From weak to strong matter-light coupling with organic materials

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FOUNDED 1413

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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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- Microscopic model . . .

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Holstein-Tavis-Cummings model

Model capable of lasing & condensation

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

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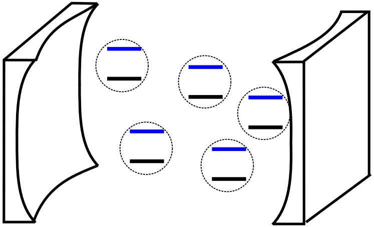

Cwik *et al.* EPL 105 '14; Spano, J. Chem. Phys '15; Galego *et al.* PRX '15; Cwik *et al.* PRA '16; Herrera & Spano PRL '16; Wu *et al.* PRB '16; Zeb *et al.* arXiv:1608.08929; Herrera & Spano arXiv:1610.04252; ...

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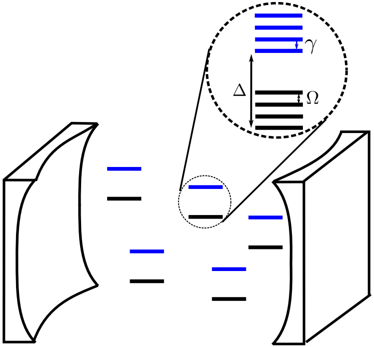

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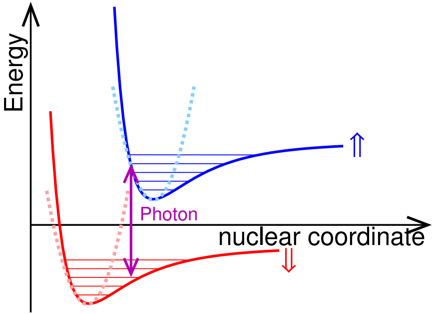

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Introduction and models

Weak coupling

Photon: Microscopic Model

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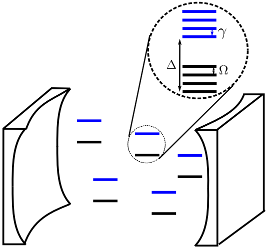

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- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
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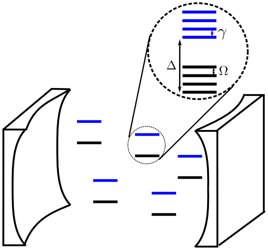

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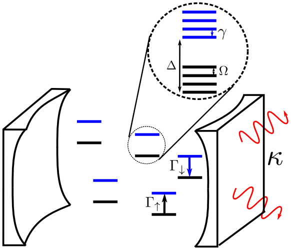

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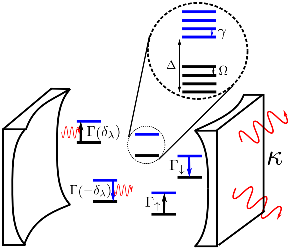

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Steady state populations and equilibrium

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

- Steady state distribution:

$$\frac{n_m}{n_m + 1} = \frac{\Gamma(-\delta_m)N_\uparrow}{\Gamma(\delta_m)N_\downarrow}$$

- Microscopic conditions for equilibrium:
  - Emission/absorption rate:

$$\Gamma(\delta) \simeq 2g^2 \,\mathrm{Re}\left[\int dt e^{-i\delta t}\langle D_\alpha^\dagger(t) D_\alpha(0)\rangle\right] \qquad D_\alpha = \exp\left(2\lambda_0(b_\alpha - b_\alpha^\dagger)\right)$$

[Kirton & JK PRL '13]

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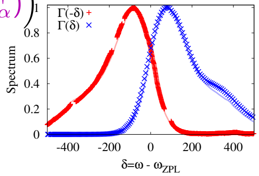

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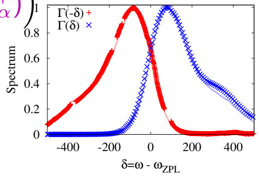

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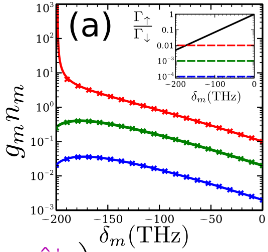

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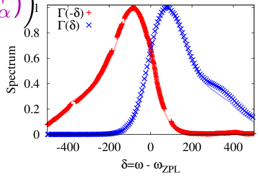

