Optical investigation of strong electronic correlations: magnetism in semiconductor moire materials

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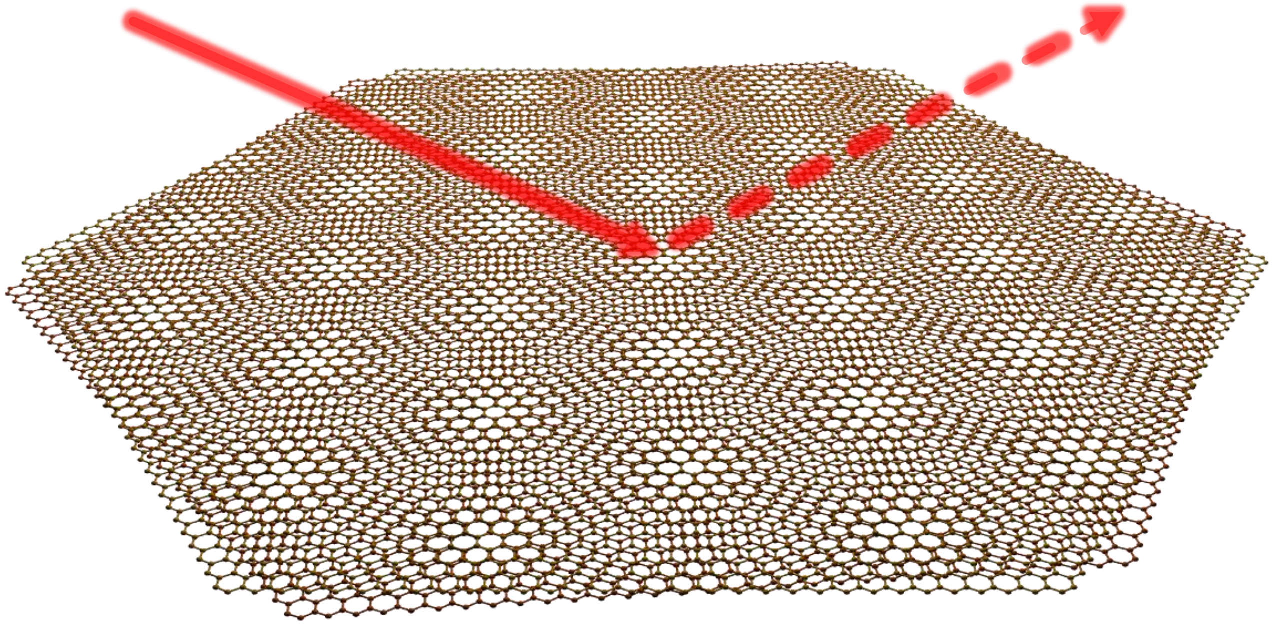
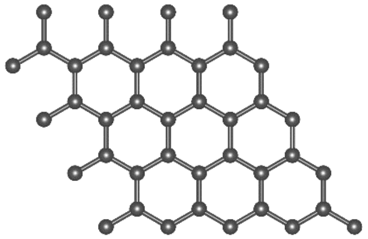


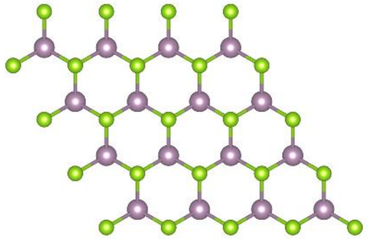
Emerging platform for studying strong correlations: van der Waals heterostructures

- Atomically-thin materials with different electronic, optical and magnetic properties can be combined to create a van der Waals heterostructure with novel hybrid functionality

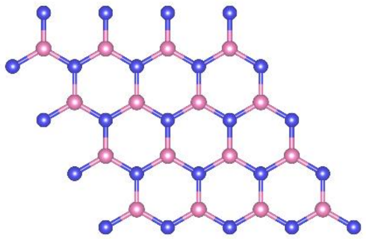
Graphene (metal)



TMD (semiconductor)

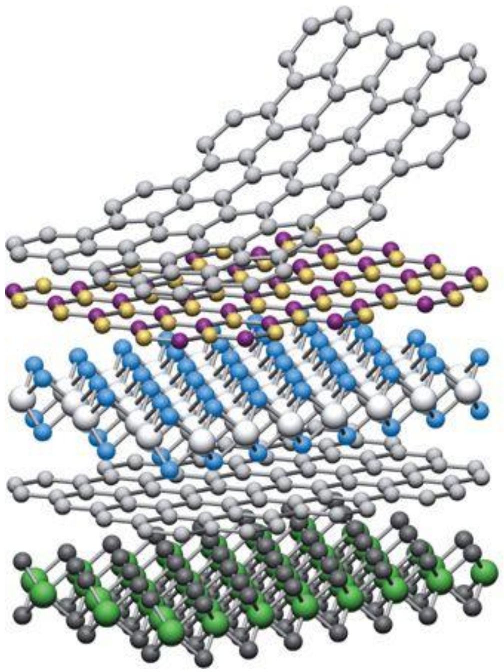


hBN (insulator)



Magnet, Superconductor, TI, …

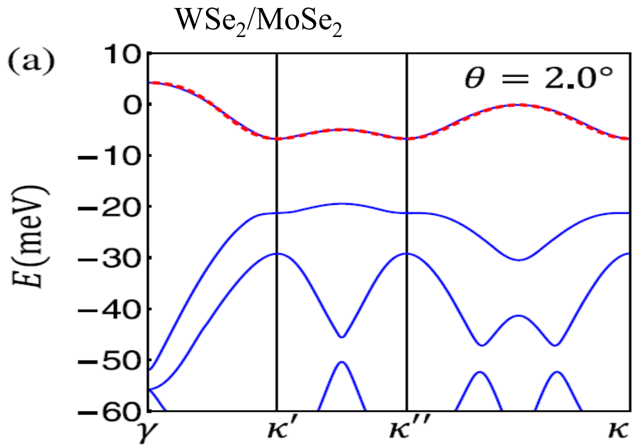
Van der Waals heterostructure



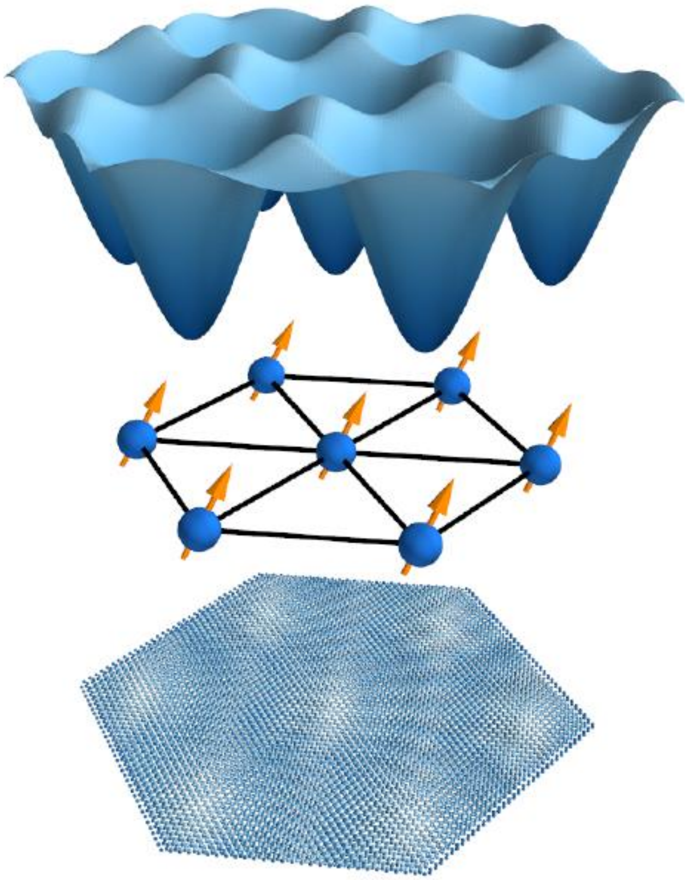
A. K. Geim *et al.*, Nature **499**, 419 (2013)

Moire fatbands in (twisted) semiconductor bilayers

Twisted bilayer transition metal dichalcogenide (TMD)



F. Wu *et al.,* Phys. Rev. Lett. **121**, 026402 (2018)



Heavy effective mass ($m^* \sim 0.7 m_e$)

→ Flat bands in wide range of angles

→ Electric field tunable moire potential

Difficulties

Electrical contact

Inhomogeneity due to strain

Local probe by optical spectroscopy

Mott-Wigner states & quantum anomalous Hall effect
(pioneering work: Wang and Mak-Shan groups)

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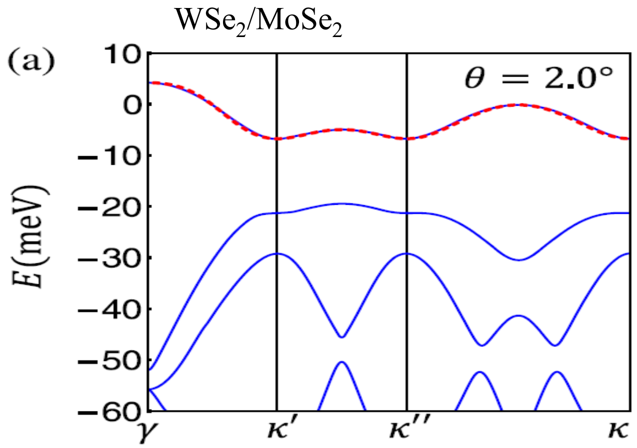

