The weak to strong light-matter coupling crossover with organic polaritons

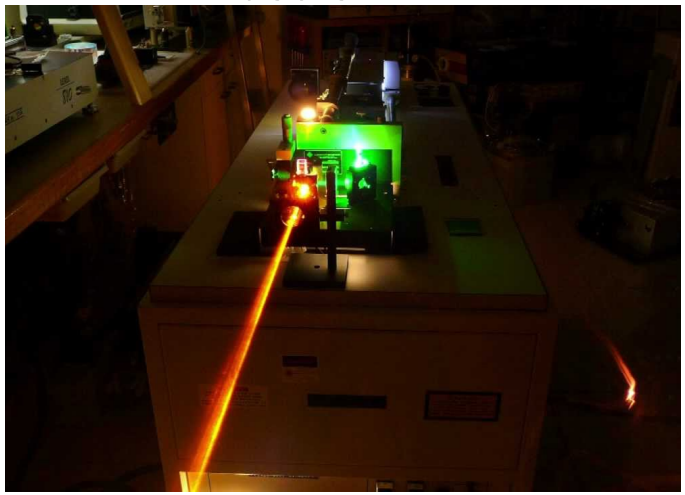
Peter Kirton

TU Wien, Austria

Molecular Polaritonics, July 2019

Generating coherent light

Lasers $T \sim ?$



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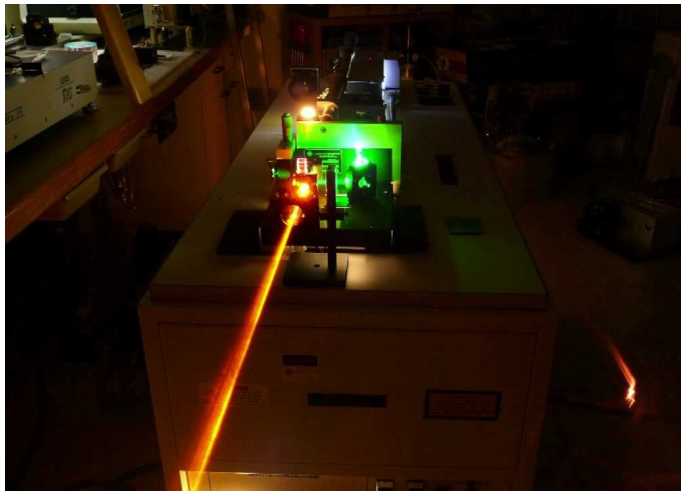
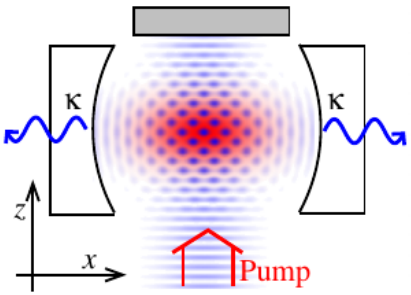
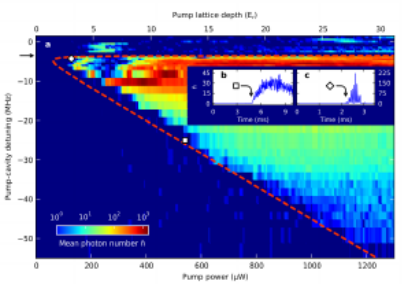


Superradiance $T \sim 0$





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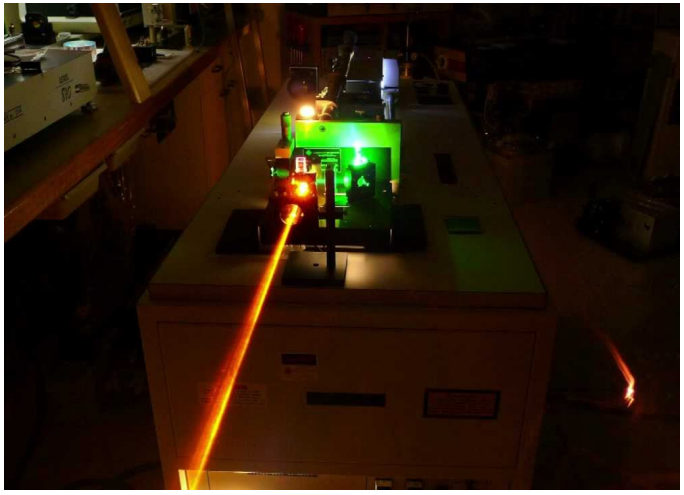
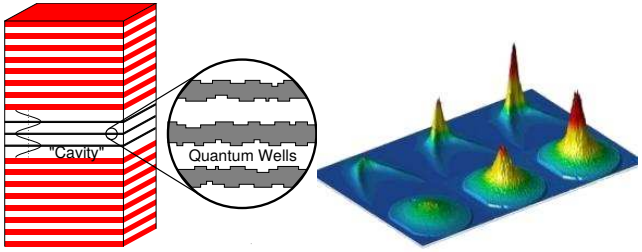


Microcavity polaritons $T \sim 20K$



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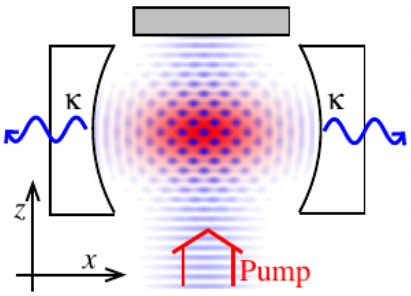

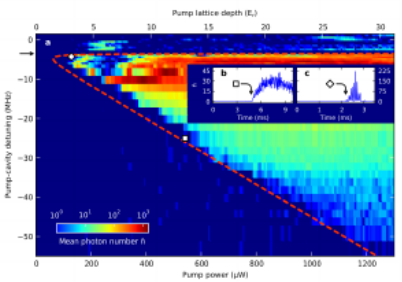

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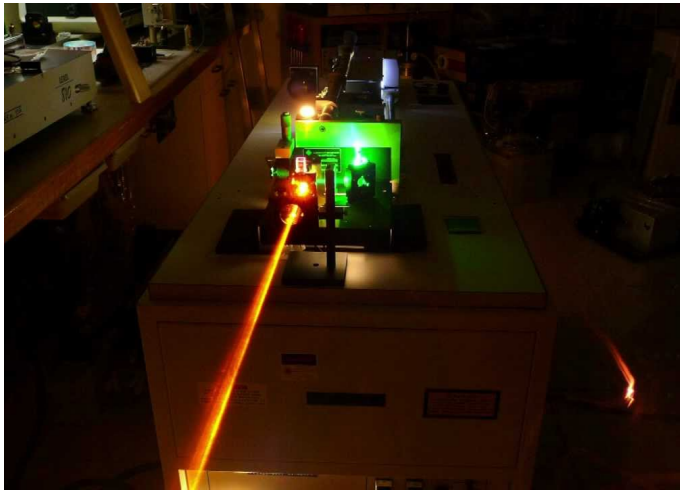

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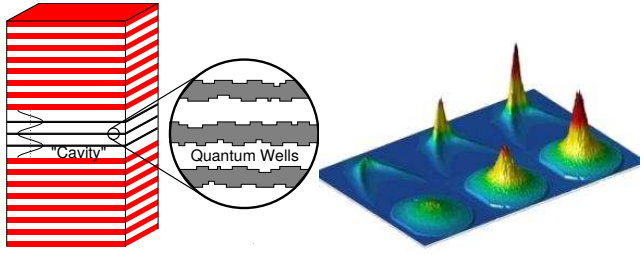
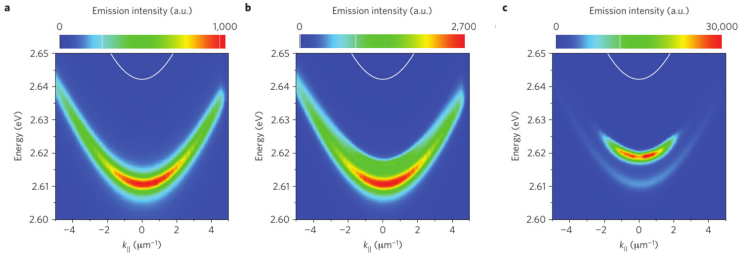
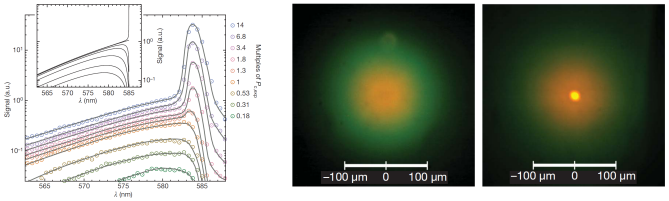


Organic polaritons, photons $T \sim 300K$





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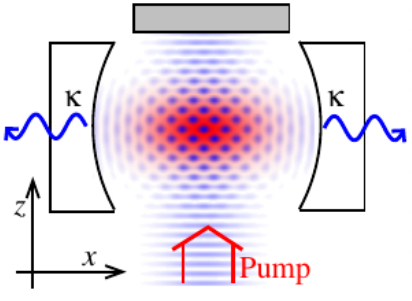
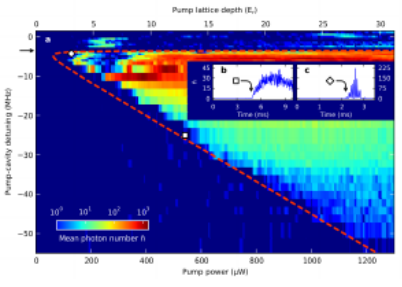


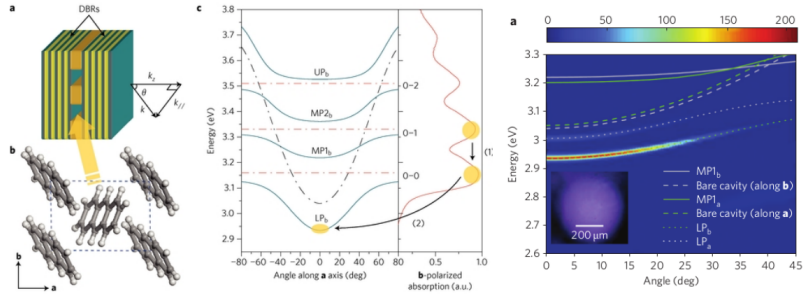


Outline

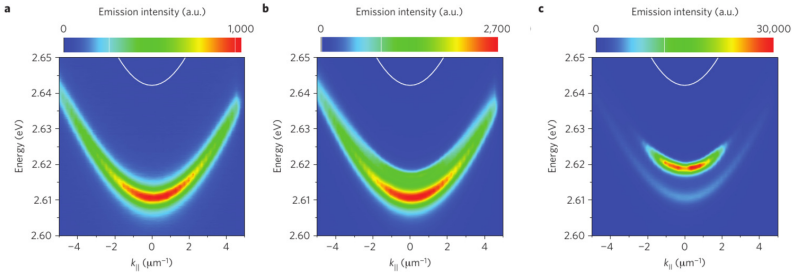
Physics of Organic Polaritons

What?

