Photon and polariton condensation with organic molecules

Jonathan Keeling

SUPA

University of St Andrews
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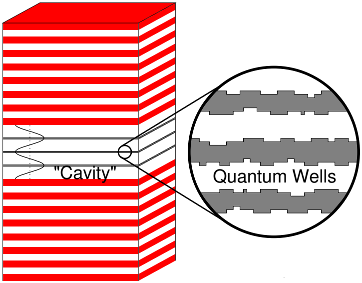
Microcavity polaritons



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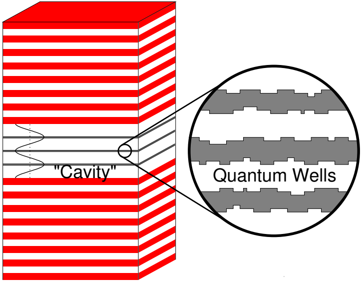
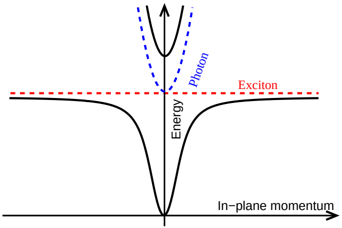


Cavity photons:

$$\omega_k = \sqrt{\omega_0^2 + c^2k^2}$$
$$\simeq \omega_0 + k^2/2m^*$$
$$m^* \sim 10^{-4} m_e$$



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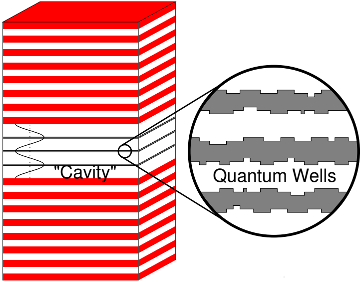

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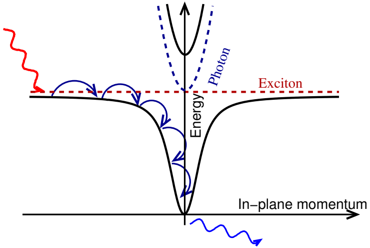

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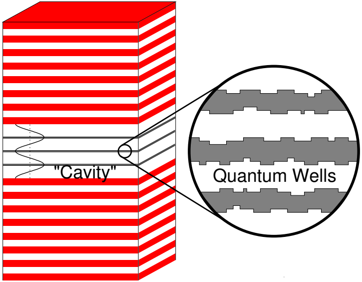

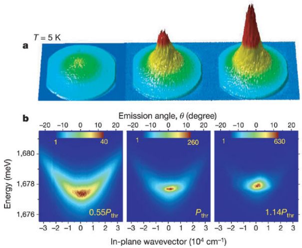

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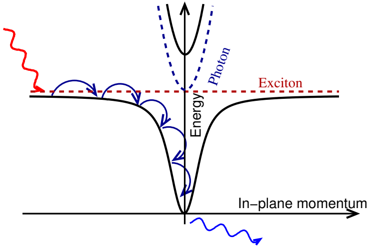
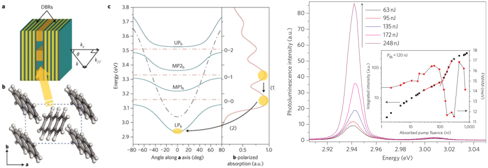


Motivation: polariton condensates

- Anthracene Polariton Lasing
  $T \sim 300$K



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

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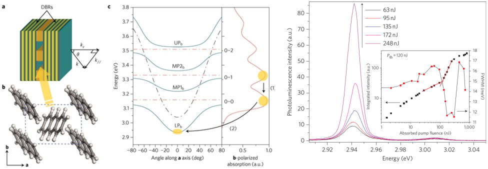

Q1. Vibrational replicas?

Q2. Relevance of disorder?

Q3. Lasing vs condensation?

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

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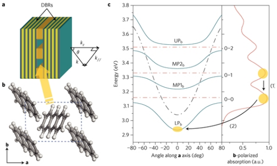

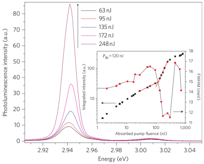

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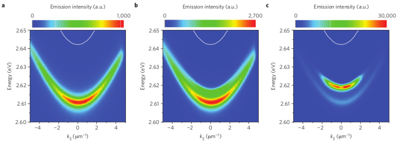

[Plumhoff *et al.* Nat. Materials ’14, Daskalakis *et al.* ibid ’14]

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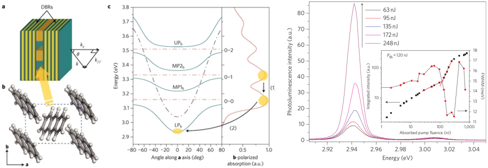

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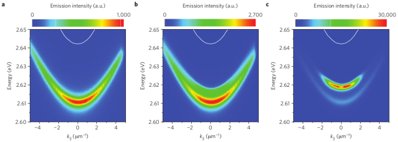


[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

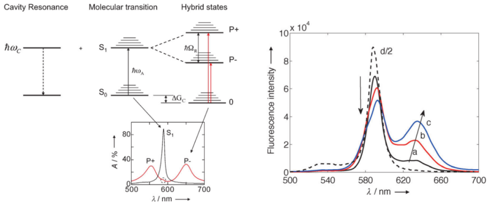
Q1. Frenkel to Wannier crossover?

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]

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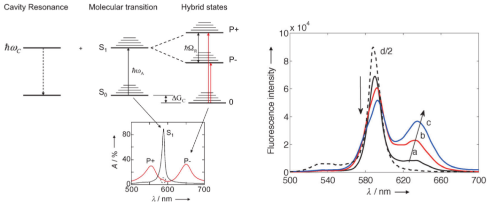


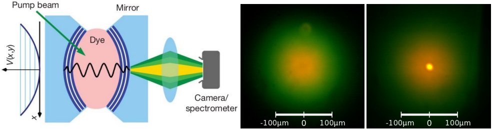
[Canaguier-Durand *et al.* Angew. Chem. '13]

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

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

Motivation: photon condensates

- Photon Condensate $T \sim 300$K



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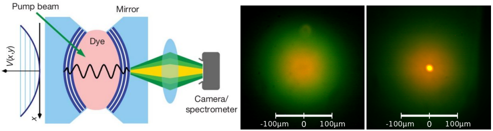


[Klaers *et al.* Nature, '10]

Q1. Relation to dye laser?

