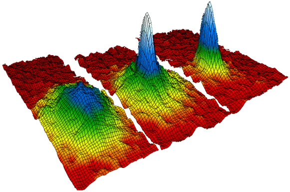
Modelling organic condensates from weak to strong coupling

Jonathan Keeling

Madrid, January 2017

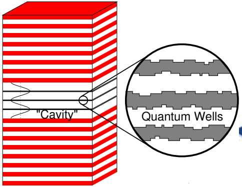
Condensation, Lasing, Superradiance

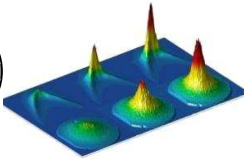
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science '95]

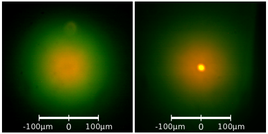
Polariton Condensate $T \sim 20$K





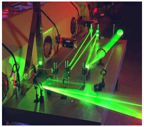
[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K

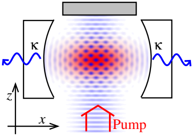


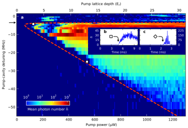
[Klaers *et al.* Nature, '10]

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



Superradiance transition
$T \sim 0$

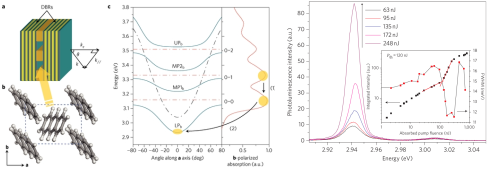




[Baumann *et al.* Nature '10]

Motivation: polariton condensates

- Anthracene Polariton Lasing
  $T \sim 300$K



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

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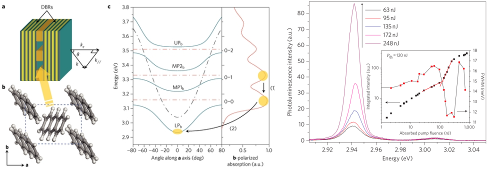

Q1. Vibrational replicas?

Q2. Relevance of disorder?

Q3. Lasing vs condensation?

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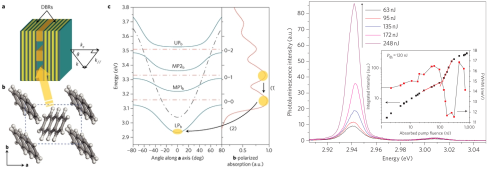

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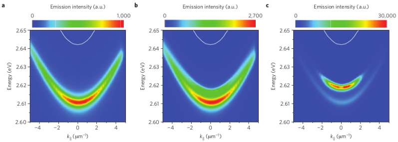

[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

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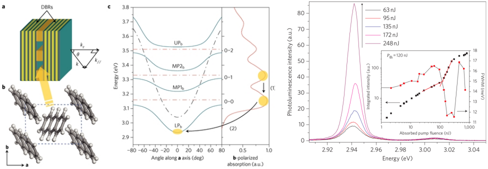

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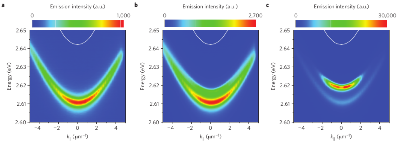

[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

Q1. Frenkel to Wannier crossover?

Q2. Optimal vibrational properties?

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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- Top-down
  - Equilibrium stat. mech.
  - (complex/stochastic/...)GPE (+ Boltzmann) $\rightarrow$ condensate
  - Rate equations $\rightarrow$ laser
- Tractable microscopic toy models
- Bottom up
  - DFT (or quantum chemistry) $\rightarrow$ electronic structure
  - Time-dependent DFT /MD $\rightarrow$ vibrational spectra
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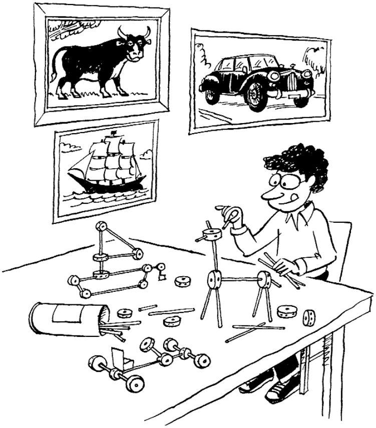

Illustration by Dick Codor.
[Auerbach, Interacting Electrons (Springer, 1998)]

[From Auerbach, Interacting electrons and quantum magnetism]

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Toy models

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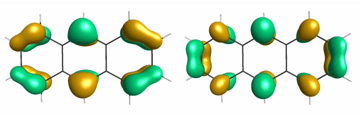

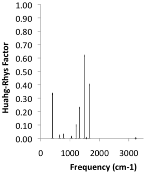

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- *Each* molecule: two DoF
  - Electronic state: 2LS
  - Vibrational state: harmonic oscillator

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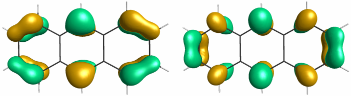
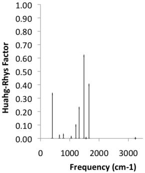
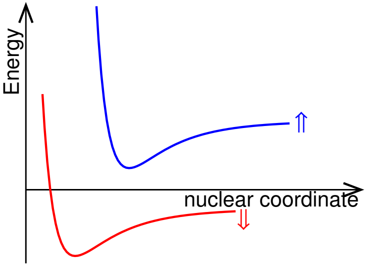




Q2. Focus on low-energy effective theory

- Two-level system, HOMO/LUMO
- Single DoF PES



See also [Galego, Garcia-Vidal, Feist. PRX '15]

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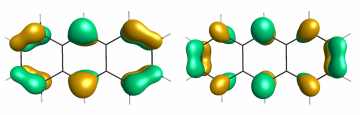

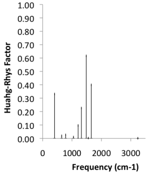

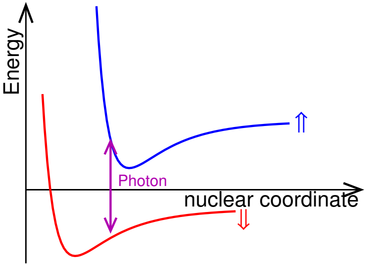

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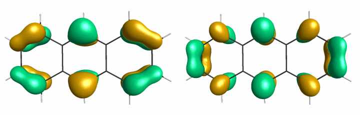

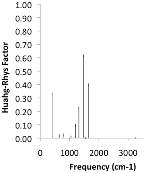

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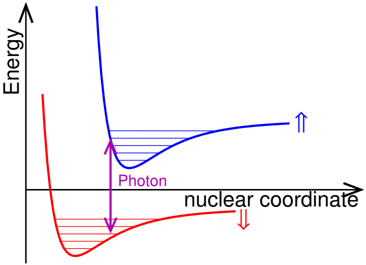

