Polaritons in some interacting exciton systems

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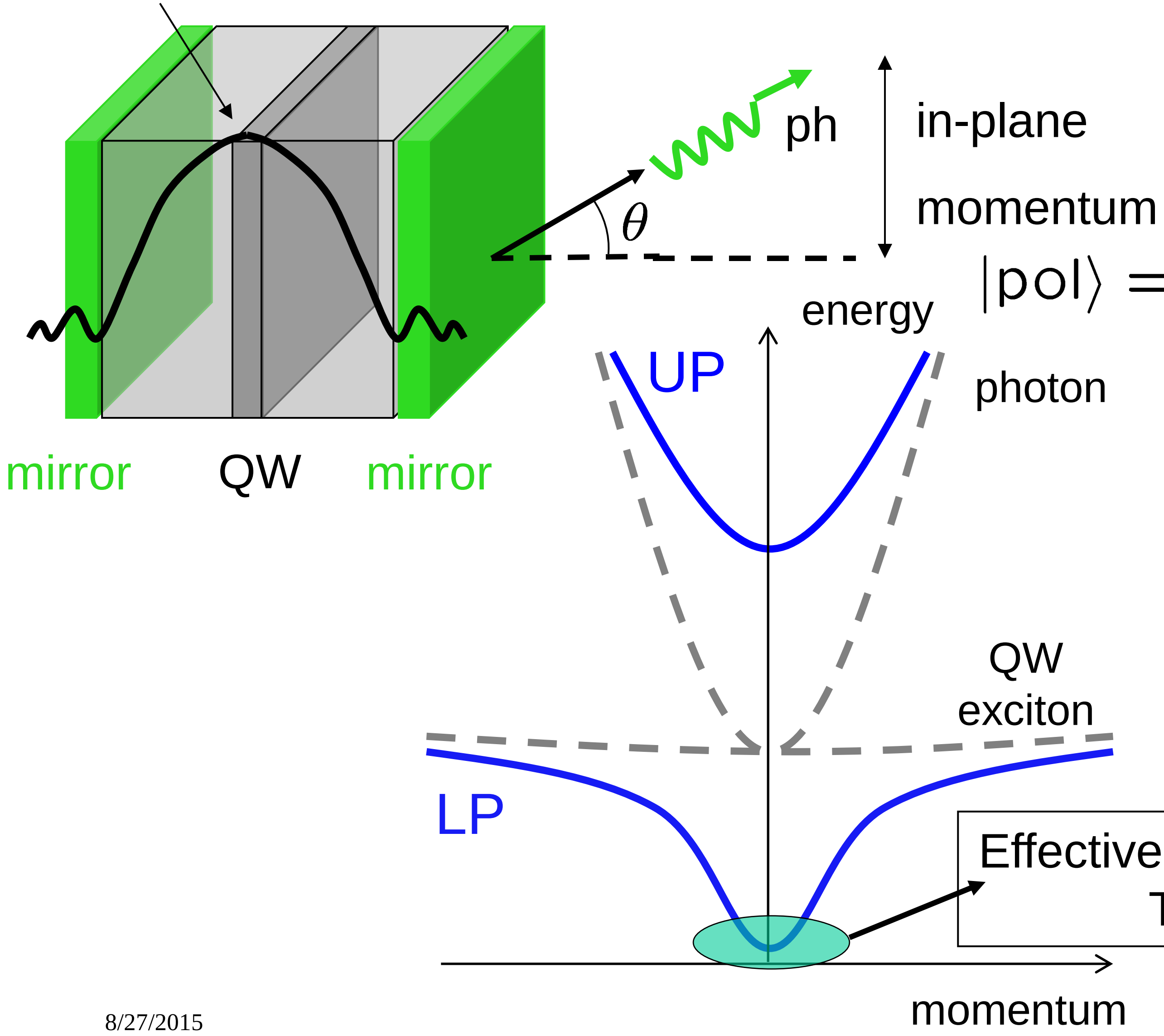
Outline

- Brief review of a microscopic model for polariton condensation and quasi-equilibrium theory
- Quantum dynamics out of equilibrium
  - pumped dynamics beyond mean field theory and dynamical instabilities
  - use of chirped pump pulses to generate non-equilibrium populations, possibly with entanglement
- Polariton systems with strong electron-phonon coupling – e.g. organic microcavities
  - Can you condense into phonon polariton states?
- Cavity – coupled Rydberg atoms
  - Competition between superfluid and quantum crystal

Polaritons: Matter-Light Composite Bosons

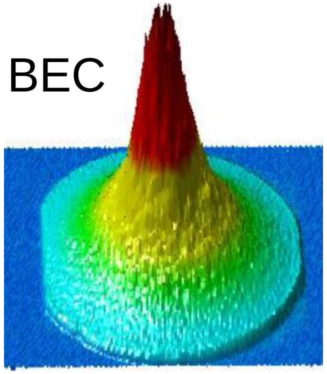
[C. Weisbuch *et al.*, *PRL* **69** 3314 (1992)]



$$|\mathrm{pol}\rangle = c_1|\mathrm{exc}\rangle + c_2|\mathrm{ph}\rangle$$

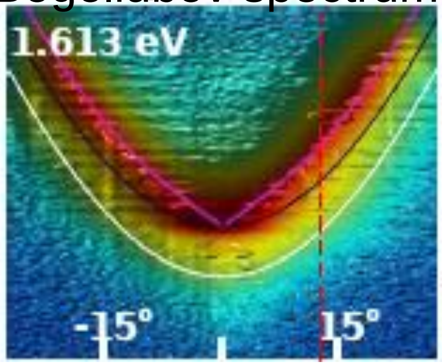
Effective Mass $m^* \sim 10^{-4}\,m_e$
$T_{BEC} \sim 1/m^*$

BEC



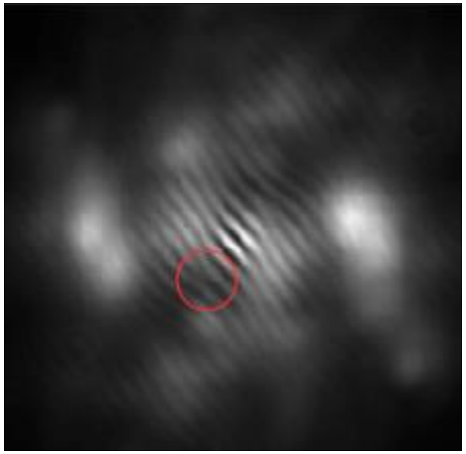
Kasprzak et al 2006

Bogoliubov spectrum



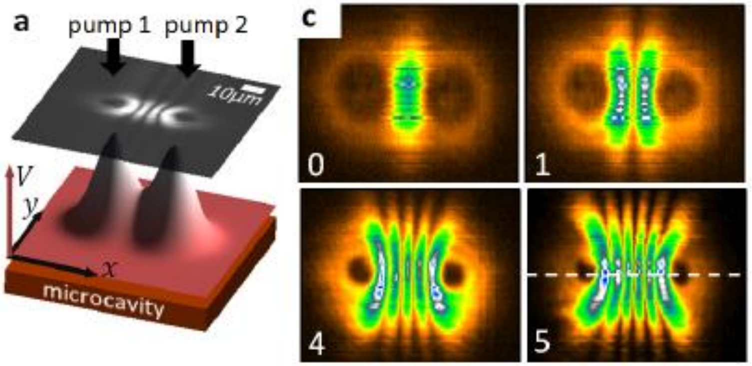
Utsunomiya et al,2008

Vortices



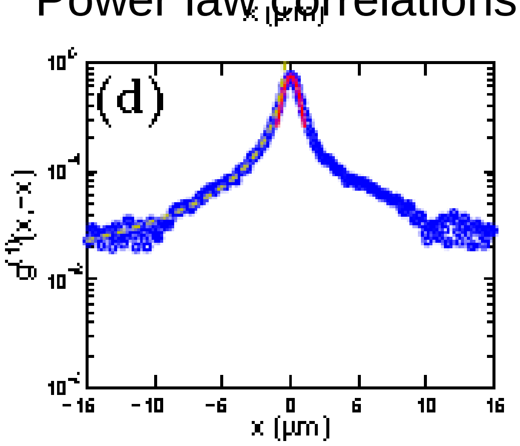
Lagoudakis et al 2008

Coupled condensates



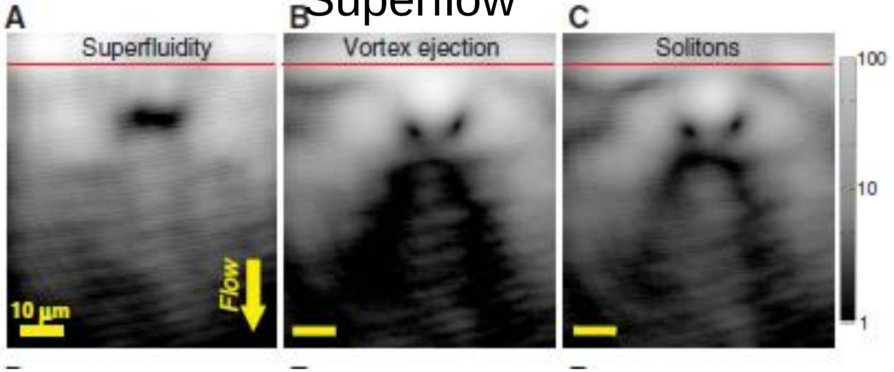
Tosi et al 2012

Power law correlations



Roumpos et al 2012

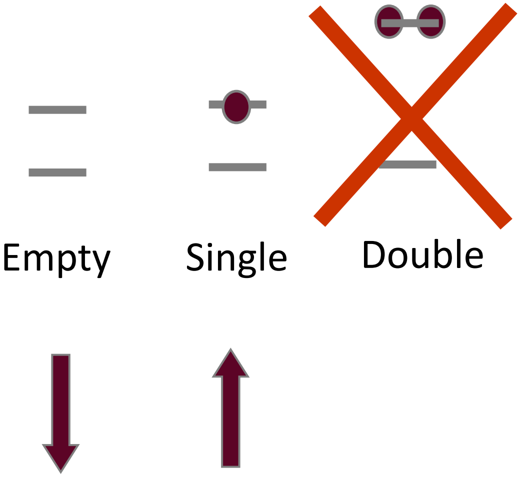
Superflow



Amo et al, 2011

Polaritons and the Dicke Model – a.k.a Jaynes-Tavis-Cummings model

Excitons as “spins”



Spins are flipped by absorption/emission of photon

$$H = \omega\psi^\dagger\psi + \sum_i \epsilon_i S_i^z + \frac{g}{\sqrt{N}} \sum_i \left[S_i^+ \psi + \psi^\dagger S_i^- \right]$$