- Anthracene polariton lasing [Kena-Cohen and Forrest, Nat. Photon '10]



- BEC in polymers [Plumhoff et al. Nat. Mater. '14, Daskalakis et al. Nat. Mater. '14]



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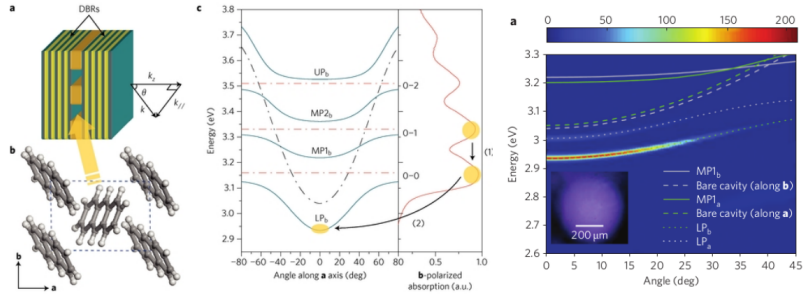

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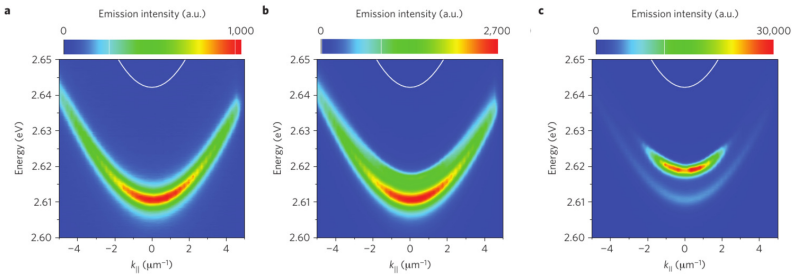

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Why?

- Ultastrong coupling: 1eV at $T = 300$K
- Low threshold lasers?

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- Top down
  - GPE/kinetic equations (assumes strong coupling)
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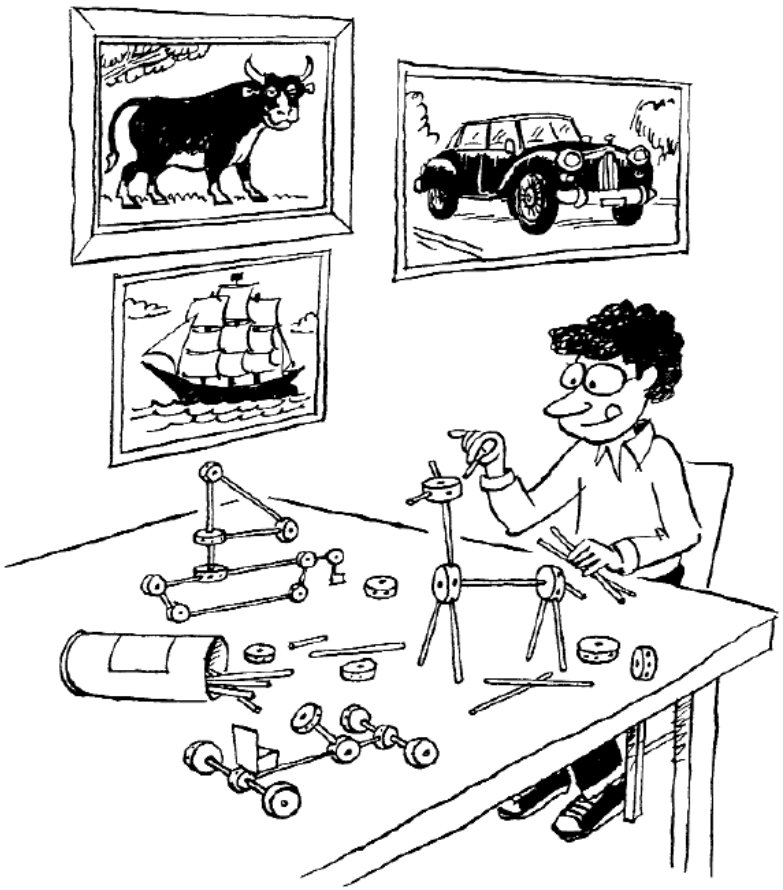

Illustration by Dick Codor.
[Auerbach, Interacting Electrons (Springer, 1998)]

- Top down
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  - Laser rate equations (assumes weak coupling)
- Bottom up
  - DFT/quantum chemistry
  - BUT only possible to treat small number of molecules
- Instead - Physically motivated microscopic models

Dicke-Holstein Model

$$H = \epsilon\sigma_n^z + \omega_v \left[b_n^\dagger b_n + \sqrt{S}\sigma_n^z (b + b^\dagger) \right]$$

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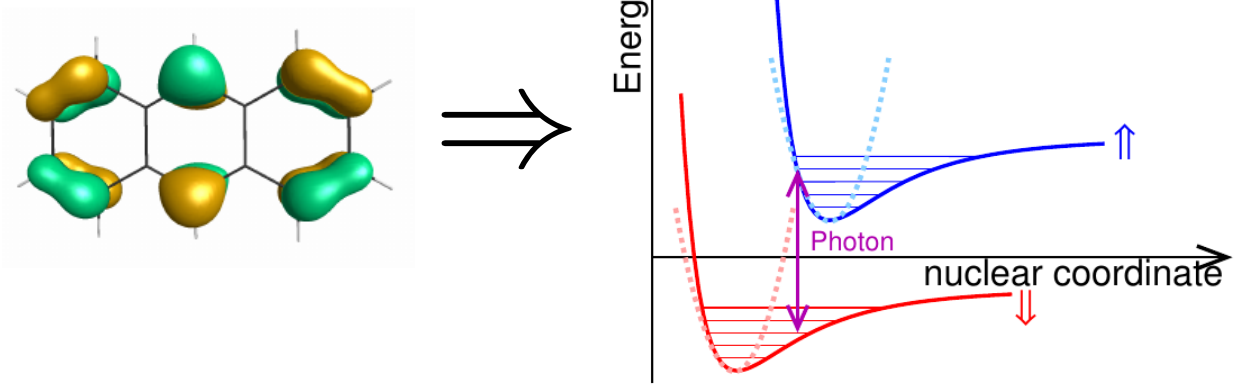

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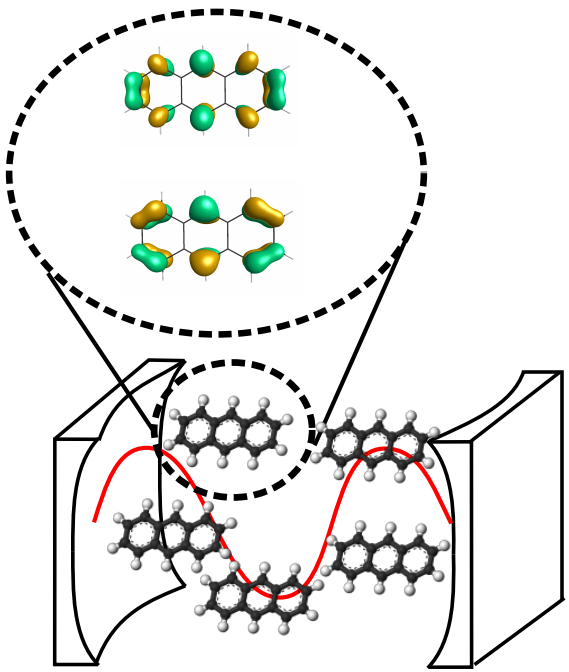
- Include many molecules and couple to a cavity



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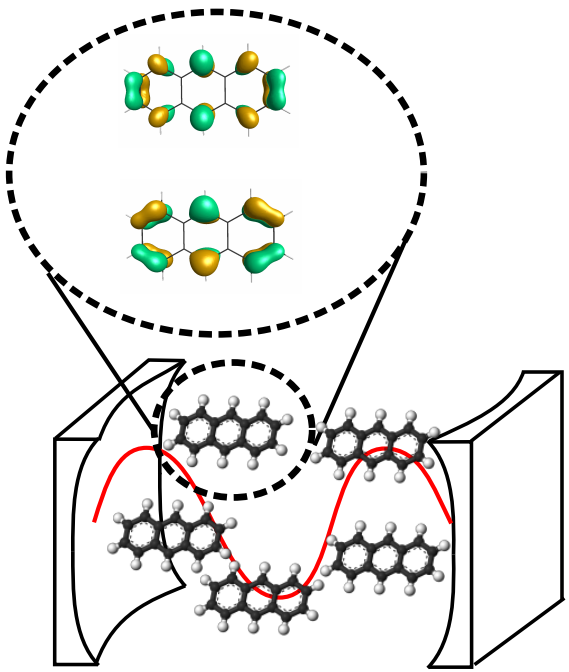

- Equilibrium: [Cwik et al. EPL 14, PRA 16; Spano JCP 15; Galego et al. PRX 15; Wu et al. PRB 16; Herrera & Spano PRL, PRA 17; Zeb et al. ACS Phot. 18 . . .]