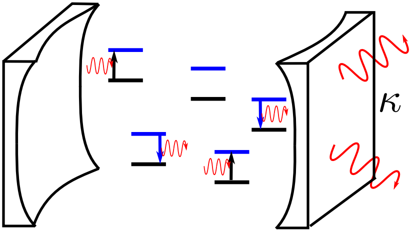
Q2. Relation to polaritons?

Q3. Thermalisation breakdown?

1. Modelling photon BEC
   - Steady state
2. Spatial profile
   - Steady state
   - Spatial oscillations
3. (Toward) strong coupling

Dicke Holstein Model

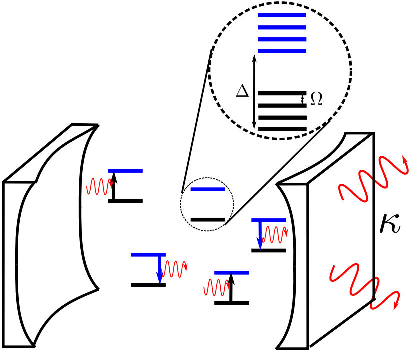
- Dicke model: 2LS $\leftrightarrow$ photons



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

Dicke Holstein Model

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



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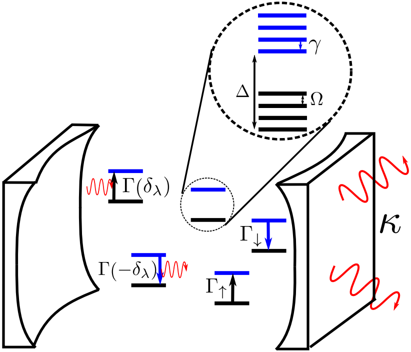
Photon: Microscopic Model

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

- **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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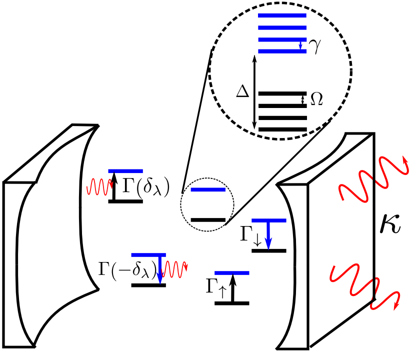

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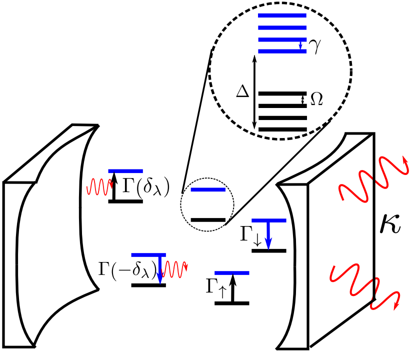

Microscopic model – all orders in S

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

$$h_\alpha = \frac{\epsilon}{2}\sigma^z_\alpha + g\left(\psi_m \sigma^+_\alpha D_\alpha + \text{H.c.}\right) + \Omega b^\dagger_\alpha b_\alpha, \qquad D_\alpha = e^{2\sqrt{S}(b^\dagger_\alpha - b_\alpha)}$$

- 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:

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

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

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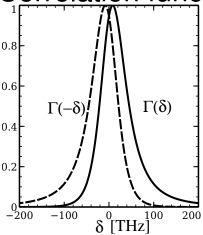

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Steady state populations and equilibrium

- Rate equation:

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- Bose-Einstein distribution without losses

[Kirton & JK PRL '13]

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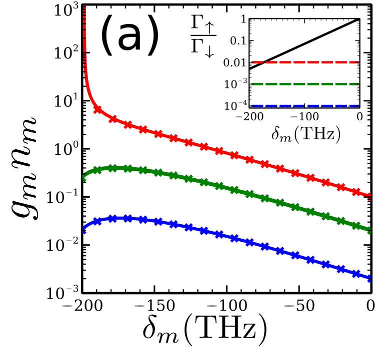

Low loss: Thermal

[Kirton & JK PRL ’13]

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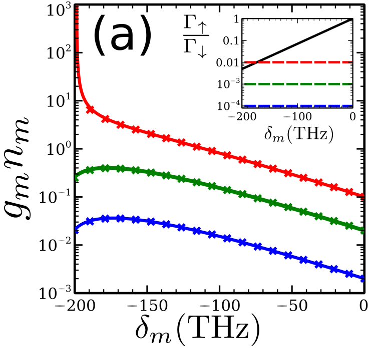

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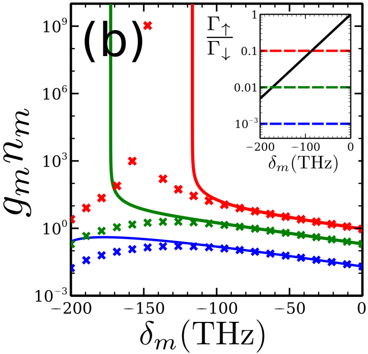
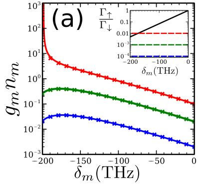


High loss → Laser

[Kirton & JK PRL '13]

Chemical potential?



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

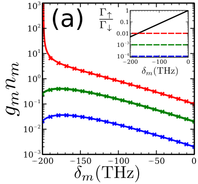
$$\frac{n_m}{n_m+1} = e^{-\beta\delta_m+\beta\mu}, \qquad e^{\beta\mu} \equiv \frac{N_\uparrow}{N_\downarrow} = \frac{\Gamma_\uparrow + \sum_m \Gamma(\delta_m)n_m}{\Gamma_\downarrow + \sum_m \Gamma(-\delta_m)(n_m+1)}$$

- Below threshold,

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

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

Chemical potential?



