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

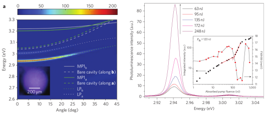
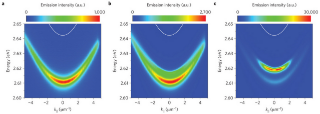
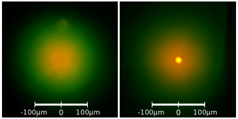
Jonathan Keeling

University of St Andrews
1413-2013

Snowbird, January 2015

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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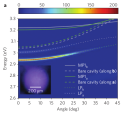

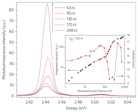

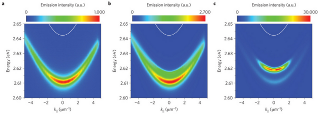

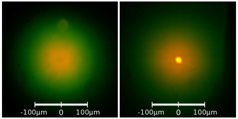

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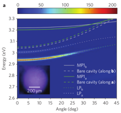
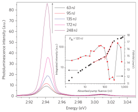
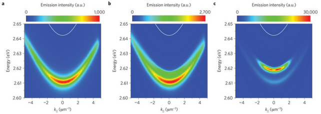
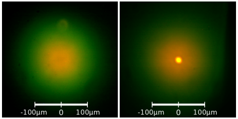

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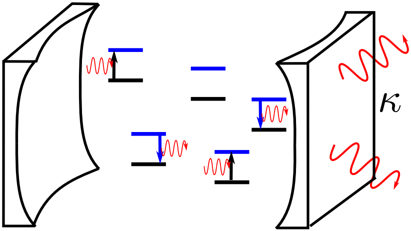
Dicke Holstein Model

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



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

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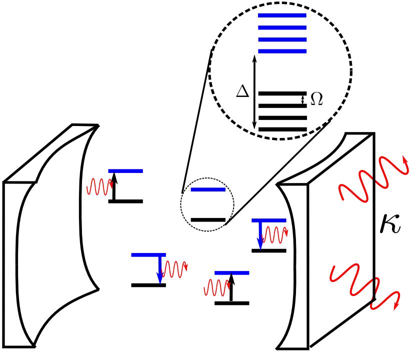

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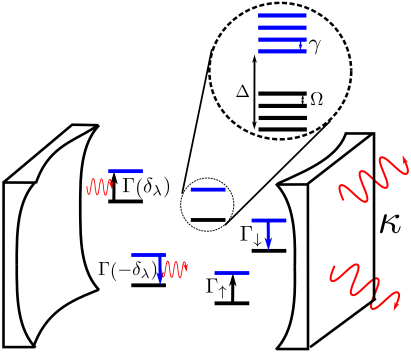
Three stories

1. Weak coupling: photon condensation
2. Strong coupling: polaritons
3. Ultra strong coupling: vibrational reconfiguration

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



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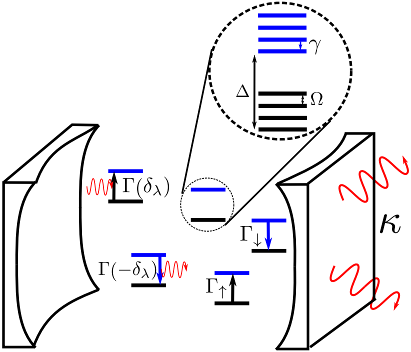

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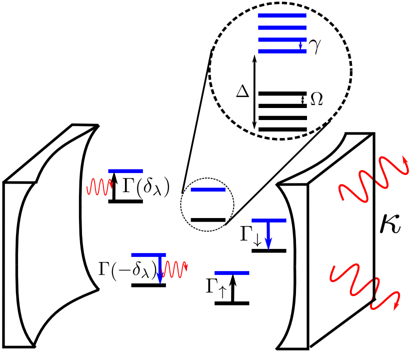
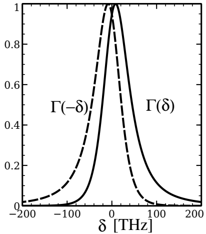


Modelling

Master equation

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

$$- \sum_{m,\alpha}\left[\frac{\Gamma(\delta_m = \omega_m - \epsilon)}{2}\mathcal{L}[\sigma^+_\alpha \psi_m] + \frac{\Gamma(-\delta_m = \epsilon - \omega_m)}{2}\mathcal{L}[\sigma^-_\alpha \psi^\dagger_m]\right]$$



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

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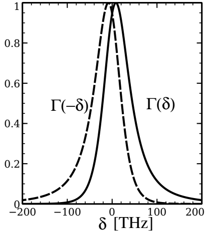


