

# Spatial dynamics, thermalization and breakdown of thermalization in photon condensates

Jonathan Keeling

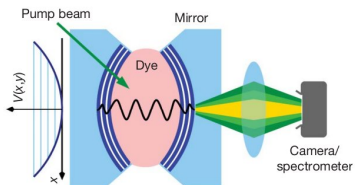


University of  
St Andrews

1413-2013

Windsor, August 2015

# Photon BEC experiments

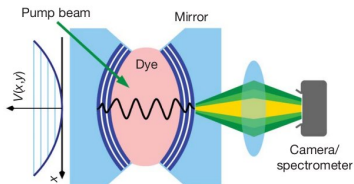


- (Curved) microcavity
- R6G dye (in solvent)

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

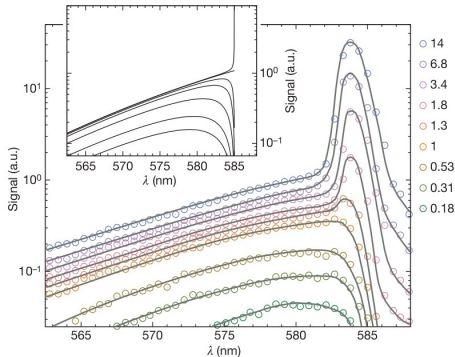
[Klaers et al, Nature, 2010]

# Photon BEC experiments



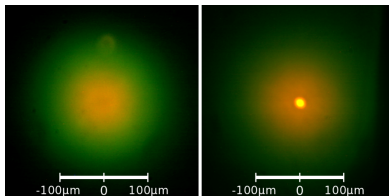
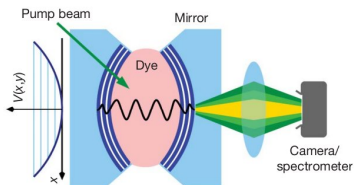
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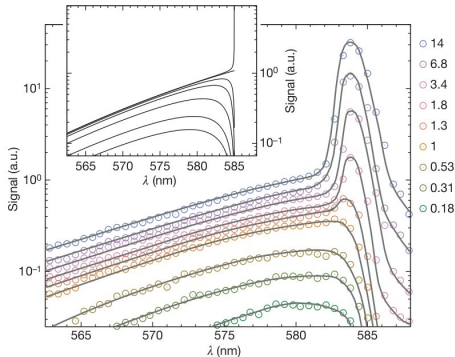


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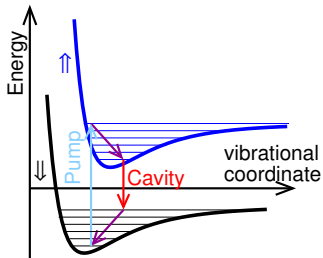
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# Relation to dye laser

## 4 Level Dye Laser

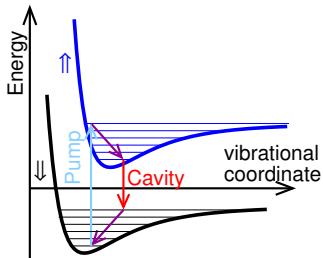


- No electronic inversion
- No strong coupling

- No single cavity mode
  - Condensate mode is not maximum gain
  - Gain/Absorption in balance
- Thermalised many-mode system

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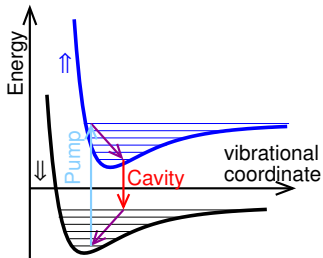


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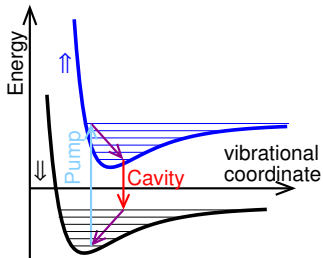
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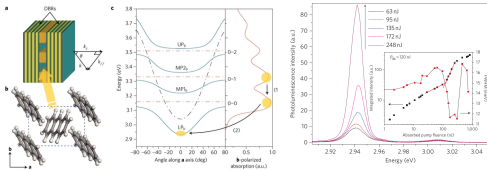
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# Motivation: organic polariton condensates

## ● Anthracene Polariton Lasing

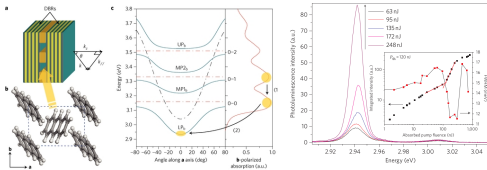
$T \sim 300\text{K}$



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

# Motivation: organic polariton condensates

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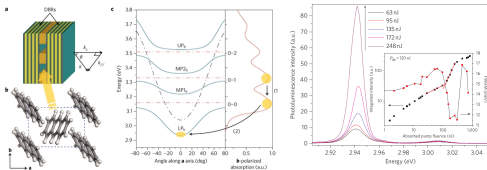


- Q1. Vibrational replicas?
- Q2. Relevance of disorder?
- Q3. Lasing vs condensation?

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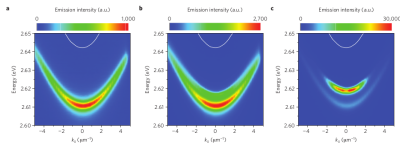
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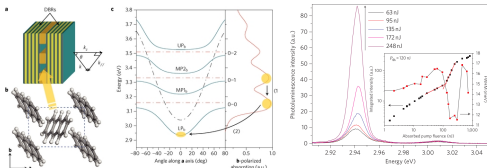
- Polariton condensates, other materials, e.g. polymers:



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

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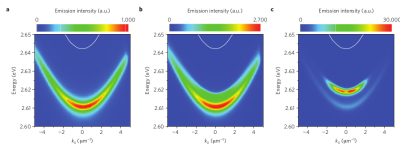
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- Polariton condensates, other materials, e.g. polymers:



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

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

## 1 Introduction to Photon BEC

## 2 Modelling photon BEC

- Steady state
- Approach to steady state

## 3 Spatial profile

- Steady state
- Spatial oscillations

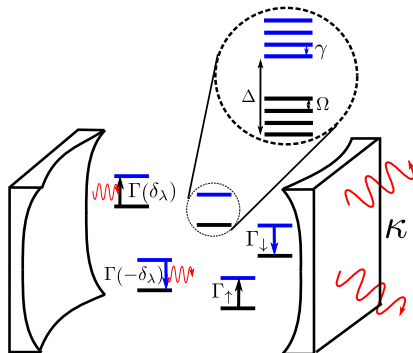
# Modelling

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

- **2D** harmonic oscillator

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

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

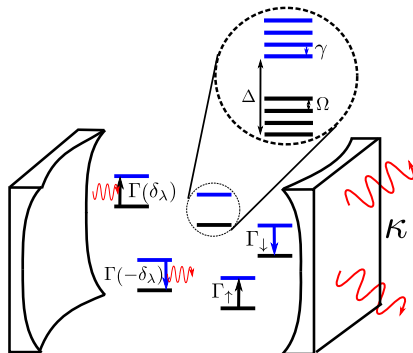


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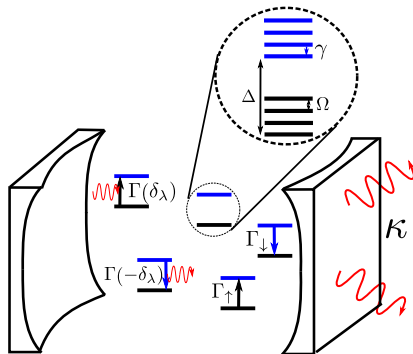
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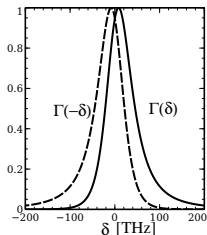




# Modelling

## Master equation

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



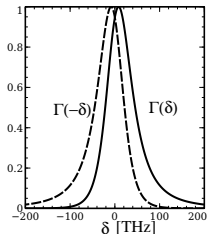
• Kennard-Stepanov  
 $\Gamma(+\delta) \simeq \Gamma(-\delta) e^{\beta\hbar\delta}$

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

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## Distribution $g_m n_m$

- Master equation  $\rightarrow$  Rate equation

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

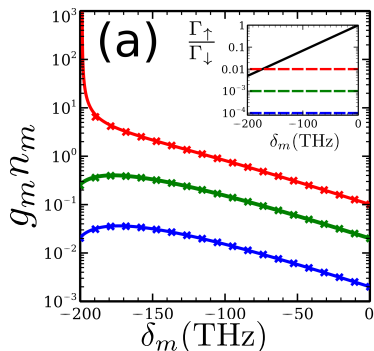
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Low loss: Thermal

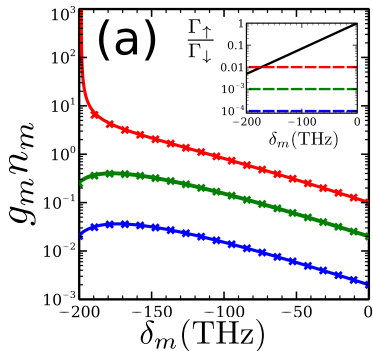
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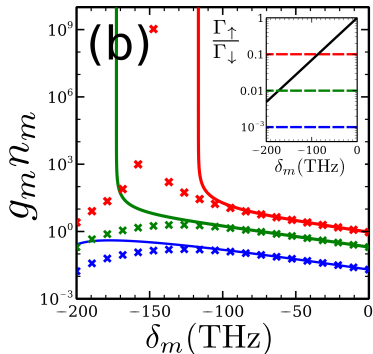
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Low loss: Thermal

[Kirton & JK PRL '13]



High loss  $\rightarrow$  Laser

# Chemical potential?

- Steady state distribution:

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

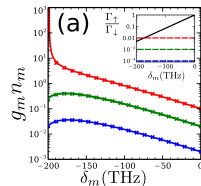
- $\kappa \ll N\Gamma(\beta)$ , Kennard-Stepanov

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

- Below threshold,

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

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



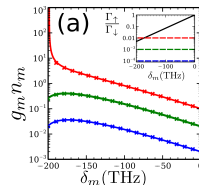
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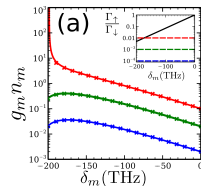
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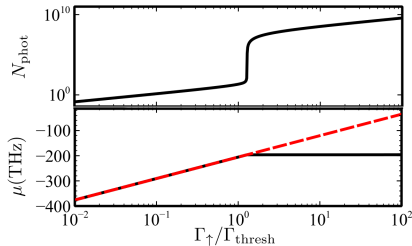
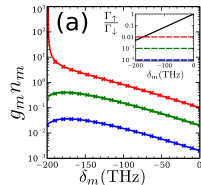
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- Below threshold,

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



# Modelling photon BEC

1 Introduction to Photon BEC

2 Modelling photon BEC

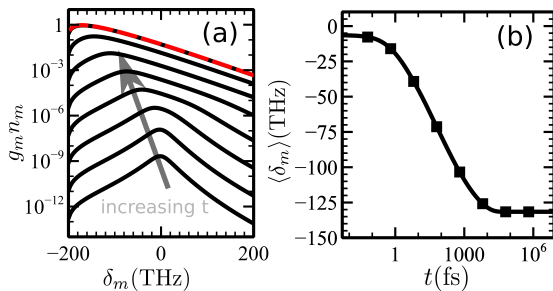
- Steady state
- Approach to steady state

3 Spatial profile

- Steady state
- Spatial oscillations

# Time evolution

- Initial state: excited molecules
  - Initial emission, follows gain peak
  - Thermalisation by repeated absorption

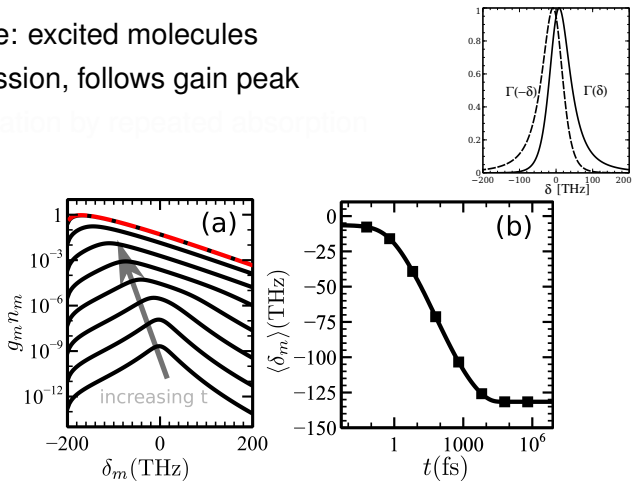


[Kirton & JK PRA '15]

