Exact and approximate eigenstates of vibrationally dressed polaritons

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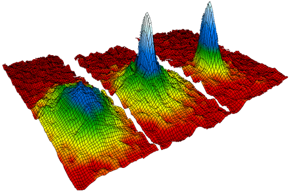
University of St Andrews
FOUNDED 1413

SUPA

Quantum Nanophotonics, February 2017

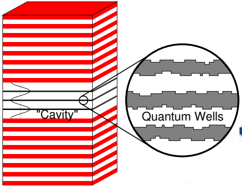
Condensation, Lasing, Superradiance

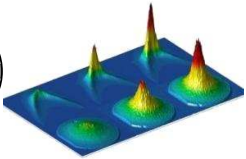
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science '95]

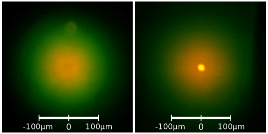
Polariton Condensate $T \sim 20$K





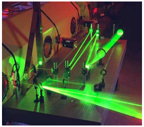
[Kasprzak *et al.* Nature, '06]

Photon Condensate $T \sim 300$K

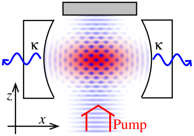


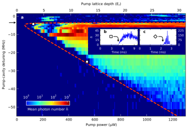
[Klaers *et al.* Nature, '10]

Laser $T \sim ?, < 0, \infty$



Superradiance transition $T \sim 0$

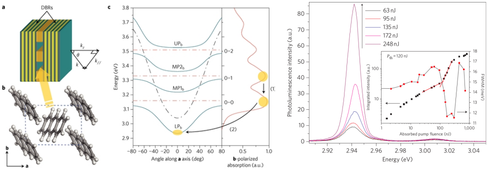




[Baumann *et al.* Nature '10]

Motivation: polariton condensates

- Anthracene Polariton Lasing
  $T \sim 300$K



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

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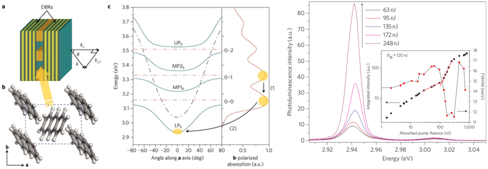

Q1. Vibrational replicas?

Q2. Relevance of disorder?

Q3. Lasing vs condensation?

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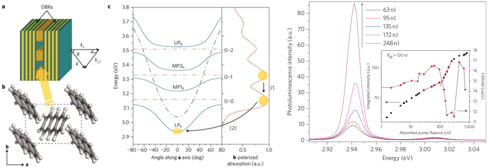

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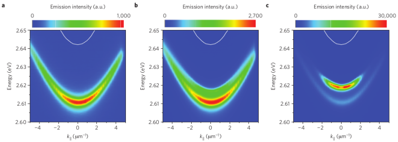

[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

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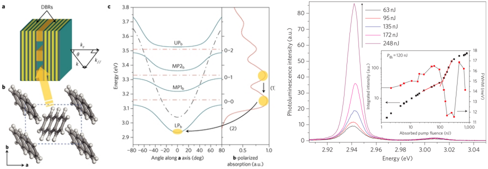

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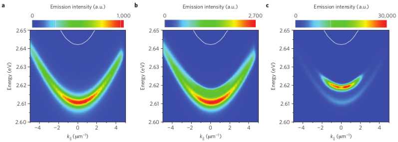

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Q1. Frenkel to Wannier crossover?

Q2. Optimal vibrational properties?

Q3. Nonlinearities?

Paradigms & Models

- Weakly interacting dilute Bose gas
  $$H = \int d^d r \hat{\psi}^\dagger(-\mu - \nabla^2)\hat{\psi} + U\hat{\psi}^\dagger\hat{\psi}^\dagger\hat{\psi}\hat{\psi}$$
  - Single field — assumes strong coupling
  - Continuum model, hard to include molecular physics

- Laser rate equations
  - Emission, absorption — assumes weak coupling, lasing.

