

Exact and approximate eigenstates of vibrationally dressed polaritons

Jonathan Keeling



University of
St Andrews

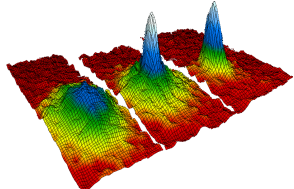
FOUNDED
1413



Quantum Nanophotonics, February 2017

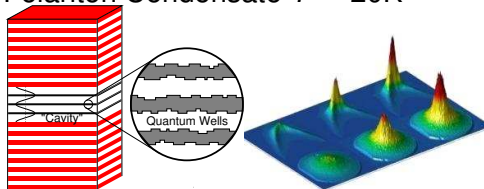
Condensation, Lasing, Superradiance

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



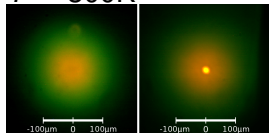
[Anderson *et al.* Science '95]

Polariton Condensate $T \sim 20K$



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

Photon Condensate
 $T \sim 300K$

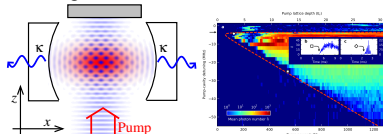


[Klaers *et al.* Nature, '10]

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



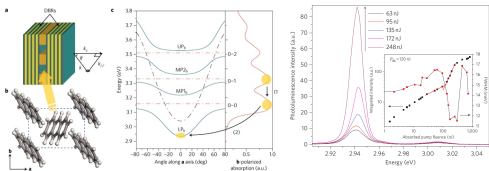
Superradiance transition
 $T \sim 0$



[Baumann *et al.* Nature '10]

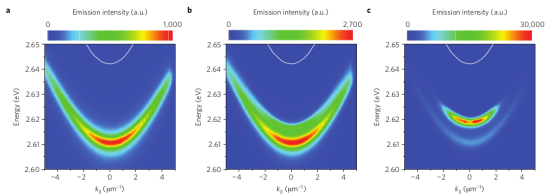
Motivation: polariton condensates

- Anthracene Polariton Lasing
 $T \sim 300\text{K}$



[Kena Cohen and Forrest, Nat. Photon '10]

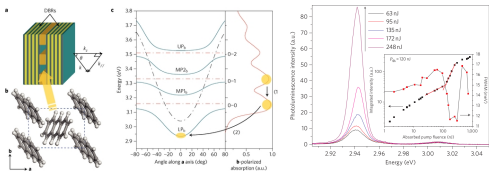
- Polariton condensates, other materials, e.g. polymers:



[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* *ibid* '14]

Motivation: polariton condensates

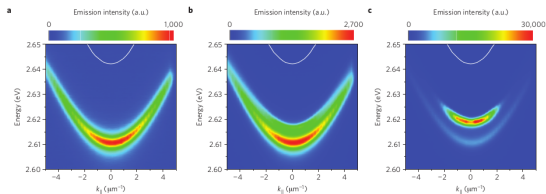
- Anthracene Polariton Lasing
 $T \sim 300\text{K}$



- Polariton condensates, other materials, e.g. polymers:

- Q1. Vibrational replicas?
- Q2. Relevance of disorder?
- Q3. Lasing vs condensation?

[Kena Cohen and Forrest, Nat. Photon '10]



[Plumhoff *et al.* Nat. Materials '14, Daskalakis *et al.* *ibid* '14]

Paradigms & Models

- Weakly interacting dilute Bose gas

$$H = \int d^d r \hat{\psi}^\dagger (-\mu - \nabla^2) \hat{\psi} + U \hat{\psi}^\dagger \hat{\psi}^\dagger \hat{\psi} \hat{\psi}$$

- ▶ Single field — assumes strong coupling
- ▶ Continuum model, hard to include molecular physics

• Laser rate equations

- ▶ Emission, absorption — assumes weak coupling, lasing

• Transfer matrix, exciton susceptibility $\chi(\nu)$

• Microscopic model ...

Paradigms & Models

- Weakly interacting dilute Bose gas

$$H = \int d^d r \hat{\psi}^\dagger (-\mu - \nabla^2) \hat{\psi} + U \hat{\psi}^\dagger \hat{\psi}^\dagger \hat{\psi} \hat{\psi}$$

- ▶ Single field — assumes strong coupling
 - ▶ Continuum model, hard to include molecular physics
- Laser rate equations
 - ▶ Emission, absorption — assumes weak coupling, lasing.

◦ Transfer matrix, exciton susceptibility $\chi(\nu)$

◦ Microscopic model ...

Paradigms & Models

- Weakly interacting dilute Bose gas

$$H = \int d^d r \hat{\psi}^\dagger (-\mu - \nabla^2) \hat{\psi} + U \hat{\psi}^\dagger \hat{\psi}^\dagger \hat{\psi} \hat{\psi}$$

- ▶ Single field — assumes strong coupling
 - ▶ Continuum model, hard to include molecular physics
- Laser rate equations
 - ▶ Emission, absorption — assumes weak coupling, lasing.
 - Transfer matrix, exciton susceptibility $\chi(\nu)$

● Microscopic model ...

Paradigms & Models

- Weakly interacting dilute Bose gas

$$H = \int d^d r \hat{\psi}^\dagger (-\mu - \nabla^2) \hat{\psi} + U \hat{\psi}^\dagger \hat{\psi}^\dagger \hat{\psi} \hat{\psi}$$

- ▶ Single field — assumes strong coupling
 - ▶ Continuum model, hard to include molecular physics
- Laser rate equations
 - ▶ Emission, absorption — assumes weak coupling, lasing.
 - Transfer matrix, exciton susceptibility $\chi(\nu)$
 - Microscopic model . . .

Tavis-Cummings & Dicke model

Model capable of lasing & condensation

- Tavis-Cummings / Dicke model

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) \right]$$

→ Weak pumping → Superradiance/BEC transition

→ High temperature: Maxwell-Bloch laser

- Including molecular physics

Tavis-Cummings & Dicke model

Model capable of lasing & condensation

- Tavis-Cummings / Dicke model + baths

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) \right]$$

- ▶ Weak pumping \rightarrow Superradiance/BEC transition
- ▶ High temperature: Maxwell-Bloch laser

Szymanska et al. PRL 06; Keeling et al. book chapter 1010.3338

• Including molecular physics

Tavis-Cummings & Dicke model

Model capable of lasing & condensation

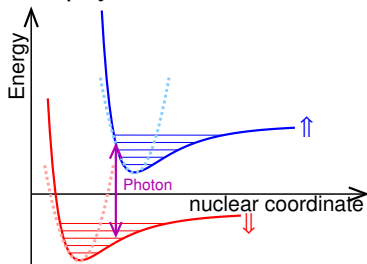
- Tavis-Cummings / Dicke model + baths

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) \right]$$

- ▶ Weak pumping \rightarrow Superradiance/BEC transition
- ▶ High temperature: Maxwell-Bloch laser

Szymanska et al. PRL 06; Keeling et al. book chapter 1010.3338

- Including molecular physics



Holstein-Tavis-Cummings & Holstein-Dicke model

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Few emitters (molecules/quantum dots)

Wilson-Rae & Imamoglu PRB 2002; McCutcheon & Nazir PRB 2011; Roy & Hughes PRB 2011; Bera *et al.* PRB 2014; Pollock *et al.* NJP 2013; Hornecker *et al.* arXiv:1609.09754; ...

