

# Non-equilibrium coherence in light-matter systems

Condensation, lasing, superradiance and more

Jonathan Keeling



University  
of  
St Andrews

600  
YEARS



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# Acknowledgements

GROUP:



COLLABORATORS: Szymanska, Littlewood, Simons, Bhaseen,  
Schmidt, Blatter, Türeci, Krüger

EXPERIMENT: Houck, Wallraff, Fink, Mylnek

FUNDING:



Engineering and Physical Sciences  
Research Council



# Coupling many atoms to light

**Old question:** *What happens to radiation when many atoms interact “collectively” with light.*

**Superradiance** — dynamical and steady state.

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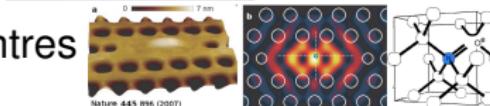
**Superradiance** — dynamical and steady state.

**New relevance**

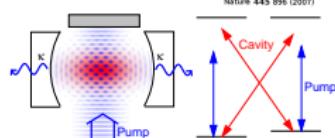
- Superconducting qubits



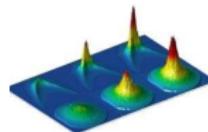
- Quantum dots & NV centres



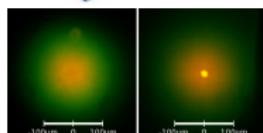
- Ultra-cold atoms



- Rydberg atoms/polaritons



- Microcavity Polaritons



- Photon condensation

# Dicke effect: Enhanced emission

PHYSICAL REVIEW

VOLUME 93, NUMBER 1

JANUARY 1, 1954

## Coherence in Spontaneous Radiation Processes

R. H. DICKE

*Palmer Physical Laboratory, Princeton University, Princeton, New Jersey*



$$H_{\text{int}} = \sum_{k,i} g_k \left( \psi_k^\dagger S_i^- e^{-i\mathbf{k}\cdot\mathbf{r}_i} + \text{H.c.} \right)$$

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Collective decay:

$$\frac{d\rho}{dt} = -\frac{\Gamma}{2} [S^+ S^- \rho - S^- \rho S^+ + \rho S^+ S^-]$$

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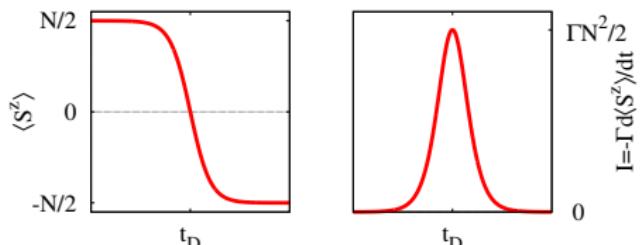


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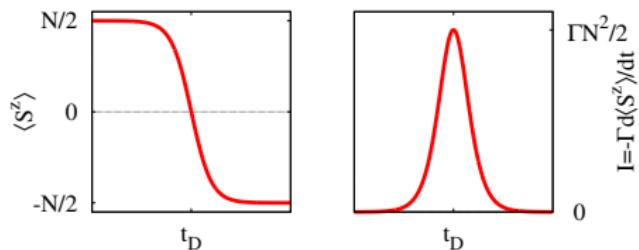


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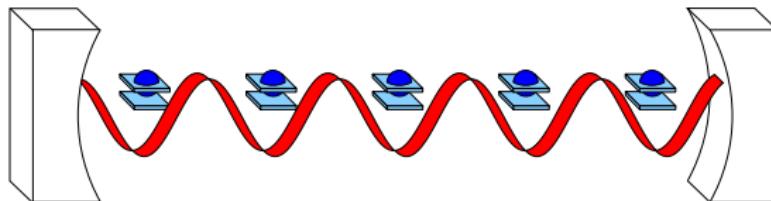
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**Problem:** dipole interactions dephase. [Friedberg et al, Phys. Lett. 1972]

# Collective radiation **with** a cavity: Dynamics

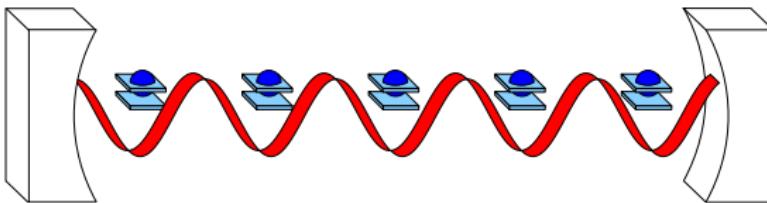


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Single cavity mode: oscillations

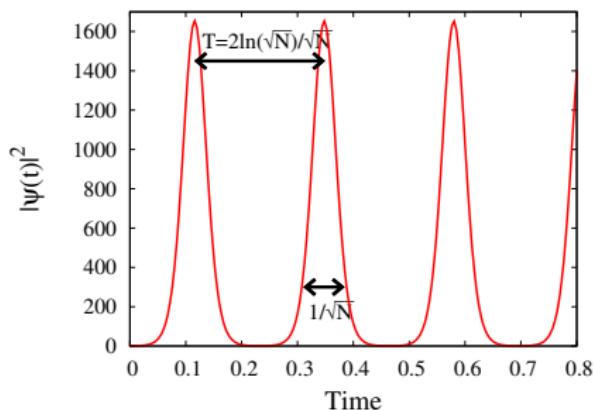
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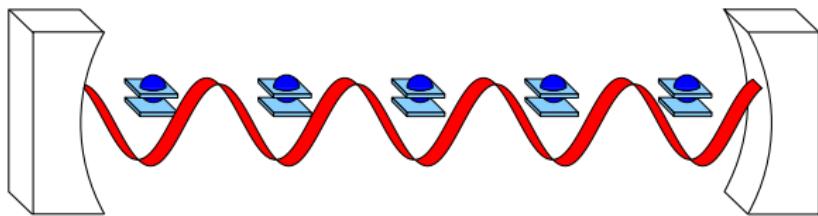
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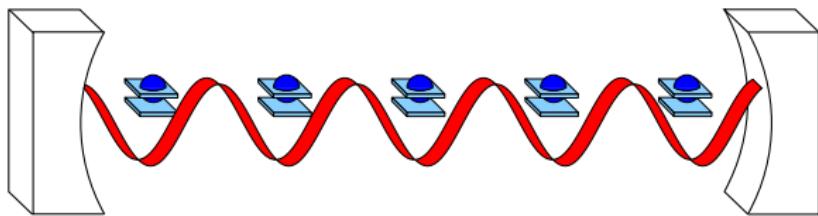
$$H = \omega \psi^\dagger \psi + \omega_0 S^z + g (\psi^\dagger S^- + \psi S^+).$$

Condon state (Pulse sequence)

- Small  $g$ , min at  $\lambda, \eta = 0$

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

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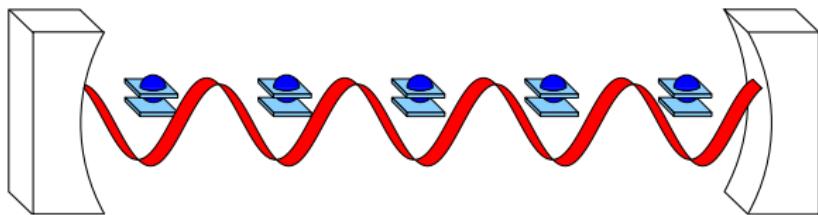
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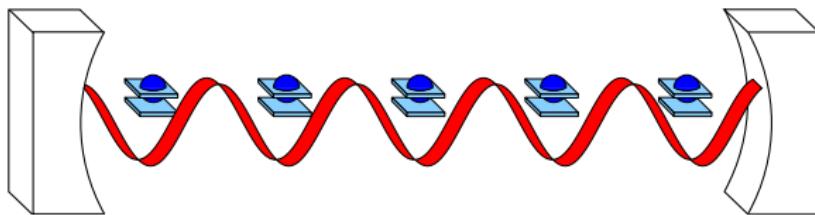
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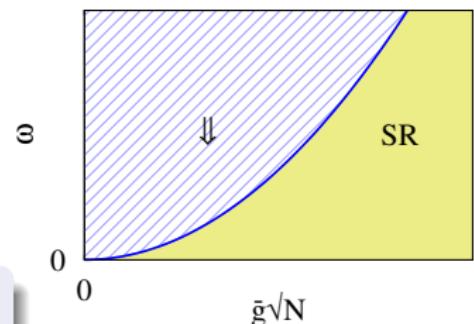
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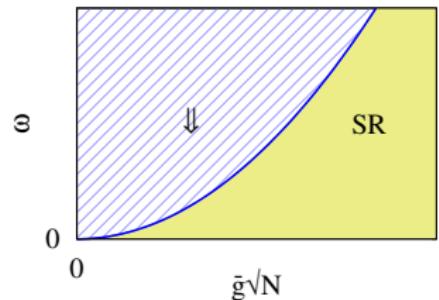
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- $T > 0$ , minimum free energy if

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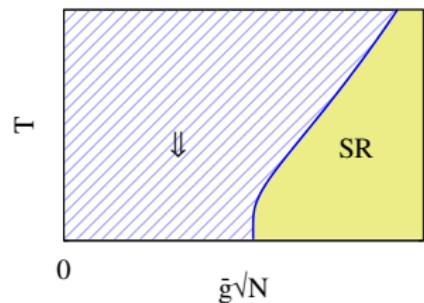
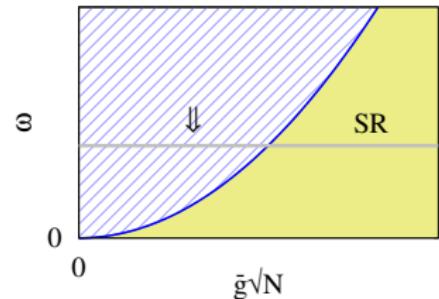
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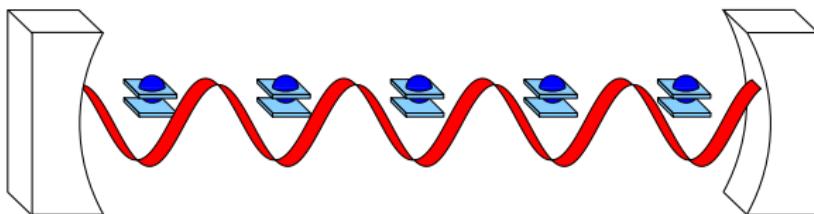
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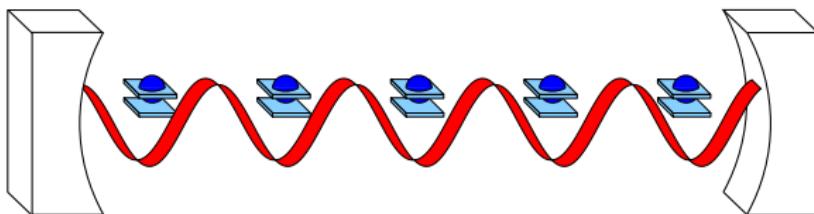
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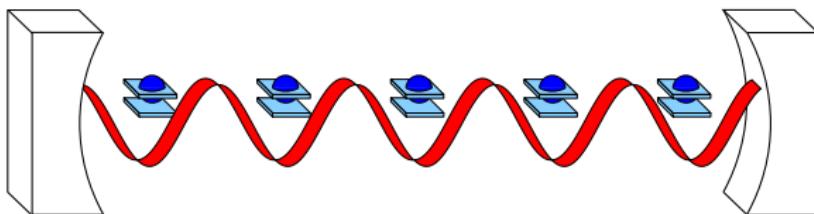
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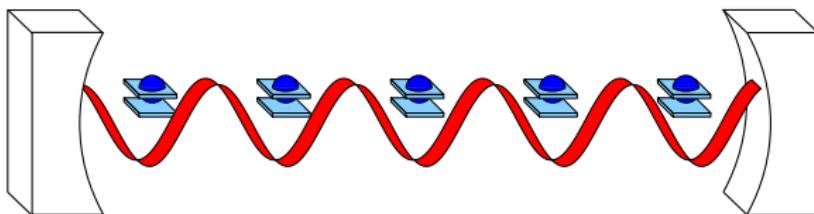
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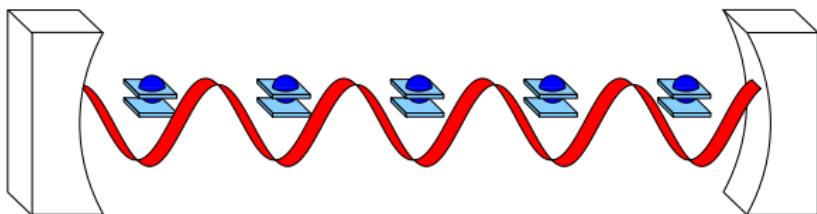
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But Thomas-Reiche-Kuhn sum rule states:  $g^2/\omega_0 < 2\zeta$ . **No transition**  
[Rzazewski *et al* PRL '75]

# Dicke phase transition: ways out

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

- Interpretation:  
    • Ferroelectric transition in D-r gauge.  
        [JK, JPCM '07; Vukics & Demokos PRA 2012]
- Circuit QED [Nataf and Ciuti, Nat. Comm. '10; Viehmann et al. PRL '11]
- Grand canonical ensemble:
  - If  $H \rightarrow H - \mu(S^z + \phi^* \phi)$ , need only:  
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e.g. Raman scheme:  $\omega_R \ll \omega$ .  
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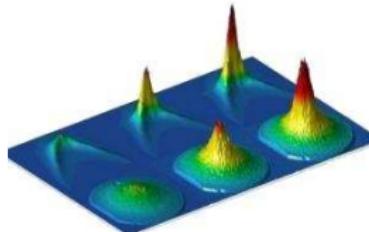
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→ Dicke phase transition

→ e.g., Raman scattering,  $\omega \ll \omega_0$

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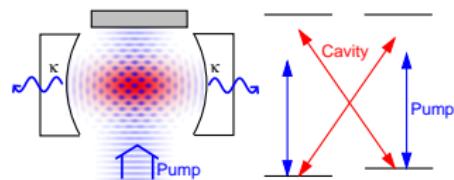
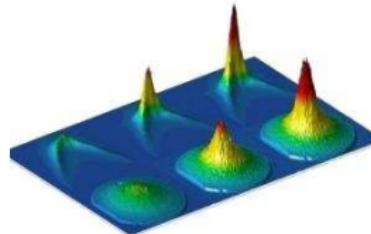
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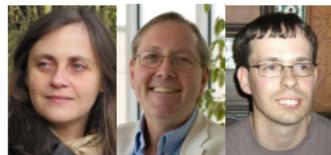
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  - Non-equilibrium condensation vs lasing
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  - Disorder
- 4 Phase transitions with SC qubits
  - Pumping without symmetry breaking
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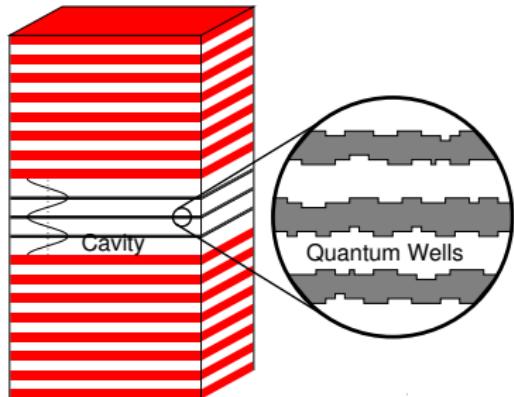
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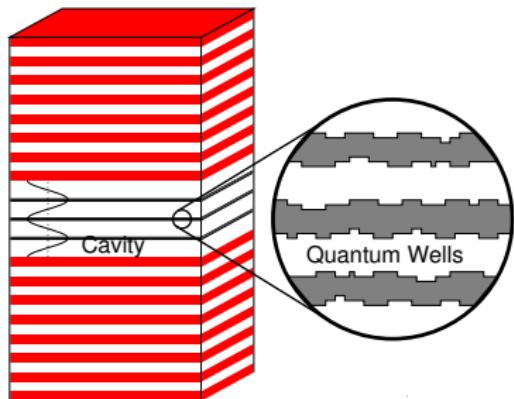
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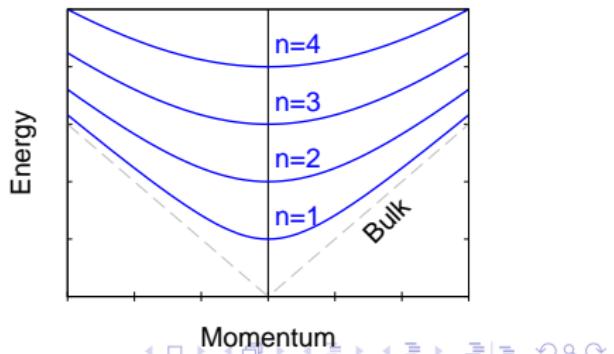


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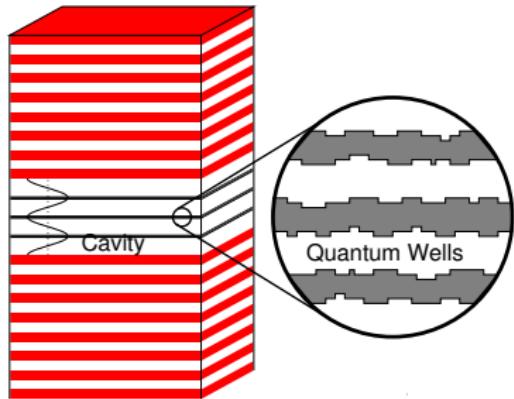


Cavity photons:

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



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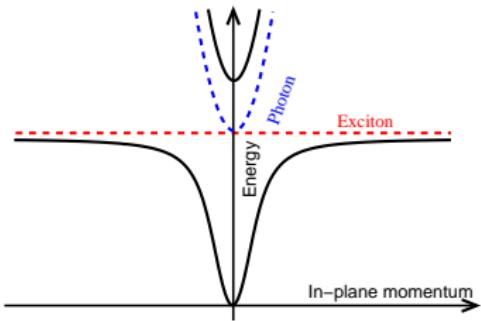


