Introduction to Polaritons as a Super-nonlinear Optical System

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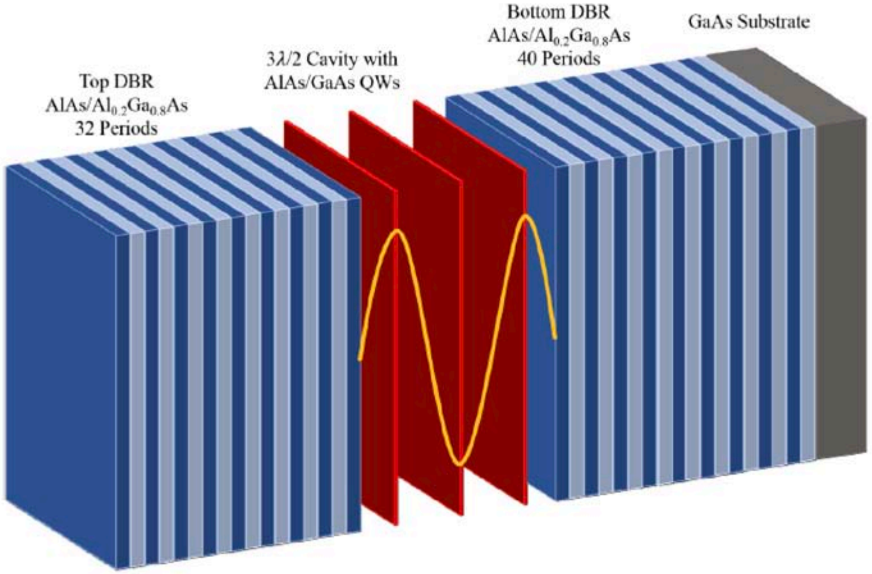
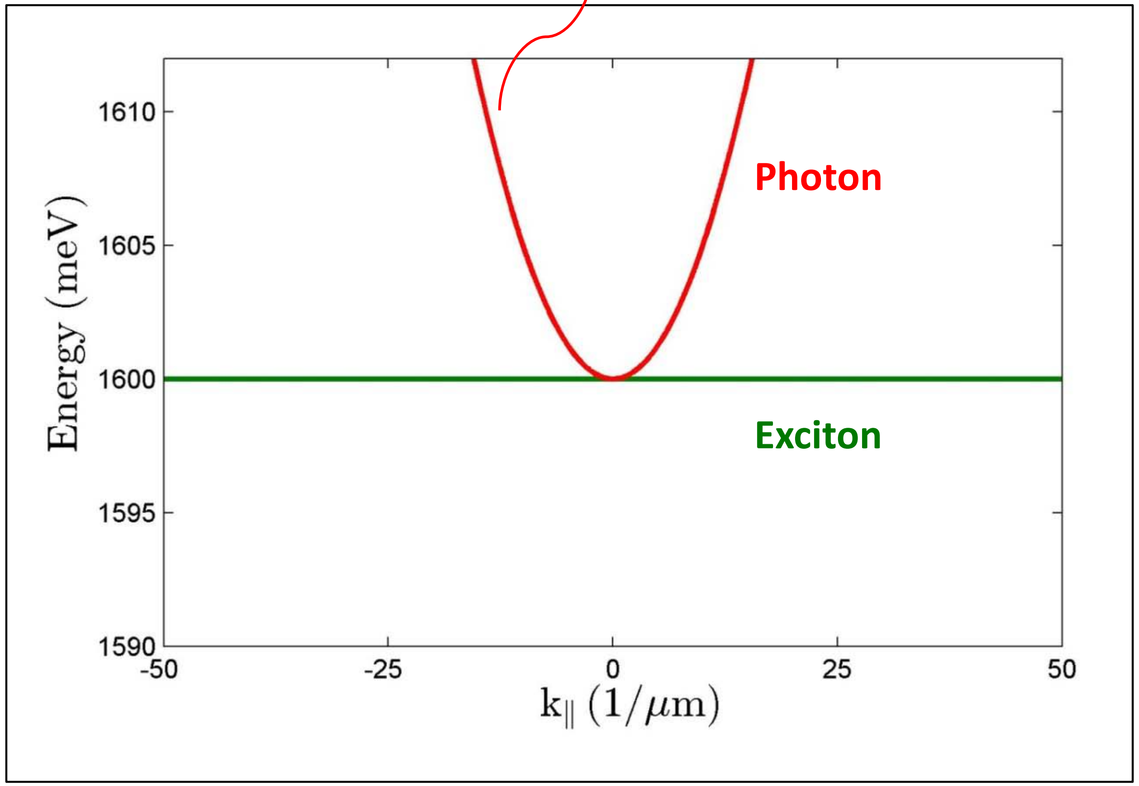
Polaritons in a microcavity

Effective mass

$$m^* = \frac{E_0}{c^2/n^2} \sim 3\times10^{-5} m_0$$



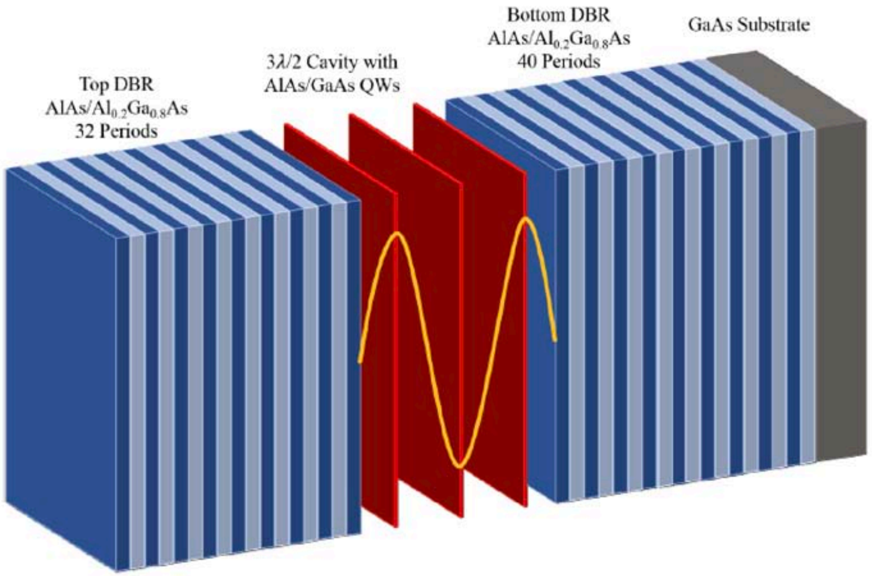


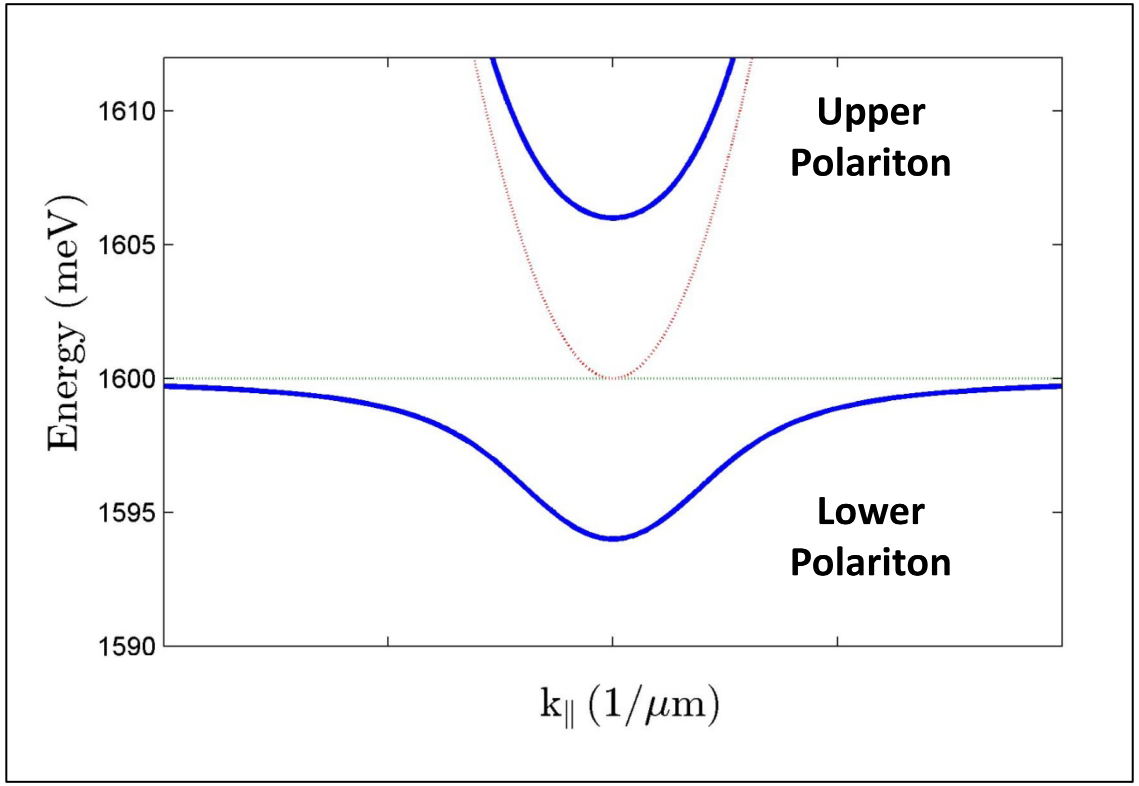
Photon energy in 2D planar cavity:

$$E = \hbar c \sqrt{k_z^2 + k_\parallel^2} = \hbar c \sqrt{(\pi/L)k_z^2 + k_\parallel^2}$$

In-plane
dispersion

Polaritons in a microcavity





Interaction of polaritons controlled by their exciton fraction.

$$\begin{pmatrix} E_{phot} & \Omega/2 \\ \Omega/2 & E_{exc} \end{pmatrix} \begin{pmatrix} \psi_{phot} \\ \psi_{exc} \end{pmatrix}$$

$$\Psi = \alpha\psi_{phot} + \beta\psi_{exc}$$

Gross-Pitaevksii equation/nonlinear wave equation

$$\nabla^2 E = \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} + 4\mu_0\chi^{(3)}\frac{\partial^2}{\partial t^2}|E|^2 E$$ Nonlinear Maxwell equation

$$\left.\begin{aligned} &E(\vec{x},t) = \psi(\vec{x},t)e^{-i\omega_0 t} \\ &\frac{\partial^2}{\partial t^2}|E|^2 E \simeq -\omega_0^2|\psi|^2\psi e^{-i\omega_0 t} \end{aligned}\right\}$$ Slowly-varying envelope approximation

$$i\hbar\frac{\partial\psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla_{\parallel}^2\psi - \frac{2\mu_0\chi^{(3)}(\hbar\omega)^2}{m}|\psi|^2\psi \quad -i\frac{\psi}{\tau} + iG(t,x)$$

Cf. $$i\hbar\frac{\partial\psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla_{\parallel}^2\psi + U|\psi|^2\psi \quad -i\frac{\psi}{\tau} + iG(t,x)$$

“Driven-dissipative G-P equation”

Gross-Pitaevskii equation is description of coherent condensate
with particle-particle interactions (classical wave).

Atoms alone are normally incoherent due to random interactions (dephasing).

Non-interacting photons alone are typically coherent.

Adding strong interactions makes polaritons behave like atoms: coherence
arises out of incoherence via Bose condensation.

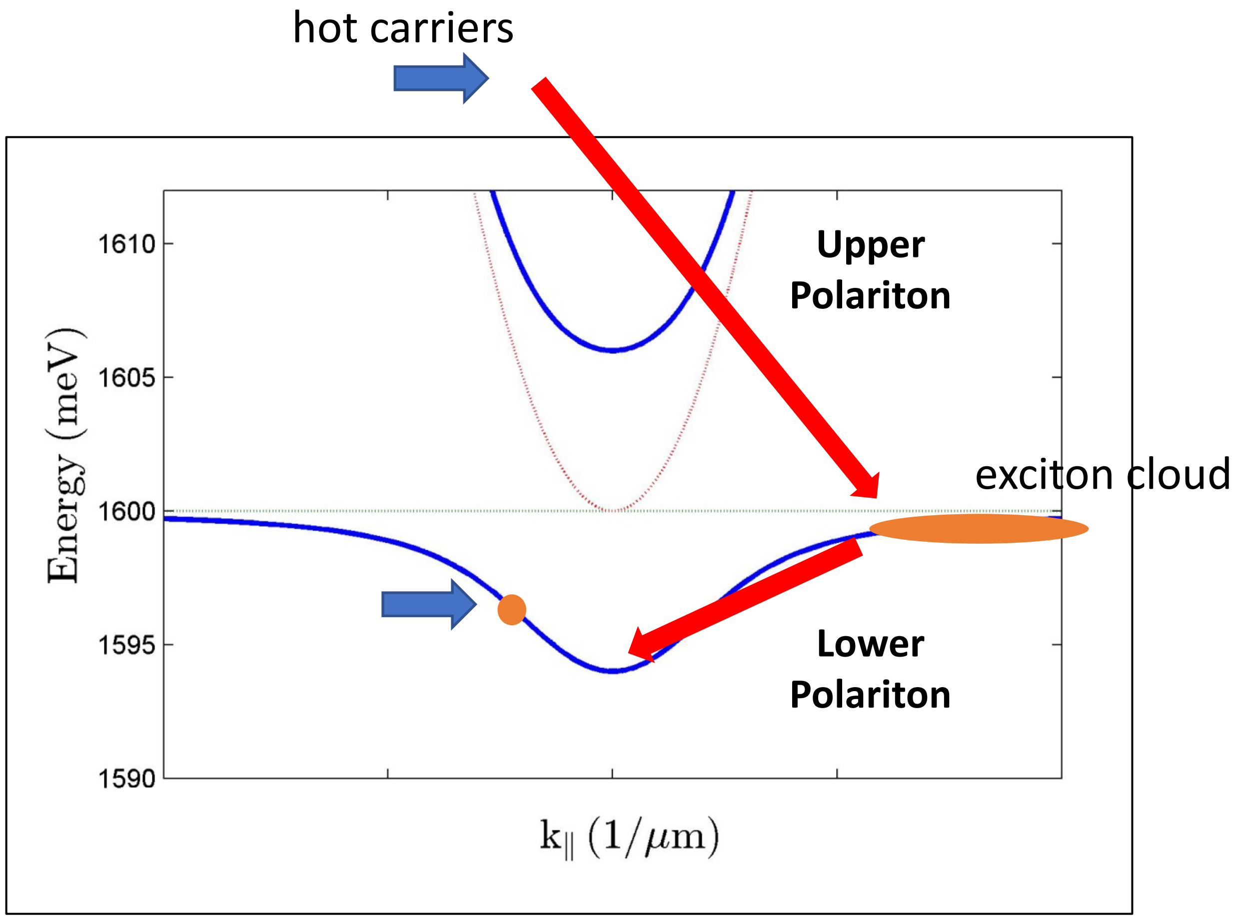
Two experiments

Resonant:

polaritons acquire coherence
of the pump laser, lose it quickly
unless above condensate threshold