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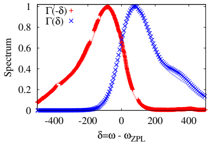
Strong coupling

1. Introduction and models
   - Holstein-Dicke model
2. Weak coupling
   - Photon BEC
3. Strong coupling
   - Exact polariton states, scaling with N
   - Spectrum
4. Ultra strong coupling
   - Vibrational reconfiguration
   - Vibrations and disorder

Strong coupling: One excitation subspace

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

Strong coupling: fate of spectrum



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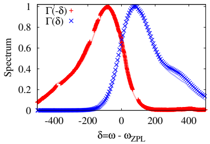

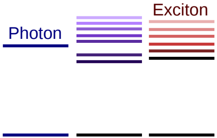

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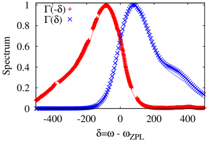

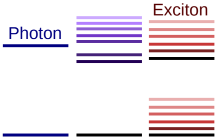

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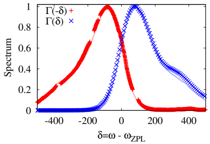

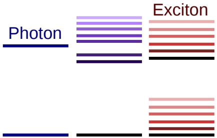

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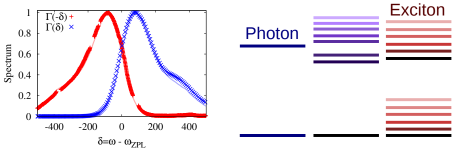

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Exact solution, $N = 20$

- Vibrational Wigner function:

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

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

- Use permutation symmetry

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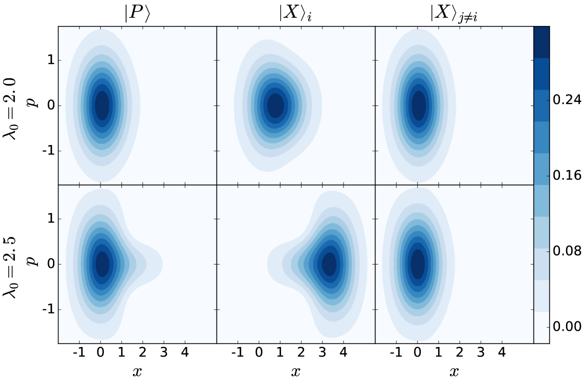

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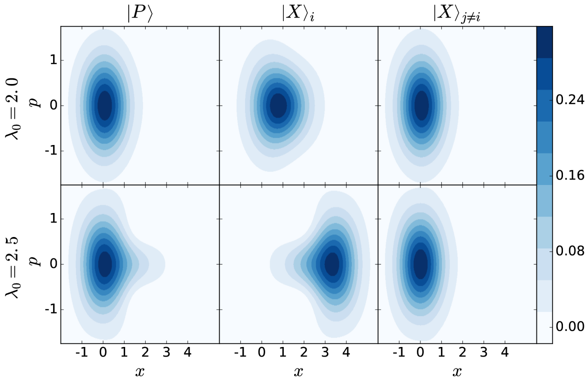

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Extending to arbitrary N, polaron ansatz

- Polaron transform, $\mathcal{D}_i(\lambda) = \exp\left(\lambda(\hat{b}_i^\dagger - \hat{b}_i)\right)$
- N site polaron ansatz

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[Wu *et al.* PRB '16, Zeb *et al.* arXiv:1608.08929]

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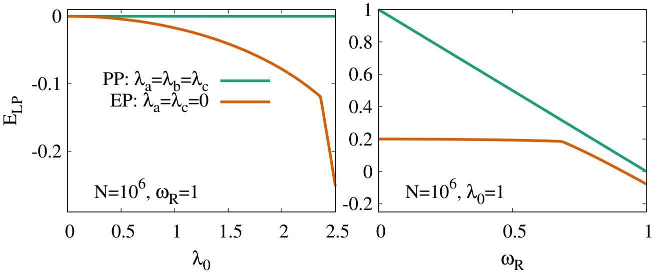

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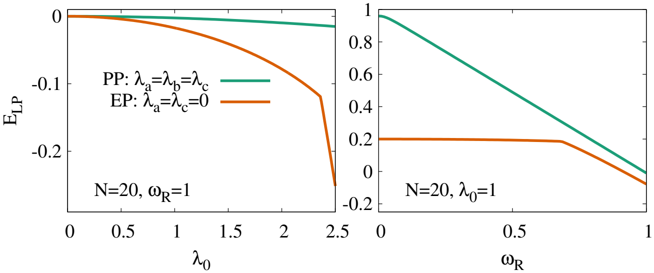