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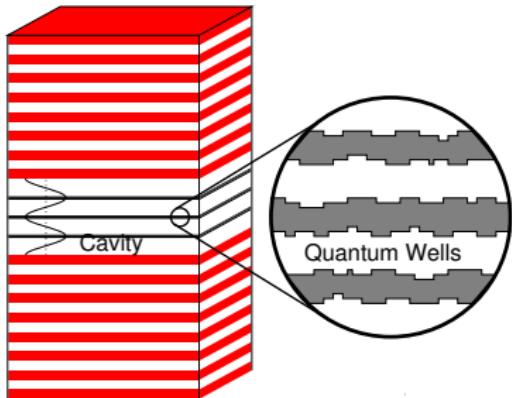
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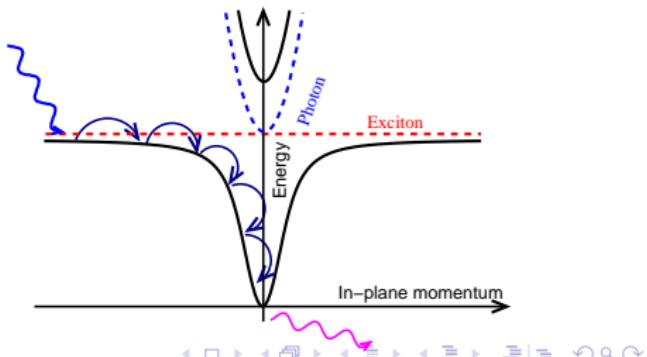


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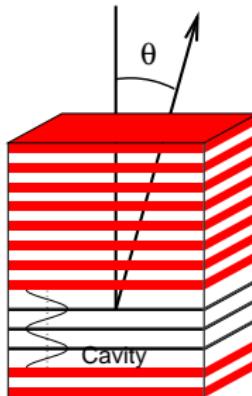
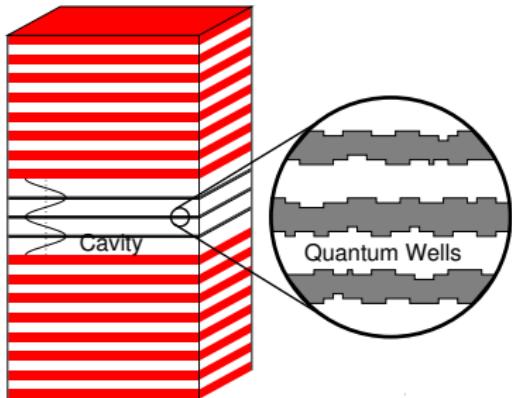
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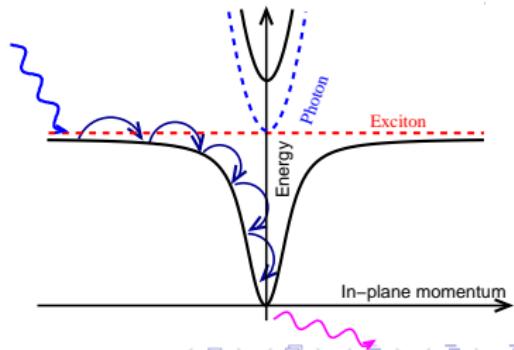


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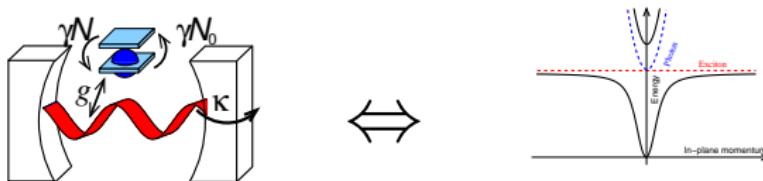
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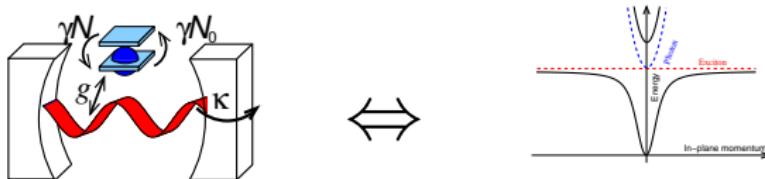
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Dicke model:

$$H_{\text{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi_{\mathbf{k}}^\dagger \psi_{\mathbf{k}} + \sum_{\alpha} \left[ \epsilon S_{\alpha}^z + \frac{(g_{\alpha, \mathbf{k}} \psi_{\mathbf{k}} S_{\alpha}^+ + \text{H.c.})}{\sqrt{A}} \right]$$

# Polariton model and equilibrium results

Localised excitons, propagating photons

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

Self-consistent polarisation and field

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

# Polariton model and equilibrium results

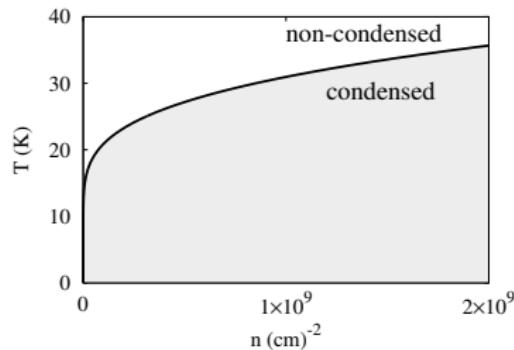
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Phase diagram:



# Polariton model and equilibrium results

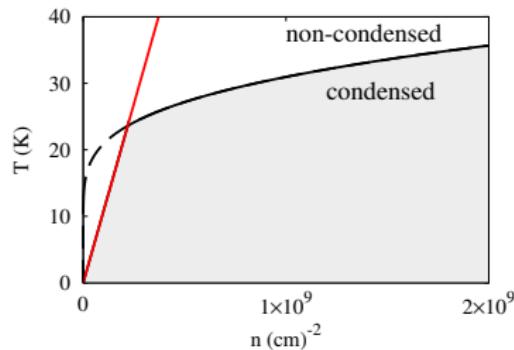
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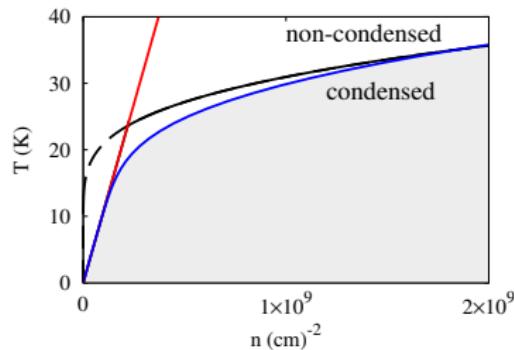
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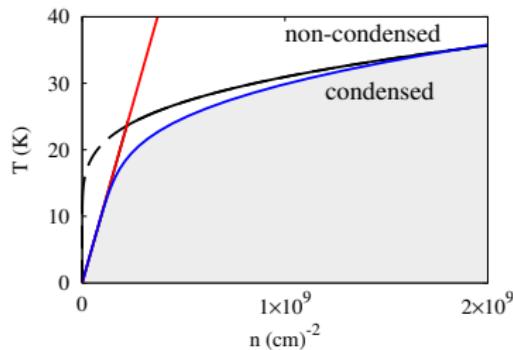
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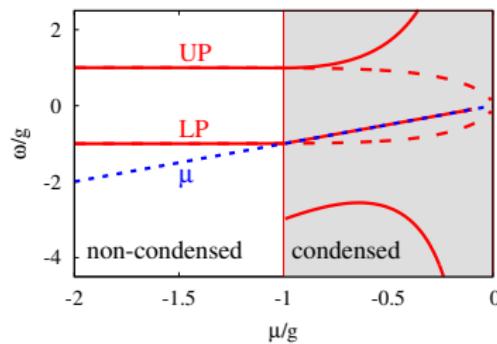
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Phase diagram:



Modes (at  $k = 0$ )



# Simple Laser: Maxwell Bloch equations

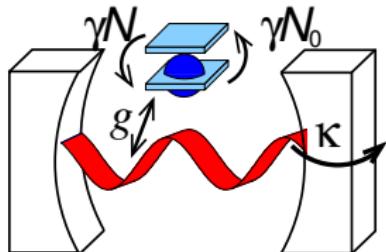
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Maxwell-Bloch eqns:  $P = -i\langle S^- \rangle$ ,  $N = 2\langle S^z \rangle$

$$\partial_t\psi = -i\omega\psi - \kappa\psi + \sum_{\alpha} g_{\alpha}P_{\alpha}$$

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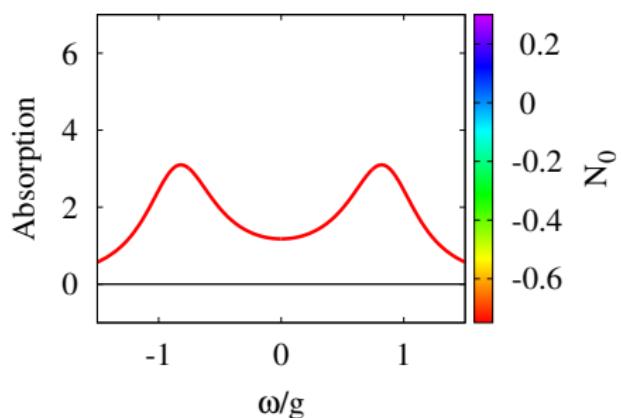
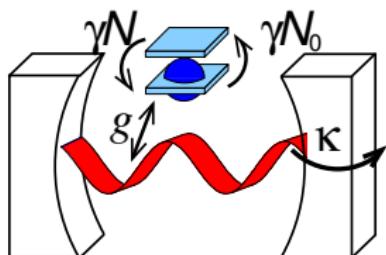
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• Strong coupling.  $\kappa, \gamma < g\sqrt{n}$

before lasing

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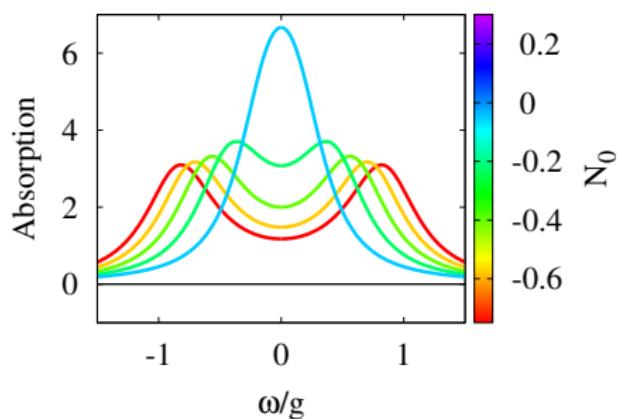
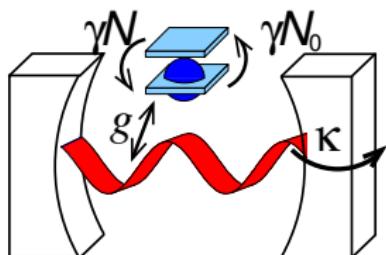
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- Inversion causes collapse before lasing

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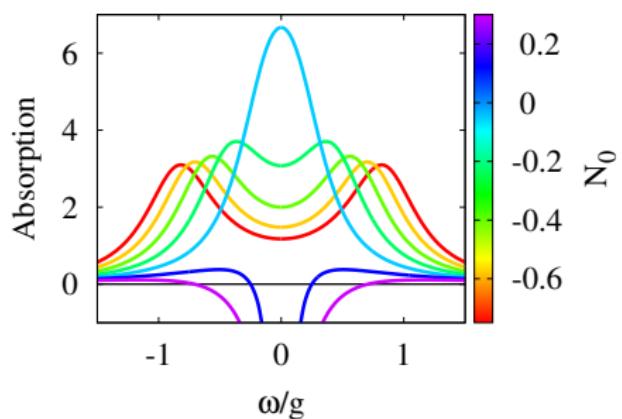
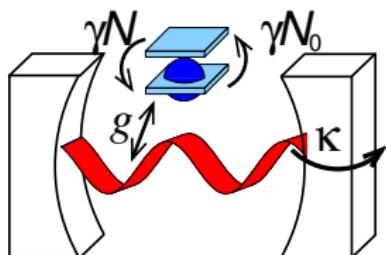
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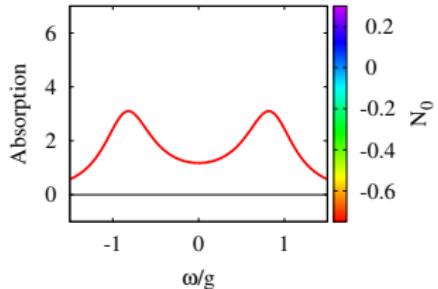
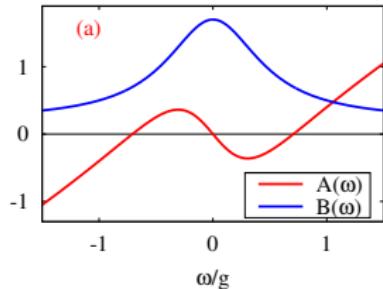
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# Poles of Retarded Green's function and gain

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

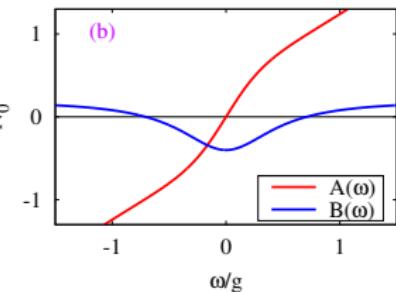
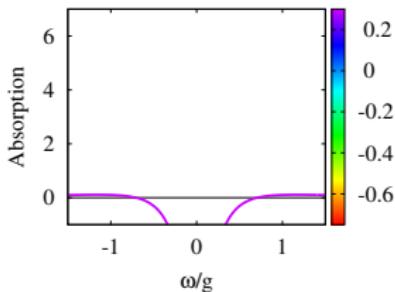
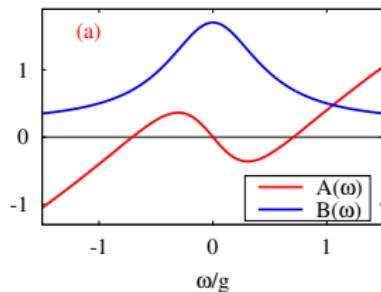
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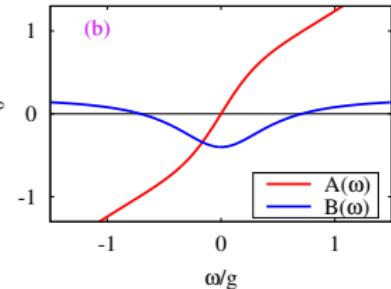
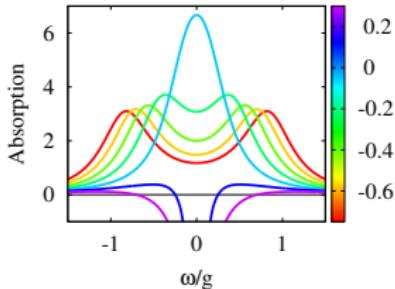
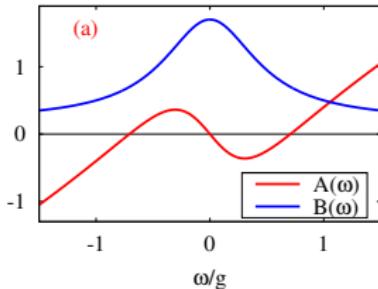
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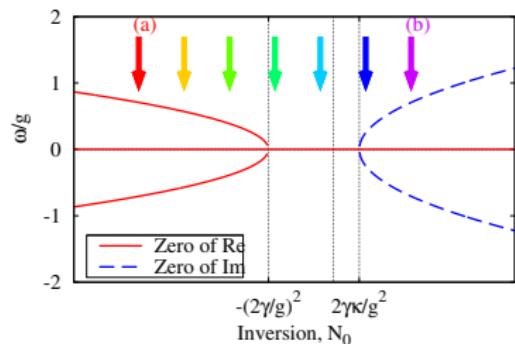


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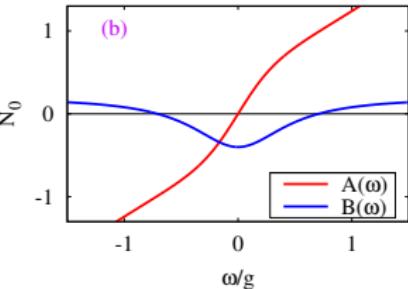
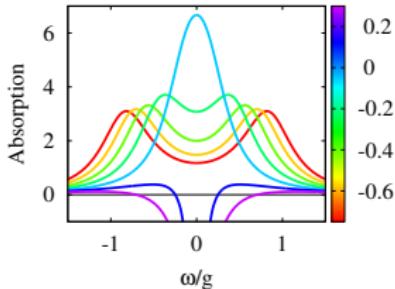
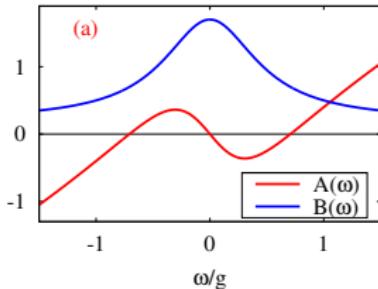


Laser:

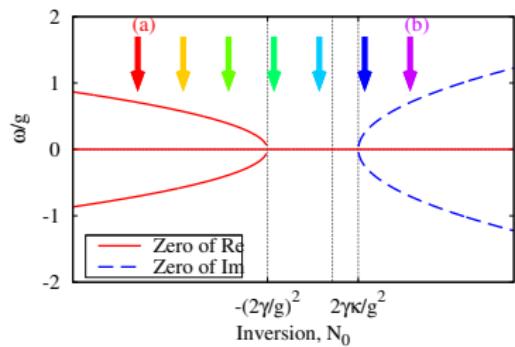


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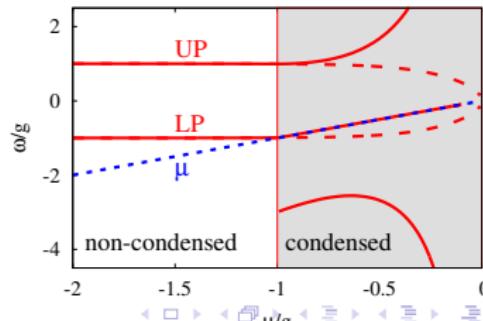
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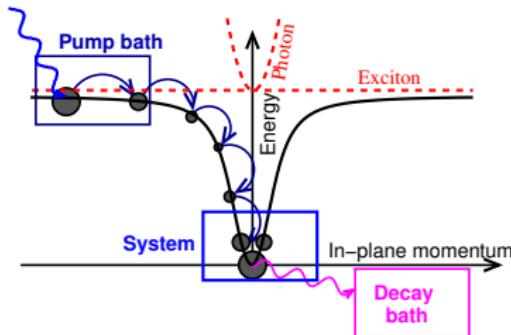
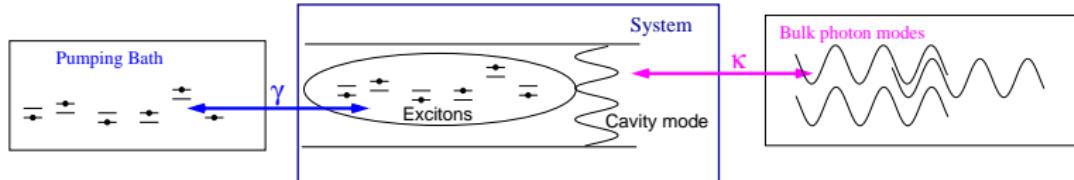
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Equilibrium:



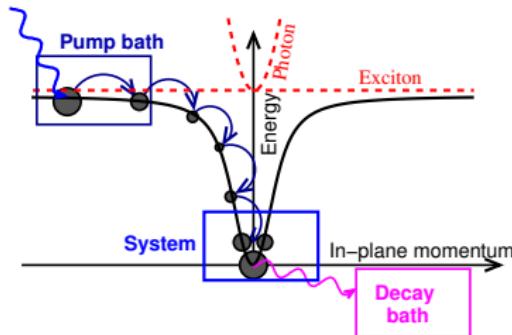
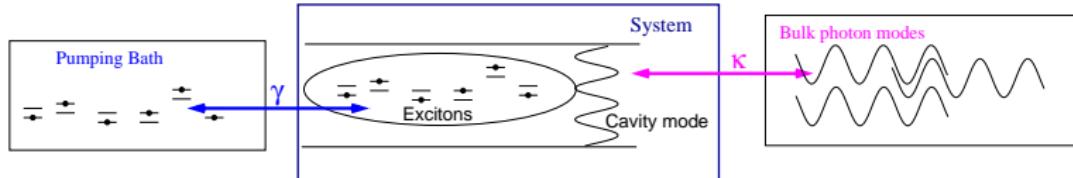
# Non-equilibrium description: baths



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

→ Decay bath: Empty ( $\mu \rightarrow -\infty$ )  
→ Pump bath: Thermal  $\mu_p, T_p$

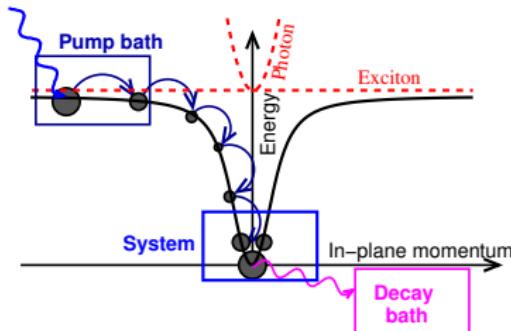
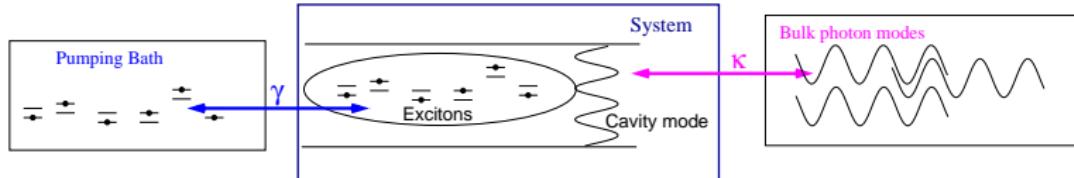
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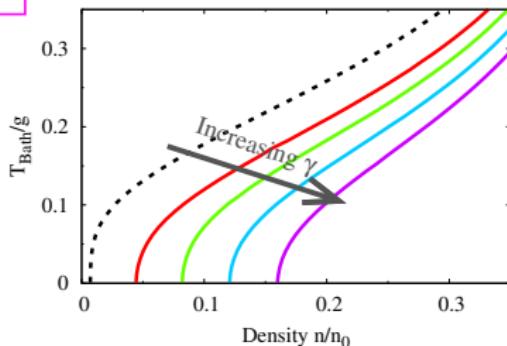
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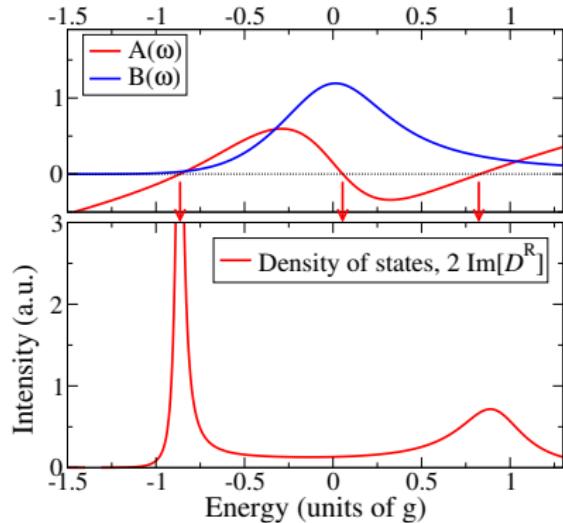
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## Mean field theory

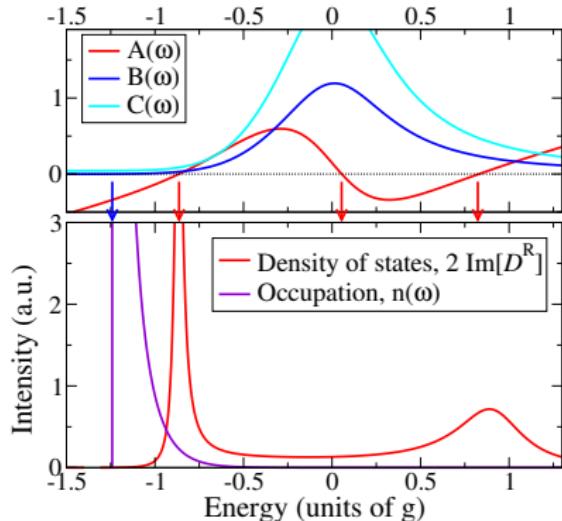


# Stability and evolution with pumping



$$[D^R(\nu)]^{-1} = A(\nu) + iB(\nu)$$

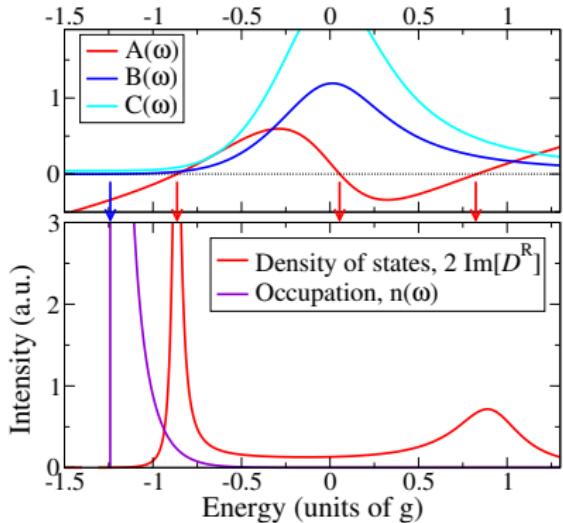
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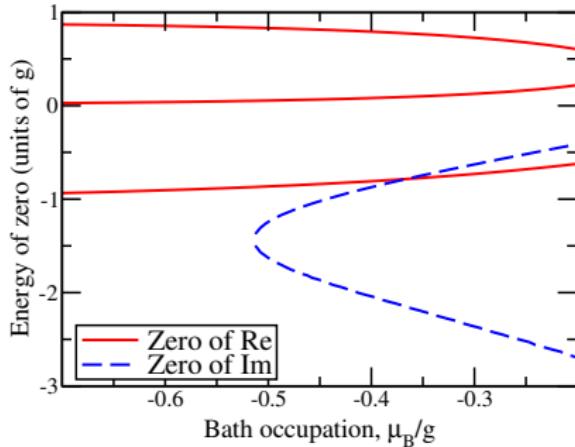
$$2n(\nu) + 1 = \frac{iD^K(\nu)}{-2\Im[D^R(\nu)]} = \frac{C(\nu)}{2B(\nu)}$$

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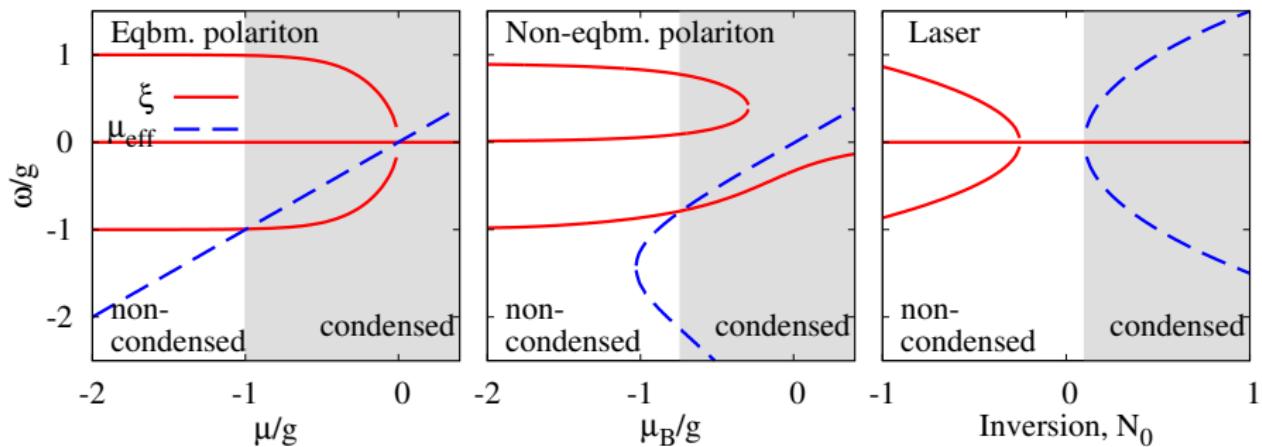


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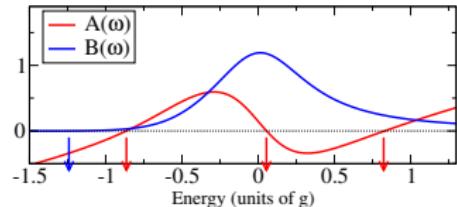
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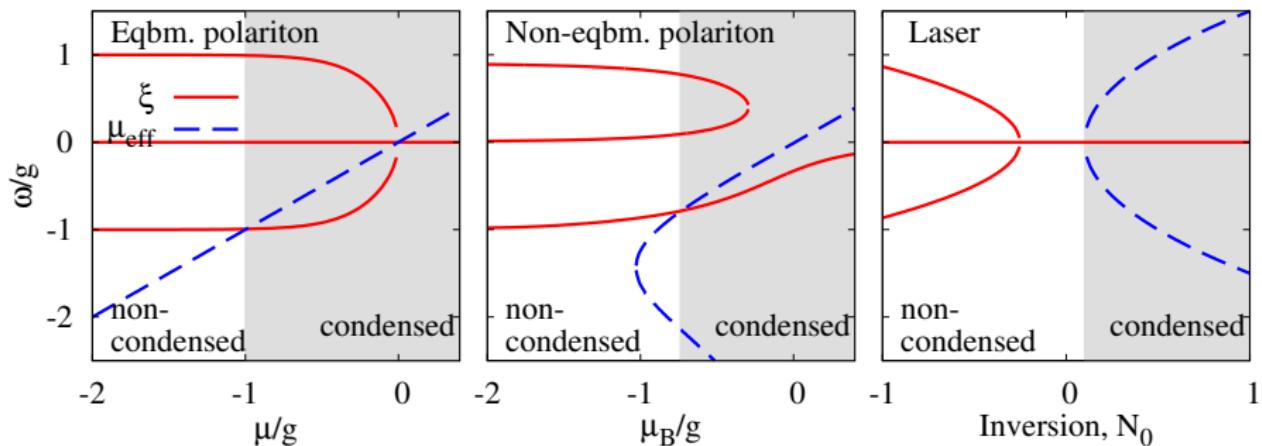
# Strong coupling and lasing — low temperature phenomenon



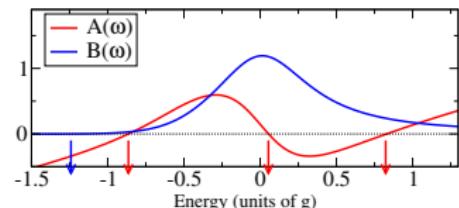
- Laser: Uniformly invert TLS



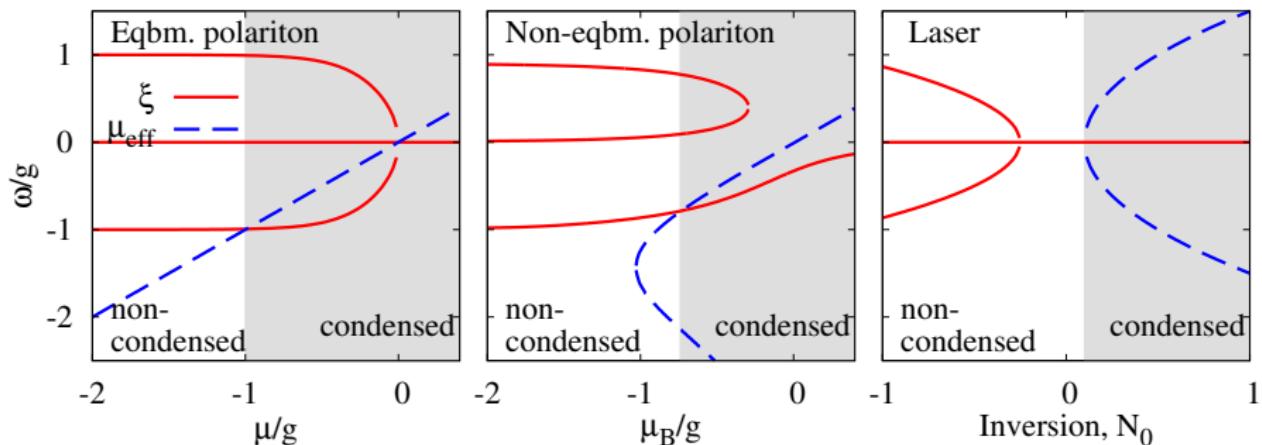
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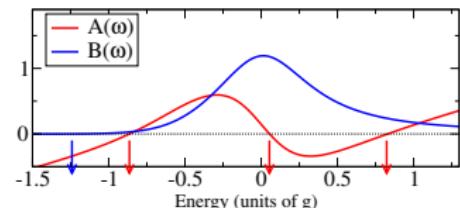
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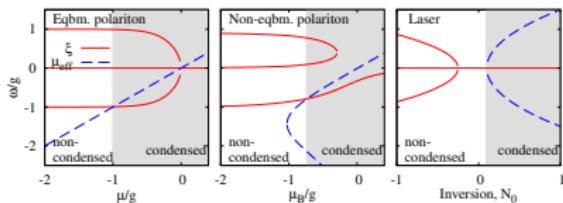
- Laser: Uniformly invert TLS
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- If  $T_B \gg \gamma \rightarrow$  Laser limit



# Coherence, inversion, strong-coupling

Polariton condensation:

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



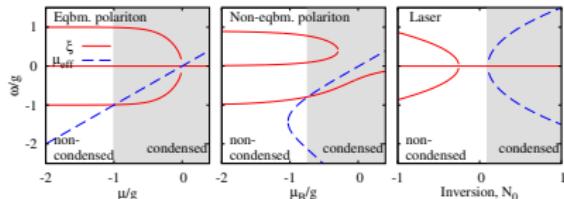
→ Circuit QED [Marthaler et al. PRL '11]

- Noise-assisted
- Off-resonant cavity
- Emission/absorption  $P^+ \sim 2n_c(\pm\delta\omega) + 1$
- Low  $T \rightarrow$  inversionless threshold

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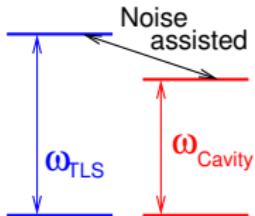
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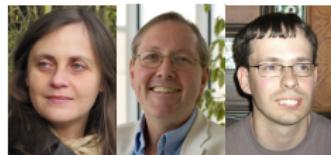
Related *weak-coupling inversionless* lasing:

- Circuit QED [Marthaler *et al.* PRL '11]



- ▶ Noise-assisted
- ▶ Off-resonant cavity
- ▶ Emission/absorption  $\Gamma^{\pm} \sim 2n_B(\pm\delta\omega) + 1$
- ▶ Low  $T \rightarrow$  inversionless threshold

# Polariton and photon Condensation



1 Dicke model and superradiance

2 Polariton and photon condensation

- Polaritons
- Non-equilibrium condensation vs lasing
- Photon condensation

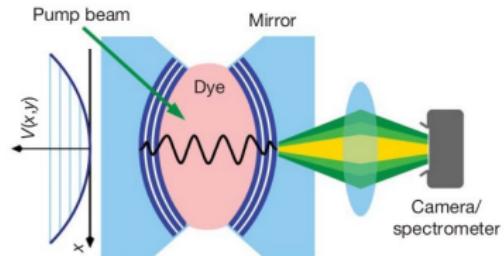
3 Jaynes Cummings Hubbard model

- JCHM vv Dicke
- Coherently driven array
- Disorder

4 Phase transitions with SC qubits

- Pumping without symmetry breaking
- Collective dephasing

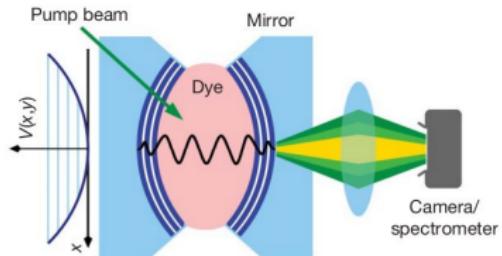
# Photon BEC experiments



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

[Klaers et al, Nature, 2010]

# Photon BEC experiments



- Dye filled microcavity
- Pump at angle

→ Strong coupling

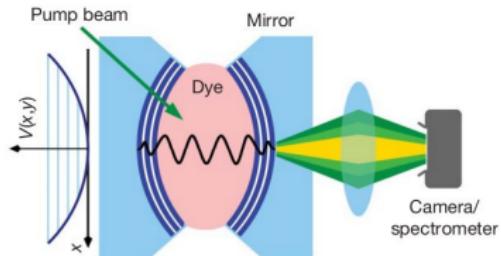
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→ Thermalised emission spectrum

[Klaers et al, Nature, 2010]