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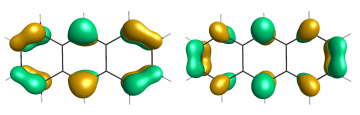

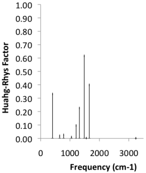

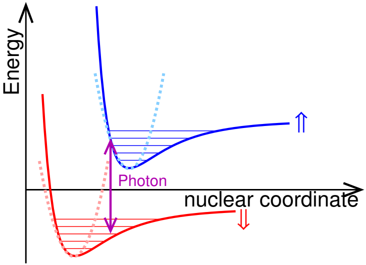

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Tavis-Cummings & Dicke model

Model capable of lasing & condensation

- Tavis-Cummings / Dicke model

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- Weak pumping $\rightarrow$ Superradiance/BEC transition
- High temperature: Maxwell-Bloch laser

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Szymanska et al. PRL 06; Keeling et al. book chapter 1010.3338

Holstein-Tavis-Cummings & Holstein-Dicke model

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- Few emitters (molecules/quantum dots)
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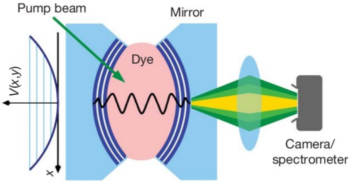
Introduction and models

Weak coupling: Photon BEC

Photon BEC experiments



- (Curved) microcavity
- R6G dye (in solvent)
- Thermalisation of light
- Condensation at $P > P_{th}$

[Klaers et al, Nature, 2010]

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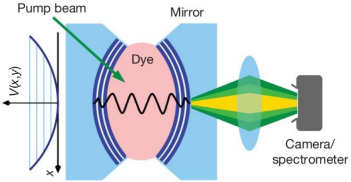

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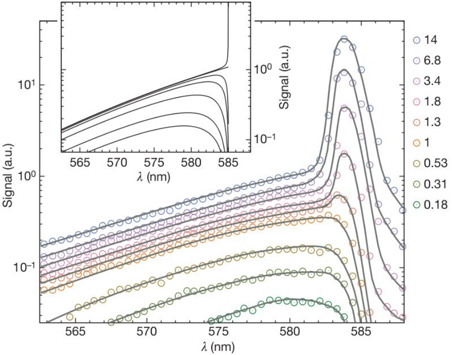

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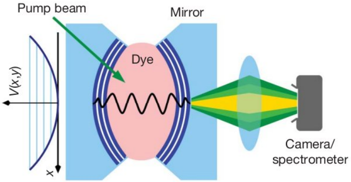

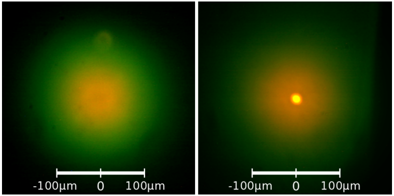

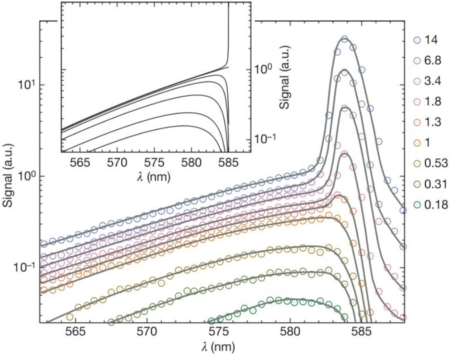

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Photon: Microscopic Model

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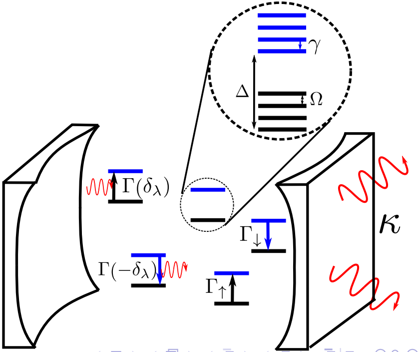
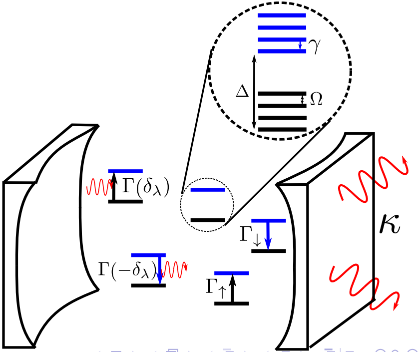
- **2D** harmonic oscillator
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in g



Photon: Microscopic Model

$$H = \sum_m \omega_m \hat{a}_m^\dagger \hat{a}_m + \sum_{i=1}^{N} \Big[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ \hat{a}_m + \text{H.c.} \right)$$
$$+ \omega_v \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \Big]$$

- **2D** harmonic oscillator
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.



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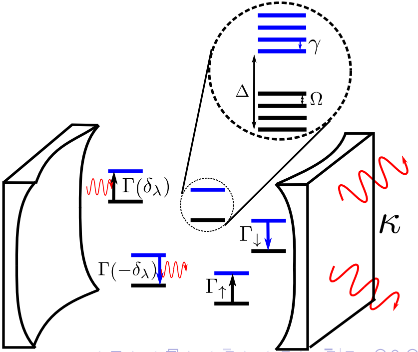

Microscopic model – all orders in λ_0

- Polaron transform (exact), $H = \sum_m \omega_m \psi_m^\dagger \psi_m + \sum_\alpha h_\alpha$,

$$h_\alpha = \frac{\omega_X}{2}\sigma_\alpha^z + g\left(\psi_m \sigma_\alpha^+ D_\alpha + \text{H.c.}\right) + \omega_v b_\alpha^\dagger b_\alpha, \qquad D_\alpha = e^{2\lambda_0(\hat{b}_\alpha - \hat{b}_\alpha^\dagger)}$$

- Master equation

$$\dot{\rho} = -i[H_0,\rho] + \sum_m \frac{\kappa}{2}\mathcal{L}[\psi_m] + \sum_\alpha \left[\frac{\Gamma_\uparrow}{2}\mathcal{L}[\sigma_\alpha^+] + \frac{\Gamma_\downarrow}{2}\mathcal{L}[\sigma_\alpha^-]\right]$$
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$$\Gamma(\delta) = 2g^2\,\mathrm{Re}\left[\int dt\, e^{-i\delta t - (\Gamma_\uparrow + \Gamma_\downarrow)t/2}\langle D_\alpha^\dagger(t) D_\alpha(0)\rangle\right]$$

[Marthaler et al PRL '11, Kirton & JK PRL '13]

