(Toward) Modelling strong coupling with organic molecules

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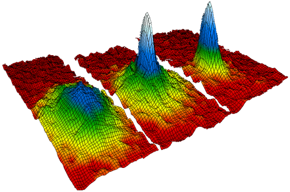
FOUNDED 1413

SUPA

SISSA, April 2017

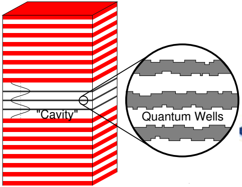
Condensation, Lasing, Superradiance

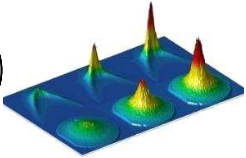
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science '95]

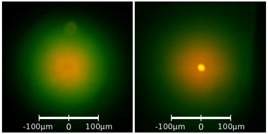
Polariton Condensate $T \sim 20$K





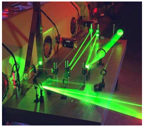
[Kasprzak *et al.* Nature, '06]

Photon Condensate $T \sim 300$K

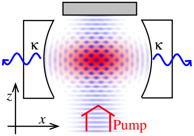


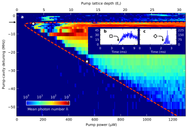
[Klaers *et al.* Nature, '10]

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



Superradiance transition $T \sim 0$

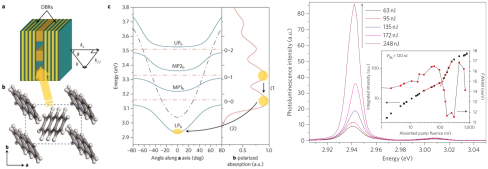




[Baumann *et al.* Nature '10]

Motivation: polariton condensates

- Anthracene Polariton Lasing
  $T \sim 300\text{K}$



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

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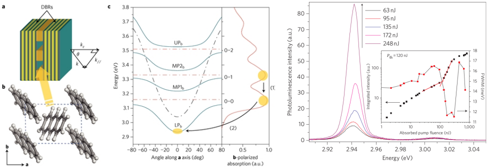

Q1. Vibrational replicas?

Q2. Relevance of disorder?

Q3. Lasing vs condensation?

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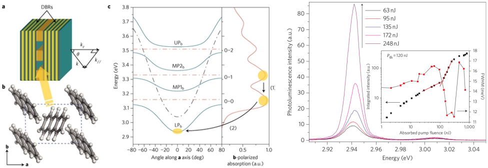

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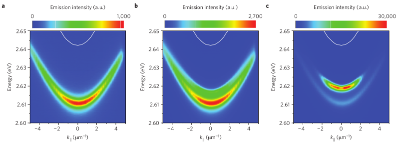

[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14, + many more]

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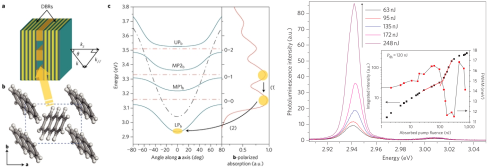

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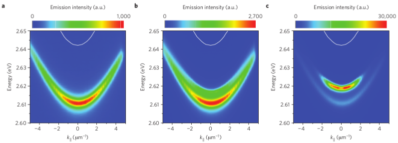


Q1. Frenkel to Wannier crossover?

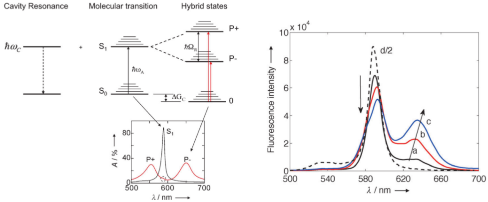
Q2. Optimal vibrational properties?

Q3. Nonlinearities?

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

Motivation: vacuum-state strong coupling

- Linear response (no pump, no condensate): effects of matter-light coupling alone.



[Canaguier-Durand *et al.* Angew. Chem. ’13; Baumberg group]

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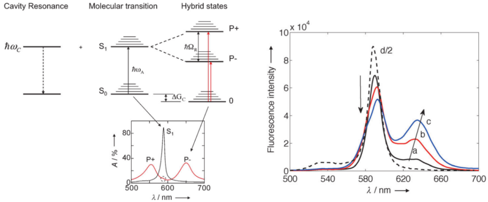


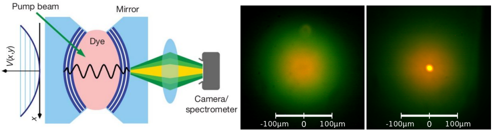
[Canaguier-Durand *et al.* Angew. Chem. '13; Baumberg group]

Q1. Can **ultra-strong** coupling to light change:
- charge distribution?
- vibrational configuration?
- molecular orientation?
- crystal structure?

Q2. Are changes collective ($\sqrt{N}$ factor) or not?

Motivation: photon condensates

- Photon Condensate $T \sim 300$K



[Klaers *et al.* Nature, '10, Marelic *et al.* '15]

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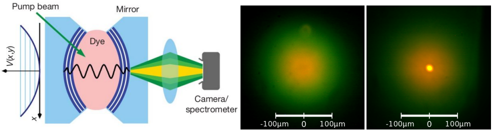

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Q1. Relation to dye laser?

Q2. Relation to polaritons?

Q3. Thermalisation breakdown?

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
  $$i\partial_t\psi = \left(-\nabla^2 + V(r) + U|\psi|^2\right)\psi + i\left(P(\psi, n, r) - \kappa\right)\psi$$
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What kinds of modelling

- 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
  - FDTD/transfer-matrix $\rightarrow$ cavity modes

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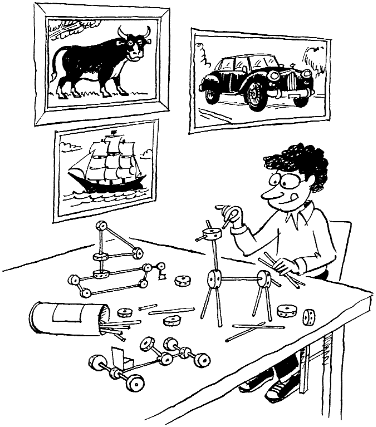

Illustration by Dick Codor.
[Auerbach, Interacting Electrons (Springer, 1998)]

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Toy models

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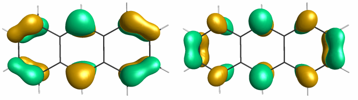

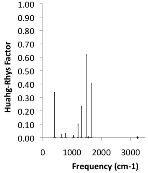

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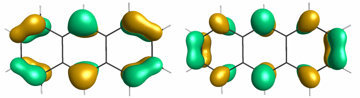

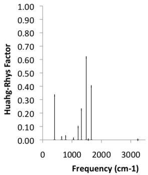

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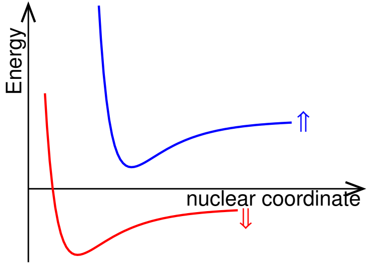
   - Two-level system, HOMO/LUMO
   - Single DoF PES



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

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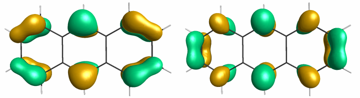

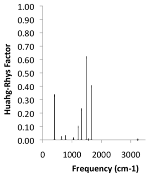

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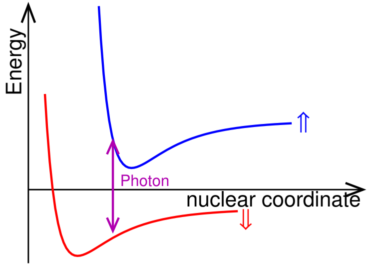

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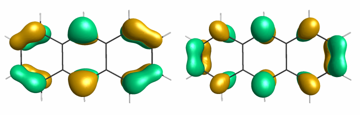