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

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

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

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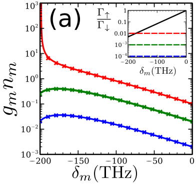

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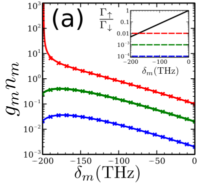

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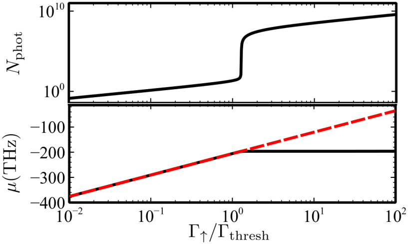

Spatial profile

1. Modelling photon BEC
   - Steady state
2. Spatial profile
   - Steady state
   - Spatial oscillations
3. (Toward) strong coupling

Spatially varying pump intensity

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

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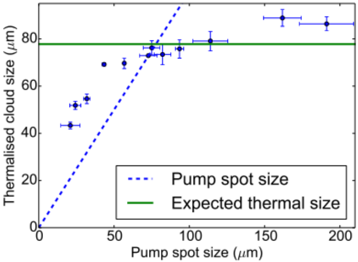


Modelling spatial profile.

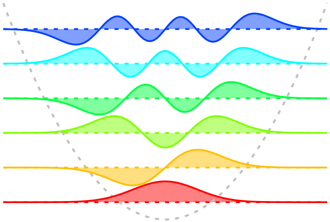
- Varying excited density – differential coupling to modes

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

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

Modelling spatial profile.

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



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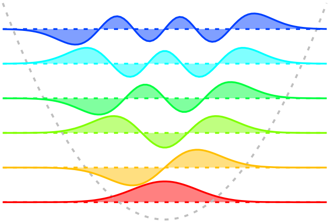

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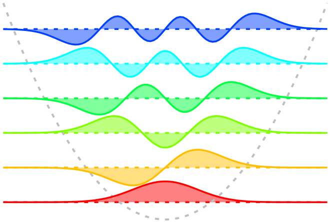
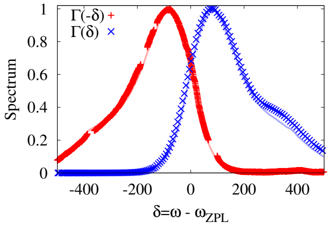


- Use exact R6G spectrum



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Spatially varying pump: below threshold

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

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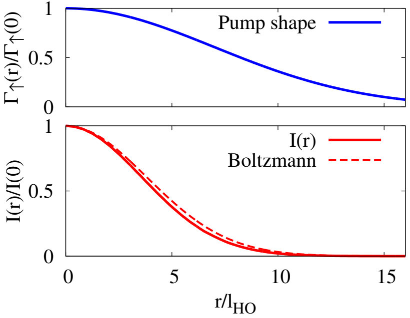

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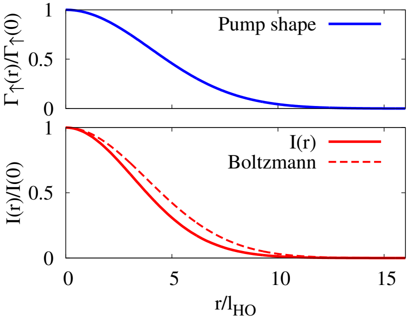

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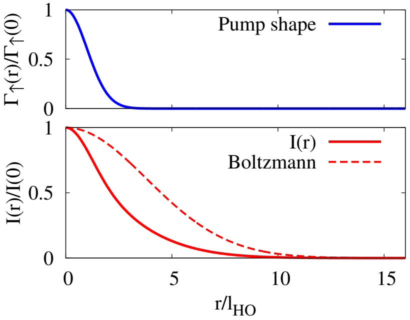

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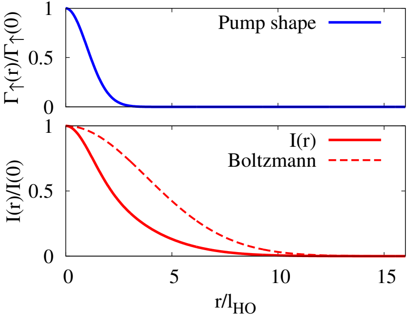
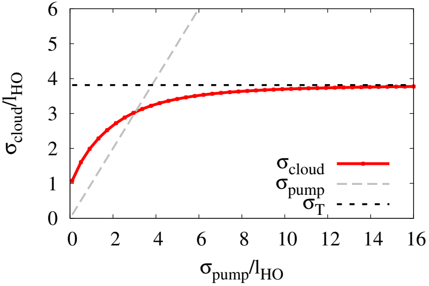
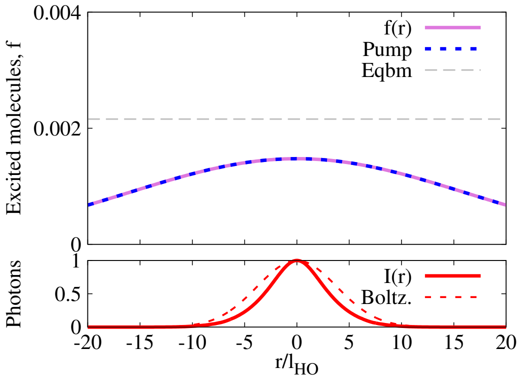




Near threshold behaviour



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

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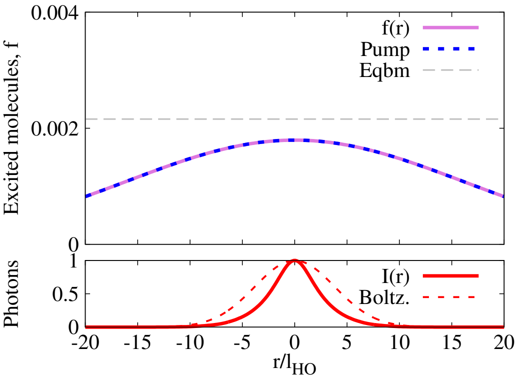

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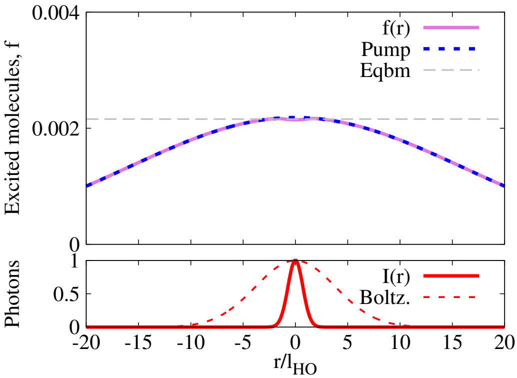

