Some theory of polariton condensation and dynamics

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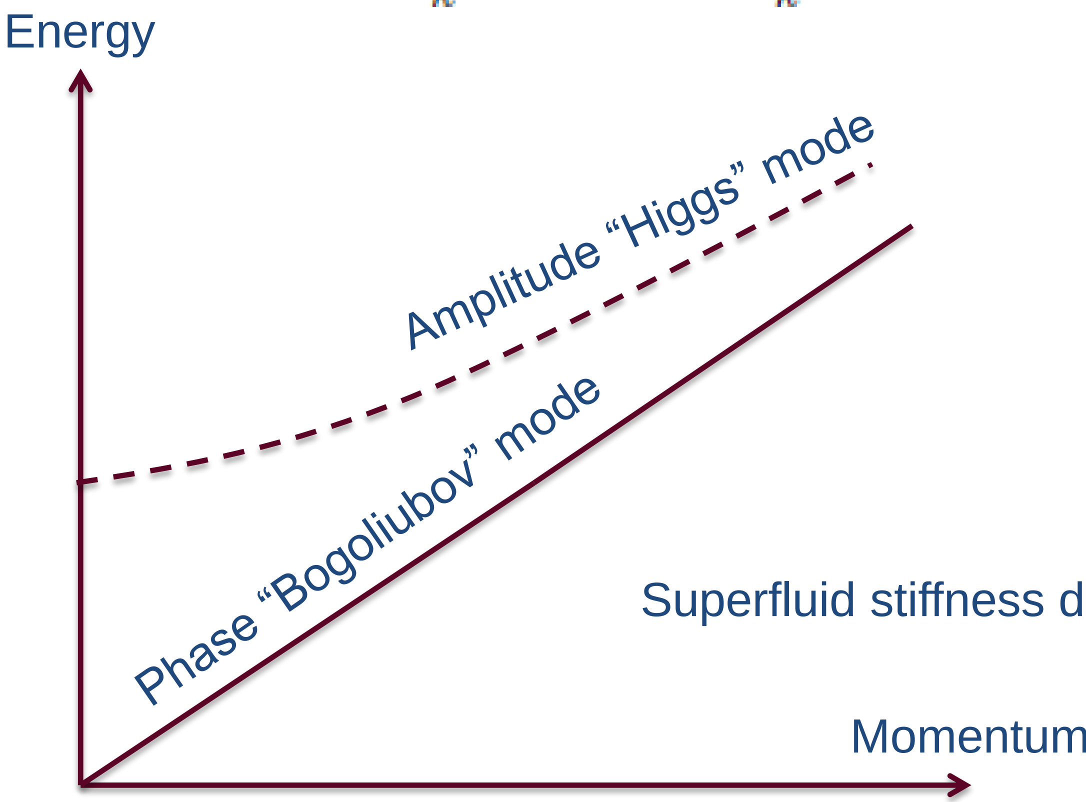
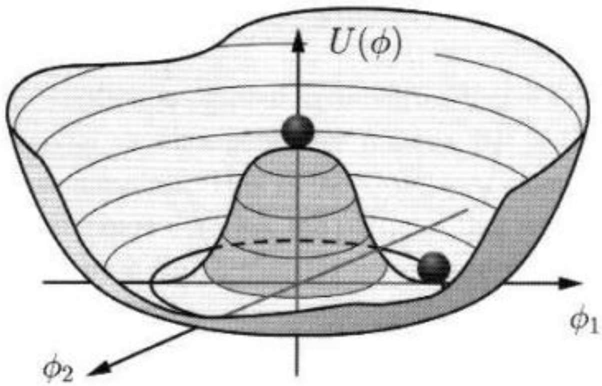
Vanilla theory of a condensate

BEC Superconductor Density wave Exciton

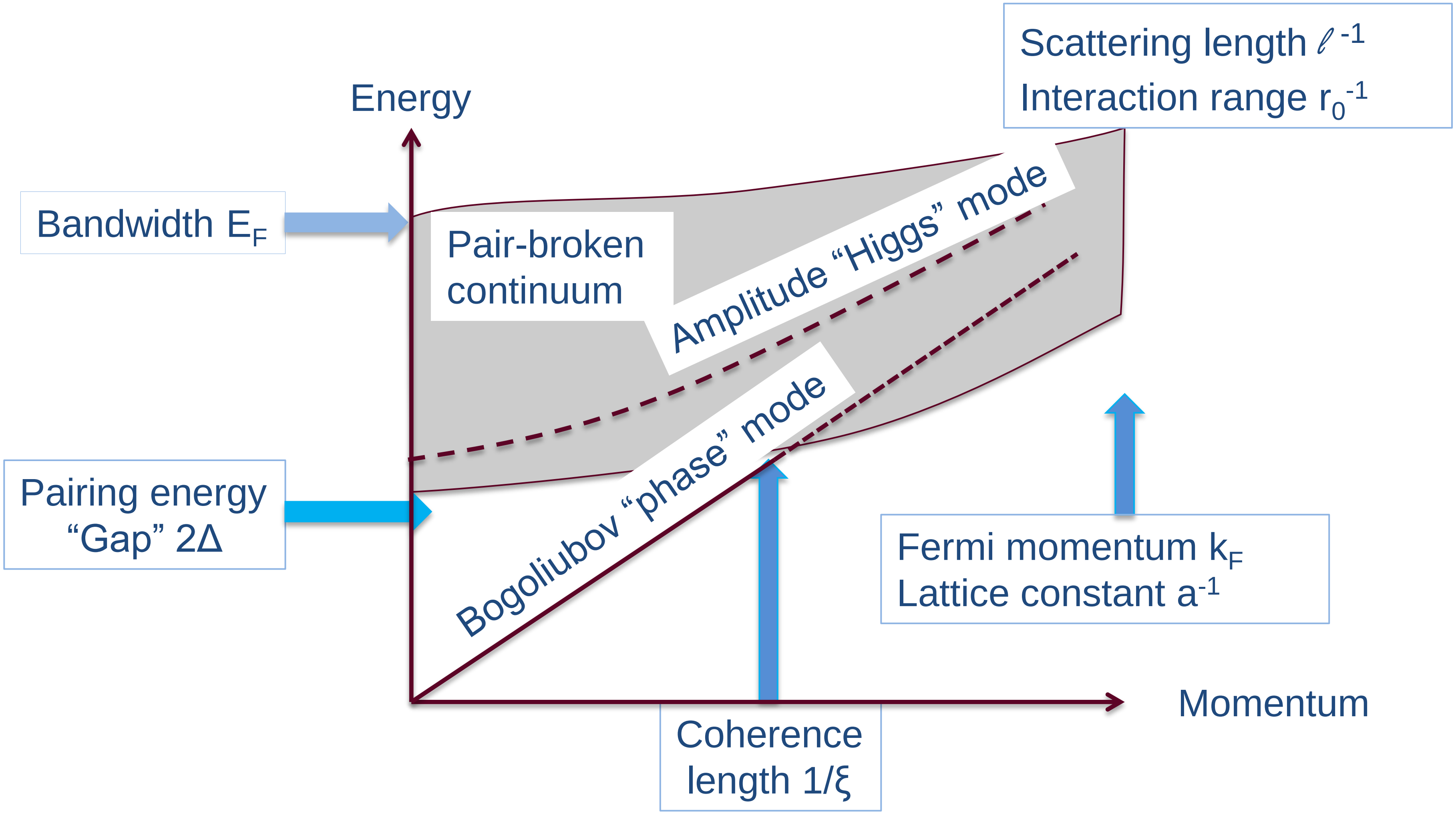
$$\langle \Psi \rangle = \langle b_0 \rangle = \sum_k \langle c_k c_{-k} \rangle = \sum_k \left\langle c^\dagger_{k+Q} c_k \right\rangle = \sum_k \left\langle c^\dagger_{1,k} c_{2,k} \right\rangle = \Delta e^{i\phi}$$





Superfluid stiffness determines phase mode velocity

In reality, there are more scales



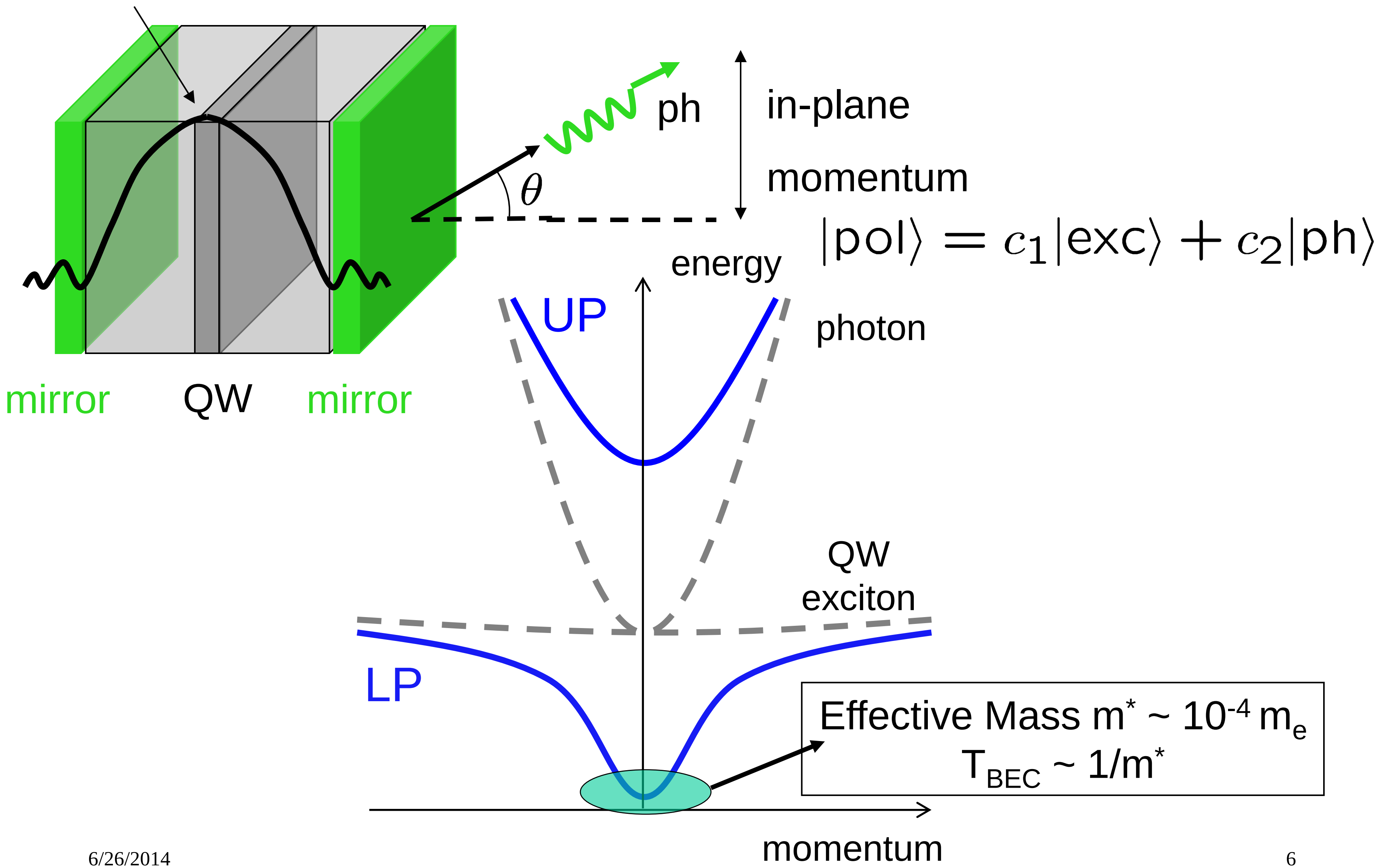
Outline

- 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?
- Preliminary thoughts on cavity – coupled Rydberg atoms