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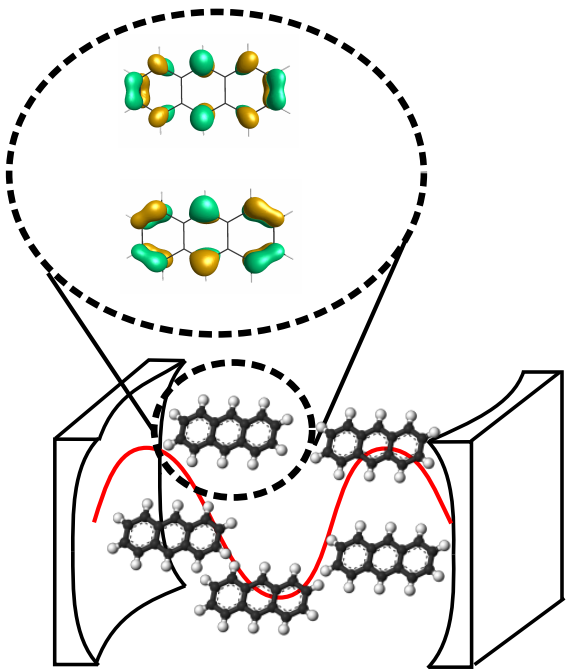

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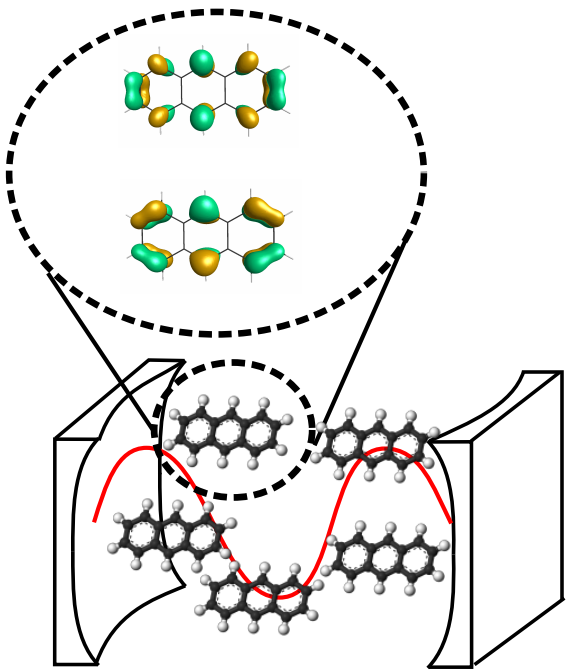

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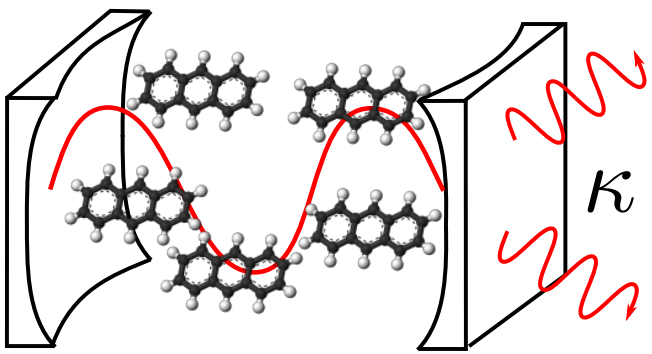

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$$\dot{\rho} = -i[H, \rho] + \kappa \mathcal{L}[a]$$

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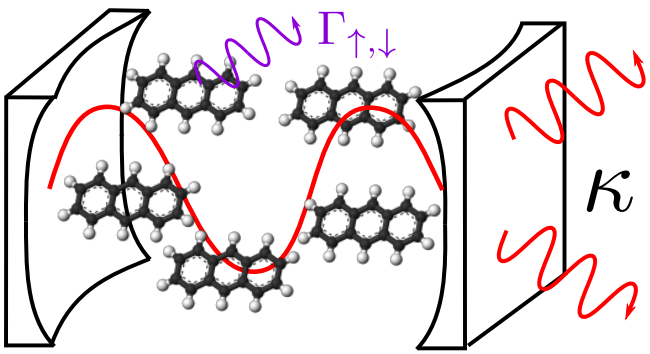

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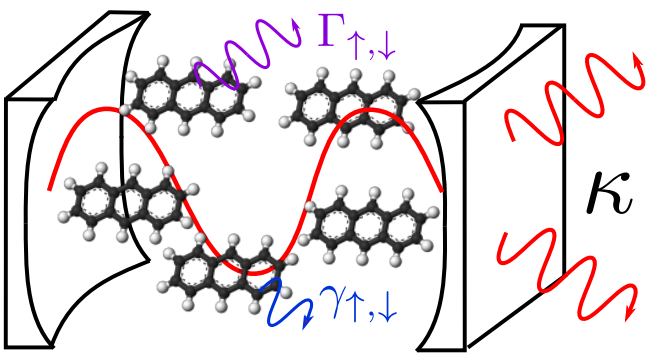

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- Describe combined electronic and vibrational state by Gell-Mann matrices λ_i
- Set of equations for $\alpha = \langle a \rangle$ and $l_i = \langle \lambda_i \rangle$

$$\partial_t \alpha = -\left(i\omega_c + \frac{\kappa}{2}\right)\alpha - 4i\frac{g^2 \mathcal{N}_m}{\varepsilon}\mathrm{Re}[\alpha] - i\mathcal{N}_m B_i \ell_i$$

$$\partial_t \ell_i = \left(\xi_{ik} + 4f_{ijk}B_j \mathrm{Re}[\alpha]\right)\ell_k + \frac{4i}{N}c_j^{\mu} c_k^{\mu *} f_{ijk}$$

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- Look at stability of $\alpha = 0$ solution: Lasing phase diagram

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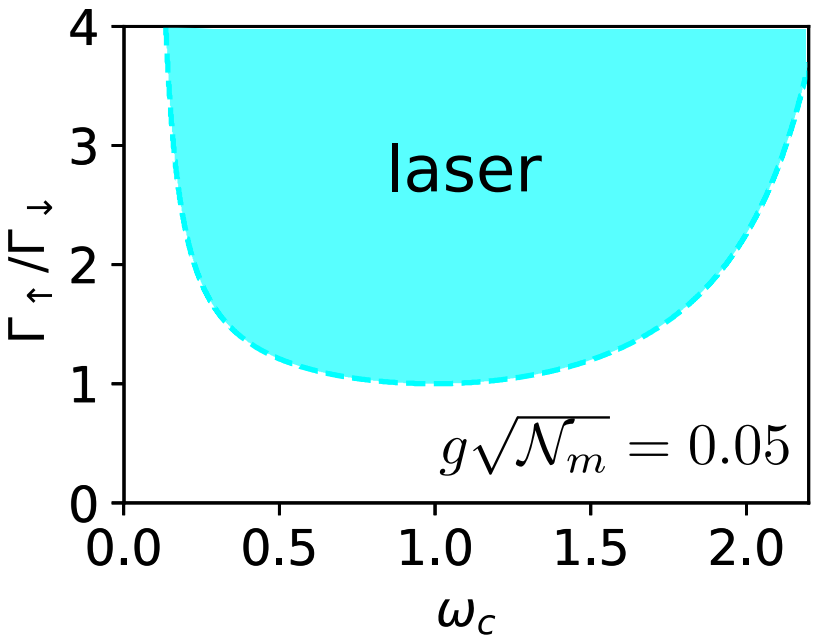

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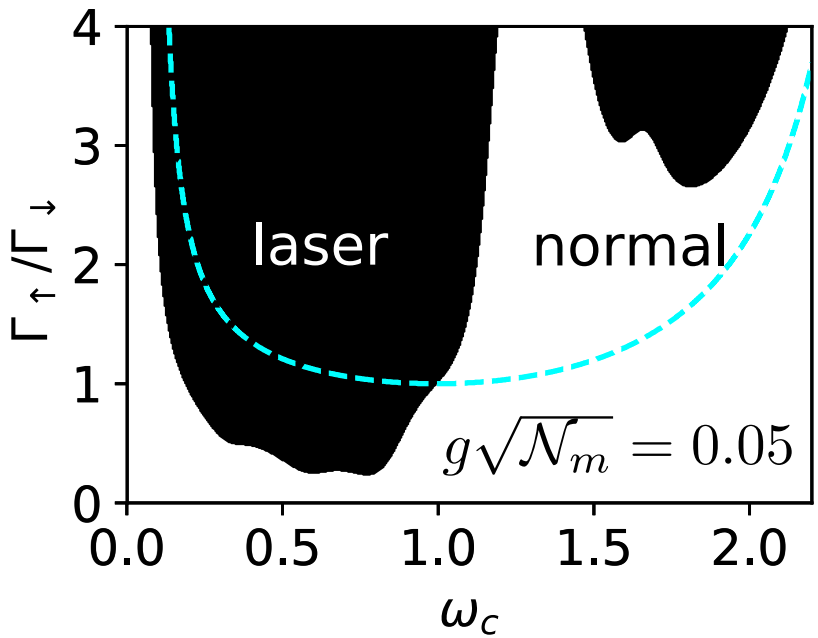

