Polariton and photon condensates in organic materials

Jonathan Keeling

Dresden,May 2013

Acknowledgements

GROUP:



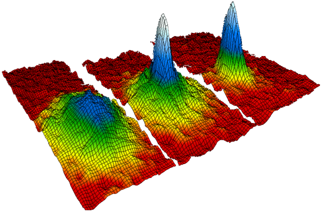

COLLABORATORS: Szymanska (Warwick), Reja (Cam.), Littlewood (ANL)

FUNDING:

EPSRC
Engineering and Physical Sciences Research Council

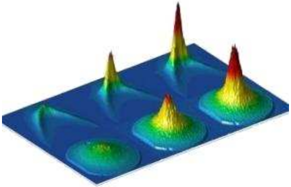
Bose-Einstein condensation: macroscopic occupation

Atoms. $\sim 10^{-7}$K



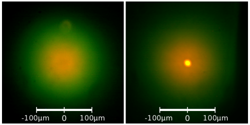
[Anderson *et al.* Science '95]

Polaritons. $\sim$ 20K



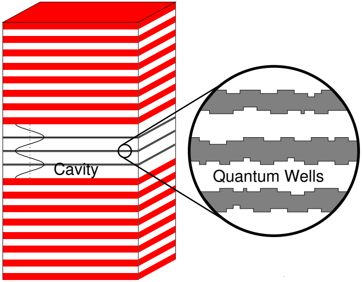
[Kasprzak *et al.* Nature, '06]

Photons. $\sim$ 300K



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

Microcavity polaritons



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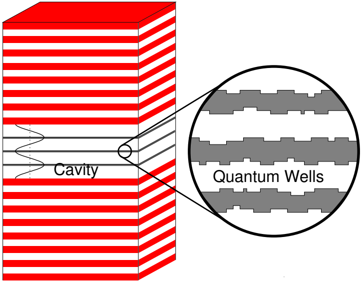
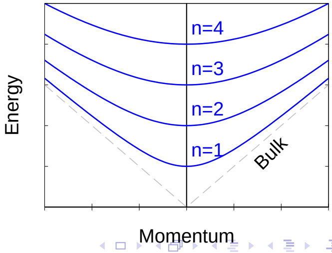


Cavity photons:

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



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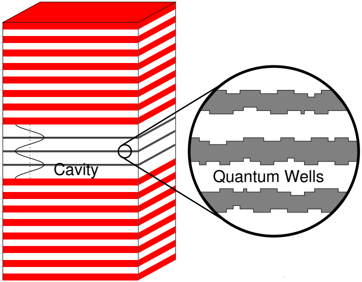

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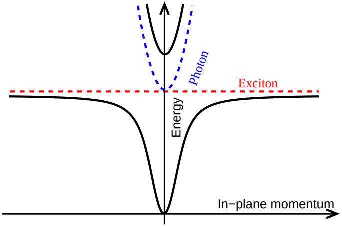

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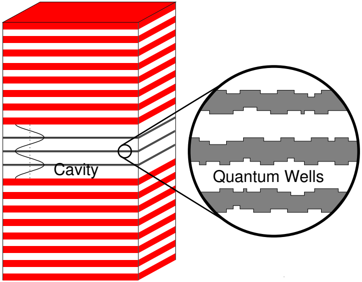

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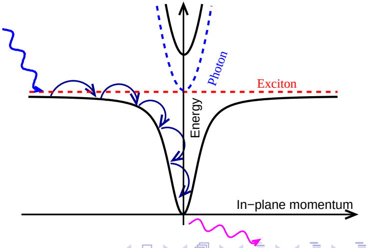

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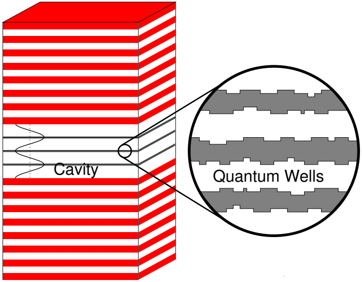

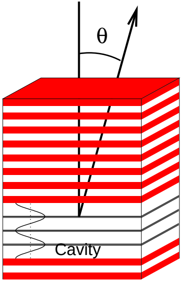

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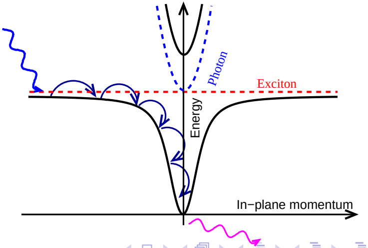
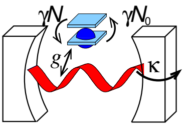


Outline

1. Polariton condensation
   - Introducton to polaritons
   - Non-equilibrium condensation vs lasing
   - Dicke model phase transition
2. Organic polaritons
   - Experiments and Dicke-Holstein model
   - Modified phase diagram and phonon sidebands
   - First order transitions
3. Photon condensation
   - Multimode rate equation
   - Critical properties from non-equilibrium model

Lasing-condensation crossover model

- Use model that can show lasing and condensation:



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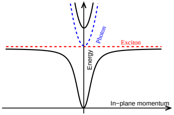

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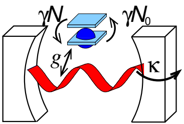

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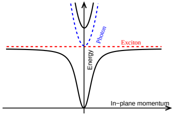
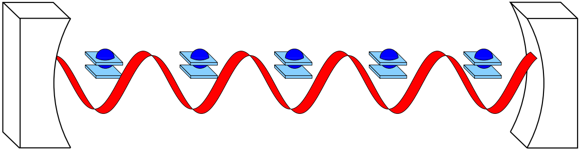


Dicke model:

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

Dicke model: Equilibrium superradiance transition



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

- 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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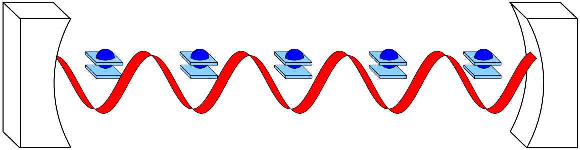

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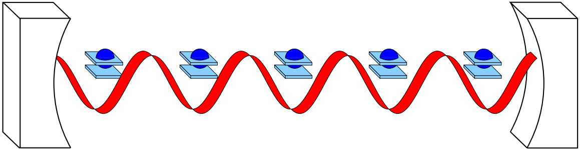

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Spontaneous polarisation if: $Ng^2 > \omega\epsilon$

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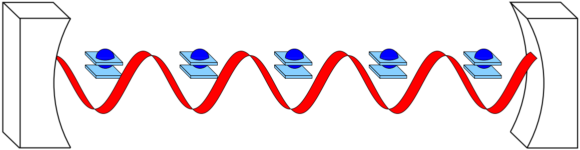

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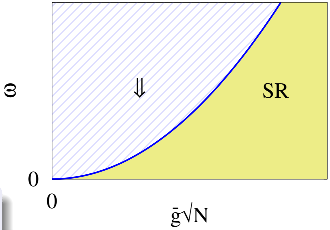

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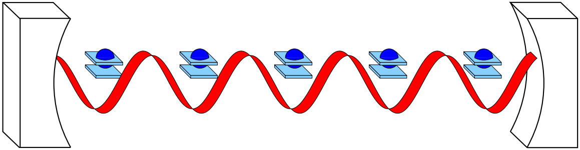
No go theorem and transition



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[Rzazewski *et al* PRL '75]

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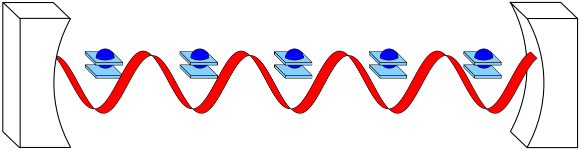

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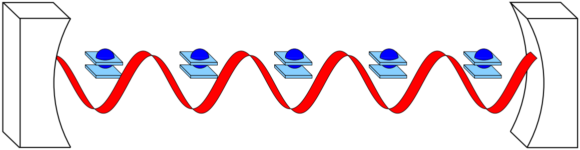