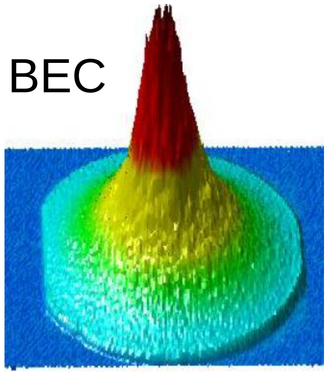
Polaritons: Matter-Light Composite Bosons

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

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

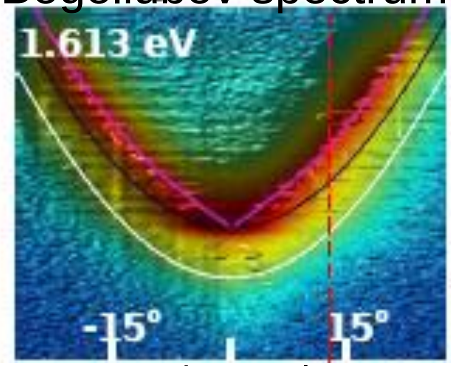


BEC



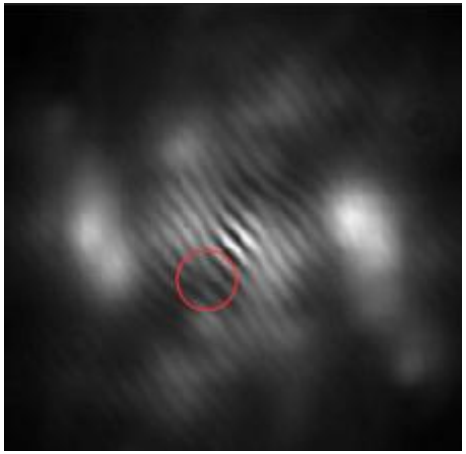
Kasprzak et al 2006

Bogoliubov spectrum



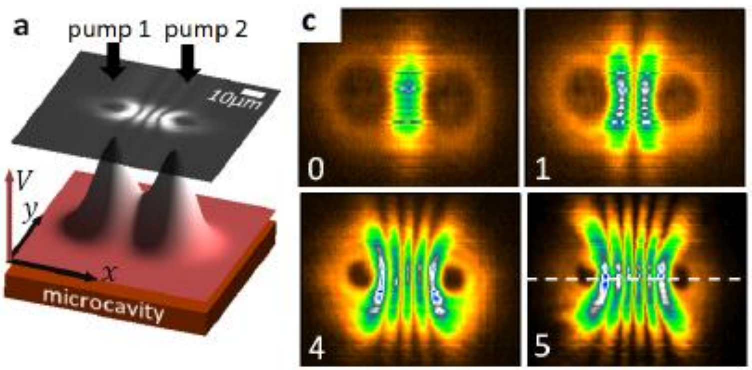
Utsunomiya et al,2008

Vortices



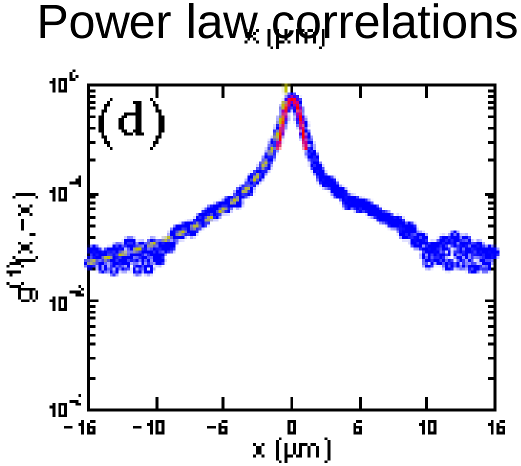
Lagoudakis et al 2008

Coupled condensates



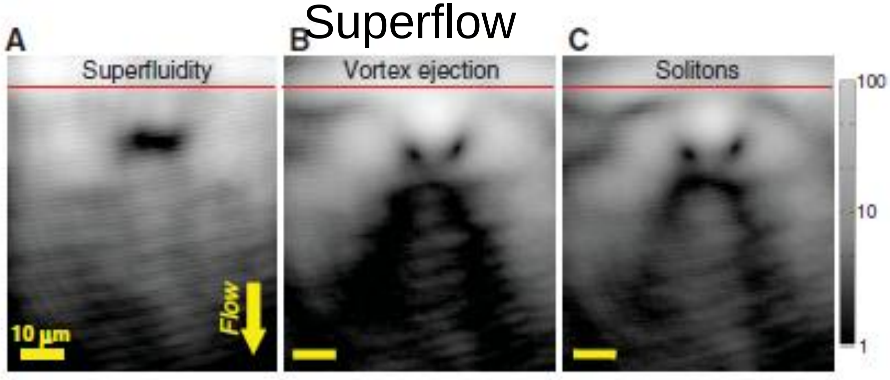
Tosi et al 2012

Power law correlations



Roumpos et al 2012

Superflow



Amo et al, 2011

What's new about a polariton condensate ?

- Composite particle – mixture of electron-hole pair and photon
- Extremely light mass (~ 10^{-5} m_e) means that polaritons are large, and overlap strongly even at low density
  - crossover from dilute gas BEC to coherent state, eventually to plasma
- Two-dimensional physics
  - Berezhinski-Kosterlitz-Thouless transition ?
- Polariton lifetime is short
  - Non-equilibrium, pumped dynamics leads to decoherence on long length scales

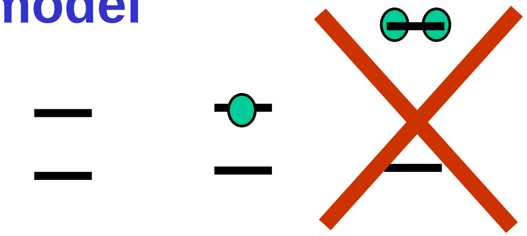
Excellent description by damped, driven Gross-Pitaevskii equation

Derivable from microscopic theory [see e.g. Keeling et al Semicond. Sci. Technol. 22 R1 (2007)]

- Can prepare out-of-equilibrium condensates
  - Quantum dynamics of many body system

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 $\gg$ 1

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

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

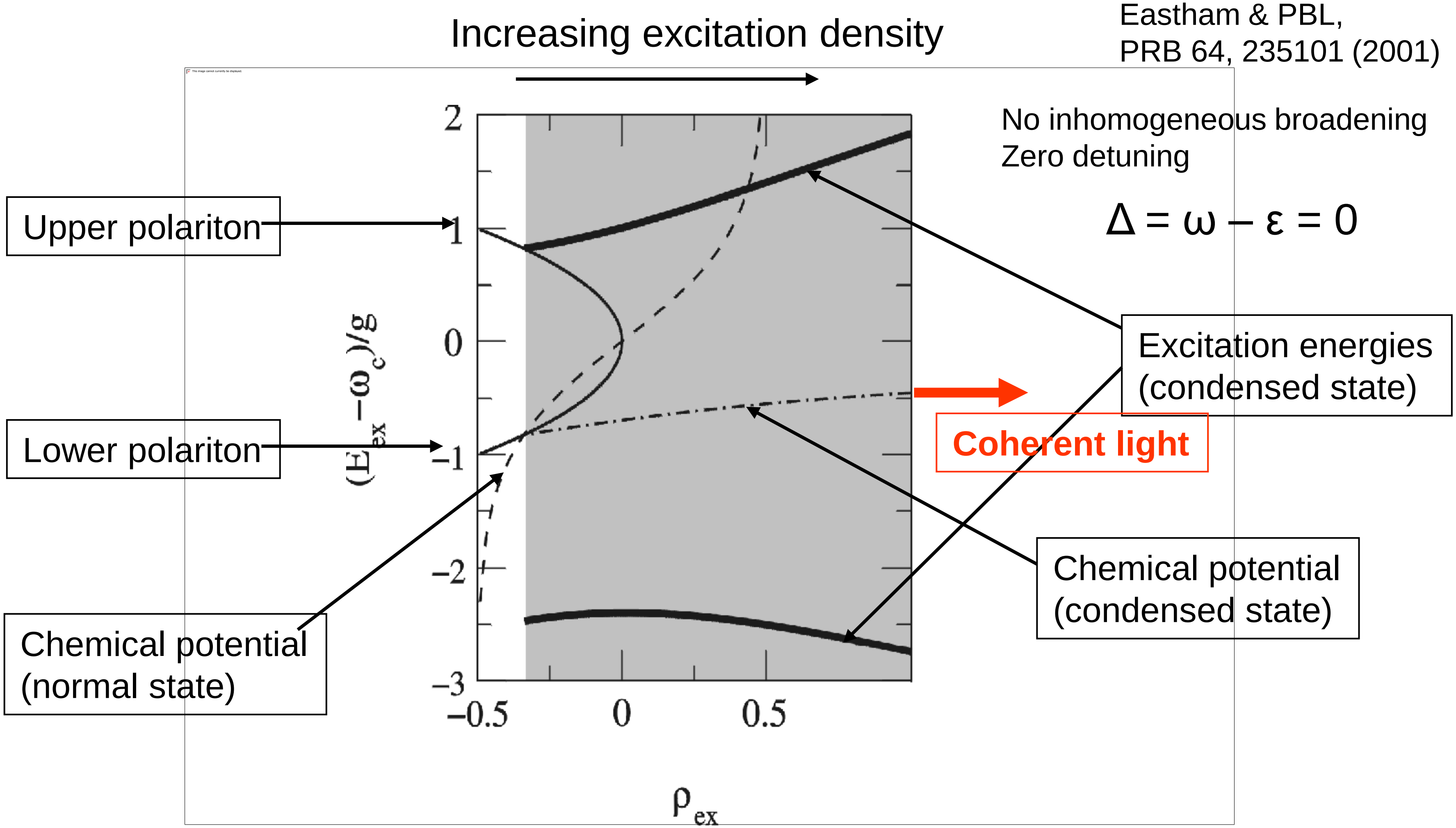
Transition temperature depends on coupling constant

Mean field theory of Condensation

Eastham & PBL,
PRB 64, 235101 (2001)