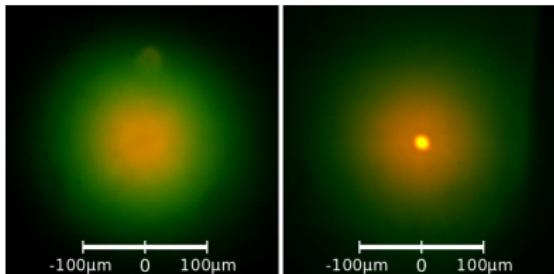
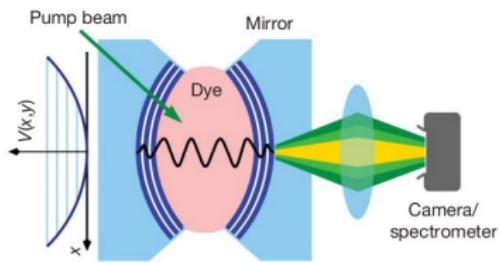
# Photon BEC experiments



- 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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# Modelling

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

]

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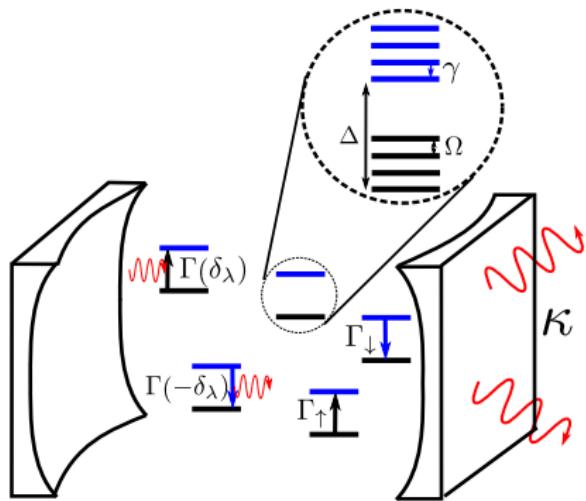
- Add local vibrational mode
- Integrate out phonon effects
  - Polaron transform
  - Perturbation theory in  $g$

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

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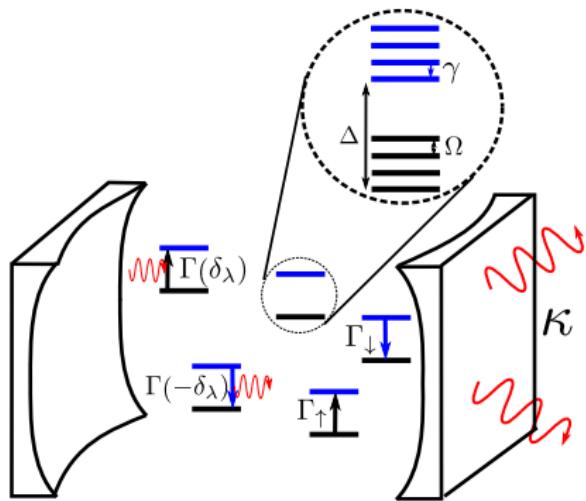
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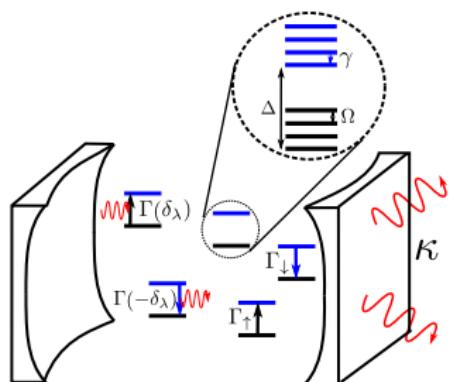
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# Modelling

## Rate equation

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



$$\Gamma(-\delta) \simeq \Gamma(-\delta) e^{-\delta/\Delta}$$

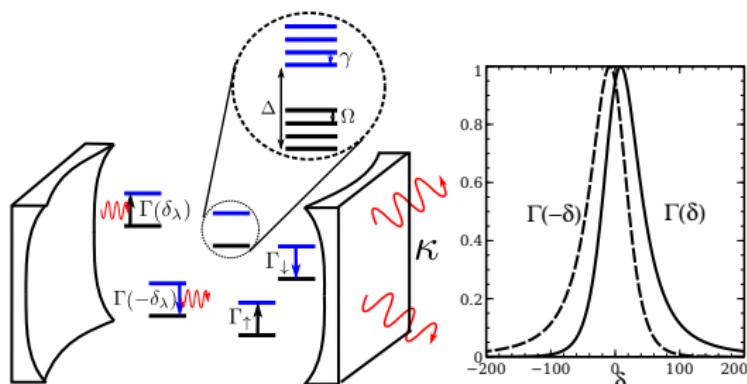
$\Gamma \rightarrow 0$  at large  $\delta$

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

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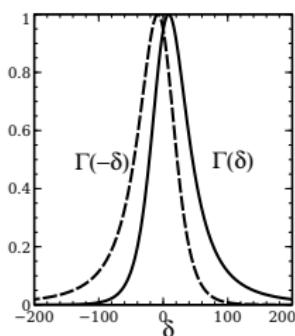
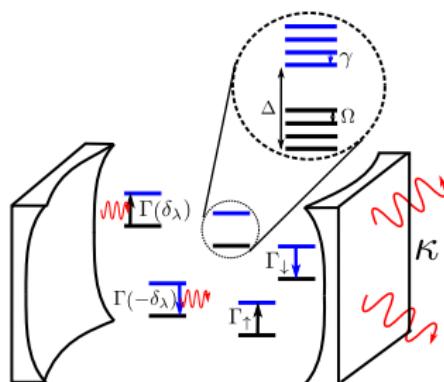


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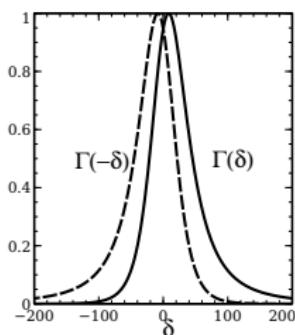
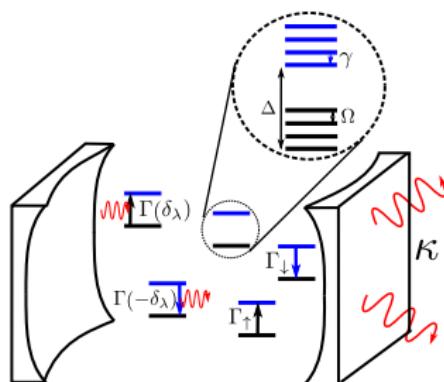
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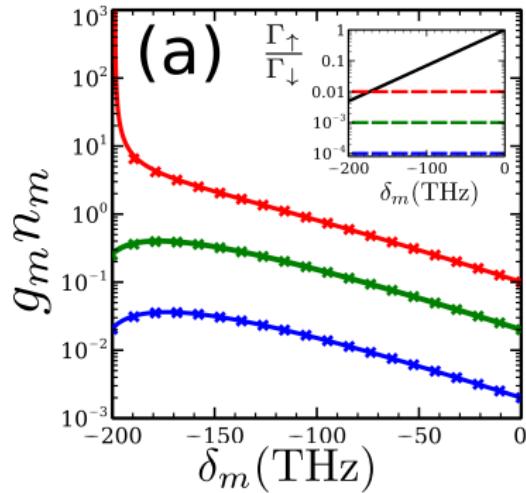
## Distribution $g_m n_m$

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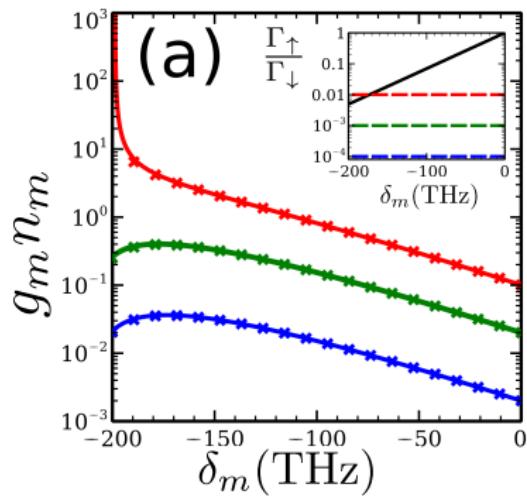


Low loss: Thermal

[Kirton & JK arXiv:1303.3459]

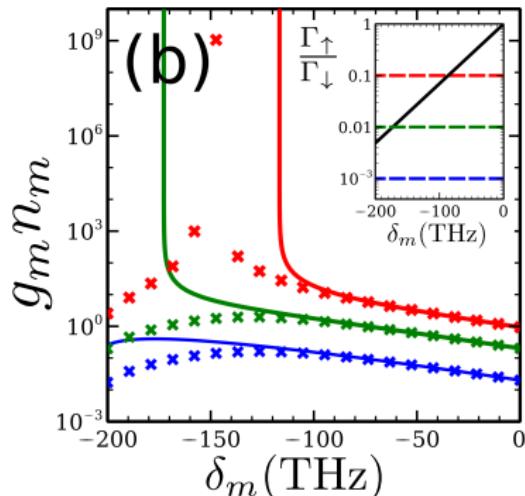
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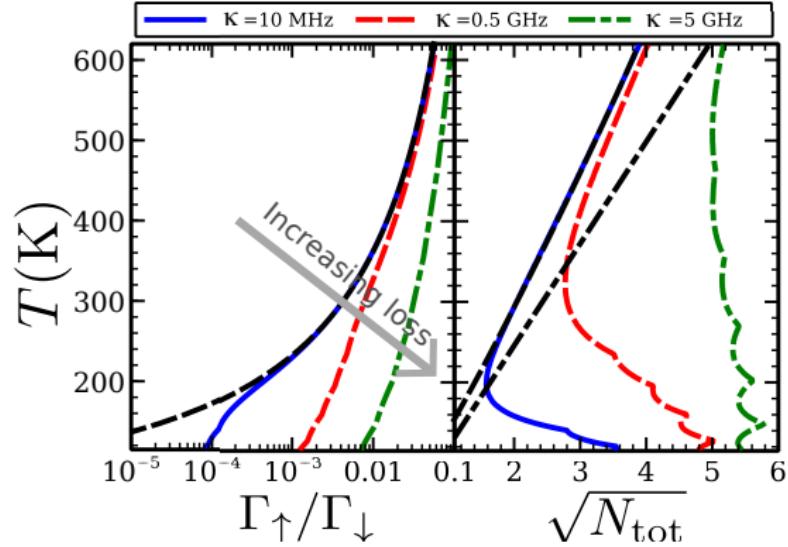
Low loss: Thermal

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High loss  $\rightarrow$  Laser

# Threshold condition



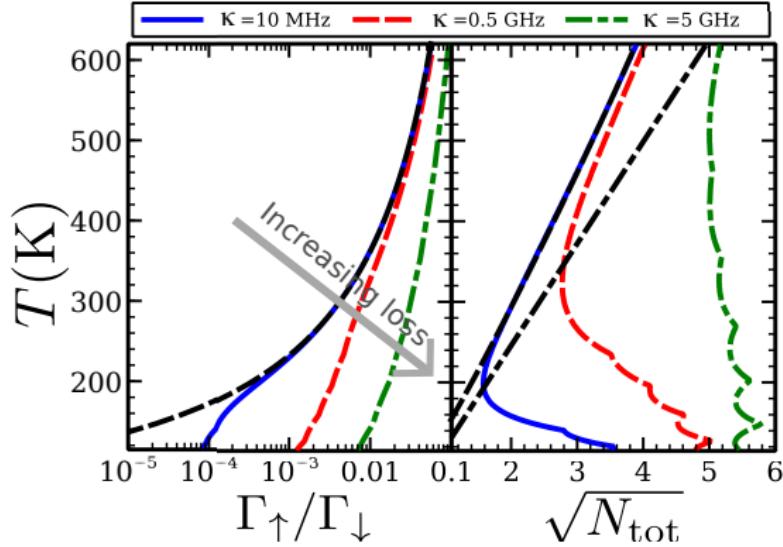
Compare threshold:

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

- Thermal at low/high temperature
- High loss,  $\kappa$  competes with  $\Gamma(\pm\delta_0)$
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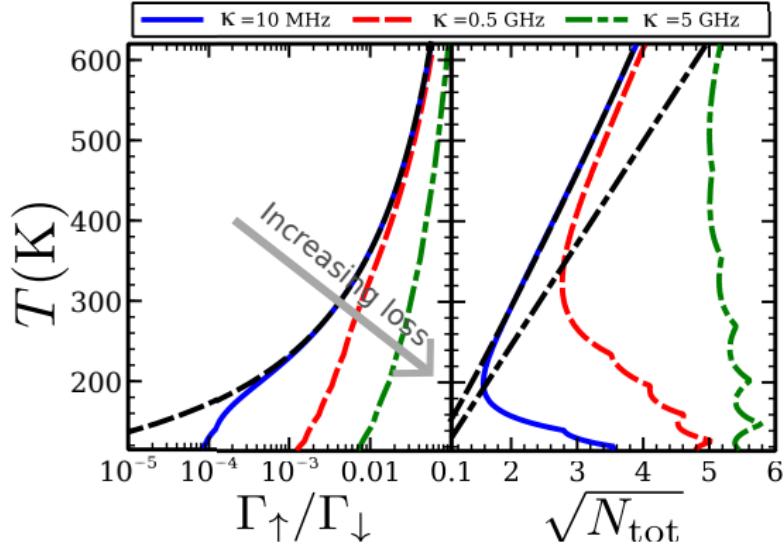
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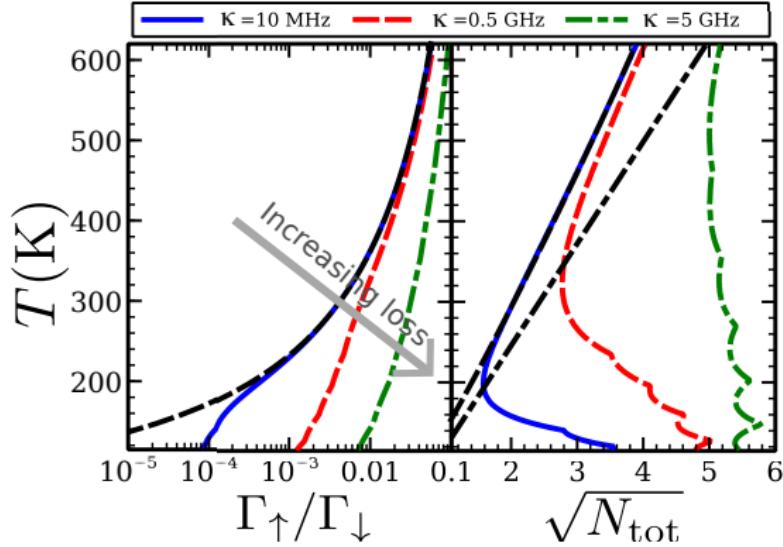
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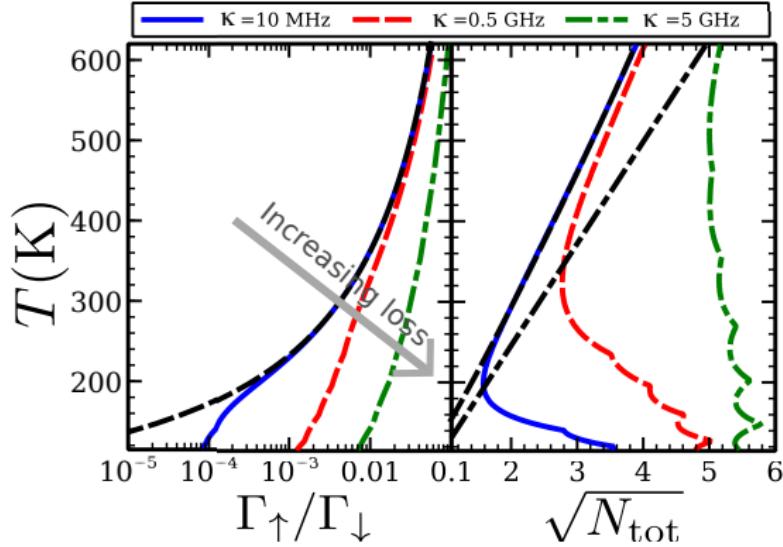
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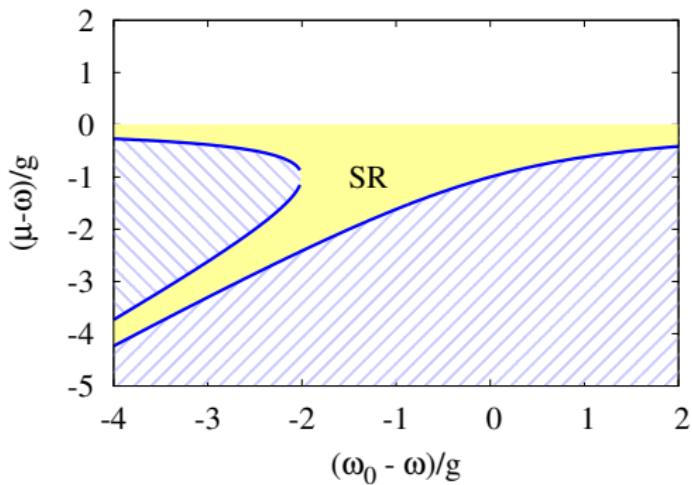
# Jaynes Cummings Hubbard model



- 1 Dicke model and superradiance
- 2 Polariton and photon condensation
  - Polaritons
  - Non-equilibrium condensation vs lasing
  - Photon condensation
- 3 Jaynes Cummings Hubbard model
  - JCHM vv Dicke
  - Coherently driven array
  - Disorder
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# Equilibrium: Dicke model with chemical potential

$$H - \mu N = (\omega - \mu)\psi^\dagger\psi + (\omega_0 - \mu)S^z + g(\psi^\dagger S^- + \psi S^+)$$

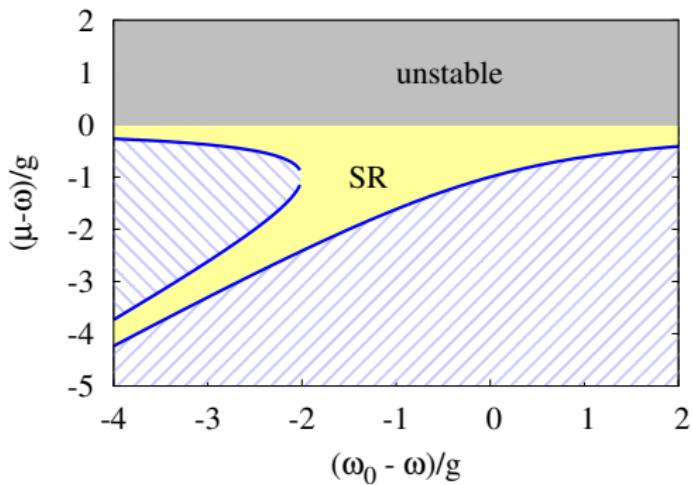


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[Eastham and Littlewood, PRB '01]