N ~ [(photon wavelength)/(exciton radius)]^d $_{>>}$ 1

Mean field theory – i.e. BCS coherent state – expected to be good approximation

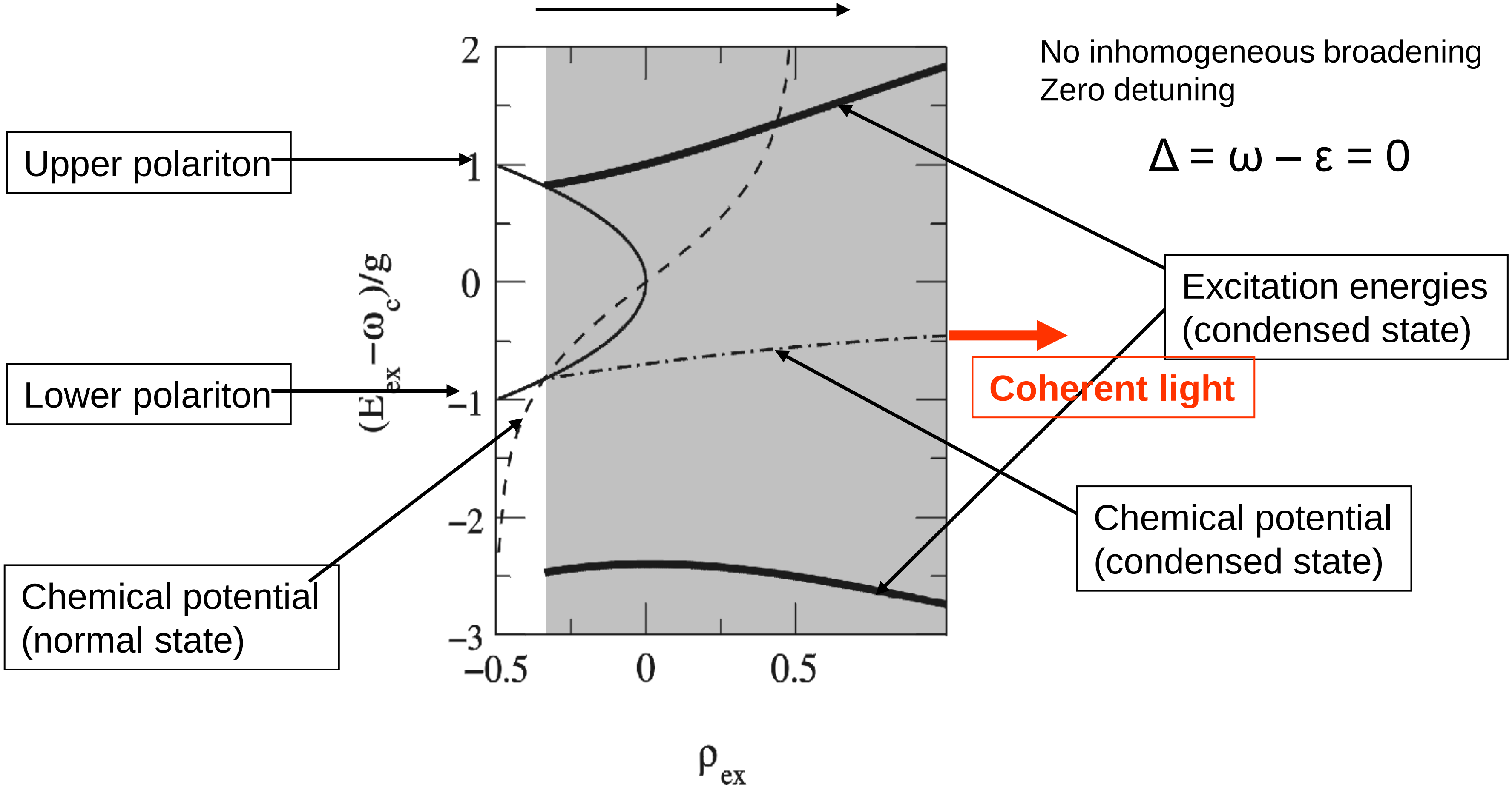
$$|\lambda, w_i\rangle = \exp\left[\lambda\psi^\dagger + \sum_i w_i S_i^+\right]|0\rangle \qquad T_c \approx g\exp(-1/gN(0))$$

Transition temperature depends on coupling constant

Mean field theory of Condensation

Eastham & PBL,
PRB 64, 235101 (2001)

Increasing excitation density



Upper polariton

Lower polariton

Chemical potential (normal state)

Excitation energies (condensed state)

Coherent light

Chemical potential (condensed state)

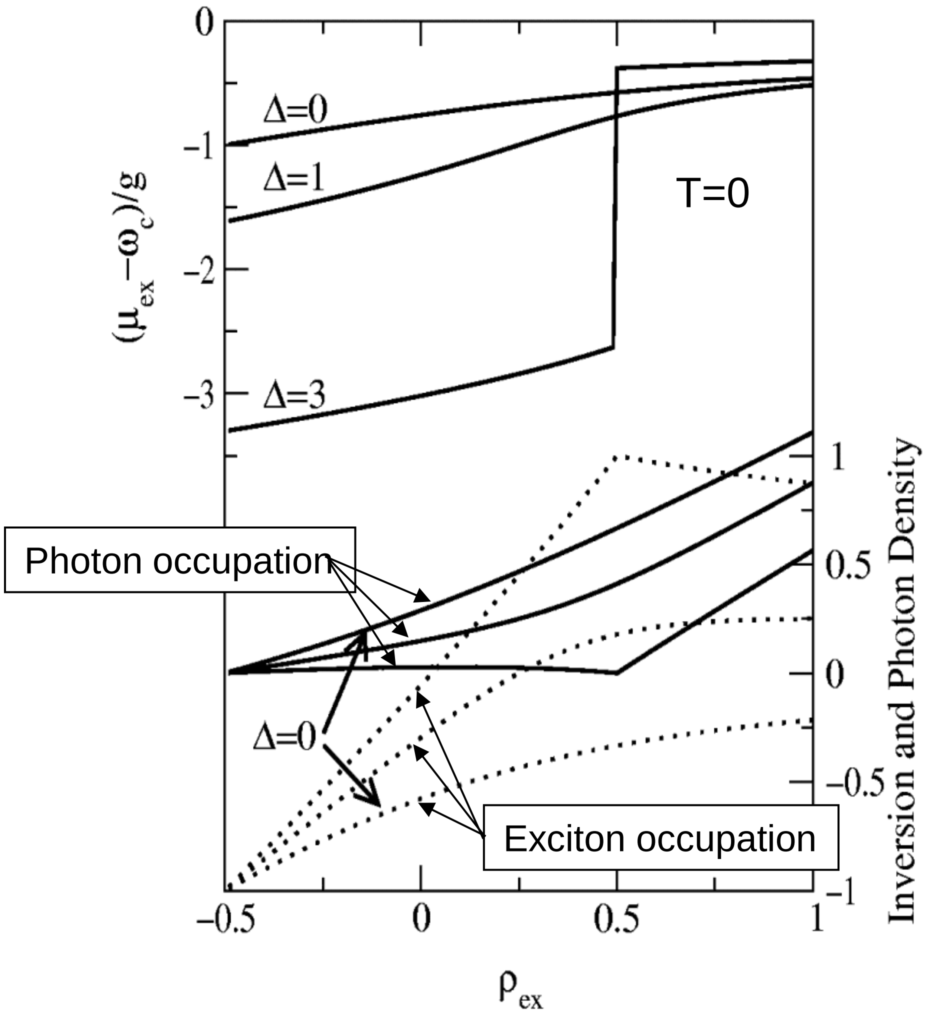
No inhomogeneous broadening
Zero detuning

$$\Delta = \omega - \varepsilon = 0$$

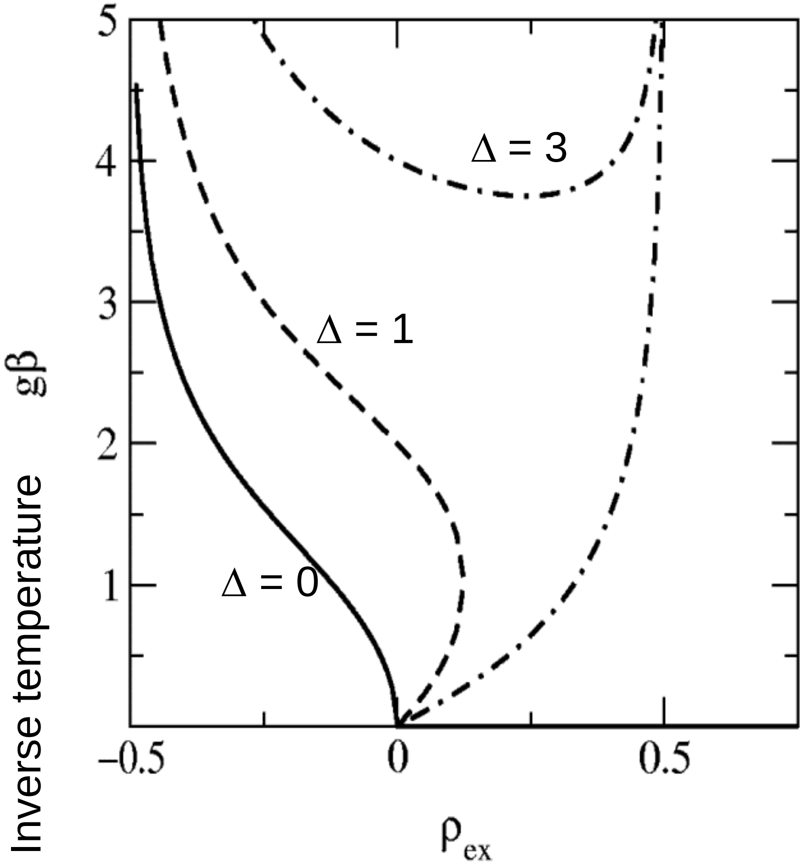
$$\rho_{ex} = \frac{1}{N}\left\langle \sum_i S_z^i + \psi^\dagger \psi \right\rangle = \frac{N_{photon} + N_{exciton}}{N} - \frac{1}{2}$$

Phase diagram with detuning: appearance of “Mott lobe”

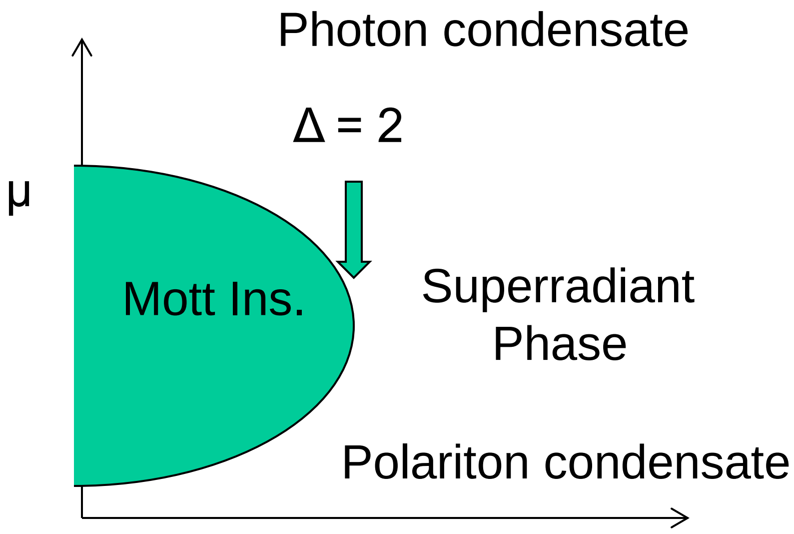
Solid State Commun, 116, 357 (2000); PRB 64, 235101 (2001)



Detuning $\Delta = (\omega - \varepsilon)/g$



Single Mott Lobe for s=1/2 state



Photon condensate

Δ = 2

μ

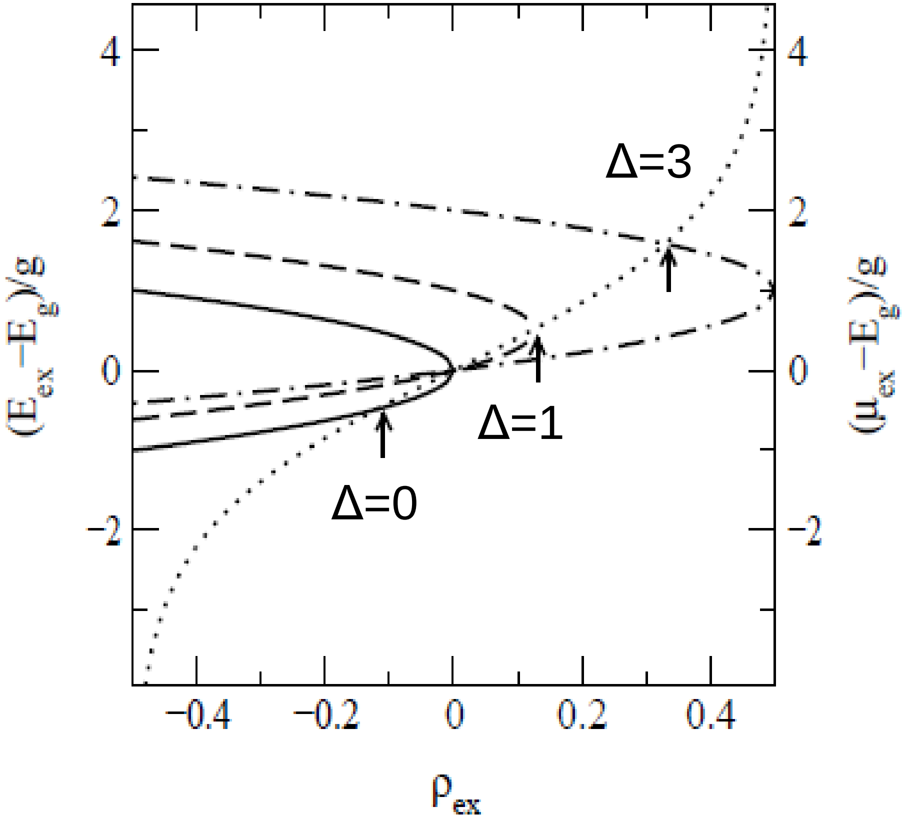
Mott Ins.

