From weak to ultra-strong matter-light coupling with organic materials

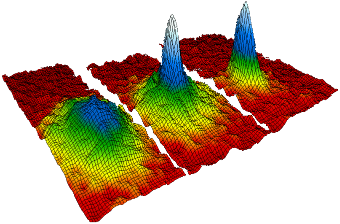
Jonathan Keeling

University of St Andrews
1413-2013

Yale, February 2015

Coherent states of matter and light

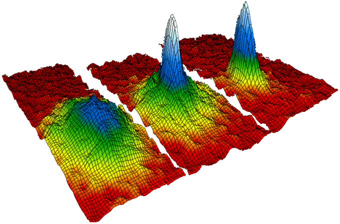
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science ’95]

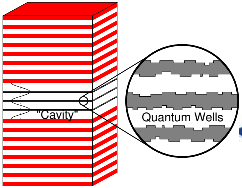
Coherent states of matter and light

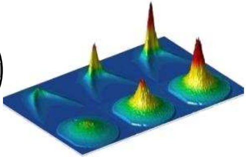
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science '95]

Polariton Condensate $T \sim 20$K

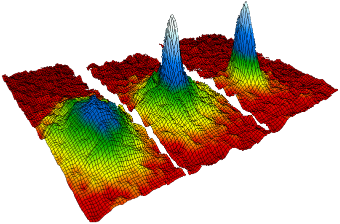




[Kasprzak *et al.* Nature, '06]

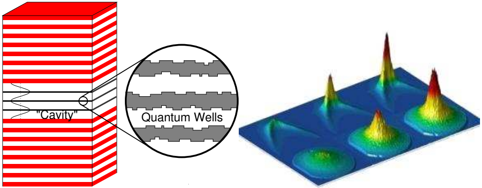
Coherent states of matter and light

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



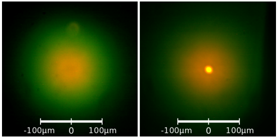
[Anderson *et al.* Science '95]

Polariton Condensate $T \sim 20$K



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

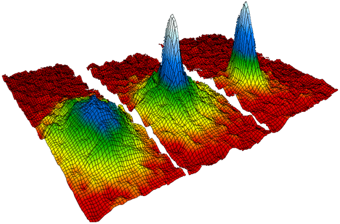
Photon Condensate
$T \sim 300$K



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

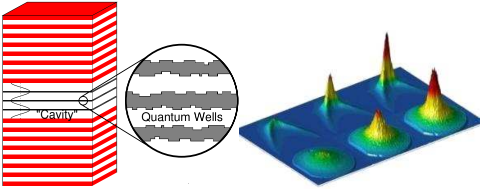
Coherent states of matter and light

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



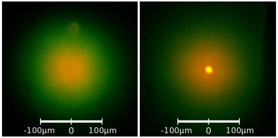
[Anderson *et al.* Science '95]

Polariton Condensate $T \sim 20$K



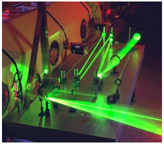
[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K



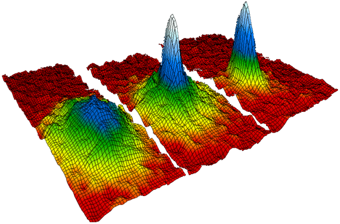
[Klaers *et al.* Nature, '10]

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



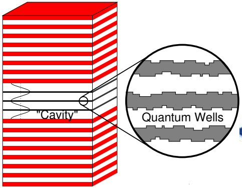
Coherent states of matter and light

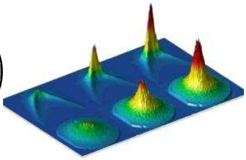
Atomic BEC $T \sim 10^{-7}$K



[Anderson *et al.* Science '95]

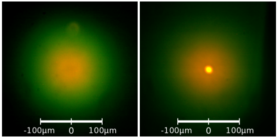
Polariton Condensate $T \sim 20$K





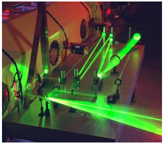
[Kasprzak *et al.* Nature, '06]

Photon Condensate
$T \sim 300$K

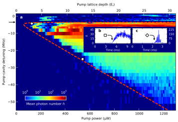


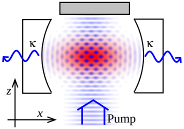
[Klaers *et al.* Nature, '10]

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



Superradiance transition
$T \sim 0$

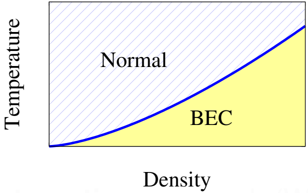


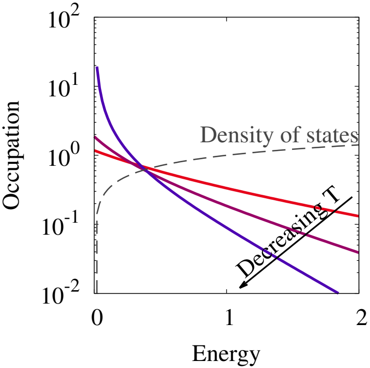


[Baumann *et al.* Nature, '10]

"Textbook" BEC

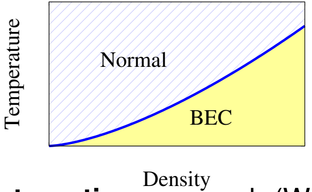
- **Non-interacting** viewpoint
  - BE distribution: $\mu < \omega_0$
  - $T_c = \frac{2\pi\hbar^2}{m}\left(\frac{n}{\xi_d}\right)^{2/d}$

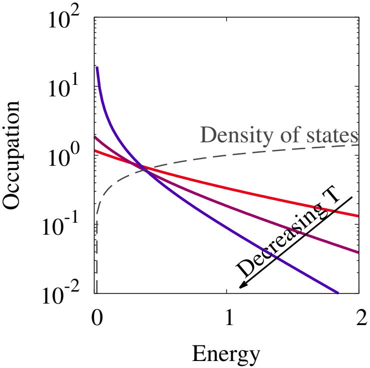




“Textbook” BEC

- **Non-interacting** viewpoint
  - BE distribution: $\mu < \omega_0$
  - $T_c = \frac{2\pi\hbar^2}{m}\left(\frac{n}{\xi_d}\right)^{2/d}$





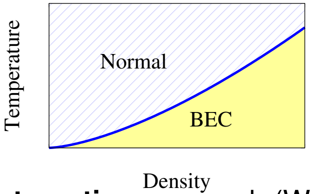
- **Interacting** approach (WIDBG)

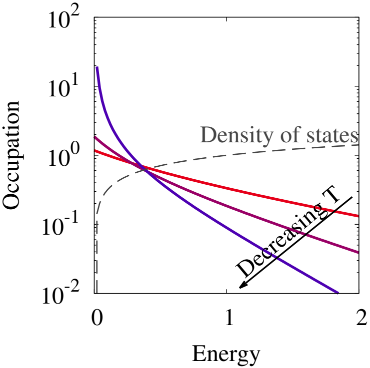
$$H = \sum_k \omega_k \psi_k^\dagger \psi_k + \frac{g}{2V} \sum_{k,k',q} \psi_{k+q}^\dagger \psi_{k'-q}^\dagger \psi_{k+q} \psi_k$$

  - Mean field: $|\psi|^2 = (\mu - \omega_0)/V$
  - Fluctuations deplete condensate, vanishes at $T > T_c$

“Textbook” BEC

- **Non-interacting** viewpoint
  - BE distribution: $\mu < \omega_0$
  - $T_c = \frac{2\pi\hbar^2}{m}\left(\frac{n}{\xi_d}\right)^{2/d}$





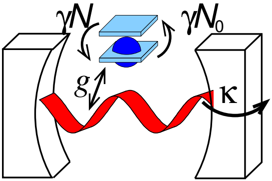
- **Interacting** approach (WIDBG)

$$H = \sum_k \omega_k \psi_k^\dagger \psi_k + \frac{g}{2V} \sum_{k,k',q} \psi_{k+q}^\dagger \psi_{k'-q}^\dagger \psi_{k+q} \psi_k$$

  - Mean field: $|\psi|^2 = (\mu - \omega_0)/V$
  - Fluctuations deplete condensate, vanishes at $T > T_c$

“Textbook” Laser: Maxwell Bloch equations

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



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

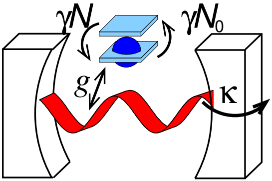
$$\partial_t \psi = -i\omega_0 \psi - \kappa \psi + \sum_\alpha g_\alpha P_\alpha$$

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

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

"Textbook" Laser: Maxwell Bloch equations

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

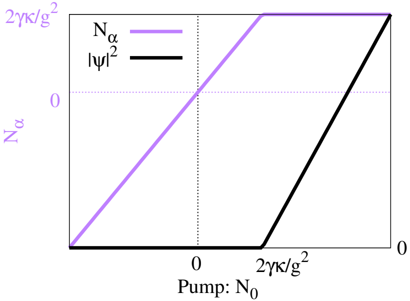


Maxwell-Bloch eqns: $P = -i\langle \sigma^- \rangle, N = 2\langle \sigma^z \rangle$

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

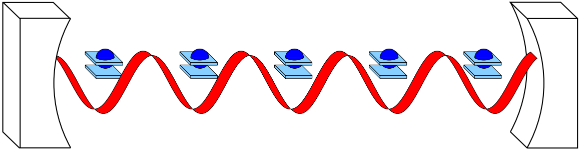
$$\partial_t P_\alpha = -2i\epsilon_\alpha P_\alpha - 2\gamma P_\alpha + g_\alpha \psi N_\alpha$$

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



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

“Textbook” Dicke-Hepp-Lieb superradiance

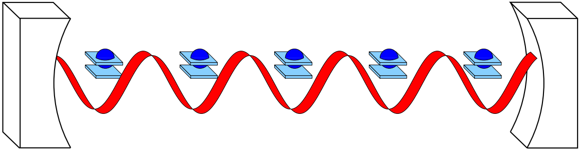


$$H = \omega \psi^\dagger \psi + \sum_\alpha \epsilon \sigma^z_\alpha + g \left(\psi^\dagger \sigma^-_\alpha + \psi \sigma^+_\alpha \right)$$

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

[Hepp, Lieb, Ann. Phys. ’73]

“Textbook” Dicke-Hepp-Lieb superradiance

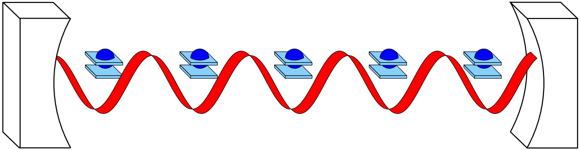


$$H = \omega\psi^\dagger\psi + \sum_\alpha \epsilon\sigma^z_\alpha + g\left(\psi^\dagger\sigma^-_\alpha + \psi\sigma^+_\alpha\right)$$

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

[Hepp, Lieb, Ann. Phys. ’73]

"Textbook" Dicke-Hepp-Lieb superradiance



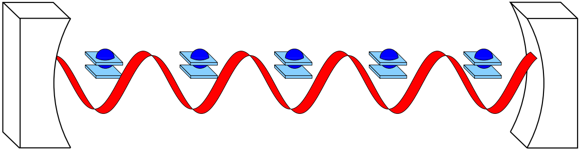
$$H = \omega\psi^\dagger\psi + \sum_\alpha \epsilon\sigma^z_\alpha + g\left(\psi^\dagger\sigma^-_\alpha + \psi\sigma^+_\alpha\right)$$

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

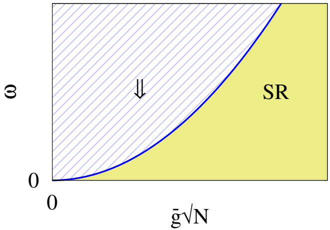
Spontaneous polarisation if: $Ng^2 > \omega\epsilon$

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

“Textbook” Dicke-Hepp-Lieb superradiance



$$H = \omega\psi^\dagger\psi + \sum_\alpha \epsilon\sigma^z_\alpha + g\left(\psi^\dagger\sigma^-_\alpha + \psi\sigma^+_\alpha\right)$$

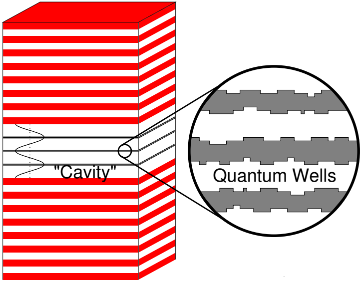


- Coherent state: $|\Psi\rangle \to e^{\lambda\psi^\dagger + \eta\sigma^+}|\Omega\rangle$
- Small g, min at $\lambda, \eta = 0$

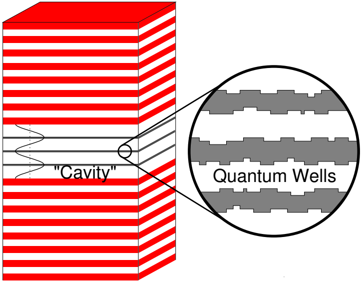
Spontaneous polarisation if: $Ng^2 > \omega\epsilon$

[Hepp, Lieb, Ann. Phys. ’73]

Microcavity polaritons

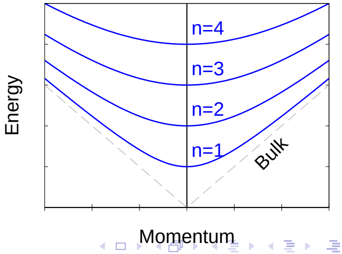


Microcavity polaritons

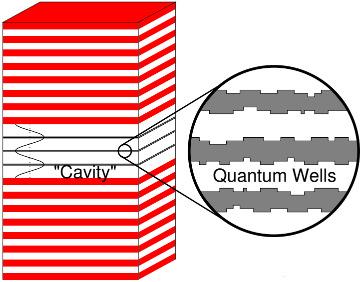


Cavity photons:

