Polariton and photon condensates in organic materials

Jonathan Keeling

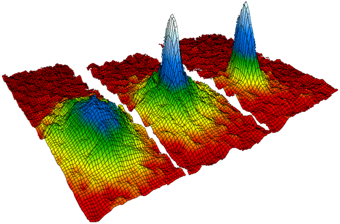
University of St Andrews
600 YEARS

SUPA

Oxford, June 2013

Coherent states of matter and light

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



[Anderson *et al.* Science '95]

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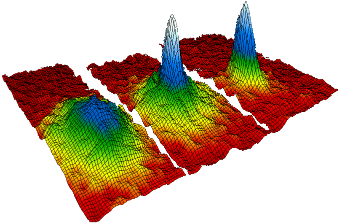

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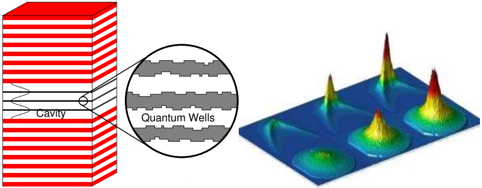
Polariton Condensate $T \sim 20$K



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

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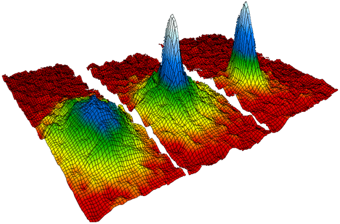

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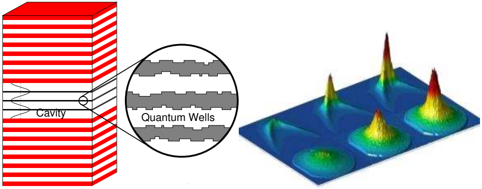
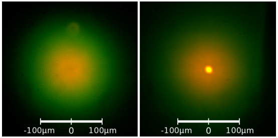


[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K



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

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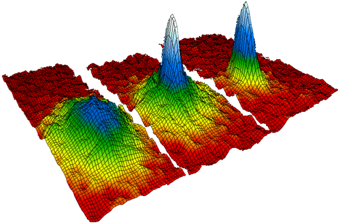

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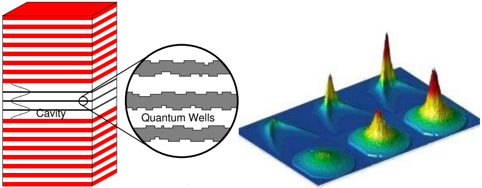

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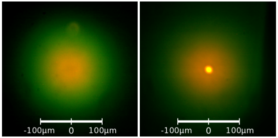
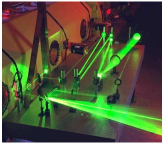


[Klaers *et al.* Nature, '10]

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



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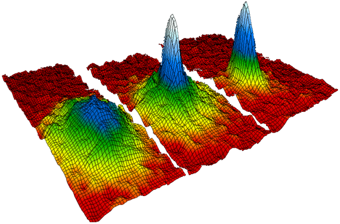

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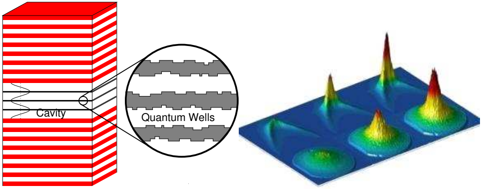

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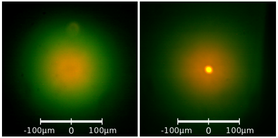

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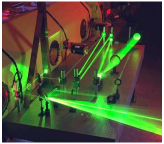
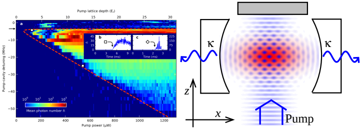


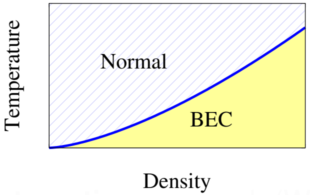
Superradiance transition
$T \sim 0$

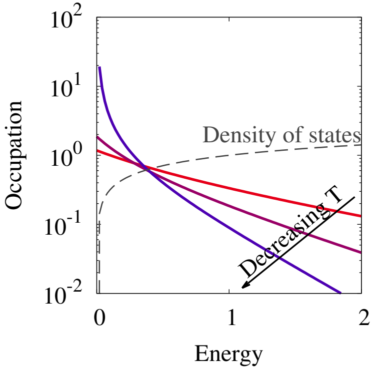


[Baumann *et al.* Nature, '10]

"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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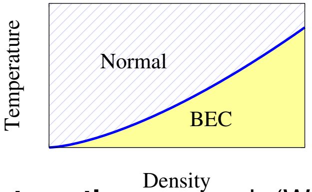

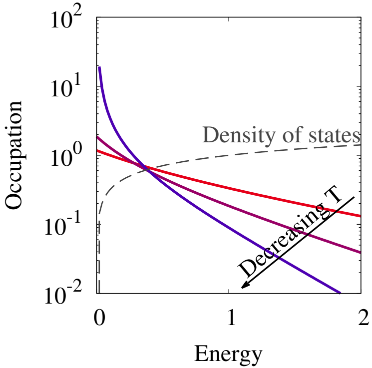

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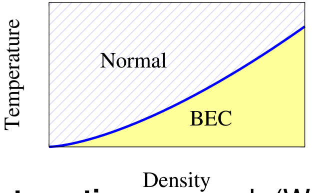

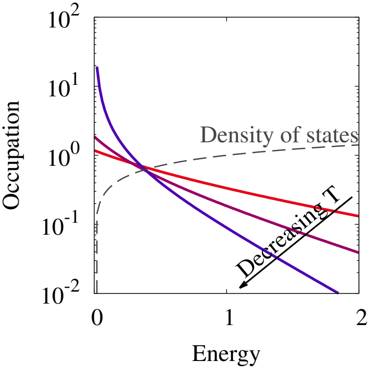

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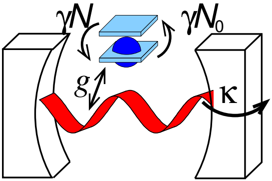
“Textbook” Laser: Maxwell Bloch equations

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



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

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

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

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

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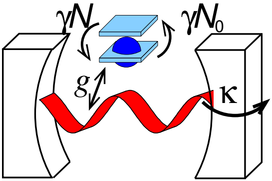

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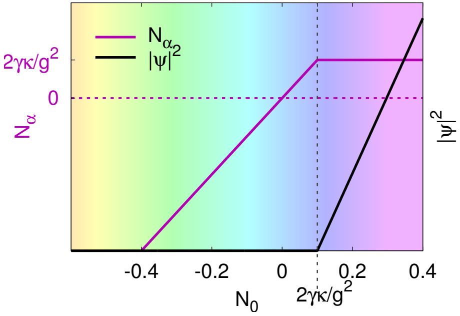
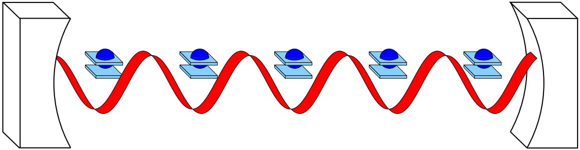


