Recent Results with Polaritons and Exciton Complexes

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GaAs/AlGaAs sample fabrication:

Loren Pfeiffer, Ken West, Kirk Baldwin, Princeton
Zbig Wasilewski, Waterloo

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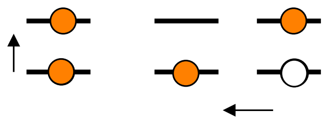
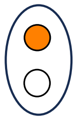
What is an exciton?

Start with a set of two-level quantum oscillators:

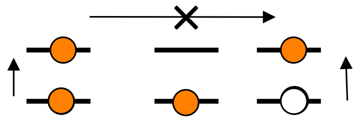


excitation is mobile: an “exciton”



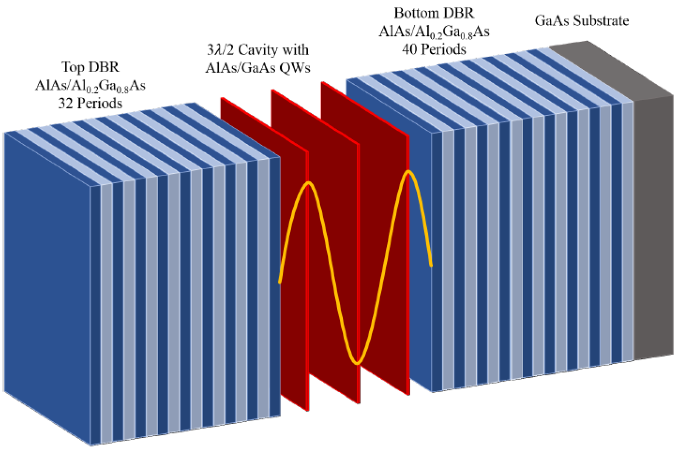
“Wannier” limit: electron and hole orbit in bound state like hydrogen atom.

Two excitations:

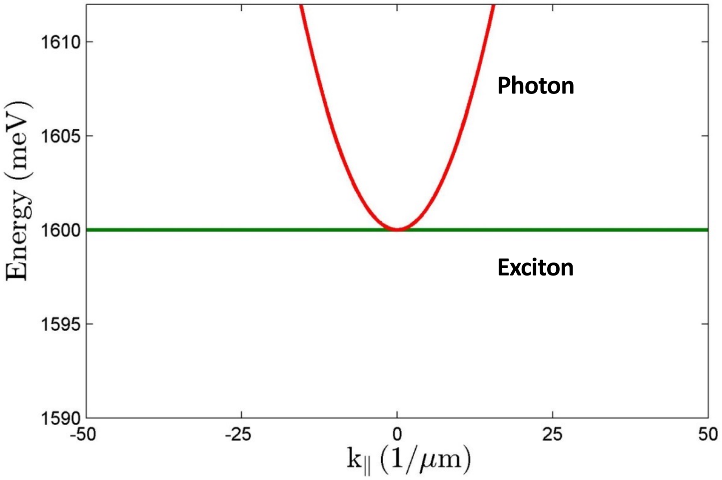


hard-core repulsion of excitons

Polariton states

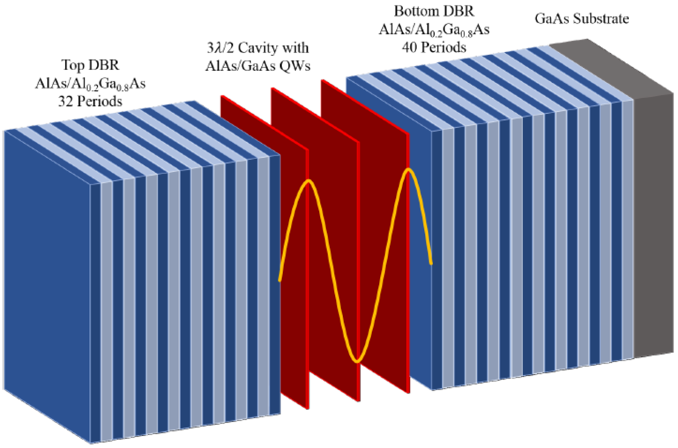


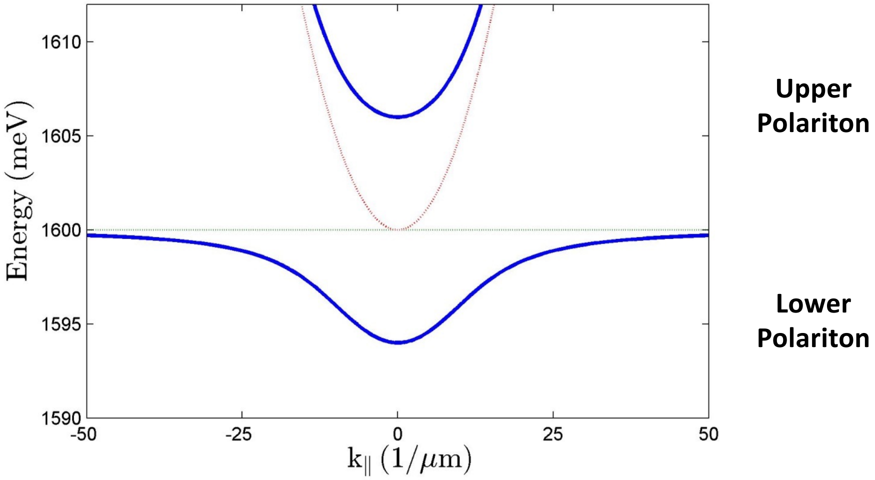
Typical microcavity structure



In-plane
dispersion

Polariton states





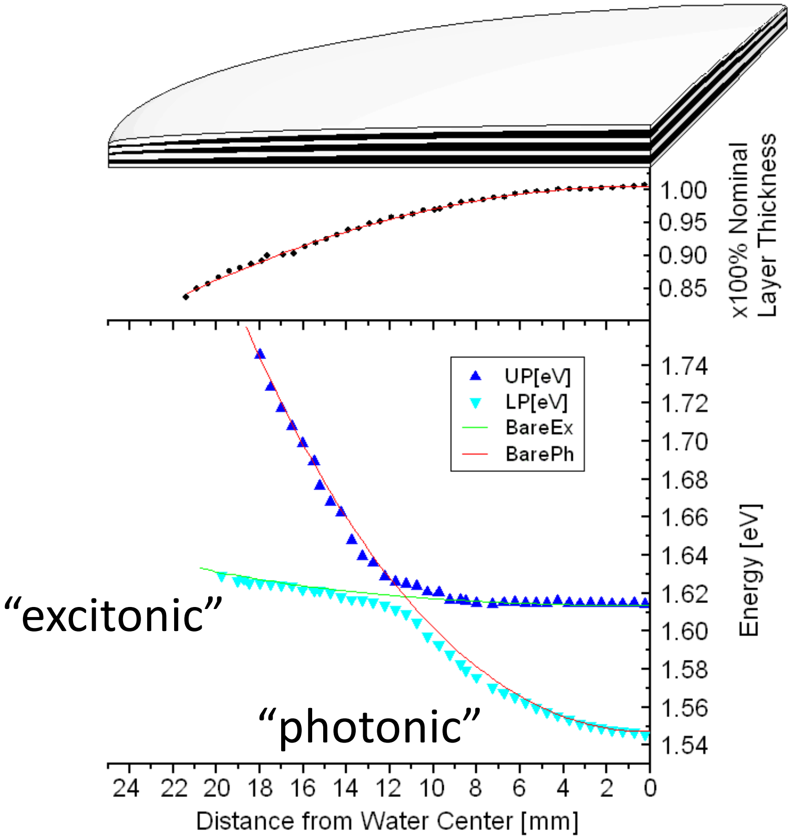
By mixing with exciton states, we have an *interacting*, light-mass boson gas. ($m \sim 10^{-4}\, m_e$)

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

“detuning”
$\delta = E_c - E_x$

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

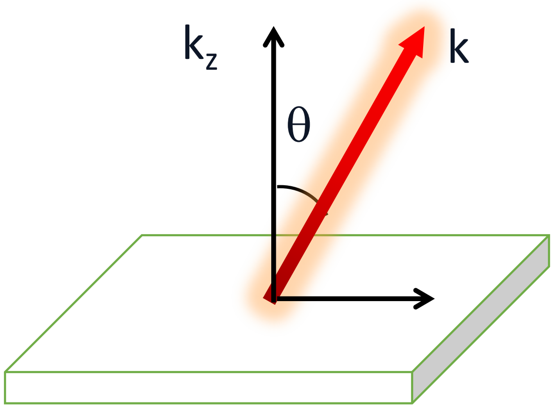
Cavity “wedge” leads to spatially varying potential energy for polaritons



“excitonic”

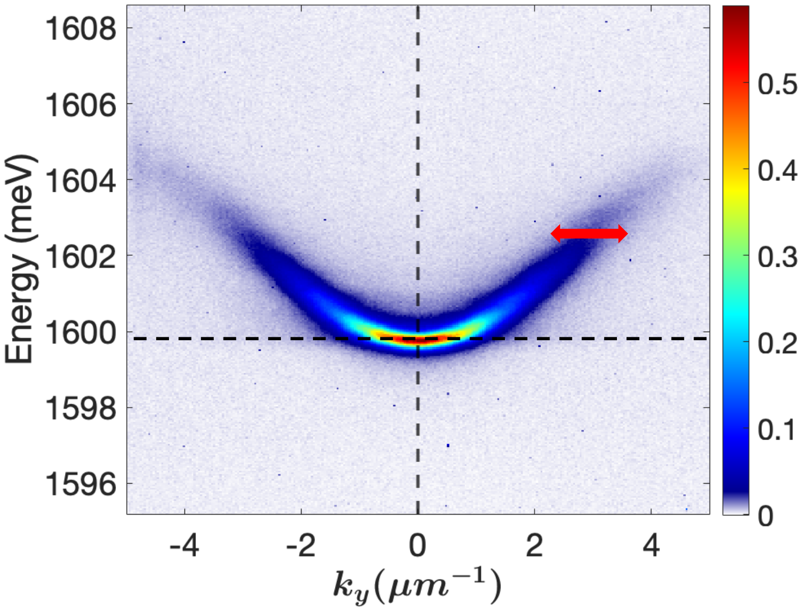
“photonic”

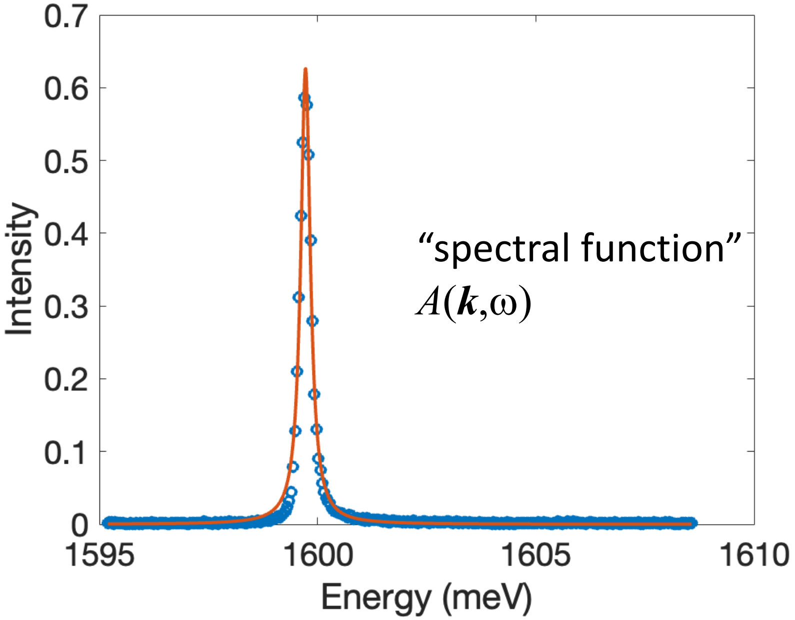
Generalized Snell's Law: Angle-resolved photon emission data give momentum distribution