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

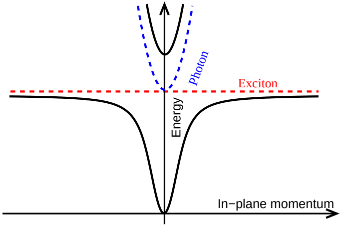


Microcavity polaritons

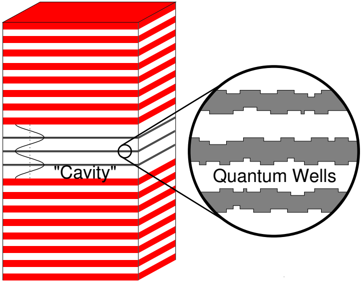


Cavity photons:

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

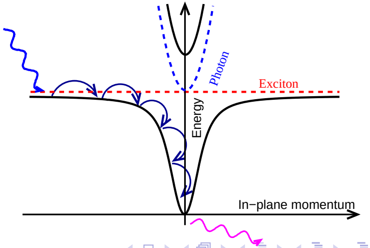


Microcavity polaritons

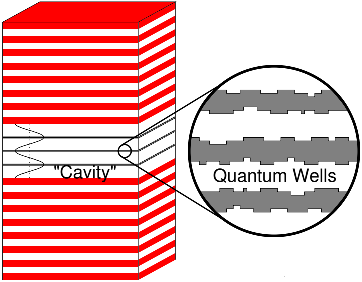


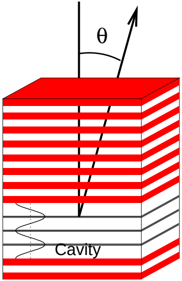
Cavity photons:

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



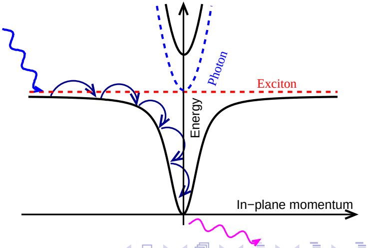
Microcavity polaritons



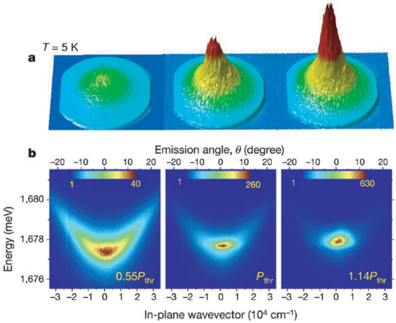


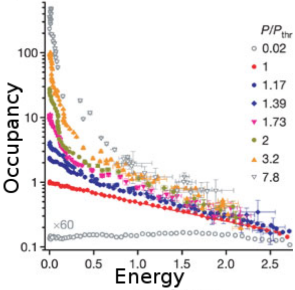
Cavity photons:

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



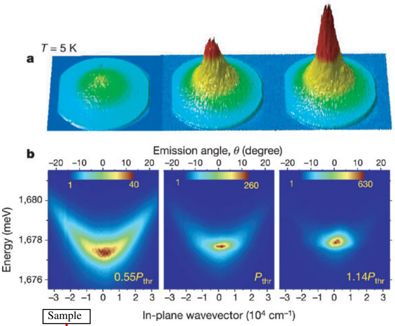
Polariton experiments: occupation and coherence

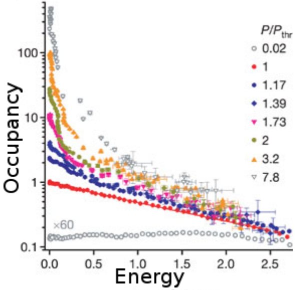


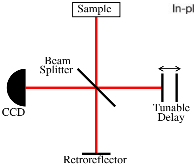


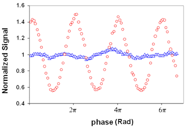
[Kasprzak, *et al.* Nature, '06]

Polariton experiments: occupation and coherence

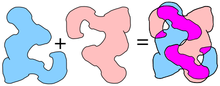


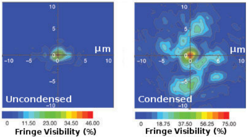






Coherence map:





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

Condensation-superradiance crossover

- Use model that can show lasing and condensation: Generalised Dicke model:

$$H_{\mathrm{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^{\dagger}_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^{z}_{\alpha} + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^{+}_{\alpha} + \psi^{\dagger}_{\mathbf{k}} \sigma^{-}_{\alpha} \right) \right]$$

- Grand canonical equilibrium, $\hat{H} - \mu \hat{N}$. Dicke superradiance:

- Many modes — fluctuations restore $T_c \propto n^{2/d}$

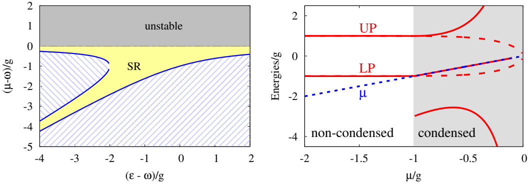
[JK et al, PRL '04, PRB '05, Review: Semicond. Sci. Tech. '07]

Condensation-superradiance crossover

- Use model that can show lasing and condensation:
  Generalised Dicke model:

$$H_{\text{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^z_\alpha + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^+_\alpha + \psi^\dagger_{\mathbf{k}} \sigma^-_\alpha \right) \right]$$

- Grand canonical equilibrium, $\hat{H} - \mu \hat{N}$. Dicke superradiance:



- Many modes — fluctuations restore $T_c \propto n^{2/d}$

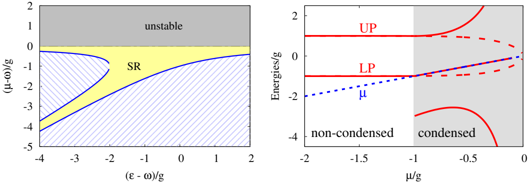
[JK et al, PRL ’04, PRB ’05, Review: Semicond. Sci. Tech. ’07]

Condensation-superradiance crossover

- Use model that can show lasing and condensation: Generalised Dicke model:

$$H_{\mathrm{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^z_\alpha + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^+_\alpha + \psi^\dagger_{\mathbf{k}} \sigma^-_\alpha \right) \right]$$

- Grand canonical equilibrium, $\hat{H} - \mu \hat{N}$. Dicke superradiance:



- Many modes — fluctuations restore $T_c \propto n^{2/d}$

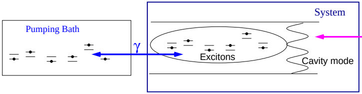
[JK et al, PRL '04, PRB '05, Review: Semicond. Sci. Tech. '07]

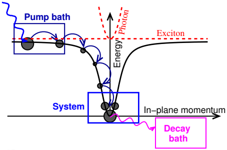
Lasing-condensation crossover

Generalised Dicke model:

$$H_{\text{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^z_\alpha + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^+_\alpha + \psi^\dagger_{\mathbf{k}} \sigma^-_\alpha \right) \right]$$

- Pumping and dissipation





- Mean field, $T_{\text{bath}} \to \infty$, Maxwell Bloch Laser
- Pump and decay $\to 0$, Equilibrium condensate

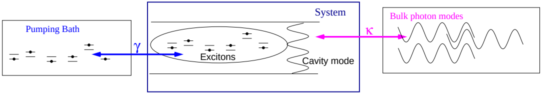
[Szymanska et al, PRL ’06, PRB ’07, Reviews: ’10, ’13

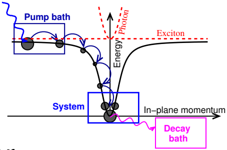
Lasing-condensation crossover

Generalised Dicke model:

$$H_{\text{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^z_\alpha + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^+_\alpha + \psi^\dagger_{\mathbf{k}} \sigma^-_\alpha \right) \right]$$

- Pumping and dissipation





- Mean field, $T_{\text{bath}} \to \infty$, Maxwell Bloch Laser
- Pump and decay $\to 0$, Equilibrium condensate

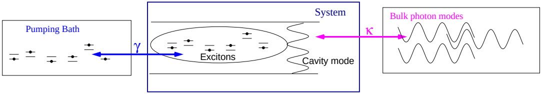
[Szymanska et al, PRL '06, PRB '07, Reviews: '10, '13

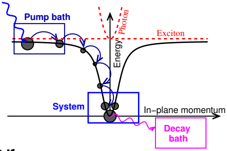
Lasing-condensation crossover

Generalised Dicke model:

$$H_{sys} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^\dagger_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^z_\alpha + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^+_\alpha + \psi^\dagger_{\mathbf{k}} \sigma^-_\alpha \right) \right]$$

- Pumping and dissipation





- Mean field, $T_{bath} \to \infty$, Maxwell Bloch Laser
- Pump and decay $\to 0$, Equilibrium condensate

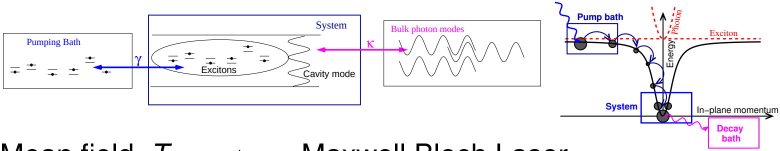
[Szymanska et al, PRL '06, PRB '07, Reviews: '10, '13

Lasing-condensation crossover

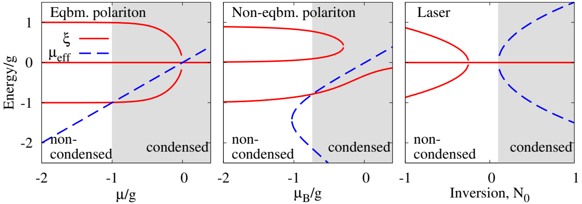
Generalised Dicke model:

$$H_{\text{sys}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \psi^{\dagger}_{\mathbf{k}} \psi_{\mathbf{k}} + \sum_{\alpha} \left[\frac{\epsilon}{2} \sigma^{z}_{\alpha} + g_{\alpha,\mathbf{k}} \left(\psi_{\mathbf{k}} \sigma^{+}_{\alpha} + \psi^{\dagger}_{\mathbf{k}} \sigma^{-}_{\alpha} \right) \right]$$

- Pumping and dissipation



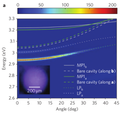
- Mean field, $T_{\text{bath}} \to \infty$, Maxwell Bloch Laser
- Pump and decay $\to 0$, Equilibrium condensate

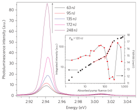


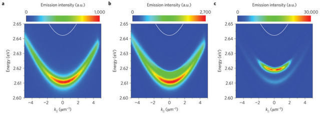
[Szymanska et al, PRL ’06, PRB ’07, Reviews: ’10, ’13

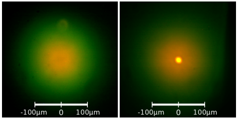
Matter-Light coupling with organic molecules

- What & why?









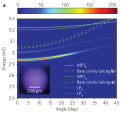
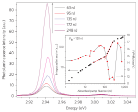
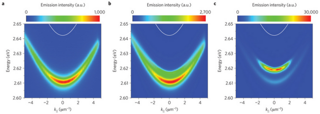
[Kena Cohen and Forrest, Nat. Photon '10; Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14] [Klaers *et al.* Nature '10]

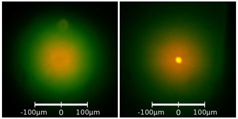
  - Wide variety of systems:
    polymers, fluorenes, J-aggregrates, molecular crystals.
  - Often large polariton splitting, $g\sqrt{N} \sim 0.1$ eV $\leftrightarrow$ 1000K

- Theory questions/challenges
  - Ultrastrong coupling
  - Vibrational modes
  - (Partial) thermalisation

Matter-Light coupling with organic molecules

- What & why?



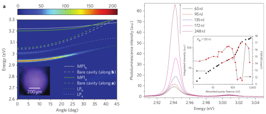
[Kena Cohen and Forrest, Nat. Photon ’10; Plumhoff *et al.* Nat. Materials ’14, Daskalakis *et al.* ibid ’14] [Klaers *et al.* Nature ’10]

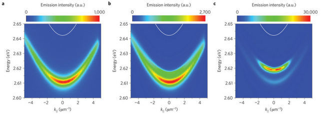
  - Wide variety of systems:
    polymers, fluorenes, J-aggregrates, molecular crystals.
  - Often large polariton splitting, $g\sqrt{N} \sim 0.1$ eV $\leftrightarrow$ 1000K

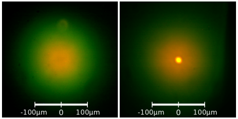
- Theory questions/challenges
  - Ultrastrong coupling
  - Vibrational modes
  - (Partial) thermalisation

Matter-Light coupling with organic molecules

- What & why?







