Polariton and photon condensates in organic materials

Jonathan Keeling

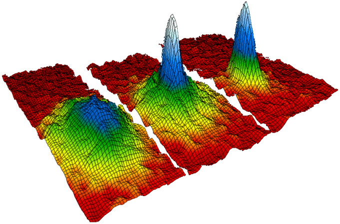
University of St Andrews

1413-2013

Sheffield, October 2014

Coherent states of matter and light

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



[Anderson *et al.* Science '95]

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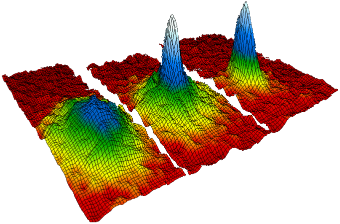

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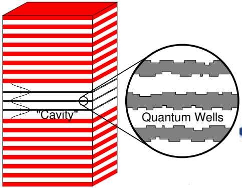
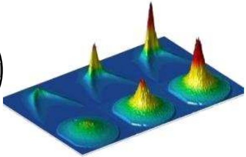
Polariton Condensate $T \sim 20$K





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

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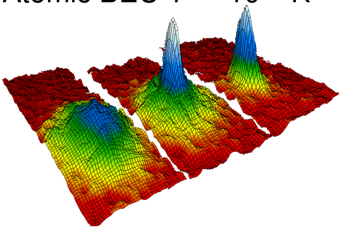

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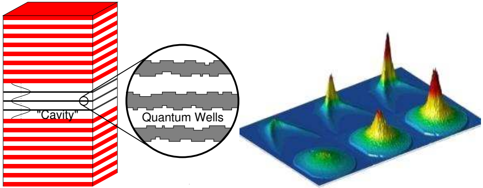
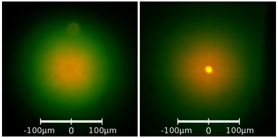


[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K



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

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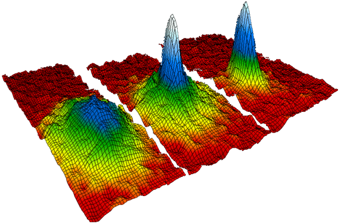

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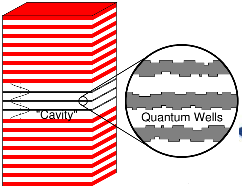

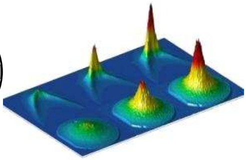

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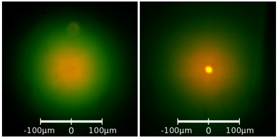
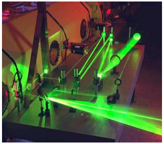


[Klaers *et al.* Nature, '10]

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



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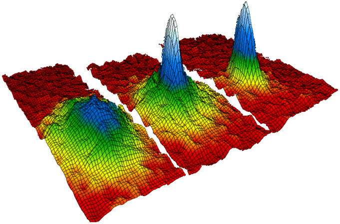

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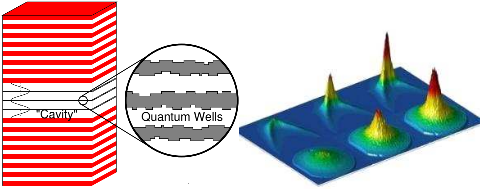

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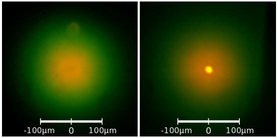

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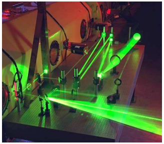
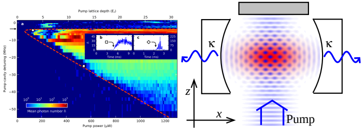


Superradiance transition
$T \sim 0$



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

Outline

1. Condensation, superradiance, lasing
   - Polariton condensation and Dicke model
   - Condensation vs superradiance transition
   - Non-equilibrium condensation vs lasing
2. Room temperature condensates: Organic polaritons
   - Dicke phase diagram with phonons
   - Condensation of phonon replicas?
3. Room temperature condensates: Photons
   - Lasing model and thermalisation
   - Phase diagram
   - Time evolution
   - Linewidth

Acknowledgements

GROUP:

COLLABORATORS: Szymanska (UCL), Reja (MPI-PKS), Littlewood (ANL), De Liberato (Soton)

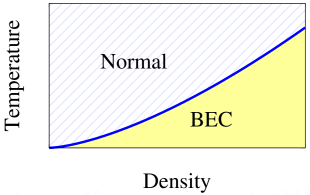
FUNDING:

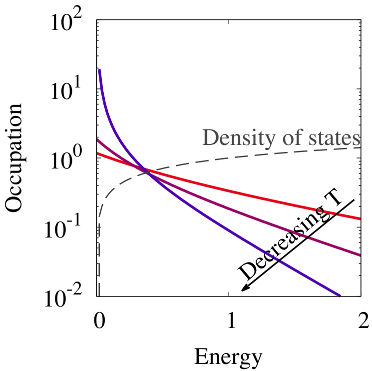
topnes
Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

EPSRC
Engineering and Physical Sciences Research Council

"Textbook" BEC

- **Non-interacting** viewpoint
  - BE distribution: $\mu < \omega_0$
  - $T_c = \frac{2\pi\hbar^2}{m}\left(\frac{n}{\xi_d}\right)^{2/d}$





- **Interacting** approach (WIDBG)

$$H = \sum_k \omega_k \psi_k^\dagger \psi_k + \frac{g}{2V}\sum_{k,k',q} \psi_{k+q}^\dagger \psi_{k'-q}^\dagger \psi_{k+q} \psi_k$$

  - Mean field: $|\psi|^2 = (\mu - \omega_0)/V$

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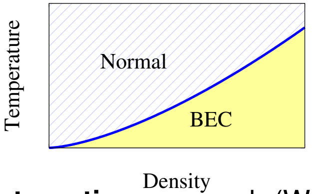

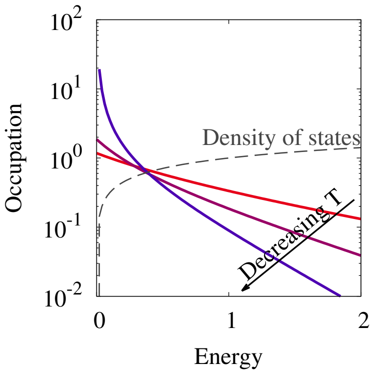

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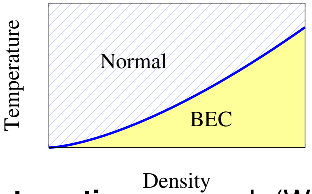

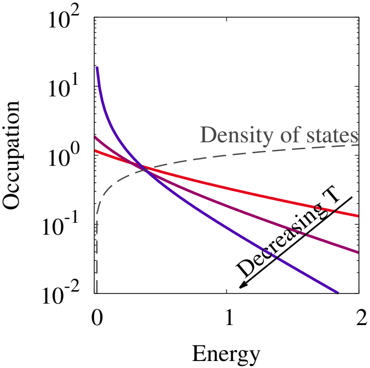

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“Textbook” Laser: Maxwell Bloch equations

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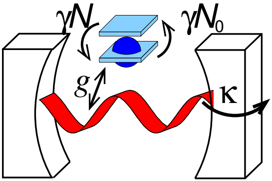

Maxwell-Bloch eqns: $P = -i\langle S^- \rangle, N = 2\langle S^z \rangle$

$$\partial_t \psi = -i\omega_0 \psi - \kappa\psi + \sum_\alpha g_\alpha P_\alpha$$

$$\partial_t P_\alpha = -2i\epsilon_\alpha P_\alpha - 2\gamma P + g_\alpha \psi N_\alpha$$

$$\partial_t N_\alpha = 2\gamma(N_0 - N_\alpha) - 2g_\alpha(\psi^* P_\alpha + P^*_\alpha \psi)$$

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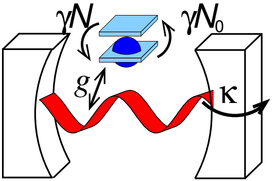

