Polariton and photon condensates in organic materials

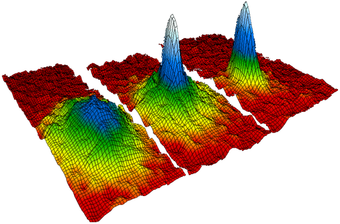
Jonathan Keeling

University of St Andrews
600 YEARS

LCN, March 2014

Coherent states of matter and light

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



[Anderson *et al.* Science '95]

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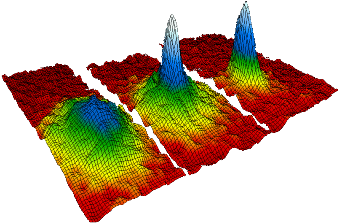

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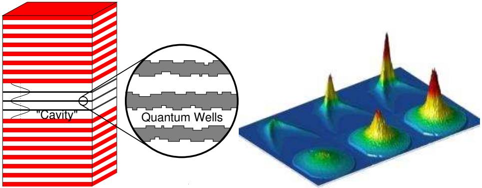
Polariton Condensate $T \sim 20$K



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

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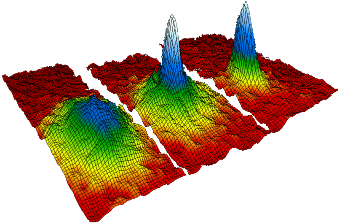

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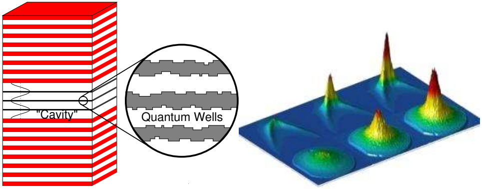
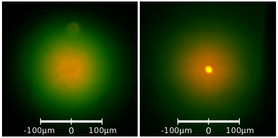


[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K



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

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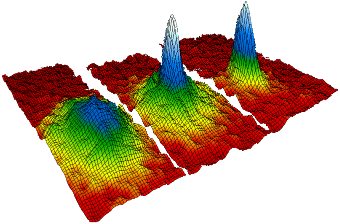

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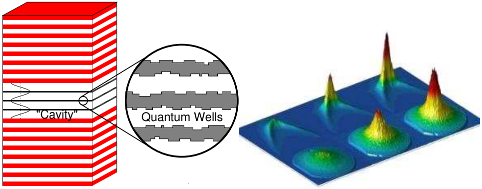

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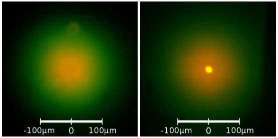
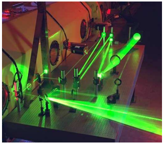


[Klaers *et al.* Nature, '10]

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



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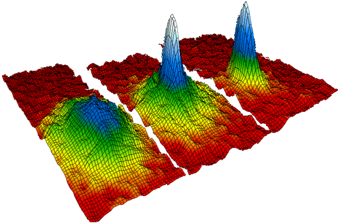

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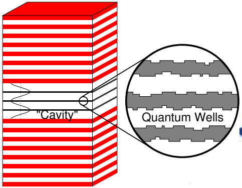

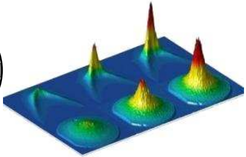

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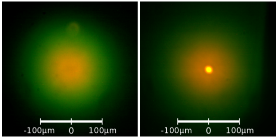

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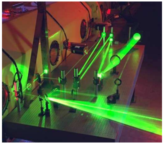
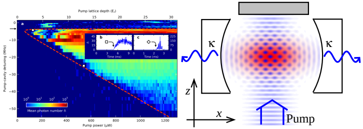


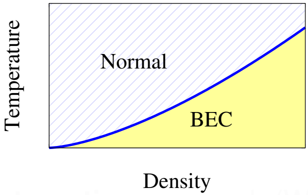
Superradiance transition
$T \sim 0$

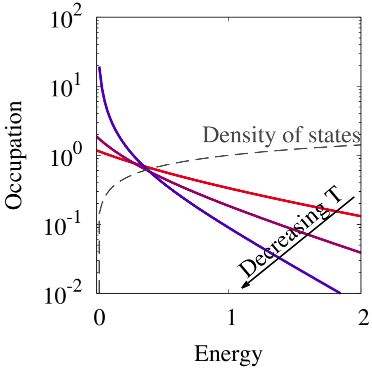


[Baumann *et al.* Nature, '10]

“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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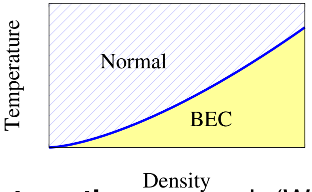

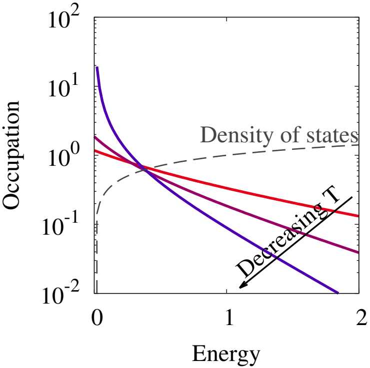

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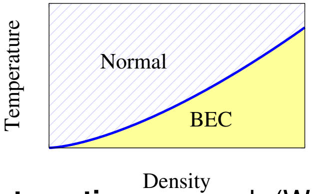

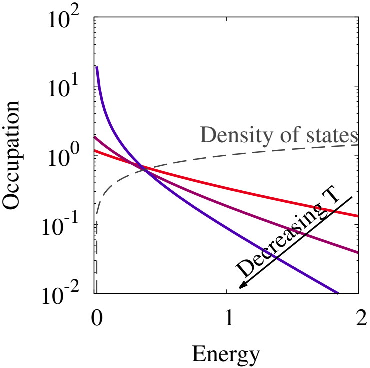

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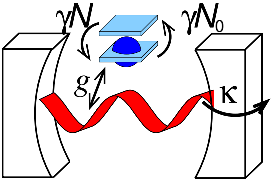
“Textbook” Laser: Maxwell Bloch equations

$$H = \omega_0 \psi^\dagger \psi + \sum_\alpha \epsilon_\alpha S^z_\alpha + g_{\alpha,\mathbf{k}} \left(\psi S^+_\alpha + \psi^\dagger S^-_\alpha \right)$$



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

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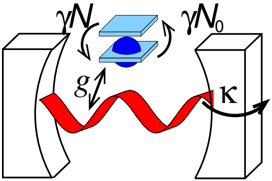

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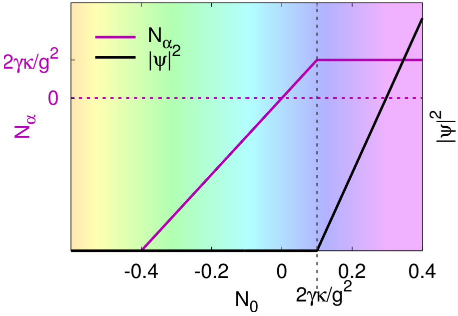
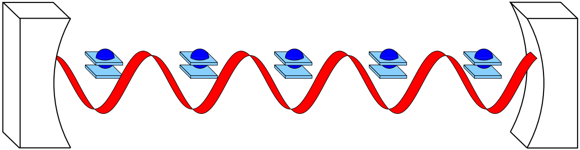


$|\psi|^2 > 0$ if $N_0 g^2 > 2\gamma\kappa$

“Textbook” Dicke-Hepp-Lieb superradiance



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

[Hepp, Lieb, Ann. Phys. ’73]

