Pairing Phases of Polaritons, and photon condensates

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600 YEARS

SUPA

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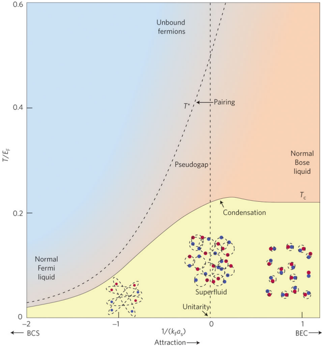
Outline

1. Pairing phases of polaritons
   - Pairing phases and Feshbach for polaritons
   - Phase diagram: Critical detunings
   - Signatures
   - Phase diagram: Critical temperatures
2. Photon condensation
   - Modelling organic molecules: Vibrational modes
   - Strong coupling?

Pairing phases of atoms

Fermions



From Randeria, Nat. Phys. '10

- BEC-BCS crossover

[Eagles, Leggett, Keldysh, Nozières, Randeria, . . .]

- BEC-BEC *transition*
- $\hat{H} = \ldots + \hat{\psi}_m^\dagger \hat{\psi}_{a_1} \hat{\psi}_{a_2} + \text{h.c.}$
  - If $\langle \hat{\psi}_m \rangle \neq 0$, MSF
  - If $\langle \hat{\psi}_{a_1} \rangle \neq 0$, $\langle \hat{\psi}_{a_2} \rangle \neq 0$, AMSF
- High density $\rightarrow$ metastability.

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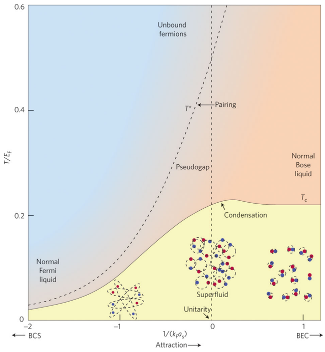

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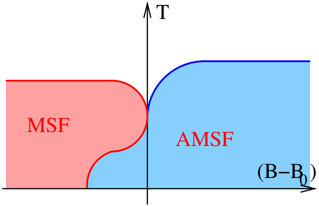

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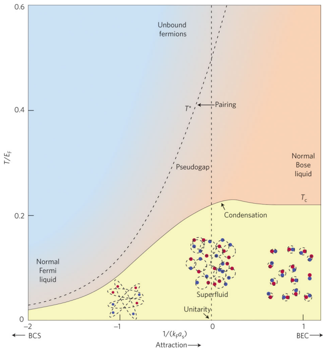

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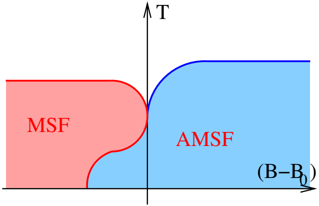

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- Hybridisation of bound states:
  - Biexciton: opposite spins (two-species): $2\omega_0^X - E_b$
  - Hybridisation with photons: $2\left[\frac{1}{2}(\omega_0^C + \omega_0^X) - \frac{1}{2}\sqrt{\Omega_r^2 + \delta^2}\right]$
- Control δ change ν, m, Interaction ...

[Ivanov, Haug, Keldysh '98], [Wouters '07], [Caursotto *et al.* '10], [Deveaud-Pledran *et al.* '13]

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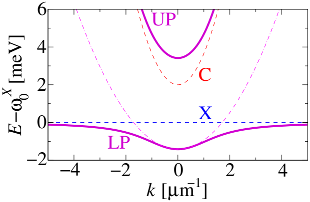

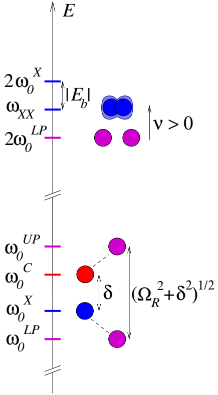

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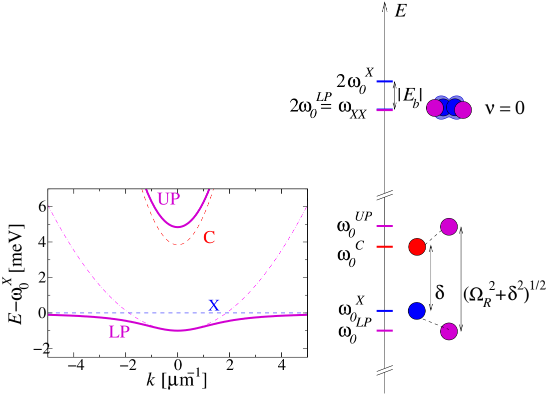

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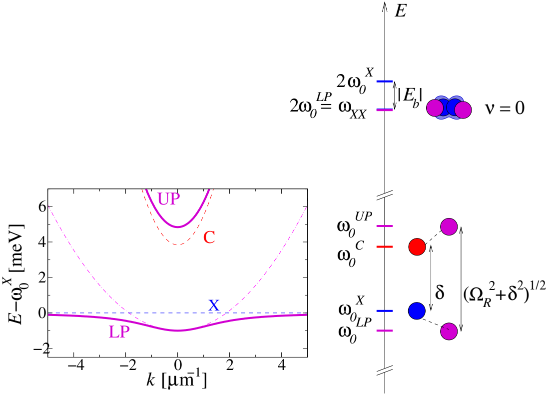

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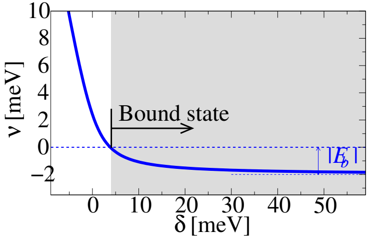

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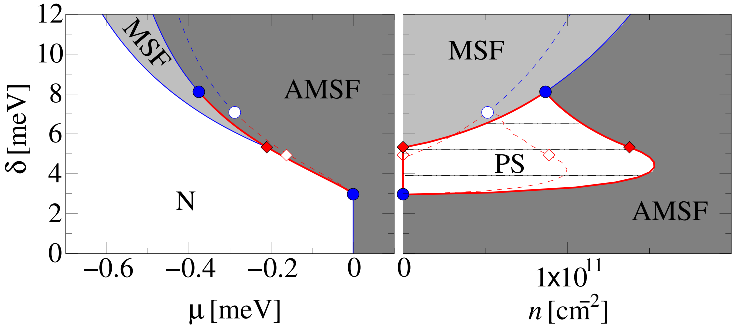

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- Small $|\delta|$: 1st order transition
  - Larkin-Pikin mechanism

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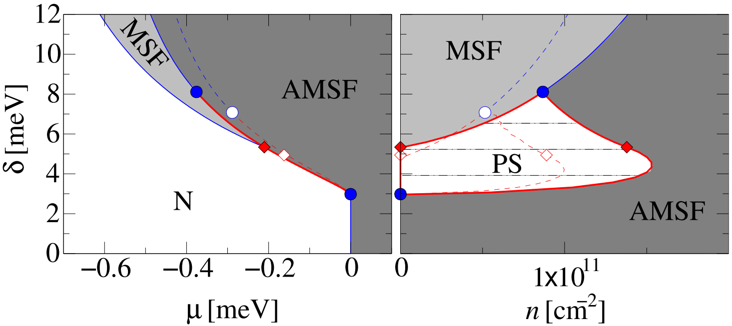