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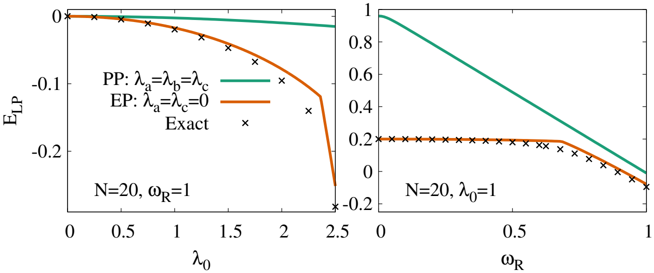

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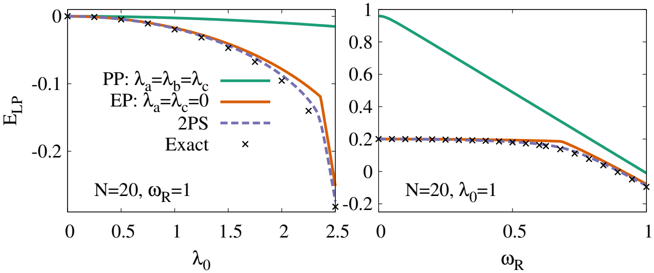

Strong coupling

Tavis-Cummings-Holstein spectrum

- Direct calculation

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

- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger|0\rangle$.
- Mean-field Green's function

- Multiple excitation $\sim 1/N$,

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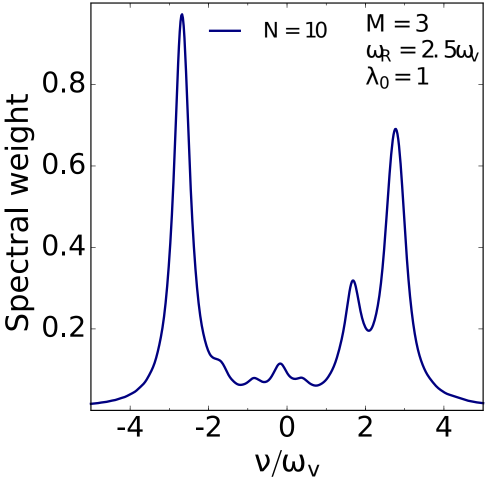

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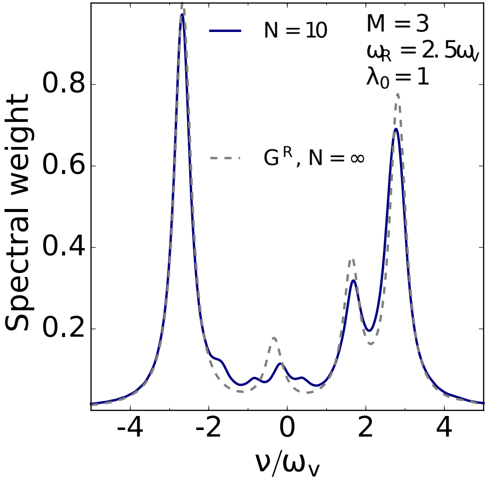
(Classical expression)



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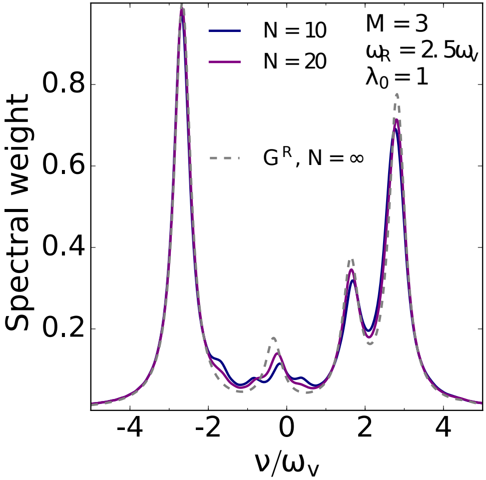