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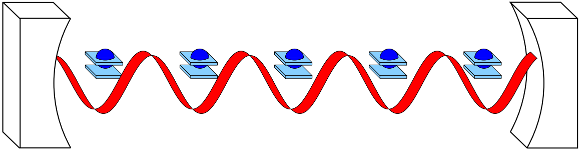

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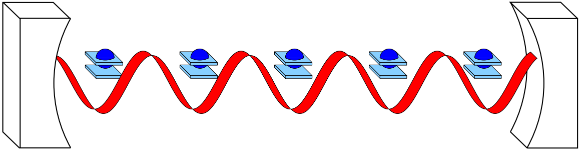

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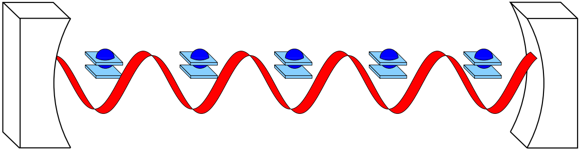

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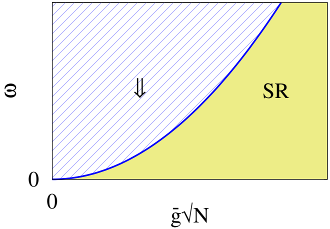

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Outline

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

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GROUP:

  

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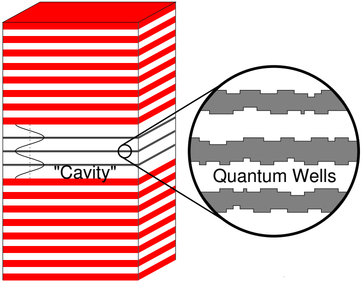
topnes — Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

EPSRC — Engineering and Physical Sciences Research Council

Microcavity polaritons



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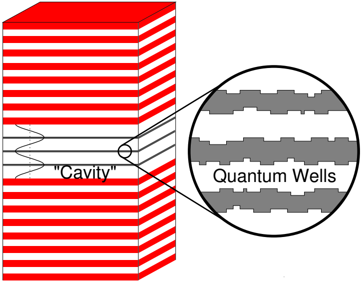
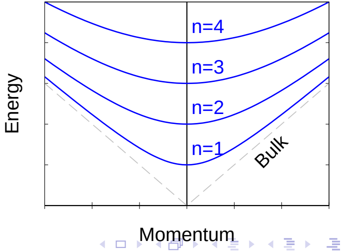


Cavity photons:

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



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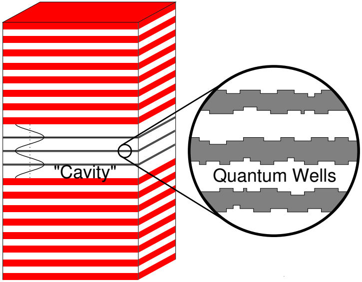

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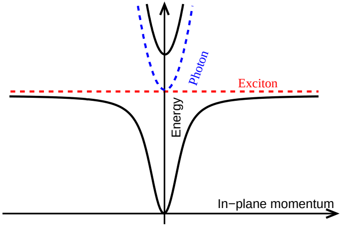

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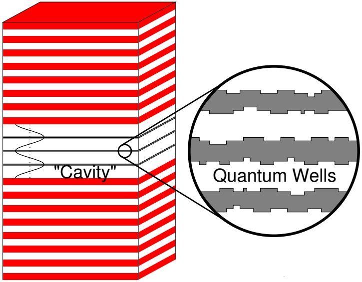

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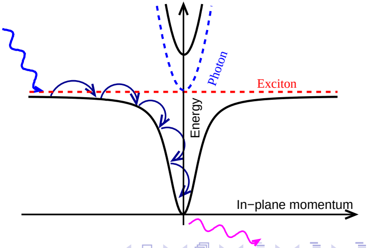

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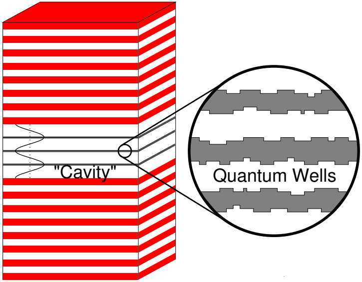

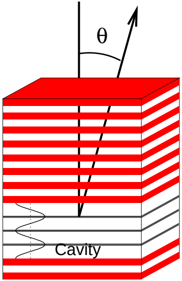

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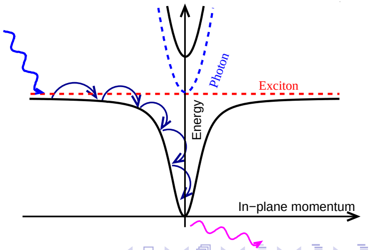
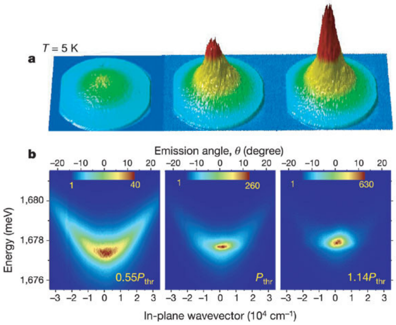
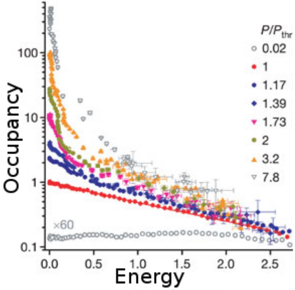


Polariton experiments: occupation and coherence





[Kasprzak, *et al.* Nature, ’06]

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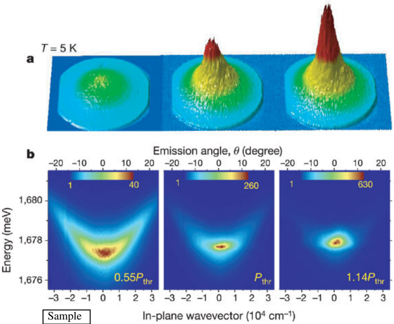
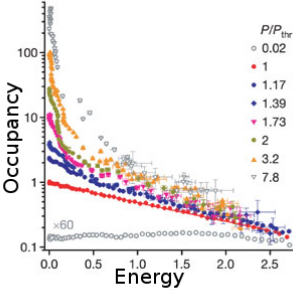
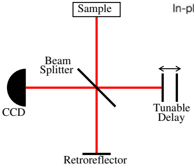
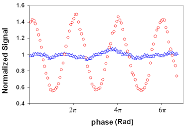
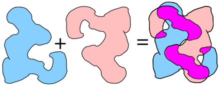


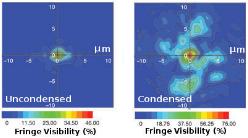






Coherence map:





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

Lasing-condensation crossover model

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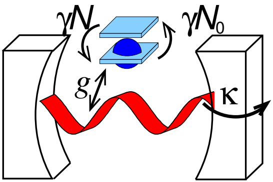

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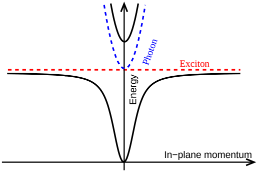

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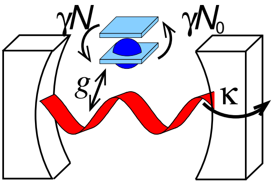

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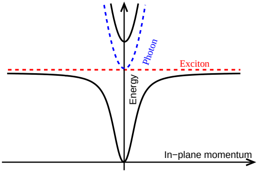

Dicke model:

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

Non-equilibrium condensation vs lasing

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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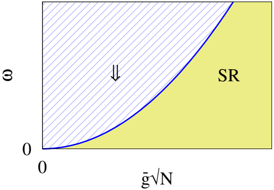
- Excitation cost E:
  $$(E-\omega)(E-\epsilon) = g^2 N$$
- Transition when $E = 0$