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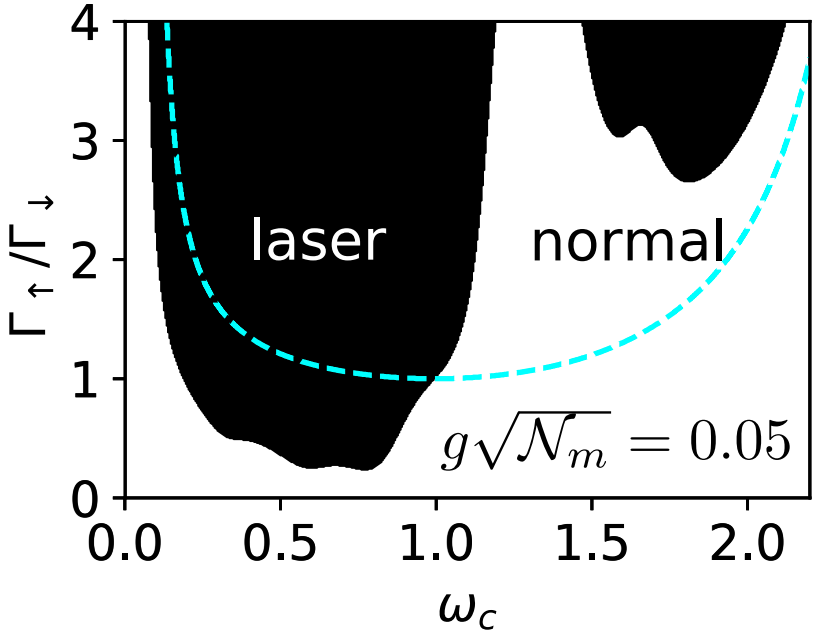
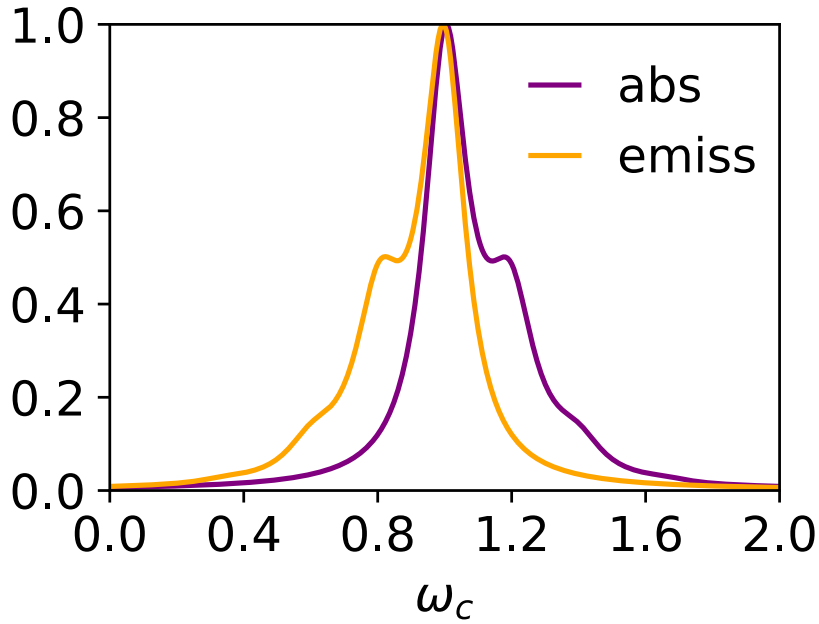
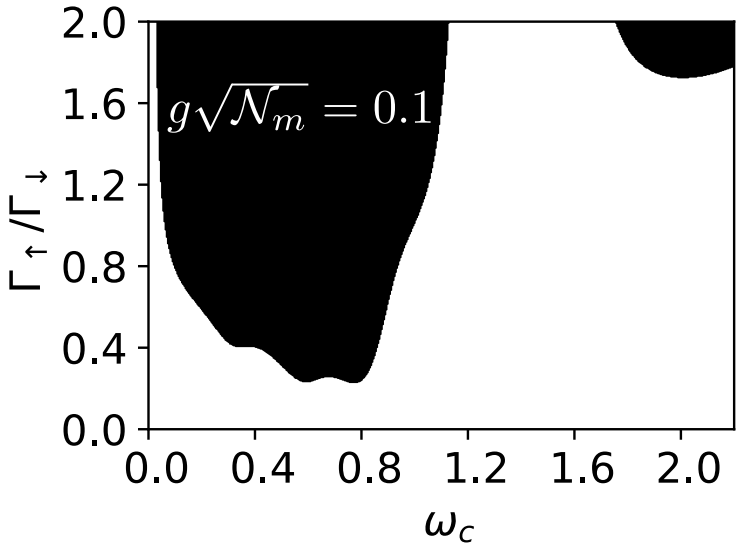


- Understand from absorption/emission



Phase Diagram - Stronger Coupling



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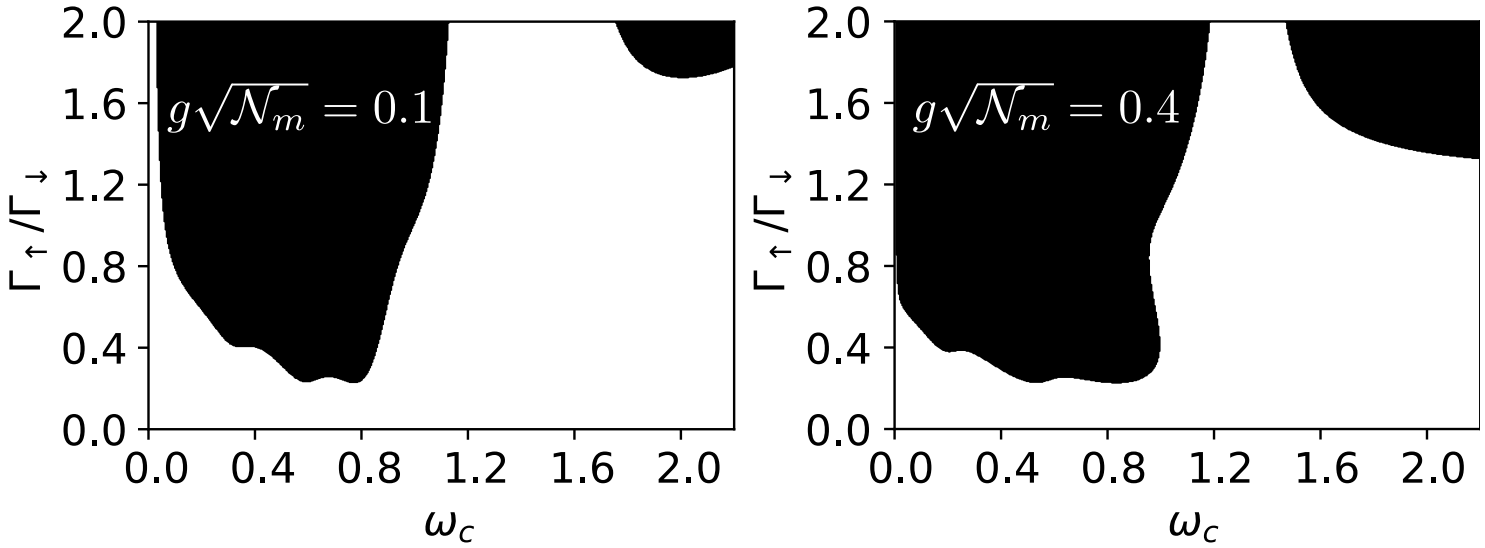

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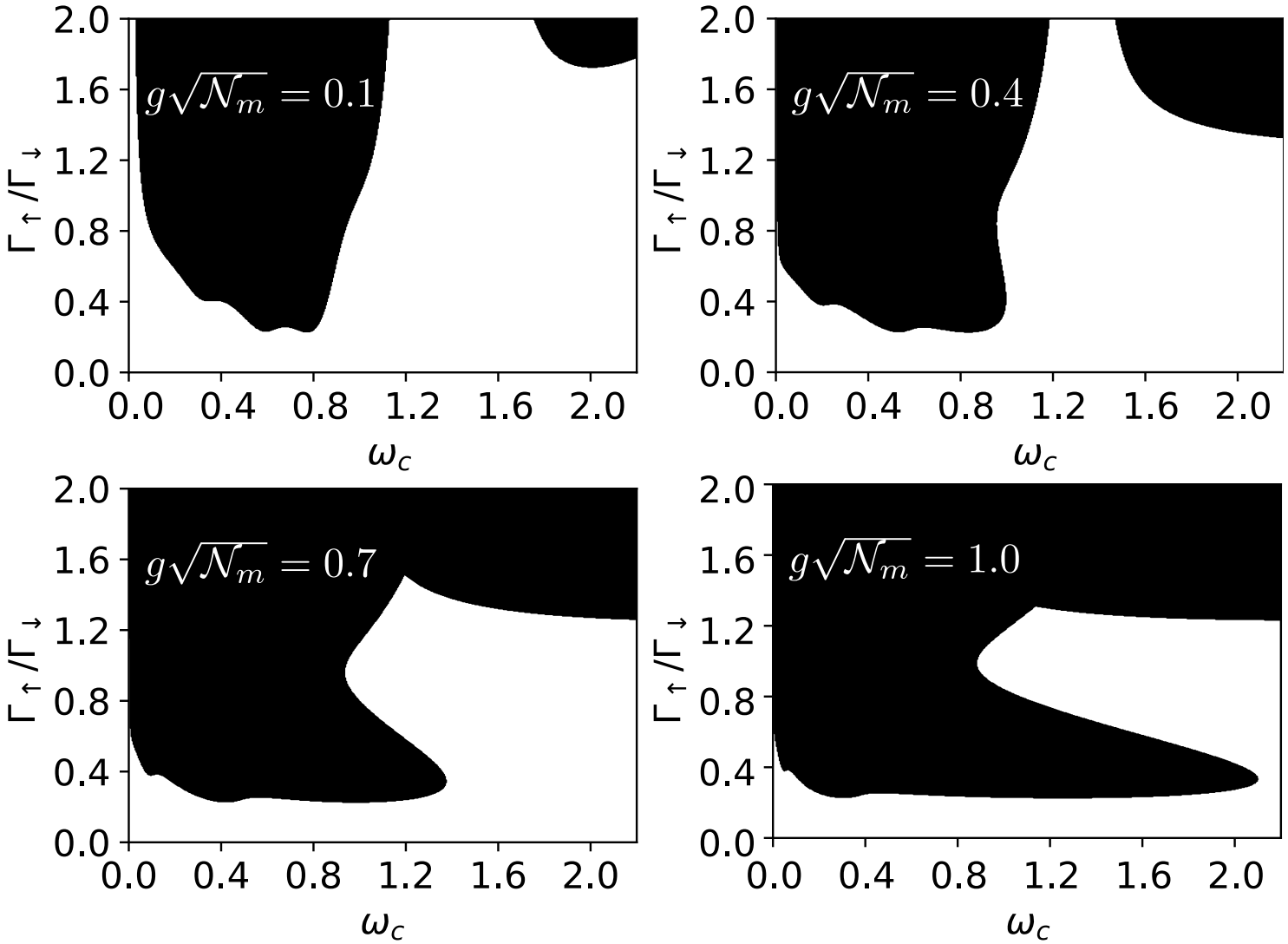