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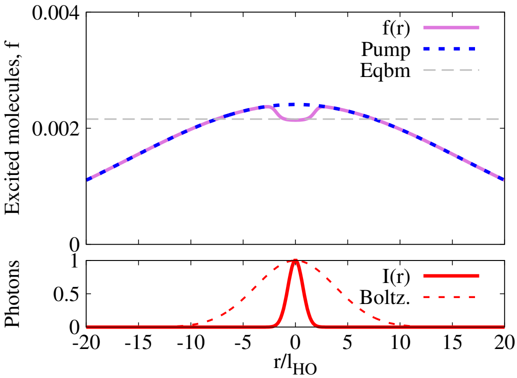

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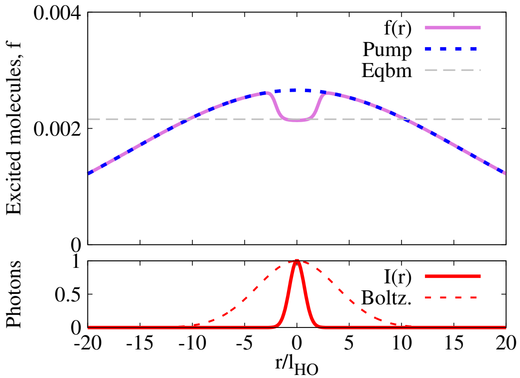

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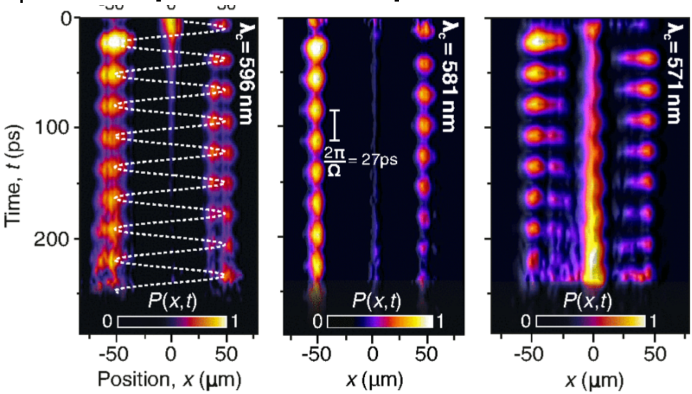
Off centre pumping; oscillations

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



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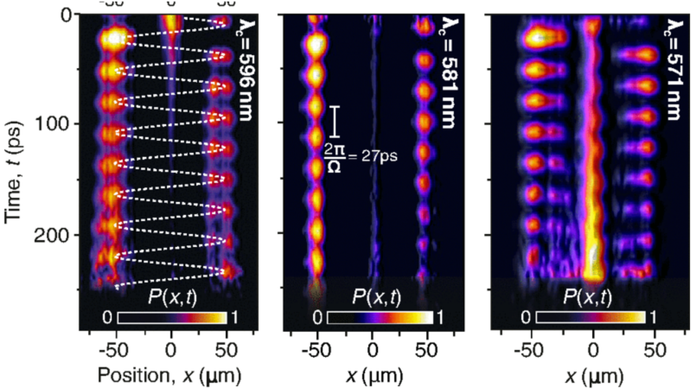

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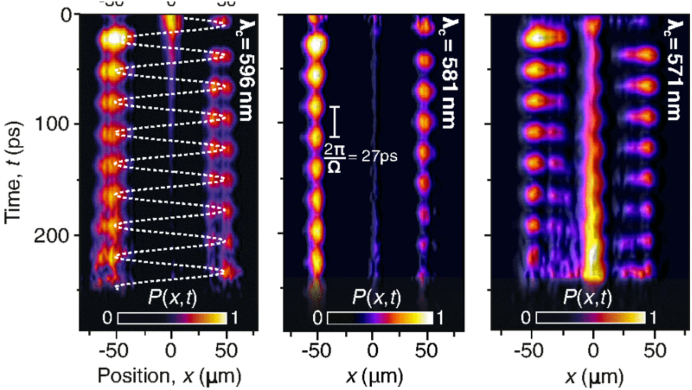

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Modelling

- Full master equation required

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

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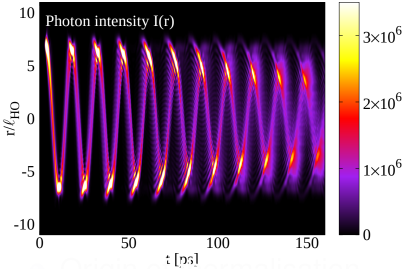
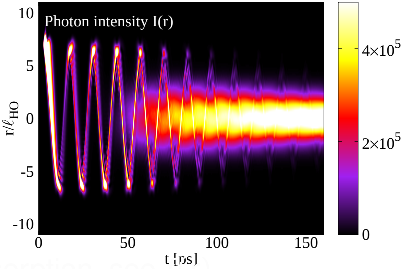
  - **Must** have emission into m, m' superposition
  - **Must** have $K = K(\delta_m)$ (Kennard-Stepanov)

Dynamics from model

Longer cavity



Shorter cavity



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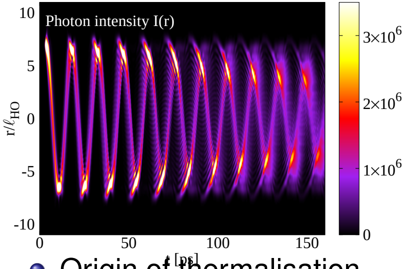

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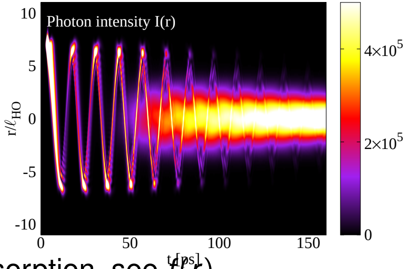

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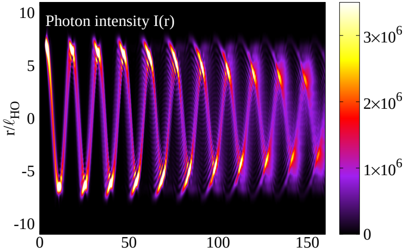

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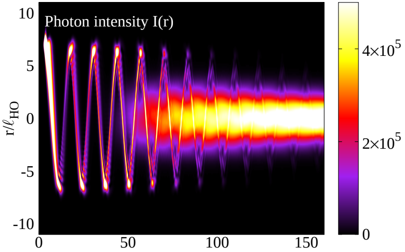