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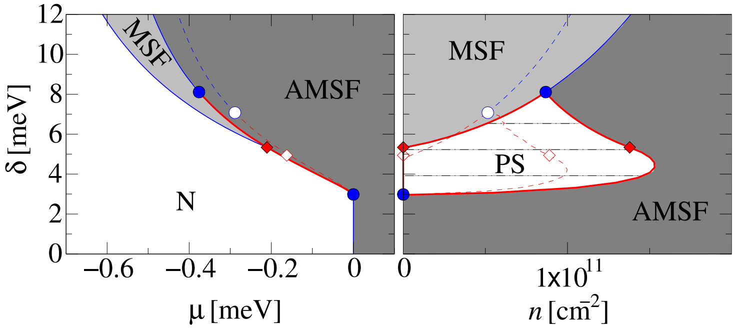

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Consequences and Signatures

- Phase separation
- Phase coherence
- Novel half vortices, $\psi_\uparrow = e^{im_\uparrow\theta}, \psi_\downarrow = e^{im_\downarrow\theta}$,
  MSF has $(m_\uparrow, m_\downarrow) = (1/2, 1/2)$

  Previous half-vortex $(m_\uparrow, m_\downarrow) = (1, 0)$ [Lagoudakis *et al.* Science '09]

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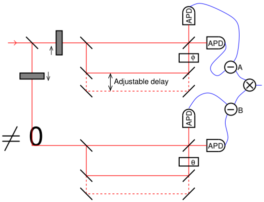

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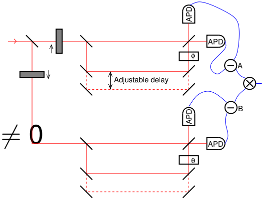

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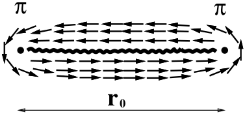

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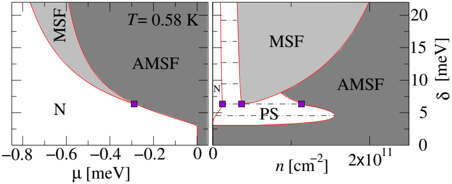

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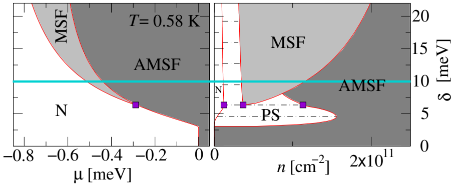

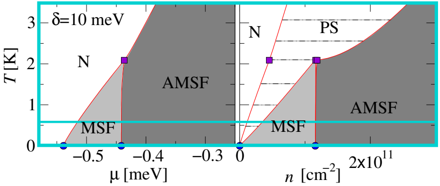

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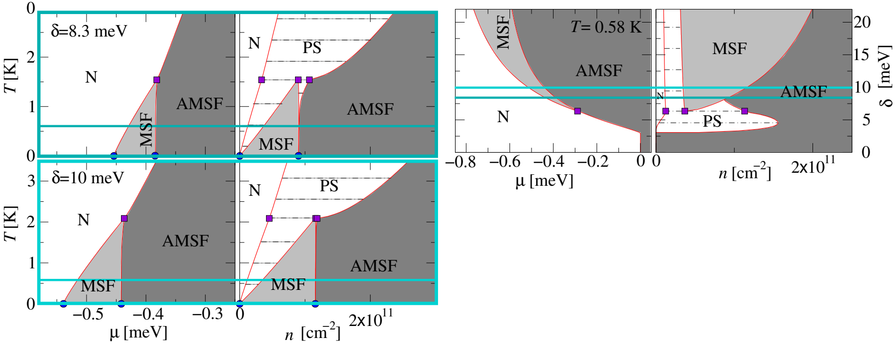

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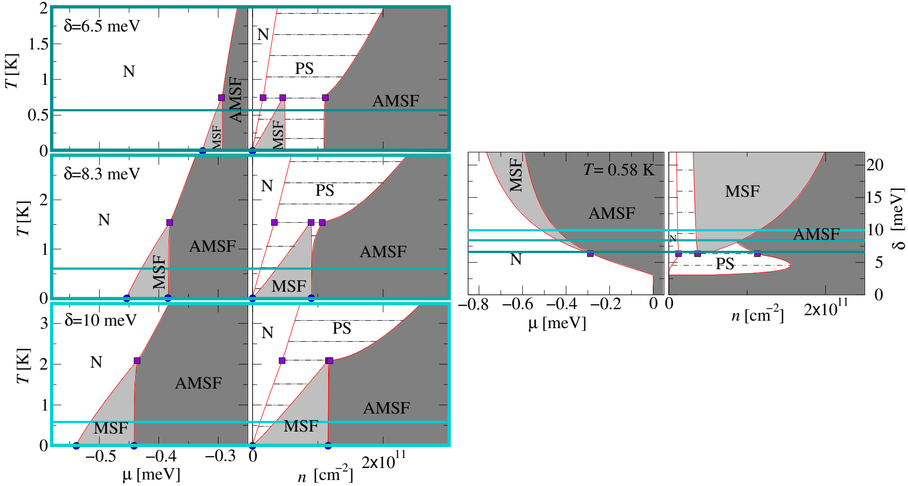

Evolution of triple point

- Exciton fraction $= \frac{1}{2}\left[1 + \frac{\delta}{\sqrt{\delta^2 + \Omega^2}}\right].$
- GaAs, need low T/high exciton fraction
- ZnO, easy to attain MSF

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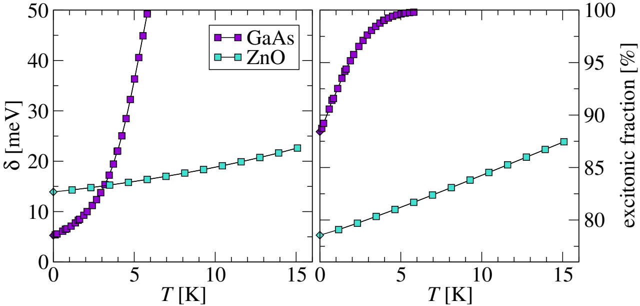

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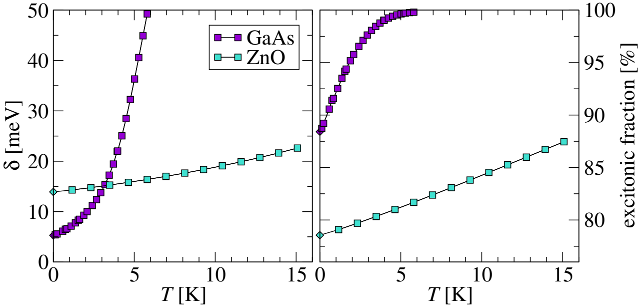

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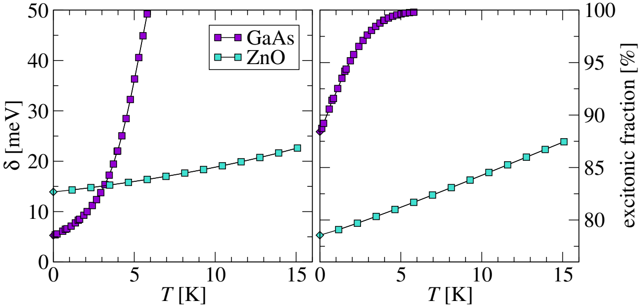