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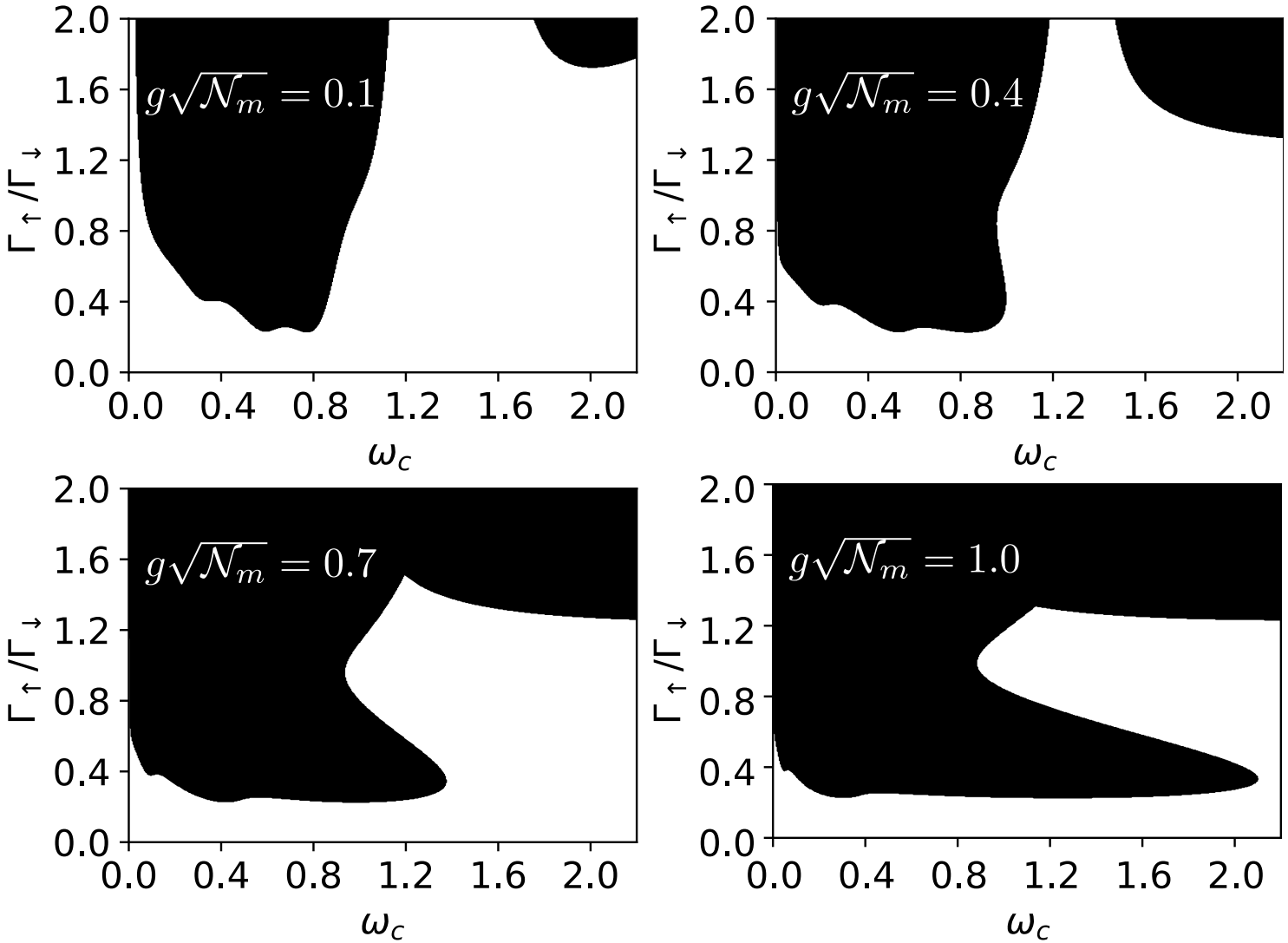
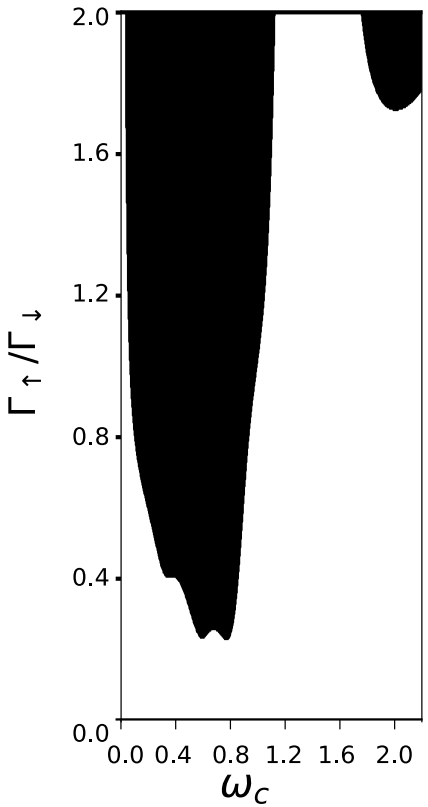


- What is the physics behind the re-entrant phase diagram?

Spectrum - weak coupling

$$g = 0.2$$



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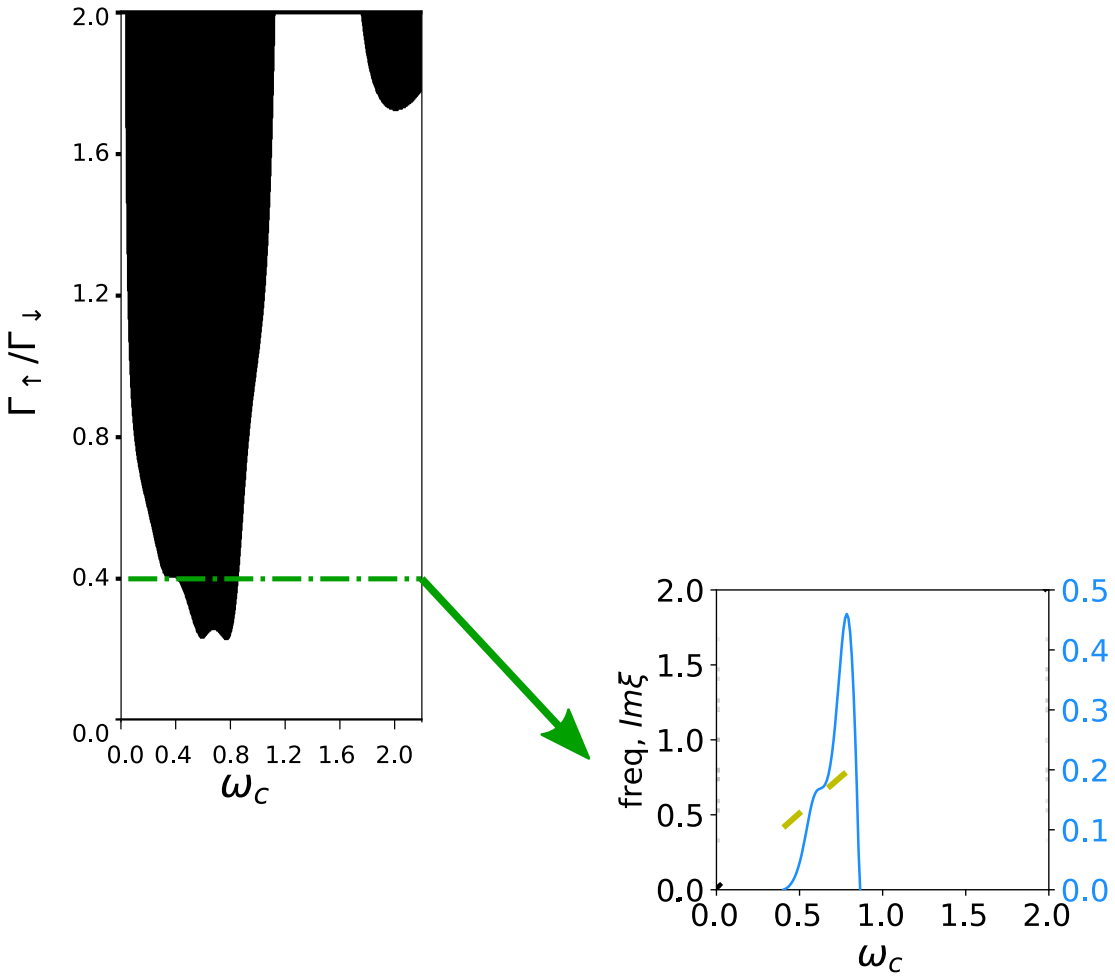