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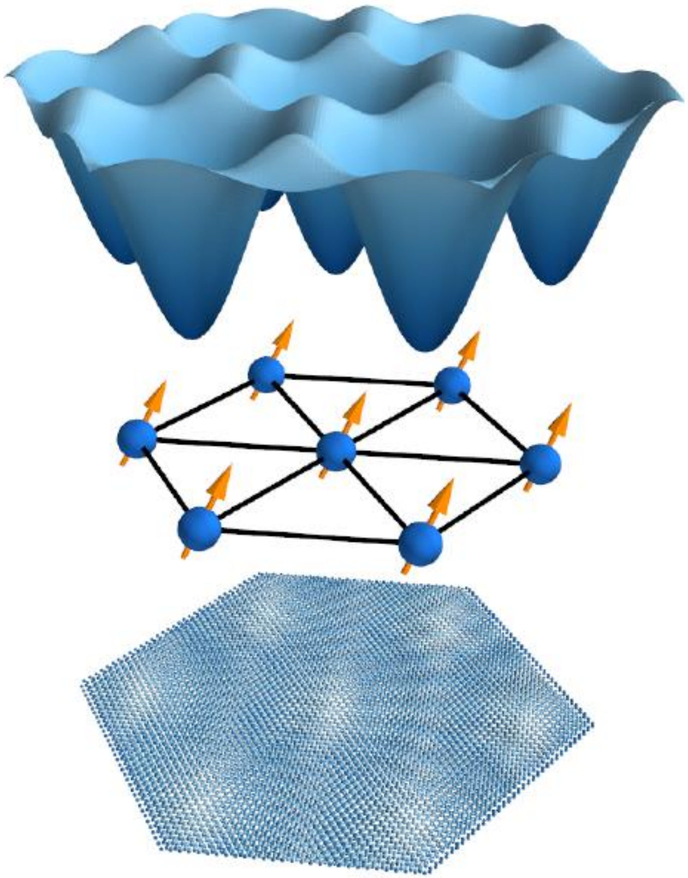


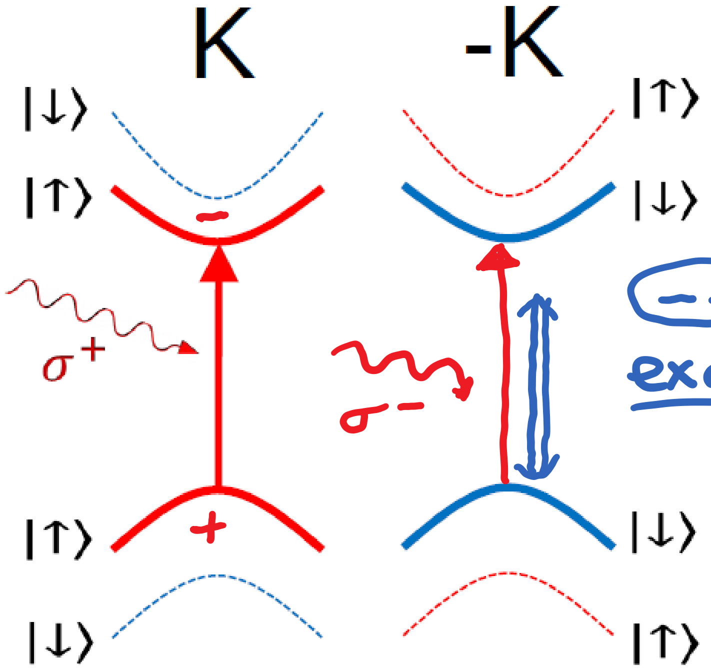
Quantum magnetism of correlated electrons:
120 Neel order or spin liquid or …?

Outline

- Optics/excitons as a spectroscopic tool for investigating ground-state electronic properties

- Kinetic magnetism in semiconductor moire materials: magnetism that stems from kinetic energy minimization, instead of interactions

Materials: Transition metal dichalcogenides (TMD) -layered 2D valley semiconductors



Strong exciton resonance below the band-gap dominates the absorption/ reflection & emission spectrum

(− +) exciton: bound state of an electron & a hole

Valley and spin are locked: low energy K valley states have spin-up

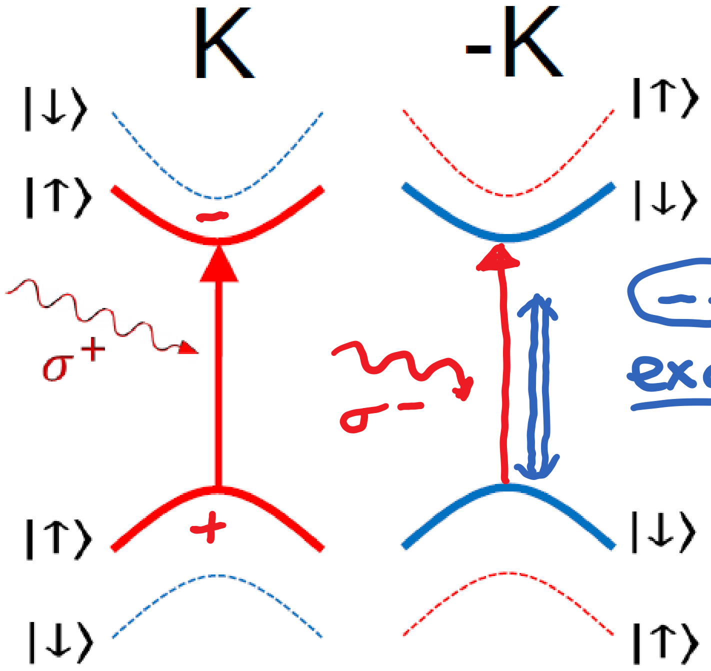
Table 1 Fundamental optoelectronic material parameters of monolayer TMD semiconductors

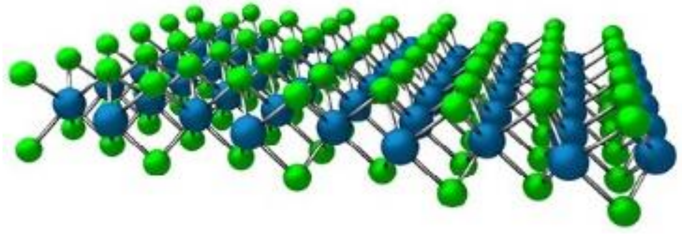
Material	m_r (m_0)	E_b (meV)	E_{gap} (eV)	κ	r_0 (nm)	r_{1s} (nm)
hBN | MoS_2| hBN	0.275 ± 0.015	221	2.160	4.45	3.4	1.2
hBN | $MoSe_2$| hBN	0.350 ± 0.015	231	1.874	4.4	3.9	1.1
hBN | $MoTe_2$| hBN	0.360 ± 0.040	177	1.352	4.4[a]	6.4	1.3
hBN | WS_2| hBN	0.175 ± 0.007	180	2.238	4.35	3.4	1.8
hBN | WSe_2| hBN[b]	0.20 ± 0.01	167	1.890	4.5	4.5	1.7

Experimentally determined values of the exciton reduced mass m_r, the 1s exciton binding energy E_b, the free-particle bandgap E_{gap}, the dielectric screening parameters r_0 and κ, and the root-mean-square radius of the 1s exciton r_{1s}. Typical error bars on experimental values of E_b and E_{gap} are ±3 meV. Typical error bars on values of r_0 and r_{1s} are ±0.1 nm, except for $MoTe_2$, where they are ±0.3 nm
[a]The value of κ for $MoTe_2$ is assumed to be 4.4 and is not a fitting parameter (see text for details)
[b]Values for hBN-encapsulated WSe_2 are taken from ref. [27]

2 NATURE COMMUNICATIONS | (2019)10:4172 | https://doi.org/10.1038/s41467-019-12180-y | www.nature.com/naturecommunications

Materials: Transition metal dichalcogenides (TMD) -monolayer valley semiconductors





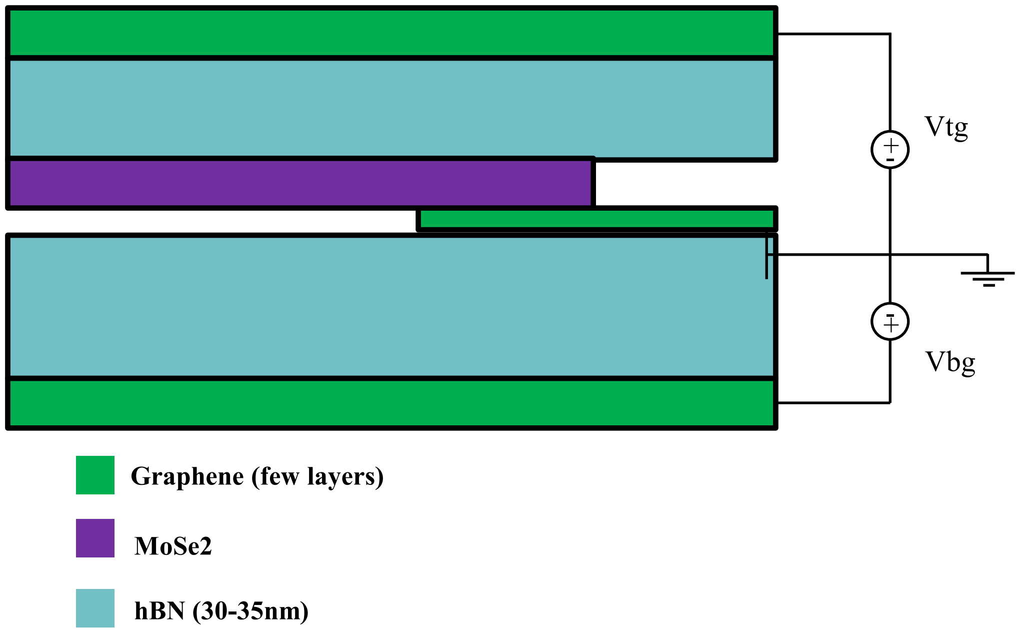
exciton: bound state of an electron & a hole