Non-Resonant:

polaritons initially incoherent
can become coherent through
condensation

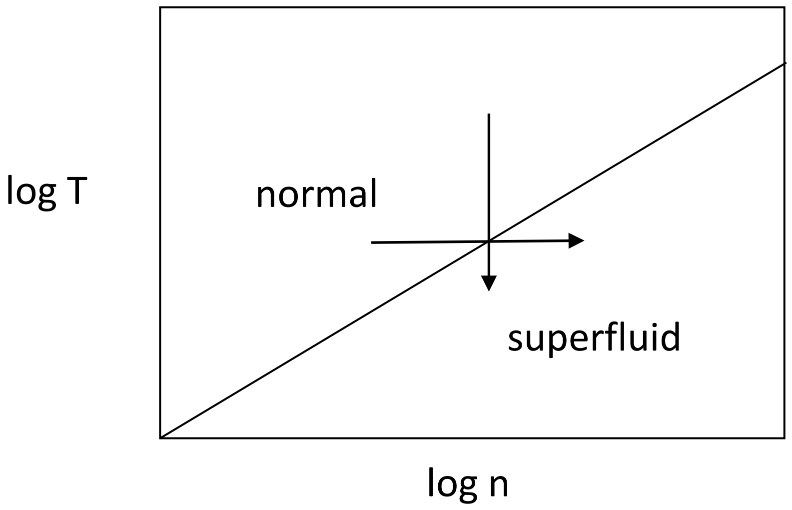


Bose-Einstein condensation of polaritons (ca. 2006-2007)

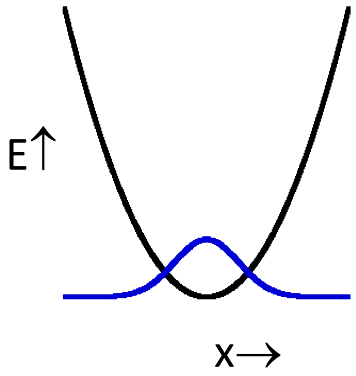
Critical threshold for quantum coherence $r \sim \lambda_{dB}$

$$n^{-1/d} \sim h / \sqrt{m k_B T}$$

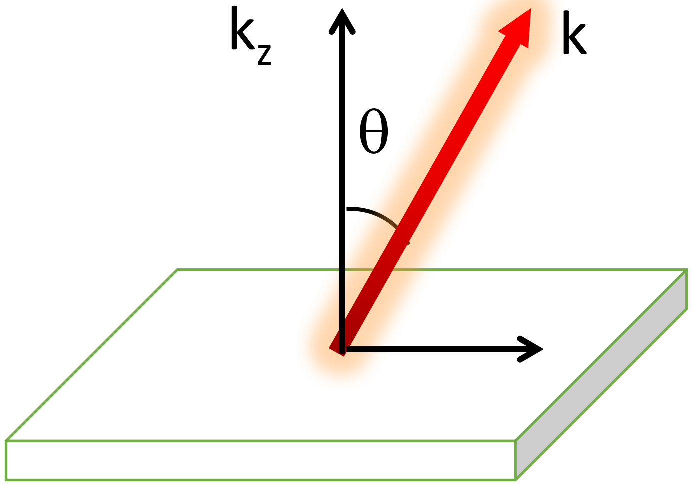
⇒ superfluid at *low T* or *high density*



trap implies *spatial* condensation

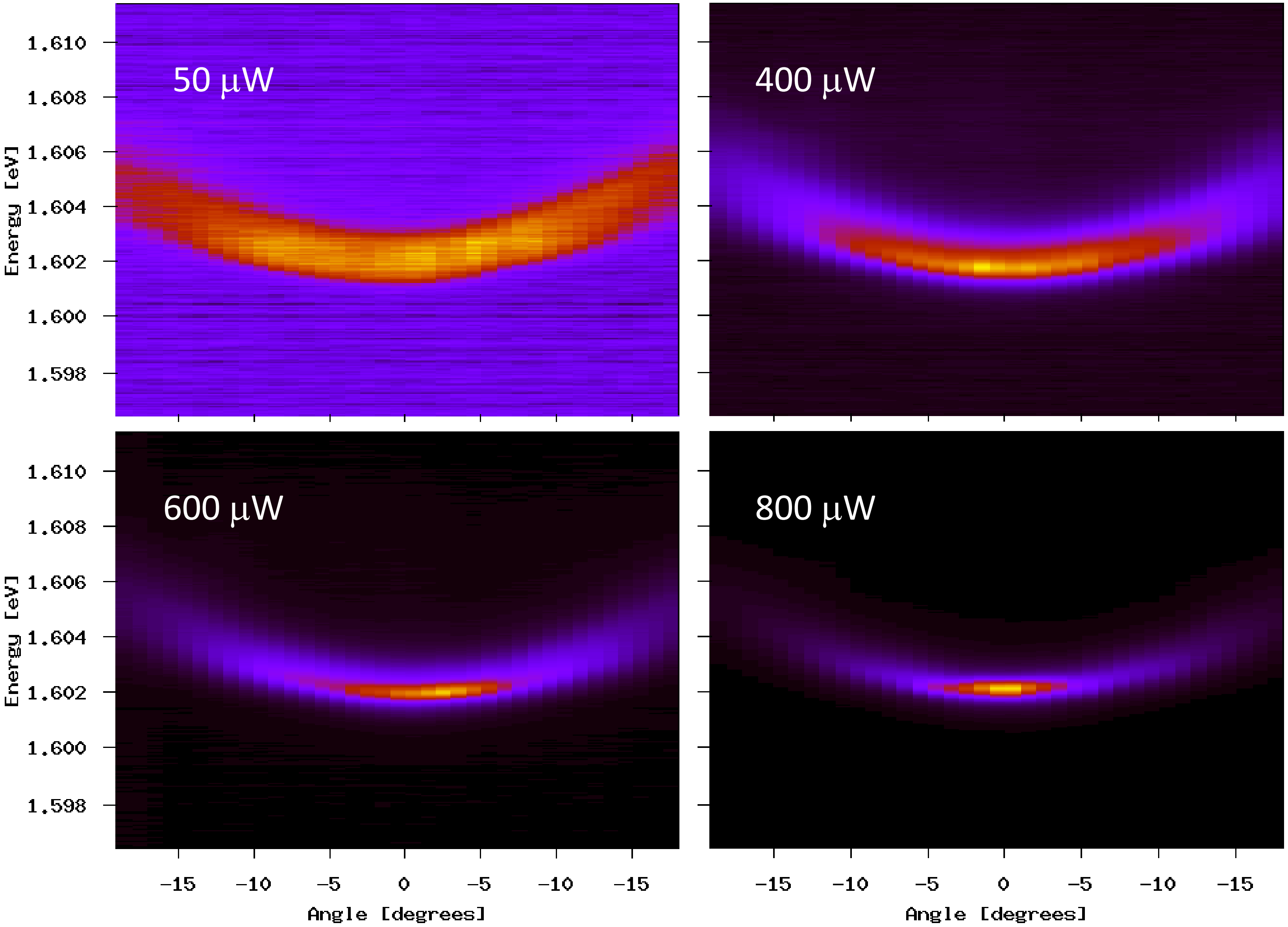


Angle-resolved photon emission data give momentum distribution.



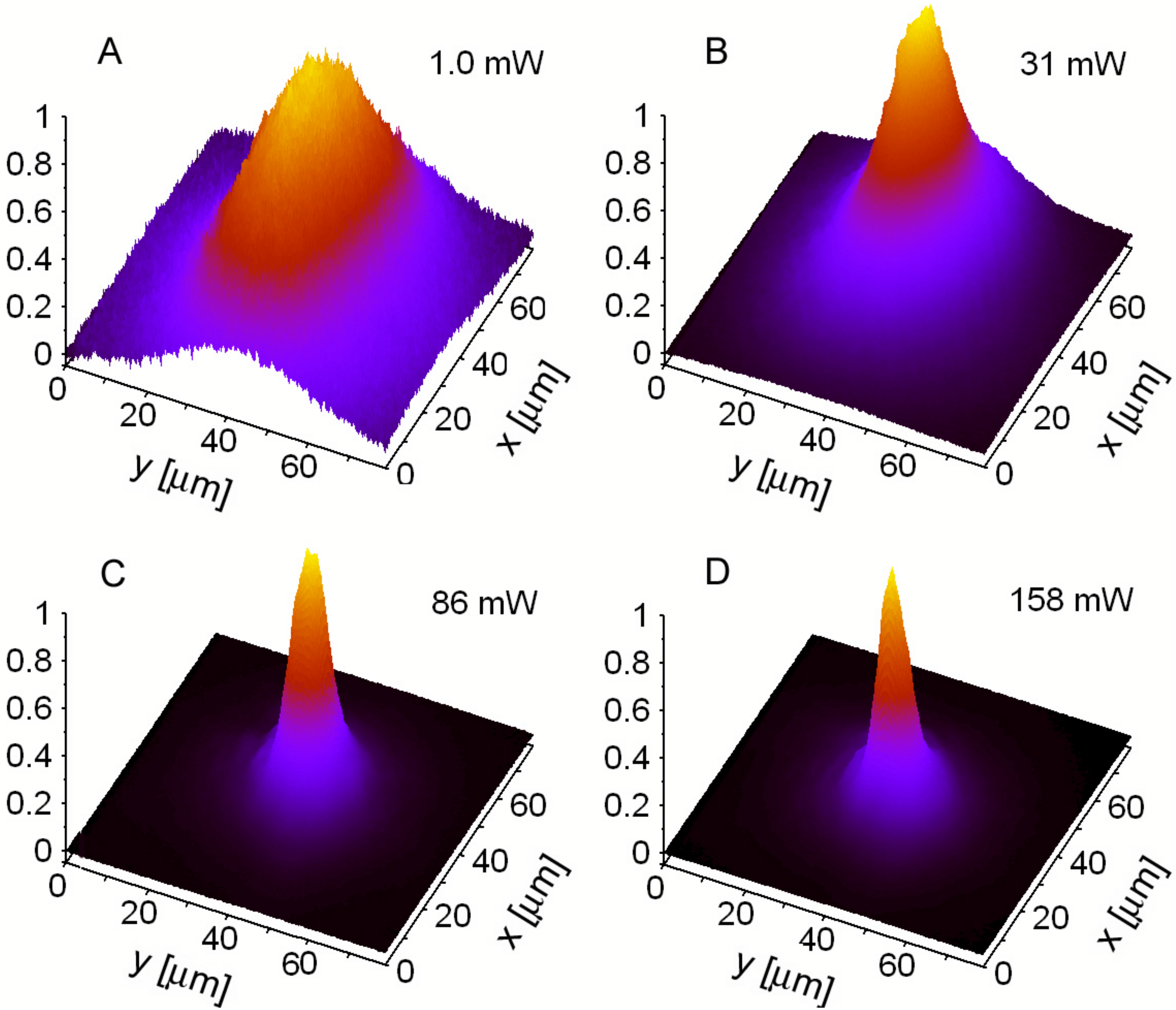
We can therefore directly image the gas in both real space and momentum space.