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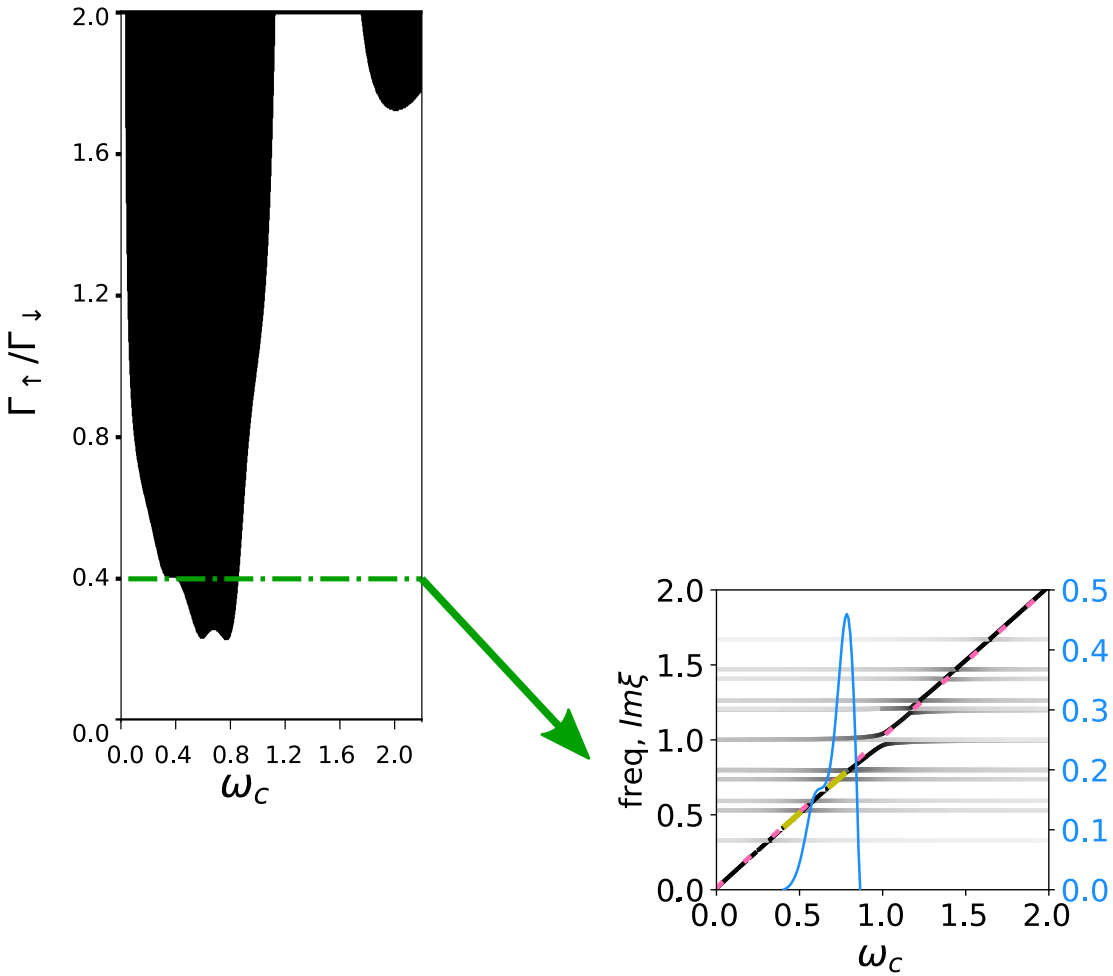

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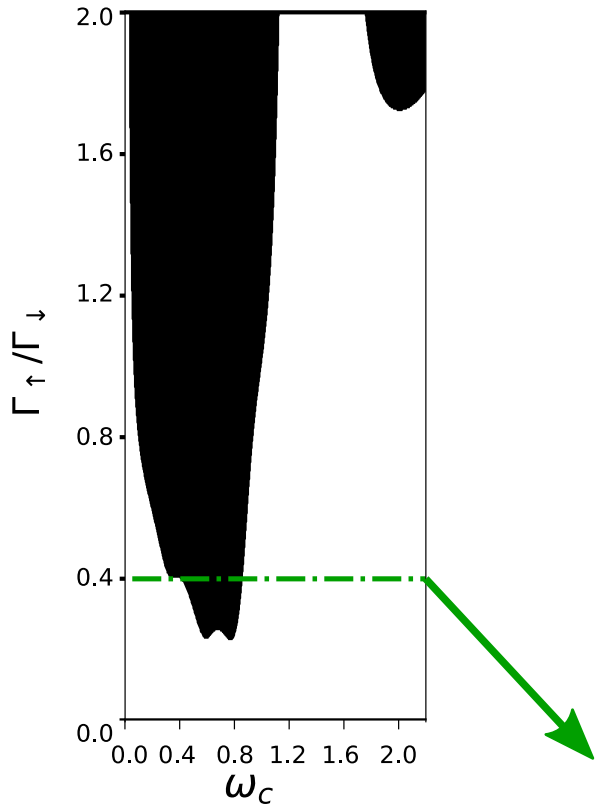

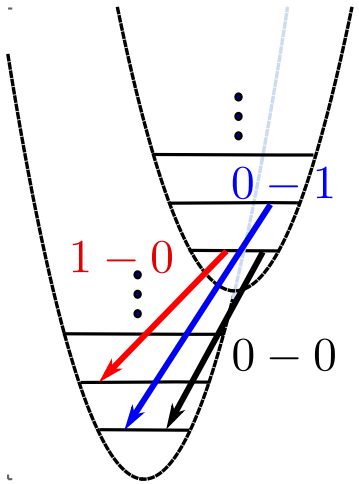

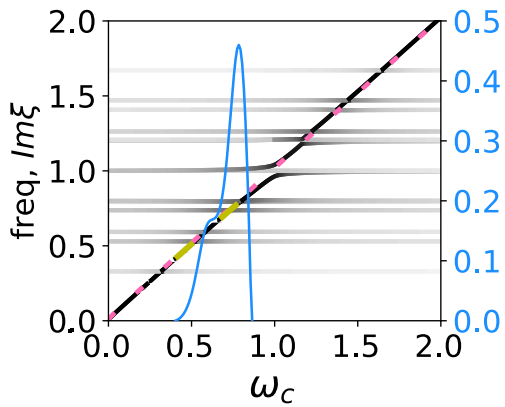

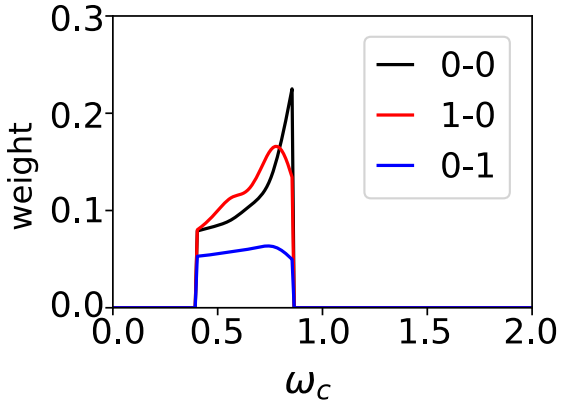

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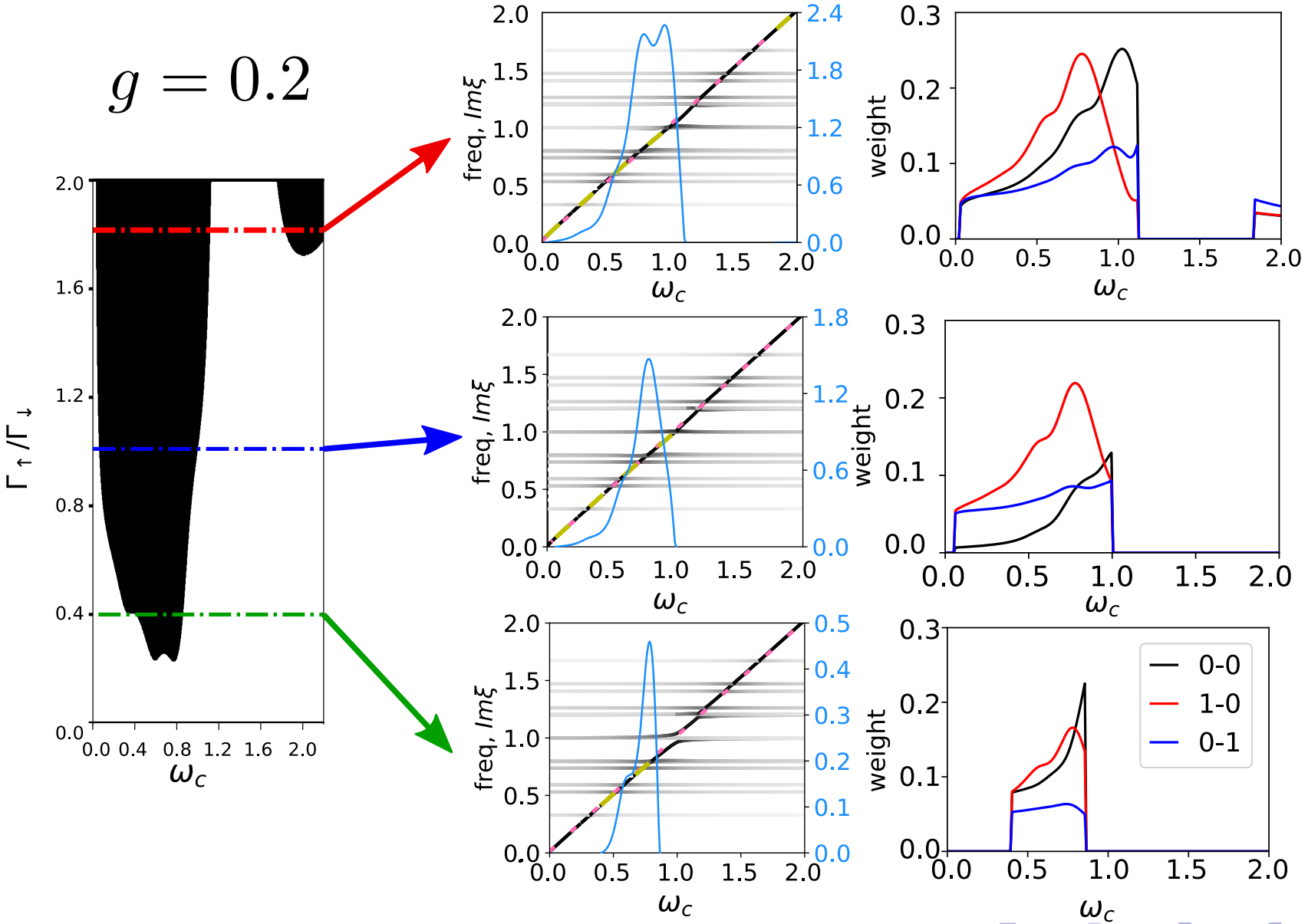
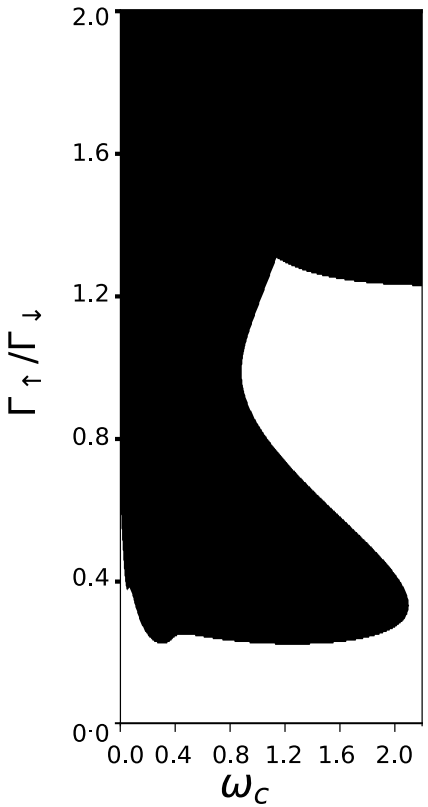