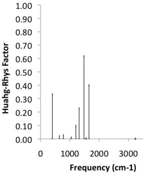

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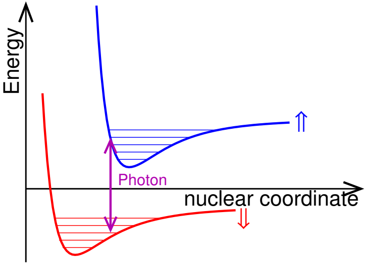

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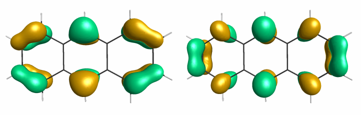

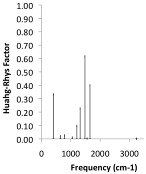

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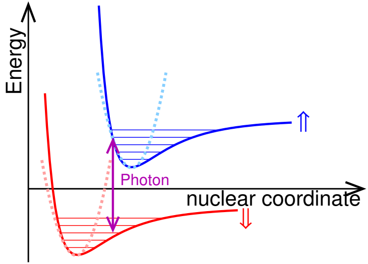

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Tavis-Cummings & Dicke model

Model capable of lasing & condensation

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  - Weak pumping → Superradiance/BEC transition
  - High temperature: Maxwell-Bloch laser

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  Szymanska et al. PRL ’06; Keeling et al. book chapter 1001.3338

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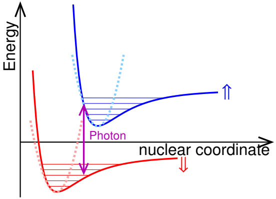

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- Few emitters (molecules/quantum dots)
  Wilson-Rae & Imamoğlu PRB 2002 McCutcheon & Nazir PRB 2011 Roy & Hughes PRB 2011; Bera *et al.* PRB 2014; Pollock *et al.* NJP 2013; Hornecker *et al.* arXiv:1609.09754; …
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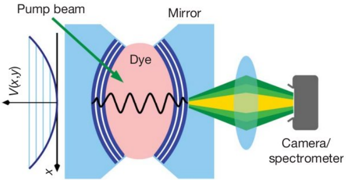
  Cwik *et al.* EPL 105 '14; Spano, J. Chem. Phys '15; Galego *et al.* PRX '15; Cwik *et al.* PRA '16; Herrera & Spano PRL '16; Wu *et al.* PRB '16; Zeb *et al.* arXiv:1608.08929; Herrera & Spano PRL, PRA '17; ...

Weak coupling

Bose-Einstein condensation of photons

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



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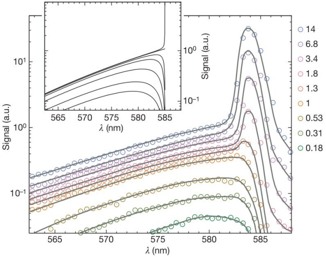

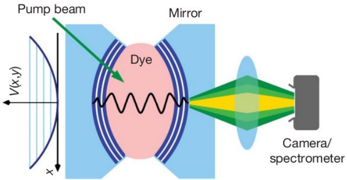

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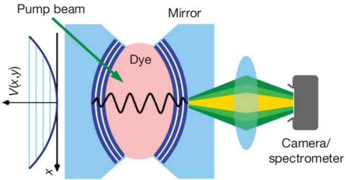

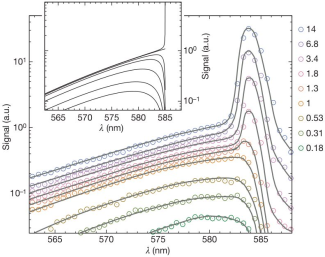

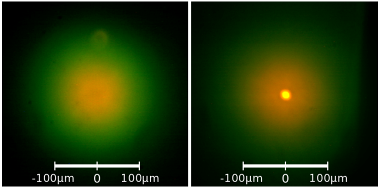

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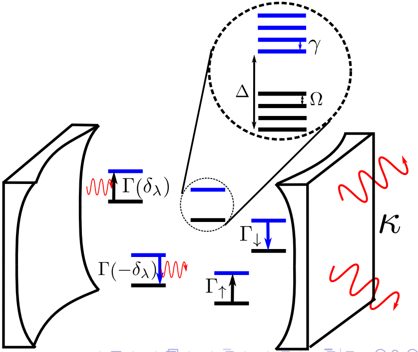
Photon: Microscopic Model

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

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



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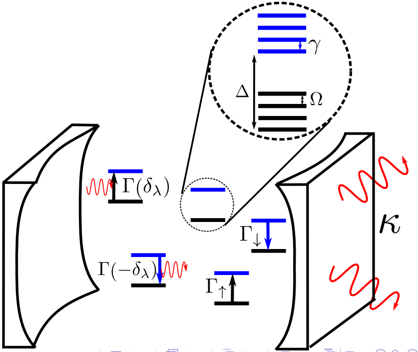

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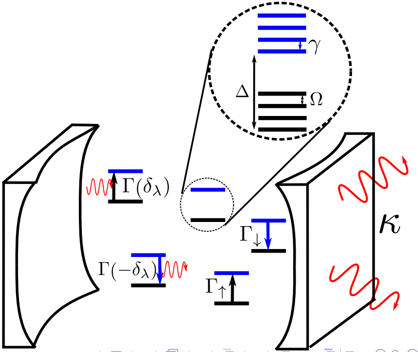

Microscopic model – all orders in λ_0

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

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- Perturbation theory in g, Born-Markov.
- 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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- Correlation function:

$$\Gamma(\delta) = 2g^2 \,\text{Re}\left[\int dt\, e^{-i\delta t - (\Gamma_\uparrow + \Gamma_\downarrow)t/2}\langle D^\dagger_\alpha(t) D_\alpha(0)\rangle\right]$$

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

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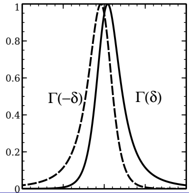

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Steady state populations and equilibrium

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:

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

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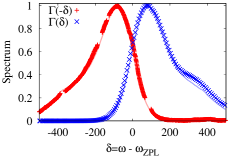

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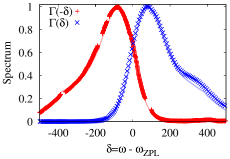

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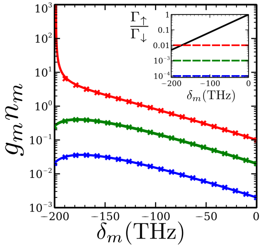

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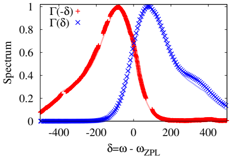

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[Kirton & JK PRL '13]

Chemical potential?

- Steady state, thermalised:

$$\frac{n_m}{n_m+1} = \frac{\Gamma(-\delta_m)N_\uparrow}{\kappa + \Gamma(\delta_m)N_\downarrow} \simeq e^{-\beta\delta_m+\beta\mu},$$

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- 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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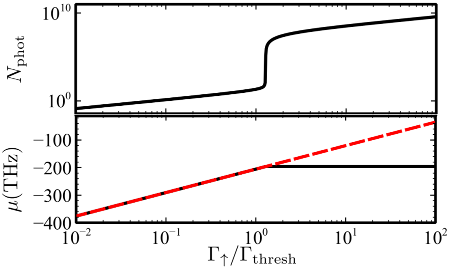

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Weak coupling

Spatially varying pump intensity

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

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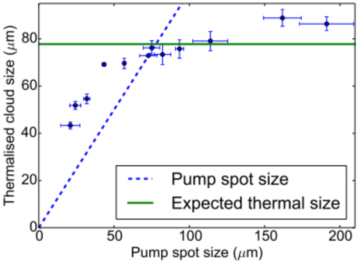

Modelling spatial profile.

