

# From weak to ultra-strong matter-light coupling with organic materials

Jonathan Keeling



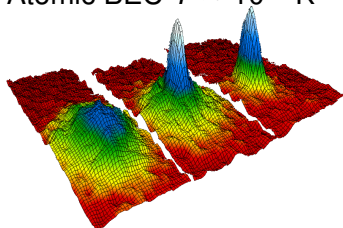
University of  
St Andrews

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Birmingham, March 2016

# Coherent states of matter and light

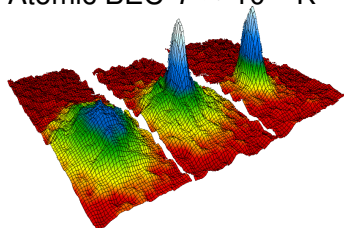
Atomic BEC  $T \sim 10^{-7}\text{K}$



[Anderson *et al.* Science '95]

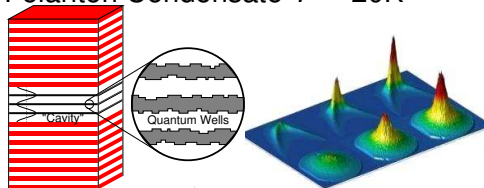
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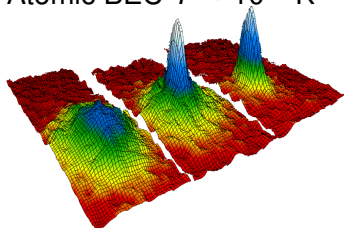
Polariton Condensate  $T \sim 20\text{K}$



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

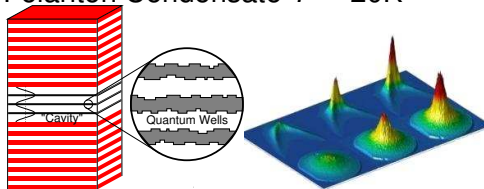
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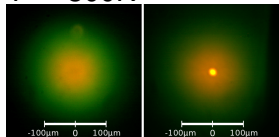
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Photon Condensate

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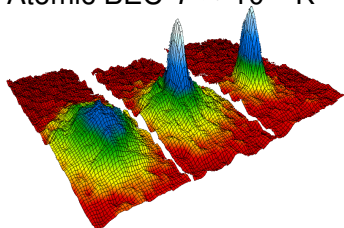


[Klaers *et al.* Nature, '10]



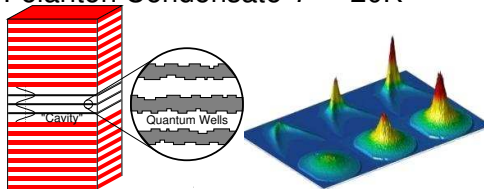
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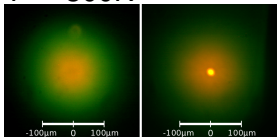
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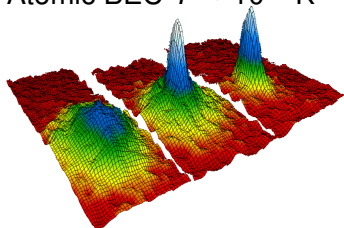
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Laser  
 $T \sim ?, < 0, \infty$



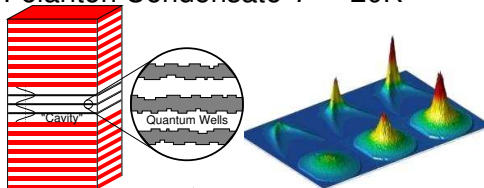
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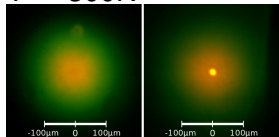
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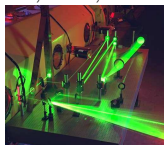
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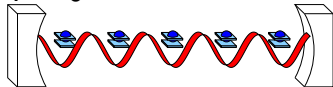


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Superradiance transition  
 $T \sim 0$



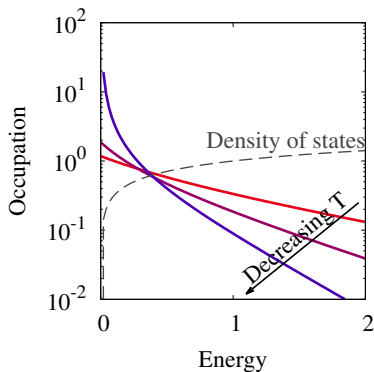
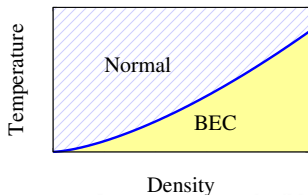
[Hepp & Lieb, Ann. Phys. '73]

# “Textbook” BEC

## ● Non-interacting viewpoint

▶ BE distribution:  $\mu < \omega_0$

▶  $T_c = \frac{2\pi\hbar^2}{m} \left( \frac{n}{\xi_d} \right)^{2/d}$



● Interacting approach (WIDBG)

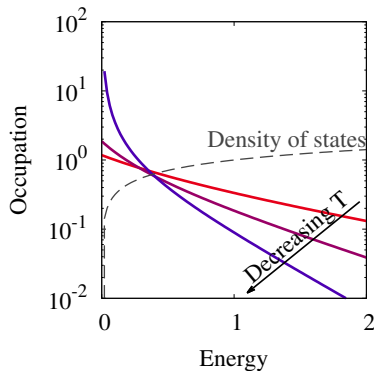
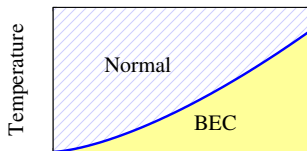
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● Mean field:  $\langle \psi \rangle^2 = (\mu - \omega_0)/g$

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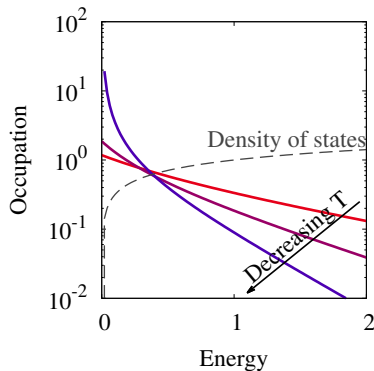
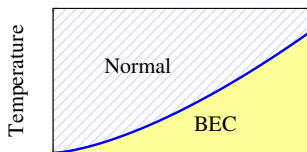
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# “Textbook” Laser: Maxwell Bloch equations

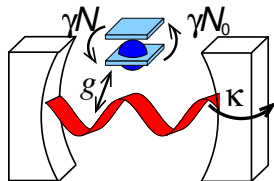
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Maxwell-Bloch eqns:  $P = -i\langle \sigma^{-} \rangle$ ,  $N = 2\langle \sigma^z \rangle$

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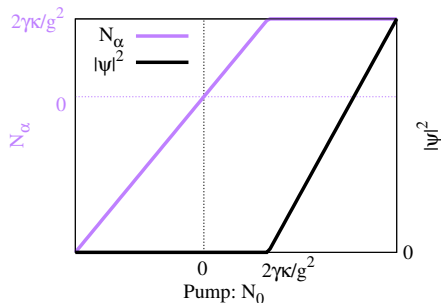
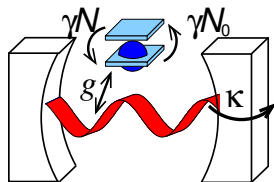
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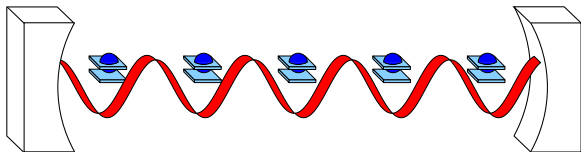
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$$|\psi|^2 > 0 \text{ if } N_0 g^2 > 2\gamma\kappa$$

# “Textbook” Dicke-Hepp-Lieb superradiance



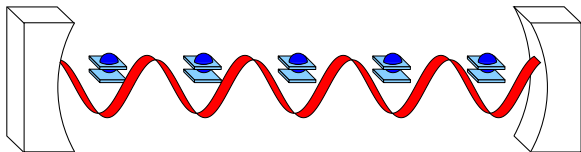
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- Coherent state:  $|\Psi\rangle \rightarrow e^{\lambda \psi^\dagger + \eta \sigma^+} |\Omega\rangle$
- Small  $g$ , min at  $\lambda, \eta = 0$

[Hepp, Lieb, Ann. Phys. '73]



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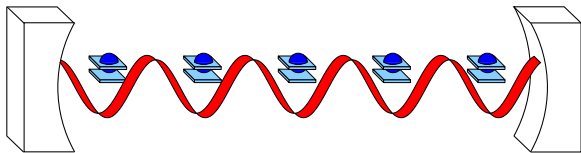
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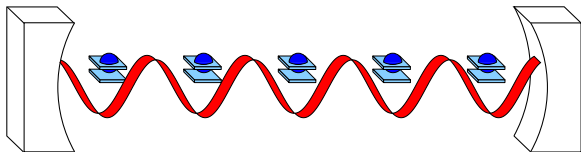
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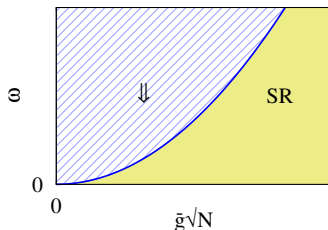
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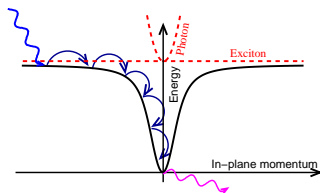
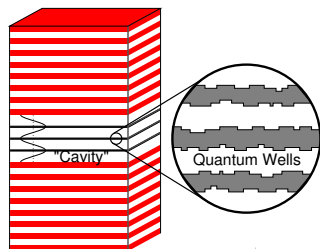
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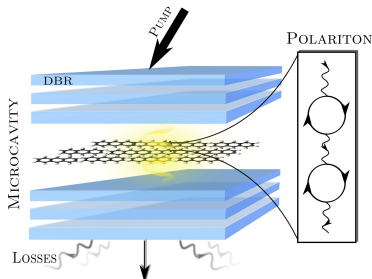
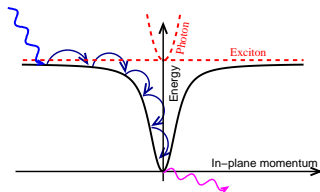
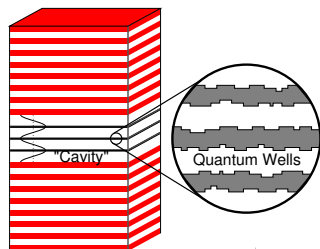


