An exciton Mott insulator?

Leon Balents, KITP

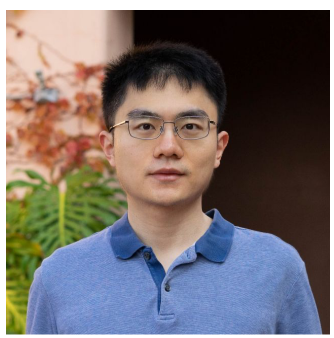
FCMP, October 2024

Collaborators

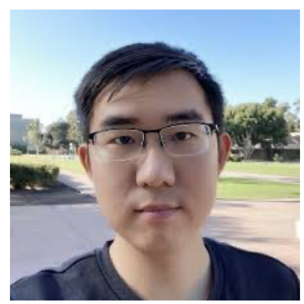
Zhenhao Song
UCSB

Tessa Cookmeyer
KITP

Thanks for the discussions



Chenhao Jin
UCSB Physics



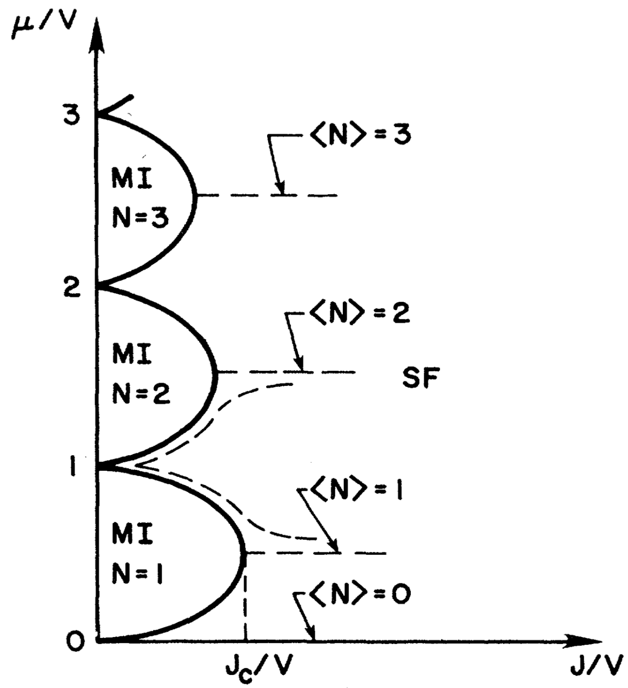
Richen Xiong
UCSB Physics

Bose Mott transition

$$H = -t\sum_{(ij)} b_i^\dagger b_j + U\sum_i n_i(n_i - 1)$$

Mott

$$|\Psi_0\rangle \approx \otimes_i |n_i = m\rangle$$

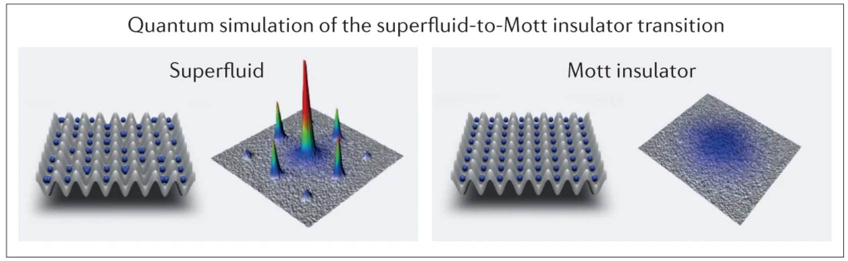


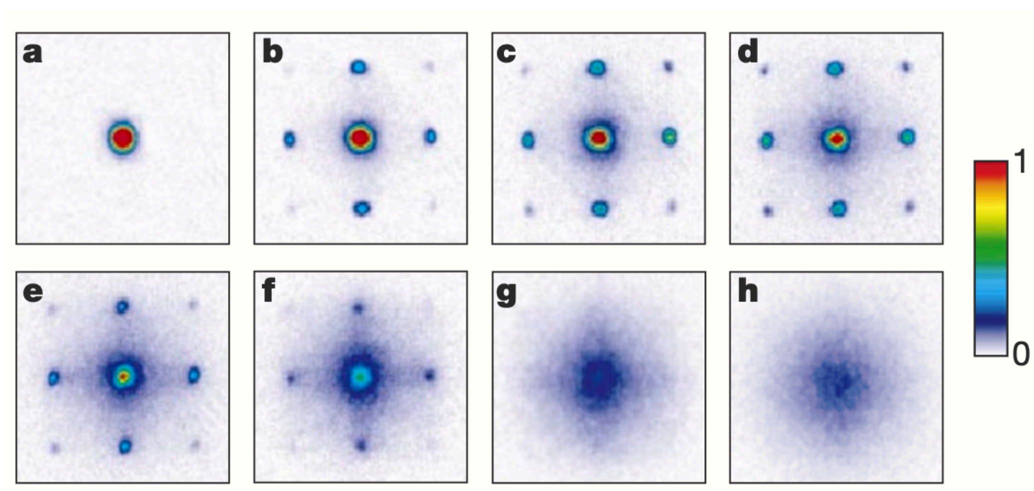
Fisher *et al*, 1989

Superfluid/BEC

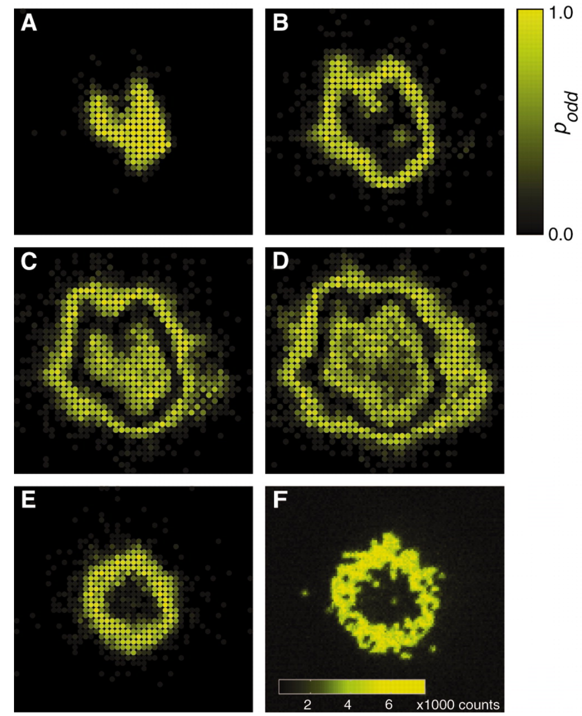
$$|\Psi_0\rangle \approx \left(\sum_i b_i^\dagger\right)^N |0\rangle$$

Bose Mott transition





Greiner *et al*, 2002

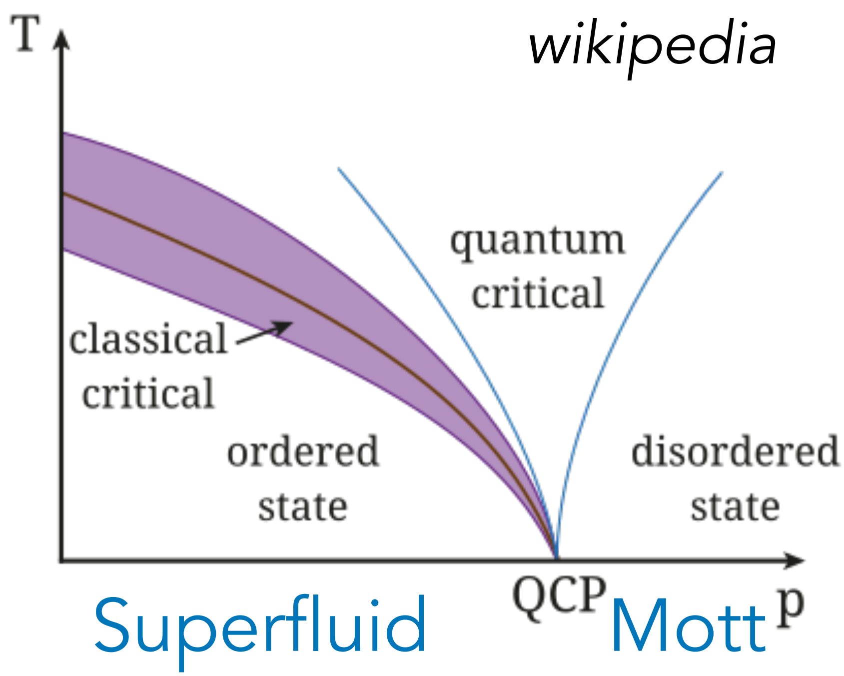


Bakr *et al*, 2010

Quantum Phase Transition

- This is an example of critical phenomena
  - The Mott and superfluid states are fully distinct, and not smoothly connected
  - The transitions are characterized by singular behavior: the density jumps, the correlation length diverges, the superfluid order parameter rises from zero non-analytically.
  - These phenomena are universal and not model dependent.
- The T=0 quantum phase transition involves Mott states, which have no order parameter and exhibit quantization. At T=0 there is no normal fluid, and the non-Mott region is superfluid.
- At T=0 the quantum transition occurs in the ground state: entropy is zero everywhere.