- Weak coupling

Kirton & JK, PRL 2013, PRA 2015; PRA 2016 ...

- Full model

Cwik *et al.* EPL 105 2014; Spano, J. Chem. Phys 2015; Galego *et al.* PRX 2015; Cwik *et al.* PRA 2016; Herrera & Spano PRL 2016; Wu *et al.* arXiv:1608.08019; Zeb *et al.* arXiv:1608.08929; Herrera & Spano arXiv:1610.04252

Holstein-Tavis-Cummings & Holstein-Dicke model

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Few emitters (molecules/quantum dots)

Wilson-Rae & Imamoglu PRB 2002 McCutcheon & Nazir PRB 2011 Roy & Hughes PRB 2011; Bera *et al.* PRB 2014; Pollock *et al.* NJP 2013; Hornecker *et al.* arXiv:1609.09754; ...

- Weak coupling

Kirton & Jk, PRL 2013, PRA 2015; PRA 2016

- Full model

Cwik *et al.* EPL 105 2014; Spano, J. Chem. Phys 2015; Galego *et al.* PRX 2015;

Cwik *et al.* PRA 2016; Herrera & Spano PRL 2016; Wu *et al.* arXiv 1608.08019;

Zeb *et al.* arXiv:1608.08829; Herrera & Spano arXiv:1610.04252

Holstein-Tavis-Cummings & Holstein-Dicke model

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Few emitters (molecules/quantum dots)

Wilson-Rae & Imamoglu PRB 2002 McCutcheon & Nazir PRB 2011 Roy & Hughes PRB 2011; Bera *et al.* PRB 2014; Pollock *et al.* NJP 2013; Hornecker *et al.* arXiv:1609.09754; ...

- Weak coupling

Kirton & JK, PRL 2013, PRA 2015; PRA 2016 ...

- Full model

Cwik *et al.* EPL 105 2014; Spano, J. Chem. Phys 2015; Galego *et al.* PRX 2015; Cwik *et al.* PRA 2016; Herrera & Spano PRL 2016; Wu *et al.* arXiv 1608.08019; Zab *et al.* arXiv:1608.08829; Herrera & Spano arXiv:1610.04252

Holstein-Tavis-Cummings & Holstein-Dicke model

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g \left(\sigma_i^+ (\hat{a} + \hat{a}^\dagger) + \text{H.c.} \right) + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Few emitters (molecules/quantum dots)

Wilson-Rae & Imamoglu PRB 2002 McCutcheon & Nazir PRB 2011 Roy & Hughes PRB 2011; Bera *et al.* PRB 2014; Pollock *et al.* NJP 2013; Hornecker *et al.* arXiv:1609.09754; ...

- Weak coupling

Kirton & JK, PRL 2013, PRA 2015; PRA 2016 ...

- Full model

Cwik *et al.* EPL 105 2014; Spano, J. Chem. Phys 2015; Galego *et al.* PRX 2015; Cwik *et al.* PRA 2016; Herrera & Spano PRL 2016; Wu *et al.* arXiv:1608.08019; Zeb *et al.* arXiv:1608.08929; Herrera & Spano arXiv:1610.04252; ...

Introduction and models

- 1 Introduction and models
 - Holstein-Dicke model
- 2 Strong coupling: polariton states
 - Exact solutions
 - Scaling with N
- 3 Strong coupling: spectrum

Strong coupling: polariton states

- 1 Introduction and models
 - Holstein-Dicke model
- 2 Strong coupling: polariton states
 - Exact solutions
 - Scaling with N
- 3 Strong coupling: spectrum

One excitation subspace, questions

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g (\sigma_i^+ \hat{a} + \text{H.c.}) \right. \\ \left. + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Rotating wave approximation — Holstein Tavis Cummings

- Restrict, $\hat{a}^\dagger \hat{a} + \sum_i \sigma_i^+ \sigma_i^- = 1$.

- Questions:

- Competition of $g\sqrt{N}$ vs $\omega_V, \omega_V \lambda_0^2$

- Scaling with N

One excitation subspace, questions

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g (\sigma_i^+ \hat{a} + \text{H.c.}) + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Rotating wave approximation — Holstein Tavis Cummings
- Restrict, $\hat{a}^\dagger \hat{a} + \sum_i \sigma_i^+ \sigma_i^- = 1$.

• Questions:

- Competition of $g\sqrt{N}$ vs $\omega_V, \omega_V \lambda_0^2$
- Scaling with N

One excitation subspace, questions

$$H = \omega \hat{a}^\dagger \hat{a} + \sum_{i=1}^N \left[\omega_X \sigma_i^+ \sigma_i^- + g (\sigma_i^+ \hat{a} + \text{H.c.}) + \omega_V \left(\hat{b}_i^\dagger \hat{b}_i - \lambda_0 \sigma_i^+ \sigma_i^- (\hat{b}_i^\dagger + \hat{b}_i) \right) \right]$$

- Rotating wave approximation — Holstein Tavis Cummings
- Restrict, $\hat{a}^\dagger \hat{a} + \sum_i \sigma_i^+ \sigma_i^- = 1$.
- Questions:
 - ▶ Competition of $g\sqrt{N}$ vs $\omega_V, \omega_V \lambda_0^2$
 - ▶ Scaling with N

Exact solution, $N = 2$

Vibrational Wigner function:

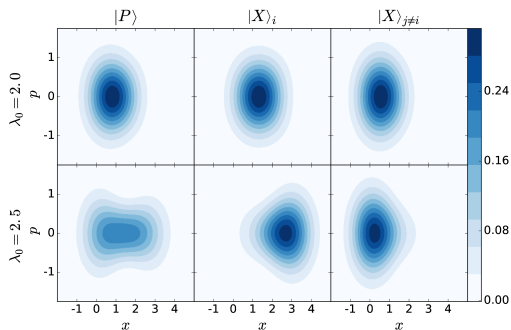
$$W(x, p) = \int dy \langle x + y/2 | \rho | x - y/2 \rangle_i e^{iyp}, \quad \left(\frac{\hat{b}_i + \hat{b}_i^\dagger}{\sqrt{2}} \right) |x\rangle_i = x|x\rangle_i$$