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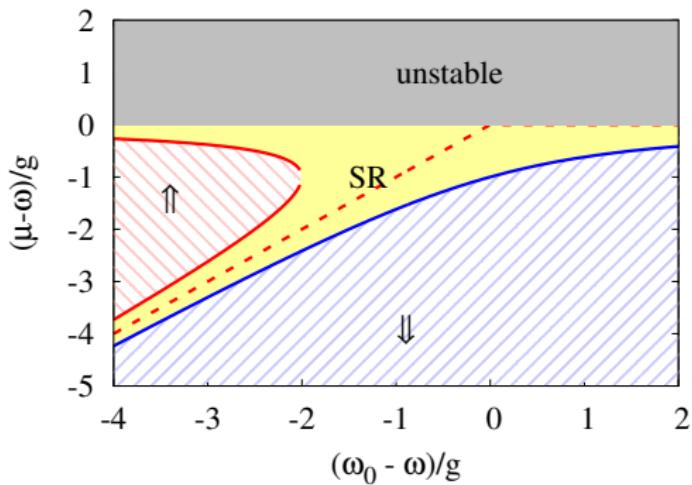


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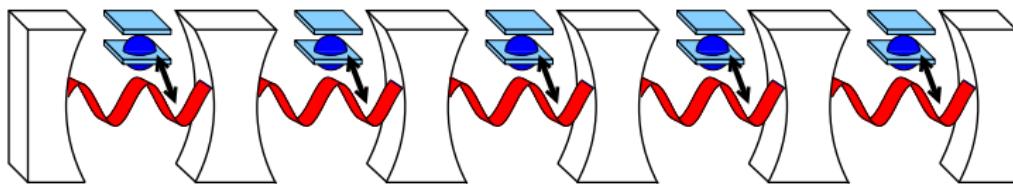
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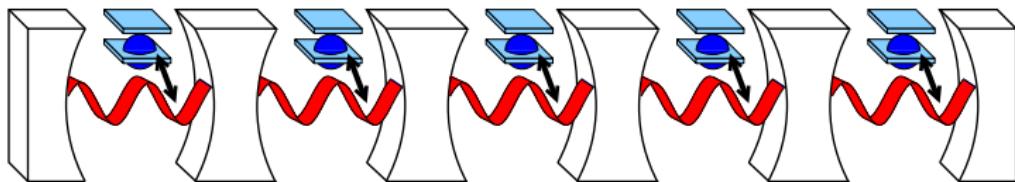
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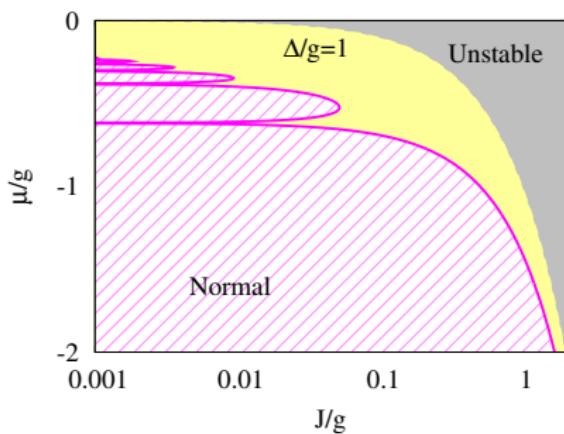


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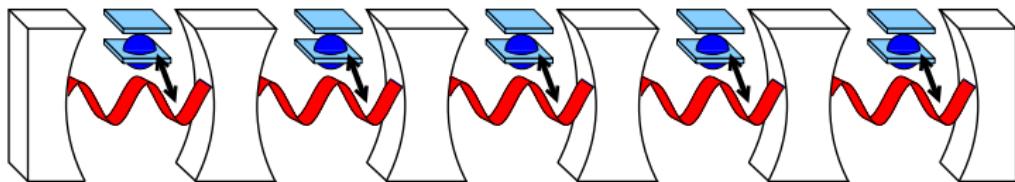
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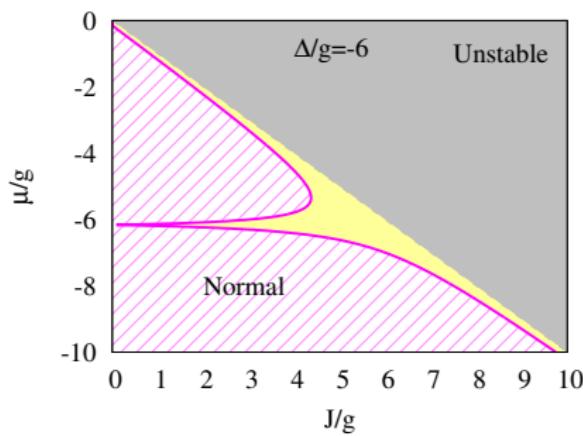
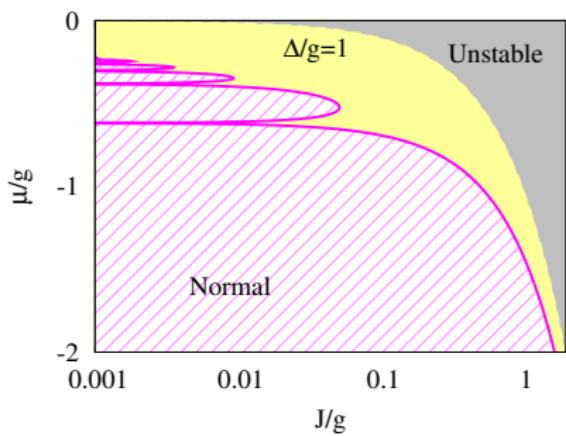
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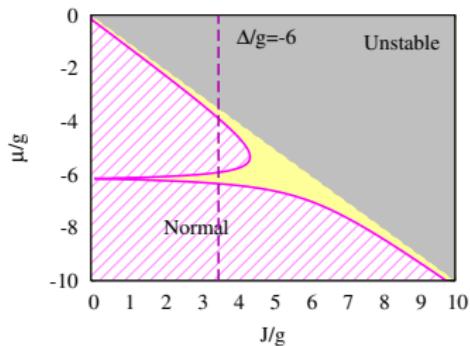


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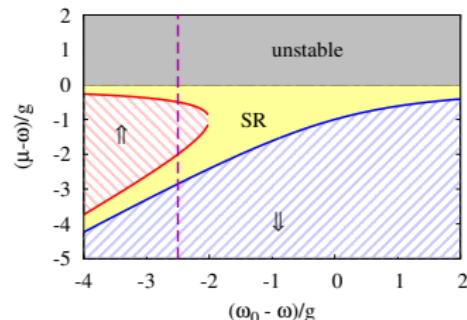


# Dicke vs JCHM

JCHM



Dicke

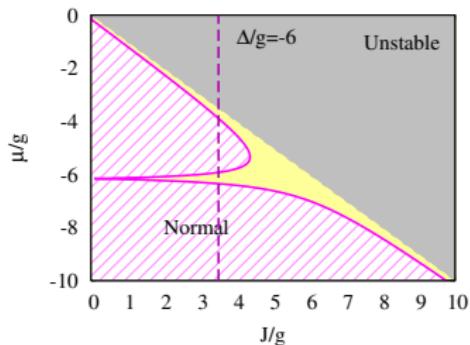


$\rightarrow$   $\downarrow$   $\rightarrow$   $\rightarrow$  Dicke photon mode

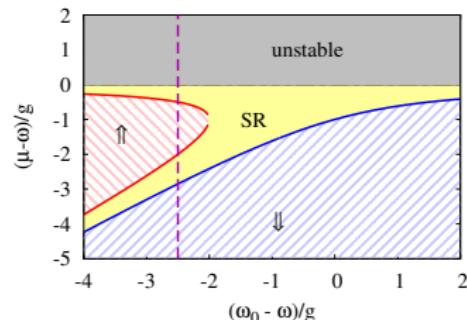
$\rightarrow$   $\uparrow$   $\leftarrow$   $\leftarrow$   $\pi = 1$  Mott lobe

# Dicke vs JCHM

JCHM



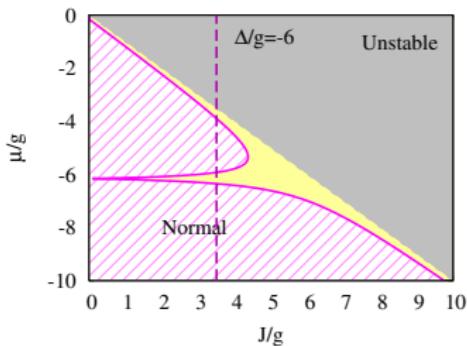
Dicke



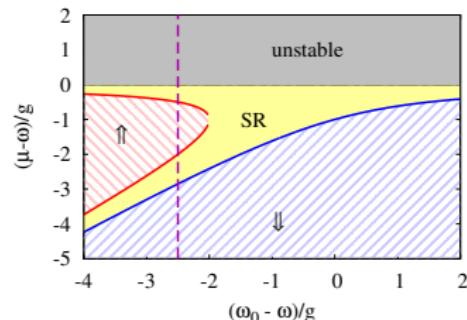
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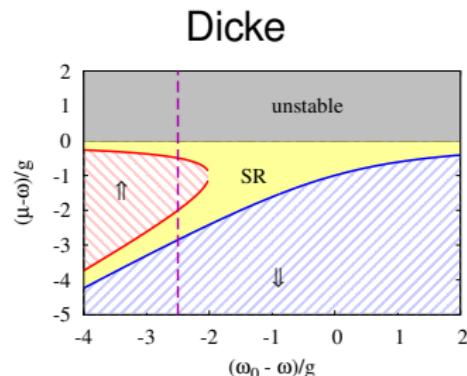
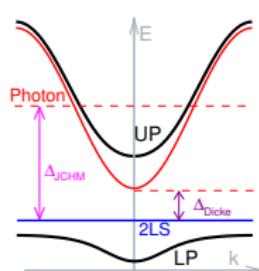
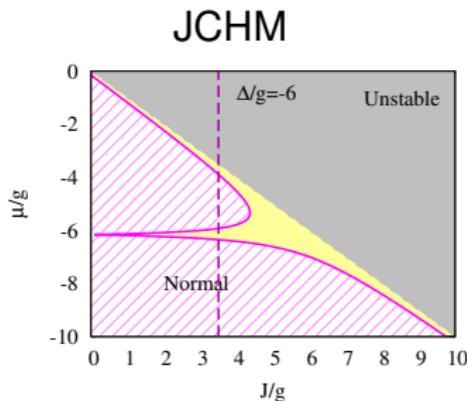


Dicke



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# Dicke vs JCHM



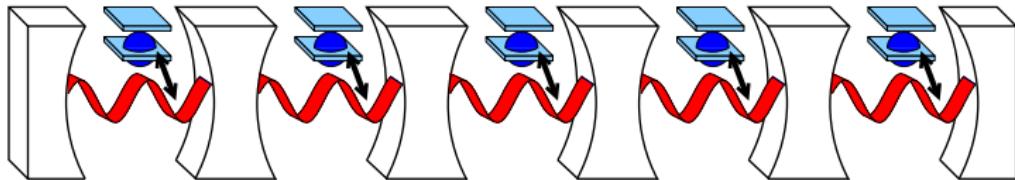
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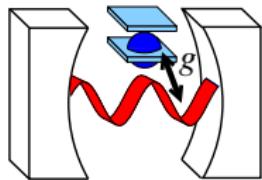
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# Coherently pumped single cavity [Bishop *et al.* Nat. Phys '09]

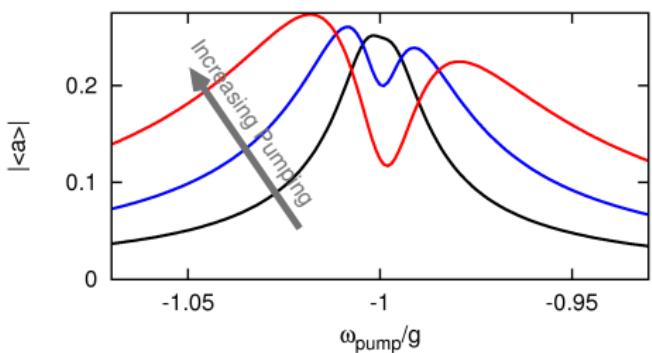


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Anti-resonance in  $\langle a^\dagger a \rangle$

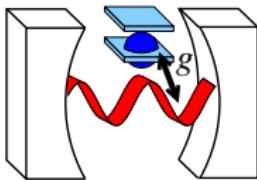
Mollow triplet fluorescence

Condensation & superradiance



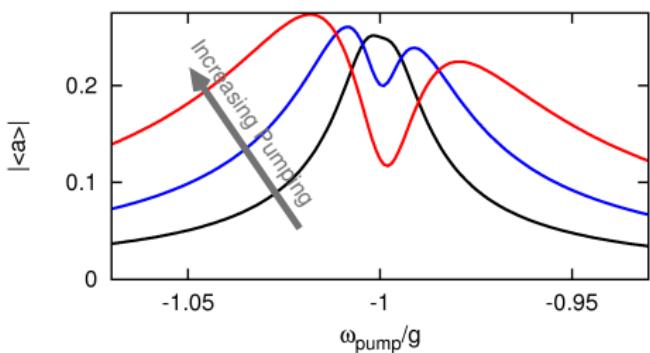
[Lang *et al.* PRL '11]

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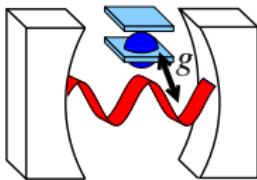
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- Anti-resonance in  $|\langle \psi \rangle|$ .
- Effective 2LS:  
 $|\text{Empty}\rangle, |\text{1 polariton}\rangle$



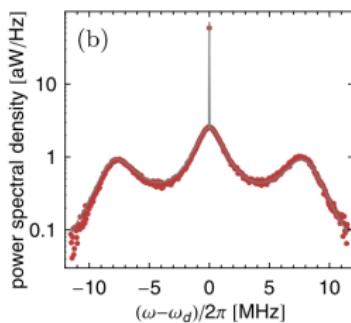
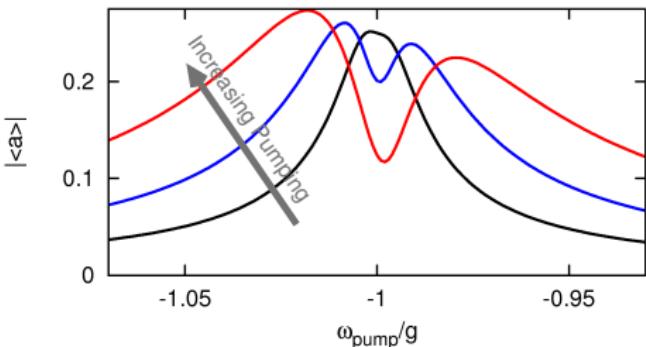
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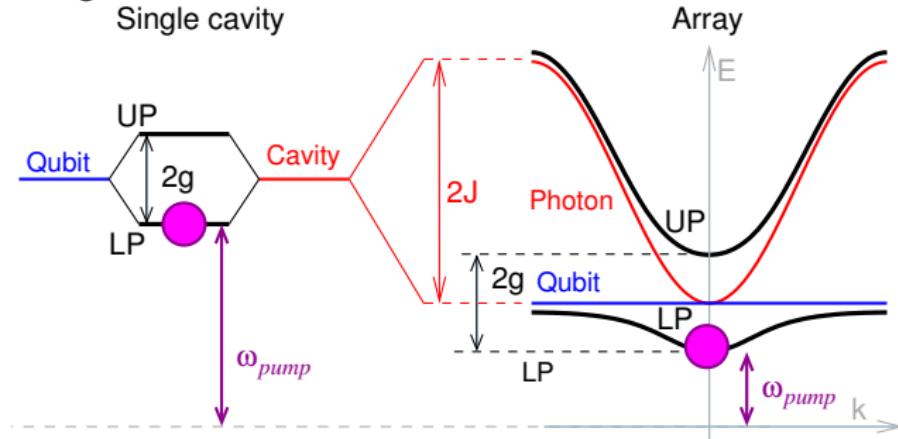
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[Lang *et al.* PRL '11]

# Coherently pumped dimer & array

Chose detuning *a la* Dicke model

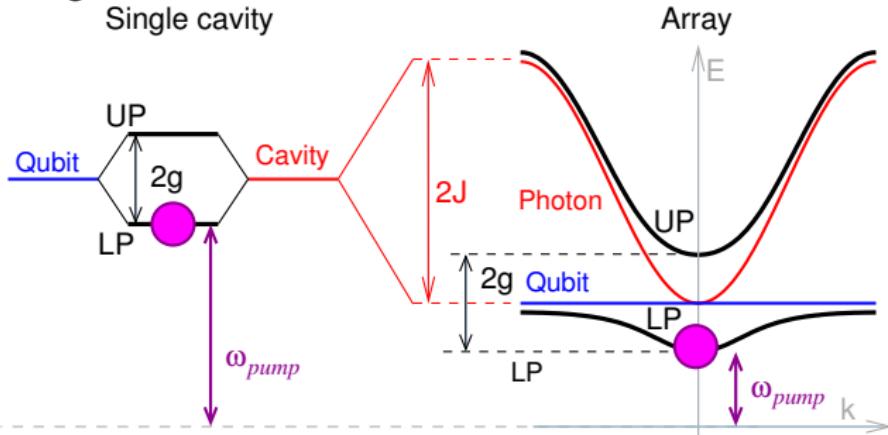


→ Bistability at intermediate  $J$   
→ More/less localised states  
→ Connection to Dicke limit

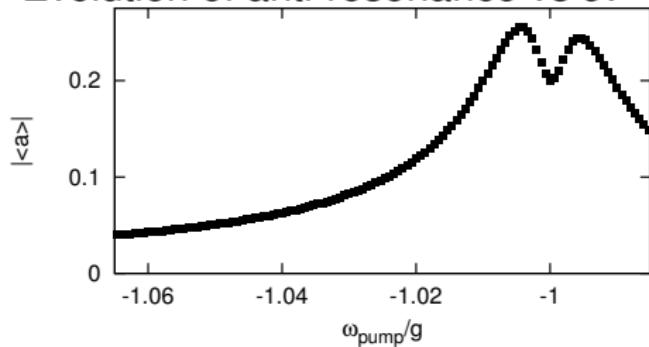
[Nissen *et al.* PRL '12]

# Coherently pumped dimer & array

Chose detuning *a la* Dicke model



Evolution of anti-resonance vs  $J$ .