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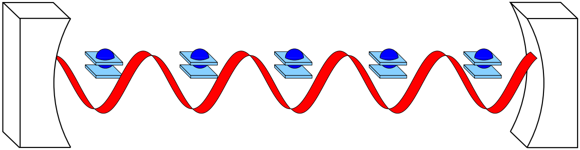

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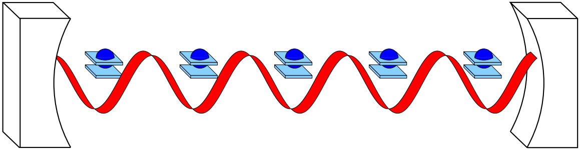

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But Thomas-Reiche-Kuhn sum rule states: $g^2/\epsilon < 2\zeta$. **No transition**

[Rzazewski *et al* PRL '75]

Grand canonical ensemble: no no-go

Problem: $g^2/\epsilon < 2\zeta$ for intrinsic parameters.
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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[Eastham and Littlewood, PRB '01]

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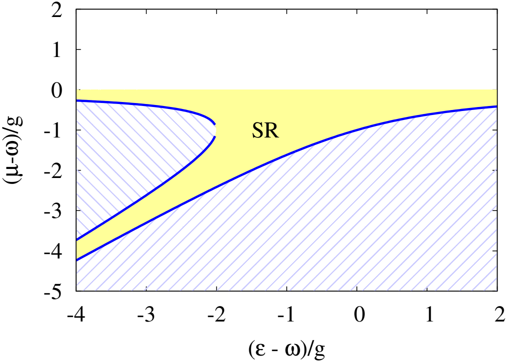

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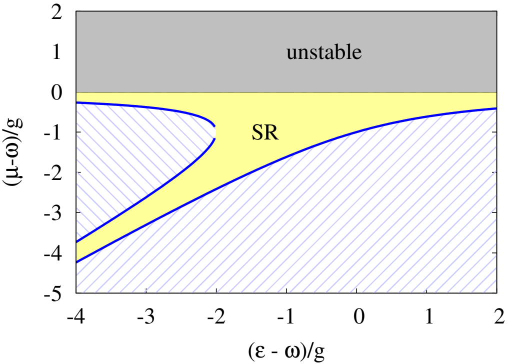

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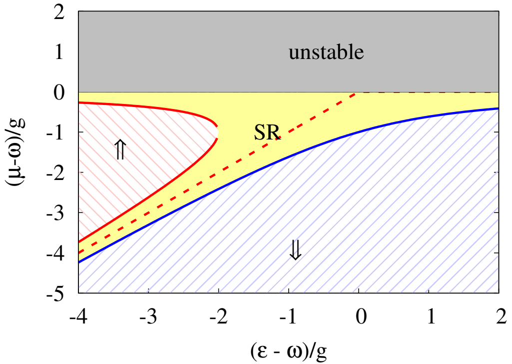

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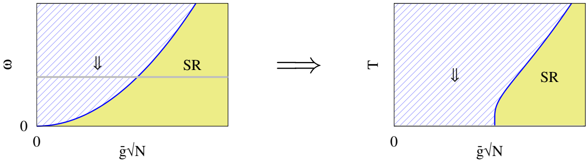
Grand canonical Dicke, finite temperature

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



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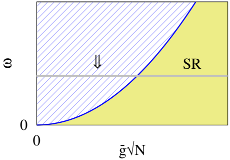

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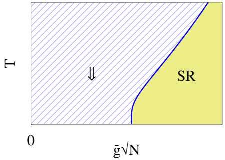

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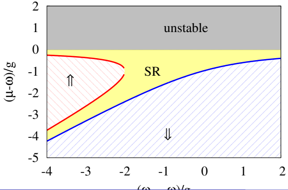

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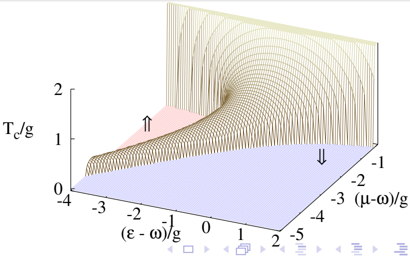

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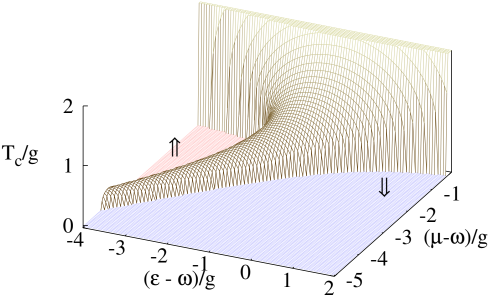

- Superradiant bubble near $\epsilon = \mu$
- T_c diverges as $\mu \to \omega$

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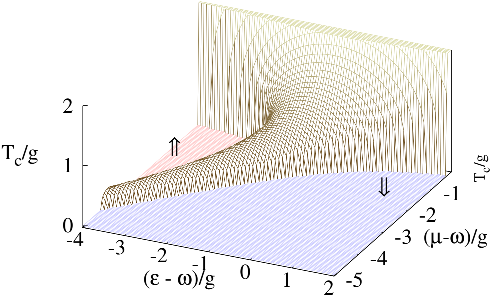

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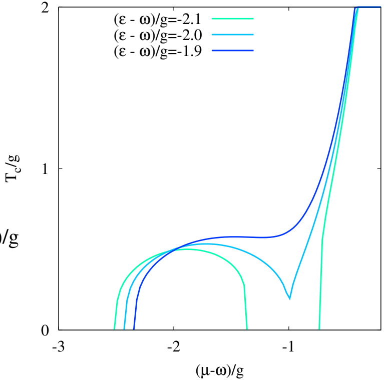

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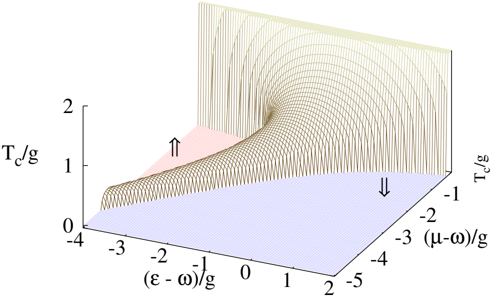

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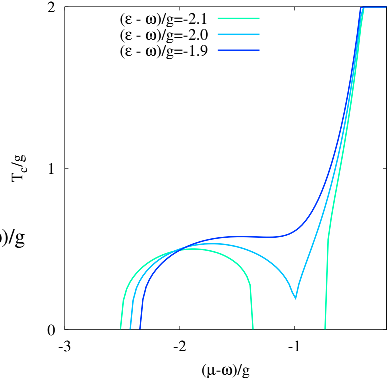

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Non-equilibrium condensation vs lasing

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Polariton model and equilibrium results

Localised excitons, propagating photons

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

Self-consistent polarisation and field

$$(\omega - \mu)\,\psi = \frac{1}{A}\sum_{\alpha} \frac{g^2_\alpha \psi}{2E_\alpha} \tanh\left(\beta E_\alpha\right), \quad {E_\alpha}^2 = \left(\frac{\epsilon_\alpha - \mu}{2}\right)^2 + g^2_\alpha |\psi|^2$$

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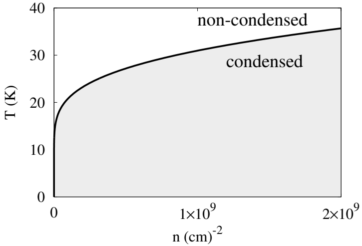
Phase diagram:



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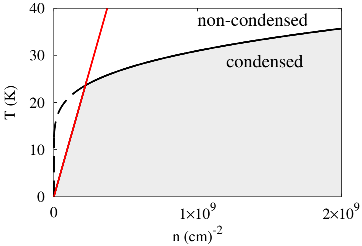

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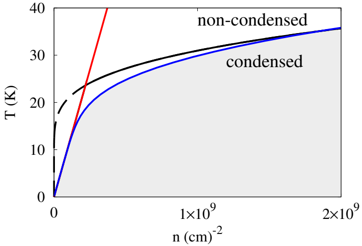
$$H - \mu N = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \mu)\psi^{\dagger}_{\mathbf{k}}\psi_{\mathbf{k}} + \sum_{\alpha} (\epsilon_\alpha - \mu) S^z_\alpha + \frac{g_{\alpha,\mathbf{k}}}{\sqrt{A}} \psi_{\mathbf{k}} S^+_\alpha + \text{H.c.}$$