- Varying excited density – differential coupling to modes

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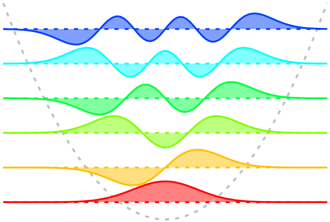
  $I(\mathbf{r}) = \sum_m n_m |\psi_m(\mathbf{r})|^2$



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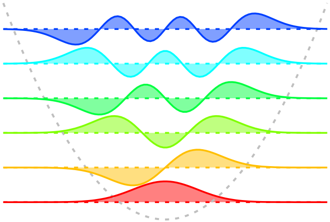

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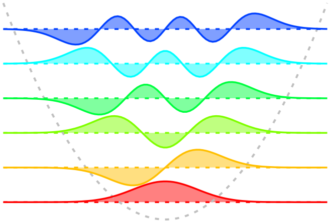
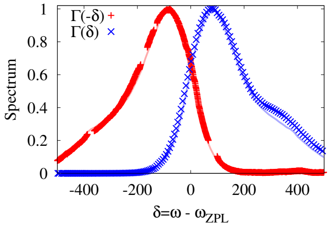


- Use exact R6G spectrum



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Spatially varying pump: below threshold

- Far below threshold:
  - If $\kappa \ll \rho_M \Gamma(\delta_m)$, $$\frac{n_m}{n_m+1} \simeq e^{-\beta\delta_m} \times \int d\mathbf{r} \frac{1}{\rho_M} \rho_\uparrow(\mathbf{r}) |\psi_m(\mathbf{r})|^2$$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$

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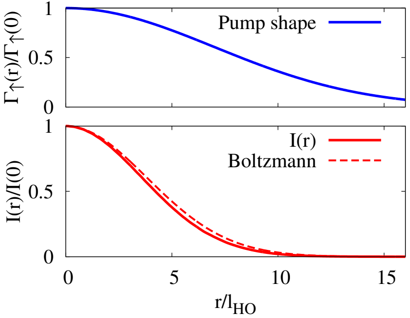

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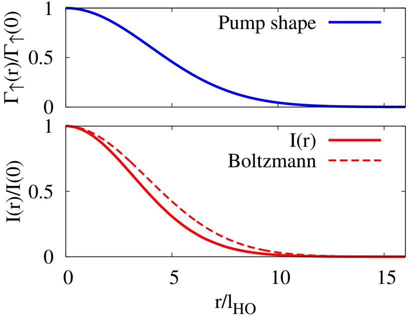

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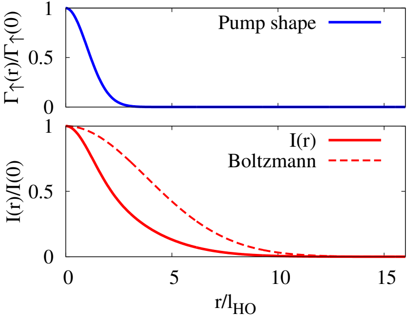

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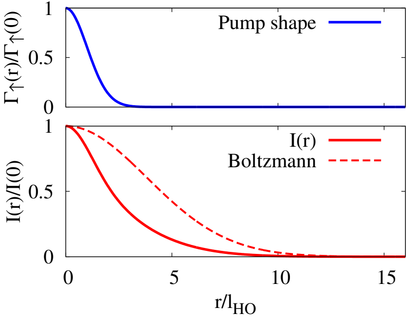
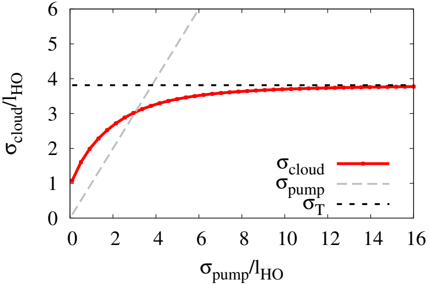
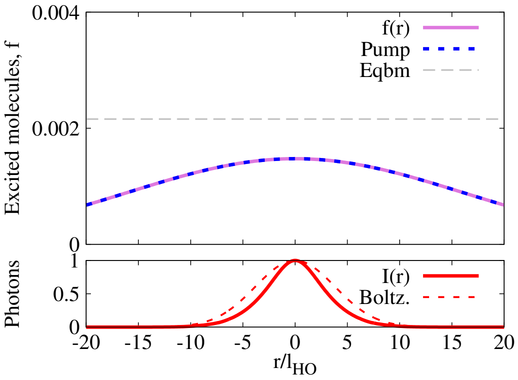




Near threshold behaviour



- Large spot, $\sigma_p \gg l_{\mathrm{HO}}$
- "Gain saturation" at centre
- Saturation of $f(r) = 1/(1 + e^{-\beta\mu})$ — spatial equilibriation

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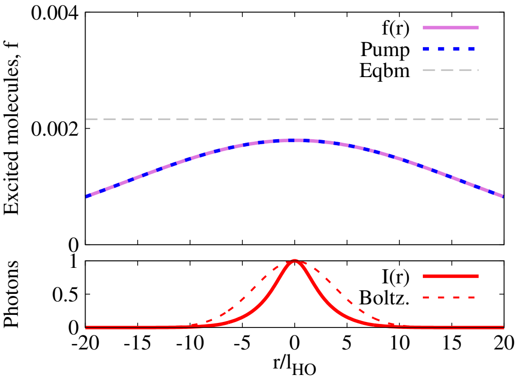

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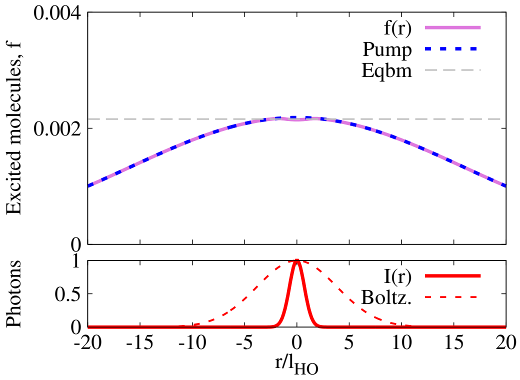

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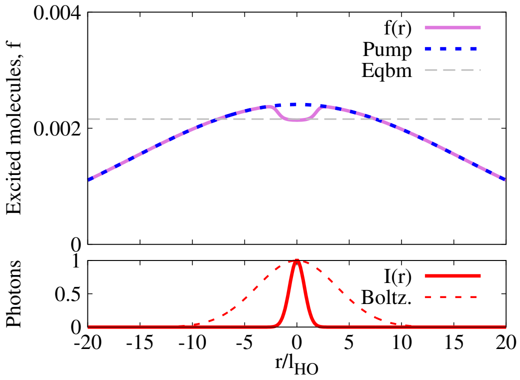

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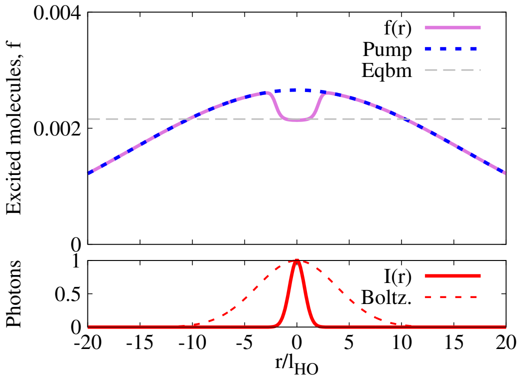

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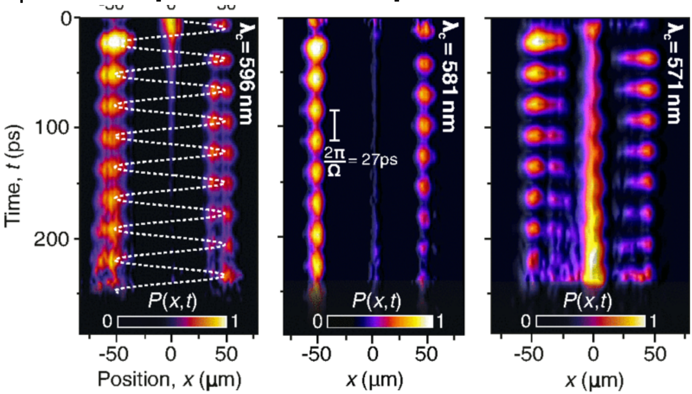
Off centre pumping; oscillations