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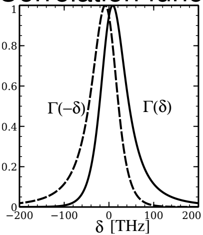

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Steady state populations and equilibrium

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[Kirton & JK PRL '13]

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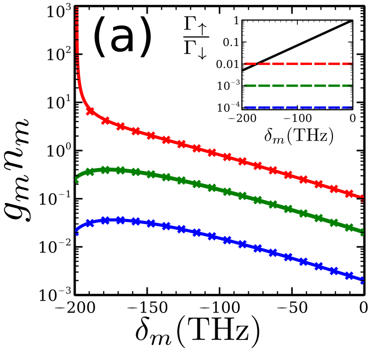

Low loss: Thermal

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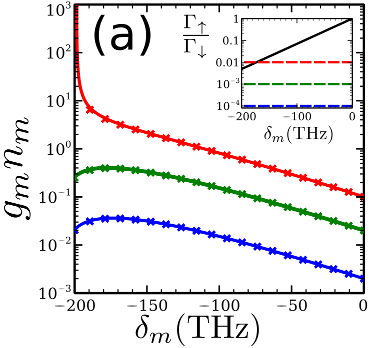

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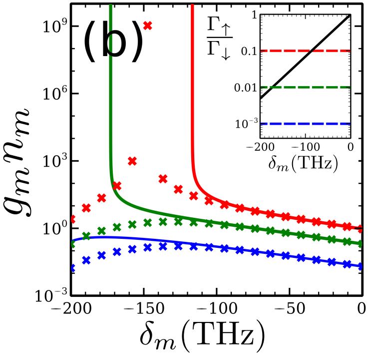

High loss → Laser

[Kirton & JK PRL '13]

Chemical potential?

- Steady state distribution:

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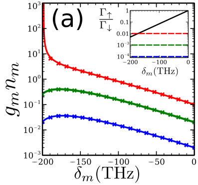

- $\kappa \ll N\Gamma(\delta)$, Kennard-Stepanov

$$\frac{n_m}{n_m+1} = e^{-\beta\delta_m+\beta\mu}, \qquad e^{\beta\mu} \equiv \frac{N_\uparrow}{N_\downarrow} = \frac{\Gamma_\uparrow + \sum_m \Gamma(\delta_m) n_m}{\Gamma_\downarrow + \sum_m \Gamma(-\delta_m)(n_m+1)}$$

- Below threshold,

$$\mu = k_B T \ln[\Gamma_\uparrow/\Gamma_\downarrow]$$

- At/above threshold, $\mu \to \delta_0$

[Kirton & JK, PRA '15]

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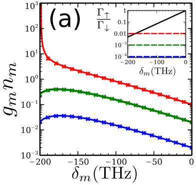

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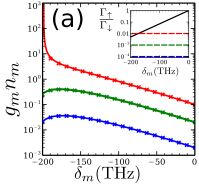

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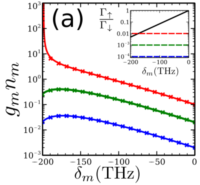

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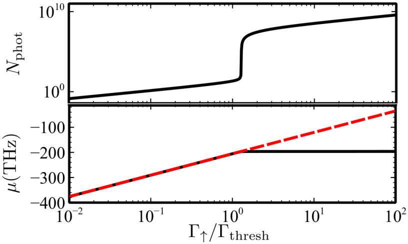

[Kirton & JK, PRA '15]

Weak coupling: Photon BEC

Spatially varying pump intensity

- Consider effects of pump profile, $\Gamma_\uparrow(\mathbf{r}) = \dfrac{\Gamma_\uparrow \exp\left(-r^2/2\sigma_p^2\right)}{(2\pi\sigma_p^2)^{d/2}}$
- Experiments: [Marelic & Nyman, PRA '15]

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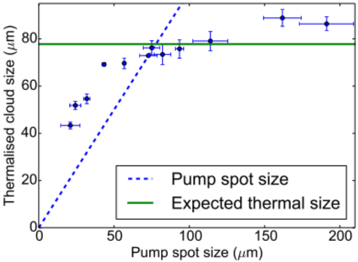

Modelling spatial profile.

- Varying excited density – differential coupling to modes

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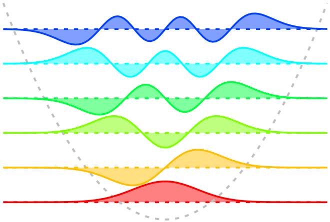
  $I(\mathbf{r}) = \sum_m n_m |\psi_m(\mathbf{r})|^2$



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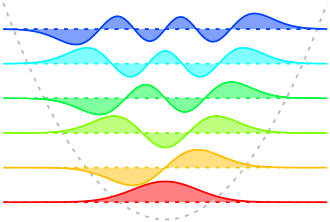

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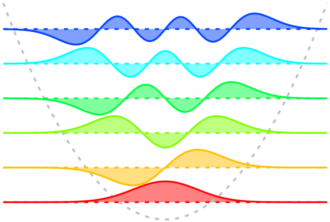
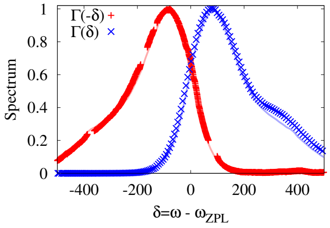


- Use exact R6G spectrum



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Spatially varying pump: below threshold

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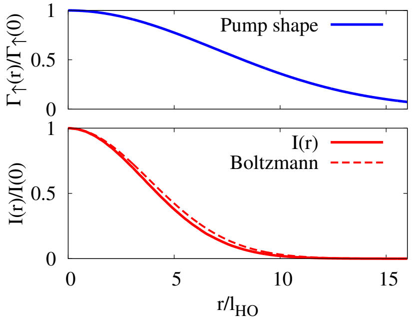

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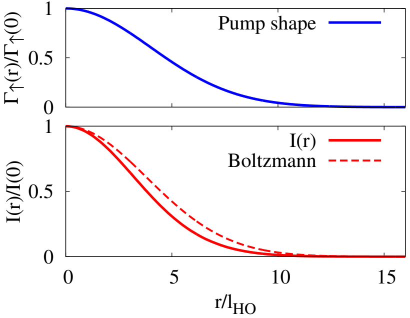

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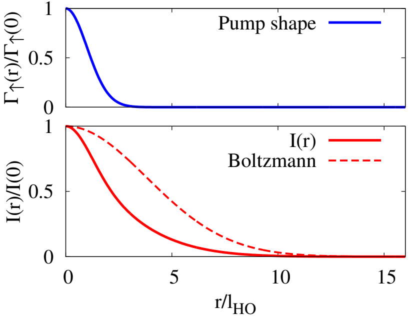