- Complex Gross-Pitaevskii/Ginzburg Landau equations
  $$i\partial_t\psi = \left(-\nabla^2\psi + V(r) + U|\psi|^2\right)\psi + i\left(P(\psi, n, r) - \kappa\right)\psi$$
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- Transfer matrix, exciton susceptibility $\chi(\omega)$

- Microscopic model …

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Model capable of lasing & condensation

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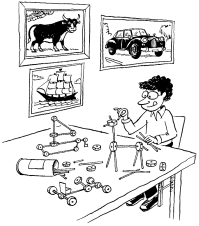
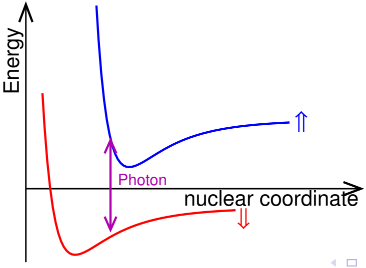


Illustration by Dick Codor.
[Auerbach, Interacting Electrons (Springer, 1998)]



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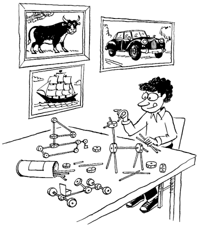

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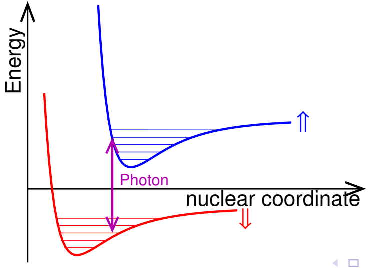

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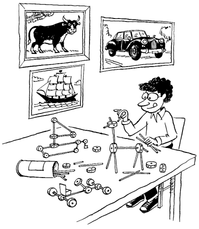

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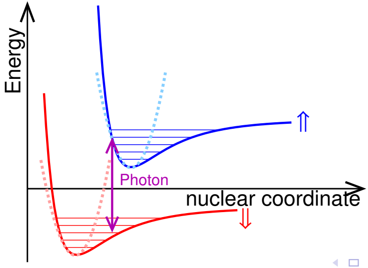

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Introduction and models

Strong coupling: polariton states

One excitation subspace, questions

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- Restrict, $\hat{a}^\dagger \hat{a} + \sum_i \sigma_i^+ \sigma_i^- = 1$.
- Questions:
  - Competition of $g\sqrt{N}$ vs ω_v, $\omega_v \lambda_0^2$
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Exact solution, $N = 2$

Vibrational Wigner function:

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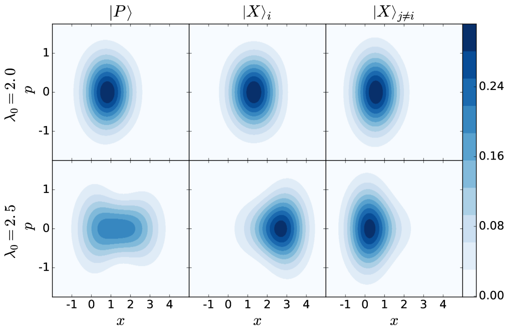
Conditioned on Photon $|P\rangle$/Exciton at i, $|X\rangle_i$/Other site $|X\rangle_{j\neq i}$



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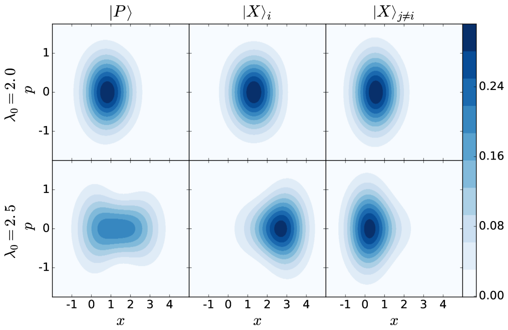

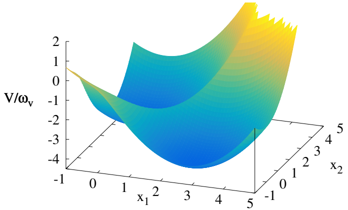

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- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5 - 6$

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- Distinct behaviour vs λ_0
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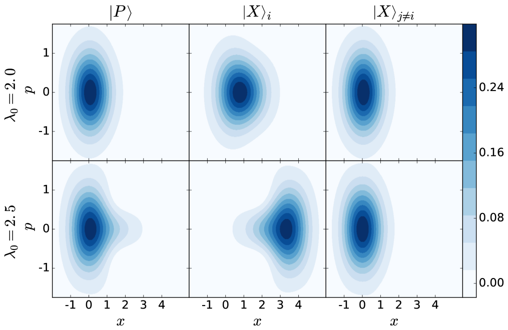