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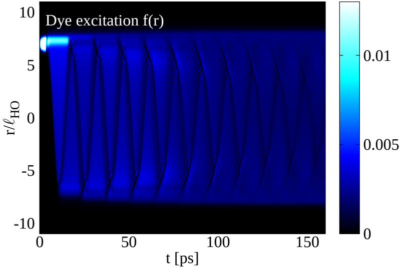
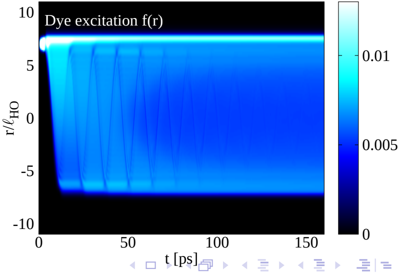
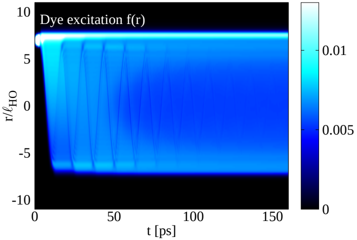




Thermalisation at late times

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



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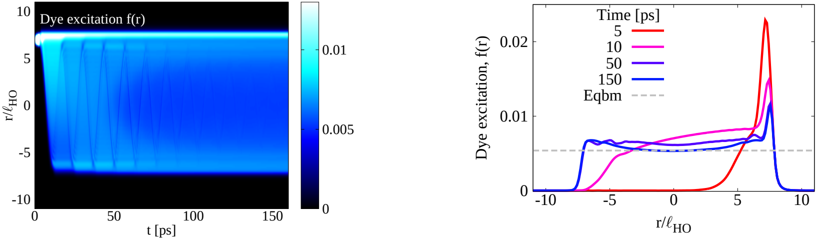


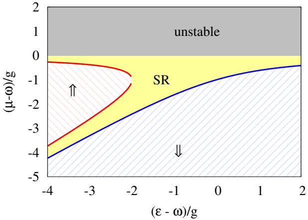
(Toward) strong coupling

Strong coupling phase diagram — mean field

- Mean field — single photon mode

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

- Dicke phase diagram vs μ



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

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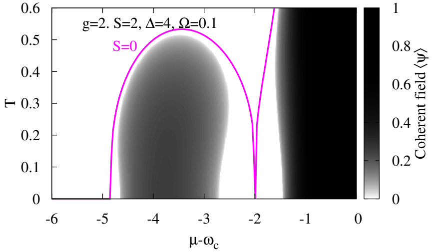

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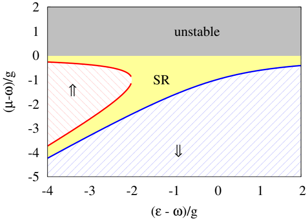

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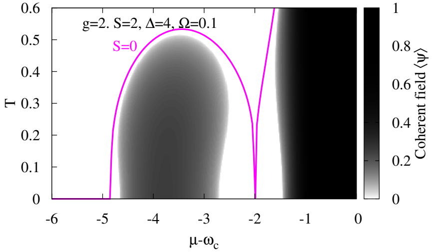

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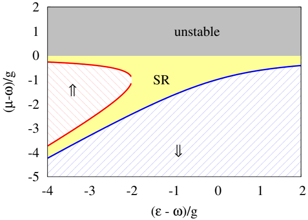

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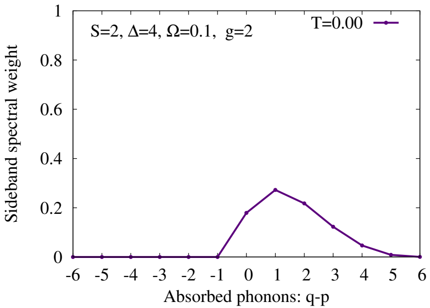
Polariton spectrum: what condensed

- Vibrational composition of condensate



- Optimal $T \sim 2\Omega$

[Cwik *et al.* EPL ’14]

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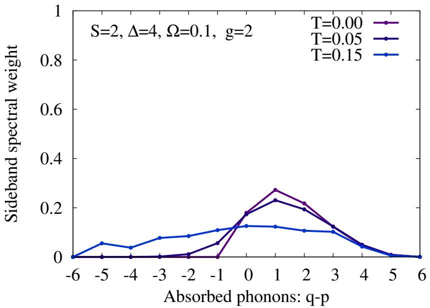

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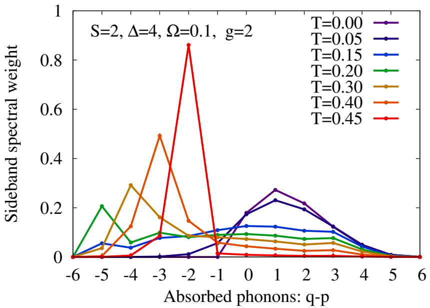

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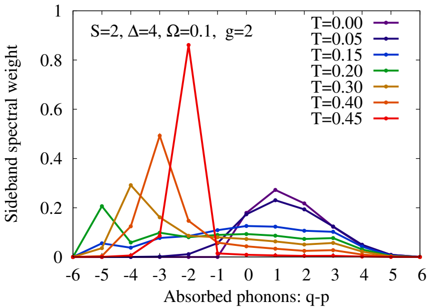

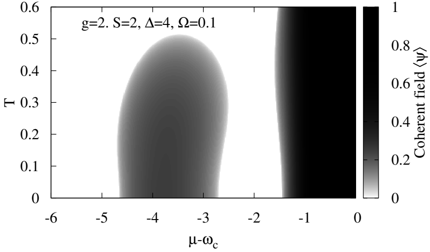

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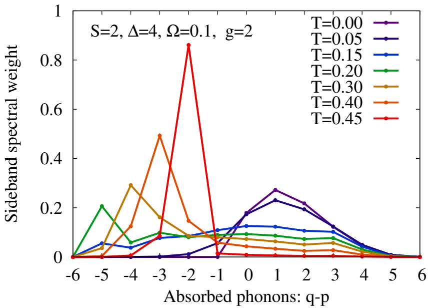
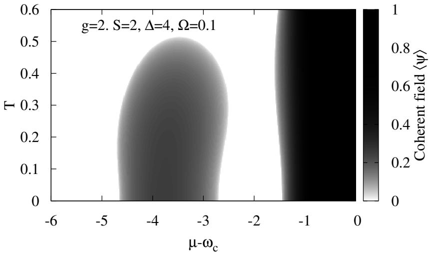
Polariton spectrum: what condensed

- Vibrational composition of condensate



- Optimal $T \sim 2\Omega$



[Cwik *et al.* EPL ’14]

Acknowledgements

GROUP:

FUNDING:

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

EPSRC Engineering and Physical Sciences Research Council

The Leverhulme Trust

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

ICSCE8

Edinburgh, 25^{th}–29^{th} 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)

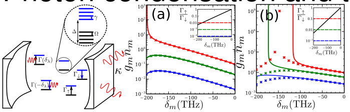
Early-bird registration & abstract deadline: 31^{st} January 2016.