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

Typical angle-resolved photon emission data

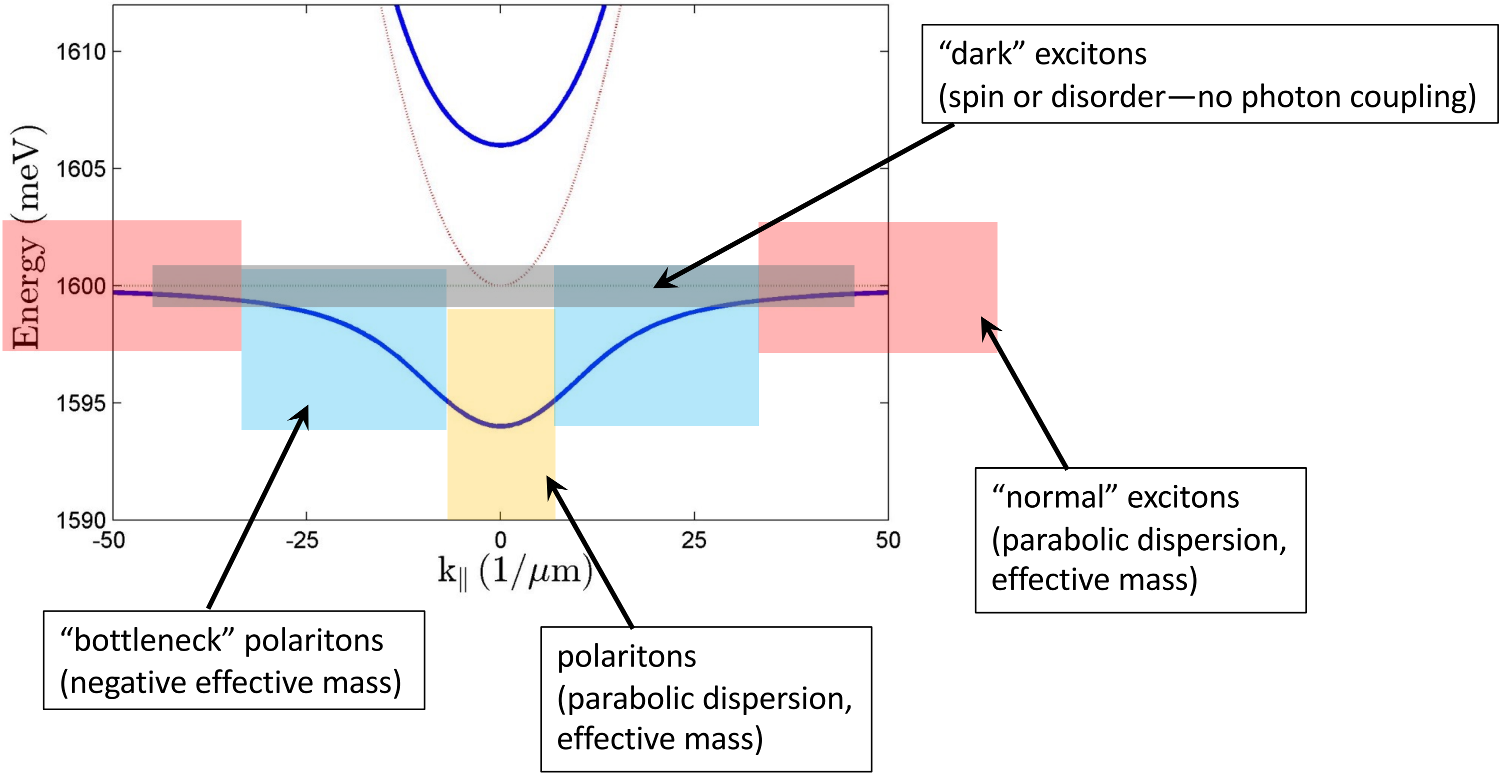




“spectral function”
$A(\boldsymbol{k},\omega)$

Disorder broadening Δk

Polariton dispersion and the “exciton reservoir”



“dark” excitons
(spin or disorder—no photon coupling)

“normal” excitons
(parabolic dispersion,
effective mass)

“bottleneck” polaritons
(negative effective mass)

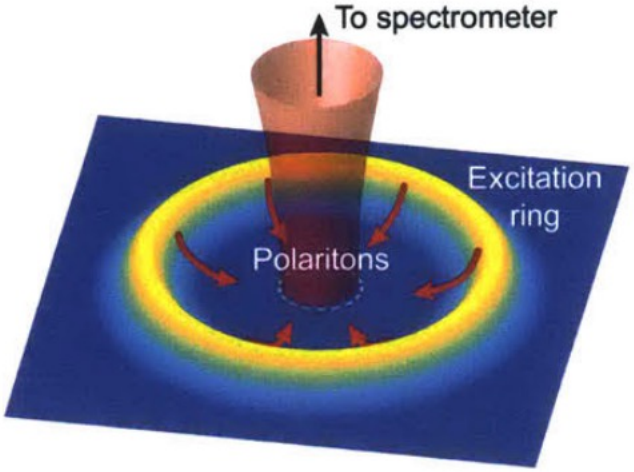
polaritons
(parabolic dispersion,
effective mass)

1. Thermal Equilibrium of Polariton Condensates

Condition for equilibrium: $\tau_{\text{lifetime}} \gg \tau_{\text{therm}}$

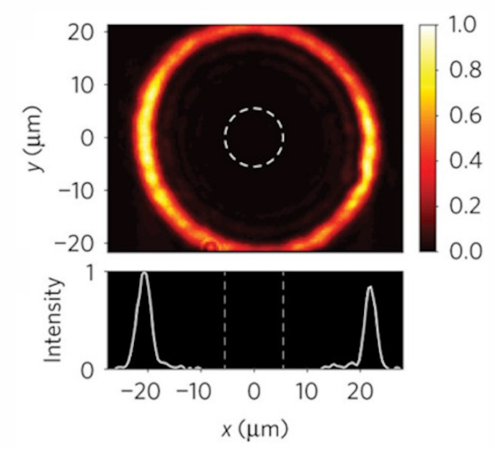
Previous work (MIT-Pitt collaboration)

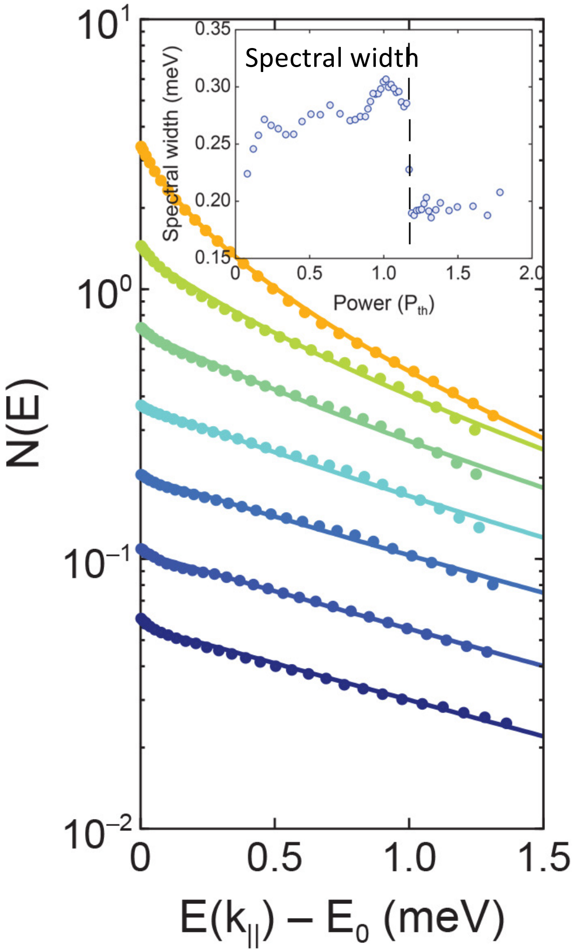
Optical trap used to create uniform condensate



(Figure taken from Yoseob Yoon Ph.D. thesis, MIT, 2019.)

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



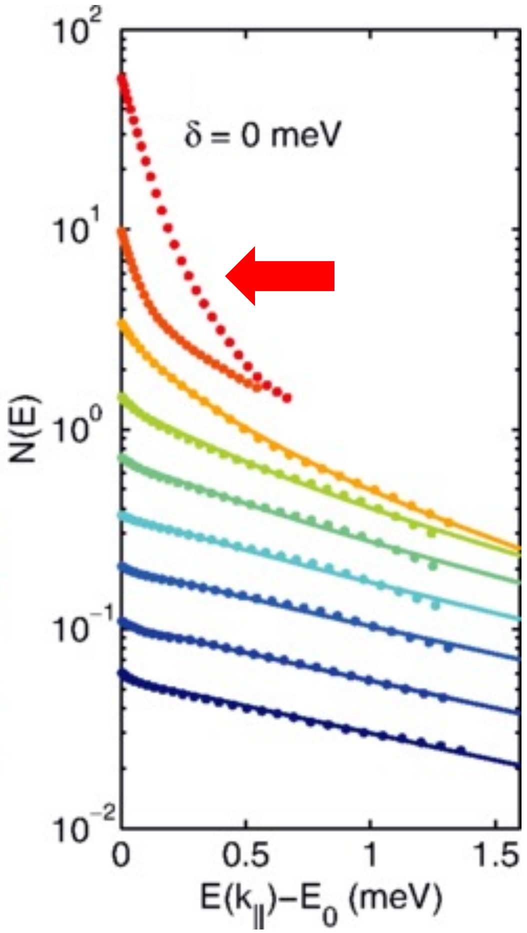


overall height is ***not*** a free parameter: fixed by μ.

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

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

Breakdown of BE fits at high density:
Spatial inhomogeneity

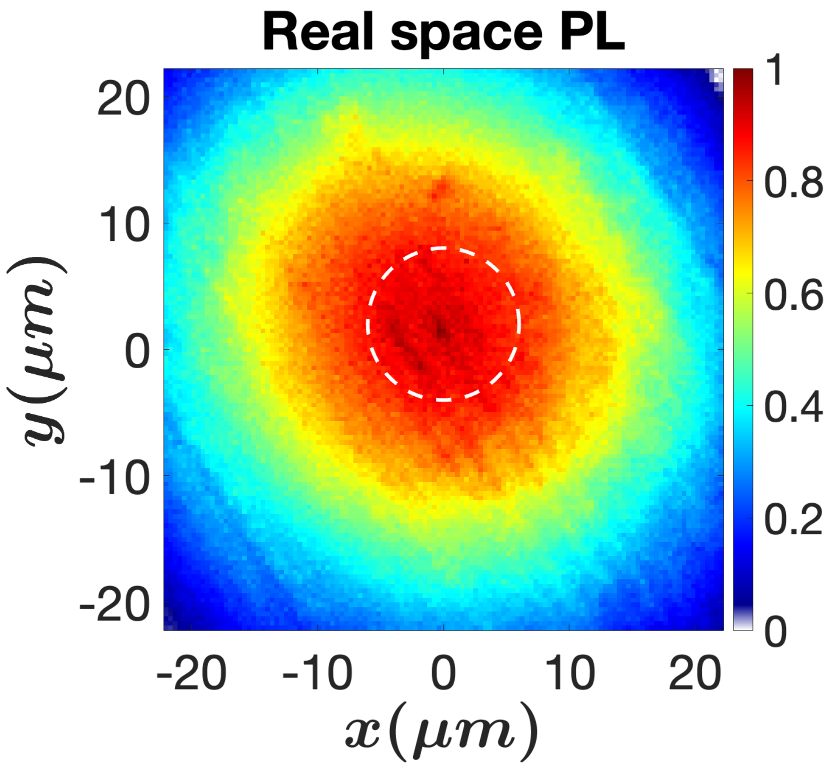


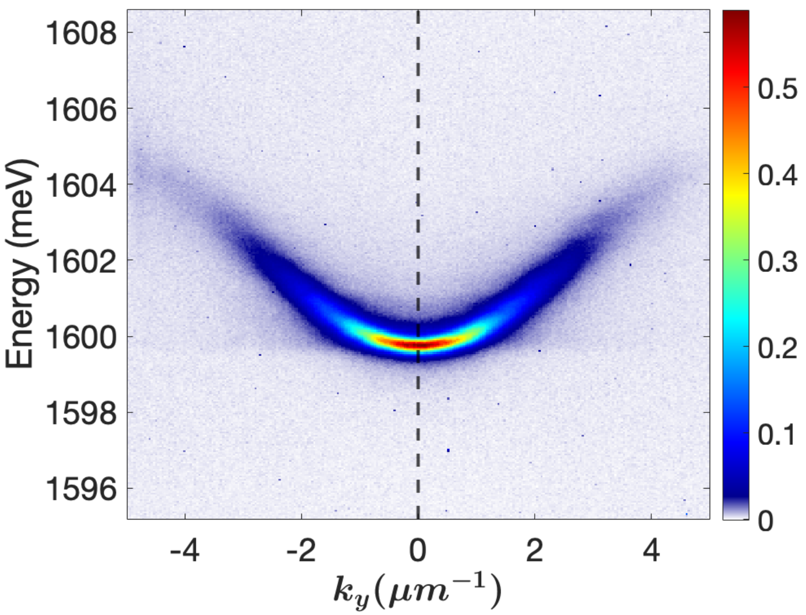
New experiments (Alnatah): wide area, homogeneous steady state

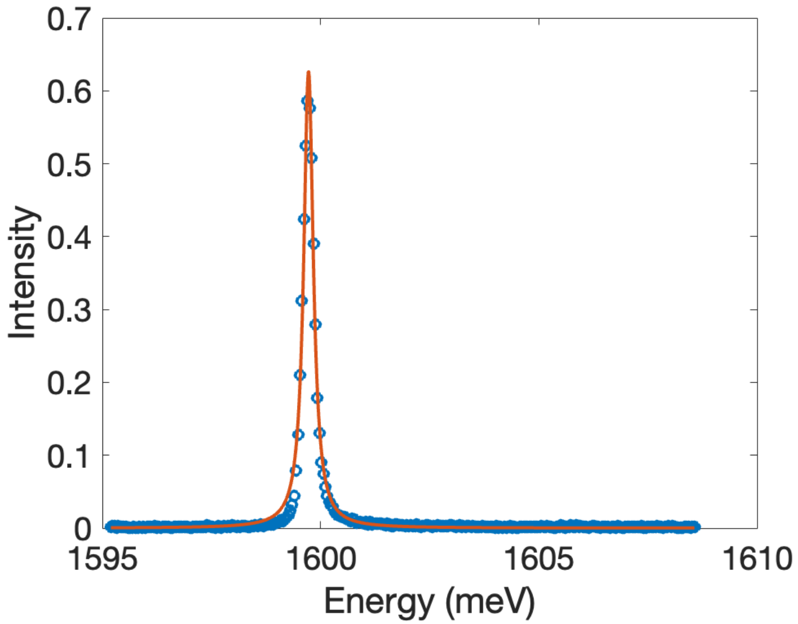
Science Advances **10**, eadk6960 (2024).