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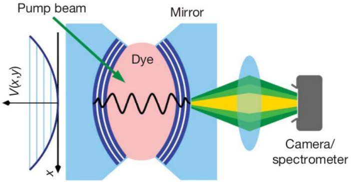

- Dye filled microcavity
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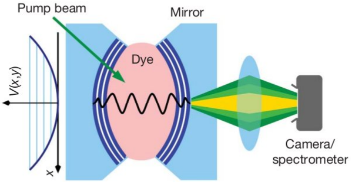

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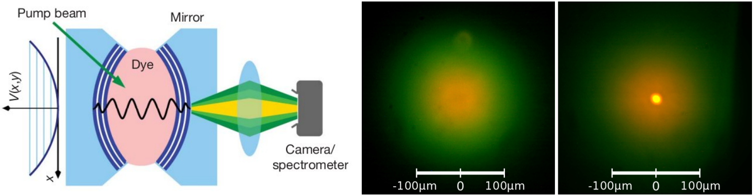

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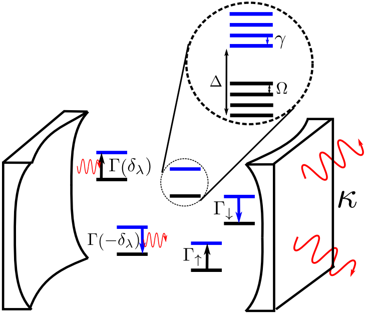

- No single cavity mode
  - Condensate mode is not maximum gain
  - Gain/Absorption in balance
- Thermalised many-mode system

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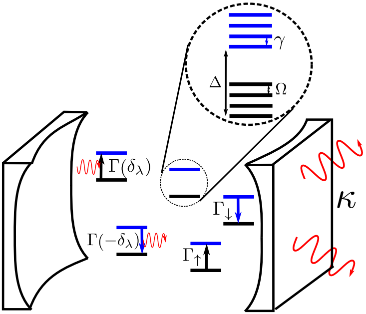

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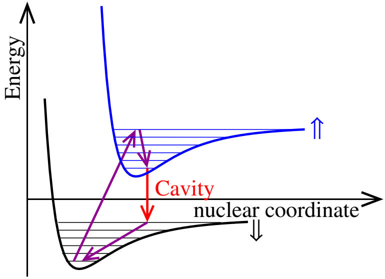

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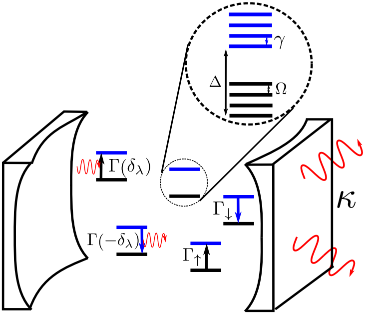

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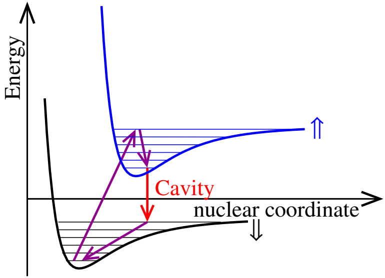

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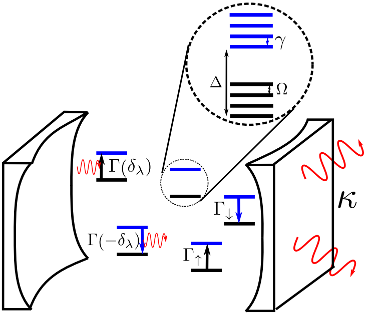

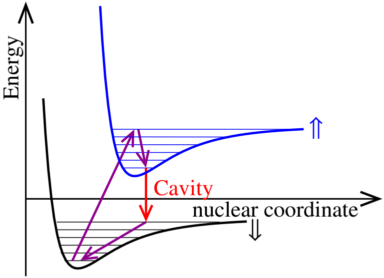

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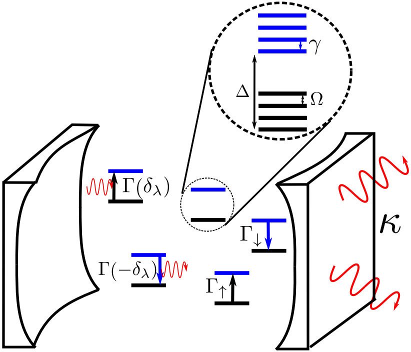
Modelling

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

- 2D harmonic cavity
  $\omega_m = \omega_{\text{cutoff}} + m\omega_{H.O.}$
  Degeneracies $g_m = m + 1$
- Local vibrational mode
  - Phonon frequency Ω
  - Huang-Rhys parameter S — phonon coupling



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$$\sum_\alpha + \Omega \left\{ b_\alpha^\dagger b_\alpha + \sqrt{S} \sigma_\alpha^z \left(b_\alpha^\dagger + b_\alpha \right\} \right)$$

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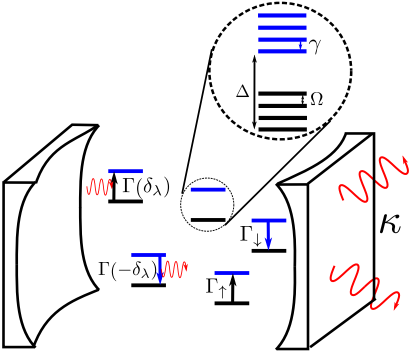
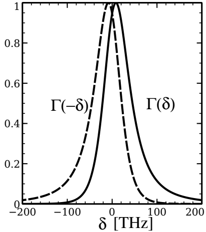


Modelling

Rate equation

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



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

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

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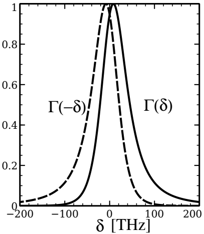
$$- \sum_{m,\alpha}\left[\frac{\Gamma(\delta_m = \omega_m - \epsilon)}{2}\mathcal{L}[\sigma_\alpha^+\psi_m] + \frac{\Gamma(-\delta_m = \epsilon - \omega_m)}{2}\mathcal{L}[\sigma_\alpha^-\psi_m^\dagger]\right]$$



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

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

Distribution $g_m n_m$

- Rate equation — include spontaneous emission
- Bose-Einstein distribution without losses

[Kirton & JK PRL '13]

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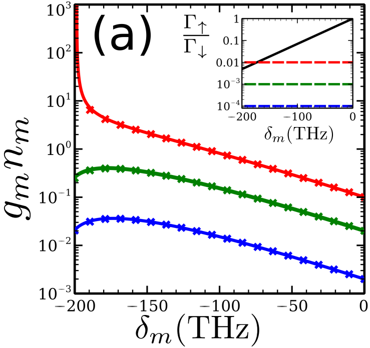

Low loss: Thermal

[Kirton & JK PRL '13]

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- Rate equation — include spontaneous emission
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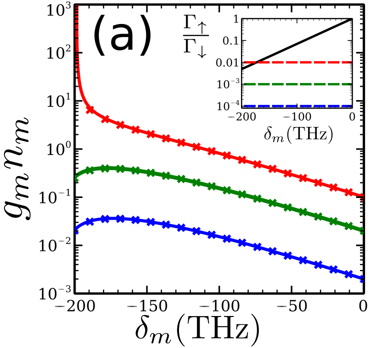

