Spatial dynamics, thermalization and breakdown of thermalization in photon condenstaes

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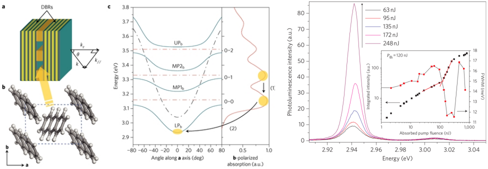
FOUNDED 1413

SUPA

Condensates of Light, January 2016

Motivation: polariton condensates

- Anthracene Polariton Lasing
  $T \sim 300$K



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

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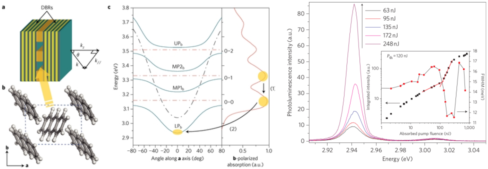

Q1. Vibrational replicas?

Q2. Relevance of disorder?

Q3. Lasing vs condensation?

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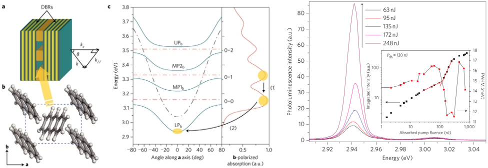

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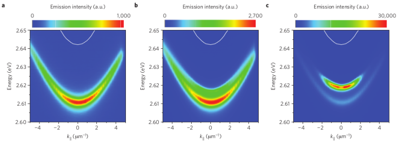

[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

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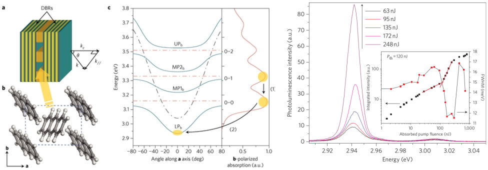

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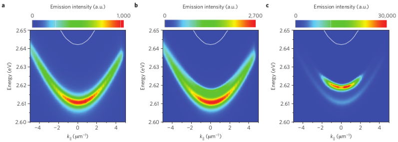

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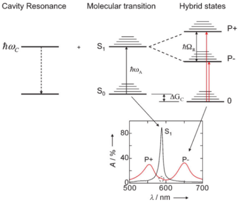
Q1. Frenkel to Wannier crossover?

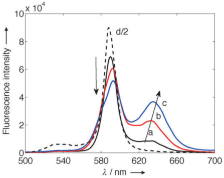
Q2. Optimal vibrational properties?

Q3. Nonlinearities?

Motivation: vacuum-state strong coupling

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





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

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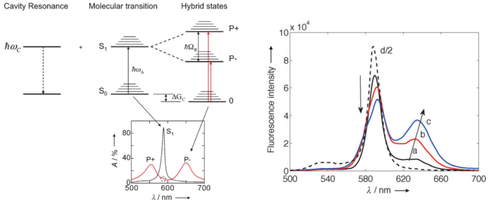

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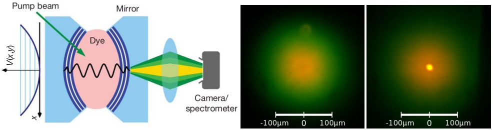
Q1. Can **ultra-strong** coupling to light change:

- charge distribution?
- vibrational configuration?
- molecular orientation?
- crystal structure?

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

Motivation: photon condensates

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



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

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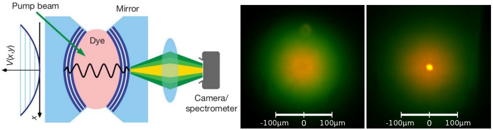

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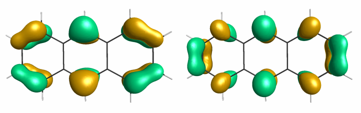
Q1. Relation to dye laser?

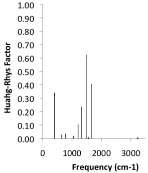
Q2. Relation to polaritons?

Q3. Thermalisation breakdown?

Toy models

1. Full molecular spectra electronic structure & Raman spectrum





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

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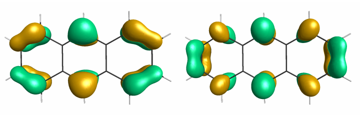

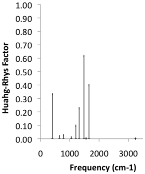

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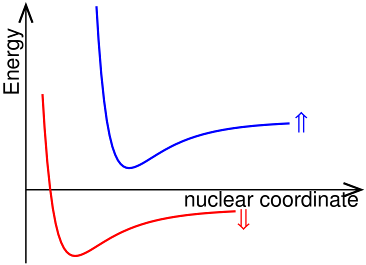
   - Two-level system, HOMO/LUMO
   - Single DoF PES



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

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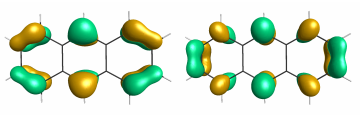

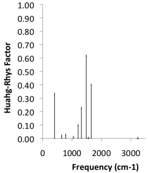

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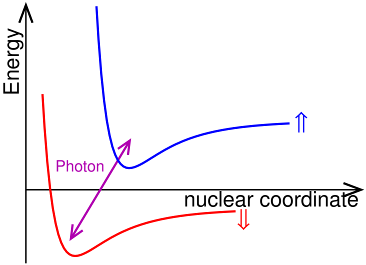

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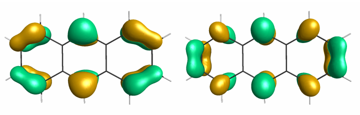

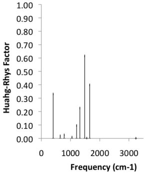

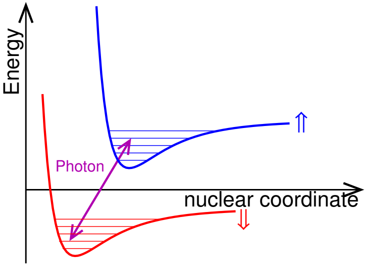

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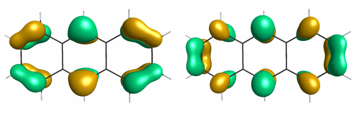

