Polariton and photon condensates with organic molecules

Jonathan Keeling

University of St Andrews 600 YEARS

SUPA

Telluride, July 2013

Polariton and photon condensates with organic molecules:

The “Dicke-Holstein” model

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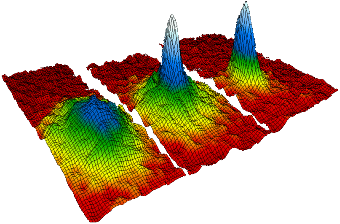
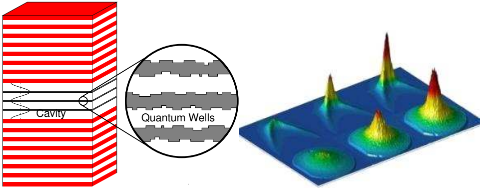
Coherent states of matter and light

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



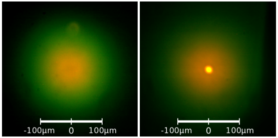
[Anderson *et al.* Science '95]

Polariton Condensate $T \sim 20$K



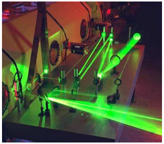
[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K

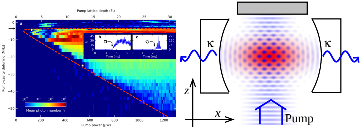


[Klaers *et al.* Nature, '10]

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



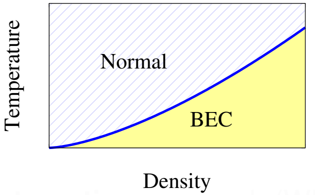
Superradiance transition
$T \sim 0$

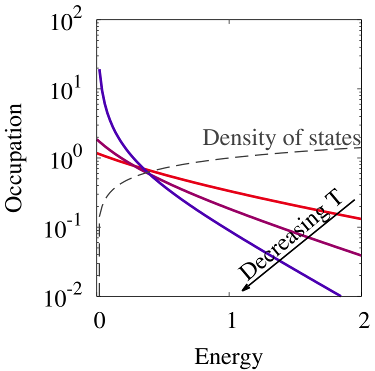


[Baumann *et al.* Nature, '10]

“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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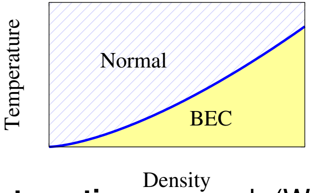

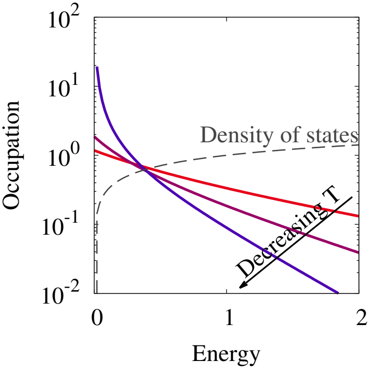

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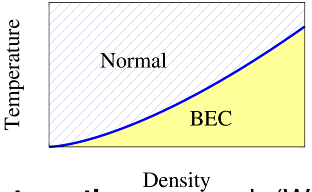

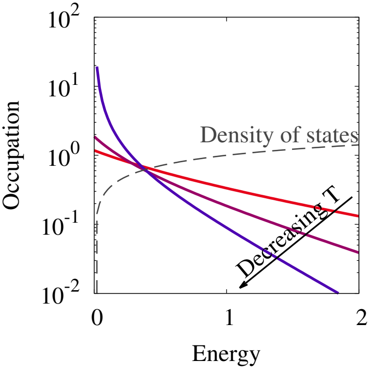

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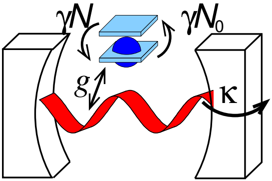
“Textbook” Laser: Maxwell Bloch equations

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



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

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

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

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

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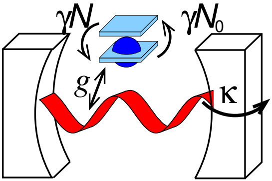

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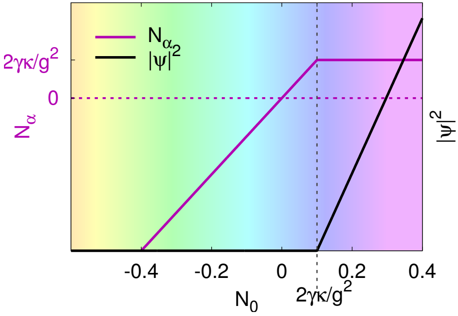
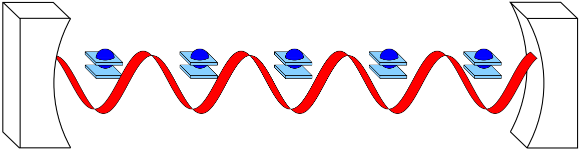


$|\psi|^2 > 0$ if $N_0 g^2 > 2\gamma\kappa$

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

[Hepp, Lieb, Ann. Phys. ’73]

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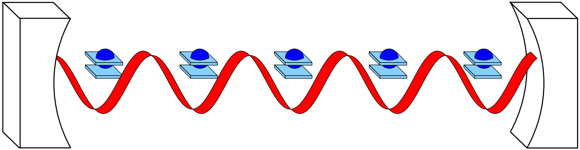

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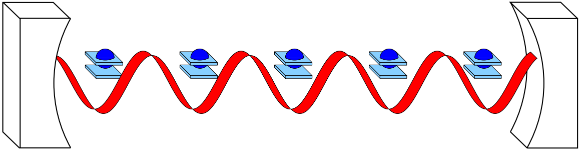

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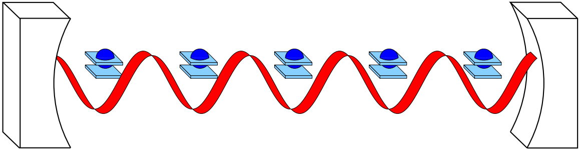

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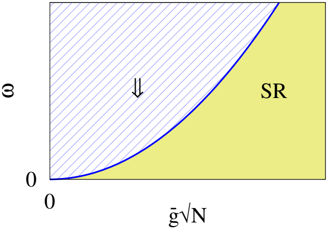

[Hepp, Lieb, Ann. Phys. ’73]

Outline

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

Acknowledgements

GROUP:




COLLABORATORS: Szymanska (Warwick), Reja (Cam.), Littlewood (ANL)

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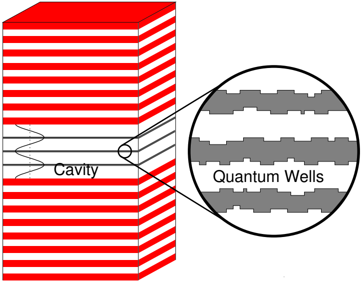
Microcavity polaritons