Low loss: Thermal

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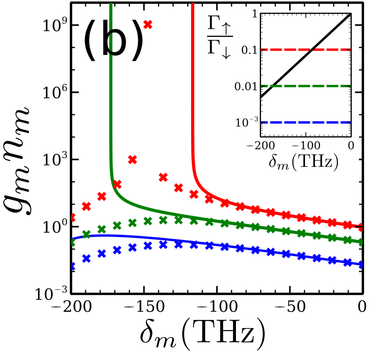
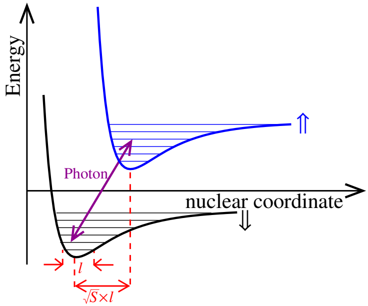


High loss → Laser

Outline

Strong coupling limit

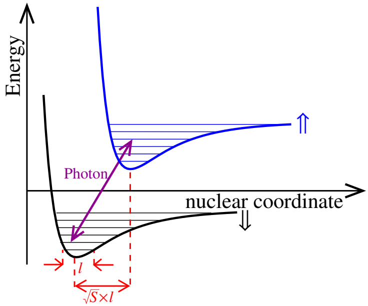


$$H = \omega\psi^\dagger\psi + \sum_\alpha \left[\frac{\epsilon}{2}\sigma^z_\alpha + g\left(\psi\sigma^+_\alpha + \psi^\dagger\sigma^-_\alpha\right)\right.$$
$$\left. + \Omega\left\{b^\dagger_\alpha b_\alpha + \sqrt{S}\left(b^\dagger_\alpha + b_\alpha\right)\sigma^z_\alpha\right\}\right]$$

- Phonon frequency Ω
- Huang-Rhys parameter S — phonon coupling

- Polaron formation (dressing by vibrational modes)
- Vibrational replicas and BEC
- Ultra-strong phonon coupling

Strong coupling limit



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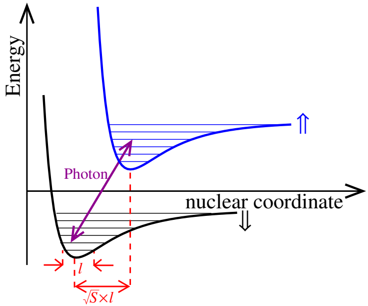

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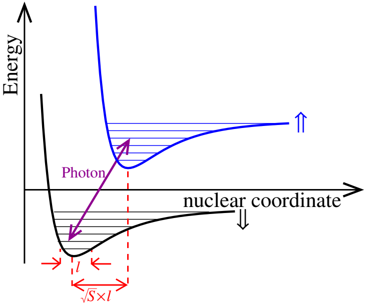

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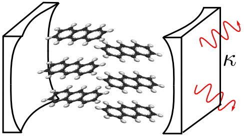
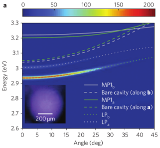
First attempt — equilibrium [Cwik *et al.* EPL '14]

Organic materials in microcavities

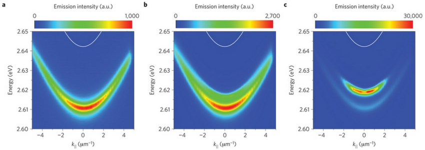
- Strong coupling with organic materials [Lidzey, Nature ’98]
- Polariton lasing





$\Omega_r \simeq 0.1$eV [Kena Cohen and Forrest, Nat. Photon 2010]

- Thermalisation, condensation interactions



$T = 300$K [Daskalakis *et al.* Nat. Photon 2014; Plumhoff *et al.* ibid.]

- Ultrastrong coupling regime

  $\Omega_r \simeq 0.6$eV! [Canaguier-Durand Ang. Chem. ’13, . . .]

Phase diagram

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

- S suppresses condensation — reduces overlap
- Reentrant behaviour — Min μ at $T \sim 0.2$

[Cwik *et al.* EPL '14]

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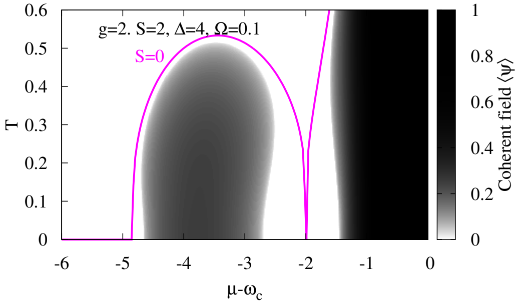

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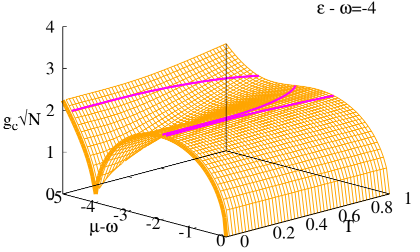
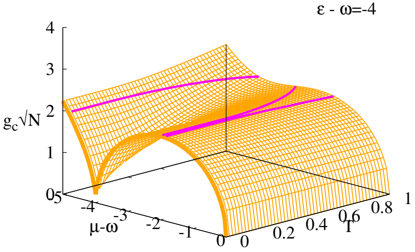
Critical coupling with increasing S

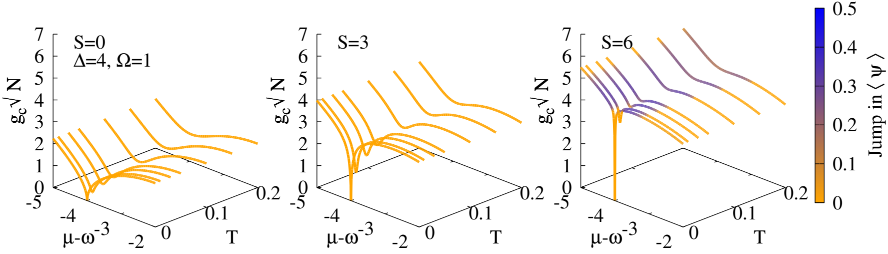
- Re-orient phase diagram
- *g* vs μ, *T*



Critical coupling with increasing S

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



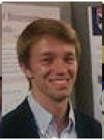
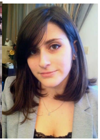


Acknowledgements

GROUP:





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Francesca Marchetti, UAM

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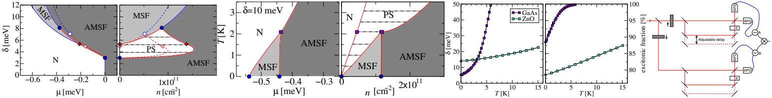
Topological Protection and Non-Equilibrium States in Strongly Correlated Electron Systems

EPSRC
Engineering and Physical Sciences Research Council

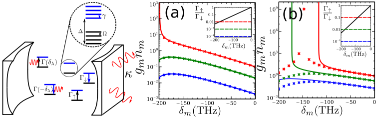
Summary

- Polaritons pairing phase feasible for ZnO, signatures in coherence and vortices



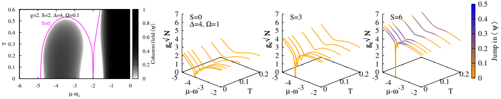
[Marchetti and Keeling, `arXiv:1308.1032`]

- Photon condensation and thermalisation; vibrational modes



[Kirton and Keeling, PRL '13]