- Kennard-Stepanov $\Gamma(+\delta) \simeq \Gamma(-\delta)e^{\beta\delta}$
- Expt: $\omega_0 < \epsilon$
- $\Gamma \rightarrow 0$ at large δ

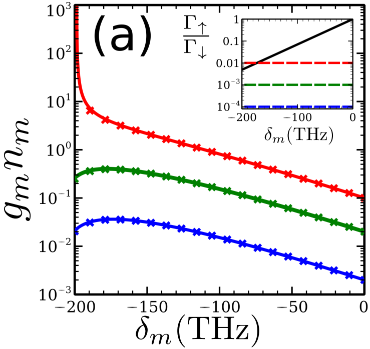
[Marthaler et al PRL ’11, Kirton & JK PRL ’13]

Distribution $g_m n_m$

- Master equation $\rightarrow$ Rate equation

$$\partial_t n_m = -\kappa n_m + N\left[\Gamma(-\delta_m)(n_m+1)\langle\sigma^{ee}\rangle - \Gamma(\delta_m) n_M \langle\sigma^{gg}\rangle\right]$$

- Bose-Einstein distribution without losses



Low loss: Thermal

[Kirton & JK PRL '13]

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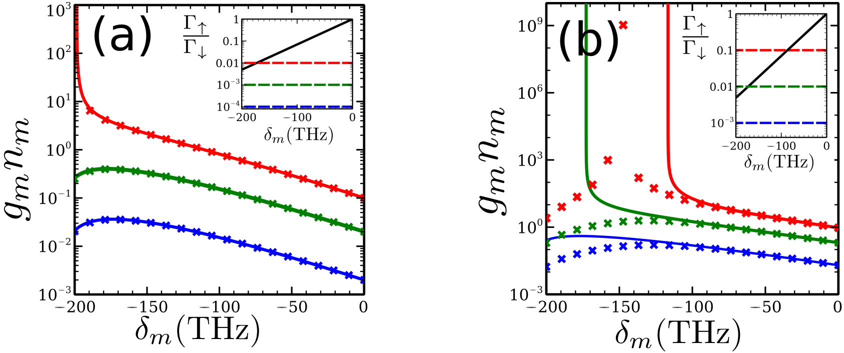


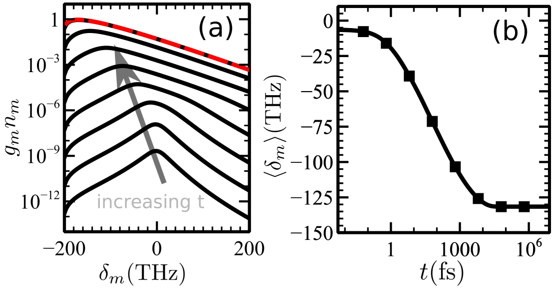
Low loss: Thermal

High loss $\rightarrow$ Laser

[Kirton & JK PRL '13]

Time evolution

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



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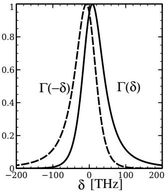

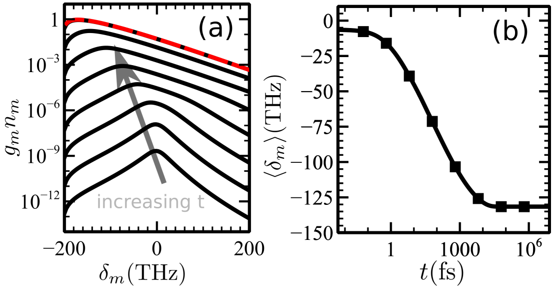

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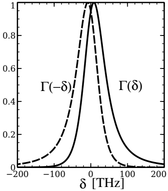

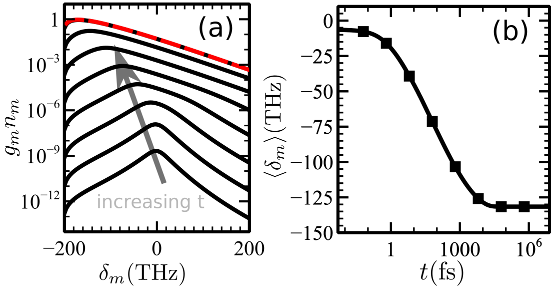

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Strong coupling: polaritons

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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 $k_B T \simeq 0.1\Omega$

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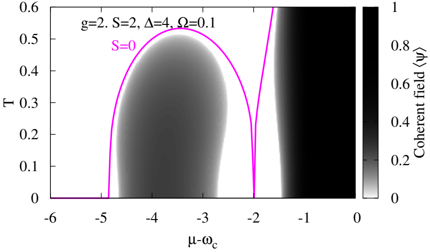