No inhomogeneous broadening
Zero detuning

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

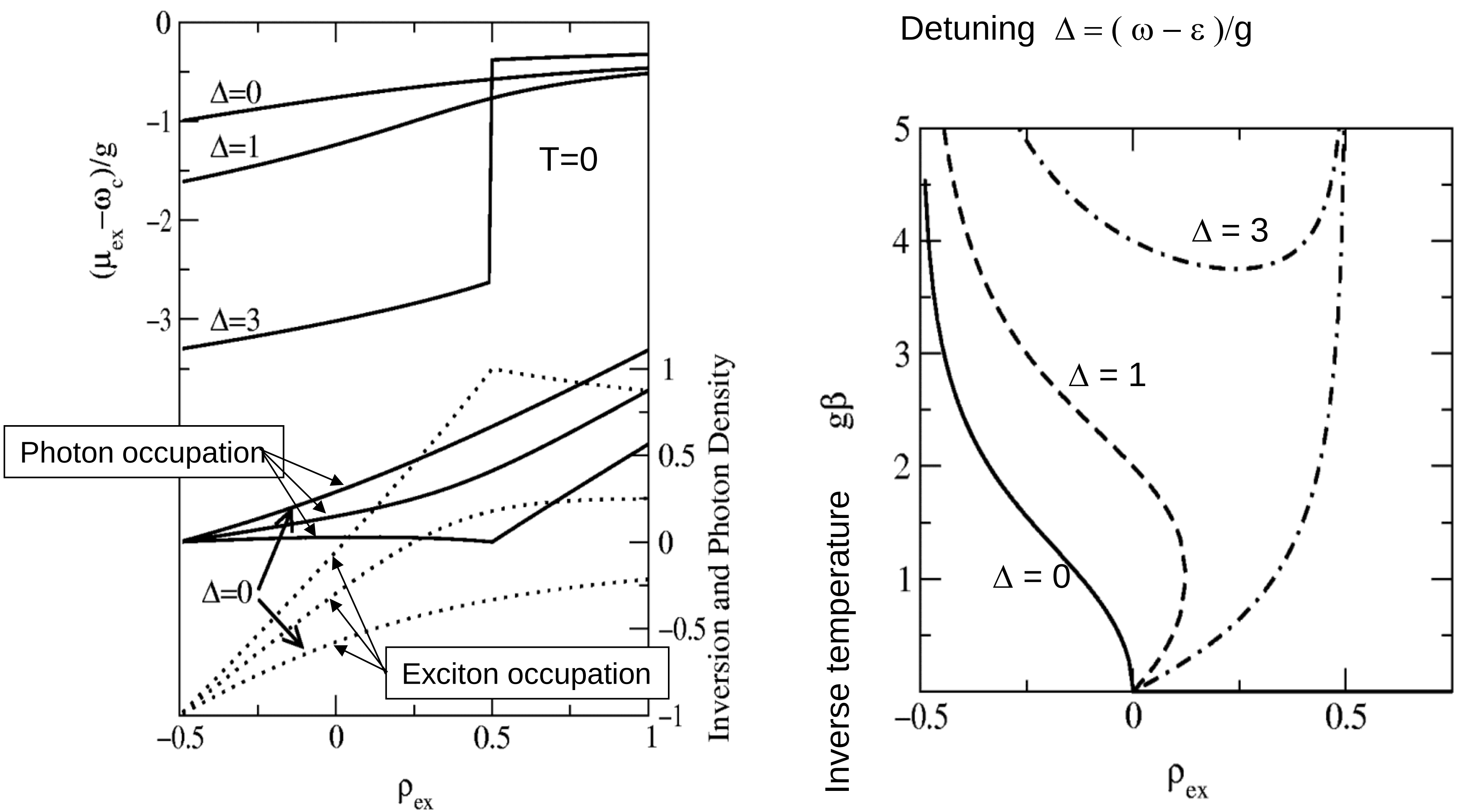


$$\rho_{ex} = \frac{1}{N}\left\langle \sum_i S_z^i + \psi^\dagger \psi \right\rangle = \frac{N_{photon} + N_{exciton}}{N} - \frac{1}{2}$$

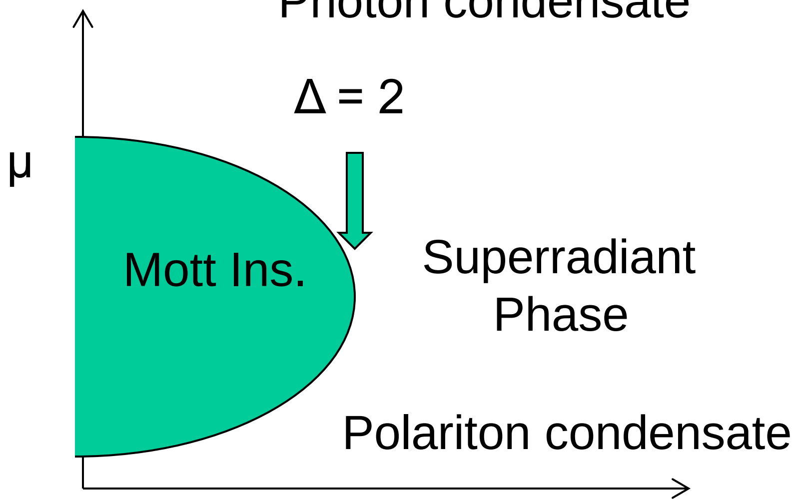
Phase diagram with detuning: appearance of “Mott lobe”

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

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

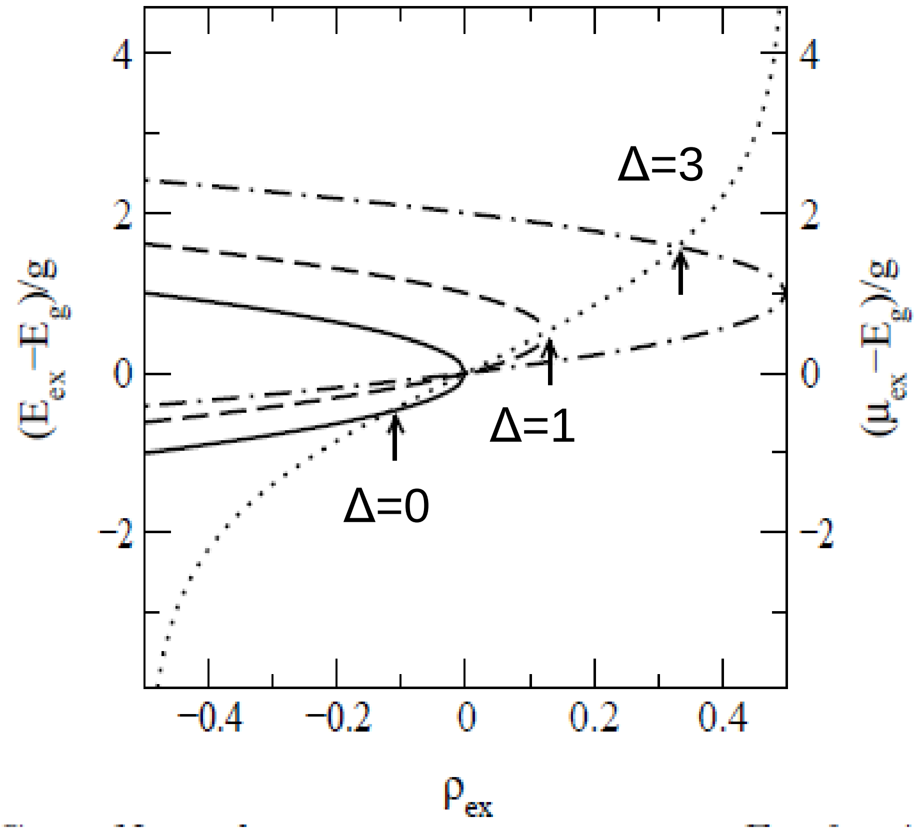


Single Mott Lobe for s=1/2 state



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

negative detuning

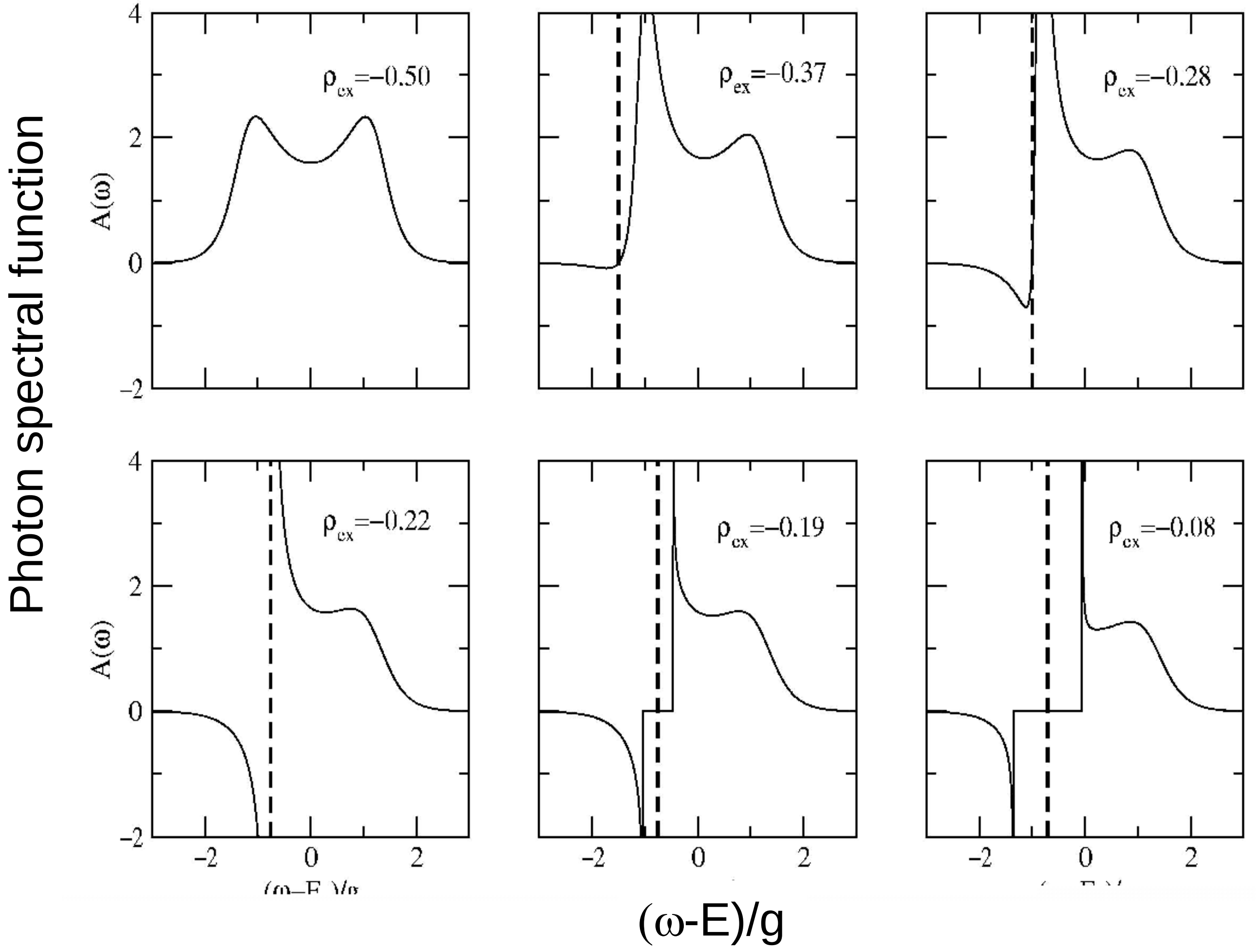


Eastham and Littlewood, Solid State Communications 116 (2000) 357--361

Excitation spectrum with inhomogeneous broadening

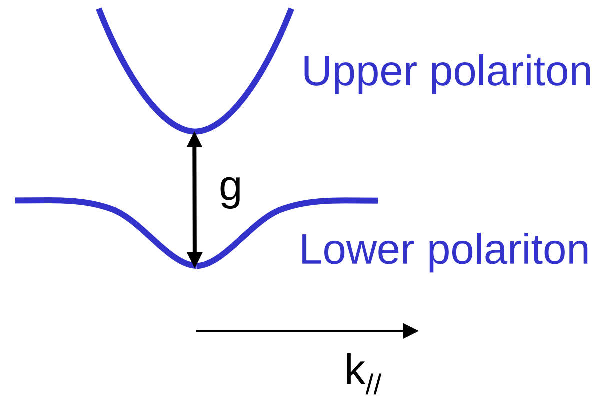
Zero detuning: $\omega = \varepsilon$

Gaussian broadening of exciton energies $\sigma = 0.5$ g



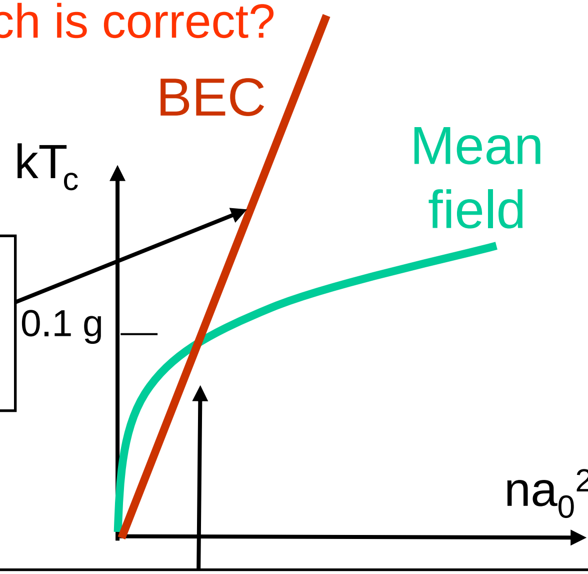
Beyond mean field: Interaction driven or dilute gas?