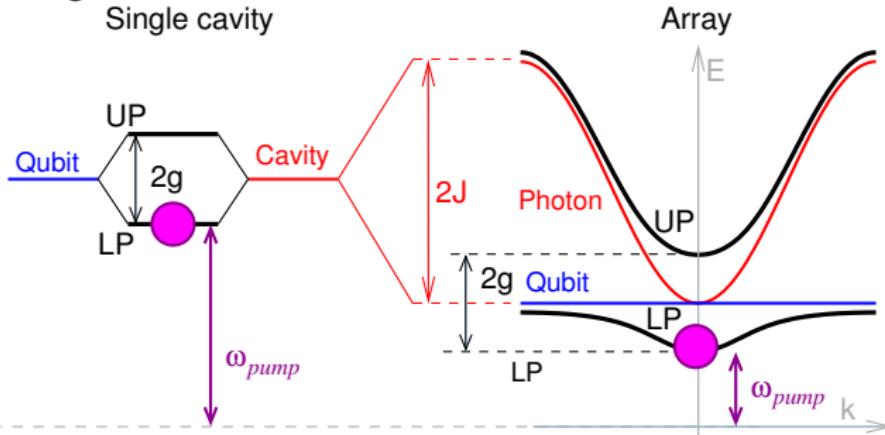


→ Bistability at intermediate  $J$   
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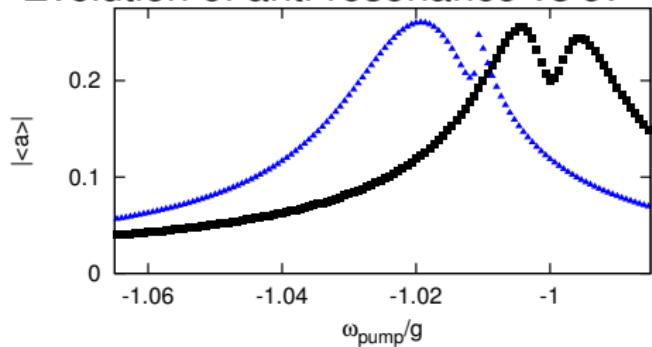
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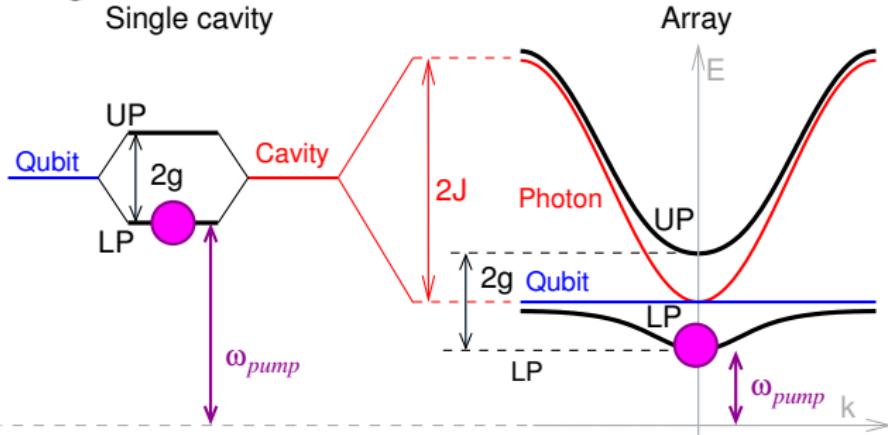


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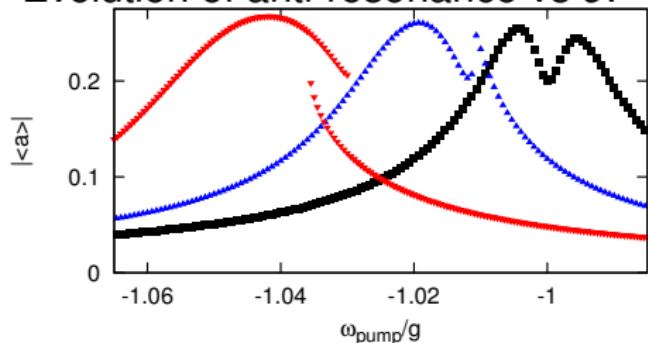
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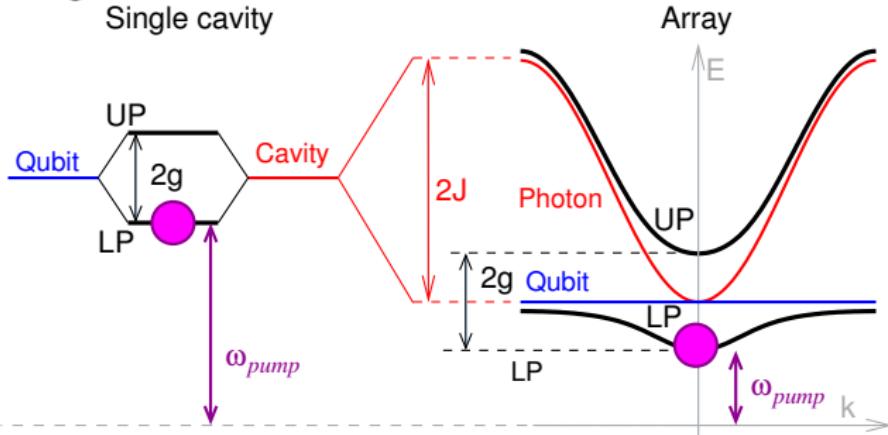


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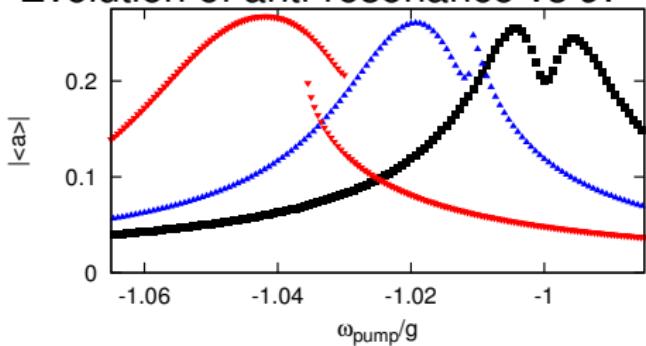
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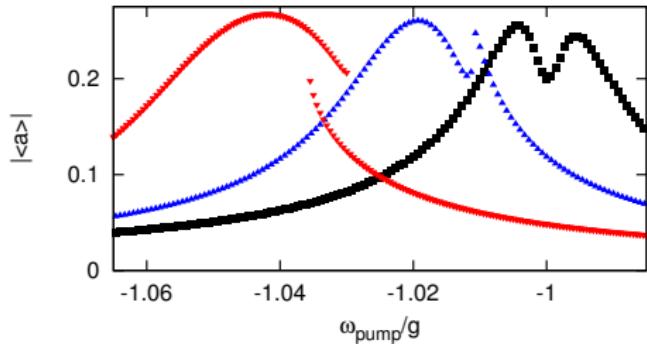
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# Photon blockade picture $J \lesssim g$

- Polariton basis
- Nonlinearity  $|\epsilon_2 - 2\epsilon_1| \propto g$ .

$$H = \sum_i \left( \frac{\epsilon}{2} \tau_i^z + \tilde{f} \tau_i^x \right)$$



[Nissen *et al.* PRL '12]

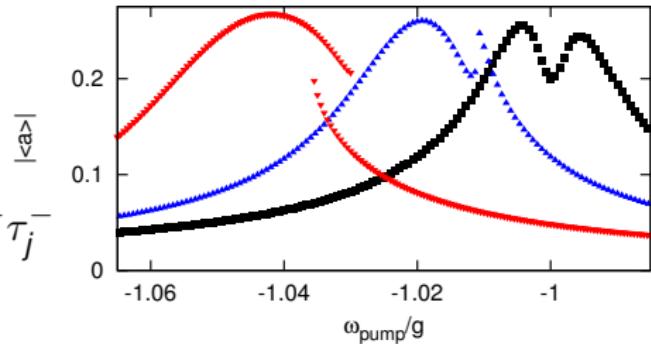
- Decouple hopping:  
 $\tau^z \tau^z \rightarrow \tau^z \tau^z + \tau^z \tau^z$
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$$J > J_c = \frac{4}{P} \left( \frac{2\delta + (g/2)^2}{3} \right)^{3/2}$$

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[Nissen et al. PRL '12]

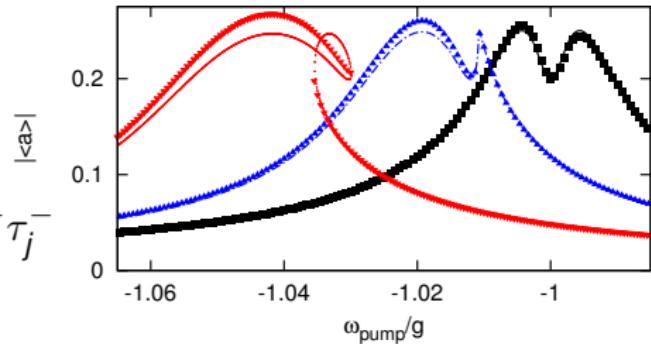
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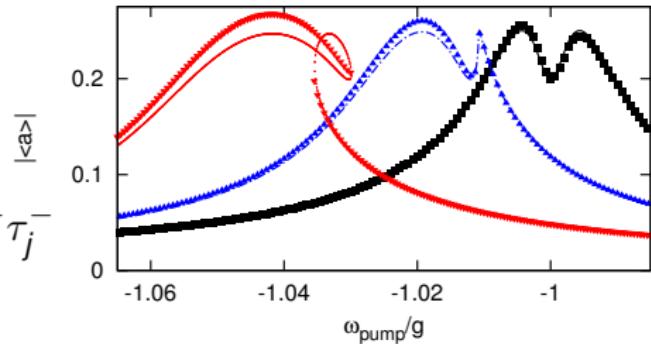
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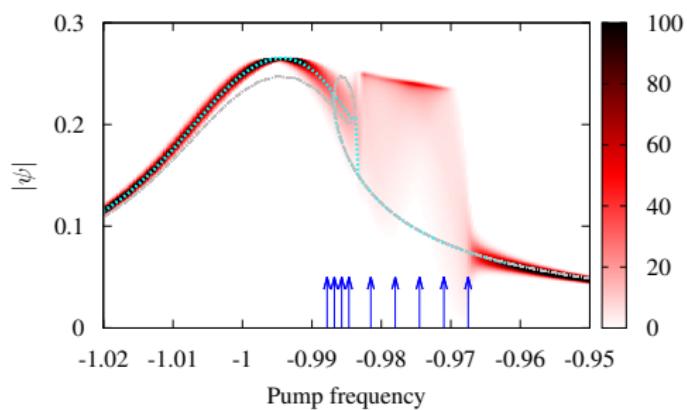
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$$J > J_c = \frac{4}{\tilde{f}^2} \left( \frac{2\tilde{f}'' + (\tilde{\kappa}/2)^2}{3} \right)^{3/2}$$

# Coherent pumped array – disorder

- Effect of disorder,  $\Delta \rightarrow \Delta_i$ 
  - ▶ Distribution of  $\psi$  – Washes out bistable jump

- Bistability disappears → phase transition on  $\Delta_i$
- Complex  $\psi$  distribution
- Superfluid phases in driven system?



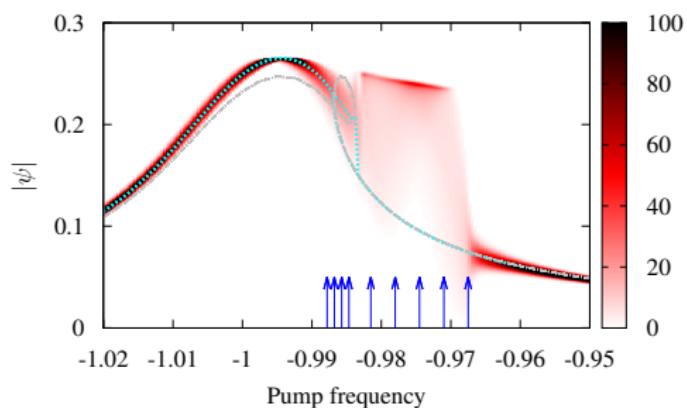
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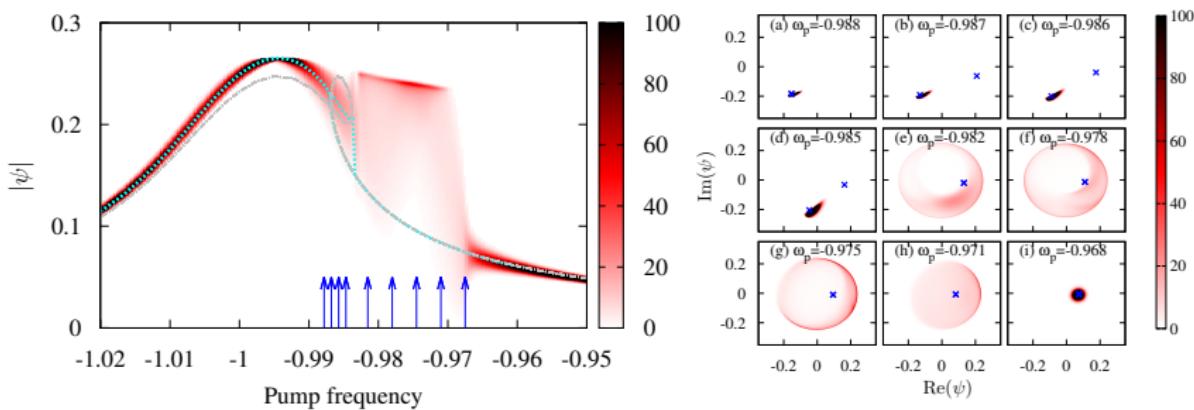


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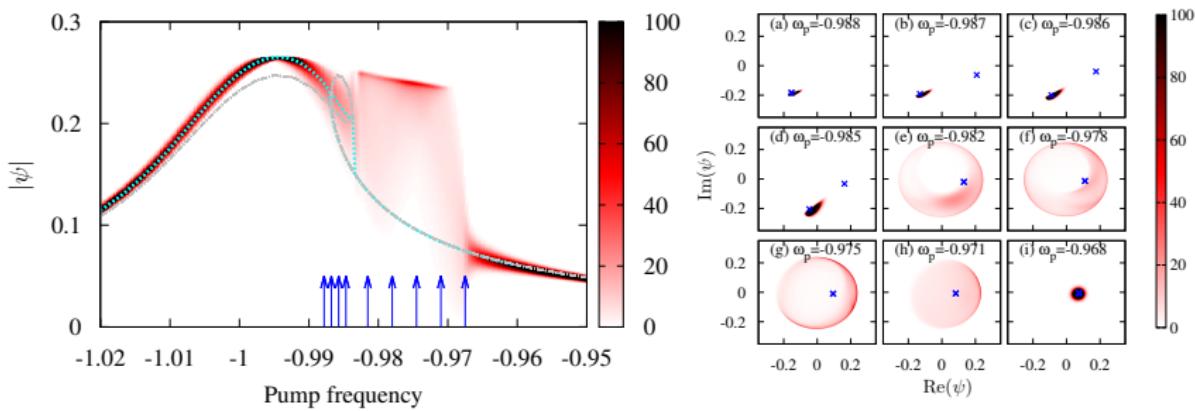
Is there more to say about driven system?



[Kulaitis et al. PRA, '13]

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# Phase transitions with SC qubits

- 1 Dicke model and superradiance
- 2 Polariton and photon condensation
  - Polaritons
  - Non-equilibrium condensation vs lasing
  - Photon condensation
- 3 Jaynes Cummings Hubbard model
  - JCHM vv Dicke
  - Coherently driven array
  - Disorder
- 4 Phase transitions with SC qubits
  - Pumping without symmetry breaking
  - Collective dephasing

# Raman pumping

- How to pump without breaking symmetry
- Counter-rotating terms — Raman pumping
  - ▶ Atom proposal [Dimer *et al.* PRA '07]
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JK, Türeci, Houck in progress

• Qubit dephasing much bigger than atom

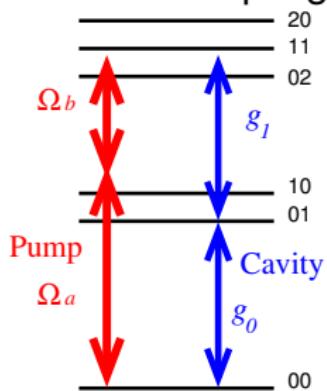
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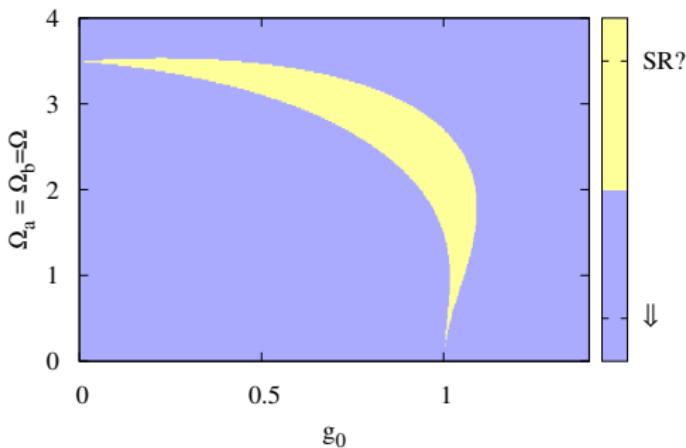
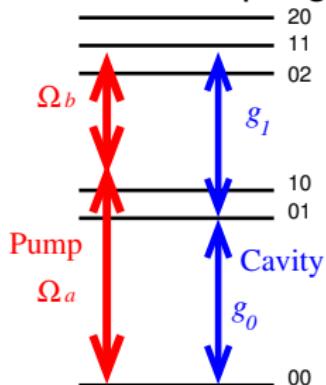


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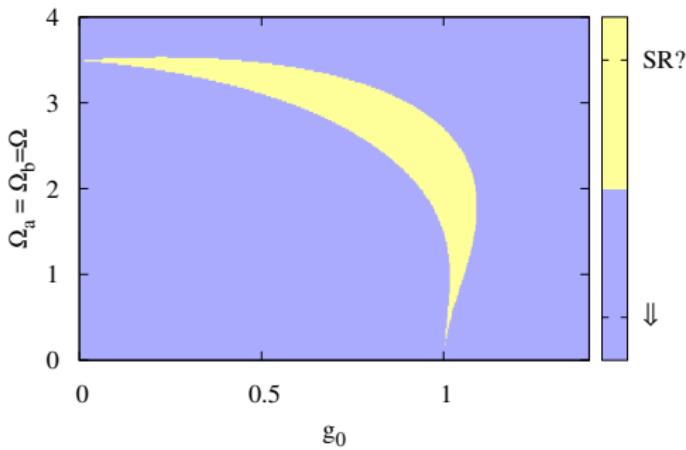
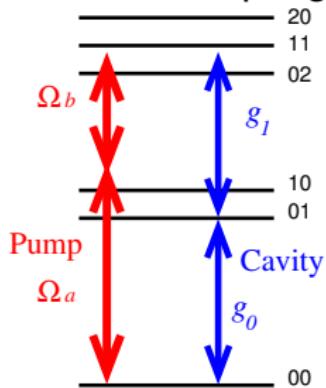