Momentum-resolved luminescence spectra: short lifetime
(cavity lifetime ~ 1 ps, average lifetime ~ 10 ps)

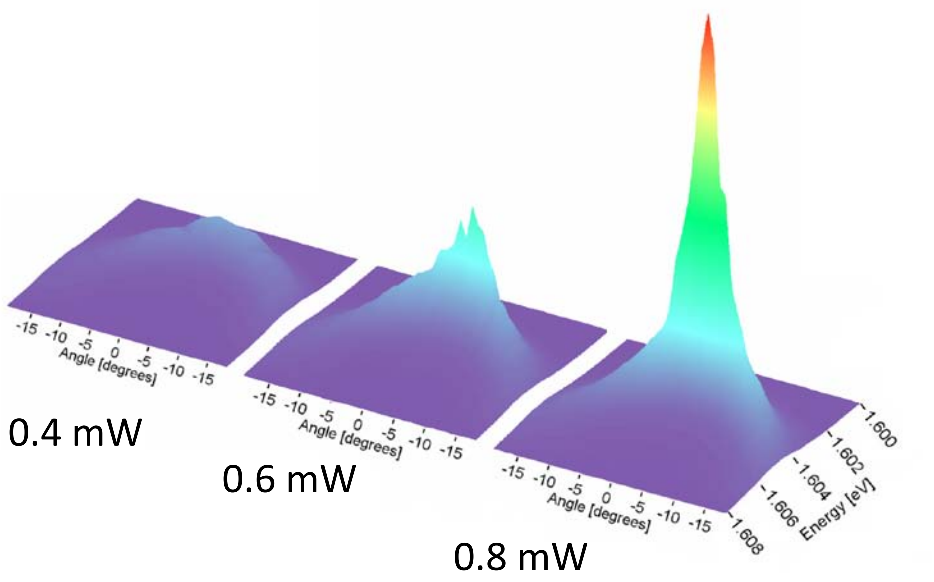


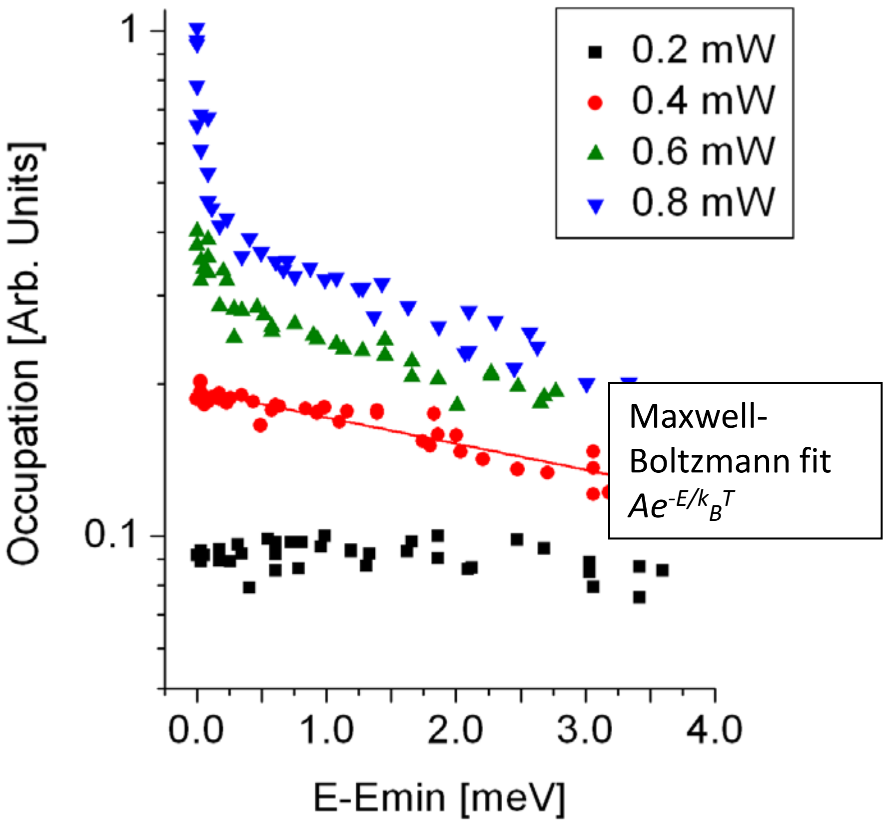
R. Balili et al., Science **316**, 1007 (2007)

Spatial profiles of polaritons in a harmonic potential trap



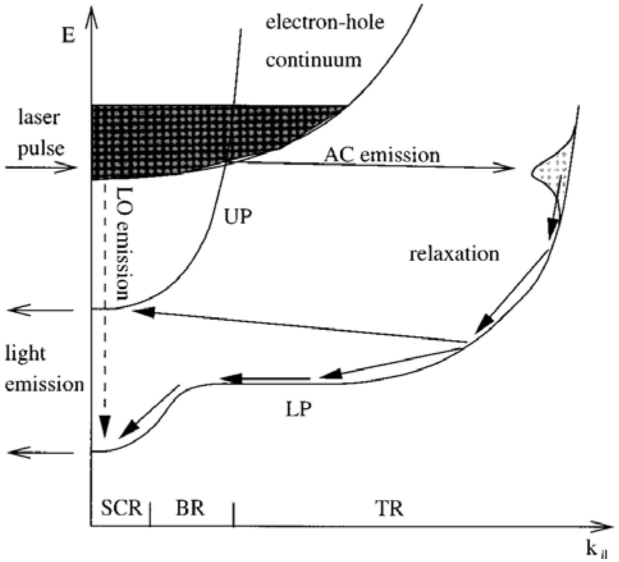
“Bimodal” momentum distribution of polaritons





2006-2007 data actually a *nonequilibrium* condensate—
Peaking due to Bose statistics, but excited states not in equilibrium.

Kinetic simulations of polariton equilibration



Tassone, *et al ,* Phys Rev B **56**, 7554 (1997).

Tassone and Yamamoto, Phys Rev B **59**, 10830 (1999).

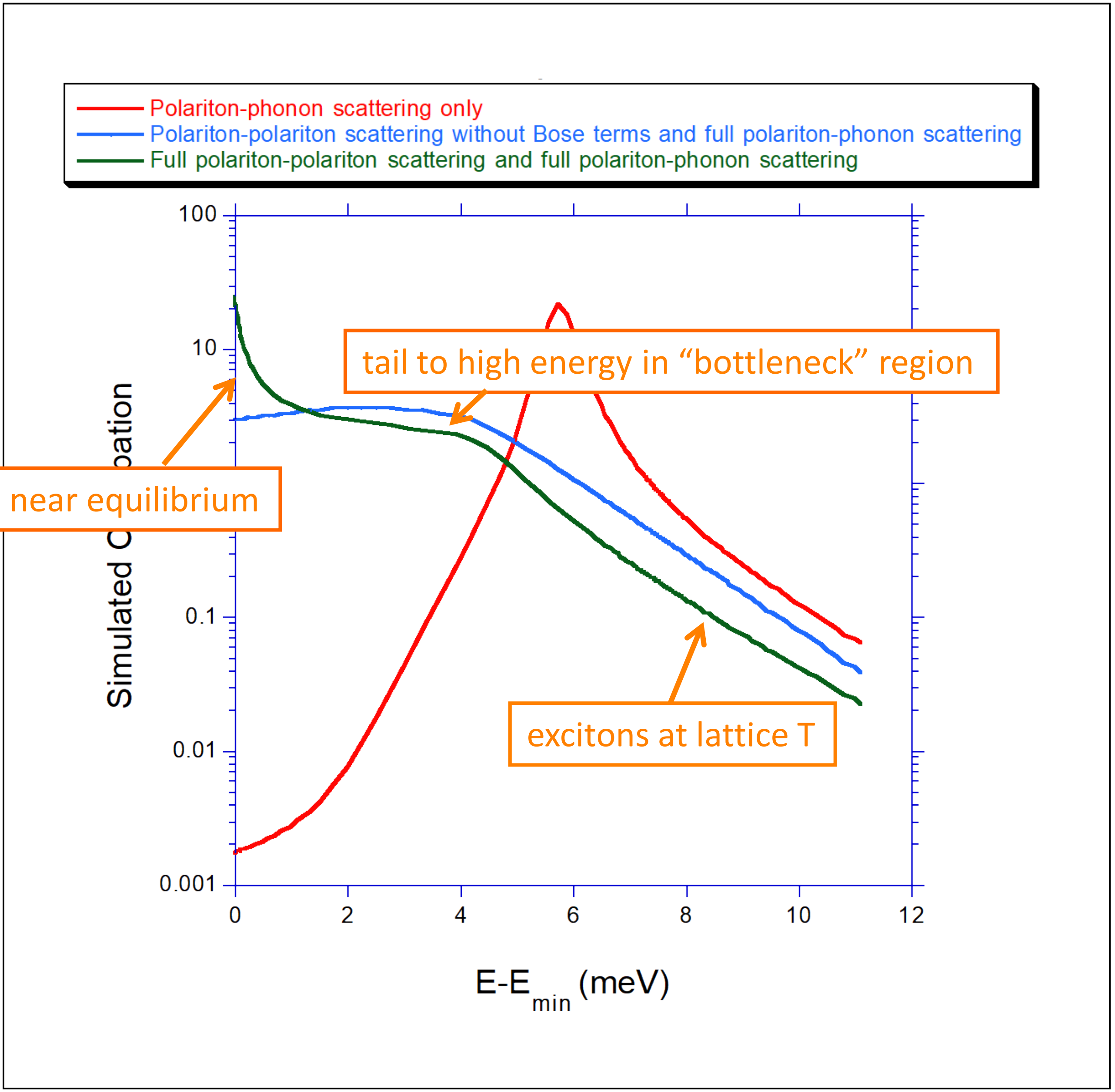
Porras et al., Phys. Rev. B **66**, 085304 (2002).

Haug et al.*,* Phys Rev B **72,** 085301 (2005).

Sarchi and Savona, Solid State Comm **144**, 371 (2007).

$$\frac{d\langle\hat{N}_k\rangle}{dt} = \frac{2\pi}{\hbar}\left(\frac{V}{(2\pi)^3}\right)^2 \frac{1}{2}\int d^3k_1\, d^3k_2\, |U_D \pm U_E|^2 \delta(E_{k_1} + E_{k_2} - E_k - E_{k'})$$
$$\times \left[\langle\hat{N}_{k_1}\rangle\langle\hat{N}_{k_2}\rangle(1 \pm \langle\hat{N}_k\rangle)(1 \pm \langle\hat{N}_{k'}\rangle) - \langle\hat{N}_k\rangle\langle\hat{N}_{k'}\rangle(1 \pm \langle\hat{N}_{k_1}\rangle)(1 \pm \langle\hat{N}_{k_2}\rangle)\right]$$

Numerical steady-state solution for occupation number



tail to high energy in "bottleneck" region

polaritons near equilibrium

excitons at lattice T

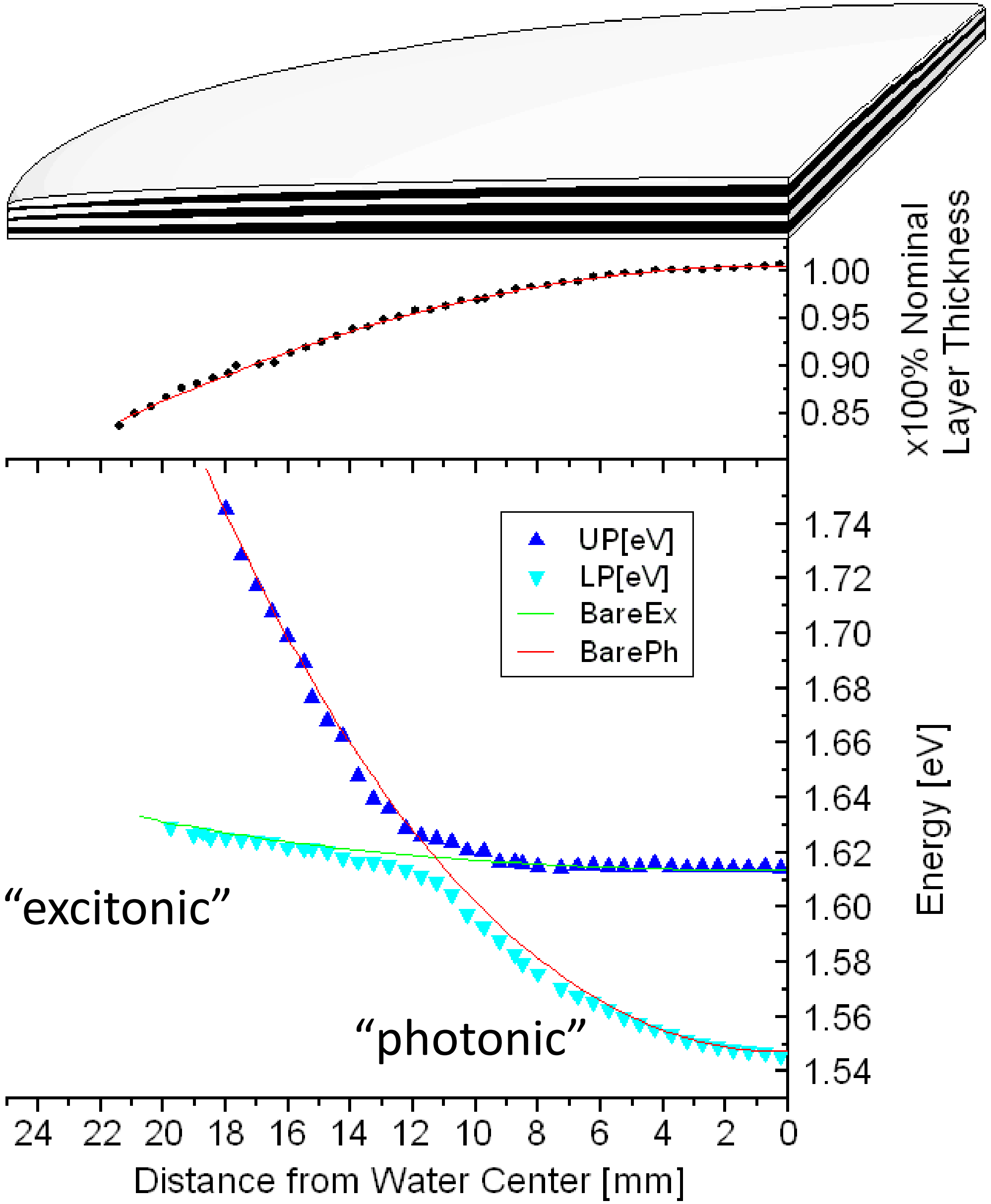
Phonon emission is so weak that particles are essentially isolated from the lattice

V. Hartwell, Ph.D. thesis (2008); PRB **82**, 075307 (2010)

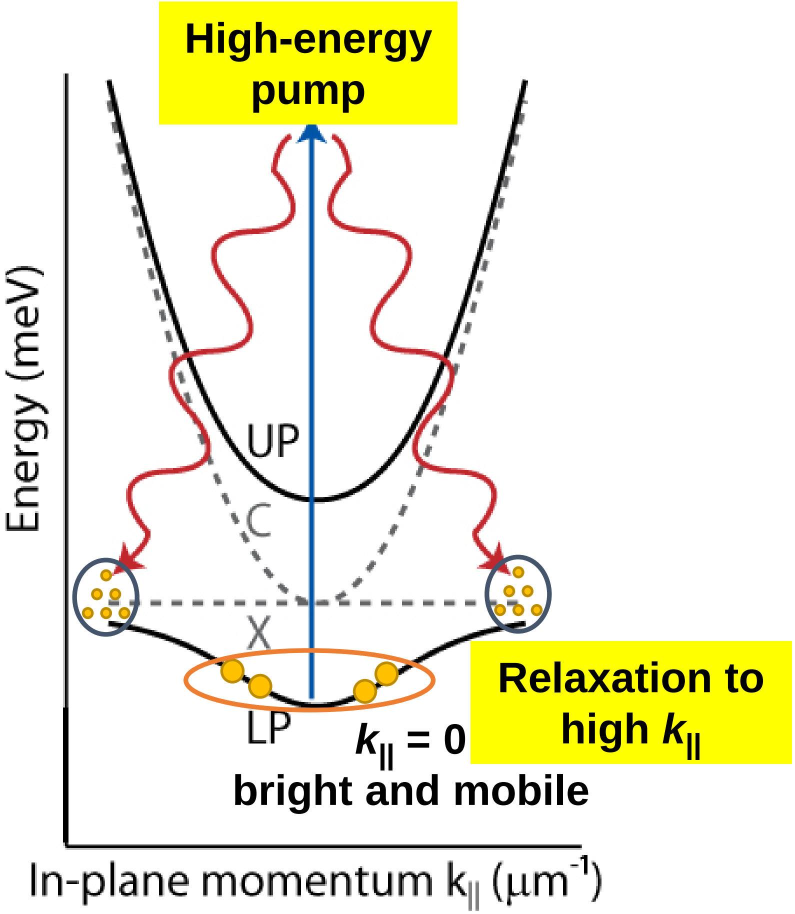
Two important details

1. Photon energy shift by cavity wedge:

Crossing of photon and
exciton energies gives
“photonic” and
“excitonic” sides
for lower polariton.