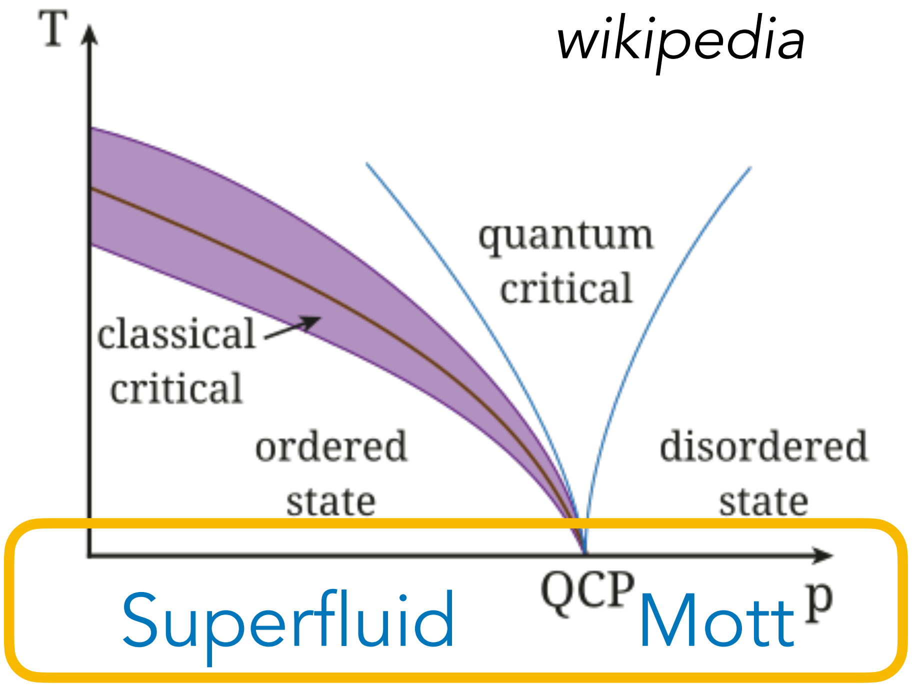
Quantum phase transition



Scaling
Criticality
Singularities
Universality
All the good stuff

Quantum phase transition

wikipedia



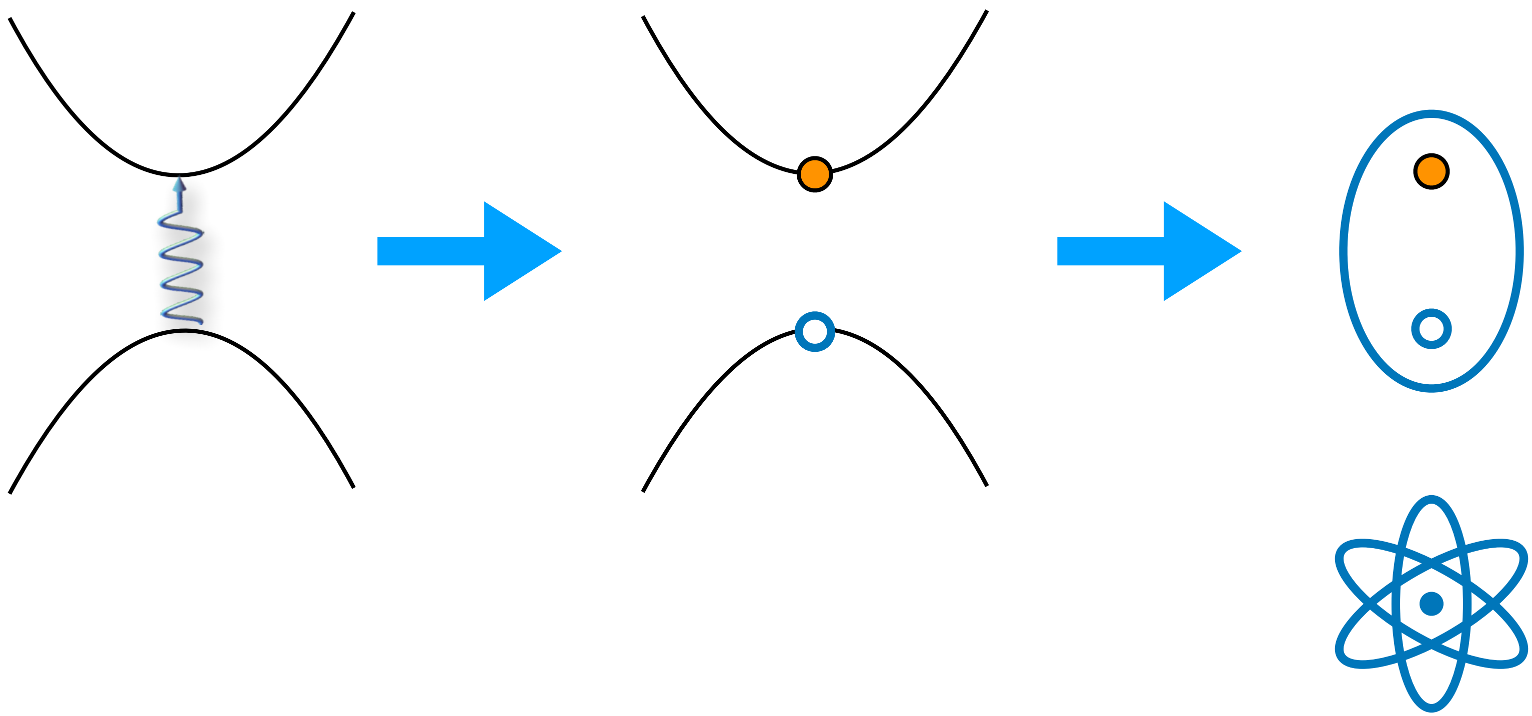
Scaling
Criticality
Singularities
Universality
All the good stuff

@T=0: no entropy

No normal fluid

Excitons

A “composite” boson in electronic systems

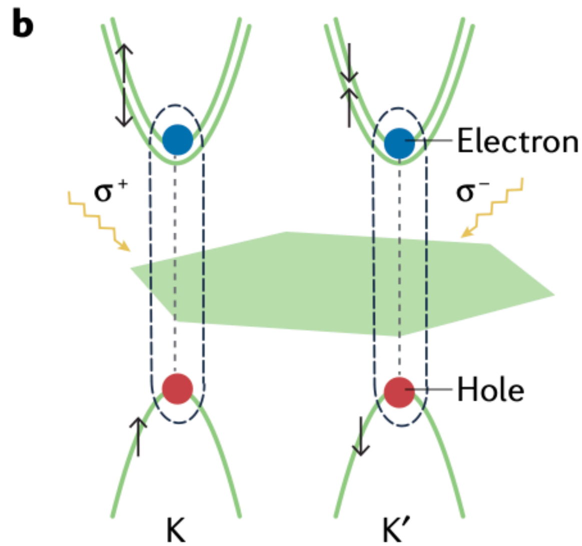


Unlike an atom an exciton is not stable and is only an excitation.

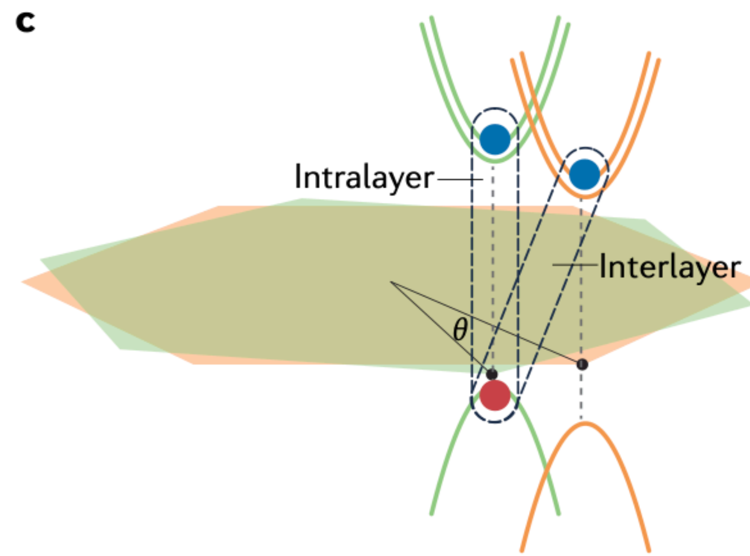
TMD excitons

Electrons have valley and spin QNs

Hole have valley QNs



E. Regan *et al*, 2022



For certain “type II” band alignments, **inter**-layer excitons have lowest energy

These have long lifetimes and are more sensitive to layer alignment/moiré

Interlayer moiré excitons

WS_2-WSe_2

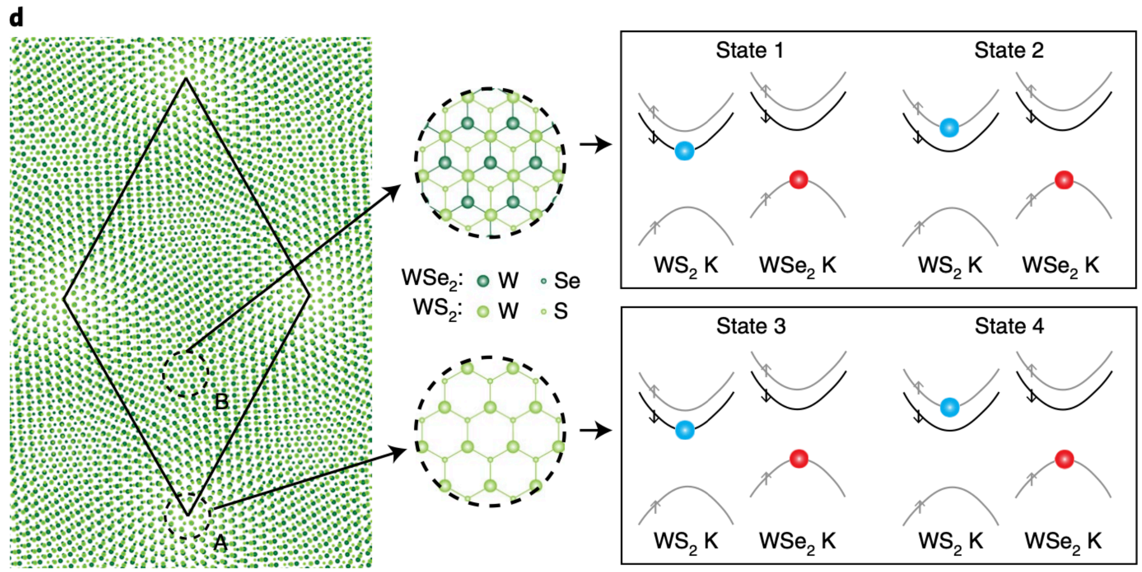
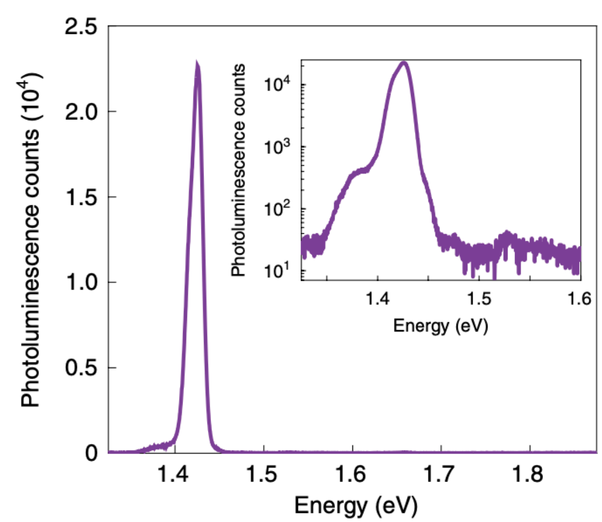


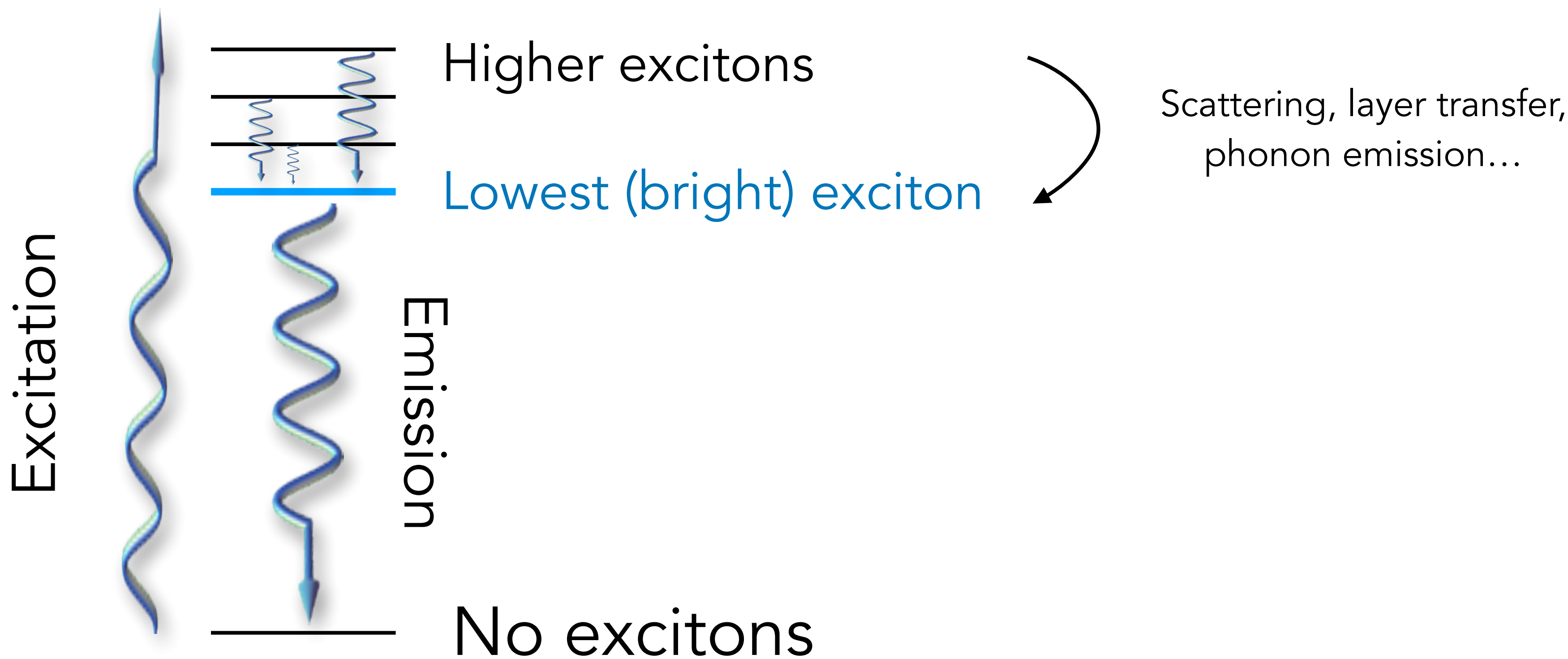
Table 1 | The nature of different interlayer moiré exciton states and the spin, valley and moiré contributions to their optical selection rules