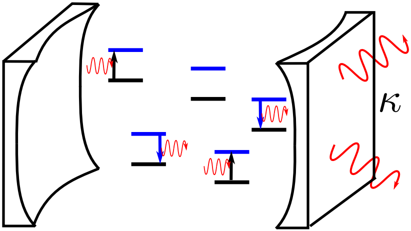
  [Kena Cohen and Forrest, Nat. Photon '10; Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* ibid '14] [Klaers *et al.* Nature '10]

  - Wide variety of systems:
    polymers, fluorenes, J-aggregrates, molecular crystals.
  - Often large polariton splitting, $g\sqrt{N} \sim 0.1$ eV $\leftrightarrow$ 1000K

- Theory questions/challenges
  - Ultrastrong coupling
  - Vibrational modes
  - (Partial) thermalisation

Dicke Holstein Model

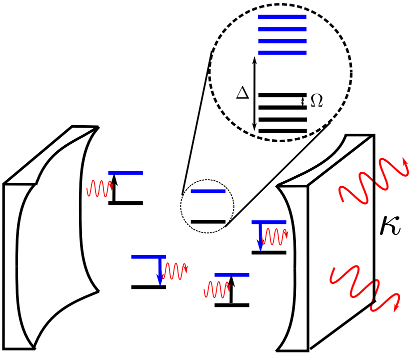
- Dicke model: 2LS $\leftrightarrow$ photons
- Molecular vibrational mode
  - Phonon frequency Ω
  - Huang-Rhys parameter S — coupling strength



$$H_{\text{sys}} = \omega\psi^{\dagger}\psi + \sum_{\alpha}\left[\frac{\epsilon}{2}\sigma^{z}_{\alpha} + g\left(\psi + \psi^{\dagger}\right)\left(\sigma^{+}_{\alpha} + \sigma^{-}_{\alpha}\right)\right]$$

Dicke Holstein Model

- Dicke model: 2LS $\leftrightarrow$ photons
- Molecular vibrational mode
  - Phonon frequency Ω
  - Huang-Rhys parameter S — coupling strength



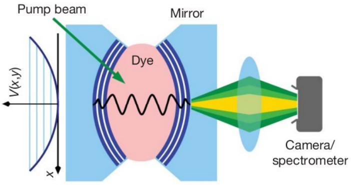
$$H_{\text{sys}} = \omega\psi^\dagger\psi + \sum_\alpha \left[\frac{\epsilon}{2}\sigma^z_\alpha + g\left(\psi + \psi^\dagger\right)\left(\sigma^+_\alpha + \sigma^-_\alpha\right)\right]$$

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

Overview

1. Introduction
2. Weak coupling: Photon BEC
   - Model & threshold
   - Time evolution
   - Pump-spot size dependence
3. Ground state spectrum
   - Ultra strong coupling & reconfiguration
   - Vibrational sidebands in spectrum

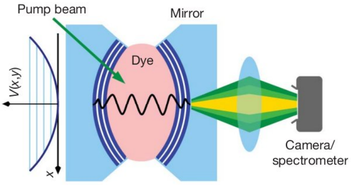
Photon BEC experiments

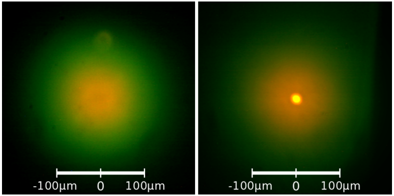


- Dye filled microcavity

[Klaers et al, Nature, 2010]

Photon BEC experiments

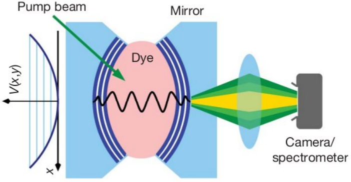


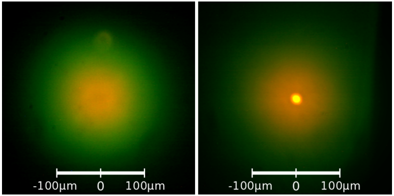


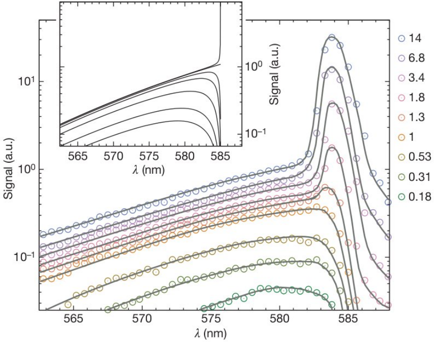
- Dye filled microcavity

[Klaers et al, Nature, 2010]

Photon BEC experiments







- Dye filled microcavity

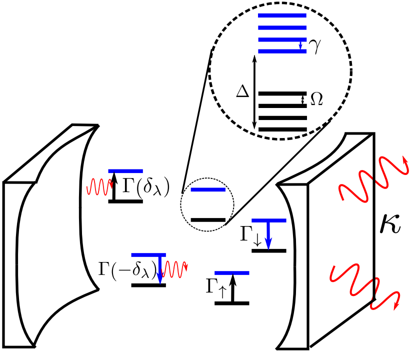
[Klaers et al, Nature, 2010]

Modelling

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

$$+ \sum_\alpha \Omega \left\{ b_\alpha^\dagger b_\alpha + \sqrt{S} \sigma_\alpha^z \left(b_\alpha^\dagger + b_\alpha \right) \right\}$$

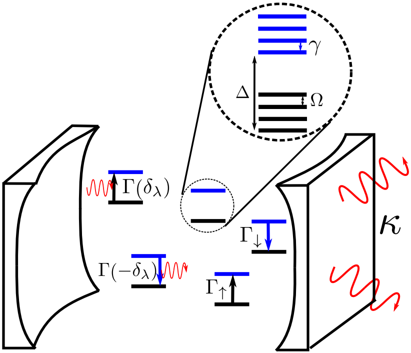
- **2D** harmonic oscillator
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in g



Modelling

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

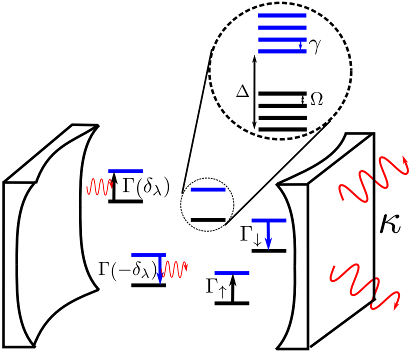
- **2D** harmonic oscillator
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in g



Modelling

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

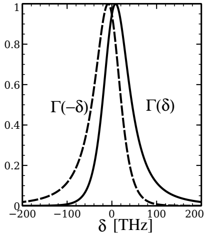
- **2D** harmonic oscillator
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in g



Modelling

Master equation

$$\dot{\rho} = -i[H_0, \rho] - \sum_m \frac{\kappa}{2}\mathcal{L}[\psi_m] - \sum_\alpha \left[\frac{\Gamma_\uparrow}{2}\mathcal{L}[\sigma^+_\alpha] + \frac{\Gamma_\downarrow}{2}\mathcal{L}[\sigma^-_\alpha]\right]$$
$$- \sum_{m,\alpha}\left[\frac{\Gamma(\delta_m = \omega_m - \epsilon)}{2}\mathcal{L}[\sigma^+_\alpha \psi_m] + \frac{\Gamma(-\delta_m = \epsilon - \omega_m)}{2}\mathcal{L}[\sigma^-_\alpha \psi^\dagger_m]\right]$$



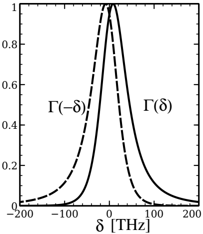
- Kennard-Stepanov $\Gamma(+\delta) \simeq \Gamma(-\delta)e^{\beta\delta}$

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

Modelling

Master equation

$$\dot{\rho} = -i[H_0, \rho] - \sum_m \frac{\kappa}{2}\mathcal{L}[\psi_m] - \sum_\alpha \left[\frac{\Gamma_\uparrow}{2}\mathcal{L}[\sigma_\alpha^+] + \frac{\Gamma_\downarrow}{2}\mathcal{L}[\sigma_\alpha^-]\right]$$
$$- \sum_{m,\alpha}\left[\frac{\Gamma(\delta_m = \omega_m - \epsilon)}{2}\mathcal{L}[\sigma_\alpha^+\psi_m] + \frac{\Gamma(-\delta_m = \epsilon - \omega_m)}{2}\mathcal{L}[\sigma_\alpha^-\psi_m^\dagger]\right]$$



- Kennard-Stepanov $\Gamma(+\delta) \simeq \Gamma(-\delta)e^{\beta\delta}$

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

Distribution $g_m n_m$

- Master equation → Rate equation

$$\partial_t n_m = -\kappa n_m + \Gamma(-\delta_m)(n_m + 1)N_\uparrow - \Gamma(\delta_m) n_m N_\downarrow$$

- Bose-Einstein distribution without losses

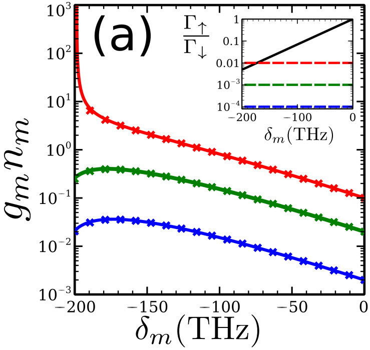
[Kirton & JK PRL '13]

Distribution $g_m n_m$

- Master equation $\to$ Rate equation

$$\partial_t n_m = -\kappa n_m + \Gamma(-\delta_m)(n_m+1)N_\uparrow - \Gamma(\delta_m) n_m N_\downarrow$$

- Bose-Einstein distribution without losses



Low loss: Thermal

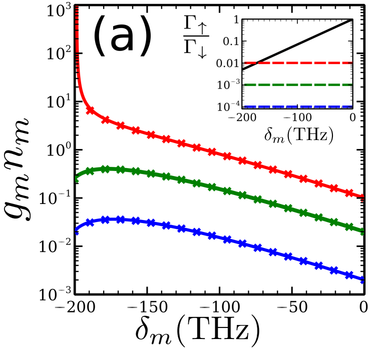
[Kirton & JK PRL '13]

Distribution $g_m n_m$

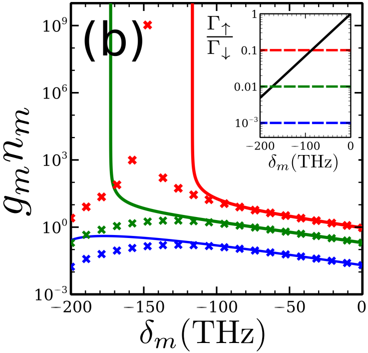
- Master equation $\rightarrow$ Rate equation

$$\partial_t n_m = -\kappa n_m + \Gamma(-\delta_m)(n_m+1)N_\uparrow - \Gamma(\delta_m) n_m N_\downarrow$$

- Bose-Einstein distribution without losses



Low loss: Thermal



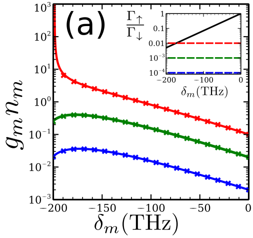
High loss $\rightarrow$ Laser

[Kirton & JK PRL '13]

Chemical potential?

- Steady state distribution:

$$\frac{n_m}{n_m+1} = \frac{\Gamma(-\delta_m)N_\uparrow}{\kappa + \Gamma(\delta_m)N_\downarrow}$$



- $\kappa \ll N\Gamma(\delta)$, Kennard-Stepanov

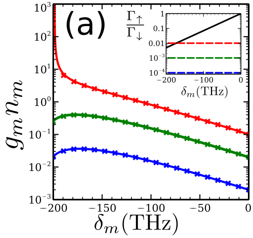
$$\frac{n_m}{n_m+1} = e^{-\beta\delta_m+\beta\mu}, \qquad e^{\beta\mu} = \frac{N_\uparrow}{N_\downarrow} = \frac{\Gamma_\uparrow + \sum_m \Gamma(\delta_m)n_m}{\Gamma_\downarrow + \sum_m \Gamma(-\delta_m)(n_m+1)}$$

- Below threshold, $\mu = k_B T \ln[\Gamma_\uparrow/\Gamma_\downarrow]$
- At/above threshold, $\mu \to \delta_0$

Chemical potential?

- Steady state distribution:

$$\frac{n_m}{n_m+1} = \frac{\Gamma(-\delta_m)N_\uparrow}{\kappa + \Gamma(\delta_m)N_\downarrow}$$



- $\kappa \ll N\Gamma(\delta)$, Kennard-Stepanov

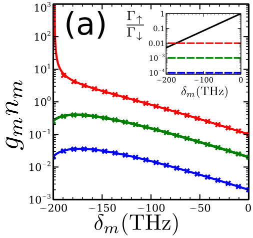
$$\frac{n_m}{n_m+1} = e^{-\beta\delta_m+\beta\mu}, \qquad e^{\beta\mu} \equiv \frac{N_\uparrow}{N_\downarrow} = \frac{\Gamma_\uparrow + \sum_m \Gamma(\delta_m)n_m}{\Gamma_\downarrow + \sum_m \Gamma(-\delta_m)(n_m+1)}$$

- Below threshold, $\mu = k_B T \ln[\Gamma_\uparrow/\Gamma_\downarrow]$
- At/above threshold, $\mu \to \delta_0$

Chemical potential?

- Steady state distribution:

$$\frac{n_m}{n_m+1} = \frac{\Gamma(-\delta_m)N_\uparrow}{\kappa + \Gamma(\delta_m)N_\downarrow}$$



- $\kappa \ll N\Gamma(\delta)$, Kennard-Stepanov

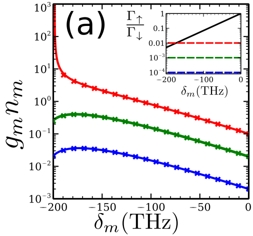
$$\frac{n_m}{n_m+1} = e^{-\beta\delta_m+\beta\mu}, \qquad e^{\beta\mu} \equiv \frac{N_\uparrow}{N_\downarrow} = \frac{\Gamma_\uparrow + \sum_m \Gamma(\delta_m)n_m}{\Gamma_\downarrow + \sum_m \Gamma(-\delta_m)(n_m+1)}$$

- Below threshold, $\mu = k_B T \ln[\Gamma_\uparrow/\Gamma_\downarrow]$
- At/above threshold, $\mu \to \delta_0$

Chemical potential?