2. “Exciton cloud” or “reservoir”



High-energy pump

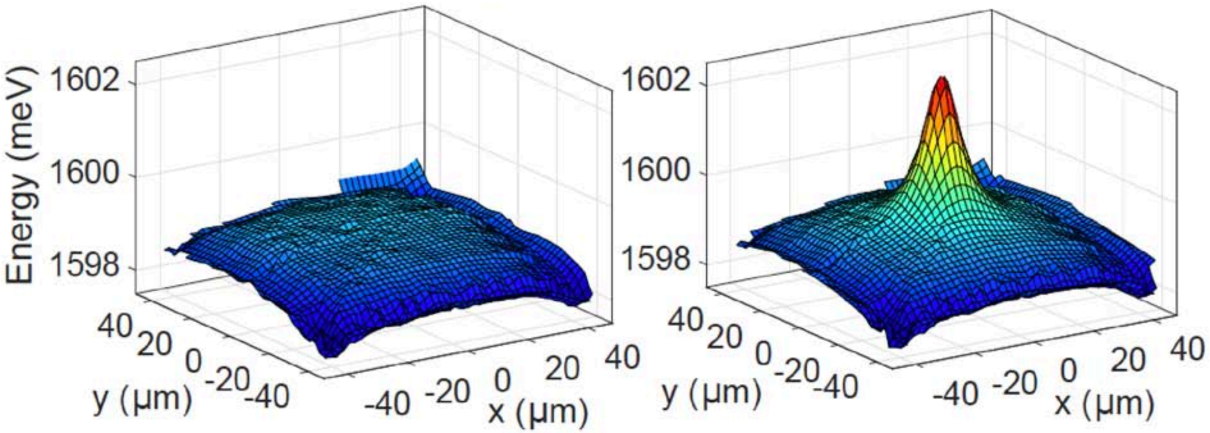
Relaxation to high $k_{||}$

$k_{||} = 0$ bright and mobile

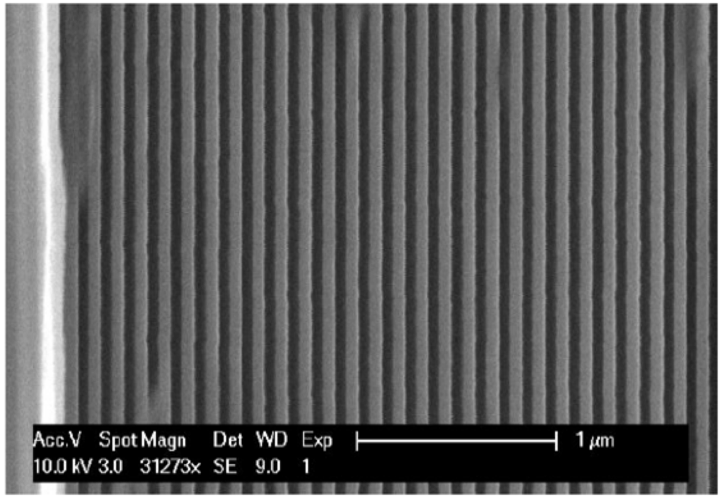
- Excitons are 10^4 more massive than the polaritons. They move very little, so that collisions of polaritons with excitons are nearly elastic– a static barrier as seen by polaritons

- Position and height controlled directly by laser

- Disadvantage: Excitons turn into polaritons. Exciton cloud acts both as potential energy and source term.



Super samples:



Q > 300,000
cf. previous samples with Q~5000

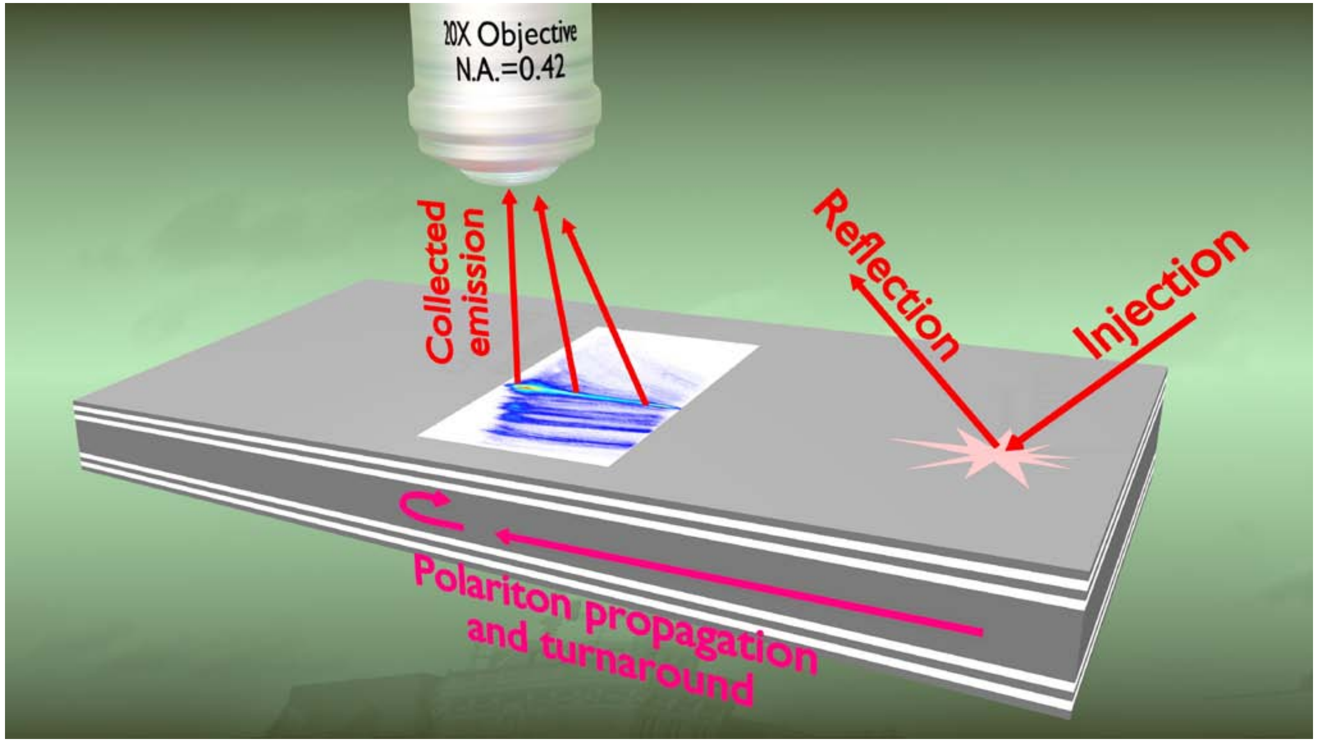
Done by using DBR mirrors with 40 layers

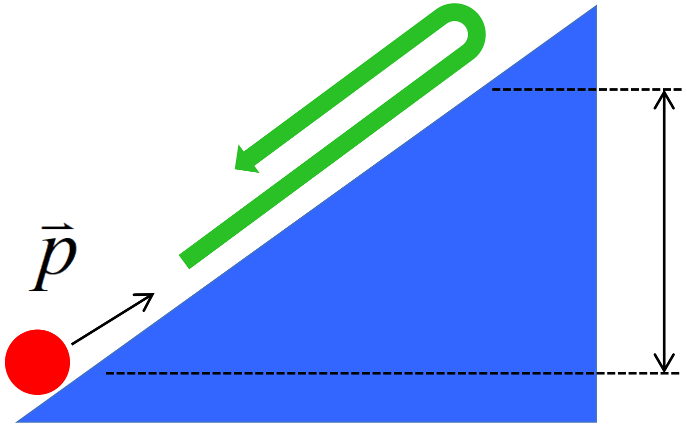
MBE growth by Pfeiffer group > 30 hours per sample.
growth rate must remain stable during this time (±1%)
disorder must remain low

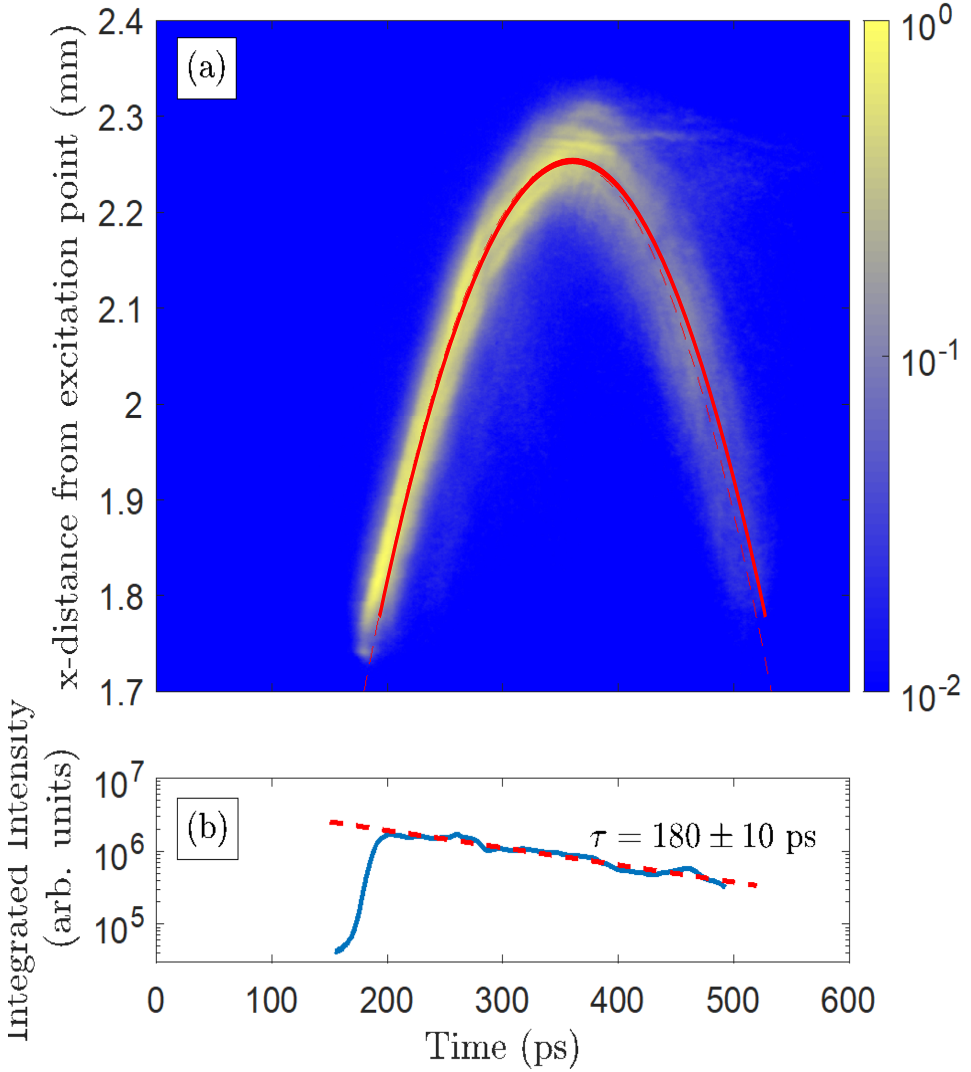
Cavity lifetime scales with *Q*: from ~ 1 ps to over 100 ps

Polariton lifetime is ~ 250 ps: decay rate is proportional to photon fraction)

Direct resonant injection: angle of injection gives in-plane p







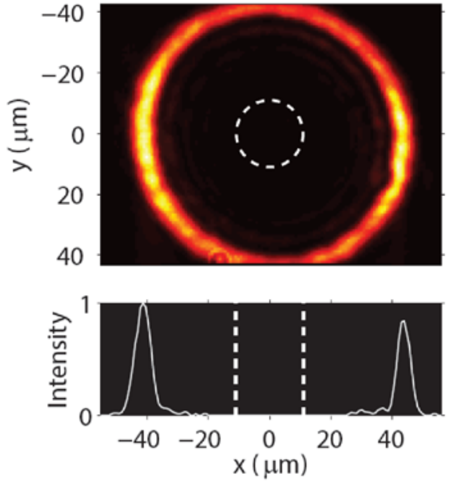
M. Steger et al., Optica **2**, 1 (2015)