- Vibrational modes and strong coupling



[Cwik, Reja, Littlewood, Keeling EPL '14]

3. Pairing phases model
   - Excitons and photons
   - Polaritons
   - Exciton spin
4. Calculation details
   - Variational wavefunction
   - Variational MFT
   - WIDBG result
5. More phase diagrams
6. Photon phase diagram
7. Organic polaritons
   - Polarons
   - Condensation of phonon replicas?
   - Anticrossing vs ρ

Exciton-photon model

- Microscopic model — coupled exciton-photon system

$$H = \sum_k \left[\sum_{\sigma=\pm2,\pm1} \left(\frac{k^2}{2m_X} - \mu \right) \hat{X}^\dagger_{k\sigma} \hat{X}_{k\sigma} + \sum_{\sigma=\pm1} \left(\delta + \frac{k^2}{2m_C} - \mu \right) \hat{C}^\dagger_{k\sigma} \hat{C}_{k\sigma} \right.$$

$$\left. + \sum_{\sigma=\pm1} \Omega_R \left(\hat{C}^\dagger_{k\sigma} \hat{X}_{k\sigma} + \hat{X}^\dagger_{k\sigma} \hat{C}_{k\sigma} \right) \right] + \iint d^2r d^2R \sum_{\sigma,\sigma',\tau,\tau'=\pm2,\pm1} U^{XX}_{\sigma'\tau'\tau\sigma}(\mathbf{r})$$

$$\times \hat{X}^\dagger_{\sigma'} \left(\mathbf{R} + \frac{\mathbf{r}}{2} \right) \hat{X}^\dagger_{\tau'} \left(\mathbf{R} - \frac{\mathbf{r}}{2} \right) \hat{X}_{\tau} \left(\mathbf{R} - \frac{\mathbf{r}}{2} \right) \hat{X}_{\sigma} \left(\mathbf{R} + \frac{\mathbf{r}}{2} \right)$$

- Interaction U^{XX} has exchange structure
- For large Ω_R, neglect $\sigma = \pm 2$
- Interaction supports bound states in $U^{XX}_{+1,-1,-1,+1}$ channel — *bipolariton*
- NB, bipolariton, bound polaritons, *but* larger exciton fraction.

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$$\left. + \sum_{\sigma=\pm1} \Omega_R \left(\hat{C}^\dagger_{k\sigma} \hat{X}_{k\sigma} + \hat{X}^\dagger_{k\sigma} \hat{C}_{k\sigma} \right) \right] + \iint d^2r d^2R \sum_{\sigma,\sigma',\tau,\tau'=\pm2,\pm1} U^{XX}_{\sigma'\tau'\tau\sigma}(\mathbf{r})$$
$$\times \hat{X}^\dagger_{\sigma'} \left(\mathbf{R} + \frac{\mathbf{r}}{2} \right) \hat{X}^\dagger_{\tau'} \left(\mathbf{R} - \frac{\mathbf{r}}{2} \right) \hat{X}_{\tau} \left(\mathbf{R} - \frac{\mathbf{r}}{2} \right) \hat{X}_{\sigma} \left(\mathbf{R} + \frac{\mathbf{r}}{2} \right)$$

- Interaction U^{XX} has exchange structure
- For large Ω_R, neglect $\sigma = \pm 2$
- Interaction supports bound states in $U^{XX}_{+1,-1,-1,+1}$ channel — *bipolariton*
- NB, bipolariton, bound polaritons, *but* larger exciton fraction.

Polariton model

$$H = \sum_k \left[\sum_{\sigma=\uparrow,\downarrow} \left(\frac{k^2}{2m} - \mu \right) \hat{\psi}^\dagger_{\sigma k} \hat{\psi}_{\sigma k} + \left(\frac{k^2}{2m_m} + \nu - 2\mu \right) \hat{\psi}^\dagger_{mk} \hat{\psi}_{mk} \right]$$

$$+ \int d^2 R \left[\sum_{\sigma=\uparrow,\downarrow,m} \frac{U_{\sigma\sigma}}{2} \hat{\psi}^\dagger_\sigma \hat{\psi}^\dagger_\sigma \hat{\psi}_\sigma \hat{\psi}_\sigma + U_{\uparrow\downarrow} \hat{\psi}^\dagger_\downarrow \hat{\psi}^\dagger_\uparrow \hat{\psi}_\uparrow \hat{\psi}_\downarrow + \frac{g}{2} \left(\hat{\psi}^\dagger_\uparrow \hat{\psi}^\dagger_\downarrow \hat{\psi}_m + \text{h.c.} \right) \right]$$

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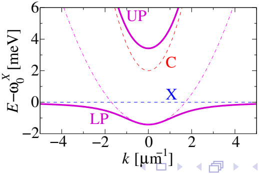
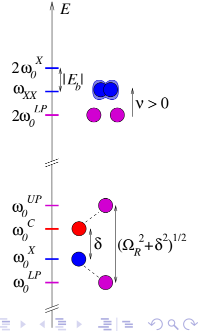
- Resonance width, dispersion derived from dressed exciton T matrix.





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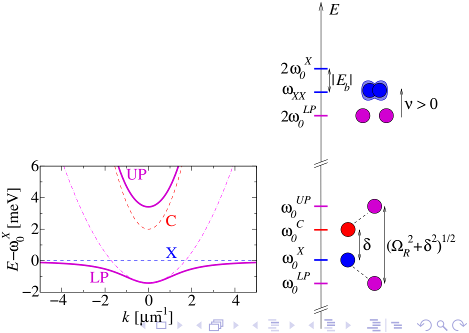

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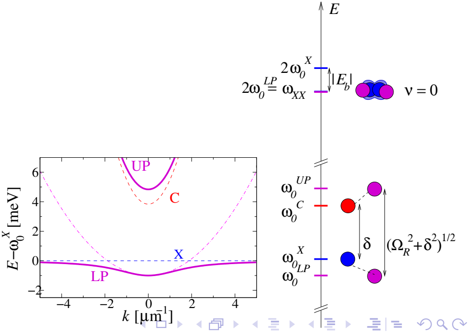

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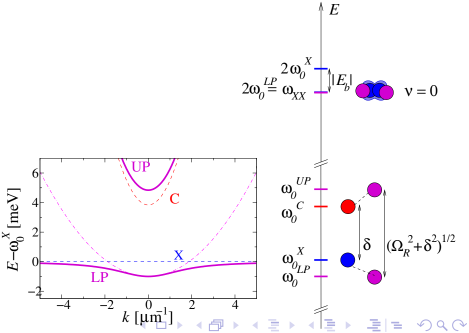

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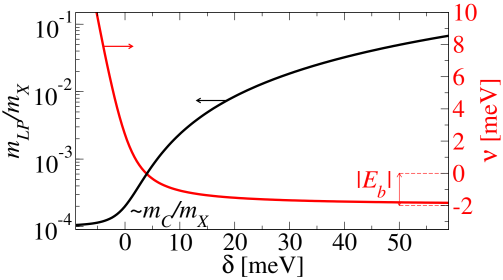
$\nu > 0$
Biexciton in continuum



$\nu < 0$
Bound biexciton.
(Excitonic limit)