Self-consistent polarisation and field

$$(\omega - \mu)\,\psi = \frac{1}{A}\sum_\alpha \frac{g_\alpha^2 \psi}{2E_\alpha} \tanh(\beta E_\alpha), \qquad {E_\alpha}^2 = \left(\frac{\epsilon_\alpha - \mu}{2}\right)^2 + g_\alpha^2 |\psi|^2$$

Phase diagram:



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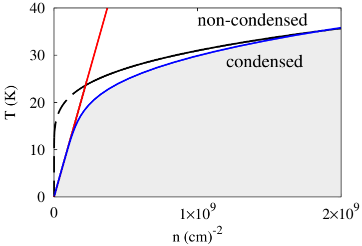
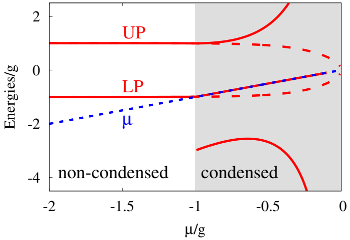


Modes (at $k = 0$)



Simple Laser: Maxwell Bloch equations

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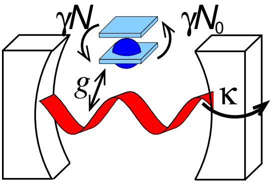

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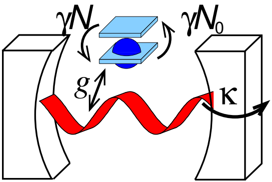

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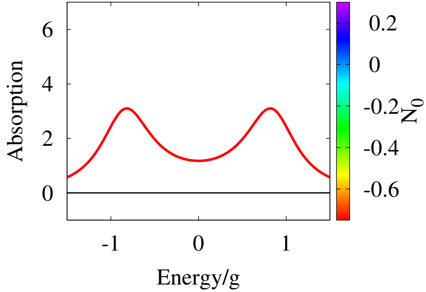

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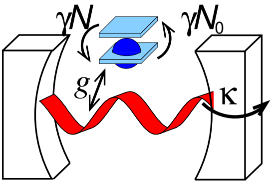

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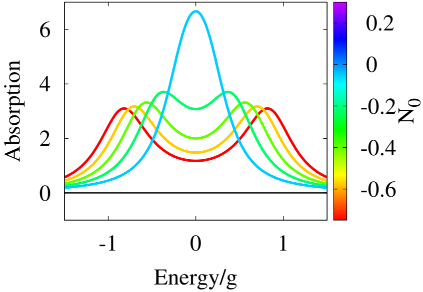

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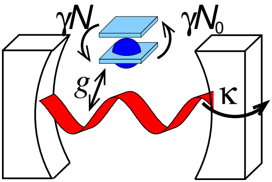

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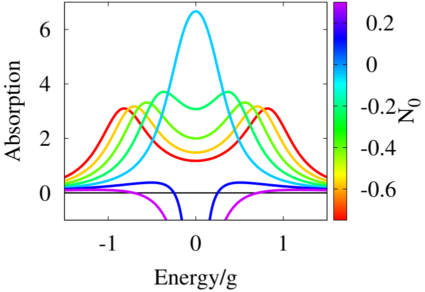

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Poles of Retarded Green's function and gain

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

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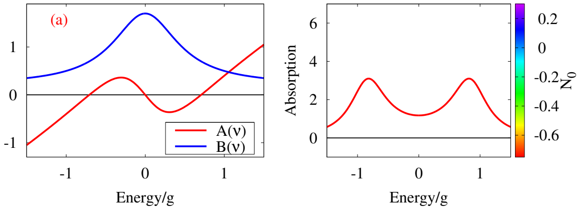

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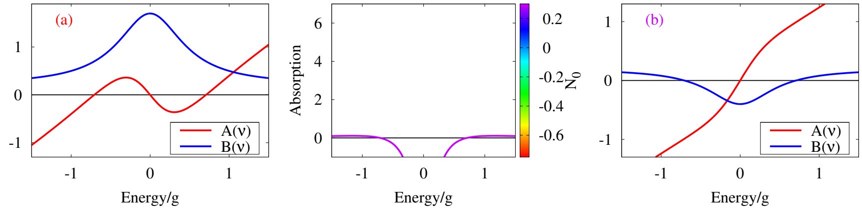

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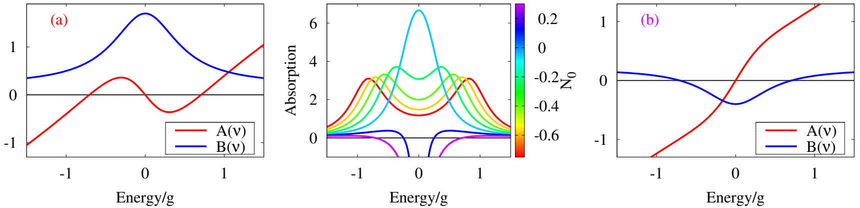
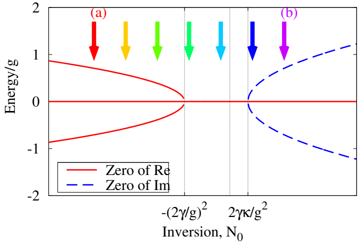


Laser:



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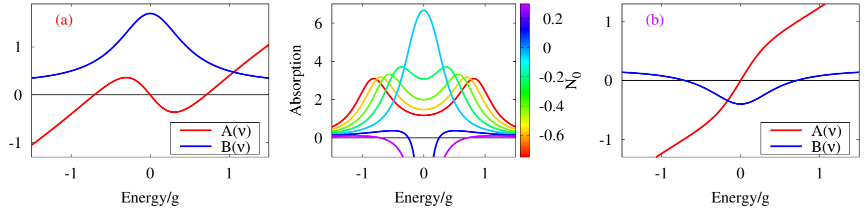

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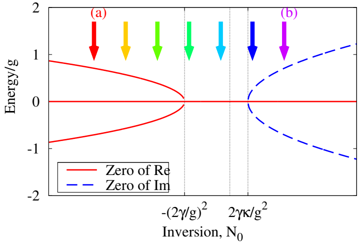
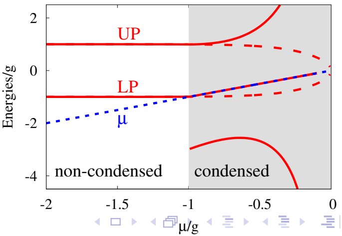
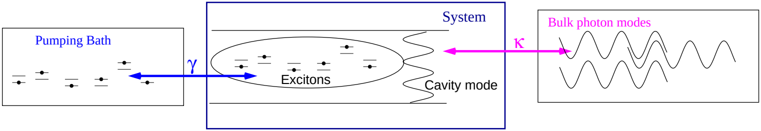
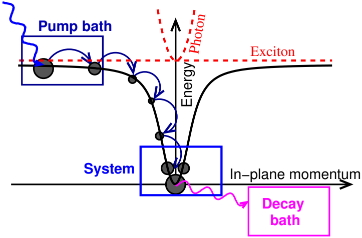


Equilibrium:



Non-equilibrium description: baths





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

- Decay bath: Empty ($\mu \to -\infty$)
- Pump bath: Thermal μ_B, T_B

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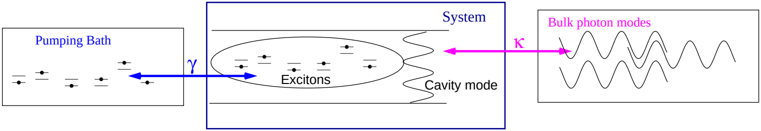