Final registration deadline: 31^{st} March 2016.

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

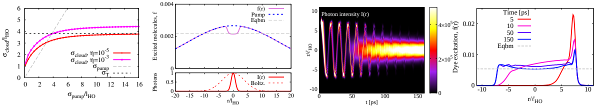
Summary

- Photon condensation and thermalisation



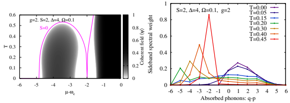
[Kirton & JK, PRL ’13, PRA ’15]

- Photon condensation, pattern formation physics



[JK & Kirton, arXiv:1506:00280]

- Reentrance, phonon assisted transition, 1st order at $S \gg 1$



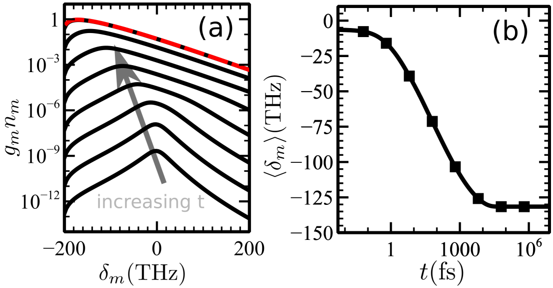
[Cwik *et al.* EPL ’14]

PDRA position available on organic polaritons

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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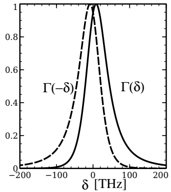

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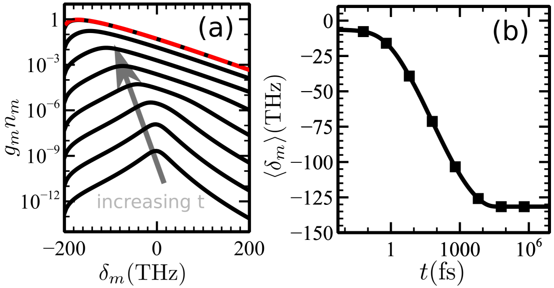

[Kirton & JK PRA '15]

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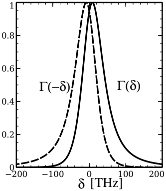

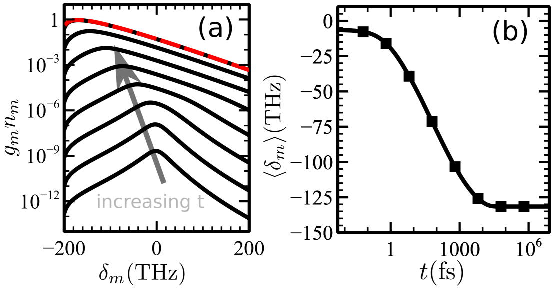

[Kirton & JK PRA '15]

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)

- Thermal at low κ/high temperature
- High loss, κ competes with $\Gamma(\pm\delta_0)$
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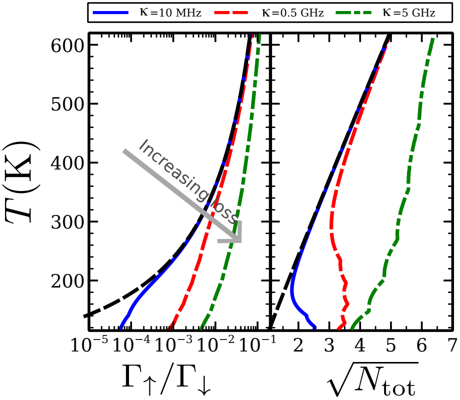

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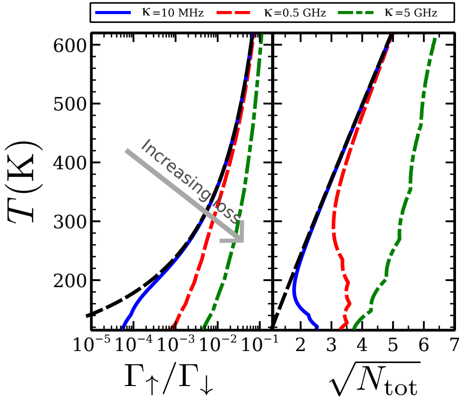

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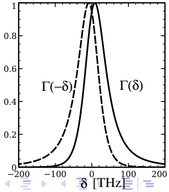

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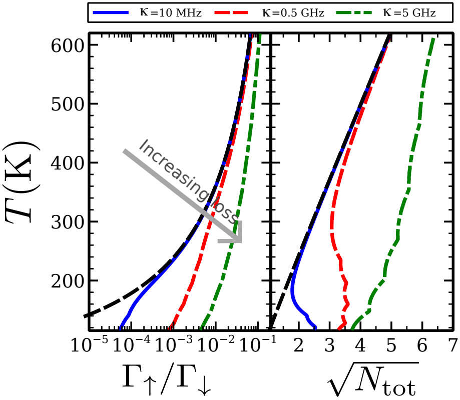

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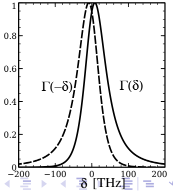

Threshold condition

- Lasing threshold, dependence on spot size.
  - Equilibrium: $\mu = \delta_c$
  - Gives $\Gamma_\uparrow(\mathbf{r}=0) = \Gamma_\downarrow e^{\beta\delta_c}$
- Dependence on ω_c — experimental spectrum

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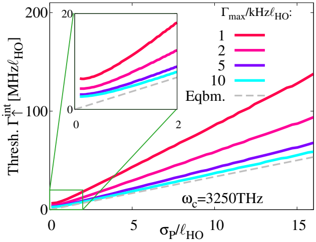