Exciton and polariton spin degrees of freedom

- Photon: two circular polarisation modes
- Exciton: bound state of electron & hole
- Exciton spin states $J_z = +2, +1, -1, -2$
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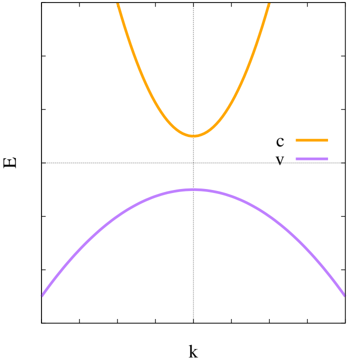

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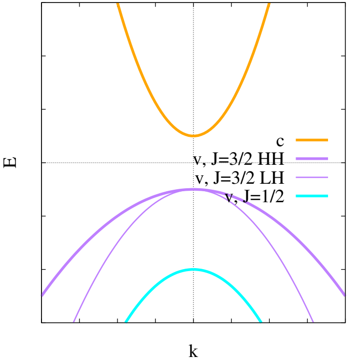

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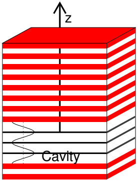

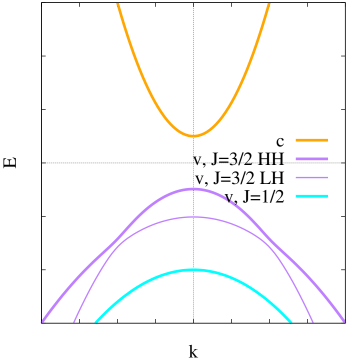

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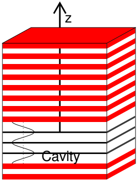

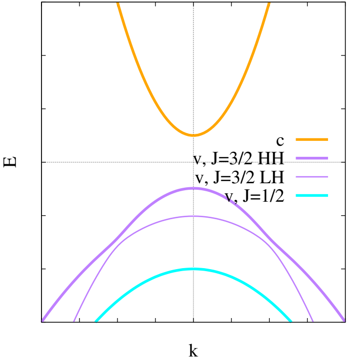

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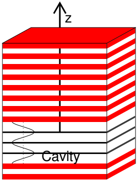

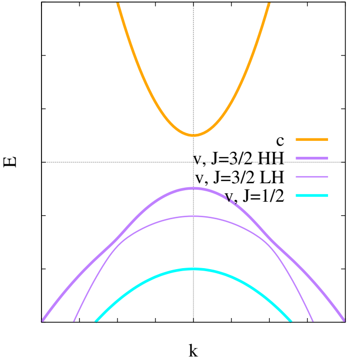

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- Fluctuation effects?
  - Polariton fluctuations irrelevant: $mU \sim 10^{-4}$.
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$$|\Psi\rangle \propto \exp\left(- \sum_{\sigma=\uparrow,\downarrow,m} \psi_\sigma \hat{\psi}_{k=0,\sigma} + \sum_{k,\gamma=a,b,m} \tanh(\theta_{k\gamma}) \hat{b}^\dagger_{k\gamma} \hat{b}^\dagger_{-k\gamma} \right).$$

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- Finite T — minimize free energy
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$$F = -k_B T \ln\left[\mathrm{Tr} e^{-\hat{H}/k_B T}\right] \le F_{\mathrm{MF}} + \langle \hat{H} - \hat{H}_{\mathrm{MF}} \rangle_{\mathrm{MF}}$$

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- Test validity. WIDBG $\hat{H} = \sum_k \frac{k^2}{2m}\psi^\dagger_k\psi_k + \frac{U}{2}\int d^2r\psi^\dagger\psi^\dagger\psi\psi$
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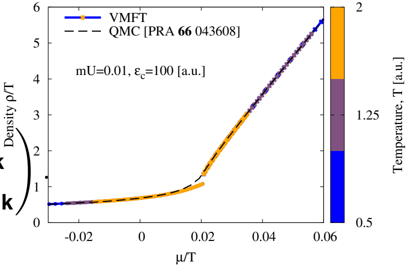

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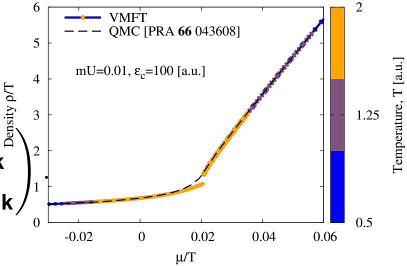

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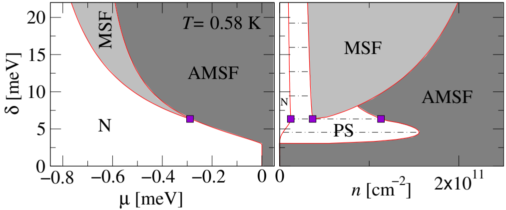
Phase diagram, finite temperature



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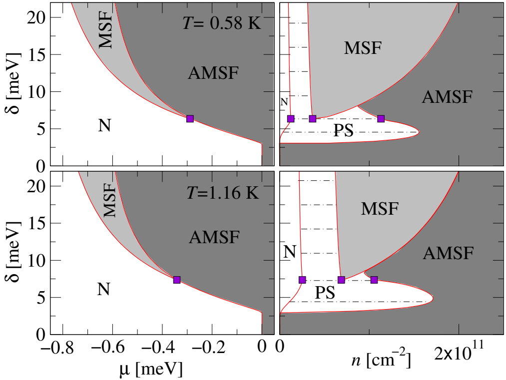

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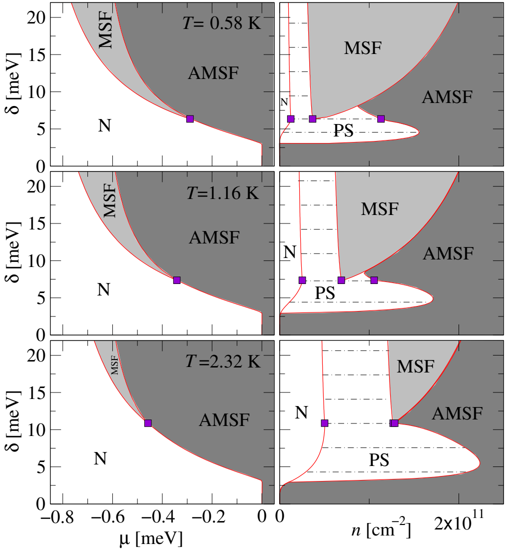

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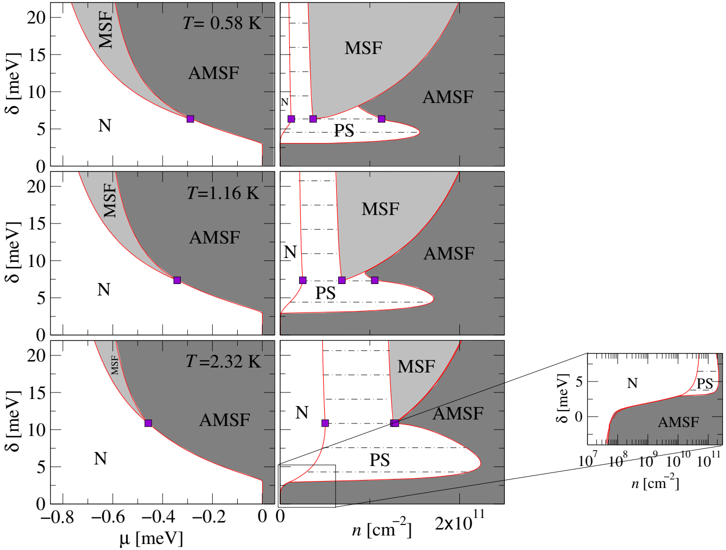
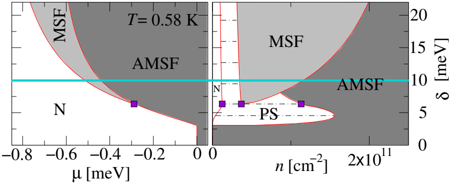
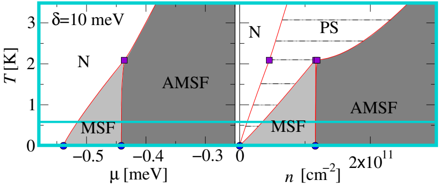