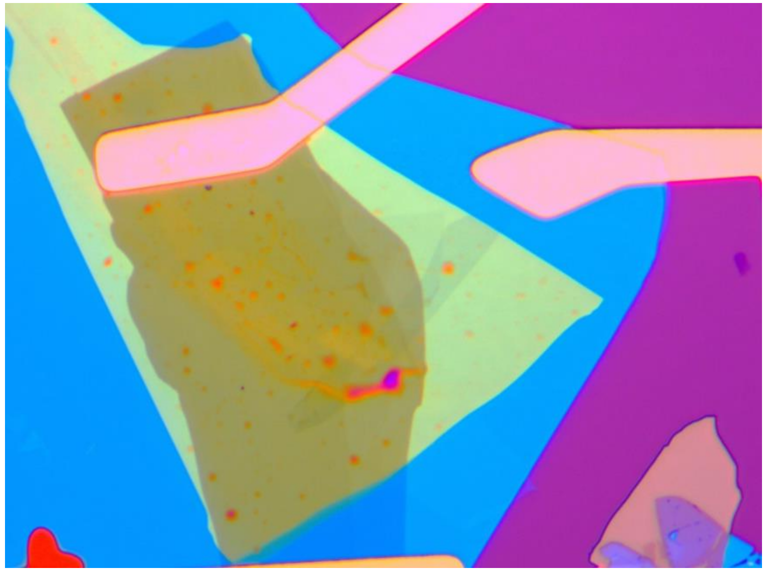
Strong exciton resonance below the band-gap dominates the absorption/ reflection & emission spectrum

Implications of small Bohr radius ($a_B \sim 1$ nm)

- robust quantum impurity (exciton 1s wavefunction unaffected by exciton-electron interactions)
- fast radiative decay of excitons: $\Gamma_x \propto 1/a_B^2$

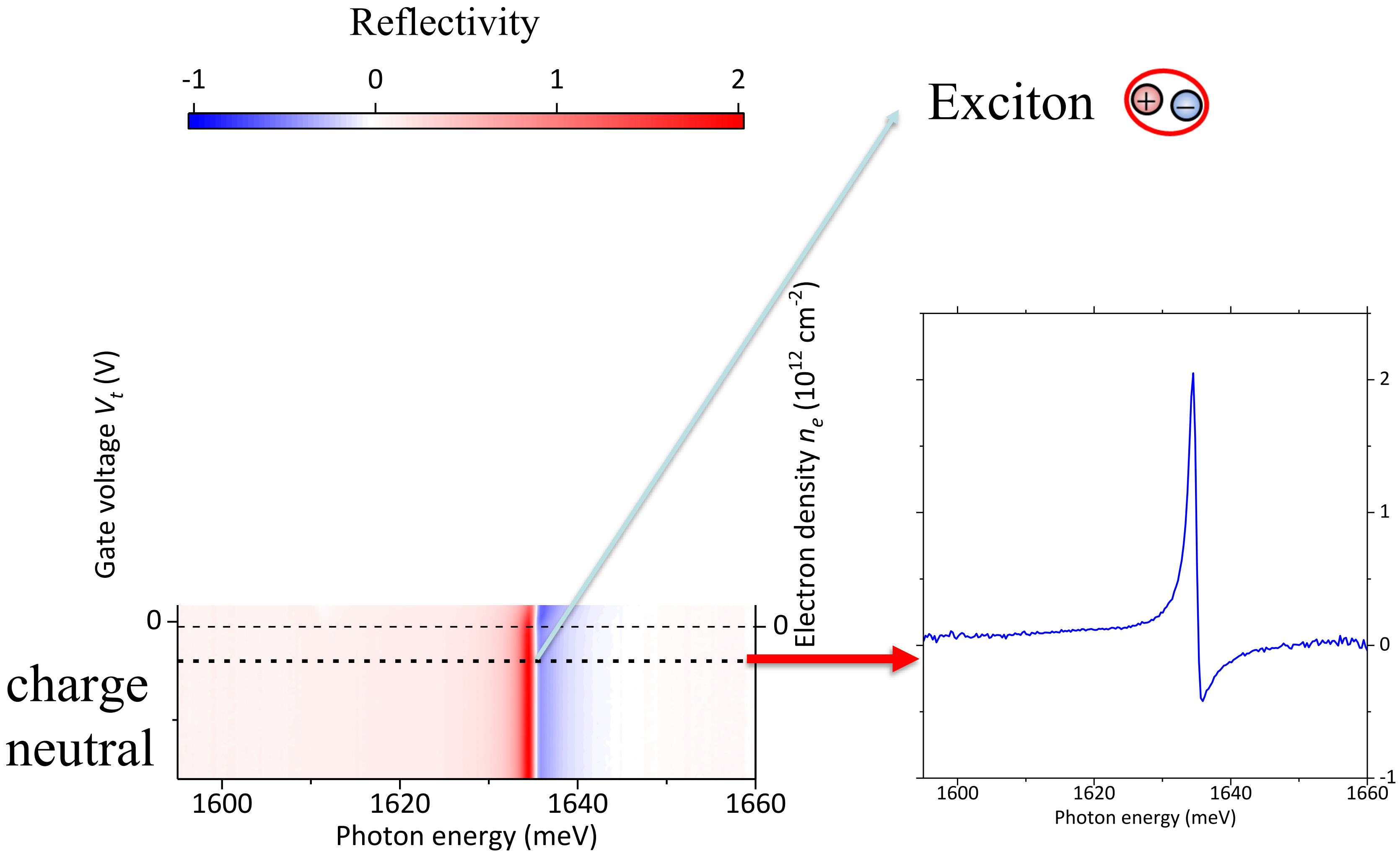
Monolayer $MoSe_2$ device with tunable charge density