Superradiant Phase

Polariton condensate

$$-\Delta = -\omega_0 + \epsilon$$

negative detuning

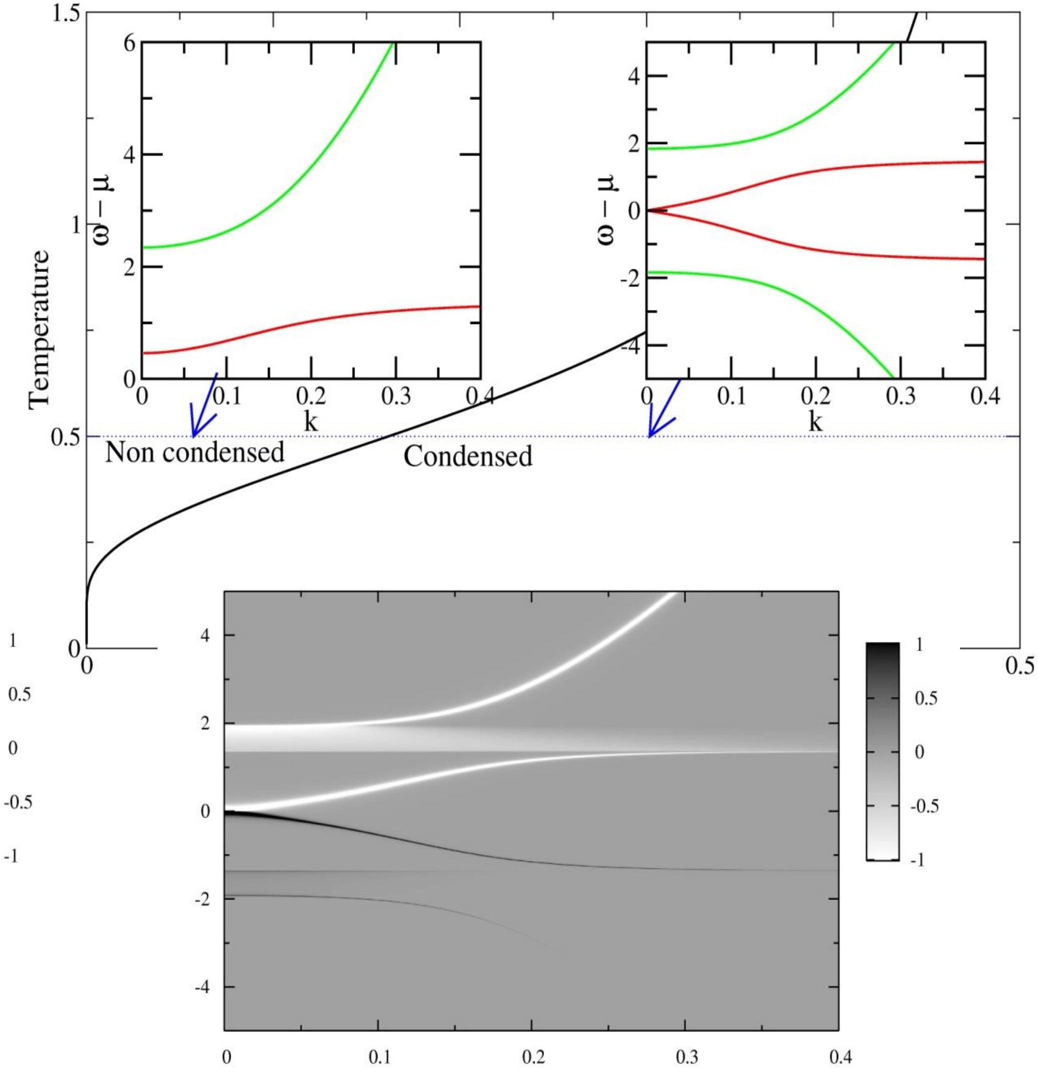


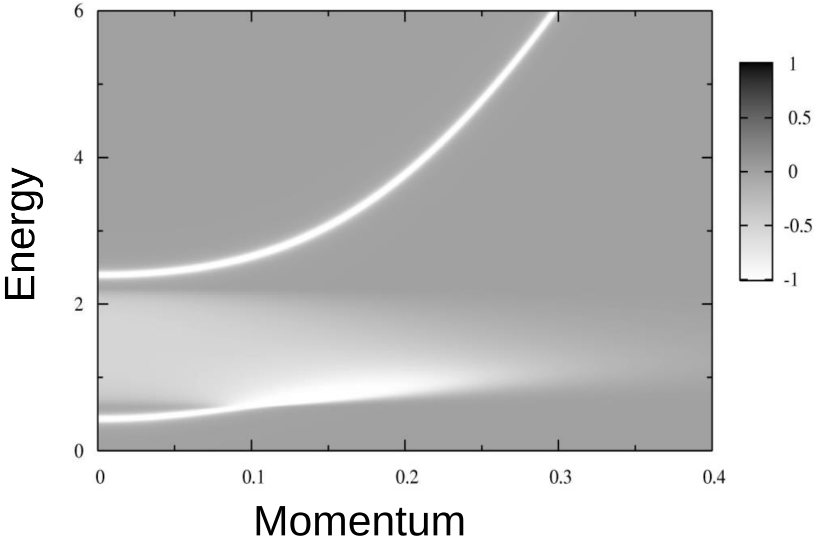
Eastham and Littlewood, Solid State Communications 116 (2000) 357--361

2D polariton spectrum

Keeling et al PRL 93, 226403 (2004)

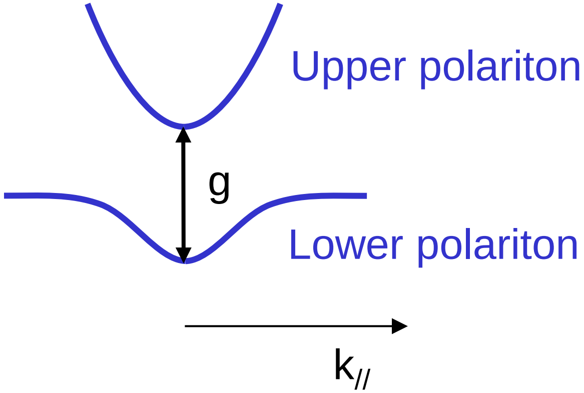
- Below critical temperature polariton dispersion is linear – Bogoliubov sound mode appears
- Include disorder as inhomogeneous broadening



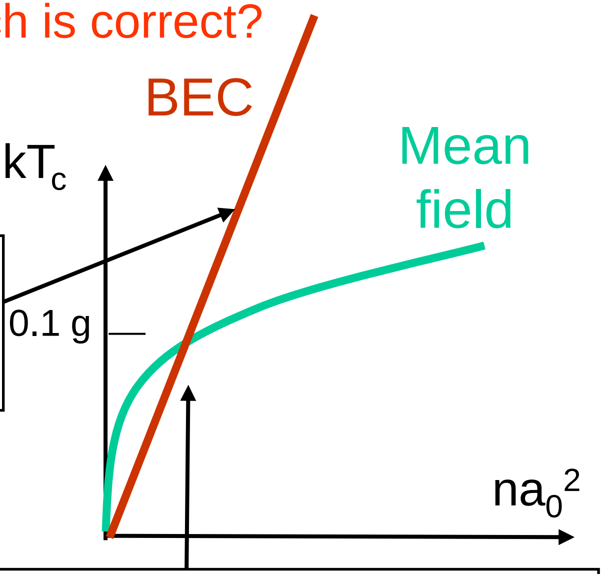


Beyond mean field: Interaction driven or dilute gas?

- Conventional “BEC of polaritons” will give high transition temperature because of light mass m^*
- Single mode Dicke model gives transition temperature ~ g



Which is correct?



$$k_B T_0 \approx \frac{\hbar^2}{2m} n$$

$$\left(\frac{g}{\hbar^2/2ma_0^2}\right) \times \left(\frac{m^*}{m}\right) \approx 10^{-4}$$

a_o = characteristic separation of excitons

a_o > Bohr radius

Dilute gas BEC only for excitation levels < 10^9 cm^{-2} or so

A further crossover to the plasma regime when $na_B^2 \sim 1$

Condensation in polaritons with strong electron-phonon coupling

Justina Cwik, Jonathan Keeling (St Andrews);

Sahinur Reja (Cambridge-> Dresden)

Europhysics Letters 105 (2014) 47009

“Dicke-Holstein” model

Cwik, Reja, Keeling, PBL
EPL 105 (2014) 47009

Exciton-polariton (as before) + coupling to local phonon mode
Mimics coupling of Frenkel exciton to optical phonon

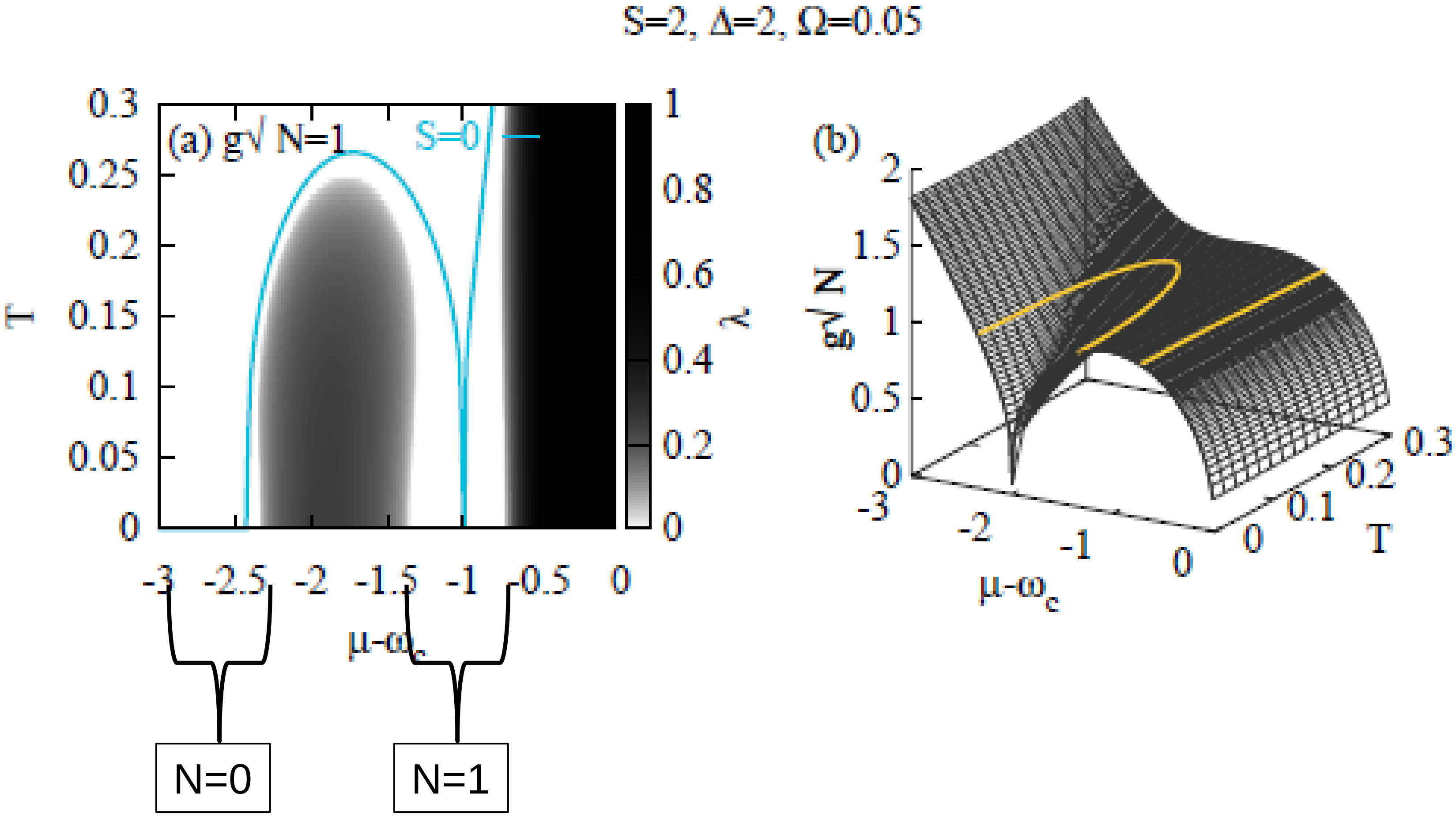
$$\hat{H} - \mu N = \overset{\text{Cavity}}{\overset{\Downarrow}{\tilde{\omega}_c \hat{\psi}^\dagger \hat{\psi}}} + \sum_n \left\{ \overset{\text{Exciton}}{\overset{\Downarrow}{\frac{\tilde{\epsilon}}{2}\sigma_n^z}} + \overset{\text{Rabi coupling}}{\overset{\Downarrow}{g(\hat{\psi}\sigma_n^+ + \hat{\psi}^\dagger \sigma_n^-)}} + \underset{\text{Phonon}}{\underset{\Uparrow}{\Omega \hat{a}_n^\dagger \hat{a}_n}} + \underset{\text{Local (Holstein) coupling}}{\underset{\Uparrow}{\frac{\Omega\sqrt{S}}{2}\sigma_n^z(\hat{a}_n + \hat{a}_n^\dagger)}} \right\}.$$

With strong exciton-phonon coupling, exciton develops sidebands
Can you have condensation into a phonon replica?