Spatial imaging/filtering selects only very uniform center.

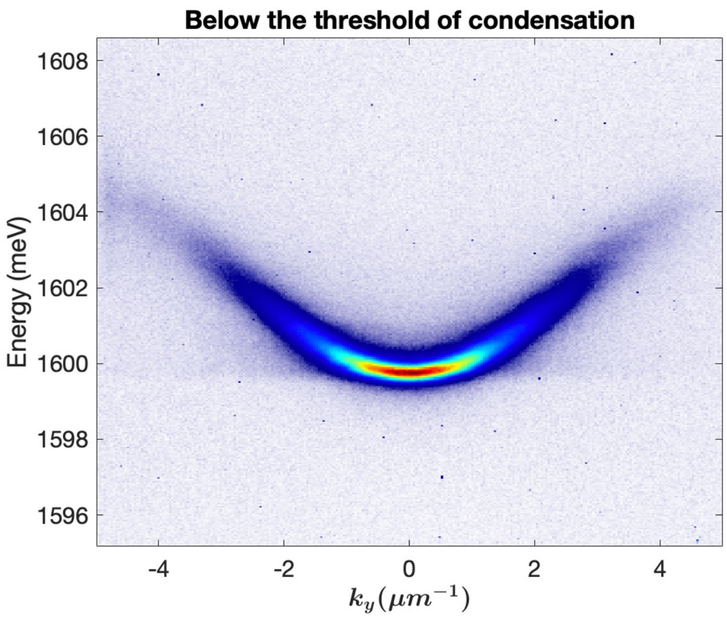
New sample has almost zero wedge in detuning range of interest.

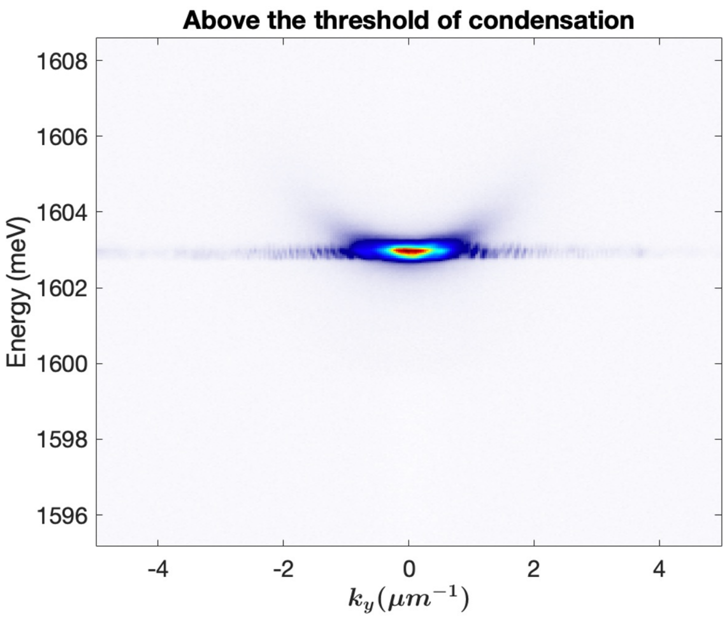




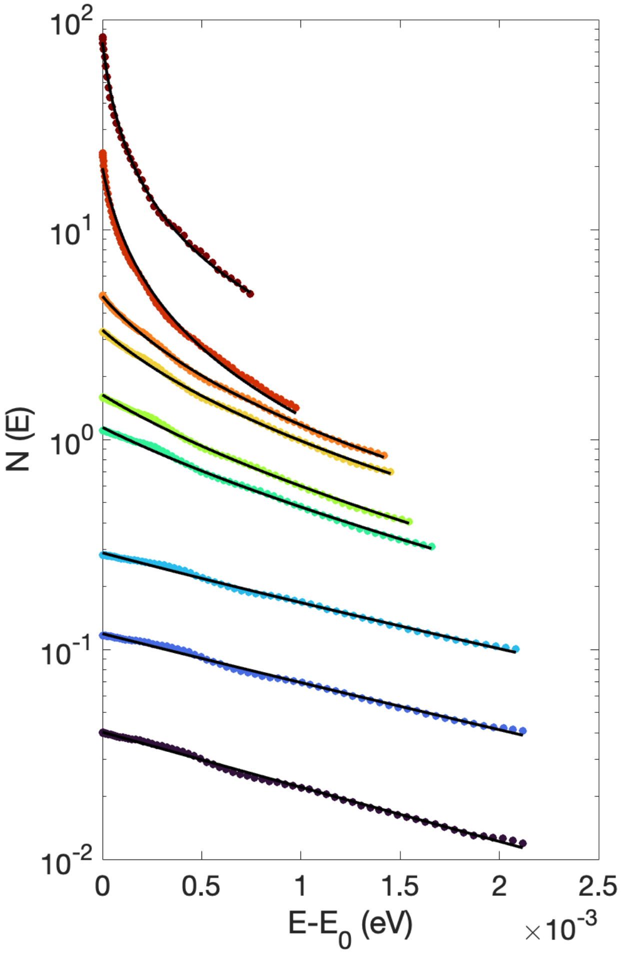


$$N(k_i) = \eta\tau(k_i)\int I(k_i, E)\,dE$$

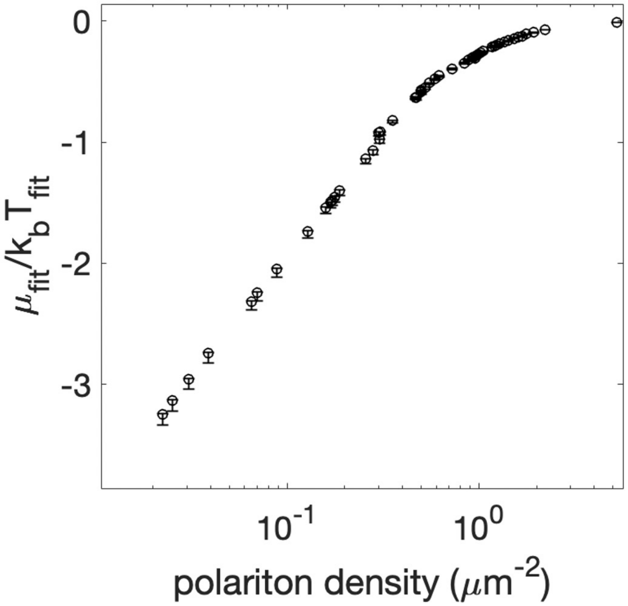

Below the threshold of condensation


Above the threshold of condensation

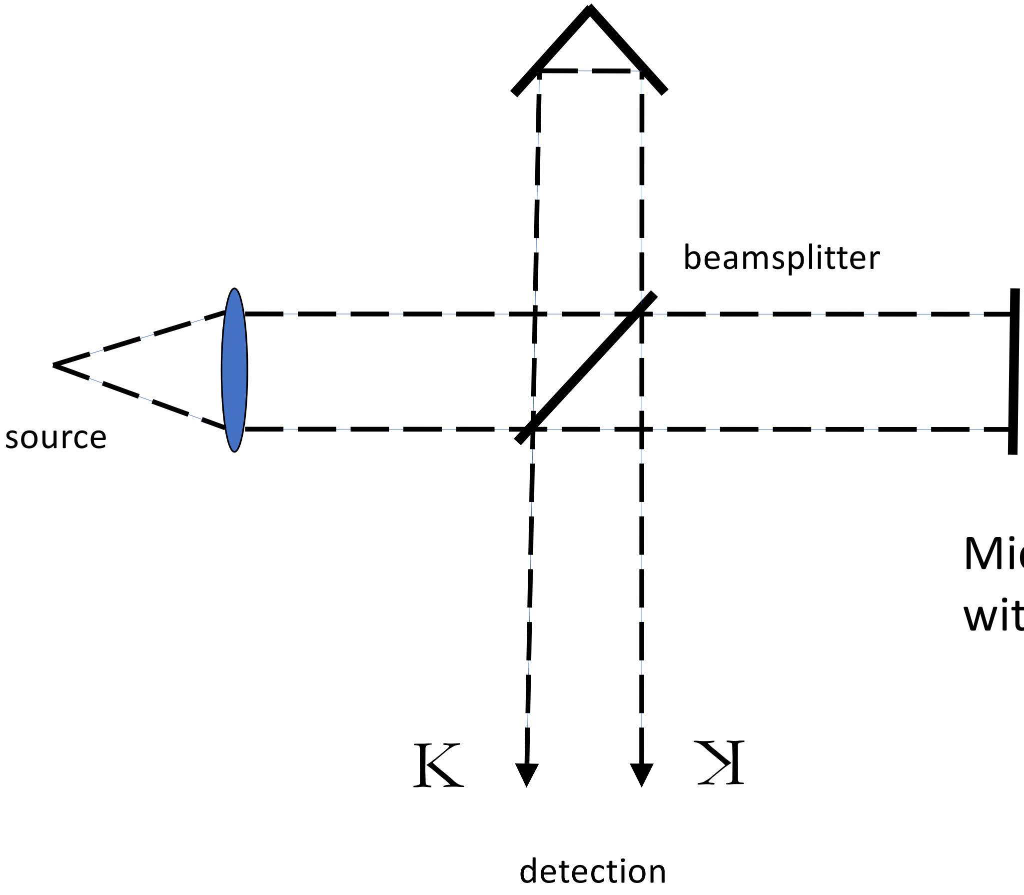
- The solid black lines are fits to the Bose-Einstein distribution.
- The best fits of the data were determined using T and μ as free parameters
- A single efficiency factor A was used for all the distributions so that the absolute occupation numbers of different distributions could be compared.