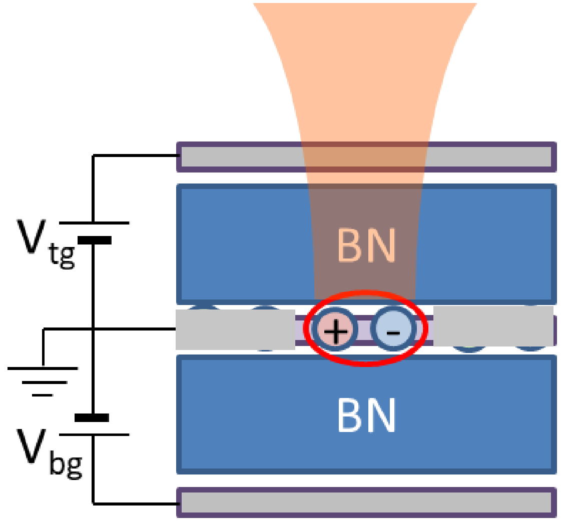




Elementary optical excitations in monolayer $MoSe_2$

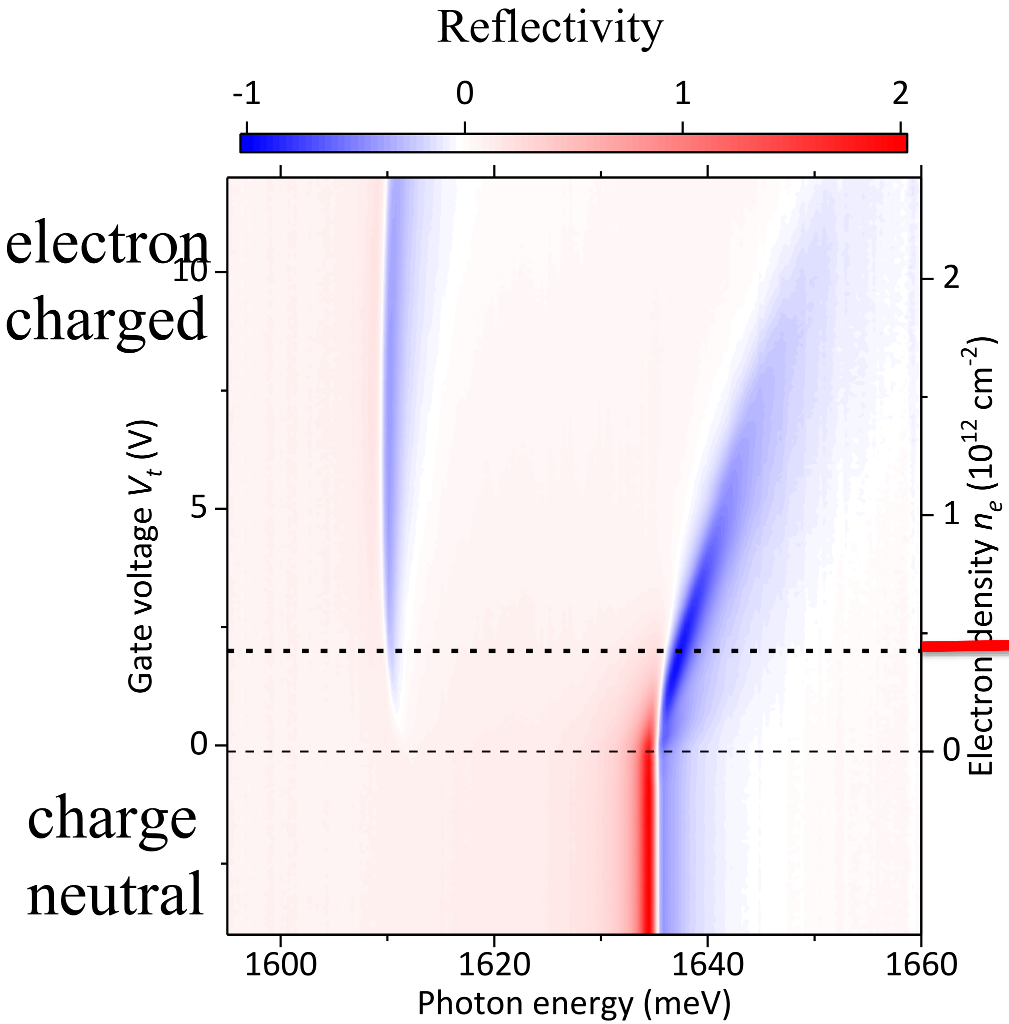
Charge neutrality: tightly bound 1s exciton dominates

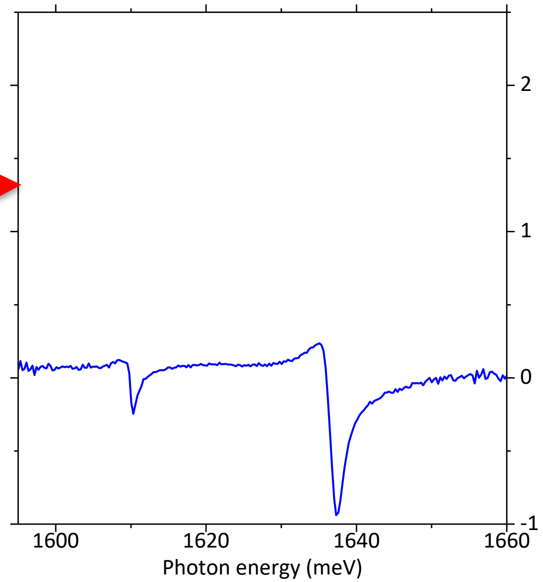


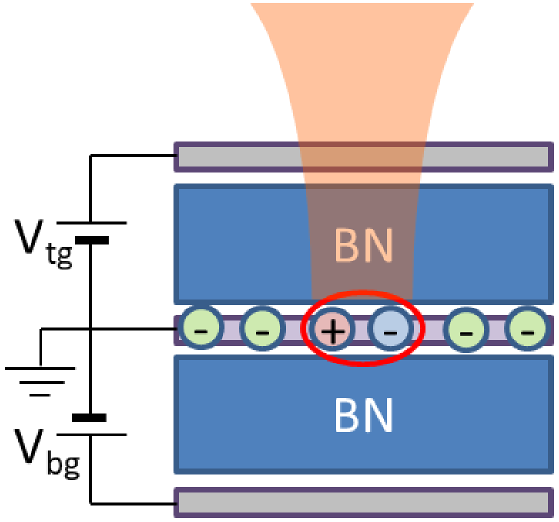


Elementary optical excitations in monolayer $MoSe_2$

Charge neutrality:	tightly bound 1s exciton dominates
Finite electron or hole density:	spectrum is drastically modified





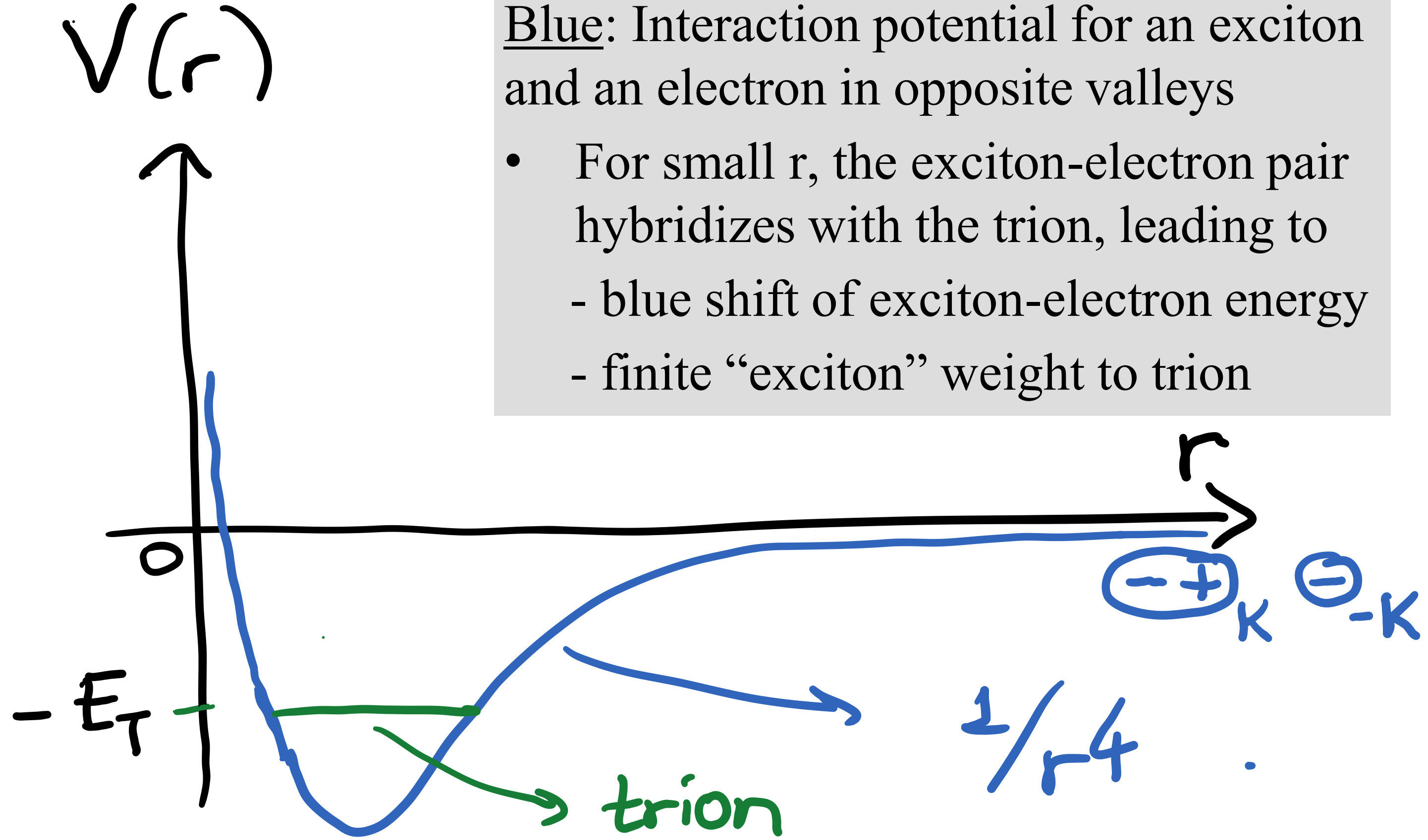


How to understand the modified spectrum: Exciton-electron scattering in a monolayer TMD

- Excitons interact with itinerant electrons or holes from the <u>opposite valley</u> and form a bound molecular state termed "<u>trion</u>"

<u>Blue</u>: Interaction potential for an exciton and an electron in opposite valleys

