Modelling organic condensates from weak to strong coupling

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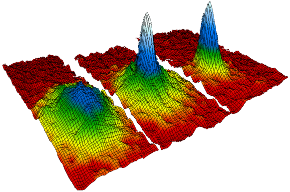
FOUNDED 1413

SUPA

SISSA, April 2017

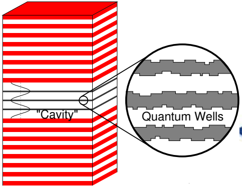
Condensation, Lasing, Superradiance

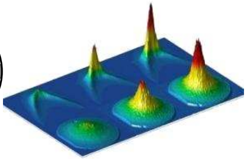
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science '95]

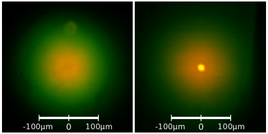
Polariton Condensate $T \sim 20$K





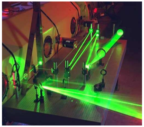
[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K

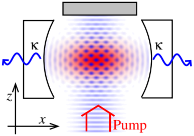


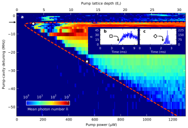
[Klaers *et al.* Nature, '10]

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



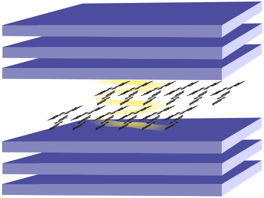
Superradiance transition
$T \sim 0$





[Baumann *et al.* Nature '10]

Organic polaritons: What & Why



- Why: Polariton splitting $\sim 1eV \gg k_B T_{\text{Room}}$

- Anthracene Polariton Lasing

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

- Polymers (MeLPPP, TDAF)

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

- Biologically produced materials (GFP)

[Dietrich *et al.* Sci. Adv. '16]

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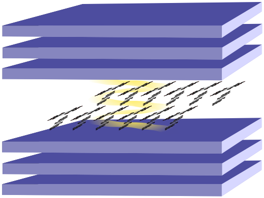

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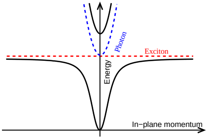

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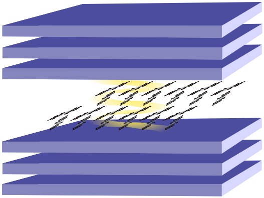

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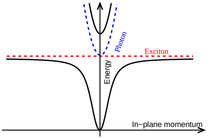
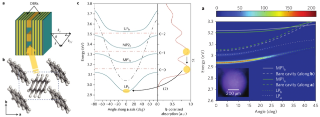


Examples:

- Anthracene Polariton Lasing



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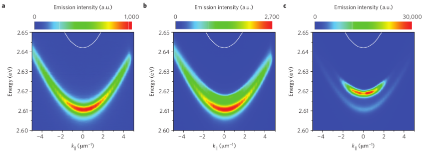

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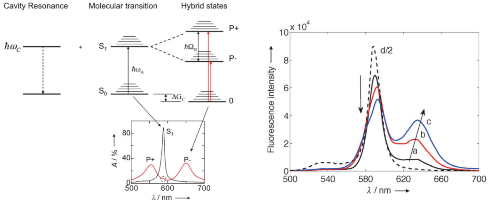
Motivation: vacuum-state strong coupling

- 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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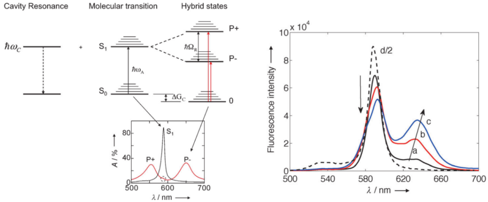

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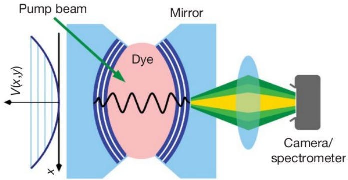
Q1. Can **ultra-strong** coupling to light change:
  - charge distribution?
  - vibrational configuration?
  - molecular orientation?
  - crystal structure?

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

Motivation: Bose-Einstein condensation of photons

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



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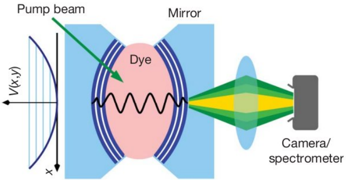

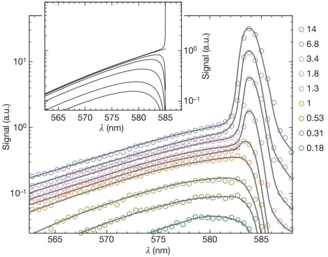

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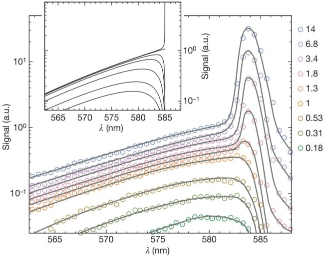