- Steady state distribution:

$$\frac{n_m}{n_m+1} = \frac{\Gamma(-\delta_m)N_\uparrow}{\kappa + \Gamma(\delta_m)N_\downarrow}$$



- $\kappa \ll N\Gamma(\delta)$, Kennard-Stepanov

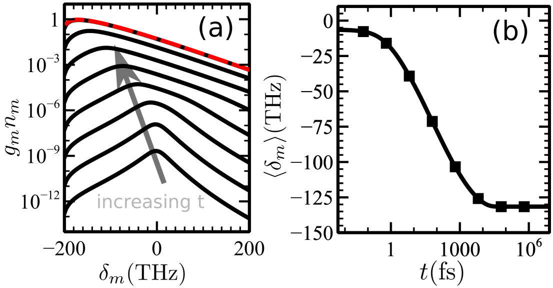
$$\frac{n_m}{n_m+1} = e^{-\beta\delta_m+\beta\mu}, \qquad e^{\beta\mu} \equiv \frac{N_\uparrow}{N_\downarrow} = \frac{\Gamma_\uparrow + \sum_m \Gamma(\delta_m)n_m}{\Gamma_\downarrow + \sum_m \Gamma(-\delta_m)(n_m+1)}$$

- Below threshold, $\mu = k_B T \ln[\Gamma_\uparrow/\Gamma_\downarrow]$
- At/above threshold, $\mu \to \delta_0$

Time evolution

Time evolution

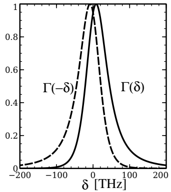
- Initial state: excited molecules
- Initial emission, follows gain peak
- Thermalisation by repeated absorption

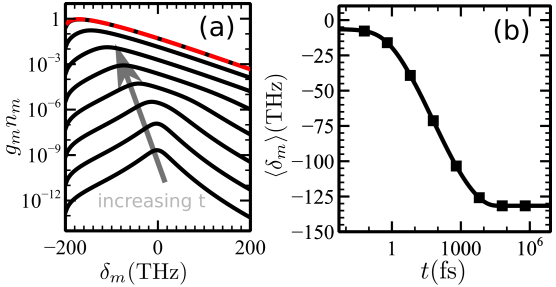


[Kirton & JK arXiv:1410.6632]

Time evolution

- Initial state: excited molecules
- Initial emission, follows gain peak
- Thermalisation by repeated absorption

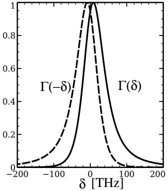


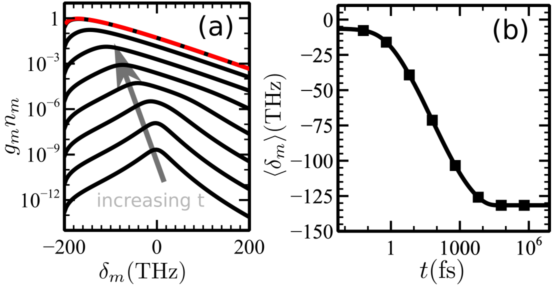


[Kirton & JK arXiv:1410.6632]

Time evolution

- Initial state: excited molecules
- Initial emission, follows gain peak
- Thermalisation by repeated absorption





[Kirton & JK arXiv:1410.6632]

Pump spot size

1. Introduction
2. Weak coupling: Photon BEC
   - Model & threshold
   - Time evolution
   - Pump-spot size dependence
3. Ground state spectrum
   - Ultra strong coupling & reconfiguration
   - Vibrational sidebands in spectrum

Spatially varying pump intensity

- Consider effects of pump profile, $\Gamma_\uparrow(\mathbf{r}) = \Gamma_\uparrow \exp(-r^2/2\sigma_{\text{pump}}^2)$
- Experiments: [Marelic & Nyman, arXiv:1410.6822]

- Varying excited density – differential coupling to modes

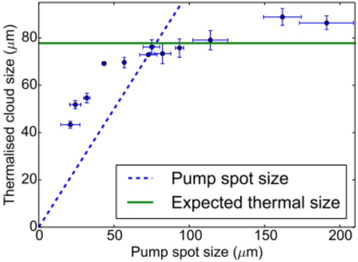
$$\partial_t \rho_\uparrow(\mathbf{r}) = -\tilde{\Gamma}_\downarrow(\mathbf{r})\rho_\uparrow(\mathbf{r}) + \tilde{\Gamma}_\uparrow(\mathbf{r})\rho_\downarrow(\mathbf{r}))$$

$$\partial_t n_m = \Gamma(-\delta_m) O_m (n_m + 1) - \left[\kappa + \Gamma(\delta_m)(\rho_m - O_m)\right] n_m$$

$$O_m = \int d\mathbf{r}\, \rho_\uparrow(\mathbf{r}) |\psi_m(\mathbf{r})|^2, \qquad \rho_\uparrow + \rho_\downarrow = \rho_m$$

Spatially varying pump intensity

- Consider effects of pump profile, $\Gamma_\uparrow(\mathbf{r}) = \Gamma_\uparrow \exp(-r^2/2\sigma^2_{\text{pump}})$
- Experiments: [Marelic & Nyman, `arXiv:1410.6822`]



- Varying excited density – differential coupling to modes

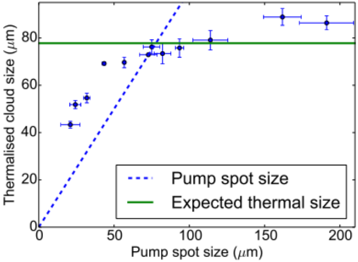
$$\partial_t \rho_\uparrow(\mathbf{r}) = -\tilde{\Gamma}_\downarrow(\mathbf{r})\rho_\uparrow(\mathbf{r}) + \tilde{\Gamma}_\uparrow(\mathbf{r})\rho_\downarrow(\mathbf{r}))$$

$$\partial_t n_m = \Gamma(-\delta_m) O_m (n_m + 1) - [\kappa + \Gamma(\delta_m)(\rho_m - O_m)] n_m$$

$$O_m = \int d\mathbf{r} \rho_\uparrow(\mathbf{r}) |\psi_m(\mathbf{r})|^2, \qquad \rho_\uparrow + \rho_\downarrow = \rho_m$$

Spatially varying pump intensity

- Consider effects of pump profile, $\Gamma_\uparrow(\mathbf{r}) = \Gamma_\uparrow \exp(-r^2/2\sigma_{\text{pump}}^2)$
- Experiments: [Marelic & Nyman, arXiv:1410.6822]



- Varying excited density – differential coupling to modes

$$\partial_t \rho_\uparrow(\mathbf{r}) = -\tilde{\Gamma}_\downarrow(\mathbf{r})\rho_\uparrow(\mathbf{r}) + \tilde{\Gamma}_\uparrow(\mathbf{r})\rho_\downarrow(\mathbf{r}))$$

$$\partial_t n_m = \Gamma(-\delta_m) O_m (n_m + 1) - [\kappa + \Gamma(\delta_m)(\rho_m - O_m)]\, n_m$$

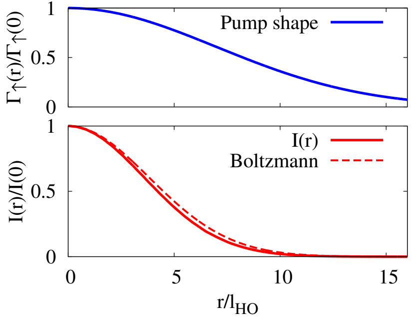
$$O_m = \int d\mathbf{r} \rho_\uparrow(\mathbf{r}) |\psi_m(\mathbf{r})|^2, \qquad \rho_\uparrow + \rho_\downarrow = \rho_m$$

Spatially varying pump: below threshold

- Far below threshold:
  - Excitation: $\rho_\uparrow(\mathbf{r}) \simeq \rho_m \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} \ll \rho_m$
  - If $\kappa \ll \rho_m \Gamma(\delta_m)$, $\frac{n_m}{n_m+1} \simeq e^{-\beta\delta_m} \times \int d\mathbf{r} \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} |\psi_m(\mathbf{r})|^2$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$

Spatially varying pump: below threshold

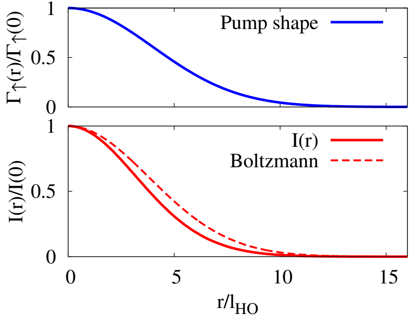
- Far below threshold:
  - Excitation: $\rho_\uparrow(\mathbf{r}) \simeq \rho_m \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} \ll \rho_m$
  - If $\kappa \ll \rho_m \Gamma(\delta_m)$, $\quad \frac{n_m}{n_m+1} \simeq e^{-\beta\delta_m} \times \int d\mathbf{r} \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} |\psi_m(\mathbf{r})|^2$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$



- $\eta = \frac{\kappa}{\rho_m \max[\Gamma(\delta)]}$

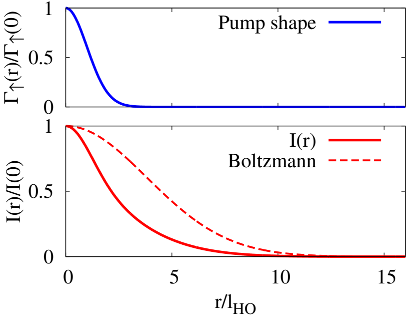
Spatially varying pump: below threshold

- Far below threshold:
  - Excitation: $\rho_\uparrow(\mathbf{r}) \simeq \rho_m \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} \ll \rho_m$
  - If $\kappa \ll \rho_m \Gamma(\delta_m)$, $\frac{n_m}{n_m+1} \simeq e^{-\beta\delta_m} \times \int d\mathbf{r} \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} |\psi_m(\mathbf{r})|^2$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$



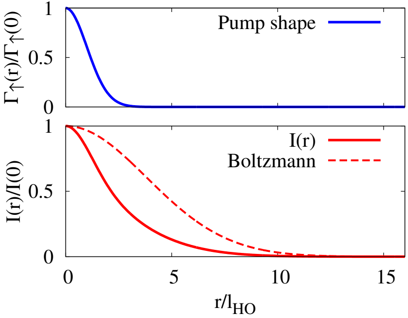
Spatially varying pump: below threshold

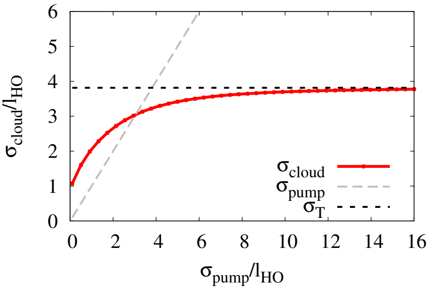
- Far below threshold:
  - Excitation: $\rho_\uparrow(\mathbf{r}) \simeq \rho_m \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} \ll \rho_m$
  - If $\kappa \ll \rho_m \Gamma(\delta_m)$, $\quad \frac{n_m}{n_m+1} \simeq e^{-\beta\delta_m} \times \int d\mathbf{r} \frac{\Gamma_\uparrow(\mathbf{r})}{\Gamma_\downarrow} |\psi_m(\mathbf{r})|^2$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$



Spatially varying pump: below threshold

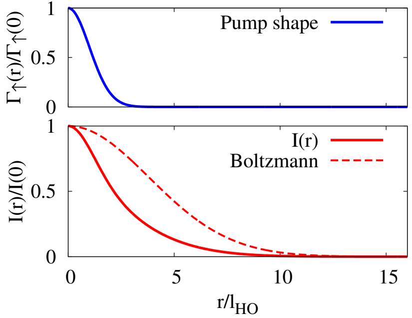
- Far below threshold:
  - Excitation: $\rho_{\uparrow}(\mathbf{r}) \simeq \rho_m \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} \ll \rho_m$
  - If $\kappa \ll \rho_m \Gamma(\delta_m)$, $\frac{n_m}{n_m + 1} \simeq e^{-\beta\delta_m} \times \int d\mathbf{r} \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} |\psi_m(\mathbf{r})|^2$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$

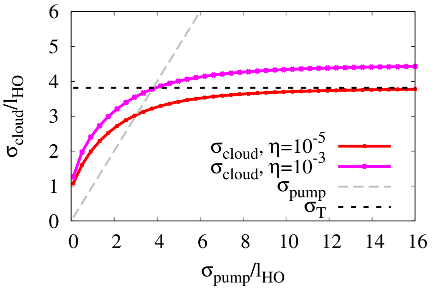




Spatially varying pump: below threshold

- Far below threshold:
  - Excitation: $\rho_{\uparrow}(\mathbf{r}) \simeq \rho_m \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} \ll \rho_m$
  - If $\kappa \ll \rho_m \Gamma(\delta_m)$, $\quad \frac{n_m}{n_m + 1} \simeq e^{-\beta \delta_m} \times \int d\mathbf{r} \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} |\psi_m(\mathbf{r})|^2$
- Resulting profile, $I(\mathbf{r}) = \sum_m n_m |\psi_m(r)|^2$

