Photon and polariton condensates with organic molecules

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University of St Andrews
1413-2013

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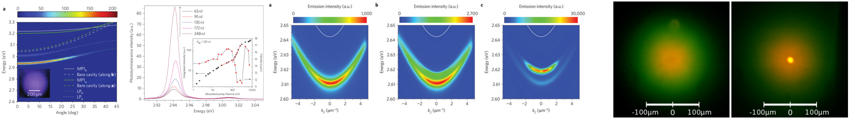
Matter-Light coupling with organic molecules

- 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

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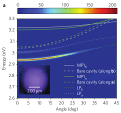
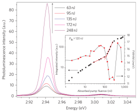
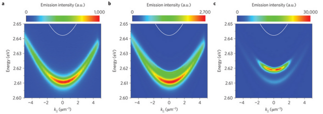

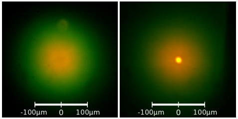

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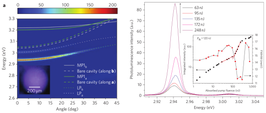

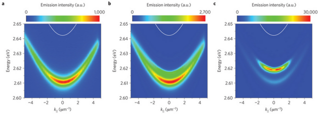

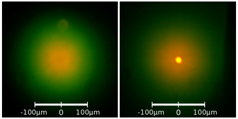

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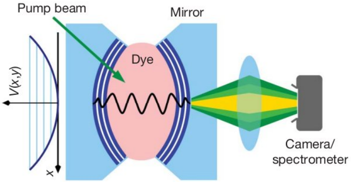

- Dye filled microcavity
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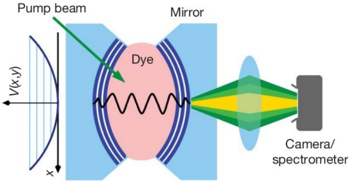

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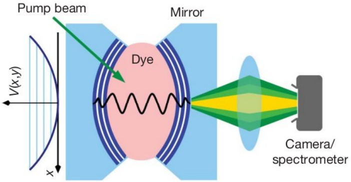

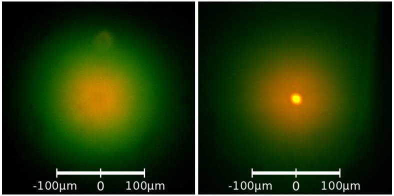

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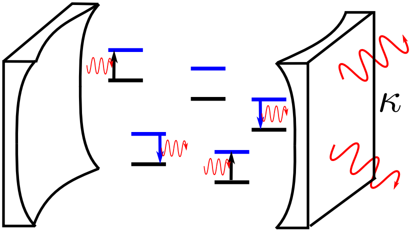
Dicke Holstein Model

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



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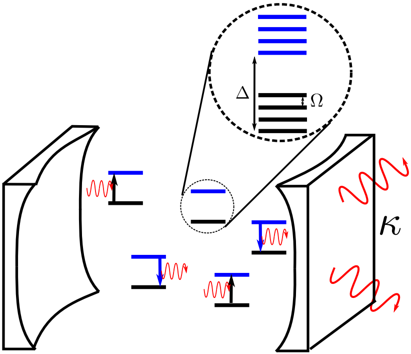

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$$+ \sum_\alpha \Omega\left\{b^\dagger_\alpha b_\alpha + \sqrt{S}\sigma^z_\alpha\left(b^\dagger_\alpha + b_\alpha\right)\right\}$$

Overview

1. Introduction: organic molecules
2. Modelling photon BEC
   - Threshold behaviour
   - Time evolution
   - Pump-spot size dependence
3. Strong coupling: polaritons
   - Polariton spectrum nature

Modelling

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

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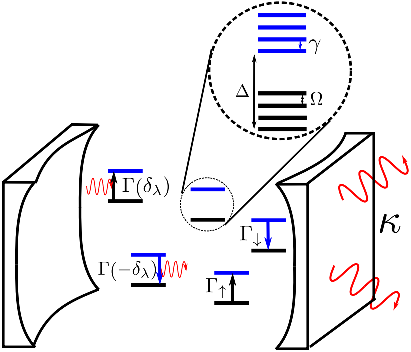
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in g



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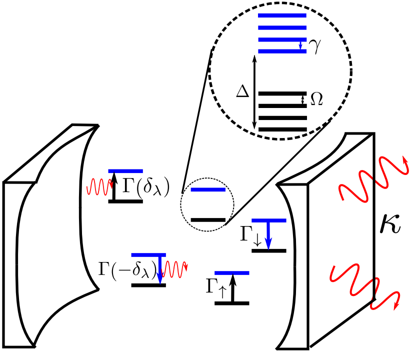