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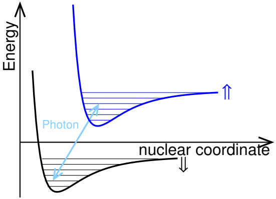

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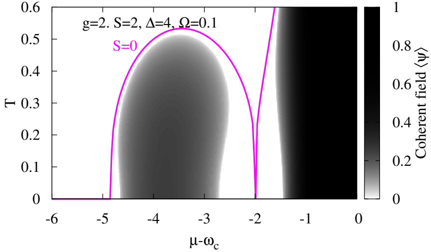

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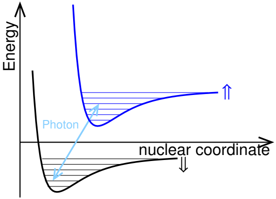

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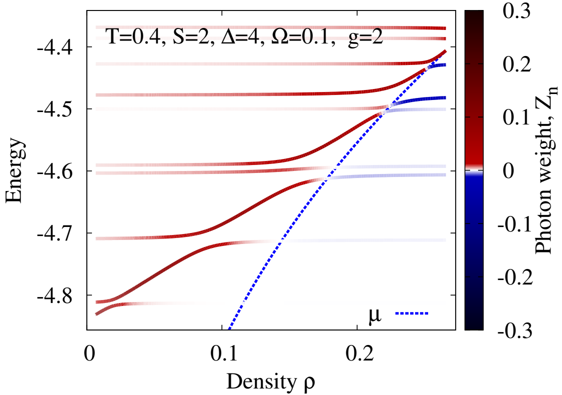
Polariton spectrum: photon weight



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

[Cwik *et al.* EPL '14]

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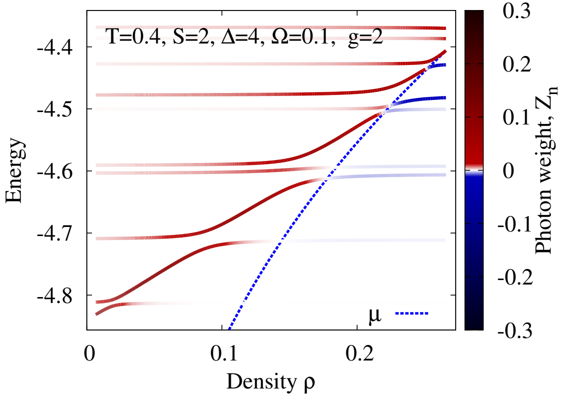

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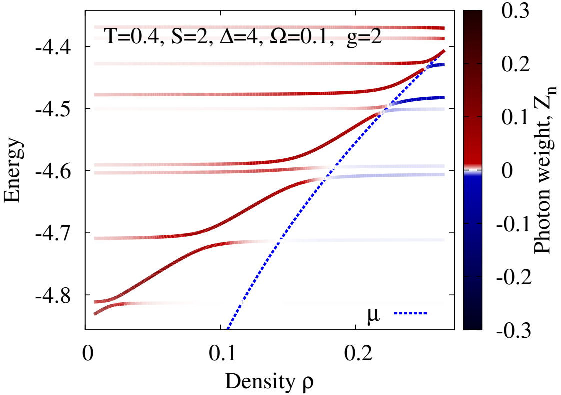

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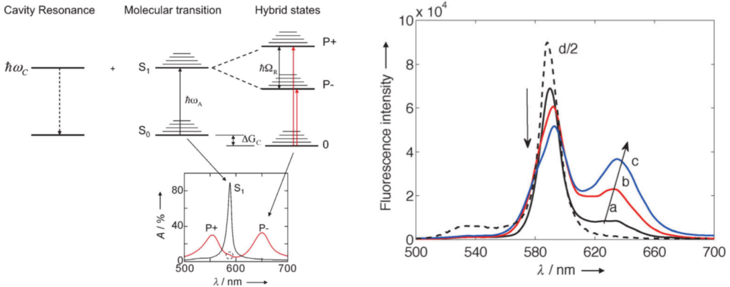
Ultra strong coupling: vibrational reconfiguration

1. Weak coupling: photon condensation
2. Strong coupling: polaritons
3. Ultra strong coupling: vibrational reconfiguration

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:

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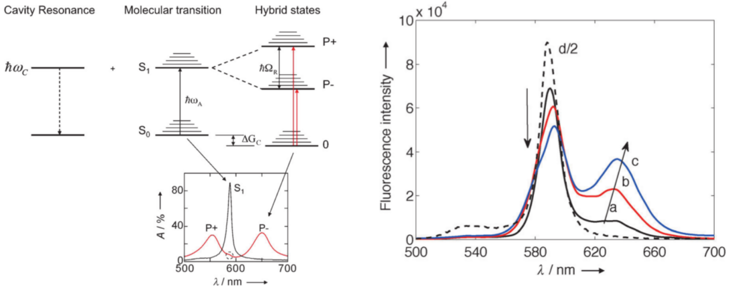