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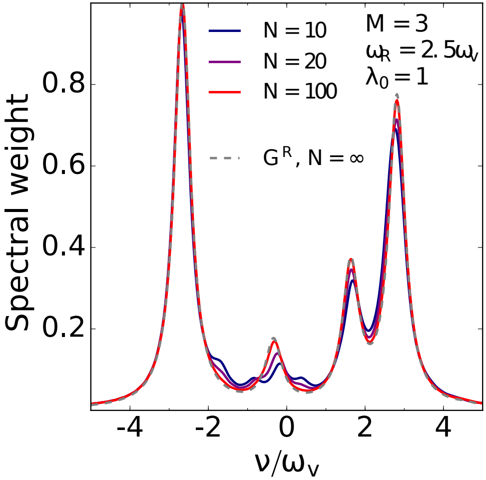

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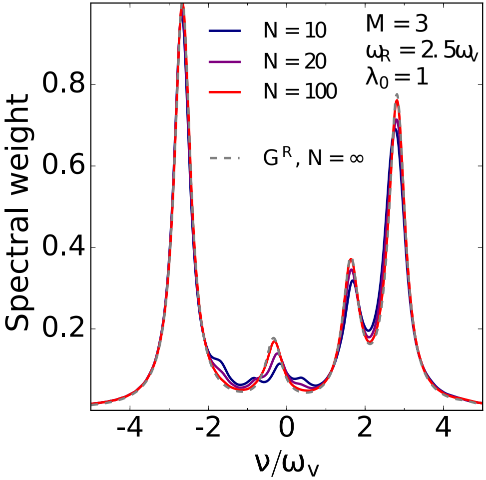

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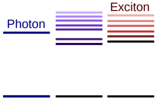

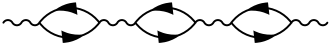

Ultra strong coupling: vibrational reconfiguration

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Vibrational reconfiguration

- Many photon modes, beyond RWA perturbatively

$$H = \sum_k \omega_k \hat{a}_k^\dagger \hat{a}_k + \sum_{i=1}^{N} \left[\omega_X \sigma_i^+ \sigma_i^- + \sum_k g_\mathbf{k} \left(\sigma_i^+ (\hat{a}_k + \hat{a}_k^\dagger) + \text{H.c.} \right) \right.$$

- Reduced vibrational offset

$$\lambda_0 \to \lambda_0 (1 - K_1), \qquad K_1 = \sum_k \frac{g_k^2}{(\omega_k + \omega_X)^2}$$

[Cwik *et al.* PRA '16]

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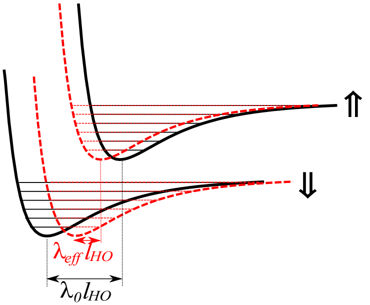

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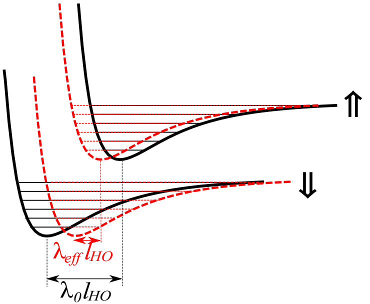
  - But, no collective effect: $\delta H \simeq K_1 N$



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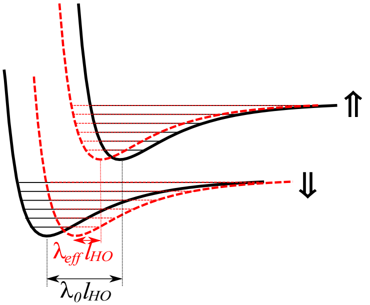
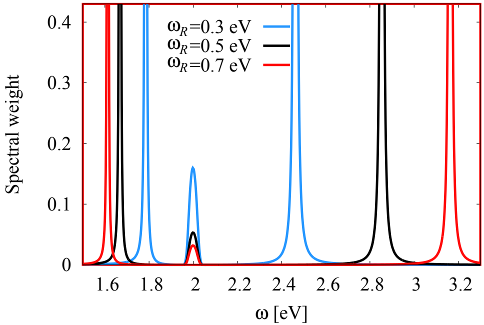


[Cwik *et al.* PRA '16]