- Experiments [Schmitt *et al.* PRA '15]



- Oscillations in space – beating of normal modes
- Thermalisation depends on cutoff

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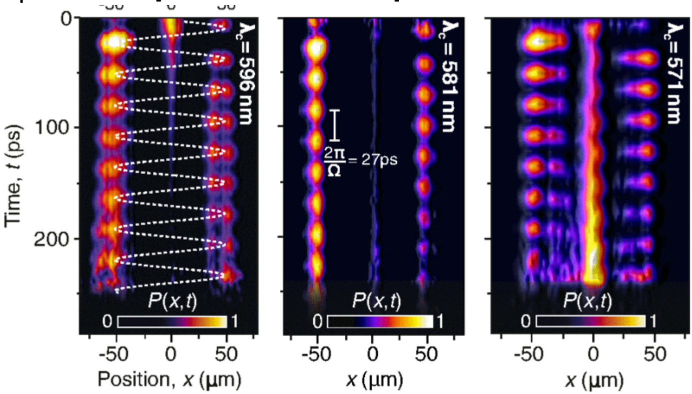

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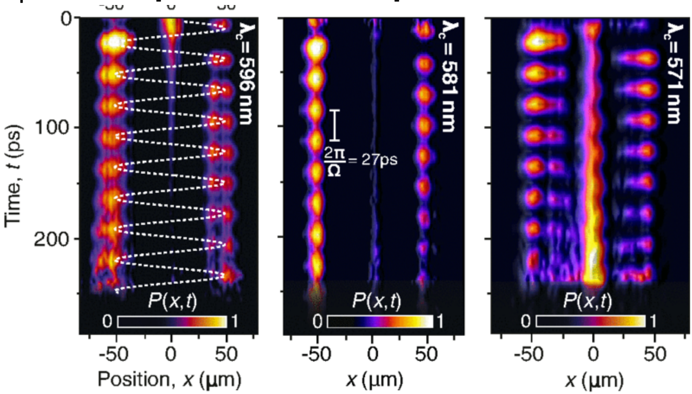

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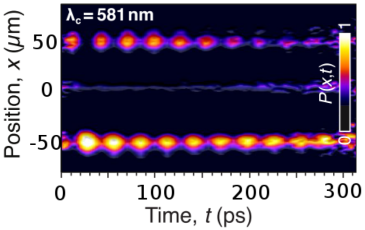
Limit of rate equations



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

- Oscillations: beating of modes.
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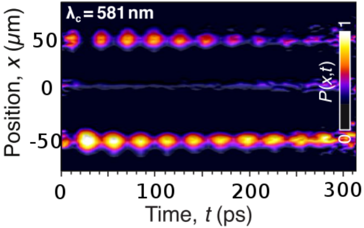

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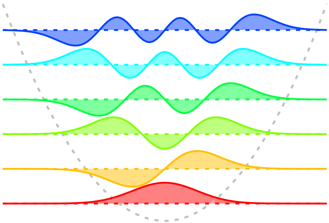
$$I(x) = \sum_m n_m |\psi_m(x)|^2$$



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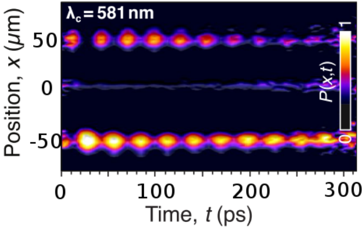

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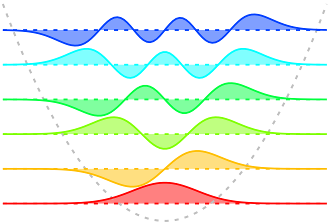

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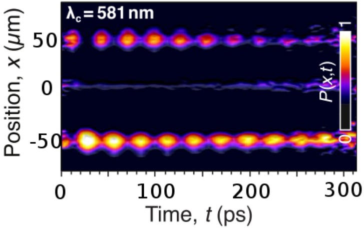

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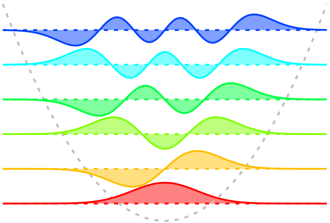

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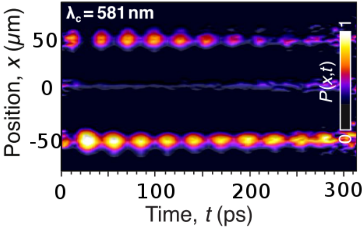

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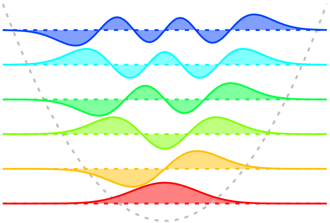

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Modelling

- Wavepacket emission: use Redfield theory:

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

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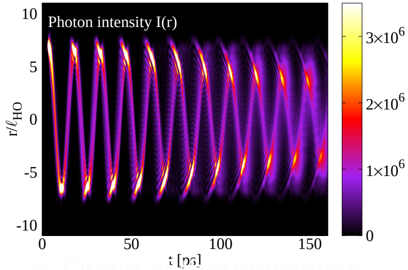
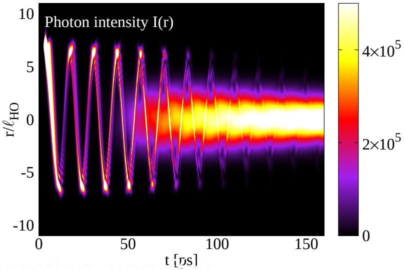
Dynamics from model

Longer cavity



Shorter cavity



- Origin of thermalisation — reabsorption, see I(ν)

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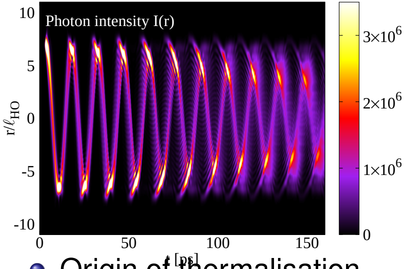

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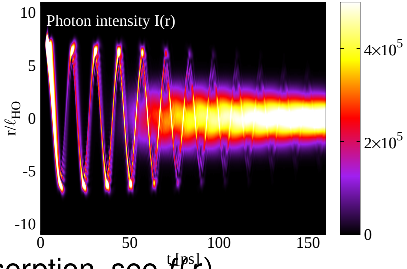

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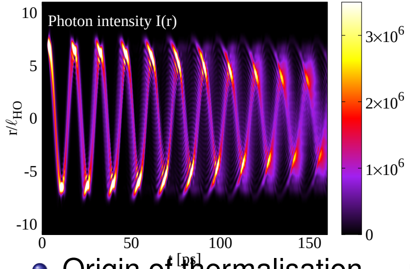

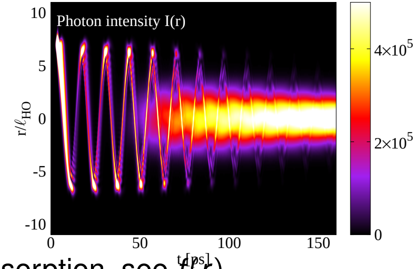

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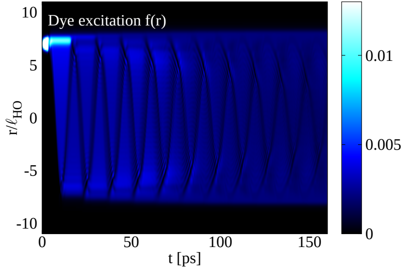
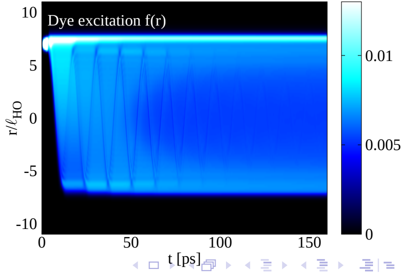
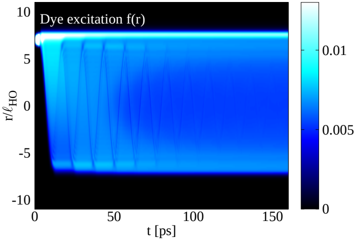