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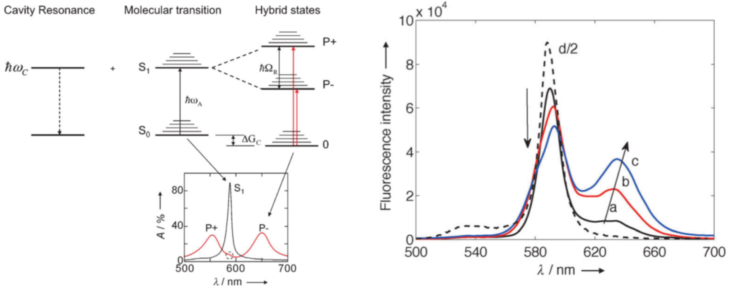

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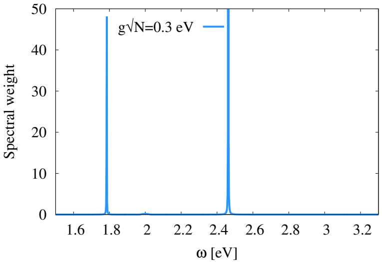
- Questions:
  - Vibrationally dressed spectrum + disorder
  - Microscopic theory – changing configuration

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



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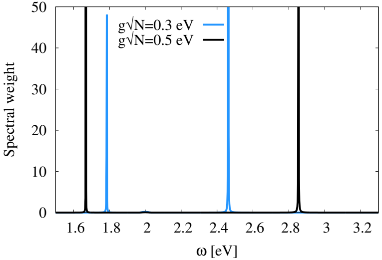

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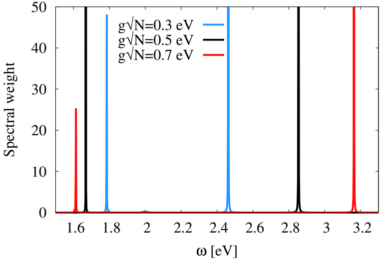

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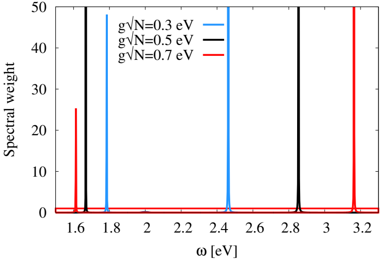

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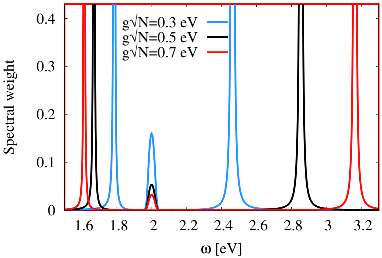

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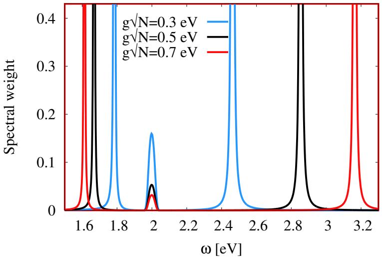

- Central peak:

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

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

[Houdré *et al.* , PRA '96]

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

Disordered molecules — spectrum

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

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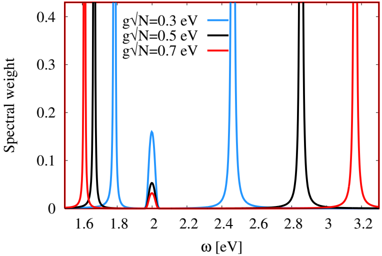

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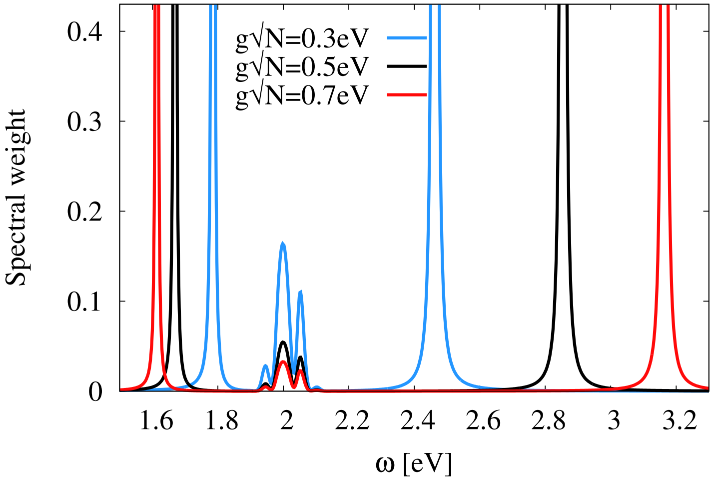
Disordered molecules — vibrational mode