- 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

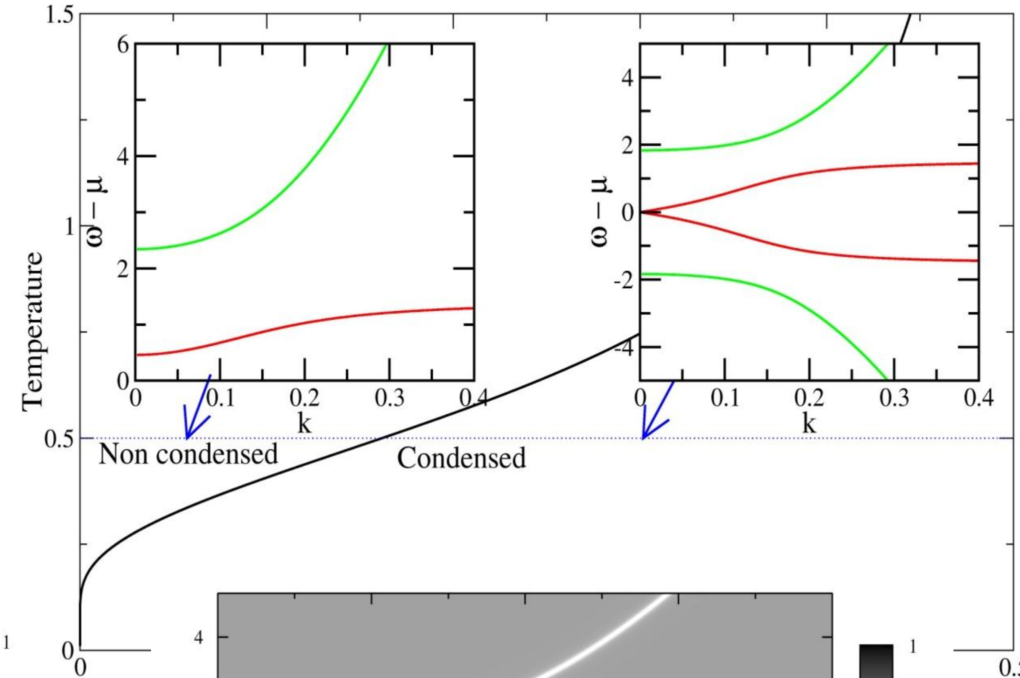
Dilute gas BEC only for excitation levels < 10^9 cm^{-2} or so

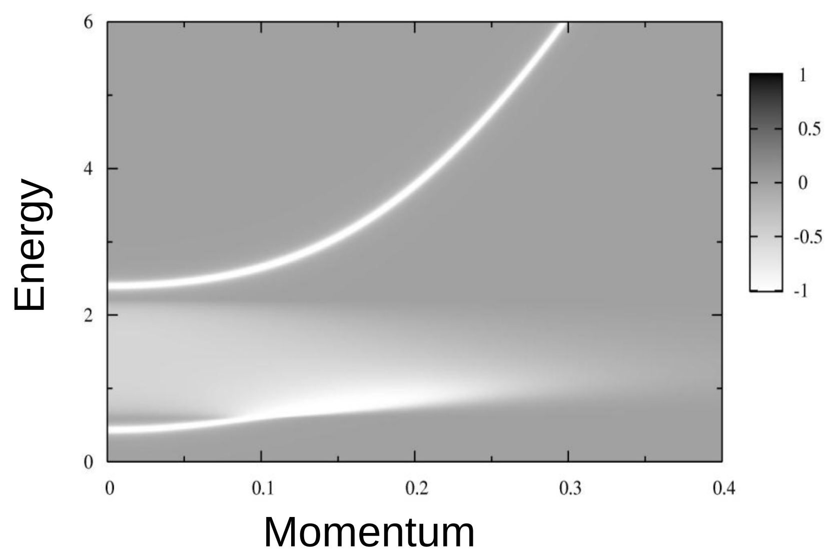
A further crossover to the plasma regime when $na_B^2 \sim 1$

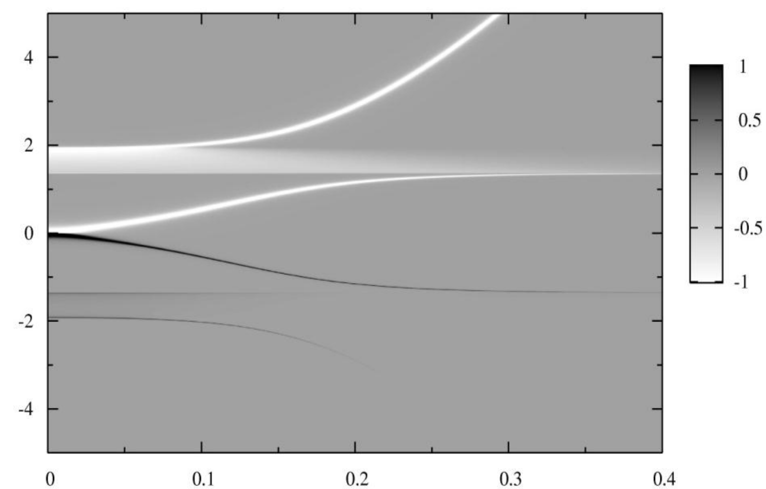
2D polariton spectrum

Keeling et al PRL 93, 226403 (2004)

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

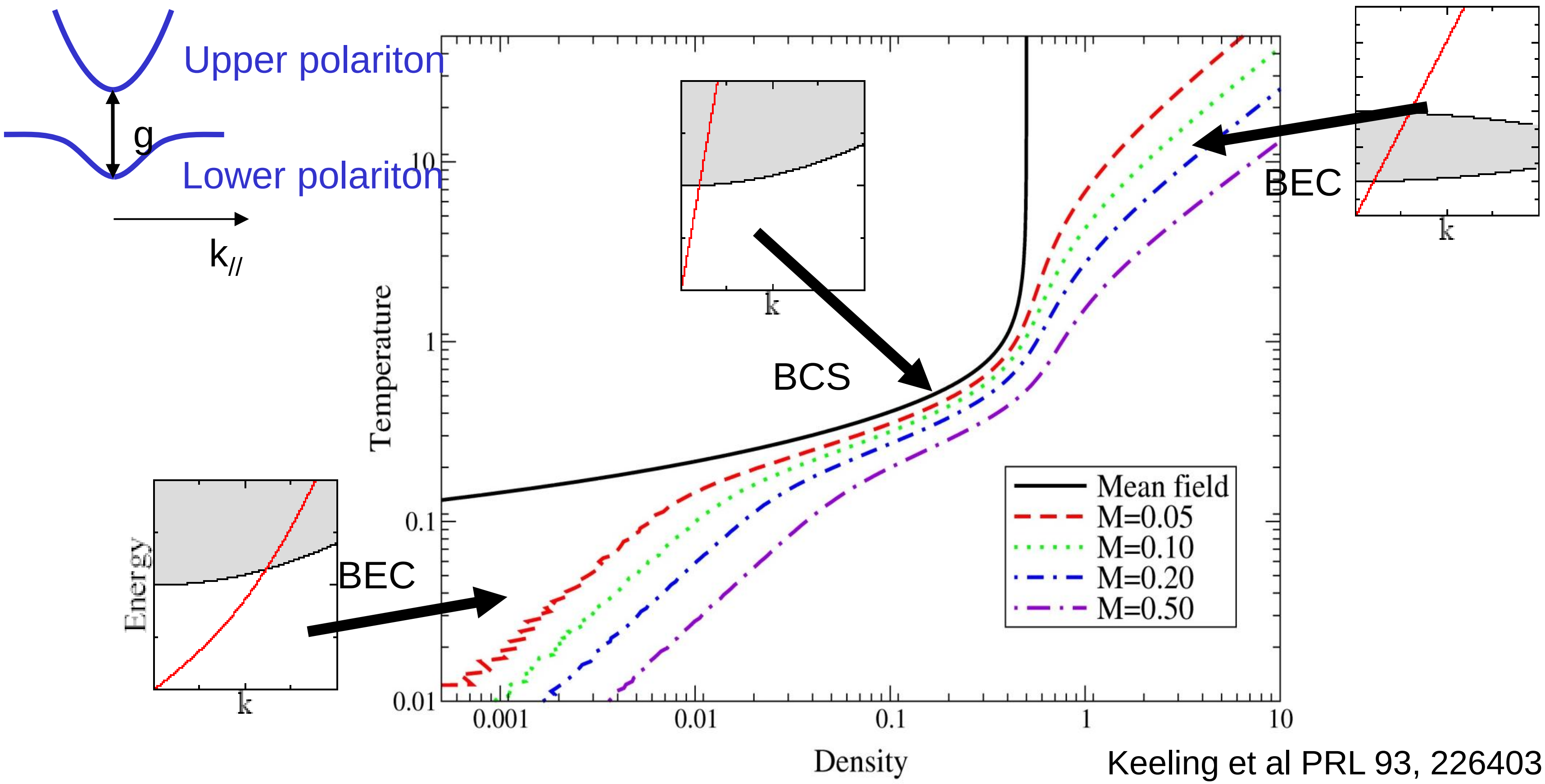






Phase diagram

- T_c suppressed in low density (polariton BEC) regime and high density (renormalised photon BEC) regimes
- For typical experimental polariton mass ~ 10^{-5} deviation from mean field is small