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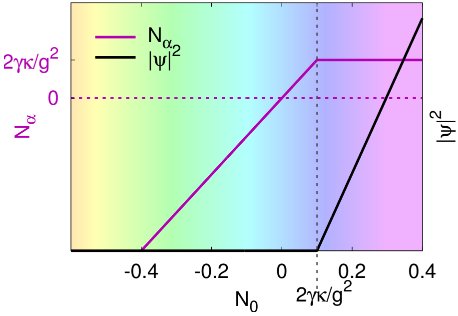
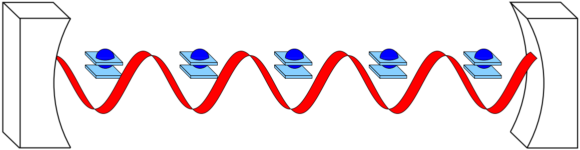


$|\psi|^2 > 0$ if $N_0 g^2 > 2\gamma\kappa$

"Textbook" Dicke-Hepp-Lieb superradiance



$$H = \omega\psi^\dagger\psi + \sum_\alpha \epsilon S^z_\alpha + g\left(\psi^\dagger S^-_\alpha + \psi S^+_\alpha\right)$$

- Coherent state: $|\Psi\rangle \to e^{\lambda\psi^\dagger + \eta S^+}|\Omega\rangle$
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[Hepp, Lieb, Ann. Phys. '73]

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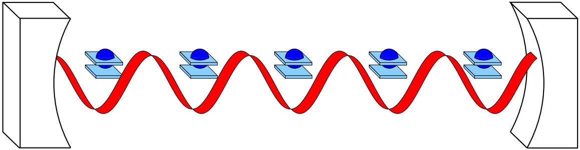

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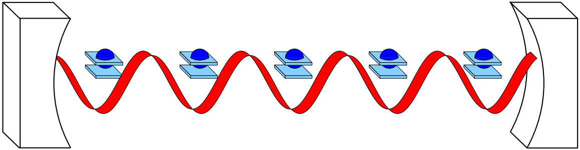

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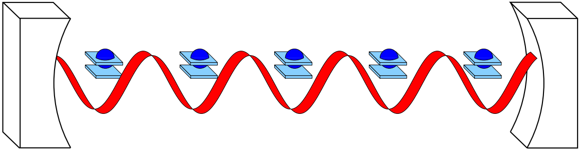

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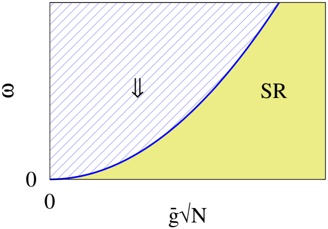

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Polariton condensation and Dicke model

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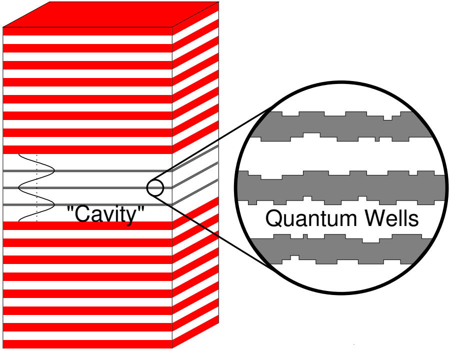

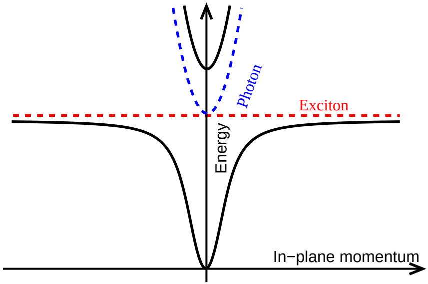

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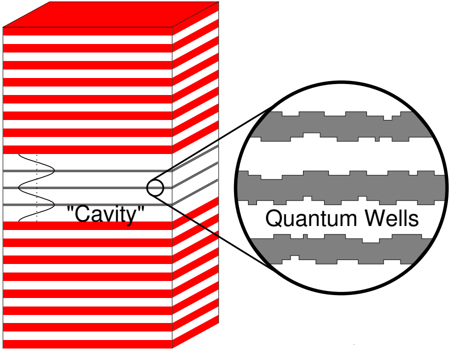

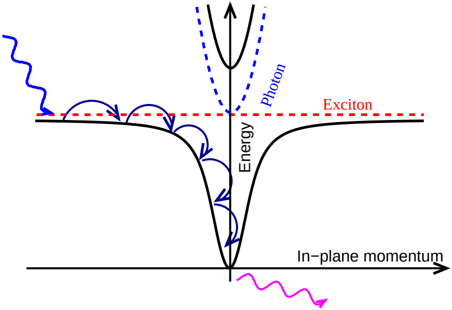

Lasing-condensation crossover model

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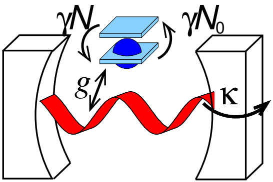

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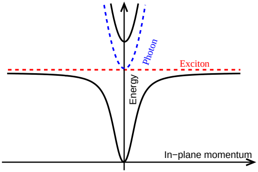

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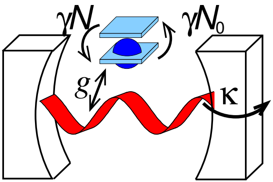

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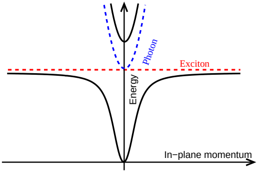

Dicke model:

$$H_{\text{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^{\dagger}_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\epsilon S^{z}_{\alpha} + g_{\alpha,\mathbf{k}} \psi_{\mathbf{k}} S^{+}_{\alpha} + \text{H.c.} \right]$$

Dicke-Hepp-Lieb superradiance and modes

$$H = \omega\psi^\dagger\psi + \epsilon S^z + g\left(\psi^\dagger S^- + \psi S^+\right)$$

Spontaneous polarisation if: $Ng^2 > \omega\epsilon$

- Normal state, $S^z = -N/2 + B^\dagger B$
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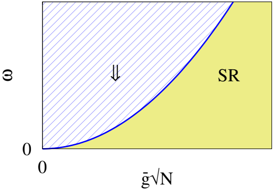
- Excitation cost E:

$$(E-\omega)(E-\epsilon) = g^2 N$$

- Transition when $E = 0$



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

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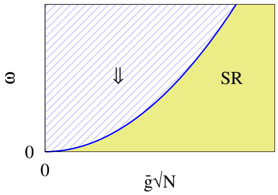

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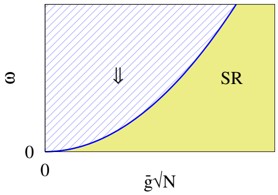

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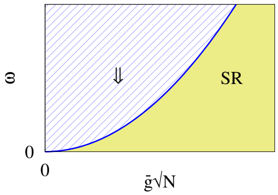

[Hepp, Lieb, Ann. Phys. '73]

Grand canonical ensemble

Grand canonical ensemble:

- If $H \to H - \mu(S^z + \psi^\dagger\psi)$, need only: $g^2 N > (\omega - \mu)|\epsilon - \mu|$
- Fix density / fix $\mu > 0$ — pumping

- Transition at: $g^2 N > (\omega - \mu)|\epsilon - \mu|$
- μ hits lowest mode

[Eastham and Littlewood, PRB '01]

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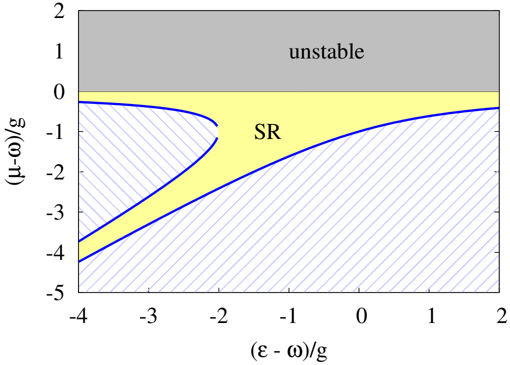

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[Eastham and Littlewood, PRB '01]

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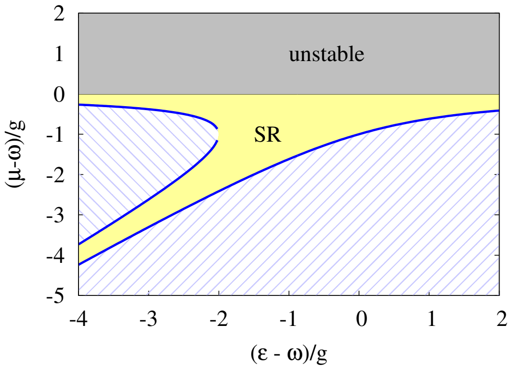

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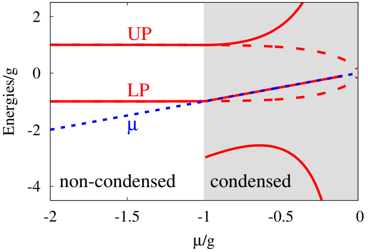