Thermalisation at late times

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



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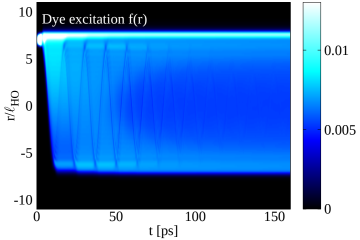

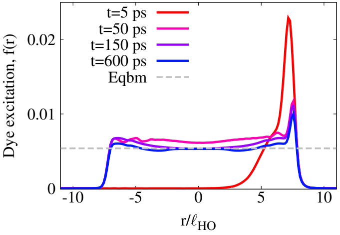

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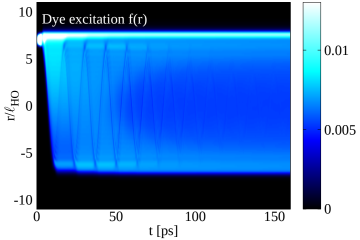

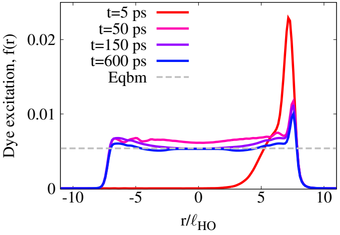

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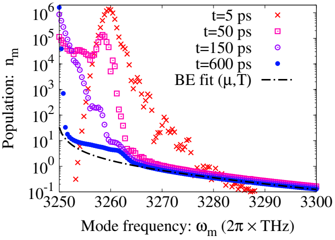


Acknowledgements

GROUP:

FUNDING:

EPSRC
Engineering and Physical Sciences Research Council

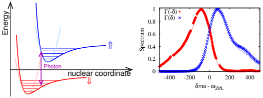
topnes
Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

Hybrid Polaritonics

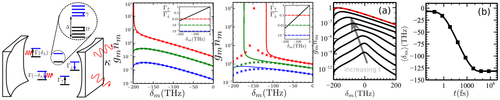
The Leverhulme Trust

Summary

- Holstein-Dicke and Holstein-Tavis-Cummings models

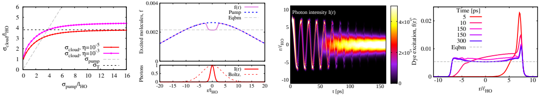


- Photon BEC and thermalisation



[Kirton & JK, PRL '13, PRA '15]

- Photon condensation, pattern formation physics



[JK & Kirton, PRA '16]

Exact states and spectra of vibrationally dressed polaritons

Jonathan Keeling

University of St Andrews
FOUNDED 1413

SUPA®

Quantum Nanophotonics, February 2017

Holstein-Tavis-Cummings & Holstein-Dicke model

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

- Few emitters (molecules/quantum dots)
  Wilson-Rae & Imamoğlu PRB 2002 McCutcheon & Nazir PRB 2011 Roy & Hughes PRB 2011; Bera *et al.* PRB 2014; Pollock *et al.* NJP 2013; Hornecker *et al.* arXiv:1609.09754; . . .
- Weak coupling
  Kirton & JK, PRL 2013, PRA 2015; PRA 2016 . . .
- Full model
  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 PRL, PRA '17; . . .

Reminder of models

Polariton states

One excitation subspace, questions

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- Rotating wave approximation — Holstein Tavis Cummings
- $\hat{x}_i = \left(\hat{b}_i + \hat{b}_i^\dagger \right) / \sqrt{2}$
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$$\left. + \frac{\hat{p}_i^2}{2} + \frac{\omega_v}{2} \left(\hat{x}_i - \sqrt{2} \lambda_0 \sigma_i^+ \sigma_i^- \right)^2 - \omega_v \lambda_0^2 \sigma_i^+ \sigma_i^- \right]$$

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Exact solution, $N = 2$

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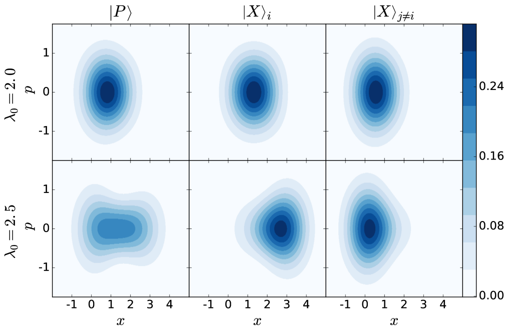

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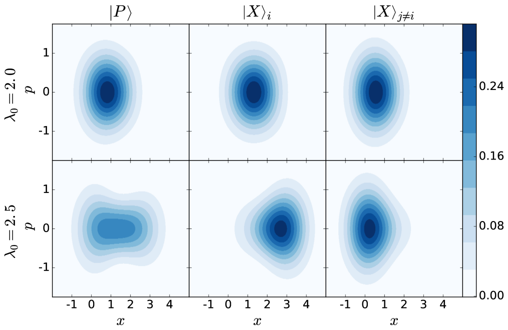

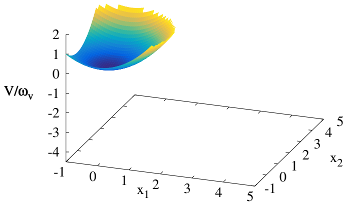

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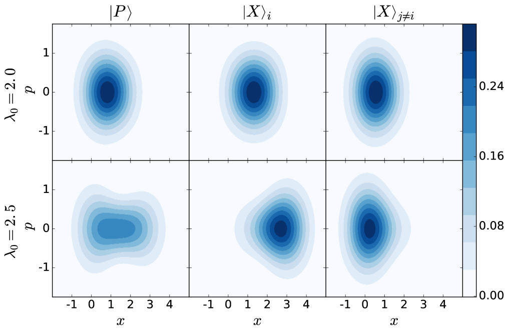

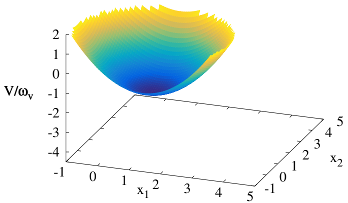

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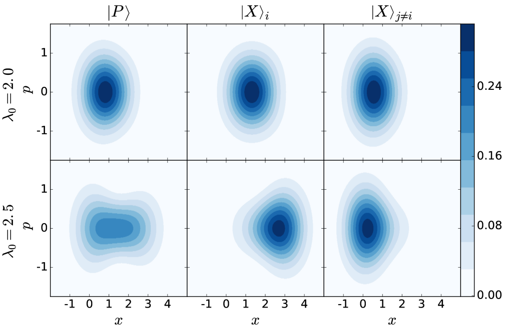

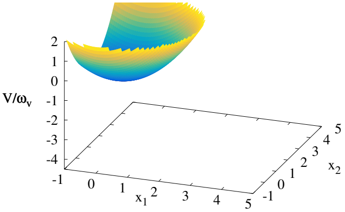

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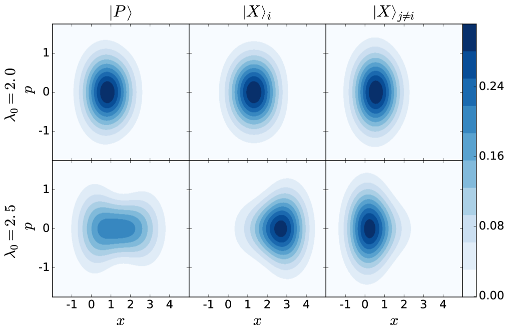

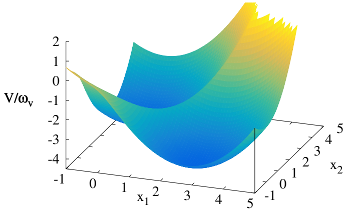

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- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$
- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5-6$