$|\psi|^2 > 0$ if $N_0 g^2 > 2\gamma\kappa$

“Textbook” Dicke-Hepp-Lieb superradiance



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

- 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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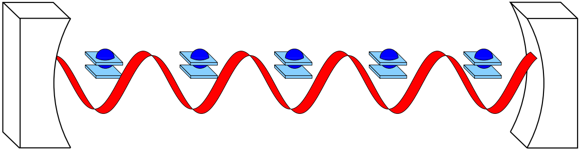

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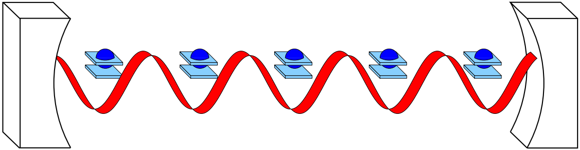

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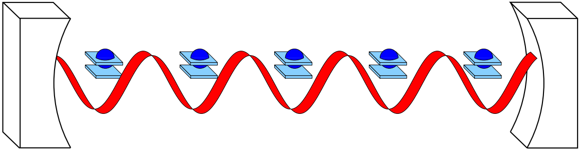

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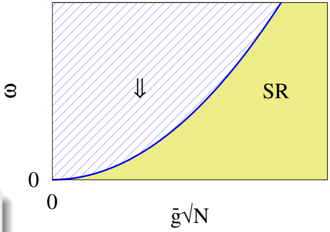


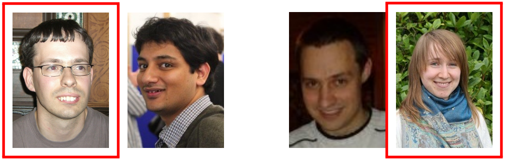
[Hepp, Lieb, Ann. Phys. ’73]

Outline

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

Acknowledgements

GROUP:



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

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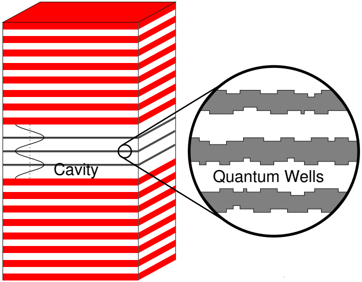
EPSRC

Engineering and Physical Sciences Research Council

Microcavity polaritons



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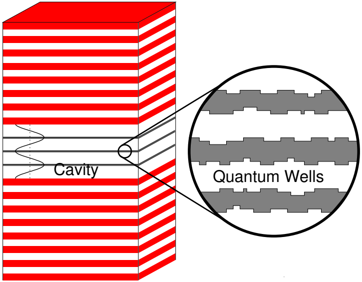
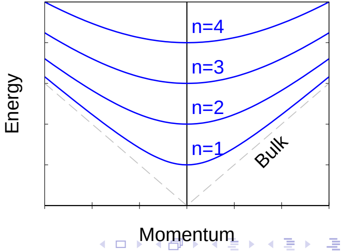


Cavity photons:

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



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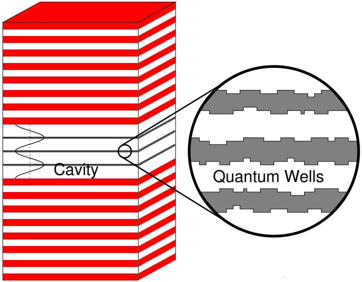

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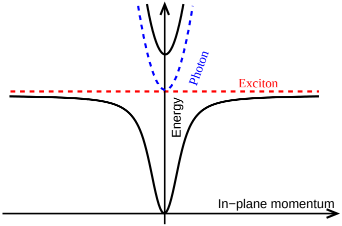

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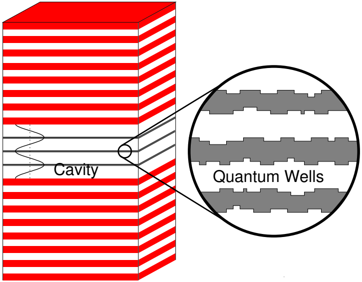

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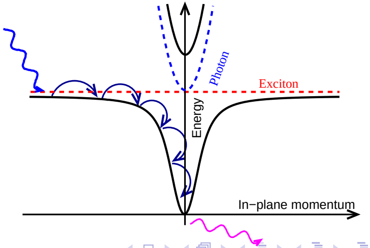

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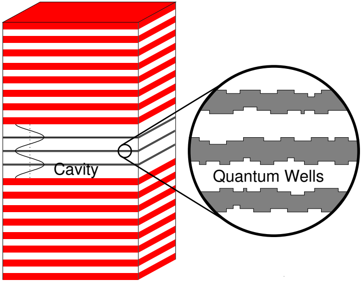

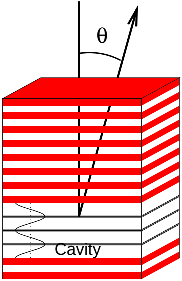

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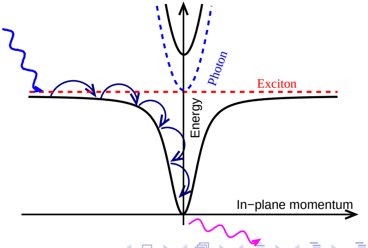

Lasing-condensation crossover model

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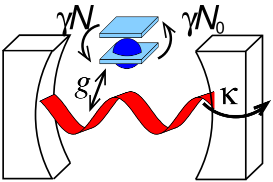
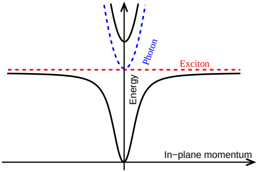


$\Leftrightarrow$



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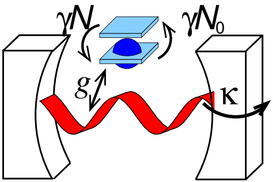

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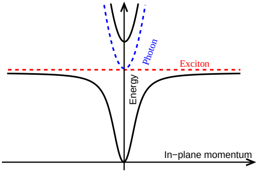


Dicke model:

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

Dicke-Hepp-Lieb superradiance and modes

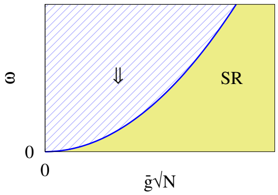
$$H = \omega\psi^\dagger\psi + \epsilon S^z + g\left(\psi^\dagger S^- + \psi S^+\right)$$

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

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

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

- Transition when $E = 0$



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

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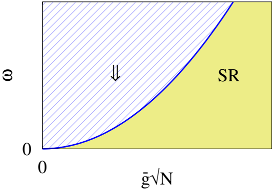

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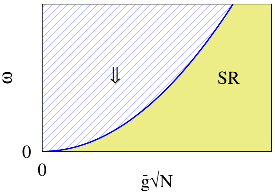

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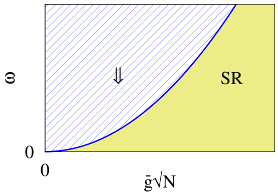
  $H = \omega\psi^\dagger\psi + \epsilon B^\dagger B + g\sqrt{N}\left(\psi^\dagger B + \psi B^\dagger\right)$
- Excitation cost E:

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

- 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: $g^2 N > (\omega - \mu)|\epsilon - \mu|$
- Fix density / fix $\mu > 0$ — pumping