Plot for $\omega = \epsilon = 0$

[Eastham and Littlewood, PRB '01]

Polariton model and equilibrium results

- Localised excitons, propagating photons

$$H - \mu N = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \mu)\psi^\dagger_{\mathbf{k}}\psi_{\mathbf{k}} + \sum_{\alpha} (\epsilon_\alpha - \mu) S^z_\alpha + g_{\alpha,\mathbf{k}}\psi_{\mathbf{k}} S^+_\alpha + \text{H.c.}$$

- Self-consistent polarisation and field

$$(\omega - \mu)\,\psi = \sum_\alpha \frac{g_\alpha^2 \psi}{E_\alpha} \tanh\left(\beta E_\alpha / 2\right), \quad {E_\alpha}^2 = (\epsilon_\alpha - \mu)^2 + 4 g_\alpha^2 |\psi|^2$$

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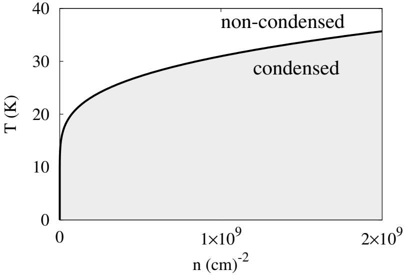

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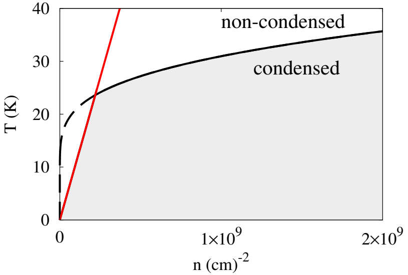

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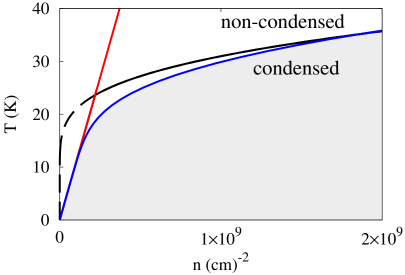

Non-equilibrium condensation vs lasing

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Simple Laser: Maxwell Bloch equations

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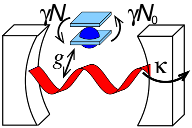

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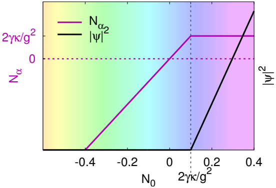

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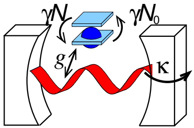

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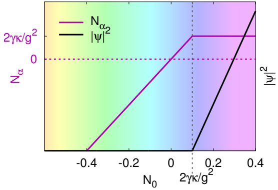

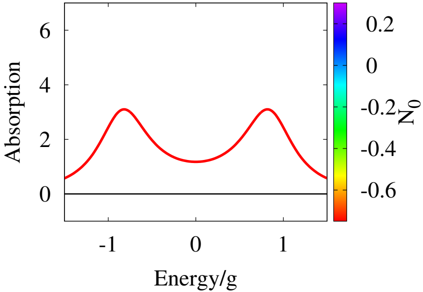

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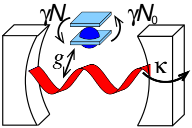

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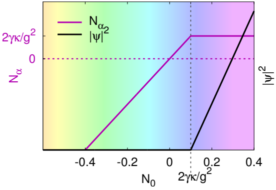

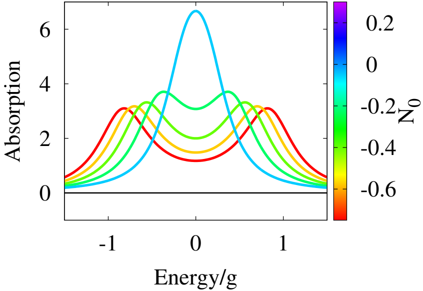

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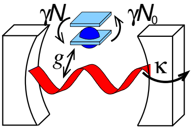

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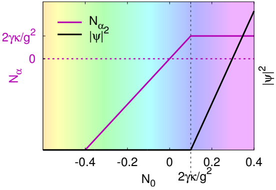

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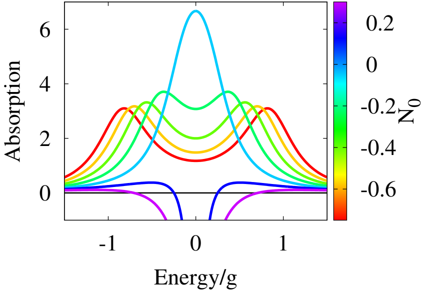

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Poles of Retarded Green's function and gain

$$\left[D^R(\nu)\right]^{-1} = \nu - \omega_k + i\kappa + \frac{g^2 N_0}{\nu - 2\epsilon + i2\gamma}$$

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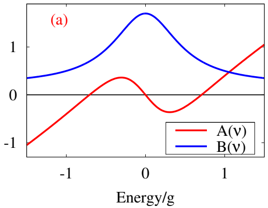

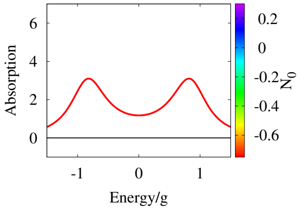

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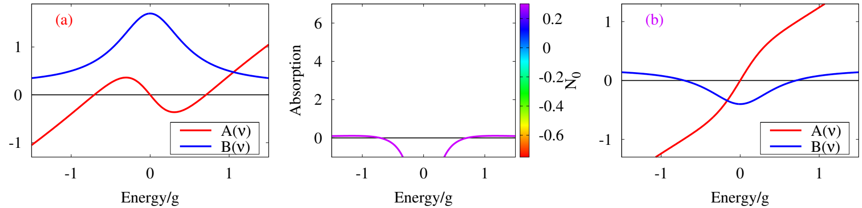

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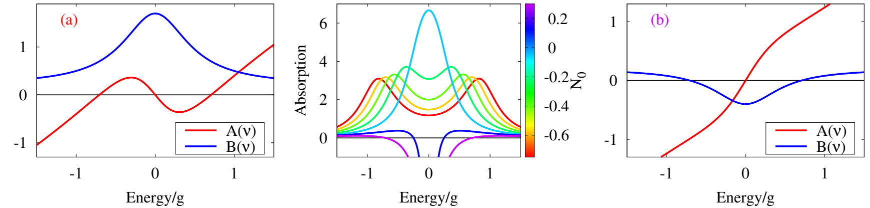
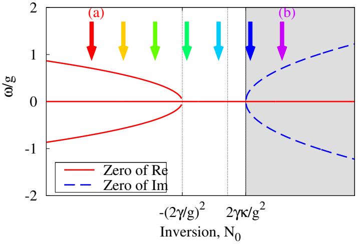


Laser:



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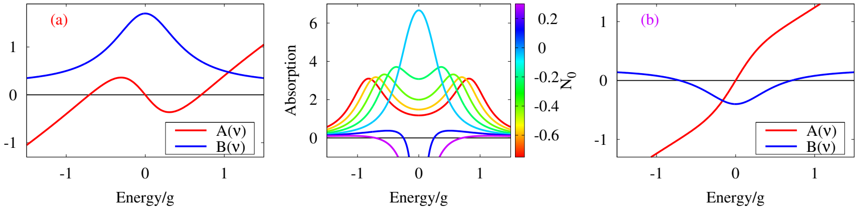

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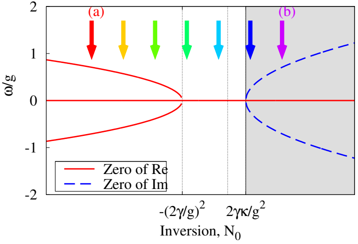

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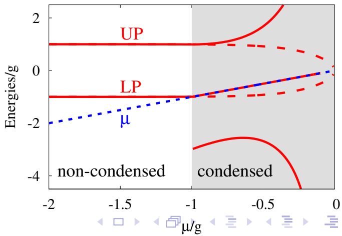
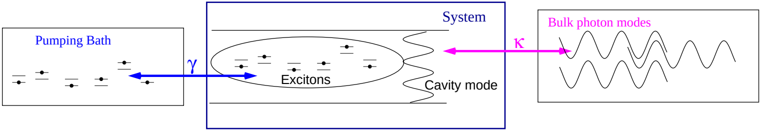
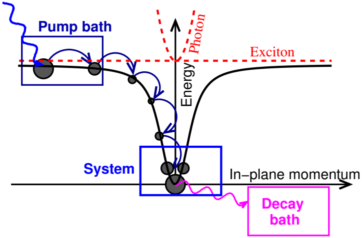