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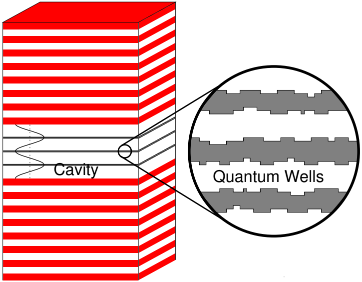
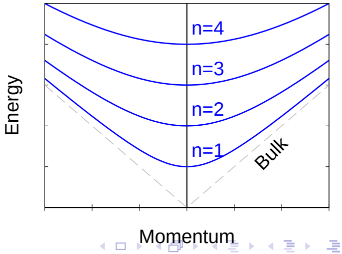


Cavity photons:

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



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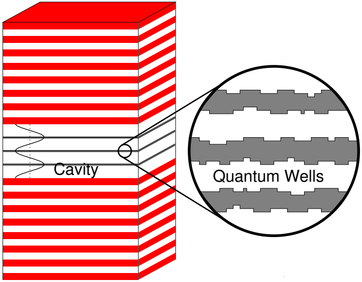

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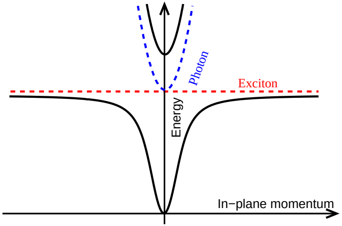

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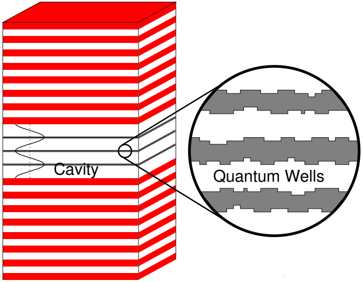

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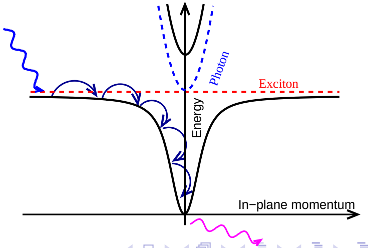

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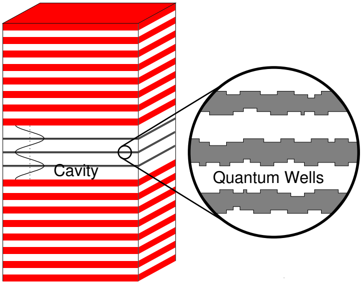

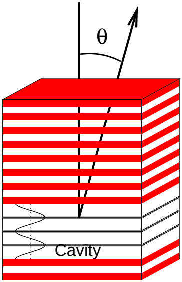

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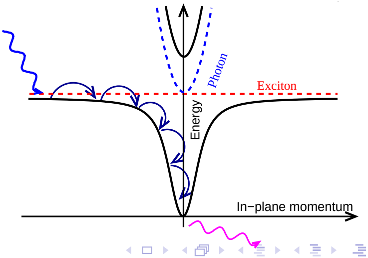
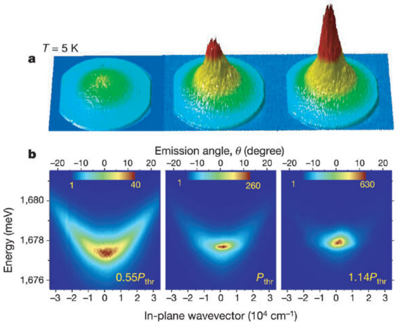
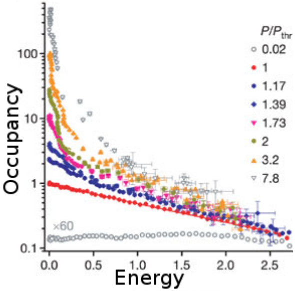


Polariton experiments: occupation and coherence





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

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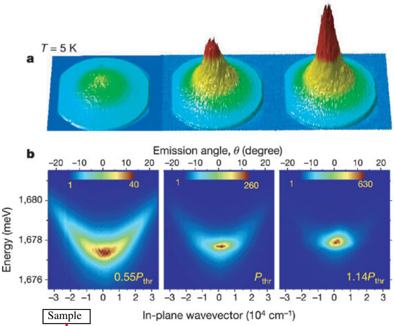
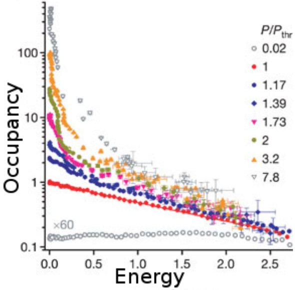
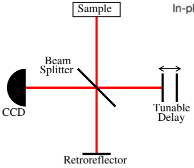
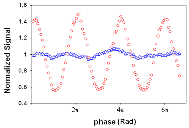
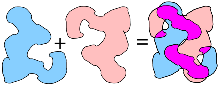


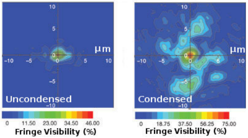






Coherence map:

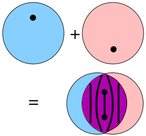


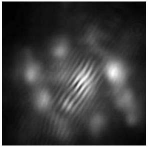


[Kasprzak, *et al.* Nature, ’06]

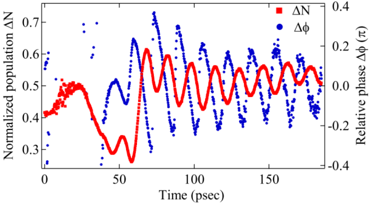
(Some) other polariton condensation experiments

- Quantised vortices
  [**Lagoudakis *et al.* Nat. Phys. '08**. Science '09, PRL '10; Sanvitto *et al.* Nat. Phys. '10; Roumpos *et al.* Nat. Phys. '10]

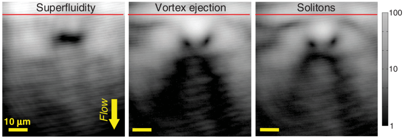




- Josephson oscillations
  [**Lagoudakis *et al.* PRL '10**]



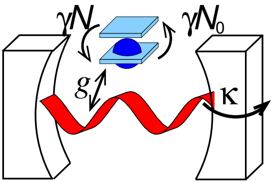
- Pattern formation/Hydrodynamics
  [**Amo *et al.* Science '11**, Nature '09; Wertz *et al.* Nat. Phys '10]



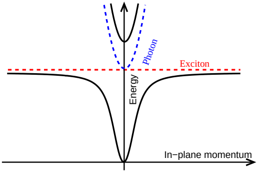
Non-equilibrium condensation vs lasing

Lasing-condensation crossover model

- Use model that can show lasing and condensation:



$\Leftrightarrow$



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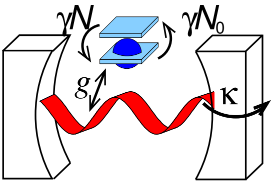

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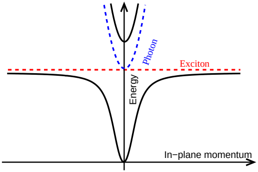


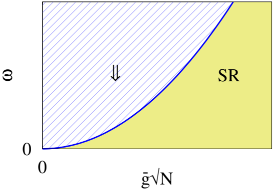
Dicke model:

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

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: $Ng^2 > \omega\epsilon$



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

- 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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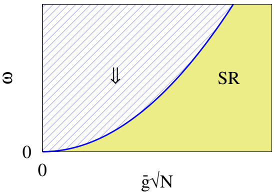