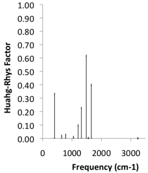

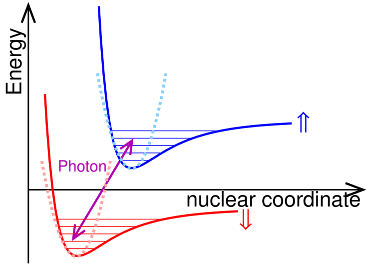

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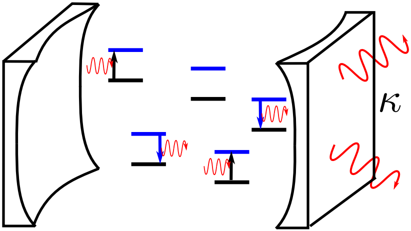
Dicke Holstein Model

- Dicke model: 2LS $\leftrightarrow$ photons
- Molecular vibrational mode
  - Phonon frequency Ω
  - Huang-Rhys parameter S — coupling strength



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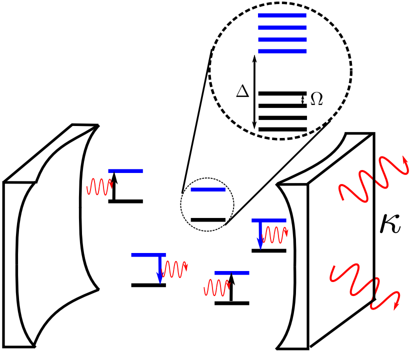

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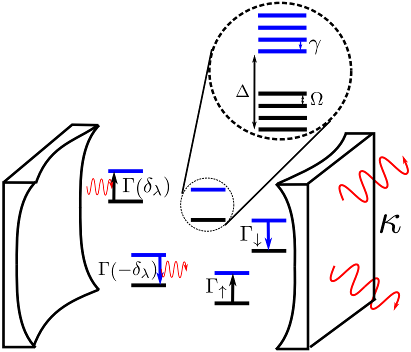
Modelling photon BEC

1. Modelling photon BEC
   - Uniform pumping results
2. Modelling steady-state spatial profile
   - Spatial profile vs spot size
   - Threshold vs spot size
3. Modelling spatial oscillations
   - Toy problem; validating model
   - Oscillation results

Photon: Microscopic Model

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

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



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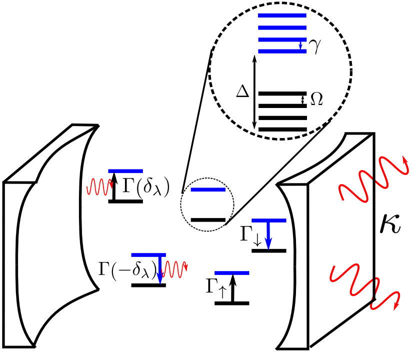

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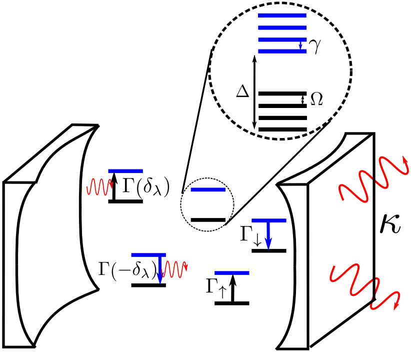

Microscopic model – all orders in S

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

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

- Master equation

$$\dot{\rho} = -i[H_0, \rho] + \sum_m \frac{\kappa}{2}\mathcal{L}[\psi_m] + \sum_\alpha \left[\frac{\Gamma_\uparrow}{2}\mathcal{L}[\sigma_\alpha^+] + \frac{\Gamma_\downarrow}{2}\mathcal{L}[\sigma_\alpha^-]\right]$$
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- Correlation function:

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

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

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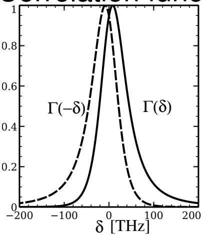

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Steady state populations and equilibrium

- Rate equation:

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Steady state populations vs loss

- Steady state distribution:

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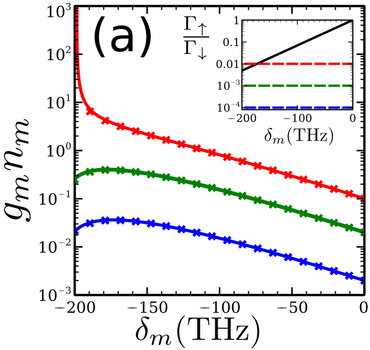

Low loss: Thermal

[Kirton & JK PRL ’13]

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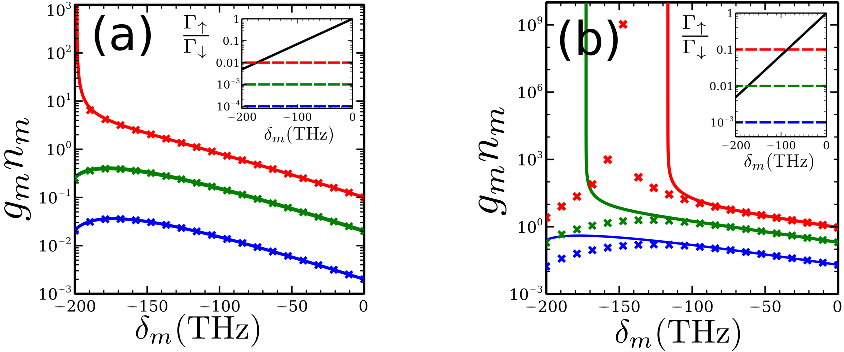

Low loss: Thermal

High loss → Laser

[Kirton & JK PRL '13]

Chemical potential?

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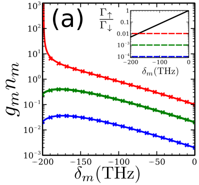

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[Kirton & JK, PRA '15]

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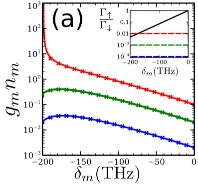

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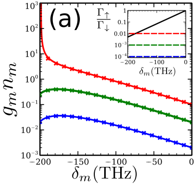

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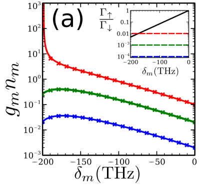

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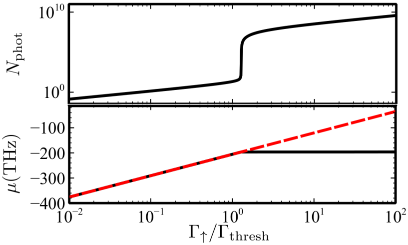

[Kirton & JK, PRA '15]

Modelling steady-state spatial profile

1. Modelling photon BEC
   - Uniform pumping results

2. Modelling steady-state spatial profile
   - Spatial profile vs spot size
   - Threshold vs spot size

3. Modelling spatial oscillations
   - Toy problem; validating model
   - Oscillation results

Spatially varying pump intensity

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

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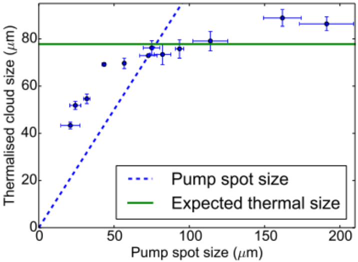

Modelling spatial profile.