	Experimental results				Inferred conclusions				
State	Energy (eV)	Oscillator strength	Total QAM	Hole valley and spin	Electron valley and spin	Spin QAM	Valley QAM	Moiré QAM	Moiré position
1	1.43	Weak	+1=−2	K↑	K↓	−1	+1	−2	B
2	1.46	Strong	−1	K↑	K↑	0	+1	−2	B
3		Not observed	0	K↑	K↓	−1	+1	0	A
4	1.51	Strong	+1	K↑	K↑	0	+1	0	A
5	1.43	Weak	−1=+2	K′↓	K′↑	1	−1	+2	B
6	1.46	Strong	+1	K′↓	K′↓	0	−1	+2	B
7		Not observed	0	K′↓	K′↑	1	−1	0	A
8	1.51	Strong	−1	K′↓	K′↓	0	−1	0	A



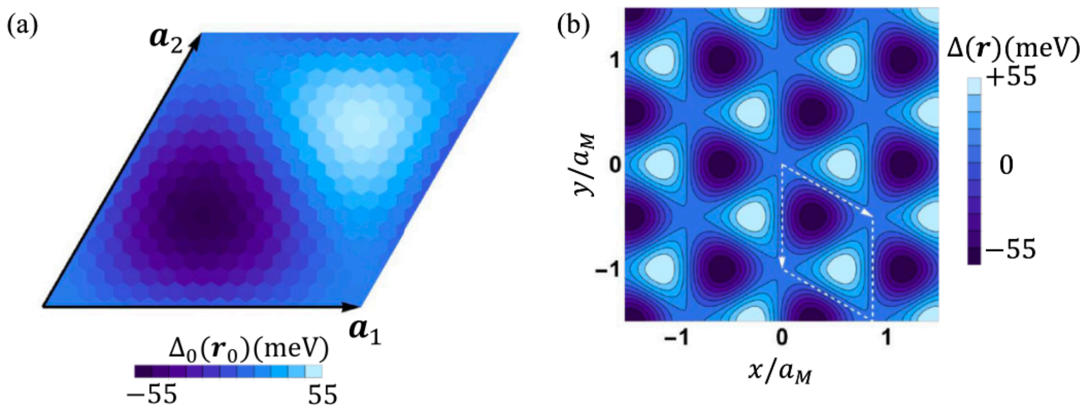
C. Jin *et al*, 2019

Emission



NOT to scale!

Moiré potential

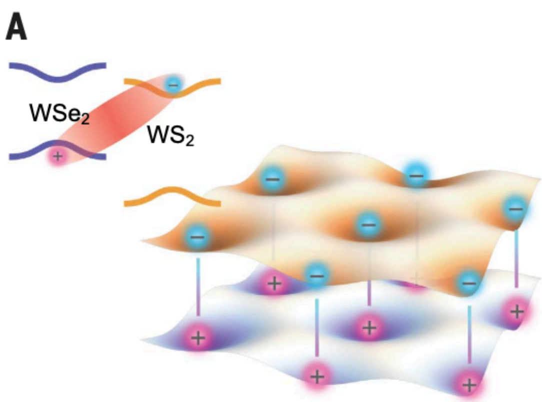


F. Wu *et al*, 2018 (plot is for WS_2-MoS_2)

Excitons localize in minima of local band gap

Experimentalists can populate excitons in these wells.

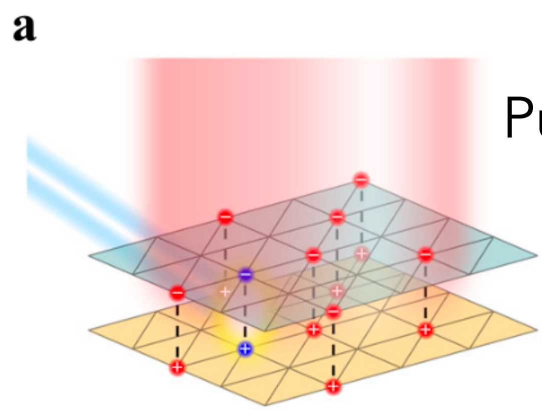
Exciton Mott insulator



Probe

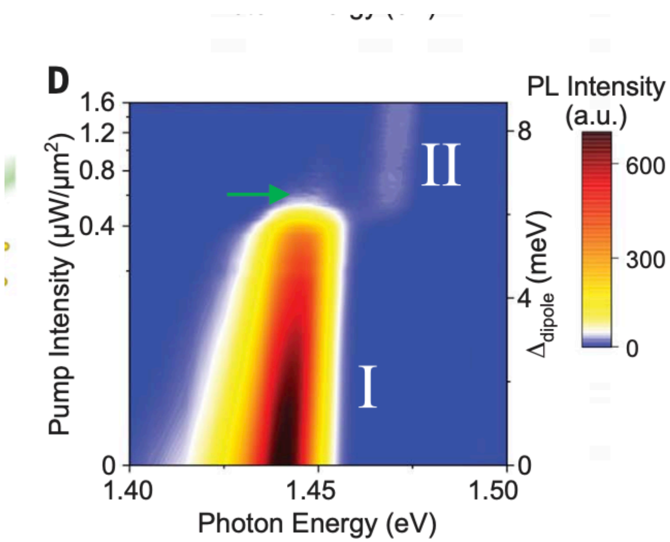
$\Delta I \cos \omega t$

(8 Hz)



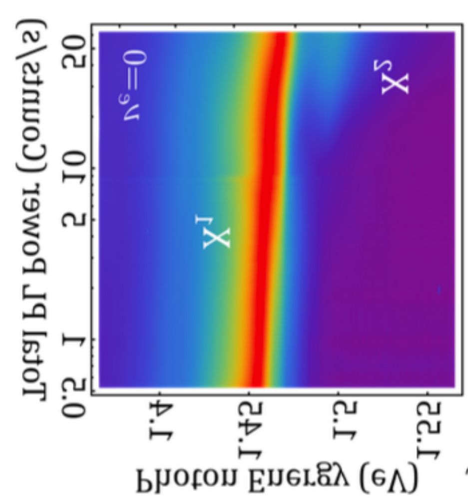
Pump intensity I_0

I_0



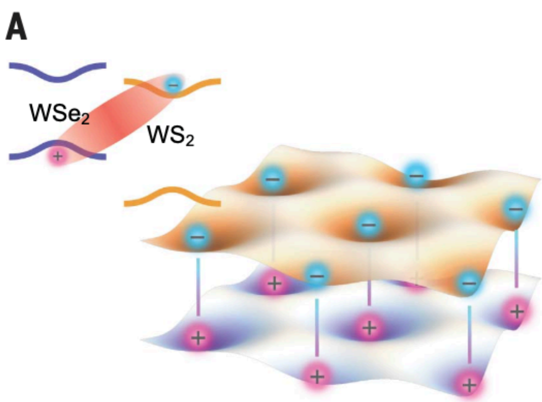
$$\frac{\Delta I_{emit}}{\Delta I}$$

R. Xiong *et al*, 2023



B. Gao *et al*, 2023

Exciton Mott insulator



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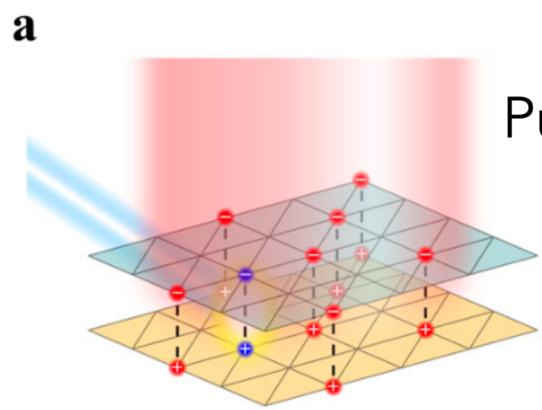

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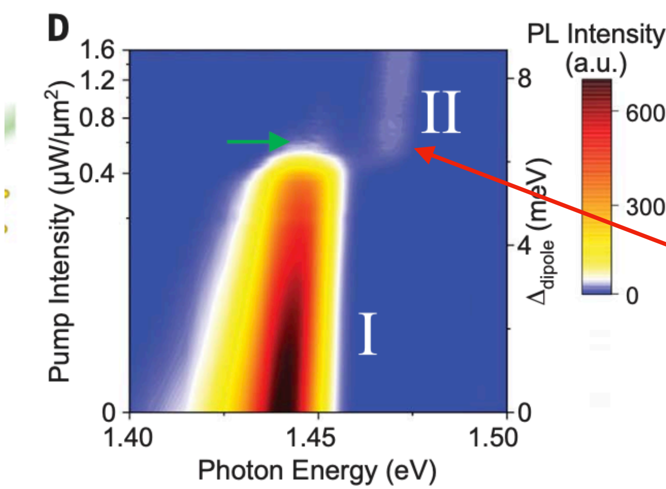
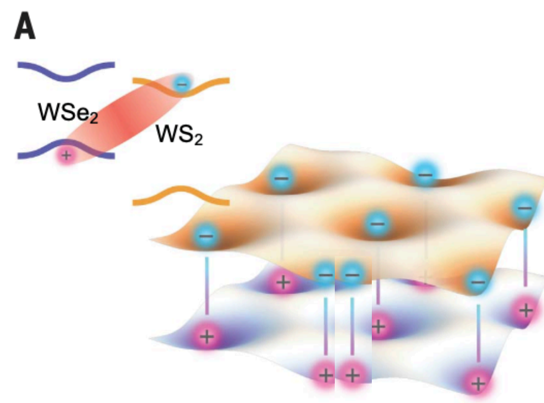


$$\frac{\Delta I_{emit}}{\Delta I}$$

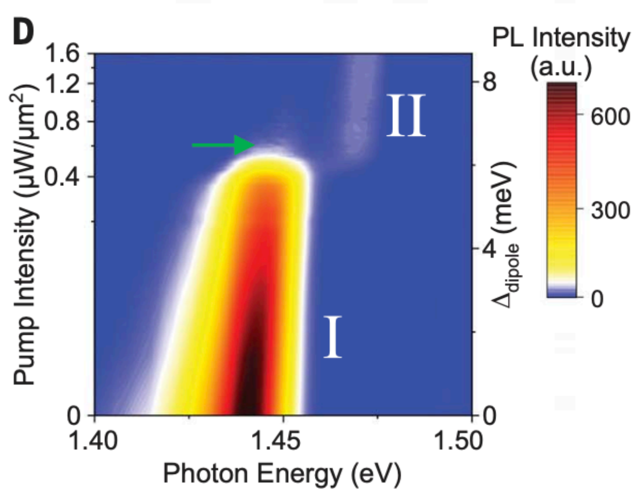


2nd exciton's energy increased by $U_{ex\text{-}ex}$

R. Xiong *et al*, 2023

Issues

- Excitons are far from equilibrium
  - under constant illumination
  - not conserved
- Is there a Mott transition under these conditions? What is its nature?