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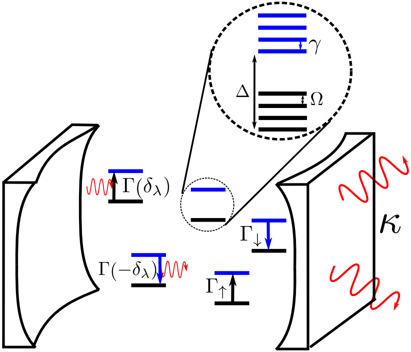
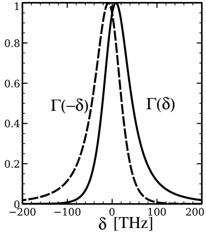


Modelling

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 $\Gamma(+\delta) \simeq \Gamma(-\delta)e^{\beta\delta}$
- Expt: $\omega_0 < \epsilon$
- $\Gamma \to 0$ at large δ

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

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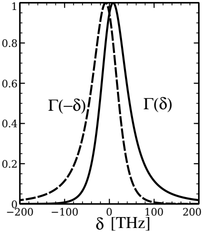

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Microscopic model – calculating $\Gamma(\delta)$

How to calculate $\Gamma(\delta)$

- Polaron transform (exact)

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

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

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

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

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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

[Kirton & JK PRL '13]

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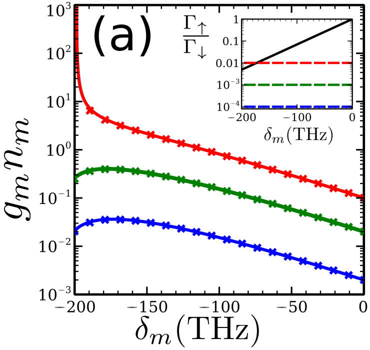

Low loss: Thermal

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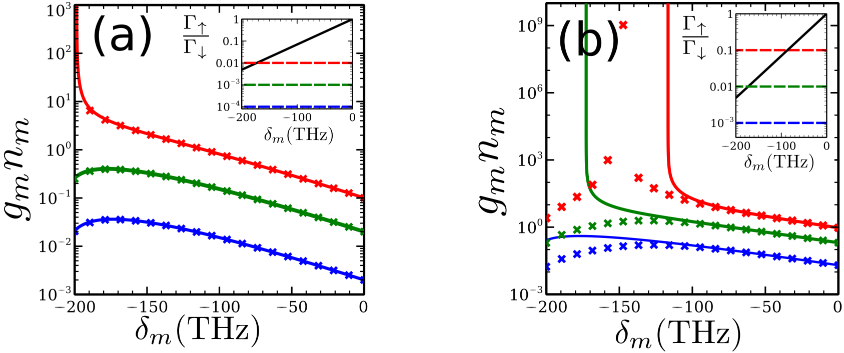


Low loss: Thermal

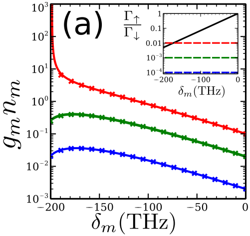
High loss → Laser

[Kirton & JK PRL '13]

Chemical potential?

- Steady state distribution:

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



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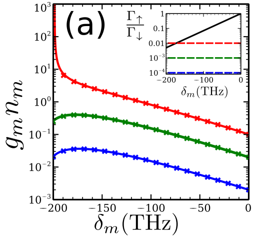

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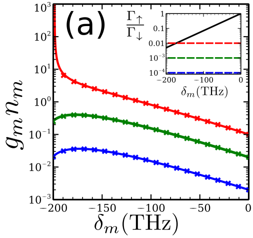

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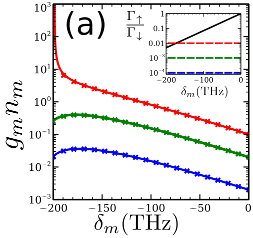

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Use: $\max[n_m] = 1/(\beta\epsilon) \quad \rightarrow \quad k_B T_c = \sqrt{6/\pi^2}\epsilon\sqrt{N}$.