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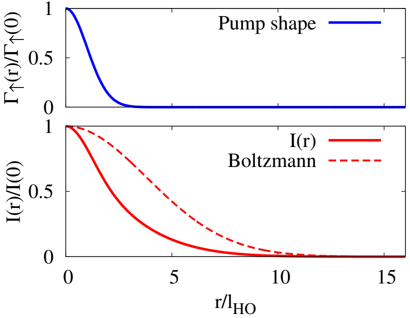
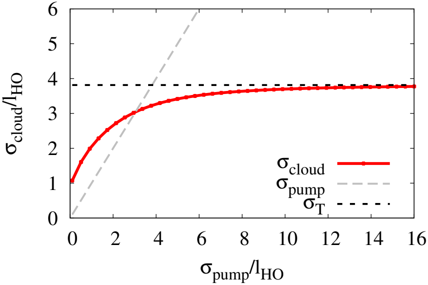
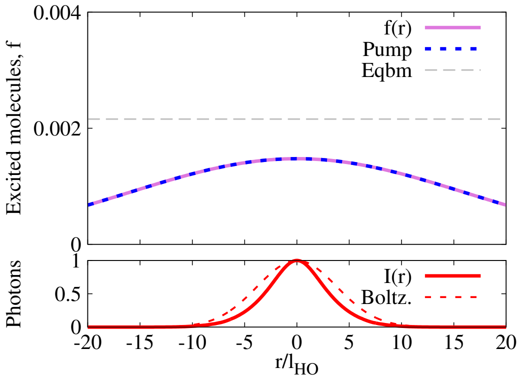




Near threshold behaviour



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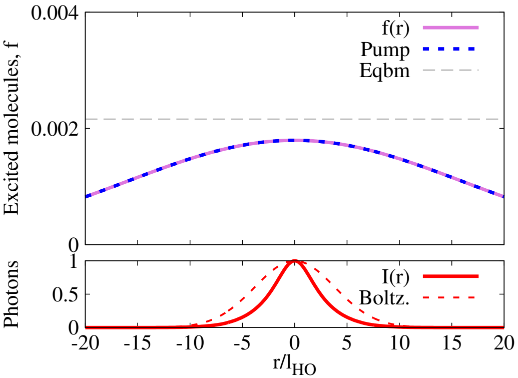

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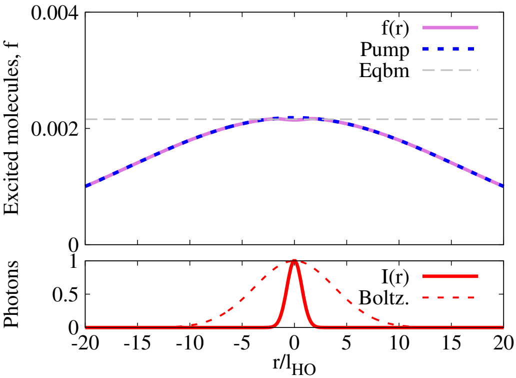

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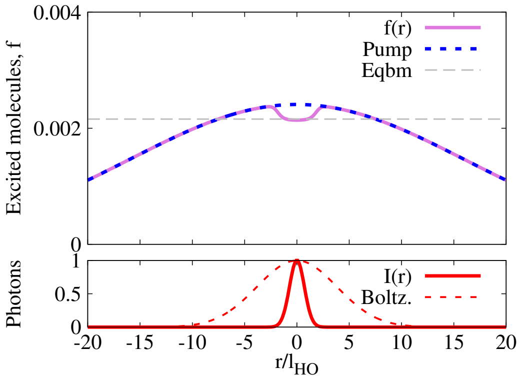

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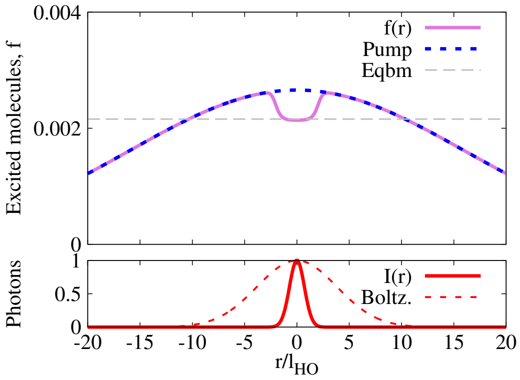

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Off centre pumping; oscillations

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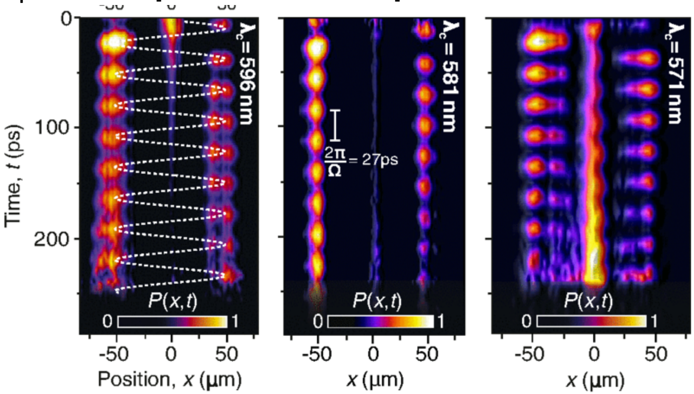

- Oscillations in space – beating of normal modes
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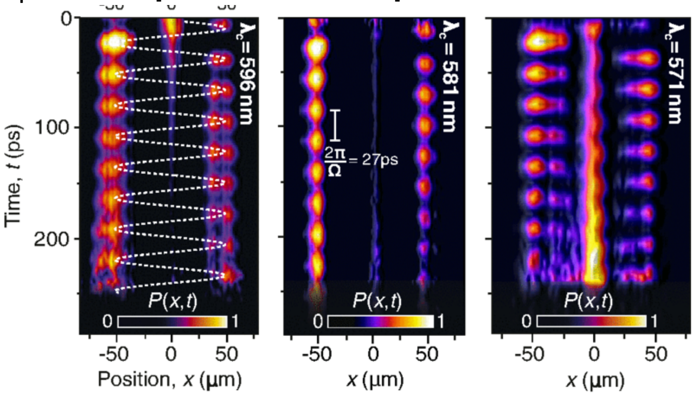