Non-equilibrium description: baths





$$H = H_{\text{sys}} + H_{\text{sys,bath}} + H_{\text{bath}}$$

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- Pump bath: Thermal μ_B, T_B

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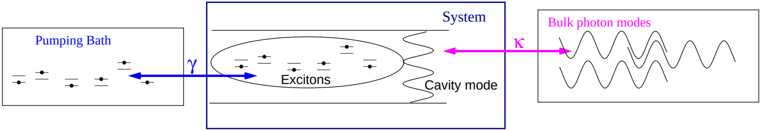

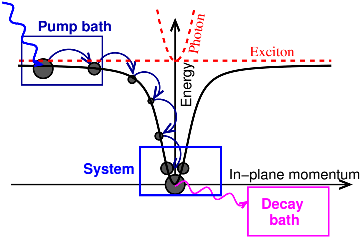

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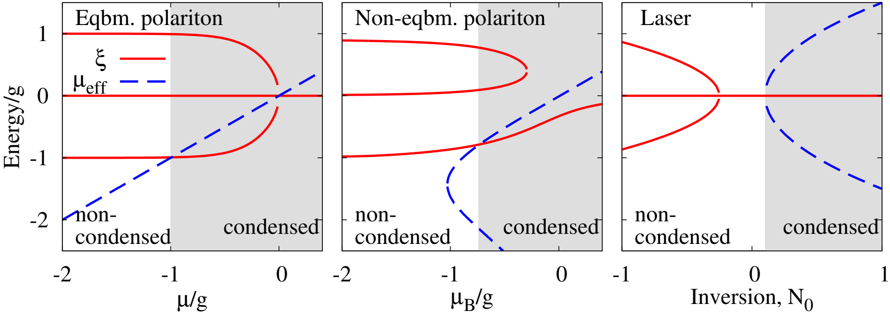
Strong coupling and lasing — low temperature phenomenon



- Inversionless
- **allows** strong coupling
- **requires** low $T \leftrightarrow$ condensation
- Related *weak-coupling* **inversionless** lasing

[Szymanska *et al.* PRL '06; Keeling *et al.* book chapter 1010.3338]

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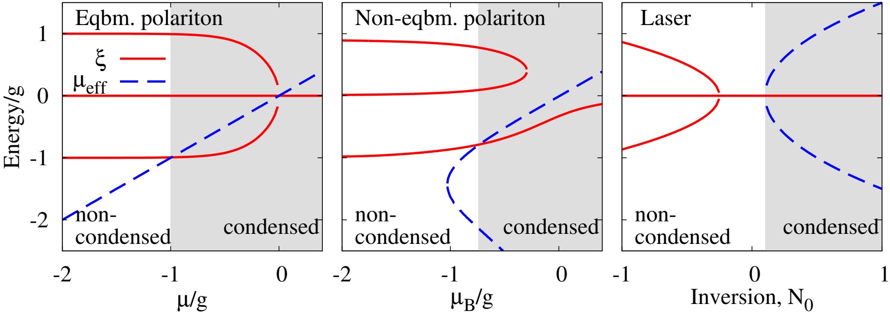

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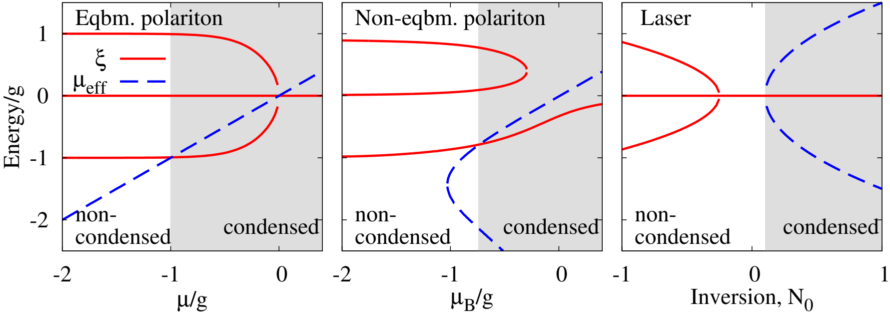

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Room temperature condensates: Organic polaritons

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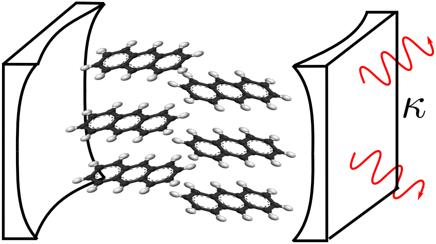
Organic materials in microcavities

- What?
- Why?



- Lasing threshold at room T

[Lidzey *et al.* , Nat. '98, Kena Cohen and Forrest, Nat. Photon '10; Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

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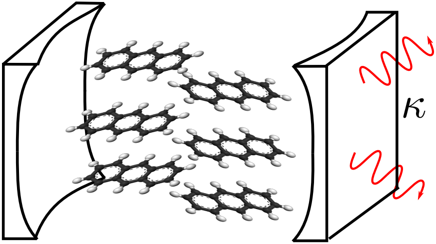

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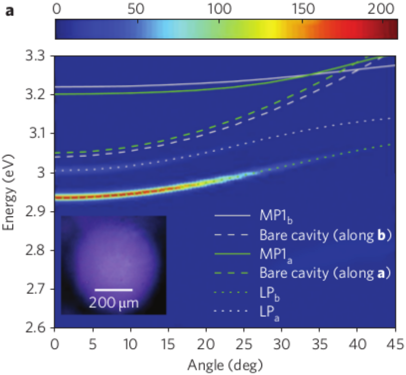

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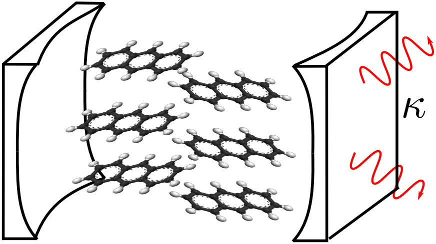

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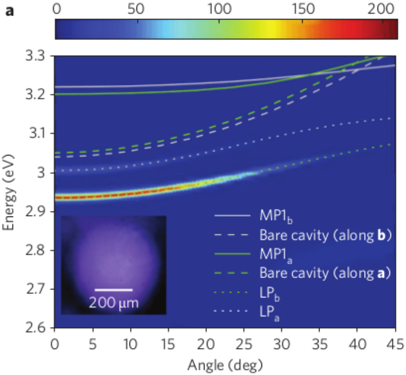

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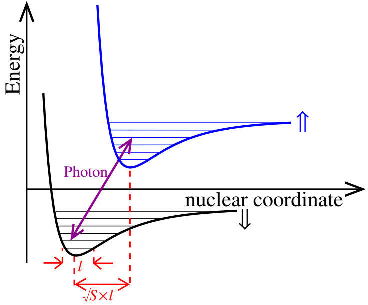
Dicke Holstein Model



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- Phonon frequency Ω
- Huang-Rhys parameter S — phonon coupling

- Phase diagram with $S \neq 0$
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- Polariton spectrum, phonon replicas
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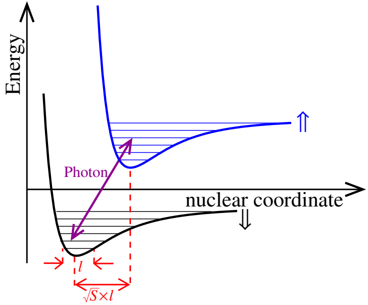

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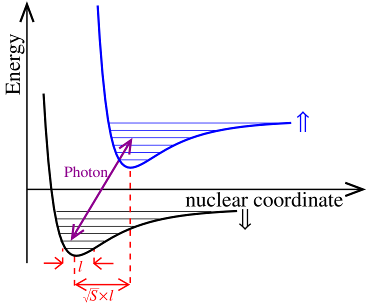

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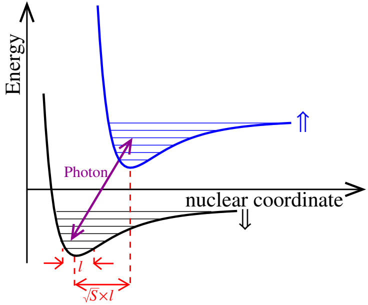

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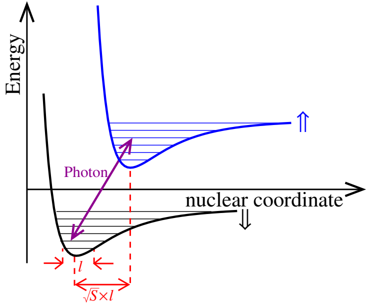
Dicke Holstein Model



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

- Phonon frequency Ω
- Huang-Rhys parameter S — phonon coupling

Questions?