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

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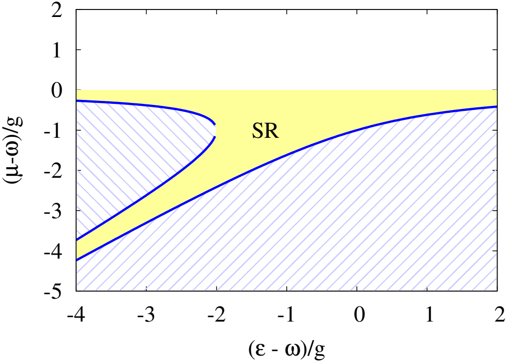

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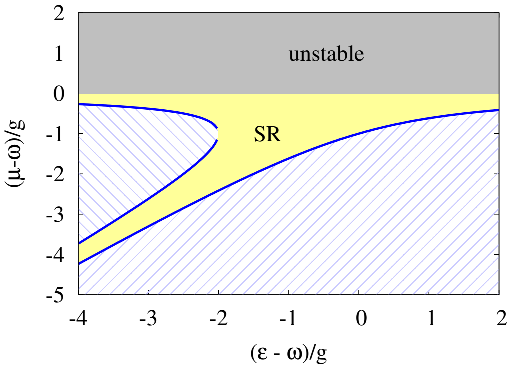

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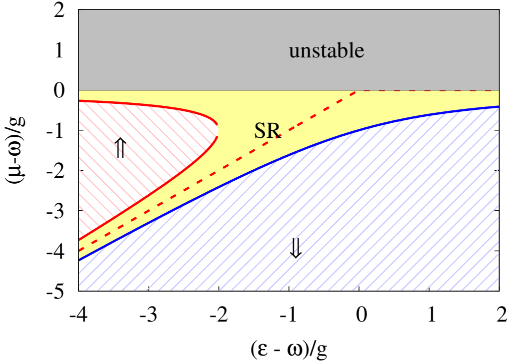

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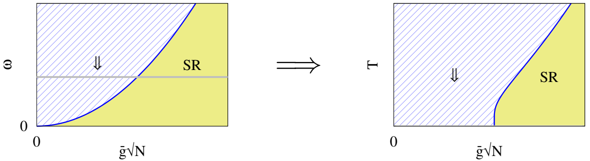
Grand canonical Dicke, finite temperature

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



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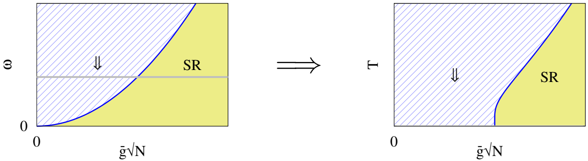

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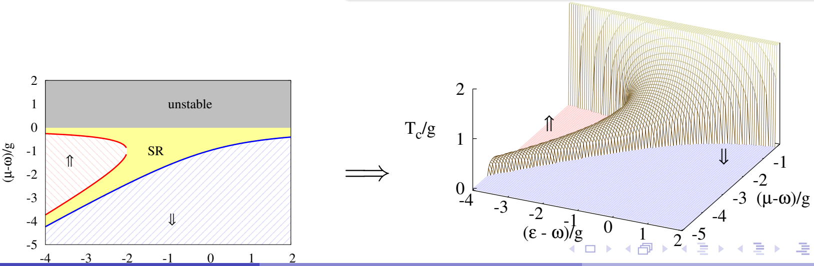

Polariton model and equilibrium results

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

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

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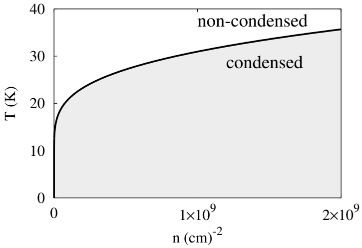

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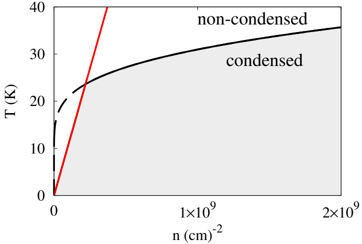

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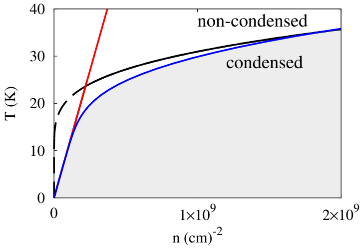

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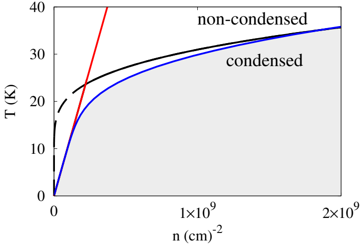
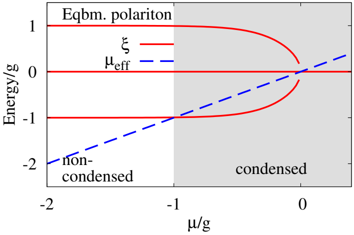
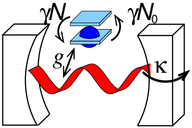


Modes (at $k = 0$)



Non-equilibrium condensation vs lasing

Simple Laser: Maxwell Bloch equations



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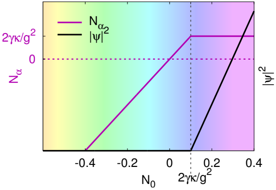

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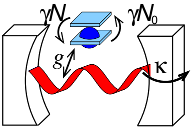

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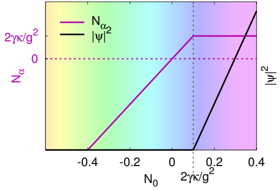

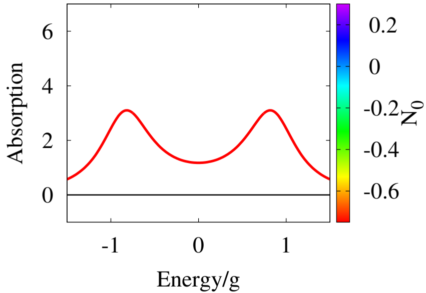

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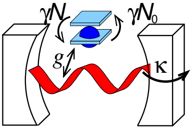

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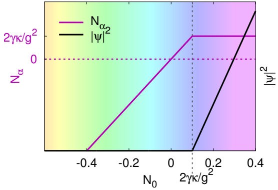

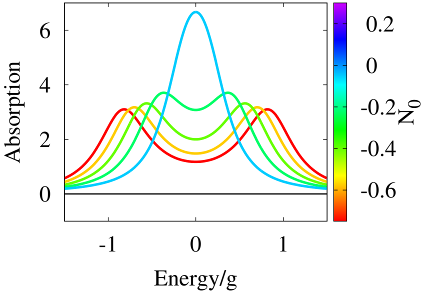

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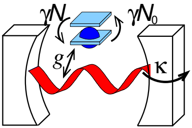

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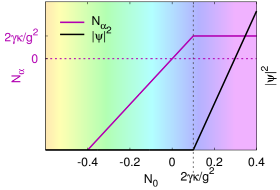

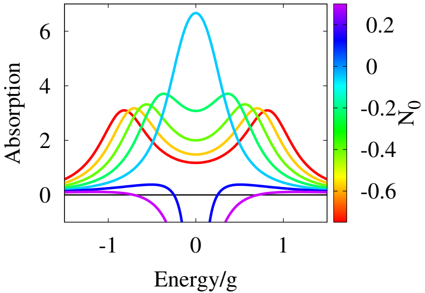

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Poles of Retarded Green's function and gain