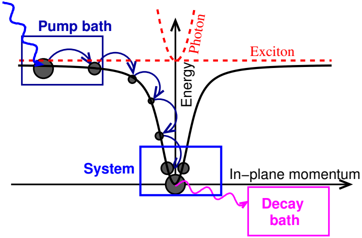

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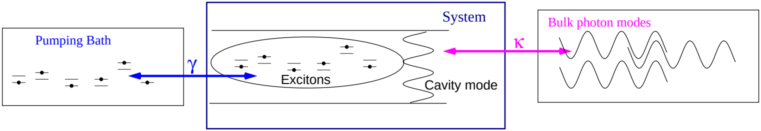

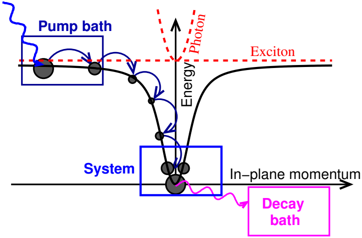

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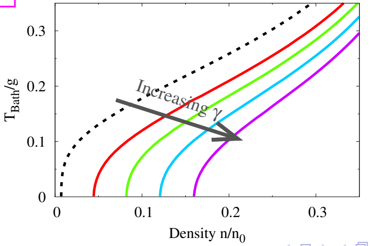
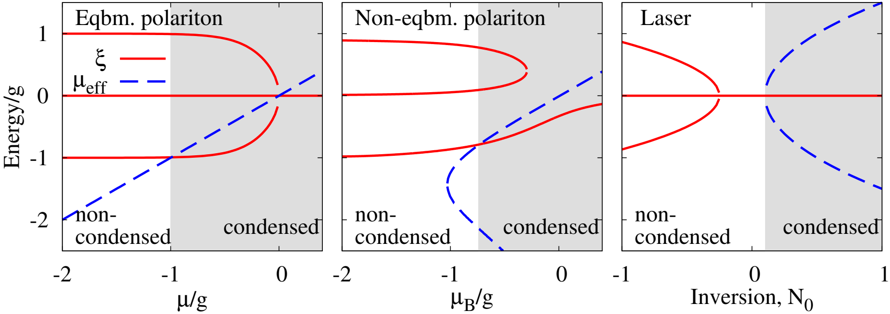
Mean field theory



Strong coupling and lasing — low temperature phenomenon



- Laser: Uniformly invert TLS
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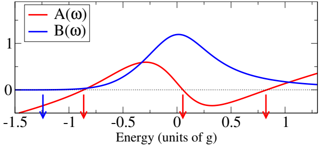
- If $T_B \gg \gamma \rightarrow$ Laser limit



[Szymanska *et al.* PRL ’06; Keeling *et al.* 1001.3338]

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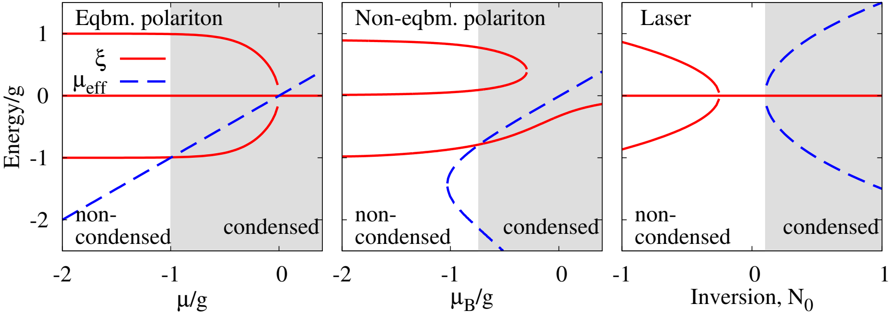

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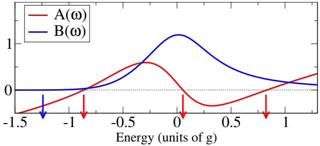

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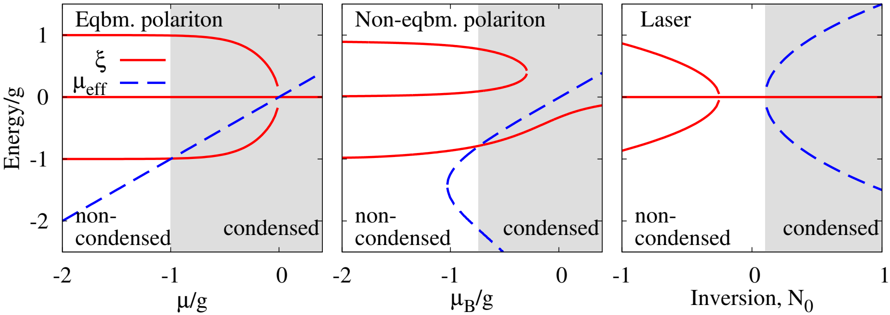

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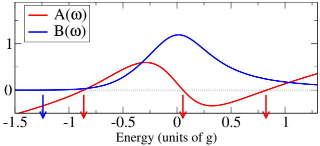

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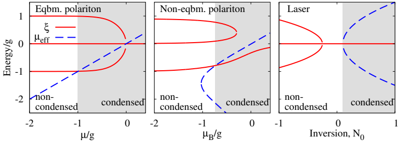
Coherence, inversion, strong-coupling

Polariton condensation:

- Inversionless
- **allows** strong coupling
- **requires** low $T \leftrightarrow$ condensation
- NB **NOT** thresholdless/single atom lasing.



- Circuit QED [Marthaler *et al.* PRL '11]
  - Noise-assisted
  - Off-resonant cavity
  - Emission/absorption $\Gamma^{\pm} \sim 2n_B(\pm\delta\omega) + 1$
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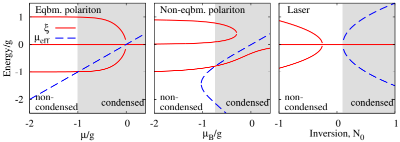

Related *weak-coupling* **inversionless** lasing:

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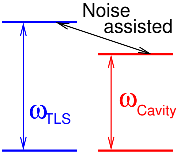

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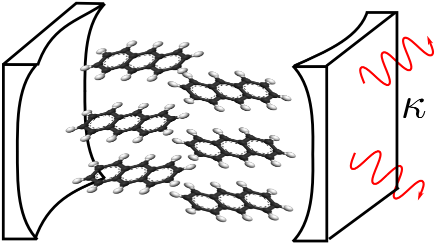
Organic polaritons: photon-exciton-phonon coupling

1. Polariton condensation
   - Introducton to polaritons
   - Non-equilibrium condensation vs lasing
   - Dicke model phase transition
2. Organic polaritons
   - Experiments and Dicke-Holstein model
   - Modified phase diagram and phonon sidebands
   - First order transitions
3. Photon condensation
   - Multimode rate equation
   - Critical properties from non-equilibrium model

Organic materials in microcavities

- What?
- Why?



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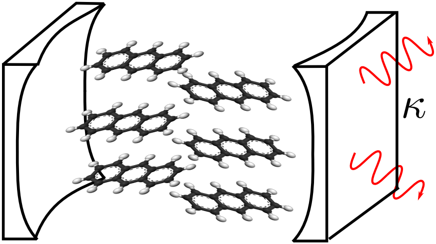

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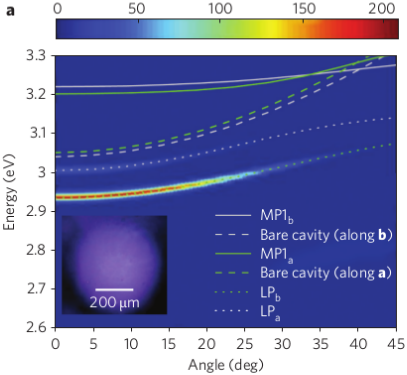

Polariton splitting: 0.1eV $\leftrightarrow$ 1000K.