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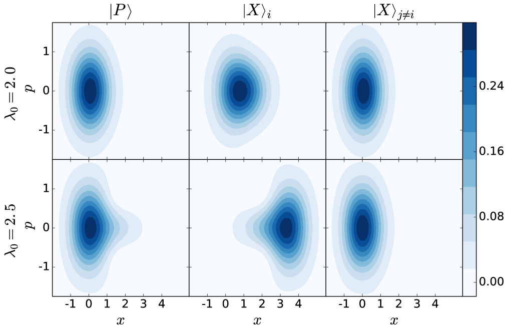

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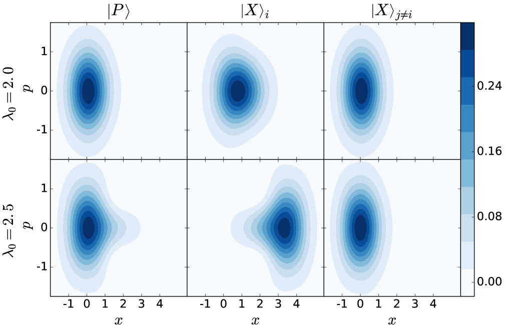

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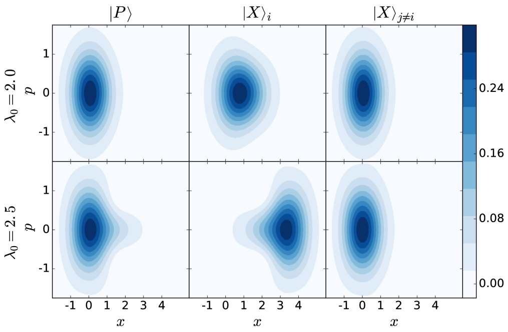

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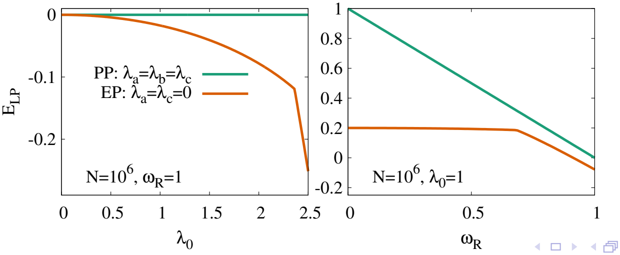

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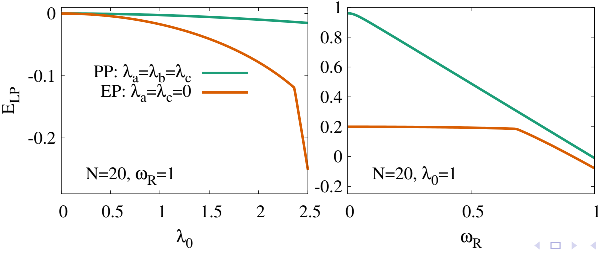

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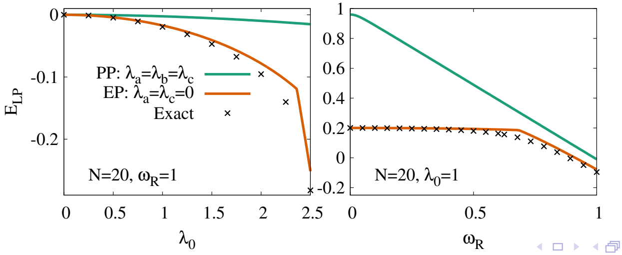

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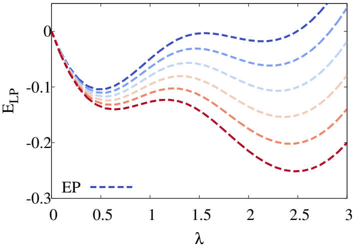

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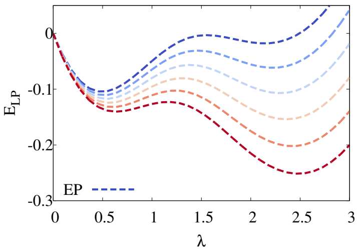

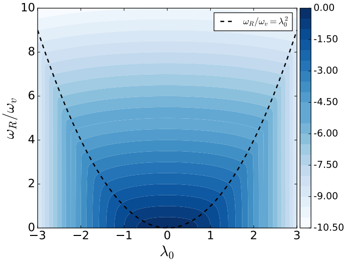

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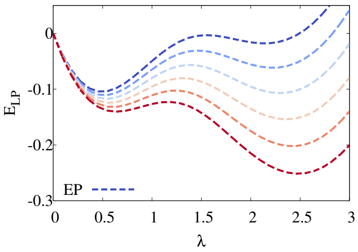

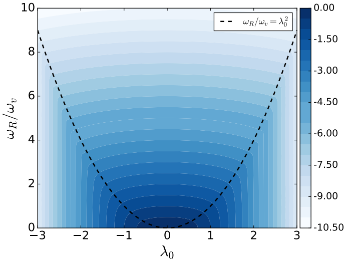

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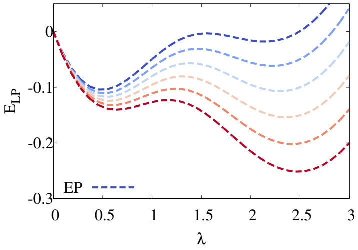

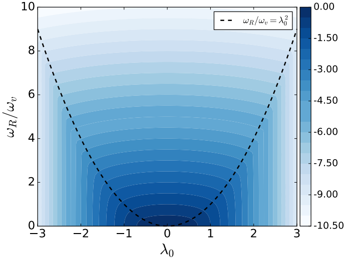

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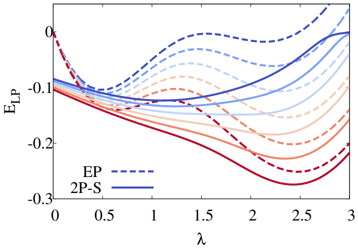
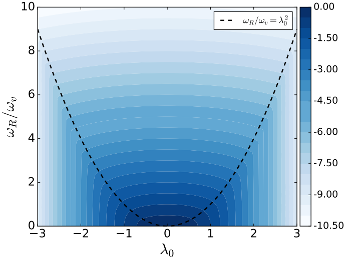


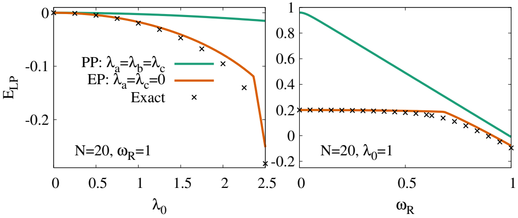


- Suggests multi-polaron ansatz [Bera *et al.* PRB 2014]
  - Superpose multiple polarons
  - Multimodal Wigner function
- Simplifed 2-polaron form [Zeb *et al.* arXiv:1608.08929]

$$|\Psi\rangle = \left[|P\rangle \frac{1}{N} \sum_i (\alpha_1 + \alpha_2 \mathcal{D}_i(\lambda)) + \frac{1}{\sqrt{N}} \sum_i |X\rangle_i (\beta_1 + \beta_2 \mathcal{D}_i(\lambda)) \right] |0\rangle_V$$

Simplified two-polaron physics

- Accurate energy & wavefunction



- Recovers Wigner function (analytic)
  - $W_{|P\rangle}(x \neq 0, p) \sim 1/N$, no other suppression

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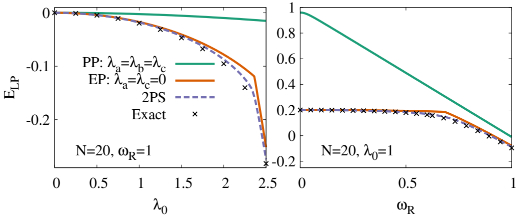

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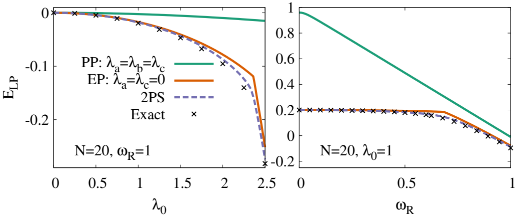