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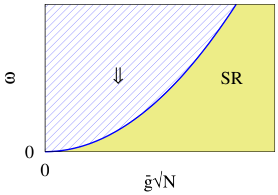

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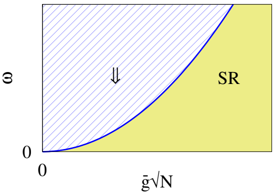

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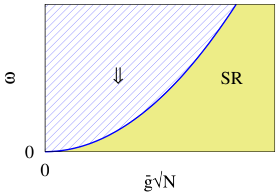

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Grand canonical ensemble

Grand canonical ensemble:

- If $H \to H - \mu(S^z + \psi^\dagger\psi)$, need only: $g^2 N > (\omega - \mu)|\epsilon - \mu|$
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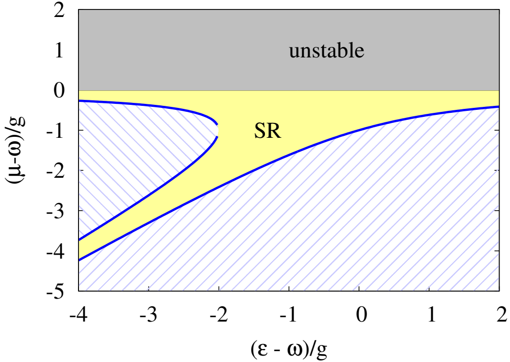

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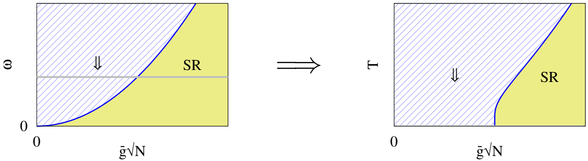
Grand canonical Dicke, finite temperature

- Finite temperature: $Ng^2 \tanh(\beta\epsilon/2) > \omega\epsilon$



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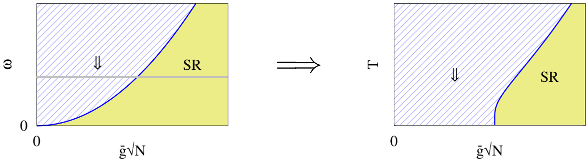

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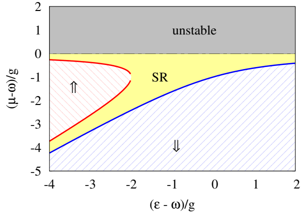

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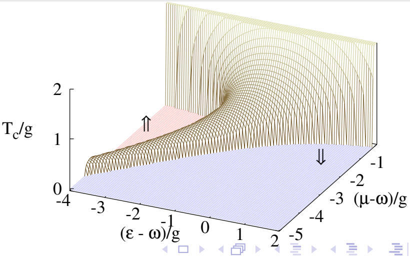

Polariton model and equilibrium results

- Localised excitons, propagating photons

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

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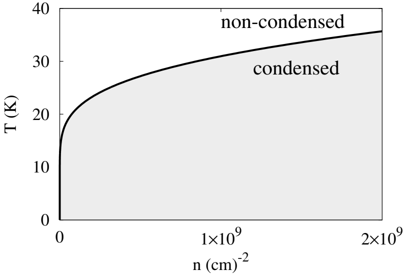

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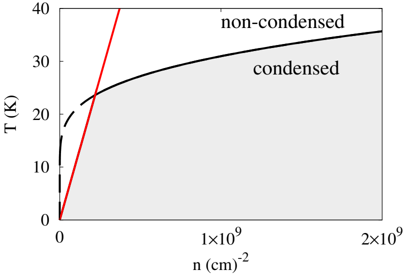

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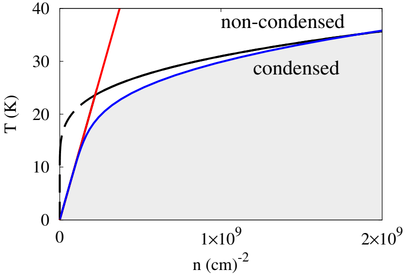

Simple Laser: Maxwell Bloch equations

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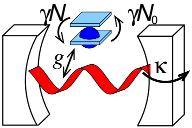

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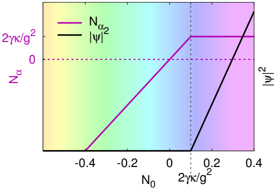

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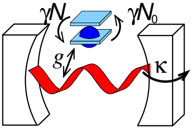

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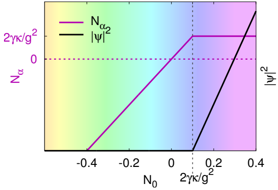

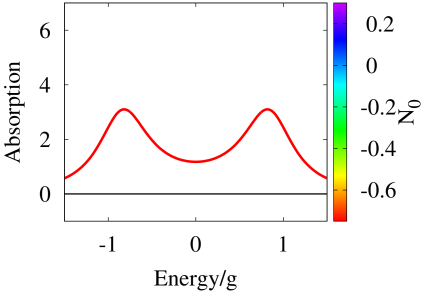

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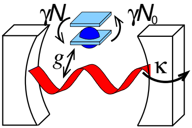

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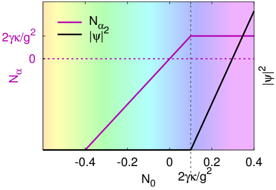

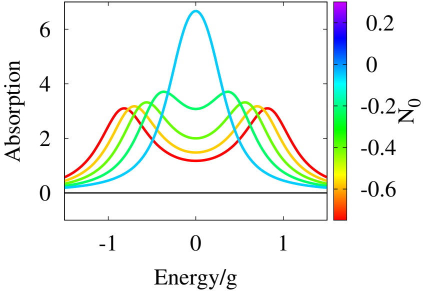

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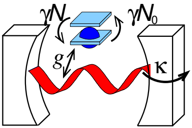

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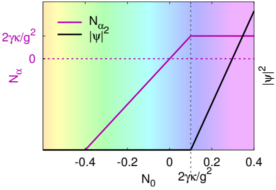

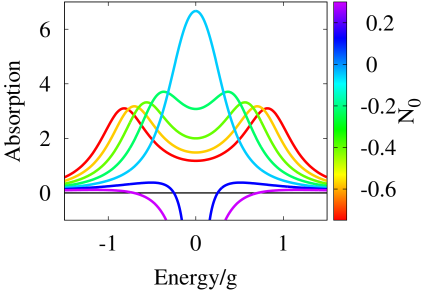

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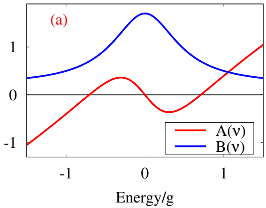

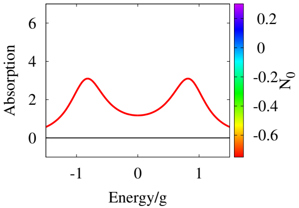

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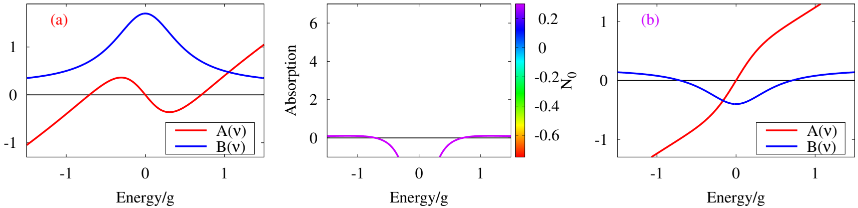