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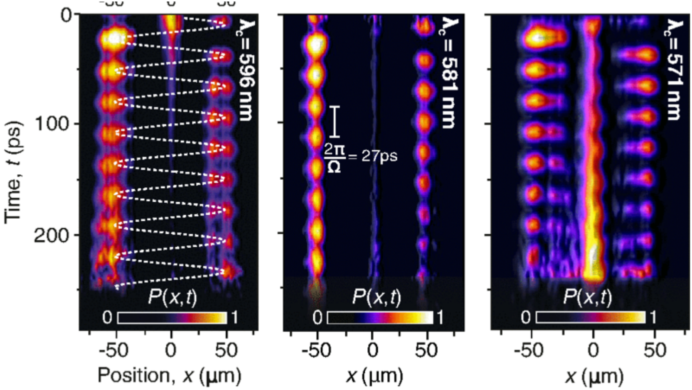

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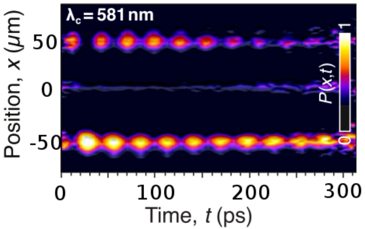
Limit of rate equations



$$\partial_t n_m = -\kappa n_m + \Gamma(-\delta_m)(n_m + 1)N_\uparrow - \Gamma(\delta_m) n_m N_\downarrow$$

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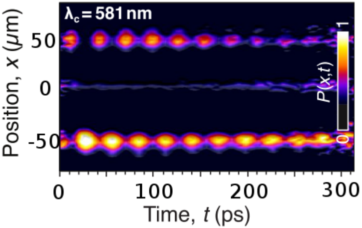

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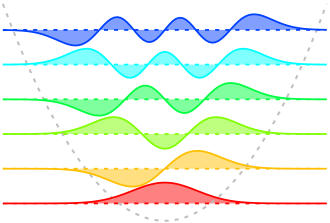
Emission into Gauss-Hermite mode m:

$$I(x) = \sum_m n_m |\psi_m(x)|^2$$



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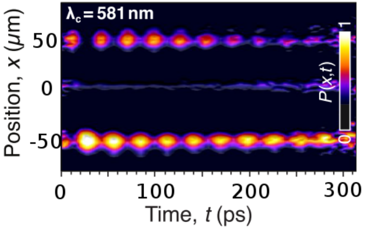

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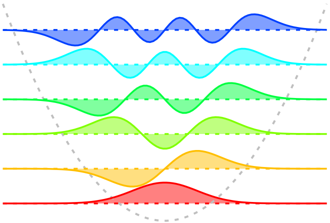

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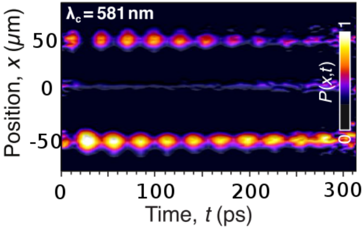

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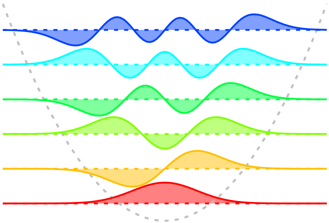

Emission must create coherence between non-degenerate modes.

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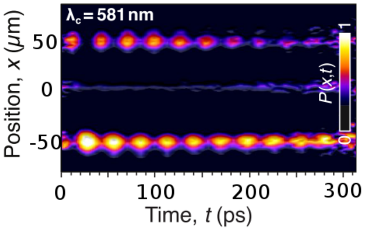

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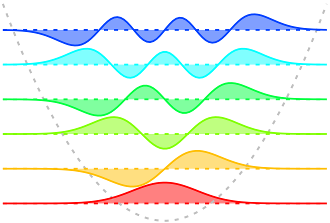

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Modelling

- Wavepacket emission: use Redfield theory:

$$\partial_t \rho = -i\left[\sum_m \omega_m a_m^\dagger a_m, \rho\right] + \sum_{m,m',i} \psi_m^*(r_i)\psi_{m'}(r_i)\Big(K(\delta_{m'})[\hat{a}_{m'}\hat{\sigma}_i^+\hat{\rho}, \hat{a}_m^\dagger\hat{\sigma}_i^-]$$

$$+ K(-\delta_m)[\hat{a}_m^\dagger\hat{\sigma}_i^-\hat{\rho}, \hat{a}_{m'}\hat{\sigma}_i^+]\Big) + \text{H.c.} + (\textit{pumping}, \textit{decay}\ldots),$$

- $K(\delta)$ analytic continuation of $\Gamma(\delta)$.
- Not secular approximation
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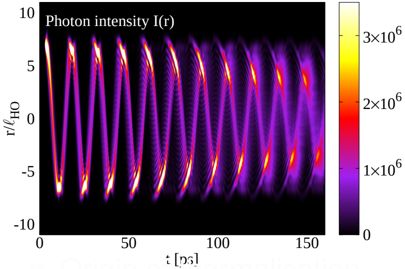
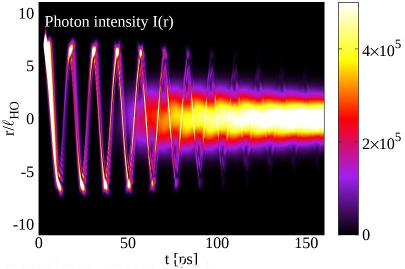
Dynamics from model

Longer cavity

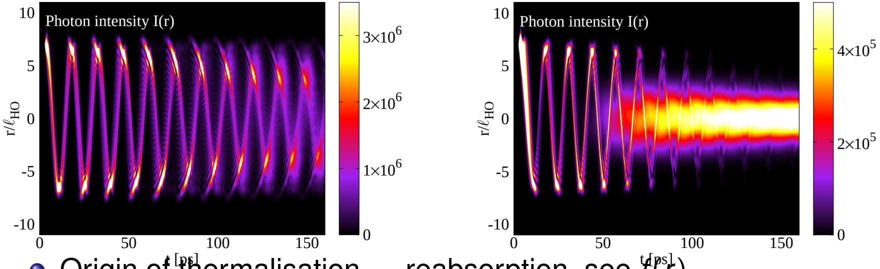


Shorter cavity



Dynamics from model

Longer cavity Shorter cavity



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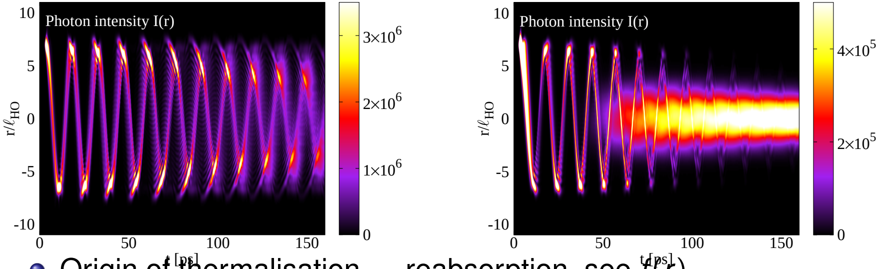