[Kena Cohen and Forrest, Nat. Photon 2010]

Organic materials in microcavities

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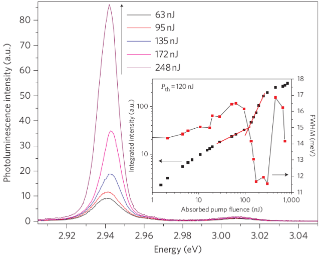

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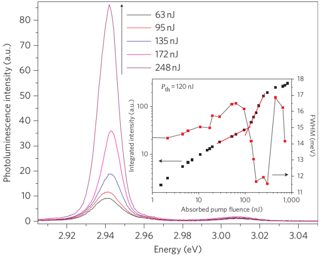

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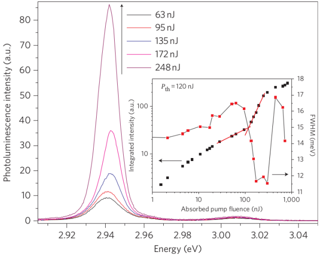

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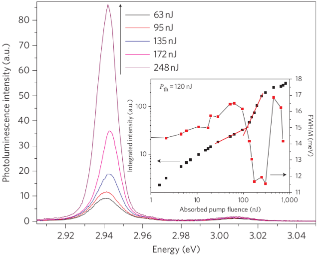

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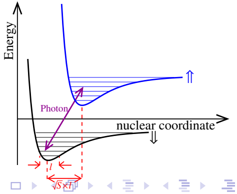
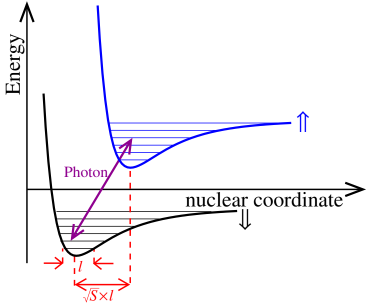


Dicke Holstein Model



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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
- Strong phonon coupling

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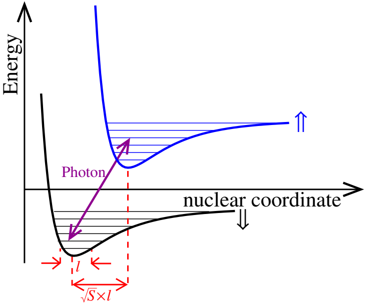

$$H = \omega\psi^\dagger\psi + \sum_\alpha \left[\epsilon S^z_\alpha + g\left(\psi S^+_\alpha + \psi^\dagger S^-_\alpha\right) + \Omega\left\{b^\dagger_\alpha b_\alpha + \sqrt{S}\left(b^\dagger_\alpha + b_\alpha\right)S^z_\alpha\right\}\right]$$

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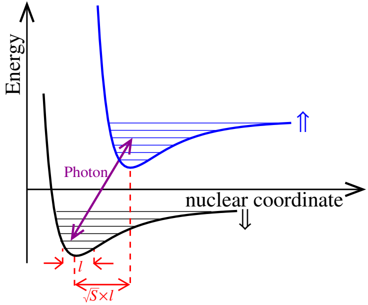

$$H = \omega\psi^\dagger\psi + \sum_\alpha \Big[\epsilon S^z_\alpha + g\left(\psi S^+_\alpha + \psi^\dagger S^-_\alpha\right) + \Omega\left\{b^\dagger_\alpha b_\alpha + \sqrt{S}\left(b^\dagger_\alpha + b_\alpha\right) S^z_\alpha\right\}\Big]$$

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

Questions?

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

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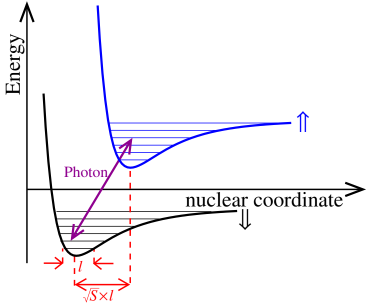

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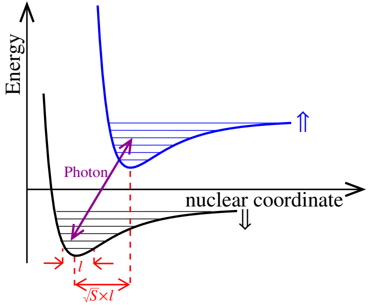

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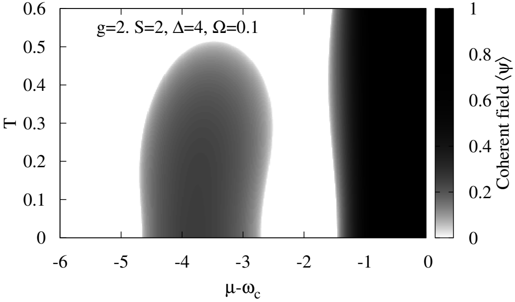

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  - Min μ at $T \sim 0.2$
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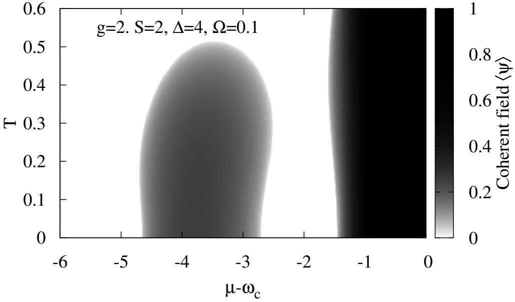

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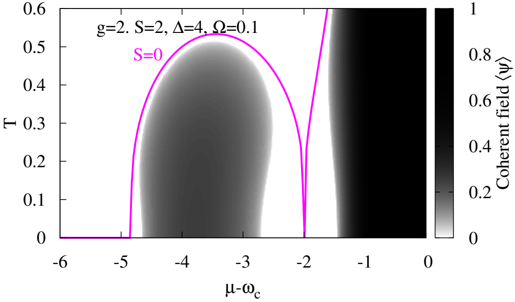

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Polariton spectrum — coupled oscillators

- Sidebands, $\epsilon - n\Omega$ coupled to photon
- Anticrossings of hybrid levels
- Coupling reduces with n
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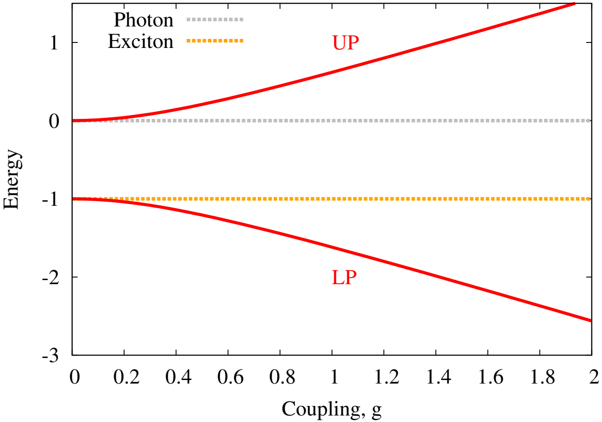

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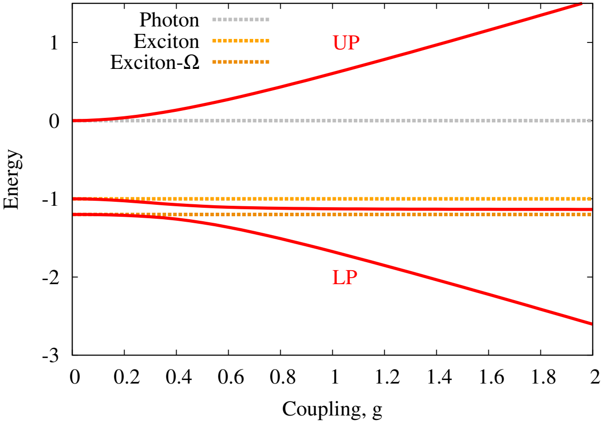

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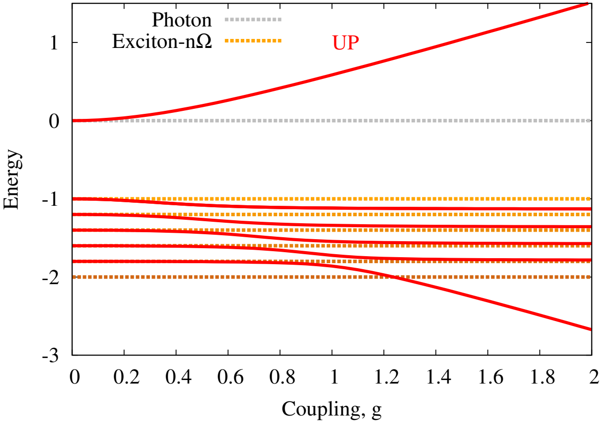