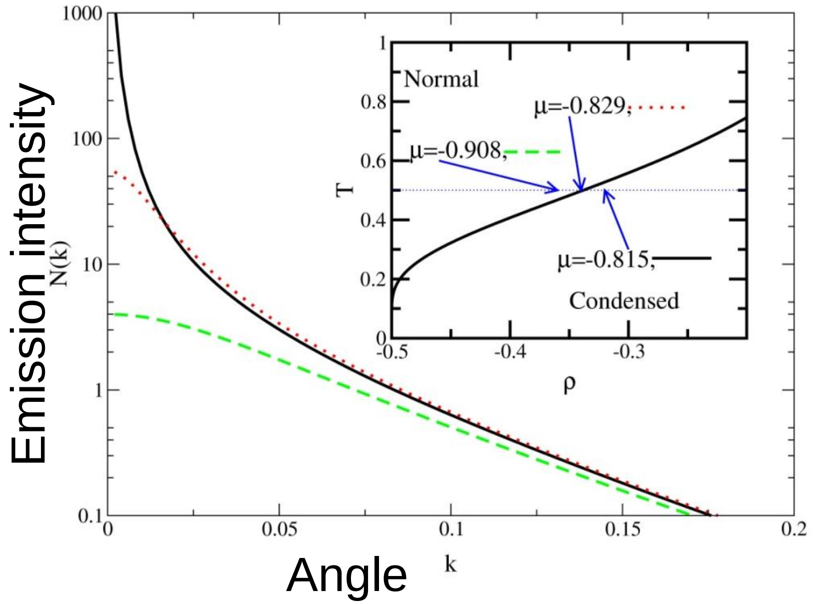


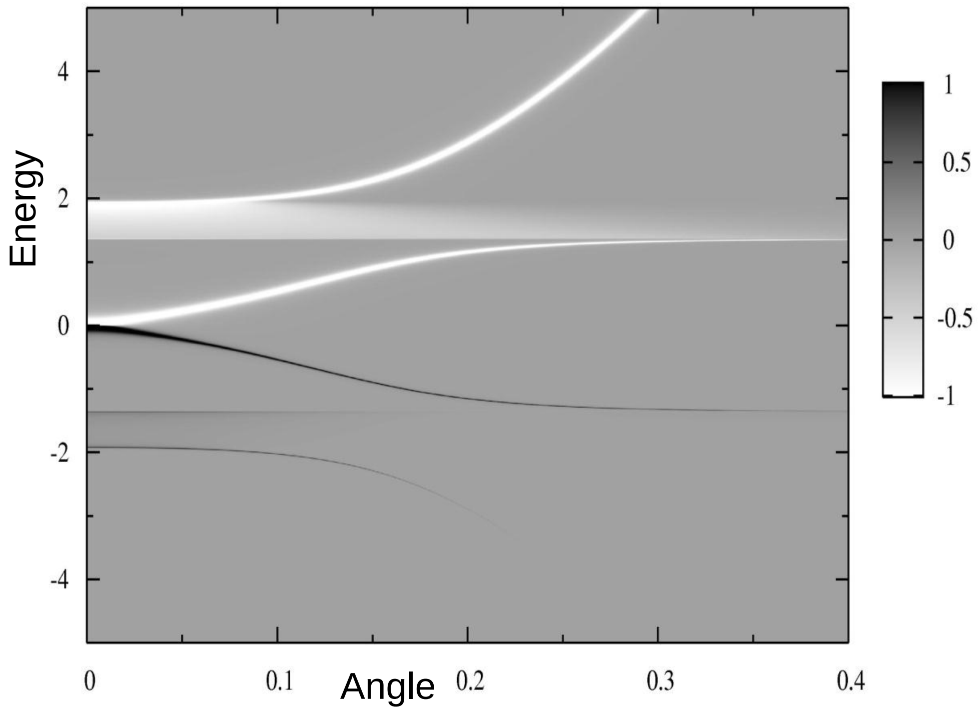
Keeling et al PRL 93, 226403 (2004)

Excitation spectra in 2D microcavities with coherence

Keeling, Eastham, Szymanska, PBL PRL 2004

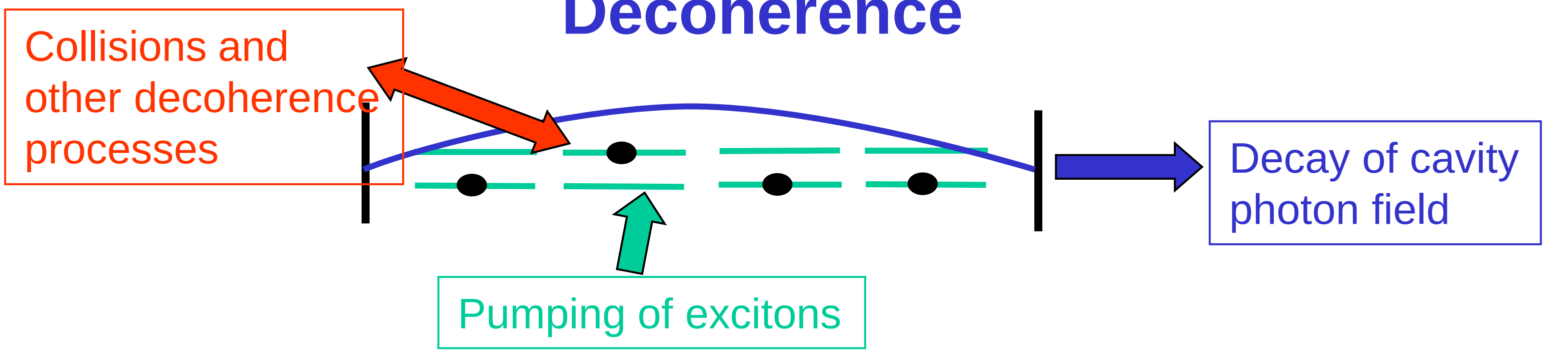
Angular dependence of luminescence becomes sharply peaked at small angles
(No long-range order because a 2D system)





Absorption(white) / Gain(black) spectrum of coherent cavity

Decoherence



Decay, pumping, and collisions may introduce "decoherence" - loosely, lifetimes for the elementary excitations - include this by coupling to bosonic "baths" of other excitations

in analogy to superconductivity, the external fields may couple in a way that is "pair-breaking" or "non-pair-breaking"

$$\sum_{i,k} g^{(1)}_{i,k} \left[b_i^\dagger b_i - a_i^\dagger a_i \right] (c^\dagger_{1,k} + c_{1,k})$$ non-pairbreaking (inhomogeneous distribution of levels)

$$\sum_{i,k} g^{(2)}_{i,k} \left[b_i^\dagger b_i + a_i^\dagger a_i \right] (c^\dagger_{2,k} + c_{2,k})$$ pairbreaking disorder

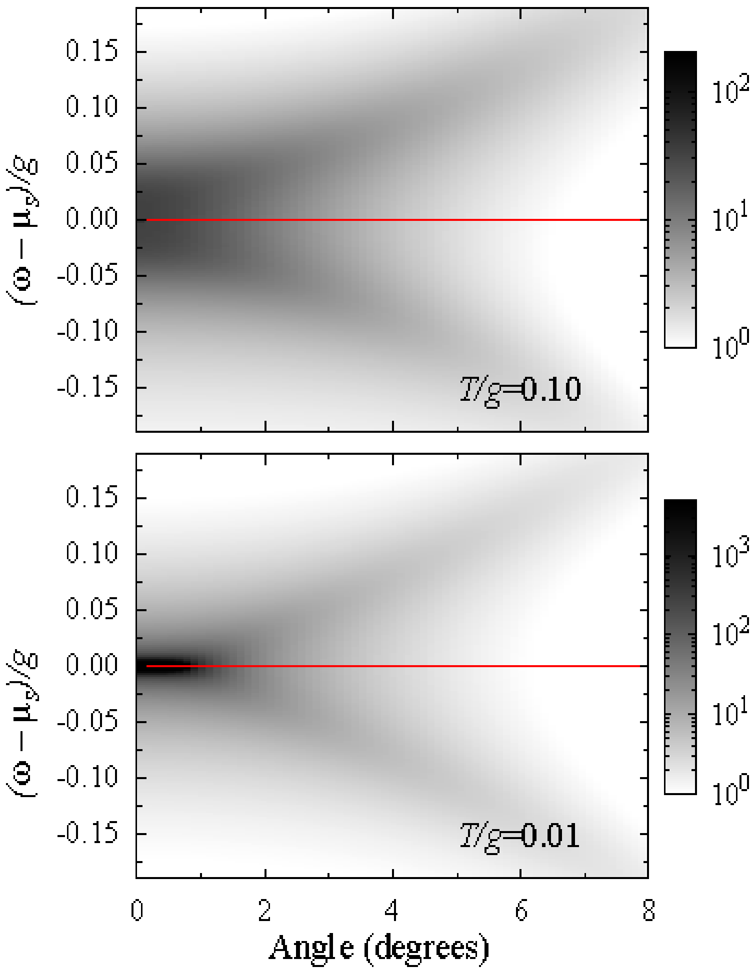
- *Conventional theory of the laser assumes that the external fields give rise to rapid decay of the excitonic polarisation - incorrect if the exciton and photon are strongly coupled*
- *Correct theory is familiar from superconductivity - Abrikosov-Gorkov theory of* superconductors with magnetic impurities

$$\sum_{i,k} g^{(3)}_{i,k} \left[b_i^\dagger a_i c^\dagger_{3,k} + a_i^\dagger b_i c_{3,k} \right]$$ symmetry breaking – XY random field destroys LRO

Generic response for "flat" bath spectrum

Szymanska et al, PRB 75, 195331 (2007)

Calculated spectrum



Phase correlation function

$$D_{\phi\phi}(\omega, p) = \frac{C}{\omega^2 - c^2p^2 + 2i\omega\gamma}$$

Overdamped mode at long wavelengths

$$\omega = -i\gamma \pm i\sqrt{\gamma^2 - c^2p^2}$$

Correlation function is power law

$$g_1 \propto e^{-f(r,\tau)}$$

$$f = \eta \ln(r/\xi) \quad r \to \infty \;, \tau \simeq 0$$

$$f = \frac{\eta}{2} \ln(c^2 t/\gamma\xi^2) \quad r \simeq 0 \;, \tau \to \infty$$

η measures "strength" of noise, and 1/ξ the momentum cutoff

$$1/\xi \simeq E_{max}/c \simeq k_B T_{eff}/c$$

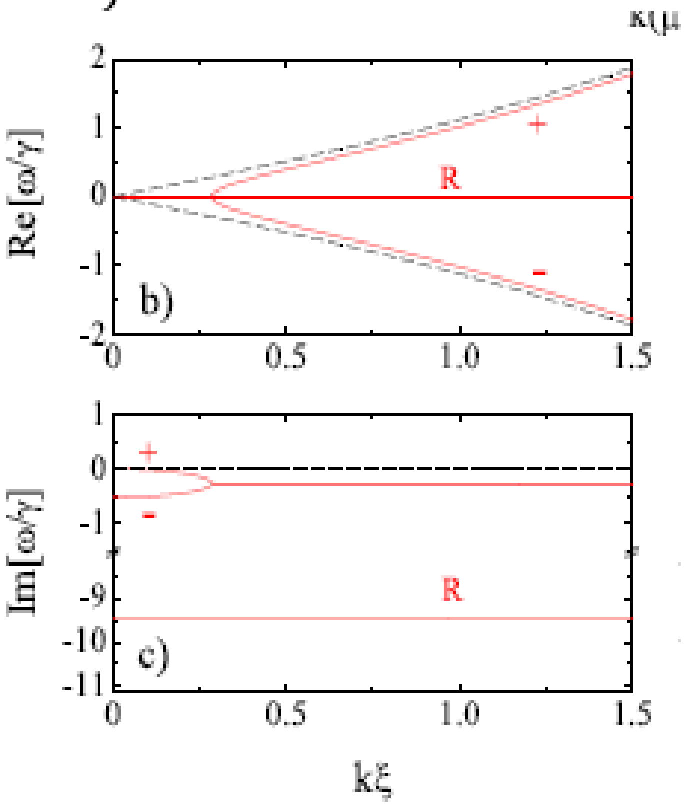
Damped, driven Gross-Pitaevski equation

- Microscopic derivation consistent with simple behavior at long wavelengths for the condensate order parameter ψ and polariton density n_R

$$i\frac{\partial \psi}{\partial t} = \left\{ -\frac{\hbar \nabla^2}{2m_{LP}} + \frac{i}{2}\left[R(n_R) - \gamma\right] + g\left|\psi\right|^2 + 2\tilde{g}\, n_R \right\} \psi.$$

$$\frac{\partial n_R}{\partial t} = P - \gamma_R\, n_R - R\left(n_R\right)\left|\psi\left(x\right)\right|^2 + D\nabla^2 n_R.$$

$$\omega_{\pm}(k) = -\frac{i\Gamma}{2} \pm \sqrt{\omega_{Bog}(k)^2 - \frac{\Gamma^2}{4}},$$



From Wouters and Carusotto, Phys. Rev. Lett. 99, 140402 (2007)
Keeling and Berloff Phys. Rev. Lett. 100, 250401 (2008)