Jump in emission line *seems* abrupt but…

Simple model

- Minimalist excitons: ignore spin, valley

$$H_0 = \mu \sum_i n_i + \sum_i \frac{U}{2} n_i(n_i - 1) - \sum_{i,j} t_{i,j} b_i^\dagger b_j,$$

- Take non-equilibrium seriously: *Lindblad equation*

$$\dot{\rho} = -i[H, \rho] + \sum \gamma_k \mathcal{L}(L_k, L_k^\dagger)[\rho]$$

$$\mathcal{L}(A, B)[\rho(t)] = A\rho(t)B - \tfrac{1}{2}\{BA, \rho(t)\}$$

"Jump operators" encode transitions generated by excitation and decay

Jump operators

- Microscopically, these arise from coupling to photons (+…)

$$\mathcal{B}$$

$$H = H_0 + H_I; \qquad H_I = \mathcal{R}\left(\sum_i b_i^\dagger\right) + \mathcal{R}^\dagger\left(\sum_i b_i\right)$$

Electromagnetic field has long wavelength >> moiré period
For simplicity we use just this bath coupling.

- Standard derivation (H.P. Breuer and F. Petruccione, 2002) gives that jump operators determined by
  - Spectral correlator of R
  - Decomposition of B into eigen-operators of H_0

$$[\mathcal{B}_\lambda, H_0] = \omega_\lambda \mathcal{B}_\lambda$$

$$\mathcal{B} = \sum_\lambda \mathcal{B}_\lambda$$

Lindblad construction

- Eigen-operators

$$[\mathcal{B}_\lambda, H_0] = \omega_\lambda \mathcal{B}_\lambda$$

In general finding these is hard!
Similar to diagonalizing H_0

Bath generates transitions that conserve total energy of bath and system.

- Lindbladian in interaction picture:

$$\dot{\rho}(t) = \sum_{\lambda,\lambda':\omega_\lambda=\omega'_\lambda} \gamma(\omega_\lambda)\mathcal{L}\left(B_\lambda(\omega_\lambda), B^\dagger_{\lambda'}(\omega_{\lambda'})\right)[\rho(t)] + \sum_{\lambda,\lambda':\omega_\lambda=\omega'_\lambda} I(\omega_\lambda)\mathcal{L}\left(B_\lambda(\omega_\lambda)^\dagger, B_{\lambda'}(\omega_{\lambda'})\right)[\rho(t)]$$

$$\gamma(\omega) = \int_{-\infty}^{\infty} ds e^{i\omega s}\langle \mathcal{R}(s)\mathcal{R}^\dagger\rangle$$

$$I(\omega) = \int_{-\infty}^{\infty} ds e^{-i\omega s}\langle \mathcal{R}(s)^\dagger\mathcal{R}\rangle$$

Eigen-operators

- Finding these is hard! $[\mathcal{B}_\lambda, H_0] = \omega_\lambda \mathcal{B}_\lambda$
- A prior we cannot restrict to low energy
- What we do: *all-all hopping model*

$$H_0 = \mu \sum_i n_i + \frac{U}{2} \sum_i n_i(n_i - 1) - \frac{t}{N} \sum_{i,j} b_i^\dagger b_j$$

In equilibrium, this affords a mean-field approximation as N → infinity

But we can work for any N using *permutation symmetry*

$$b_i \to b_{P(i)}$$

Eigen-operators

- We can restrict to states with full permutation symmetry

Basis: $$|\vec{n}\rangle = \mathcal{N}_{\vec{n}} \sum_{\sigma \in S_N} \sigma |(n_0, n_1, ..., n_M)\rangle$$ n_0 empty sites, n_1 singly occupied sites, n_2 doubly occupied sites…

There are relatively few states. Can diagonalize H_0 in this basis.

Result: $$H_0|\alpha, N_B\rangle = E_{\alpha,N_B}|\alpha, N_B\rangle$$ Numerically except special cases

➡ Eigenoperators:

$$B_{\alpha,\beta,N_B} = \langle\alpha, N_B - 1|\mathcal{B}|\beta, N_B\rangle|\alpha, N_B - 1\rangle\langle\beta, N_B|$$

$$\omega_{\alpha,\beta,N_B} = E_{\beta,N_B} - E_{\alpha,N_B-1}$$

Lindblad

- Using these states, we find the density matrix takes the form

$$\rho = \sum_{n,\alpha} \rho_{n,\alpha} |\alpha, n\rangle\langle\alpha, n|$$

Diagonal in this basis.

- Then the Lindblad equation becomes

$$\dot{\rho}_{an} = \sum_{\lambda=(\alpha,a,n)} |B_\lambda|^2 \left(I(\omega_\lambda)\rho_{\alpha,n-1} - \gamma(\omega_\lambda)\rho_{an}\right) + \sum_{\lambda=(a,\beta,n+1)} |B_\lambda|^2 \left(\gamma(\omega_\lambda)\rho_{\beta,n+1} - I(\omega_\lambda)\rho_{an}\right)$$

This is just a fancy rate equation for a mixed state.

But we can evaluate it all using just μ, U, t and the functions $\gamma(\omega), I(\omega)$

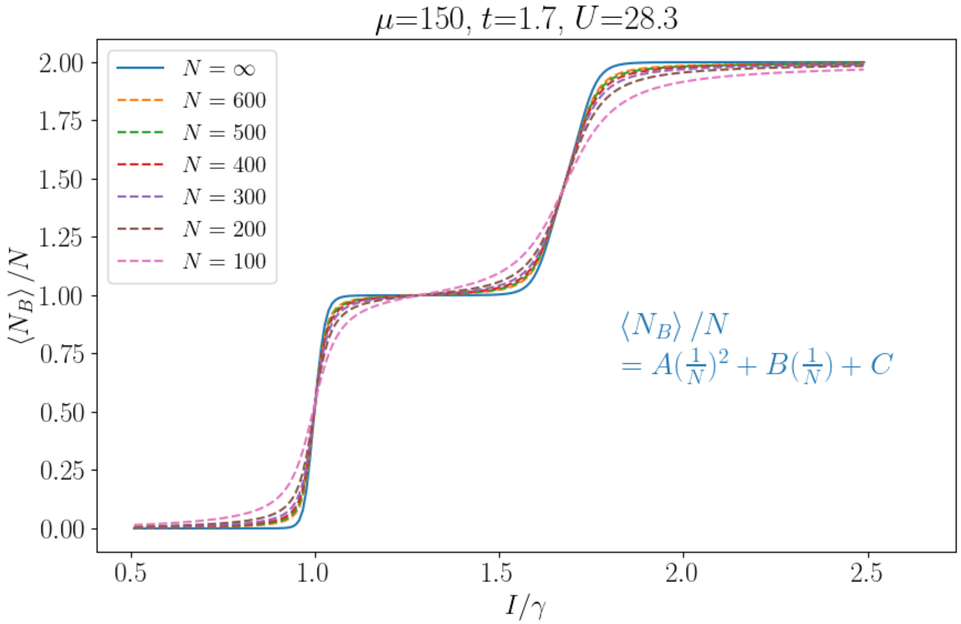
n.b. we assume no coherent pumping (typical for inter-layer excitons)

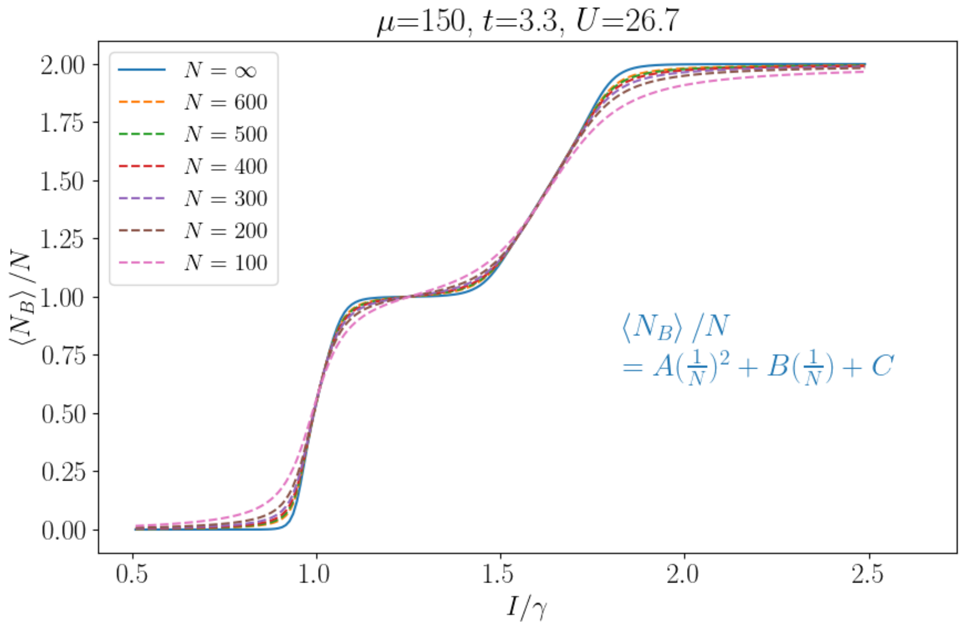
Results

- We studied several cases:
  - Zero hopping: exactly soluble
  - Hard-core bosons: exactly soluble
  - n_i=0,1,2 (n_{max}=2) numerical solution
- In all cases, the solution is a non-thermal mixed state. There are, however, clearly phases.