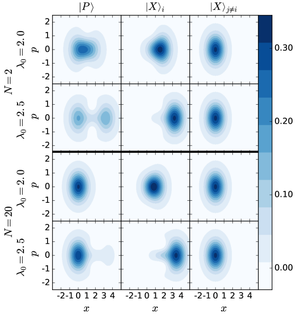

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Spectrum

Calculating spectra: Input-Output formalism

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

- Scattering matrix gives:

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

- Green's function:

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

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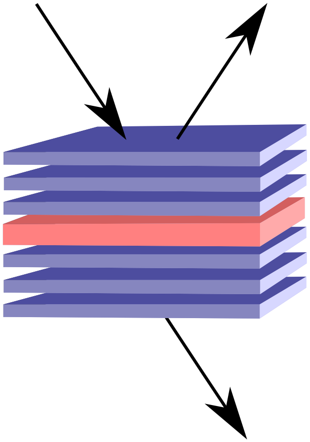

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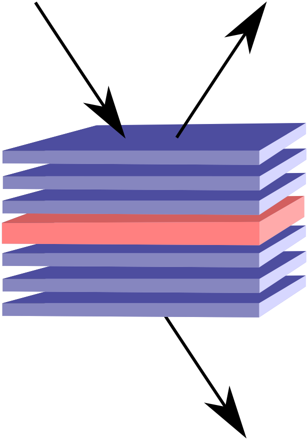

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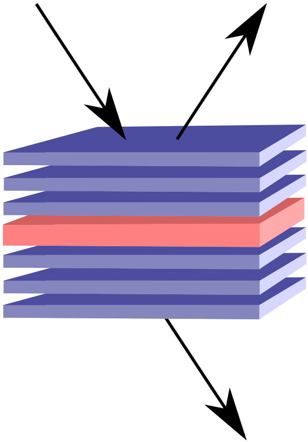

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Tavis-Cummings-Holstein vs Coupled Oscillators

- 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,

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

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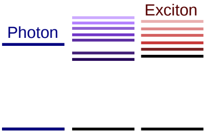

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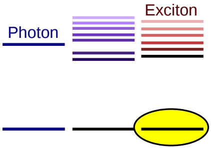

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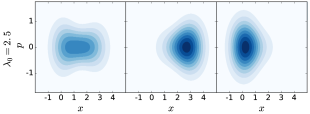

Tavis-Cummings-Holstein spectrum

- 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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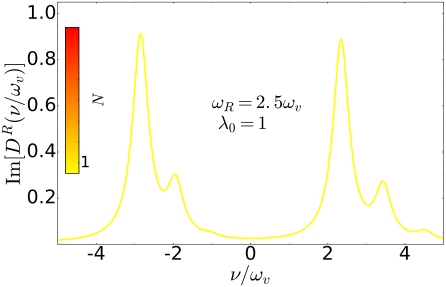

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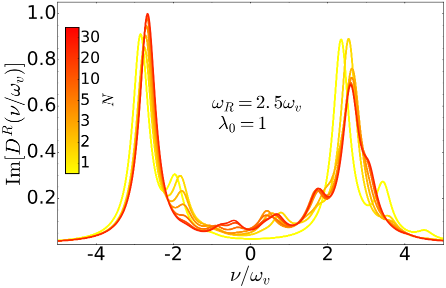

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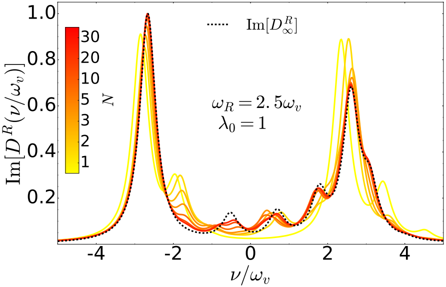

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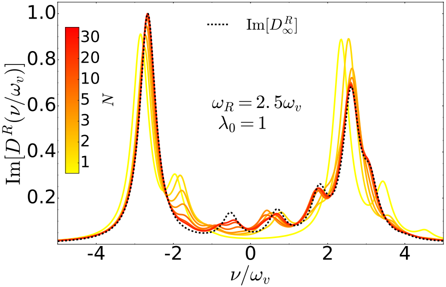

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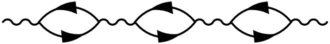
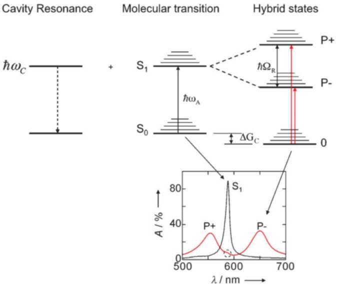
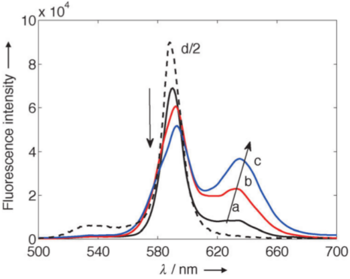
- Why? Multiple excitation $\sim 1/N$, 

Ultrastrong coupling, ground-state reconfiguration

Ultra strong coupling experimental features

- Ultra-strong coupling: $\omega, \omega_X \sim g\sqrt{N} \propto \sqrt{\text{concentration}}$
- Normal state: configuration of molecules





[Canaguier-Durand *et al.* Angew. Chem. ’13]

- Polariton *vs* molecular spectral weight – chemical eqbm
- (Weakly) temperature dependent

- Questions:

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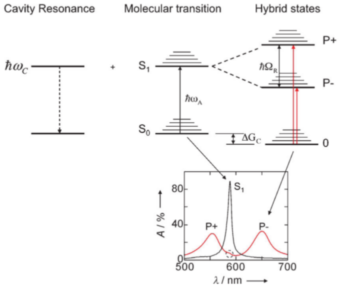

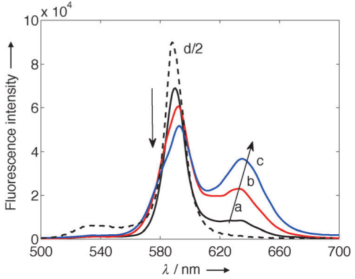

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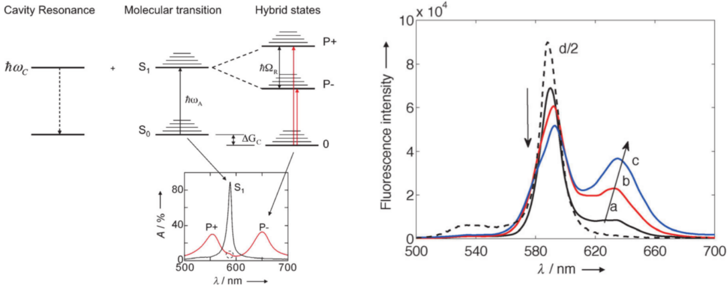

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- Questions:
  - Can USC change ground state configuration
  - Disorder + vibrations + USC

Ground state molecular reconfiguration

- Dicke model: beyond rotating wave approximation

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

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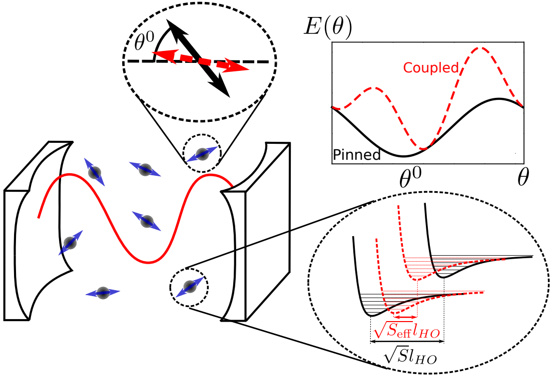
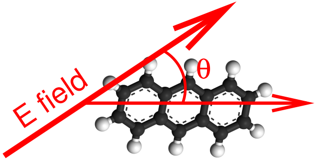