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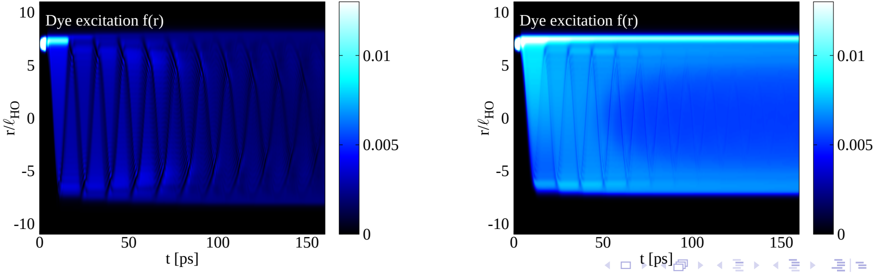
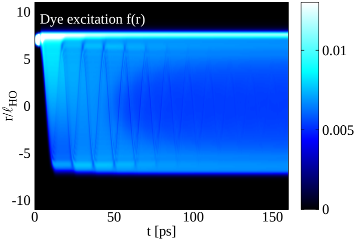


Thermalisation at late times

- Reabsorption “fills-in” excited molecules
- Reach thermal equilibrium, $f = [e^{-\beta\delta_0} + 1]^{-1}$



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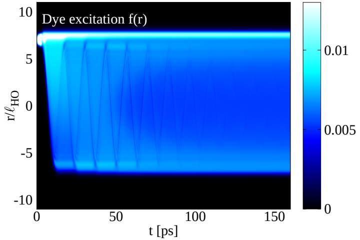

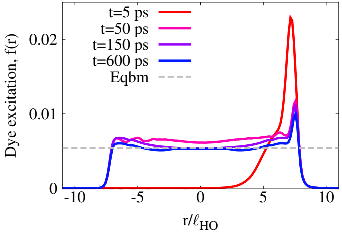

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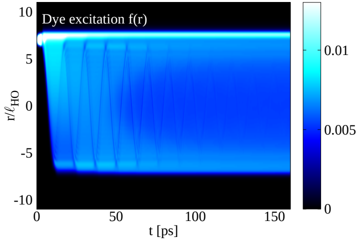

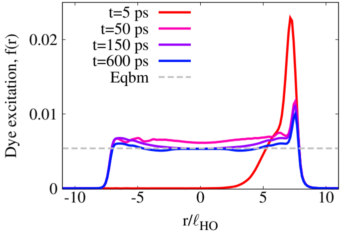

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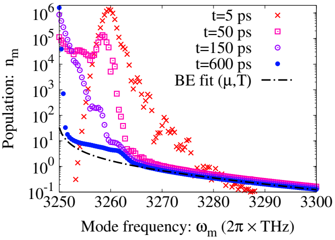

Strong coupling: polariton states

1. Introduction and models
   - Holstein-Dicke model

2. Weak coupling: Photon BEC
   - Homogeneous model & threshold
   - Spatial profile
   - Spatial dynamics

3. Strong coupling: polariton states
   - Exact solutions
   - Scaling with N

One excitation subspace, questions

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^{N} \left[\omega_X \sigma_i^+ \sigma_i^- + g\left(\sigma_i^+ \hat{a} + \text{H.c.}\right) \right.$$
$$\left. + \omega_v \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Rotating wave approximation — Holstein Tavis Cummings
- 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:

$$W(x,p) = \int dy \langle x + y/2|\rho|x - y/2\rangle_i e^{iyp}, \qquad \left(\frac{\hat{b}_i + \hat{b}_i^\dagger}{\sqrt{2}}\right)|x\rangle_i = x|x\rangle_i$$

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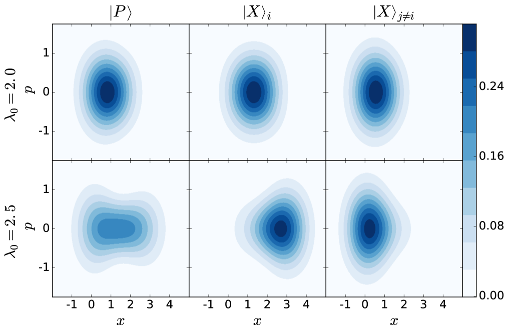

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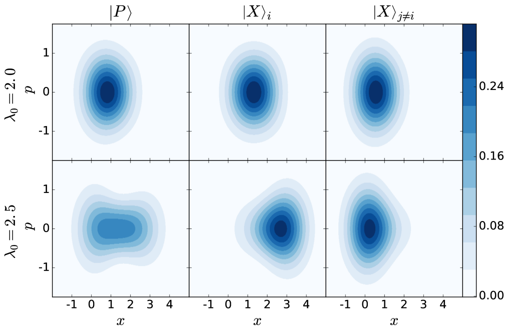

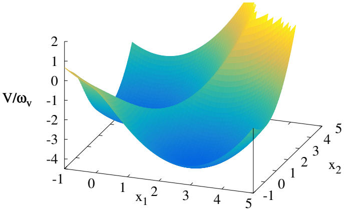

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Exact solution, larger N

- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$
- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5-6$

- Increasing N, suppress $W_{|P\rangle}(x \neq 0)$
- Distinct behaviour vs λ_0
- Exact energy and state vs ω_R, λ_0 for validation

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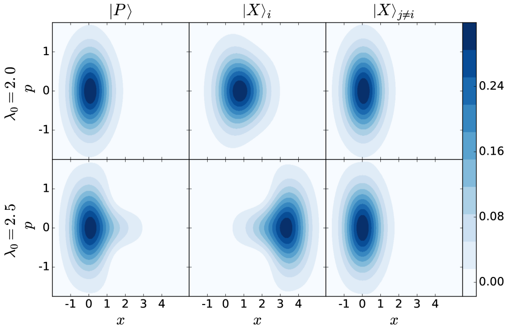

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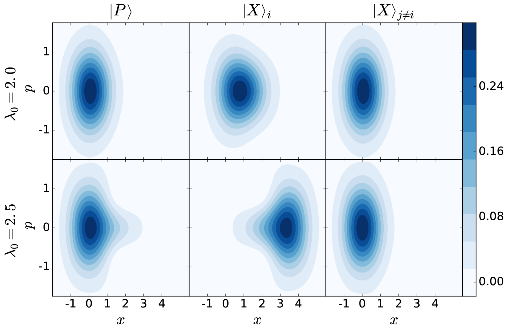

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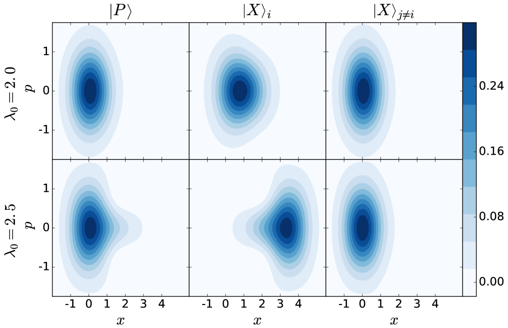

