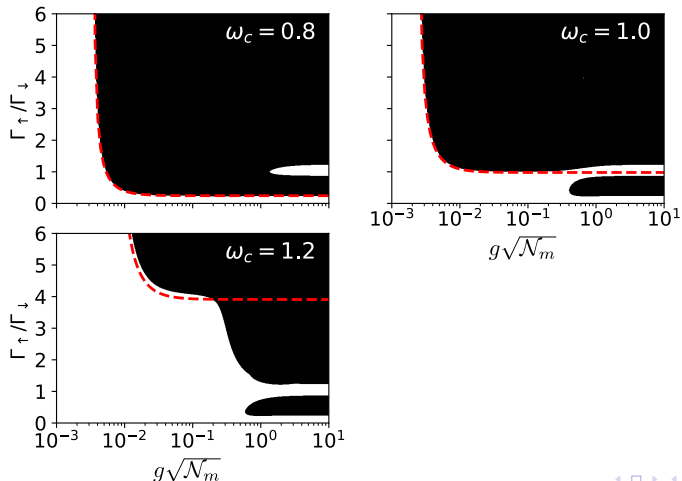
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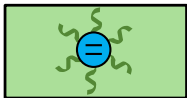
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QUAPI [Makri and Makarov, J. Chem Phys '95]

- System + harmonic bath



$$H = H_S + O \sum_k \xi_k (b_k^\dagger + b_k) + H_B$$

- Write $\rho_f(t)$ as path sum/integral

- ▶ Discrete sum for system

$$1 = \sum_j |j\rangle\langle j|$$

- ▶ Coordinate path integral for bath

$$1 = \int dx_n |x_n\rangle\langle x_n|$$

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- ADT; products $\rightarrow$ Growth.

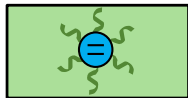
$$A_{b,b,h}^{k,h,b,h} = B_{b,b,h}^{k,h,b,h} A_{b,b,h}^{b,b,h}$$

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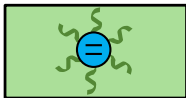
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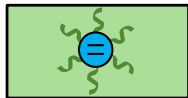
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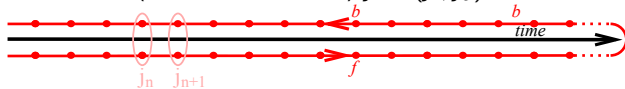
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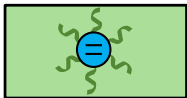
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$$\mathbb{1} = \sum_j |j\rangle \langle j|$$

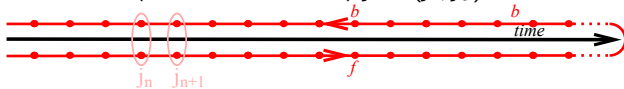
- Coordinate path integral for bath

$$\mathbb{1} = \int dx_n |x_n\rangle \langle x_n|$$

- Discretize times, $t_N = N\Delta$

- Integrate out bath.

- Path sum (doubled indices) $j = (j_f, j_b)$:



$$\rho_{j_N}(t_N) = \sum_{j_1 \dots j_{N-1}} A^{j_N, j_{N-1}, \dots, j_1}$$

- ADT; products $\rightarrow$ Growth.

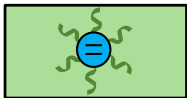
$$A^{j_4, j_3, j_2, j_1} = B^{j_4, j_3, j_2, j_1}_{j_3, j_2, j_1} A^{j_3, j_2, j_1}$$

$$B^{j_n, j_{n-1}, \dots, j_1}_{j_{n-1}, \dots, j_1} = \left(\prod_{k=1}^{n-1} \delta_{j_{n-k}}^{j_{n-k-1}} \right) \prod_{k=0}^{n-1} I_k(j_n, j_{n-k}).$$

Problem: n^2 Tensor Size $\sim j^2$

QUAPI [Makri and Makarov, J. Chem Phys '95]