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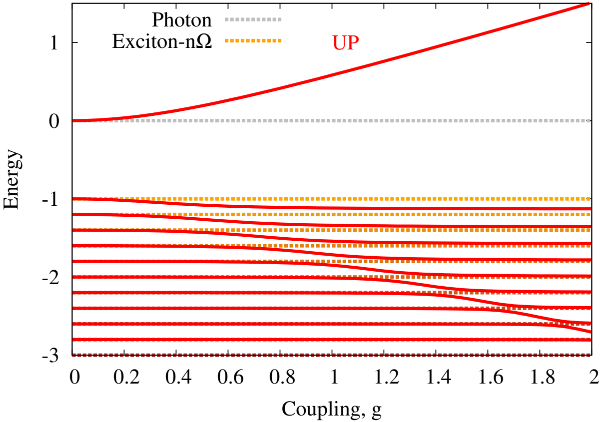

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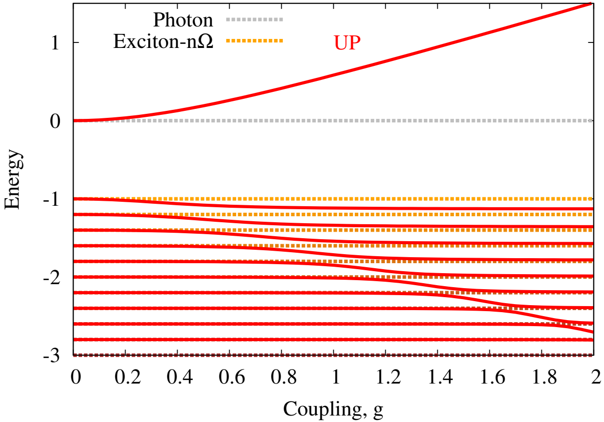

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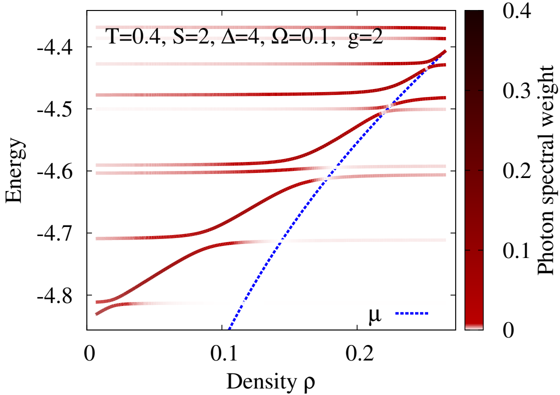

- Saturating 2LS: $g_{\text{eff}}^2 \sim g^2(1-2\rho)$
- What is nature of polariton mode?
- $\mathcal{D}(t) = -i\langle\psi^\dagger(t)\psi(0)\rangle, \qquad \mathcal{D}(\omega) = \sum_n \frac{Z_n}{\omega - \omega_n}$

[Cwik *et al.* arXiv:1303.3702]

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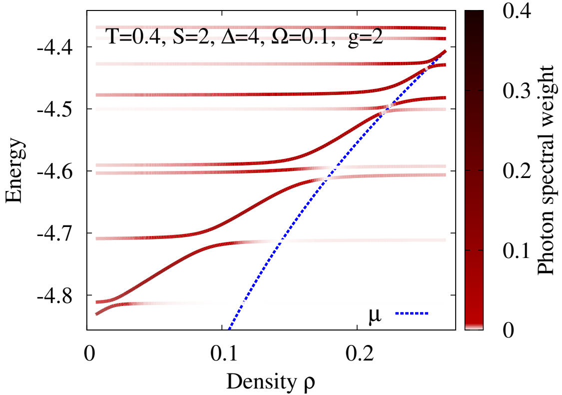

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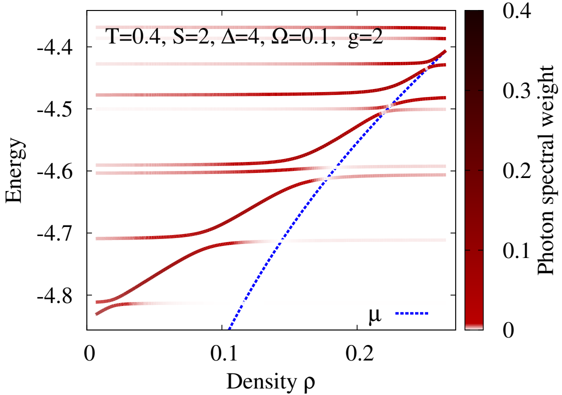

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Polariton spectrum: what condensed

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

- Optimal $T \sim 2\Omega$

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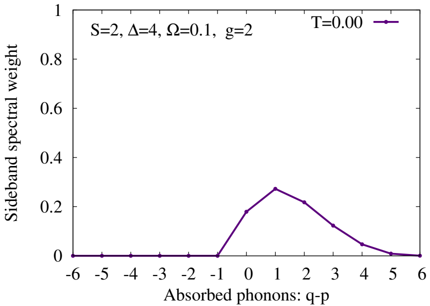

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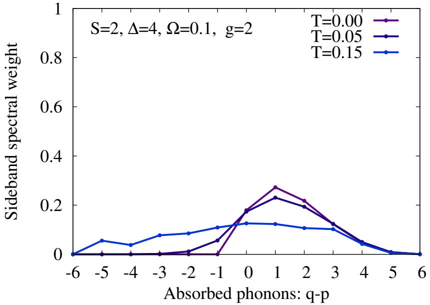

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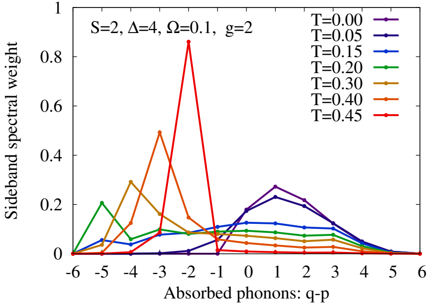

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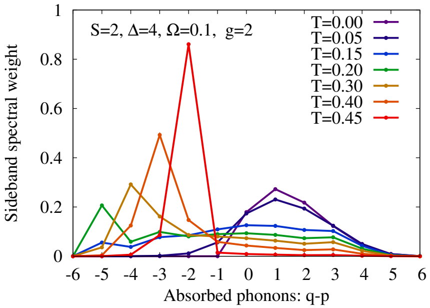

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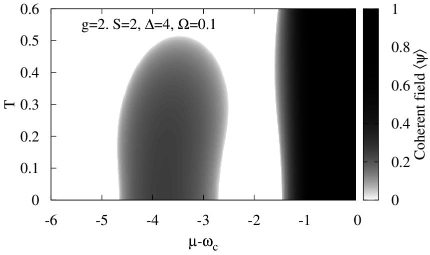

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Organic polaritons

Reorientation: Critical coupling strength

$$Ng^2 \tanh(\beta(\epsilon - \mu)) > (\omega - \mu)(\epsilon - \mu)$$

- At $\mu = \epsilon$
  - $g_c \to 0$ at $T = 0$
  - Superradiant bubble if $\epsilon < \omega$

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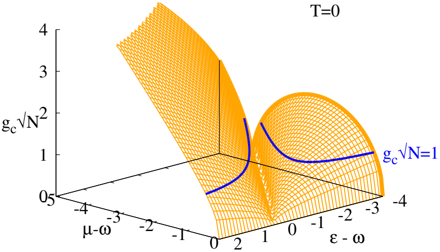

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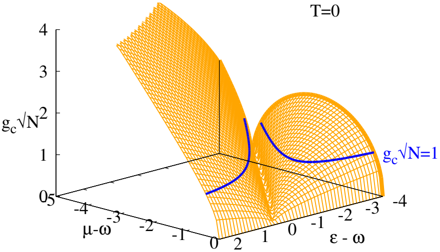