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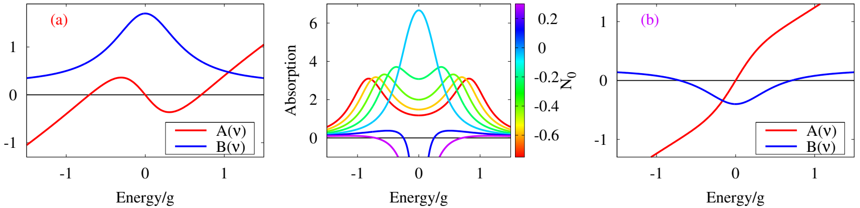
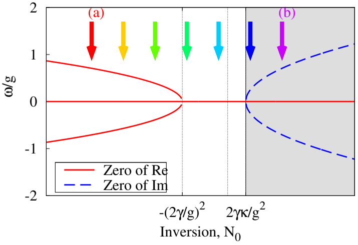


Laser:



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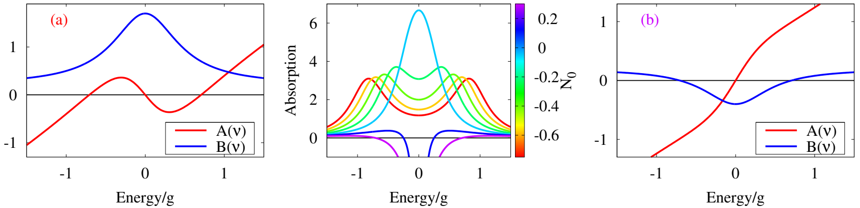

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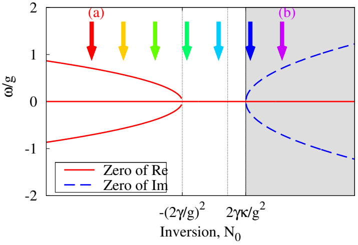

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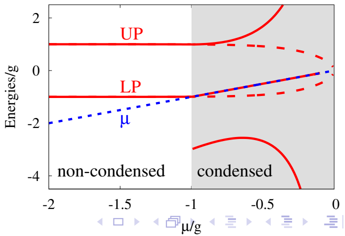
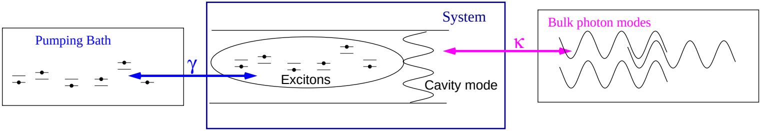
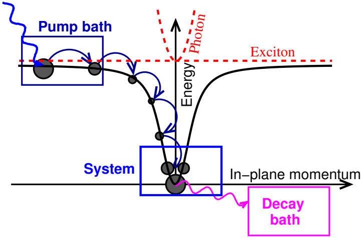


Non-equilibrium description: baths





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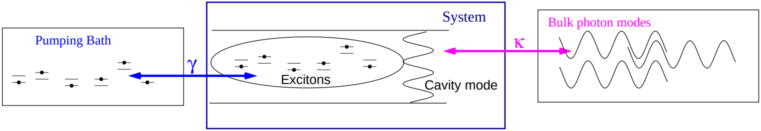

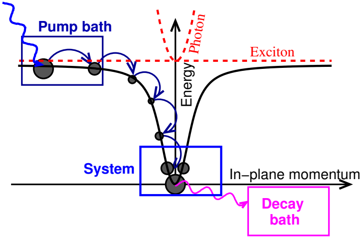

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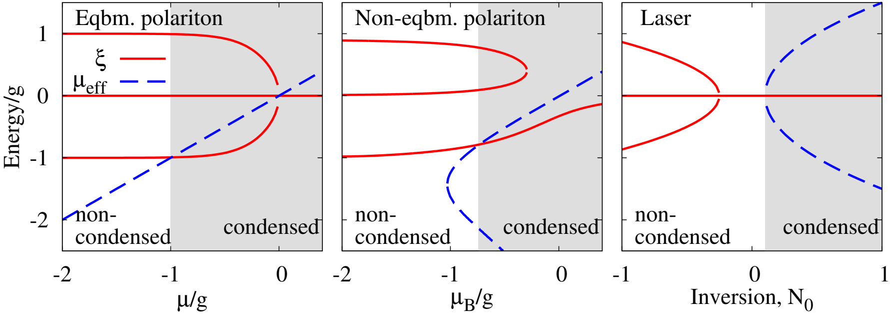
Strong coupling and lasing — low temperature phenomenon



- Inversionless
- **allows** strong coupling
- **requires** low $T \leftrightarrow$ condensation
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[Szymanska *et al.* PRL '06; Keeling *et al.* book chapter 1010.3338]

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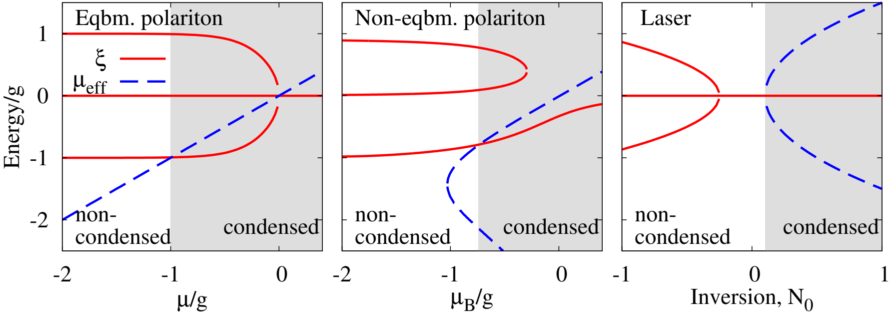

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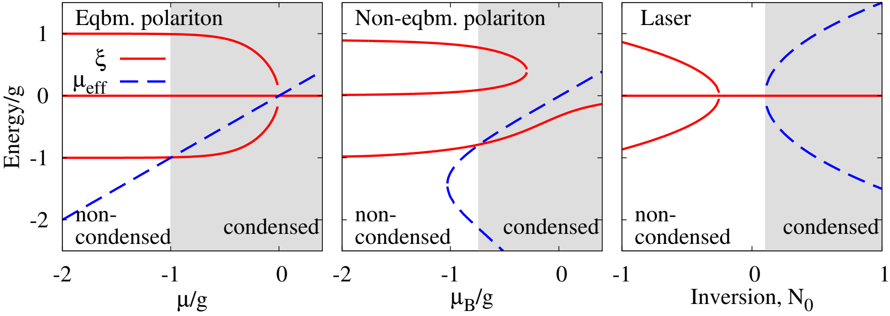

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Organic polaritons: photon-exciton-phonon coupling

1. Condensation, superradiance, lasing

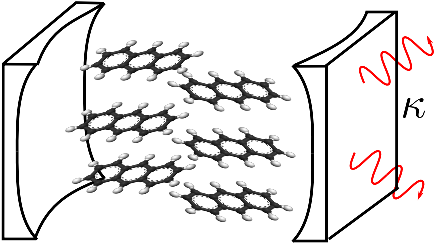
2. Polariton condensation and Dicke model
   - Condensation vs superradiance transition
   - Non-equilibrium condensation vs lasing

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

4. Room temperature condensates: Photons
   - Lasing model and thermalisation
   - Critical properties

Organic materials in microcavities

- What?

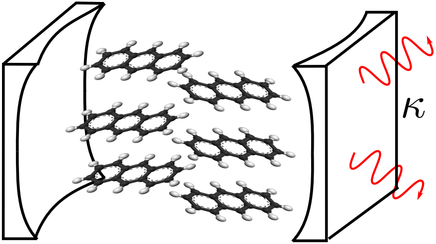


- Why?
- Lasing threshold at room T

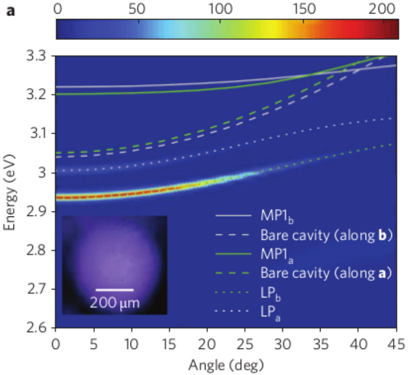
[Kena Cohen and Forrest, Nat. Photon '10; Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14]

Organic materials in microcavities

- What?