Bumps in the middle of the spectrum

- Orgin of bumps in middle of spectrum: Disorder



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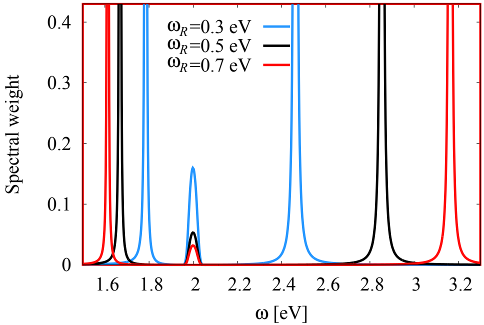


- Central peak:

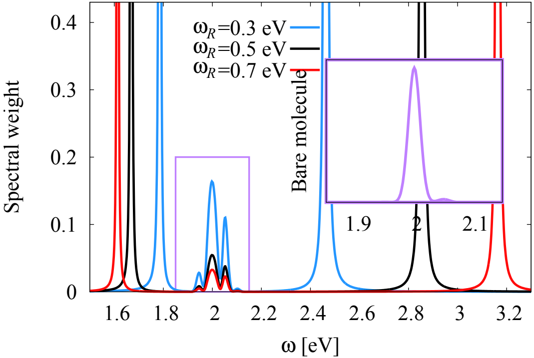
$$D^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_k + \Sigma_X(\nu)}$$

$$\Sigma_X(\nu) = -\int dx \rho(x) \frac{\omega_R^2 |f_m(\lambda_0)|^2}{\nu + i\gamma/2 - x}$$

Gaussian $\rho(x)$, variance σ_x [Houdré *et al.* , PRA '96]

Disorder + Vibrations + Strong coupling

- Disordered spectrum + vibrations, $\lambda_0^2 = 0.02 \ll 1, \sigma_x = 0.01\text{eV}$



[Cwik *et al.* PRA '16]

- Stronger disorder, $\lambda_0^2 = 0.5, \sigma = 0.025\text{eV}$

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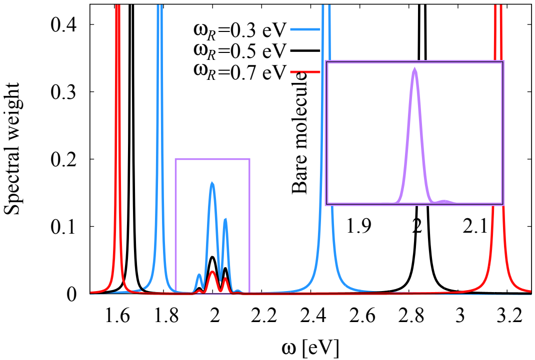

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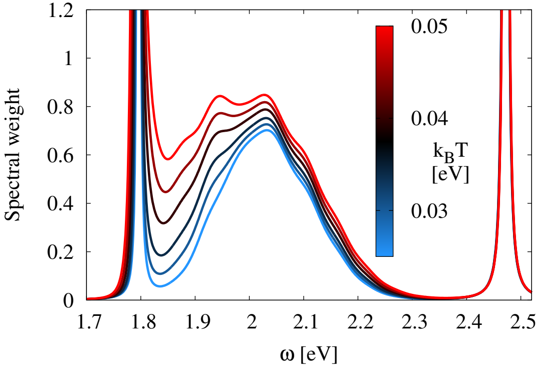
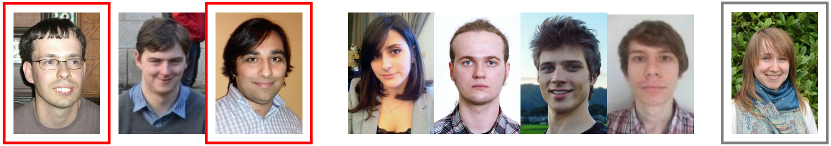


Acknowledgements

GROUP:



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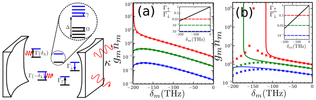
Hybrid
Polaritonics

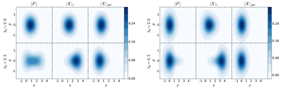
The Leverhulme Trust

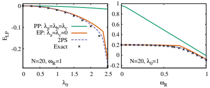
Conclusions

- Matter-light coupling & organic molecules: Holstein-Tavis-Cummings model
- Photon BEC and thermalisation [Kirton & JK, PRL ’13, PRA ’15]

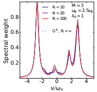


- Strong coupling, one excitation
  - Exact solution & Polaron Ansatz



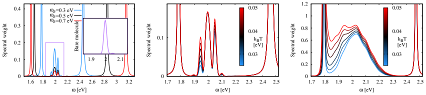


  - Validity of mean-field Green’s functions



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

- Vibrations + disorder + USC



[Cwik *et al.* PRA ’16]

Conclusions

5. Photon BEC Chemical potential
6. Spatial profile
7. Spectrum

Chemical potential?