Spectrum - strong coupling

$$g = 1.0$$



Spectrum - strong coupling

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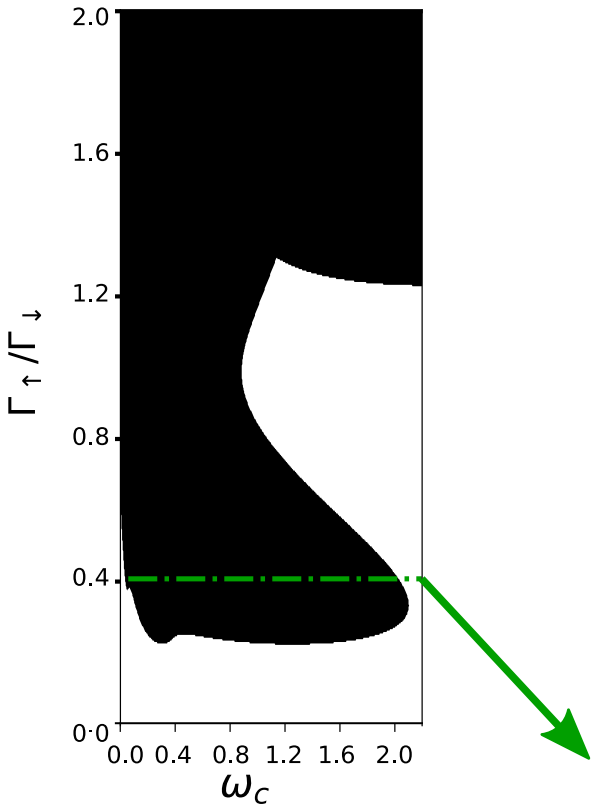

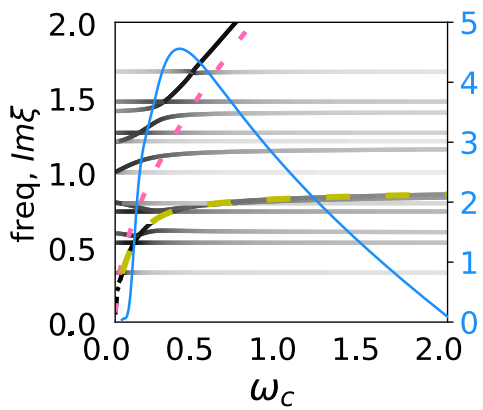

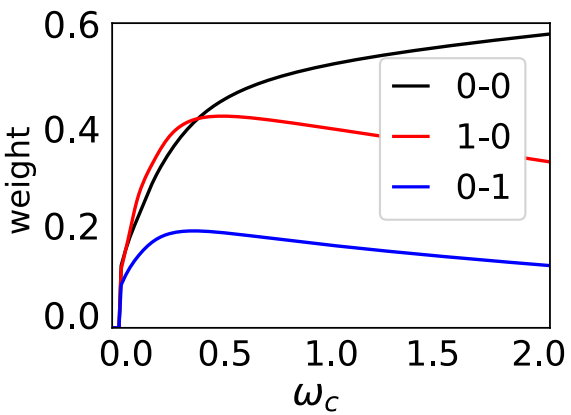

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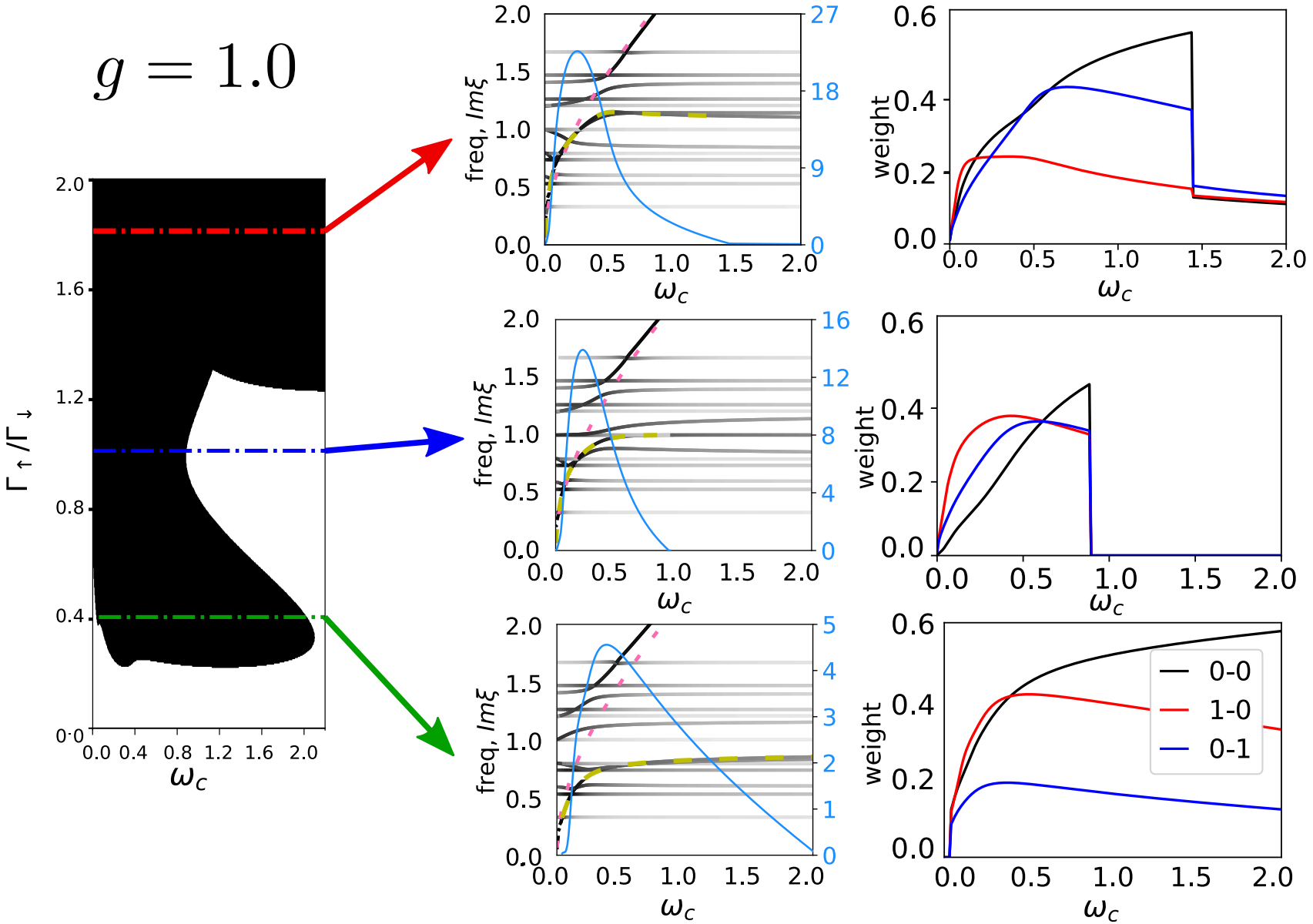

Optimising lasing

- Can strong coupling help with low threshold lasing?

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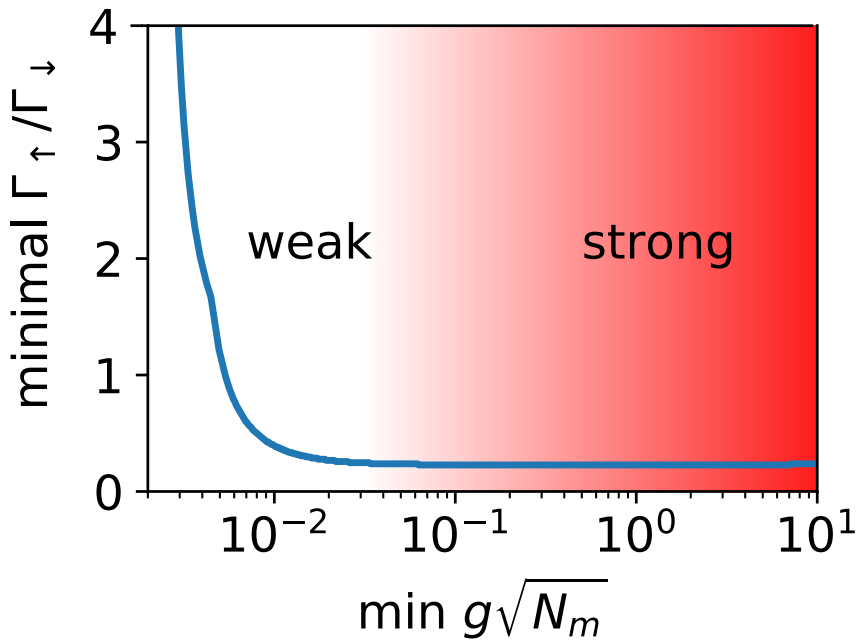
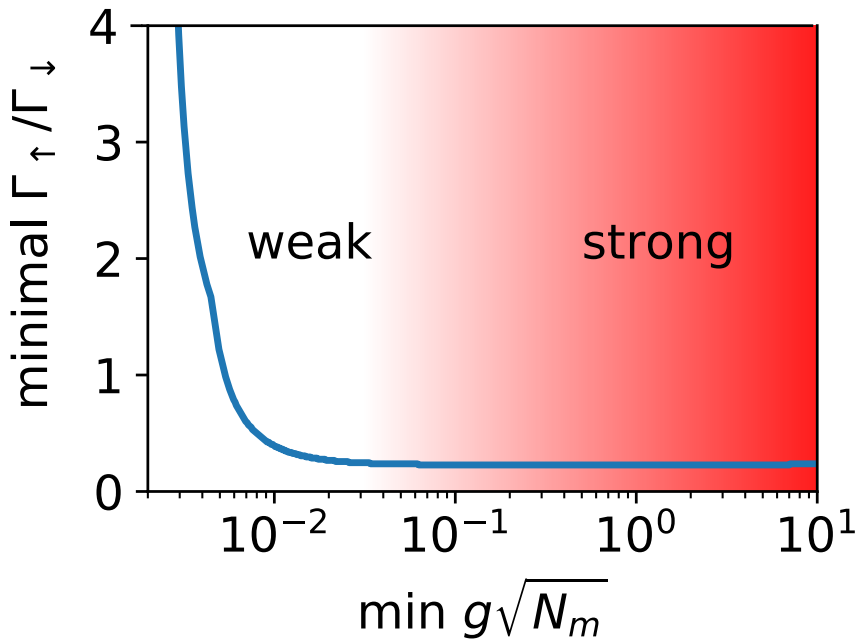


Optimising lasing

- Can strong coupling help with low threshold lasing?