Density

$\langle n_i \rangle$ should be quantized to an integer in a Mott insulator





n_{max}=2

Density

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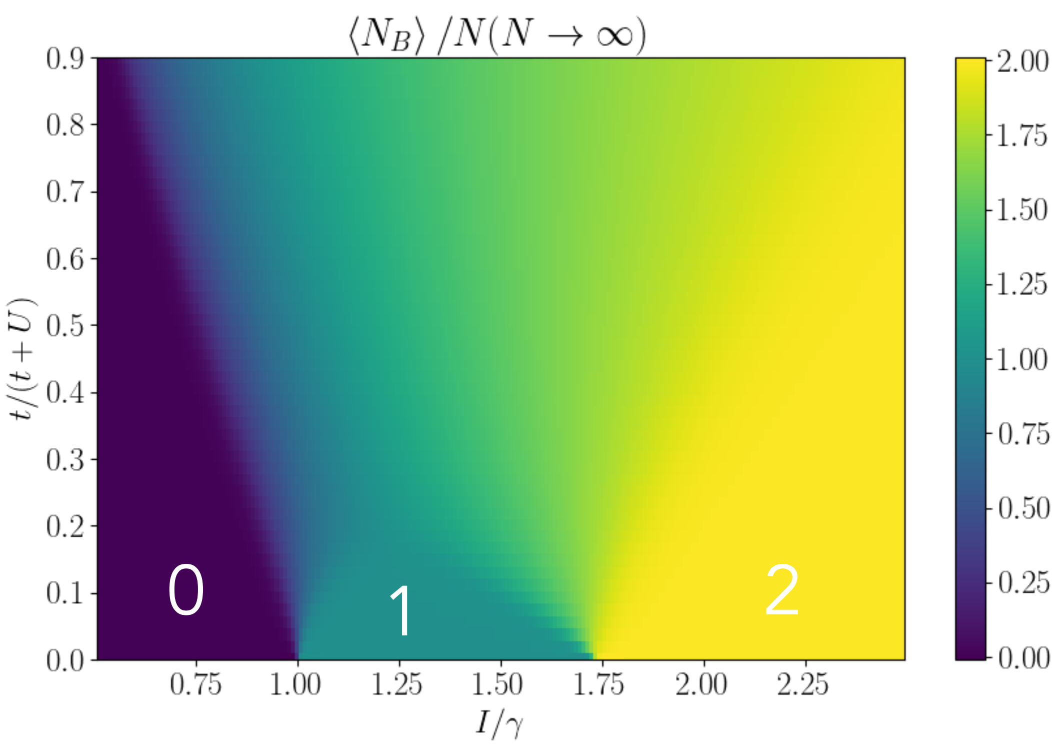
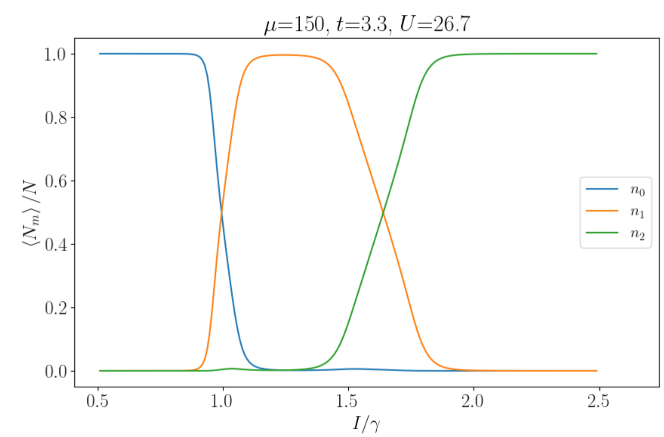
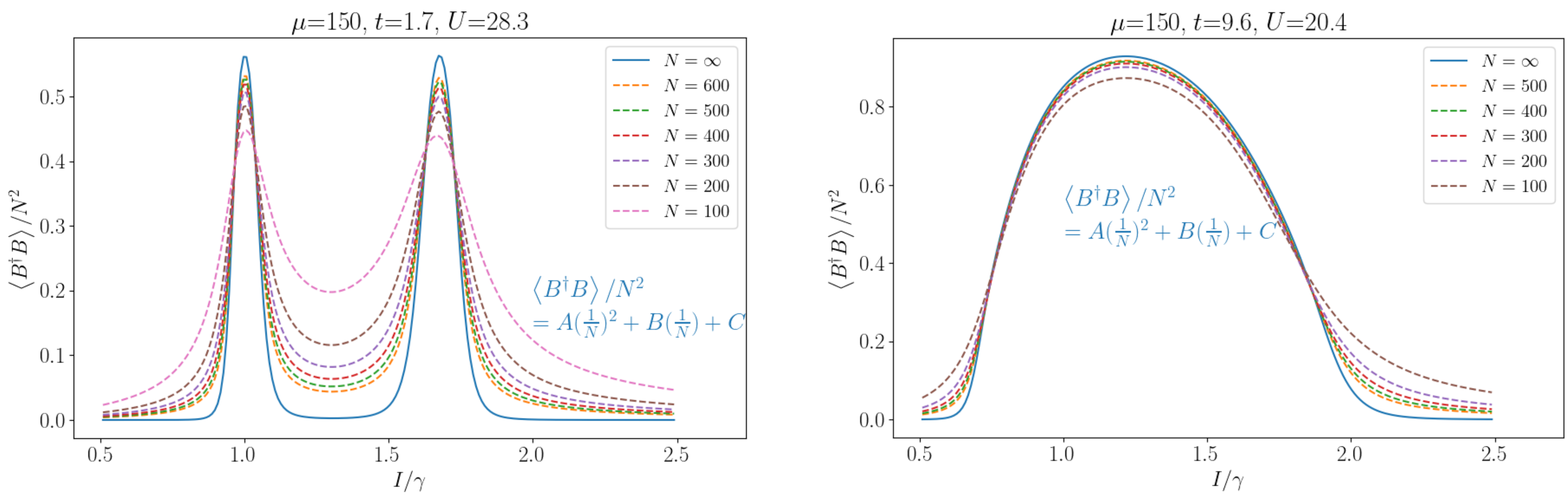




Condensation

We examine

$$C = \frac{1}{N^2} \sum_{ij} \langle b_i^\dagger b_j \rangle = \frac{1}{N^2} \langle \mathcal{B}^\dagger \mathcal{B} \rangle$$



Weak hopping

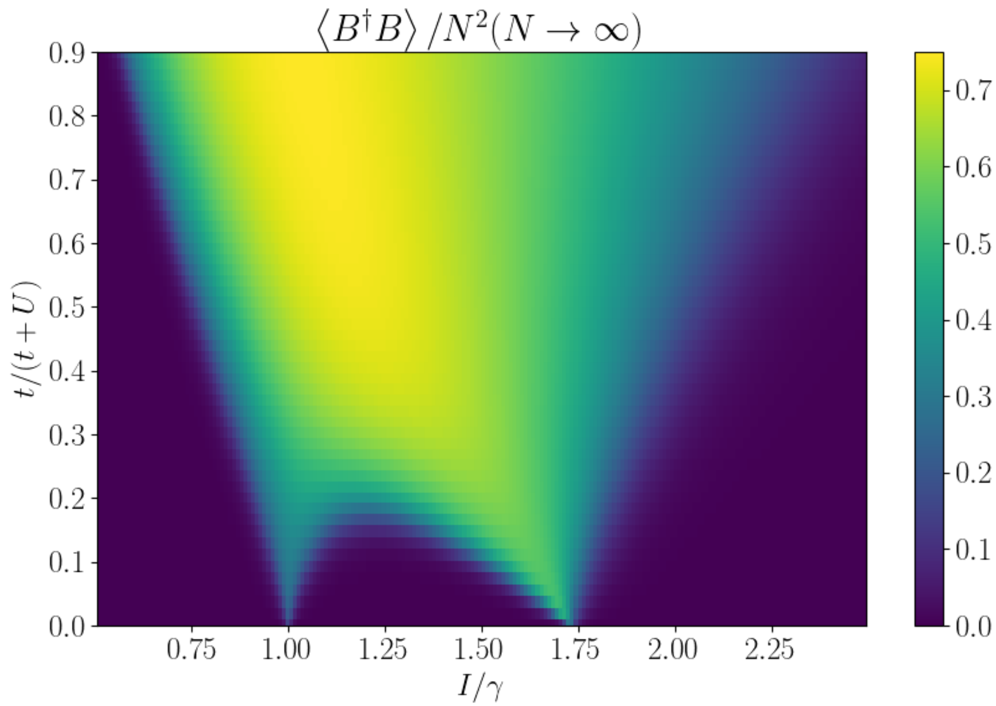
Strong hopping

Condensation

We examine

$$C = \frac{1}{N^2}\sum_{ij}\langle b_i^\dagger b_j\rangle = \frac{1}{N^2}\langle \mathcal{B}^\dagger \mathcal{B}\rangle$$