- Varying excited density – differential coupling to modes

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

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

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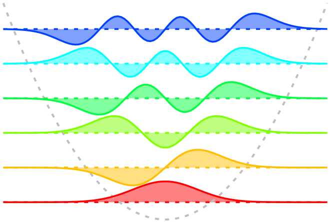
  $I(\mathbf{r}) = \sum_m n_m |\psi_m(\mathbf{r})|^2$



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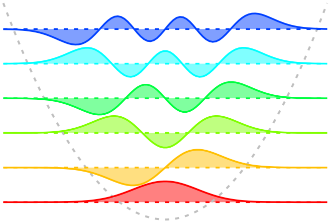

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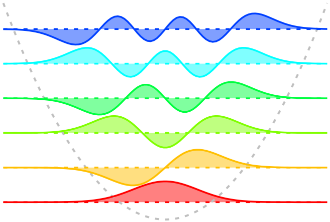
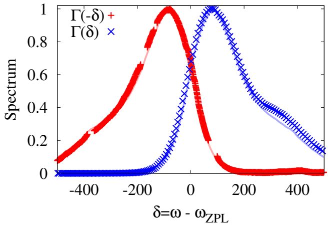


- Use exact R6G spectrum



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Spatially varying pump: below threshold

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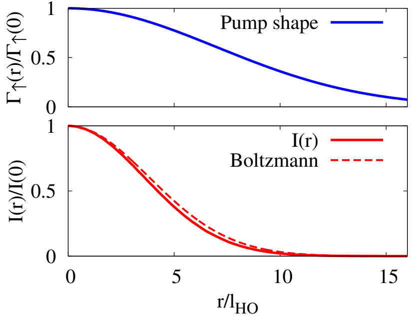

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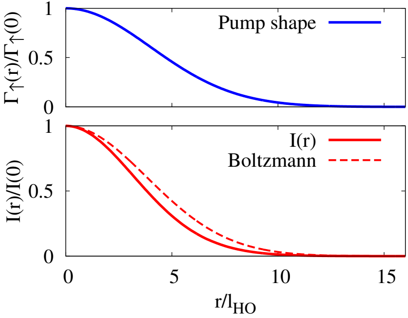

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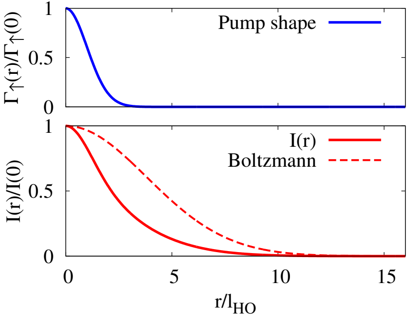

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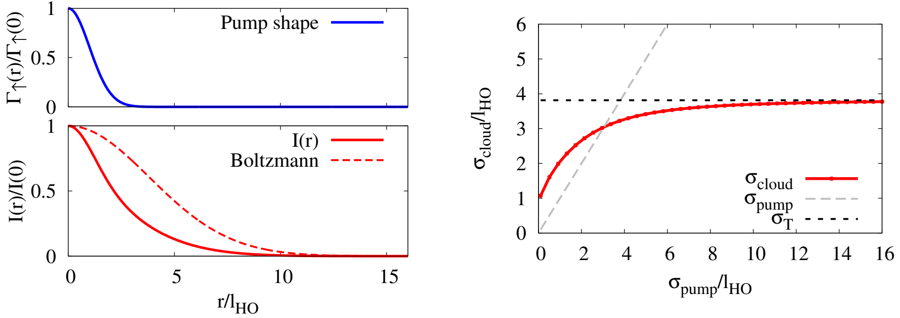
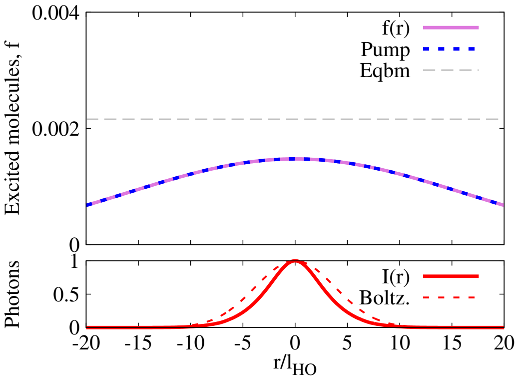


Near threshold behaviour



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

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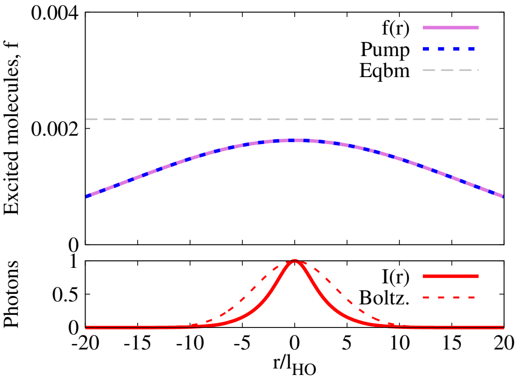

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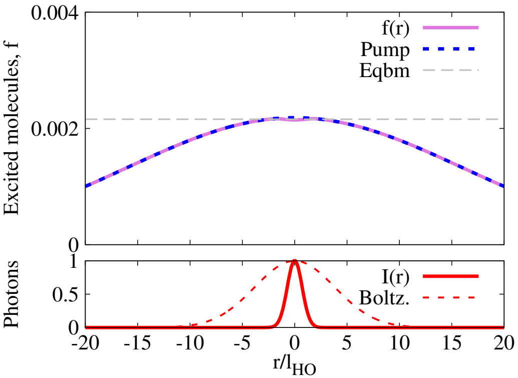

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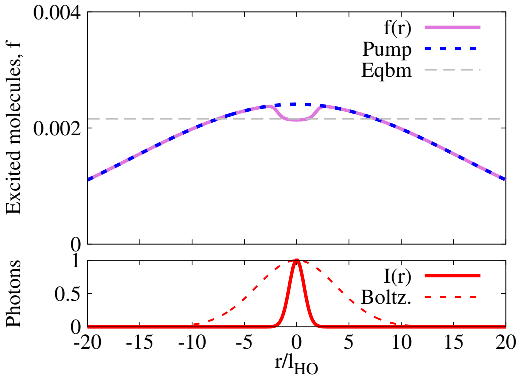

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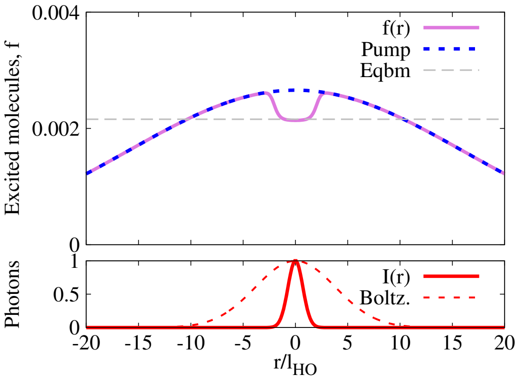

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- Lasing threshold, dependence on spot size.