- Phase diagram with $S \neq 0$
  - 2LS energy $\epsilon - n\Omega$
- Polariton spectrum, phonon replicas
- Ultra-strong phonon coupling?

Phase diagram

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- S suppresses condensation — reduces overlap
- Reentrant behaviour — Min μ at $T \sim 0.2$
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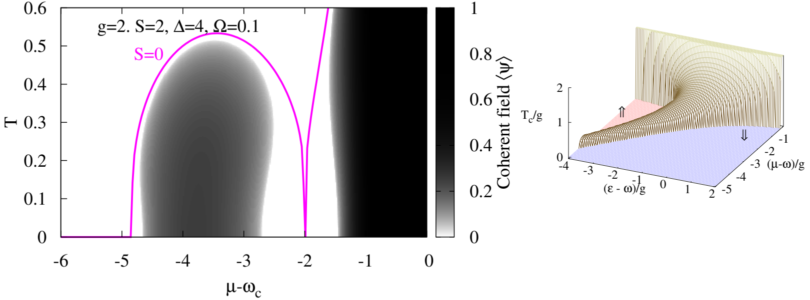

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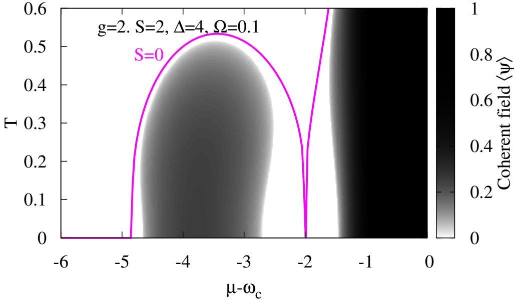

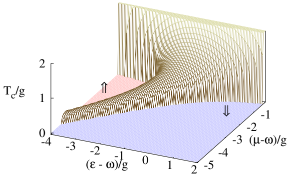

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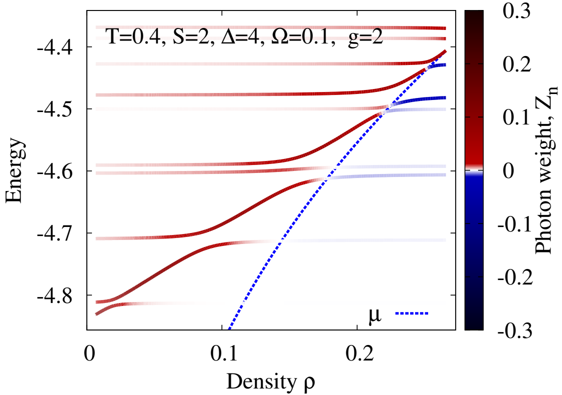
Polariton spectrum: photon weight



- Saturating 2LS: $g_{\text{eff}}^2 \sim g^2(1-2\rho)$

[Cwik *et al.* EPL '14]

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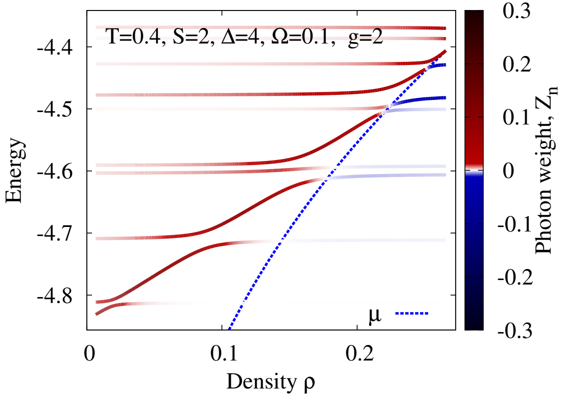

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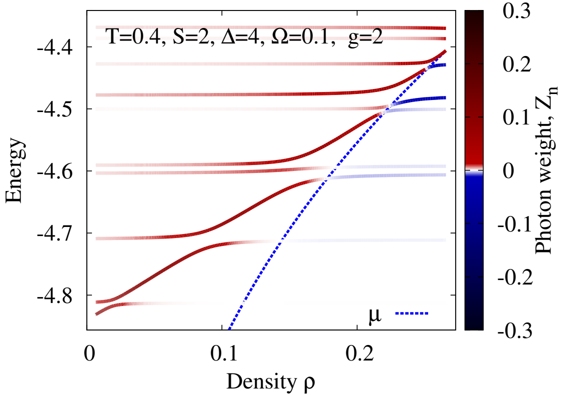

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[Cwik *et al.* EPL '14]

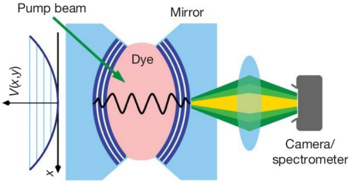
Room temperature condensates: Photons

1. Condensation, superradiance, lasing
   - Polariton condensation and Dicke model
   - Condensation vs superradiance transition
   - Non-equilibrium condensation vs lasing

2. Room temperature condensates: Organic polaritons
   - Dicke phase diagram with phonons
   - Condensation of phonon replicas?

3. Room temperature condensates: Photons
   - Lasing model and thermalisation
   - Phase diagram
   - Time evolution
   - Linewidth

Photon BEC experiments



- Dye filled microcavity
- No strong coupling

[Klaers et al, Nature, 2010]

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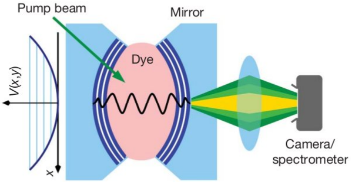

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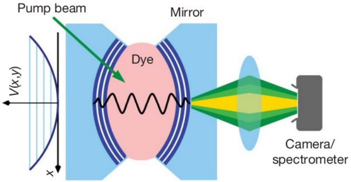

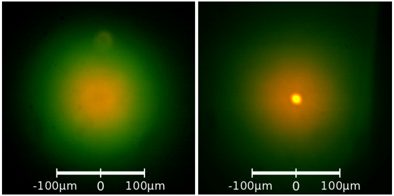

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Relation to dye laser

- 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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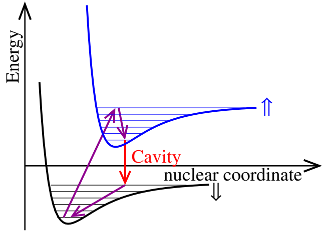
4 Level Dye Laser



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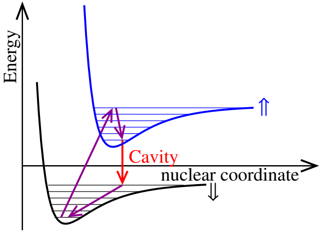

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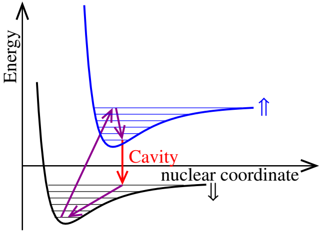

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Modelling

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

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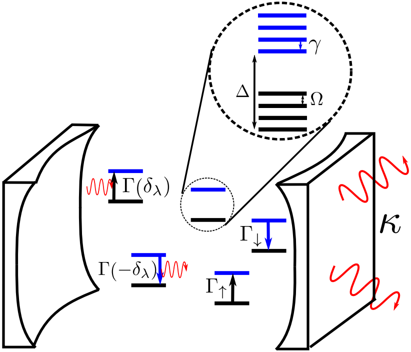

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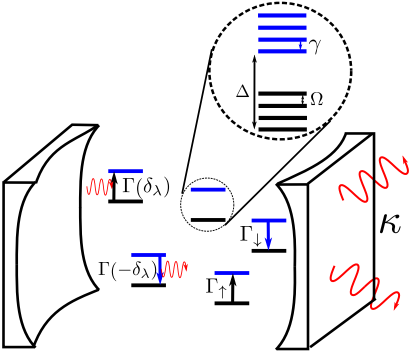

Modelling

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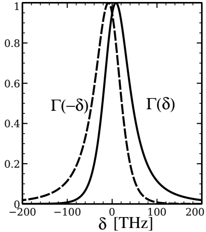

- Kennard-Stepanov $\Gamma(+\delta) \simeq \Gamma(-\delta)e^{\beta\delta}$
- Expt: $\omega_0 < \epsilon$
- $\Gamma \to 0$ at large δ