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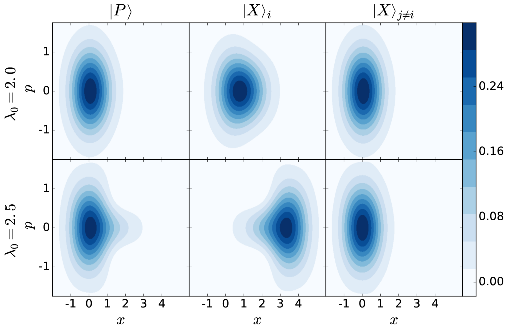

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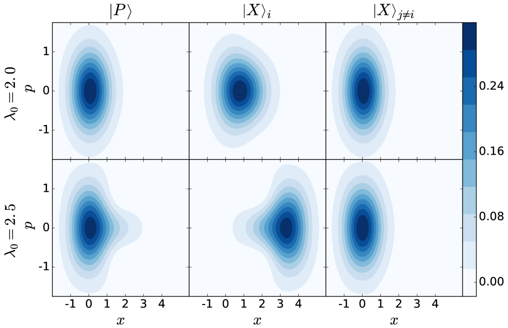

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Extending to arbitrary N, polaron ansatz

- Polaron transform, $\mathcal{D}_i(\lambda) = \exp\left(\lambda(\hat{b}_i^\dagger - \hat{b}_i)\right)$
- Single molecule ansatz:

$$|\Psi\rangle = [\alpha\mathcal{D}(\lambda_\uparrow)|\uparrow\rangle + \beta\mathcal{D}(\lambda_\downarrow)|\downarrow\rangle]|0\rangle_V$$

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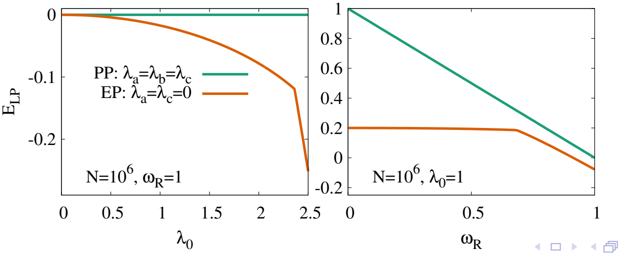

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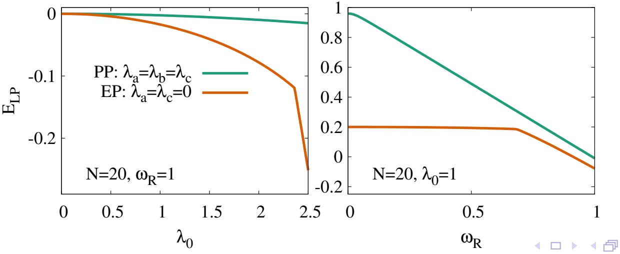

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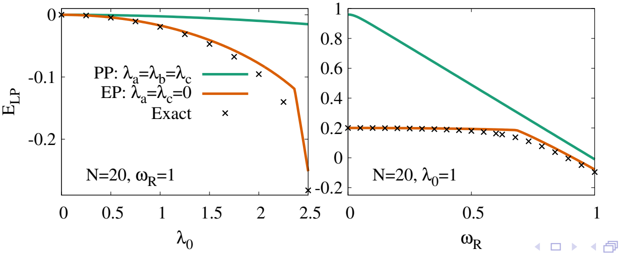

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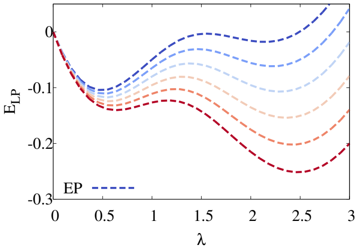

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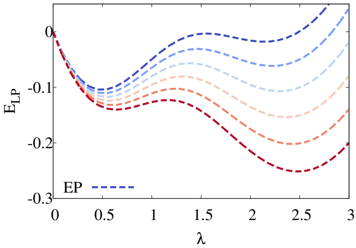