- System + harmonic bath



$$H = H_S + O \sum_k \xi_k (b_k^\dagger + b_k) + H_B$$

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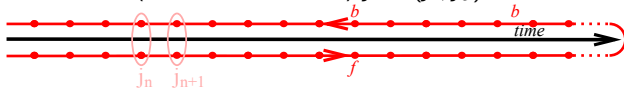
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- Problem: n^{th} Tensor size $\sim d^{2n}$

QUAPI as tensor network: TEMPO

So far ...:
$$\rho_{j_N}(t_N) = \sum_{j_1 \dots j_{N-1}} A^{j_N, j_{N-1}, \dots, j_1} \rho_{j_1}(t_1), \quad \mathbb{A} \rightarrow \mathbb{B} \cdot \mathbb{A}$$

• B is tensor network: $B_{j_{n-1}, \dots, j_1}^{j_n, j_{n-1}, \dots, j_1} = [b_0]_{\alpha_1}^{j_n} \left(\prod_{k=1}^{n-2} [b_k]_{\alpha_{k+1}, j_{n-k}}^{\alpha_k, j_{n-k}} \right) [b_{n-1}]_{j_1}^{\alpha_{n-1}, j_{n-1}},$

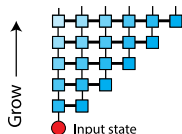
• Finite memory approx — neglect correlations K steps ago

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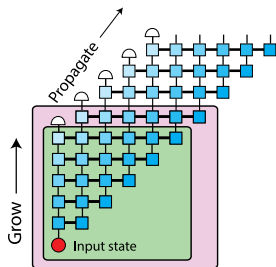
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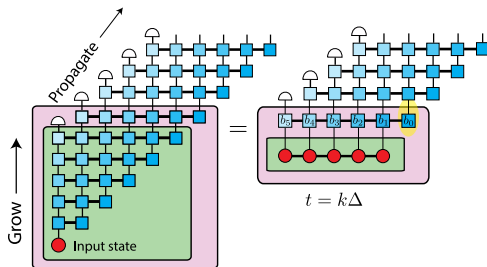
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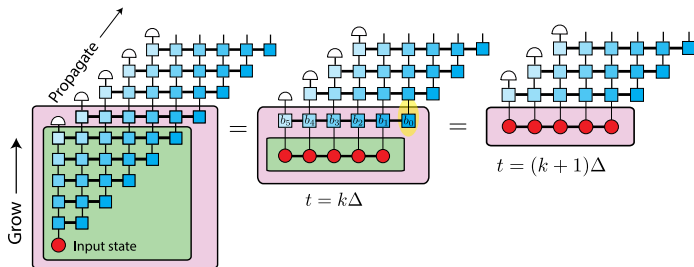
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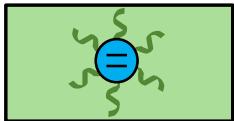
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Ohmic Spin Boson model

- Archetypal non-Markovian model:



$$H = \Omega S_x + \sum_i S_z (g_i a_i + g_i^* a_i^\dagger) + \omega_i a_i^\dagger a_i,$$

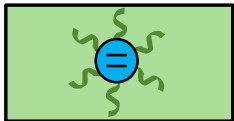
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• Known behaviour [Leggett *et al.* RMP '87], localization if $\alpha > 1$.