- Steady state, thermalised:

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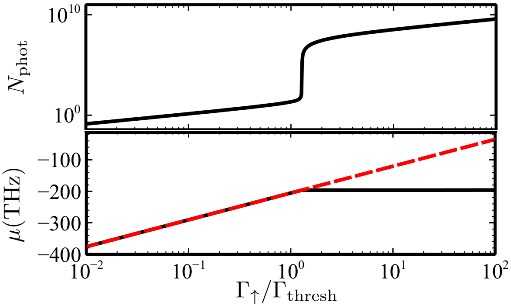

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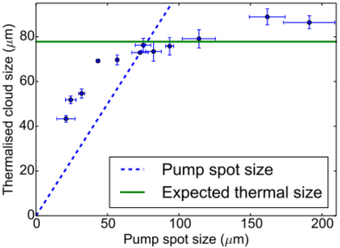
Spatial profile, pump-spot dependence

- Dependence on pump profile, $\Gamma_\uparrow(\mathbf{r}) = \frac{\Gamma_\uparrow \exp\left(-r^2/2\sigma_p^2\right)}{(2\pi\sigma_p^2)^{d/2}}$

  [Marelic & Nyman, PRA '15]



- Use Gauss-Hermite modesÂÿ $I(\mathbf{r}) = \sum_m n_m |\psi_m(\mathbf{r})|^2$
- Varying excited density – differential coupling to modes

$$\partial_t n_m = -\kappa n_m + \Gamma(-\delta_m) O_m (n_m + 1) - \Gamma(\delta_m)(\rho_M - O_m) n_m$$

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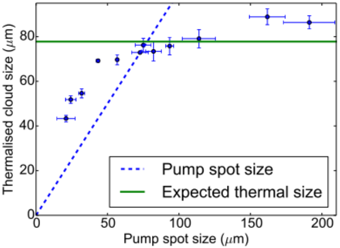

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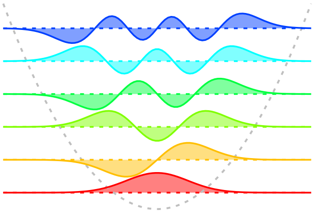

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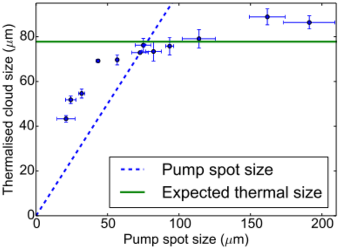

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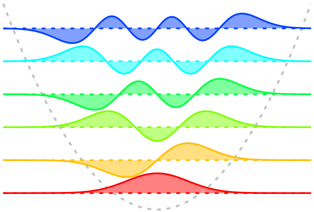

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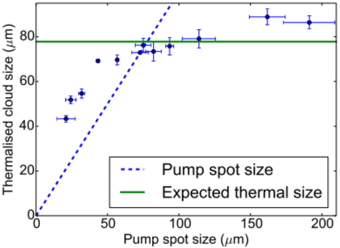

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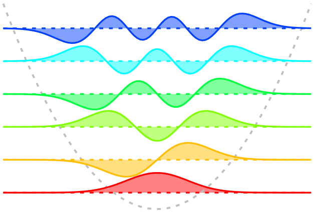

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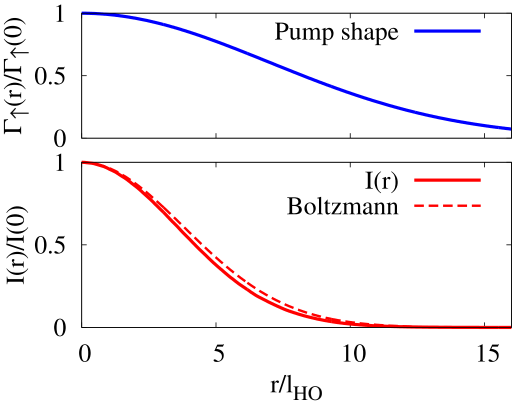