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

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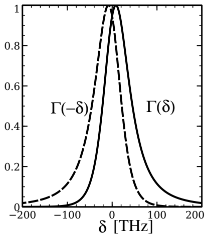

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Distribution $g_m n_m$

- Master equation $\rightarrow$ Rate equation

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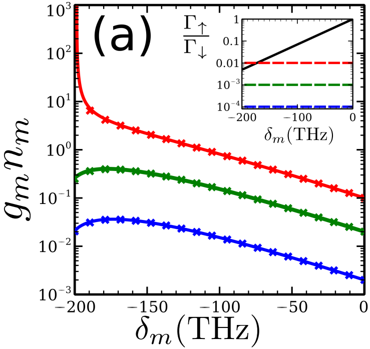

Low loss: Thermal

[Kirton & JK PRL '13]

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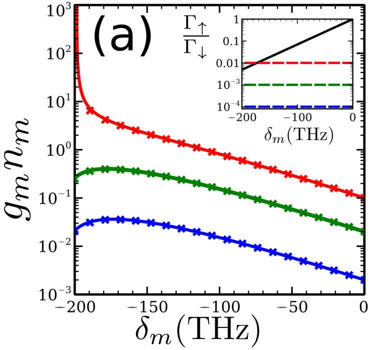

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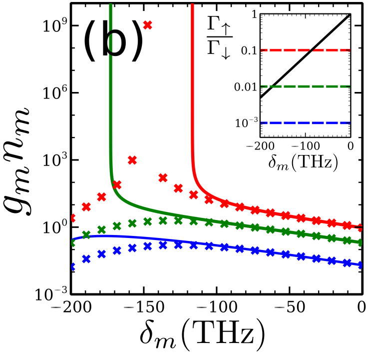
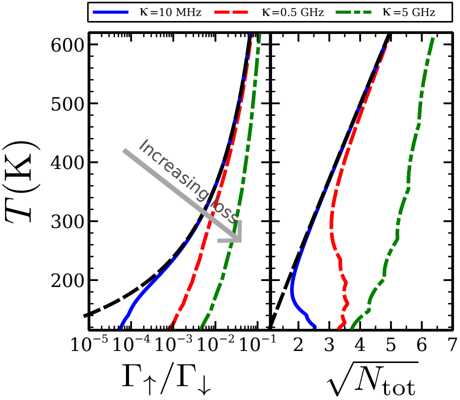


High loss $\to$ Laser

[Kirton & JK PRL '13]

Threshold condition



Compare threshold:

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

- Thermal at low κ/high temperature
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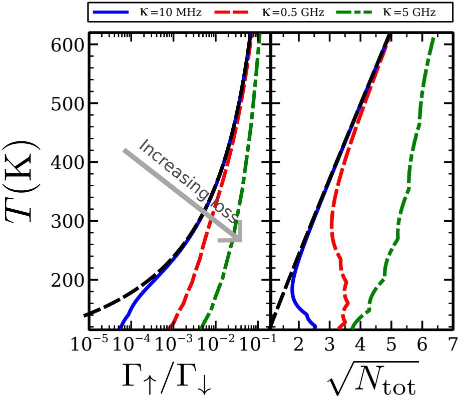

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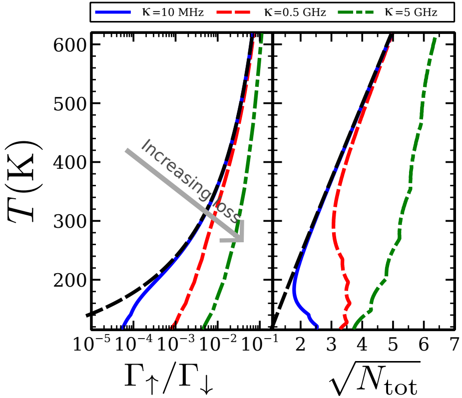

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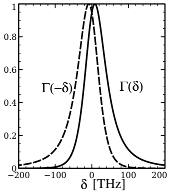

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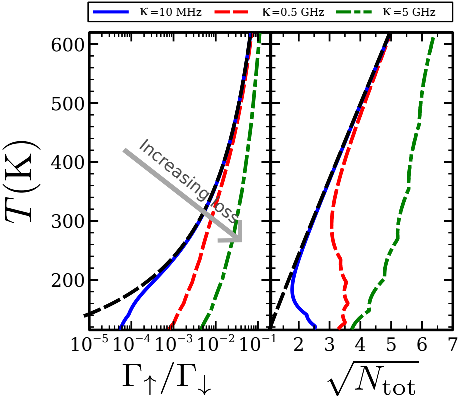

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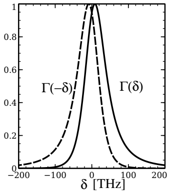
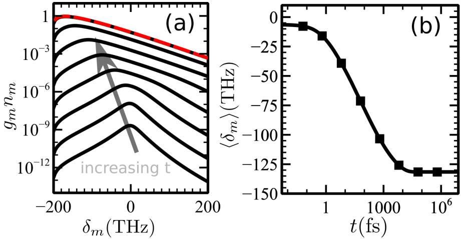


[Kirton & JK PRL '13]

Time evolution

- Initial state: excited molecules
- Initial emission, follows gain peak
- Thermalisation by repeated absorption
- Above threhsold, jump to condensate



[Kirton & JK arXiv:1410.XXXX]

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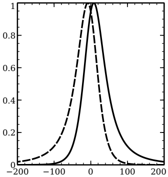

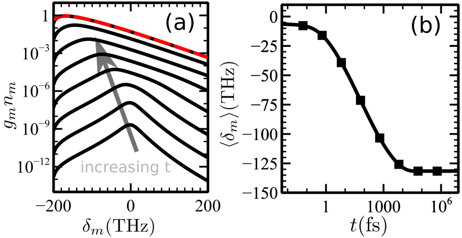

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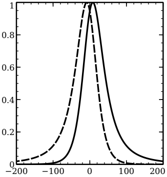

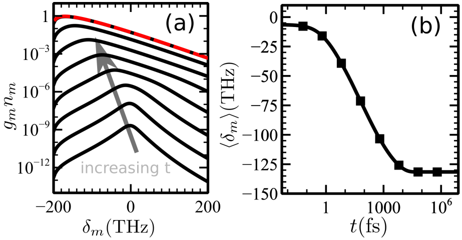

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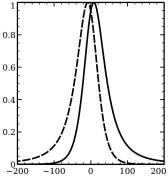

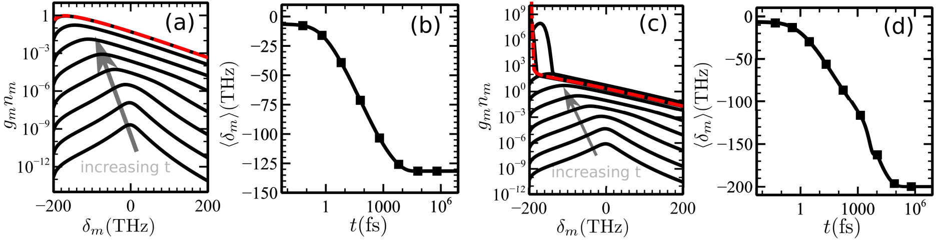

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Linewidth

1. Condensation, superradiance, lasing
   - Polariton condensation and Dicke model
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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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- Quantum regression theorm $\rightarrow$ linewidth

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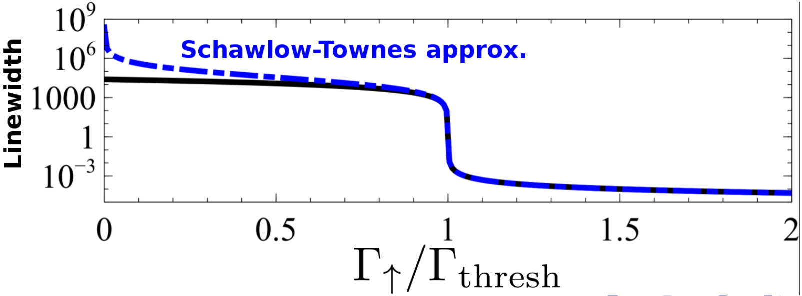
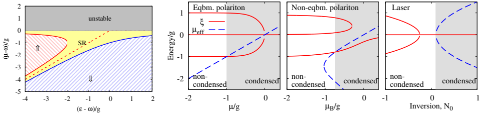
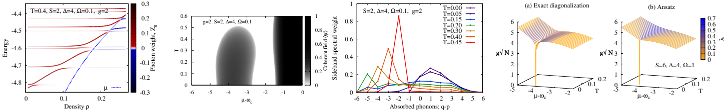