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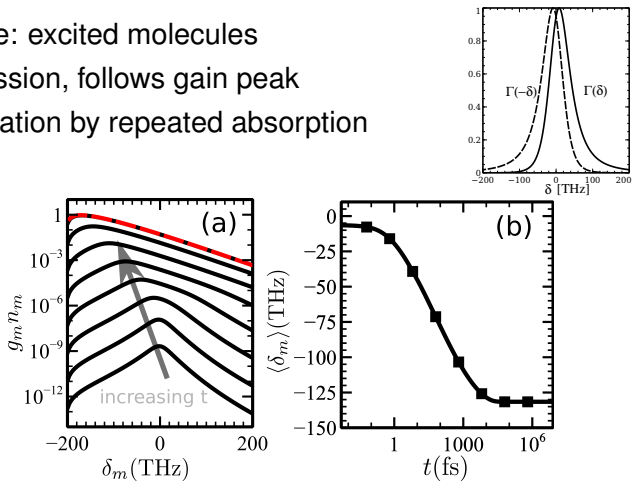
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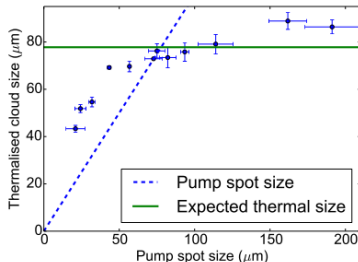
# Spatially varying pump intensity

- 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

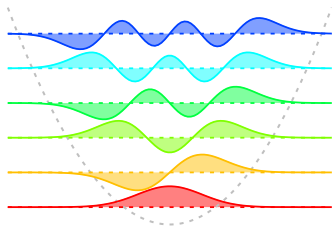
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$$O_m = \int d\mathbf{r} \rho_{\uparrow}(\mathbf{r}) |\psi_{m1}(\mathbf{r})|^2, \quad \rho_{\uparrow} + \rho_{\downarrow} = \rho_m$$

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- Gauss-Hermite modes

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



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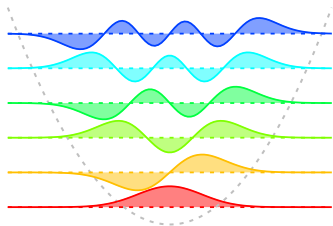
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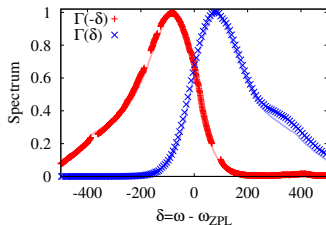
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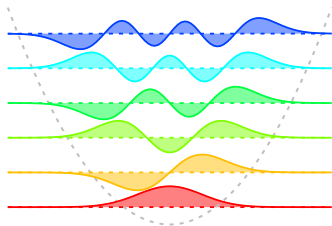
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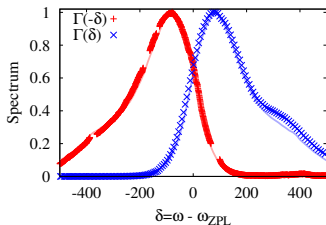
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$$\partial_t \rho_{\uparrow}(\mathbf{r}) = -\tilde{\Gamma}_{\downarrow}(\mathbf{r}) \rho_{\uparrow}(\mathbf{r}) + \tilde{\Gamma}_{\uparrow}(\mathbf{r}) \rho_{\downarrow}(\mathbf{r})$$

# Spatially varying pump: below threshold

- Far below threshold:

- ▶ Excitation:  $\rho_{\uparrow}(\mathbf{r}) \simeq \rho_m \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} \ll \rho_m$

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

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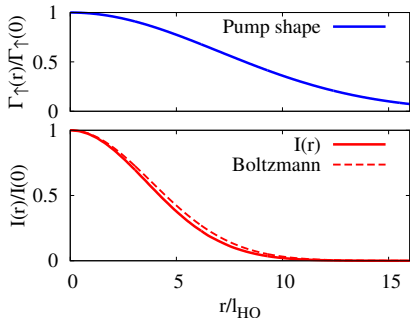
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$$\mu = \frac{\kappa}{\rho_m \max[\Gamma(\delta)]}$$

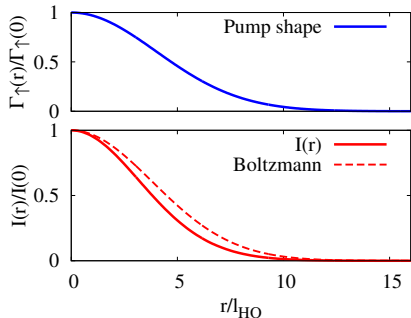
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- Excitation:  $\rho_{\uparrow}(\mathbf{r}) \simeq \rho_m \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} \ll \rho_m$

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

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



$$\mu = \frac{\kappa}{\rho_m \max_i |\Gamma_i(r)|}$$

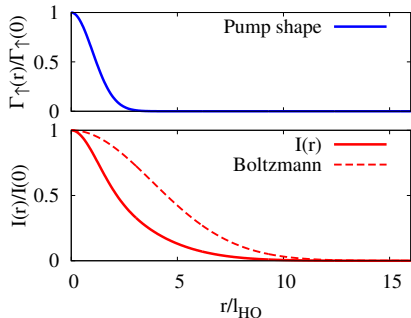
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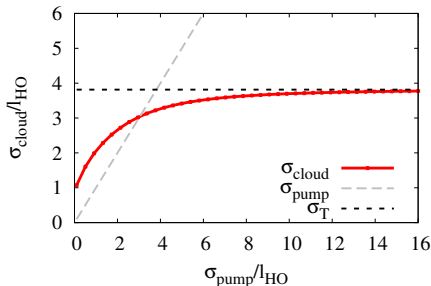
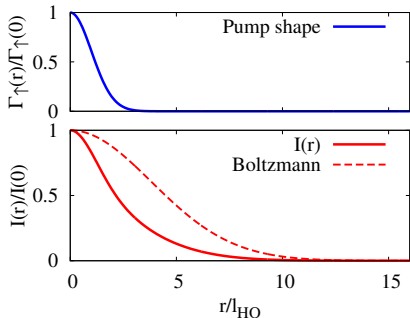
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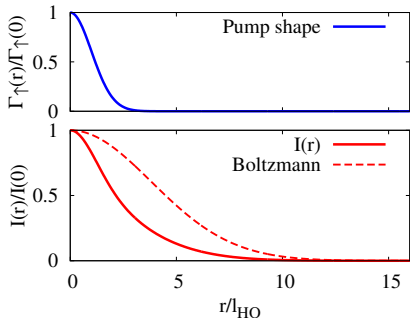
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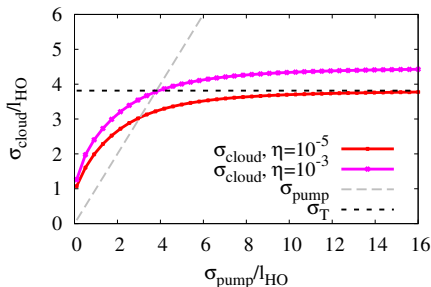
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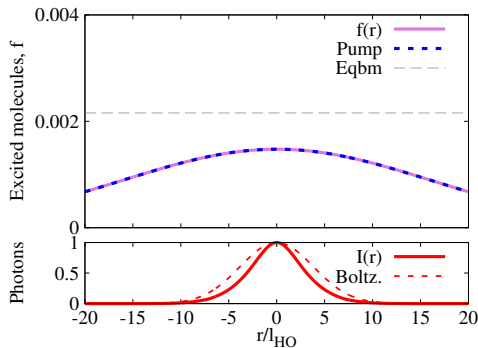
- $\eta = \frac{\kappa}{\rho_m \max[\Gamma(\delta)]}$