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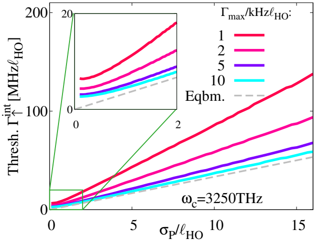

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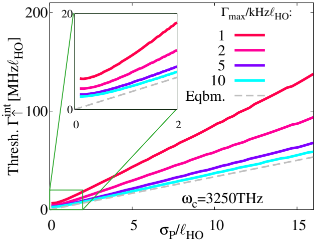

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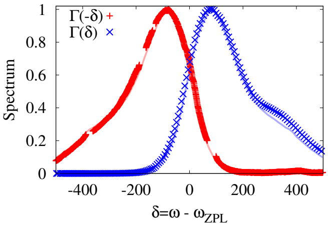

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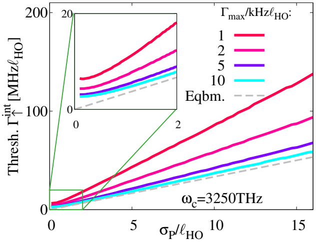

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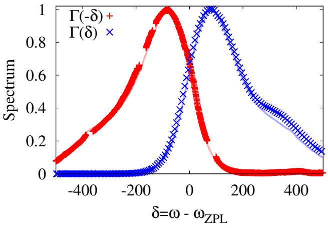
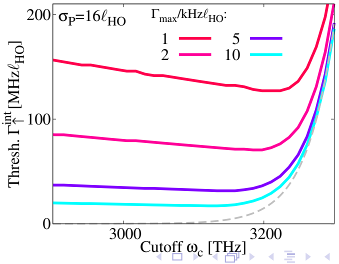
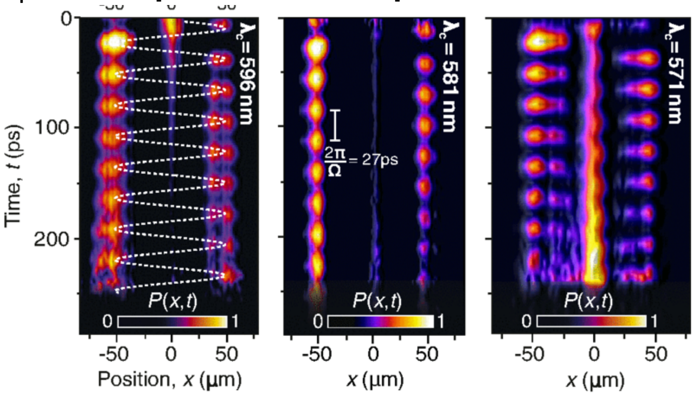




Threshold vs spot size

Off centre pumping; oscillations

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



- Oscillations in space – beating of normal modes
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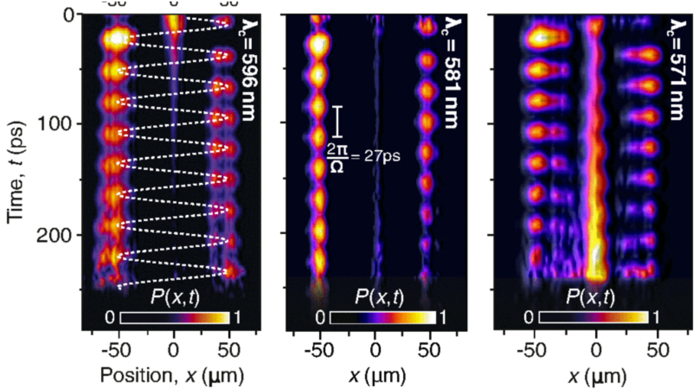

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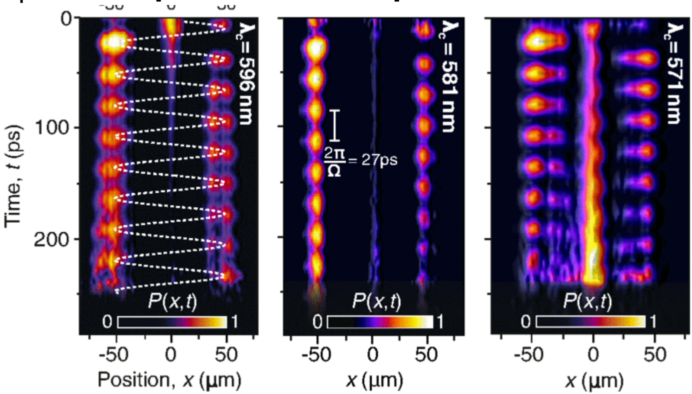

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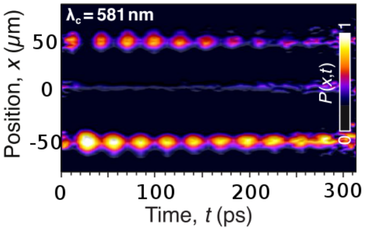
Limit of rate equations



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

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- Need $I(x) = \sum_{m,m'} n_{m,m'} \psi_m(x)\psi_{m'}(x)$
- Thermalisation from $\Gamma(\pm\delta)$

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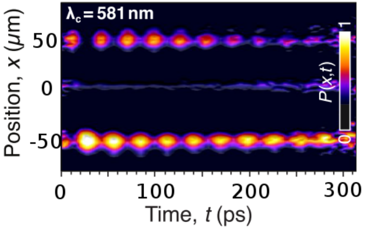

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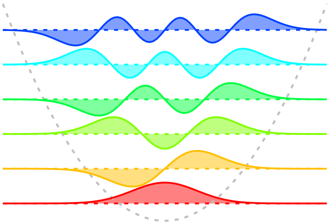
Emission into Gauss-Hermite mode m:

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



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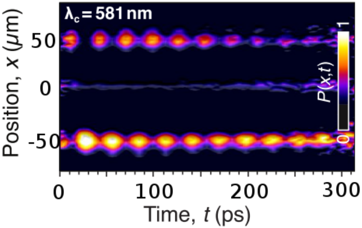

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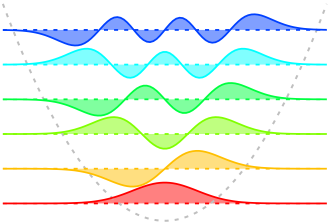

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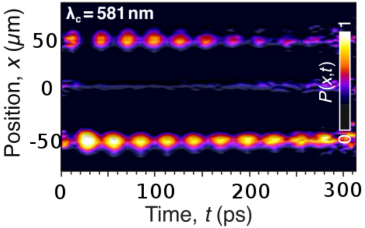

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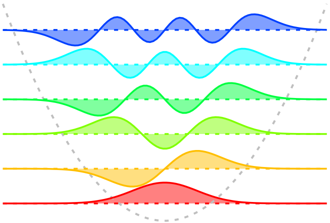
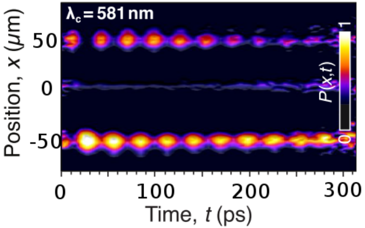


Emission must create coherence between non-degenerate modes.

Limit of rate equations



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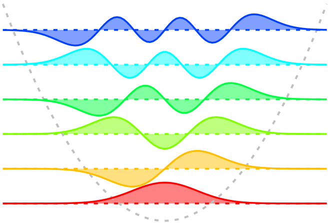
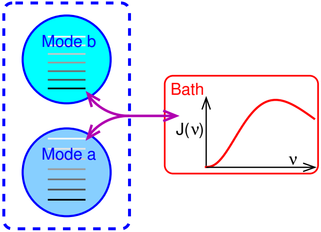


- Oscillations: beating of modes.
- Need $I(x) = \sum_{m,m'} n_{m,m'} \psi_m(x) \psi_{m'}(x)$
- Thermalisation from $\Gamma(\pm\delta)$

Emission must create coherence between non-degenerate modes.

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

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

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- 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
  - (individual always wrong)
- $F_{ab} \sim F_{aa}, F_{bb}$ only at $\Delta = 0$

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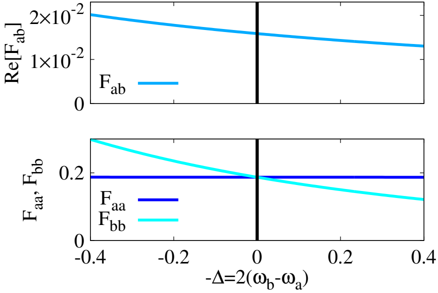

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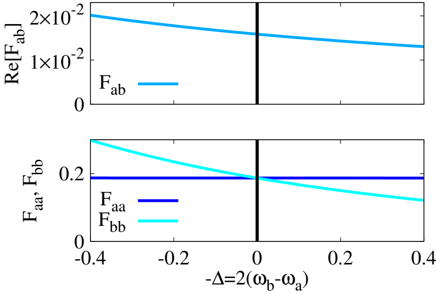

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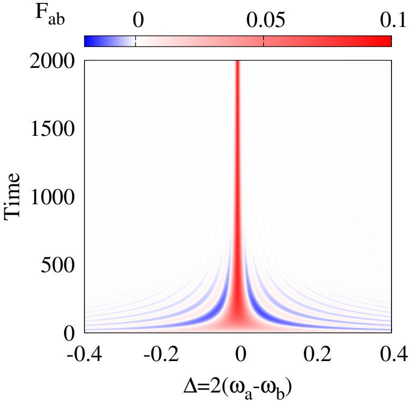

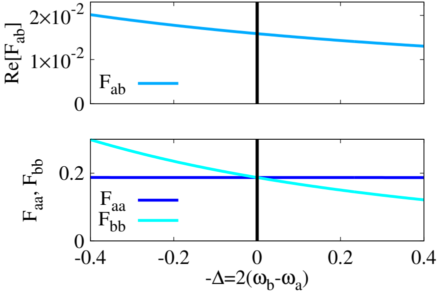

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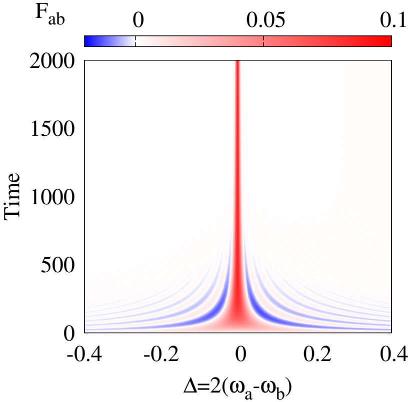

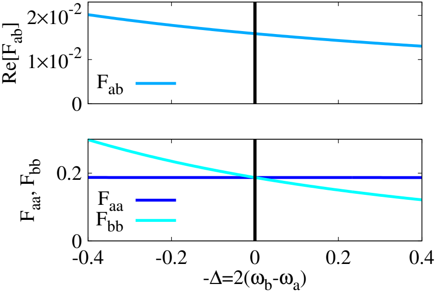

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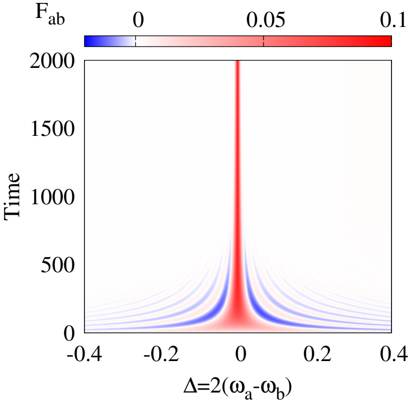

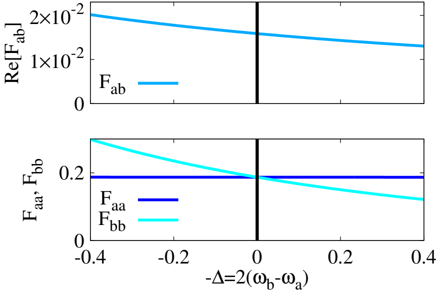

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Toy problem: Redfield theory

Unsecularised Redfield theory:

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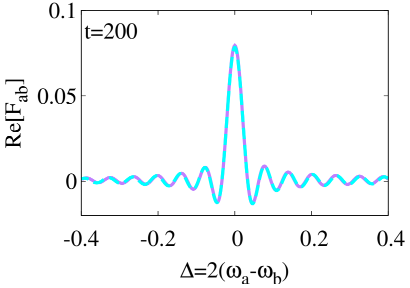

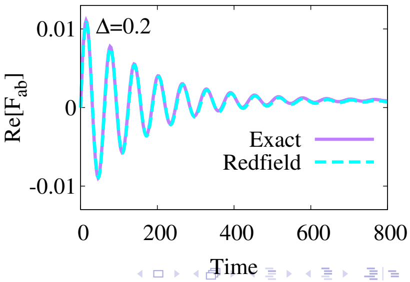

Toy problem: Secularisation

- Secularisation (in eigenbasis of $\hat{H}$): $L_{ij}^{\uparrow,\downarrow} \to L_{ii}^{\uparrow,\downarrow}\delta_{ij} \to F_{ab} = 0$
- Secularisation often invoked to cure negative eigenvalues of $L_{ij}^{\uparrow,\downarrow}$.
- Check stability: consider $f = (F_{aa}, F_{bb}, \Re[F_{ab}], \Im[F_{ab}])$

$$\partial_t \mathbf{f} = -\mathbf{M}\mathbf{f} + \mathbf{f}_0$$

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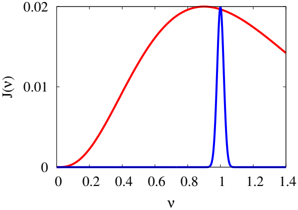

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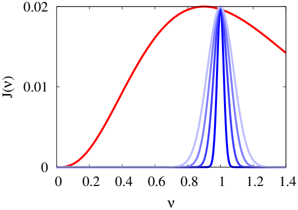

Beyond Redfield: Schrödinger picture Bloch Redfield

- Is BR the best (time-local) theory we can find?
- Hints it is not:
  - Eigenvalues of $\mathbf{M}$ vs exact sol'n near $\Delta = 0$.
  - Sum rule [Salmilehto *et al.* PRA '12; Hell *et al.* PRB '14]:

    "For $\hat{X}$ s.t. $[\hat{X}, \hat{H}_{\text{system-bath}}] = 0$, then $\partial_t \langle \hat{X} \rangle$ should match closed system."
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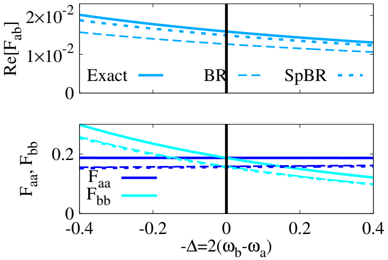

Modelling spatial oscillations

1. Modelling photon BEC
   - Uniform pumping results
2. Modelling steady-state spatial profile
   - Spatial profile vs spot size
   - Threshold vs spot size
3. Modelling spatial oscillations
   - Toy problem; validating model
   - Oscillation results

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- Following toy model, use Redfield theory:

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

- $K(\delta)$ analytic continuation of $\Gamma(\delta)$.
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- $K(\delta)$ analytic continuation of $\Gamma(\delta)$.
- Not secular approximation
  - **Must** have emission into m, m' superposition
  - **Must** have $K = K(\delta_m)$ (Kennard-Stepanov)
- Semiclassical equations for $n_{m,m'} = \langle a_m^\dagger a_{m'} \rangle$ and $f(r)$.

Modelling

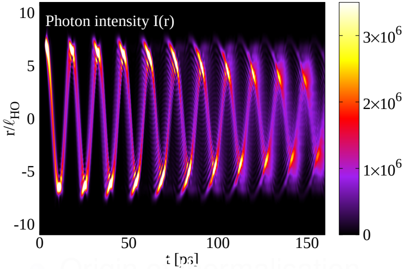
- Following toy model, use Redfield theory:

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

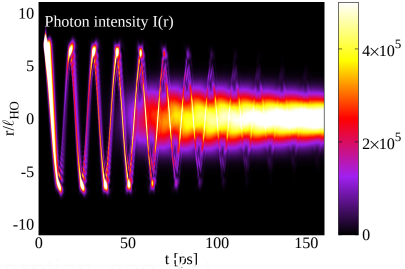
- $K(\delta)$ analytic continuation of $\Gamma(\delta)$.
- Not secular approximation
  - **Must** have emission into m, m' superposition
  - **Must** have $K = K(\delta_m)$ (Kennard-Stepanov)
- Semiclassical equations for $n_{m,m'} = \langle a_m^\dagger a_{m'} \rangle$ and $f(r)$.

Dynamics from model

Longer cavity

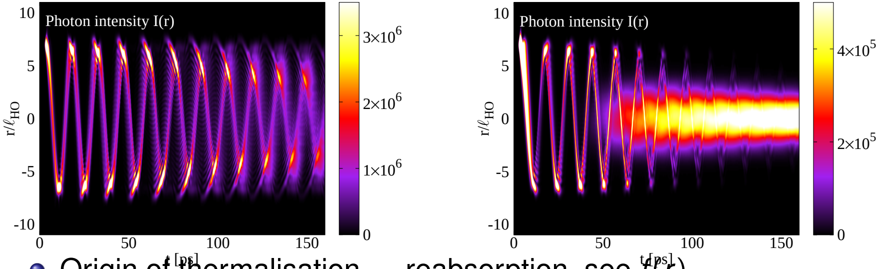


Shorter cavity



Dynamics from model

Longer cavity | Shorter cavity



- Origin of thermalisation — reabsorption, see $f(r)$

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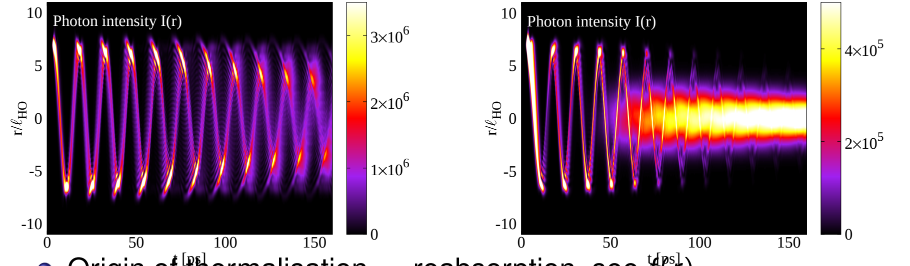