Exact solution, $N = 2$

Vibrational Wigner function:

$$W(x, p) = \int dy \langle x + y/2 | \rho | x - y/2 \rangle_i e^{iyp}, \quad \left(\frac{\hat{b}_i + \hat{b}_i^\dagger}{\sqrt{2}} \right) |x\rangle_i = x|x\rangle_i$$

Conditioned on Photon $|P\rangle$ /Exciton at i , $|X\rangle_i$ /Other site $|X\rangle_{j \neq i}$



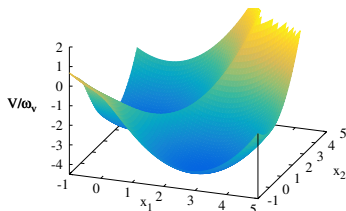
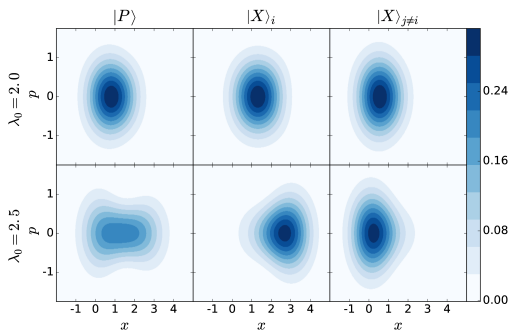
$$N = 2, \omega = \omega_X, \omega_R \equiv g/\sqrt{N} = 1$$

Exact solution, $N = 2$

Vibrational Wigner function:

$$W(x, p) = \int dy \langle x + y/2 | \rho | x - y/2 \rangle_i e^{iyp}, \quad \left(\frac{\hat{b}_i + \hat{b}_i^\dagger}{\sqrt{2}} \right) |x\rangle_i = x|x\rangle_i$$

Conditioned on Photon $|P\rangle$ /Exciton at i , $|X\rangle_i$ /Other site $|X\rangle_{j \neq i}$



$$N = 2, \omega = \omega_X, \omega_R \equiv g/\sqrt{N} = 1$$

Exact solution, larger N

- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$

• Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5 - 6$

- Increasing N , suppress $W_{(P)}(x \neq 0)$
- Distinct behaviour vs λ_0
- Exact energy and state vs ω_R, λ_0 for validation

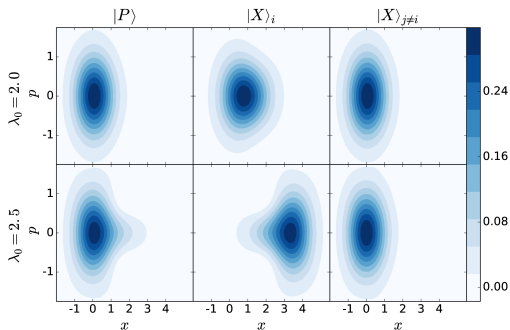
Exact solution, larger N

- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$
- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5 - 6$

- Increasing N , suppress $W_{(P)}(x \neq 0)$
- Distinct behaviour vs λ_0
- Exact energy and state vs ω_R, λ_0 for validation

Exact solution, larger N

- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$
- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5 - 6$



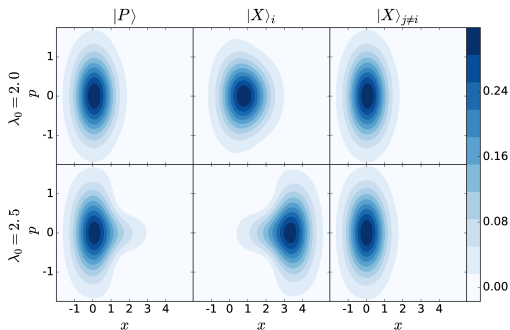
- Increasing N , suppress $W_{|P\rangle}(x \neq 0)$

- Distinct behaviour vs λ_0
- Exact energy and state vs ω_R, λ_0 for validation

$$N = 20, \omega = \omega_X, \omega_R \equiv g/\sqrt{N} = 1$$

Exact solution, larger N

- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$
- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5 - 6$



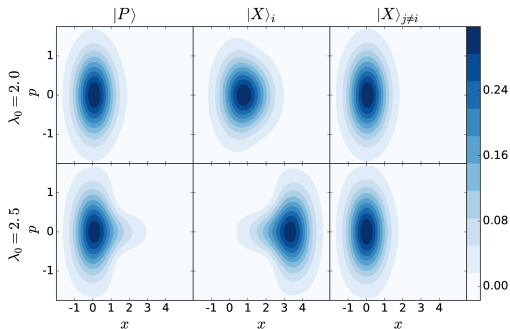
- Increasing N , suppress $W_{|P\rangle}(x \neq 0)$
- Distinct behaviour vs λ_0

• Exact energy and state vs ω_R, λ_0 for validation

$$N = 20, \omega = \omega_X, \omega_R \equiv g/\sqrt{N} = 1$$

Exact solution, larger N

- Brute force approach, N sites, $\hat{b}^\dagger \hat{b} < M$, $D_{\text{Hilbert}} = M^N$
- Permutation symmetry. $D_{\text{Hilbert}} \sim N^M$, typical $M \sim 5 - 6$



- Increasing N , suppress $W_{|P\rangle}(x \neq 0)$
- Distinct behaviour vs λ_0
- Exact energy and state vs ω_R, λ_0 for validation

$$N = 20, \omega = \omega_X, \omega_R \equiv g/\sqrt{N} = 1$$

Extending to arbitrary N , polaron ansatz

- Polaron transform, $\mathcal{D}_i(\lambda) = \exp(\lambda(\hat{b}_i^\dagger - \hat{b}_i))$

- Single molecule ansatz:

$$|\Psi\rangle = [\alpha \mathcal{D}(\lambda_+) |1\rangle + \beta \mathcal{D}(\lambda_-) |0\rangle] |0\rangle_V$$

- Extend to N sites

$$|\Psi\rangle = \left[\alpha |F\rangle \prod_j \mathcal{D}_j(\lambda_j) + \frac{\beta}{\sqrt{N}} \sum_j |X\rangle_j \mathcal{D}_j(\lambda_j) \prod_{I \neq j} \mathcal{D}_I(\lambda_I) \right] |0\rangle_V$$