- For small r, the exciton-electron pair hybridizes with the trion, leading to
  - blue shift of exciton-electron energy
  - finite "exciton" weight to trion



exciton (−+)
electron ⊖
r = separation of an exciton & electron

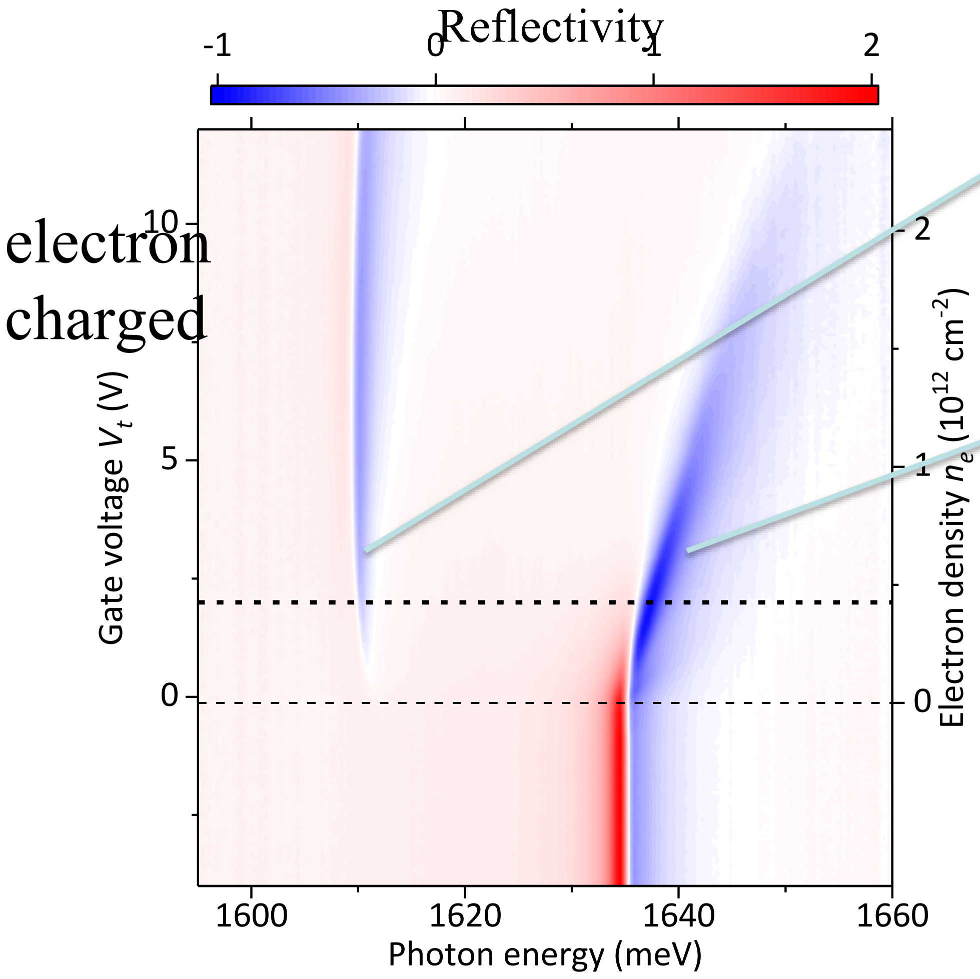
<u>A free electron-exciton pair</u>:
blue-shift - effective repulsive interaction

<u>Excitons injected below the trion resonance</u>:
red-shift - effective attractive interaction

Elementary optical excitations in monolayer $MoSe_2$

<u>Charge neutrality:</u> tightly bound 1s exciton dominates

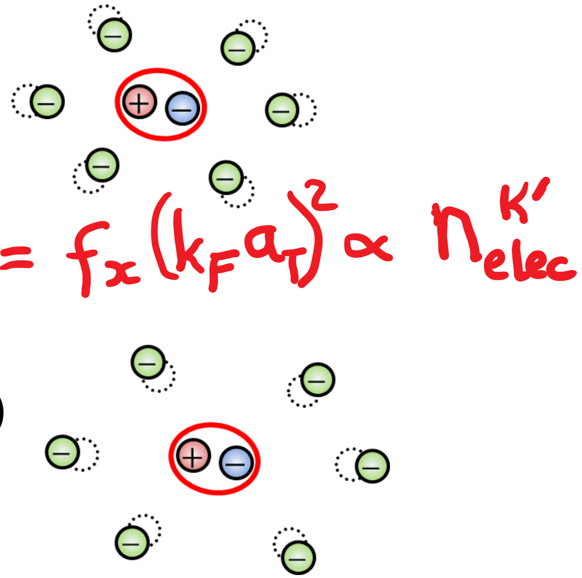
<u>Finite electron or hole density:</u> exciton-polarons (many-body excitations)

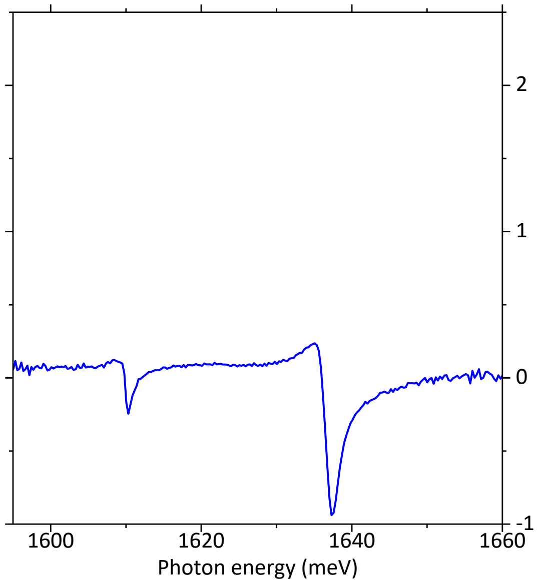


Attractive-polaron (AP)

AP oscillator strength: $f_{AP}^{K} = f_x (k_F a_T)^2 \propto n_{elec}^{K'}$

Repulsive-polaron (RP)

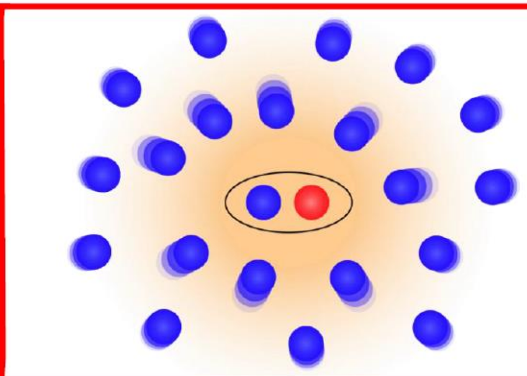




Energy shift of Exciton
(Repulsive polaron)
→ Charge sensing

Attractive polaron as an electronic spin sensor in $MoSe_2$ monolayer

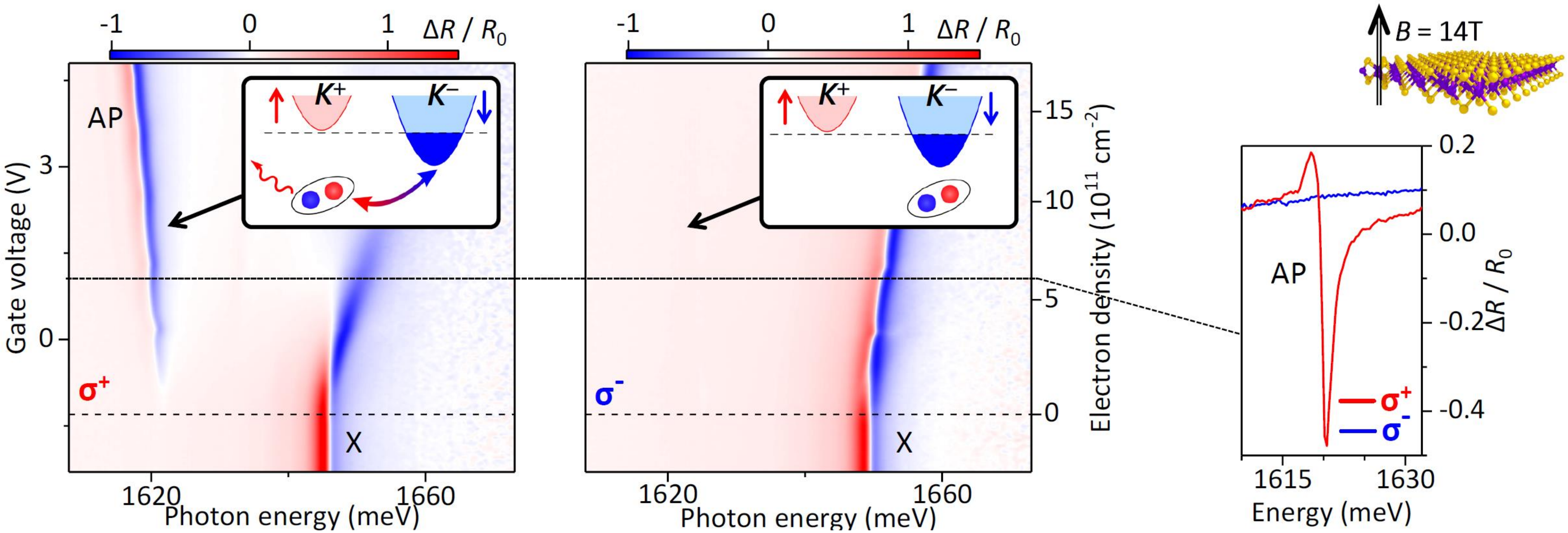
Attractive polaron (AP)