Extracted chemical potential from fits

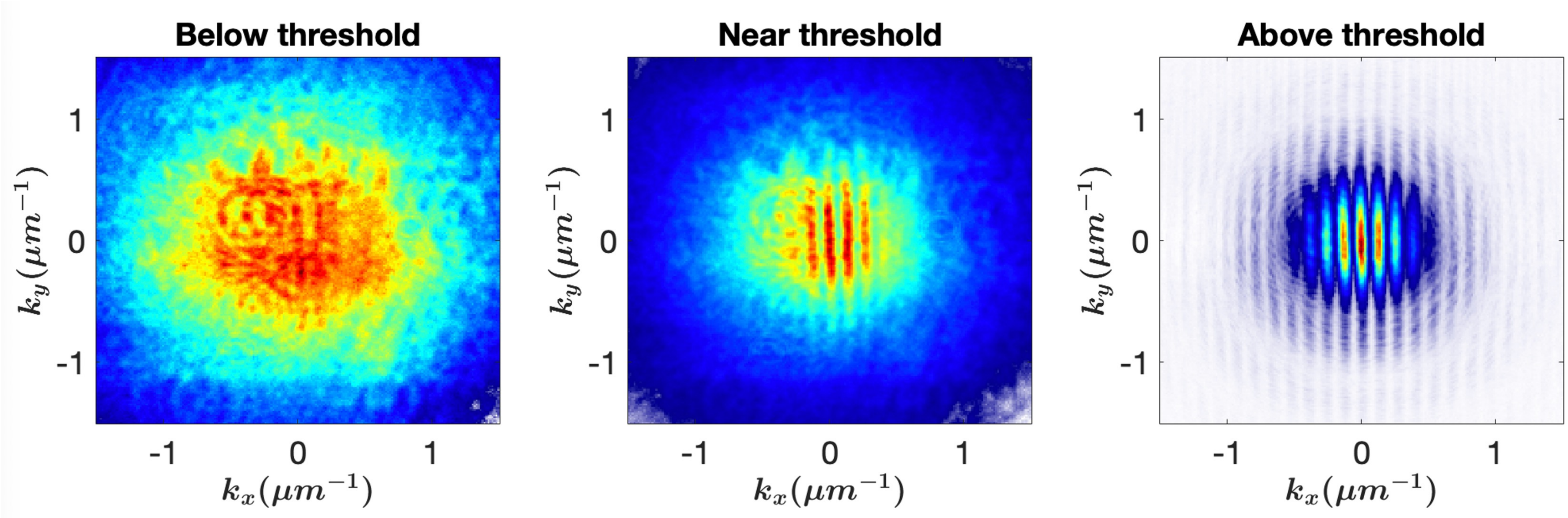


Interferometry: a measure of coherence

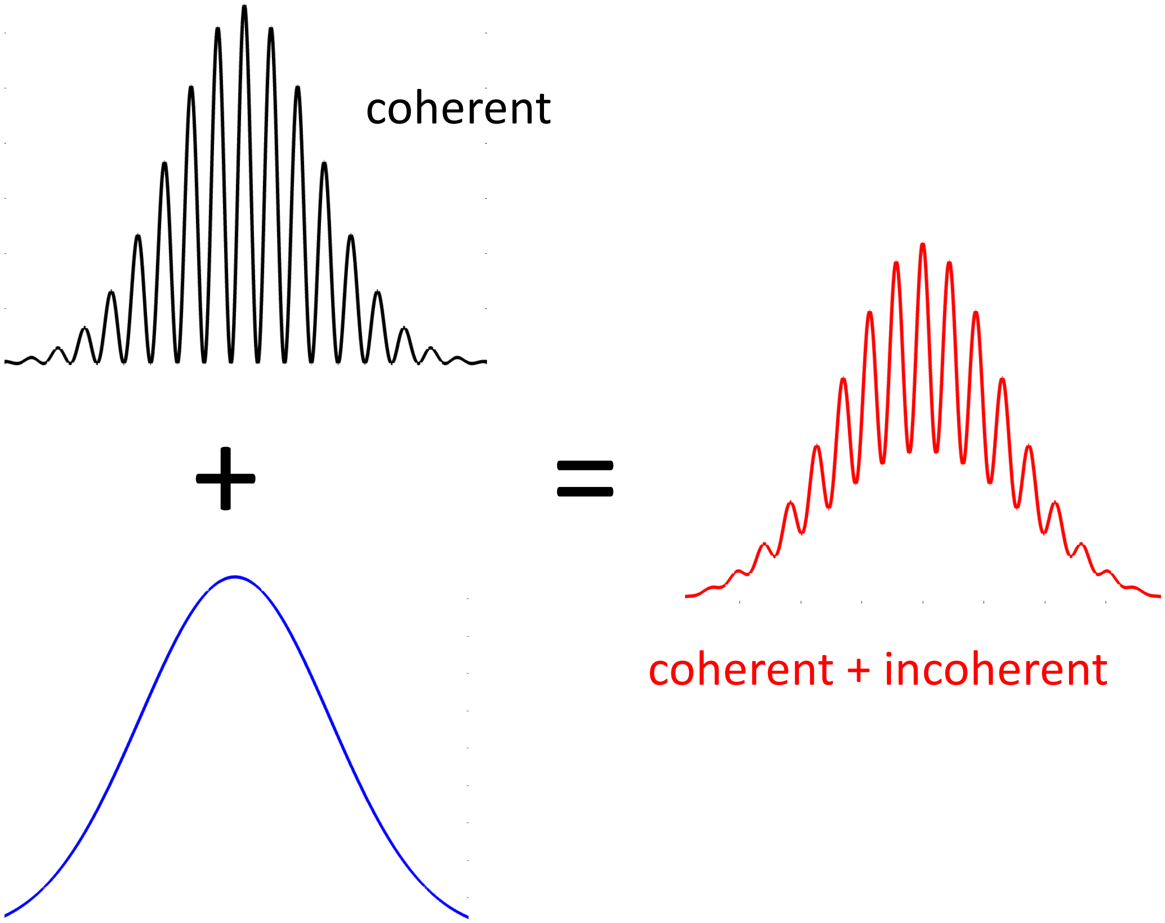


Michelson interferometer
with flip of x-axis

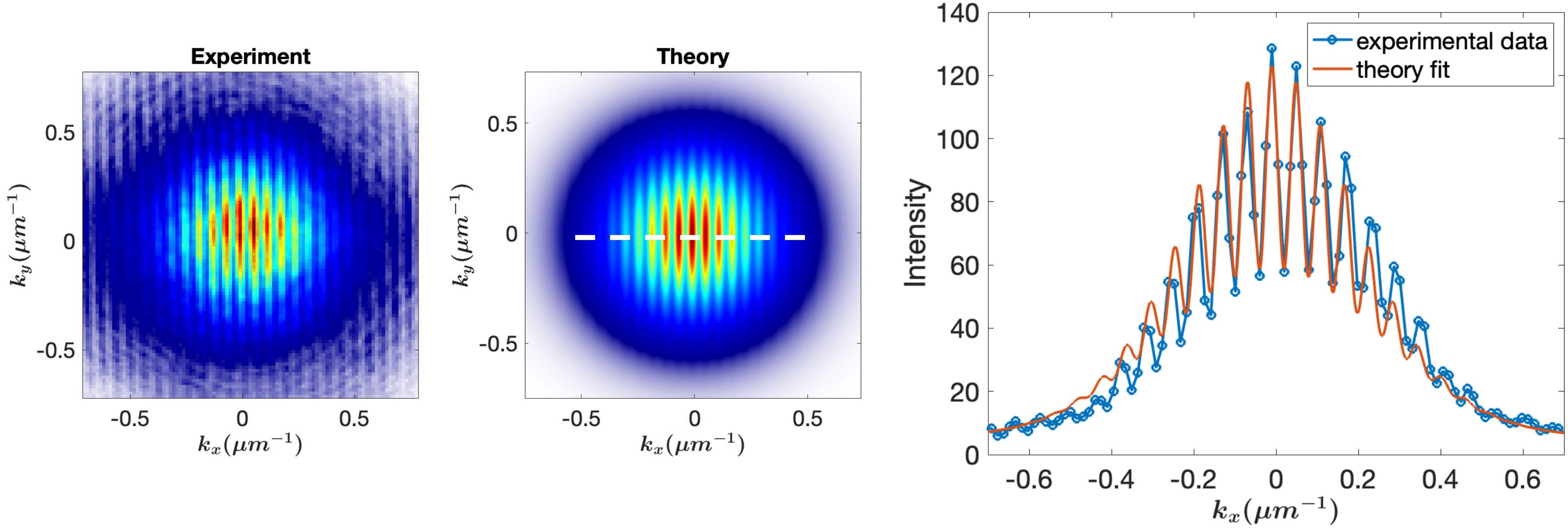
Measuring coherence in k-space



- Assume that the thermal particles are a radial Gaussian, which is <u>unaffected</u> by interferometry.
- Assume the condensate amplitude is a radial Gaussian that gives interference fringes.
- Fit the sum of these two to the data.

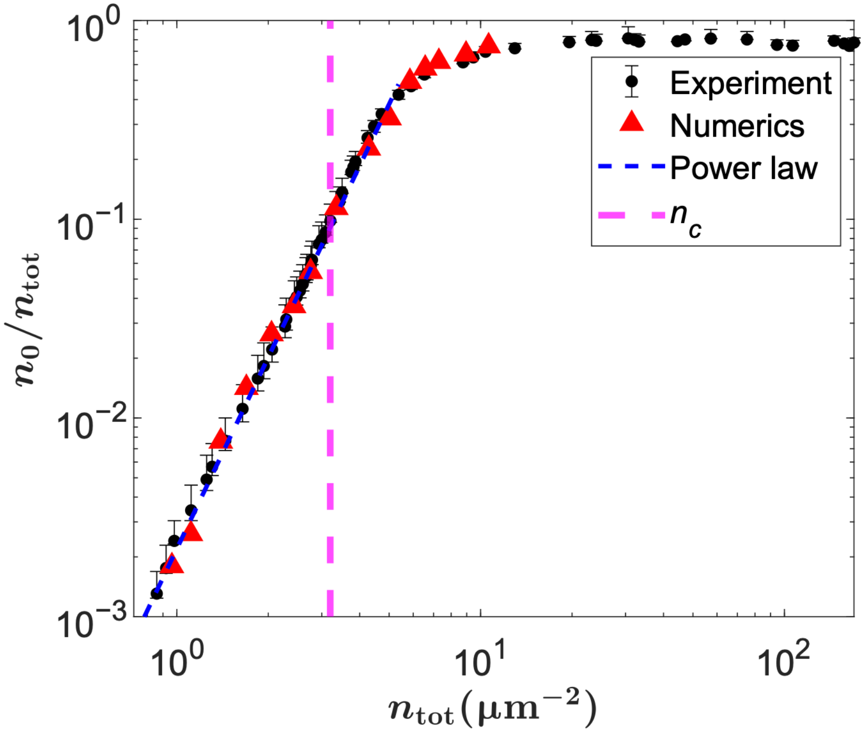


Linewidth and condensate fraction



Power Law of Coherence Fraction at Low Density

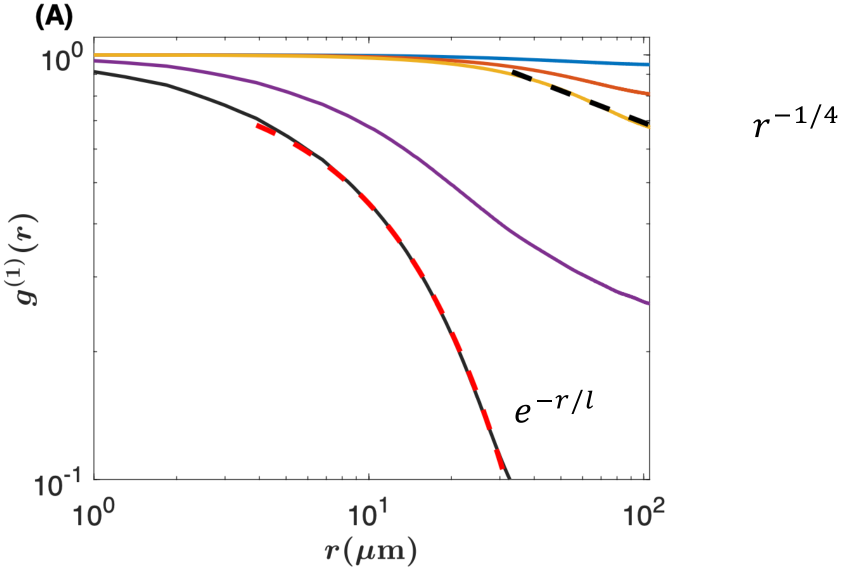
$$\frac{n_{coh}}{n_{tot}} \propto {n_{coh}}^{3.2}$$



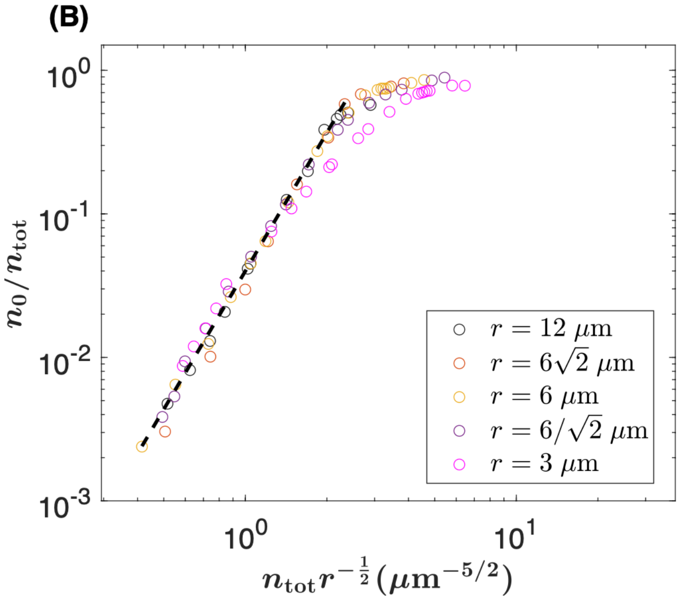
Numerical Simulation using G-P equation with *no gain or loss*

$$i\hbar\frac{\partial\psi(\mathbf{r},t)}{\partial t} = \left[-\frac{\hbar^2\nabla^2}{2m} + g_c|\psi(\mathbf{r},t)|^2\right]\psi(\mathbf{r},t)$$

Correlation as density is varied



Scaling with aperture size



Weakly interacting analytical theory

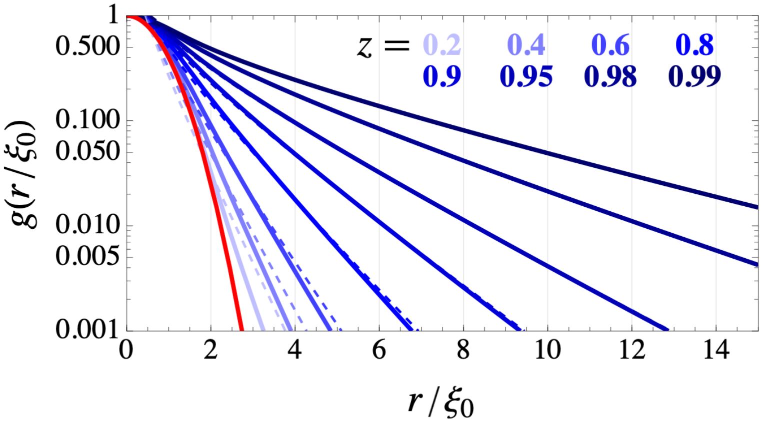
Joseph Jachinowski and Peter Littlewood

$$G^{(1)}\left(\boldsymbol{r},\boldsymbol{r}'\right)=\frac{1}{\lambda_T^2}g_1\left(z,e^{-\pi\left(\boldsymbol{r}-\boldsymbol{r}'\right)^2/\lambda_T^2}\right)$$