Rotational reorientation

- Rotational degrees of freedom



- Effective Hamiltonian

$$H = \ldots + \sum_{i,k}\left[\ldots + g_{i,\mathbf{k}}\cos(\theta_i)(\hat{a}^\dagger_k + \hat{a}_{-k})\sigma^x_i + E_0(\theta_i)\right]$$

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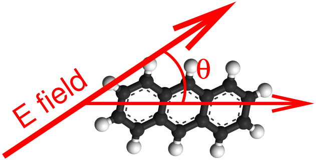

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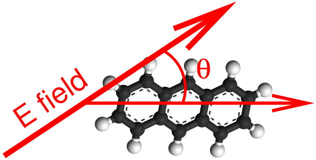

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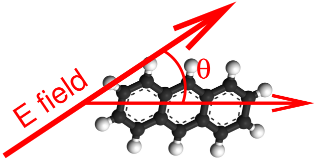

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Vibrational reconfiguration

- Schrieffer-Wolff – mixes vibrational states

$$\delta H = -\sum_{i,k} \frac{g_{\mathbf{k}}^2}{2(\omega_X + \omega_k)} \left\{ 1 - \frac{\omega_v \lambda_0 (b_i + b_i^\dagger)}{\omega_X + \omega_k} + \mathcal{O}\left[\left(\frac{\omega_v}{\omega_X}\right)^2, \frac{g\sqrt{N}}{\omega_X} \right] \right\}$$

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$$\lambda_0 \to \lambda_0(1 - K_1), \quad K_1 = \sum_k \frac{g_{\mathbf{k}}^2}{(\omega_k + \omega_X)^2}$$

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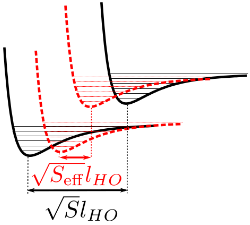

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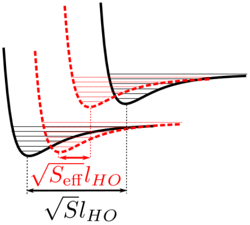

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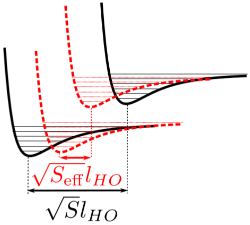
$$\lambda_0 \to \lambda_0 (1 - K_1), \quad K_1 = \sum_k \frac{g_{\mathbf{k}}^2}{(\omega_k + \omega_X)^2}$$

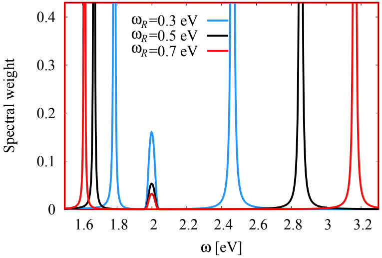


  - Increased effective coupling: $g_{\text{eff}}^2 = g^2 \exp(-S)$
  - Again, $K_1 \ll 1$, independent of density.

Ultrastrong coupling, ground-state reconfiguration

Bumps in the middle of the spectrum

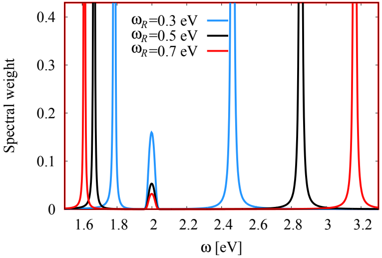
- Orgin of bumps in middle of spectrum: Disorder



- Central peak:

Bumps in the middle of the spectrum

- Orgin of bumps in middle of spectrum: Disorder



- Central peak:

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

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

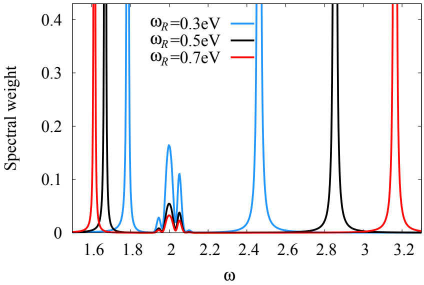
Gaussian $\rho(x)$, variance σ_x

[Houdré *et al.* , PRA '96]

Disorder + Vibrations + Strong coupling

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

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



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

[Cwik *et al.* PRA '16]

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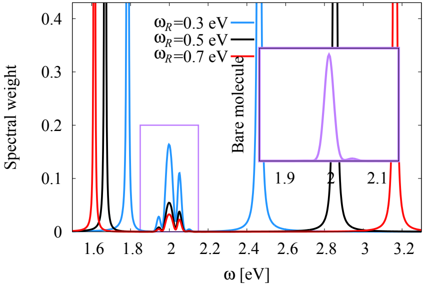

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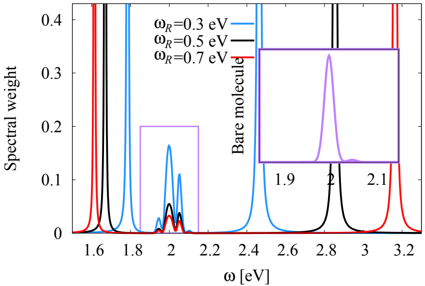

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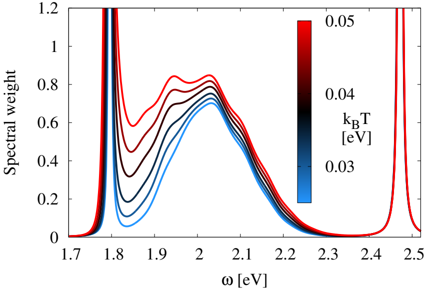

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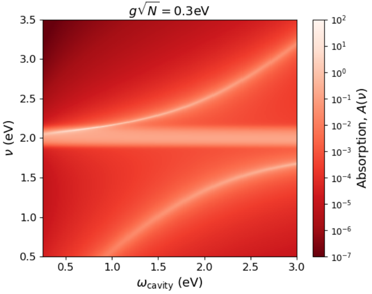
[Cwik *et al.* PRA '16]

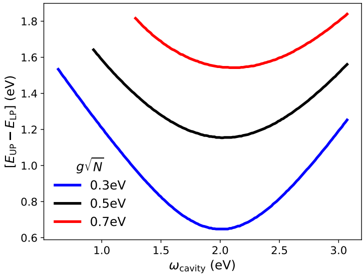
Tavis-Cummings-Holstein Spectrum Redux

Green's function absorption

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

Strong coupling ($\omega_R \gg$ linewidth) — polariton splitting





- Extract optimal splitting vs ω_v, λ_0

Green's function absorption

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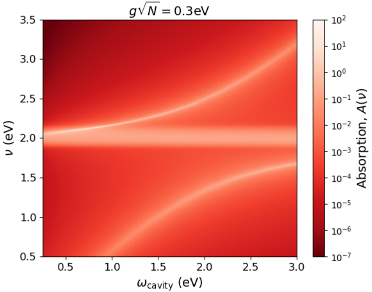

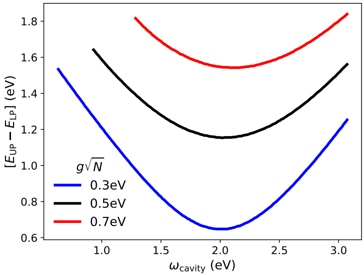

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Hybrid
Polaritonics

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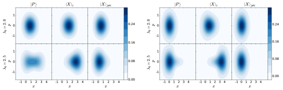
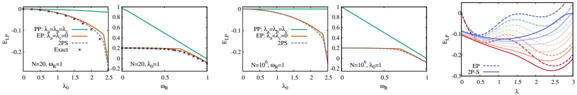
Summary

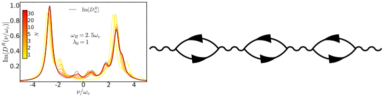
- Single polariton state [Zeb, Kirton, JK, `arXiv:1608.08929`]
  - Exact solution



  - Polaron ansatz



  - Validity of mean-field Green's functions



- Vibrations + disorder + USC [Cwik *et al.* PRA '16]