- Collective trion excitation
- Oscillator strength f_{AP} proportional to the electron density

$$f_{\mathrm{AP}}^{\pm} = f_{\mathrm{X}} \cdot (4\pi a_T^2) \cdot n_e^{\mp}$$

Due to large spin-orbit coupling at K,K' valleys, spin and valley are locked



Attractive polaron as an electronic spin sensor in $MoSe_2$ monolayer

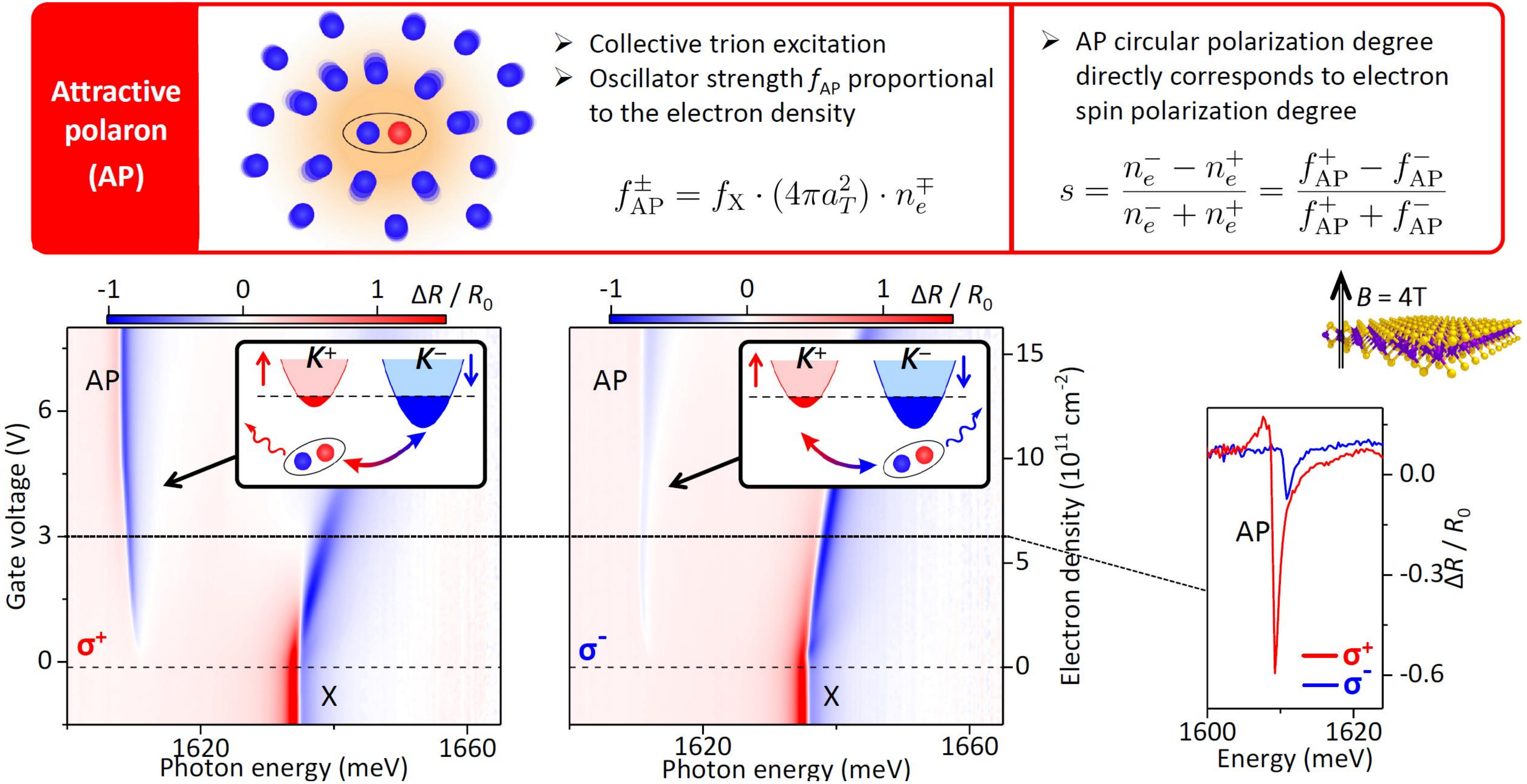
Attractive polaron (AP)

- Collective trion excitation
- Oscillator strength f_{AP} proportional to the electron density

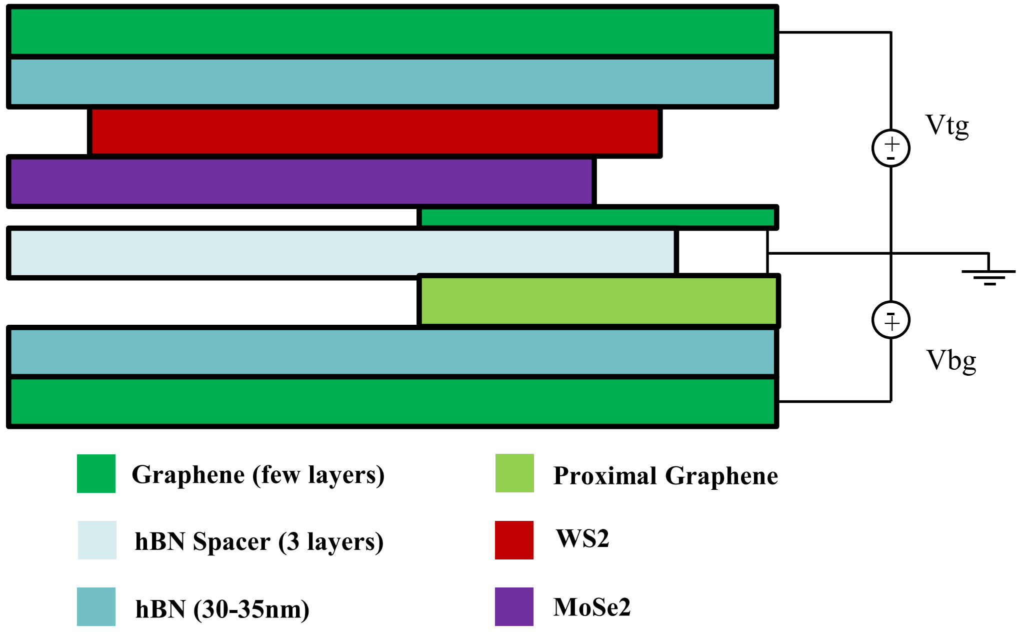
$$f_{\mathrm{AP}}^{\pm} = f_{\mathrm{X}} \cdot (4\pi a_T^2) \cdot n_e^{\mp}$$

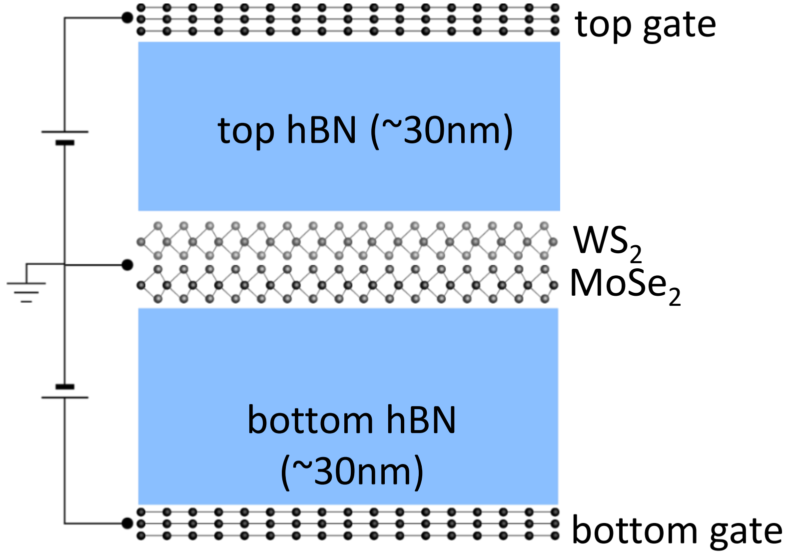
- AP circular polarization degree directly corresponds to electron spin polarization degree

$$s = \frac{n_e^- - n_e^+}{n_e^- + n_e^+} = \frac{f_{\mathrm{AP}}^+ - f_{\mathrm{AP}}^-}{f_{\mathrm{AP}}^+ + f_{\mathrm{AP}}^-}$$



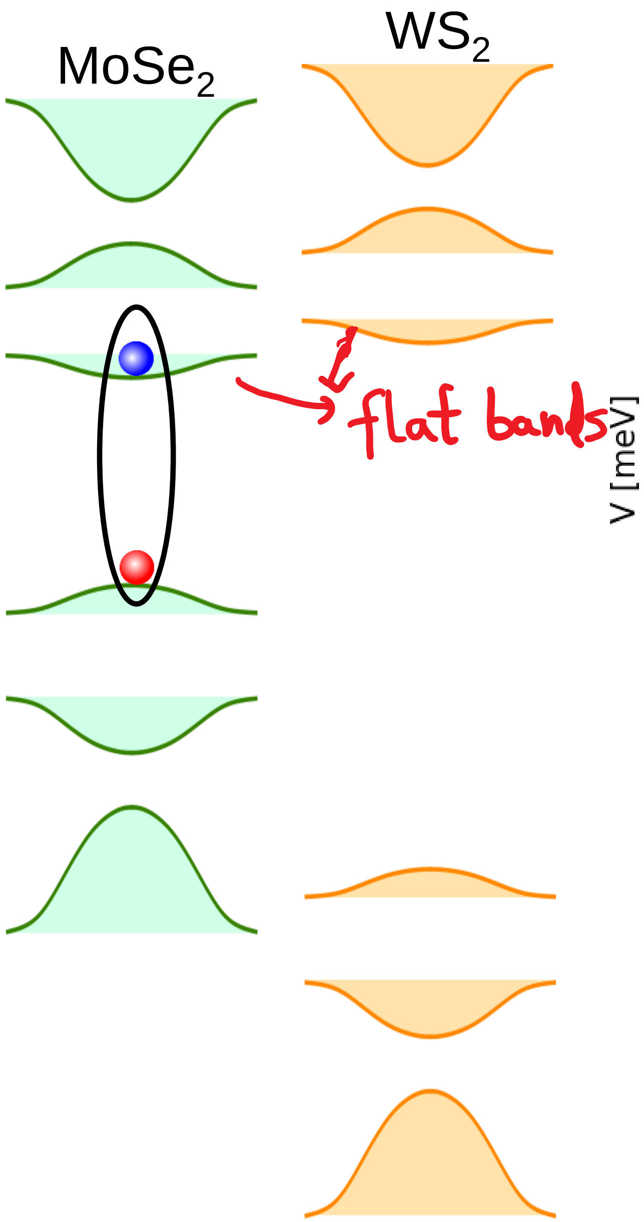
A semiconducting moire system: $MoSe_2/WS_2$