$$g_\nu\left(x,y\right)=\sum_{j=1}^{\infty}\frac{x^j y^{1/j}}{j^\nu}$$

Series expansion of Bose-Einstein distribution

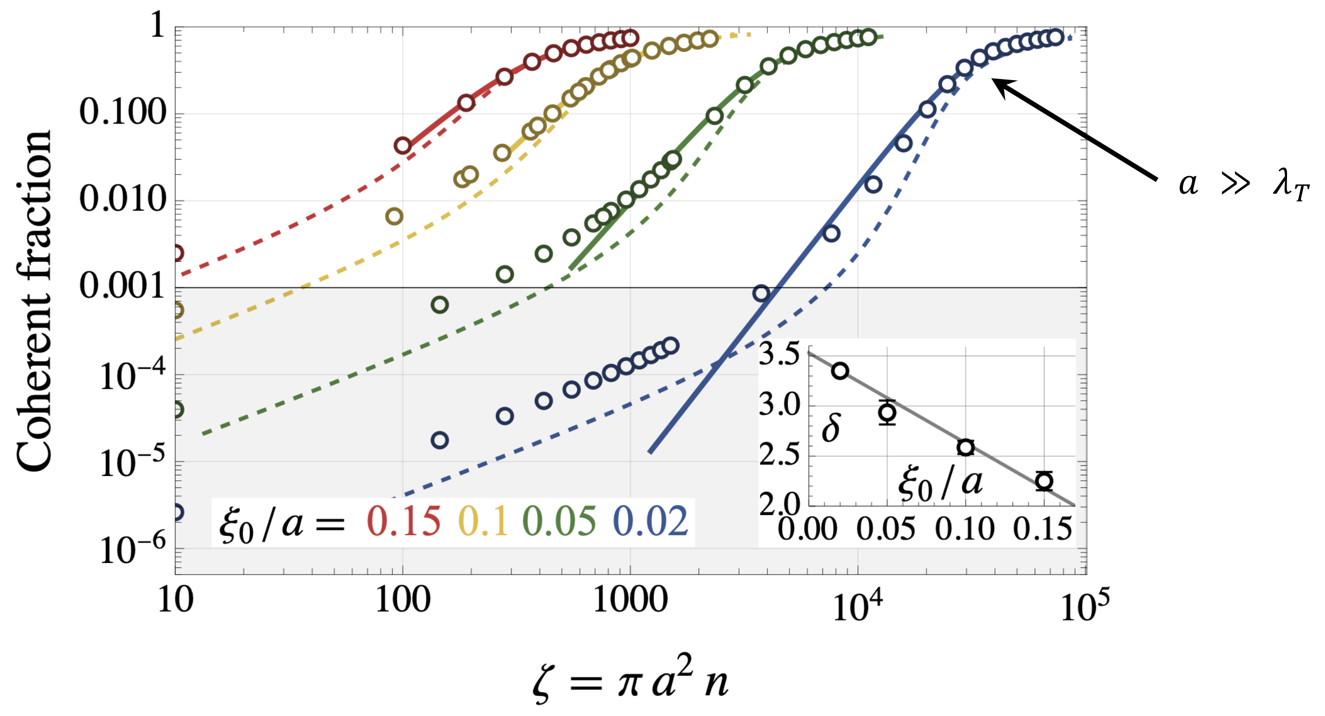
Correlation as density is varied



Visibility function:

$$\mathcal{V}\left(\boldsymbol{r},\tau\right)=\frac{\epsilon_0 c}{2\left\langle I_S\left(\boldsymbol{r},\tau\right)\right\rangle}\mathrm{Re}\left\{G^{(1)}\left(\left(x,y,z\right),\left(-x,y,z\right),\tau\right)\right\}$$

Numerical calculation of analytical solution accounting for finite radius of observation area

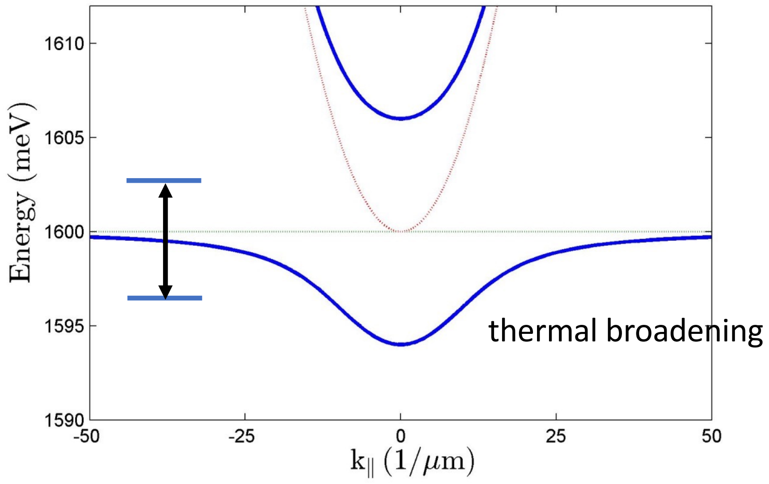


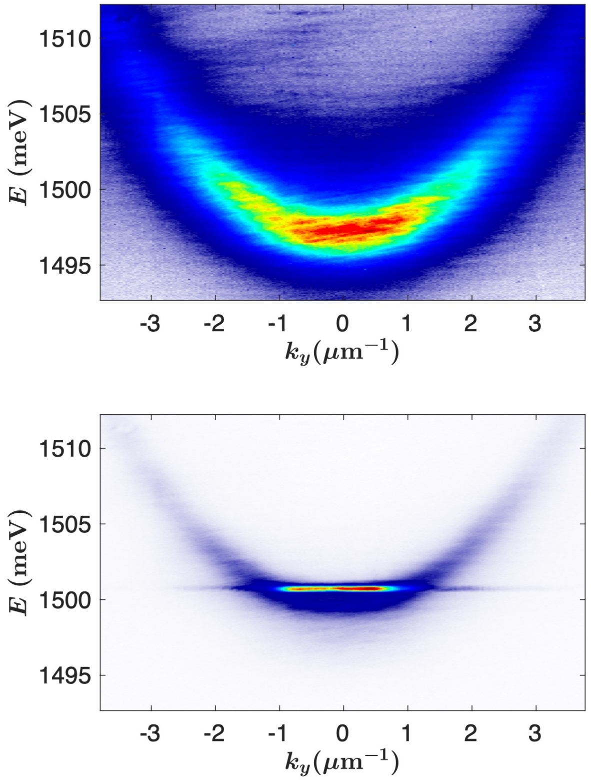
Notes on 2D Bose condensates:

- Coherence length can be much larger than the size of the system
- No sharp cutoff in behavior of k=0 state and nearby k-states
- No sharp change of coherence at superfluid threshold
- In superfluid regime, coherent fraction = superfluid fraction

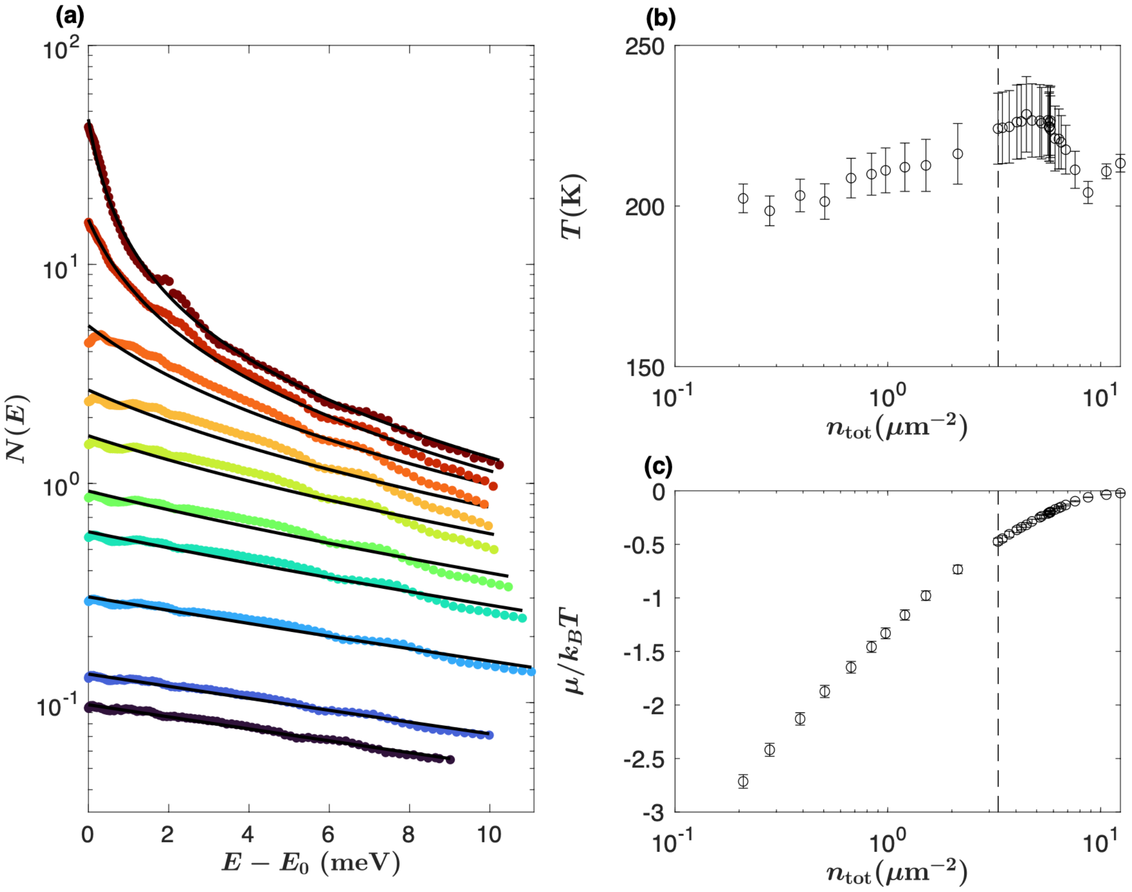
Room Temperature Polariton Condensate in the Weak-Coupling Regime

Alnatah et al., 2024 (arXiv:2406.13689)





GaAs exciton polaritons at room temperature!

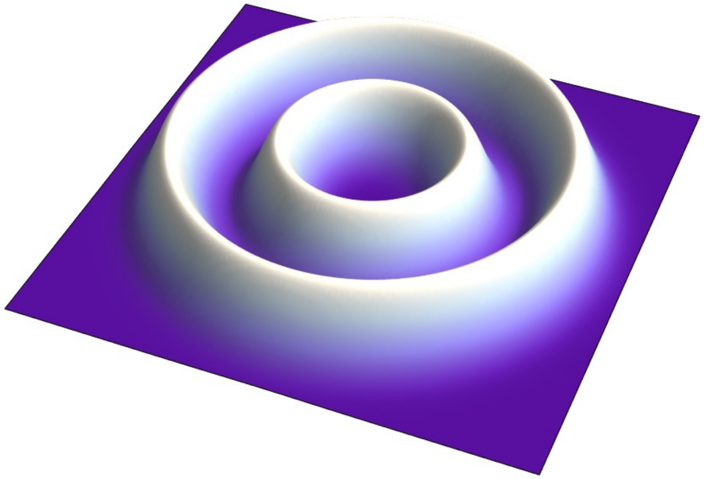


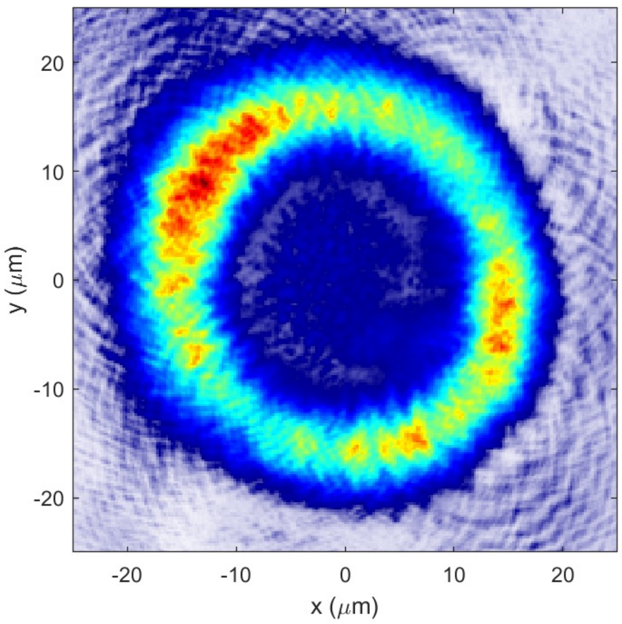
T < 300 K!