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# Coupled matter-light system: polaritons



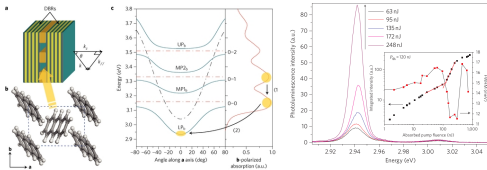
# Coupled matter-light system: polaritons



# Motivation: polariton condensates

## ● Anthracene Polariton Lasing

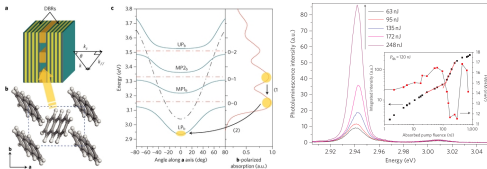
$T \sim 300\text{K}$



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

# Motivation: polariton condensates

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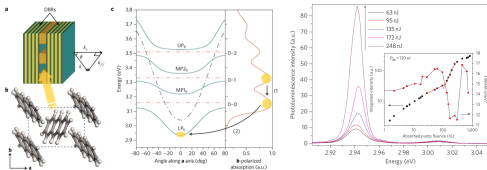


- Q1. Vibrational replicas?
- Q2. Relevance of disorder?
- Q3. Lasing vs condensation?

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

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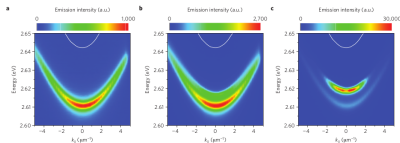
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- Polariton condensates, other materials, e.g. polymers:

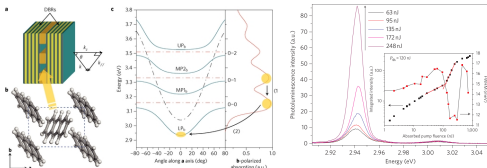


[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* *ibid* '14]



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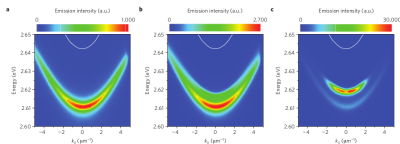
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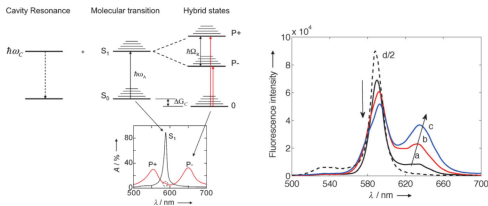


- Q1. Frenkel to Wannier crossover?
- Q2. Optimal vibrational properties?
- Q3. Nonlinearities?

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

# Motivation: vacuum-state strong coupling

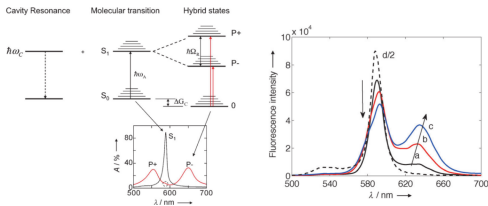
- Linear response (no pump, no condensate): effects of matter-light coupling alone.



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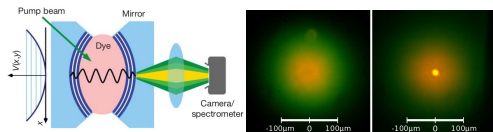


[Canaguier-Durand *et al.* Angew. Chem. '13;  
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- Q1. Can **ultra-strong** coupling to light change:
- ▶ charge distribution?
  - ▶ vibrational configuration?
  - ▶ molecular orientation?
  - ▶ crystal structure?
- Q2. Are changes collective ( $\sqrt{N}$  factor) or not?

# Motivation: photon condensates

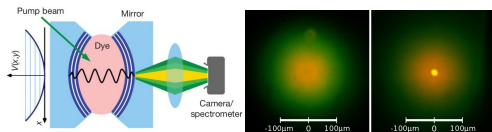
- Photon Condensate  $T \sim 300\text{K}$



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

# Motivation: photon condensates

## ● Photon Condensate $T \sim 300\text{K}$



- Q1. Relation to dye laser?
- Q2. Relation to polaritons?
- Q3. Thermalisation breakdown?

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

# Overview

## 1 Introduction

- Condensation, lasing and superradiance
- Modelling photon BEC & organic polaritons

## 2 Weak coupling: Photon BEC

- Homogeneous model & threshold
- Spatial profile and dynamics

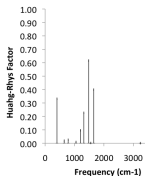
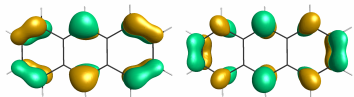
## 3 Strong coupling

- Superradiance transition
- Vibrational dressing in normal-state spectrum

## 4 Ultrastrong coupling: vibrational reconfiguration

# Toy models

## 1 Full molecular spectra electronic structure & Raman spectrum



## 2 Simplified archetypal model: Dicke-Holstein

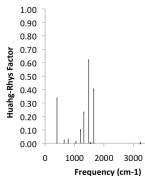
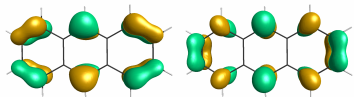
### • Each molecule: two DoF

→ Electronic state: 2LS

→ Vibrational state: harmonic oscillator

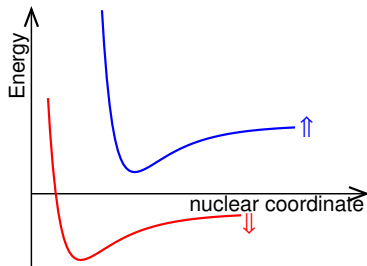
# Toy models

- 1 Full molecular spectra electronic structure & Raman spectrum



- 2 Focus on low-energy effective theory

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



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

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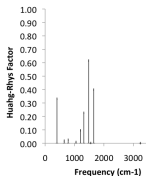
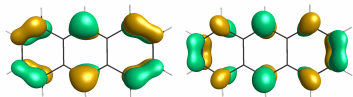
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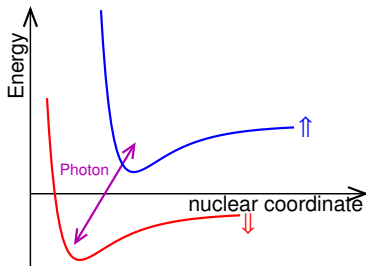
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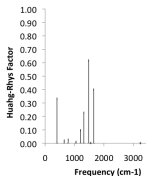
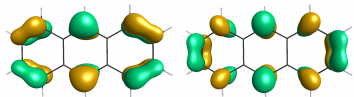
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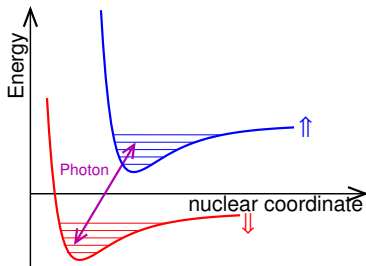
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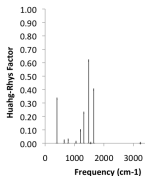
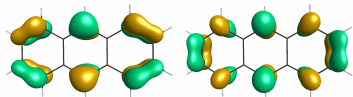
• Each molecule: two DoF

• Electronic state: 2LS

• Vibrational state: harmonic oscillator

# Toy models

- 1 Full molecular spectra electronic structure & Raman spectrum

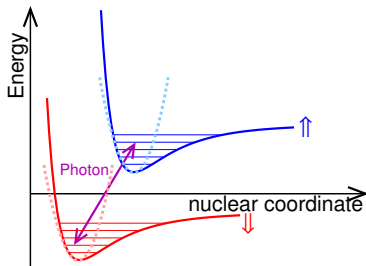


- 2 Focus on low-energy effective theory

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

- 3 Simplified archetypal model: Dicke-Holstein

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

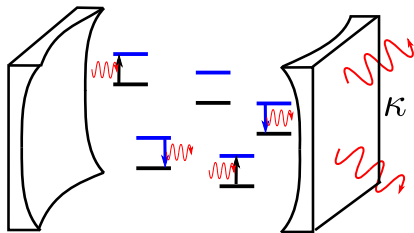


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# Dicke Holstein Model

- Dicke model: 2LS  $\leftrightarrow$  photons

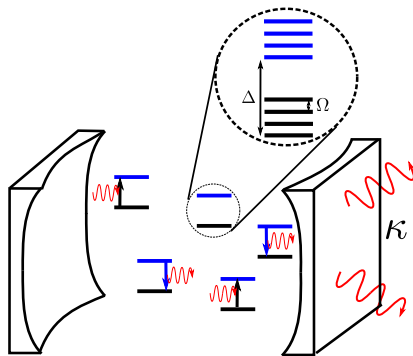
- Molecular vibrational mode
  - Phonon frequency  $\Omega$
  - Huang-Rhys parameter  $S$  — coupling strength



$$H_{\text{sys}} = \omega \psi^\dagger \psi + \sum_{\alpha} \left[ \frac{\epsilon}{2} \sigma_{\alpha}^z + g (\psi + \psi^\dagger) (\sigma_{\alpha}^+ + \sigma_{\alpha}^-) \right]$$