- Why?



Polariton splitting: 0.1eV $\leftrightarrow$ 1000K.

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

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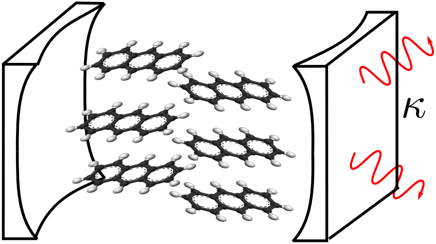
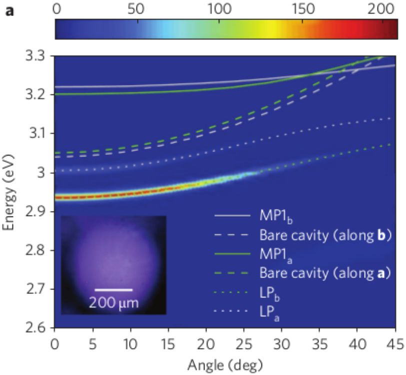


- Lasing threshold at room T

- Why?



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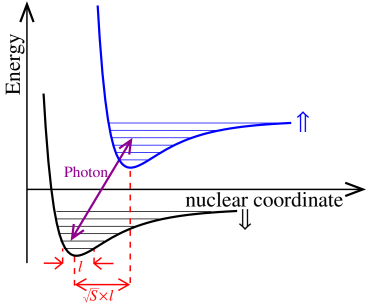
Dicke Holstein Model



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

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

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

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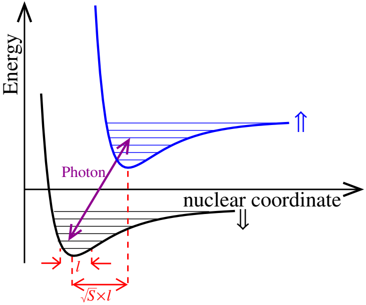

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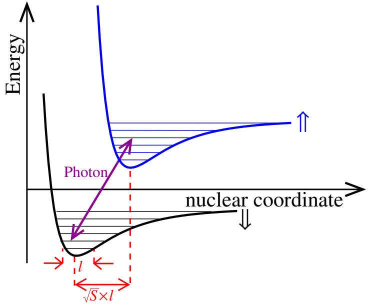

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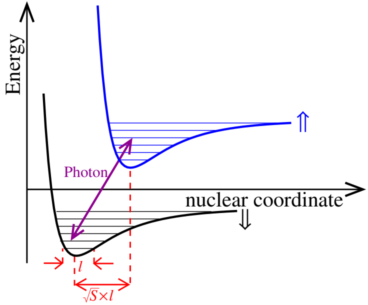

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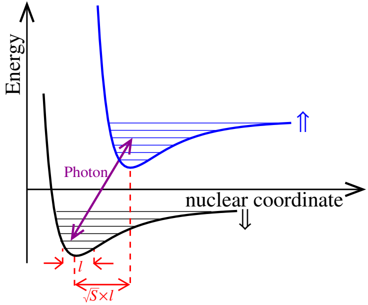

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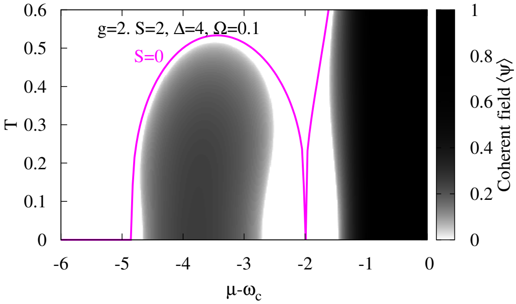

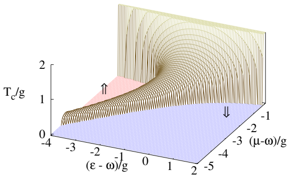

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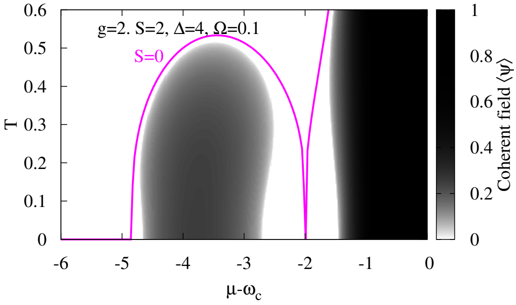

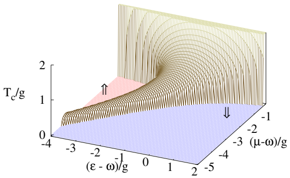

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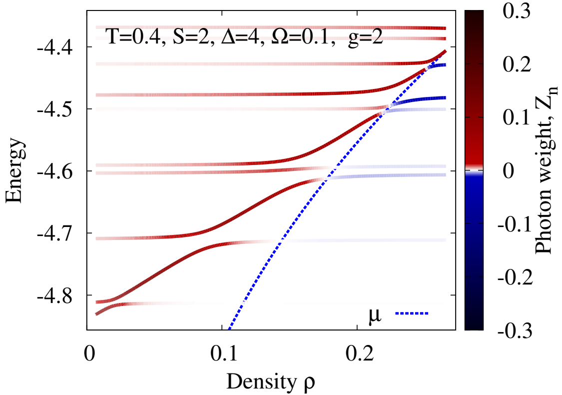
Polariton spectrum: photon weight



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- $\mathcal{D}(t) = -i\langle\psi^\dagger(t)\psi(0)\rangle, \qquad \mathcal{D}(\omega) = \sum_n \frac{Z_n}{\omega - \omega_n}$

[Cwik *et al.* EPL '14]

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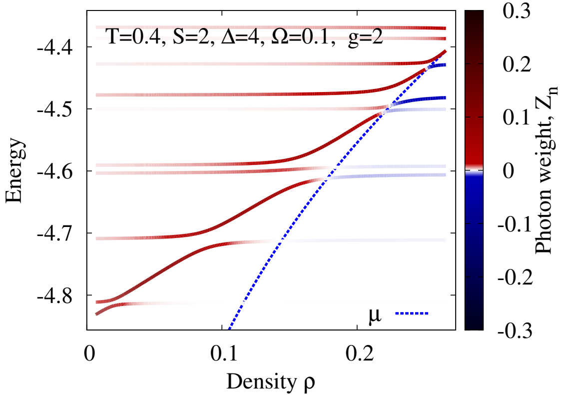

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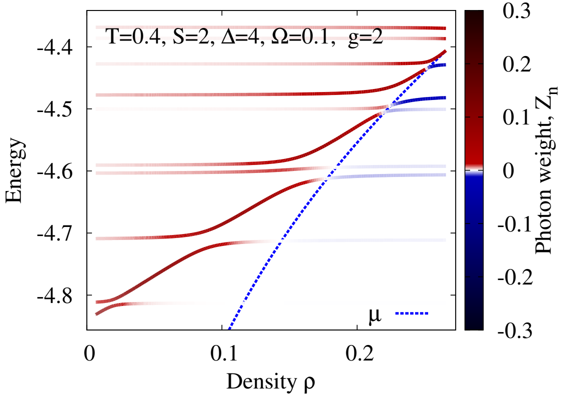

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Polariton spectrum: what condensed