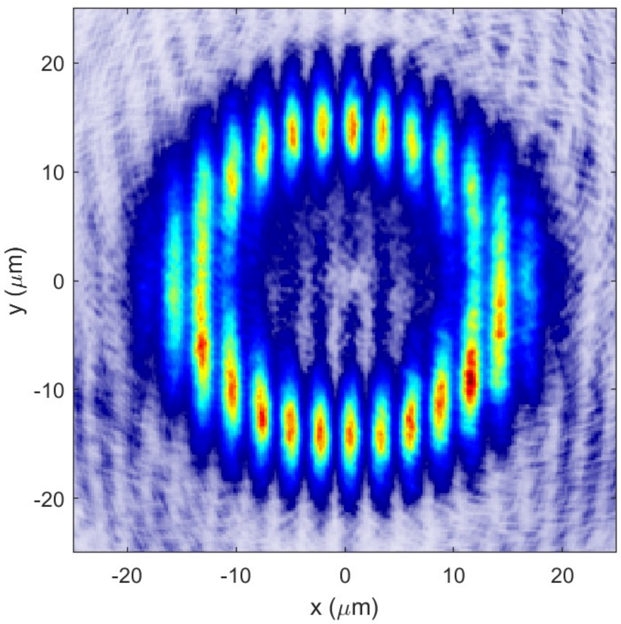
cf. M. Pieczarka, et al., 2023
(arXiv:2307.00081)

2. Persistent circulation in a ring trap

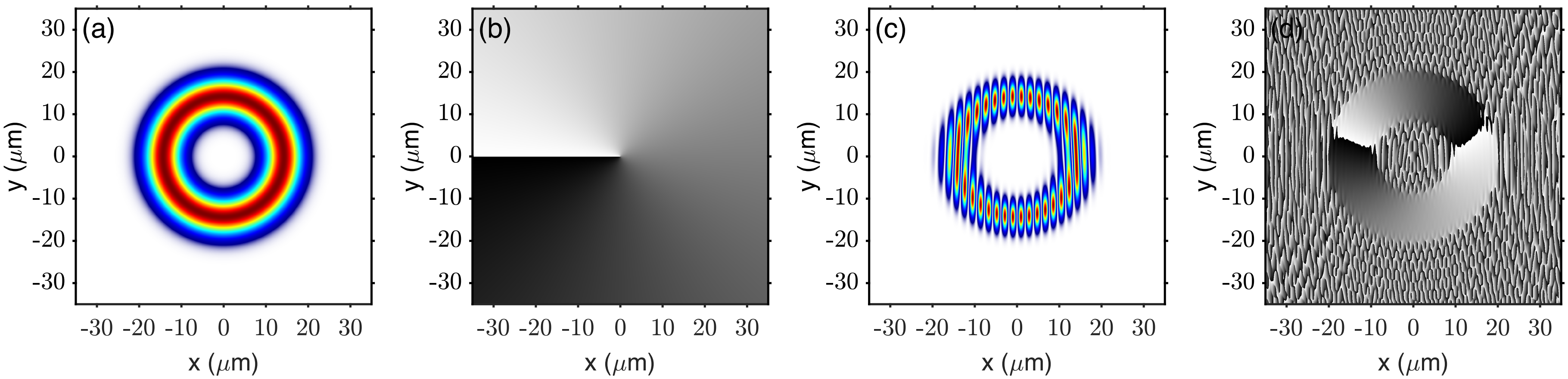
Q. Yao et, arXiv:2302.07803




Steady-state condensate


Real-space interference

Theory prediction for circulating condensate



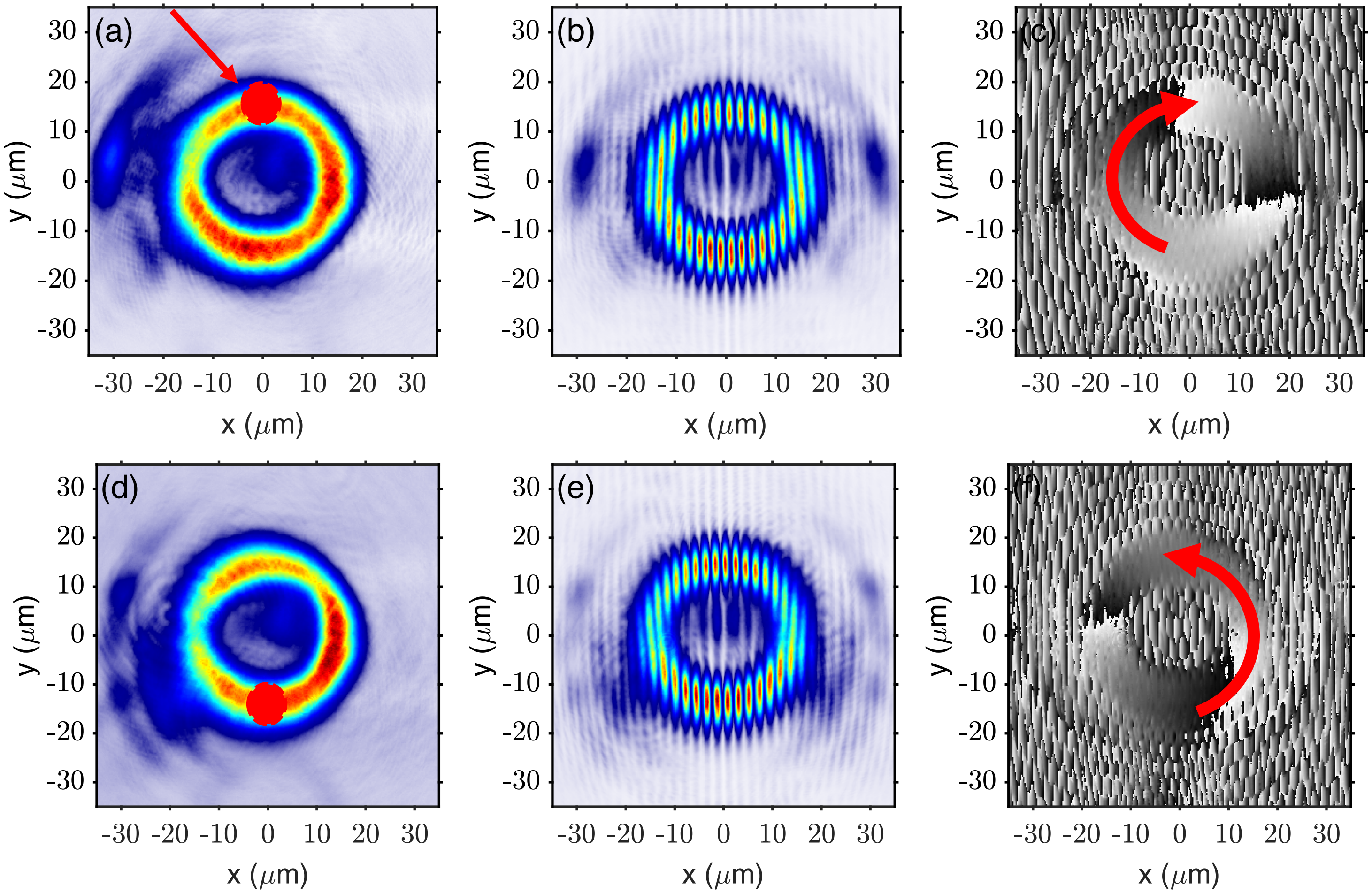
Condensate density

Phase of condensate

Predicted interference pattern

Phase map

Excite with short (2 ps) nonresonant pulse: circulation



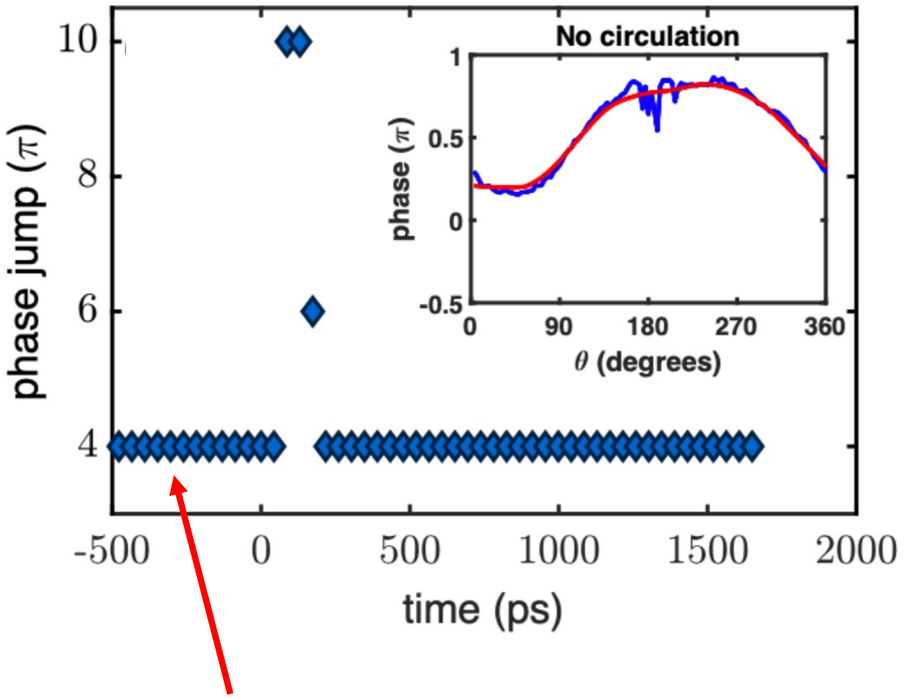
Pulsed excitation

Circulating in one direction

Circulating in other direction

Circulates *only* when probe pulse kicks it.

Long-lasting phase stability: no discernible degradation



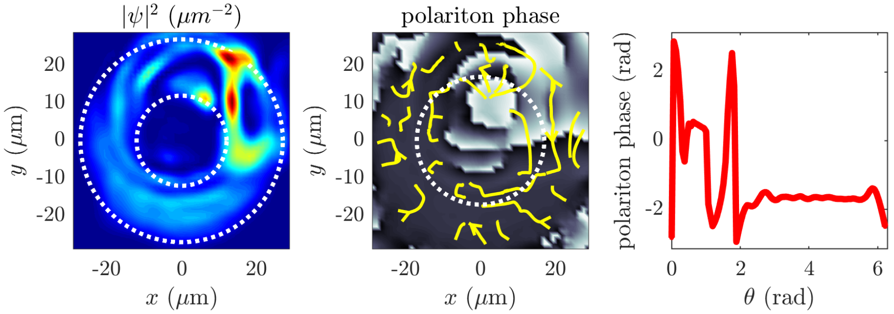
Phase winding is absolutely stable for 14 ns!

No prior experiments have shown zero decay of circulation!

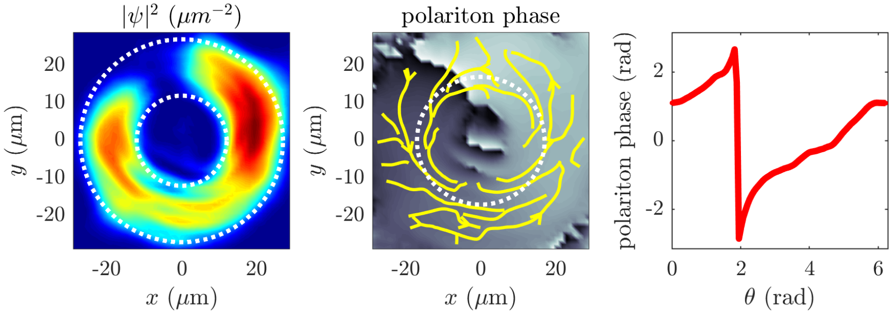
Theoretical model of symmetry breaking by a scalar potential

Comaron and Szymanska,
truncated Wigner approximation
with exciton reservoir

$t = 0$
asymmetric probe



$t = .3$ ns
asymmetric probe



Many vortices created by the “blast,” all recombine except two; one stays in the ring and the other migrates away.

Symmetry breaking due to position of pulse relative to slight density variations.

How many circulations?

$$v = \frac{\hbar}{m}\nabla\theta = \frac{\hbar}{m}\frac{2\pi}{2\pi R} \approx 0.1\ \mu\text{m/ps}$$

$$\Delta T = \frac{2\pi R}{v} \approx 800\ \text{ps}$$

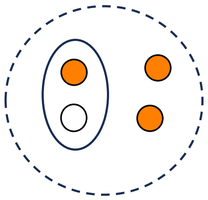
15-20 circulations between laser pulses!

(cf. particle lifetime ≈ 400 ps)

3. A new field: “Quaternions”

Doubly charged excitons = charged bosons

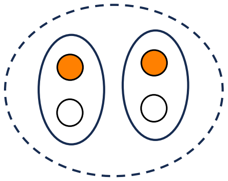
Charged bosons = preformed pairs, can lead to new type of superconductivity!



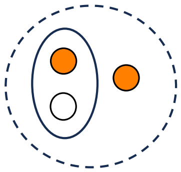
(Not in a cavity – no polariton effect)

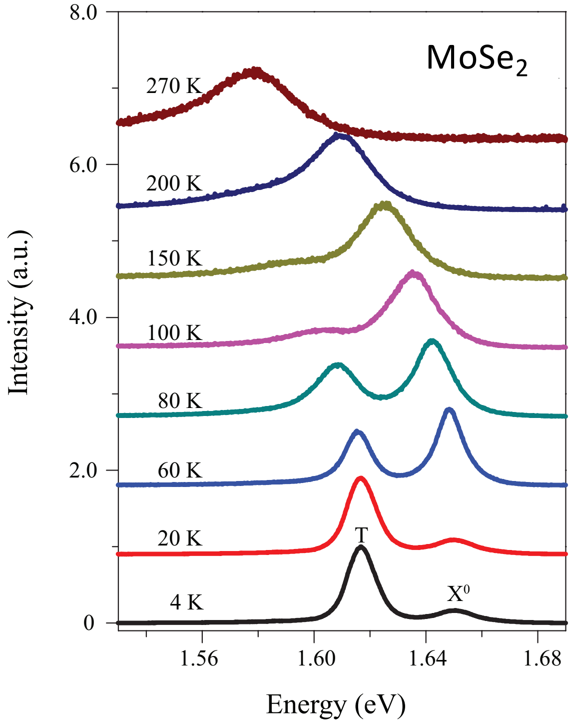
Well-known exciton complexes

Biexcitons (a.k.a. excitonic molecules) (1970s)

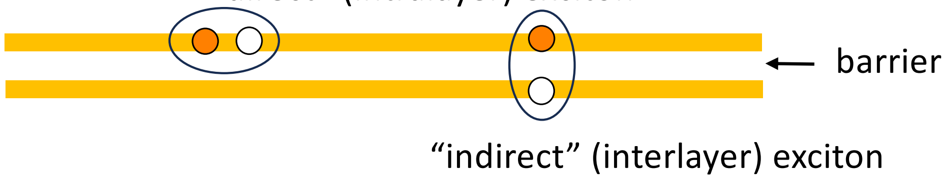


Trions (1990s)





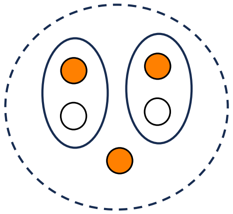
New possibilities with bilayer systems

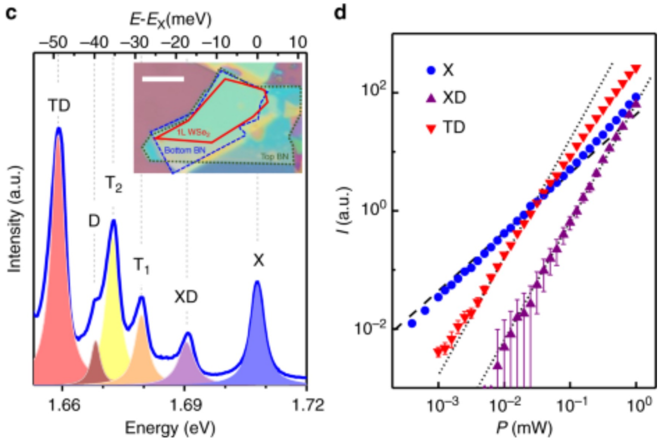


Moiré lattice effects generally occur for very thin barrier (a few lattice constants); washed out for thick barrier.