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# Weak coupling: Photon BEC

## 1 Introduction

- Condensation, lasing and superradiance
- Modelling photon BEC & organic polaritons

## 2 Weak coupling: Photon BEC

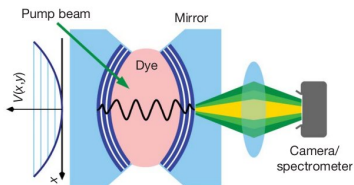
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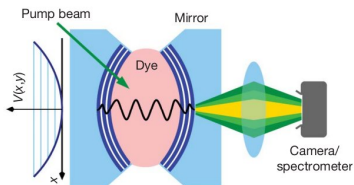


- (Curved) microcavity
- R6G dye (in solvent)

- Thermalisation of light
- Condensation at  $P > P_{th}$

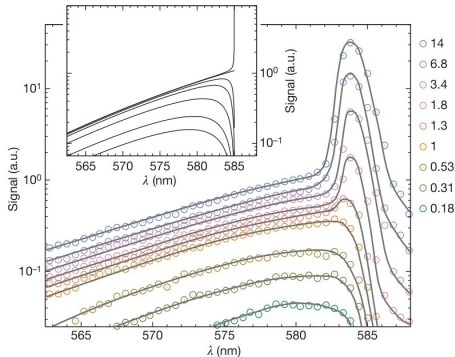
[Klaers et al, Nature, 2010]

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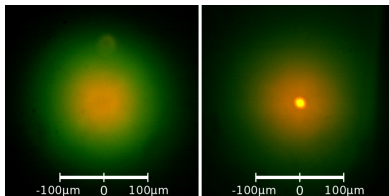
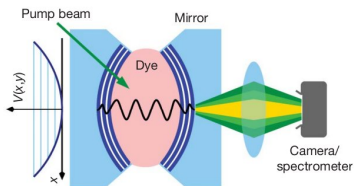
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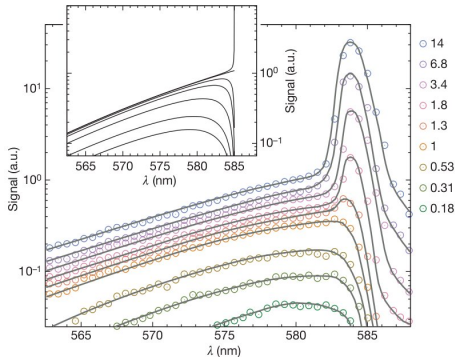
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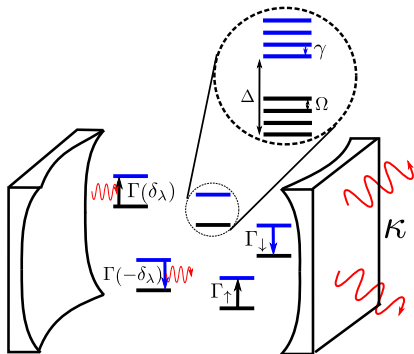
# Photon: Microscopic Model

$$H_{\text{sys}} = \sum_m \omega_m \psi_m^\dagger \psi_m + \sum_\alpha \left[ \frac{\epsilon}{2} \sigma_\alpha^z + g (\psi_m \sigma_\alpha^+ + \text{H.c.}) \right] \\ + \sum_\alpha \Omega \left\{ b_\alpha^\dagger b_\alpha + \sqrt{S} \sigma_\alpha^z (b_\alpha^\dagger + b_\alpha) \right\}$$

- **2D** harmonic oscillator

$$\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$$

- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in  $g$

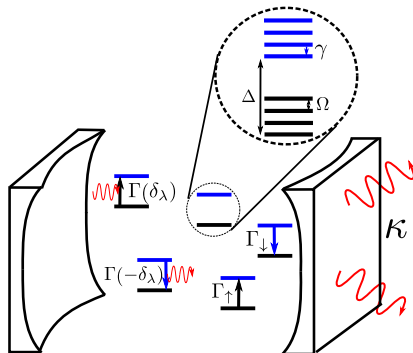


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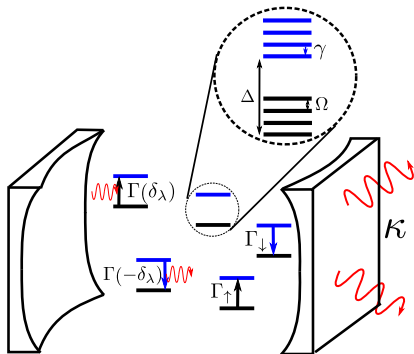
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# Microscopic model – all orders in $S$

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

$$h_\alpha = \frac{\epsilon}{2} \sigma_\alpha^z + g (\psi_m \sigma_\alpha^+ D_\alpha + \text{H.c.}) + \Omega b_\alpha^\dagger b_\alpha, \quad D_\alpha = e^{2\sqrt{S}(b_\alpha^\dagger - b_\alpha)}$$

- Master equation

$$\dot{\rho} = -i[H_0, \rho] + \sum_m \frac{\kappa}{2} \mathcal{L}[\psi_m] + \sum_\alpha \left[ \frac{\Gamma_1}{2} \mathcal{L}[\sigma_\alpha^+] + \frac{\Gamma_1}{2} \mathcal{L}[\sigma_\alpha^-] \right] + \sum_{m,\alpha} \left[ \frac{\Gamma(\delta_m - \omega_m - \epsilon)}{2} \mathcal{L}[\sigma_\alpha^+ \psi_m] + \frac{\Gamma(-\delta_m - \epsilon - \omega_m)}{2} \mathcal{L}[\sigma_\alpha^- \psi_m^\dagger] \right]$$

- Correlation function:

$$r(\delta) = 2g^2 \Re \left[ \int dt e^{-i\delta t - (\Gamma_+ + \Gamma_-)t/2} \langle \sigma_\alpha^+(t) \sigma_\alpha^-(0) \rangle \right]$$

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

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$$G(\delta) = 2\sigma^2 \pi \int dt e^{-i\delta t - (\Gamma_\uparrow + \Gamma_\downarrow)|t|^2/2} \langle \sigma_\alpha^-(t) \sigma_\alpha^-(0) \rangle$$

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

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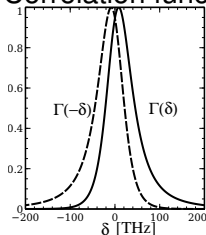
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# Steady state populations and equilibrium

- Rate equation:

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

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- Bose-Einstein distribution without losses

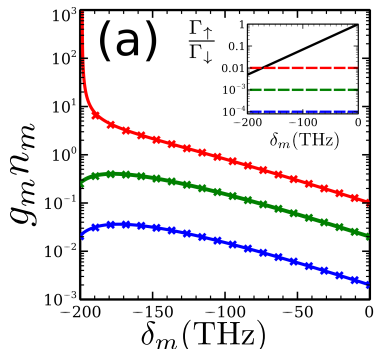
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Low loss: Thermal

[Kirton & JK PRL '13]

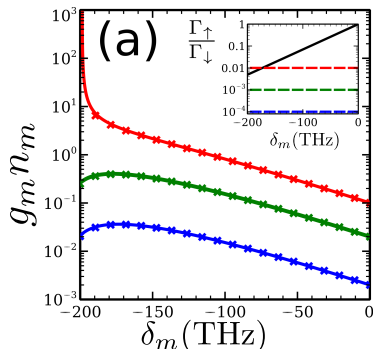
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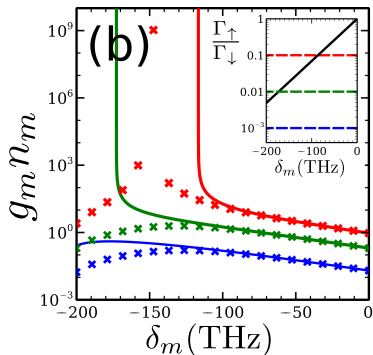
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High loss  $\rightarrow$  Laser

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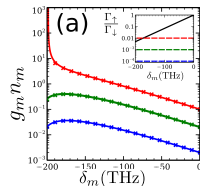
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- Below threshold,

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

- Above threshold,  $\mu \rightarrow \delta_0$



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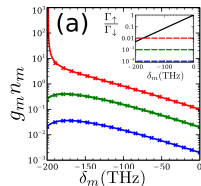
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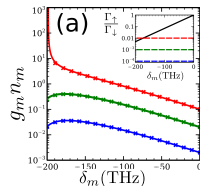
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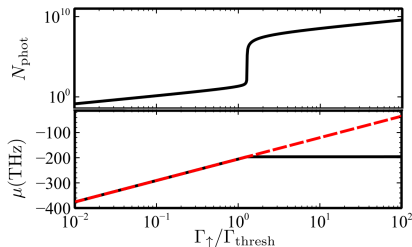
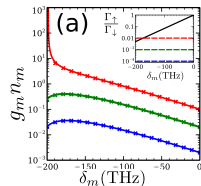
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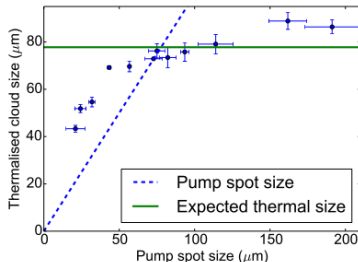
# Spatially varying pump intensity

- Consider effects of pump profile,  $\Gamma_{\uparrow}(\mathbf{r}) = \frac{\Gamma_{\uparrow} \exp(-r^2/2\sigma_p^2)}{(2\pi\sigma_p^2)^{d/2}}$

• Experiments: [Marelic & Nyman, PRA 15]

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# Modelling spatial profile.