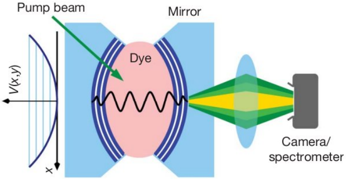

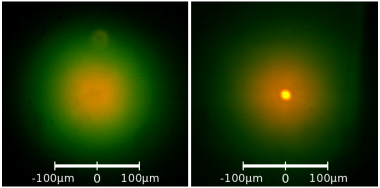

[Klaers et al, Nature, 2010]

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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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
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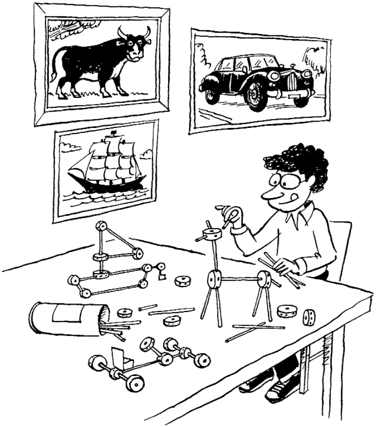

Illustration by Dick Codor.
[Auerbach, Interacting Electrons (Springer, 1998)]

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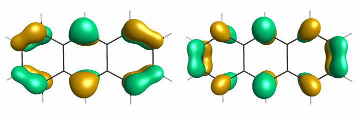
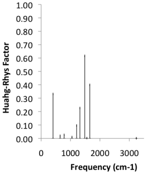
Toy models

1. Full molecular spectra electronic structure & Raman spectrum





2. Simplified archetypal model: Dicke-Holstein

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

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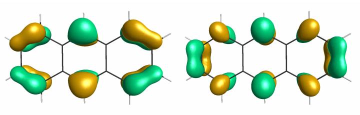

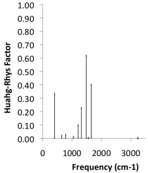

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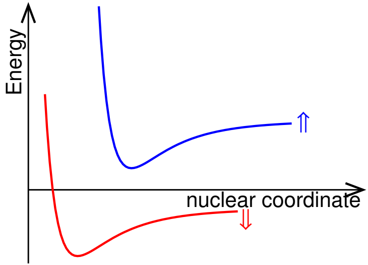
- Two-level system, HOMO/LUMO
- Single DoF PES



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

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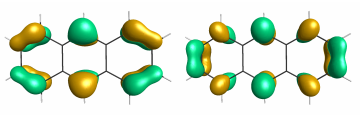

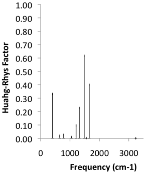

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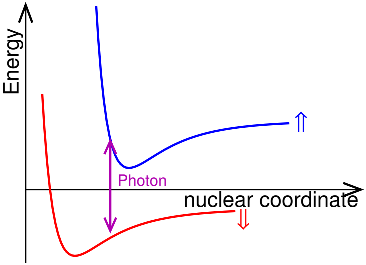

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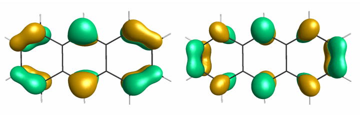

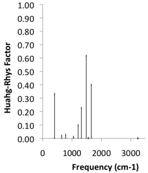

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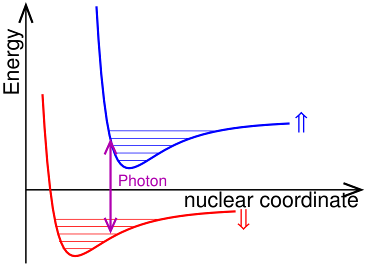

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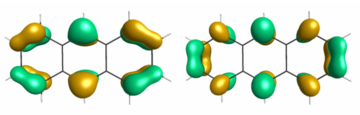

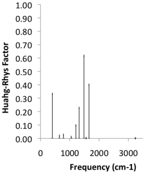

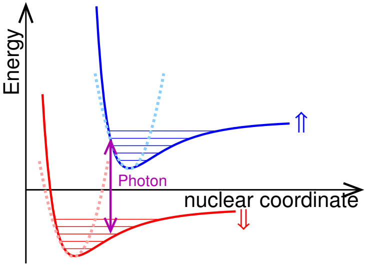

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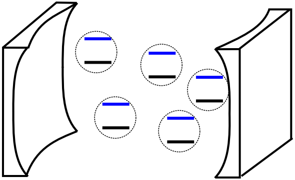
Holstein-Tavis-Cummings model

Model capable of lasing & condensation

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



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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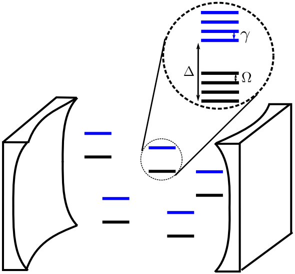
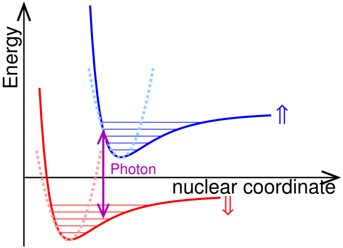
$$\left. + \omega_v \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$