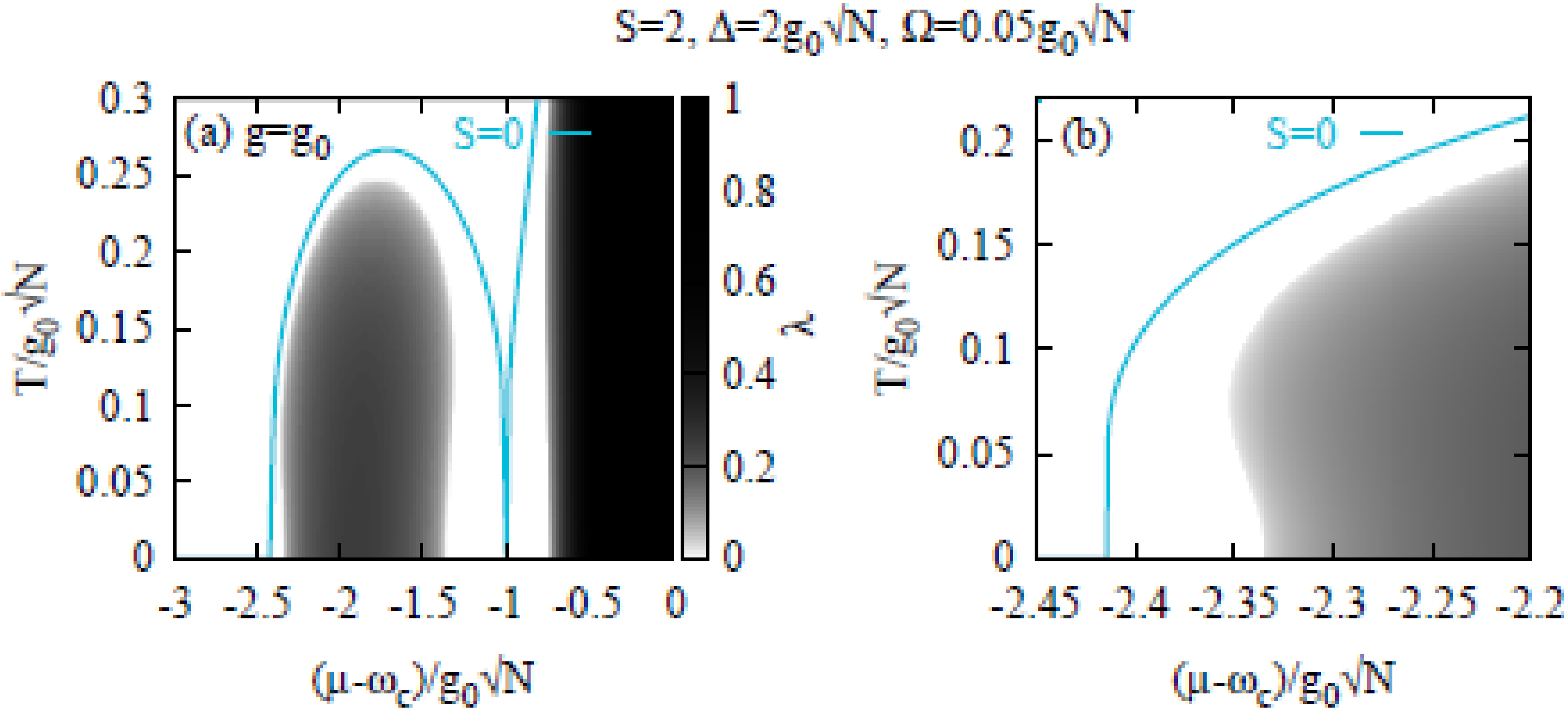
Method: mean field for photons; numerical diagonalization of phonon

Phase diagram – critical detuning

Re-entrant 'Mott lobes'

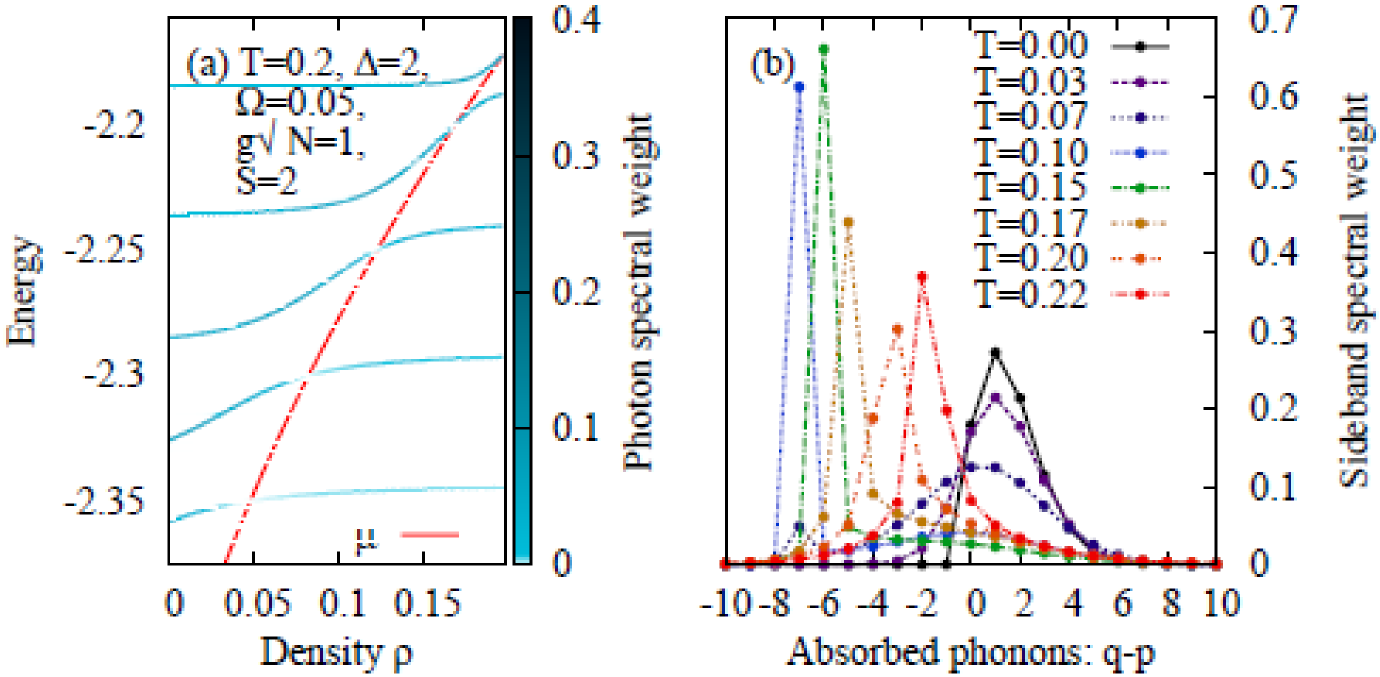


Super-radiant phase stabilized by raising temperature



Vibrational replicas

- Paradox? Infinite number of vibrational replicas at energies ε-nΩ – some of which must be therefore below the chemical potential.
- Resolution: Photon spectral weight vanishes at most level crossings



Interacting excitons – Rydberg atoms in optical cavities

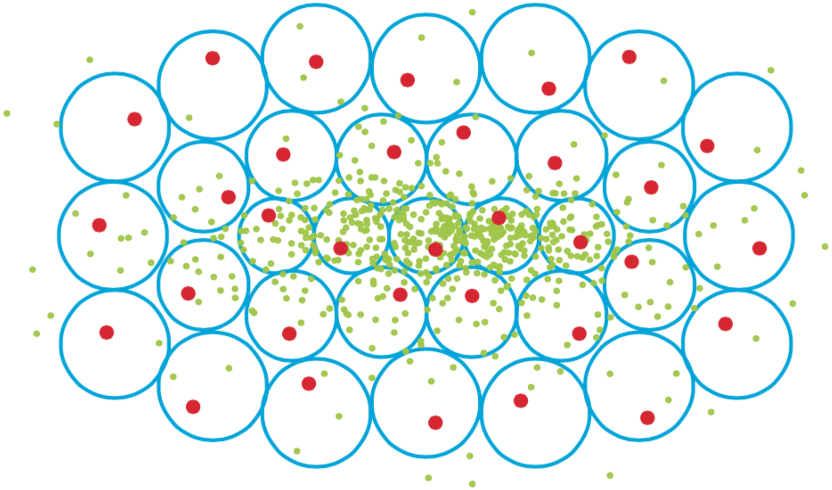
Alex Edelman (Chicago)

Ingredients

Ingredients:

2-level systems
Cavity photons } Coupled
Nearly Resonant
…and interacting
…on a lattice – Why a lattice?

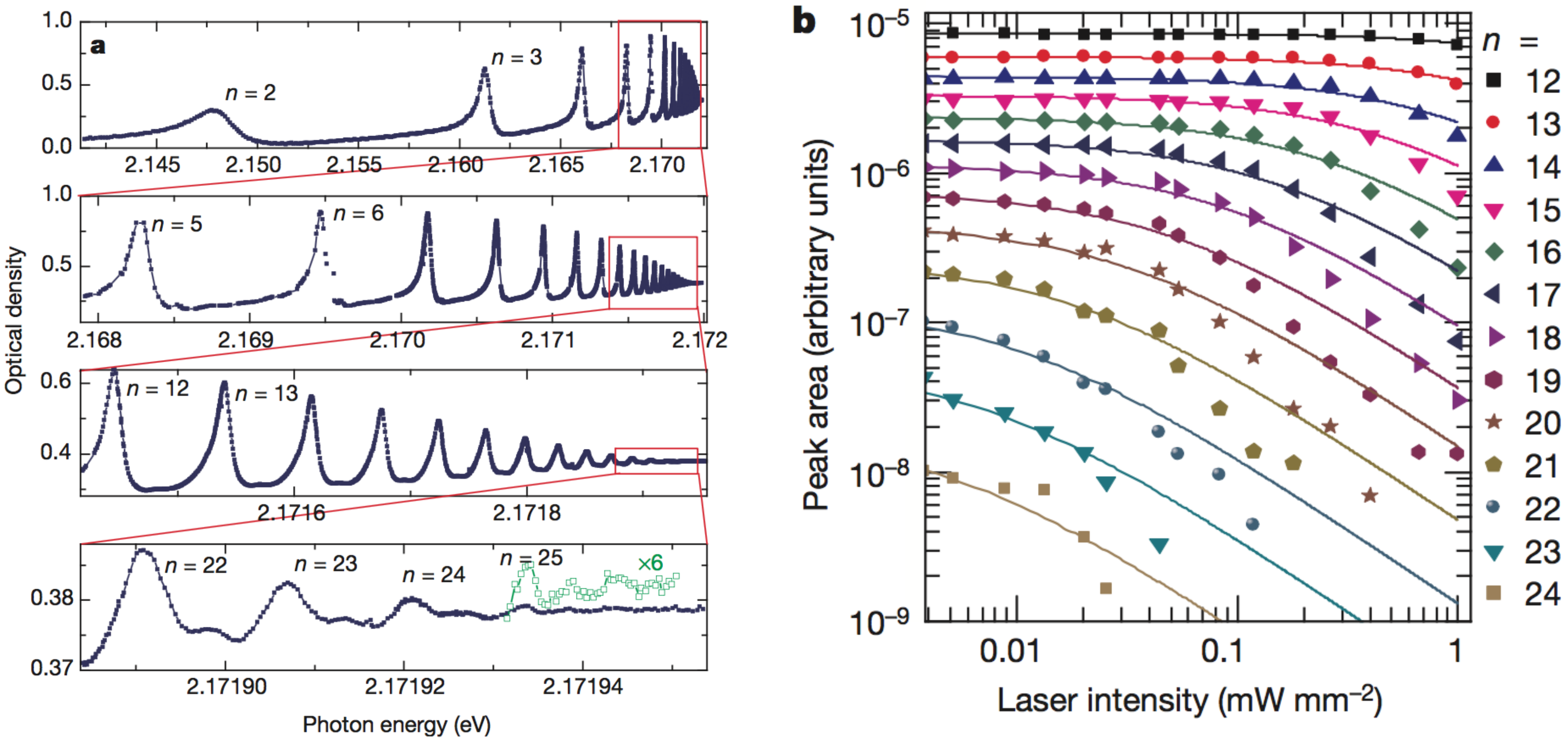
Blockade Effect:



- As an emergent lattice structure
- As a dilute limit

Löw, J Phys B (2012)

A detour: Cu$_2$O



Kazimierczuk, T., Fröhlich, D., Scheel, S., Stolz, H. & Bayer, M.
Giant Rydberg excitons in the copper oxide Cu2O. *Nature* **514,** 343–347 (2014).

Rydberg polaritons

$$H = \sum_q \omega_q \psi_q^\dagger \psi_q + \sum_i \frac{1}{2}\epsilon_i \left[a_i^\dagger a_i - b_i^\dagger b_i\right] + \sum_{i,q} g_{i,q}\left[\psi_q^\dagger b_i^\dagger a_i + \psi_q a_i^\dagger b_i\right]$$

$$+ \sum_{i,j} a_i^\dagger a_i U(i-j) a_j^\dagger a_j \qquad a_i^\dagger a_i + b_i^\dagger b_i = 1$$

Represent exciton as two fermionic levels with a constraint of single occupancy
Here, simplify as 2D lattice model with

(a) nearest neighbour interactions,
(b) generalised long range interactions

Consider instability of the superradiant polariton state.
No weak coupling instability if U(q) > 0
In strong coupling expect an effective interaction that generates a (short) length scale from the density itself.
Mixing of amplitude and phase modes only allowed at non-zero momentum.

See also Zhang et al PRL 110, 090402 (2013)

Model and Method

$$Z = \int \mathcal{D}(\underbrace{\bar{a}_j, a_j, \bar{b}_j, b_j}_{\text{Excitons (as fermions)}}, \underbrace{\bar{\psi}_{\mathbf{k}}, \psi_{\mathbf{k}}}_{\text{Photons}}) e^{-\int_0^\beta d\tau\, H - \mu N} \rightarrow \int \mathcal{D}(\underbrace{\bar{\psi}_{\mathbf{k}}, \psi_{\mathbf{k}}}_{\text{Photons}}, \underbrace{\bar{\phi}_{\mathbf{k}}, \phi_{\mathbf{k}}}_{\text{Density waves}}) e^{-S_{\text{eff}}[\psi,\phi]}$$

Mean Field:

$$\frac{\delta S_{\text{eff}}}{\delta \psi_{\mathbf{k}}} = 0$$

$$\frac{\delta S_{\text{eff}}}{\delta \phi_{\mathbf{k}}} = 0$$

$$\psi_{k \neq 0} = 0$$

$$\phi_{k \neq 0} = 0$$

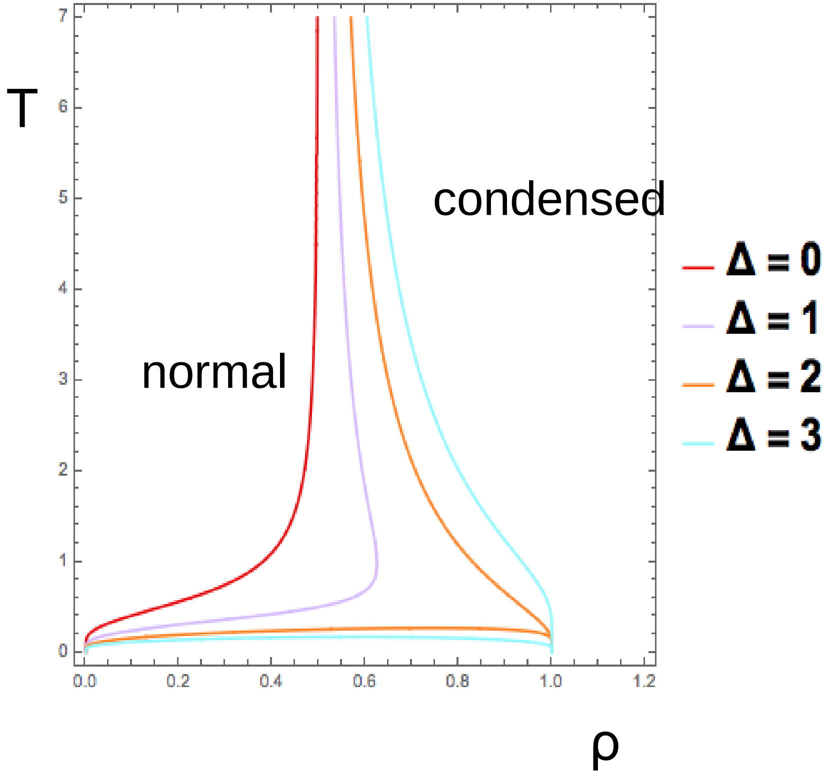
Fluctuations:

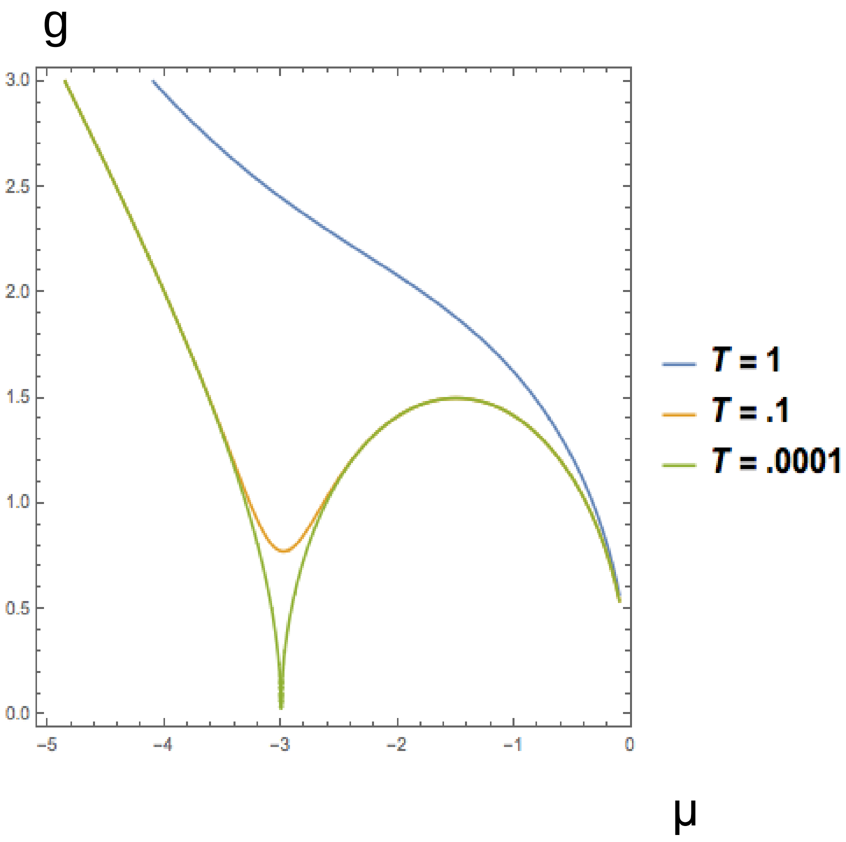
$$\psi_0 \rightarrow \psi_0 + \textstyle\sum_k \delta\psi_{\mathbf{k}}$$

$$\phi_0 \rightarrow \phi_0 + \textstyle\sum_k \delta\phi_{\mathbf{k}}$$

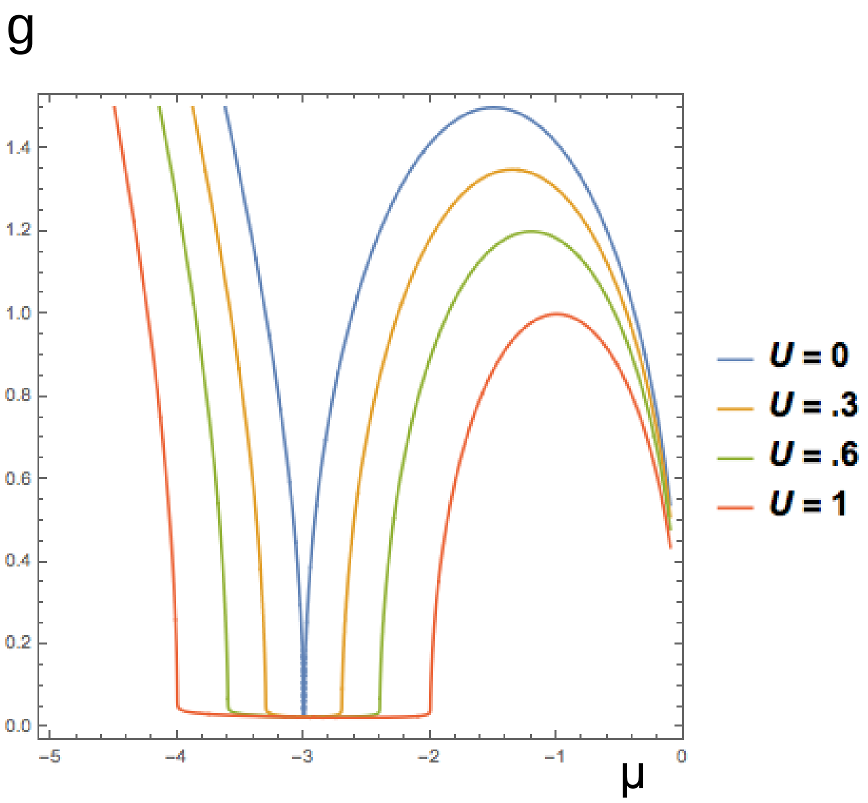
$$f = f_0 + \textstyle\sum_k \begin{pmatrix} \delta\psi_{-k} & \delta\phi_{-k} \end{pmatrix} Q(\omega, k) \begin{pmatrix} \delta\psi_k \\ \delta\phi_k \end{pmatrix}$$