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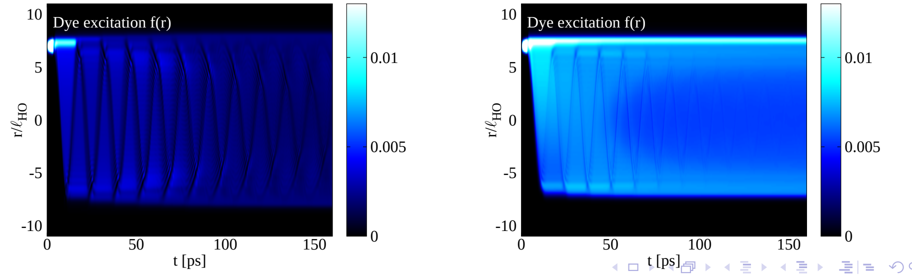
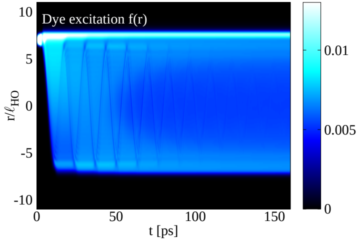


Thermalisation at late times

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



- Photon occupation thermalises later

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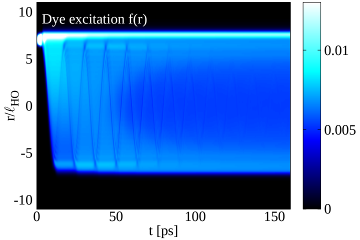

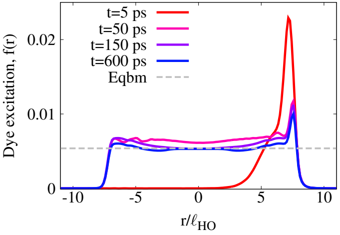

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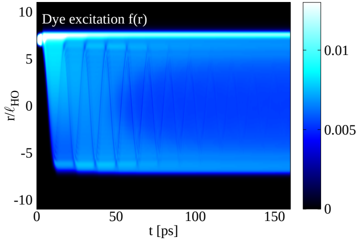

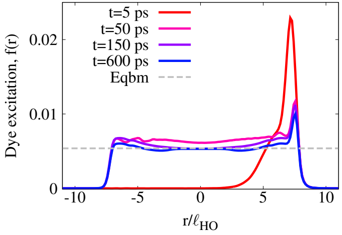

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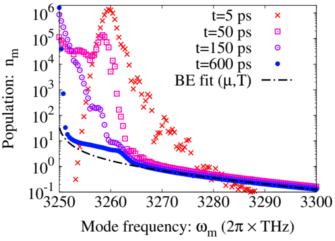
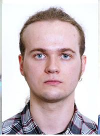


Acknowledgements

GROUP:




FUNDING:

EPSRC
Engineering and Physical Sciences
Research Council

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

Hybrid
Polaritonics

The Leverhulme Trust

I C S C E 8

Edinburgh, 25th–29th April, 2016.

Plenary speakers: Ataç İmamoğlu, Peter Zoller.

Invited speakers: Ehud Altman, Mete Atatüre, Natasha Berloff, Charles Bardyn, Jacqueline Bloch, Iacopo Carusotto, Cristiano Ciuti, Michele Devoret†, Thomas Ebbesen, Thiery Giamarchi, Jan Klärs, Dmitry Krizhanovskii, Xiaoqin (Elaine) Li, Peter Littlewood, Allan MacDonald, Francesca Marchetti, Keith Nelson, Pavlos Lagoudakis, Vivien Zapf. († To be confirmed)

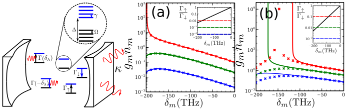
Early-bird registration & abstract deadline: 31st January 2016.

Final registration deadline: 31st March 2016.

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

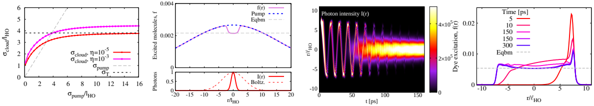
Summary

- Photon condensation and thermalisation



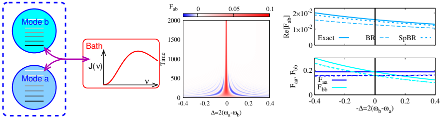
[Kirton & JK, PRL ’13, PRA ’15]

- Photon condensation, pattern formation physics



[JK & Kirton, PRA ’16]

- Modelling incoherent emission into non-degenerate modes

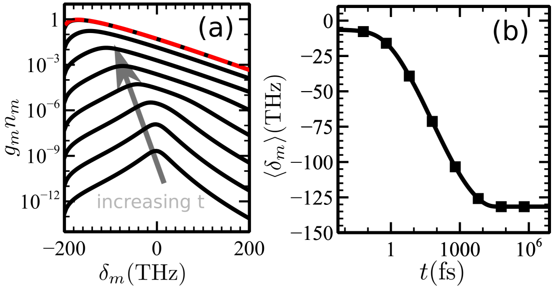


[Eastham, Kirton, Cammack, Lovett, JK `arXiv:1508.04744`]

Extra Slides

Time evolution

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



[Kirton & JK PRA '15]

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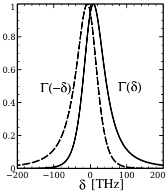

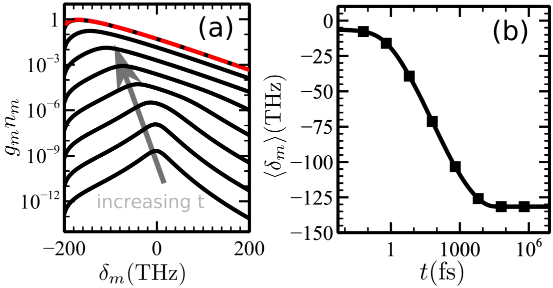

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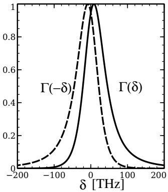

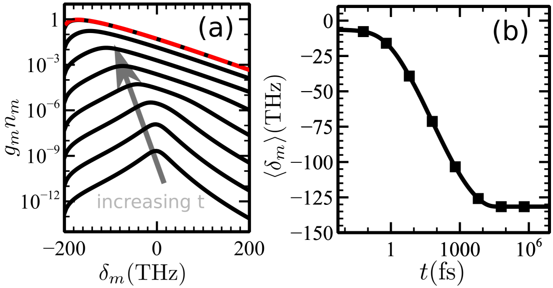

[Kirton & JK PRA '15]