[Wu *et al.* arXiv:1608.08019, Zeb *et al.* arXiv:1608.08929]

Extending to arbitrary N , polaron ansatz

- Polaron transform, $\mathcal{D}_i(\lambda) = \exp(\lambda(\hat{b}_i^\dagger - \hat{b}_i))$
- Single molecule ansatz:

$$|\Psi\rangle = [\alpha\mathcal{D}(\lambda_\uparrow)|\uparrow\rangle + \beta\mathcal{D}(\lambda_\downarrow)|\downarrow\rangle]|\mathbf{0}\rangle_V$$

• Extend to N sites

$$|\Psi\rangle = \left[\alpha|F\rangle \prod_i \mathcal{D}_i(\lambda_\alpha) + \frac{\beta}{\sqrt{N}} \sum_i |X\rangle_i \mathcal{D}_i(\lambda_\beta) \prod_{i \neq j} \mathcal{D}_j(\lambda_\alpha) \right] |\mathbf{0}\rangle_V$$

[Wu *et al.* arXiv:1605.08019, Zeb *et al.* arXiv:1608.08929]

Extending to arbitrary N , polaron ansatz

- Polaron transform, $\mathcal{D}_i(\lambda) = \exp(\lambda(\hat{b}_i^\dagger - \hat{b}_i))$
- Single molecule ansatz:

$$|\Psi\rangle = [\alpha\mathcal{D}(\lambda_\uparrow)|\uparrow\rangle + \beta\mathcal{D}(\lambda_\downarrow)|\downarrow\rangle] |0\rangle_V$$

- Extend to N sites

$$|\Psi\rangle = \left[\alpha |P\rangle \prod_j \mathcal{D}_j(\lambda_a) + \frac{\beta}{\sqrt{N}} \sum_i |X\rangle_i \mathcal{D}_i(\lambda_b) \prod_{j \neq i} \mathcal{D}_j(\lambda_c) \right] |0\rangle_V$$

[Wu *et al.* arXiv:1608.08019, Zeb *et al.* arXiv:1608.08929]

Allows distinct Wigner functions $|P\rangle, |X\rangle_{i1}, |X\rangle_{i2}$

Extending to arbitrary N , polaron ansatz

- Polaron transform, $\mathcal{D}_i(\lambda) = \exp(\lambda(\hat{b}_i^\dagger - \hat{b}_i))$
- Single molecule ansatz:

$$|\Psi\rangle = [\alpha\mathcal{D}(\lambda_\uparrow)|\uparrow\rangle + \beta\mathcal{D}(\lambda_\downarrow)|\downarrow\rangle] |0\rangle_V$$

- Extend to N sites

$$|\Psi\rangle = \left[\alpha |P\rangle \prod_j \mathcal{D}_j(\lambda_a) + \frac{\beta}{\sqrt{N}} \sum_i |X\rangle_i \mathcal{D}_i(\lambda_b) \prod_{j \neq i} \mathcal{D}_j(\lambda_c) \right] |0\rangle_V$$

[Wu *et al.* arXiv:1608.08019, Zeb *et al.* arXiv:1608.08929]

- ▶ Allows distinct Wigner functions $|P\rangle, |X\rangle_i, |X\rangle_{j \neq i}$

Polaron ansatz energy

- Polaron energy: $E_{LP} = \frac{\tilde{\omega}_X + \tilde{\omega}_P}{2} - \sqrt{\left(\frac{\tilde{\omega}_X + \tilde{\omega}_P}{2}\right)^2 + \tilde{\omega}_R^2}$

$$\tilde{\omega}_X = \omega_X + \omega_V(\lambda_b^2 - 2\lambda_0\lambda_b + (N-1)\lambda_c^2), \quad \tilde{\omega}_P = \omega + \omega_V N \lambda_a^2$$

$$\tilde{\omega}_R^2 = \omega_R^2 \exp\left[-(\lambda_a - \lambda_b)^2 - (N-1)(\lambda_a - \lambda_c)^2\right]$$

• At $N \rightarrow \infty$ Suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$

• If $\omega_R \gg \omega_V$, suggests $\lambda_a = \lambda_b = \lambda_c \sim 1/\sqrt{N}$ — factorisation

[Herrera and Spano PRL 2016]

• Minimisation:

Polaron ansatz energy

- Polaron energy: $E_{LP} = \frac{\tilde{\omega}_X + \tilde{\omega}_P}{2} - \sqrt{\left(\frac{\tilde{\omega}_X + \tilde{\omega}_P}{2}\right)^2 + \tilde{\omega}_R^2}$

$$\tilde{\omega}_X = \omega_X + \omega_V(\lambda_b^2 - 2\lambda_0\lambda_b + (N-1)\lambda_c^2), \quad \tilde{\omega}_P = \omega + \omega_V N \lambda_a^2$$

$$\tilde{\omega}_R^2 = \omega_R^2 \exp\left[-(\lambda_a - \lambda_b)^2 - (N-1)(\lambda_a - \lambda_c)^2\right]$$

- At $N \rightarrow \infty$ Suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$

• If $\omega_R \gg \omega_V$, suggests $\lambda_a = \lambda_b = \lambda_c \sim 1/\sqrt{N}$ — factorisation

[Herrera and Spano PRL 2015]

• Minimisation:

Polaron ansatz energy

- Polaron energy:
$$E_{LP} = \frac{\tilde{\omega}_X + \tilde{\omega}_P}{2} - \sqrt{\left(\frac{\tilde{\omega}_X + \tilde{\omega}_P}{2}\right)^2 + \tilde{\omega}_R^2}$$

$$\tilde{\omega}_X = \omega_X + \omega_V(\lambda_b^2 - 2\lambda_0\lambda_b + (N-1)\lambda_c^2), \quad \tilde{\omega}_P = \omega + \omega_V N \lambda_a^2$$

$$\tilde{\omega}_R^2 = \omega_R^2 \exp\left[-(\lambda_a - \lambda_b)^2 - (N-1)(\lambda_a - \lambda_c)^2\right]$$

- At $N \rightarrow \infty$ Suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$
- If $\omega_R \gg \omega_V$, suggests $\lambda_a = \lambda_b = \lambda_c \sim 1/\sqrt{N}$ — factorisation

[Herrera and Spano PRL 2016]

• Minimisation:

Polaron ansatz energy

- Polaron energy:
$$E_{LP} = \frac{\tilde{\omega}_X + \tilde{\omega}_P}{2} - \sqrt{\left(\frac{\tilde{\omega}_X + \tilde{\omega}_P}{2}\right)^2 + \tilde{\omega}_R^2}$$