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

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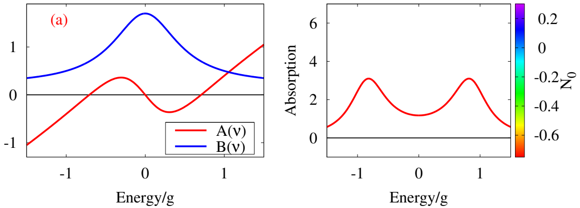

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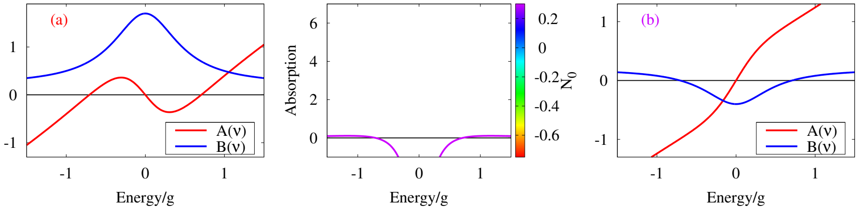

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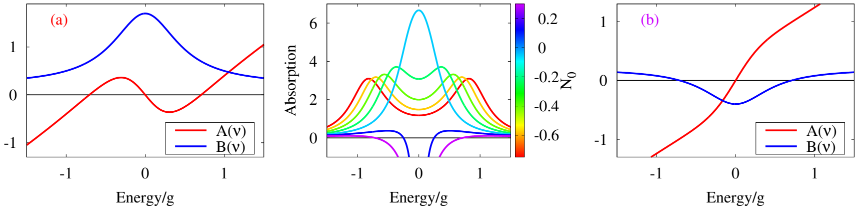
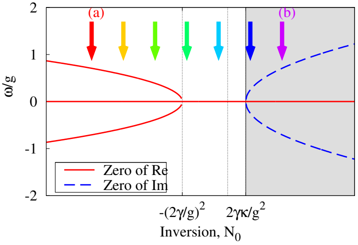


Laser:



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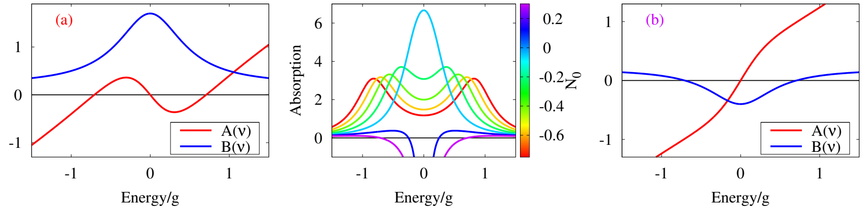

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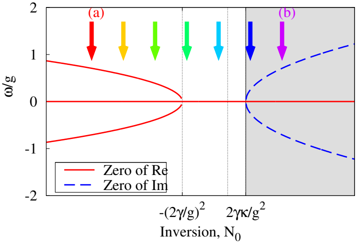
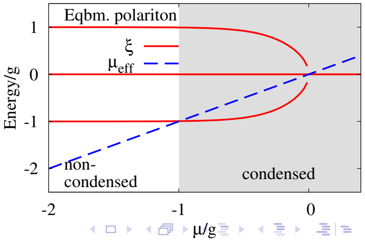
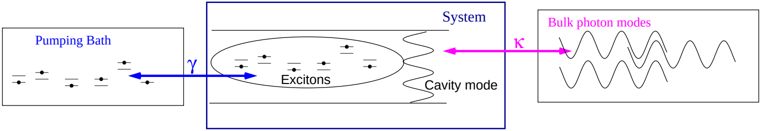
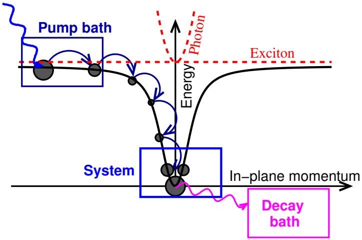


Equilibrium:



Non-equilibrium description: baths





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- Pump bath: Thermal μ_B, T_B

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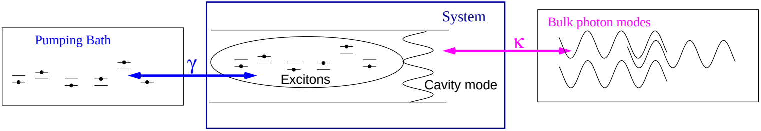

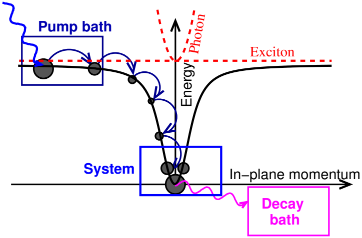

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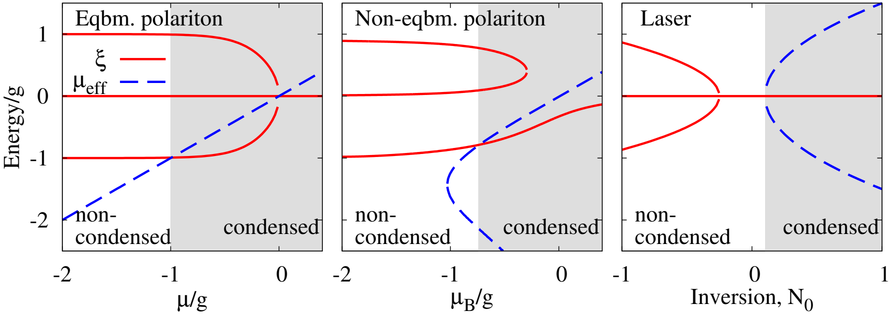
Strong coupling and lasing — low temperature phenomenon



- Inversionless
- **allows** strong coupling
- **requires** low T ↔ condensation
- Related *weak-coupling* **inversionless** lasing

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

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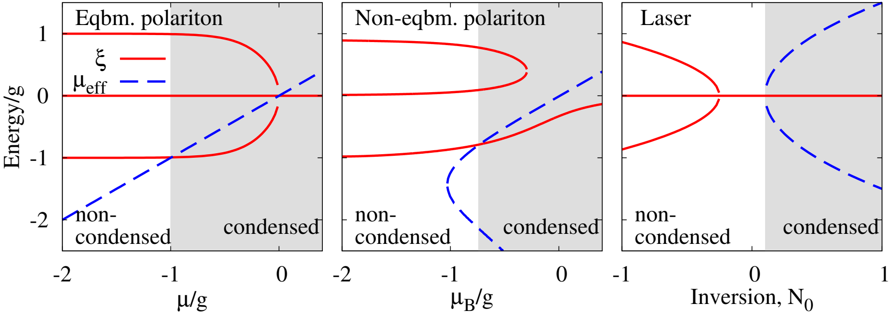