- Varying excited density – differential coupling to modes

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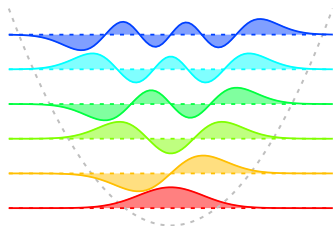
$$O_m = \int d\mathbf{r} \rho_{\uparrow}(\mathbf{r}) |\psi_{mi}(\mathbf{r})|^2, \quad \rho_{\uparrow} + \rho_{\downarrow} = \rho_m$$



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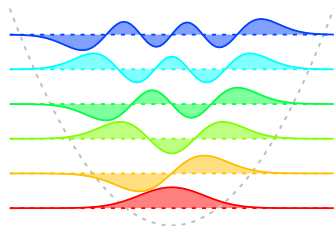
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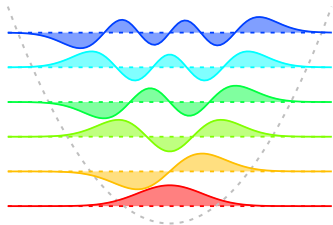
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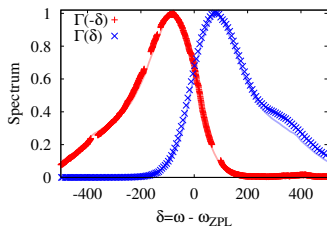
# Modelling spatial profile.

- Gauss-Hermite modes

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



- Use exact R6G spectrum



- Varying excited density – differential coupling to modes

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# Spatially varying pump: below threshold

- Far below threshold:

- ▶ If  $\kappa \ll \rho_m \Gamma(\delta_m)$ , 
$$\frac{n_m}{n_m + 1} \simeq e^{-\beta \delta_m} \times \int d\mathbf{r} \rho_{\uparrow}(\mathbf{r}) |\psi_m(\mathbf{r})|^2$$

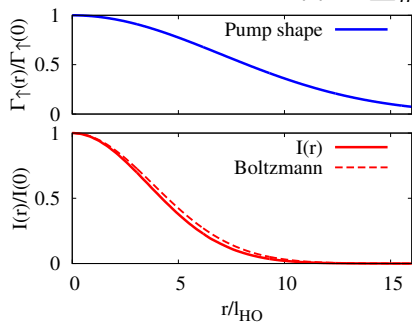
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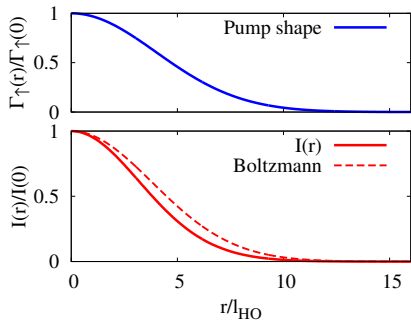


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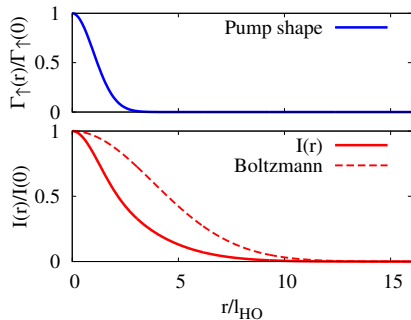


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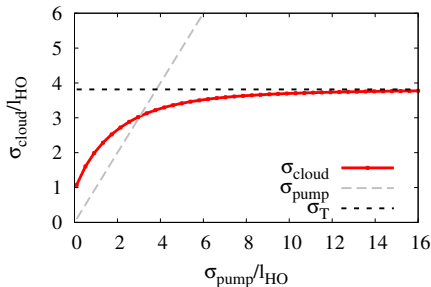
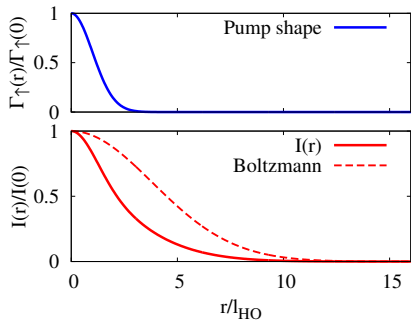


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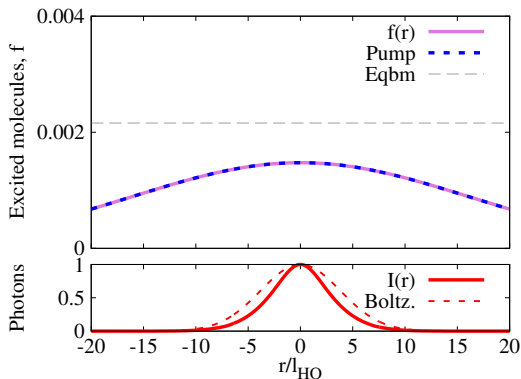
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# Near threshold behaviour

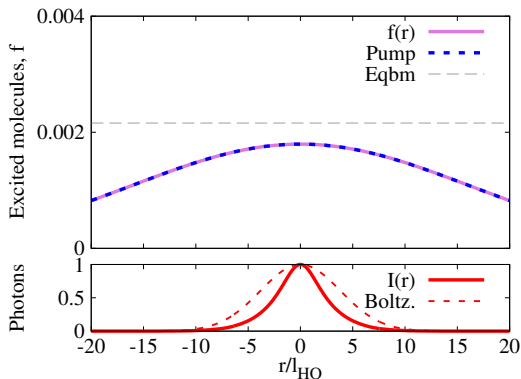


- Large spot,  $\sigma_p \gg l_{\text{HO}}$

- "Gain saturation" at centre

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

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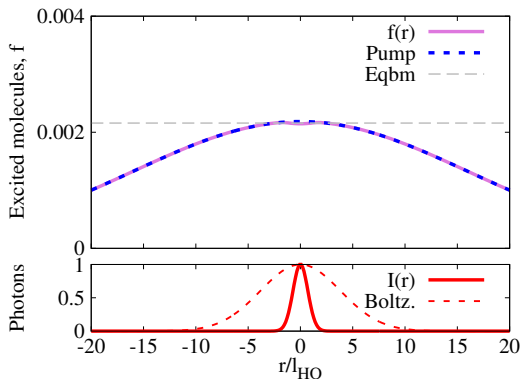


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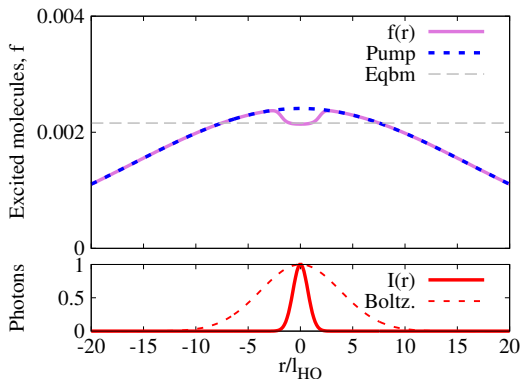
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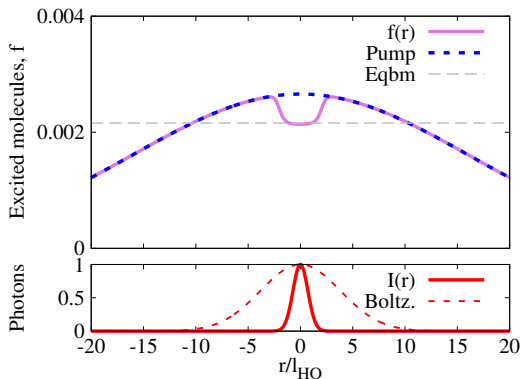
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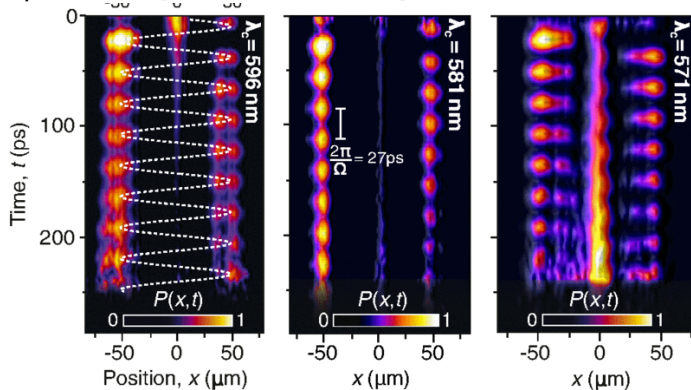
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# Off centre pumping; oscillations

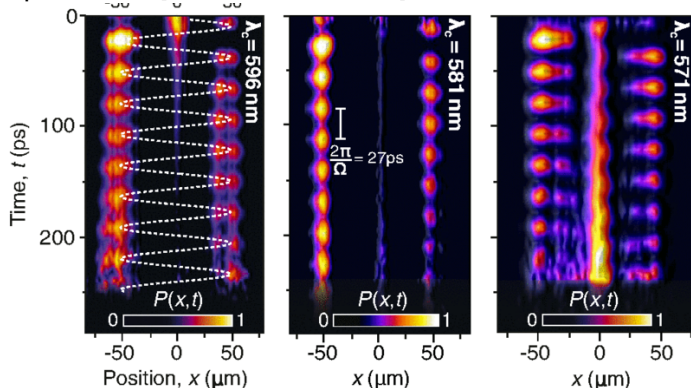
- Experiments [Schmitt *et al.* PRA '15]