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Introduction and models

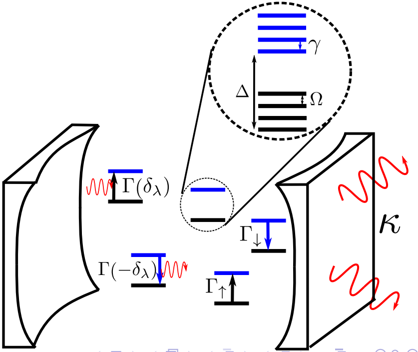
1. Introduction and models
   - Holstein-Dicke model
2. Weak coupling
   - Photon BEC
   - Spatial profile
3. Strong coupling
   - Exact eigenstates
   - Spectrum
4. Ultra strong coupling
   - Vibrational reconfiguration
   - Vibrations and disorder

Weak coupling

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.
- Weak coupling, perturbative in g



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

- **2D** harmonic oscillator
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
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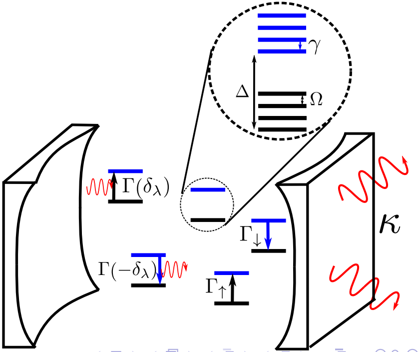

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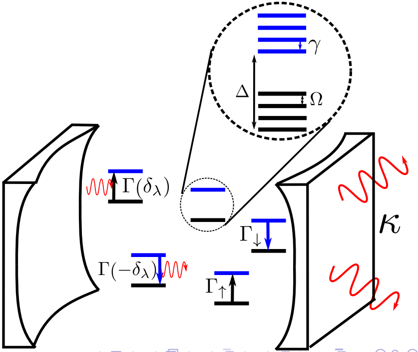

Steady state populations and equilibrium

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:

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

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[Kirton & JK PRL '13]

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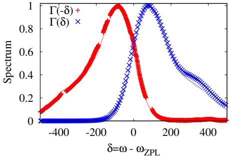

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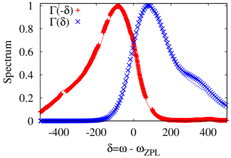

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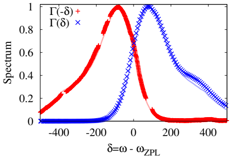

  - Equilibrium, $\rightarrow$ Kubo-Martin-Schwinger condition:

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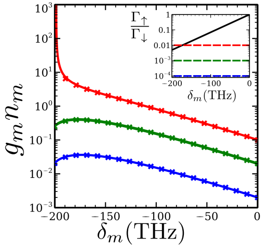

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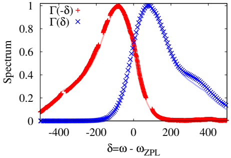

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Chemical potential?

- 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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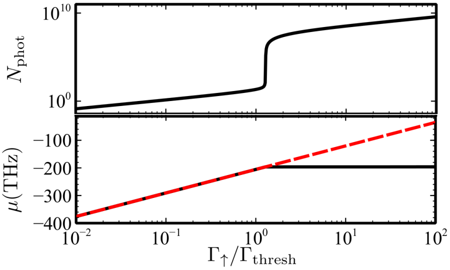

[Kirton & JK, PRA '15]

Weak coupling

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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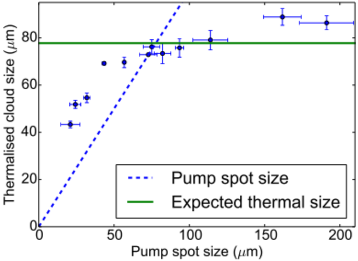

Modelling spatial profile.

- Varying excited density – differential coupling to modes

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

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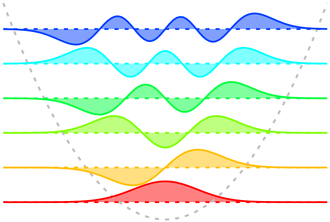

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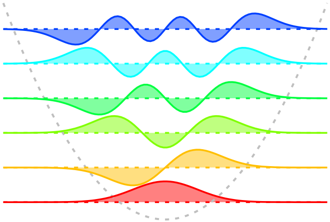

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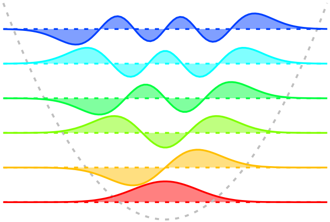
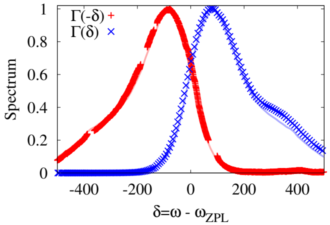


- Use exact R6G spectrum



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$$\partial_t \rho_\uparrow(\mathbf{r}) = -\tilde{\Gamma}_\downarrow(\mathbf{r})\rho_\uparrow(\mathbf{r}) + \tilde{\Gamma}_\uparrow(\mathbf{r})\rho_\downarrow(\mathbf{r}))$$