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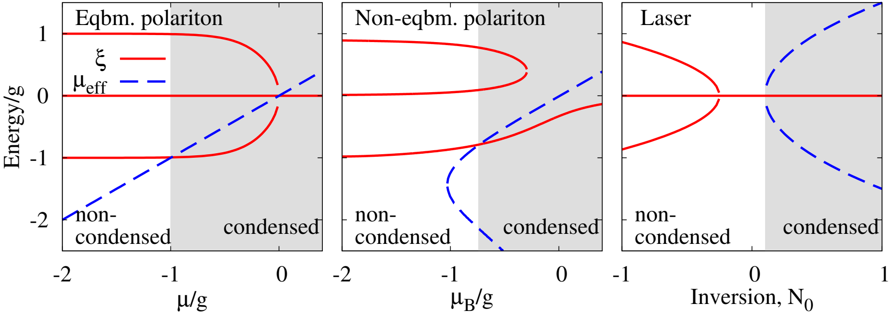

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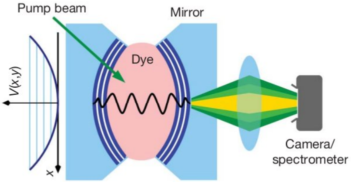
Polariton and photon Condensation

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

Photon BEC experiments



- Dye filled microcavity
- No strong coupling

[Klaers et al, Nature, 2010]

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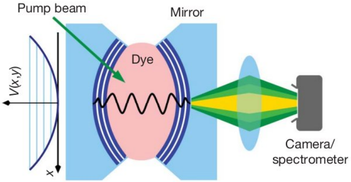

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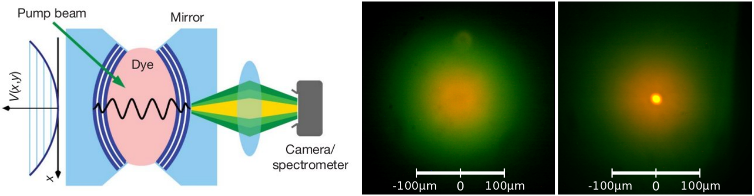

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Relation to dye laser

- No electronic inversion
- No strong coupling

- No single cavity mode
  - Condensate mode is not maximum gain
  - Gain/Absorption in balance
- Thermalised many-mode system

Relation to dye laser

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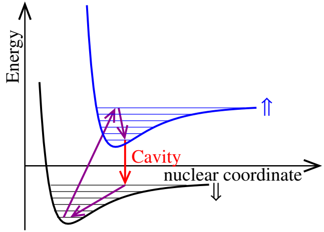
4 Level Dye Laser



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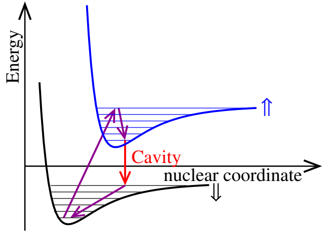

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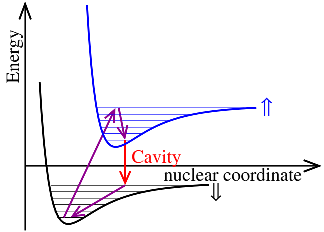

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$$H_{\text{sys}} = \sum_m \omega_m \psi_m^\dagger \psi_m + \sum_\alpha \left[\epsilon S_\alpha^z + g \left(\psi_m S_\alpha^+ + \text{H.c.} \right) \right]$$

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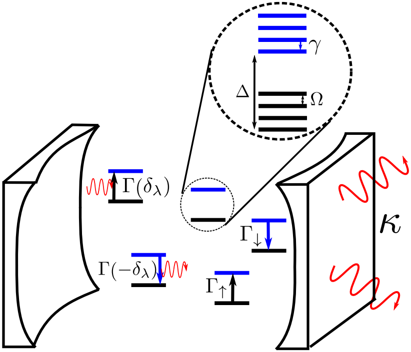

Modelling

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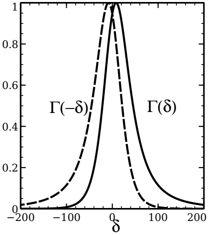

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[Marthaler et al PRL ’11, Kirton & JK arXiv:1303.3459]

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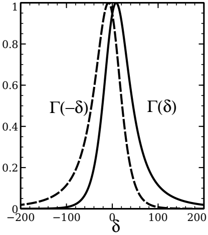

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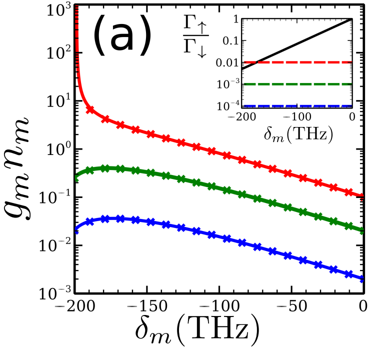

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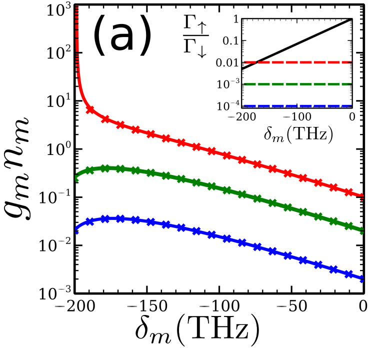

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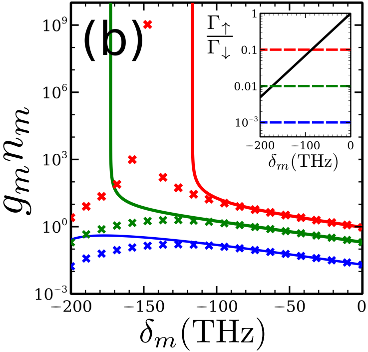
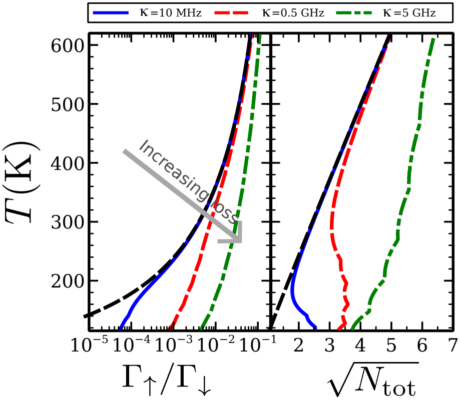


High loss → Laser

[Kirton & JK arXiv:1303.3459]

1

Threshold condition



Compare threshold:

- Pump rate (Laser)
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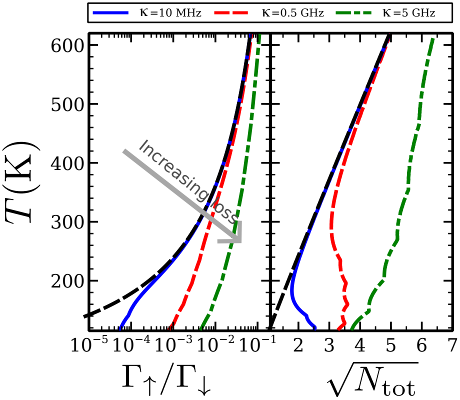

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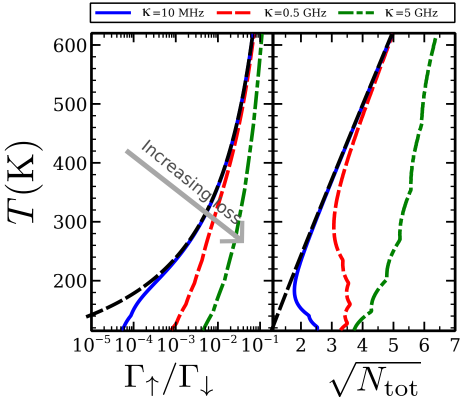