- With vibrational sidebands, $S = 0.02$



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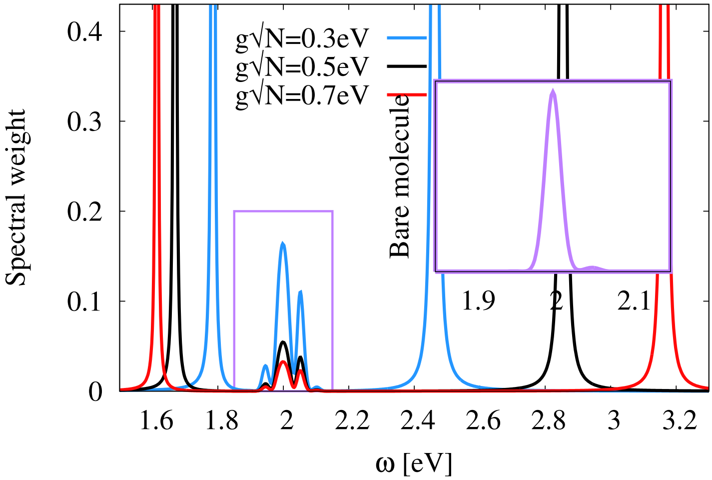
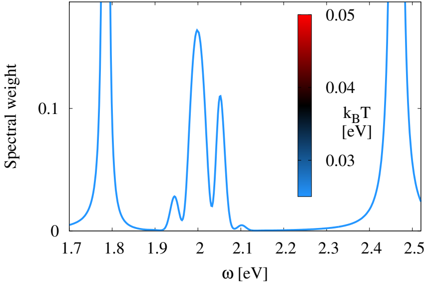


Disordered molecules + vibrations – *vs* temperature

- *vs* vs temperature



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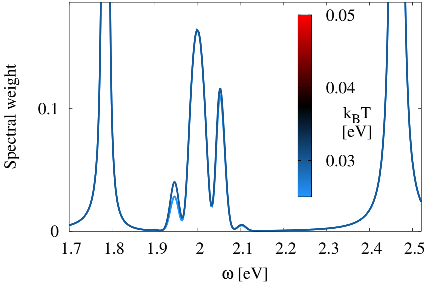

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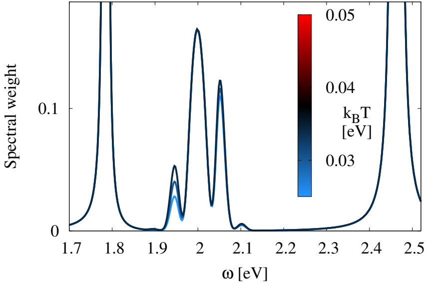

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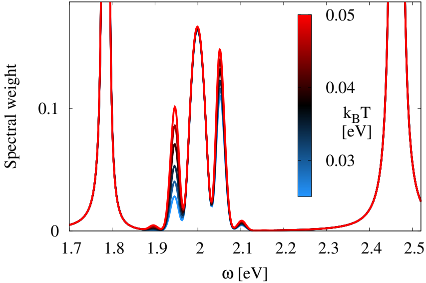

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Disordered molecules + vibrations – *vs* temperature

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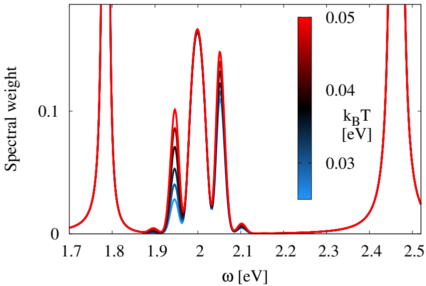

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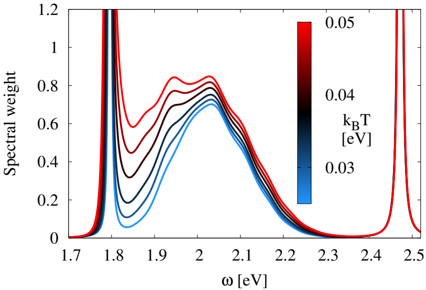


Acknowledgements

GROUP:




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

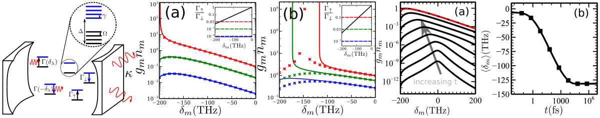
FUNDING:

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

EPSRC
Engineering and Physical Sciences Research Council

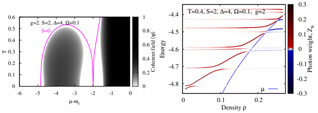
Summary

- Photon condensation and thermalisation



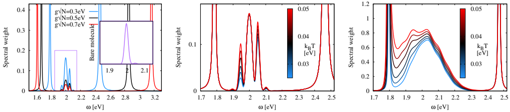
[Kirton & Keeling, PRL '13, arXiv:1410.6632]

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



[Cwik *et al.* EPL '14]

- Vibrational configuration

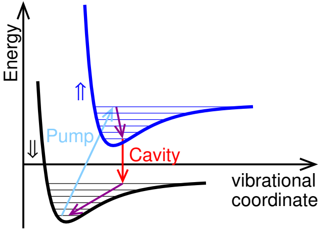


[Cwik *et al.* in preparation]

Extra Slides

Dicke-Holstein model: dye laser

4 Level Dye Laser



- No strong coupling
- No electronic inversion — vibrational inversion.

- Multiple photon modes
  - Condensate mode is not maximum gain
  - Gain/Absorption in balance
  - Thermalisation
- (Ultra)strong matter-light coupling

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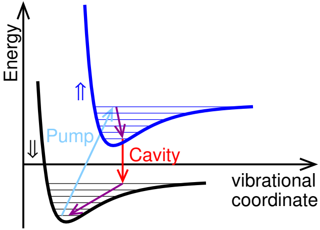

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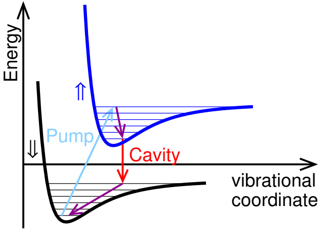

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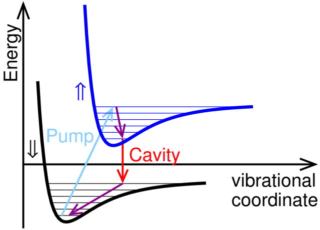

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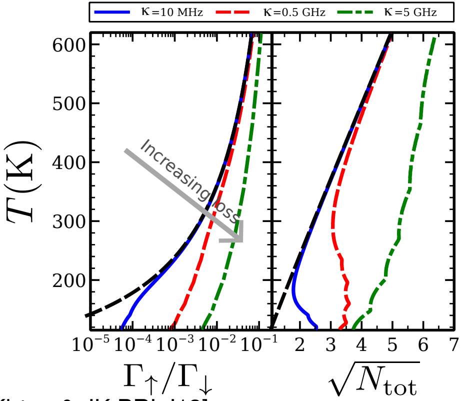
Threshold condition



[Kirton & JK PRL '13]

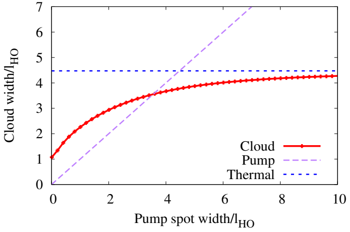
Compare threshold:

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

Spatially varying pump intensity

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

$$\partial_t n_m = \Gamma(\delta_m) \int d\vec{r} \rho_\uparrow |\psi_m(r)|^2 (n_m + 1) - \left(\kappa + \Gamma(\delta_m) \int d\vec{r} (\rho_m - \rho_\uparrow) |\psi_m(r)|^2 \right) n_m$$



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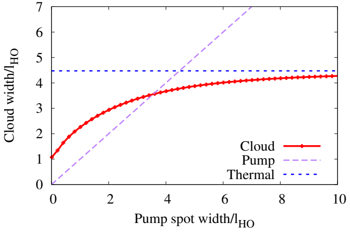
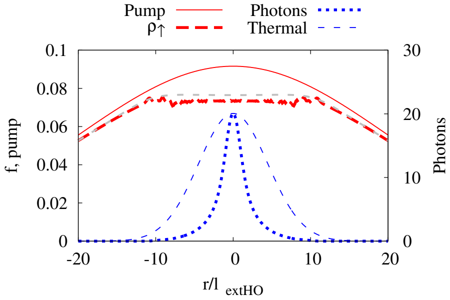
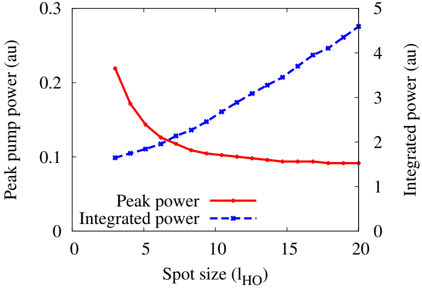
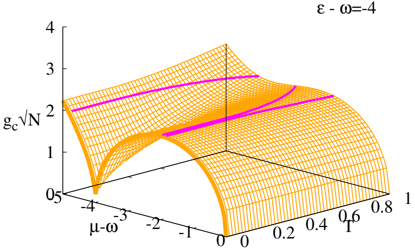