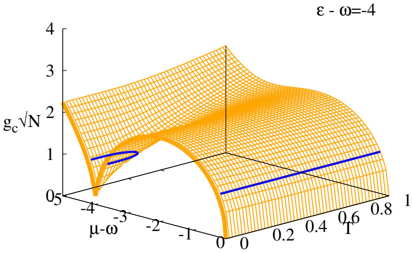

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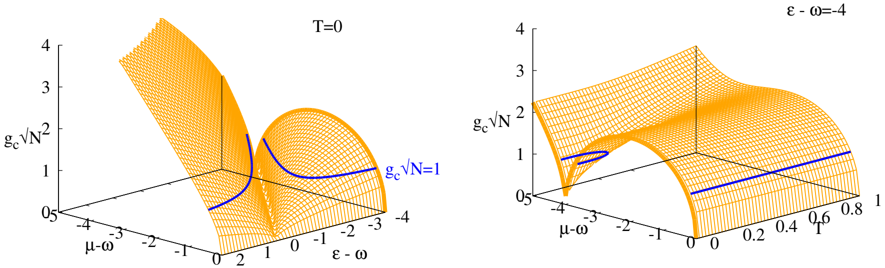

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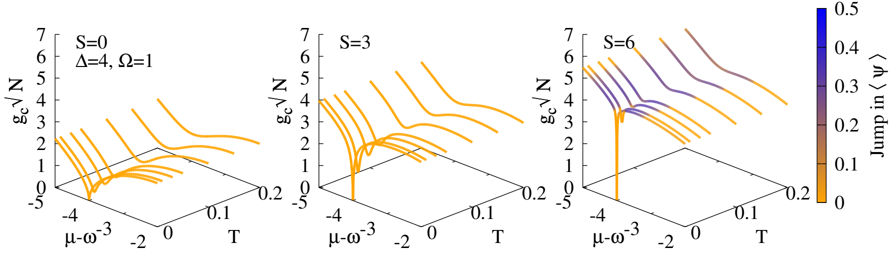

- Colors → Jump of $\langle\psi\rangle$

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 $S^\pm$

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

- Different optimal phonon displacements, $\sim \sqrt{S}$
- Reduced $g_{\text{eff}} \sim g \times \exp(-S/2)$
- For non-zero ψ, variational approx:
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Collective polaron formation

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

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

Effective 2LS energy in field:

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

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

[Cwik *et al.* arXiv:1303.3702]

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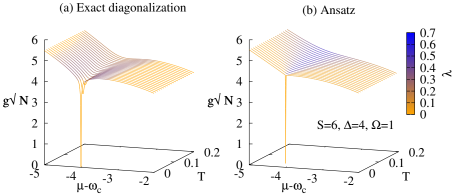
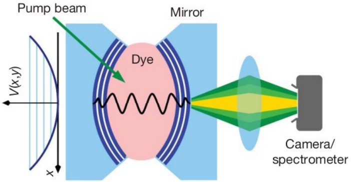


Polariton and photon Condensation

Photon BEC experiments



- Dye filled microcavity
- Pump at angle
- No strong coupling
- Condensation:
  - Far below inversion
  - Thermalised emission spectrum

[Klaers et al, Nature, 2010]

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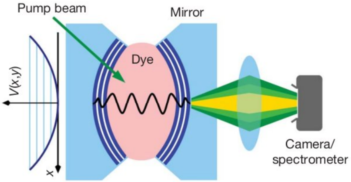

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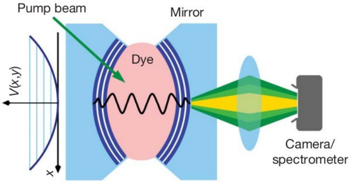

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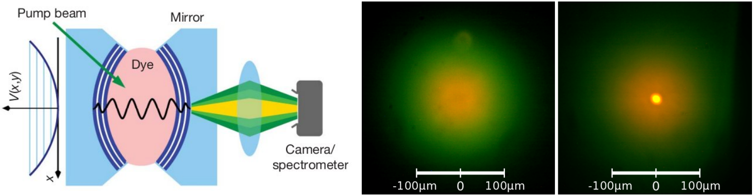

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Relation to dye laser

- No electronic inversion
- No strong coupling

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

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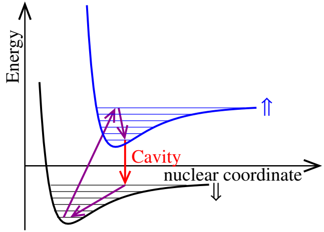
4 Level Dye Laser



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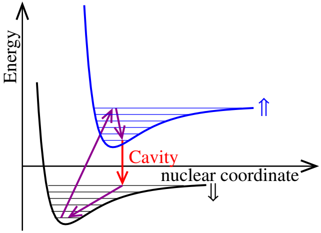

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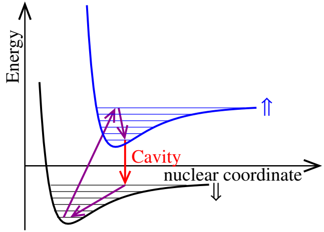

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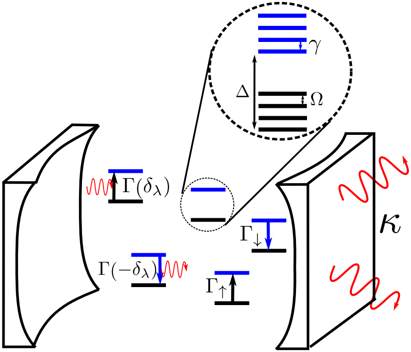
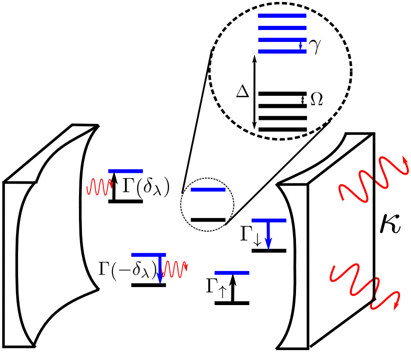


Modelling

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

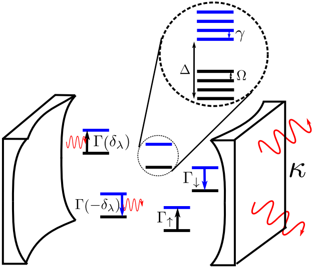
- Consider harmonic cavity modes
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Add local vibrational mode
- Integrate out phonon effects
  - Polaron transform
  - Perturbation theory in g



Modelling

Rate equation

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



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

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

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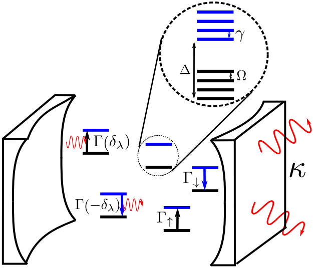

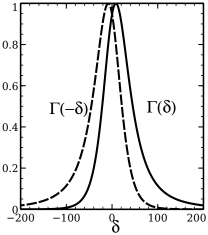

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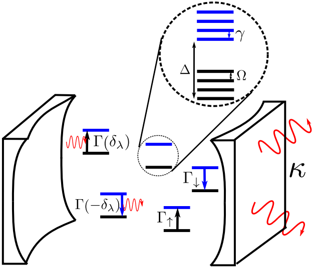

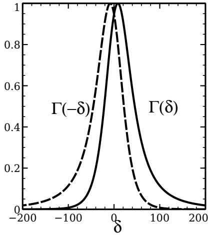

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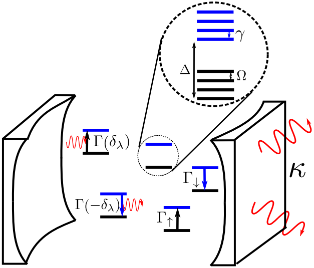

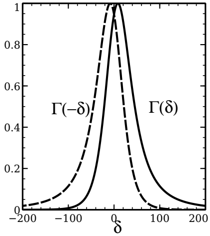

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- Rate equation — include spontaneous emission
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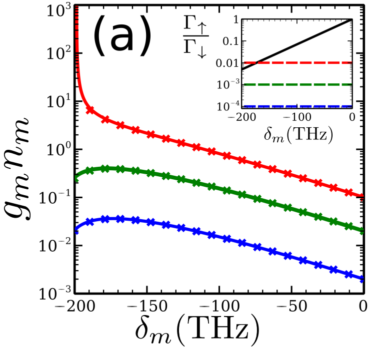