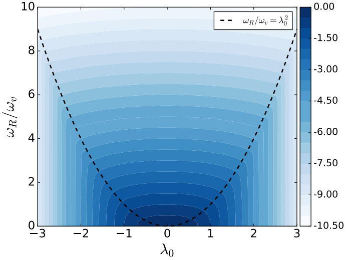

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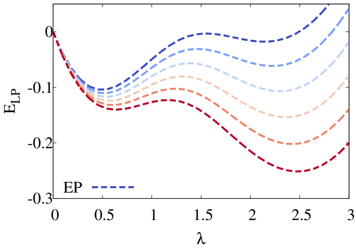

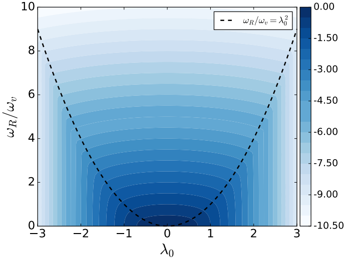

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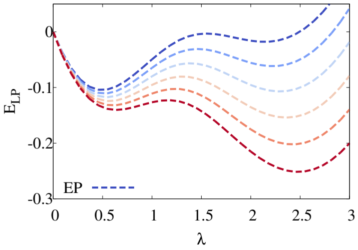

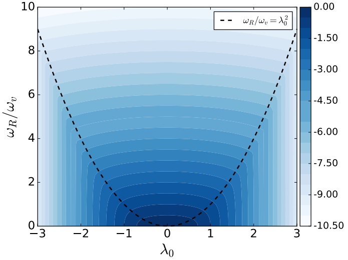

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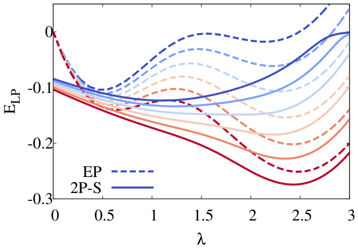

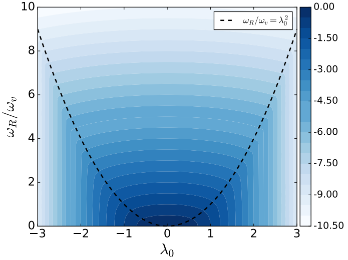

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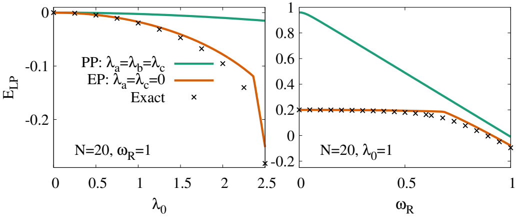
Simplified two-polaron physics

- Accurate energy & wavefunction



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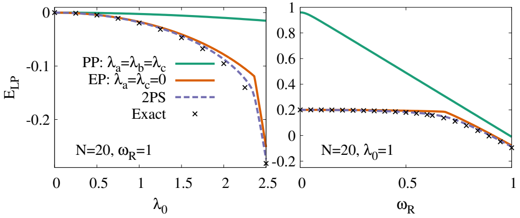

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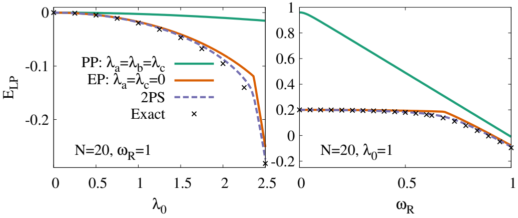

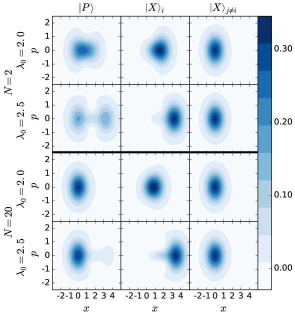

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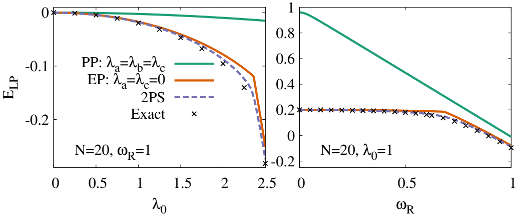