Summary

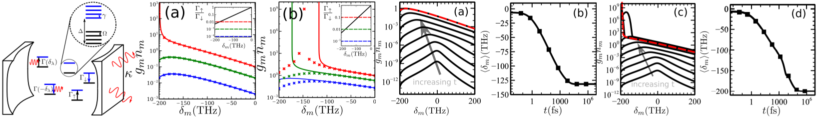
- Polariton condensation vs lasing; superradiance



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

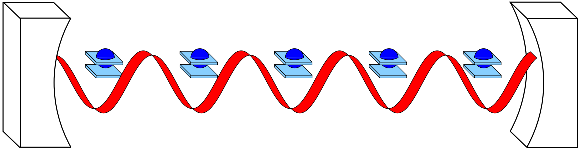


- Photon condensation and thermalisation



Extra slides

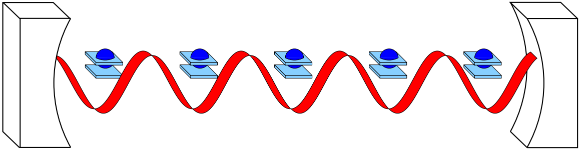
No go theorem and transition



Spontaneous polarisation if: $Ng^2 > \omega\epsilon$

[Rzazewski *et al* PRL '75]

No go theorem and transition



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No go theorem:. Minimal coupling $(p - eA)^2/2m$

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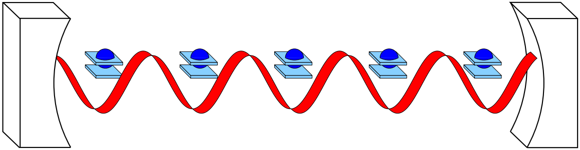

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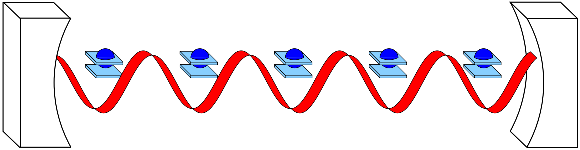

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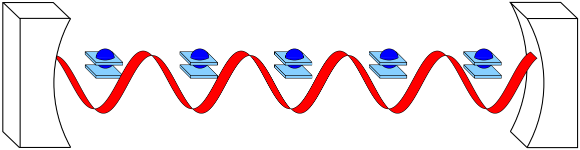

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Dicke phase transition: ways out

Problem: $g^2/\omega_0 < 2\zeta$ for intrinsic parameters. **Solutions**:

- **Interpretation**
  Ferroelectric transition in $\mathbf{D} \cdot \mathbf{r}$ gauge.
  [JK JPCM '07, Vukics & Domokos PRA 2012]
- Circuit QED [Nataf and Ciuti, Nat. Comm. '10; Viehmann *et al.* PRL '11]
- Grand canonical ensemble:
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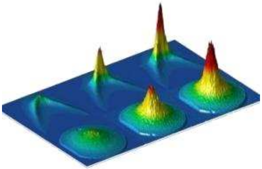

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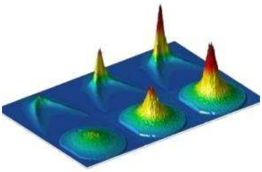

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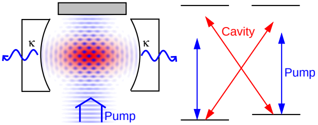

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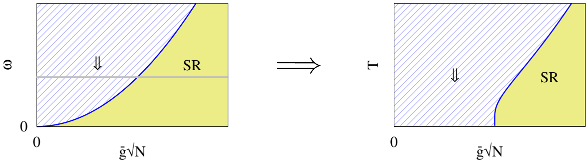

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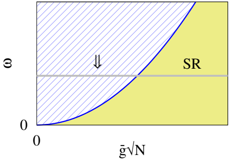

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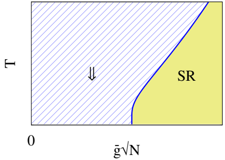

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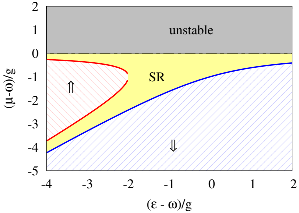

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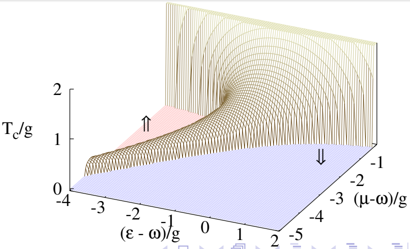
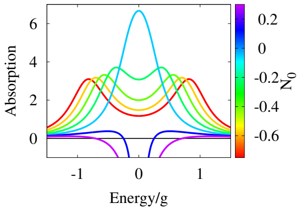


Maxwell-Bloch Equations: Retarded Green's function



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  Response to perturbation

- Absorption $= -2\Im[D^R(\omega)]$

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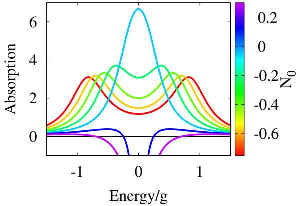

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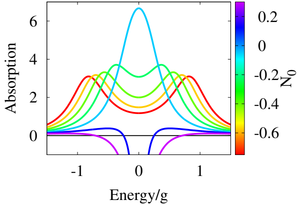

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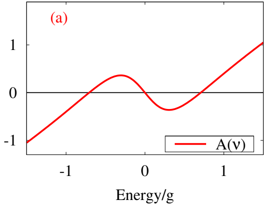

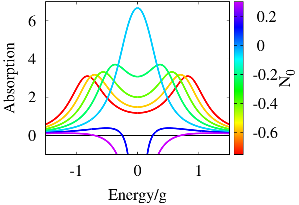

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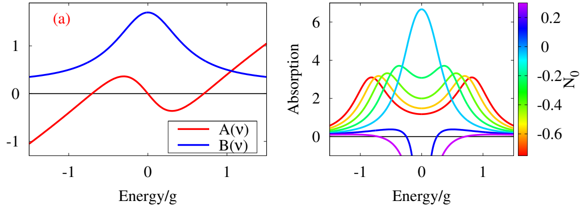

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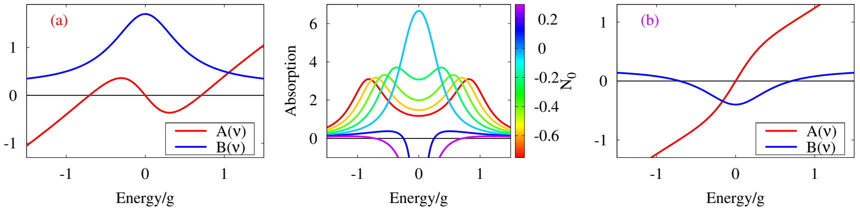

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Organic materials in microcavities

- State of art:
  - Strong coupling:
    - ★ J aggregates [Bulovic *et al.*]
    - ★ Crystaline anthracene [Forrest *et al.*]
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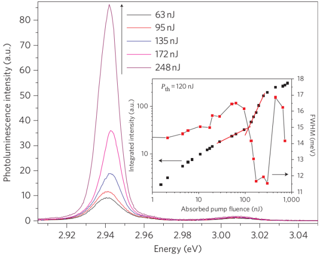

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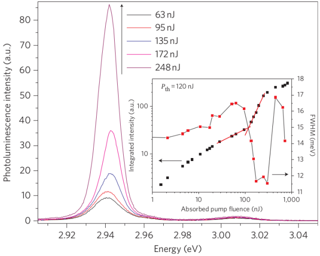

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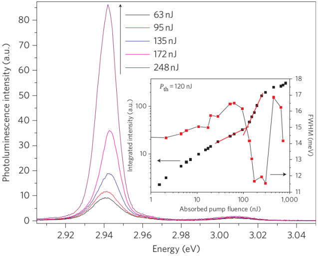

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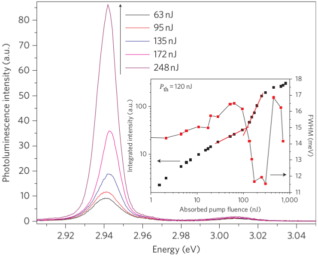

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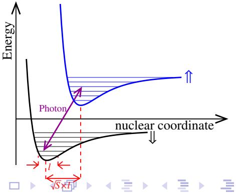