# Near threshold behaviour

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

- Non Boltzmann peak — BEC
- "Gain saturation" at centre

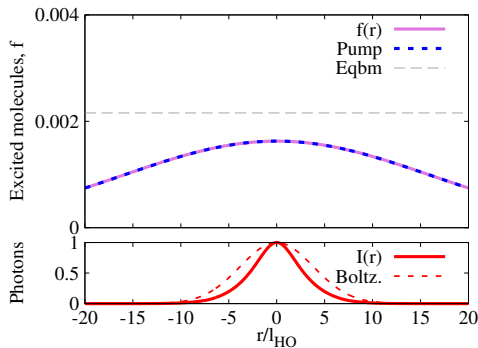


- Saturation of  $f(r) = 1/(1 + e^{-\beta \mu})$  — spatial equilibration

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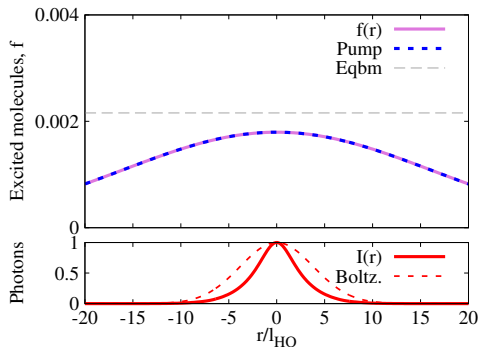


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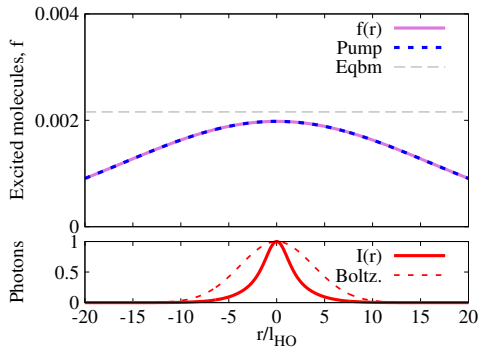


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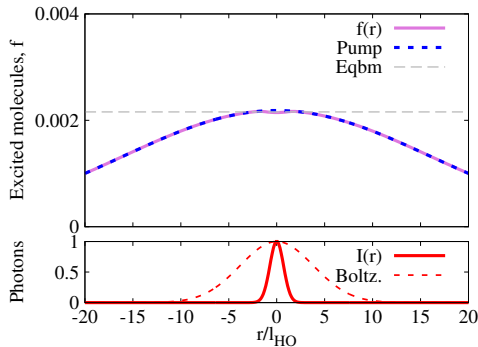
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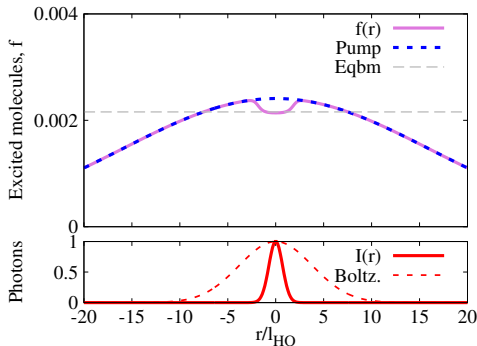


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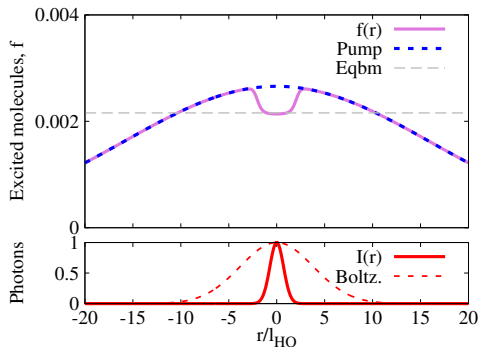


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# Near threshold behaviour

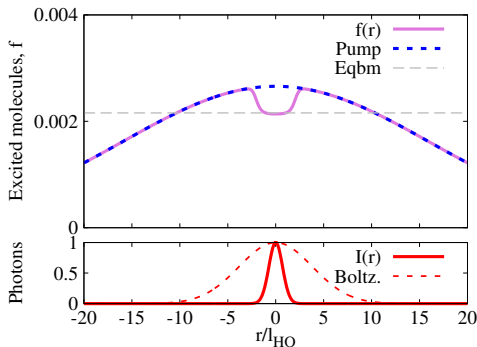
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# Spatial oscillations

1 Introduction to Photon BEC

2 Modelling photon BEC

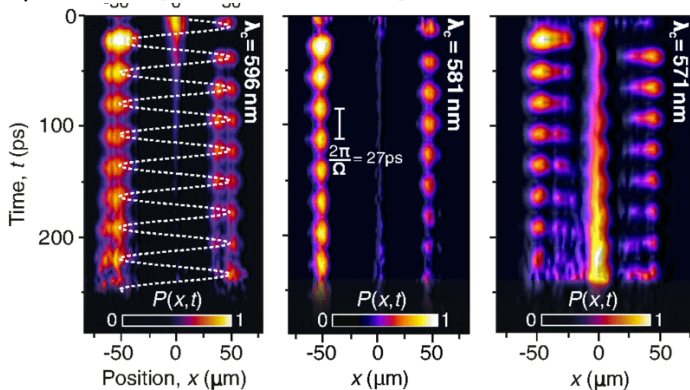
- Steady state
- Approach to steady state

3 Spatial profile

- Steady state
- **Spatial oscillations**

# Off centre pumping; oscillations

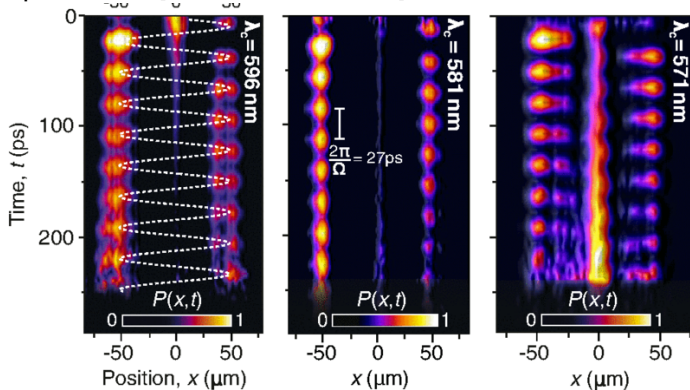
- Experiments [Schmitt *et al.* PRA '15]