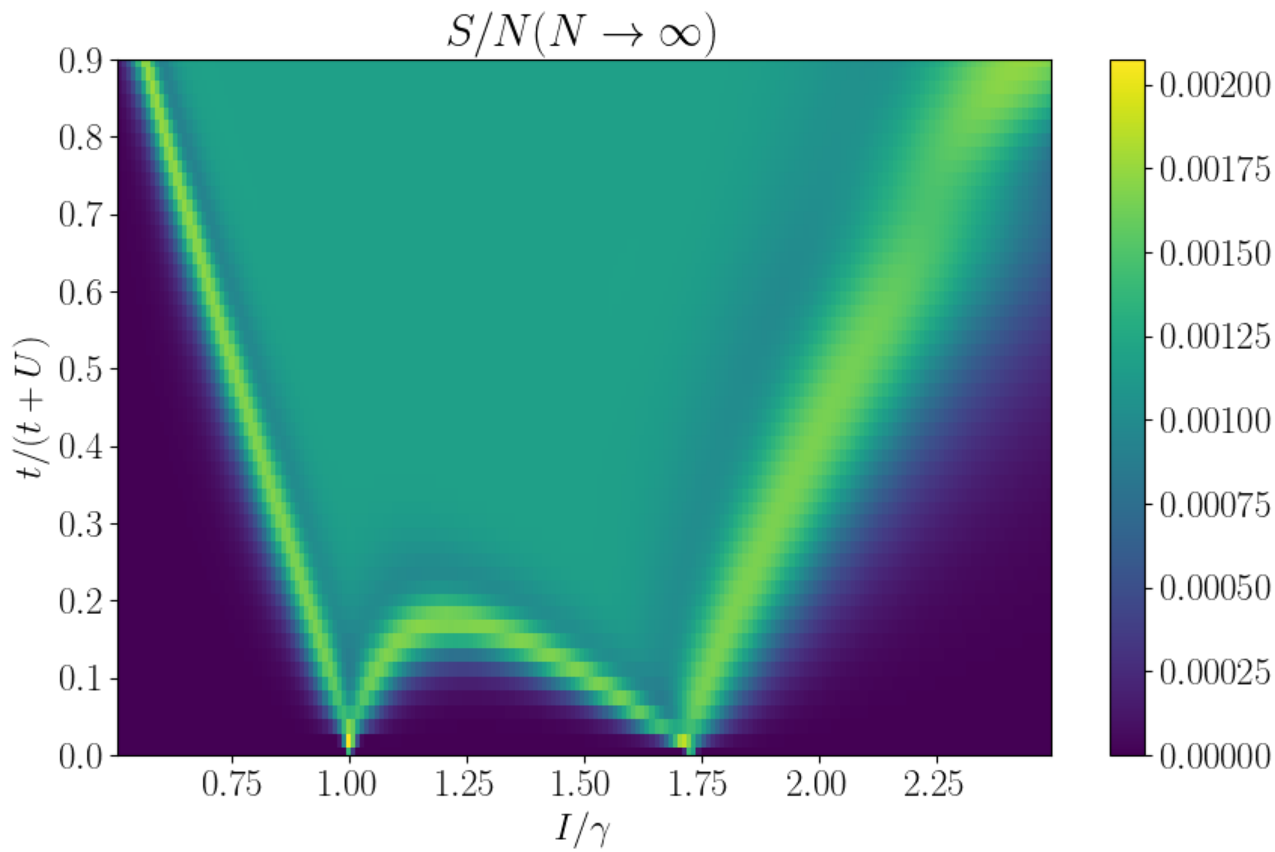
n_{max}=2



Clearly 3 “Mott” lobes are visible

Entropy

The entropy per particle is a sensitive estimator of the transitions



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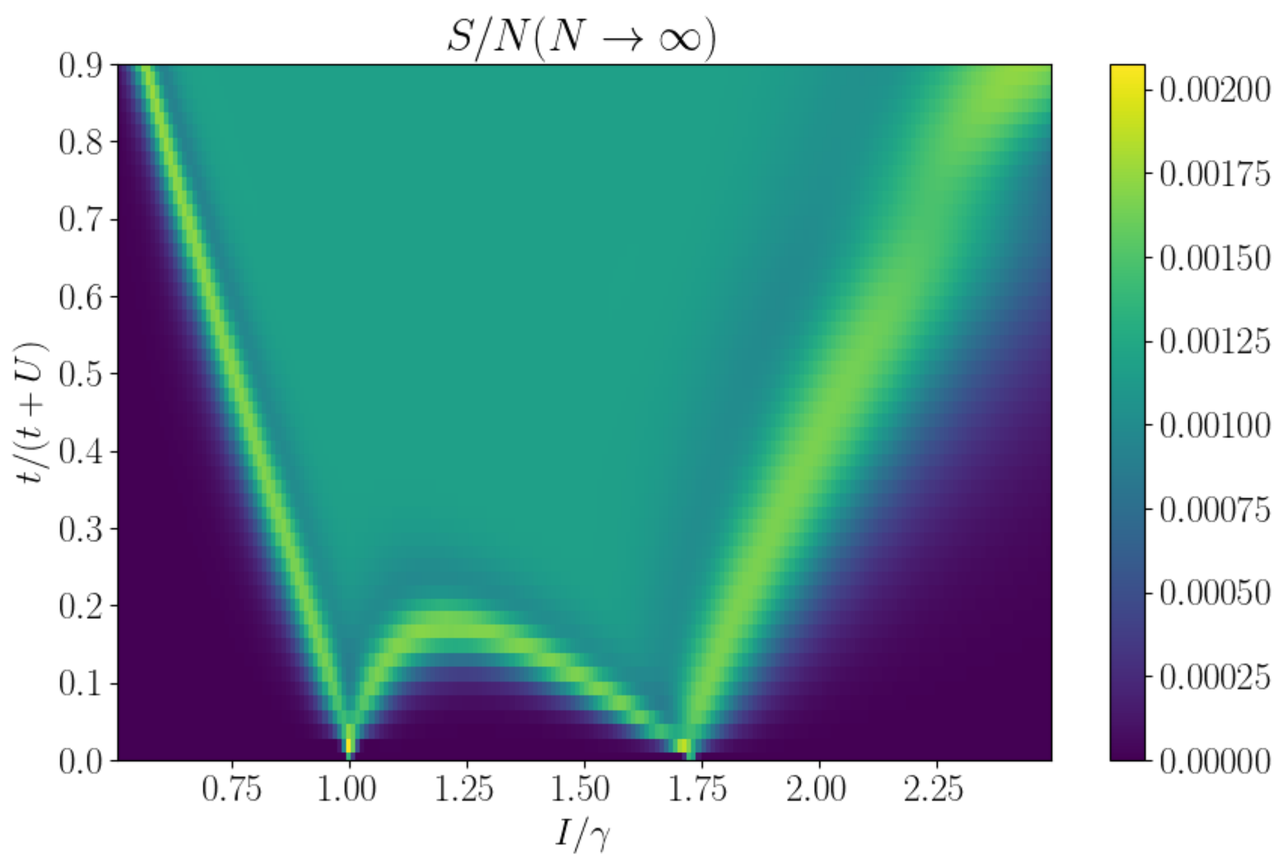
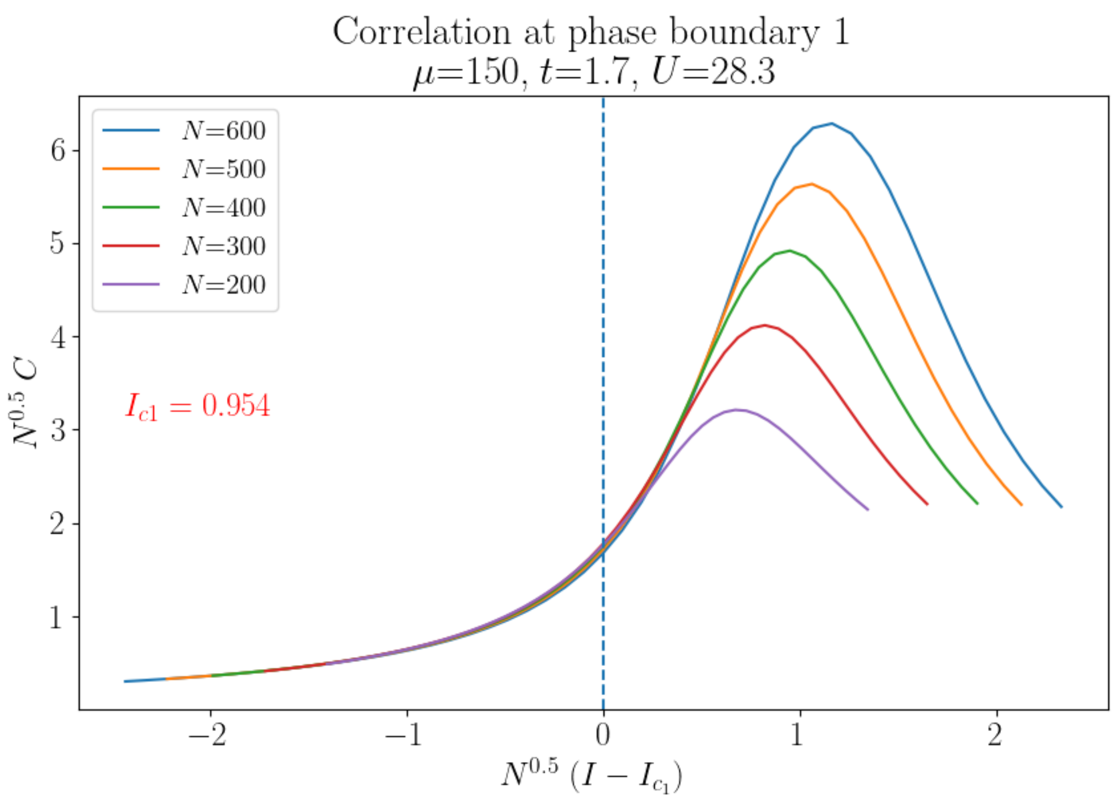


Non-zero entropy clarifies this is *not* the usual Bose-Mott transition

Critical properties

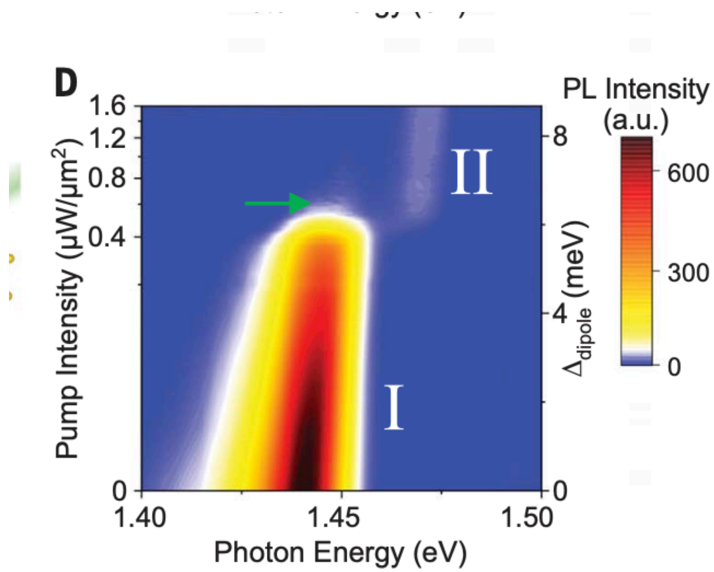


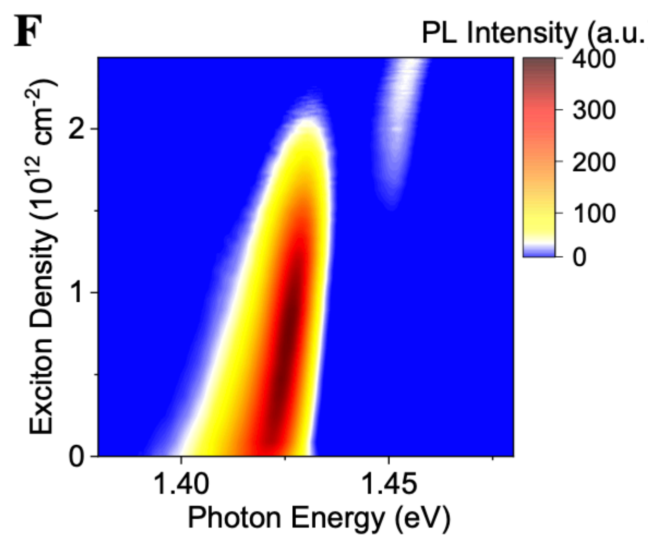
Mean-like exponents appear in finite-size scaling.

Summary

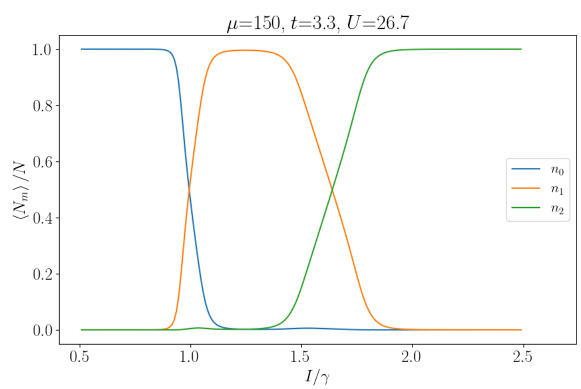
- The Lindbladian Bose-Hubbard model shows behavior very reminiscent of the equilibrium analog
  - Mott phases appear to be stable
  - Superfluidity also appears in the sense of ODLRO
  - However these states are not described by equilibrium density matrices
    - e.g. the “superfluid” has higher entropy rather than lower entropy.

Back to experiments





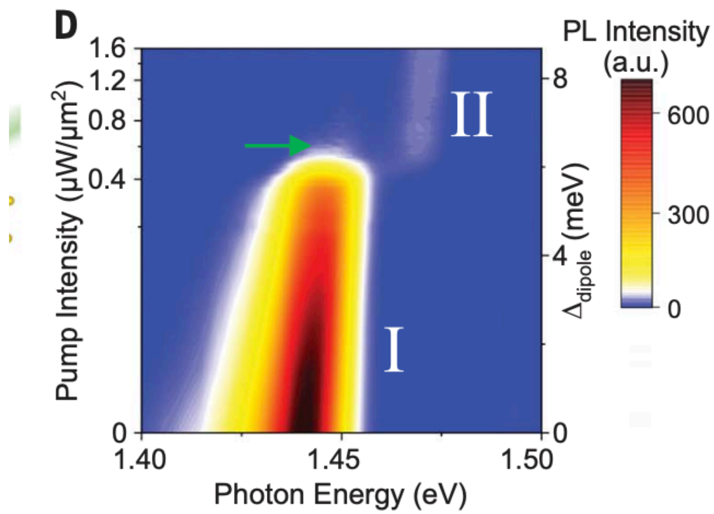
Compare this with

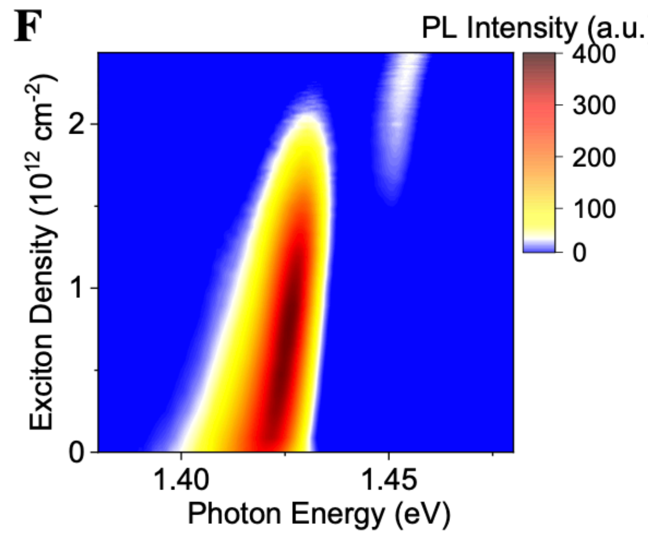


?