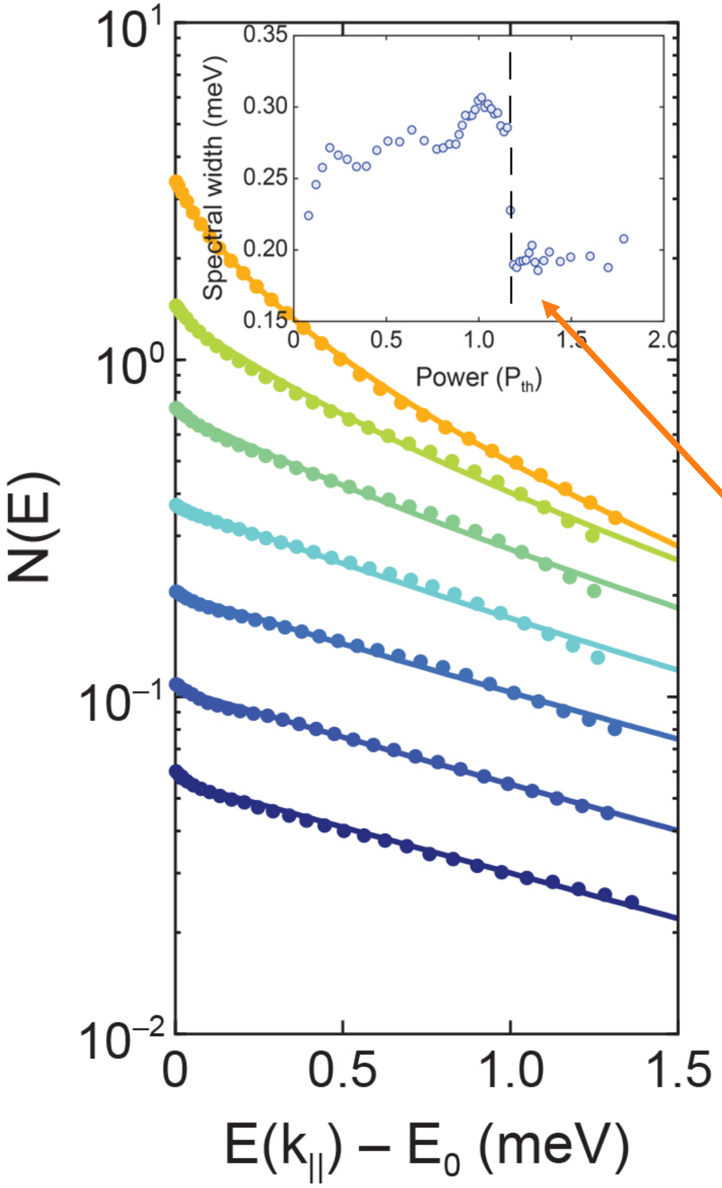
The quality of our samples
is shown by these measurements:
> 200 ps lifetime, > 2 mm transport

Time-resolved
polariton motion
in a potential gradient

“slow reflection”

Equilibrium distributions of polaritons in laser trap





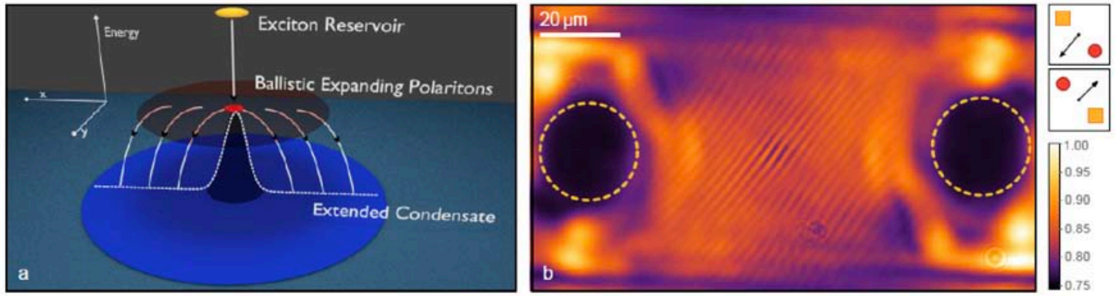
$$N(E) = \frac{1}{e^{(E-\mu)/kT} - 1}$$

Spectral width drops sharply at BEC transition (spontaneous coherence)

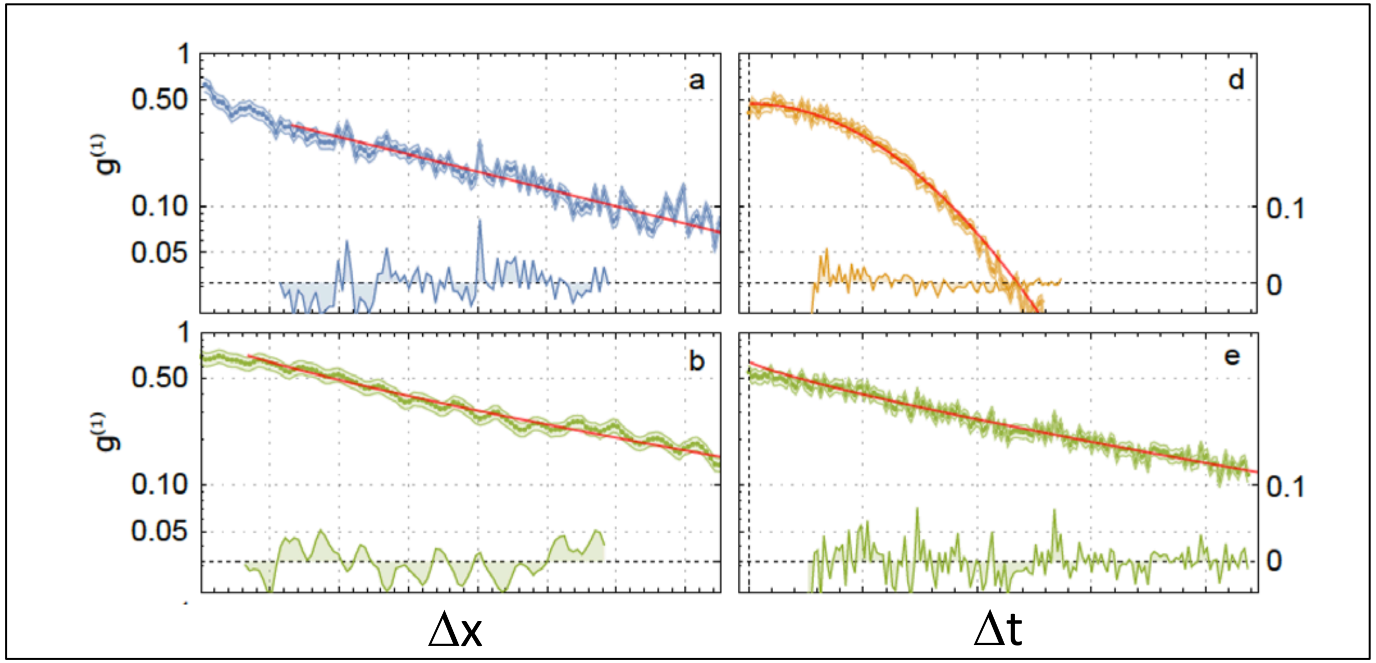
Maxwell-Boltzmann at low density Ae^{-E/k_BT}