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

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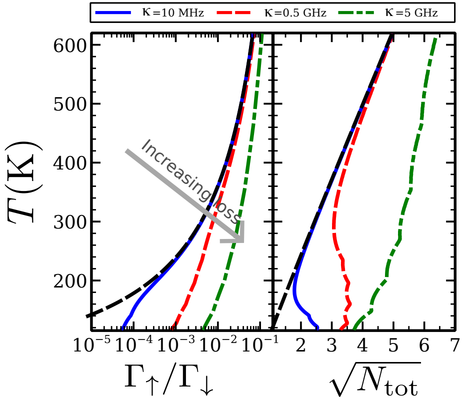

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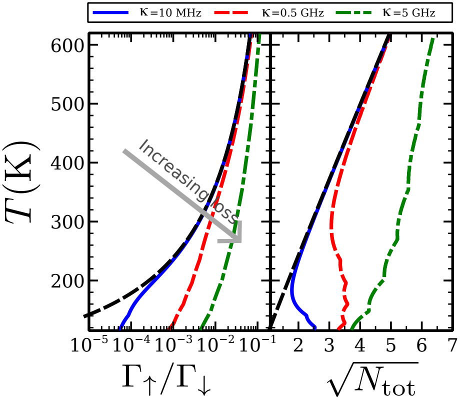

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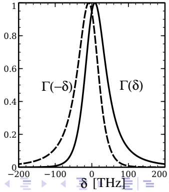

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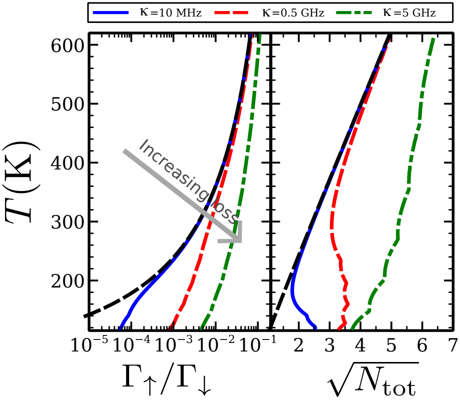

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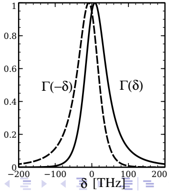
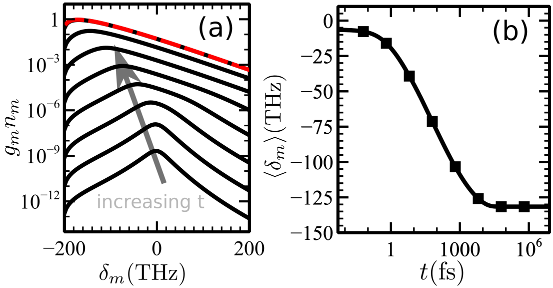


Time evolution

Time evolution

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



[Kirton & JK arXiv:1410.6632]

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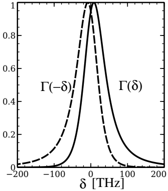

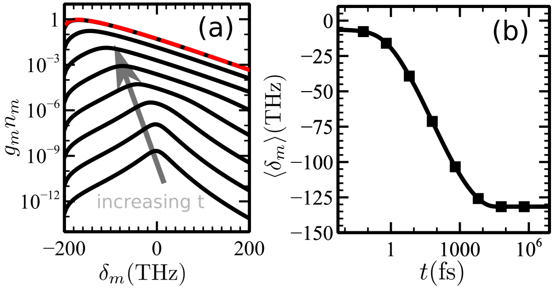

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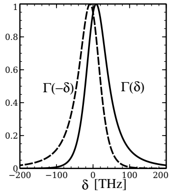

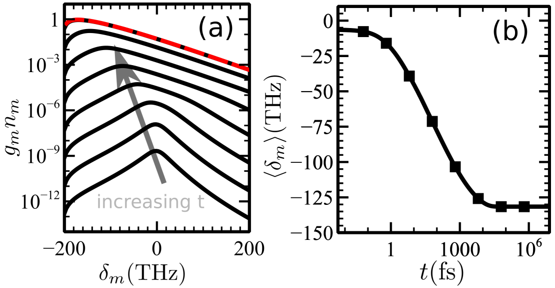

[Kirton & JK arXiv:1410.6632]

Pump spot size

Spatially varying pump intensity

- Consider effects of pump profile, $\Gamma_\uparrow(\mathbf{r}) = \Gamma_\uparrow \exp(-r^2/2r_{\text{spot}}^2)$

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

- Varying excited density – differential coupling to modes

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

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

- Experiments: [Marelic & Nyman, arXiv:1410.6822]

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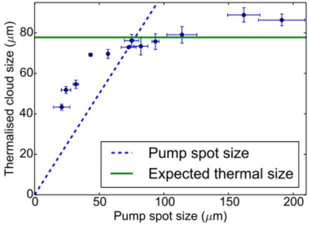

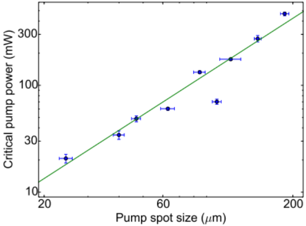

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Spatially varying pump: below threshold