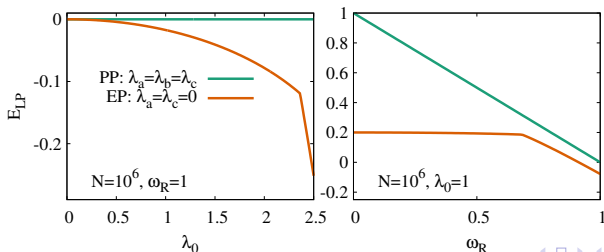
$$\tilde{\omega}_X = \omega_X + \omega_V(\lambda_b^2 - 2\lambda_0\lambda_b + (N-1)\lambda_c^2), \quad \tilde{\omega}_P = \omega + \omega_V N \lambda_a^2$$

$$\tilde{\omega}_R^2 = \omega_R^2 \exp\left[-(\lambda_a - \lambda_b)^2 - (N-1)(\lambda_a - \lambda_c)^2\right]$$

- At $N \rightarrow \infty$ Suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$
- If $\omega_R \gg \omega_V$, suggests $\lambda_a = \lambda_b = \lambda_c \sim 1/\sqrt{N}$ — factorisation

[Herrera and Spano PRL 2016]

- Minimisation:



Polaron ansatz energy

- Polaron energy:
$$E_{LP} = \frac{\tilde{\omega}_X + \tilde{\omega}_P}{2} - \sqrt{\left(\frac{\tilde{\omega}_X + \tilde{\omega}_P}{2}\right)^2 + \tilde{\omega}_R^2}$$

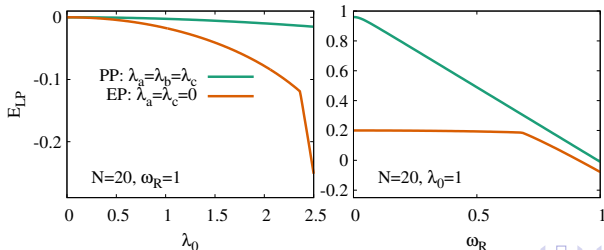
$$\tilde{\omega}_X = \omega_X + \omega_V(\lambda_b^2 - 2\lambda_0\lambda_b + (N-1)\lambda_c^2), \quad \tilde{\omega}_P = \omega + \omega_V N \lambda_a^2$$

$$\tilde{\omega}_R^2 = \omega_R^2 \exp\left[-(\lambda_a - \lambda_b)^2 - (N-1)(\lambda_a - \lambda_c)^2\right]$$

- At $N \rightarrow \infty$ Suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$
- If $\omega_R \gg \omega_V$, suggests $\lambda_a = \lambda_b = \lambda_c \sim 1/\sqrt{N}$ — factorisation

[Herrera and Spano PRL 2016]

- Minimisation:



Polaron ansatz energy

- Polaron energy:
$$E_{LP} = \frac{\tilde{\omega}_X + \tilde{\omega}_P}{2} - \sqrt{\left(\frac{\tilde{\omega}_X + \tilde{\omega}_P}{2}\right)^2 + \tilde{\omega}_R^2}$$

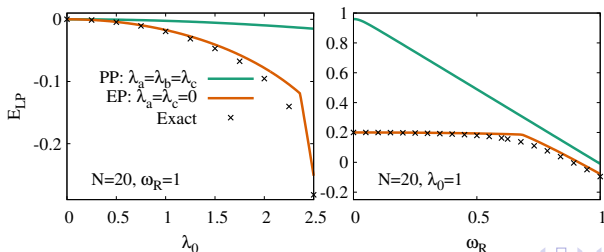
$$\tilde{\omega}_X = \omega_X + \omega_V(\lambda_b^2 - 2\lambda_0\lambda_b + (N-1)\lambda_c^2), \quad \tilde{\omega}_P = \omega + \omega_V N \lambda_a^2$$

$$\tilde{\omega}_R^2 = \omega_R^2 \exp\left[-(\lambda_a - \lambda_b)^2 - (N-1)(\lambda_a - \lambda_c)^2\right]$$

- At $N \rightarrow \infty$ Suggests $\lambda_a = \lambda_c \sim 1/\sqrt{N} \rightarrow 0$
- If $\omega_R \gg \omega_V$, suggests $\lambda_a = \lambda_b = \lambda_c \sim 1/\sqrt{N}$ — factorisation

[Herrera and Spano PRL 2016]

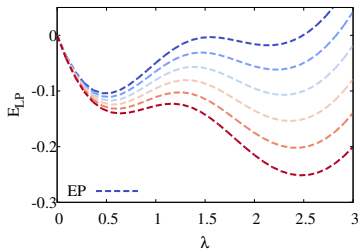
- Minimisation:



Polaron crossover

- Crossover near $\omega_R \simeq \omega_V \lambda_0^2$

[Silbey and Harris, J. Chem. Phys. 1984]

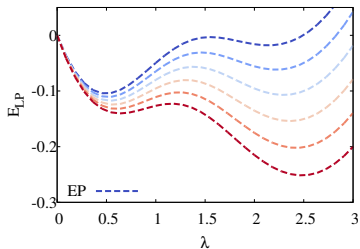


- Suggests multi-polaron ansatz [Bera *et al.* PRB 2014]
 - Superpose multiple polarons
 - Multimodal Wigner function
- Simplified 2-polaron form [Zeb *et al.* arXiv:1608.08929]

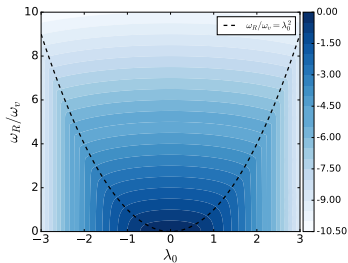
$$|\Psi\rangle = \left[|P\rangle \frac{1}{\sqrt{N}} \sum_I (\alpha_1 + \alpha_2 D_I(\lambda)) + \frac{1}{\sqrt{N}} \sum_I |X\rangle_I (\beta_1 + \beta_2 D_I(\lambda)) \right] |0\rangle_V$$

Polaron crossover

- Crossover near $\omega_R \simeq \omega_V \lambda_0^2$



[Silbey and Harris, J. Chem. Phys. 1984]

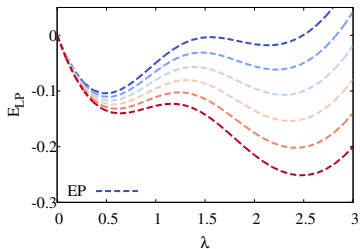


- Suggests multi-polaron ansatz [Bera *et al.* PRB 2014]
 - Superpose multiple polarons
 - Multimodal Wigner function
- Simplified 2-polaron form [Zeb *et al.* arXiv:1608.08929]