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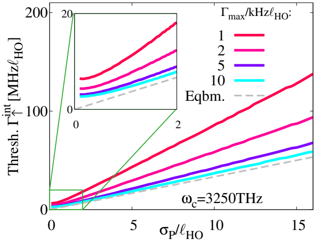

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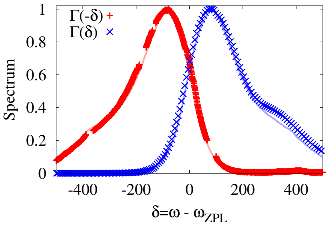

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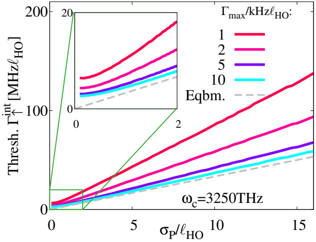

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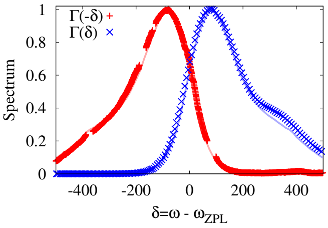

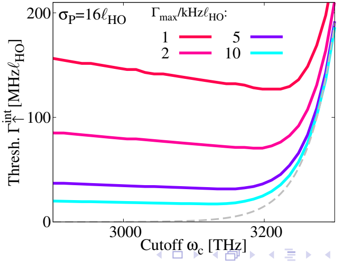

Quantum model, linewidth

Full Master equation:

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

- Factorise $\rho(t) \simeq \rho_{\text{ph}}(t) \bigotimes_i \rho_{m,i}(t)$
- Quantum regression theorm $\rightarrow$ linewidth

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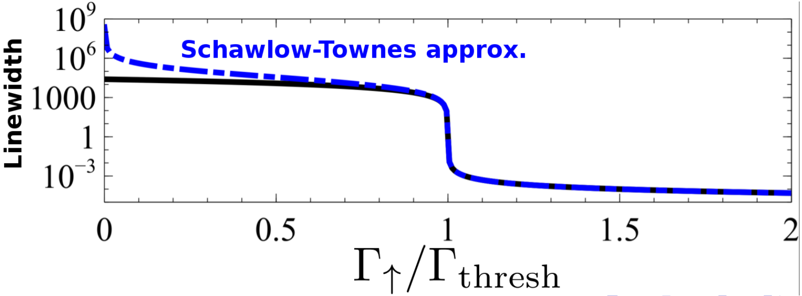

Thermalisation of spectrum:

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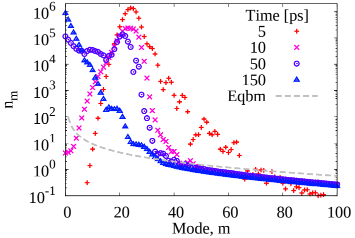

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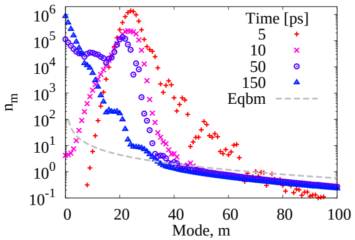
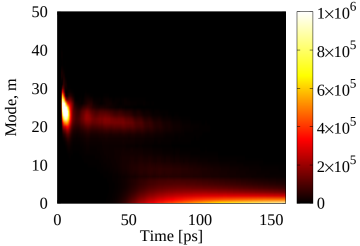
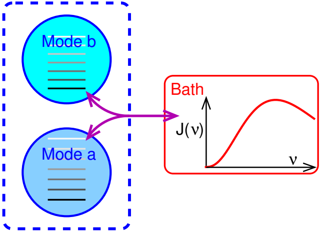




Toy problem: two bosonic modes

- 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

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

- Far from resonance: pointer states are eigenstates

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

- Questions:
  - How does crossover work?
  - Are these actually right?

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

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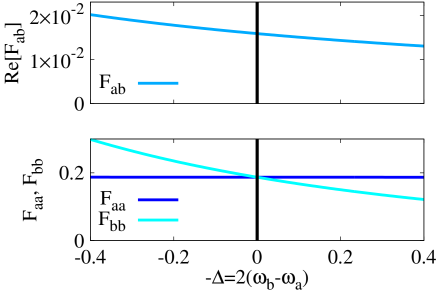

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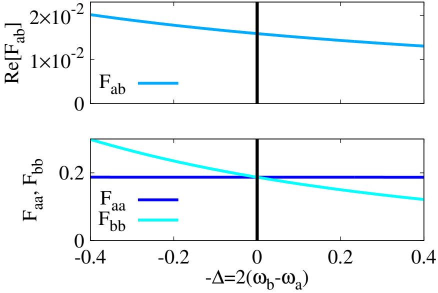

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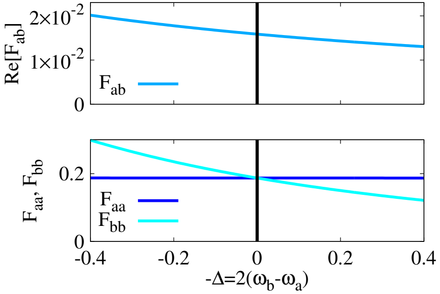

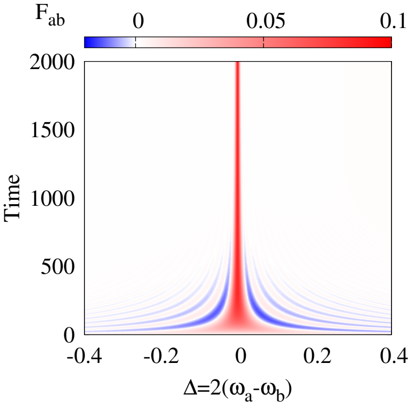

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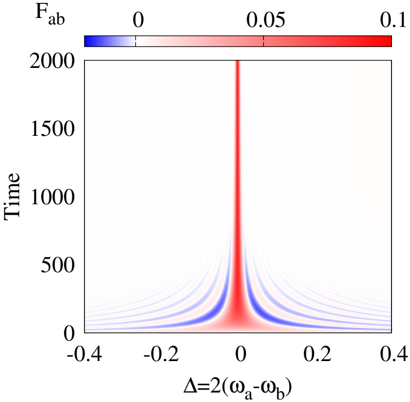