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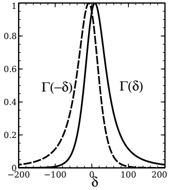

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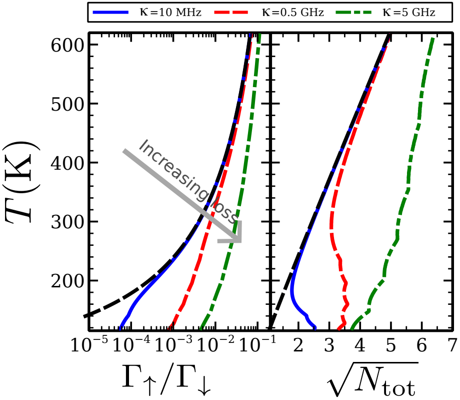

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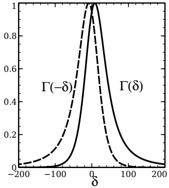

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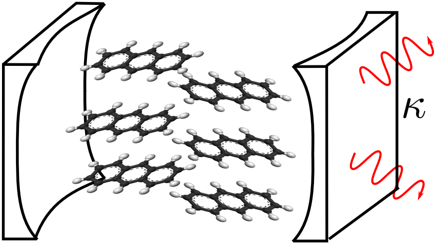
Organic polaritons: photon-exciton-phonon coupling

Organic materials in microcavities

- What?



- Why?

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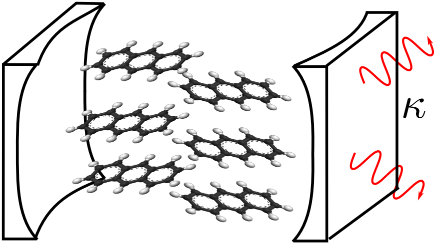

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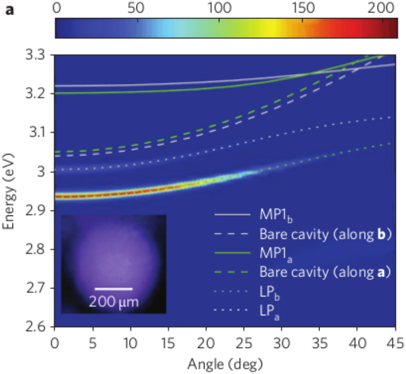

Polariton splitting: 0.1eV $\leftrightarrow$ 1000K.

[Kena Cohen and Forrest, Nat. Photon 2010]

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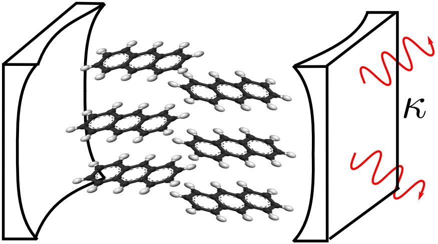

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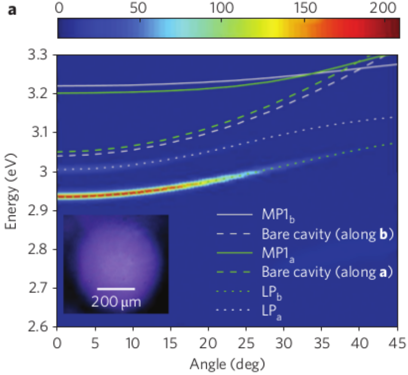

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Phase diagram

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

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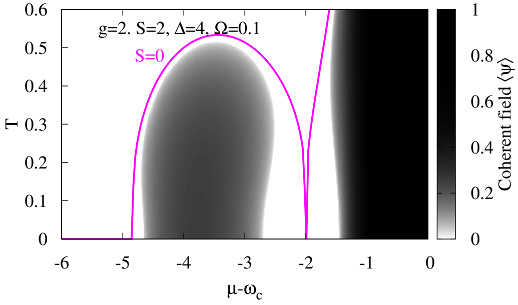

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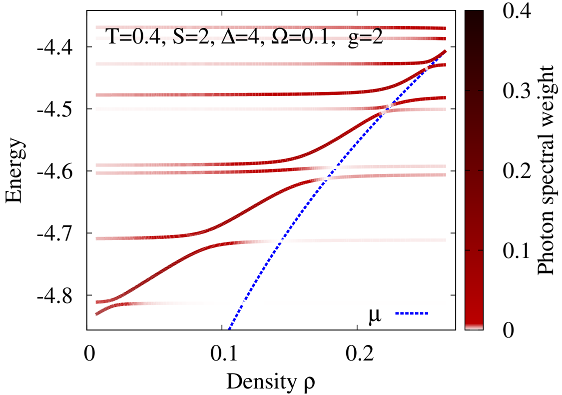
Polariton spectrum: photon weight



- 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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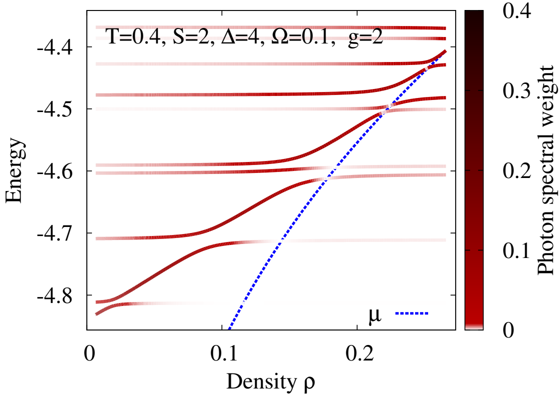

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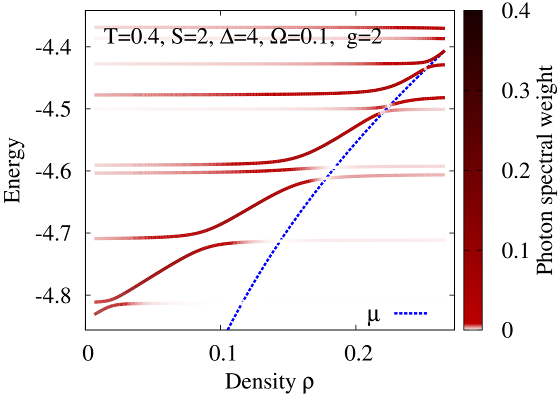

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Polariton spectrum: what condensed

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

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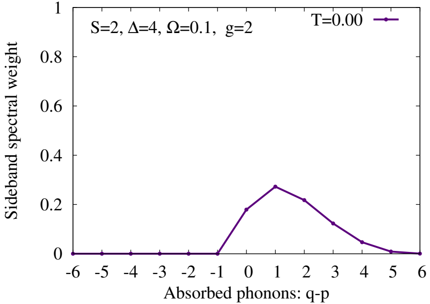

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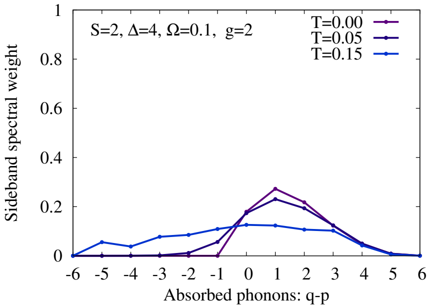