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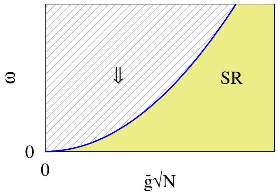


[Hepp, Lieb, Ann. Phys. '73]

Dicke-Hepp-Lieb superradiance and modes

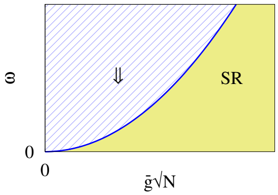
$$H = \omega\psi^\dagger\psi + \epsilon S^z + g\left(\psi^\dagger S^- + \psi S^+\right)$$

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

- Normal state, $S^z = -N/2 + B^\dagger B$
  $H = \omega\psi^\dagger\psi + \epsilon B^\dagger B + g\sqrt{N}\left(\psi^\dagger B + \psi B^\dagger\right)$
- Excitation cost E:

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

- Transition when $E = 0$



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

Grand canonical ensemble

Grand canonical ensemble:

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

- Transition at: $g^2N > (\omega - \mu)(\epsilon - \mu)$
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[Eastham and Littlewood, PRB '01]

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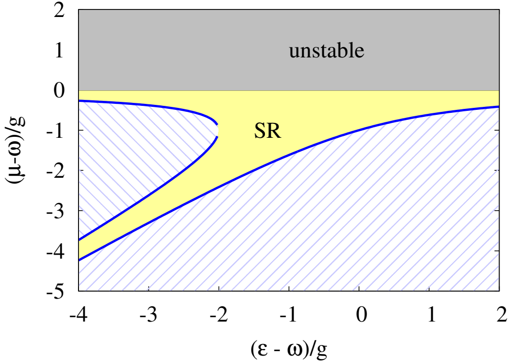

[Eastham and Littlewood, PRB '01]

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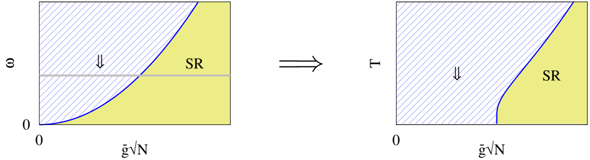
Grand canonical Dicke, finite temperature

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



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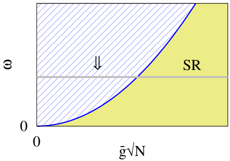
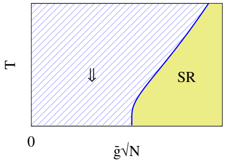


$\Longrightarrow$



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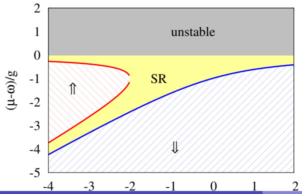

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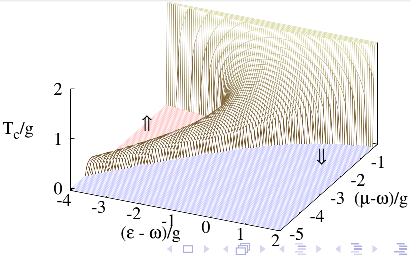

Polariton model and equilibrium results

Localised excitons, propagating photons

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

Self-consistent polarisation and field

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

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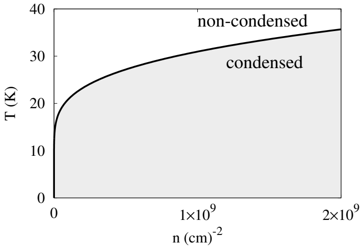

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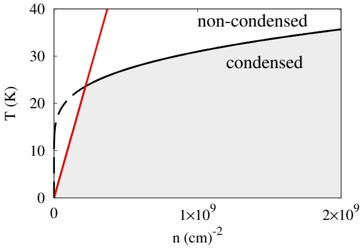

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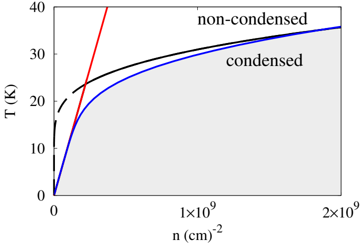

Simple Laser: Maxwell Bloch equations

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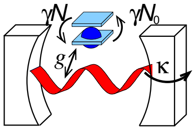

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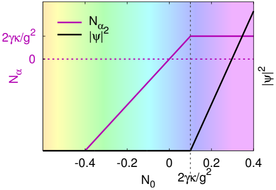

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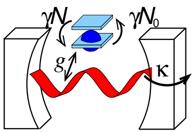

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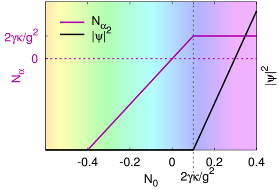

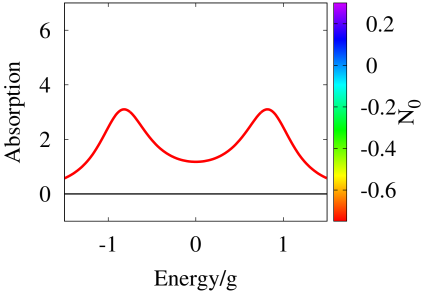

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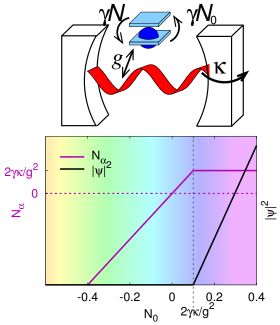

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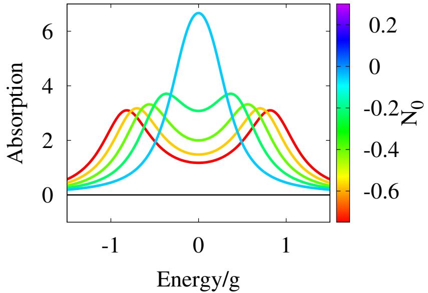

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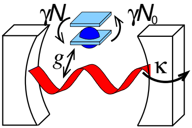

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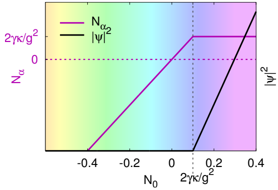

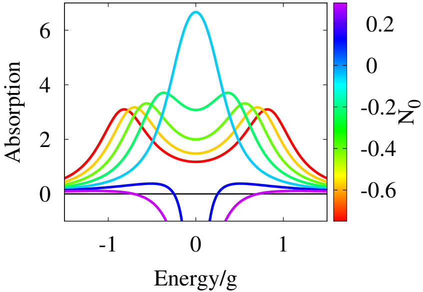

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Poles of Retarded Green's function and gain

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

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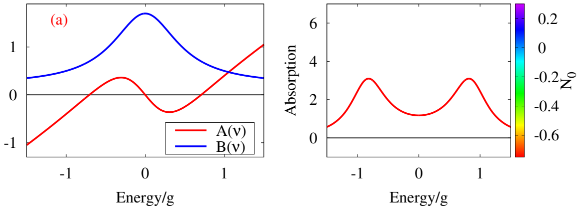