- Far below threshold:
  - Excitation: $\rho_{\uparrow}(\mathbf{r}) \simeq \rho_m \frac{\Gamma_{\uparrow}(\mathbf{r})}{\Gamma_{\downarrow}} \ll \rho_m$
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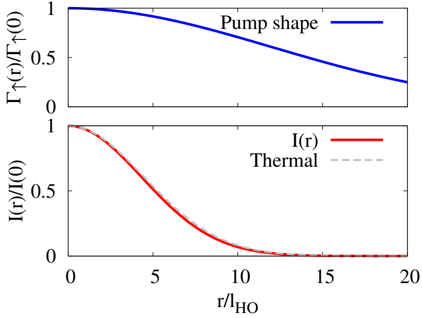

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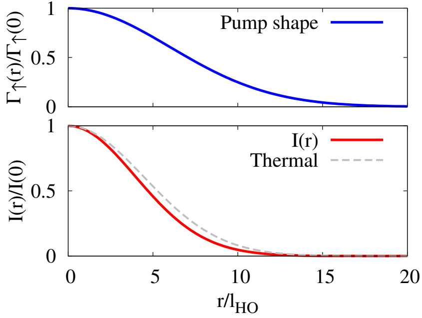

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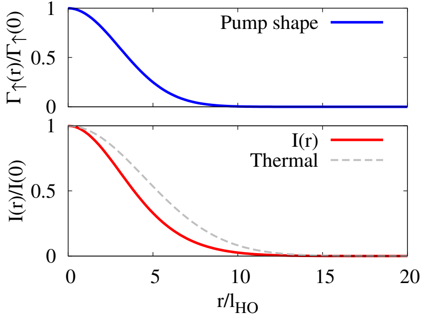

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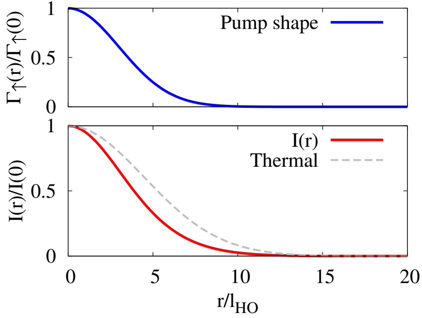

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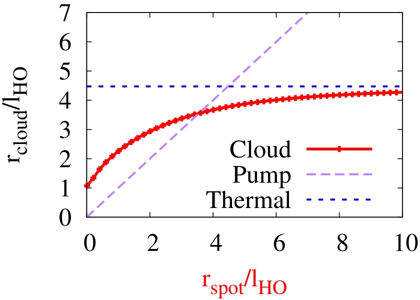

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- Large spot, $r_{spot}/l_{HO} = 20$
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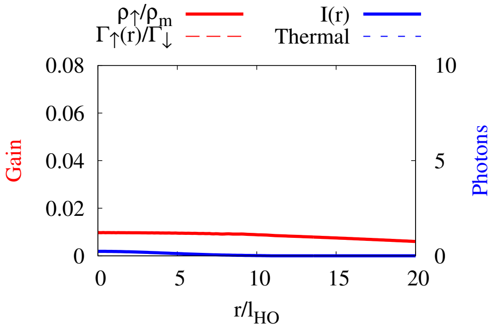

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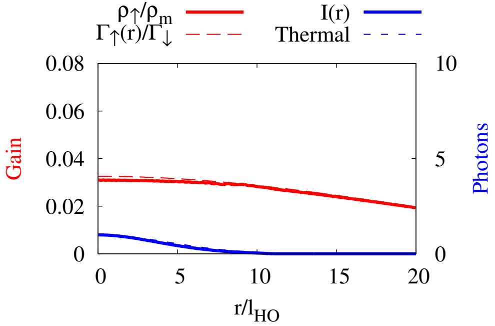

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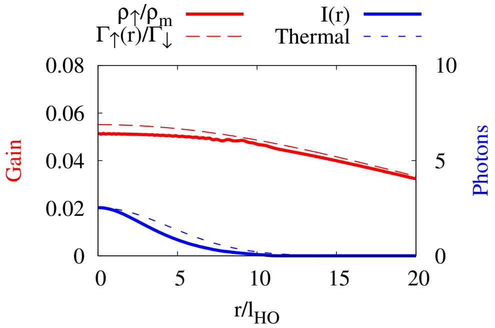

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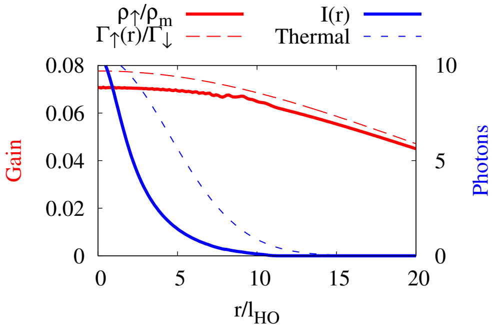