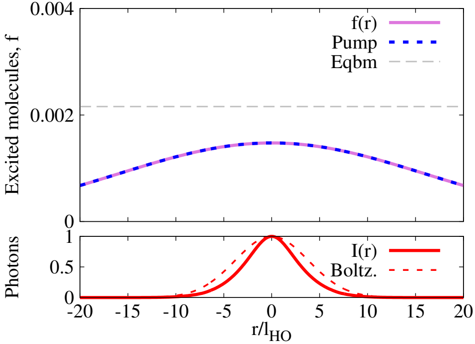




  - $\eta = \frac{\kappa}{\rho_m \max[\Gamma(\delta)]}$

Near threshold behaviour

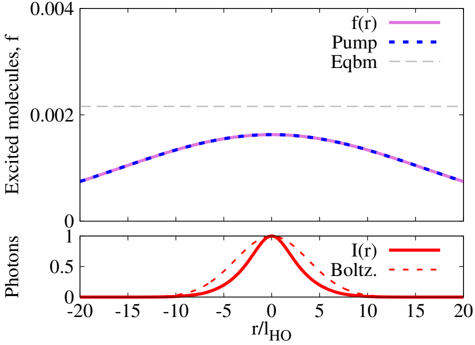
- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$
- Non Boltzmann peak — BEC
- "Gain saturation" at centre



- Saturation of $f(r) = 1/(1+e^{-\beta\mu})$ — spatial equilibriation

Near threshold behaviour

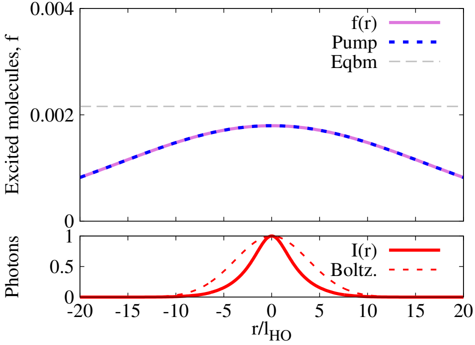
- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$
- Non Boltzmann peak — BEC
- "Gain saturation" at centre



- Saturation of $f(r) = 1/(1 + e^{-\beta\mu})$ — spatial equilibriation

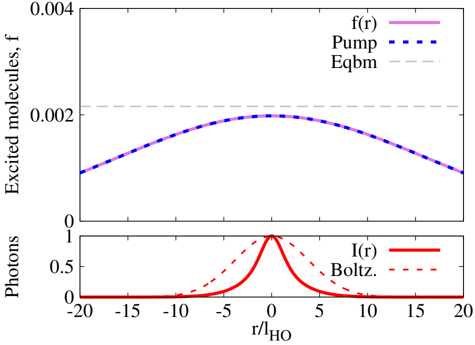
Near threshold behaviour

- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$



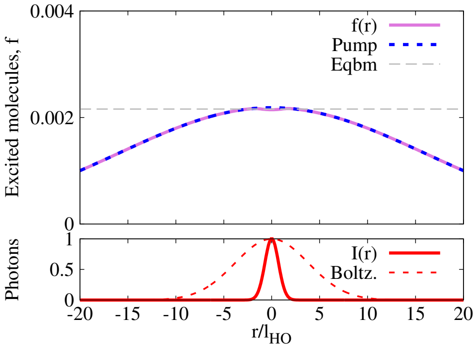
Near threshold behaviour

- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$



Near threshold behaviour

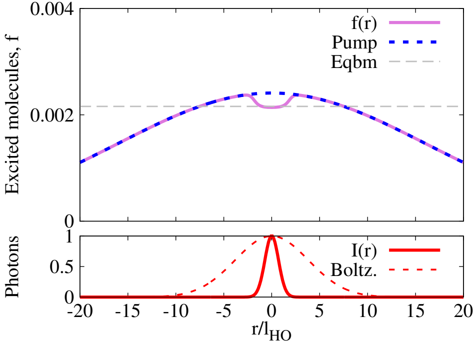
- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$
- Non Boltzmann peak — BEC
- "Gain saturation" at centre



- Saturation of $f(r) = 1/(1 + e^{-\beta\mu})$ — spatial equilibriation

Near threshold behaviour

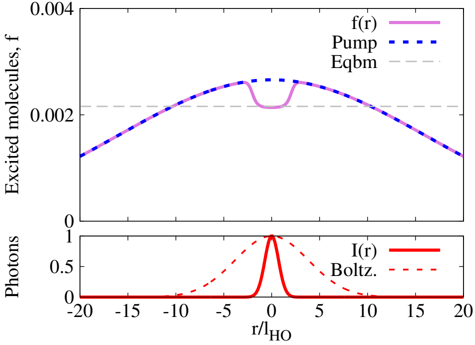
- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$
- Non Boltzmann peak — BEC
- “Gain saturation” at centre



- Saturation of $f(r) = 1/(1 + e^{-\beta\mu})$ — spatial equilibriation

Near threshold behaviour

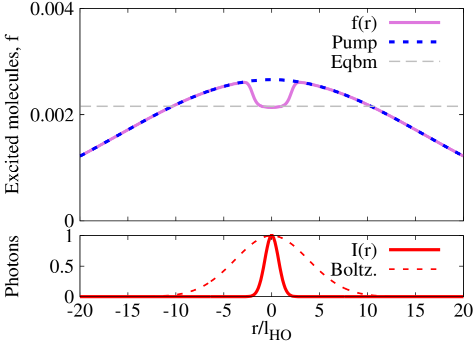
- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$
- Non Boltzmann peak — BEC
- “Gain saturation” at centre



- Saturation of $f(r) = 1/(1+e^{-\beta\mu})$ — spatial equilibriation

Near threshold behaviour

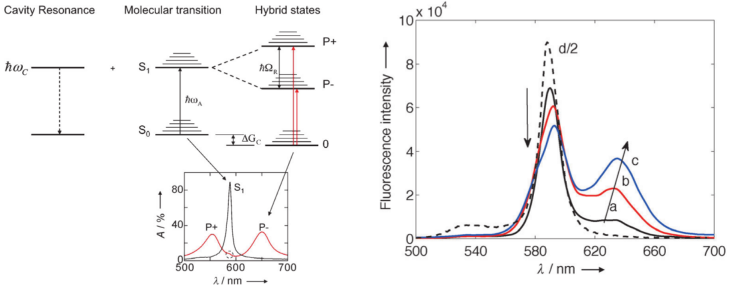
- Large spot, $\sigma_{\text{pump}} \gg l_{\text{HO}}$
- Non Boltzmann peak — BEC
- "Gain saturation" at centre



- Saturation of $f(r) = 1/(1 + e^{-\beta\mu})$ — spatial equilibriation

Ultra-strong coupling, changing configuration

- Ultra-strong coupling: $\omega, \epsilon \sim g\sqrt{N} \propto \sqrt{\text{concentration}}$
- Normal state: configuration of molecules



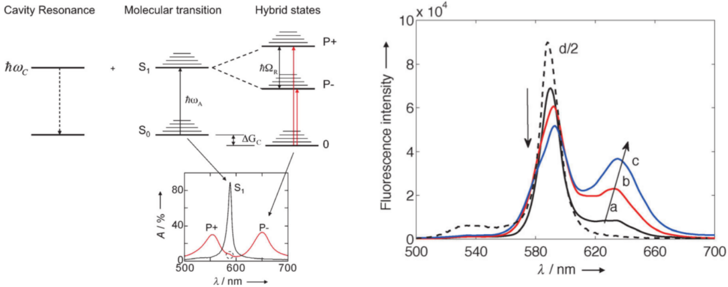
[Canaguier-Durand *et al.* Angew. Chem. '13]

- Polariton *vs* molecular spectral weight – chemical eqbm
- Temperature dependent

- Questions:

Ultra-strong coupling, changing configuration

- Ultra-strong coupling: $\omega, \epsilon \sim g\sqrt{N} \propto \sqrt{\text{concentration}}$
- Normal state: configuration of molecules

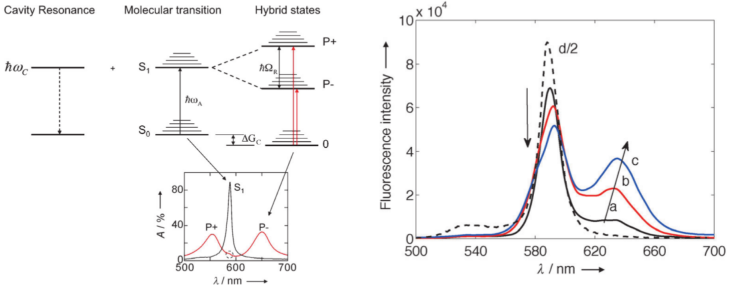


[Canaguier-Durand *et al.* Angew. Chem. '13]

- Polariton *vs* molecular spectral weight – chemical eqbm
- Temperature dependent

Ultra-strong coupling, changing configuration

- Ultra-strong coupling: $\omega, \epsilon \sim g\sqrt{N} \propto \sqrt{\text{concentration}}$
- Normal state: configuration of molecules



[Canaguier-Durand *et al.* Angew. Chem. '13]

- Polariton *vs* molecular spectral weight – chemical eqbm
- Temperature dependent

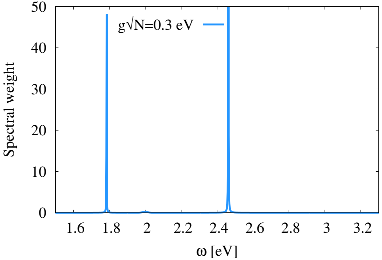
- Questions:
  - Microscopic picture?
  - Vibrationally dressed spectrum + disorder

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + (\text{interference})$$

- Ultra-strong coupling — renormalised photon



- Central peak:

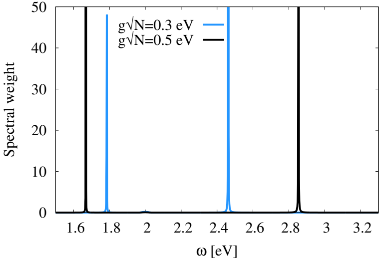
- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + \text{(interference)}$$

- Ultra-strong coupling — renormalised photon



- Central peak:

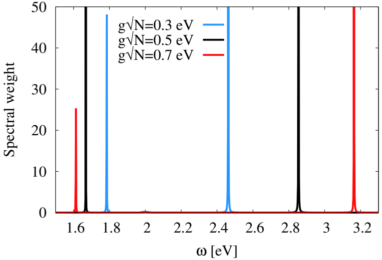
- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + \text{(interference)}$$

- Ultra-strong coupling — renormalised photon



- Central peak:

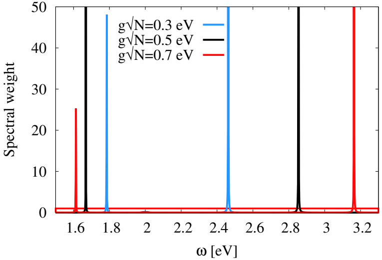
- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + (\text{interference})$$

- Ultra-strong coupling — renormalised photon



- Central peak:

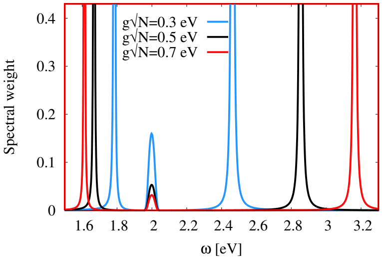
- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + \text{(interference)}$$

- Ultra-strong coupling — renormalised photon



- Central peak:

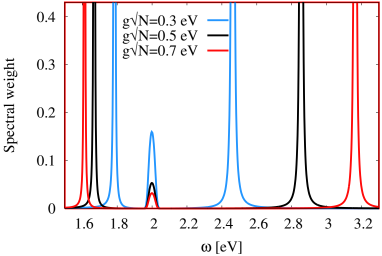
- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + (\text{interference})$$

- Ultra-strong coupling — renormalised photon



- Central peak:

$$G^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_k - g^2 G^R_{Exc.}(\nu)}$$

$$A(\nu) \sim \left(\frac{\kappa}{2} - \Im[G^R_{\text{Exc.}}]\right) \left|G^R(\nu)\right|^2$$

[Houdré *et al.* , PRA '96]

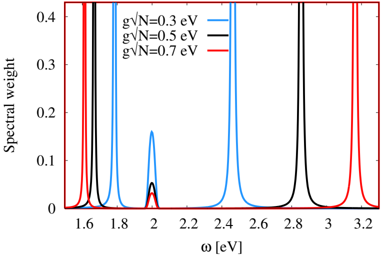
- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Disordered molecules — spectrum

- Calculate Green's function $G^R(\nu)$:

$$T(\nu) \propto |G^R(\nu)|^2, \qquad A(\nu) \propto -\Im[G^R(\nu)] + (\text{interference})$$

- Ultra-strong coupling — renormalised photon



- Central peak:

$$G^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_k - g^2 G^R_{Exc.}(\nu)}$$

$$A(\nu) \sim \left(\frac{\kappa}{2} - \Im[G^R_{\text{Exc.}}]\right) \left|G^R(\nu)\right|^2$$

[Houdré *et al.* , PRA '96]

- Temperature independent (for $k_B T \ll g\sqrt{N}$)

Molecular reconfiguration

- Central peak — depends on g, not T.
- Can g_{eff} depend on T?
- Rotational degrees of freedom

$$H = \ldots + \sum_{\alpha} \left[\ldots + g_{\alpha,k} \cos(\theta_\alpha)(\psi^\dagger_k + \psi_{-k})\sigma^x_\alpha + E_0(\theta_\alpha) \right]$$

- Schrieffer-Wolff, $\delta H = \sum_{\alpha,k} g_{\alpha,k}(\psi^\dagger_k \sigma^+_\alpha + \text{H.c.})$:

$$H_{\text{eff}} = \ldots + \sum_{\alpha} \left[K_0 \cos^2(\theta_\alpha) + E_0(\theta) \right], \quad K_0 = \sum_k \frac{g_k^2}{\omega_k + \epsilon}$$

Molecular reconfiguration

- Central peak — depends on g, not T.
- Can g_{eff} depend on T?