Critical coupling with increasing S

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



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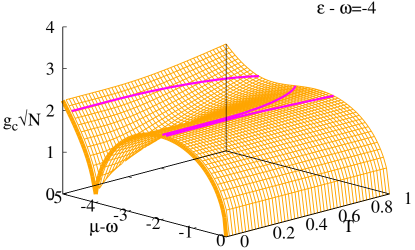

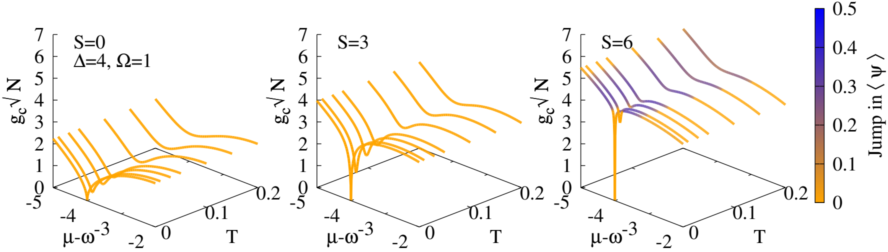

Explanation: Polaron formation

- 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 $g_{\text{eff}} \sim g \times \exp(-S/2)$
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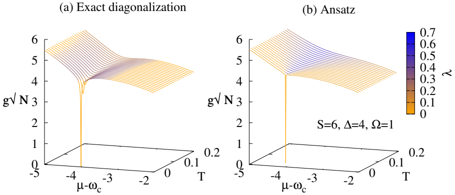
Collective polaron formation

- 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]

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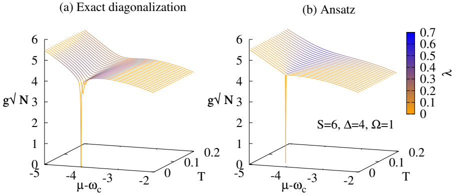

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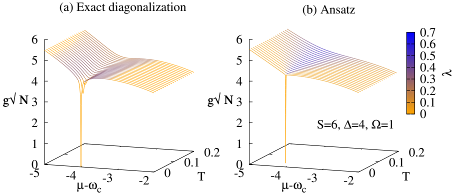

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Polariton spectrum — coupled oscillators

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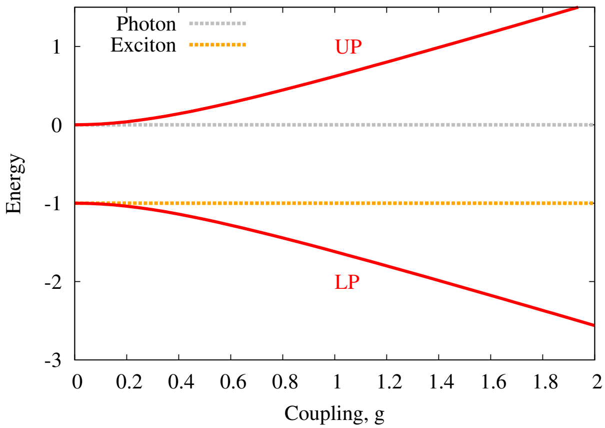

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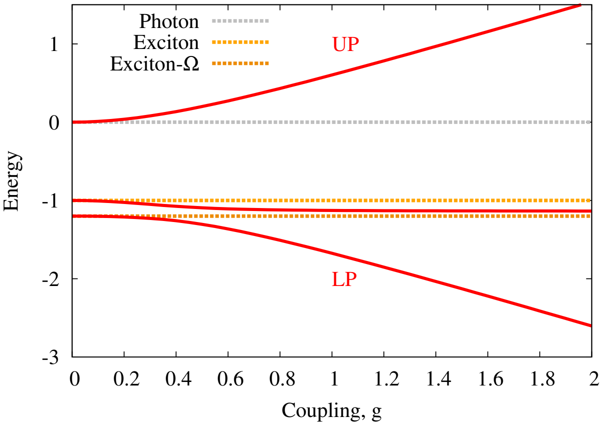