Spatially varying pump: below threshold

- Far below threshold:
  - If $\kappa \ll \rho_M \Gamma(\delta_m)$,
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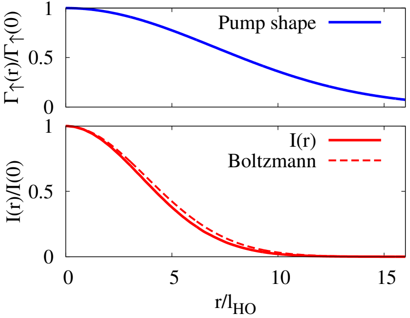

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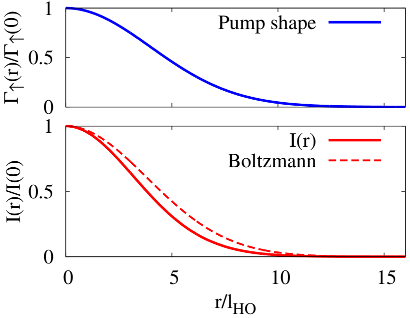

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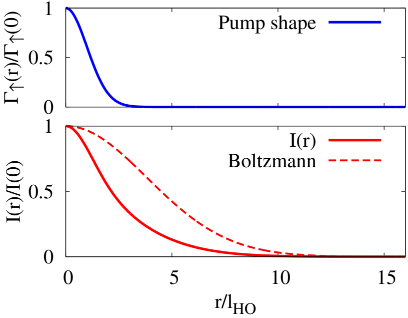

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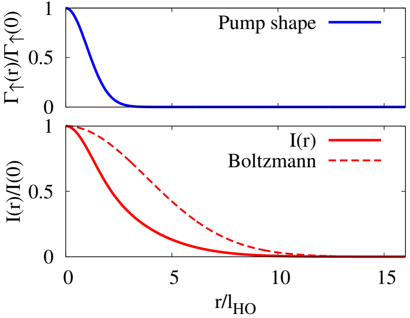
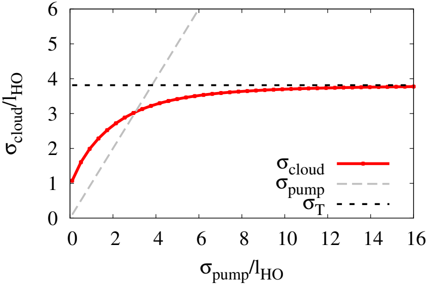
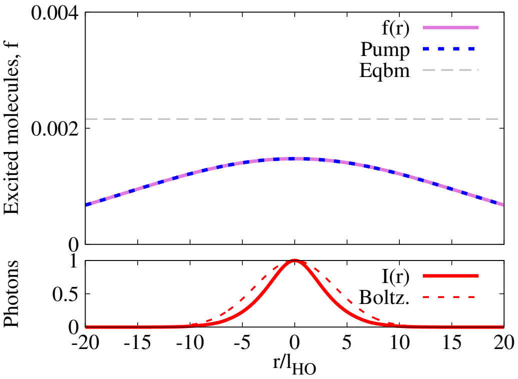




Near threshold behaviour



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- Saturation of $f(r) = 1/(1 + e^{-\beta\mu})$ — spatial equilibriation

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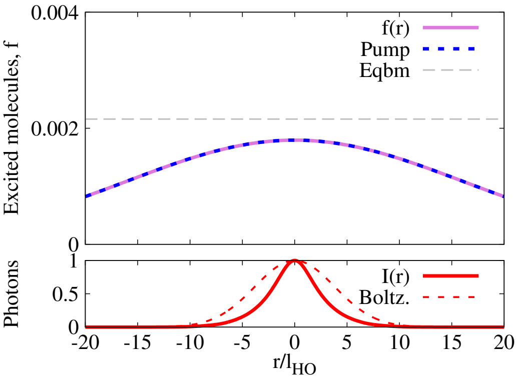

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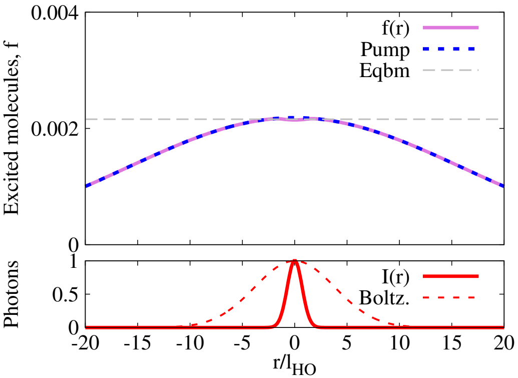

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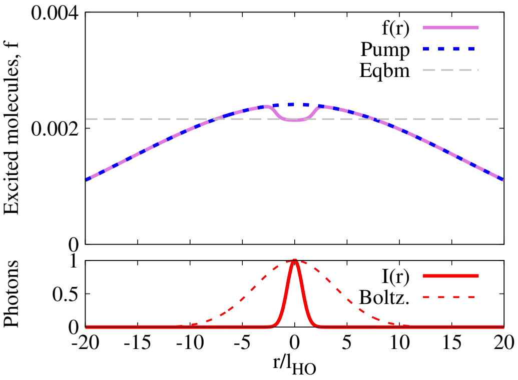