Y. Sun, et al., PRL **118**, 016602 (2017).

Equilibrium also seen in temporal and spatial correlation functions



D. Caputo et al., Nature Materials **17**, 145 (2018).



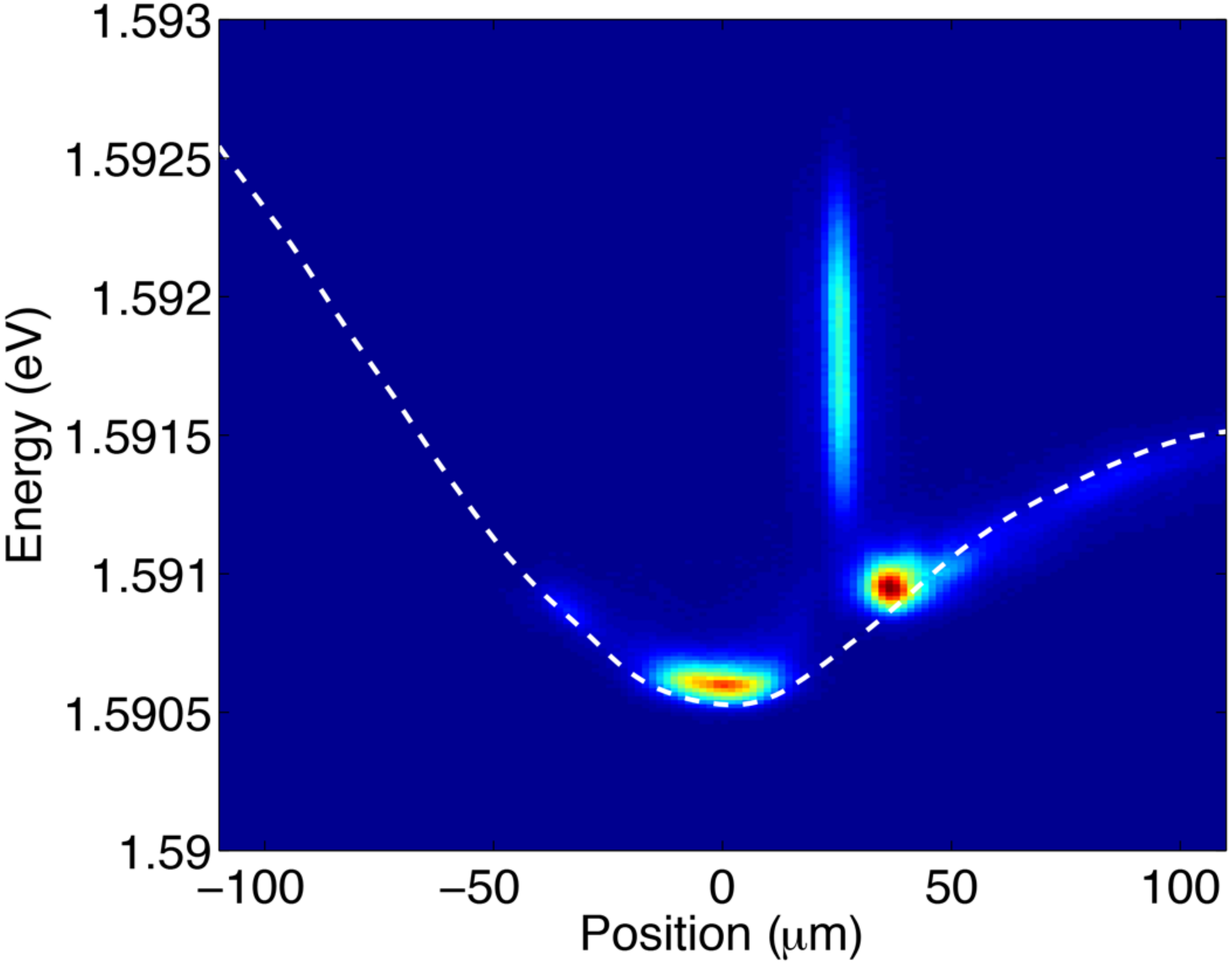
low density:
no condensate

high density:
BKT power law

Phase coherence in a ring

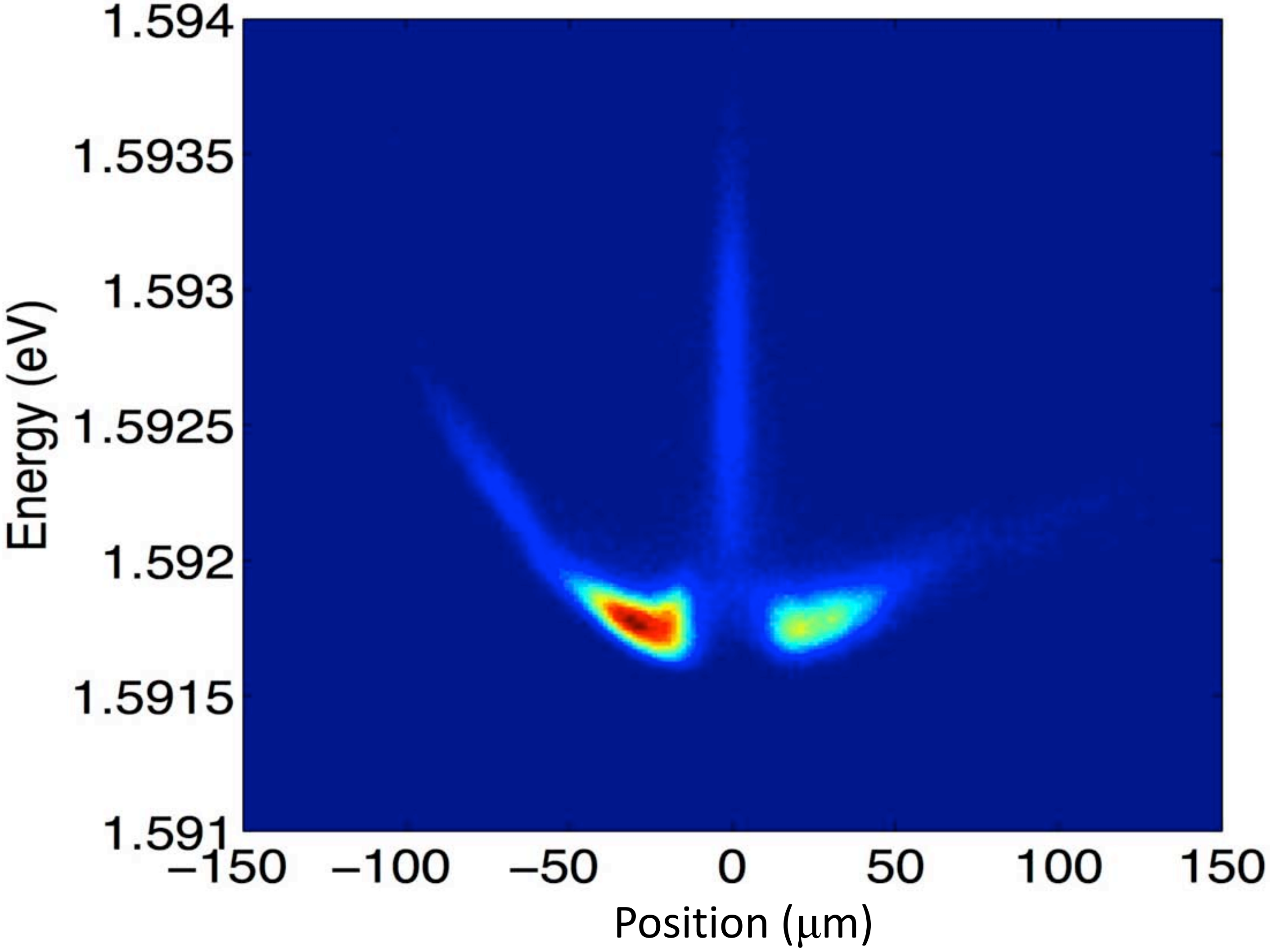
focused laser, to side

defocused laser

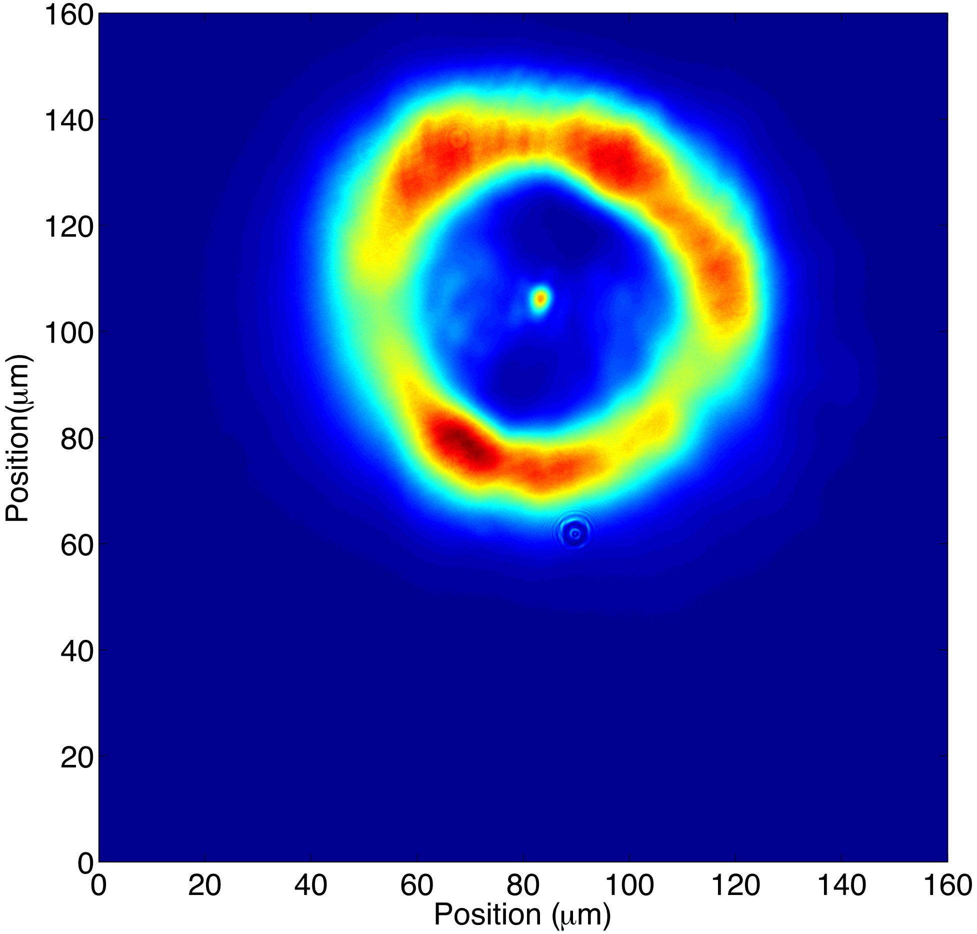


Phase coherence in a ring

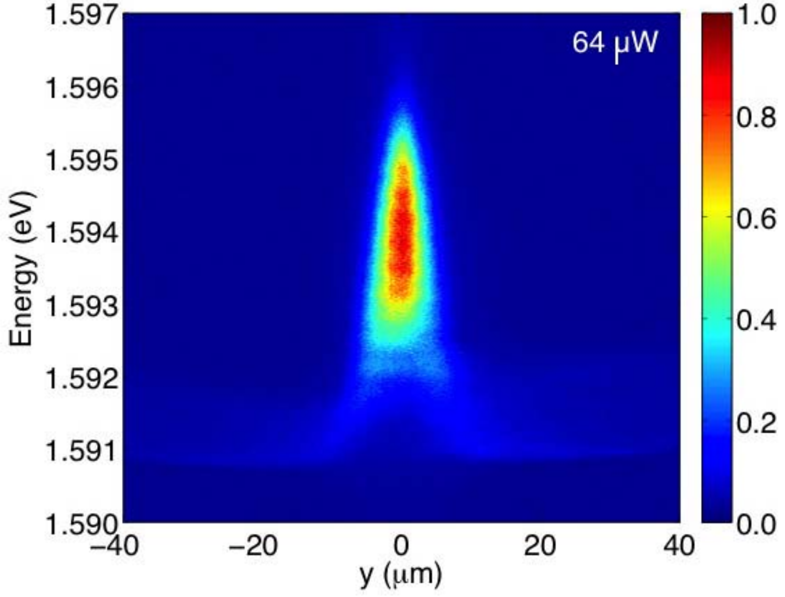
focused laser, in center



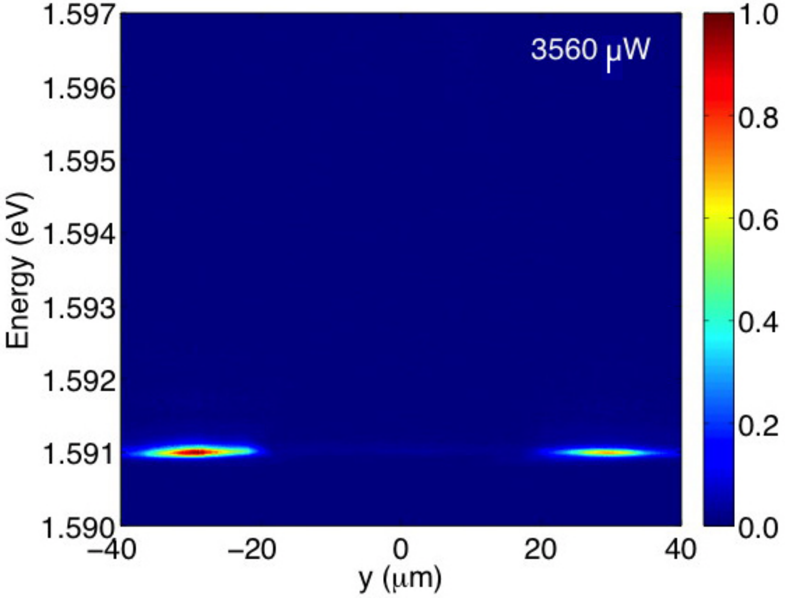
2D spatial image



Spectral narrowing in the ring:

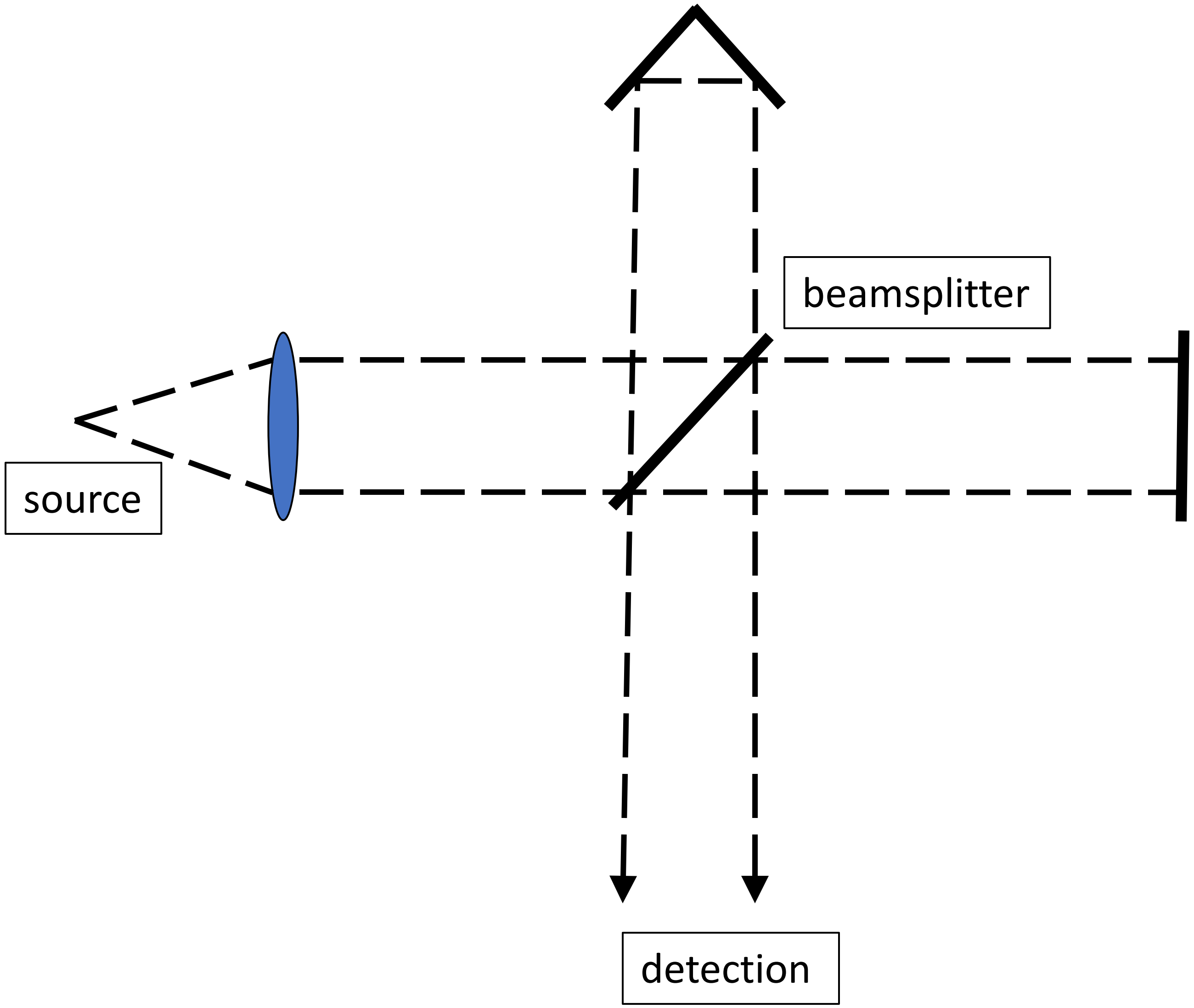


below threshold



above threshold

Michelson interferometer with flip of x-axis

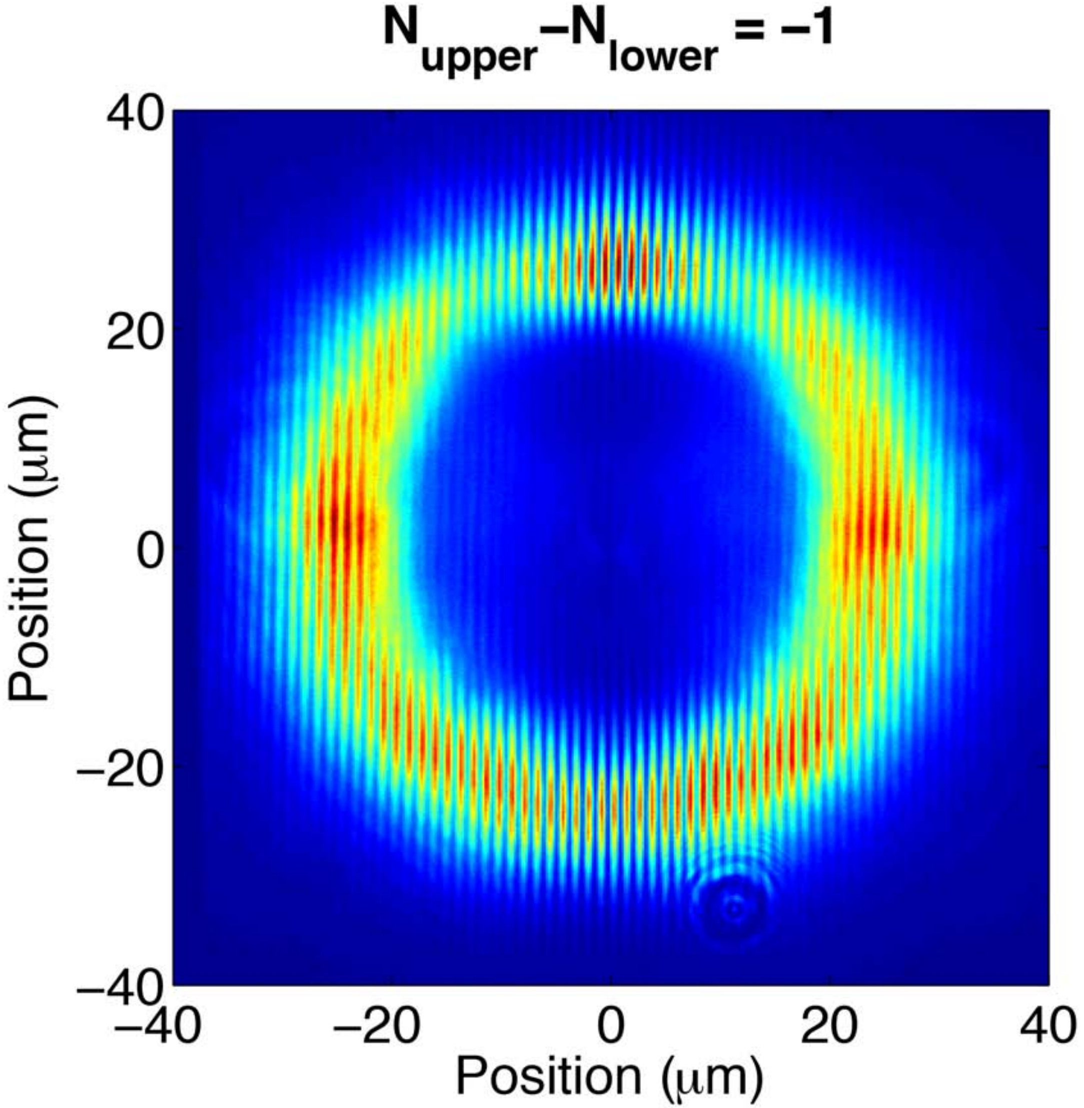


beamsplitter

source

detection

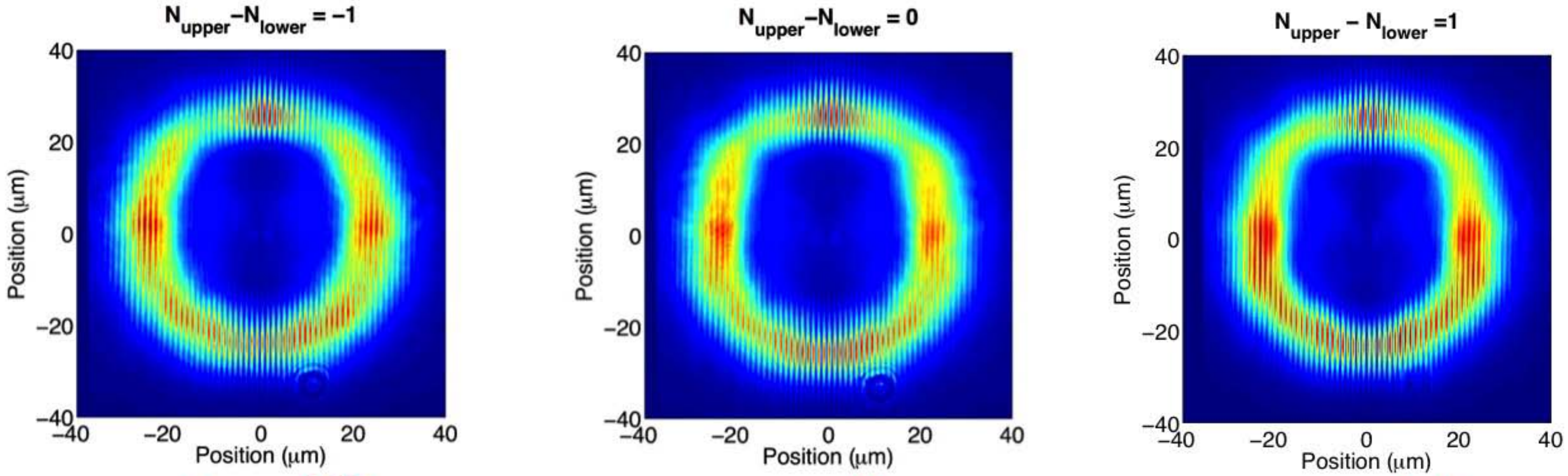
Interference patterns- interference shows quantized circulation.



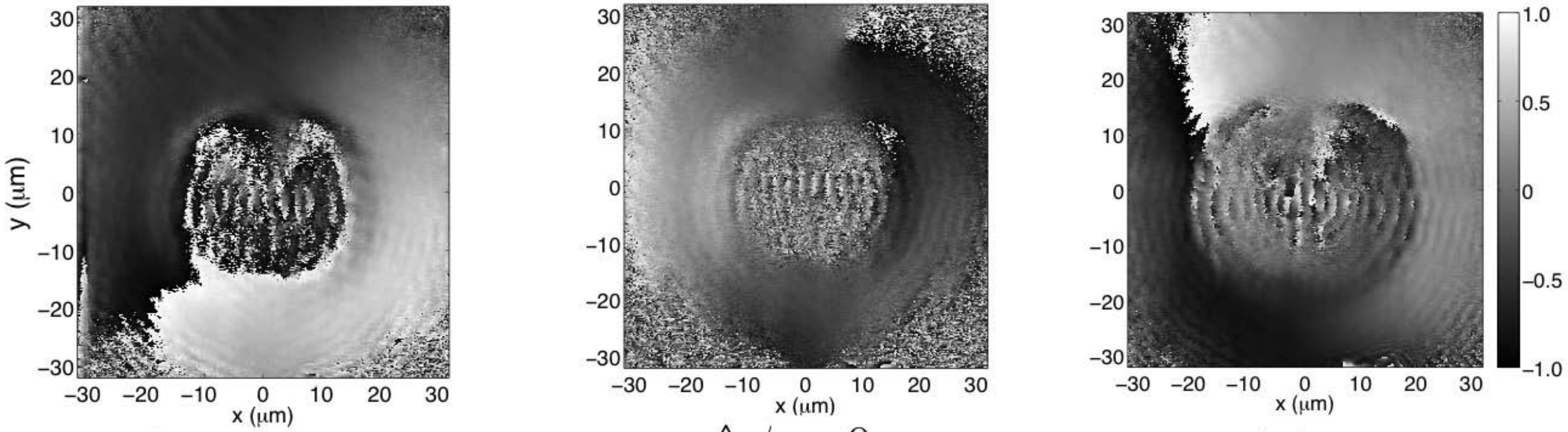
G.-Q. Liu, D.W. Snoke, A.J. Daley, L.N. Pfeiffer, and K. West, PNAS **112** , 2676 (2015).

Phase winding

45% 10% 45%



m = -1/2 m = 0 m = 1/2



$\triangle\phi = -\pi$ $\triangle\phi = 0$ $\triangle\phi = \pi$

Phase maps extracted from fringe patterns

Polariton-polariton interactions

- Important parameter for applications that use nonlinear shift of energy states
  - Single-photon blockade
  - Optical switching/optical transistor

- Controls both renormalization of energy states (real part of self energy) and scattering rate/thermalization rate (imaginary part of self energy)

- Very hard to calculate exactly: electron-electron exchange, hole-hole exchange, electron-hole exchange

- In general, expected from unit analysis to be $U \sim (Ry_{ex})a_B^2$ (Tassone/Yamamoto standard estimate ~ 6 μeV-μm^2 in GaAs)

Relation of real self-energy and imaginary self-energy

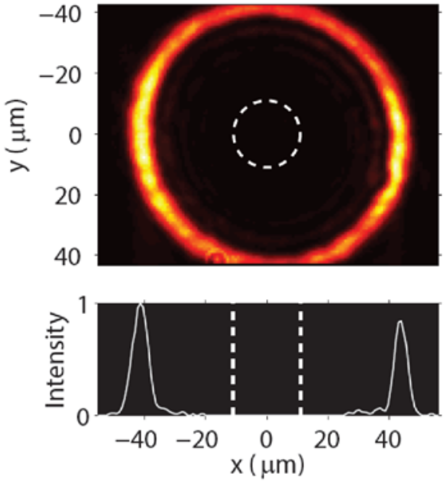
$$\langle i|\psi_t\rangle = \exp\left[-(i/\hbar)\left(E_i + \langle i|V_{\text{int}}|i\rangle + \sum_{m\neq i}\frac{|\langle m|V_{\text{int}}|i\rangle|^2}{E_i - E_m + i\eta} + \ldots\right)t\right]$$

$$\begin{aligned}\sum_{m\neq i}\frac{|\langle m|V_{\text{int}}|i\rangle|^2}{E_i - E_m + i\eta} &= \mathcal{P}\left(\sum_{m\neq i}\frac{|\langle m|V_{\text{int}}|i\rangle|^2}{E_i - E_m}\right) - i\pi \underbrace{\sum_{m\neq i}|\langle m|V_{\text{int}}|i\rangle|^2\delta(E_i - E_m)}_{\text{scattering rate}} \\ &\equiv \Delta^{(2)} - i\Gamma^{(2)}.\end{aligned}$$

MIT-Pitt experiment to measure interactions from blue shift

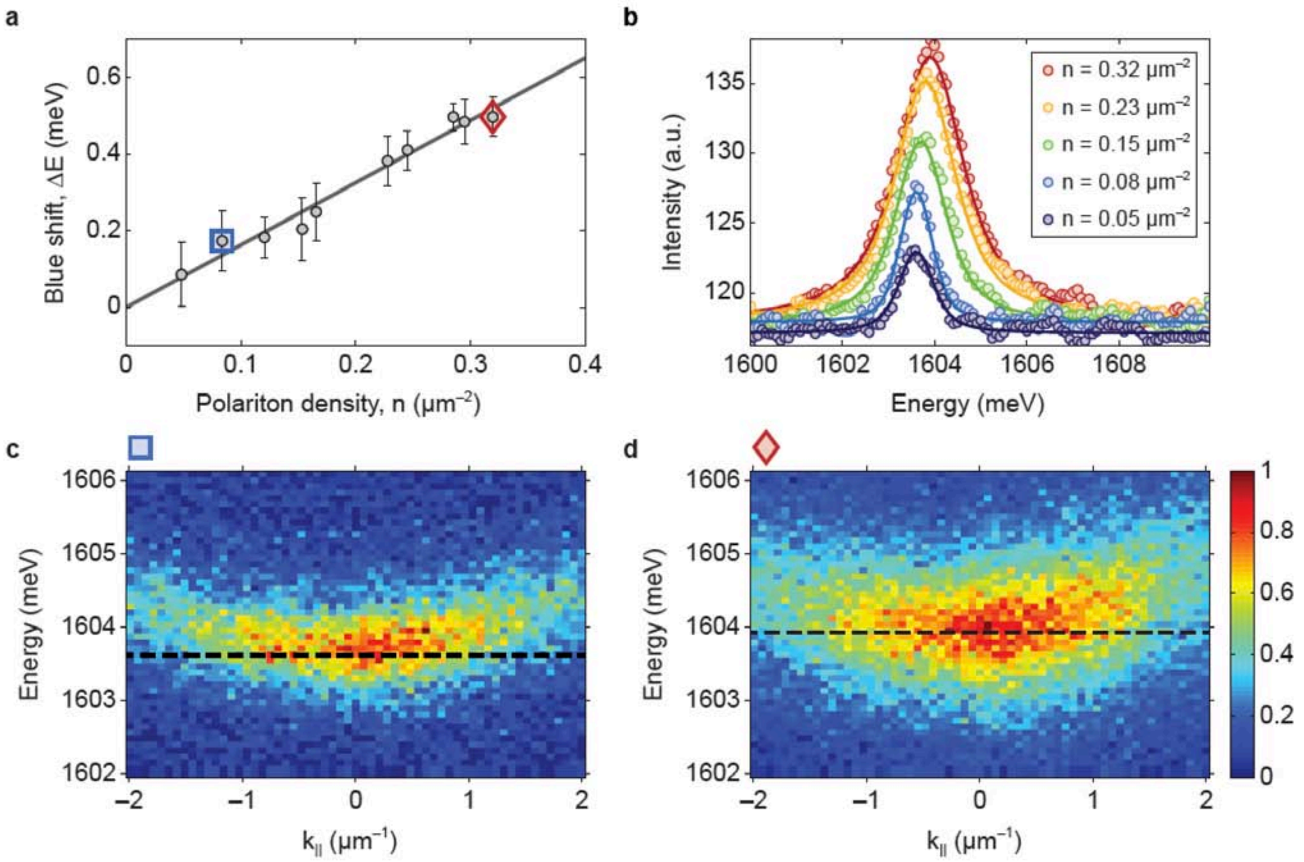
Y. Sun et al., Nature Phys. **13**, 870 (2017).

Large ring trap



Blue shift ∝ U

Linewidth (collision rate) ∝ U^2



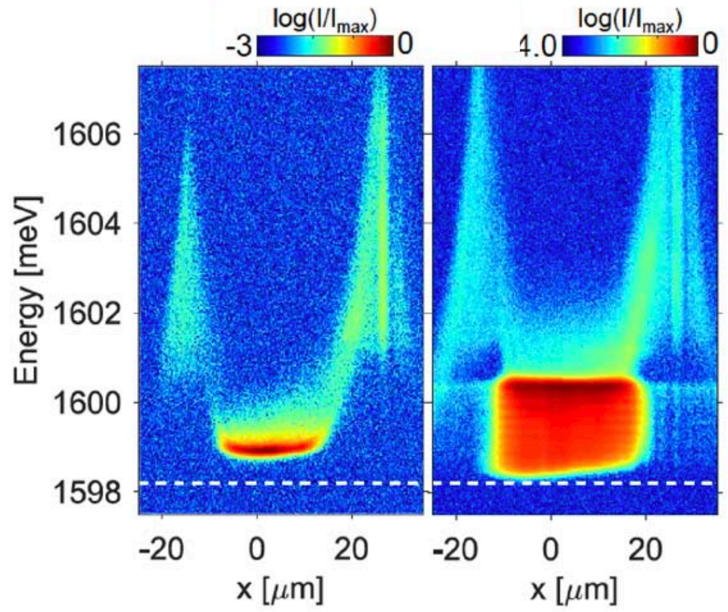
MIT results overestimated interaction strength due to ignoring larger-than-expected exciton “reservoir” diffusion.