Polariton spectrum: what condensed

- Repeat weight for *n*-phonon channel
- Eigenvector that is macroscopically occupied

- Optimal $T \sim 2\Omega$

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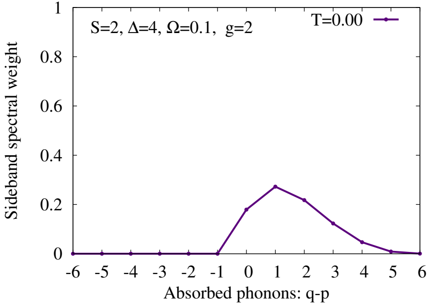

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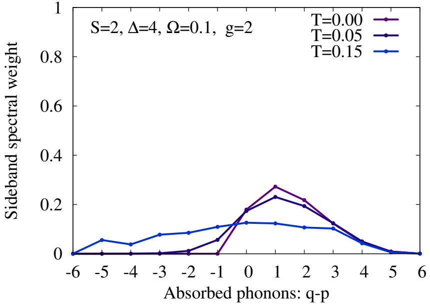

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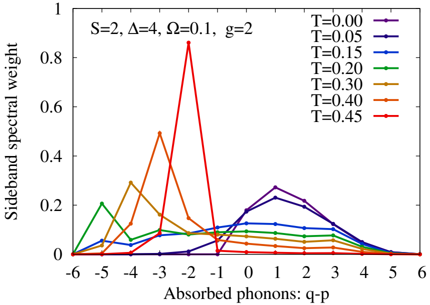

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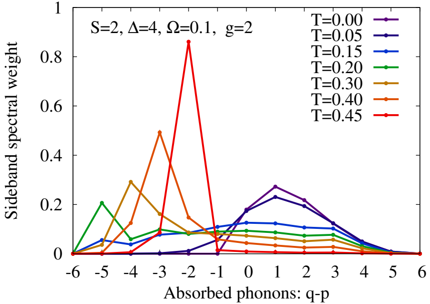

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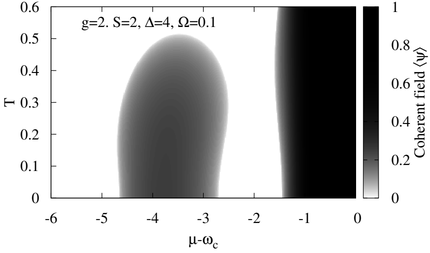

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Organic polaritons

Critical coupling with increasing S

- Re-orient phase diagram
- g vs μ, T
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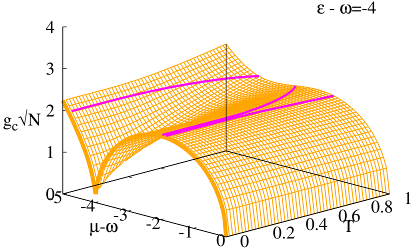

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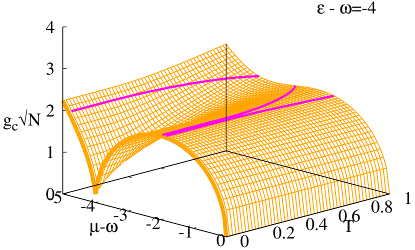

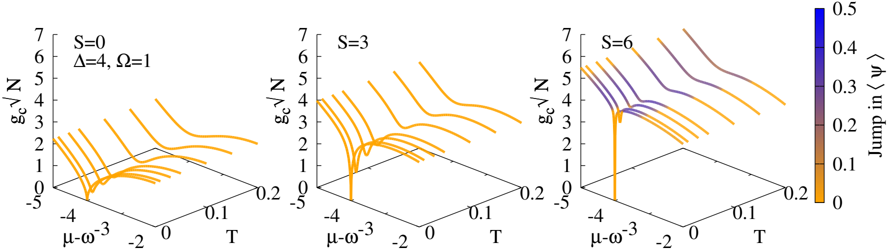

Explanation: Polaron formation

- Unitary transform

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- Coupling moves to $S^\pm$

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- Optimal phonon displacements, $\sim \sqrt{S}$
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Explanation: Polaron formation

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$$H_\alpha \to \tilde{H}_\alpha = e^{K_\alpha} H_\alpha e^{-K_\alpha} \qquad K = \sqrt{S} S^z_\alpha (b^\dagger_\alpha - b_\alpha)$$

- Coupling moves to $S^\pm$

$$\tilde{H}_\alpha = \text{const.} + \epsilon S^z_\alpha + \Omega b^\dagger_\alpha b_\alpha + g \left[\psi S^+_\alpha e^{\sqrt{S}(b^\dagger_\alpha - b_\alpha)} + \text{H.c.} \right]$$

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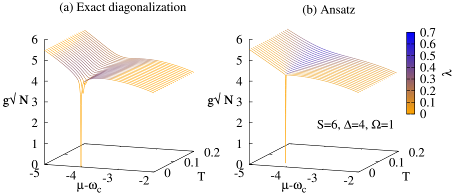
Collective polaron formation

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



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

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

Effective 2LS energy in field:

$$\zeta^2 = \left(\frac{\epsilon - \mu}{2} + \Omega\sqrt{S}(1-\eta)\zeta\right)^2 + g^2\lambda^2 e^{-S\eta^2}$$

[Cwik *et al.* EPL '14]

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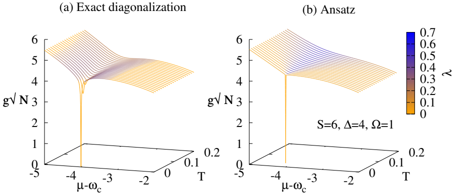

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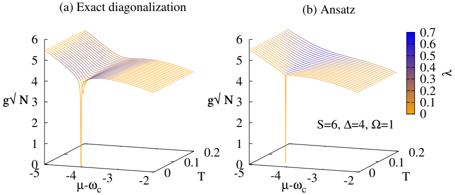

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Polariton spectrum — coupled oscillators

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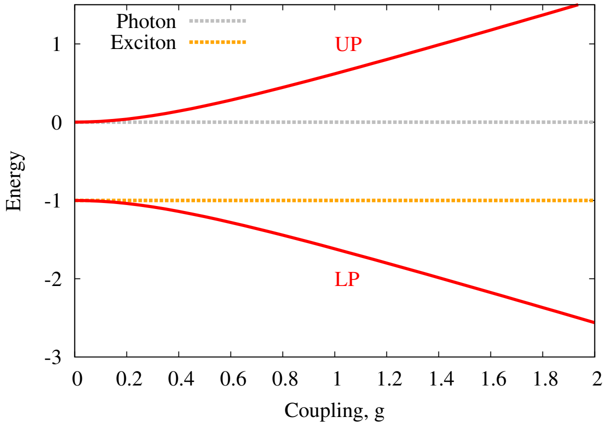

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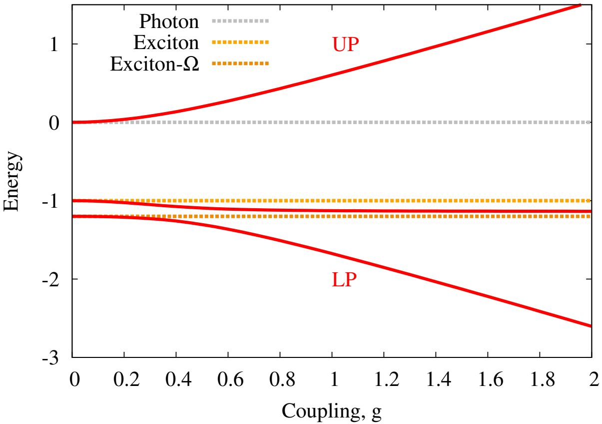

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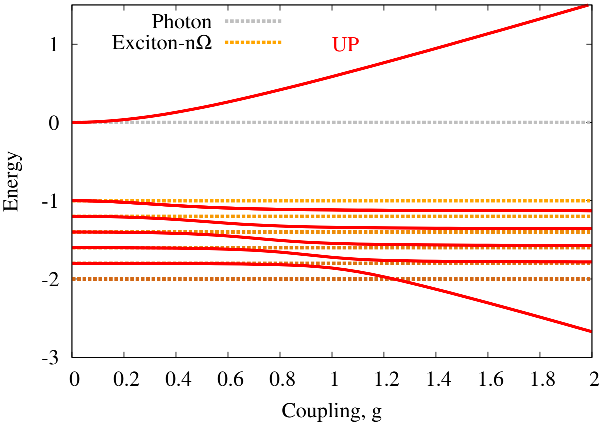

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