- Rotational degrees of freedom

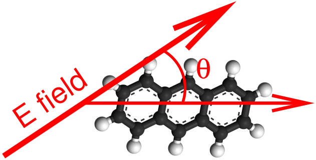
$$H = \ldots + \sum_{\alpha} \left[\ldots + g_{\alpha,k} \cos(\theta_\alpha)(\psi_k^\dagger + \psi_{-k})\sigma_\alpha^x + E_0(\theta_\alpha) \right]$$

- Schrieffer-Wolff, $\delta H = \sum_{\alpha,k} g_{\alpha,k}(\psi_k^\dagger \sigma_\alpha^+ + \text{H.c.})$:

$$H_{\text{eff}} = \ldots + \sum_{\alpha} \left[K_0 \cos^2(\theta_\alpha) + E_0(\theta) \right], \quad K_0 = \sum_k \frac{g_k^2}{\omega_k + \epsilon}$$

Molecular reconfiguration

- Central peak — depends on g, not T.
- Can g_{eff} depend on T?



- Rotational degrees of freedom

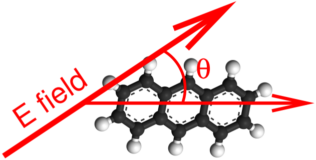
$$H = \ldots + \sum_{\alpha} \left[\ldots + g_{\alpha,k} \cos(\theta_\alpha)(\psi_k^\dagger + \psi_{-k})\sigma_\alpha^x + E_0(\theta_\alpha) \right]$$

- Schrieffer-Wolff, $\delta H = \sum_{\alpha,k} g_{\alpha,k}(\psi_k^\dagger \sigma_\alpha^- + \text{H.c.})$:

$$H_{\text{eff}} = \ldots + \sum_{\alpha} \left[K_0 \cos^2(\theta_\alpha) + E_0(\theta) \right], \quad K_0 = \sum_k \frac{g_k^2}{\omega_k + \epsilon}$$

Molecular reconfiguration

- Central peak — depends on g, not T.
- Can g_{eff} depend on T?



- Rotational degrees of freedom

$$H = \ldots + \sum_{\alpha} \left[\ldots + g_{\alpha,k} \cos(\theta_\alpha)(\psi_k^\dagger + \psi_{-k})\sigma_\alpha^x + E_0(\theta_\alpha) \right]$$

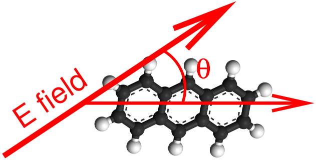
- Schrieffer-Wolff, $\delta H = \sum_{\alpha,k} g_{\alpha,k}(\psi_k^\dagger \sigma_\alpha^+ + \text{H.c.})$:

$$H_{\text{eff}} = \ldots + \sum_{\alpha} \left[K_0 \cos^2(\theta_\alpha) + E_0(\theta) \right], \quad K_0 = \sum_k \frac{g_{\mathbf{k}}^2}{\omega_k + \epsilon}$$

  - No $\sqrt{N}$ enhancement — K_0 small, independent of density

Molecular reconfiguration

- Central peak — depends on g, not T.
- Can g_{eff} depend on T?



- Rotational degrees of freedom

$$H = \ldots + \sum_{\alpha} \left[\ldots + g_{\alpha,k} \cos(\theta_\alpha)(\psi_k^\dagger + \psi_{-k})\sigma_\alpha^x + E_0(\theta_\alpha) \right]$$

- Schrieffer-Wolff, $\delta H = \sum_{\alpha,k} g_{\alpha,k}(\psi_k^\dagger \sigma_\alpha^+ + \text{H.c.})$:

$$H_{\text{eff}} = \ldots + \sum_{\alpha} \left[K_0 \cos^2(\theta_\alpha) + E_0(\theta) \right], \quad K_0 = \sum_k \frac{g_{\mathbf{k}}^2}{\omega_k + \epsilon}$$

  - No $\sqrt{N}$ enhancement — K_0 small, independent of density

Vibrational reconfiguration

- Schrieffer-Wolff – mixes vibrational states

$$H_{\text{eff}} = H_0 - \frac{g^2 N}{2(\epsilon+\omega)}\left\{1 - \frac{\Omega\sqrt{S}(b+b^\dagger)}{\epsilon+\omega} + \mathcal{O}\left[\left(\frac{\Omega}{\epsilon}\right)^2, \frac{g\sqrt{N}}{\epsilon}\right]\right\}$$

- Reduced vibrational offset

$$S \rightarrow S(1-2K_1), \qquad K_1 = \sum_k \frac{g_k^2}{(\omega_k+\epsilon)^2}$$

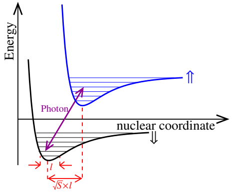
Vibrational reconfiguration

- Schrieffer-Wolff – mixes vibrational states

$$H_{\text{eff}} = H_0 - \frac{g^2 N}{2(\epsilon + \omega)} \left\{ 1 - \frac{\Omega\sqrt{S}(b + b^\dagger)}{\epsilon + \omega} + \mathcal{O}\left[\left(\frac{\Omega}{\epsilon}\right)^2, \frac{g\sqrt{N}}{\epsilon} \right] \right\}$$

- Reduced vibrational offset

$$S \to S(1 - 2K_1), \quad K_1 = \sum_k \frac{g_{\mathbf{k}}^2}{(\omega_k + \epsilon)^2}$$



- Increased effective coupling: $g_{\text{eff}}^2 = g^2 \exp(-S)$
- Again, $K_1 \ll 1$, independent of density.

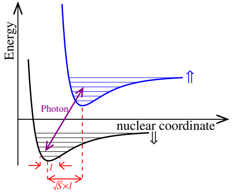
Vibrational reconfiguration

- Schrieffer-Wolff – mixes vibrational states

$$H_{\mathrm{eff}} = H_0 - \frac{g^2 N}{2(\epsilon+\omega)} \left\{ 1 - \frac{\Omega\sqrt{S}(b+b^\dagger)}{\epsilon+\omega} + \mathcal{O}\left[\left(\frac{\Omega}{\epsilon}\right)^2, \frac{g\sqrt{N}}{\epsilon} \right] \right\}$$

- Reduced vibrational offset

$$S \to S(1-2K_1), \quad K_1 = \sum_k \frac{g_{\mathbf{k}}^2}{(\omega_k+\epsilon)^2}$$



  - Increased effective coupling: $g_{\mathrm{eff}}^2 = g^2 \exp(-S)$
  - Again, $K_1 \ll 1$, independent of density.

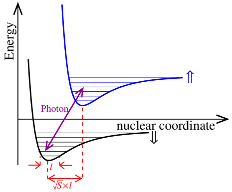
Vibrational reconfiguration

- Schrieffer-Wolff – mixes vibrational states

$$H_{\text{eff}} = H_0 - \frac{g^2 N}{2(\epsilon+\omega)} \left\{ 1 - \frac{\Omega\sqrt{S}(b+b^\dagger)}{\epsilon+\omega} + \mathcal{O}\left[\left(\frac{\Omega}{\epsilon}\right)^2, \frac{g\sqrt{N}}{\epsilon} \right] \right\}$$

- Reduced vibrational offset

$$S \to S(1-2K_1), \quad K_1 = \sum_k \frac{g_{\mathbf{k}}^2}{(\omega_k+\epsilon)^2}$$



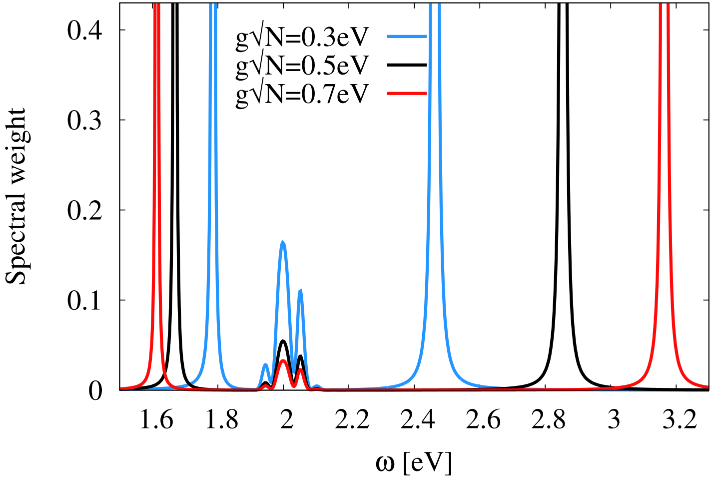
  - Increased effective coupling: $g_{\text{eff}}^2 = g^2 \exp(-S)$
  - Again, $K_1 \ll 1$, independent of density.

Disordered molecules — vibrational mode

- But: spectrum with vibrational sidebands, $S = 0.02$

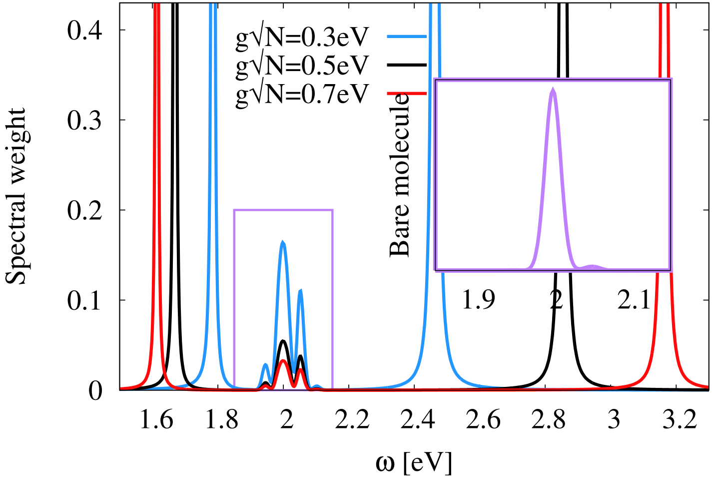
Disordered molecules — vibrational mode

- But: spectrum with vibrational sidebands, $S = 0.02$



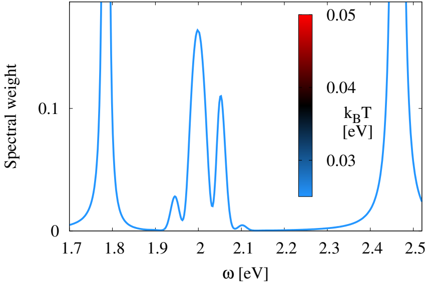
Disordered molecules — vibrational mode

- But: spectrum with vibrational sidebands, $S = 0.02$



Disordered molecules + vibrations – *vs* temperature

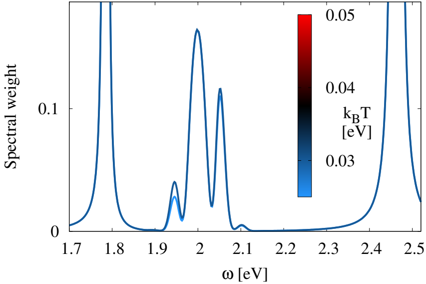
- *vs* vs temperature



- Stronger disorder & $S = 0.5, \sigma = 0.025$eV

Disordered molecules + vibrations – *vs* temperature

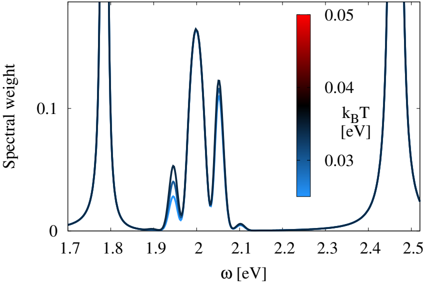
- *vs* vs temperature



- Stronger disorder & $S = 0.5, \sigma = 0.025$eV

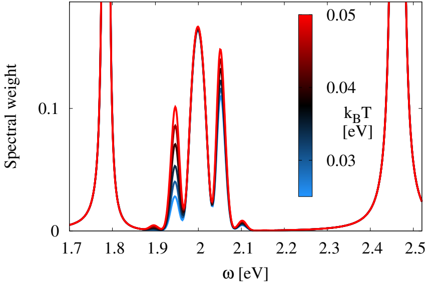
Disordered molecules + vibrations – *vs* temperature

- *vs* vs temperature
- Stronger disorder & $S = 0.5, \sigma = 0.025$eV



Disordered molecules + vibrations – *vs* temperature

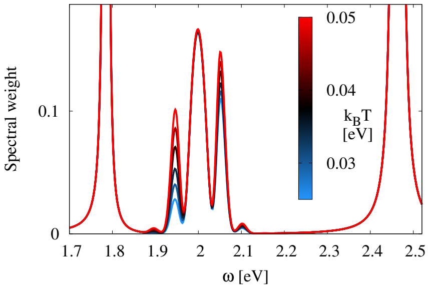
- *vs* vs temperature



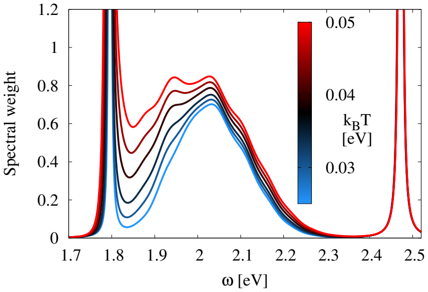
- Stronger disorder & $S = 0.5, \sigma = 0.025$eV

Disordered molecules + vibrations – *vs* temperature

- *vs* vs temperature
  $S = 0.02, \sigma = 0.01$eV



- Stronger disorder &
  $S = 0.5, \sigma = 0.025$eV



Acknowledgements

GROUP:

COLLABORATORS: Szymanska (UCL), Littlewood (ANL & Chicago), De Liberato (Southhampton)

FUNDING:

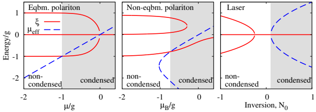
topnes
Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

EPSRC
Engineering and Physical Sciences Research Council

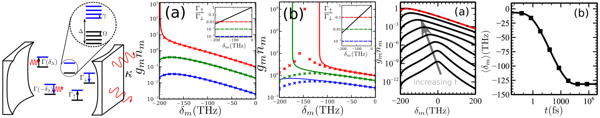
The Leverhulme Trust

Summary

- Lasing, condensation, superradiance

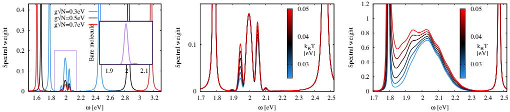


- Photon condensation and thermalisation



[Kirton & JK, PRL ’13, arXiv:1410.6632]

- Vibrational configuration



[Cwik, Kirton, De Liberato, JK in preparation]

Extra Slides

4. Microscopic calculation of $\Gamma(\delta)$
5. Threshold vs temperature
6. Threshold vs pump size
7. Strong coupling: polaritons
8. Anticrossing vs ρ
   - Polariton spectrum & sidebands
9. Ultra-strong phonon coupling?