Threshold condition

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

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

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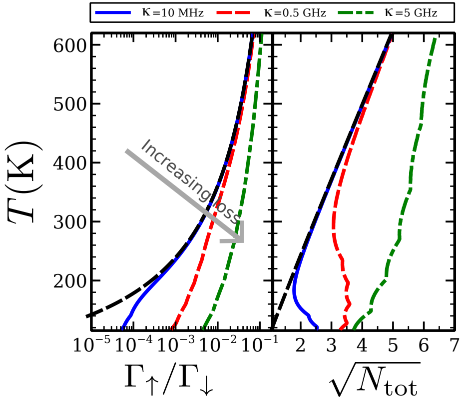

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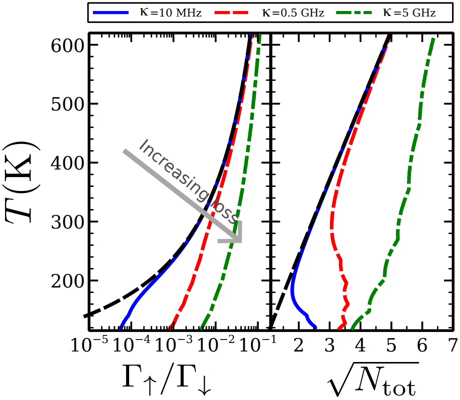

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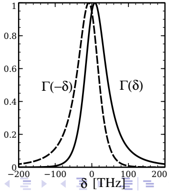

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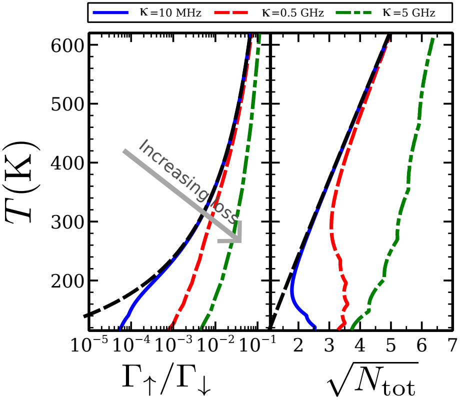

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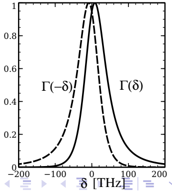

Quantum model, linewidth

Full Master equation:

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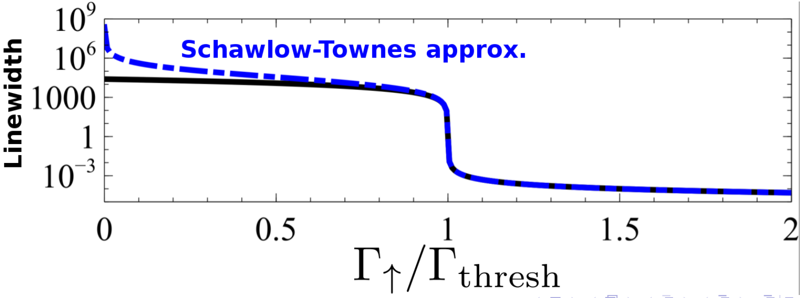

Beyond Redfield: Schrödinger picture Bloch Redfield

- Is BR the best (time-local) theory we can find?
- Hints it is not:
  - Eigenvalues of $\mathbf{M}$ vs exact sol'n near $\Delta = 0$.
  - Sum rule [Salmilehto *et al.* PRA '12; Hell *et al.* PRB '14]:

    "For $\hat{X}$ s.t. $[\hat{X}, \hat{H}_{\text{system-bath}}] = 0$, then $\partial_t \langle \hat{X} \rangle$ should match closed system."
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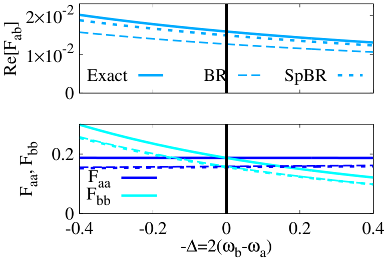

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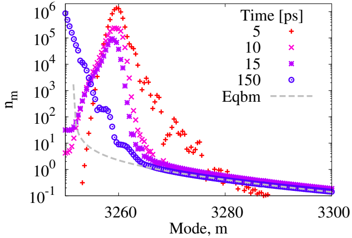

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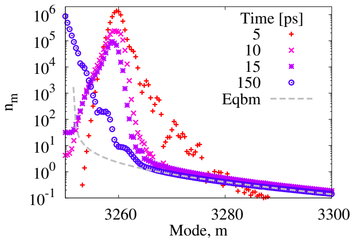
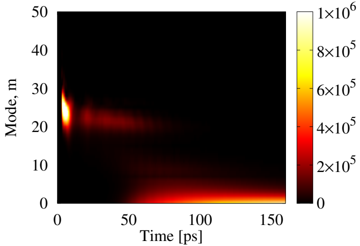
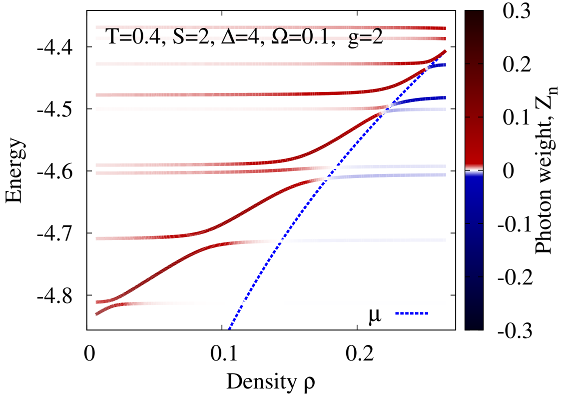




Polariton spectrum: photon weight



- What is nature of polariton mode?

$$G^R(t) = -i\langle\psi^\dagger(t)\psi(0)\rangle, \qquad G^R(\nu) = \sum_n \frac{Z_n}{\nu - \omega_n}$$

[Cwik *et al.* EPL '14]

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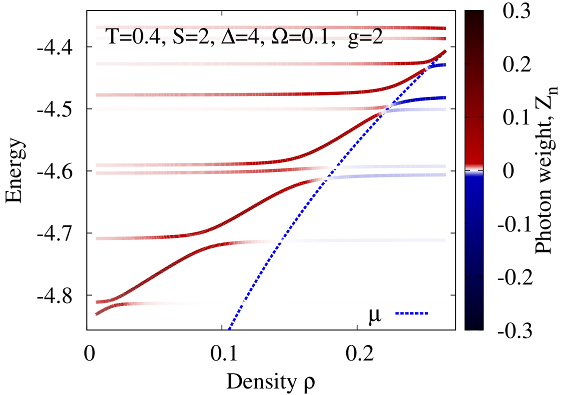

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