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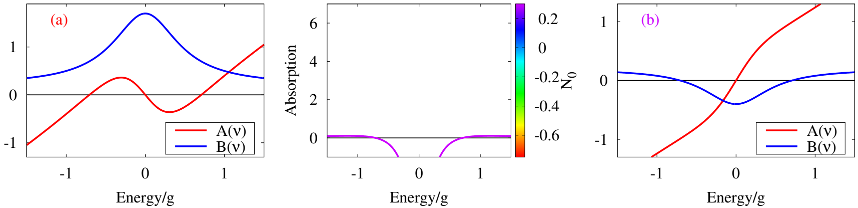

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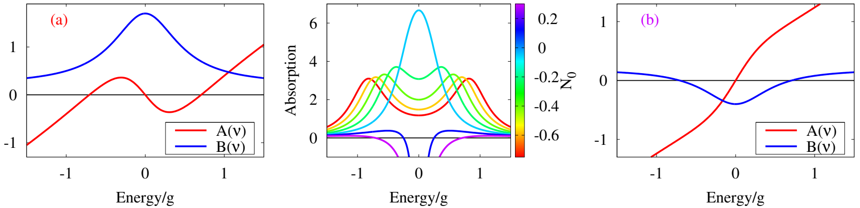
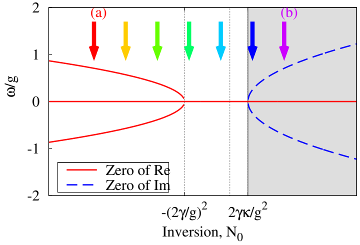


Laser:



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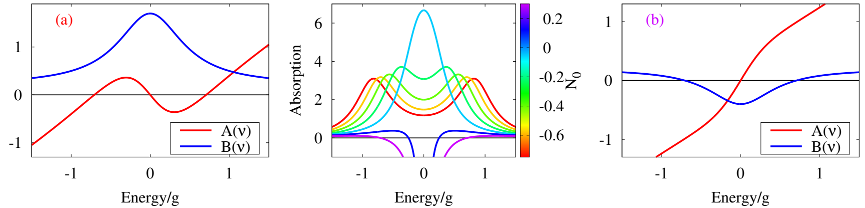

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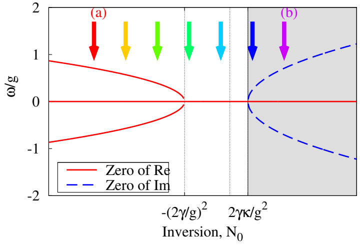
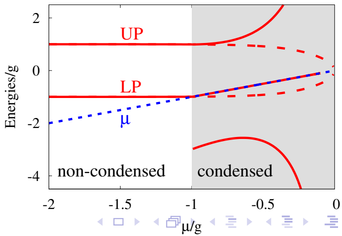
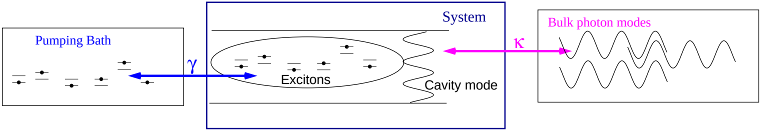
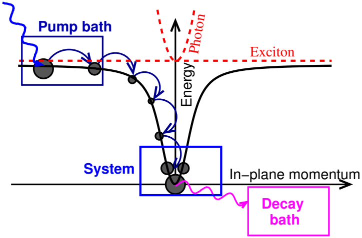


Equilibrium:



Non-equilibrium description: baths





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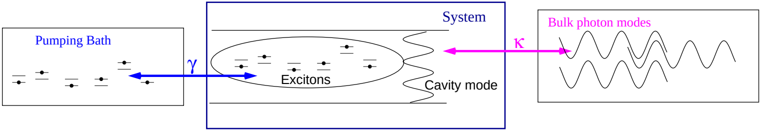

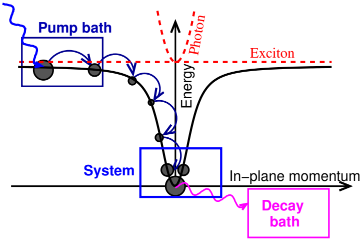

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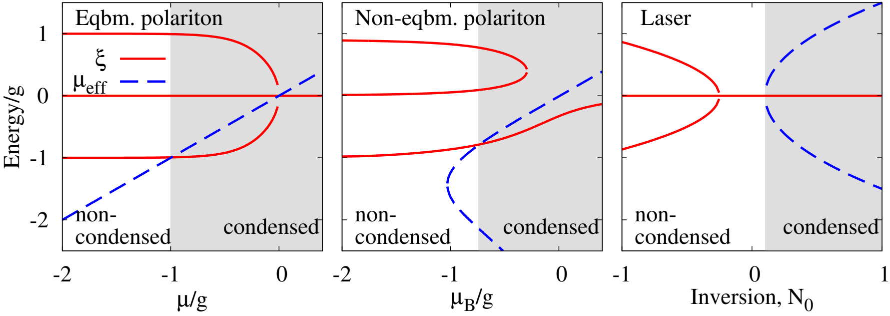
Strong coupling and lasing — low temperature phenomenon



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

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

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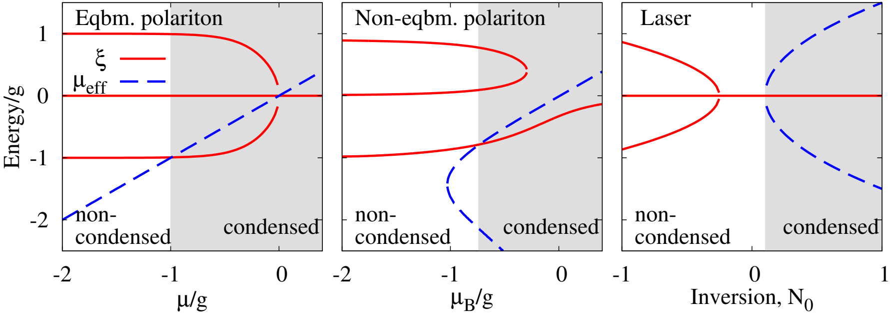

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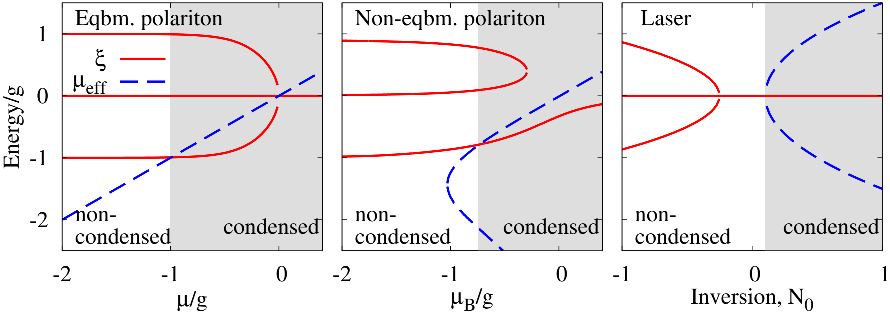

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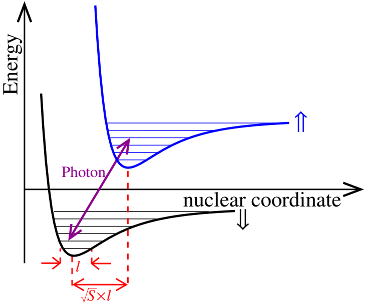
Organic polaritons: photon-exciton-phonon coupling