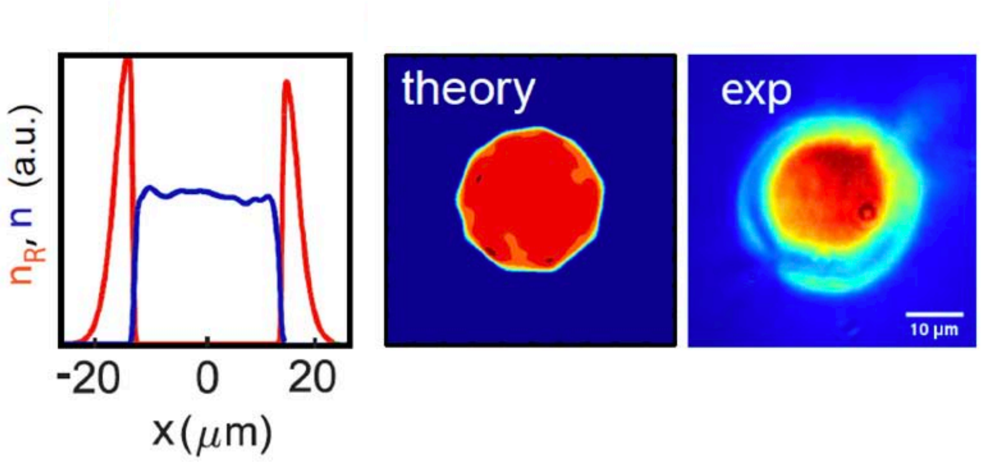
With recalibration, number is ~ 40 μeV-μm^2

Still a wide range of numbers for the interaction strength in the literature: 0.1-40 μeV-μm^2

Two recent, independent experiments in the condensate regime

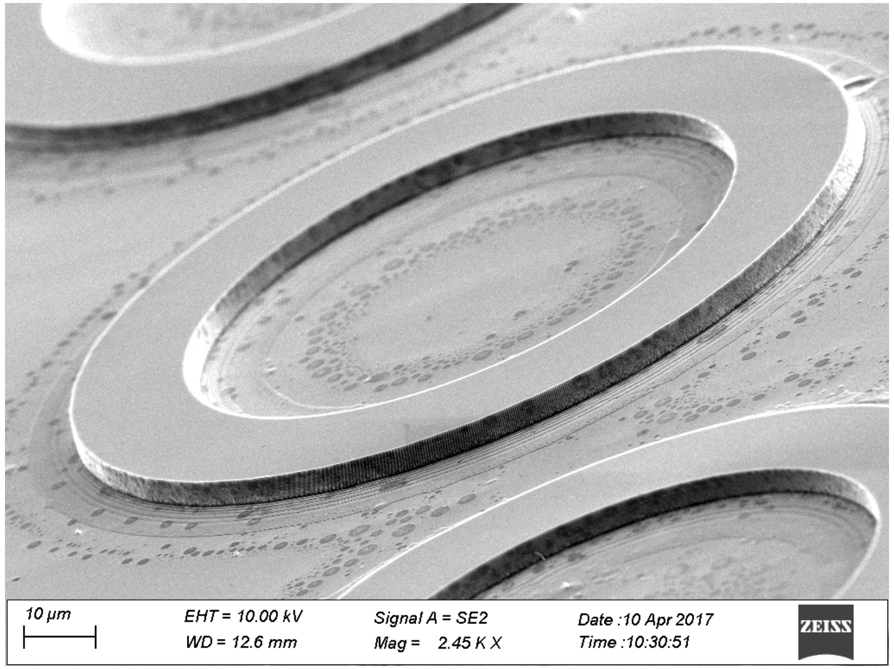
1. Estrecho et al. (ANU/Pitt collab):
Complete depletion of dark excitons in high density regime

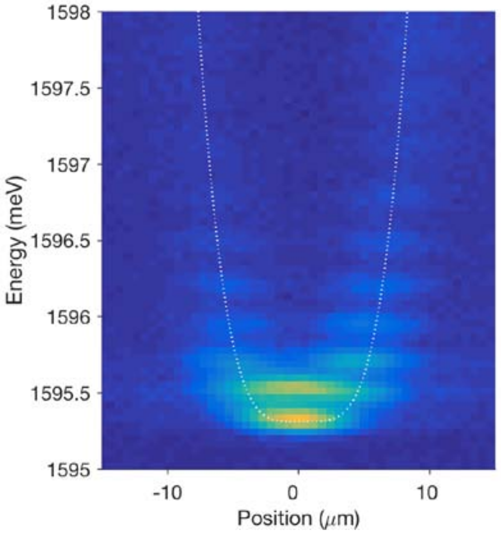




Yields $U \approx 0.2\ \mu\text{eV-}\mu\text{m}^2$

2. Mukherjee et al. (Pitt):
Oscillation of condensate in a ring



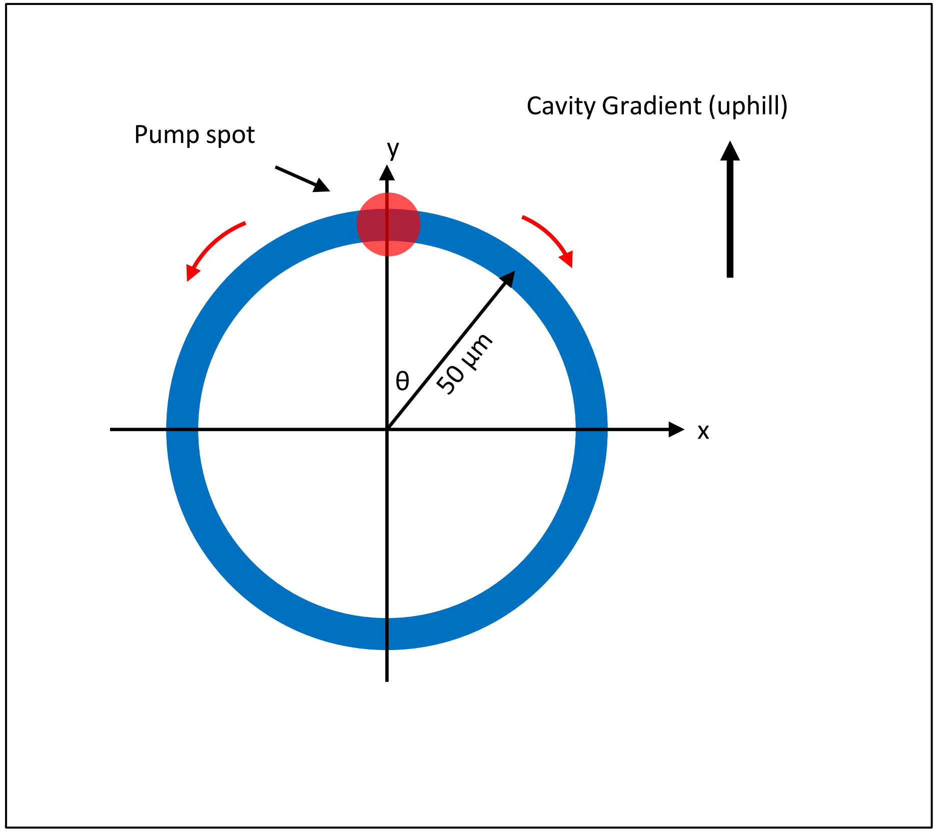


Typical width (outer – inner radius) ≈ 15 µm

Typical Center radius = 50 µm

Time-resolved measurements with streak camera and pulsed excitation

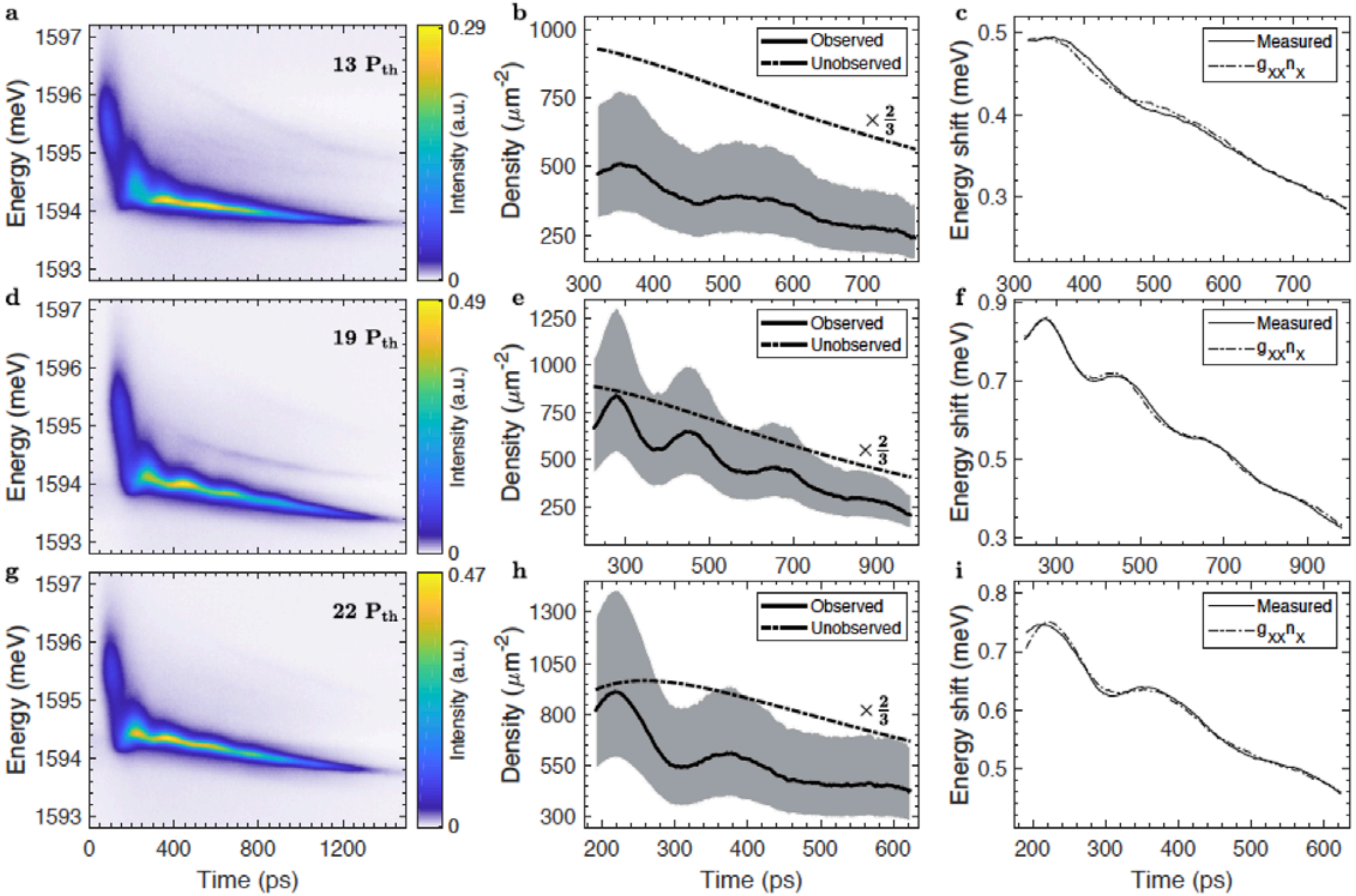
Hamiltonian of the system is the same as a rigid pendulum.



Time-resolved measurements with streak camera and pulsed excitation

Mukherjee et al., arXiv:1901.05608

Only the polariton condensate oscillates at the natural frequency.
Tight constraints since both energy and density oscillate.



Fits yield U ≈ 1 μeV-μm^2

What we know about interaction measurements

- Experimental values of the interaction parameter for polaritons (at resonance, with 10 quantum wells) cluster around two values:

  20-70 μeV-μm^2 at low density

  0.2-1.0 μeV-μm^2 at high density (condensate)

- The possibility of a density-dependent interaction strength cannot be ruled out.

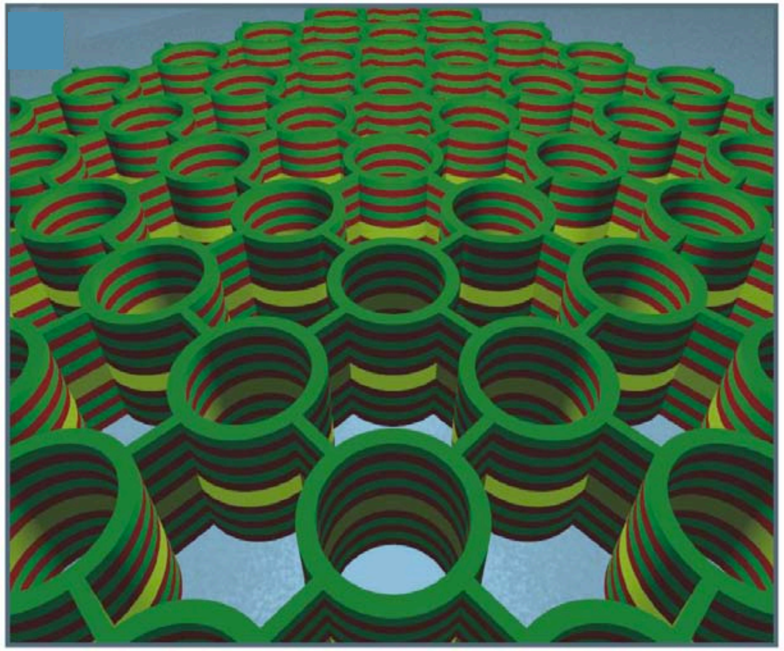
  Screening/many-body physics reduces interaction at high density?

  Disorder effects at low density?
  Taylor, Keeling: clustering of excitons in low energy minima

Polariton lattices

Interaction strength is low compared to kinetic energy of confinement in typical structures.

➡ Coupled condensates, not single particles at lattice sites.



Berloff et al.:
ground state of lattice of coupled condensates can solve NP-hard problems.

Classical or quantum?

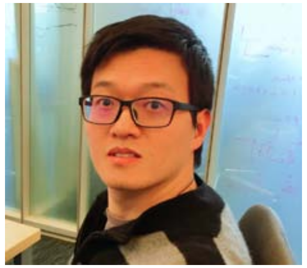
Experimental team

Dr. David Myers

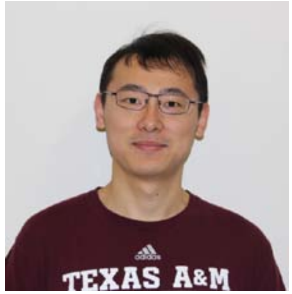
Shouvik Mukherjee

Jonathan Beaumariage

Dr. Burcu Ozden



Dr. Zheng Sun



Qi Yao

Dr. Ryan Balili