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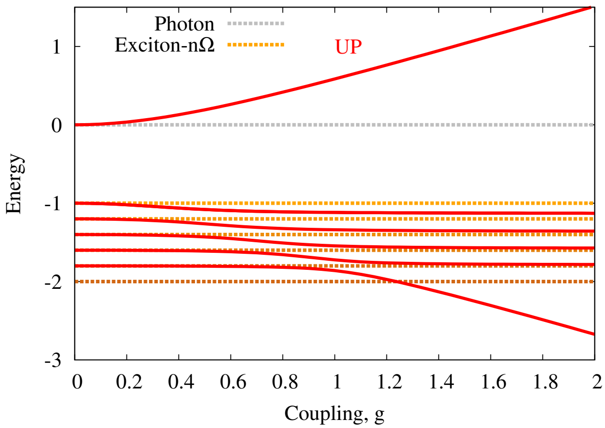

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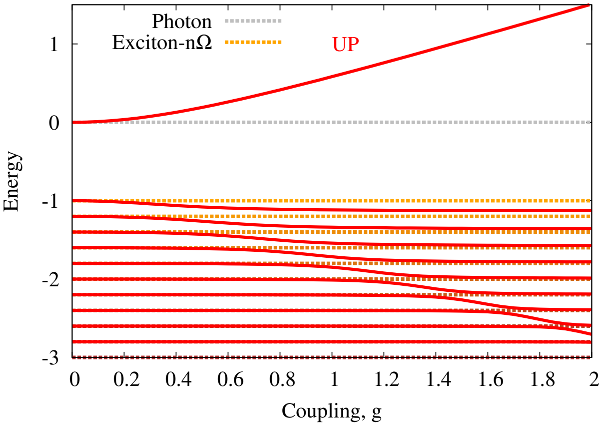

Polariton spectrum: what condensed

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

- Optimal $T \sim 2\Omega$

[Cwik *et al.* EPL ’14]

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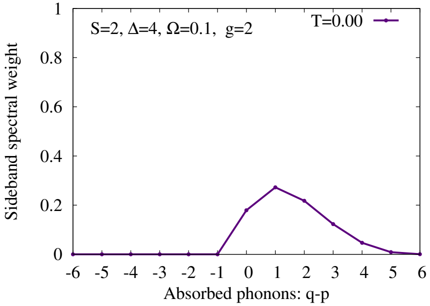

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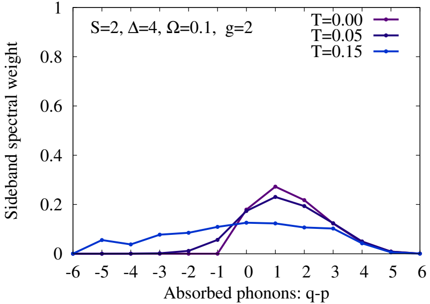

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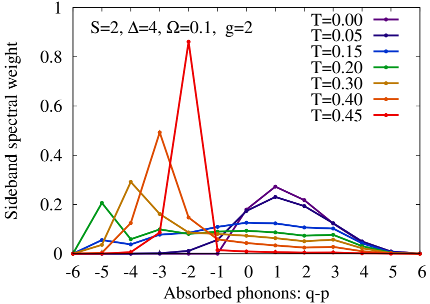

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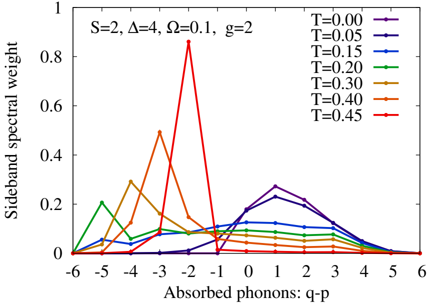

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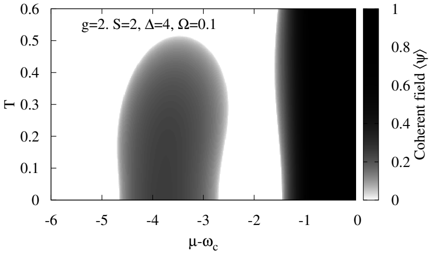

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Vibrational reconfiguration

- $H = H_0 + H_1, H_1 = \sum_{n,k} g_{n,k}(\psi^\dagger_k \sigma^+_n + \text{H.c.})$
- 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 $\sqrt{S} \to \sqrt{S}(1 - g^2 N/(\epsilon+\omega))$
- Increased effective coupling:

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

- Numerically tiny effect, $\Omega \ll \epsilon$

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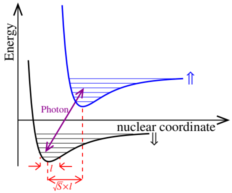

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$$g_{\text{eff}}^2 = g^2 \exp(-S)$$

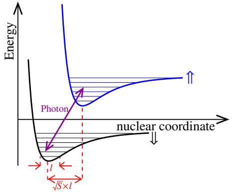
- Numerically tiny effect, $\Omega \ll \epsilon$

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 $\sqrt{S} \to \sqrt{S}(1 - g^2 N/(\epsilon + \omega))$



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