Dicke Holstein Model



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$$\left. + \Omega\left\{b^\dagger_\alpha b_\alpha + \sqrt{S}\left(b^\dagger_\alpha + b_\alpha\right) S^z_\alpha\right\}\right]$$

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

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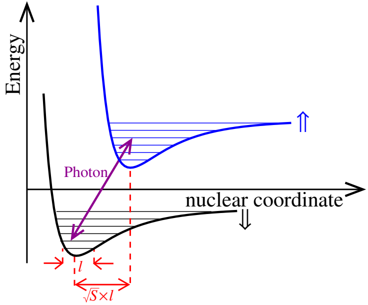

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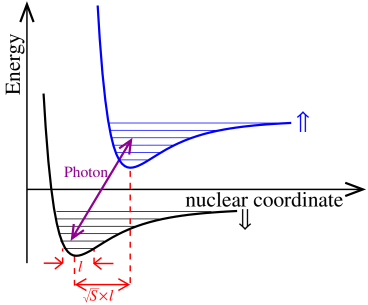

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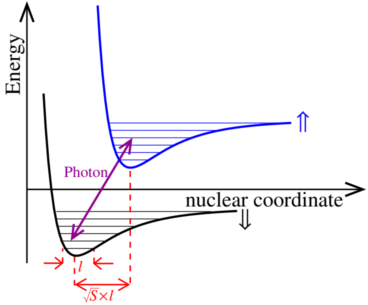

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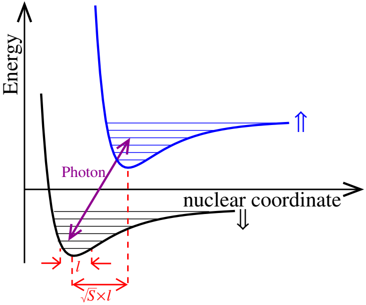

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Phase diagram

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

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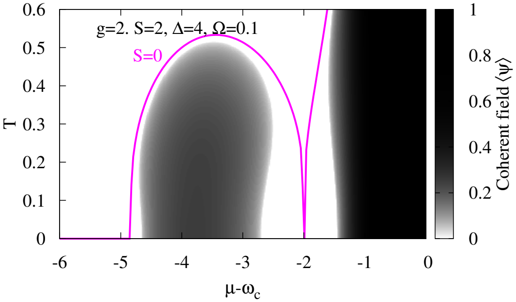

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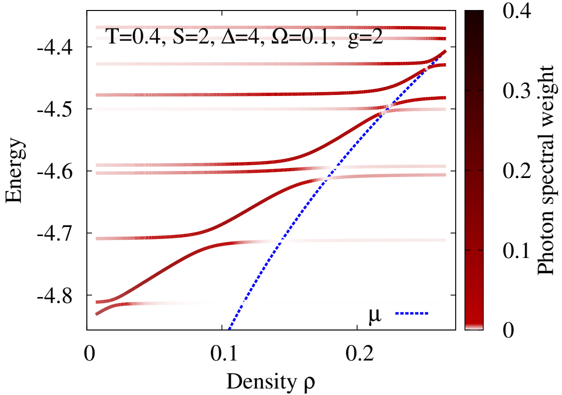
Polariton spectrum: photon weight



- Saturating 2LS: $g_{\text{eff}}^2 \sim g^2(1-2\rho)$
- What is nature of polariton mode?
- $\mathcal{D}(t) = -i\langle\psi^\dagger(t)\psi(0)\rangle, \qquad \mathcal{D}(\omega) = \sum_n \frac{Z_n}{\omega - \omega_n}$

[Cwik *et al.* arXiv:1303.3702]

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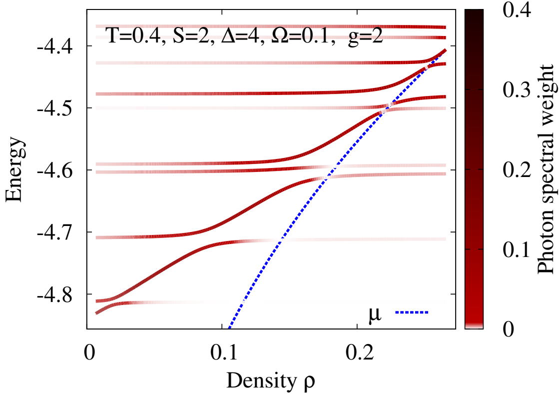

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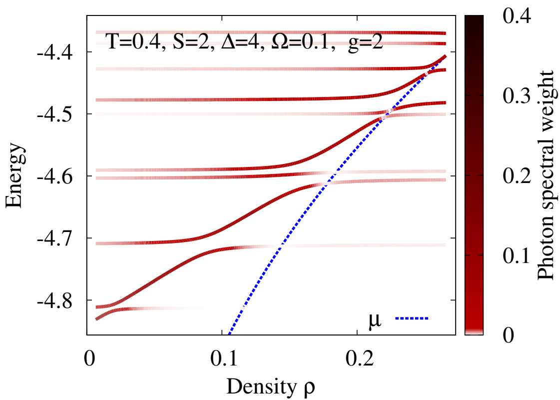

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- Eigenvector that is macroscopically occupied
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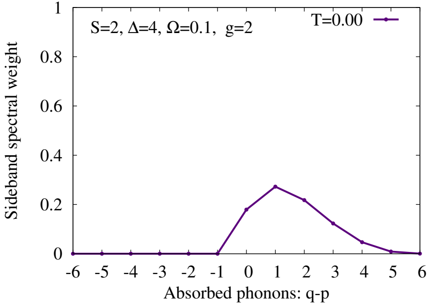

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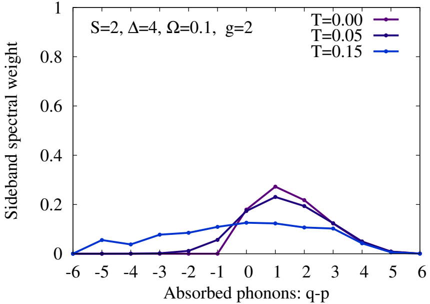

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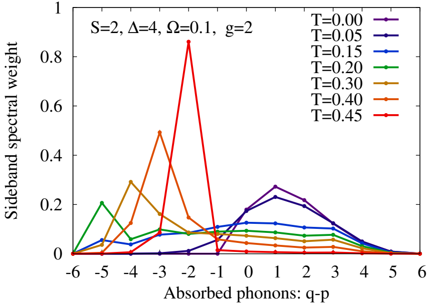

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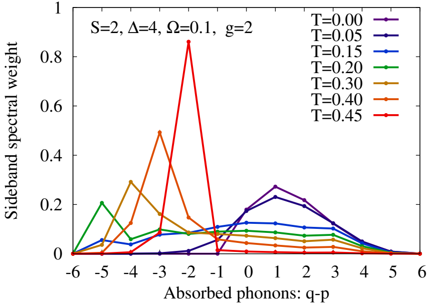

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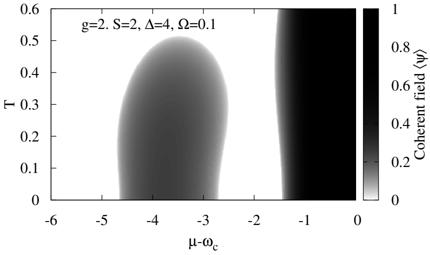