Driven dynamics

On time scales < few psec, not in thermal equilibrium

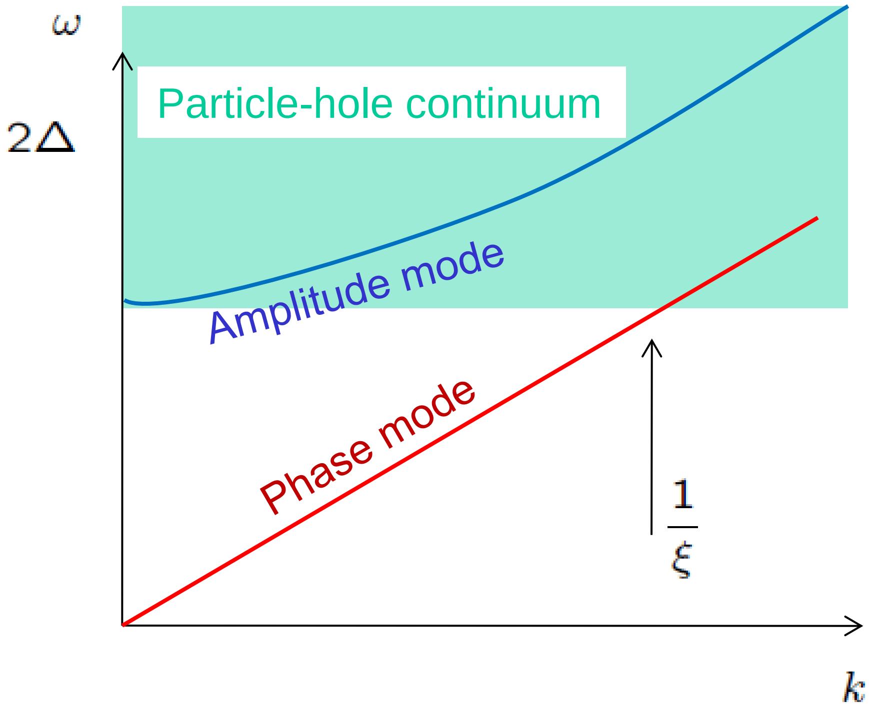
Coupling to light allows driven dynamics

Collective dynamics

- Use pump pulse to prepare non-equilibrium population
- Follow dynamics (typically on time scales faster than dephasing times)
- Similar to a “quantum quench” – where parameters of the Hamiltonian are abruptly changed
- Project an initial state onto the exact (time-dependent) eigenstates:
  - If the perturbation is small, expect to see a linear superposition of a few excitations – separate into single-particle like, and collective (e.g. Phase/amplitude)
  - Large?

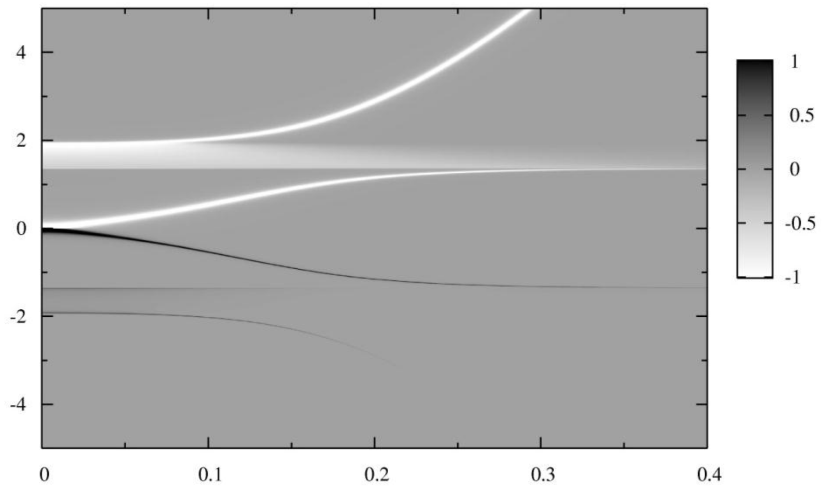
Compare condensed polaritons to superconductor

BCS s-wave superconductor



NB $2\Delta/E_F \ll 1; \quad k_F\xi \ll 1$

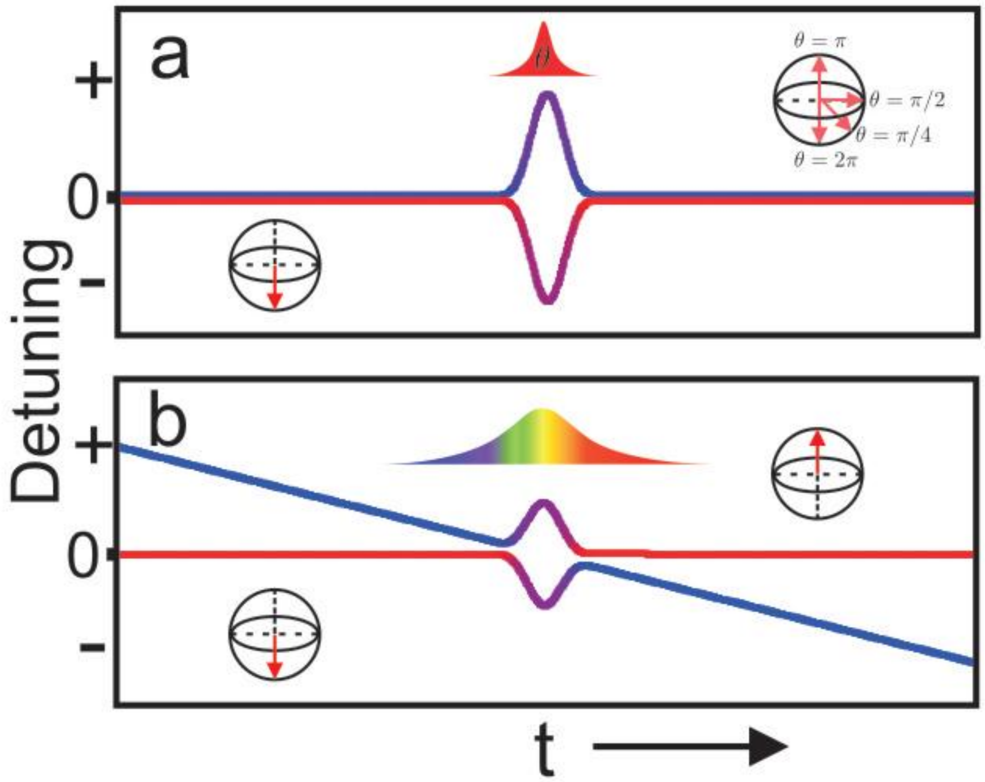
Polaritons



Phase mode – LP
Amplitude mode – UP
Continuum – inhom. broadening

Keeling 2006

Adiabatic Rapid Passage on two level system

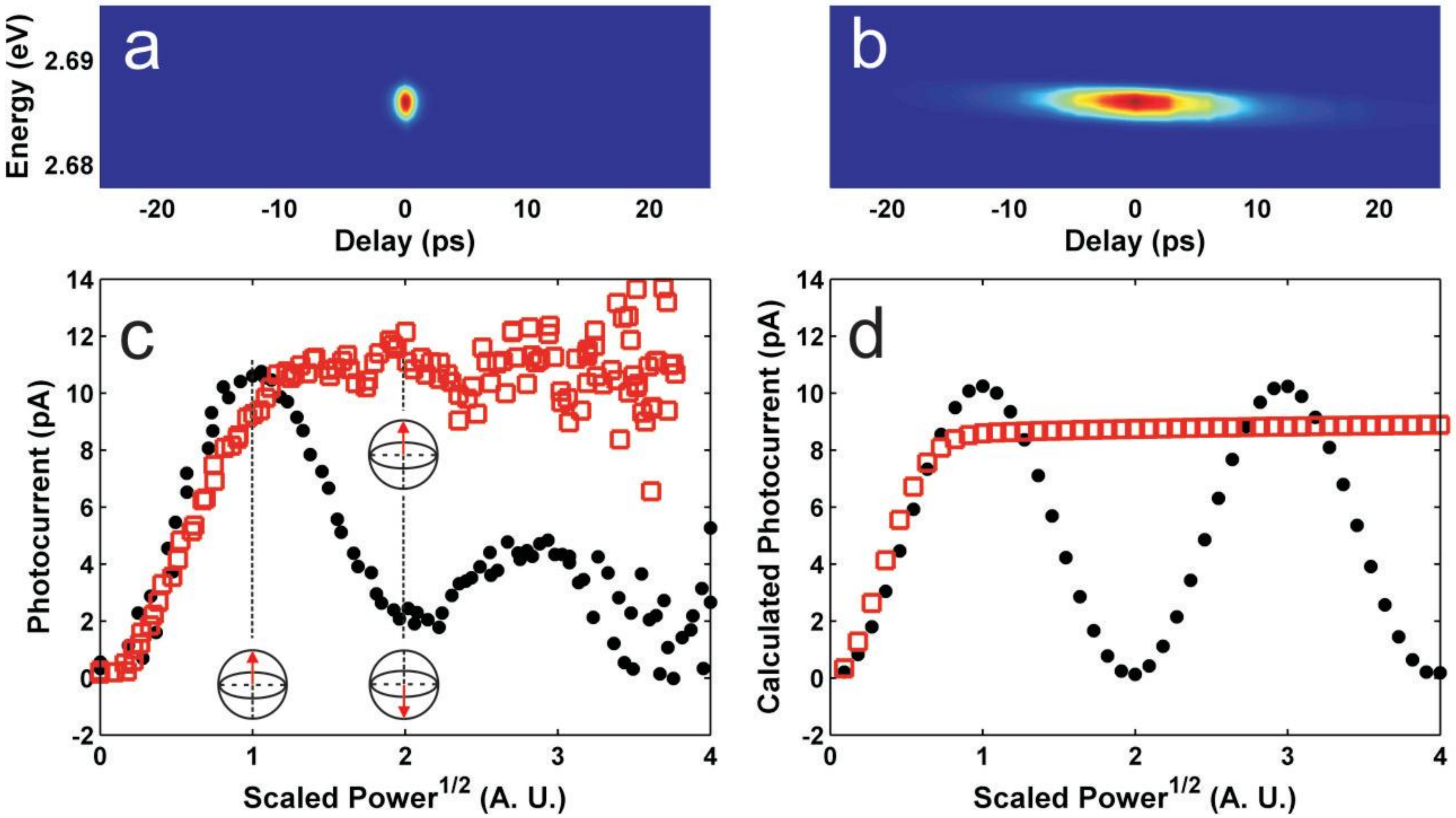


Conventional Rabi flopping requires accurate pulse areas

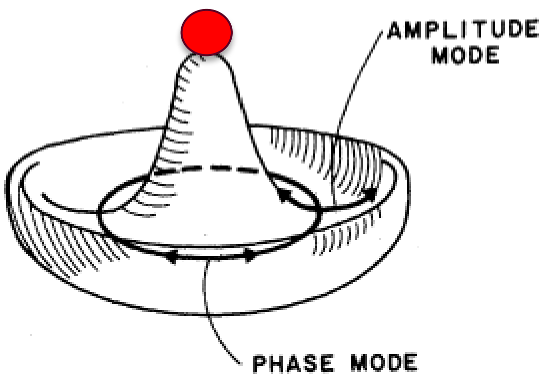
- Chirped pulse produces anticrossing of levels
- Weight of wavefunction transfers from one state to the other
- Robust population inversion independent of pulse area