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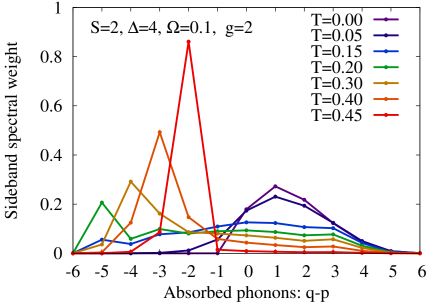

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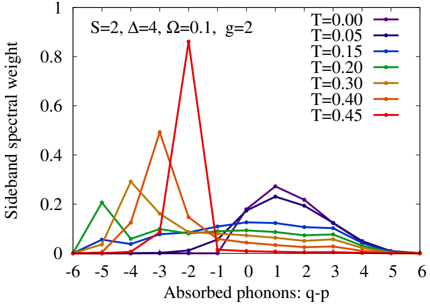

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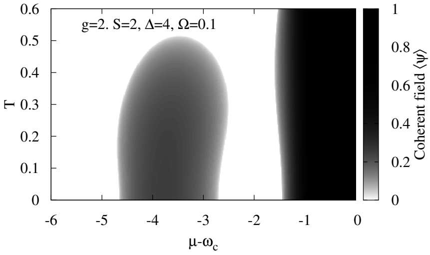

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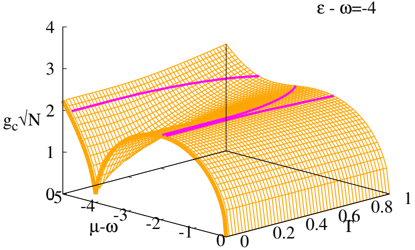
Organic polaritons

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

Critical coupling with increasing S

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



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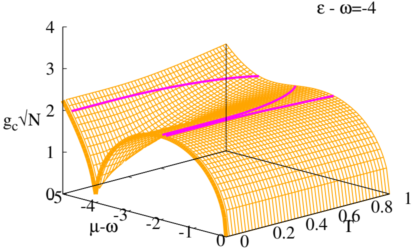

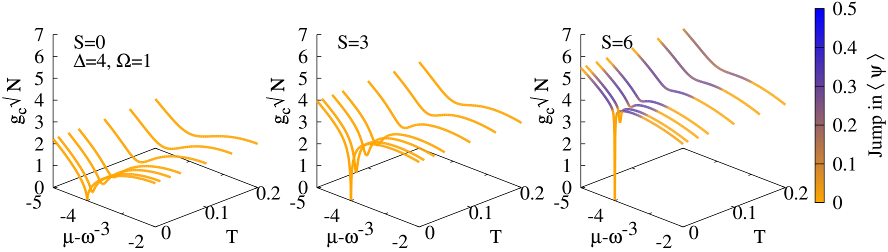

Explanation: Polaron formation

- 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 $S^\pm$

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- Optimal phonon displacements, $\sim \sqrt{S}$
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- 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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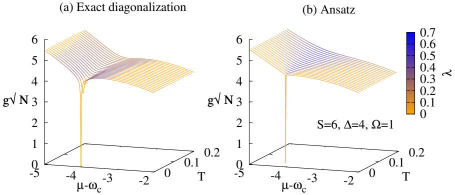
Collective polaron formation

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



- Feedback: Large/small $g_{eff} \leftrightarrow \lambda = \langle\psi\rangle$
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$$F = (\omega_c - \mu)\lambda^2 + N\left\{\Omega\left[\zeta^2 - S\frac{\eta(2-\eta)}{4}\right] - T\ln\left[2\cosh\left(\frac{\zeta}{T}\right)\right]\right\}$$

Effective 2LS energy in field:

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[Cwik *et al.* arXiv:1303.3702]

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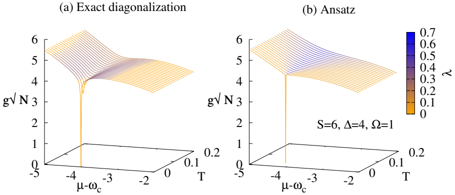

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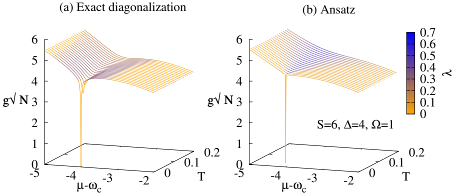

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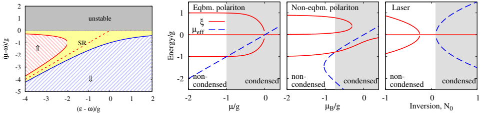
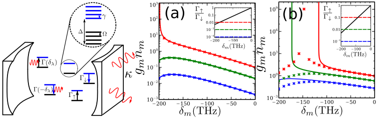
[Cwik *et al.* arXiv:1303.3702]

Summary

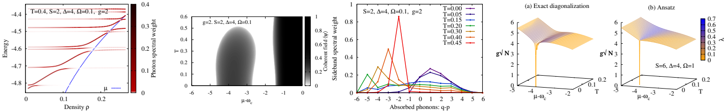
- Polariton condensation vs lasing; superradiance



- Photon condensation and thermalisation



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

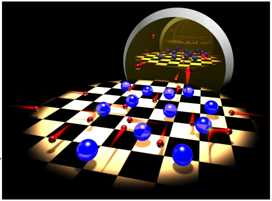


Many body quantum optics and correlated states of light

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

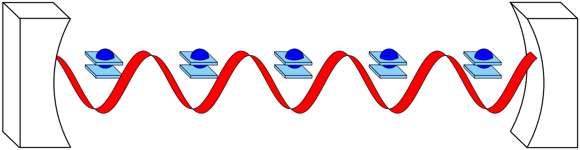
Professor Iacoppo Carusotto, Professor Andrew Cleland, Professor Hui Deng, Professor Tilman Esslinger, Professor Rosario Fazio, Professor Ed Hinds, Professor Andrew Houck, Professor Ataç İmamoğlu, Professor Jens Koch, Professor Misha Lukin, Professor Martin Plenio, Professor Arno Rauschenbeutel, Professor 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

No go theorem and transition



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

[Rzazewski *et al* PRL '75]

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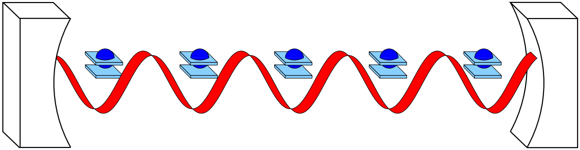

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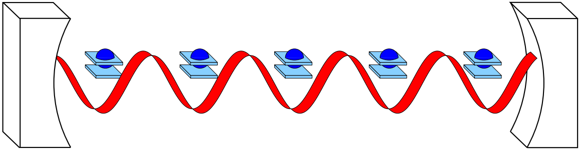

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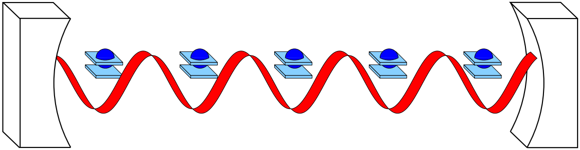