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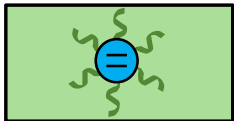
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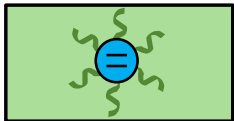
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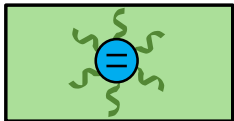
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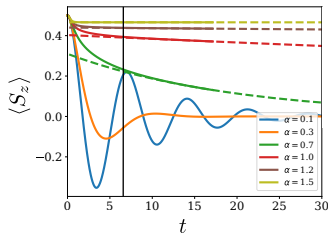
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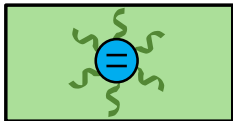
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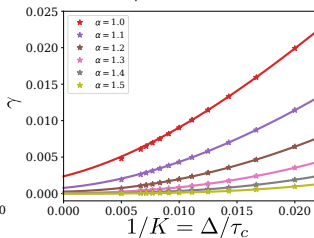
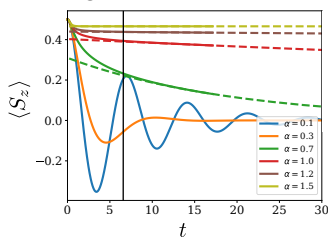
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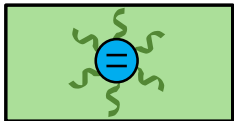
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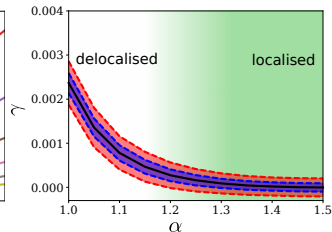
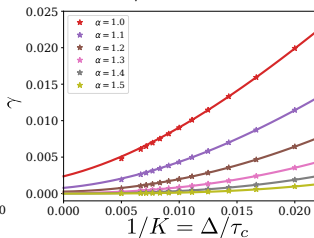
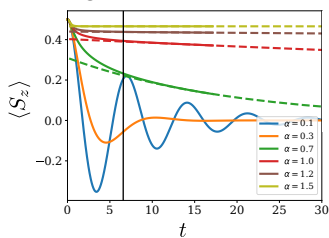
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- 1 Introduction
 - Dicke Holstein model
- 2 Polaritonic eigenstates
- 3 Lasing from organic molecules
- 4 TEMPO: methods for non-Markovian models
- 5 Outlook and conclusions

Outlook

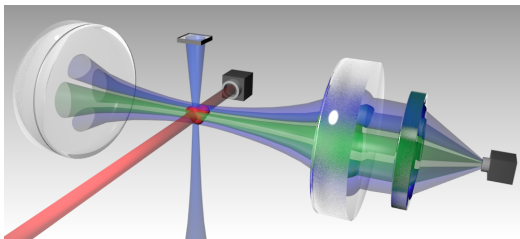
Cavity-induced change of properties & Driven-dissipative phases of matter

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Other contexts: Many Body Cavity QED & Cold atoms



- Engineering novel phases
- Beyond mean-field — multimode short-range interactions
- Dissipative computing & memory recall
- Strong cavity mediated interactions — new tool for simulation

Acknowledgements

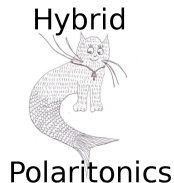
GROUP:



ALUMNI AND COLLABORATORS:

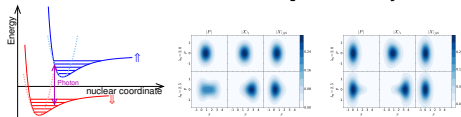


FUNDING:

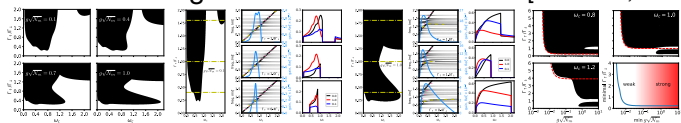


Summary

- Holstein-Dicke model [Cwik, Reja, Littlewood, JK, EPL '14; Zeb, Kirton, JK, ACS Phot. '17]



- Polariton lasing of Holstein-Dicke model [Strashko, Kirton, JK, PRL '18]



- TEMPO: algorithm for structured baths [Strathearn, Kirton, Kilda, JK & Lovett, Nat. Comm. '18]