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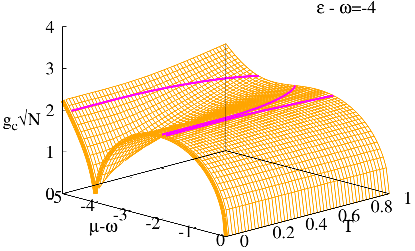
Organic polaritons

Critical coupling with increasing S

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



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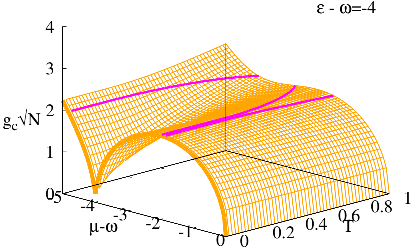

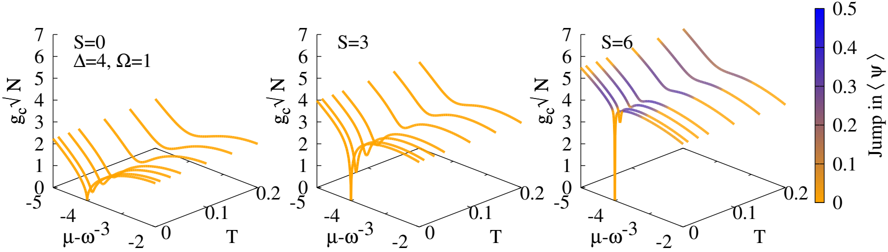

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

- Optimal phonon displacements, $\sim \sqrt{S}$
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- For $\psi \neq 0$, competition
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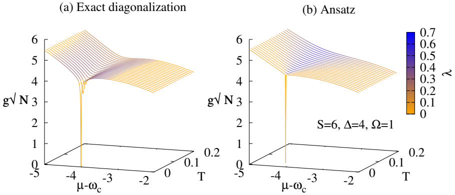
Collective polaron formation

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



- Feedback: Large/small $g_{\mathrm{eff}} \leftrightarrow \lambda = \langle\psi\rangle$
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Effective 2LS energy in field:

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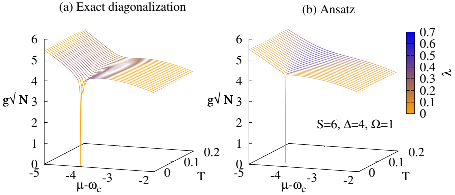

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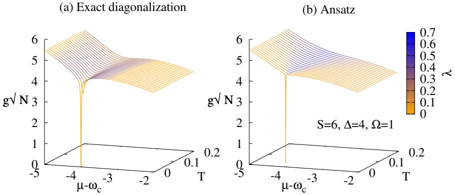

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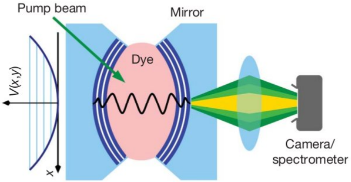
Polariton and photon Condensation

1. Introduction
   - Polariton condensation
   - Condensation, superradiance, lasing
     - Condensation vs superradiance transition
     - Non-equilibrium condensation vs lasing

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

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

Photon BEC experiments



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[Klaers et al, Nature, 2010]

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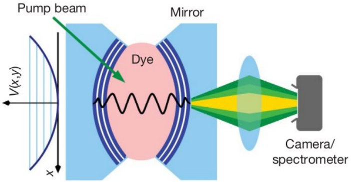

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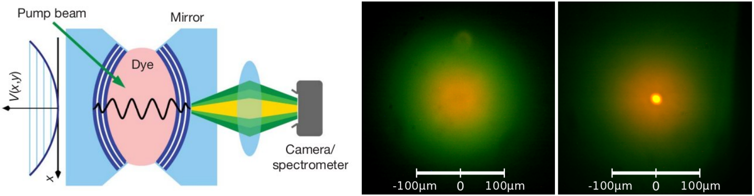

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Relation to dye laser

- No electronic inversion
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- Thermalised many-mode system

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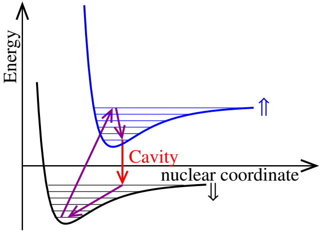

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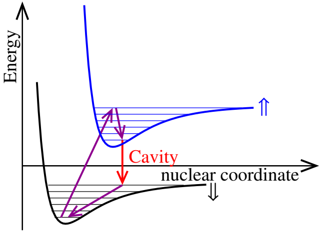

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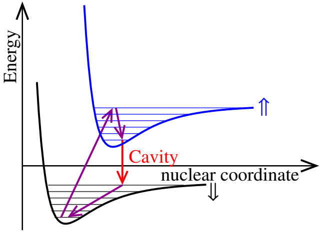

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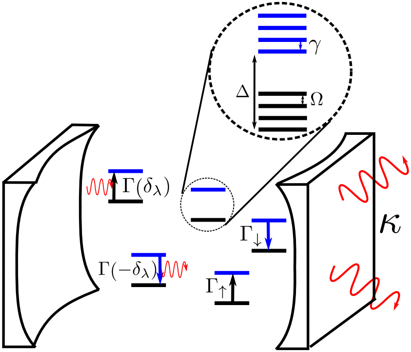

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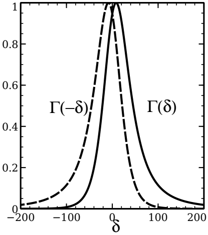

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- $\Gamma \to 0$ at large δ

[Marthaler et al PRL ’11, Kirton & JK arXiv:1303.3459]

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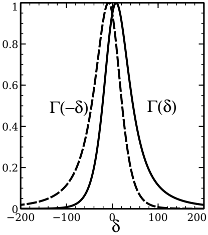

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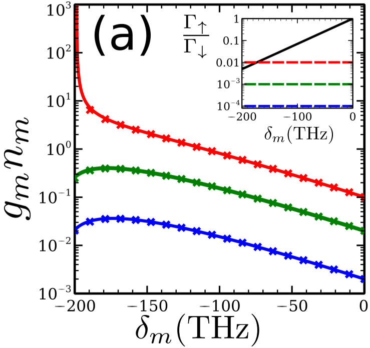