Phase diagram, vs temperature





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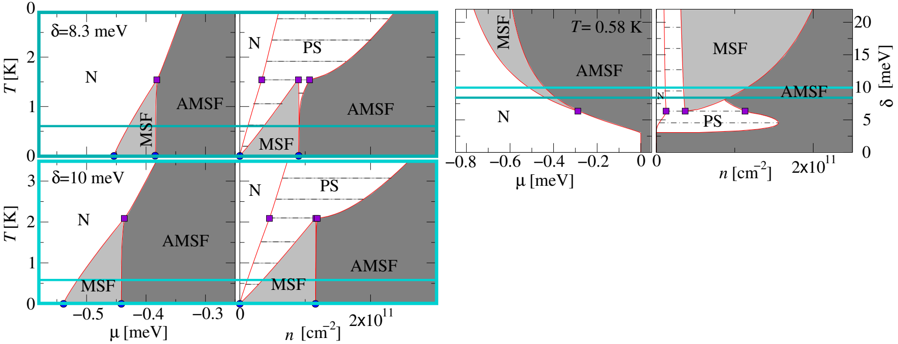

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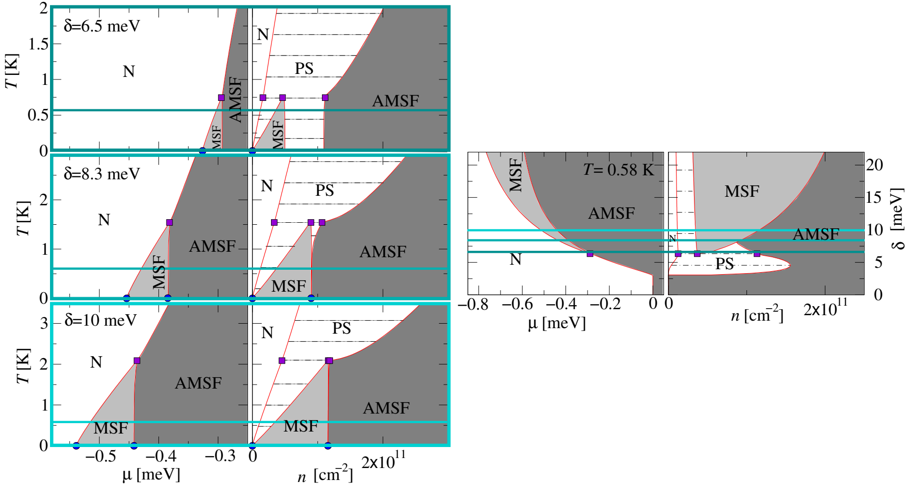
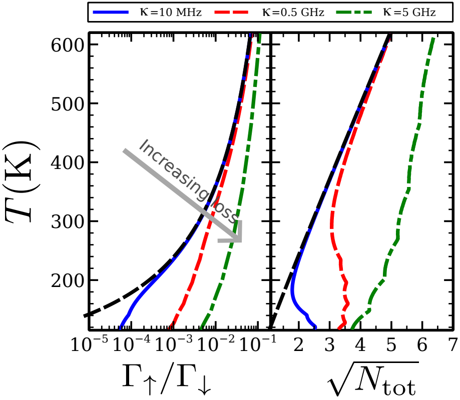


Threshold condition



Compare threshold:

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

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

[Kirton & JK PRL '13]

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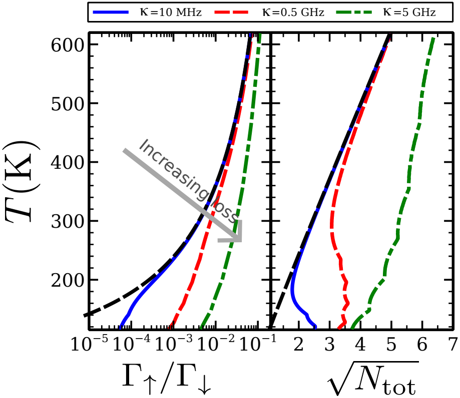

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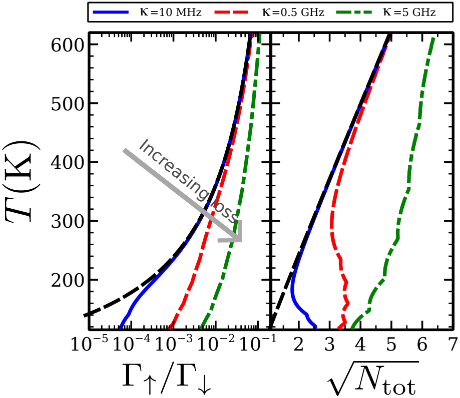

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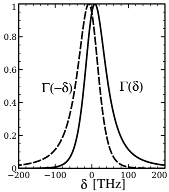

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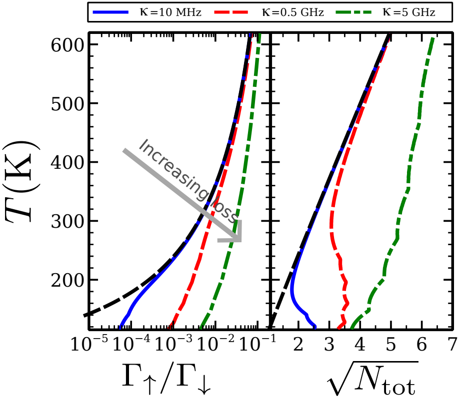

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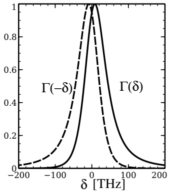

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[Kirton & JK PRL '13]

Explanation: Polaron formation

- Unitary transform

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

- Coupling moves to $S^\pm$

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

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

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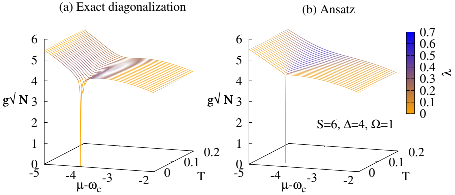
Collective polaron formation

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



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

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

Effective 2LS energy in field:

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

[Cwik *et al.* EPL '14]

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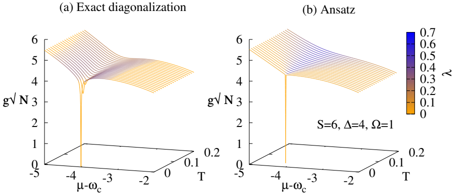

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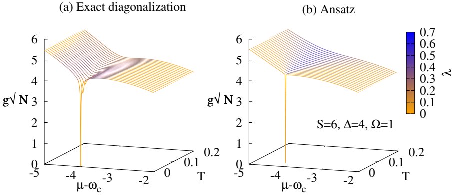