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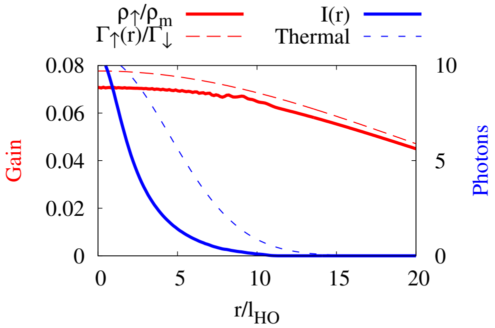

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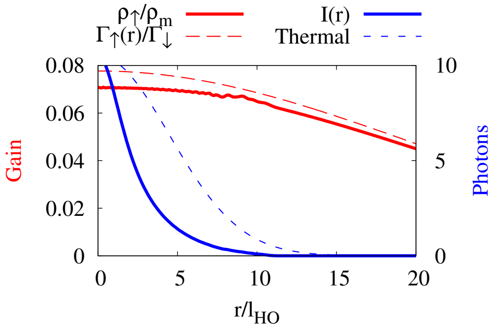

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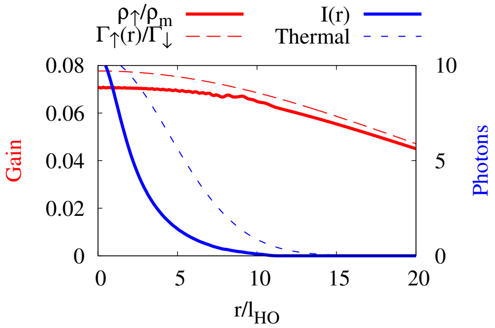

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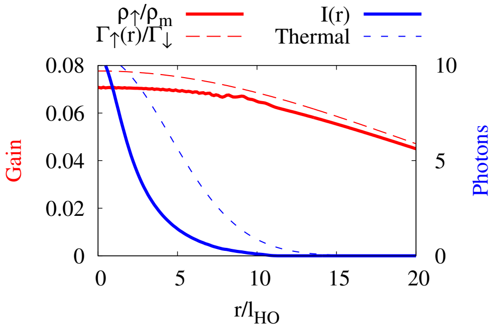

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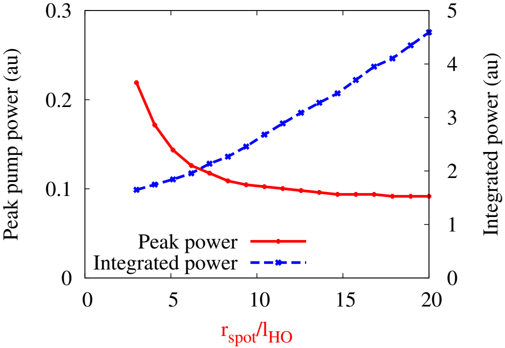

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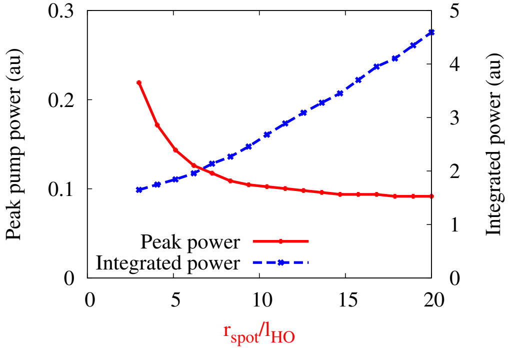

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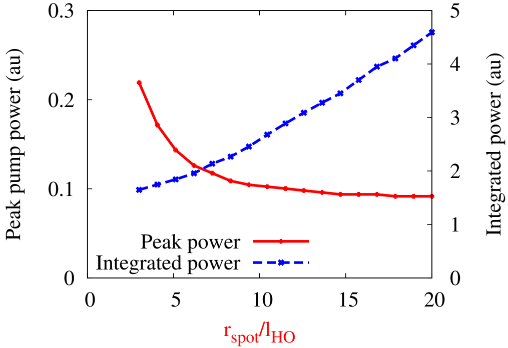

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Strong coupling: polaritons

Strong coupling phase diagram — mean field

- Mean field — single photon mode

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

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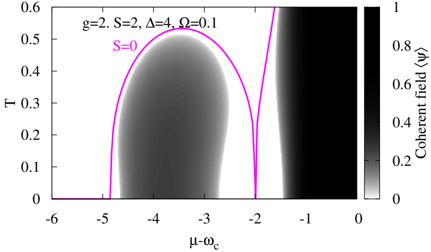