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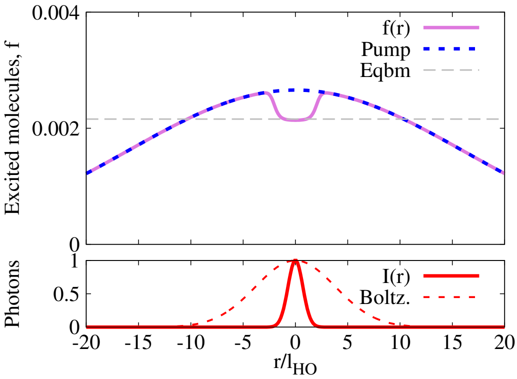

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Strong coupling

Strong coupling: One excitation subspace

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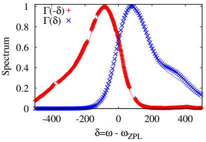
Strong coupling: fate of spectrum



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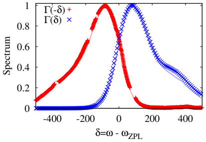

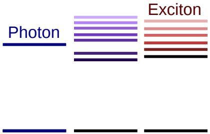

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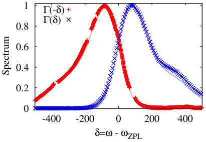

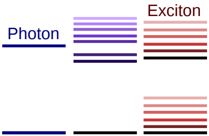

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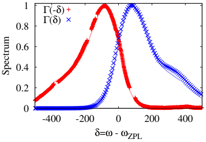
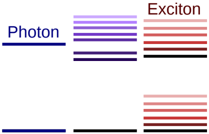
Strong coupling: fate of spectrum





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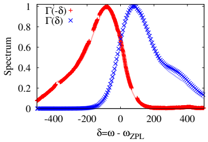

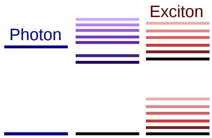

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Exact solution

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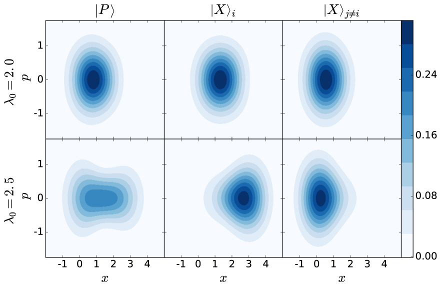

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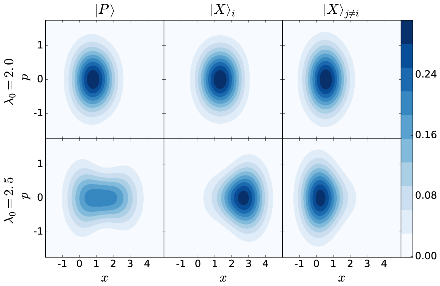

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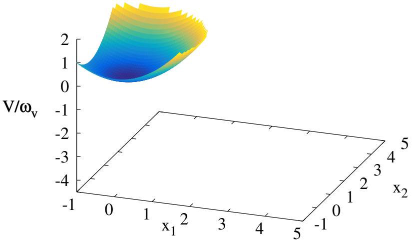

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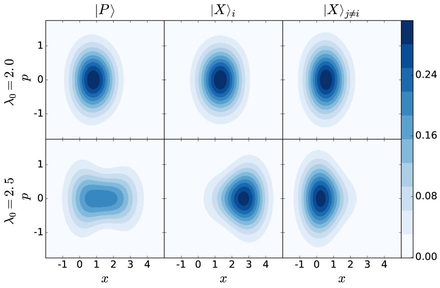

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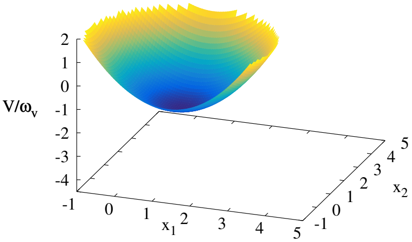

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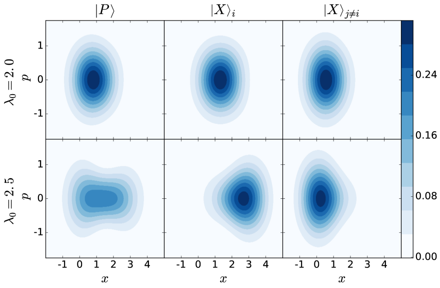

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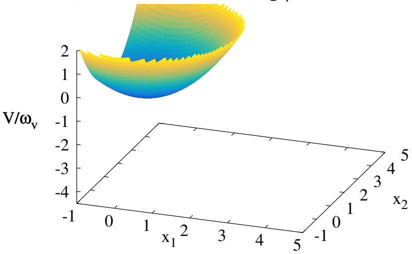

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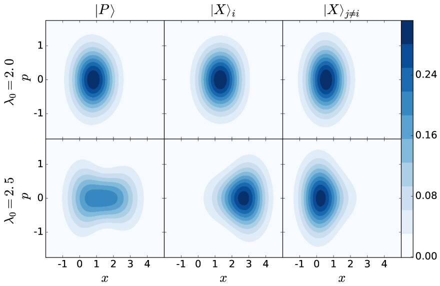