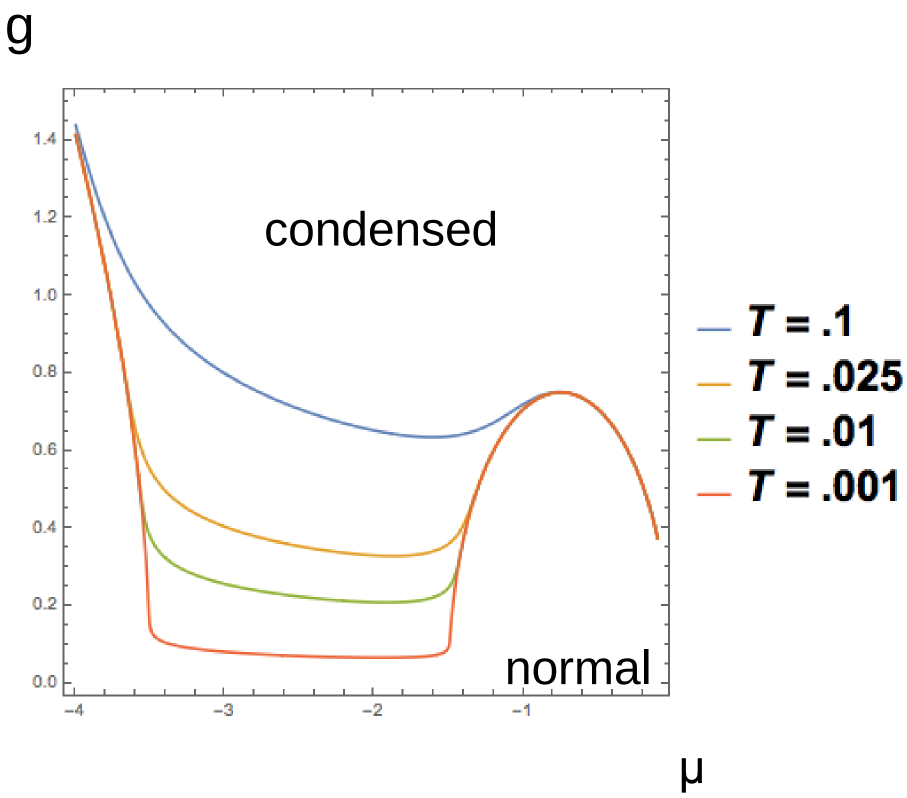
Phase Diagram: U = 0





Phase Diagram: Finite U (mean field)

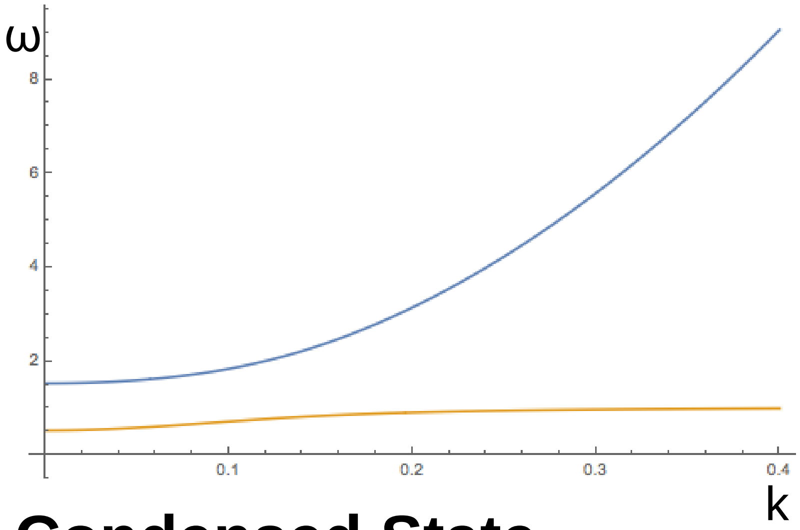




Trivial effect – occupancy + interactions shifts excitons closer to resonance with photon

Mean Field Excitation Spectrum (U=0)

Normal State



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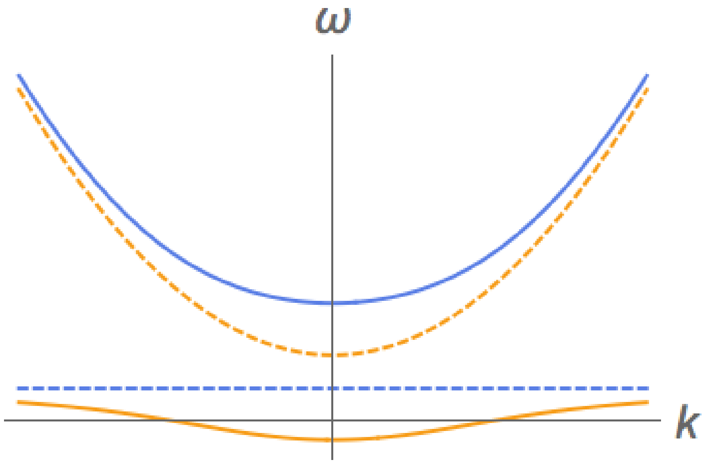
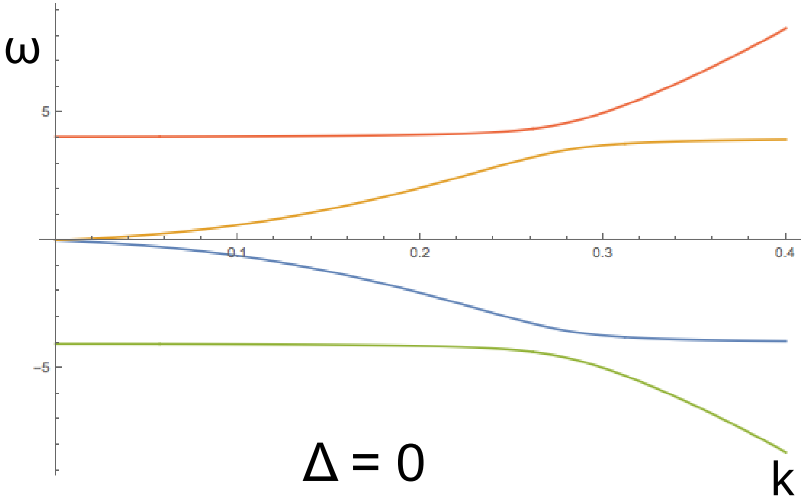
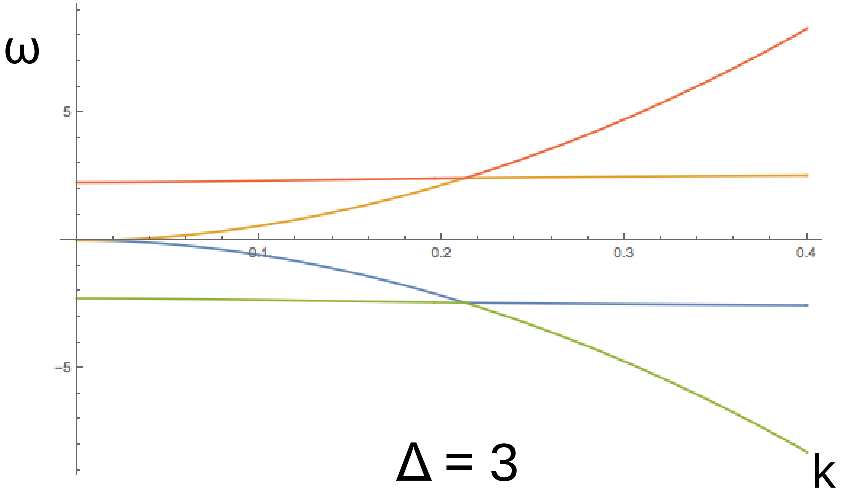


Condensed State



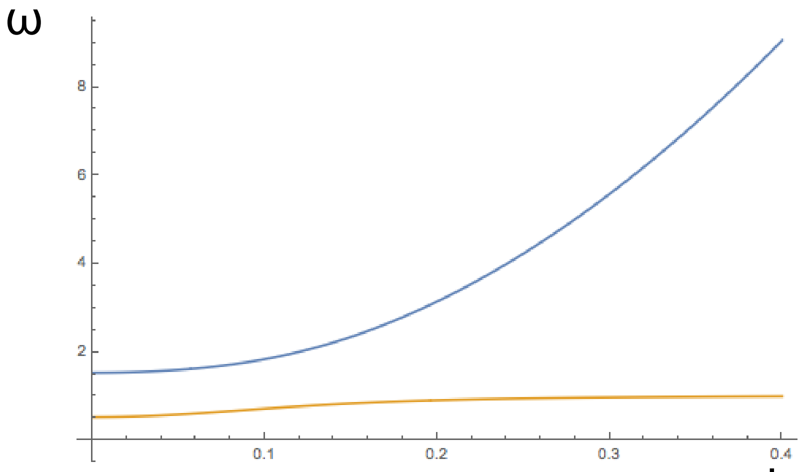
Δ = 0



Δ = 3

Excitation Spectrum – With Interactions

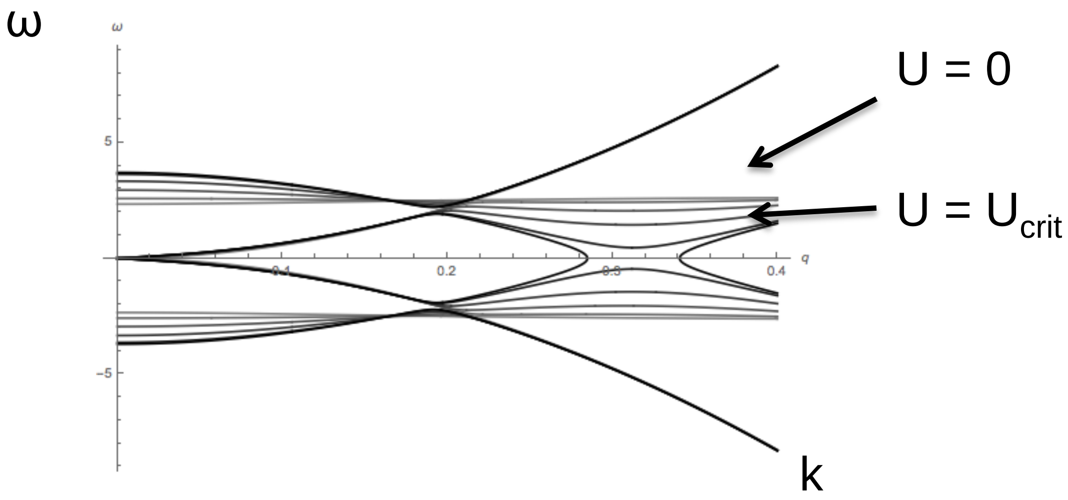
Normal State



Completely unmodified!

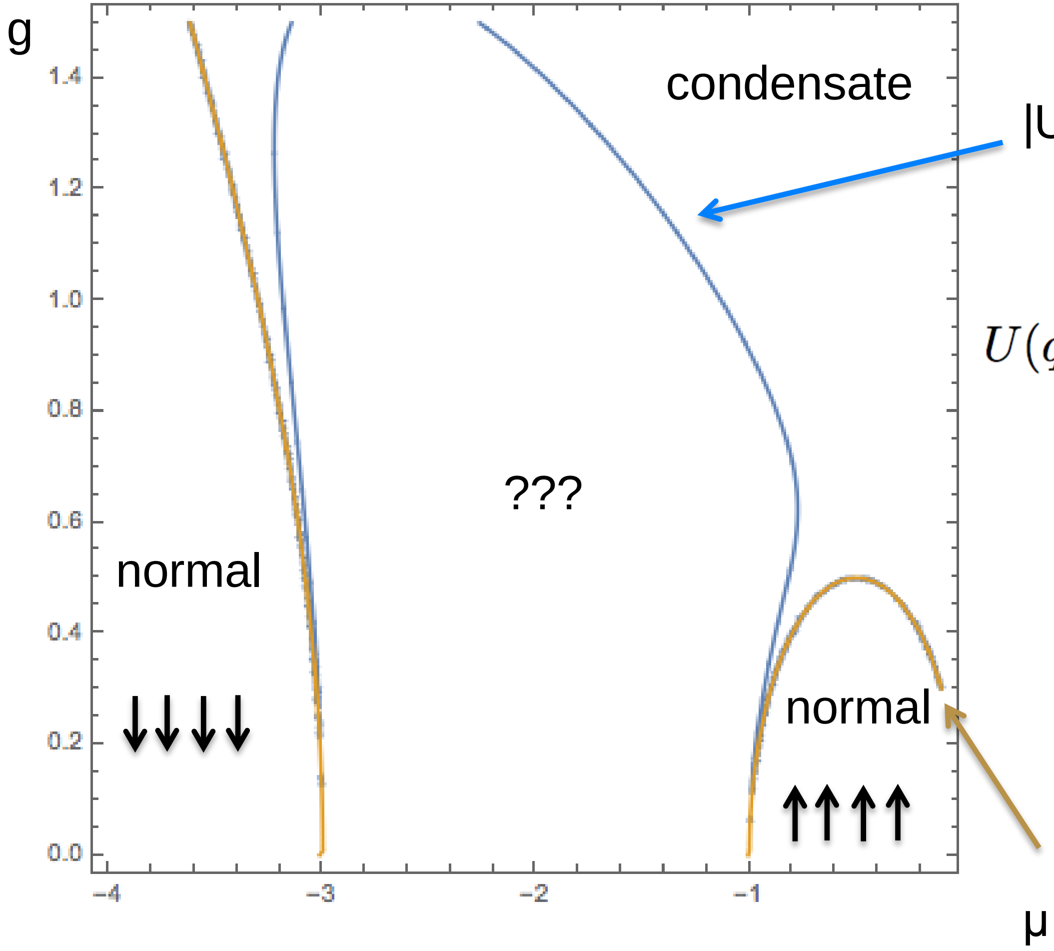
- Low-density limit
- Dispersionless excitons (spins)