Naively $PL_I \sim n_1$, $PL_{II} \sim n_2$

Back to experiments





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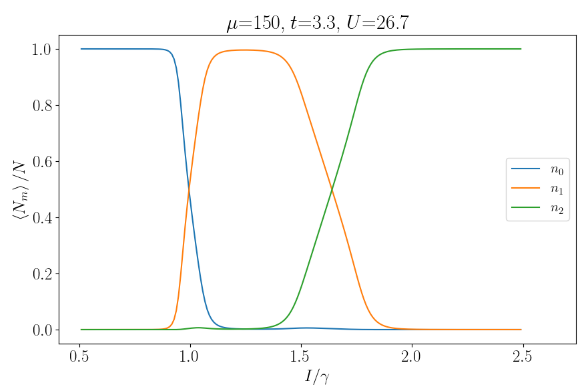


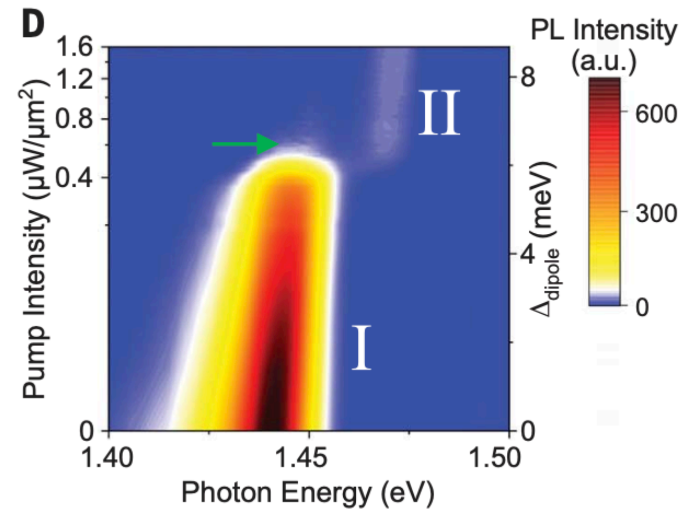
?

Naively $PL_I \sim n_1$, $PL_{II} \sim n_2$

n.b. Pump intensity is probably a different variable than our I_0, which is the exciton creation rate in our effective model. Can perhaps compare better versus density.

Back to experiments

What does this theory really show us for these experiments?



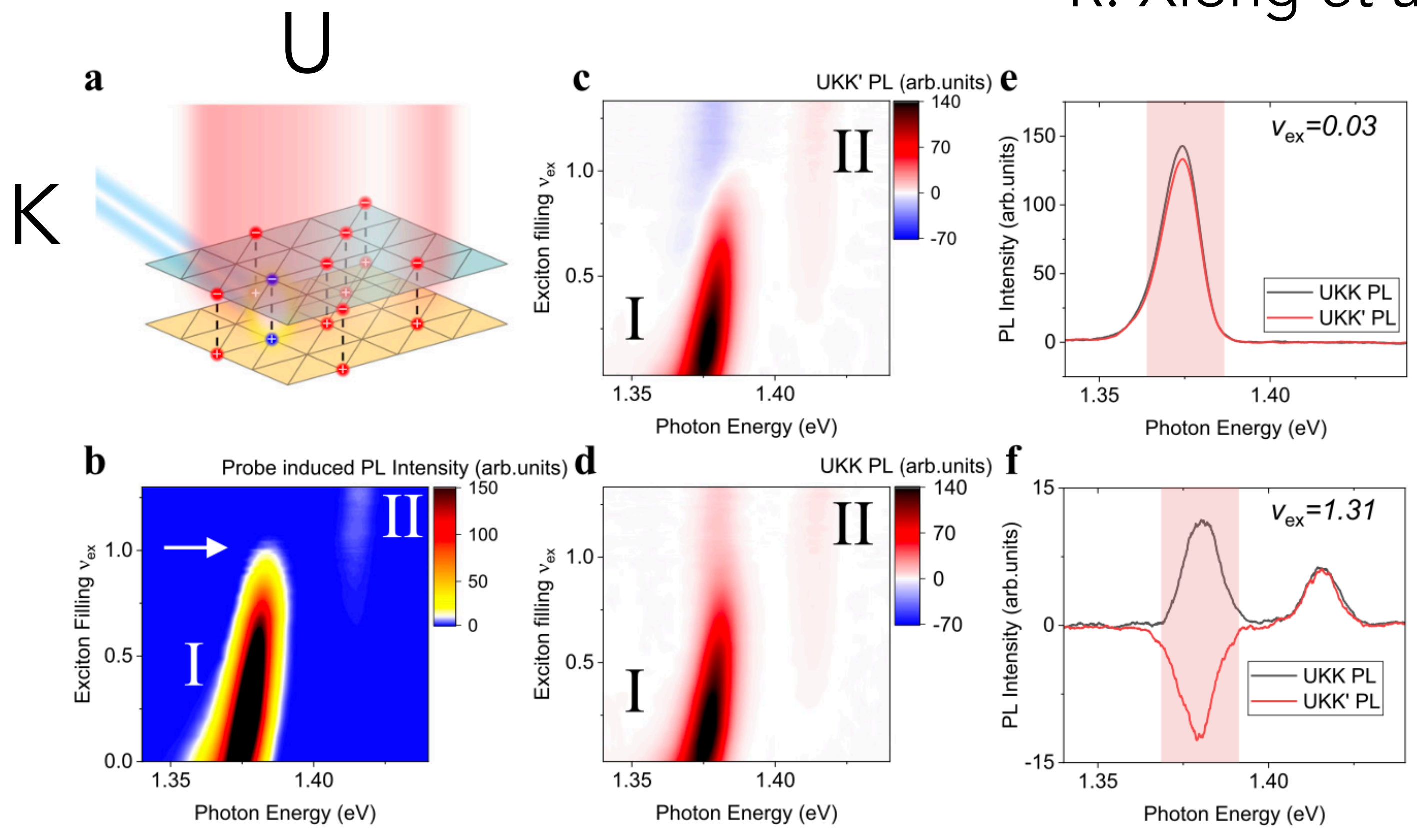
- Establishes that stable Bose Mott state can exist under illumination
- Any more detailed connection needs either more experiments or more developed theory
- To be honest, these experiments give no direct evidence for any exciton hopping $t\, b_i^\dagger b_j$ whatsoever

Interesting issues

- Roles of local versus global dissipation, tunneling
- Gating/doping with free electrons/holes.
- Valley degree of freedom of excitons

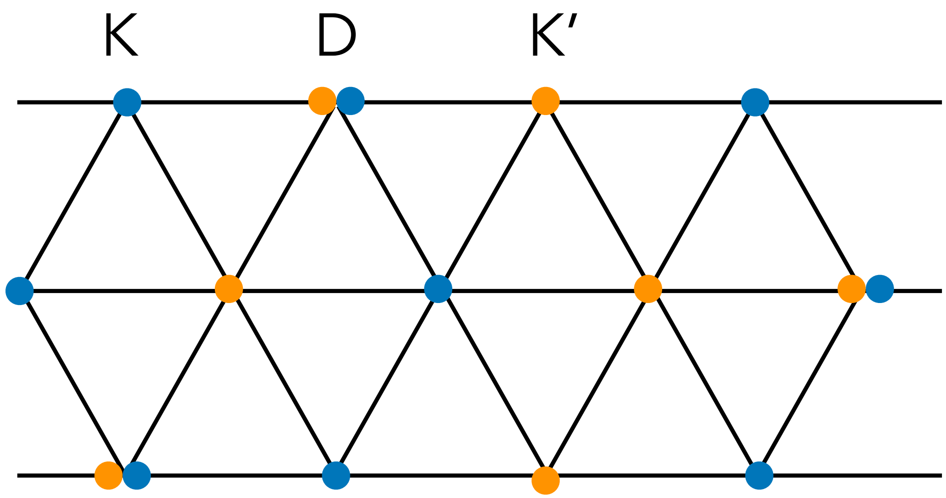
Valley physics

R. Xiong *et al*, 2024.



Polarized experiments detect response that is very sensitive to exciton density.

Picture for n>1



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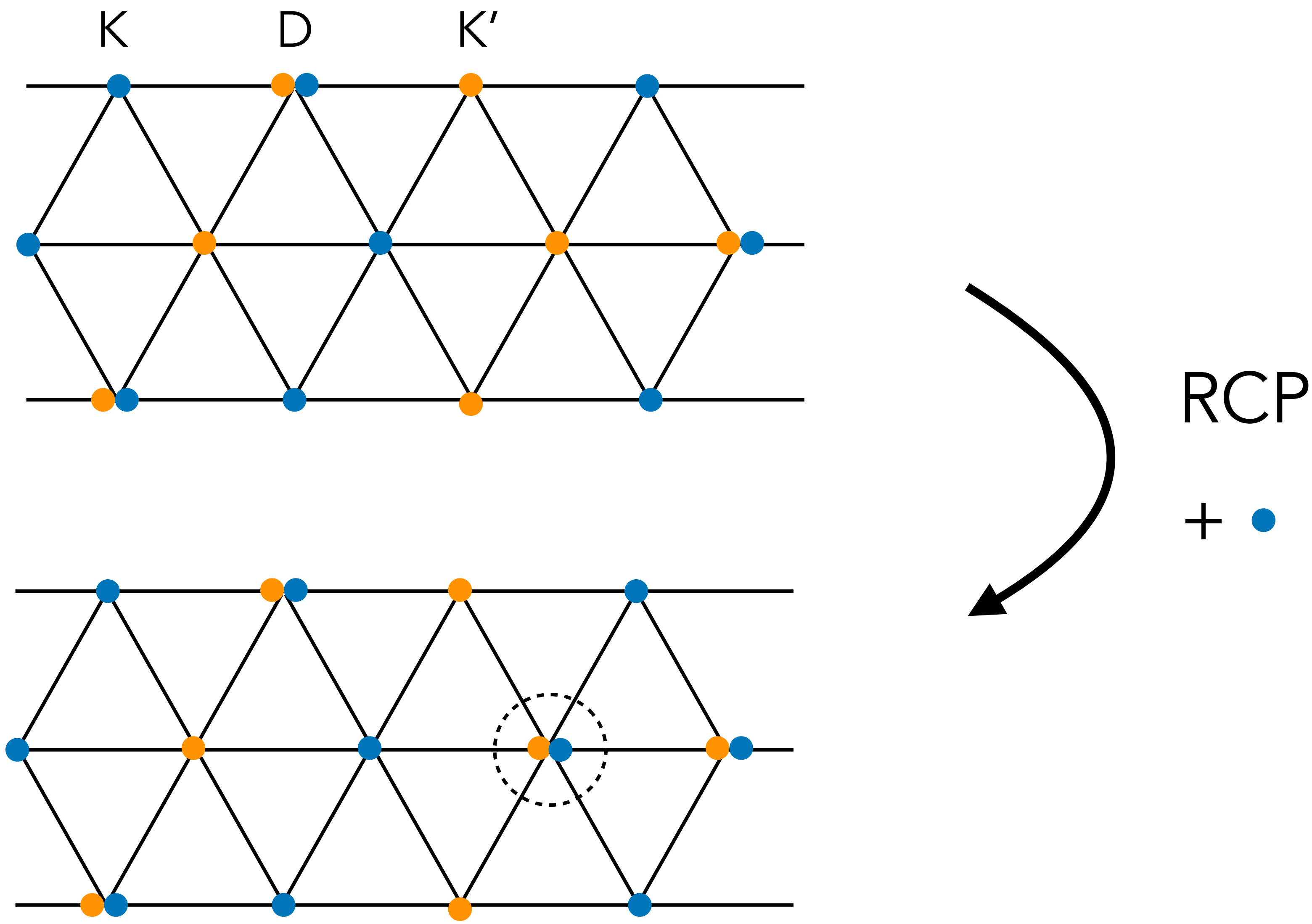