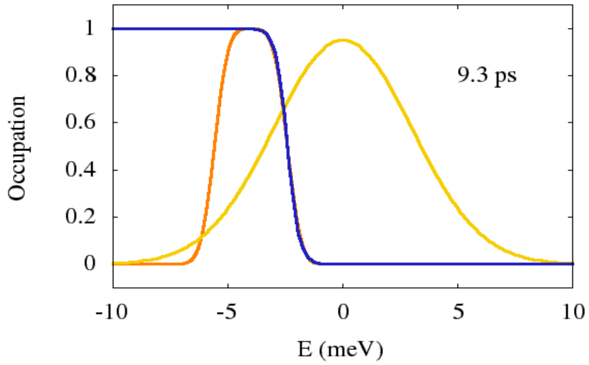
Single Dot Experiment

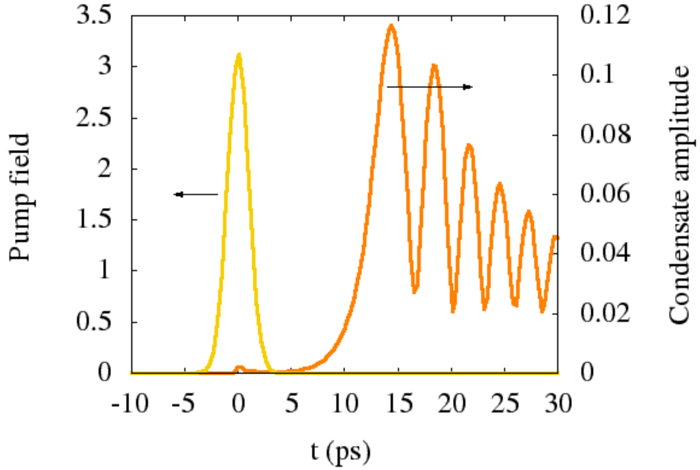
Wu et al, PRL 106 067401 (2011)



Spontaneous dynamical coherence







Pump generates non-eq. distribution of excitons without coherence

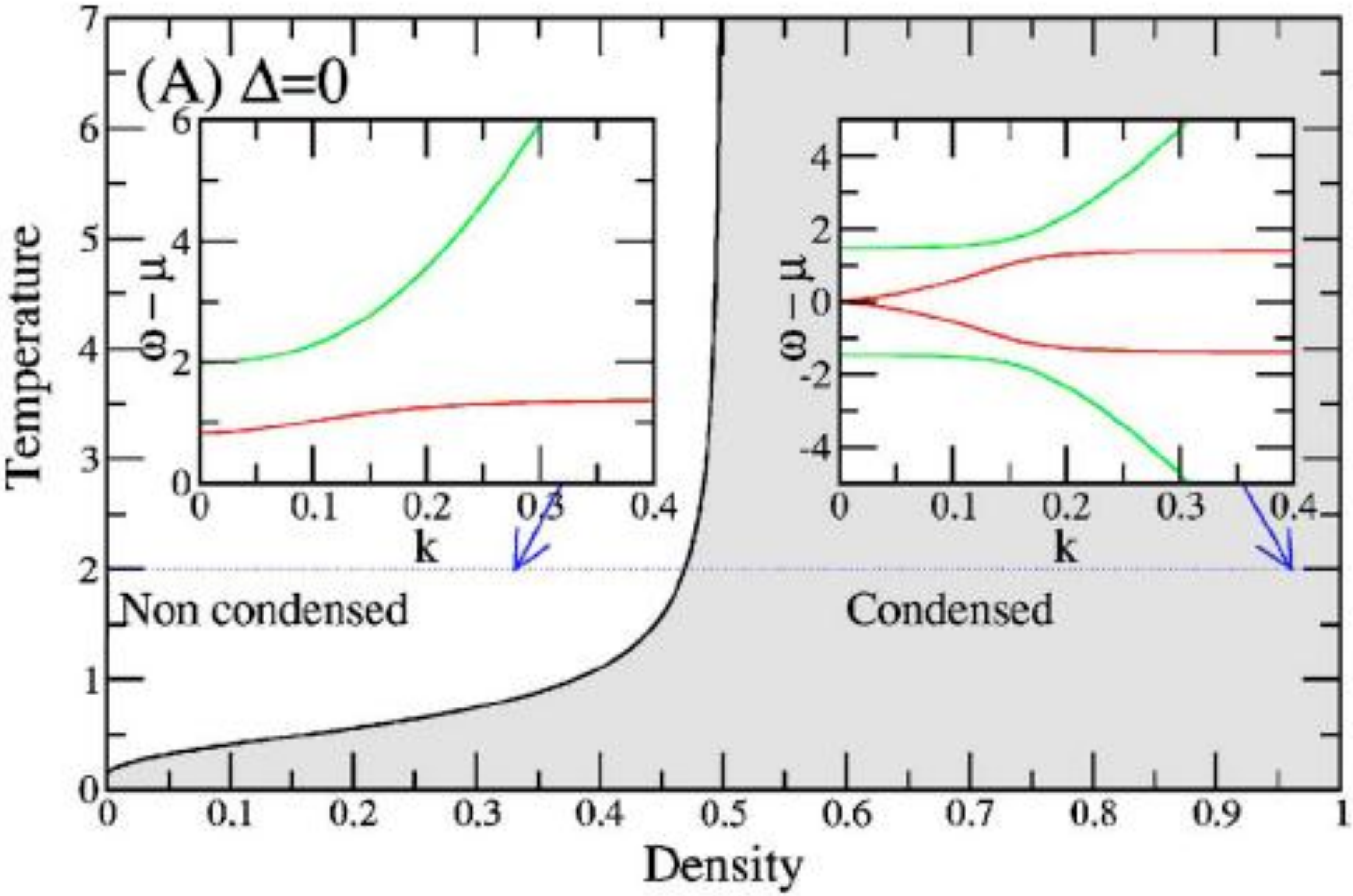
$\langle P_{k=0} \rangle$
$\langle \psi_{k=0} \rangle$ are macroscopic - scaling with $N^{1/2}$

$\Rightarrow$ A condensate of both photons and k=0 excitons
$\Rightarrow$ Ringing produced by dynamical amplitude oscillations
$\Rightarrow$ Mean field assumed: i.e. keep only momenta of pump and k=0

Paul Eastham and Richard Phillips PRB 79 165303 (2009)

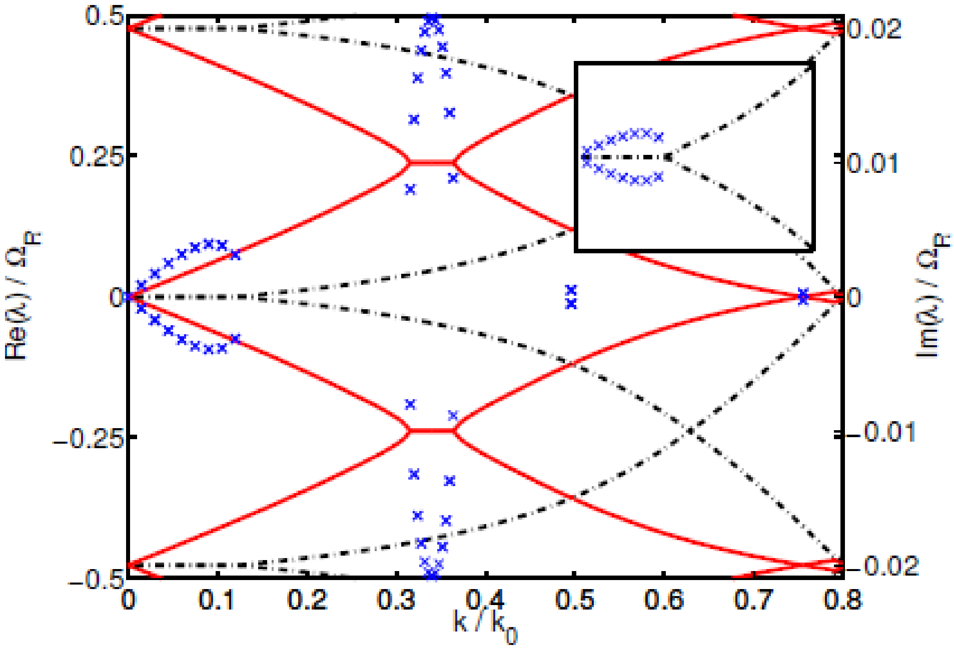
Full nonlinear semiclassical dynamics ....

Brierley et al., Phys. Rev. Lett. 107, 040401 (2011)



$\Omega_R,$

Quasienergy spectrum of oscillating system



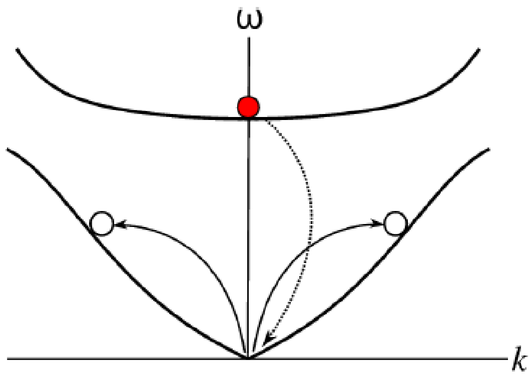
Red lines – derived from phase modes (LP)

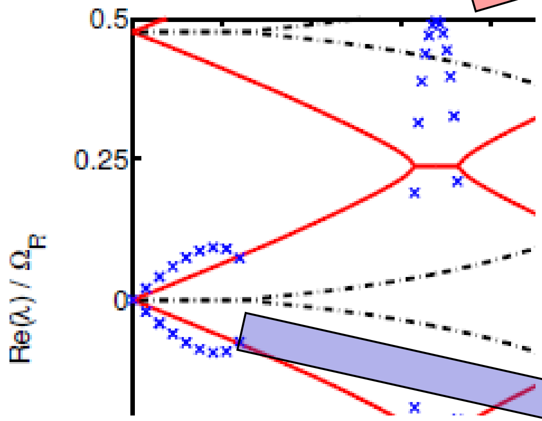
Black lines – amplitude modes (UP)

Unstable regimes when *Im λ* nonzero (Blue crosses)

Unstable regimes

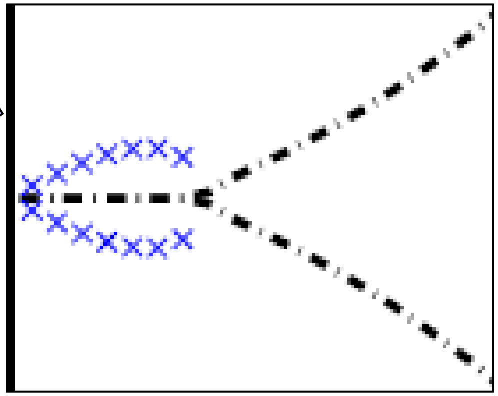
OPO – like instability
amplitude modes pump phase





Spectrum of dilute Bose gas with
weak attractive interactions

Attractive interaction between
amplitude fluctuations



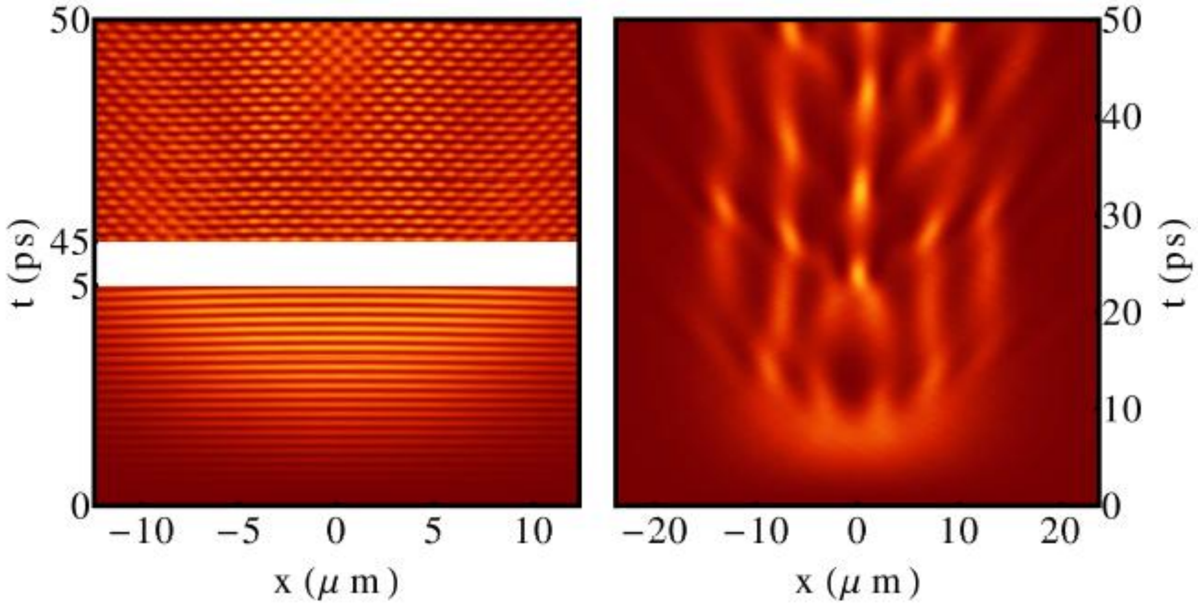
Ginzburg – Landau analysis

$$i\frac{\partial\psi}{\partial t} = \left(\omega_0 - \frac{\hbar^2}{2m_{\rm ph}}\nabla^2\right)\psi + \frac{\Omega_R}{2}\left(1-\lambda|P|^2\right)P$$
$$- i\gamma\psi + \xi + F,$$

$$i\frac{\partial P}{\partial t} = EP + \frac{\Omega_R}{2}(1-\lambda|P|^2)\psi.$$