- Optimise over frequency and find lowest threshold
- No significant g dependance
- BUT low threshold over wider frequency range



And now for something completely different....

TEMPO

- Time-Evolving Matrix Product Operators
- New method for solving dynamics of few-level quantum systems strongly coupled to environment

[Strathearn *et al.* Nat. Commun. (2018)]

TEMPO

- Time-Evolving Matrix Product Operators
- New method for solving dynamics of few-level quantum systems strongly coupled to environment

$$H = H_S + S\sum_i (g_i a_i + g_i^* a_i^\dagger) + \sum_i \omega_i a_i^\dagger a_i$$

- Specify: Spectral function $J(\omega) = \sum_i |g_i|^2 \delta(\omega - \omega_i)$, H_S and S

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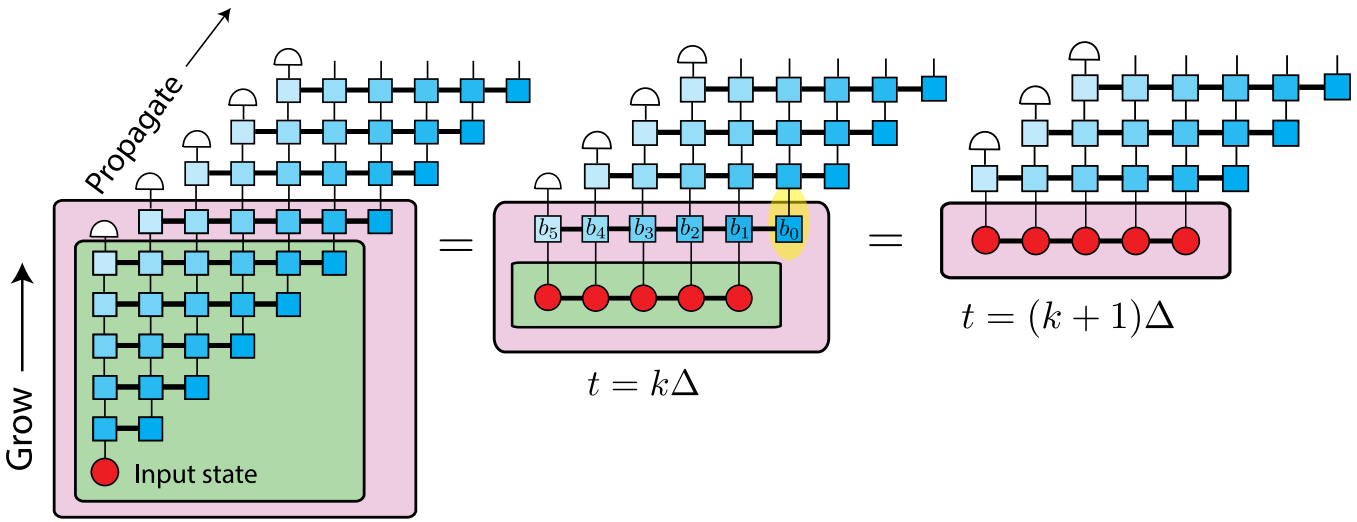
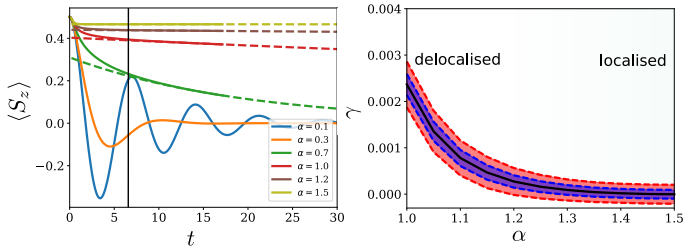
- Specify: Spectral function $J(\omega) = \sum_i |g_i|^2 \delta(\omega - \omega_i)$, H_S and S
- Evolve tensor network to find system dynamics



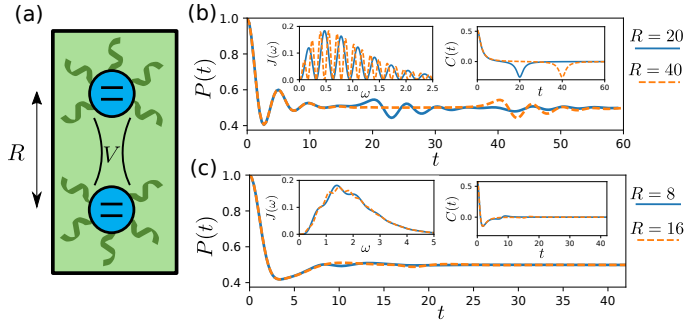
[Strathearn *et al.* Nat. Commun. (2018)]

Examples

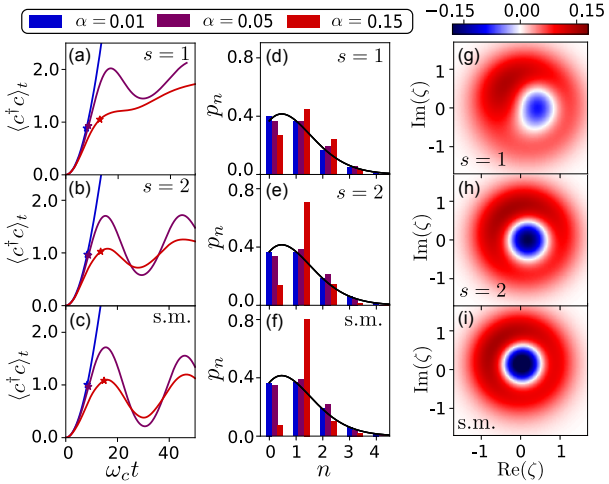
- Localisation phase transition in the spin boson model



- Spatially separated spins

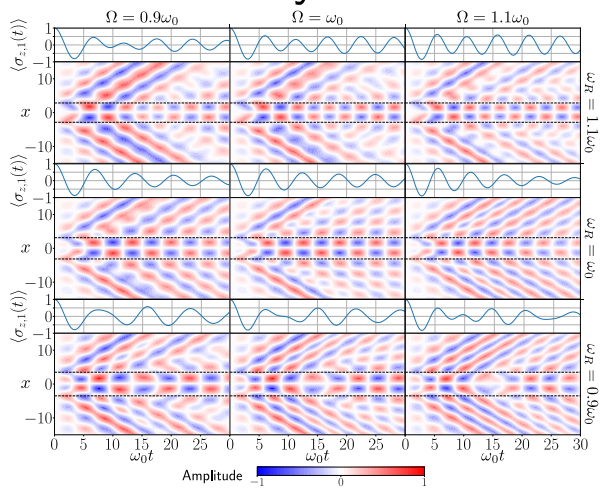


- Nonlinearities in optomechanics



[Minoguchi et al. arXiv1904.02164]

- Bath dynamics



[Gribben et al. arXiv:1907.02932]

Possible applications for polaritons?

Acknowledgements

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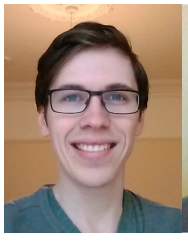
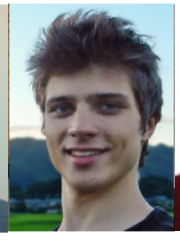
University of St Andrews





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Yuri Minoguchi, Peter Rabl

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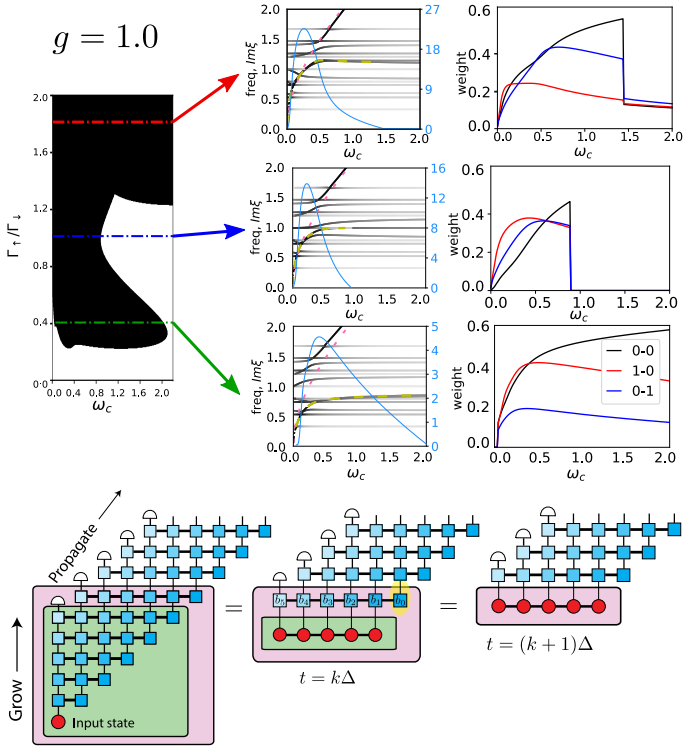
Hybrid Polaritonics

EPSRC Engineering and Physical Sciences Research Council

Conclusions

- The Dicke-Holstein model provides a relatively simple model of organic polaritons
- Complex physics emerges: competition between electronic and vibrational transitions, Hamiltonian and dissipative terms
- TEMPO provides efficient and accurate simulations of non-Markovian systems



Strashko, Kirton, Keeling PRL, **121**, *193601 (2018)*
Strathearn, Kirton, Kilda, Keeling, Lovett Nat. Commun. **9** *3322 (2018)*