- Oscillations in space – beating of normal modes
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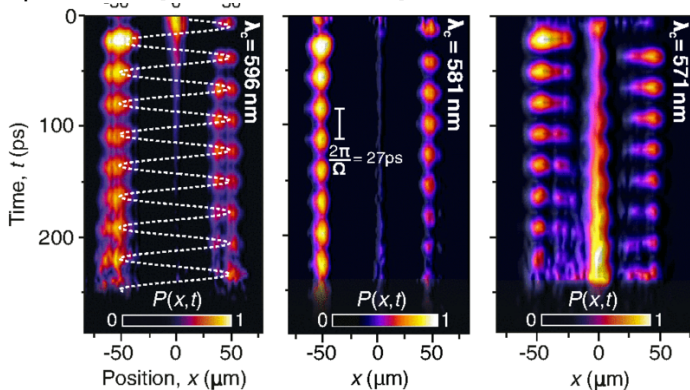


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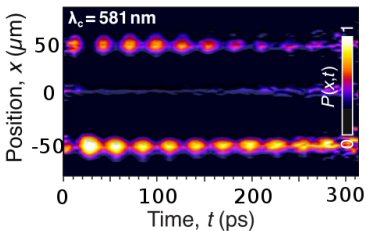
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# Limit of rate equations



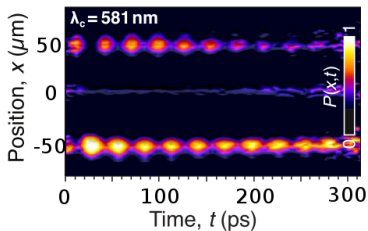
$$\partial_t n_m = -\kappa n_m + \Gamma(-\delta_m)(n_m + 1)N_\uparrow - \Gamma(\delta_m)n_m N_\downarrow$$

- Describes emission into Gauss-Hermite mode  $m$

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

- Oscillations: beating of modes.
- Need  $I(x) = \sum_{m, m'} n_{m, m'} \psi_m(x) \psi_{m'}(x)$

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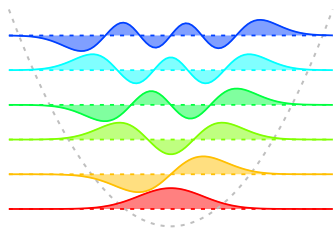


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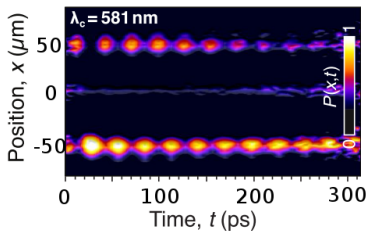
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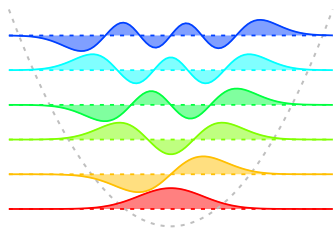
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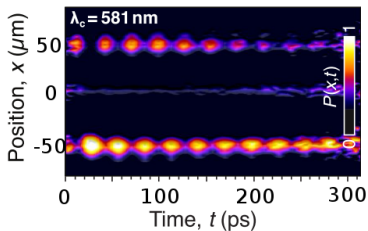
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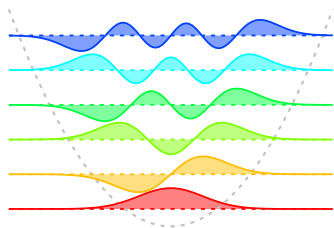


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Emission must create coherence between non-degenerate modes.

# Modelling

- Full master equation required

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

- Not secular approximation

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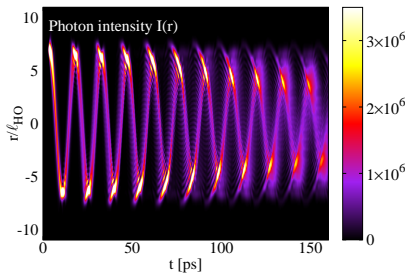
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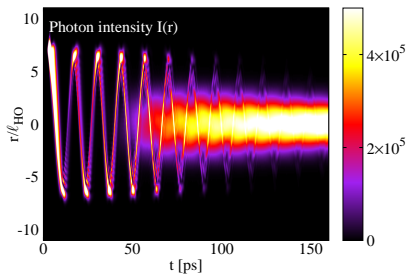
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# Dynamics from model

Smaller  $\omega_C$



Larger  $\omega_C$

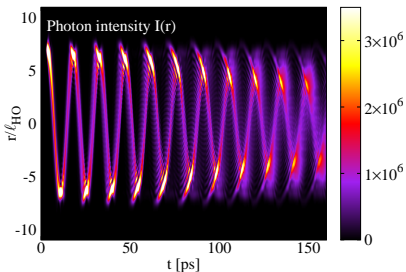


• Origin of thermalisation — reabsorption, see  $I(r)$

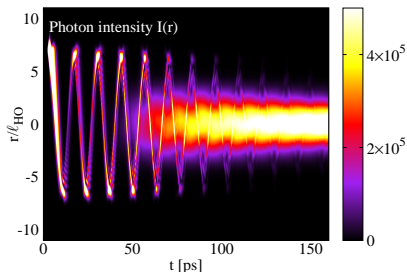


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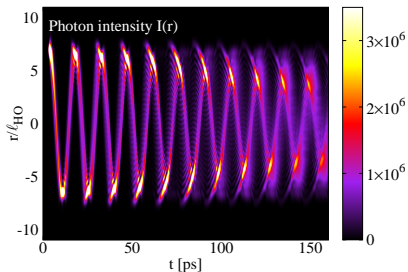
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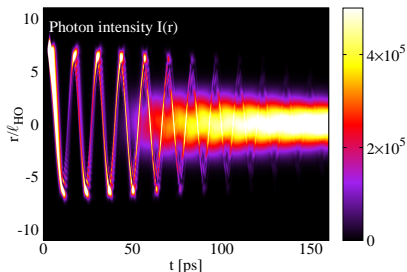
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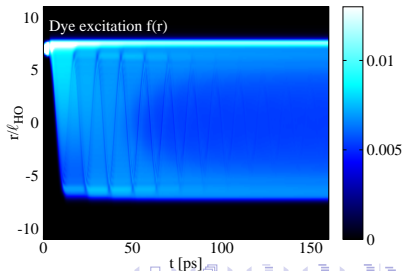
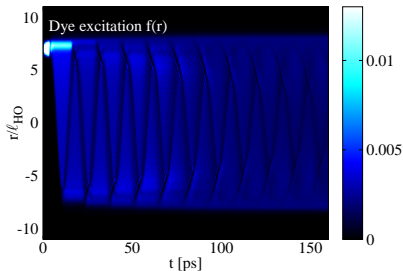
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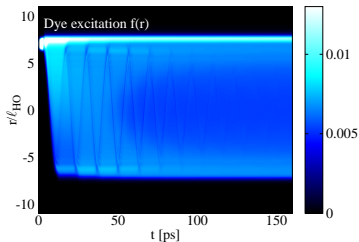
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# Thermalisation at late times