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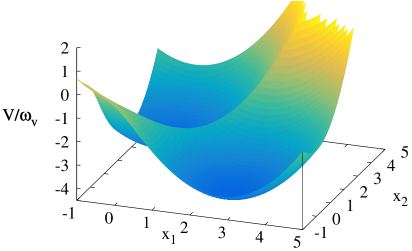

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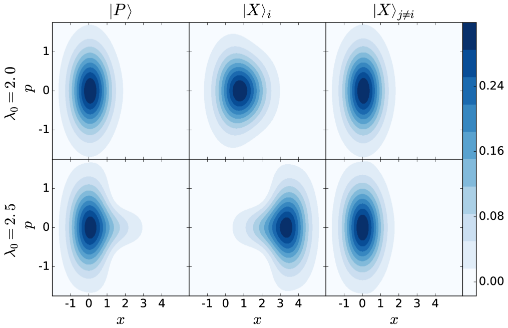

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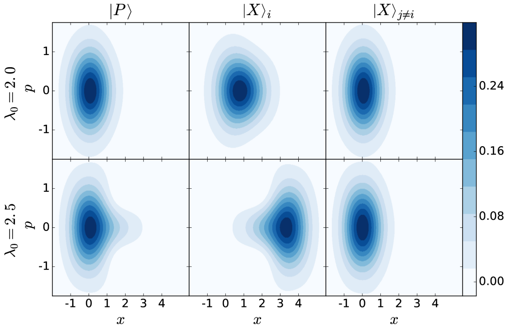

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Extending to arbitrary N, polaron ansatz

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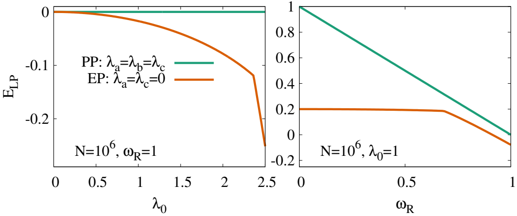

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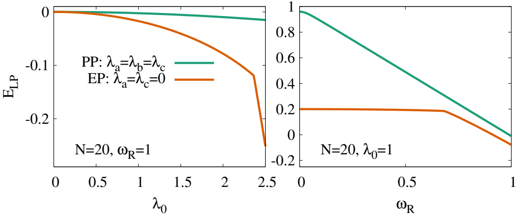

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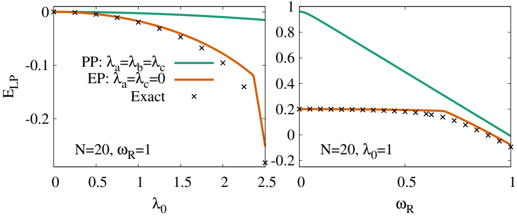

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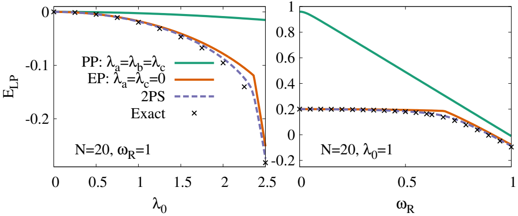

Strong coupling

Calculating spectra: Input-Output formalism

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

- Scattering matrix gives:

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

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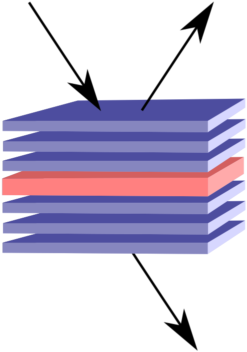

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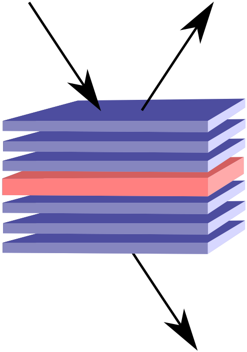

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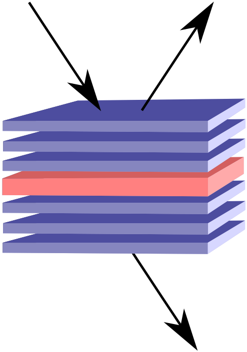

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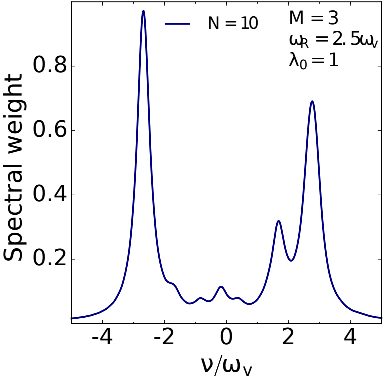

Tavis-Cummings-Holstein spectrum

- Direct calculation

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

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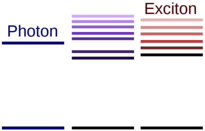

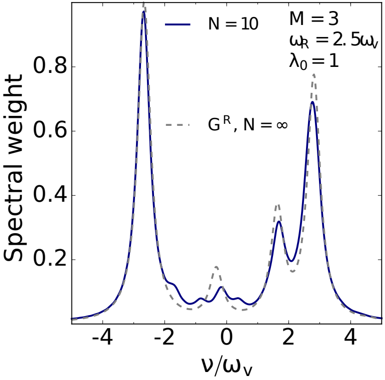

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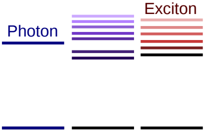