- $\epsilon = \omega - \Delta$,
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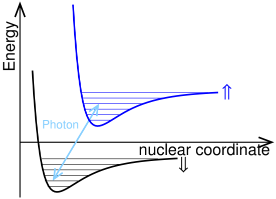

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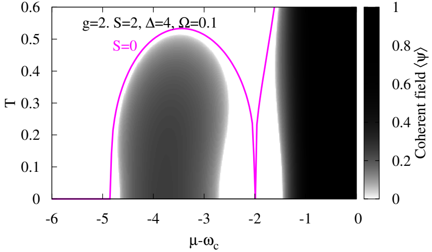

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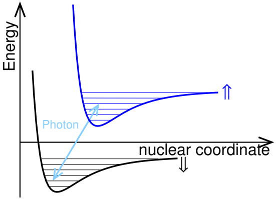

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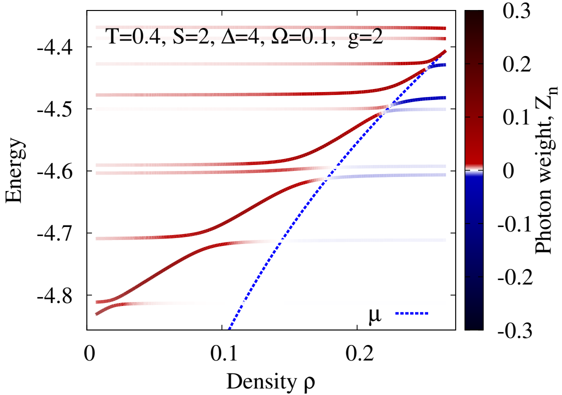
Polariton spectrum: photon weight



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[Cwik *et al.* EPL '14]

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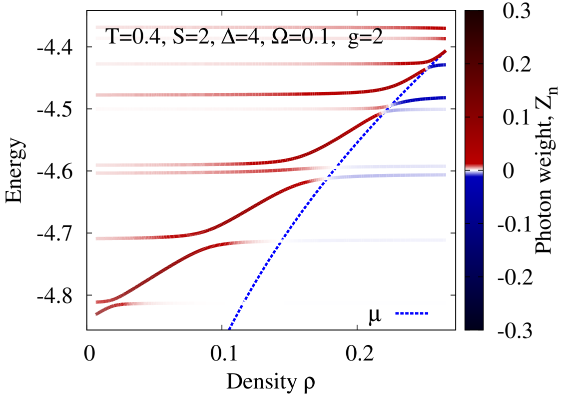

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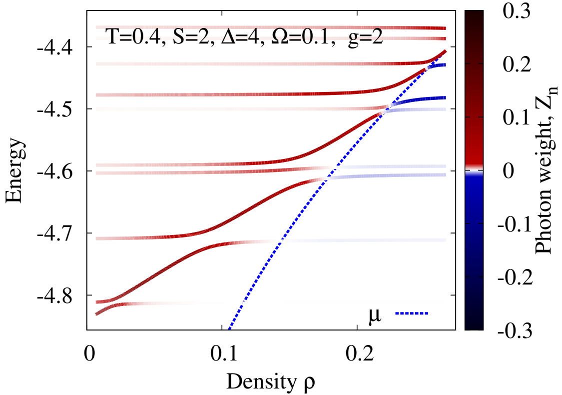

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- $G^R(t) = -i\langle\psi^\dagger(t)\psi(0)\rangle, \qquad G^R(\nu) = \sum_n \frac{Z_n}{\nu - \omega_n}$

[Cwik *et al.* EPL '14]

Polariton spectrum: what condensed

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

- Optimal $T \sim 2\Omega$

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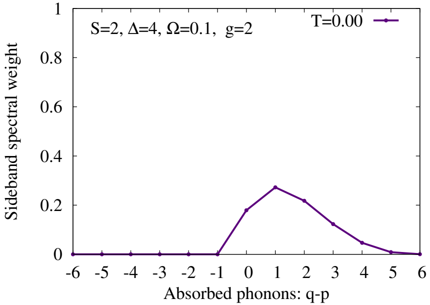

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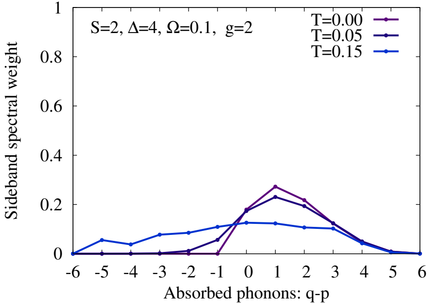