Lower and upper polariton resonantly pumped

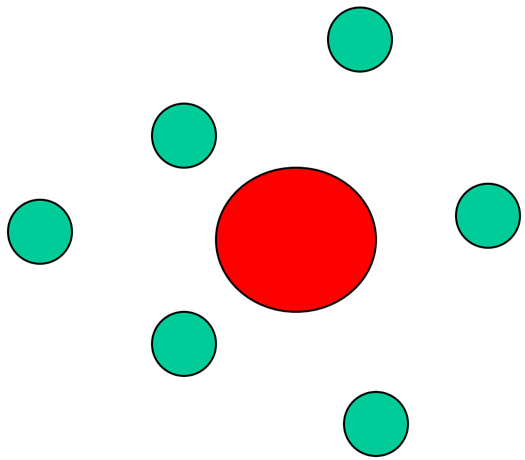
Upper polariton resonantly pumped



Long-wavelength instability appears to develop spatio-temporal chaos

Rydberg atom polaritons

Suppose the upper level of the exciton is an excited Rydberg state of an atom – has substantial long-range interactions $U(r) \sim 1/r^6$ with another excited atom



- Excited atom detunes neighbors from cavity resonance
- Favors inhomogeneous or crystalline state
- Competition between interactions and kinetic energy of cavity mode

Rydberg polaritons

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

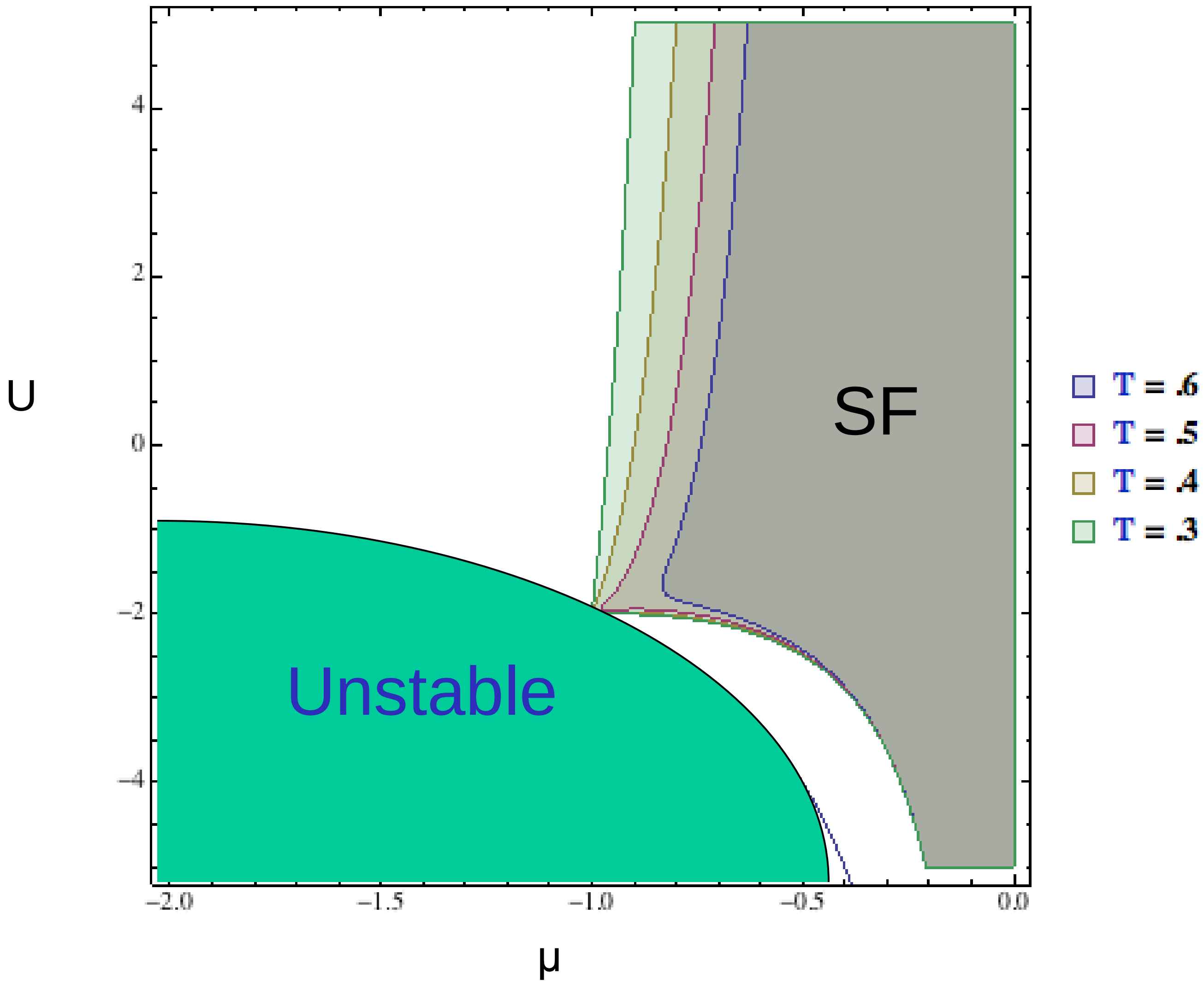
$$+ \sum_{i,j} a_i^\dagger a_i U(i-j) a_j^\dagger a_j \qquad\qquad a_i^\dagger a_i + b_i^\dagger b_i = 1$$

Represent exciton as two fermionic levels with a constraint of single occupancy

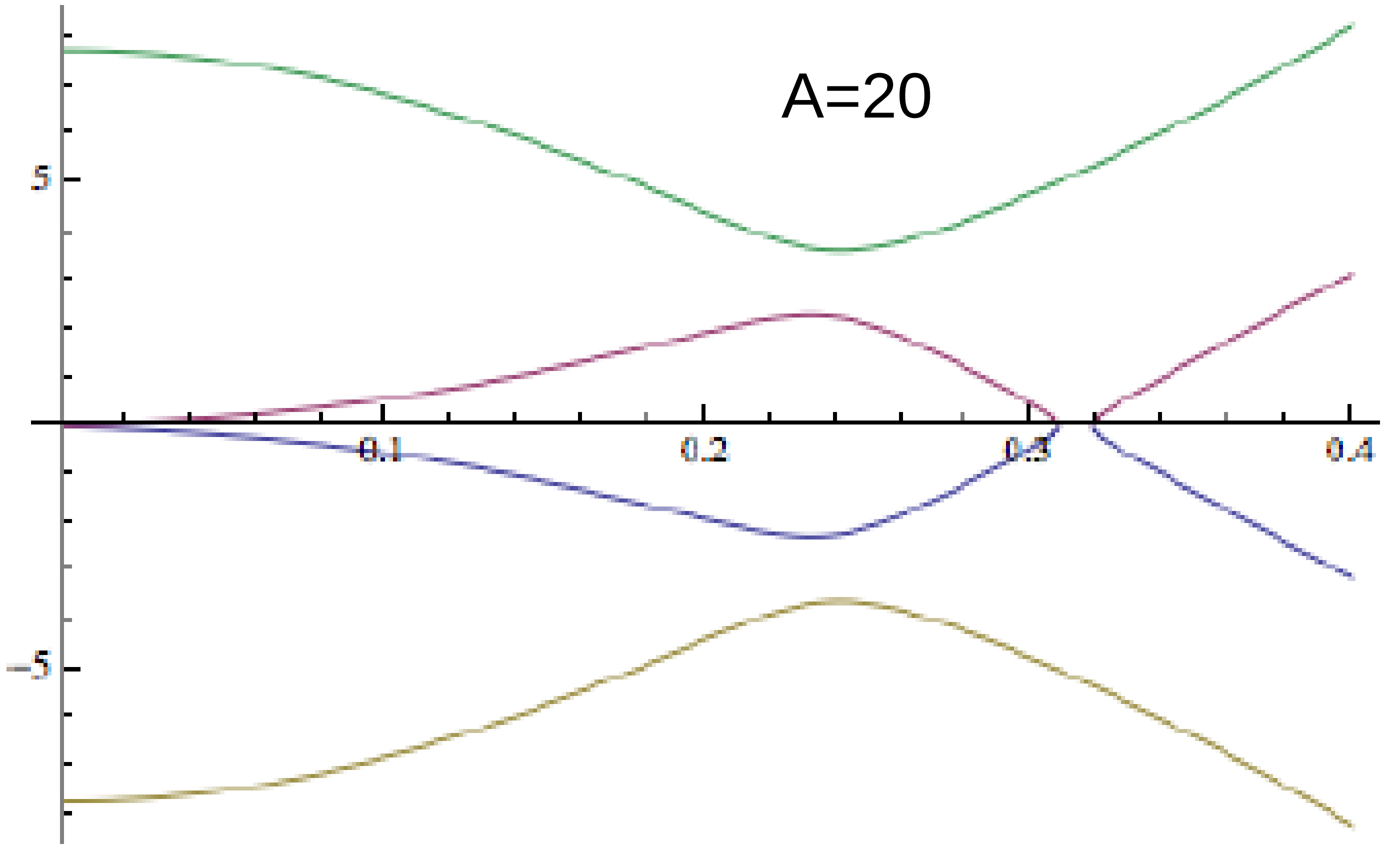
Consider instability of the superradiant polariton state.
No weak coupling instability if U(q) > 0
In strong coupling expect an effective interaction that generates a (short) length scale from the density itself.
Mixing of amplitude and phase modes only allowed at non-zero momentum.

See Zhang et al PRL 110, 090402 (2013)

Momentum-independent interaction U



Toy model: U=A cos (10q)



‘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 $NbSe_2$?
- Inhomogeneous phases?

Conclusions

Cavity polaritons are a new correlated many body system for “cold” “atoms” that show condensation phenomena analogous to BEC

- Strong and long-range coupling – transition temperature set by interaction energy, not density
- Like a laser – but matter and light remain strongly coupled
- Far from equilibrium physics – quantum dynamics?
- State preparation possible using optical control

Acknowledgements

Paul Eastham (Trinity College Dublin)
Jonathan Keeling (St Andrews)
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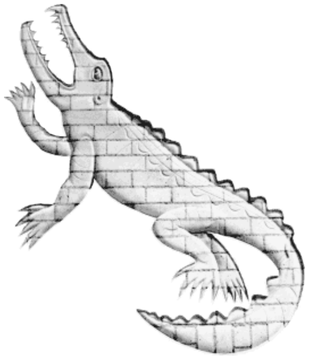
Richard Brierley (Cambridge/Yale)
Sahinur Reja (Dresden)
Alex Edelman (Chicago)
Cele Creatore (Cambridge)



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