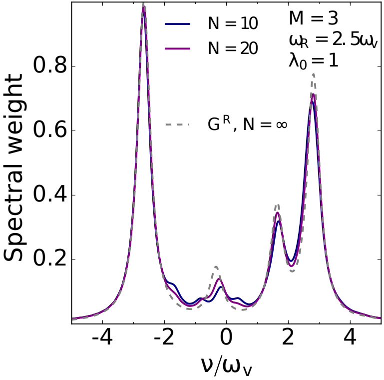

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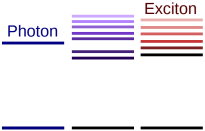

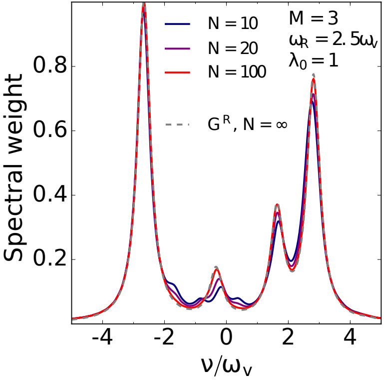

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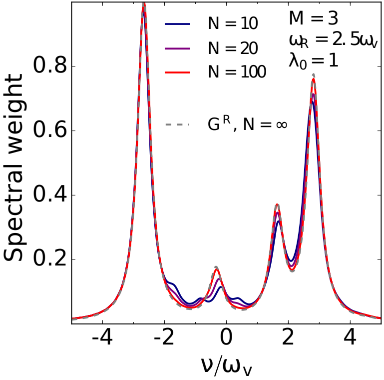
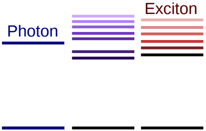
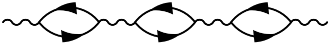




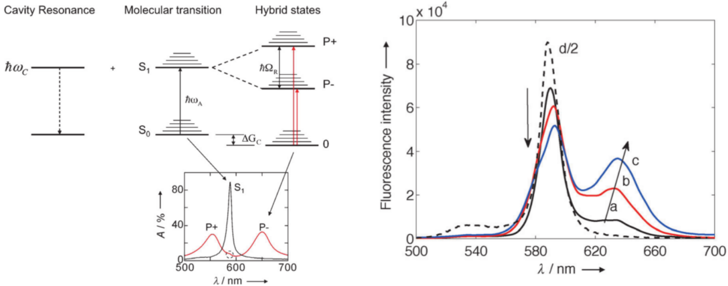
Multiple excitation $\sim 1/N$,



Ultra strong coupling

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

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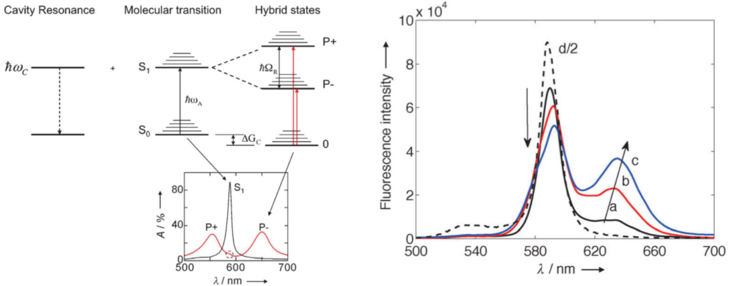

[Canaguier-Durand *et al.* Angew. Chem. '13]

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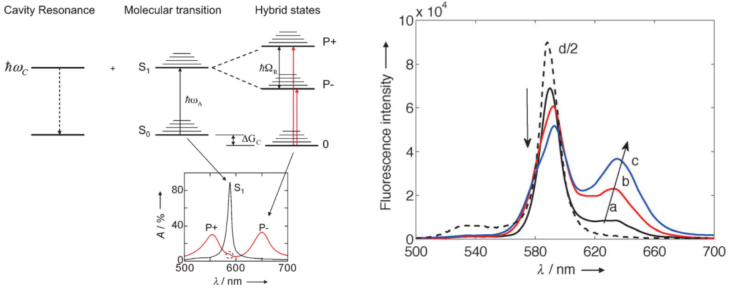

[Canaguier-Durand *et al.* Angew. Chem. '13]

  - Polariton *vs* molecular spectral weight – chemical eqbm
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- Questions:
  - Can USC change ground state configuration
  - Disorder + vibrations + USC

Vibrational reconfiguration

- Many photon modes, beyond RWA perturbatively

$$H = \sum_k \omega_k \hat{a}_k^\dagger \hat{a}_k + \sum_{i=1}^{N} \Big[\omega_X \sigma_i^+ \sigma_i^- + \sum_k g_{\mathbf{k}} \left(\sigma_i^+ (\hat{a}_k + \hat{a}_k^\dagger) + \text{H.c.} \right) + \omega_v \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \Big]$$

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

[Cwik *et al.* PRA '16]