Low loss: Thermal

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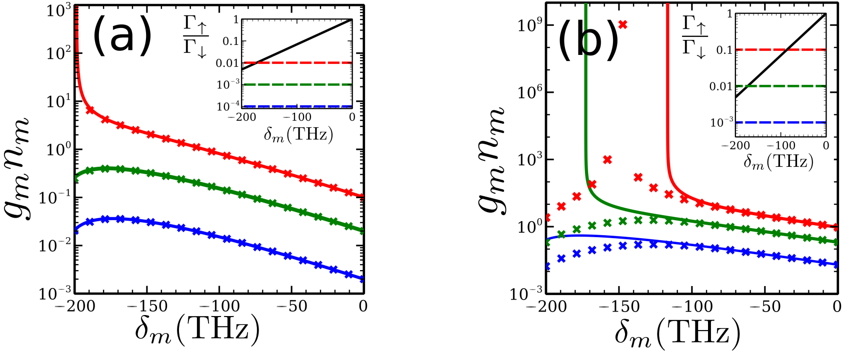
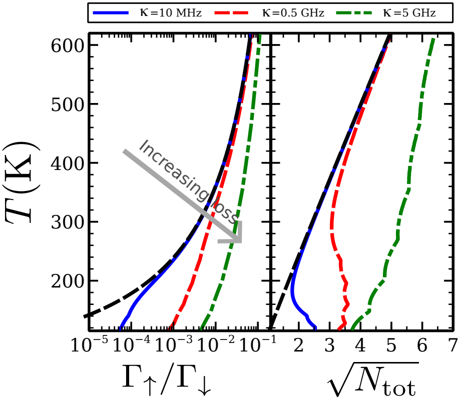


Low loss: Thermal

High loss → Laser

[Kirton & JK arXiv:1303.3459]

Threshold condition



Compare threshold:

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

- Thermal at low κ/high temperature
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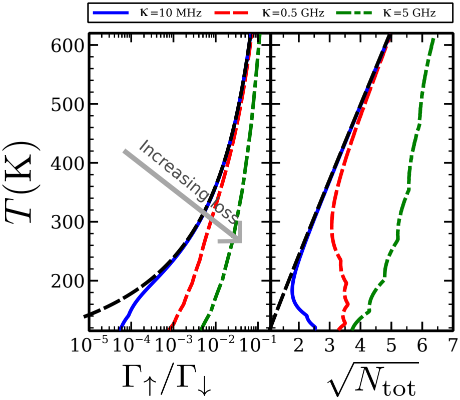

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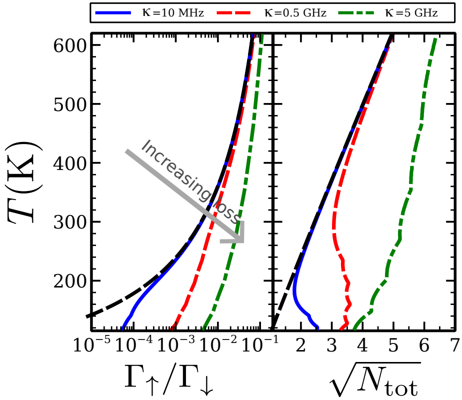

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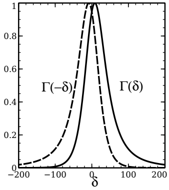

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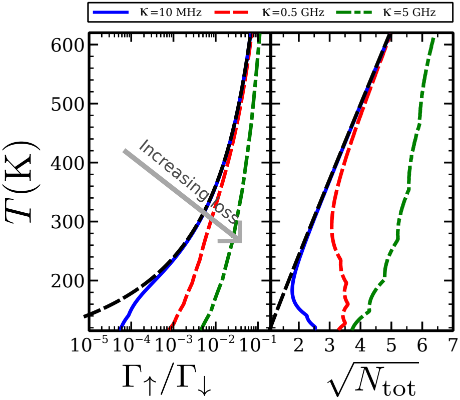

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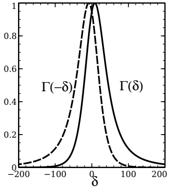

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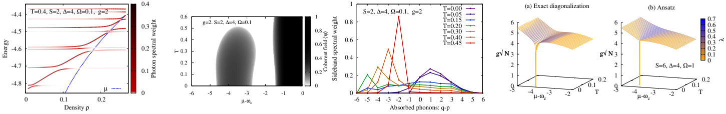
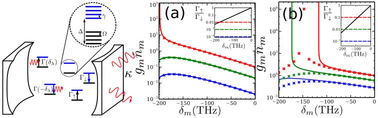
Summary

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



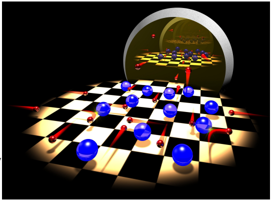
- Photon condensation and thermalisation



Many body quantum optics and correlated states of light

9:00 am on Monday 28 October 2013 – 5:00 pm on Tuesday 29 October 2013

at:The Royal Society at Chicheley Hall, home of the Kavli Royal Society International Centre, Buckinghamshire



Theo Murphy international scientific meeting organised by Dr Jonathan Keeling, Professor Steven Girvin, Dr Michael Hartmann and Professor Peter Littlewood FRS.

List of speakers and chairs

Professor Iacoppo Carusotto, Professor Andrew Cleland, Professor Hui Deng, Professor Tilman Esslinger, Professor Rosario Fazio, Professor Ed Hinds, Professor Andrew Houck, Professor Ataç İmamoğlu, Professor Jens Koch, Professor Misha Lukin, Professor Martin Plenio, Professor Arno Rauschenbeutel, Professor Timothy Spiller, Professor Jacob Taylor, Professor Hakan Tureci, Professor Andreas Wallraff

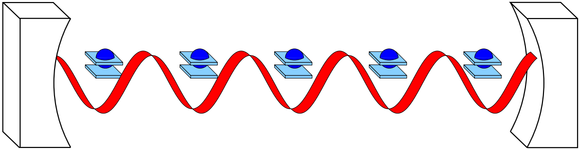
Attending this event

This is a residential conference which allows for increased discussion and networking. It is free to attend, however participants need to cover their accommodation and catering costs if required. Places are limited and therefore pre-registration is essential.

Extra slides

4. No go theorem
5. Retarded Green's function for laser
6. Organic properties
7. Anticrossing vs ρ

No go theorem and transition



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

[Rzazewski *et al* PRL ’75]

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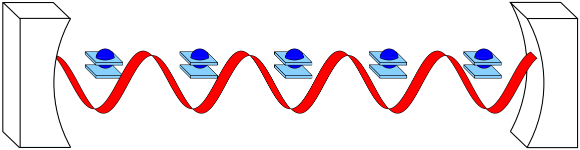

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No go theorem:. Minimal coupling $(p - eA)^2/2m$

$$-\sum_i \frac{e}{m} A \cdot p_i \Leftrightarrow g(\psi^\dagger S^- + \psi S^+), \qquad \sum_i \frac{A^2}{2m} \Leftrightarrow N\zeta(\psi + \psi^\dagger)^2$$

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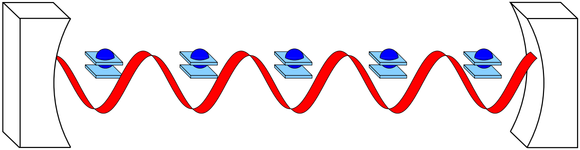

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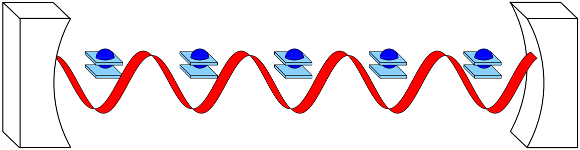

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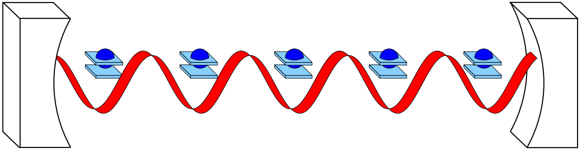