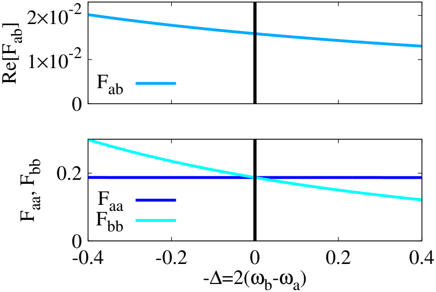

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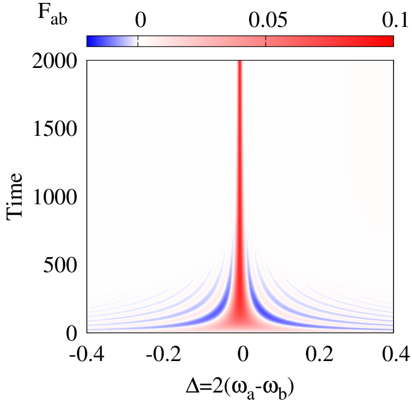

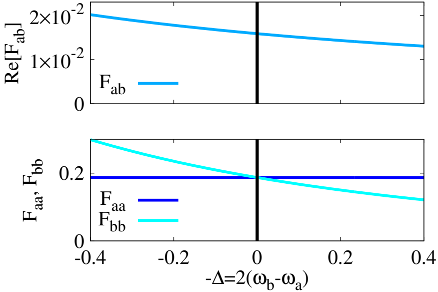

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Toy problem: Bloch-Redfield theory

Unsecularised Bloch-Redfield theory:

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

- Compare to exact solution:

Toy problem: Bloch-Redfield theory

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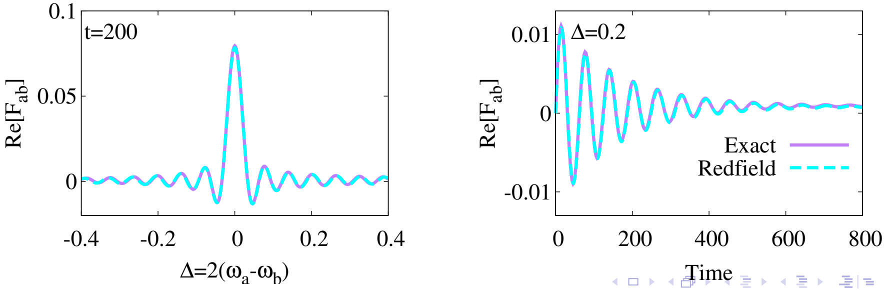
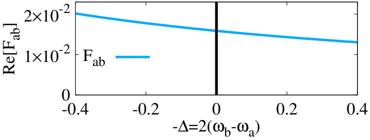
- Compare to exact solution:



Toy problem: Secularisation

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



- Leads to $F_{ab}(t \to \infty) = 0$. Exact:
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- Check stability: consider $f = (F_{aa}, F_{bb}, \Re[F_{ab}], \Im[F_{ab}])$

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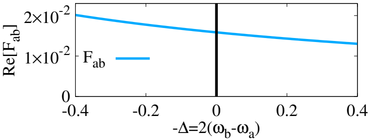

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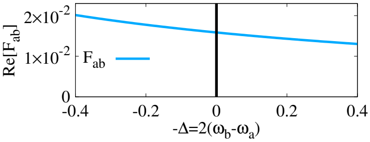

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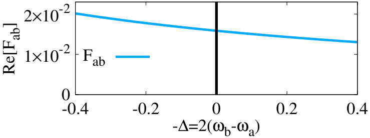

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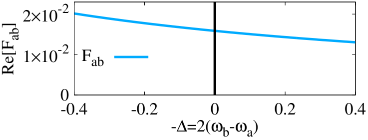

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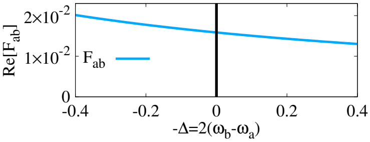

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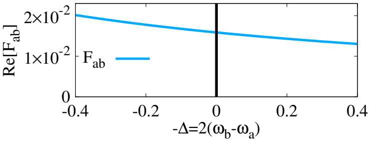

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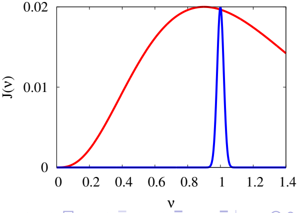

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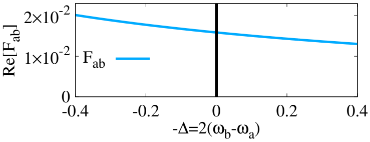

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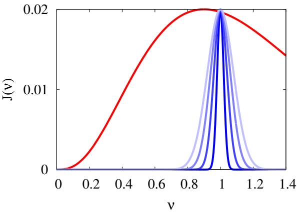
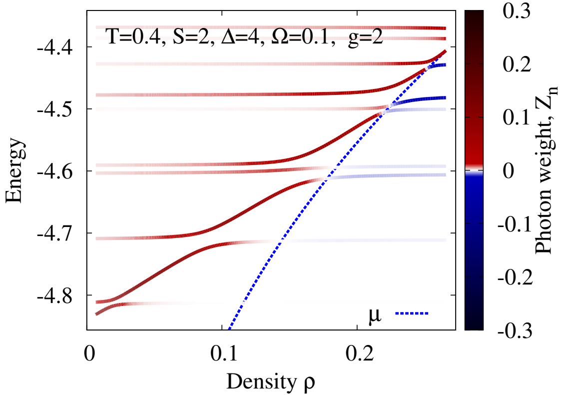


Polariton spectrum: photon weight



- 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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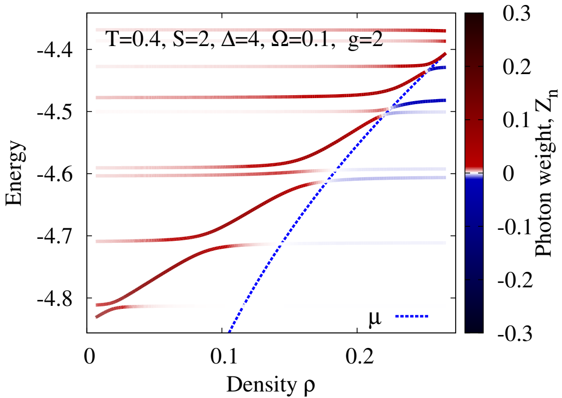

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