Microscopic model – calculating $\Gamma(\delta)$

How to calculate $\Gamma(\delta)$

- Polaron transform (exact)

$$h_\alpha = \frac{\epsilon}{2}\sigma^z_\alpha + g\left(\psi_m \sigma^+_\alpha D_\alpha + \text{H.c.}\right) + \Omega b^\dagger_\alpha b_\alpha,$$

$$D_\alpha = \exp\left[2\sqrt{S}(b^\dagger_\alpha - b_\alpha)\right]$$

- Correlation function:

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D^\dagger_\alpha(t) D_\alpha(0)\rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2}\right] e^{-i\delta t}$$

- Exponential of bosonic correlations $\langle D^\dagger_\alpha(t) D_\alpha(0)\rangle$

Microscopic model – calculating $\Gamma(\delta)$

How to calculate $\Gamma(\delta)$

- Polaron transform (exact)

$$h_\alpha = \frac{\epsilon}{2}\sigma^z_\alpha + g\left(\psi_m \sigma^+_\alpha D_\alpha + \text{H.c.}\right) + \Omega b^\dagger_\alpha b_\alpha,$$
$$D_\alpha = \exp\left[2\sqrt{S}(b^\dagger_\alpha - b_\alpha)\right]$$

- Correlation function:

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D^\dagger_\alpha(t) D_\alpha(0)\rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2}\right] e^{-i\delta t}$$

- Exponential of bosonic correlations $\langle D^\dagger_\alpha(t) D_\alpha(0)\rangle$

Microscopic model – calculating $\Gamma(\delta)$

How to calculate $\Gamma(\delta)$

- Polaron transform (exact)

$$h_\alpha = \frac{\epsilon}{2}\sigma^z_\alpha + g\left(\psi_m \sigma^+_\alpha D_\alpha + \text{H.c.}\right) + \Omega b^\dagger_\alpha b_\alpha,$$

$$D_\alpha = \exp\left[2\sqrt{S}(b^\dagger_\alpha - b_\alpha)\right]$$

- Correlation function:

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D^\dagger_\alpha(t) D_\alpha(0)\rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2}\right] e^{-i\delta t}$$

- Exponential of bosonic correlations $\langle D^\dagger_\alpha(t) D_\alpha(0)\rangle$

Microscopic model – requirements for Kennard-Stepanov

- Correlation function

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D^\dagger_\alpha(t) D_\alpha(0)\rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2}\right] e^{-i\delta t}$$

- Kubo-Martin-Schwinger condition:

$$\langle D^\dagger_\alpha(t) D_\alpha(0)\rangle = \langle D^\dagger_\alpha(-t - i\beta) D_\alpha(0)\rangle$$

- $\Gamma(+\delta) = \Gamma(-\delta)e^{\beta\delta}$

Microscopic model – requirements for Kennard-Stepanov

- Correlation function

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D^\dagger_\alpha(t) D_\alpha(0) \rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2} \right] e^{-i\delta t}$$

- Kubo-Martin-Schwinger condition:

$$\langle D^\dagger_\alpha(t) D_\alpha(0) \rangle = \langle D^\dagger_\alpha(-t - i\beta) D_\alpha(0) \rangle$$

- $\Gamma(+\delta) = \Gamma(-\delta)e^{\beta\delta}$

Microscopic model – requirements for Kennard-Stepanov

- Correlation function

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D^\dagger_\alpha(t) D_\alpha(0) \rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2}\right] e^{-i\delta t}$$

- Kubo-Martin-Schwinger condition:

$$\langle D^\dagger_\alpha(t) D_\alpha(0) \rangle = \langle D^\dagger_\alpha(-t - i\beta) D_\alpha(0) \rangle$$

- $\Gamma(+\delta) = \Gamma(-\delta)e^{\beta\delta}$

Microscopic model – requirements for Kennard-Stepanov

- Correlation function

$$\Gamma(\delta) = 2g^2 \Re \int dt \langle D_\alpha^\dagger(t) D_\alpha(0) \rangle \exp\left[-(\Gamma_\uparrow + \Gamma_\downarrow)\frac{t}{2} \right] e^{-i\delta t}$$

- Kubo-Martin-Schwinger condition:

$$\langle D_\alpha^\dagger(t) D_\alpha(0) \rangle = \langle D_\alpha^\dagger(-t - i\beta) D_\alpha(0) \rangle$$

- $\Gamma(+\delta) = \Gamma(-\delta) e^{\beta\delta}$

Threshold condition

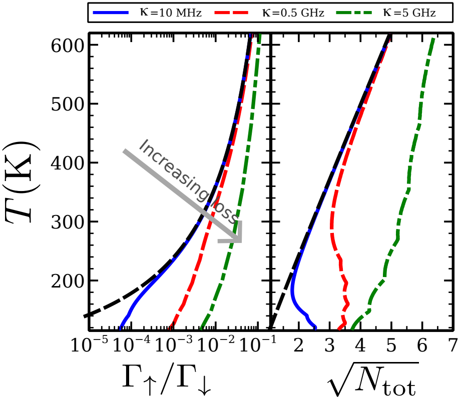
Use: $\max[n_m] = 1/(\beta\epsilon) \quad \rightarrow \quad k_B T_c = \sqrt{6/\pi^2}\epsilon\sqrt{N}$.

- 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

Threshold condition

Use: $\max[n_m] = 1/(\beta\epsilon) \quad \rightarrow \quad k_B T_c = \sqrt{6/\pi^2}\epsilon\sqrt{N}$.



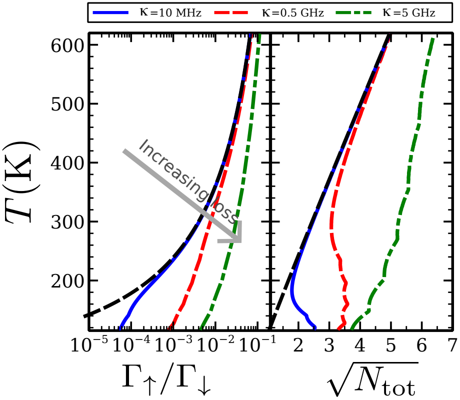
Compare threshold:

- 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

Threshold condition

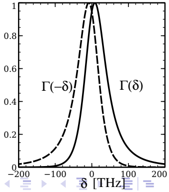
Use: $\max[n_m] = 1/(\beta\epsilon) \quad \rightarrow \quad k_B T_c = \sqrt{6/\pi^2}\epsilon\sqrt{N}$.



Compare threshold:

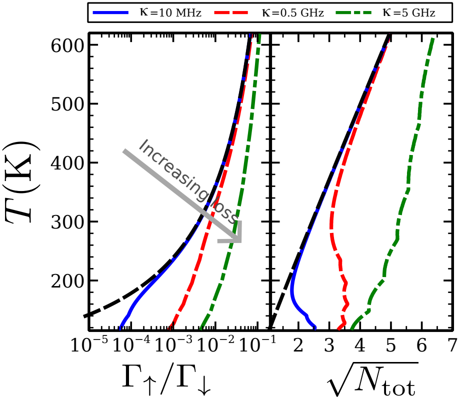
- Pump rate (Laser)
- Critical density (condensate)

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



Threshold condition

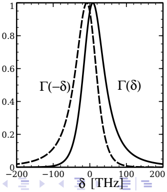
Use: $\max[n_m] = 1/(\beta\epsilon) \quad \rightarrow \quad k_B T_c = \sqrt{6/\pi^2 \epsilon \sqrt{N}}.$



Compare threshold:

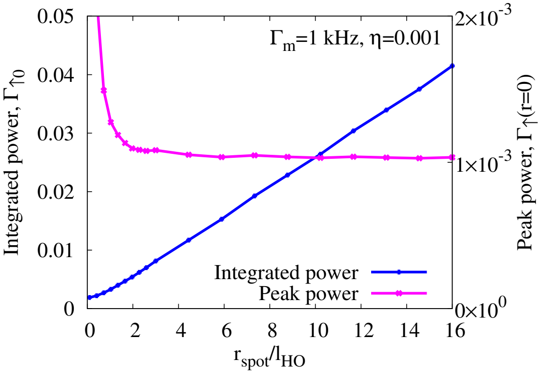
- Pump rate (Laser)
- Critical density (condensate)

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



Effect of spot size on threshold

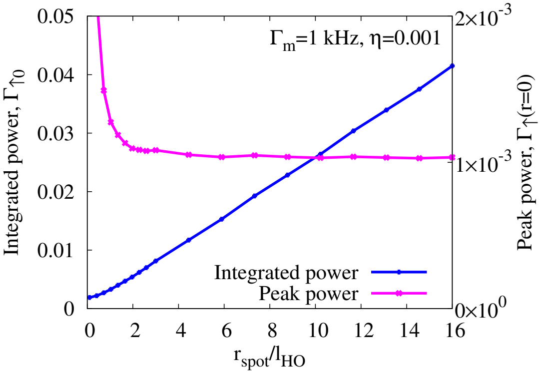
Threshold power: $\Gamma_{\uparrow}(r) = \Gamma_{\uparrow,0} \dfrac{\exp(-(r/\sigma_p)^2/2)}{(2\pi\sigma_p^2)^{d/2}}$



- Small spot, integrated power saturates
- Large spot, peak power saturates

Effect of spot size on threshold

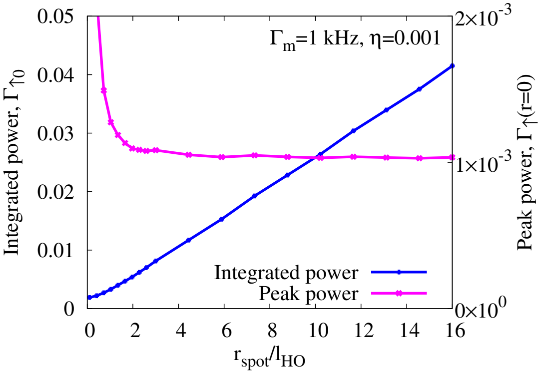
Threshold power: $\Gamma_{\uparrow}(r) = \Gamma_{\uparrow,0} \dfrac{\exp(-(r/\sigma_p)^2/2)}{(2\pi\sigma_p^2)^{d/2}}$



- Small spot, integrated power saturates
- Large spot, peak power saturates

Effect of spot size on threshold

Threshold power: $\Gamma_{\uparrow}(r) = \Gamma_{\uparrow,0} \frac{\exp(-(r/\sigma_p)^2/2)}{(2\pi\sigma_p^2)^{d/2}}$



- Small spot, integrated power saturates
- Large spot, peak power saturates

Strong coupling: polaritons

Strong coupling phase diagram — mean field

- Mean field — single photon mode

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

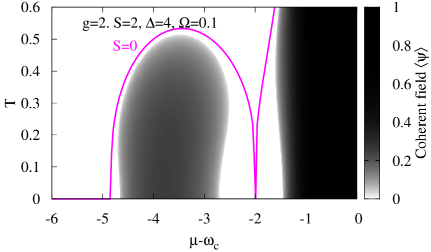
- $\epsilon = \omega - \Delta$,
  Mott lobes if $\epsilon < \omega - 2g$
- S reduces g_{eff}

- Reentrant behaviour — Min μ at $k_B T \sim 0.1\Omega$

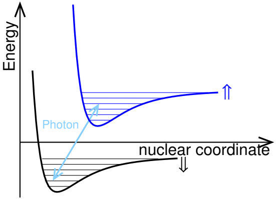
Strong coupling phase diagram — mean field

- Mean field — single photon mode

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



- $\epsilon = \omega - \Delta$,
  Mott lobes if $\epsilon < \omega - 2g$
- S reduces g_{eff}

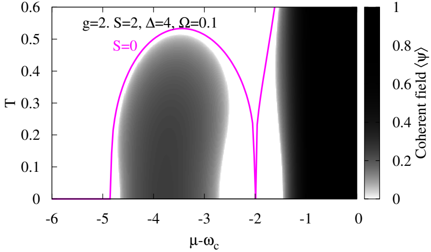


- Reentrant behaviour — Min μ at $k_B T \sim 0.1\Omega$

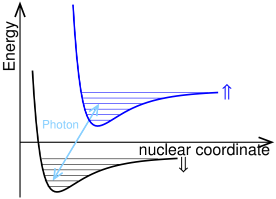
Strong coupling phase diagram — mean field