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Extending to arbitrary *N*, polaron ansatz

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

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

- Extend to *N* sites

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

[Wu *et al.* arXiv:1608.08019, Zeb *et al.* arXiv:1608.08929]

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[Wu *et al.* arXiv:1608.08019, Zeb *et al.* arXiv:1608.08929]

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[Herrera and Spano PRL 2016]

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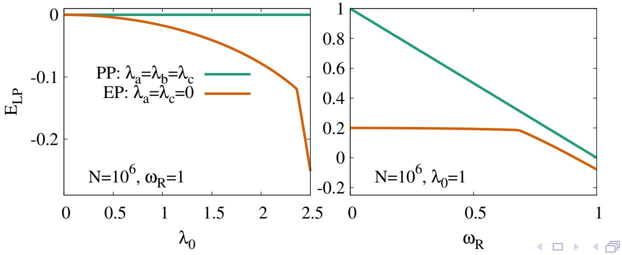

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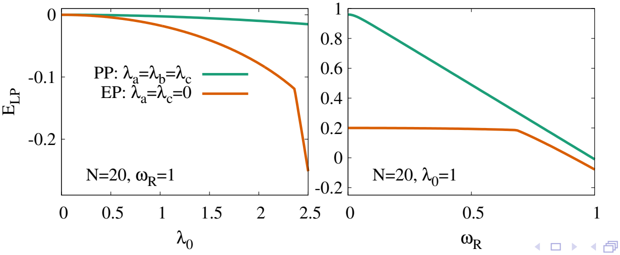

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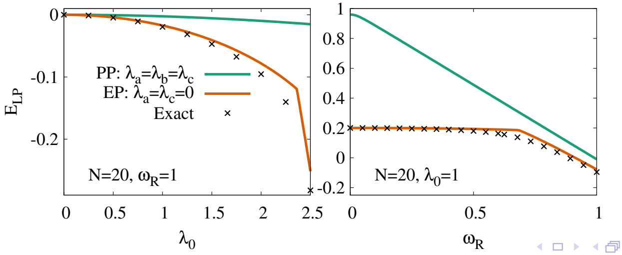

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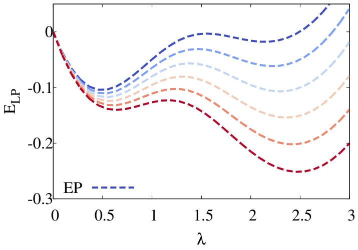

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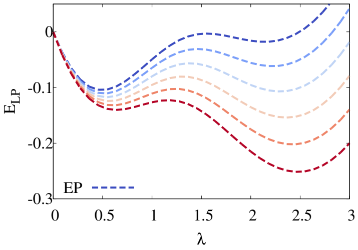

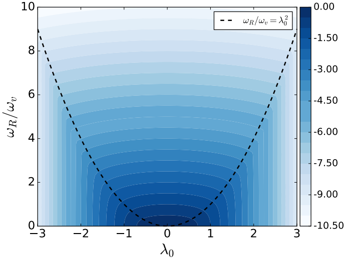

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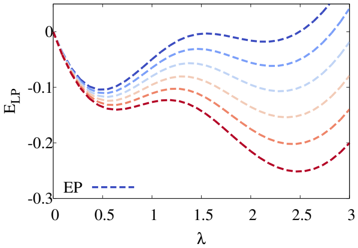

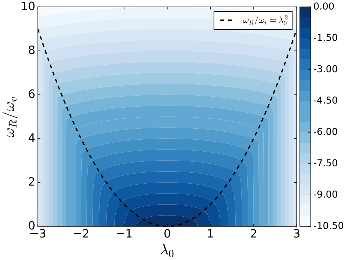

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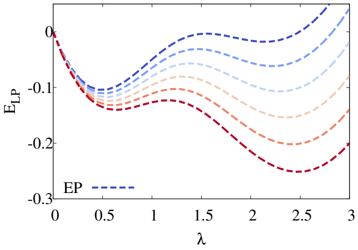

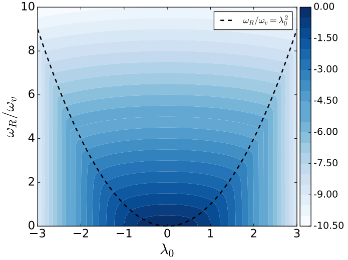

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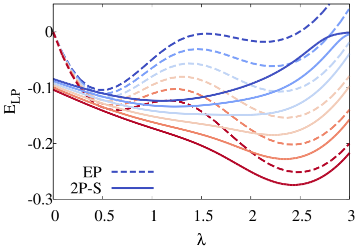

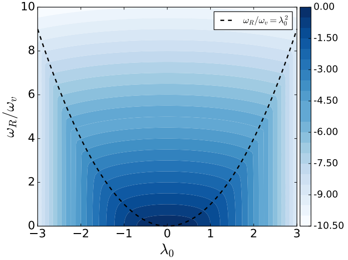

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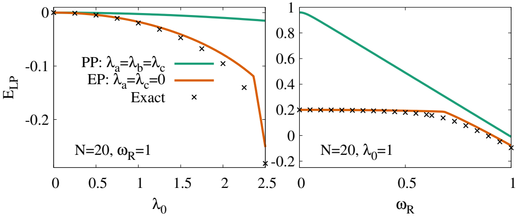
- Accurate energy & wavefunction



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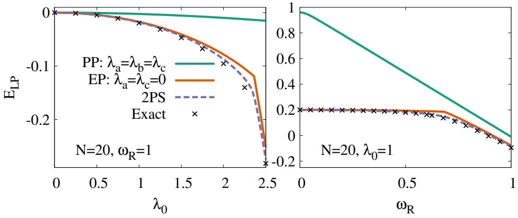

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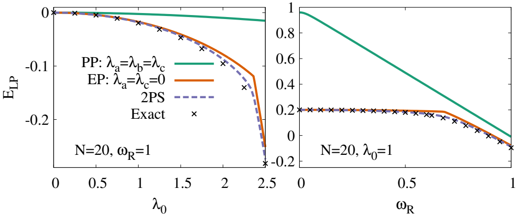

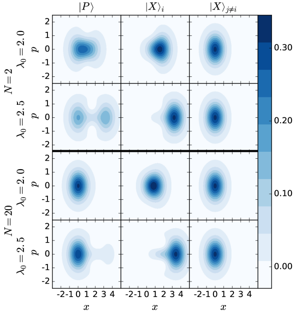

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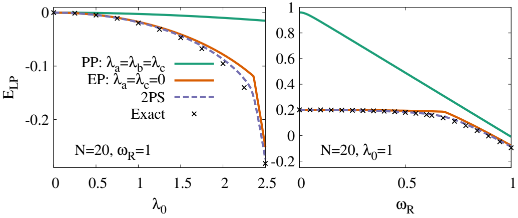