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

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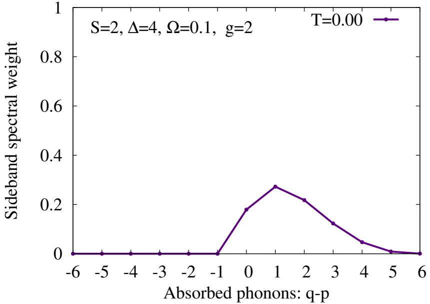

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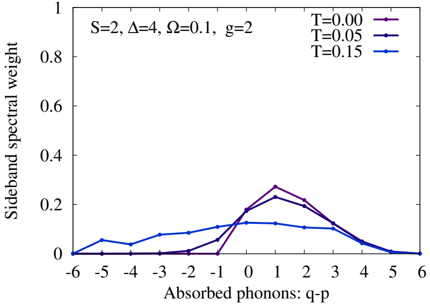

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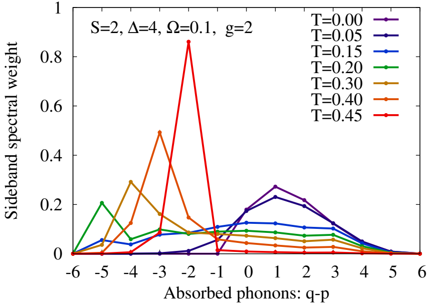

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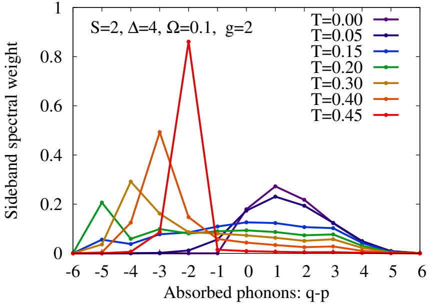

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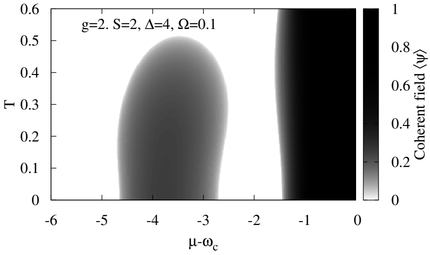

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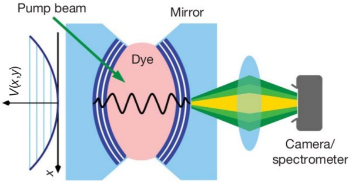
Polariton and photon Condensation

Photon BEC experiments



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- No strong coupling

[Klaers et al, Nature, 2010]

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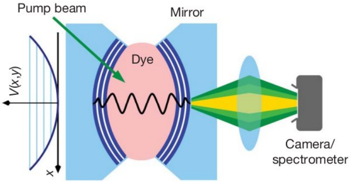

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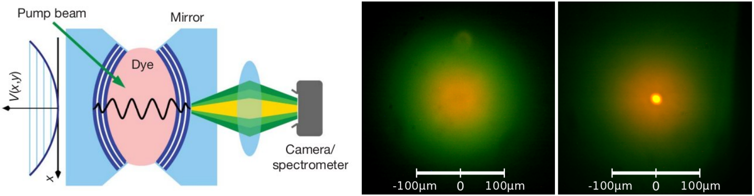

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Relation to dye laser

- No electronic inversion
- No strong coupling

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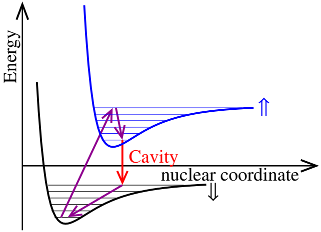
4 Level Dye Laser



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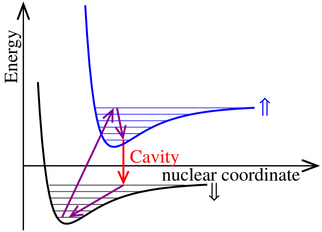

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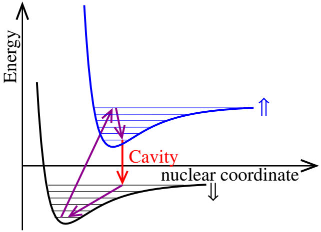

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- 2D harmonic cavity
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
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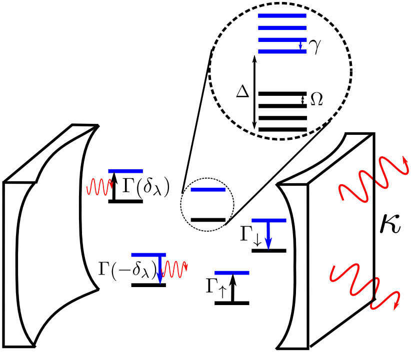

Modelling

Rate equation

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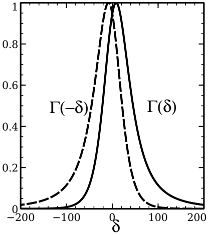

- $\Gamma(+\delta) \simeq \Gamma(-\delta)e^{\beta\delta}$
- $\Gamma \to 0$ at large δ

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

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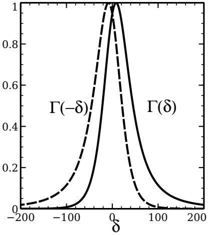

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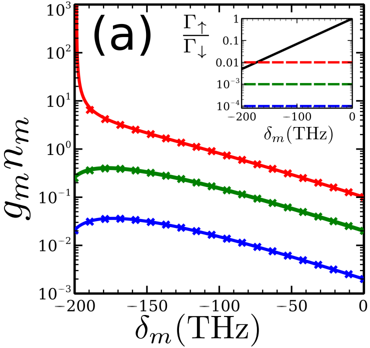

Low loss: Thermal

[Kirton & JK PRL '13]

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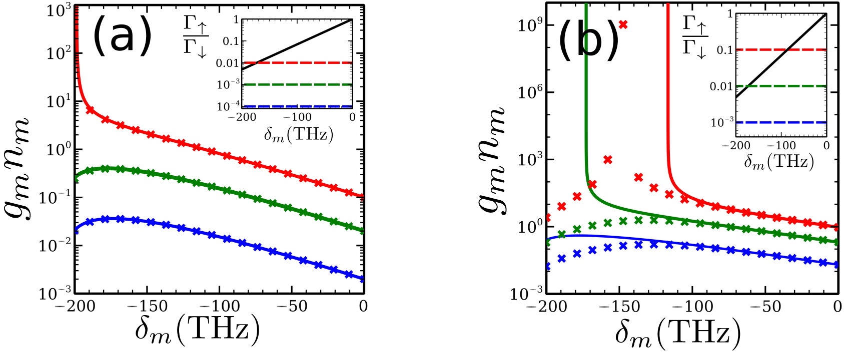
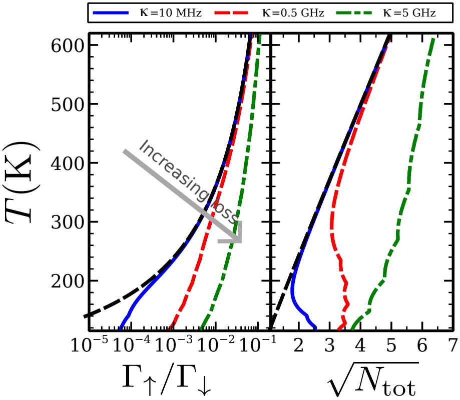


Low loss: Thermal

High loss → Laser

[Kirton & JK PRL '13]

Threshold condition



Compare threshold:

- 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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[Kirton & JK PRL '13]

