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

Jonathan Keeling

Snowbird, January 2015
Matter-Light coupling with organic molecules

**What & why?**

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

Theory questions/challenges

- Ultrastrong coupling
- Vibrational modes
- (Partial) thermalisation

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

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

$$H_{\text{sys}} = \omega \psi^\dagger \psi + \sum_\alpha \left[ \frac{\epsilon}{2} \sigma_\alpha^z + g \left( \psi + \psi^\dagger \right) \left( \sigma_\alpha^+ + \sigma_\alpha^- \right) \right]$$
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$$+ \sum_\alpha \Omega \left\{ b^{\dagger}_\alpha b_\alpha + \sqrt{S} \sigma^z_\alpha \left( b^{\dagger}_\alpha + b_\alpha \right) \right\}$$
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
  \( \omega_m = \omega_{\text{cutoff}} + m \omega_{\text{H.O.}} \)
- Incoherent processes: excitation, decay, loss, vibrational thermalisation.
- Weak coupling, perturbative in \( g \)

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Modelling

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Master equation

\[
\dot{\rho} = -i[H_0, \rho] - \sum_m \frac{\kappa}{2} \mathcal{L}[\psi_m] - \sum_\alpha \left[ \frac{\Gamma^{\uparrow}}{2} \mathcal{L}[\sigma^{\uparrow}_\alpha] + \frac{\Gamma^{\downarrow}}{2} \mathcal{L}[\sigma^{\downarrow}_\alpha] \right] - \sum_{m,\alpha} \left[ \Gamma(\delta_m = \omega_m - \epsilon) \frac{1}{2} \mathcal{L}[\sigma^{\uparrow}_\alpha \psi_m] + \Gamma(-\delta_m = \epsilon - \omega_m) \frac{1}{2} \mathcal{L}[\sigma^{\downarrow}_\alpha \psi_m^\dagger] \right]
\]

\begin{center}
\includegraphics[width=0.4\textwidth]{graph.png}
\end{center}

- Kennard-Stepanov: \(\Gamma(\delta) \approx \Gamma(-\delta) e^{\beta \delta}\)
- Expt: \(\omega_0 < \epsilon\)
- \(\Gamma \to 0\) at large \(\delta\)

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

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[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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- Bose-Einstein distribution without losses

Low loss: Thermal $\rightarrow$ Laser

[Kirton & JK PRL '13]
Time evolution

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

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Strong coupling: polaritons

1. Weak coupling: photon condensation

2. Strong coupling: polaritons

3. Ultra strong coupling: vibrational reconfiguration
Strong coupling phase diagram — mean field

- Mean field — single photon mode

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

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

- Reentrant behaviour — \( \text{Min } \mu \text{ at } k_B T \sim 0.1 \Omega \)
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Polariton spectrum: photon weight

Saturating 2LS: $g_{\text{eff}}^2 \sim g^2 (1 - 2\rho)$

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?

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

[Cwik et al. EPL '14]
Polariton spectrum: photon weight

![Graph showing polariton spectrum with photon weight values and energy density.](image)

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

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

[Cwik et al. EPL '14]
Ultra strong coupling: vibrational reconfiguration

1. Weak coupling: photon condensation

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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]

[Diagram showing cavity resonance, molecular transition, and hybrid states.

Questions:
- Polariton vs. molecular spectral weight – chemical eqbm
- Temperature dependent

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Questions:

- Vibrationally dressed spectrum + disorder
- Microscopic theory – changing configuration
Disordered molecules — spectrum

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

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

- Ultra-strong coupling — renormalised photon

- Central peak:

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

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

Spectral weight

\[
g\sqrt{N} = 0.3 \text{ eV}
\]

\[
\text{Spectral weight}
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\[
\omega [\text{eV}]
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- Central peak:

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\[ g\sqrt{N}=0.3 \text{ eV} \]
\[ g\sqrt{N}=0.5 \text{ eV} \]
Disordered molecules — spectrum

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

[Houdré et al., PRA ’96]

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[Houdré et al., PRA ’96]

- Temperature independent (for $k_B T \ll g\sqrt{N}$)
Disordered molecules — vibrational mode

- With vibrational sidebands, $S = 0.02$

![Graph showing spectral weight vs. energy with vibrational sidebands for different $g\sqrt{N}$ values.](image)
Disordered molecules — vibrational mode

- With vibrational sidebands, $S = 0.02$

![Graph showing spectral weight against energy (ω [eV]) for different values of $g\sqrt{N}$: g$\sqrt{N}=0.3$ eV, g$\sqrt{N}=0.5$ eV, g$\sqrt{N}=0.7$ eV. The graph includes a zoom-in on the bare molecule region.](image)
Disordered molecules + vibrations – vs temperature

- vs vs temperature

- Stronger disorder &
  \[ S = 0.5, \sigma = 0.025 \text{eV} \]
Disordered molecules + vibrations – vs temperature

- vs vs temperature

Stronger disorder &

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Disordered molecules + vibrations – vs temperature

- vs vs temperature

S = 0.02, σ = 0.01 eV

Stronger disorder &
S = 0.5, σ = 0.025 eV

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**vs vs temperature**

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[EPSRC logo]

[Topnes logo]
Summary

- Photon condensation and thermalisation
  

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

  ![Graphs and diagrams related to photon condensation and thermalisation]

- Vibrational configuration

  [Cwik et al. EPL ’14]

- [Cwik et al. in preparation]
Extra Slides

4. Dye laser
5. Photon BEC threshold
6. Photon BEC with spatial profile
7. Ultra-strong phonon coupling?
8. Anticrossing vs $\rho$
9. Polariton spectrum nature
10. Vibrational reconfiguration
Dicke-Holstein model: dye laser

4 Level Dye Laser

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

- No strong coupling
- No electronic inversion — vibrational inversion.
Dicke-Holstein model: dye laser

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

In this talk:
- Multiple photon modes
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Threshold condition

Compare threshold:
- Pump rate (Laser)
- Critical density (condensate)

[Kirton & JK PRL '13]

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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 $\mu, T$
- Colors $\rightarrow$ Jump of $\langle \psi \rangle$

\[ \sqrt{N} g_c = 4, \quad \Omega = 1 \]
\[ S = 0 \]
\[ \mu - \omega, T \]

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Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to \( S^+ \)

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

- 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

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

Variational free energy

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

Effective 2LS energy in field:

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

[Cwik et al. EPL ’14]
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- Compares well at $S \gg 1$
- Coherent bosonic state

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

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

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]
Polariton spectrum — coupled oscillators
Polariton spectrum — coupled oscillators

-3
-2
-1
0
1
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2
Energy
Coupling, g
LP
UP
Photon
Exciton
Exciton-Ω

Jonathan Keeling
Weak, strong, ultra-strong
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Polariton spectrum — coupled oscillators

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Weak, strong, ultra-strong

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Polariton spectrum — coupled oscillators

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Weak, strong, ultra-strong

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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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\[ S=2, \Delta=4, \Omega=0.1, \ g=2 \]

Optimal \( T \sim 2\Omega \)

\[ \langle \psi \rangle \]

[Cwik et al. EPL ’14]
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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:

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

- Numerically tiny effect, \( \Omega \ll \epsilon \)
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