Condensed State



An instability develops at q=π (in units of the lattice)

Diagnosing the Instability



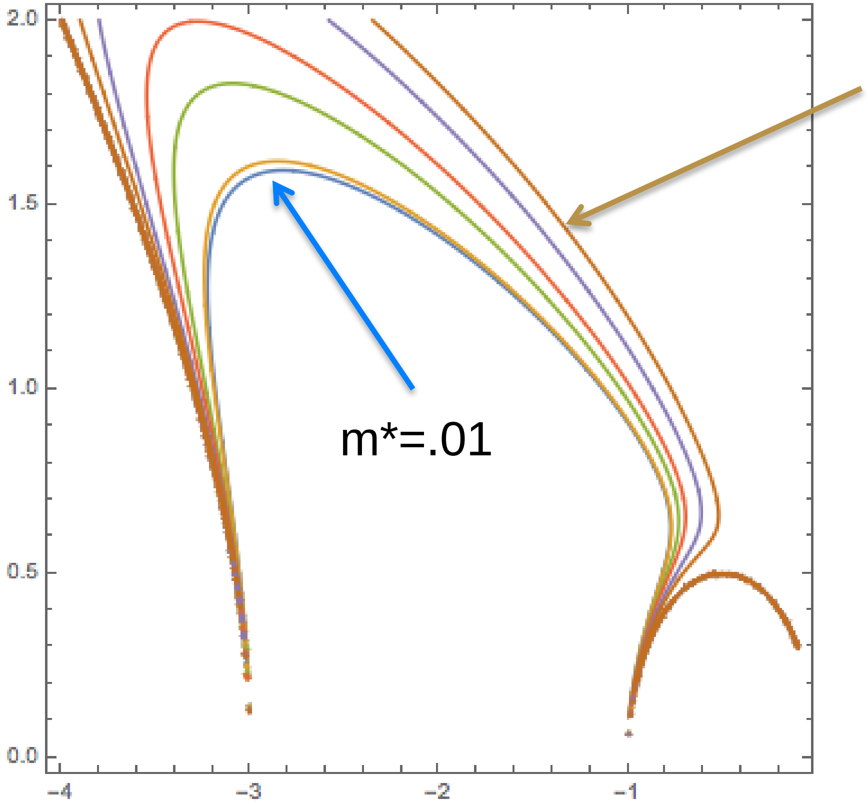
|U|=|U$_{crit}$| instability line

Mean-field phase boundary

$$U(q) = -2\frac{\underline{g^2}\,\underline{|\psi_0|^2}\,\underline{\bar{\omega}_q} + \underline{(\bar{\omega}_q - \bar{\omega}_0)}\underline{\bar{\xi}^2}}{\underline{|\psi_0|^2}\,\underline{\bar{\omega}_0\bar{\omega}_q}}$$

Coupling
Condensate Amplitude
Photon Dispersion
Exciton energy

Diagnosing the Instability



Infinite-bandwidth limit:

$$U(q) = -\frac{E^2}{2\omega_0|\psi_0|^2}$$

Compare:

Fermions

$$U(q) = -\frac{1}{\Pi(\omega{=}0,q)} \approx -\frac{\epsilon_F}{n}$$

Bosons

$$U(q) = -\frac{k^2/2m}{2\rho_0}$$

Staggered Mean Field

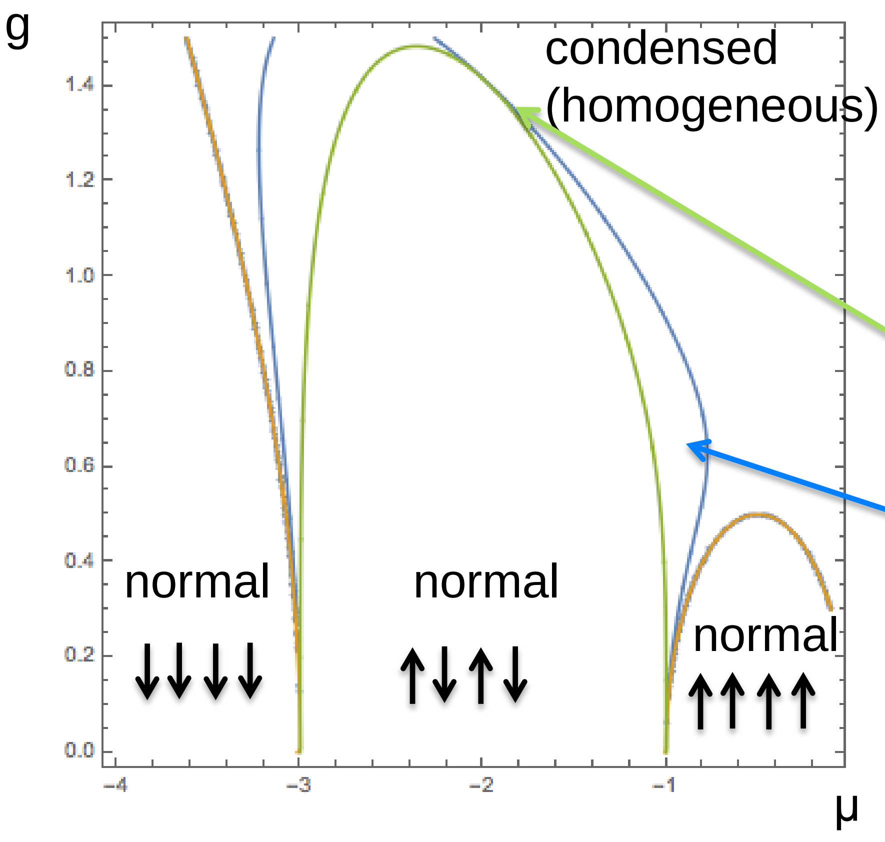
$$Z = \int \mathcal{D}(\underbrace{\bar{a}_j, a_j, \bar{b}_j, b_j}_{\text{Spins (as fermions)}}, \underbrace{\bar{\psi}_{\mathbf{k}}, \psi_{\mathbf{k}}}_{\text{Photons}}) e^{-\int_0^\beta d\tau\, H - \mu N} \rightarrow \int \mathcal{D}(\underbrace{\bar{\psi}_{\mathbf{k}}, \psi_{\mathbf{k}}}_{\text{Photons}}, \underbrace{\bar{\phi}_{\mathbf{k}}, \phi_{\mathbf{k}}}_{\text{Density waves}}) e^{-S_{\text{eff}}[\psi,\phi]}$$

$$\frac{\delta S_{\text{eff}}}{\delta \psi_{\mathbf{k}}} = 0$$

$$\frac{\delta S_{\text{eff}}}{\delta \phi_{\mathbf{k}}} = 0$$

$$\psi_{k \neq 0} = 0$$

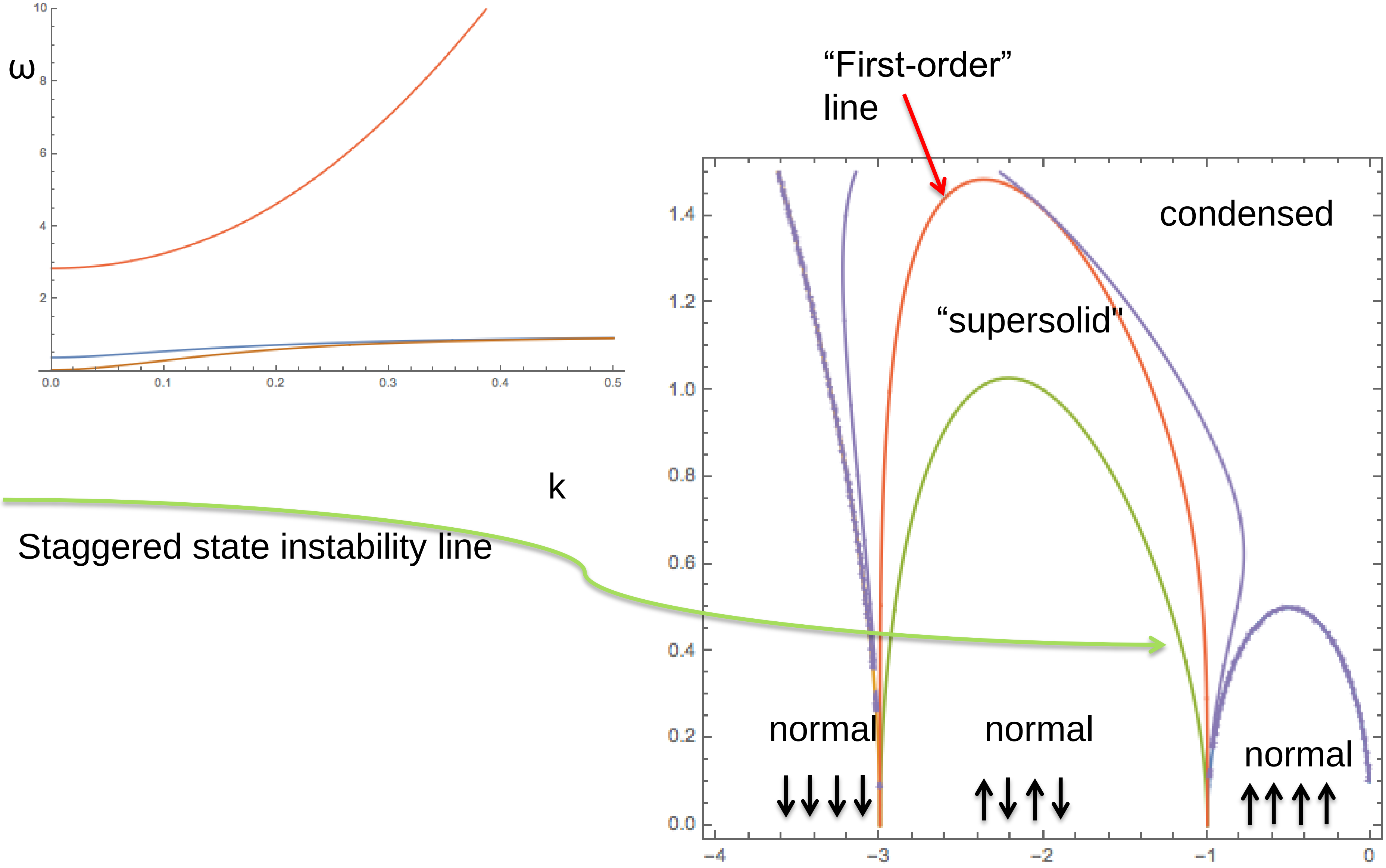
$$\phi_{k \neq 0, \pi} = 0$$



First-order transition

Coexistence of condensate and ordered phase "supersolid"

Staggered Stability Analysis



“First-order” line

condensed

“supersolid”