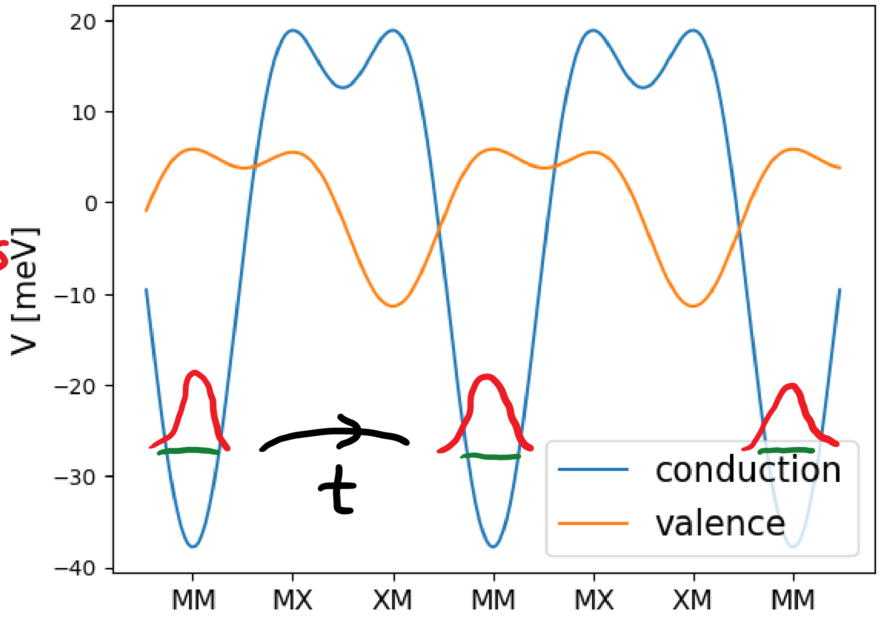


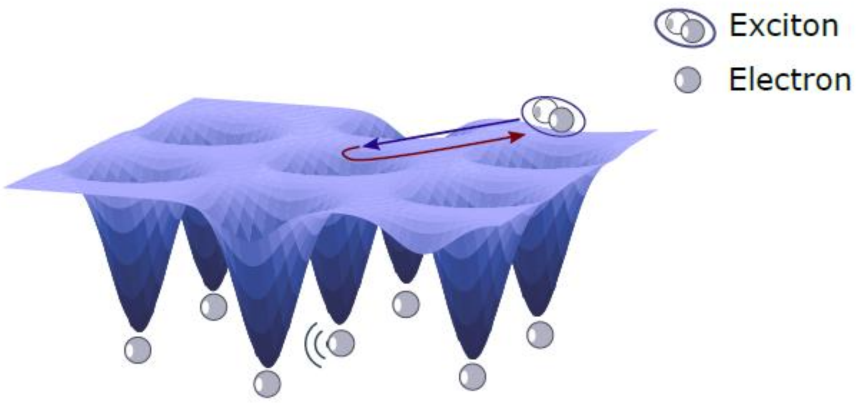
- Moire potential due to lattice mismatch of ~ 4%
- Type I band alignment – both CB minimum and VB maximum are in MoSe2
- Electronic bands are trivial but E-field tuning may yield Chern bands

A semiconducting moire system: $MoSe_2/WS_2$



(Yang Zhang)

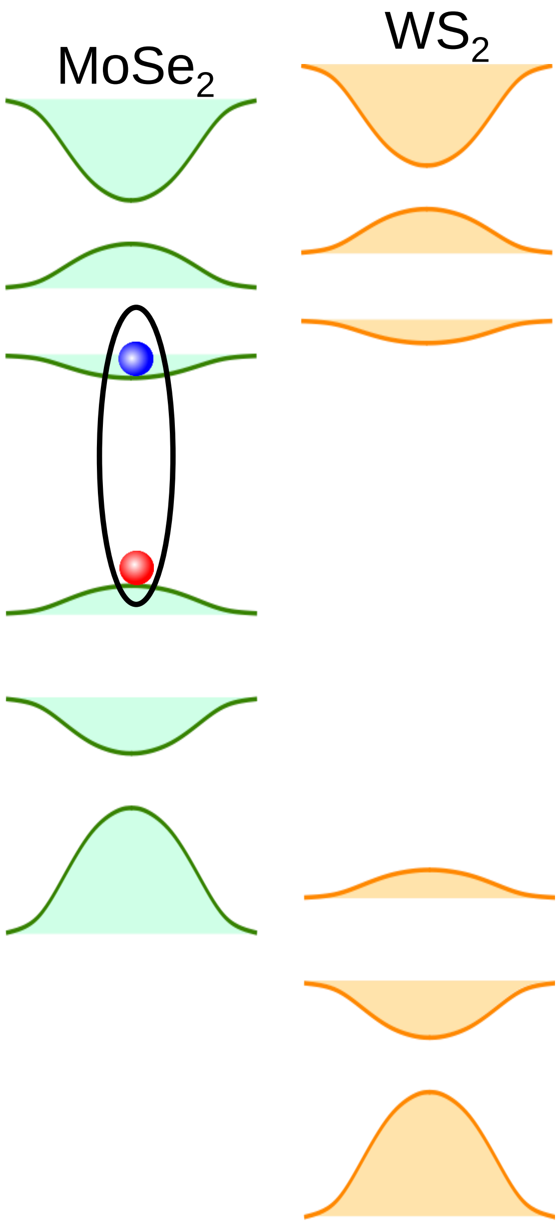


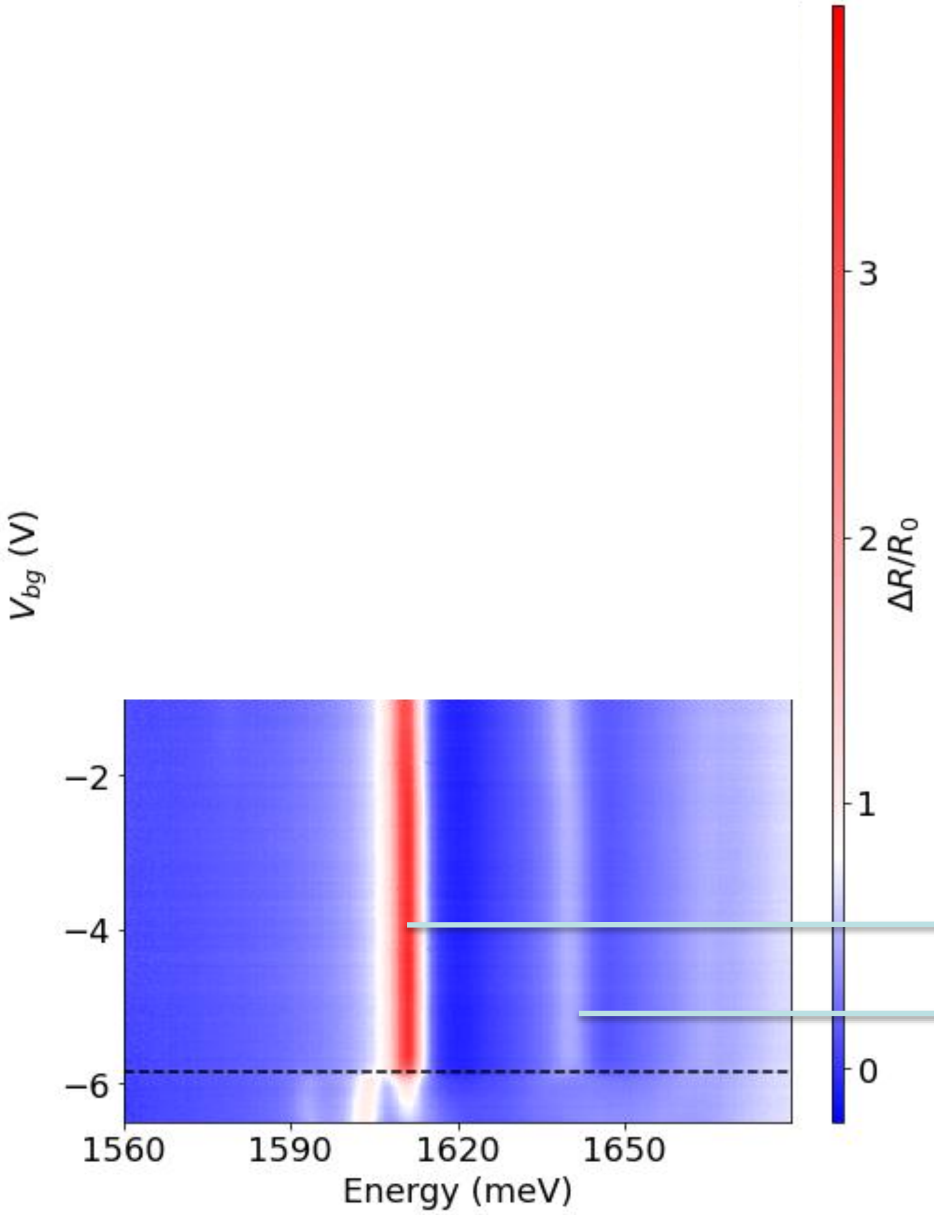


$U^c_{on\text{-}site} \sim 50$ meV
hopping: $t \sim 1$ meV

- Potential landscape for electrons and excitons are generically different.