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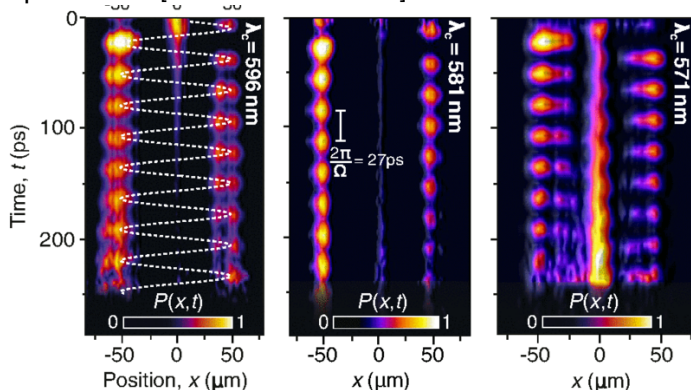


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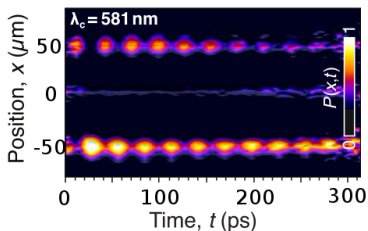
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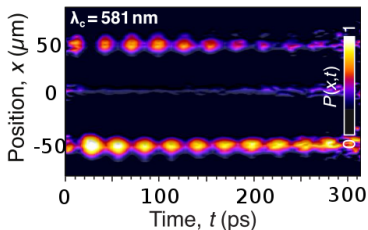
# Limit of rate equations



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

- Oscillations: beating of modes.
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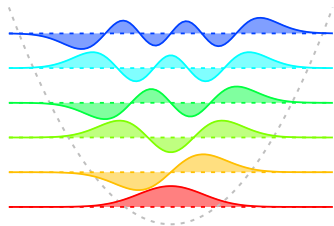


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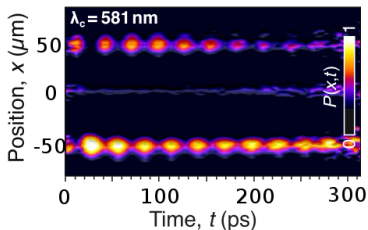
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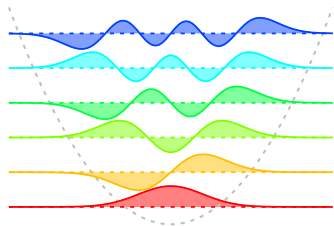
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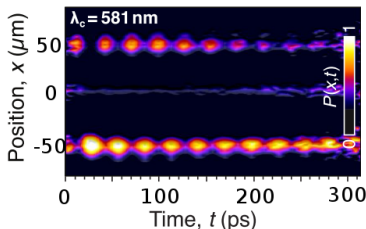
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# Limit of rate equations



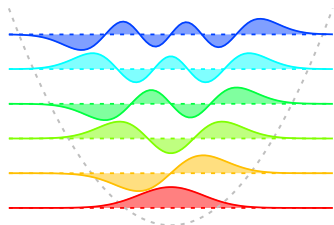
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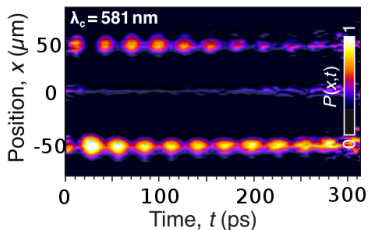
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Emission must create coherence between non-degenerate modes.

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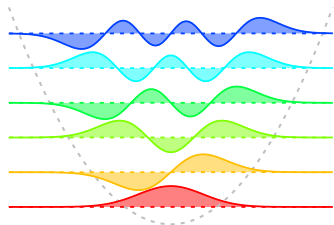


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- Following toy model, use Redfield theory:

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• Semiclassical equations for  $n_{m,m'} = \langle \hat{a}_m^\dagger \hat{a}_{m'} \rangle$  and  $f(r)$ .

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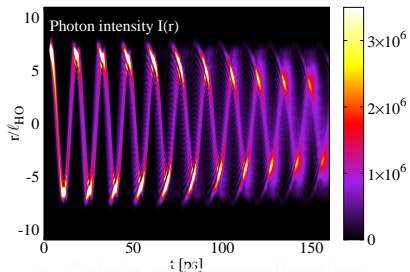
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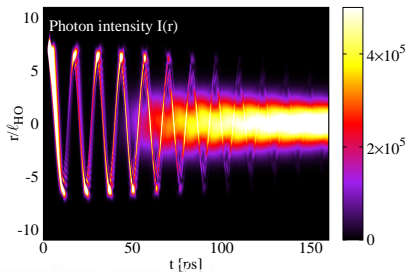


# Dynamics from model

## Longer cavity



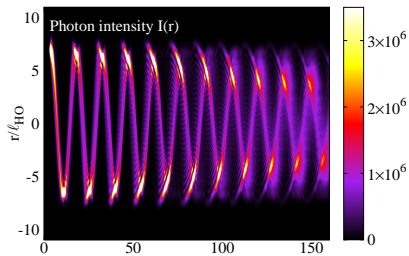
## Shorter cavity



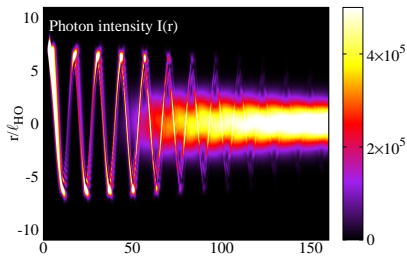
Origin of thermalisation — reabsorption, see [17]

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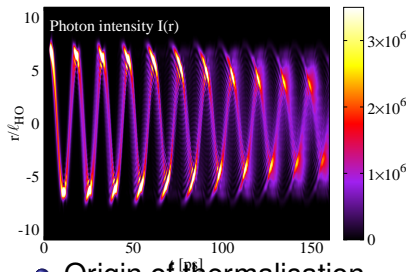
Shorter cavity



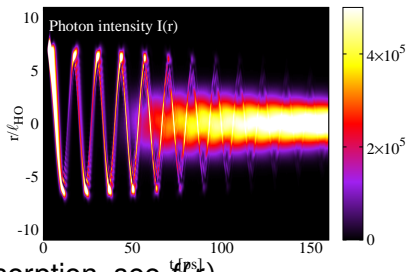
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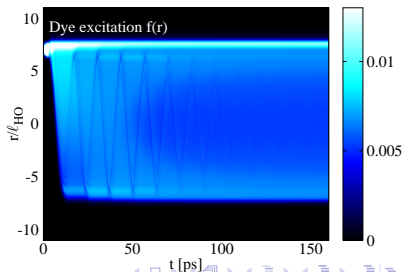
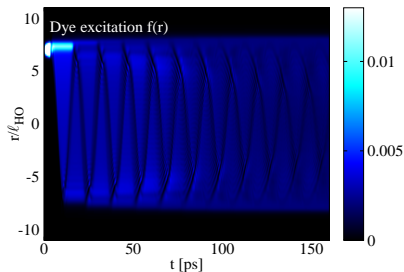
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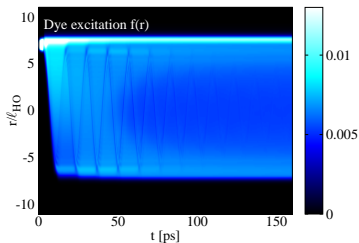
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# Thermalisation at late times

- Reabsorption “fills-in” excited molecules

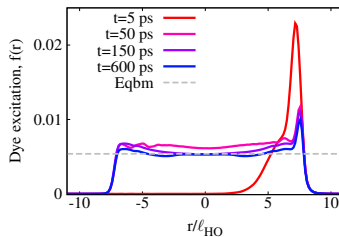
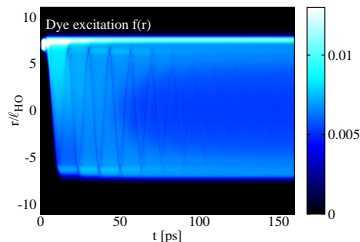
● Reach thermal equilibrium,  $f = [e^{-\beta\epsilon} + 1]^{-1}$



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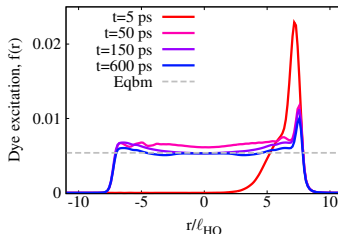
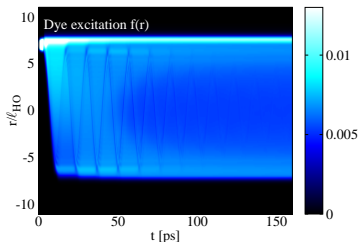
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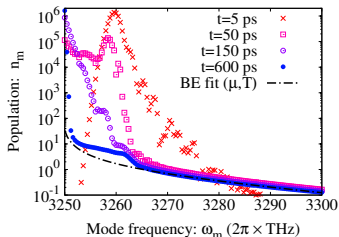
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# Strong coupling

## 1 Introduction

- Condensation, lasing and superradiance
- Modelling photon BEC & organic polaritons