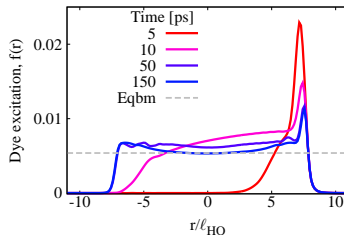
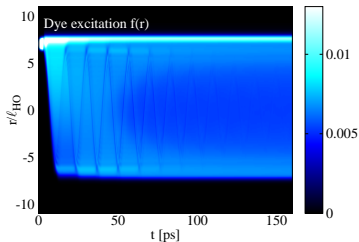
- Reabsorption “fills-in” excited molecules

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



# Thermalisation at late times

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- Reach thermal equilibrium,  $f = [e^{-\beta\delta_0} + 1]^{-1}$



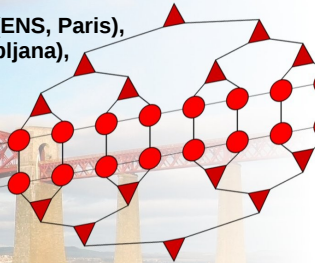
# Classical and Quantum matrix product states:

Exploring the structure of non-equilibrium states

10th-11th September 2015, Higgs Centre for Theoretical Physics

## Speakers:

M. Banuls (MPQ, Garching), X. Chen (Caltech), B. Derrida (ENS, Paris),  
K. Mallick (Saclay), J. Moore (UC Berkeley), T. Prosen (Ljubljana),  
E. Ragoucy (Annecy), G. Schutz (Julich),  
F. Verstraete (Vienna),  
S. White (UC Irvine)



The aim of this workshop is to bring together those working on quantum and classical concepts involving matrix product states, with a particular focus on non-equilibrium states.

To register, visit the Higgs centre website <https://higgs.ph.ed.ac.uk>

This meeting is made possible by funding from the EPSRC UK network on emergence and physics far from Equilibrium, the Higgs Centre for Theoretical Physics, and the EPSRC TOPNES programme grant.

Photograph by Andrew Bell. Licensed under CC BY-SA 3.0 via Wikimedia Commons

**EPSRC**

Engineering and Physical Sciences  
Research Council



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



# CONDENSATES OF LIGHT

13TH-15TH JANUARY 2016, CHICHELEY HALL, BUCKINGHAMSHIRE, UK

The scope of the workshop covers all physical realisations of condensates of light, including:

- Bose-Einstein condensation of light in dye-filled microcavities
- Condensates of semiconductor exciton-polaritons
- Organic polariton condensation
- Classical condensation phenomena in optics
- Superfluid light

## Keynote speakers:

- Jason Fleischer (Princeton, USA)
- Elisabeth Giacobino (LKB, Paris, France)
- Stephane Kena-Cohen (Polytechnique, Montreal, Canada)
- Henk Stoof (Utrecht, Netherlands)
- Martin Weitz (Bonn, Germany)

## Invited speakers:

- Alberto Amo (Marcoussis, France)
- Iacopo Carusotto (Trento, Italy)
- Natalia Berloff (Cambridge, UK/Moscow)
- Baruch Fischer (Technion, Haifa, Israel)
- Jonathan Keeling (St Andrews, UK)
- Pavlos Lagoudakis (Southampton, UK)
- Rainer Mahrt (IBM Zurich, Switzerland)
- Gian-Luca Oppo (Strathclyde, UK)
- Antonio Picozzi (Bourgogne, France)
- Daniele Sanvitto (Lecce, Italy)
- David Snoke (Pittsburgh, USA)
- Marzena Szymanska (UCL, UK)
- Jacob Taylor (Maryland, USA)

## Important Dates:

- 31st October 2015: abstract submission deadline
- 30th November 2015: registration deadline

## Organising committee:

- Robert Nyman (Imperial College, London)
- Peter Kirton (St Andrews)
- Jonathan Keeling (St Andrews)
- Martin Weitz (Bonn)

<http://condensates-of-light.org>

# Acknowledgements

GROUP:



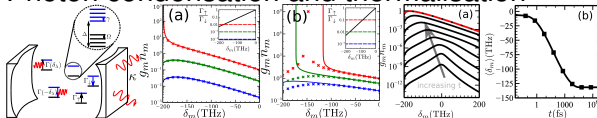
FUNDING:



**The Leverhulme Trust**

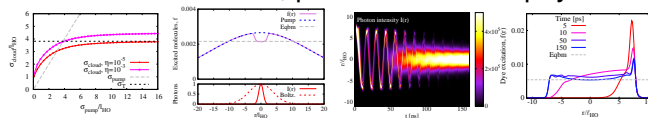
# Summary

- Photon condensation and thermalisation



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

- Photon condensation, pattern formation physics



[JK & Kirton, arXiv:1506:00280]



# Extra Slides

- 4 Microscopic calculation of  $\Gamma(\delta)$
- 5 Threshold vs temperature
- 6 Threshold vs spot size
- 7 Beyond semiclassics
- 8 More oscillations
- 9 Toy problem – two bosonic modes

# Microscopic model – calculating $\Gamma(\delta)$

How to calculate  $\Gamma(\delta)$

- Polaron transform (exact)

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

$$D_\alpha = \exp \left[ 2\sqrt{S} (b_\alpha^\dagger - b_\alpha) \right]$$

- Correlation function:

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

- Exponential of bosonic correlations  $\langle D_\alpha^\dagger(t) D_\alpha(0) \rangle$

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$$h_\alpha = \frac{\epsilon}{2} \sigma_\alpha^z + g (\psi_m \sigma_\alpha^+ D_\alpha + \text{H.c.}) + \Omega b_\alpha^\dagger b_\alpha,$$
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## Threshold condition

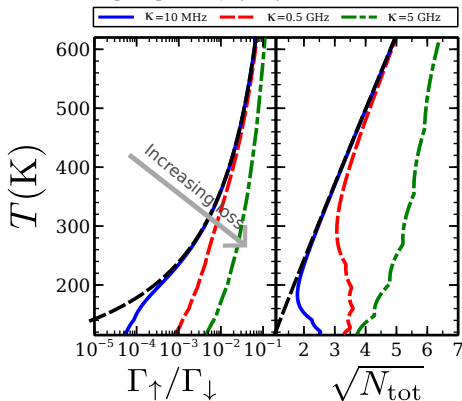
Use:  $\max[n_m] = 1/(\beta\epsilon) \rightarrow k_B T_c = \sqrt{6/\pi^2\epsilon}\sqrt{N}$ .