- Mean field — single photon mode

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

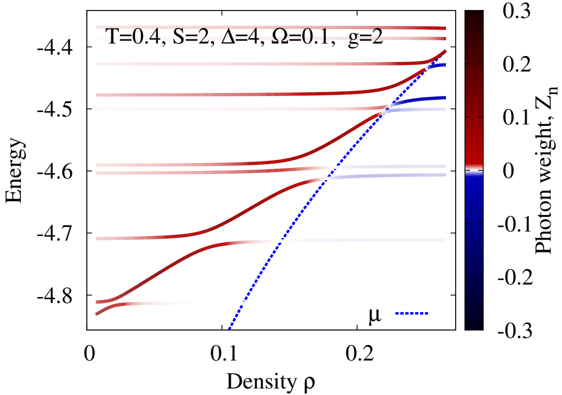


- $\epsilon = \omega - \Delta$, Mott lobes if $\epsilon < \omega - 2g$
- S reduces g_{eff}



- Reentrant behaviour — Min μ at $k_B T \sim 0.1\Omega$

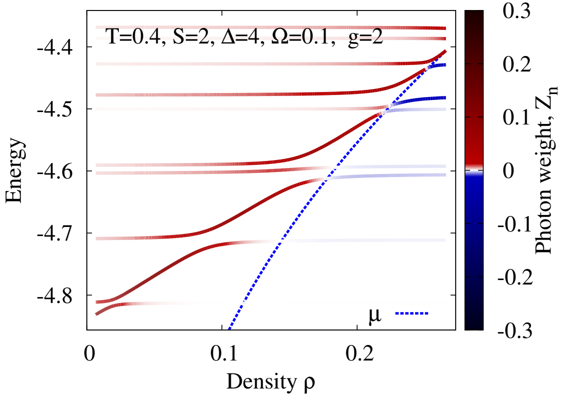
Polariton spectrum: photon weight



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

[Cwik *et al.* EPL '14]

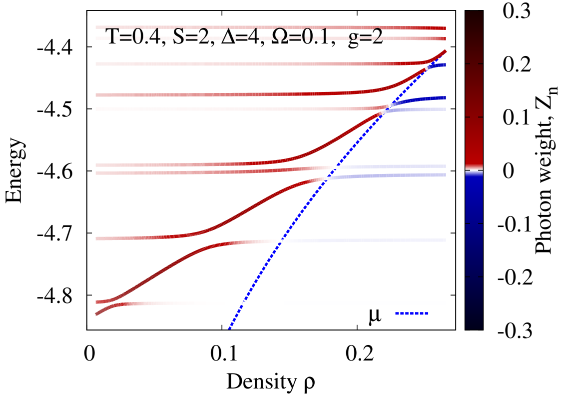
Polariton spectrum: photon weight



- Saturating 2LS: $g_{\text{eff}}^2 \sim g^2(1 - 2\rho)$
- What is nature of polariton mode?

[Cwik *et al.* EPL '14]

Polariton spectrum: photon weight

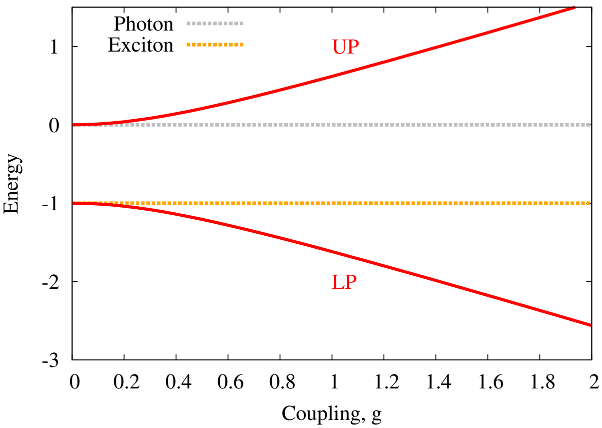


- Saturating 2LS: $g_{\mathrm{eff}}^2 \sim g^2(1-2\rho)$
- What is nature of polariton mode?
- $G^R(t) = -i\langle\psi^\dagger(t)\psi(0)\rangle, \qquad G^R(\nu) = \sum_n \frac{Z_n}{\nu - \omega_n}$

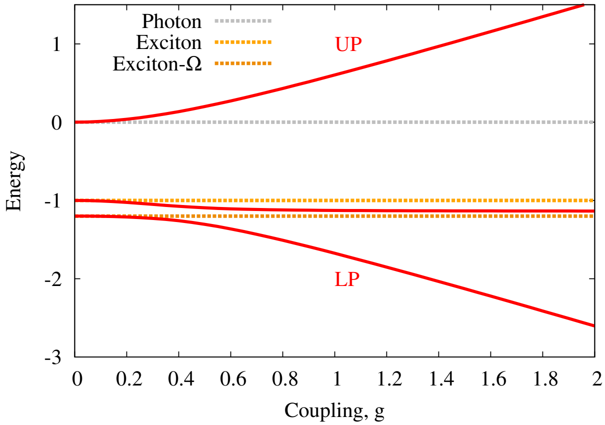
[Cwik *et al.* EPL '14]

Polariton spectrum — coupled oscillators

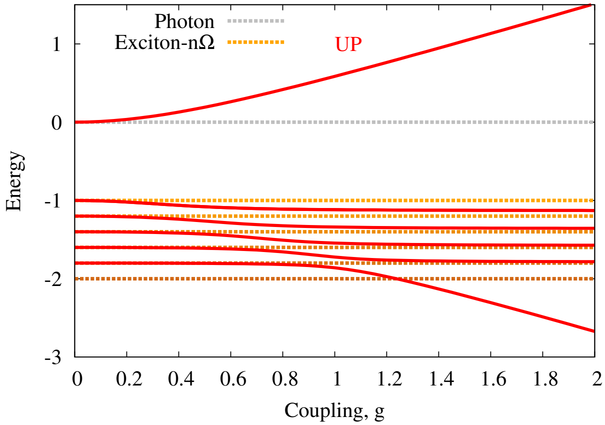
Polariton spectrum — coupled oscillators



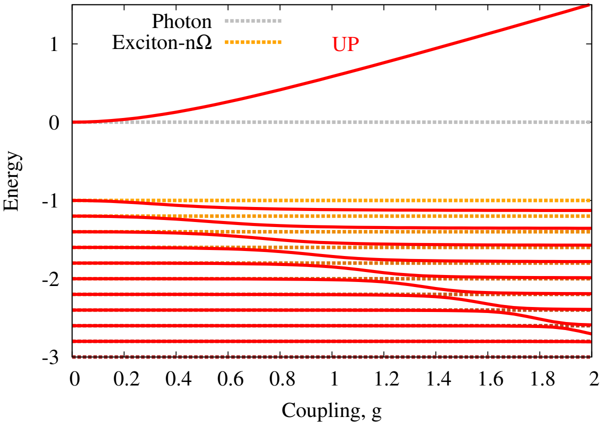
Polariton spectrum — coupled oscillators



Polariton spectrum — coupled oscillators



Polariton spectrum — coupled oscillators



Polariton spectrum: what condensed

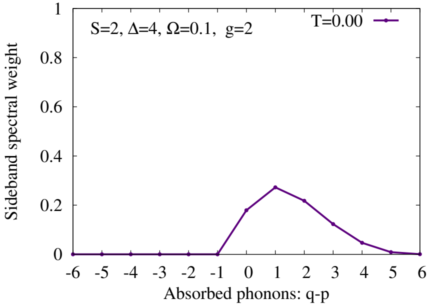
- Repeat weight for *n*-phonon channel
- Eigenvector that is macroscopically occupied

  - Optimal $T \sim 2\Omega$

[Cwik *et al.* EPL '14]

Polariton spectrum: what condensed

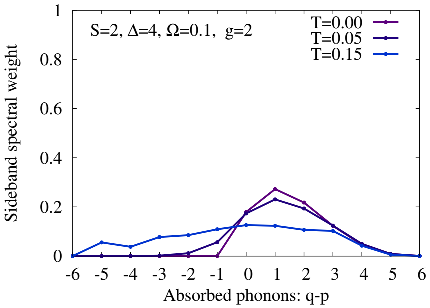
- Repeat weight for *n*-phonon channel
- Eigenvector that is macroscopically occupied



[Cwik *et al.* EPL '14]

Polariton spectrum: what condensed

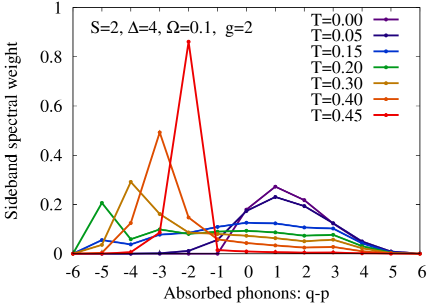
- Repeat weight for *n*-phonon channel
- Eigenvector that is macroscopically occupied
- Optimal $T \sim 2\Omega$



[Cwik *et al.* EPL '14]

Polariton spectrum: what condensed

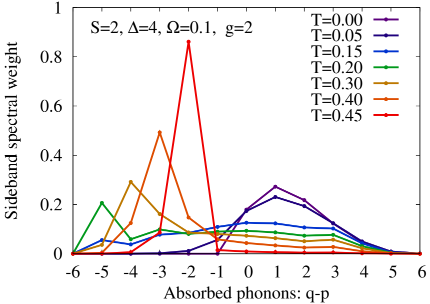
- Repeat weight for *n*-phonon channel
- Eigenvector that is macroscopically occupied



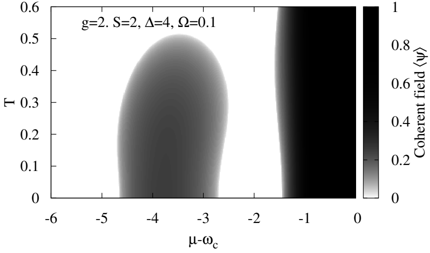
[Cwik *et al.* EPL '14]

Polariton spectrum: what condensed

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



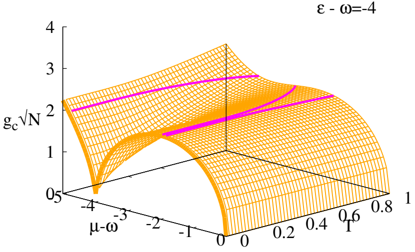
- Optimal $T \sim 2\Omega$



[Cwik *et al.* EPL '14]

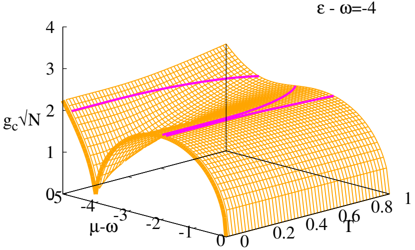
Critical coupling with increasing S

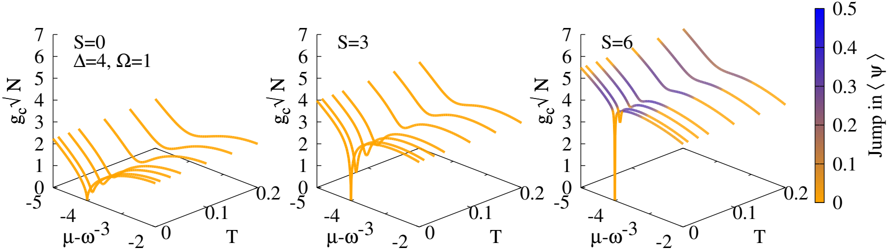
- Re-orient phase diagram
- *g* vs μ, *T*
- Colors → Jump of $\langle\psi\rangle$



Critical coupling with increasing S

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





Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

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

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

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

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

- Optimal phonon displacements, $\sim \sqrt{S}$
- Reduced $g_{\text{eff}} \sim g \times \exp(-S/2)$
- For $\psi \neq 0$, competition

  Variational MFT $|\psi\rangle_\alpha \sim \exp(-\eta K_\alpha - \zeta b^\dagger_\alpha)|0, S\rangle_\alpha$

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

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

Explanation: Polaron formation

- Unitary transform

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

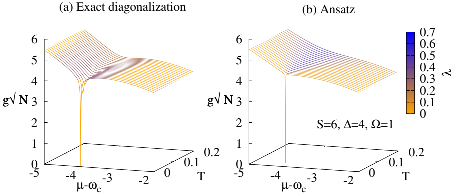
- Coupling moves to $S^\pm$

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

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

Collective polaron formation

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



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

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

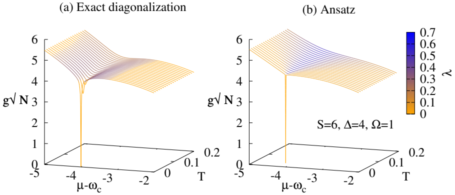
Effective 2LS energy in field:

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

[Cwik *et al.* EPL '14]

Collective polaron formation

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



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

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

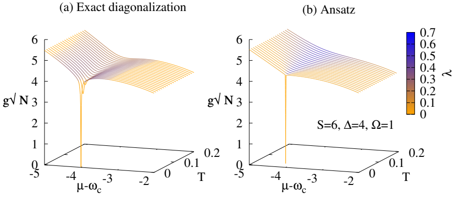
Effective 2LS energy in field:

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

[Cwik *et al.* EPL '14]

Collective polaron formation

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



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

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

Effective 2LS energy in field:

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

[Cwik *et al.* EPL '14]