## 2 Weak coupling: Photon BEC

- Homogeneous model & threshold
- Spatial profile and dynamics

## 3 Strong coupling

- Superradiance transition
- Vibrational dressing in normal-state spectrum

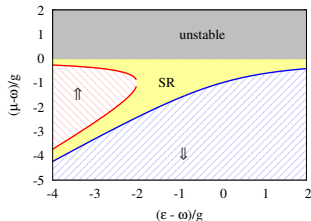
## 4 Ultrastrong coupling: vibrational reconfiguration

# Strong coupling phase diagram — mean field

- Mean field — single photon mode

$$H = \omega \psi^\dagger \psi + \sum_{\alpha} \left[ \frac{\epsilon}{2} \sigma_{\alpha}^z + g \left( \psi \sigma_{\alpha}^+ + \psi^\dagger \sigma_{\alpha}^- \right) + \Omega \left\{ b_{\alpha}^\dagger b_{\alpha} + \sqrt{S} \left( b_{\alpha}^\dagger + b_{\alpha} \right) \sigma_{\alpha}^z \right\} \right]$$

- Dicke phase diagram vs  $\mu$



- $S$  reduces  $g_{\text{eff}}$
- Reentrant behaviour — Min  $\mu$  at  $k_B T \sim 0.1\Omega$

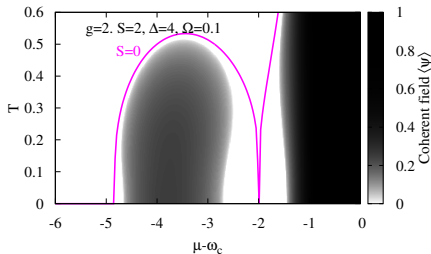
[Cwik *et al.* EPL '14]



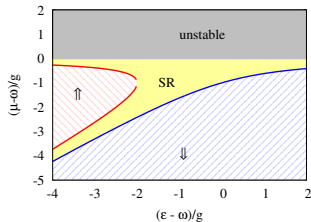
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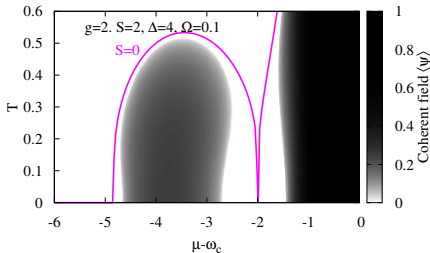
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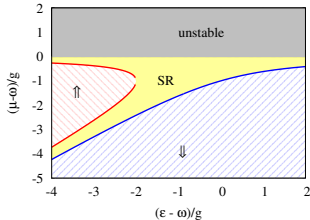
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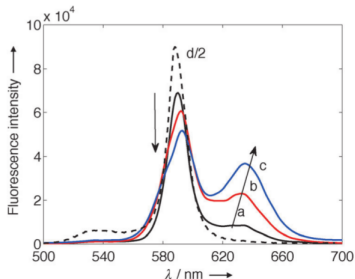
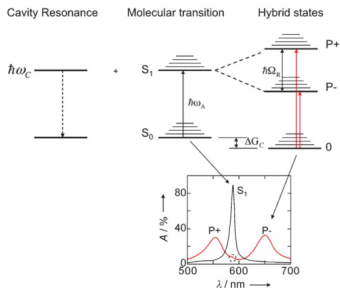
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# Strong coupling experimental features

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



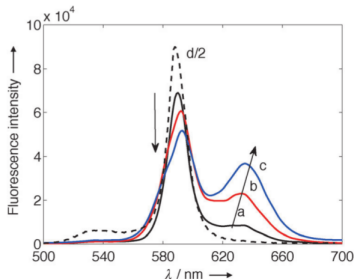
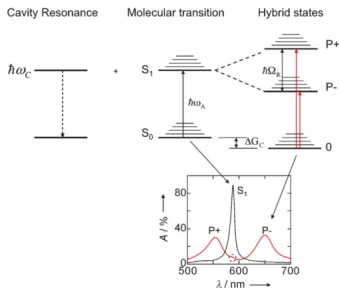
[Canaguier-Durand *et al.* Angew. Chem. '13]

- Polariton vs molecular spectral weight – chemical eqbm
- Temperature dependent

● Questions:

# Strong coupling experimental features

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



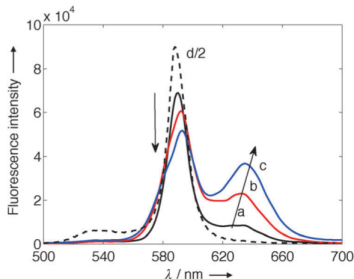
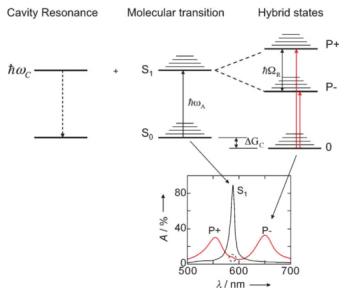
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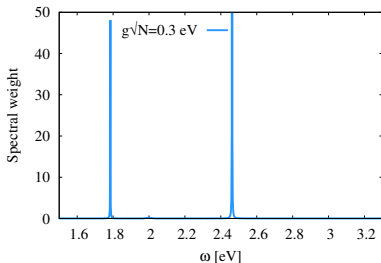
- ▶ Polariton vs molecular spectral weight – chemical eqbm
- ▶ Temperature dependent
- Questions:
  - ▶ Microscopic picture?
  - ▶ Vibrationally dressed spectrum + disorder

# Disordered molecules — spectrum

- Calculate Green's function  $G^R(\nu)$ :

$$T(\nu) \propto |G^R(\nu)|^2, \quad A(\nu) \propto -\Im[G^R(\nu)] + (\text{interference})$$

• Ultra-strong coupling — renormalised photon



• Central peak:

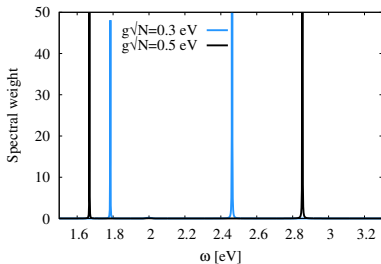
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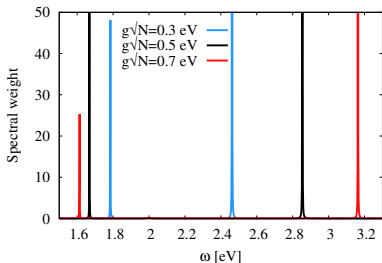


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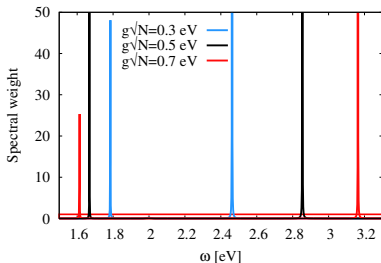
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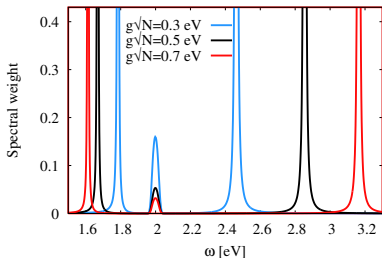


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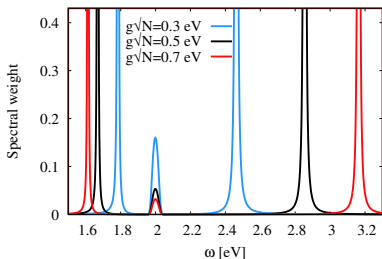
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[Houdré *et al.*, PRA '96]

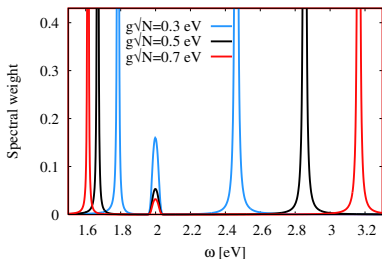
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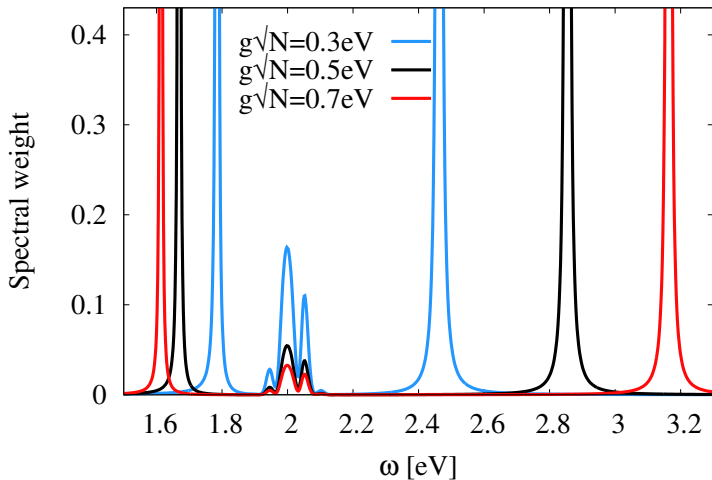
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# Disordered molecules — vibrational mode