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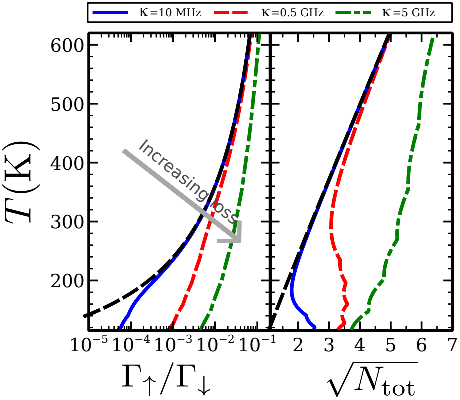

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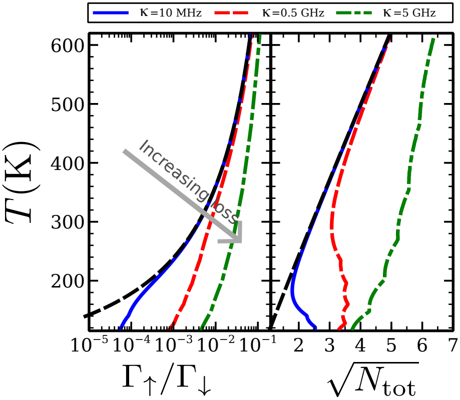

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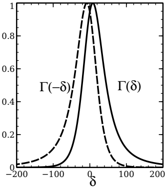

[Kirton & JK PRL '13]

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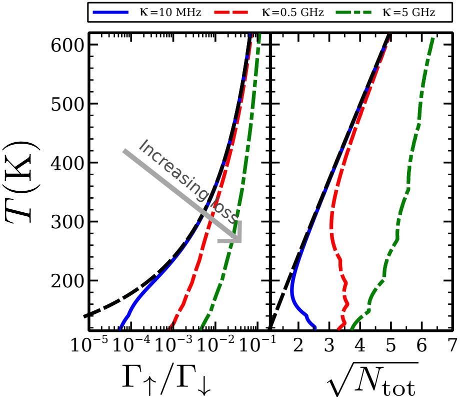

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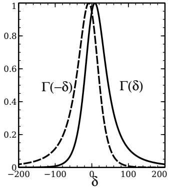
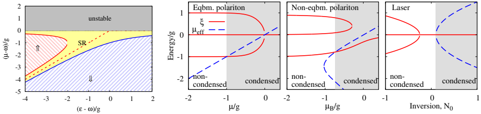


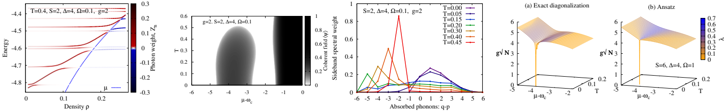
[Kirton & JK PRL '13]

Summary

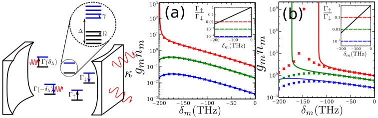
- Polariton condensation vs lasing; superradiance



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



- Photon condensation and thermalisation



Extra slides

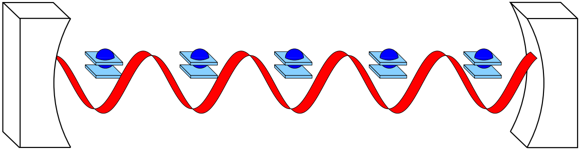
5. No go theorem

6. Retarded Green's function for laser

7. Organic properties
   - Ultra-strong phonon coupling?

8. Anticrossing vs ρ

No go theorem and transition



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

[Rzazewski *et al* PRL '75]

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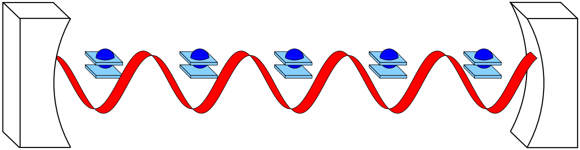

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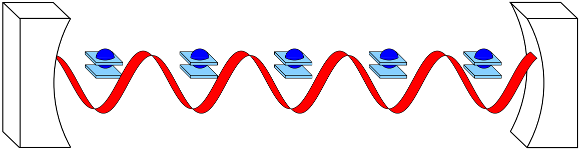

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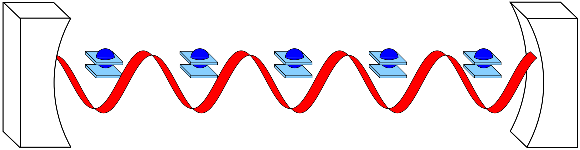

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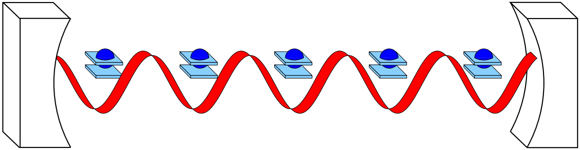

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But Thomas-Reiche-Kuhn sum rule states: $g^2/\epsilon < 2\zeta$. **No transition**

[Rzazewski *et al* PRL '75]

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:
  - If $H \to H - \mu(S^z + \psi^\dagger\psi)$, need only: $g^2 N > (\omega - \mu)(\omega_0 - \mu)$
  - Incoherent pumping — polariton condensation.
- Dissociate g, ω_0,
  e.g. Raman scheme: $\omega_0 \ll \omega$.
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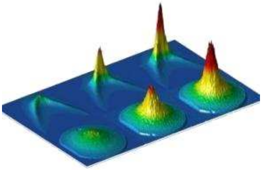

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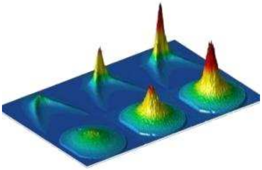

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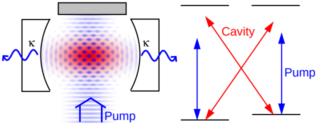
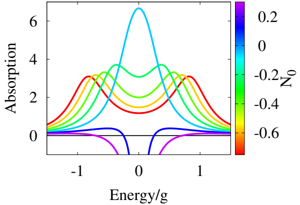


Maxwell-Bloch Equations: Retarded Green's function



- Introduce $D^R(\omega)$:
  Response to perturbation

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

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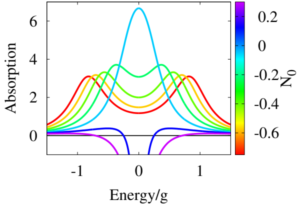

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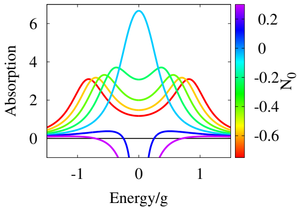

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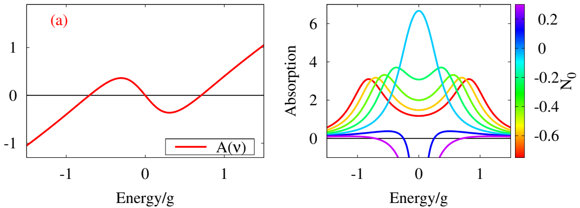

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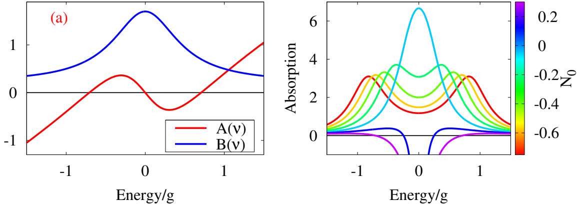

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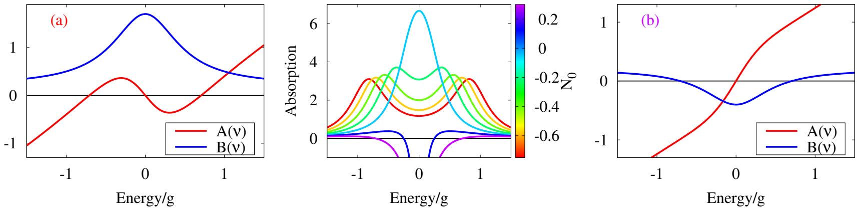