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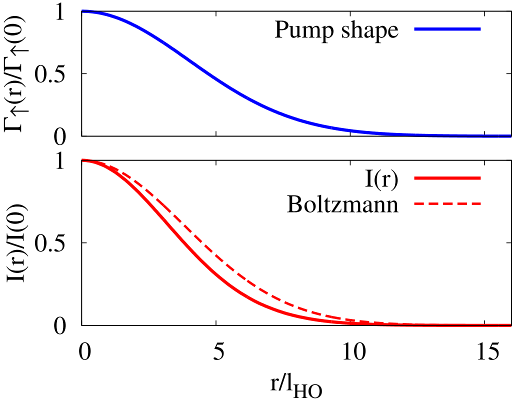

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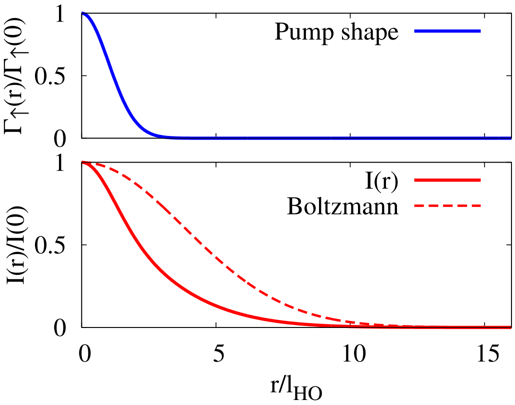

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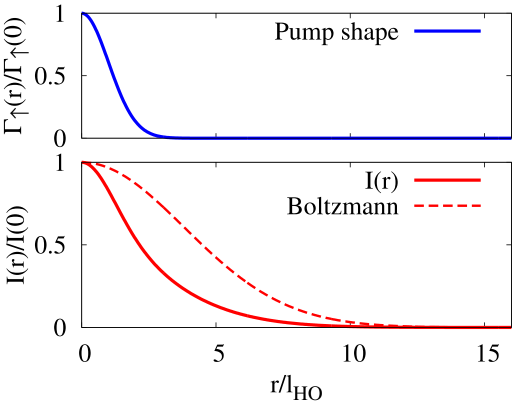

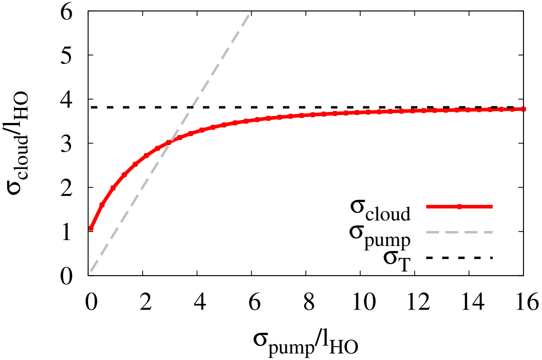

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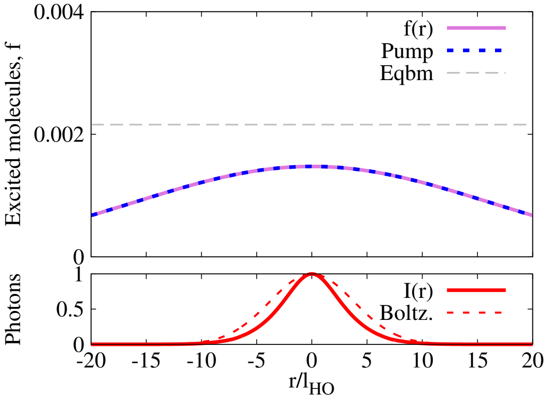

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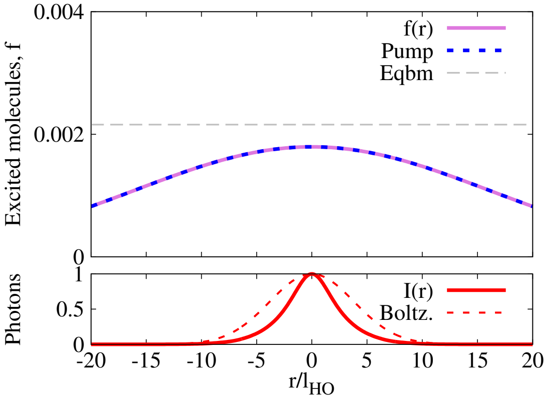