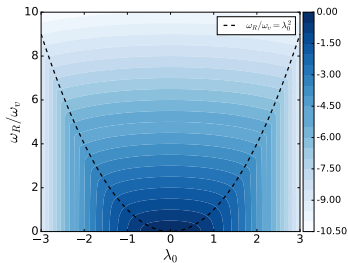
$$|\Psi\rangle = \left[(P) \frac{1}{\sqrt{N}} \sum_T (\alpha_1 + \alpha_2 D_T(\lambda)) + \frac{1}{\sqrt{N}} \sum_T (X)_T (\beta_1 + \beta_2 D_T(\lambda)) \right] |0\rangle_V$$

Polaron crossover

- Crossover near $\omega_R \simeq \omega_V \lambda_0^2$



[Silbey and Harris, J. Chem. Phys. 1984]



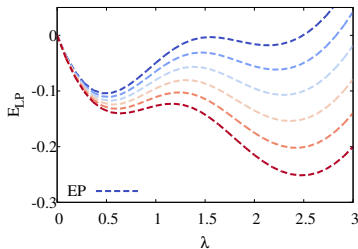
- Suggests multi-polaron ansatz [Bera *et al.* PRB 2014]
 - ▶ Superpose multiple polarons
 - ▶ Multimodal Wigner function

• Simplified 2-polaron form [Zeb *et al.* arXiv:1608.08929]

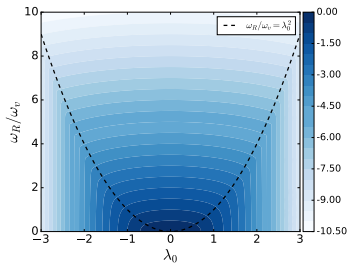
$$|\Psi\rangle = \left[\frac{1}{\sqrt{N}} \sum_T (\alpha_1 + \alpha_2 D_T(\lambda)) + \frac{1}{\sqrt{N}} \sum_T (\beta_1 + \beta_2 D_T(\lambda)) \right] |0\rangle_V$$

Polaron crossover

- Crossover near $\omega_R \simeq \omega_V \lambda_0^2$



[Silbey and Harris, J. Chem. Phys. 1984]

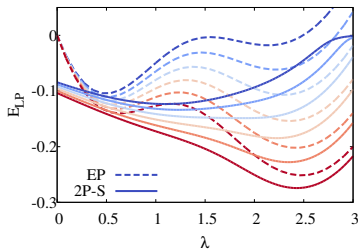


- Suggests multi-polaron ansatz [Bera *et al.* PRB 2014]
 - ▶ Superpose multiple polarons
 - ▶ Multimodal Wigner function
- Simplified 2-polaron form [Zeb *et al.* arXiv:1608.08929]

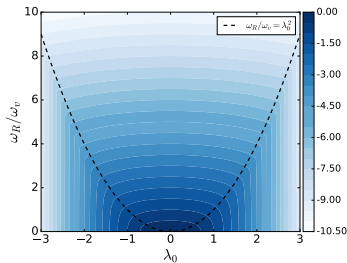
$$|\Psi\rangle = \left[|P\rangle \frac{1}{N} \sum_i (\alpha_1 + \alpha_2 \mathcal{D}_i(\lambda)) + \frac{1}{\sqrt{N}} \sum_i |X\rangle_i (\beta_1 + \beta_2 \mathcal{D}_i(\lambda)) \right] |0\rangle_V$$

Polaron crossover

- Crossover near $\omega_R \simeq \omega_V \lambda_0^2$



[Silbey and Harris, J. Chem. Phys. 1984]

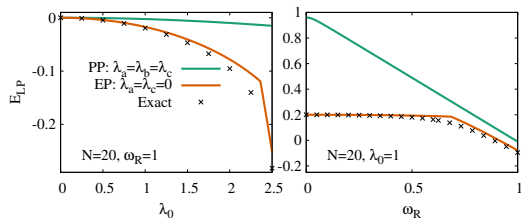


- Suggests multi-polaron ansatz [Bera *et al.* PRB 2014]
 - ▶ Superpose multiple polarons
 - ▶ Multimodal Wigner function
- Simplified 2-polaron form [Zeb *et al.* arXiv:1608.08929]

$$|\Psi\rangle = \left[|P\rangle \frac{1}{N} \sum_i (\alpha_1 + \alpha_2 \mathcal{D}_i(\lambda)) + \frac{1}{\sqrt{N}} \sum_i |X\rangle_i (\beta_1 + \beta_2 \mathcal{D}_i(\lambda)) \right] |0\rangle_V$$

Simplified two-polaron physics

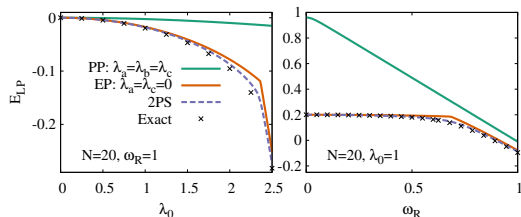
- Accurate energy & wavefunction



- Recovers Wigner function (analytic)
 - $W(p, q) \sim 1/N$, no other suppression

Simplified two-polaron physics

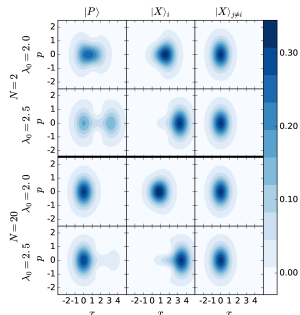
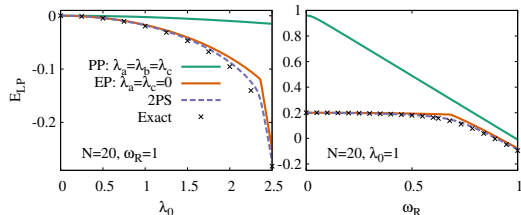
- Accurate energy & wavefunction



- Recovers Wigner function (analytic)
 - $W(p, q) \sim 1/N$, no other suppression

Simplified two-polaron physics

- Accurate energy & wavefunction



- Recovers Wigner function (analytic)
 - ▶ $W_{|P\rangle}(x \neq 0, p) \sim 1/N$, no other suppression

Strong coupling: spectrum

- 1 Introduction and models
 - Holstein-Dicke model
- 2 Strong coupling: polariton states
 - Exact solutions
 - Scaling with N
- 3 Strong coupling: spectrum

Calculating spectra: Input-Output formalism

- Observable features: absorption spectrum, $A(\nu) = 1 - T(\nu) - R(\nu)$

- Scattering matrix gives:

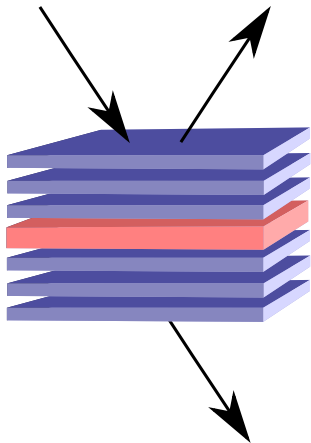
$$A(\nu) = -\kappa_i \left[2 \operatorname{Im}[D^R(\nu)] + (\kappa_i + \kappa_b) |D^R(\nu)|^2 \right]$$

- Green's function:

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

Calculating spectra: Input-Output formalism

- Obsevable features: absorption spectrum, $A(\nu) = 1 - T(\nu) - R(\nu)$



- Scattering matrix gives:

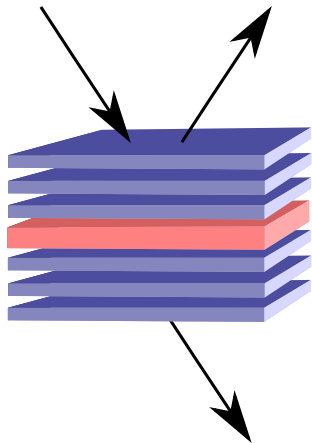
$$A(\nu) = -\pi_i \left[2 \operatorname{Im}[D^R(\nu)] + (\kappa_i + \kappa_b) |D^R(\nu)|^2 \right]$$

- Green's function:

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

Calculating spectra: Input-Output formalism

- Obsevable features: absorption spectrum, $A(\nu) = 1 - T(\nu) - R(\nu)$



- Scattering matrix gives:

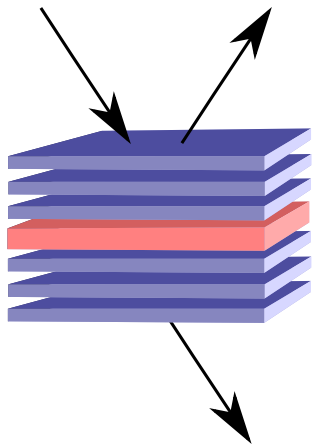
$$A(\nu) = -\kappa_t \left[2 \operatorname{Im}[D^R(\nu)] + (\kappa_t + \kappa_b) |D^R(\nu)|^2 \right]$$

- Green's function:

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

Calculating spectra: Input-Output formalism

- Observable features: absorption spectrum, $A(\nu) = 1 - T(\nu) - R(\nu)$



- Scattering matrix gives:

$$A(\nu) = -\kappa_t \left[2 \operatorname{Im}[D^R(\nu)] + (\kappa_t + \kappa_b) |D^R(\nu)|^2 \right]$$

- Green's function:

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

Tavis-Cummings-Holstein vs Coupled Oscillators

- Coupled oscillator model:

$$H = \omega_P \hat{a}^\dagger \hat{a} + \sum_i \left[\frac{\omega_R}{\sqrt{N}} \left(\hat{a} \sum_n f_n(\lambda_0) \sigma_i^{n0} + \text{H.c.} \right) + \omega_n \sigma_i^{nn} \right]$$

$$\omega_n = \omega_X + n\omega_V, \quad f_n(\lambda_0) = \langle n | D(\lambda_0) | 0 \rangle$$

- Corresponds to classical susceptibility,

$$\chi(\nu) = - \sum_n \frac{\omega_R^2 |f_n(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_n}$$

- Ignores vibrational dressing of unexcited molecules

Tavis-Cummings-Holstein vs Coupled Oscillators

- Coupled oscillator model:

$$H = \omega_P \hat{a}^\dagger \hat{a} + \sum_i \left[\frac{\omega_R}{\sqrt{N}} \left(\hat{a} \sum_n f_n(\lambda_0) \sigma_i^{n0} + \text{H.c.} \right) + \omega_n \sigma_i^{nn} \right]$$

$$\omega_n = \omega_X + n\omega_V, \quad f_n(\lambda_0) = \langle n | D(\lambda_0) | 0 \rangle$$



- Corresponds to classical susceptibility,

$$\chi(\nu) = - \sum_n \frac{\omega_R^2 |f_n(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_n}$$

- Ignores vibrational dressing of unexcited molecules

Tavis-Cummings-Holstein vs Coupled Oscillators

- Coupled oscillator model:

$$H = \omega_P \hat{a}^\dagger \hat{a} + \sum_i \left[\frac{\omega_R}{\sqrt{N}} \left(\hat{a} \sum_n f_n(\lambda_0) \sigma_i^{n0} + \text{H.c.} \right) + \omega_n \sigma_i^{nn} \right]$$

$$\omega_n = \omega_X + n\omega_V, \quad f_n(\lambda_0) = \langle n | D(\lambda_0) | 0 \rangle$$



- Corresponds to classical susceptibility,

$$\chi(\nu) = - \sum_n \frac{\omega_R^2 |f_n(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_n}$$

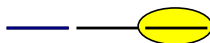
- Ignores vibrational dressing of unexcited molecules

Tavis-Cummings-Holstein vs Coupled Oscillators

- Coupled oscillator model:

$$H = \omega_P \hat{a}^\dagger \hat{a} + \sum_i \left[\frac{\omega_R}{\sqrt{N}} \left(\hat{a} \sum_n f_n(\lambda_0) \sigma_i^{n0} + \text{H.c.} \right) + \omega_n \sigma_i^{nn} \right]$$

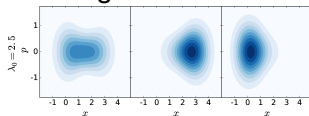
$$\omega_n = \omega_X + n\omega_V, \quad f_n(\lambda_0) = \langle n | D(\lambda_0) | 0 \rangle$$



- Corresponds to classical susceptibility,

$$\chi(\nu) = - \sum_n \frac{\omega_R^2 |f_n(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_n}$$

- Ignores vibrational dressing of unexcited molecules



Tavis-Cummings-Holstein spectrum

- Direct calculation

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

- Time-evolve $|v_0\rangle = \hat{a}^\dagger|0\rangle$
- Fourier transform
- Mean-field Green's function