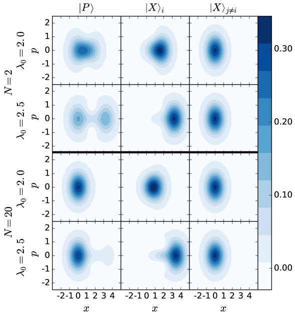

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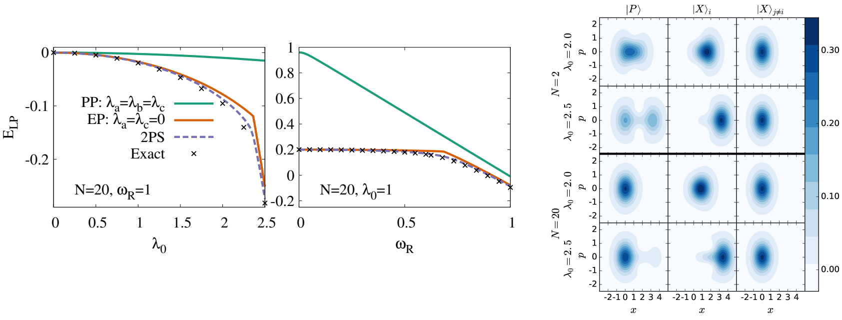

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Strong coupling: spectrum

Calculating spectra: Input-Output formalism

- Obsevable features: absorption spectrum, $A(\nu) = 1 - T(\nu) - R(\nu)$

- Scattering matrix gives:

$$A(\nu) = -\kappa_t \left[2\,\text{Im}[D^R(\nu)] + (\kappa_t + \kappa_b)|D^R(\nu)|^2 \right]$$

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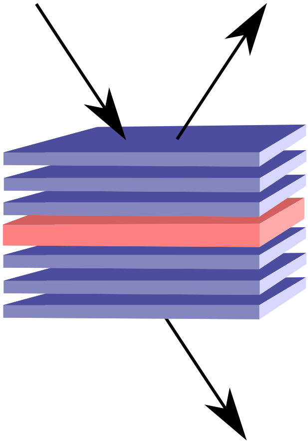

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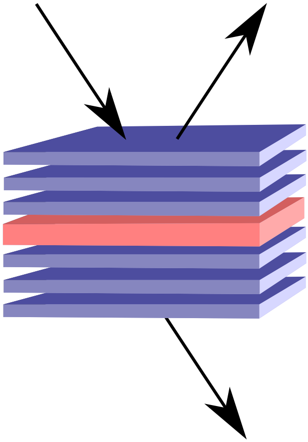

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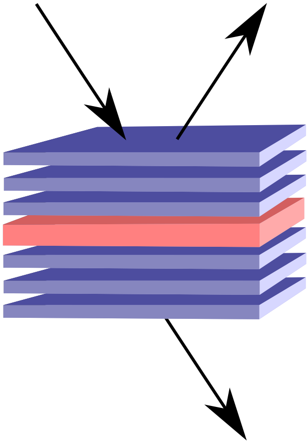

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Tavis-Cummings-Holstein vs Coupled Oscillators

- Coupled oscillator model:

$$H = \omega_P \hat{a}^\dagger \hat{a} + \sum_i \left[\frac{\omega_R}{\sqrt{N}} \left(\hat{a} \sum_n f_n(\lambda_0) \sigma_i^{n0} + \text{H.c.} \right) + \omega_n \sigma_i^{nn} \right]$$

$$\omega_n = \omega_X + n\omega_V, \quad f_n(\lambda_0) = \langle n | D(\lambda_0) | 0 \rangle$$

- Corresponds to classical susceptibility,

$$\chi(\nu) = -\sum_n \frac{\omega_R^2 |f_n(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_n}$$

- Ignores vibrational dressing of unexcited molecules

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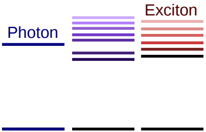

- Corresponds to classical susceptibility,

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Tavis-Cummings-Holstein vs Coupled Oscillators

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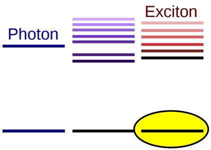

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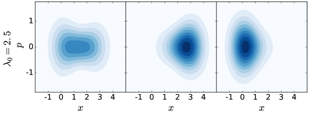

Tavis-Cummings-Holstein spectrum

- Direct calculation

$$D^R(t) = -i \left\langle 0 \middle| \left[\hat{a}(t), \hat{a}^\dagger(0) \right] \middle| 0 \right\rangle \theta(t)$$