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But Thomas-Reiche-Kuhn sum rule states: $g^2/\epsilon < 2\zeta$. **No transition**

[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: $\omega_0 \ll \omega$.
  [Dimer *et al.* PRA '07; Baumann *et al.* Nature '10. Also, Black *et al.* PRL '03]

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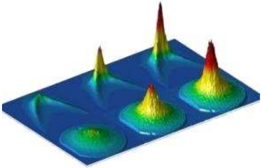

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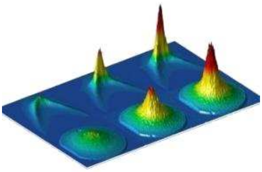

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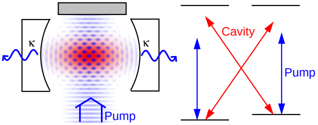
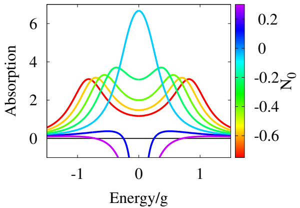


Maxwell-Bloch Equations: Retarded Green's function



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

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

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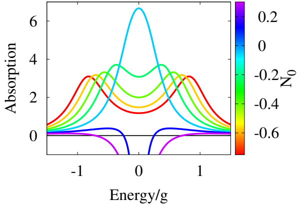

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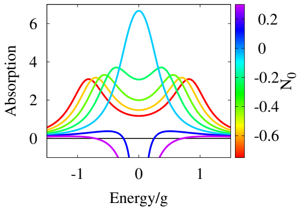

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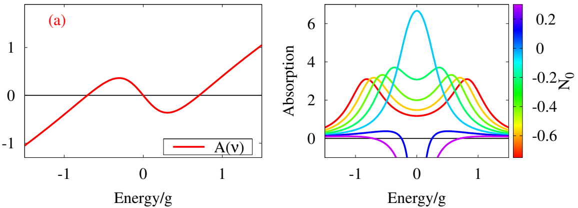

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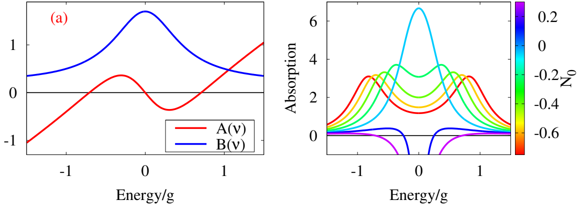

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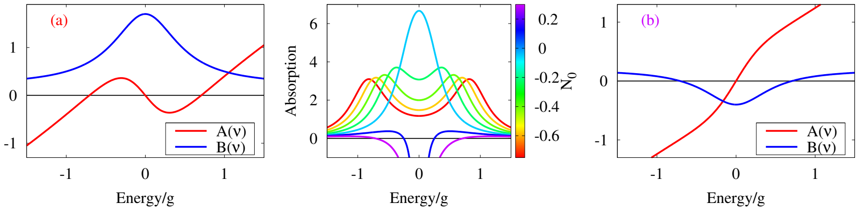

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Organic materials in microcavities

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

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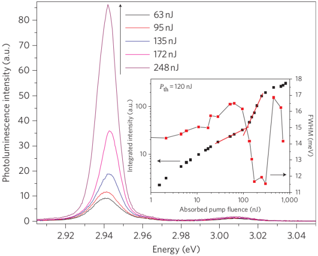

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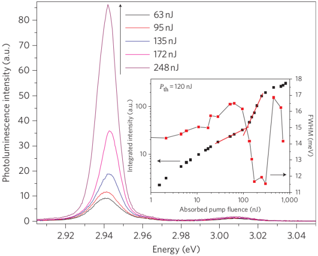

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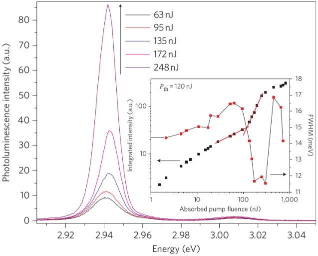

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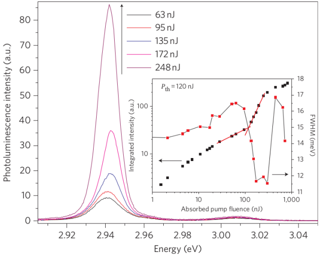

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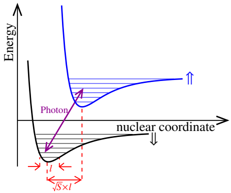

Polariton spectrum — coupled oscillators

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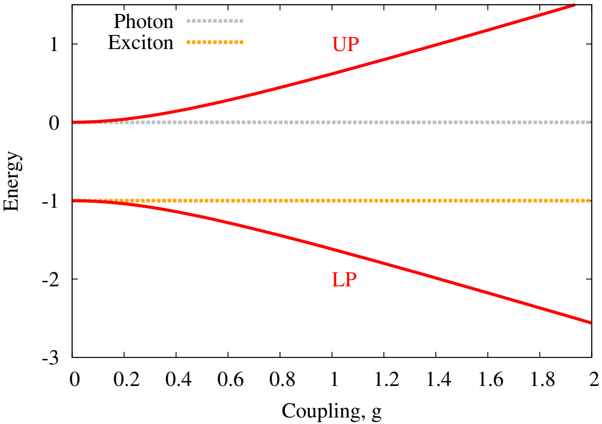

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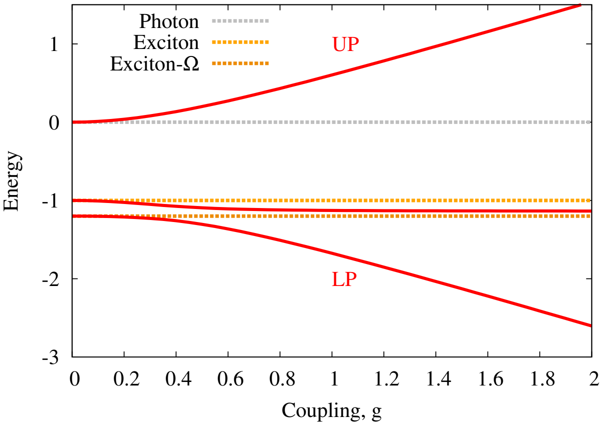

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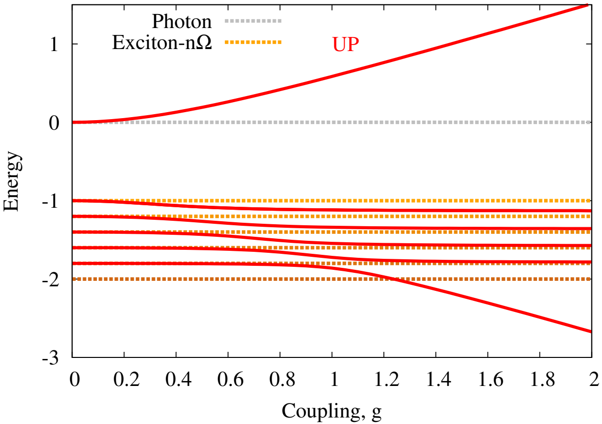

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