Staggered state instability line

normal

↓↓↓↓

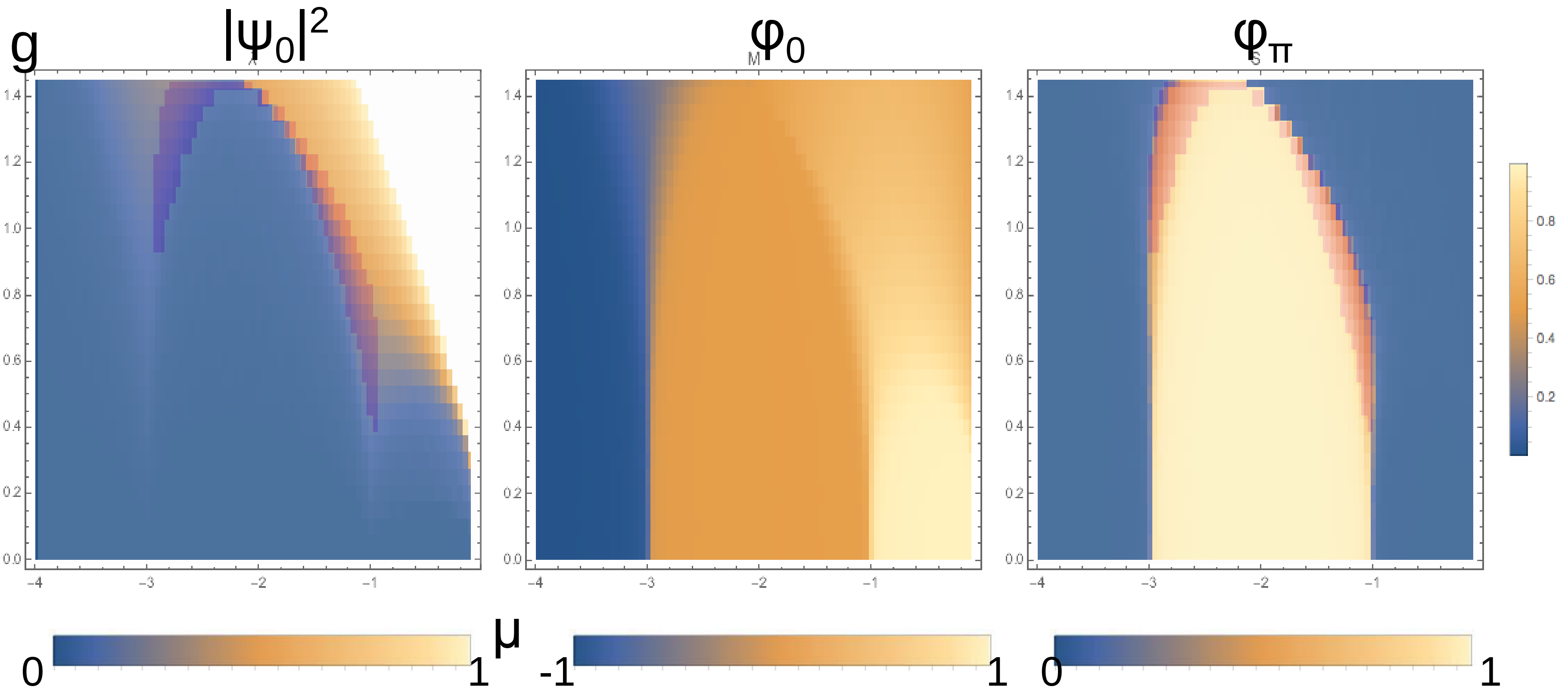
normal

↑↓↑↓

normal

↑↑↑↑

Variational Monte Carlo Data Confirm



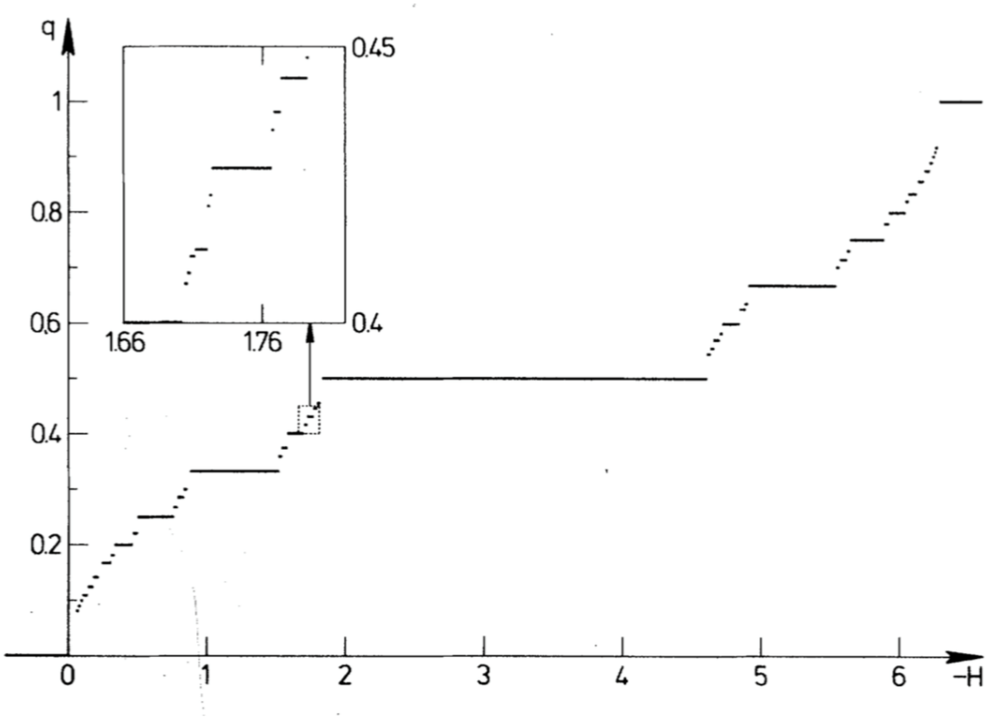
🟪 Coexistence

Long-range interactions

$U_{|i-j|} = U\ \delta_{<ij>} \longrightarrow U_0/|i-j|^{\alpha}$

(α=6 for van der Waals forces in Rydberg atoms)

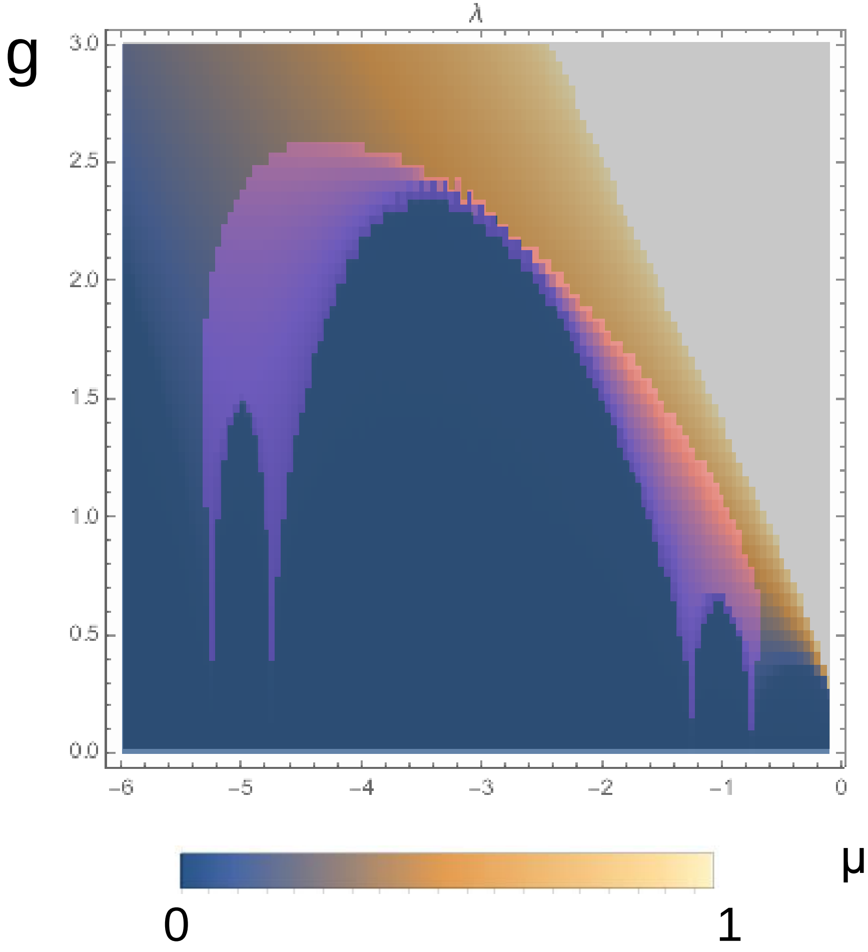
$= U_0$ (n.n.) $+U_0/2$ (n.n.n.) +…

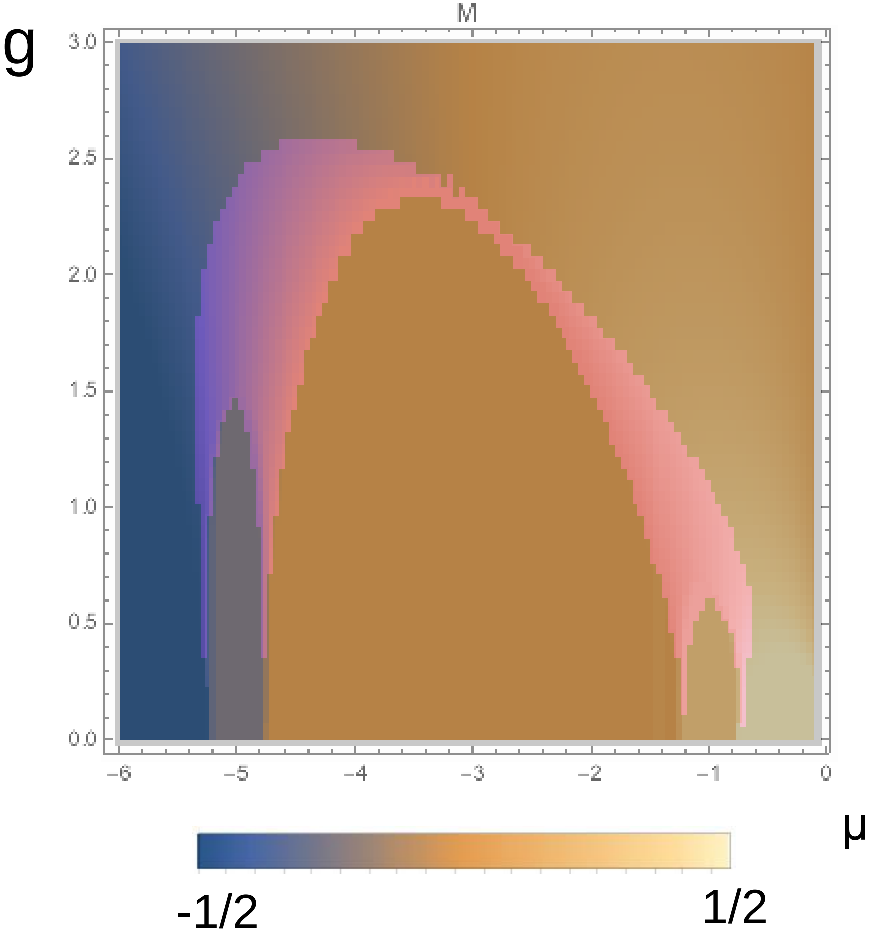


g = 0 ground state:
Complete Devil’s Staircase
(Bak and Bruinsma 1982)

Long-range interactions

g > 0: half-filling story repeats “self-similarly” at other fillings


$|\psi_0|^2$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‘Supersolid’ phase?

- Possibility of phase with both superfluid and charge order
- Has three acoustic modes (two sound and Bogoliubov)
- Has two amplitude modes (upper polariton and CDW amplitude mode)
- Amplitude modes mix; sound modes do not (not a gauge theory)
- Cold atom version of $NbSe_2$?
- Does this phase continue to be present without the lattice ?

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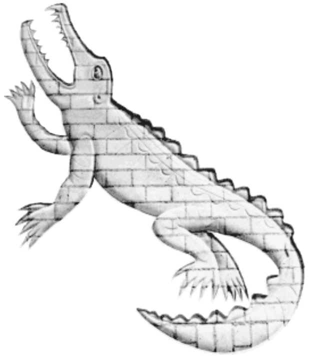

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University of Cambridge

Argonne NATIONAL LABORATORY

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The Seven Ages of Physicists

In *As You Like It* Shakespeare outlined the seven ages of man

From the infant,

to the schoolboy,

the lover,

the soldier and then ....

The Fifth Age

"And then the Justice,
In fair round belly with good capon lined,
With eyes severe, and beard of formal cut,
Full of wise saws and modern instances"