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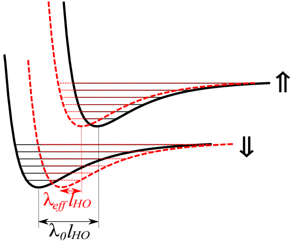

[Cwik *et al.* PRA '16]

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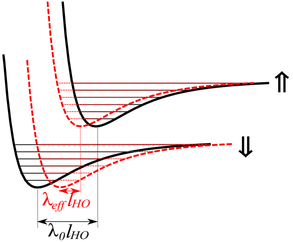

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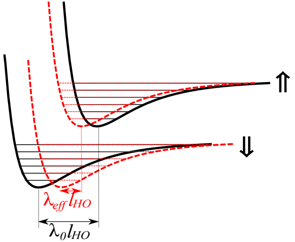
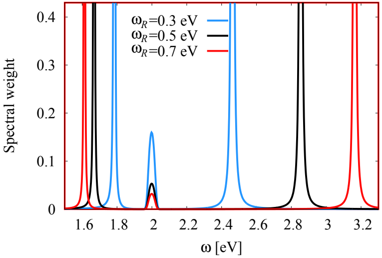


[Cwik *et al.* PRA '16]

Ultra strong coupling

Bumps in the middle of the spectrum

- Orgin of bumps in middle of spectrum: Disorder



- Central peak:

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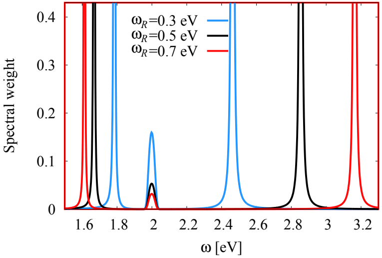


- Central peak:

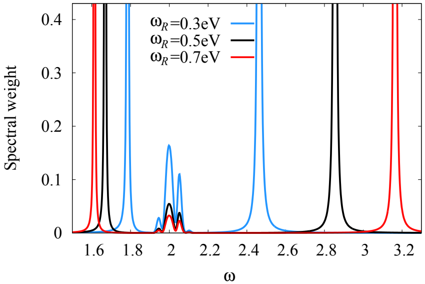
$$D^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_k + \Sigma_X(\nu)}$$

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

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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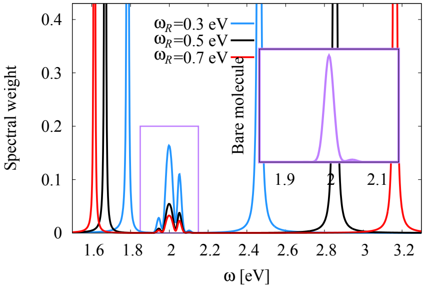

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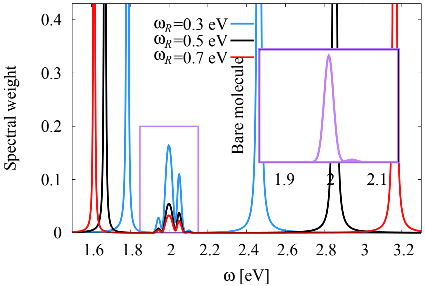

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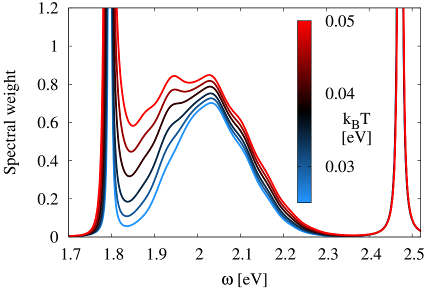
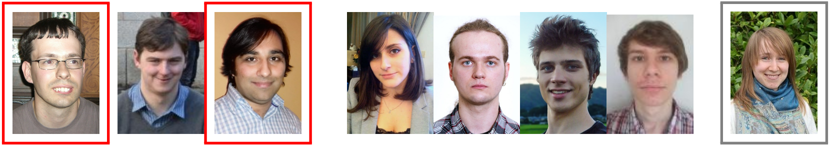


Acknowledgements

GROUP:



COLLABORATION: S. De Liberato (Southhampton).

FUNDING:

EPSRC
Engineering and Physical Sciences
Research Council

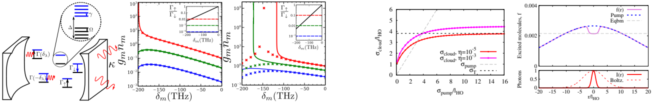
topnes
Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

Hybrid
Polaritonics

The Leverhulme Trust

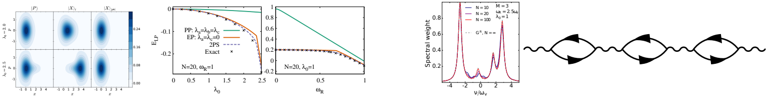
Summary

- Photon BEC and thermalisation



[Kirton & JK, PRL '13, PRA '15, JK & Kirton, PRA '16]

- Single polariton state, Exact solution vs Polaron ansatz



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

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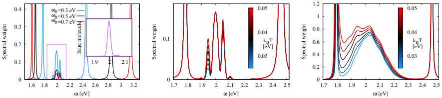

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