- But: spectrum with vibrational sidebands,  $S = 0.02$

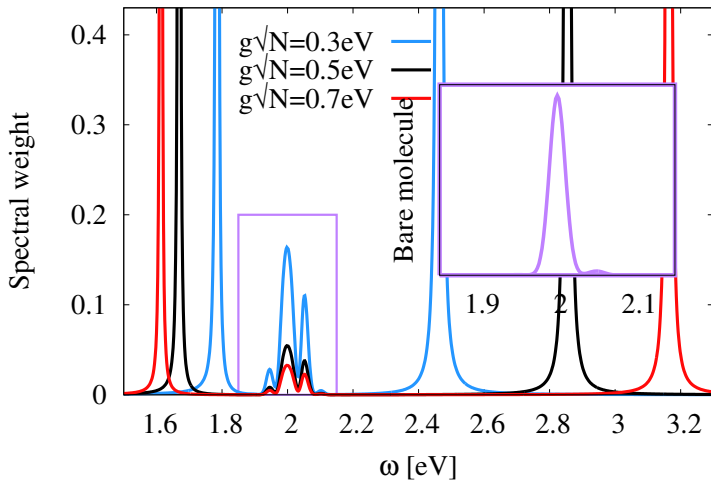
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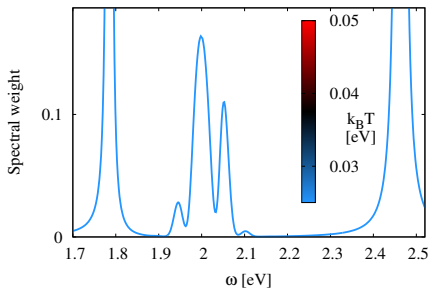




# Disordered molecules + vibrations – vs temperature

- vs vs temperature

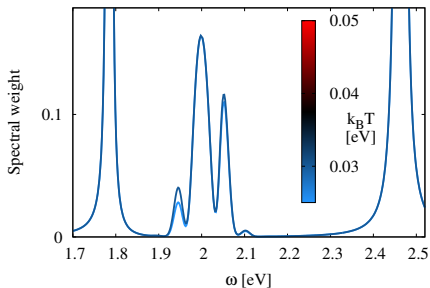
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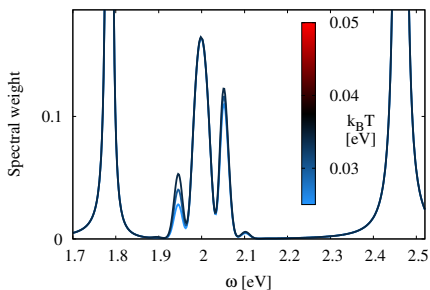
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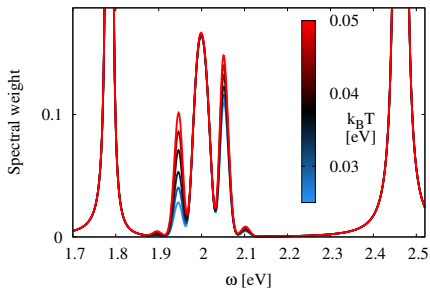
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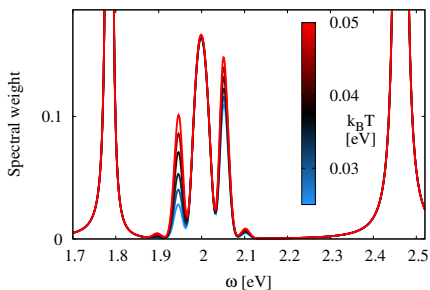
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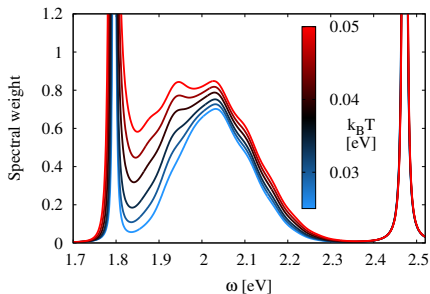


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# Ultrastrong coupling: vibrational reconfiguration

## 1 Introduction

- Condensation, lasing and superradiance
- Modelling photon BEC & organic polaritons

## 2 Weak coupling: Photon BEC

- Homogeneous model & threshold
- Spatial profile and dynamics

## 3 Strong coupling

- Superradiance transition
- Vibrational dressing in normal-state spectrum

## 4 Ultrastrong coupling: vibrational reconfiguration

# Molecular reconfiguration

- Central peak — depends on  $g$ , not  $T$ .
- Can  $g_{\text{eff}}$  depend on  $T$ ?

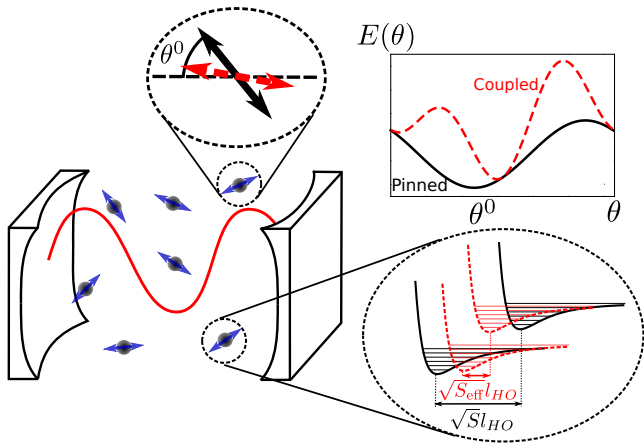
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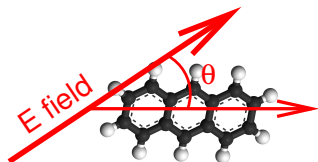
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# Rotational reorientation

- Rotational degrees of freedom



- Effective Hamiltonian

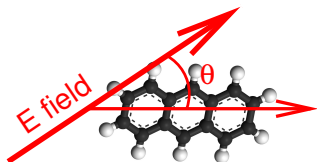
$$H = \dots + \sum_{\alpha} \left[ \dots + g_{\alpha,k} \cos(\theta_{\alpha}) (\psi_k^{\dagger} + \psi_{-k}) \sigma_{\alpha}^x + E_0(\theta_{\alpha}) \right]$$

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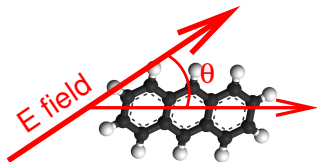
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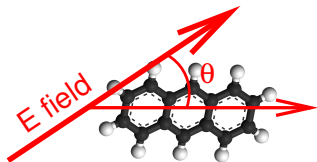
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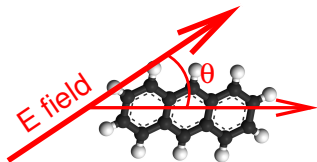
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$$S \rightarrow S(1 - 2K_1), \quad K_1 = \sum_k \frac{g_k^2}{(\omega_k + \epsilon)^2}$$

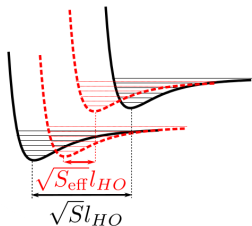
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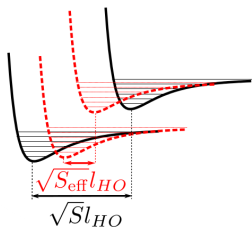
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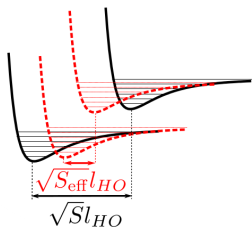
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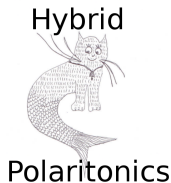
# Acknowledgements

GROUP:



COLLABORATORS: S. De Liberato (Southampton)

FUNDING:



# ICSCE 8

Edinburgh, 25<sup>th</sup>–29<sup>th</sup> April, 2016.



*Plenary speakers:* Ataç İmamoğlu, Peter Zoller.

*Invited speakers:* Alberto Amo, Mete Atatüre, Natasha Berloff, Charles Bardyn, Cristiano Ciuti, Thomas Ebbesen, Thierry Giamarchi, Jan Klärs, Dmitry Krizhanovskii, Xiaoqin (Elaine) Li, Peter Littlewood, Allan MacDonald, Francesca Marchetti, Keith Nelson, Pavlos Lagoudakis, Lukas Sieberer, Vivien Zapf.

*Early-bird registration & abstract deadline: 31st January 2016.*

**Final registration deadline: March 2016.**

<http://www.st-andrews.ac.uk/~icsce8>

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Workshop on Engineering Quantum Matter: From  
Understanding to Control

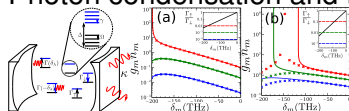
June 8-10, St Andrews, Scotland, UK



<http://eqm2016.co.uk>

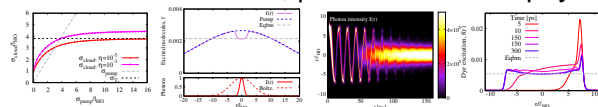
# Summary

- Photon condensation and thermalisation



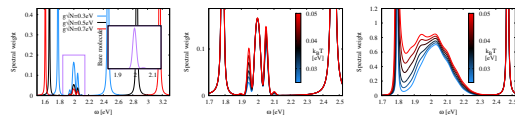
[Kirton & JK, PRL '13, PRA '15]

- Photon condensation, pattern formation physics



[JK & Kirton, PRA '16]

- Vibrational dressing, and possible reconfiguration



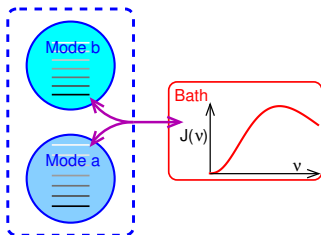
[Cwik *et al.* PRA '16]

# Extra Slides

- 5 Toy problem – two bosonic modes
- 6 First order transitions due to phonons

# Toy problem: two bosonic modes

- Basic problem: Emission from thermal bath



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



# Toy problem: naïve solutions

- Two “expected” behaviours:
  - ▶ At resonance: “weak lasing” — coupling to bath dominates