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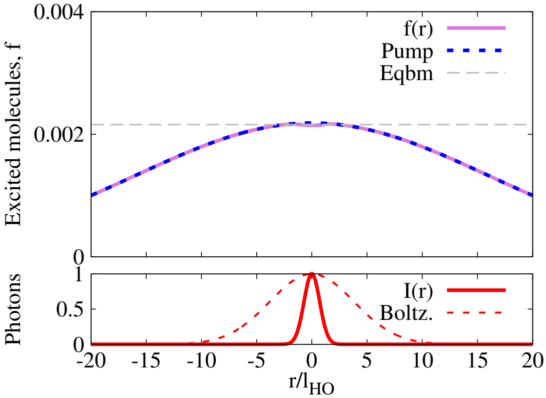

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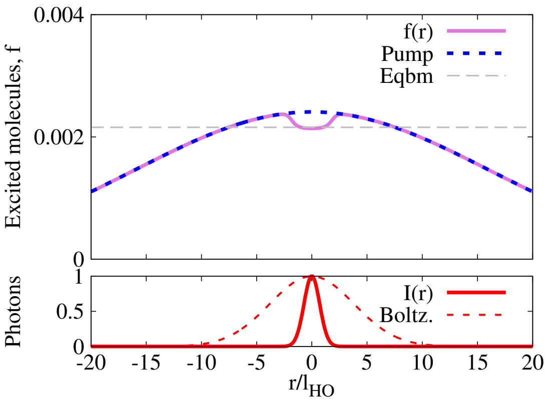

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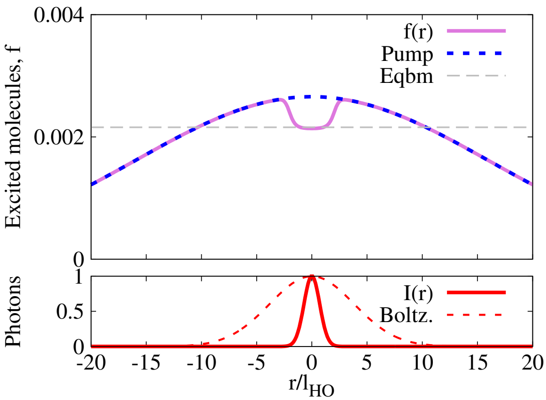

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Calculating spectra: Input-Output formalism

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

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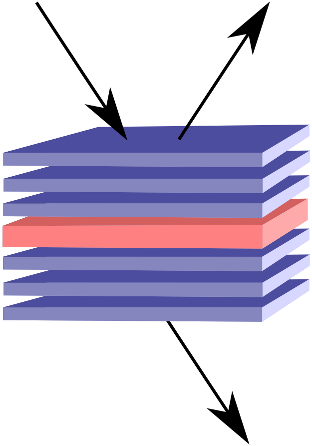

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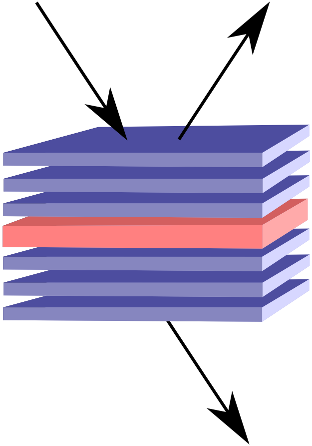

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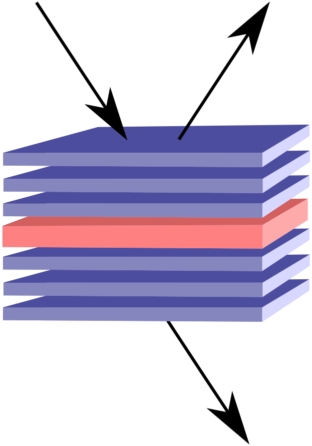

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Tavis-Cummings-Holstein vs Coupled Oscillators

- Diagrammatic approach — equivalent to coupled oscillator model:

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

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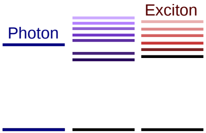

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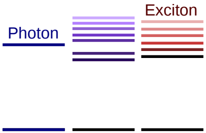

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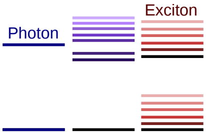

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