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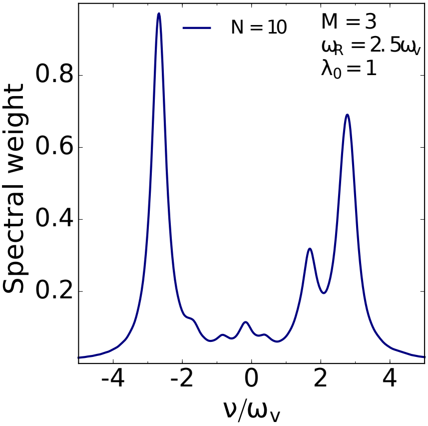

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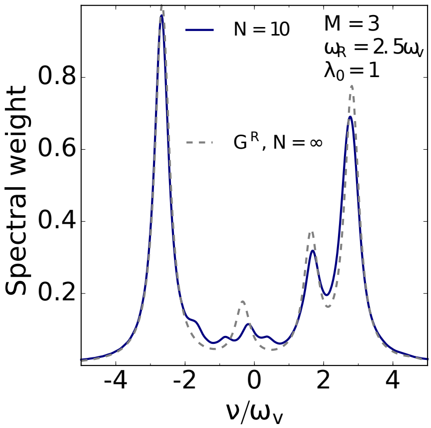

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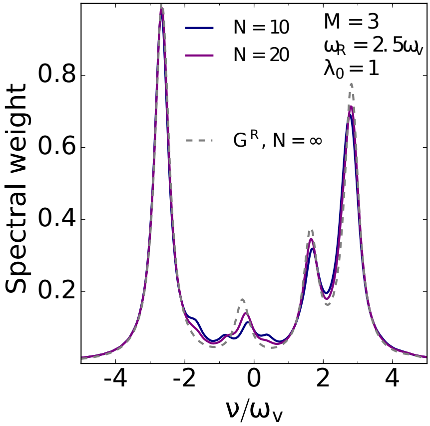

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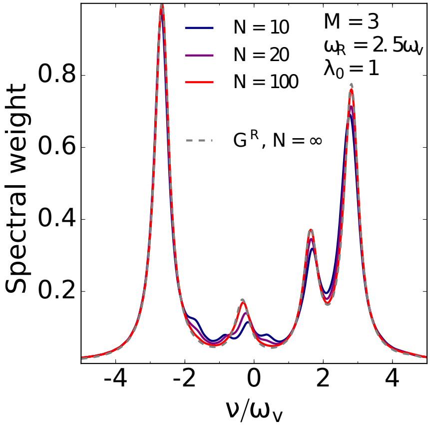

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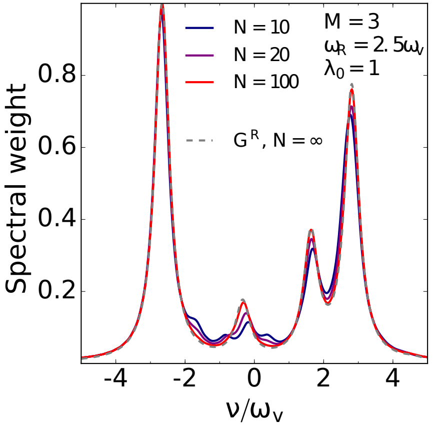

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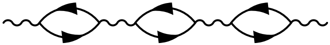
- Why? Multiple excitation $\sim 1/N$, 

Acknowledgements

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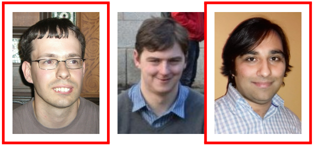

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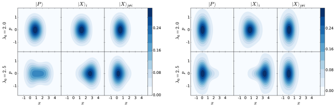
Hybrid
Polaritonics

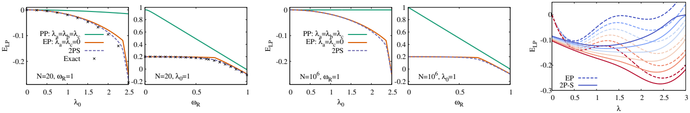
The Leverhulme Trust

Summary

- Holstein-Dicke and Holstein-Tavis-Cummings models
- Single polariton state
  - Exact solution



  - Polaron ansatz



[Zeb, Kirton, JK, `arXiv:1608.08929`]

- Validity of mean-field Green's functions