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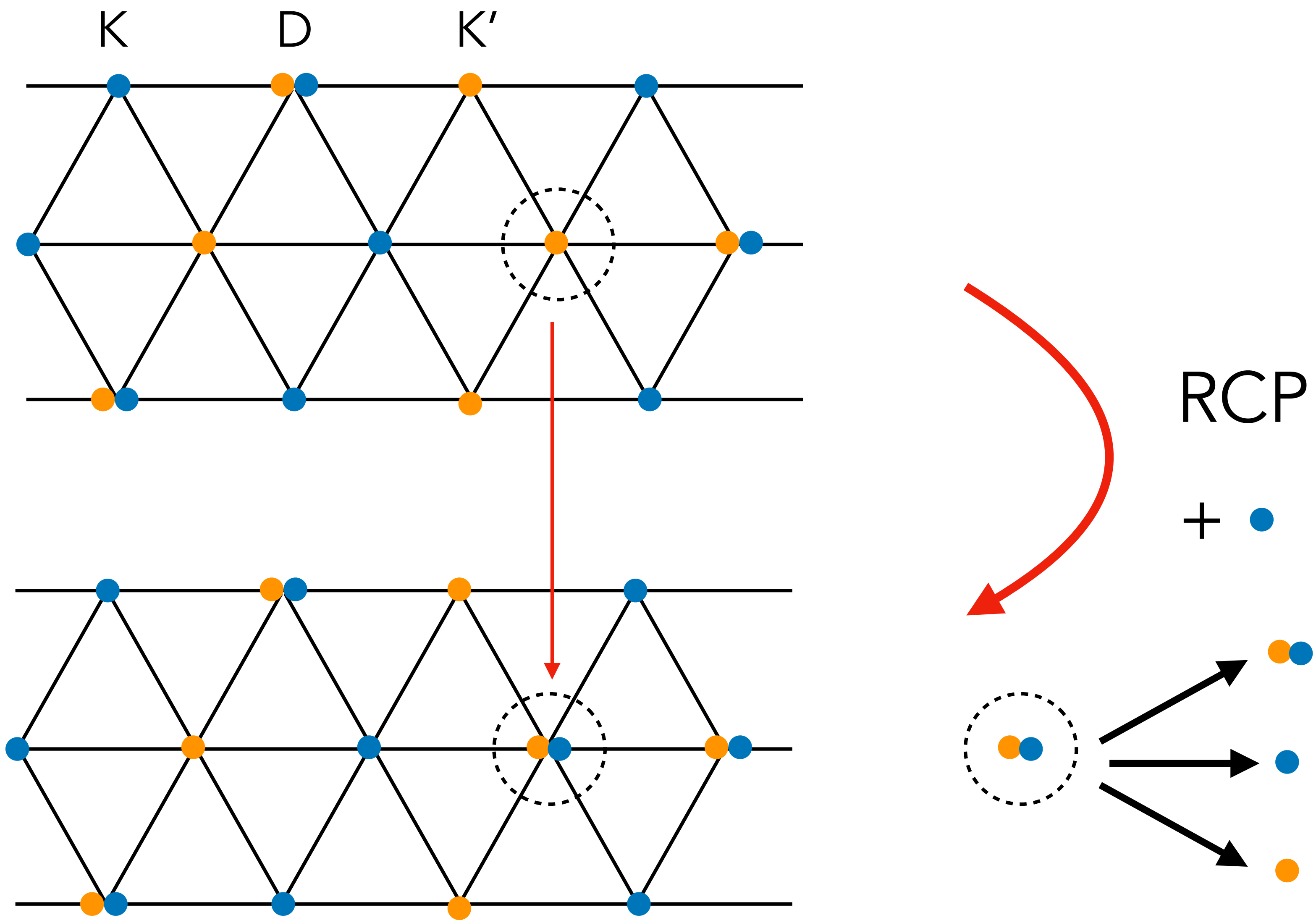
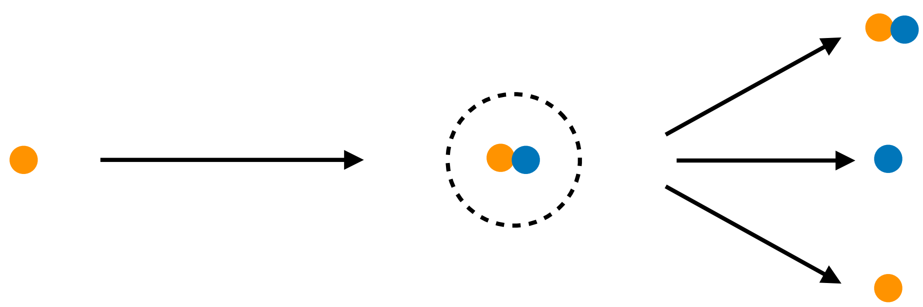
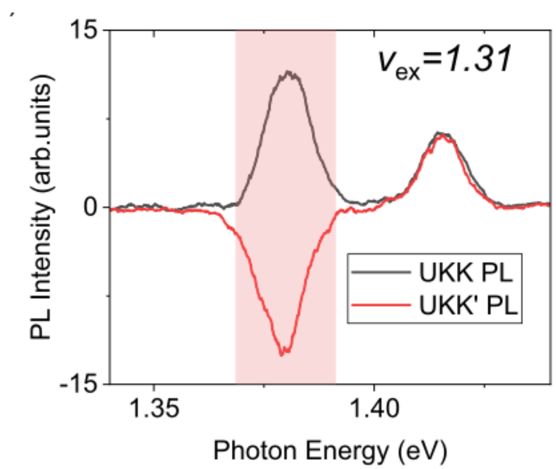


Net result:



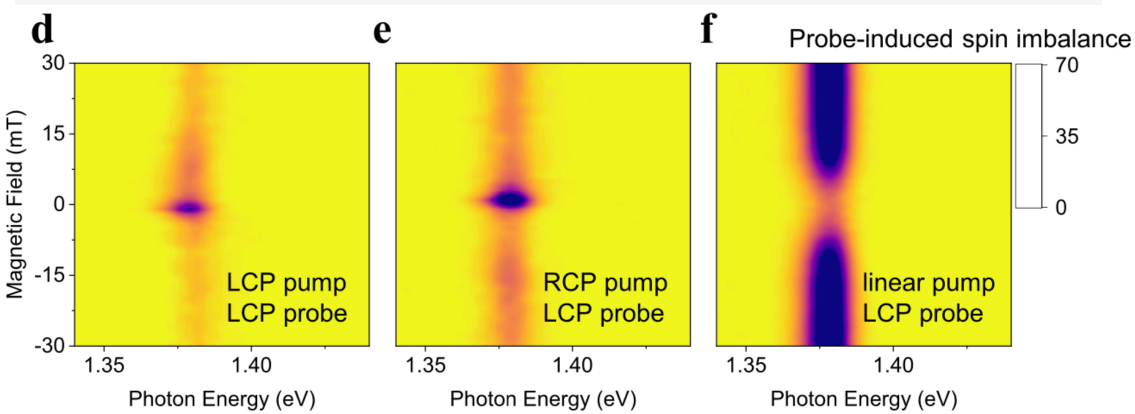
- Doublons increased
- K' valley excitons decreased
- K valley excitons increased



Valley Ferromagnetism?

R. Xiong *et al*, 2024.

Excitons in two-dimensional (2D) semiconductors have offered an attractive platform for optoelectronic and valleytronic devices. Further realizations of correlated phases of excitons promise device concepts not possible in the single particle picture. Here we report tunable exciton "spin" orders in WSe_2/WS_2 moiré superlattices. We find evidence of an in-plane (*xy*) order of exciton "spin"—here, valley pseudospin—around exciton filling $\nu_{ex} = 1$, which strongly suppresses the out-of-plane "spin" polarization. Upon increasing ν_{ex} or applying a small magnetic field of ~10 mT, it transitions into an out-of-plane ferromagnetic (FM-*z*) spin order that spontaneously enhances the "spin" polarization, i.e., the circular helicity of emission light is higher than the excitation. The phase diagram is qualitatively captured by a spin-1/2 Bose–Hubbard model and is distinct from the fermion case. Our study paves the way for engineering exotic phases of matter from correlated spinor bosons, opening the door to a host of unconventional quantum devices.



Polarization signal affected by few mT field!

~mK / exciton!

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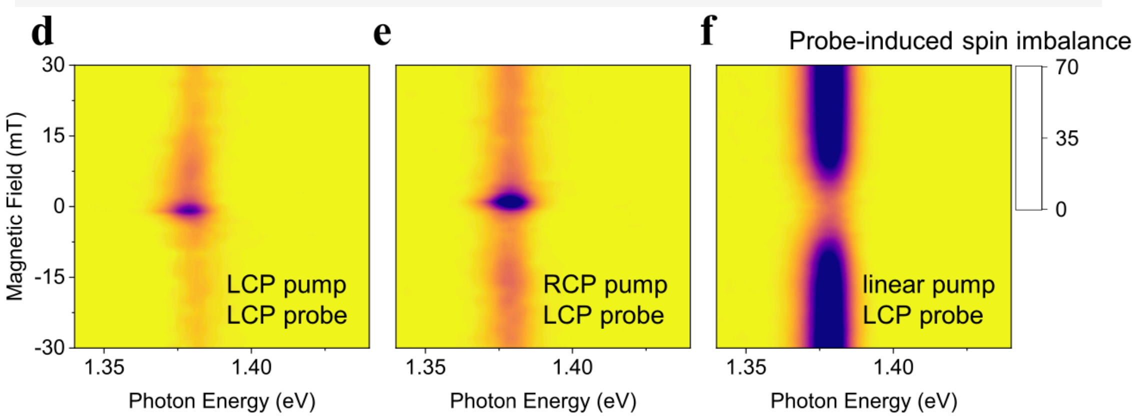

FM: energy scale is multiplied by number of electrons in a domain

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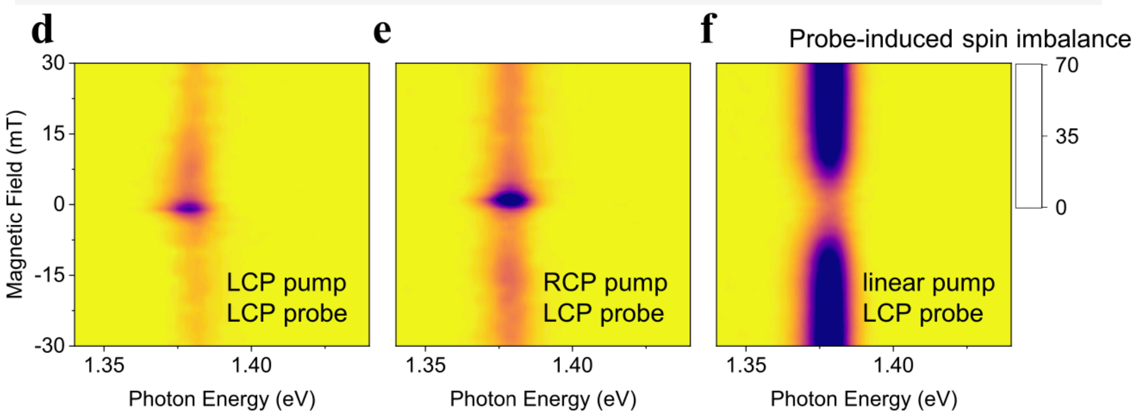

Why ferromagnetism?

- bosonic superexchange?

$$H = \sum_{\langle i,j\rangle,\alpha} -t b^\dagger_{i,\alpha} b_{j,\alpha} + h.c. + \sum_{i,\alpha} U(n_{i,\alpha} - 1/2)^2 + \sum_i V n_{i,1} n_{i,2}$$

- another mechanism?

FM: energy scale is multiplied by number of electrons in a domain

Thanks

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