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

• Far from resonance: pointer states are eigenstates

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

- Explicit derivation → Redfield theory

$$\begin{aligned} \partial_t \rho = & -i[H, \rho] + \sum_j \mathcal{L}_j^\downarrow \left( 2\hat{\psi}_j \rho \hat{\psi}_j^\dagger - [\rho, \hat{\psi}_j^\dagger \hat{\psi}_j]_+ \right) \\ & + \sum_j \mathcal{L}_j^\uparrow \left( 2\hat{\psi}_j^\dagger \rho \hat{\psi}_j - [\rho, \hat{\psi}_j \hat{\psi}_j^\dagger]_+ \right). \end{aligned}$$

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# Toy problem: exact solution

- Solve via Laplace transform. Find  $F_{ij}(t) = \langle \hat{\psi}_i^\dagger(t) \hat{\psi}_j(t) \rangle$

- Steady state:

- Time evolution —

$$F_{ab}(t) \sim \exp(-\alpha \Delta^2 t)$$

- Always some coherence

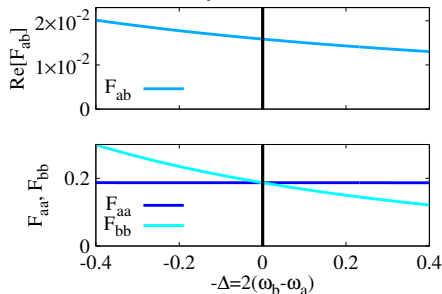
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- Solve via Laplace transform. Find  $F_{ij}(t) = \langle \hat{\psi}_i^\dagger(t) \hat{\psi}_j(t) \rangle$
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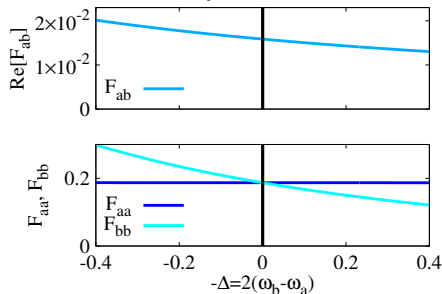


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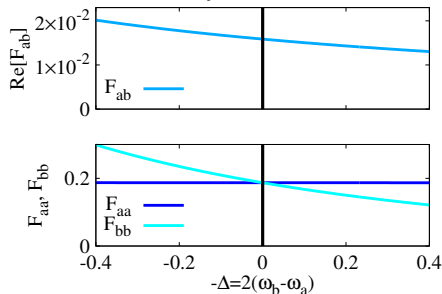
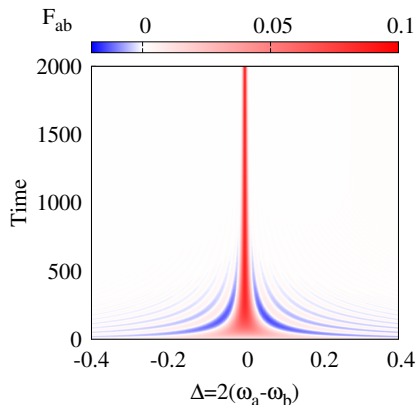
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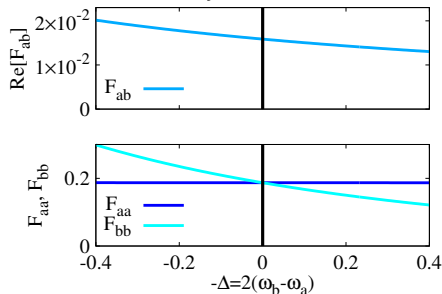
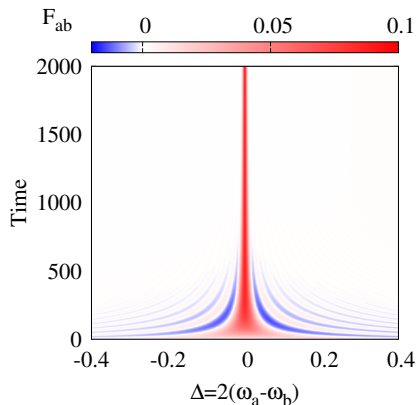
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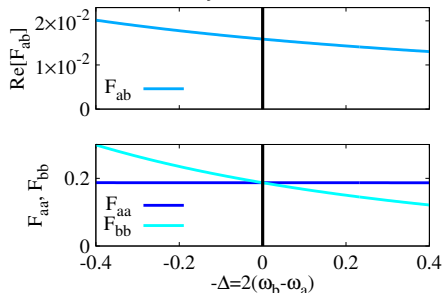
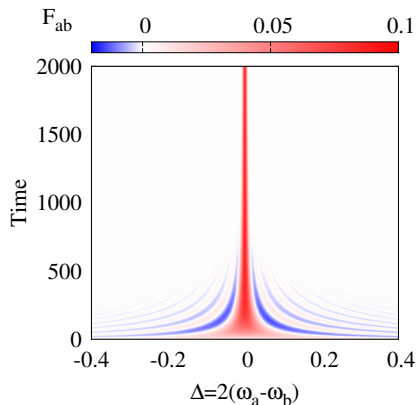
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Unsecularised Redfield theory:

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

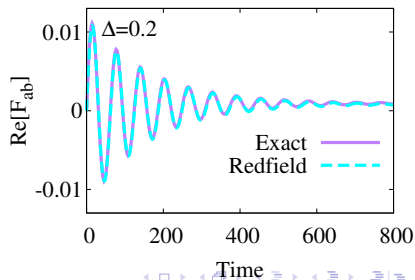
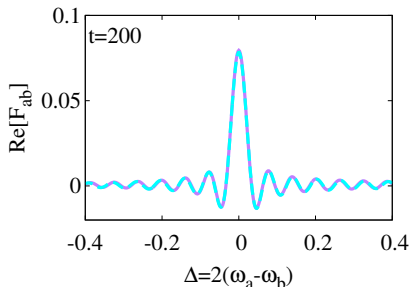
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- Secularisation (in eigenbasis of  $\hat{H}$ ):  $L_{ij}^{\uparrow,\downarrow} \rightarrow L_{ii}^{\uparrow,\downarrow} \delta_{ij} \rightarrow F_{ab} = 0$

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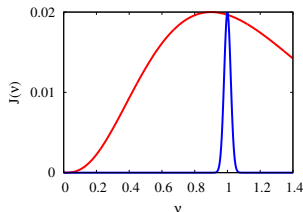
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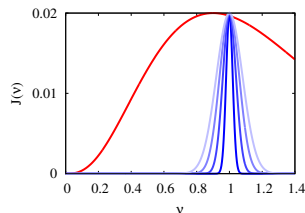


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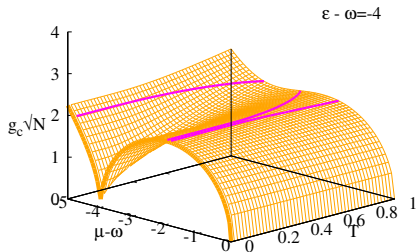
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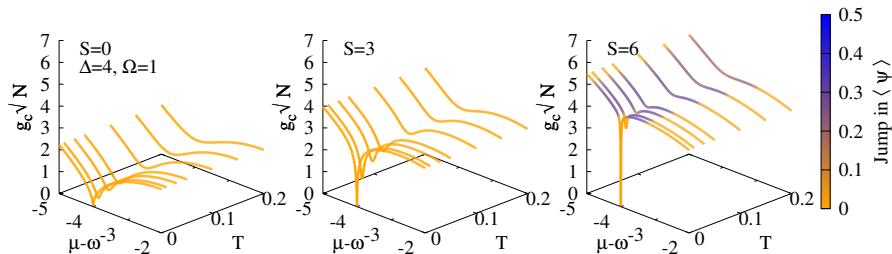
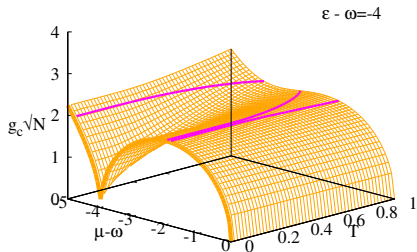
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$$H_\alpha \rightarrow \tilde{H}_\alpha = e^{K_\alpha} H_\alpha e^{-K_\alpha} \quad K = \sqrt{S} S_\alpha^z (b_\alpha^\dagger - b_\alpha)$$

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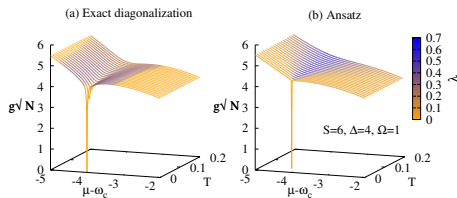
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# Collective polaron formation

- Compares well at  $S \gg 1$
- Coherent bosonic state



- Feedback: Large/small  $g_{\text{eff}} \leftrightarrow \lambda = \langle \psi \rangle$
- Variational free energy

$$F = (\omega_c - \mu)\lambda^2 + N \left\{ \Omega \left[ \zeta^2 - S \frac{\eta(2 - \eta)}{4} \right] - T \ln \left[ 2 \cosh \left( \frac{\xi}{T} \right) \right] \right\}$$

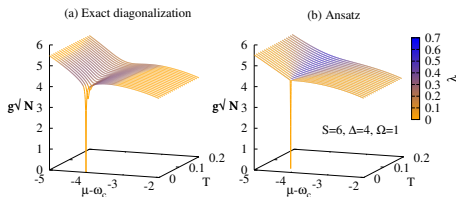
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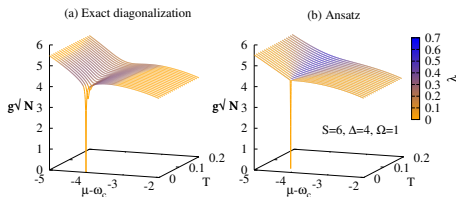
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