New excitonic complexes in 2D materials

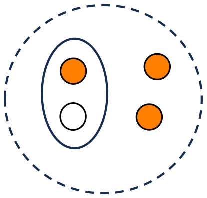
Five-particle complex: charged biexciton





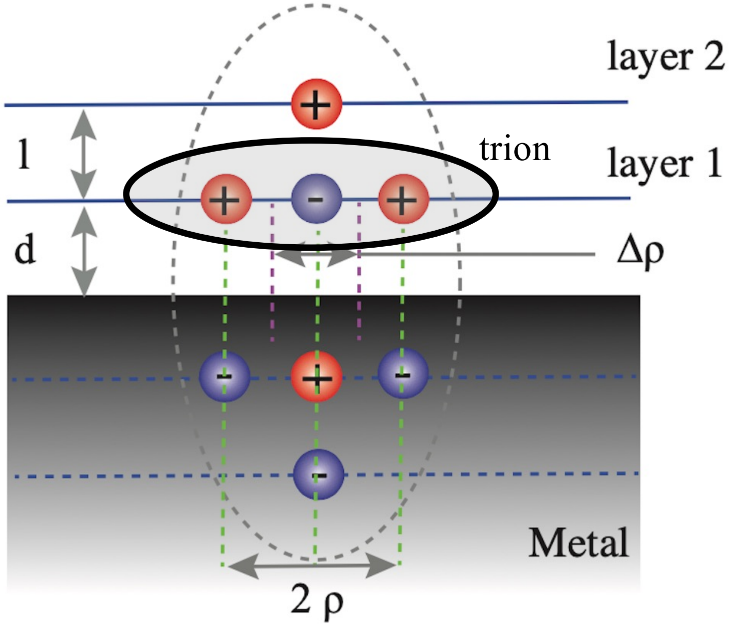
S.-Y. Chen et al., *Nature Comm.* **9**, 3717 (2018).

Doubly charged exciton (“quaternion,” a.k.a. “tetramer,” “quadron”)



Analogous state in hydrogen is metastable,
lies above hydrogen energy

Metal layer provides image charge to cancel most repulsion (screening)



Trions are stable in doped 2D layers.
e.g., Philip Kim group, *Science* **366**, 870 (2019)

Theory of trions:
I.V. Bondarev, M.R. Vladimirova, *Phys. Rev. B* **97**, 165419 (2018);
I.V. Bondarev, O.L. Berman, R.Ya. Kezerashvili, and Y.E. Lozovik, arXiv:2002.09988.

Treat quaternion as a bound state of a trion and a spatially separated free carrier.

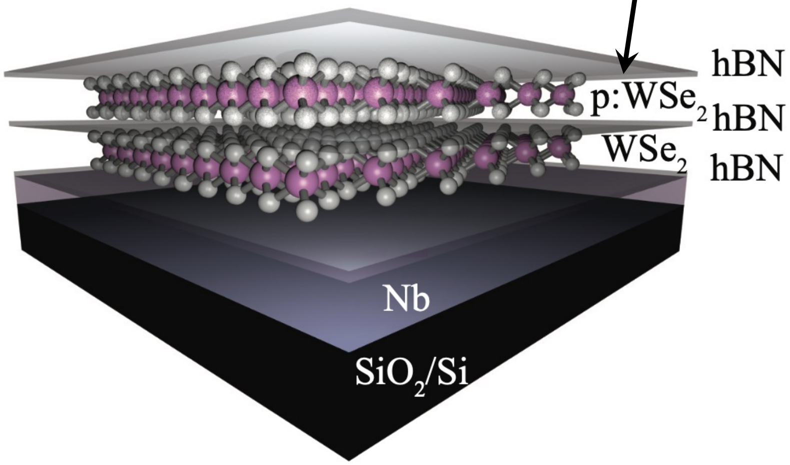
Experimental results

Z. Sun et al., *Nano Letters* **21**, 7669 (2021).

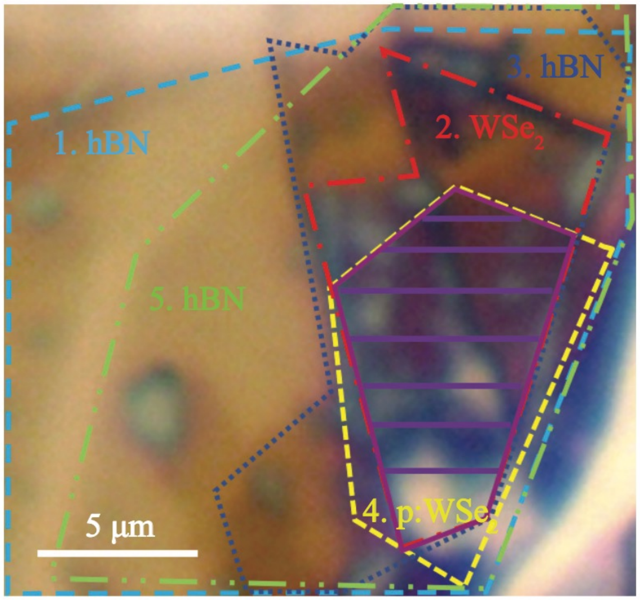
microscope image

Exfoliated stack:

doped layer gives trion formation

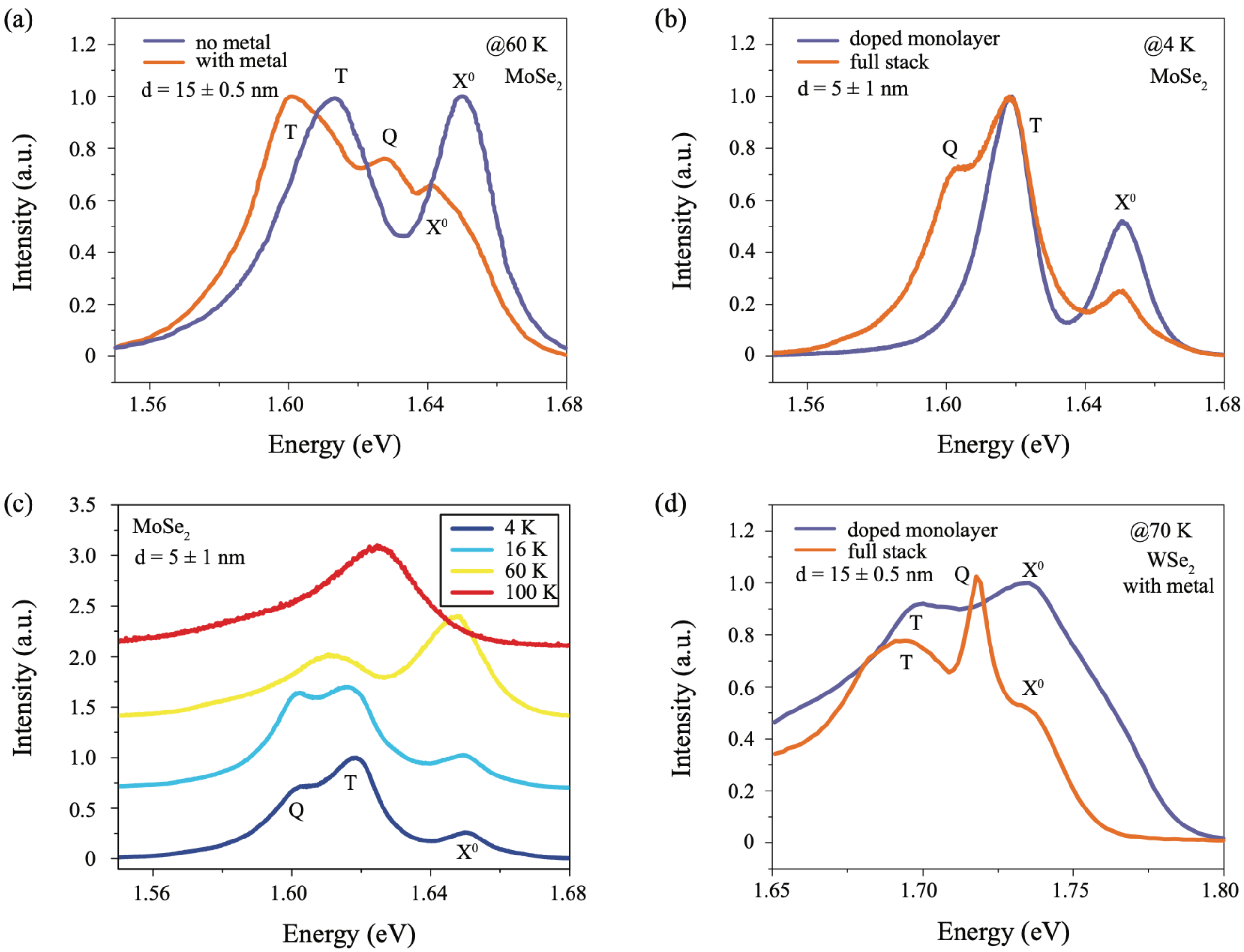


hBN gives high interlayer barrier

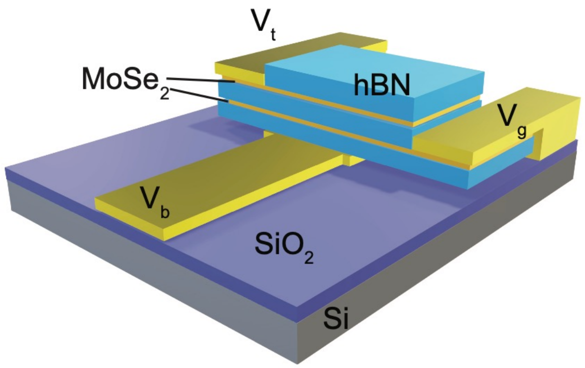


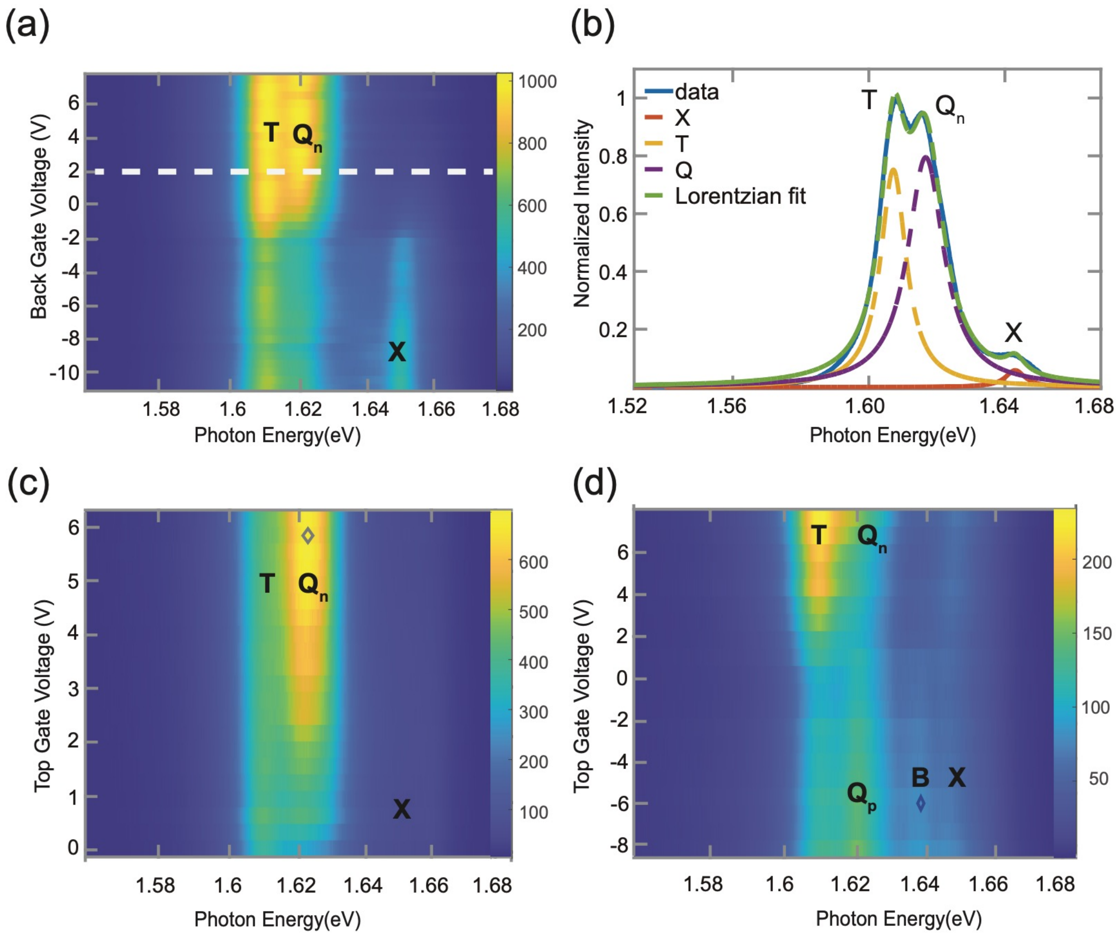
reproduced in $p:WSe_2$ and $n:MoSe_2$

many control samples

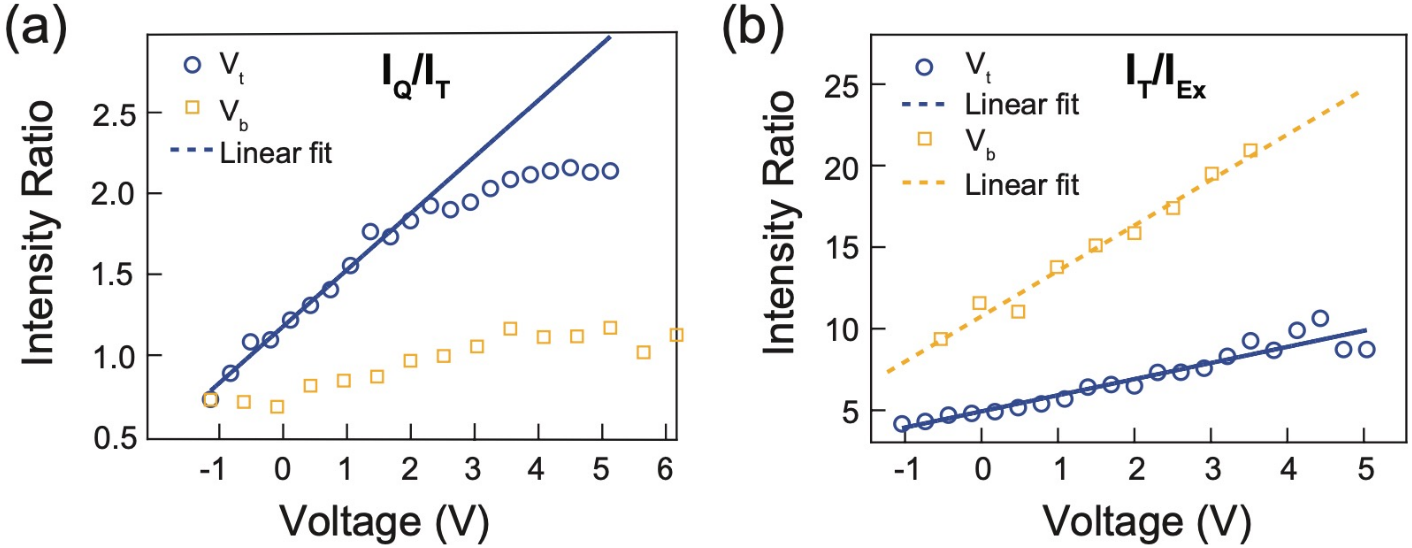


Vary background charge density with electric gating:

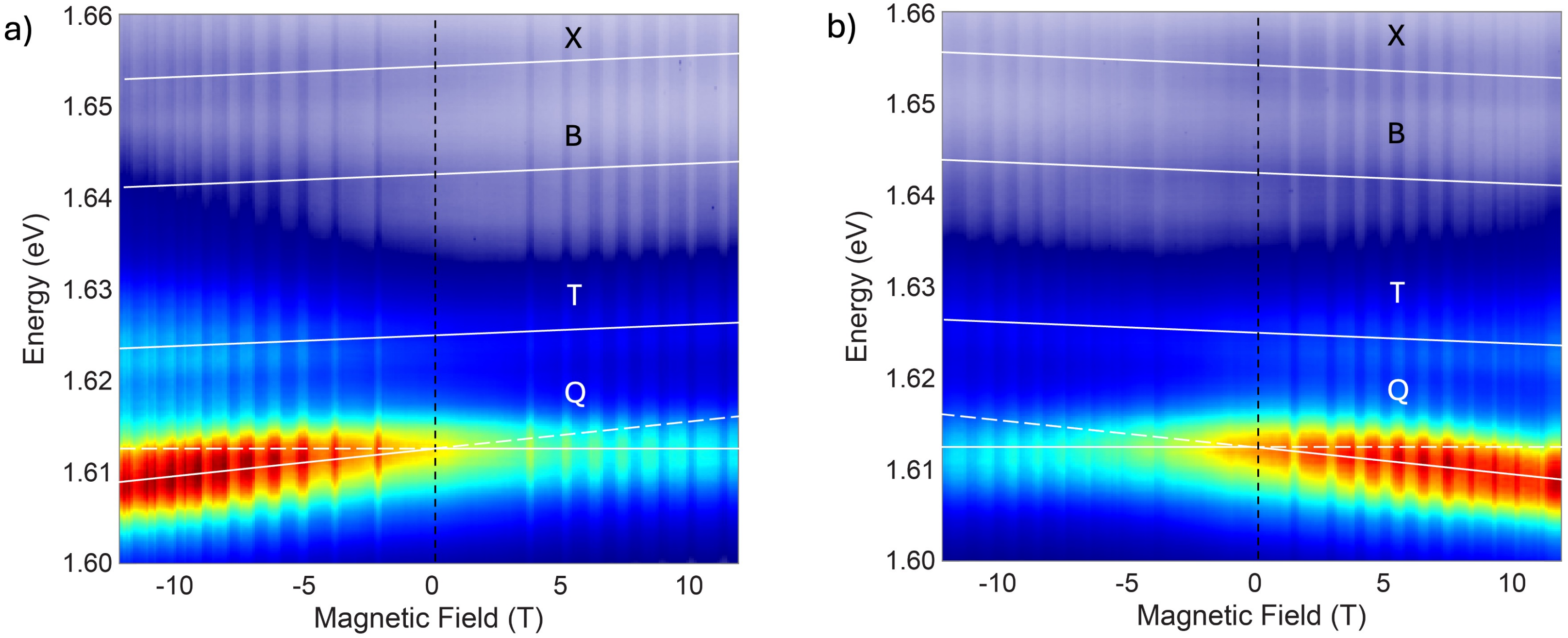




Quaternion grows relative to trion, and trion grows relative to exciton, as background charge increases



Magnetic field measurements show it is a **triplet** state, as expected



A quaternion is a ***charged boson,*** and therefore can be a ***superconductor.***

exciton	integer spin	neutral
trion	half-integer spin	charge $\pm e$
biexciton	integer spin	neutral
charged biexciton	half-integer spin	charge $\pm e$
quaternion	integer spin	charge $\pm 2e$

Open questions:

- Photon couples initial state of charged boson to final state of two free electrons. Should the optical emission be coherent?
- What is T_c for a boson gas of preformed pairs with repulsive Coulomb interaction?
- Can it be a bosonic Wigner crystal?

Summary

- We now have very good fits to the equilibrium N(E) all the way up into the condensate region with N(0) ~ 1000.
- We have clear data for coherent fraction as function of total density, which agrees with equilibrium theory.
- Equilibrium polariton condensation in the weak-coupling limit can be seen at room temperature.
- We see persistent circulation of a steady-state ring condensate with no discernable decay after a picosecond pulse excites it.
- New field: charged bosons!


Qiaochu Wan
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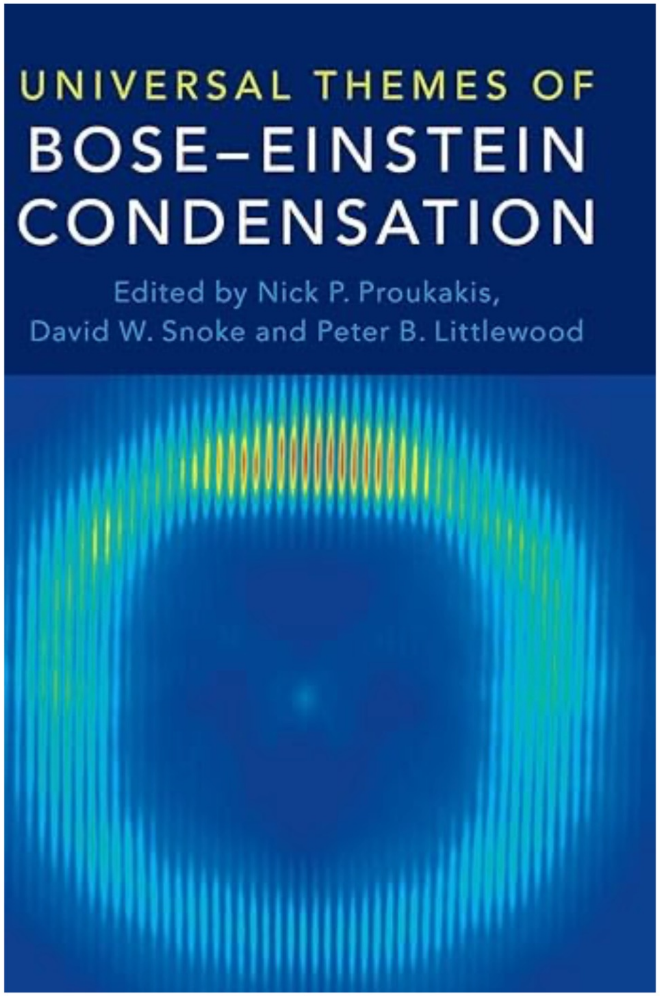
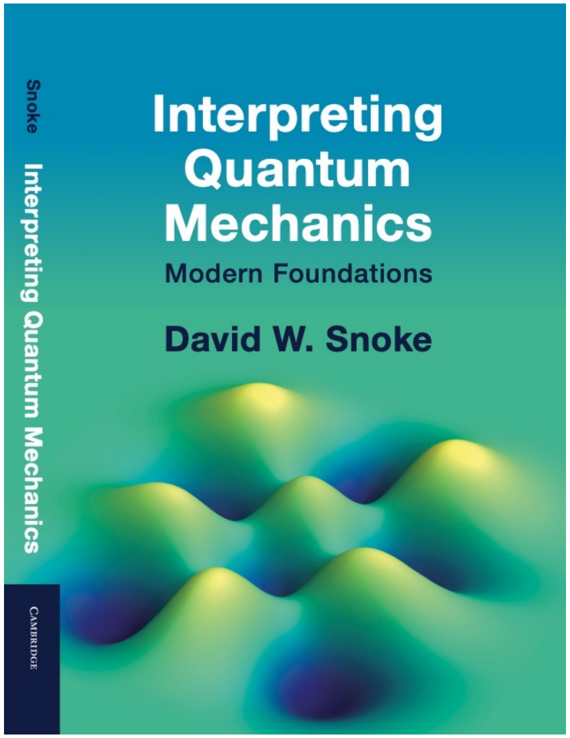


“The blue book”



I. Nonmathematical Exposition of Quantum Mechanics And Quantum Field Theory

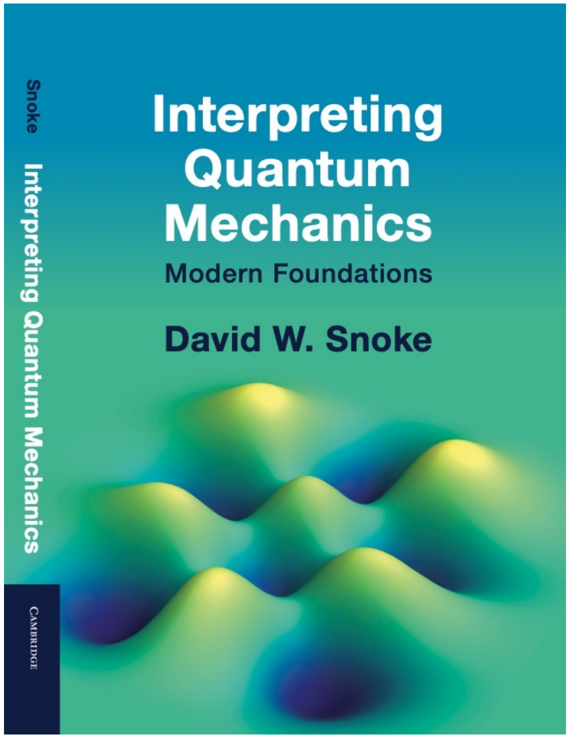
- **How fields generate particles**
- **Nonlocality in particle detection**
- **Alternative interpretations of QM**
- **Decoherence**
- **Quantum mechanics and religion/philosophy**
- **Intro to quantum technology**

II. Basic Math of Quantum Mechanics

- **Schrödinger equation**
- **Basic examples (atoms, solids)**
- **Chaos theory**

III. A Short Course in Quantum Field Theory

- **Photons, Phonons, and Electrons**
- **Qubits and two-level systems**
- **Feynman diagrams**



IV. Mathematical Considerations for Philosophy

- **Bell's theorem**
- **Nonlocality in the many-worlds interpretation**
- **Entanglement**

V. Theory of Coherence and Decoherence

- **Quantum trajectories**
- **Proposed spontaneous collapse model**
- **Superfluids and Superconductors**