- Why? Multiple excitation $\sim 1/N$,

Tavis-Cummings-Holstein spectrum

- Direct calculation

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger |0\rangle$

- Fourier transform

• Mean-field Green's function

• Why? Multiple excitation $\sim 1/N$,

Tavis-Cummings-Holstein spectrum

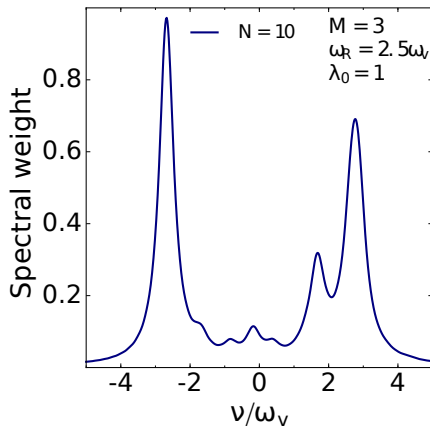
- Direct calculation

$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger |0\rangle$

- Fourier transform

• Mean-field Green's function



• Why? Multiple excitation $\sim 1/N$,

Tavis-Cummings-Holstein spectrum

- Direct calculation

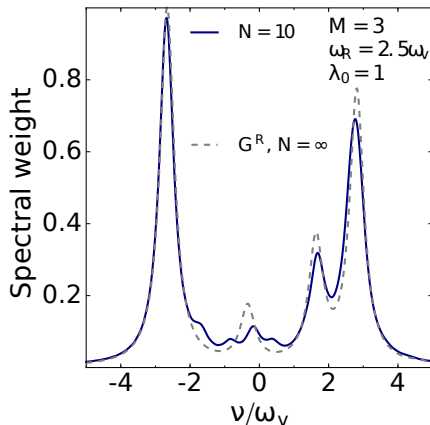
$$D^R(t) = -i \langle 0 | \left[\hat{a}(t), \hat{a}^\dagger(0) \right] | 0 \rangle \theta(t)$$

- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger |0\rangle$
- Fourier transform
- Mean-field Green's function

$$D^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_P + \Sigma_X(\nu)}$$

$$\sigma_X = - \sum_m \frac{\omega_R^2 |f_m(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_m}$$

(Classical expression)



• Why? Multiple excitation $\sim 1/N$.

Tavis-Cummings-Holstein spectrum

- Direct calculation

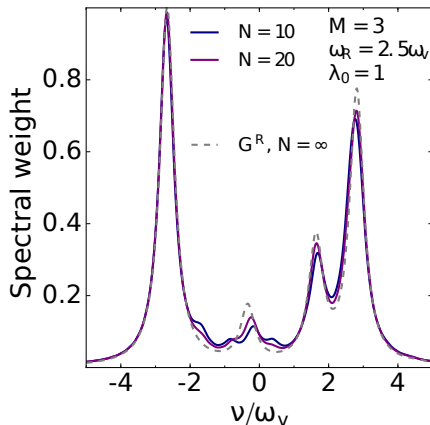
$$D^R(t) = -i \langle 0 | \left[\hat{a}(t), \hat{a}^\dagger(0) \right] | 0 \rangle \theta(t)$$

- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger |0\rangle$
- Fourier transform
- Mean-field Green's function

$$D^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_P + \Sigma_X(\nu)}$$

$$\sigma_X = - \sum_m \frac{\omega_R^2 |f_m(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_m}$$

(Classical expression)



• Why? Multiple excitation $\sim 1/N$.

Tavis-Cummings-Holstein spectrum

- Direct calculation

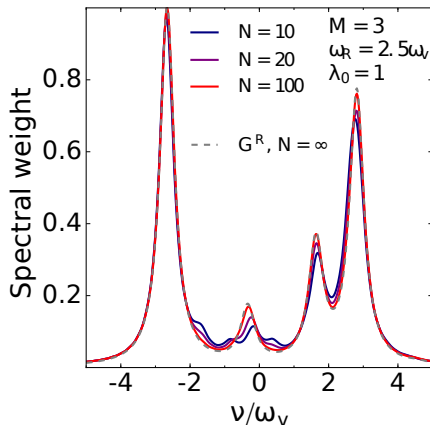
$$D^R(t) = -i \langle 0 | [\hat{a}(t), \hat{a}^\dagger(0)] | 0 \rangle \theta(t)$$

- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger |0\rangle$
- Fourier transform
- Mean-field Green's function

$$D^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_P + \Sigma_X(\nu)}$$

$$\sigma_X = - \sum_m \frac{\omega_R^2 |f_m(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_m}$$

(Classical expression)



Why? Multiple excitation $\sim 1/N$.

Tavis-Cummings-Holstein spectrum

- Direct calculation

$$D^R(t) = -i \langle 0 | \left[\hat{a}(t), \hat{a}^\dagger(0) \right] | 0 \rangle \theta(t)$$

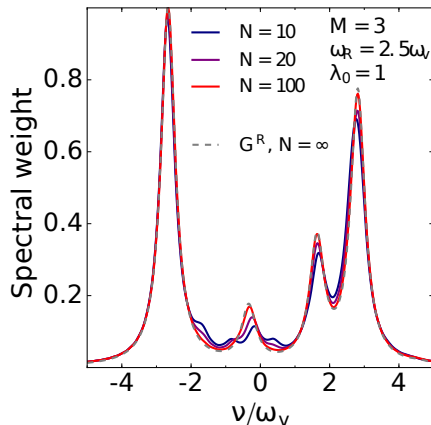
- Time-evolve $|\psi_0\rangle = \hat{a}^\dagger |0\rangle$
- Fourier transform
- Mean-field Green's function

$$D^R(\nu) = \frac{1}{\nu + i\kappa/2 - \omega_P + \Sigma_X(\nu)}$$

$$\sigma_X = - \sum_m \frac{\omega_R^2 |f_m(\lambda_0)|^2}{\nu + i\gamma/2 - \omega_m}$$

(Classical expression)

- Why? Multiple excitation $\sim 1/N$,

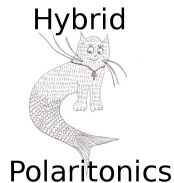


Acknowledgements

GROUP:

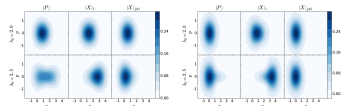


FUNDING:

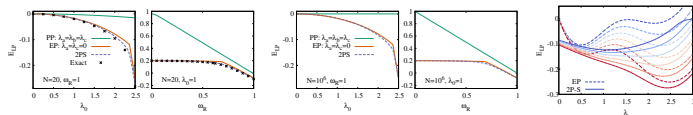


Summary

- Holstein-Dicke and Holstein-Tavis-Cummings models
- Single polariton state
 - ▶ Exact solution



- ▶ Polaron ansatz



[Zeb, Kirton, JK, arXiv:1608.08929]

- Validity of mean-field Green's functions