Low loss: Thermal

[Kirton & JK arXiv:1303.3459]

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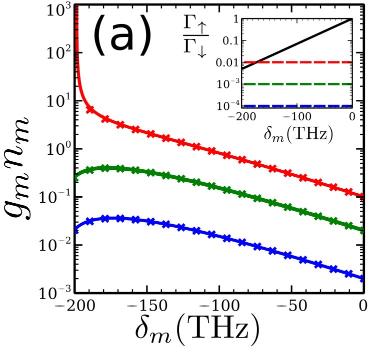

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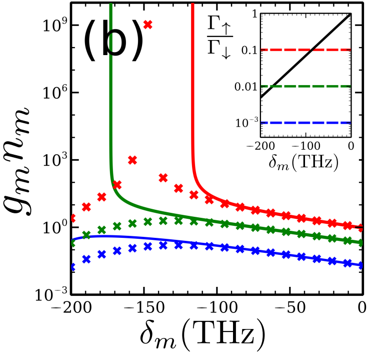
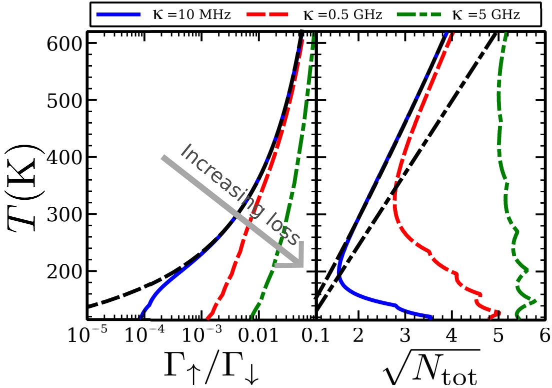


High loss → Laser

Threshold condition



Compare threshold:

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

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

[Kirton & JK arXiv:1303.3459]

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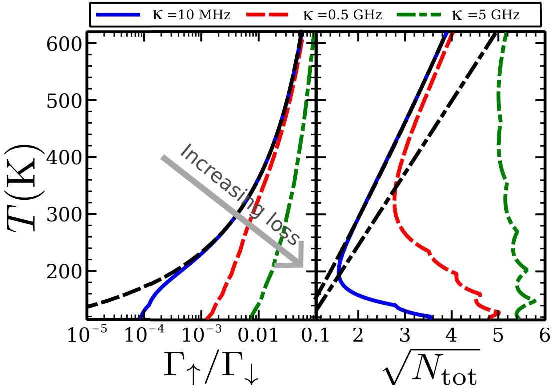

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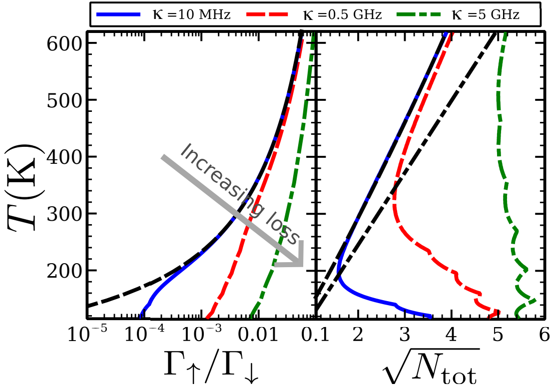

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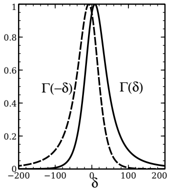

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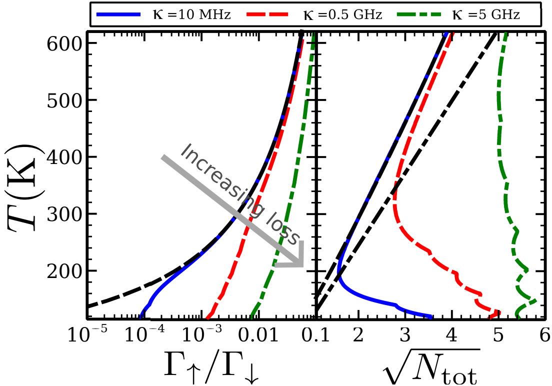

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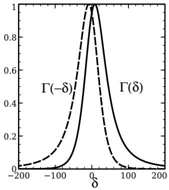

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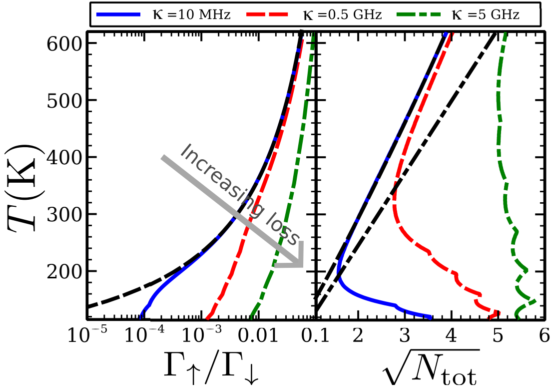

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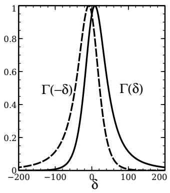

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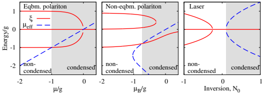
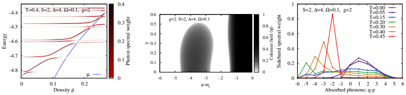
Summary

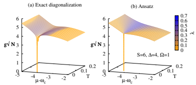
- Polariton condensation vs lasing



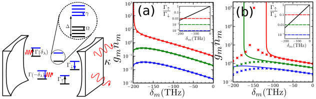
- Reentrance and phonon assisted transition



- First order transitions at very strong coupling



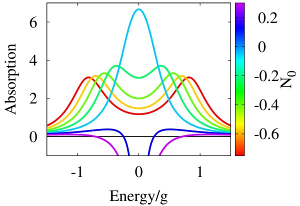
- Photon condensation and thermalisation



Extra slides

4 Retarded Green's function for laser

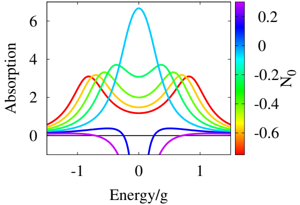
Maxwell-Bloch Equations: Retarded Green's function



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

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

Maxwell-Bloch Equations: Retarded Green's function



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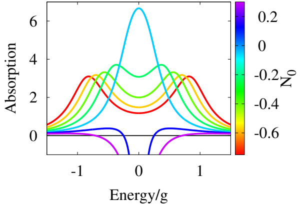

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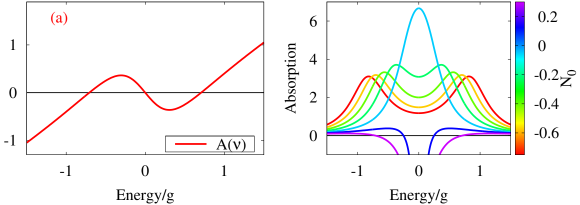

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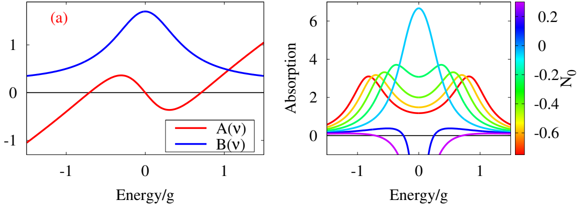

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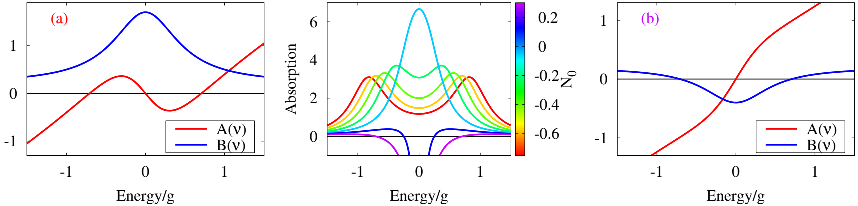

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