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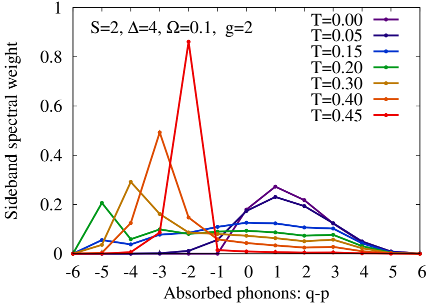

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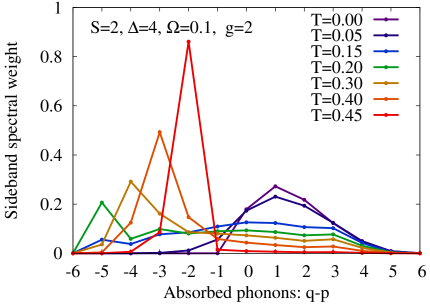

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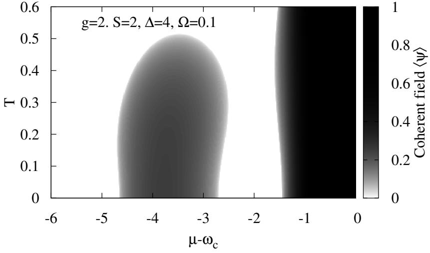

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Acknowledgements

GROUP:

COLLABORATORS: Reja (MPI-PKS), Littlewood (ANL & Chicago)

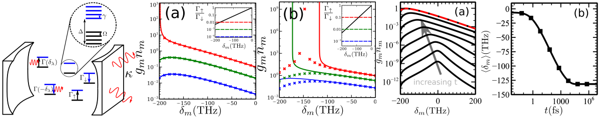
FUNDING:

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

EPSRC — Engineering and Physical Sciences Research Council

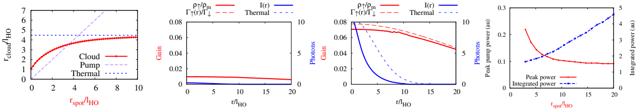
Summary

- Photon condensation and thermalisation

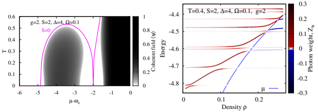


[Kirton & Keeling, PRL '13, arXiv:1410.6632]

- Effects of finite spot size



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



[Cwik *et al.* EPL '14]

Extra Slides

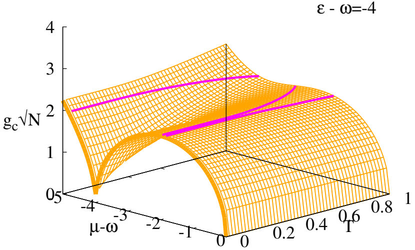
4. Ultra-strong phonon coupling?

5. Anticrossing vs ρ

6. Vibrational reconfiguration

Critical coupling with increasing S

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



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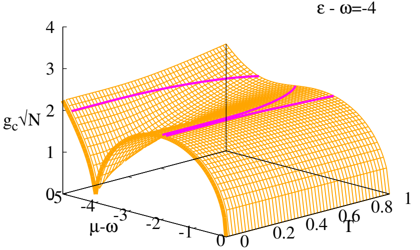

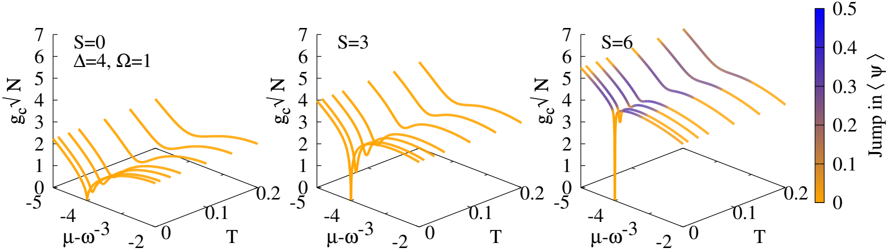

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

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

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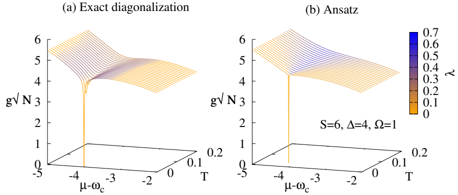
Collective polaron formation

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



[Cwik *et al.* EPL '14]

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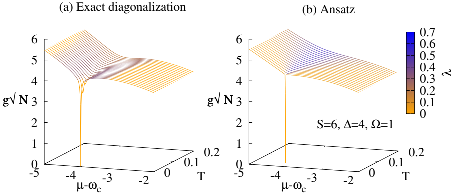

- Feedback: Large/small $g_{\text{eff}} \leftrightarrow \lambda = \langle \psi \rangle$
- Variational free energy

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

Effective 2LS energy in field:

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

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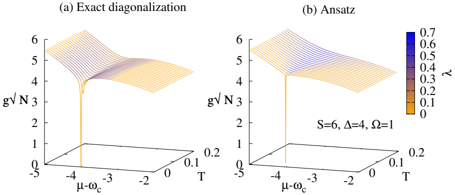

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Polariton spectrum — coupled oscillators

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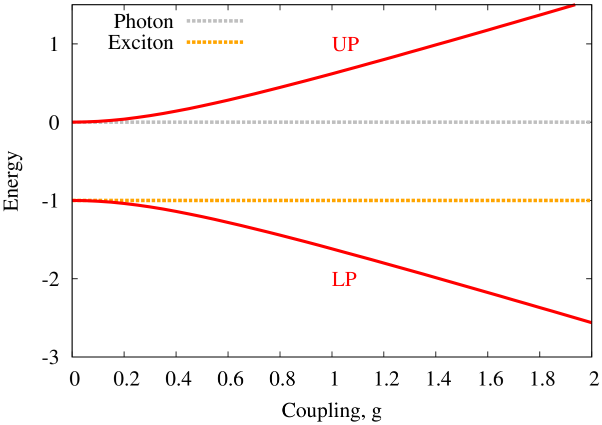

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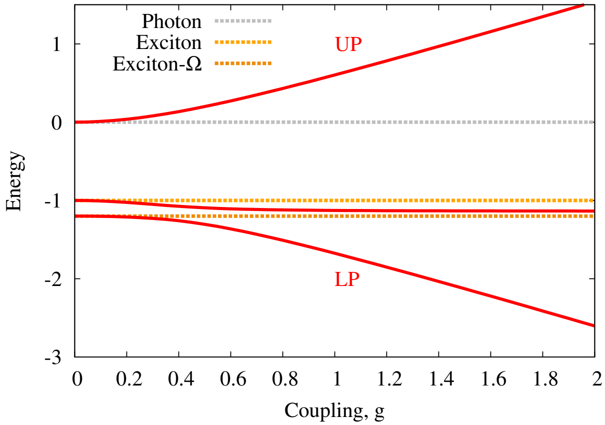

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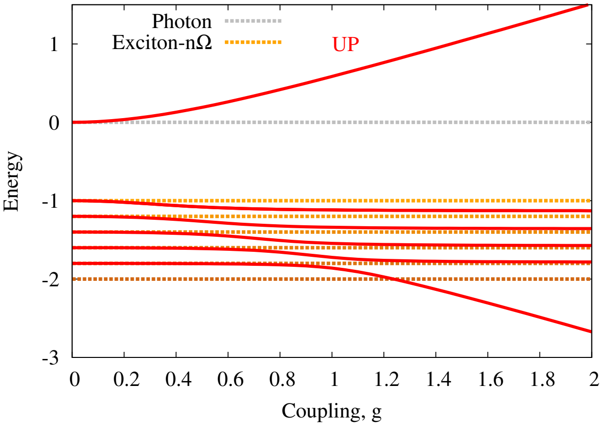

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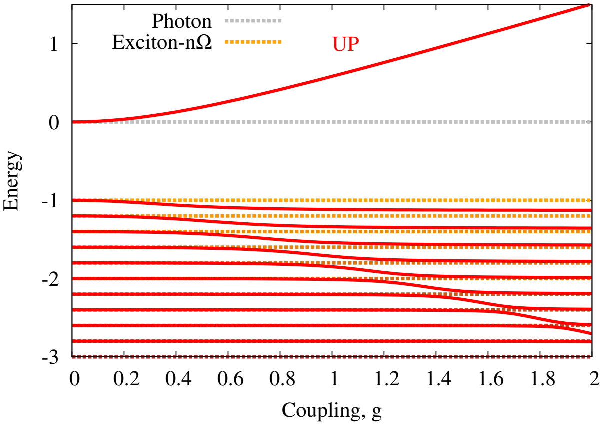

Vibrational reconfiguration

- $H = H_0 + H_1, H_1 = \sum_{n,k} g_{n,k}(\psi_k^\dagger \sigma_n^+ + \text{H.c.})$
- Schrieffer-Wolff: admixture of excited state

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

- Reduced vibrational offset $\sqrt{S} \rightarrow \sqrt{S}(1 - g^2 N/(\epsilon+\omega))$
- Increased effective coupling:

$$g_{\text{eff}}^2 = g^2 \exp(-S)$$

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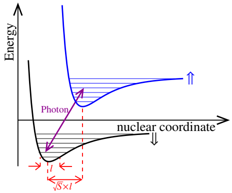

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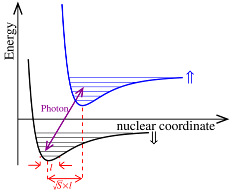

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