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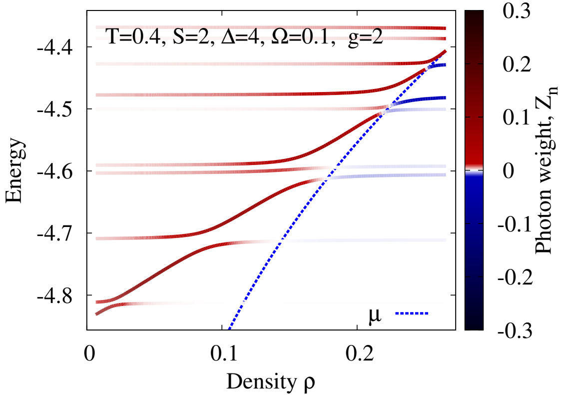
Polariton spectrum: photon weight



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

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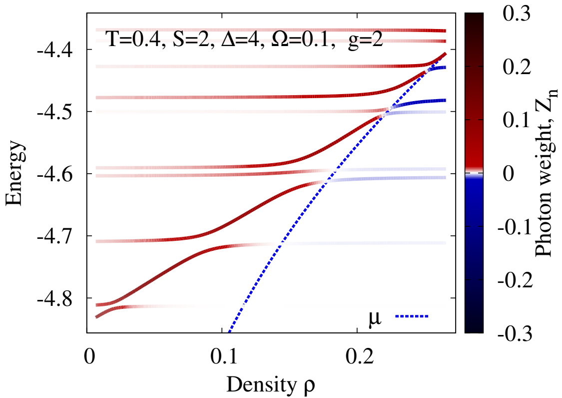

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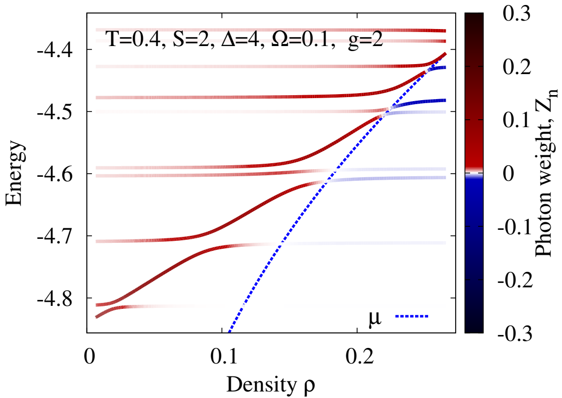

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Polariton spectrum: what condensed

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

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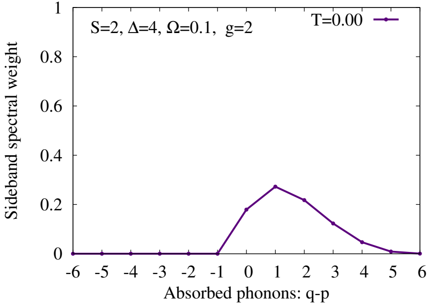

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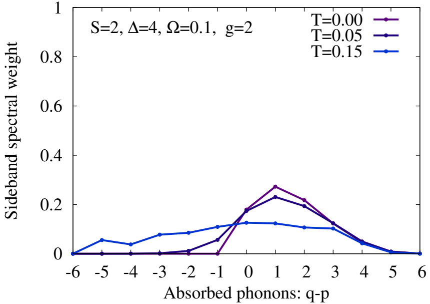

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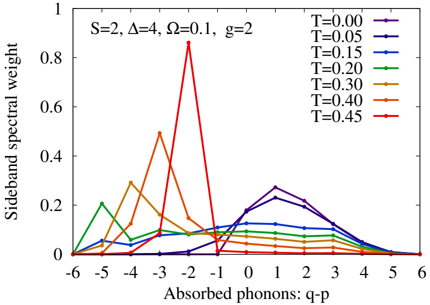

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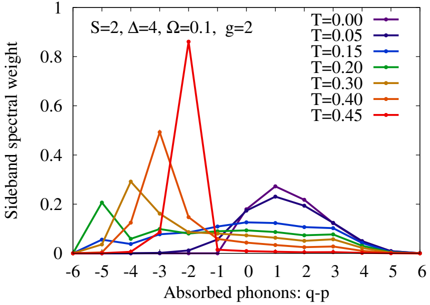

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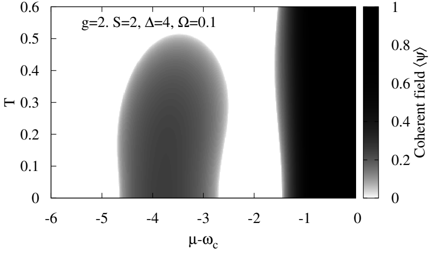

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Polariton spectrum — coupled oscillators

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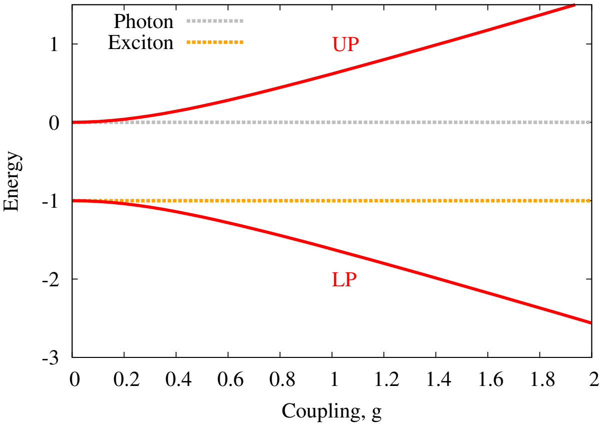

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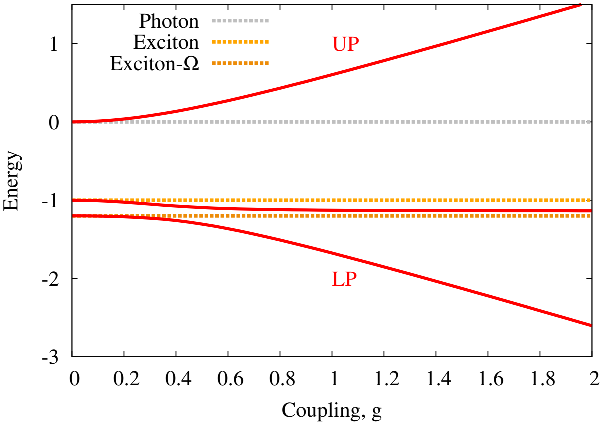

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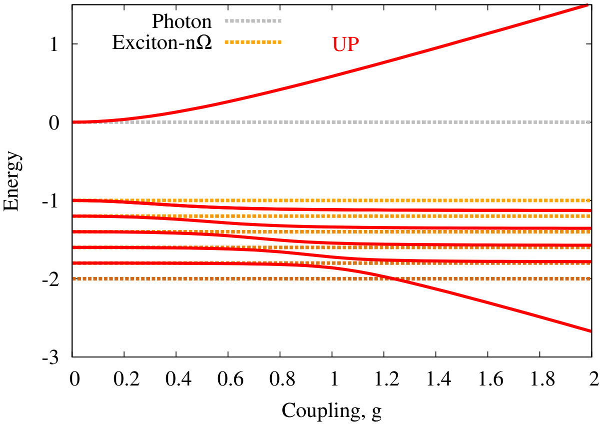

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