- Pump rate (Laser)
- Critical density (condensate)

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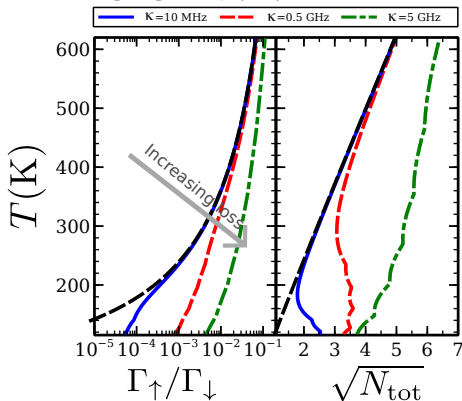
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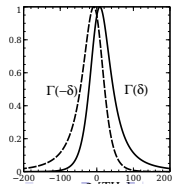


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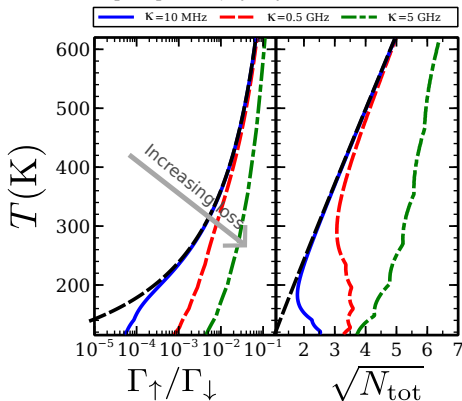
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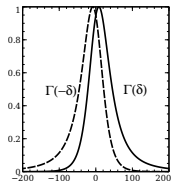
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- Lasing threshold, dependence on spot size.

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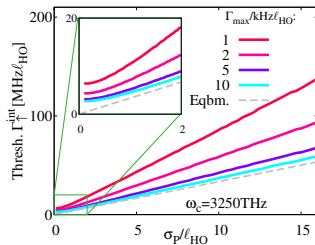
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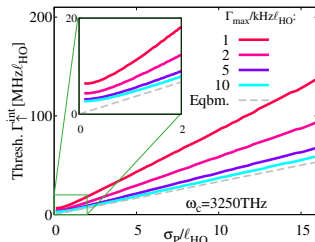


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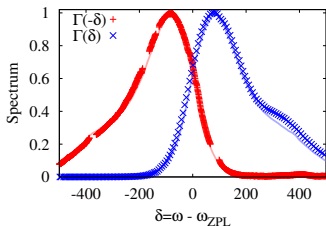
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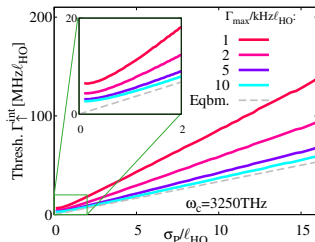




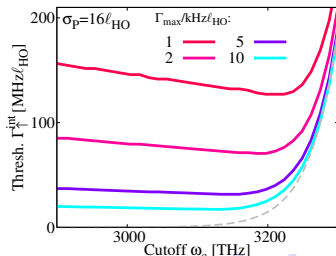
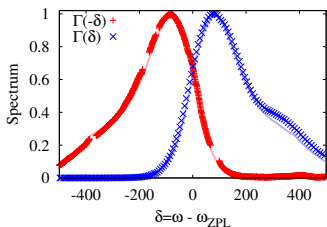
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# Quantum model, linewidth

Full Master equation:

$$\dot{\rho} = -i[H_0, \rho] - \frac{\kappa}{2} \mathcal{L}[\psi] - \sum_{\alpha} \left[ \frac{\Gamma_{\uparrow}}{2} \mathcal{L}[\sigma_{\alpha}^{+}] + \frac{\Gamma_{\downarrow}}{2} \mathcal{L}[\sigma_{\alpha}^{-}] \right] \\ - \sum_{\alpha} \left[ \frac{\Gamma(\delta = \omega - \epsilon)}{2} \mathcal{L}[\sigma_{\alpha}^{+} \psi] + \frac{\Gamma(-\delta = \epsilon - \omega)}{2} \mathcal{L}[\sigma_{\alpha}^{-} \psi^{\dagger}] \right]$$

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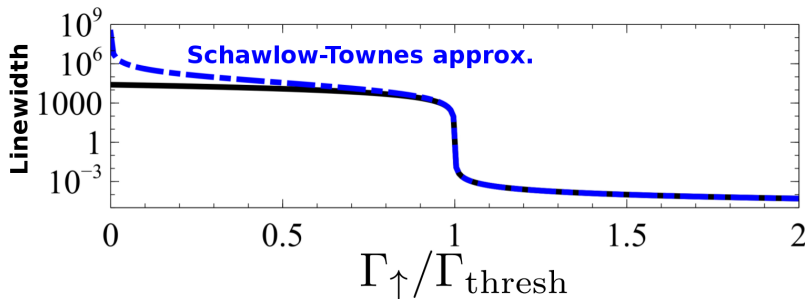
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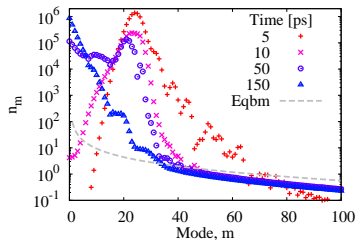
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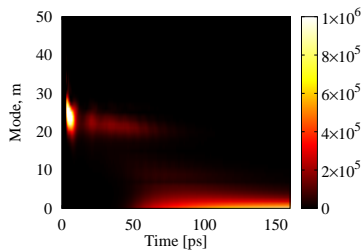
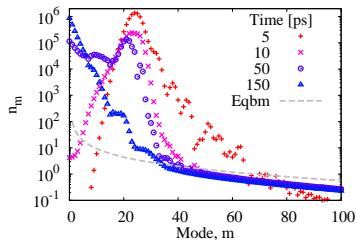
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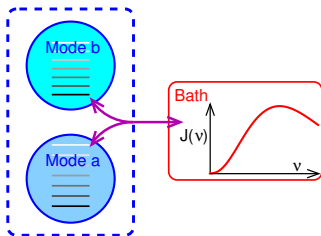
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# Toy problem: two bosonic modes

- 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

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

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

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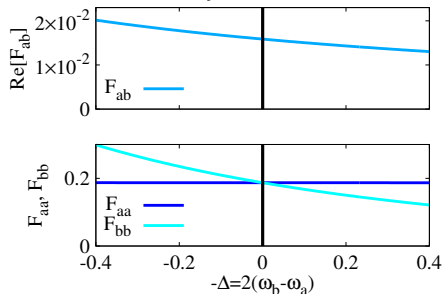
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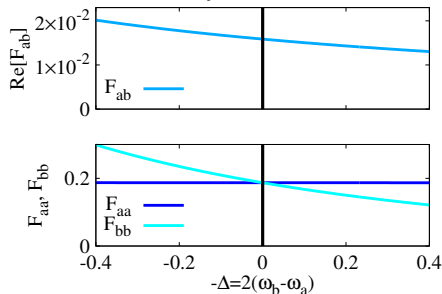