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Organic materials in microcavities

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

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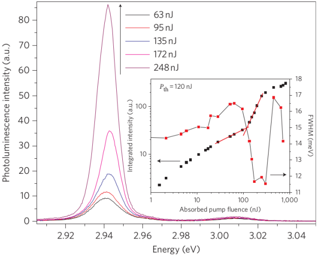

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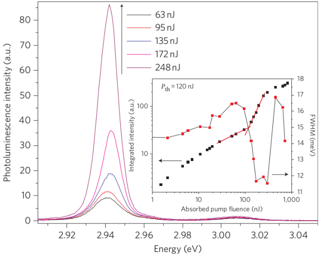

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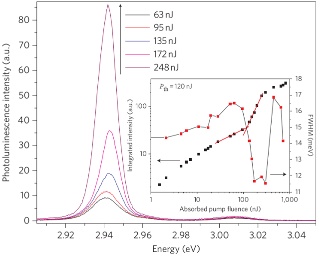

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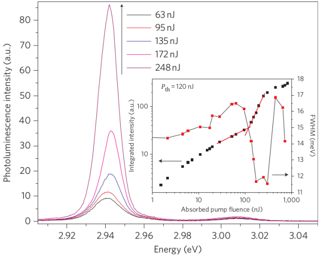

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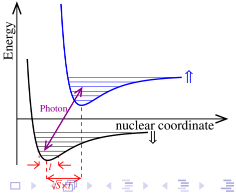


Organic polaritons

5 No go theorem

6 Retarded Green's function for laser

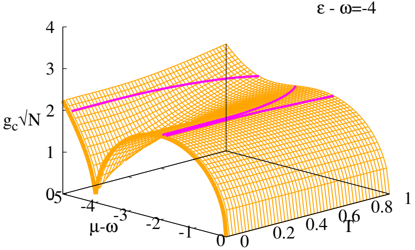
7 Organic properties

- Ultra-strong phonon coupling?

8 Anticrossing vs ρ

Critical coupling with increasing S

- Re-orient phase diagram
- *g* vs μ, *T*
- Colors → Jump of $\langle\psi\rangle$



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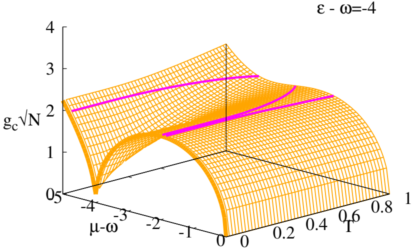

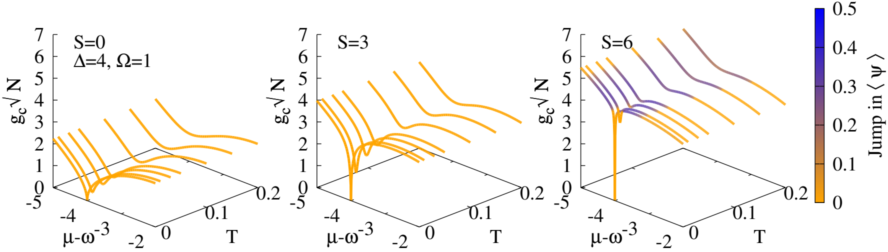

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

- Optimal phonon displacements, $\sim \sqrt{S}$
- Reduced $g_{\text{eff}} \sim g \times \exp(-S/2)$
- For $\psi \neq 0$, competition
  Variational MFT $|\psi\rangle_\alpha \sim \exp(-\eta K_\alpha - \zeta b^\dagger_\alpha)|0, S\rangle_\alpha$

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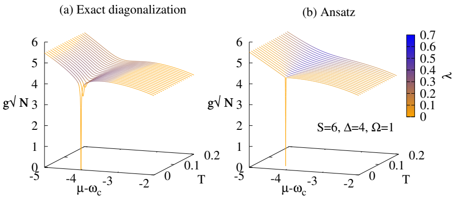
Collective polaron formation

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



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

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

Effective 2LS energy in field:

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

[Cwik *et al.* EPL '14]

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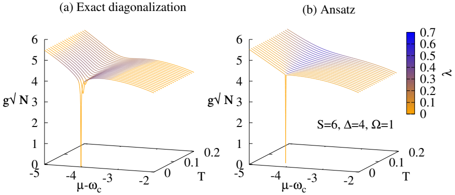

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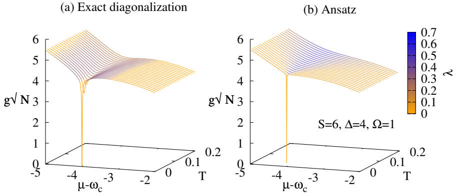

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- Feedback: Large/small $g_{\mathrm{eff}} \leftrightarrow \lambda = \langle \psi \rangle$
- Variational free energy

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

Effective 2LS energy in field:

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

[Cwik *et al.* EPL ’14]

Polariton spectrum — coupled oscillators

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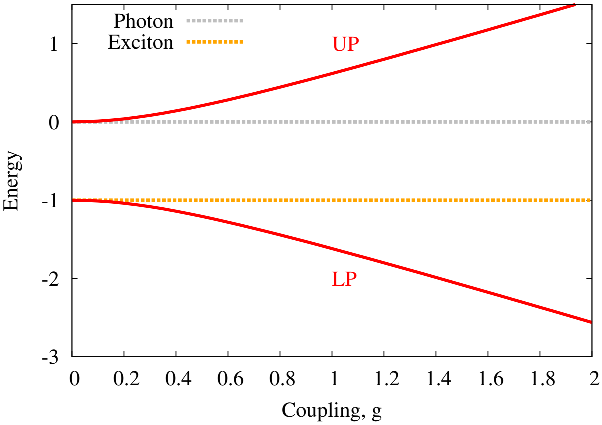
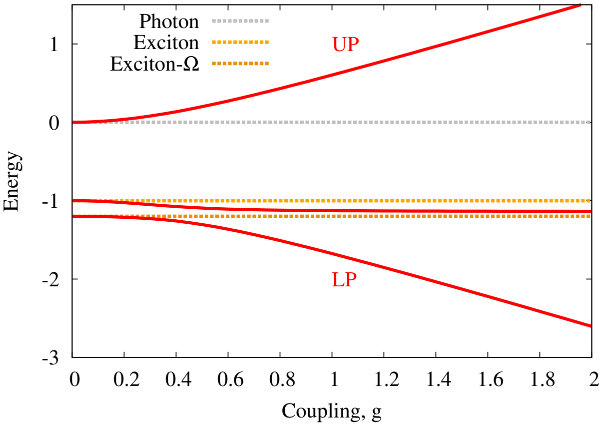
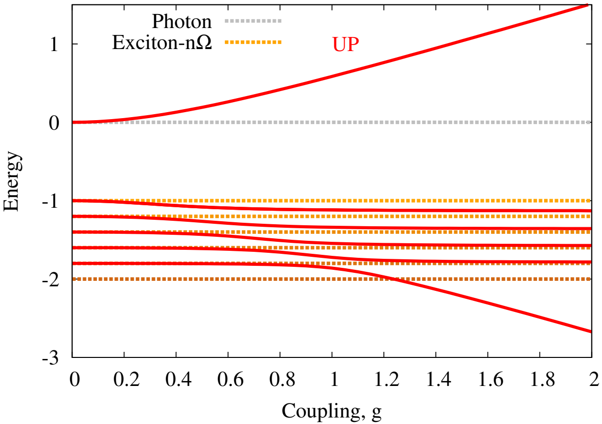


Polariton spectrum — coupled oscillators



Polariton spectrum — coupled oscillators



Polariton spectrum — coupled oscillators