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# Collective dephasing

- Real environment is not Markovian
  - ▶ [Carmichael & Walls JPA '73] Requirements for correct equilibrium
  - ▶ [Ciuti & Carusotto PRA '09] Dicke SR and emission
- Carmichael's requirements:
  - ▶ Phase transition  $\rightarrow$  soft modes
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Dicke model linewidth:

$$H = \omega\psi^\dagger\psi + \sum_{i=1}^N \frac{\epsilon_i}{2}\sigma_i^z + g(\sigma_i^+\psi + \text{h.c.}) \\ + \sum_i \sigma_i^z \sum_q \gamma_q (b_q^\dagger + b_q) + \sum_q \beta_q b_{iq}^\dagger b_q.$$

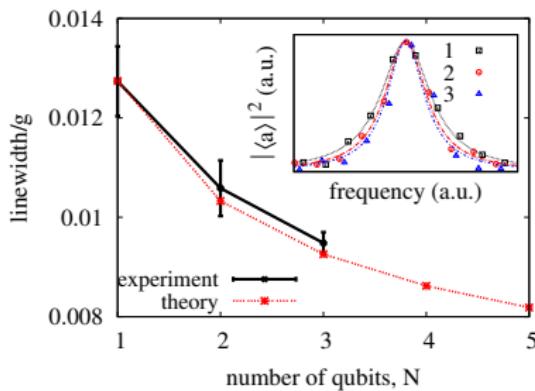
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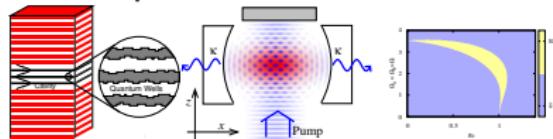
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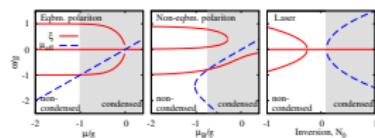
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# Summary

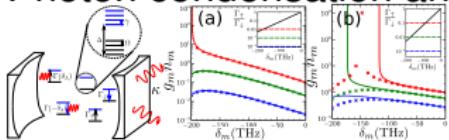
- Non-equilibrium Dicke relevant to increasing number of systems



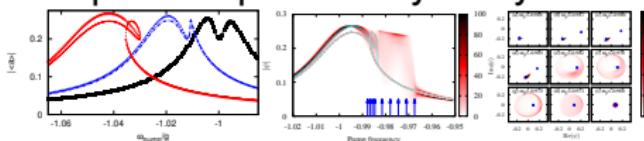
- Polariton condensation vs lasing



- Photon condensation and thermalisation



- Pumped coupled cavity array — bistability and disorder



- Future prospects – SC cavity array transitions



# Extra slides

- 5 Ferroelectric transition
- 6 Pumped JCHM correlations
- 7 Retarded Green's function for laser
- 8 Timescales for Raman pumped experiment

# Ferroelectric transition

Atoms in Coulomb gauge

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Gauge transform to dipole gauge  $\mathbf{D} \cdot \mathbf{r}$

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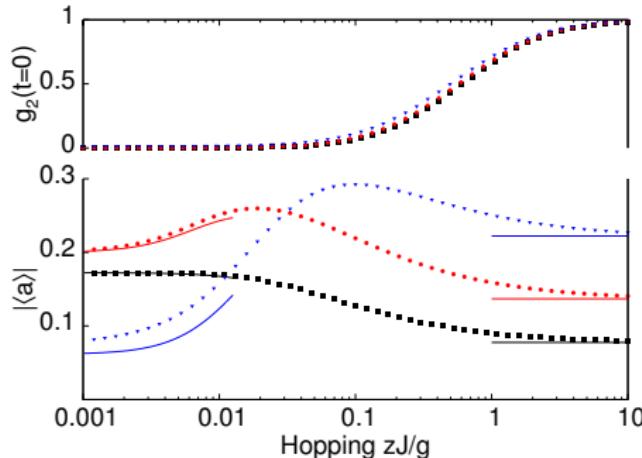
“Dicke” transition at  $\omega_0 < N\bar{g}^2/\omega \equiv 2\eta N$

But,  $\psi$  describes electric displacement

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# Coherently pumped array: correlations & fluorescence

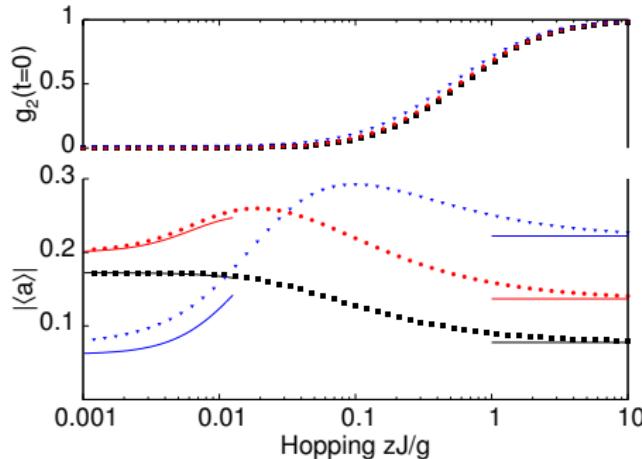


## Correlations

- $g_2 : 0 \rightarrow 1$  crossover.

- Small J: Mollow triplet
- Large J: Off resonance fluorescence
- Pump at collective resonance
- Mismatch if  $J \neq 0$

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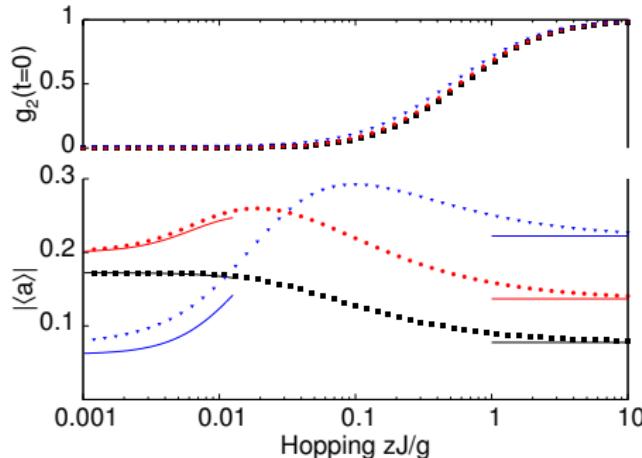


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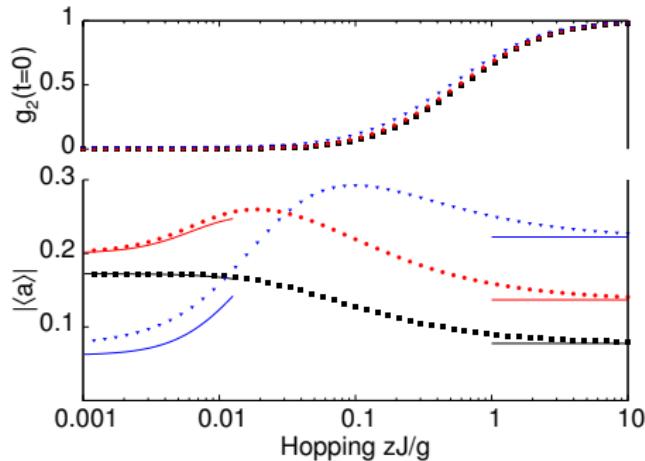
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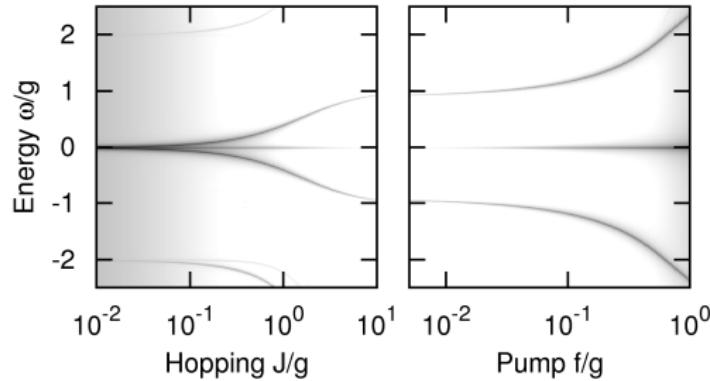


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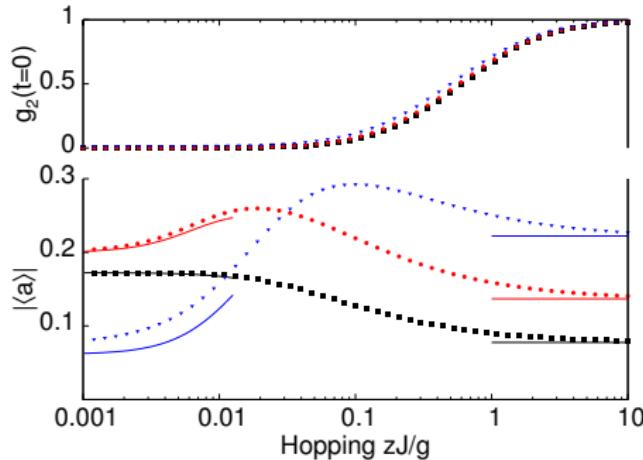
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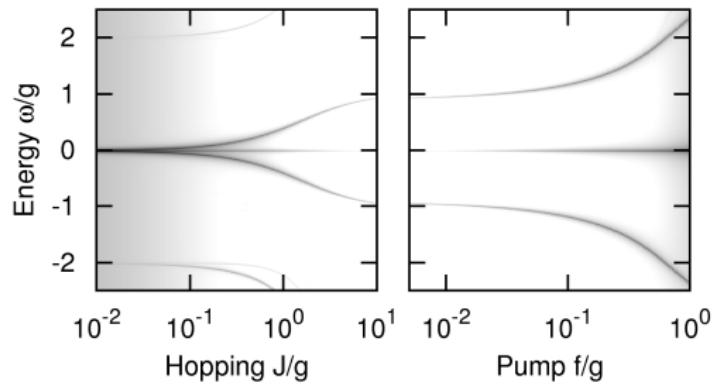


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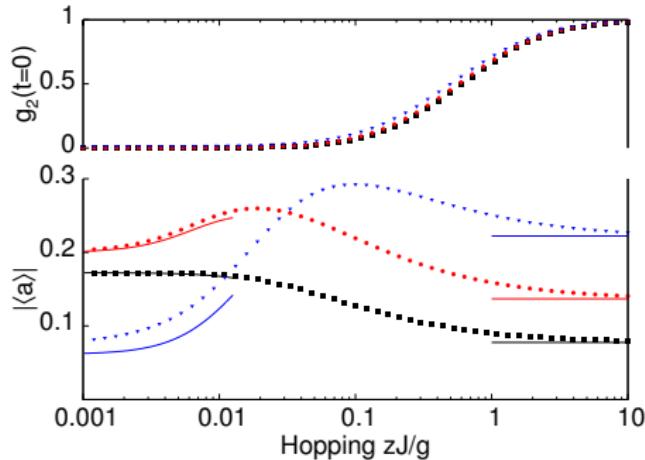
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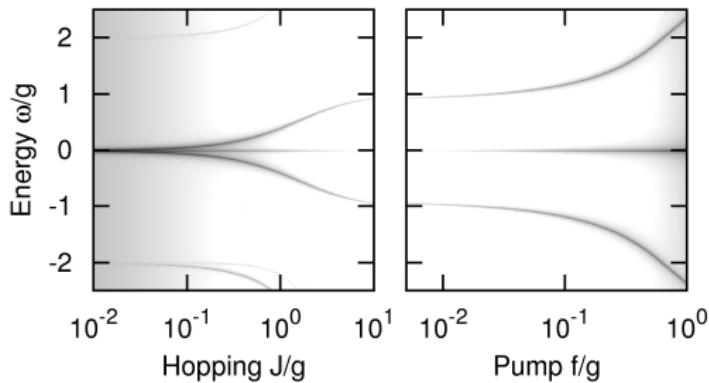


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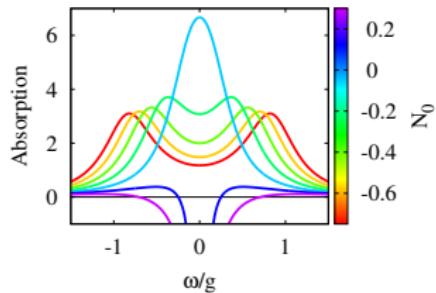
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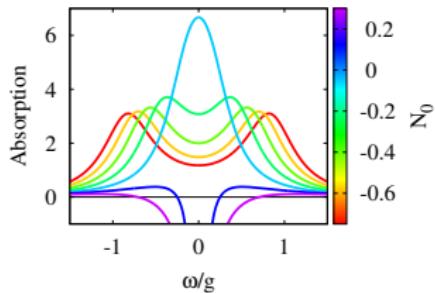
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# Maxwell-Bloch Equations: Retarded Green's function



- Introduce  $D^R(\omega)$ :  
Response to perturbation
- Absorption =  $-2\Im[D^R(\omega)]$

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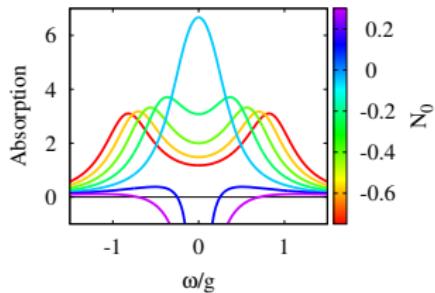
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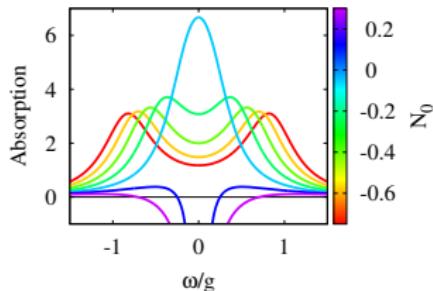
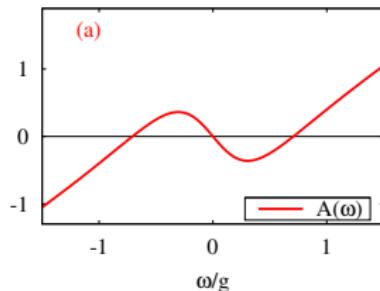
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- Absorption =  $-2\Im[D^R(\omega)] = \frac{2B(\omega)}{A(\omega)^2 + B(\omega)^2}$

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

# Maxwell-Bloch Equations: Retarded Green's function



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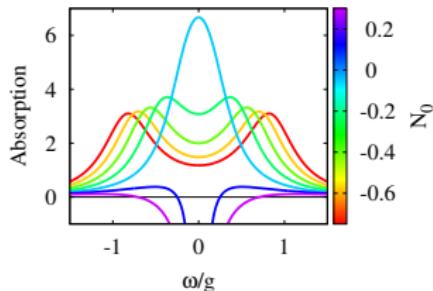
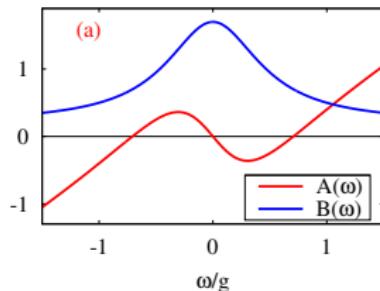
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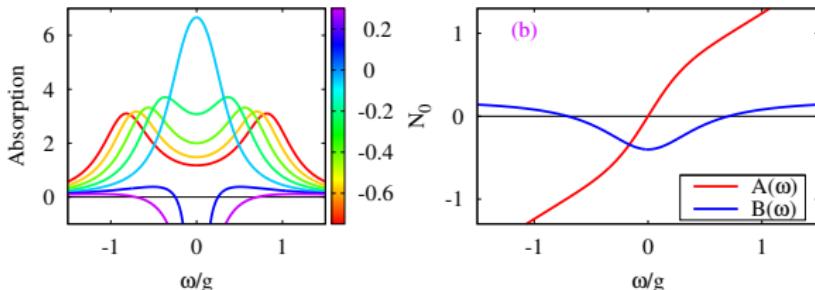
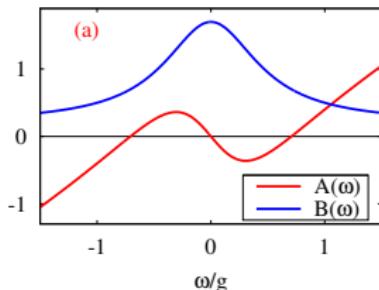
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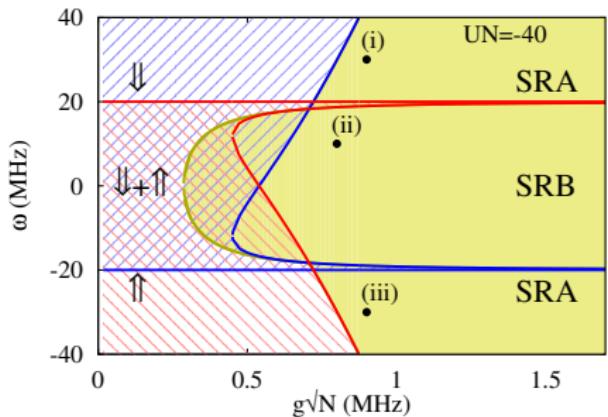
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# Extra slides

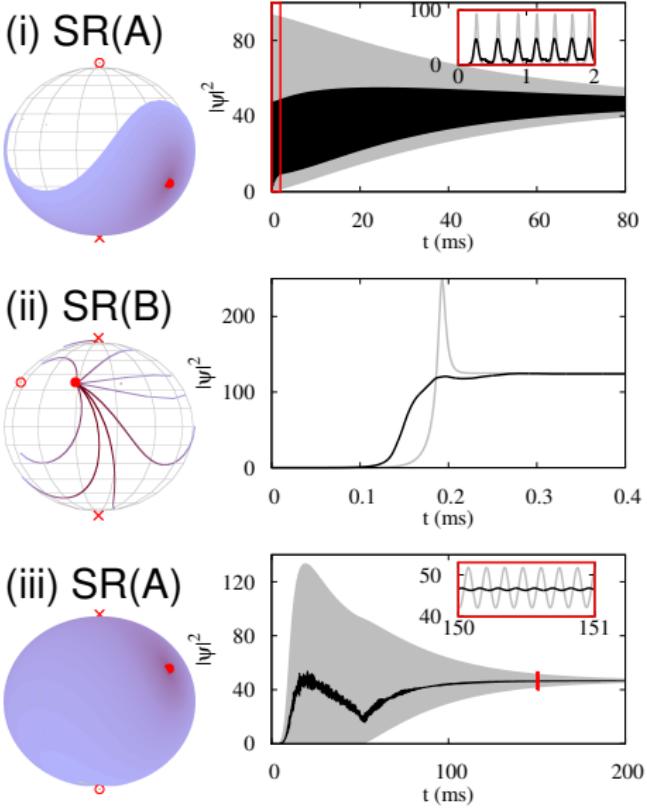
- 5 Ferroelectric transition
- 6 Pumped JCHM correlations
- 7 Retarded Green's function for laser
- 8 Timescales for Raman pumped experiment

# Dynamics: Evolution from normal state

Gray:  $\mathbf{S} = (\sqrt{N}, \sqrt{N}, -N/2)$   
Black: Wigner distribution of  $\mathbf{S}, \psi$



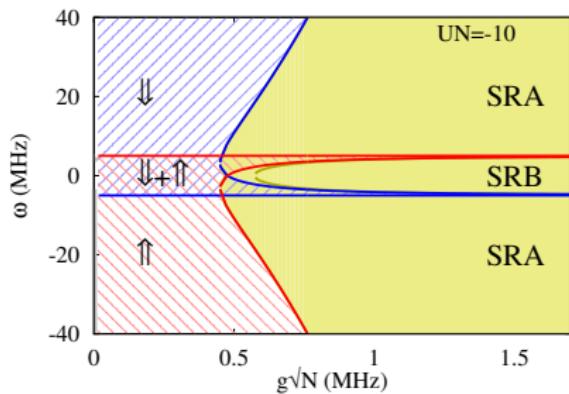
Oscillations:  $\sim 0.1\text{ms}$   
Decay:  $20\text{ms}, 0.1\text{ms}, 20\text{ms}$



# Asymptotic state: Evolution from normal state

(Near to experimental  $UN = -13\text{MHz}$ ).