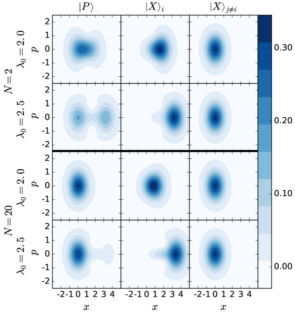

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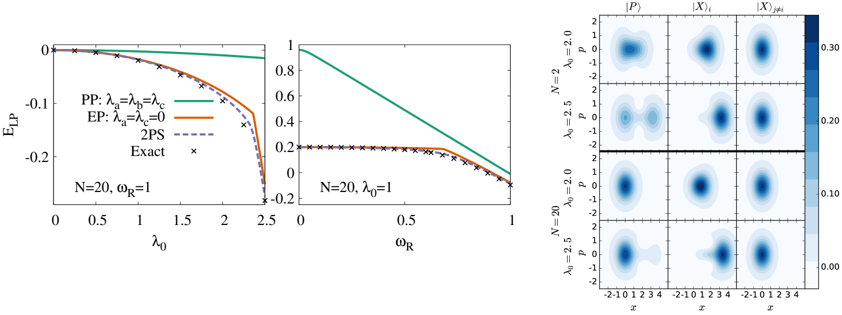

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Engineering and Physical Sciences Research Council

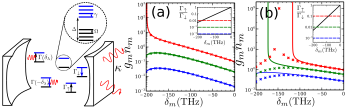
topnes
Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

Hybrid Polaritonics

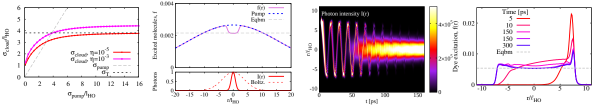
The Leverhulme Trust

Summary

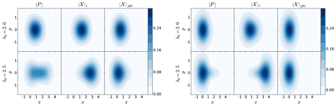
- Photon BEC and thermalisation [Kirton & JK, PRL '13, PRA '15]



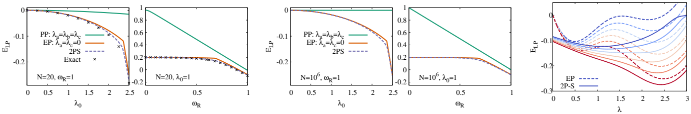
- Photon BEC pattern formation physics [JK & Kirton, PRA '16]



- Single polariton state, Exact solution vs Polaron ansatz

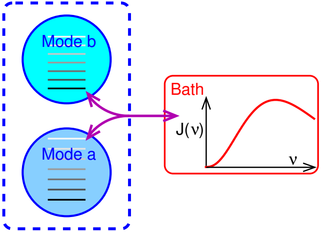


[Zeb, Kirton, JK, `arXiv:1608.08929`]



Toy problem: two bosonic modes

- Basic problem: Emission from thermal bath



$$H = \omega_a \hat{\psi}_a^\dagger \hat{\psi}_a + \omega_b \hat{\psi}_b^\dagger \hat{\psi}_b + H_{\text{Bath}} + (\varphi_a^* \hat{\psi}_a^\dagger + \varphi_b^* \hat{\psi}_b^\dagger) \sum_i g_i \hat{c}_i + \text{H.c.}$$

Toy problem: naïve solutions

- Two "expected" behaviours:
  - At resonance: "weak lasing" — coupling to bath dominates

$$\frac{d}{dt}\rho = \Gamma^{\downarrow}\mathcal{L}[\varphi_a\hat{\psi}_a + \varphi_b\hat{\psi}_b] + \Gamma^{\uparrow}\mathcal{L}[\varphi_a^*\hat{\psi}_a^\dagger + \varphi_b^*\hat{\psi}_b^\dagger]$$

  - Far from resonance: pointer states are eigenstates

$$\frac{d}{dt}\rho = \sum_{i=a,b} \Gamma_i^{\downarrow}\mathcal{L}[\hat{\psi}_i] + \Gamma_i^{\uparrow}\mathcal{L}[\hat{\psi}_i^\dagger]$$

- Explicit derivation → Redfield theory

$$\partial_t\rho = -i[\hat{H},\rho] + \sum_{ij} L_{ij}^{\downarrow}\left(2\hat{\psi}_j\rho\hat{\psi}_i^\dagger - [\rho, \hat{\psi}_i^\dagger\hat{\psi}_j]_+\right) + \sum_{ij} L_{ij}^{\uparrow}\left(2\hat{\psi}_j^\dagger\rho\hat{\psi}_i - [\rho, \hat{\psi}_i\hat{\psi}_j^\dagger]_+\right).$$

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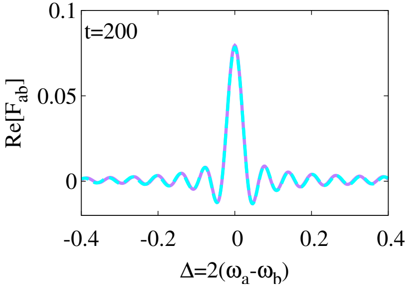

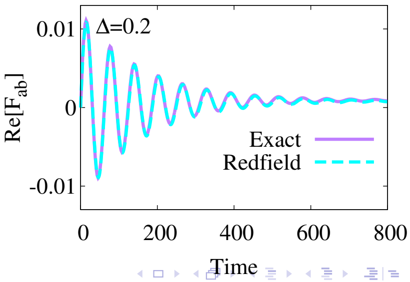

Toy problem: Secularisation

- Secularisation (in eigenbasis of $\hat{H}$): $L_{ij}^{\uparrow,\downarrow} \to L_{ii}^{\uparrow,\downarrow}\delta_{ij} \to F_{ab} = 0$
- Secularisation often invoked to cure negative eigenvalues of $L_{ij}^{\uparrow,\downarrow}$.
- Check stability: consider $f = (F_{aa}, F_{bb}, \mathrm{Re}[F_{ab}], \mathrm{Im}[F_{ab}])$

$$\partial_t \mathbf{f} = -\mathbf{M}\mathbf{f} + \mathbf{f}_0$$

- Eigenvalues of $\mathbf{M}$ exist in closed form:
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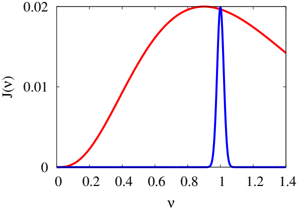

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