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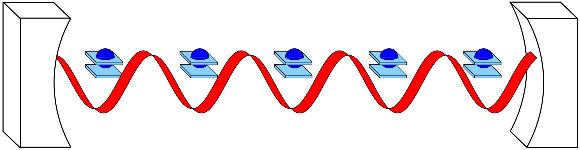

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But Thomas-Reiche-Kuhn sum rule states: $g^2/\epsilon < 2\zeta$. **No transition**

[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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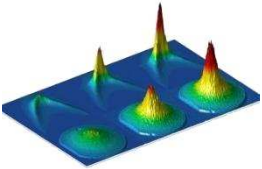

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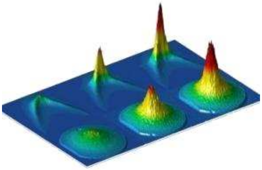

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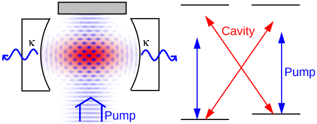
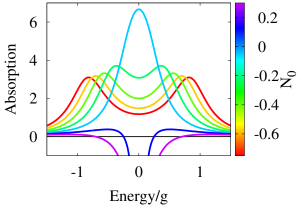


Maxwell-Bloch Equations: Retarded Green's function



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

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

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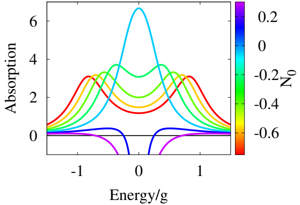

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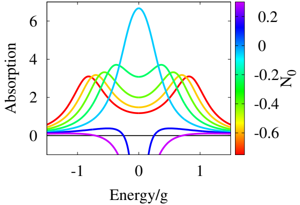

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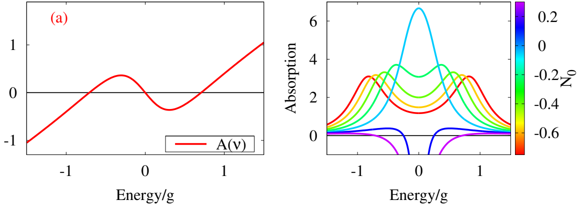

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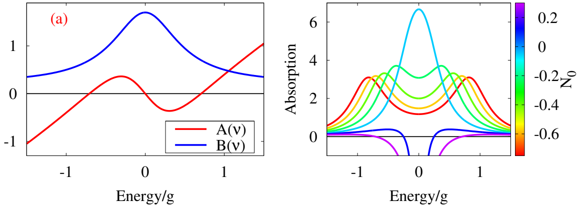

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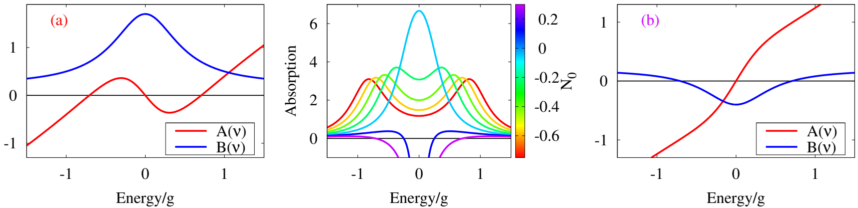

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Organic materials in microcavities

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

    [Kena Cohen and Forrest, Nat. Photon 2010]
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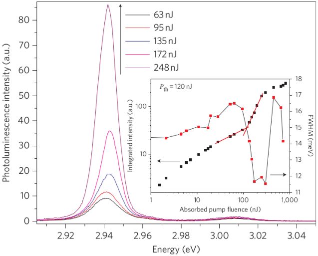

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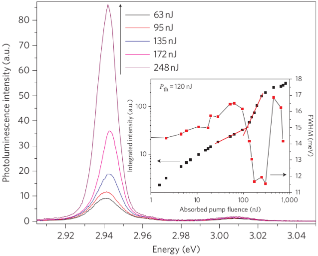

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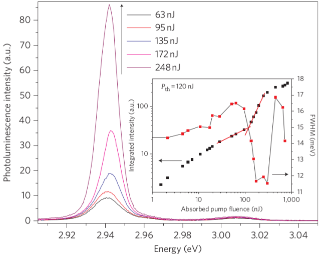

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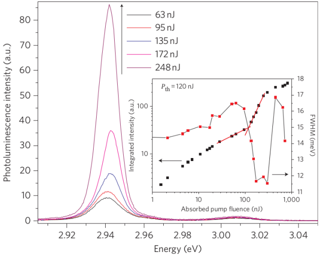

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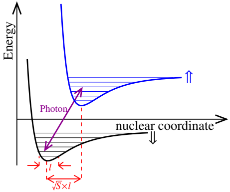

Polariton spectrum — coupled oscillators

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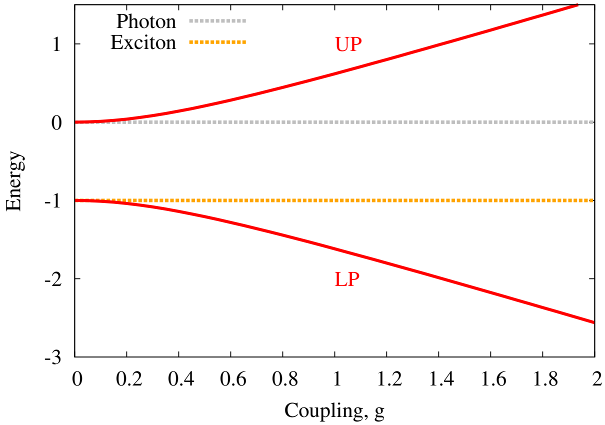

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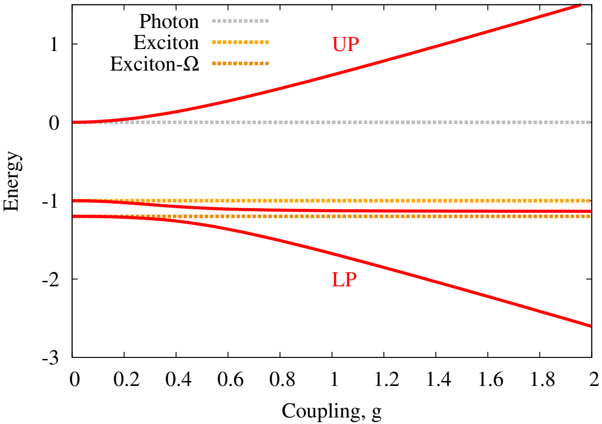

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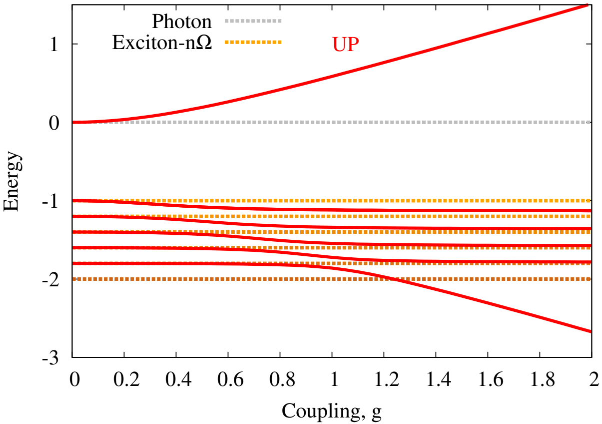

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