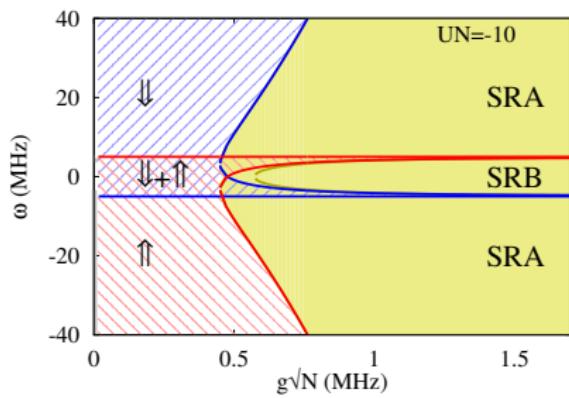
All stable attractors:



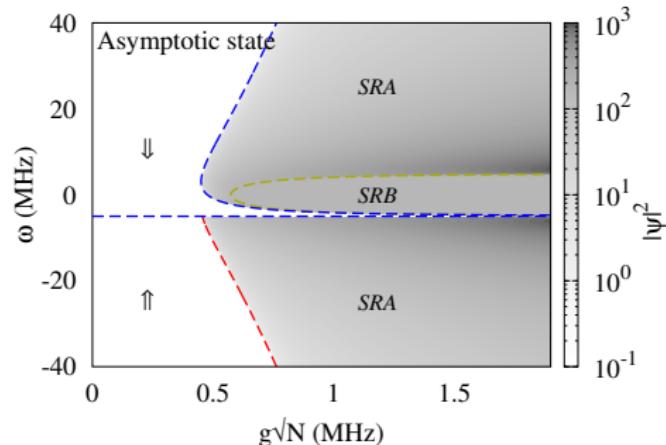
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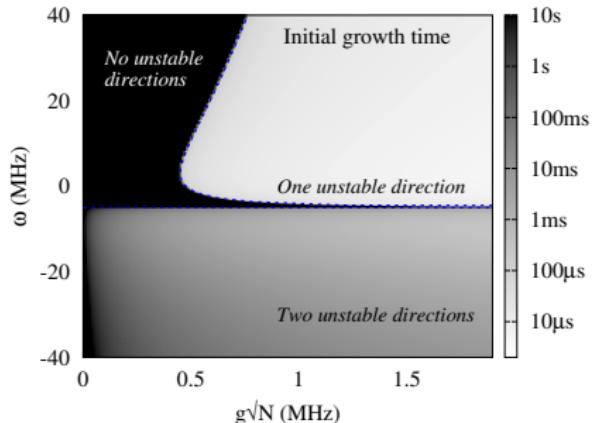
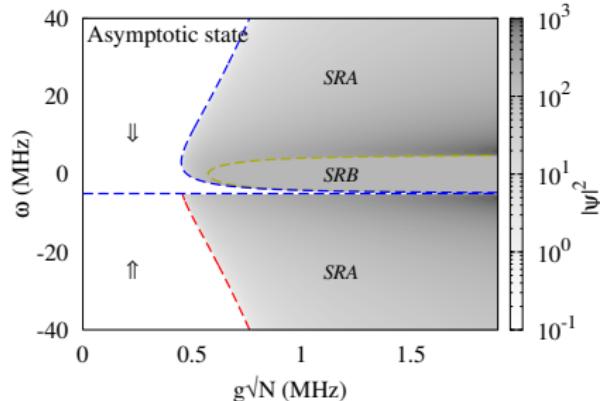
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Starting from  $\downarrow$



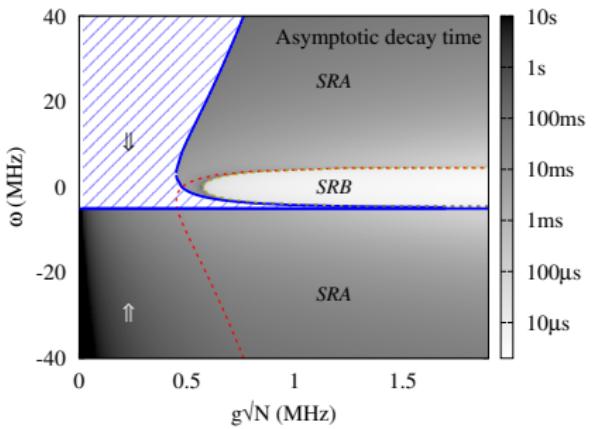
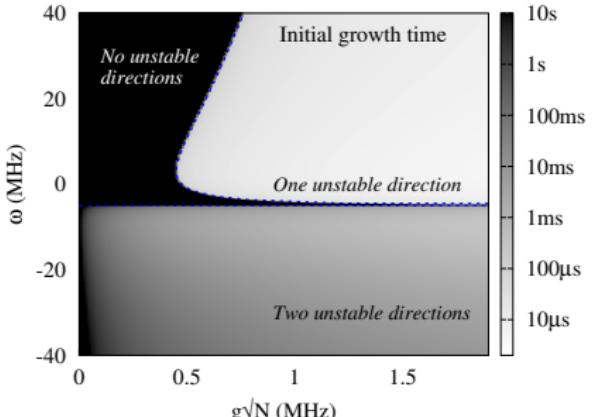
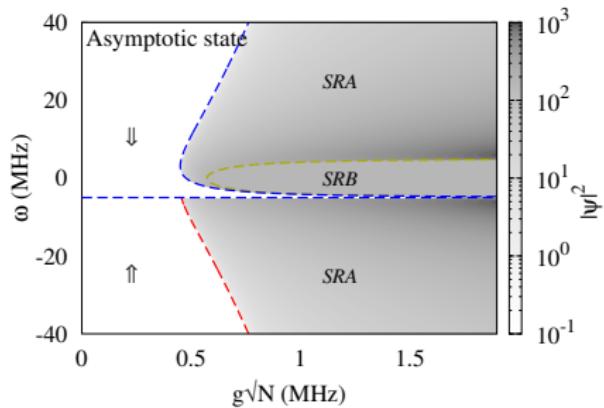
# Timescales for dynamics: What are they?



Growth Most unstable eigenvalues near  $\mathbf{S} = (0, 0, -N/2)$

Decay Slowest stable eigenvalues near final state

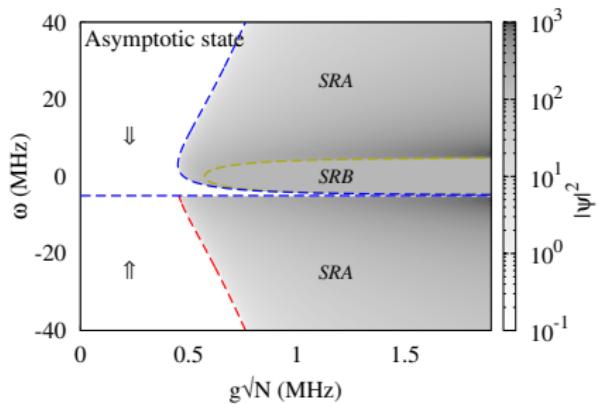
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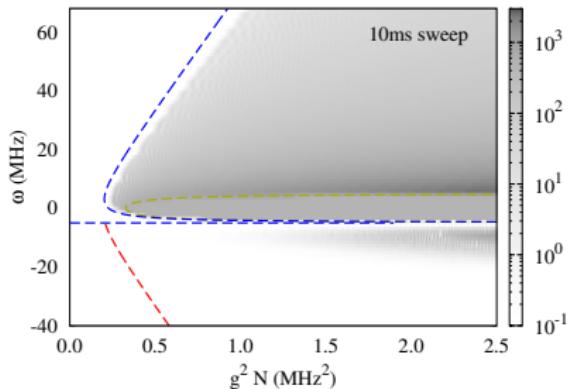
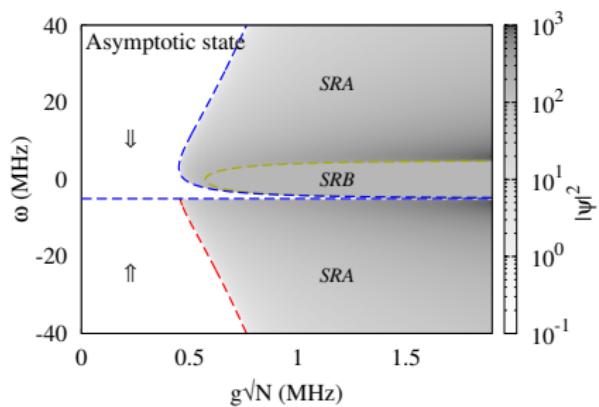
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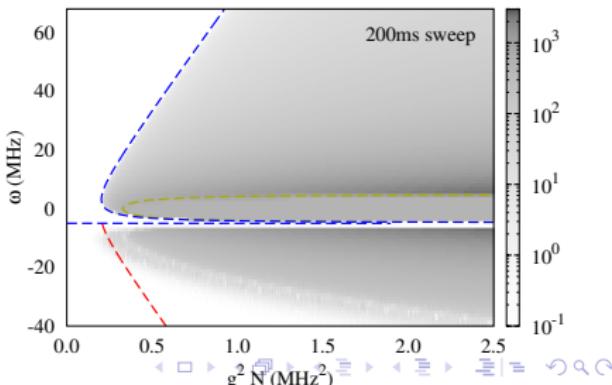
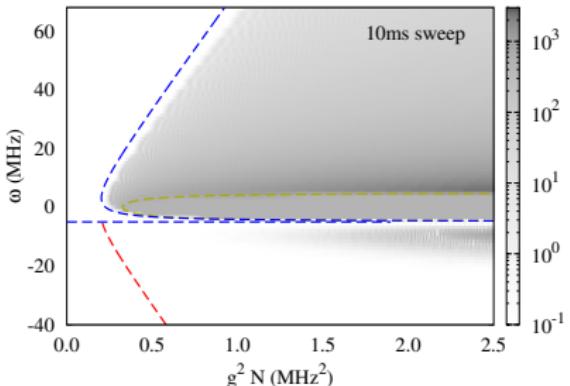
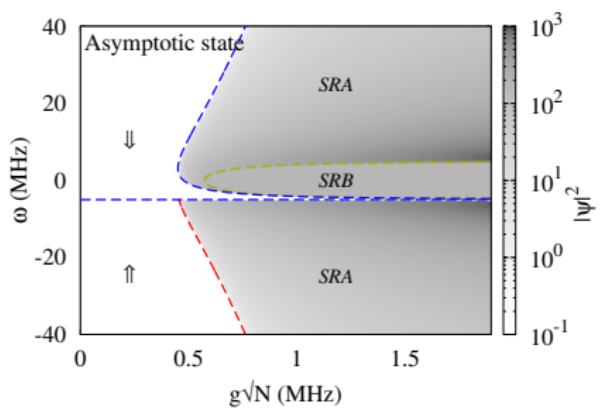
# Timescales for dynamics: Consequences for experiment



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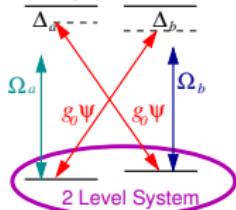


# Timescales for dynamics: Consequences for experiment



# Timescales for dynamics: Why so slow and varied?

Suppose co- and counter-rotating terms differ

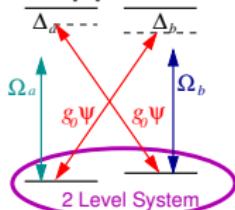


$$H = \dots + g(\psi^\dagger S^- + \psi S^+) + g'(\psi^\dagger S^+ + \psi S^-) + \dots$$

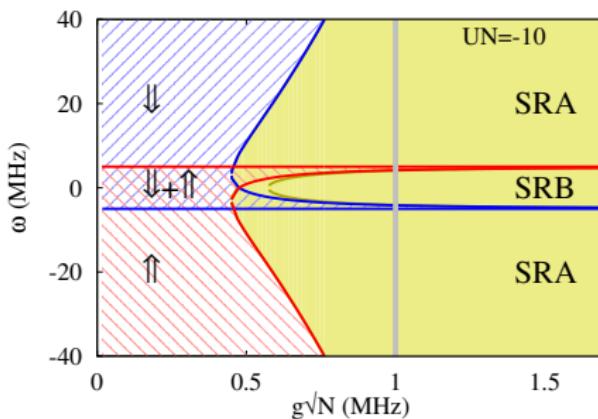
- SR(A) near phase boundary at small  $\delta g \rightarrow$  Critical slowing down
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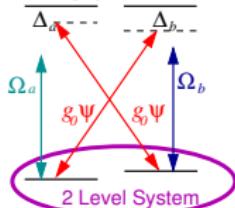
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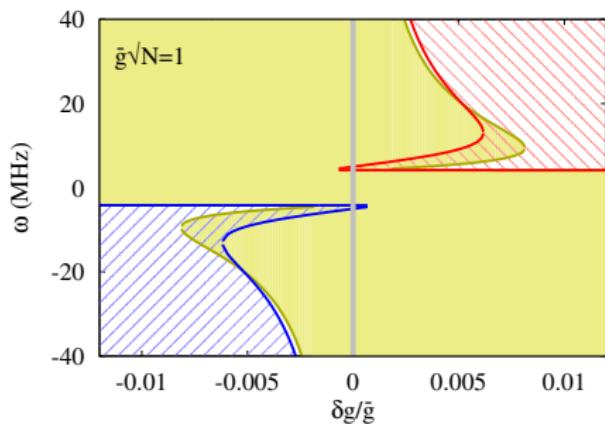
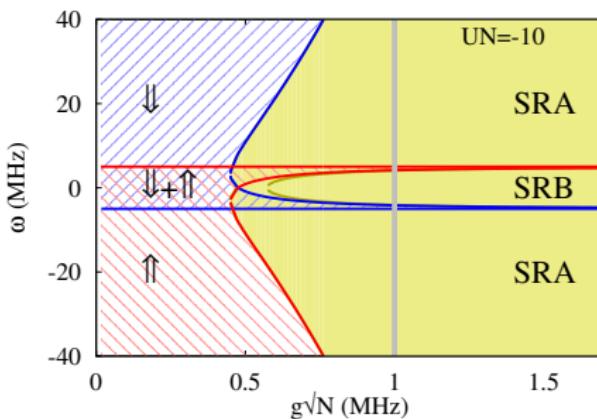
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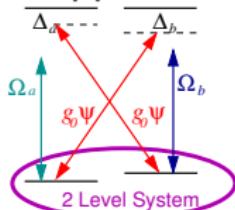
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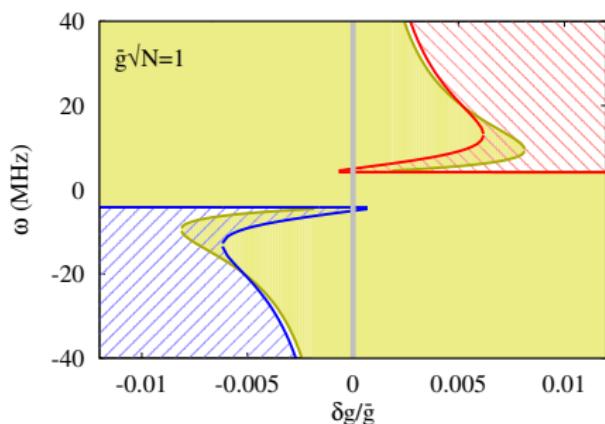
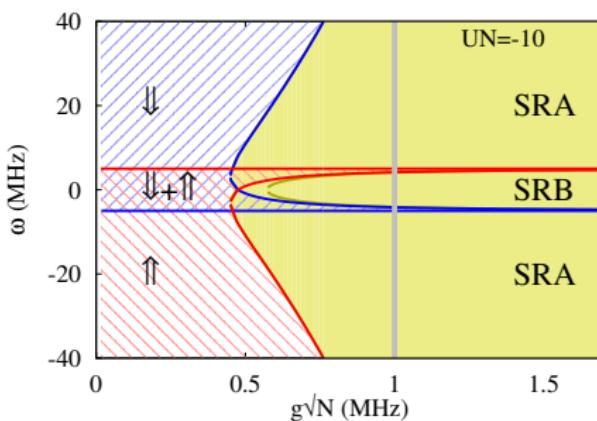
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