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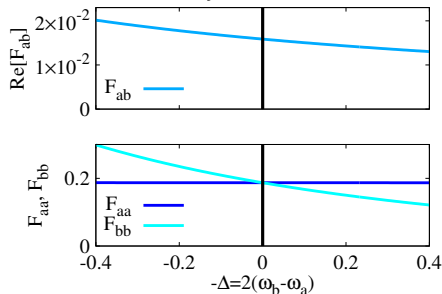
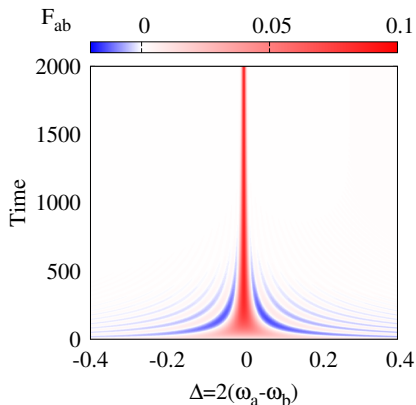
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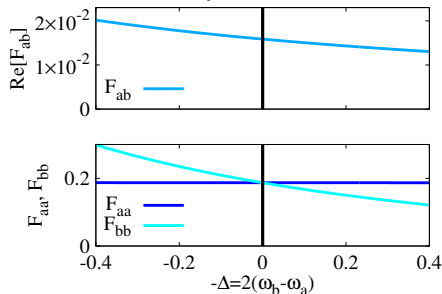
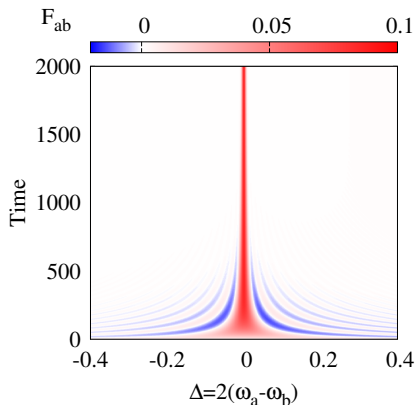


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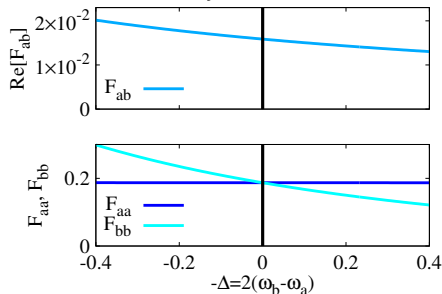
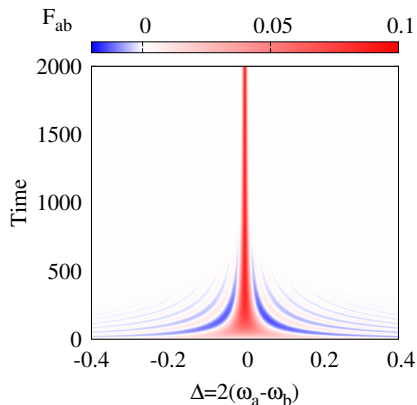


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Unsecularised Bloch-Redfield theory:

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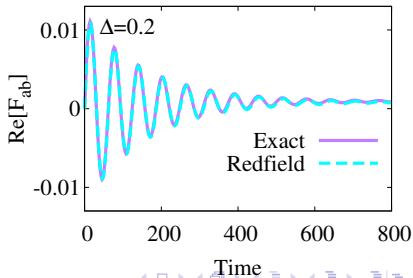
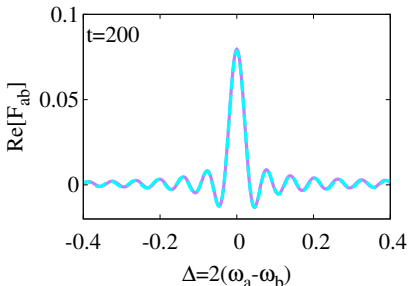
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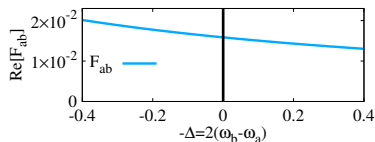
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- Secularisation (in eigenbasis of  $\hat{H}$ ):  $L_{ij}^{\uparrow,\downarrow} \rightarrow L_{ij}^{\uparrow,\downarrow} \delta_{ij}$



- Leads to  $F_{ab}(t \rightarrow \infty) = 0$ . Exact.

- Secularisation often invoked to cure negative eigenvalues of  $L_{ij}^{\uparrow,\downarrow}$ .

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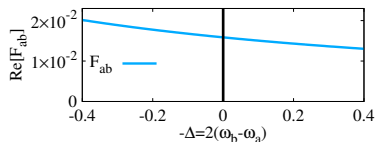
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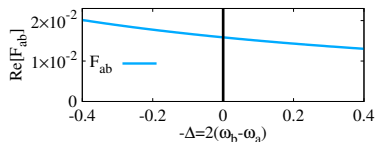
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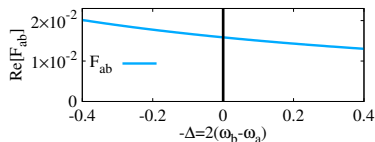
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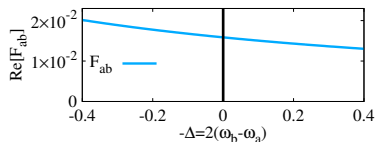
- Unstable (negative only if  $dJ(\nu)/d\nu \gg 1$   
→ Markov breakdown)



# Toy problem: Secularisation

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

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  - Non-positivity of density matrix,
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• Check stability: consider  $f = (F_{ab}, F_{ba}, \Re[F_{ab}], \Im[F_{ab}])$

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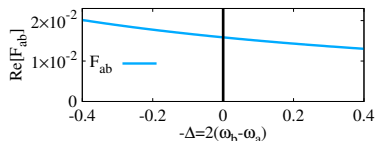
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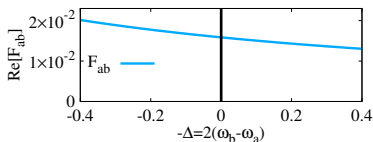
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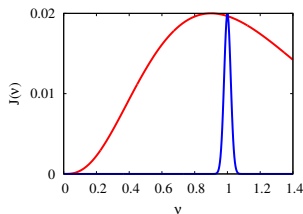
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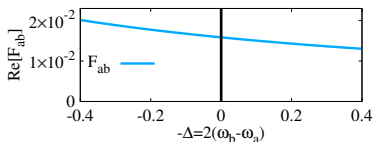
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