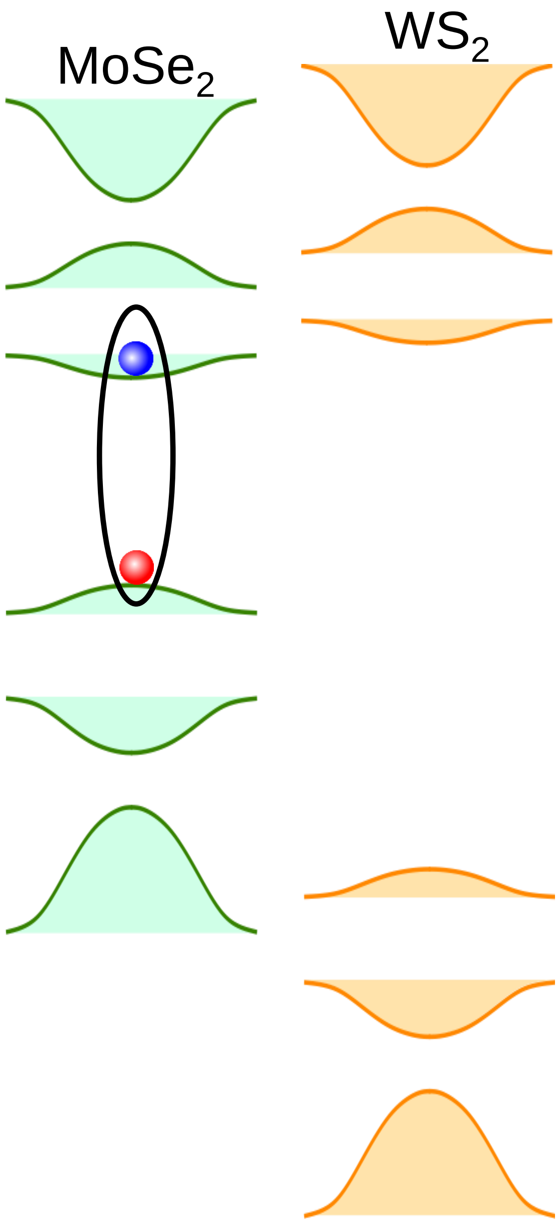
Semiconductor moire system: $MoSe_2/WS_2$

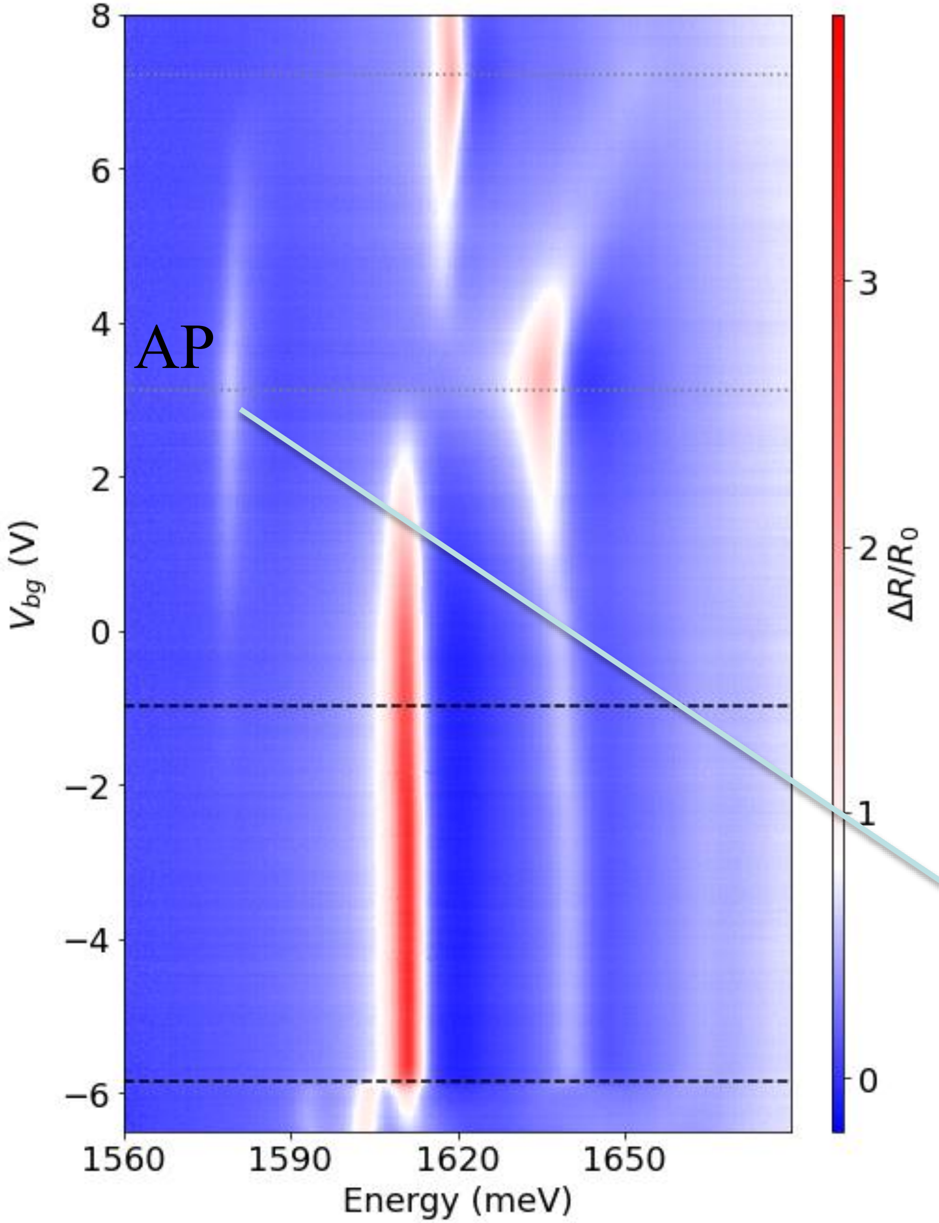


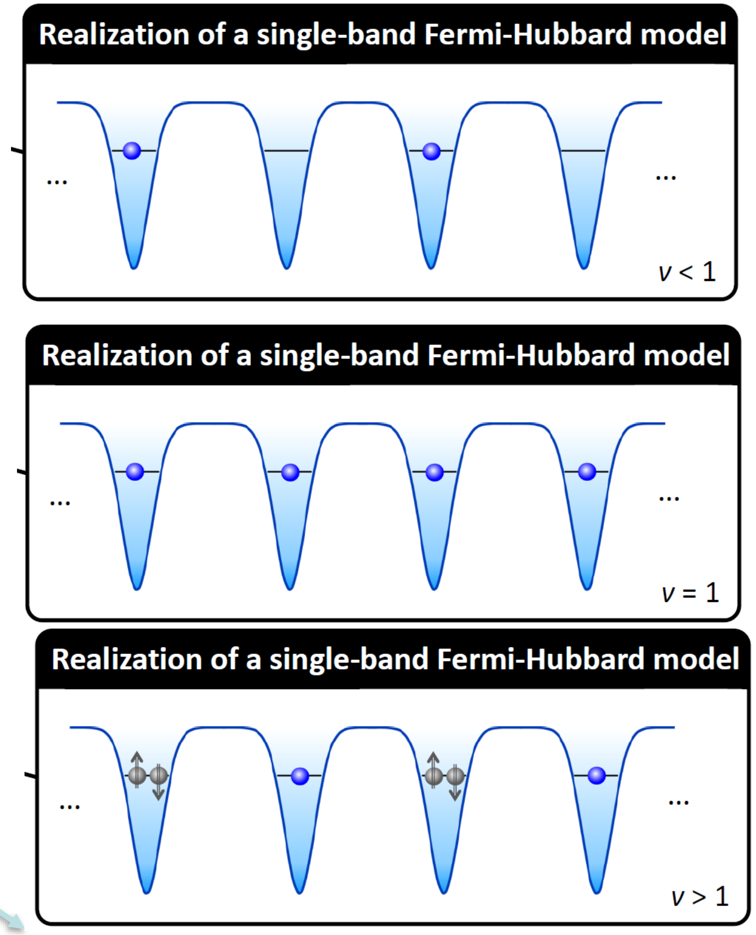


Moiré excitons: periodic potential leads to formation of flat exciton bands in charge neutrality

Semiconductor moire system: $MoSe_2/WS_2$

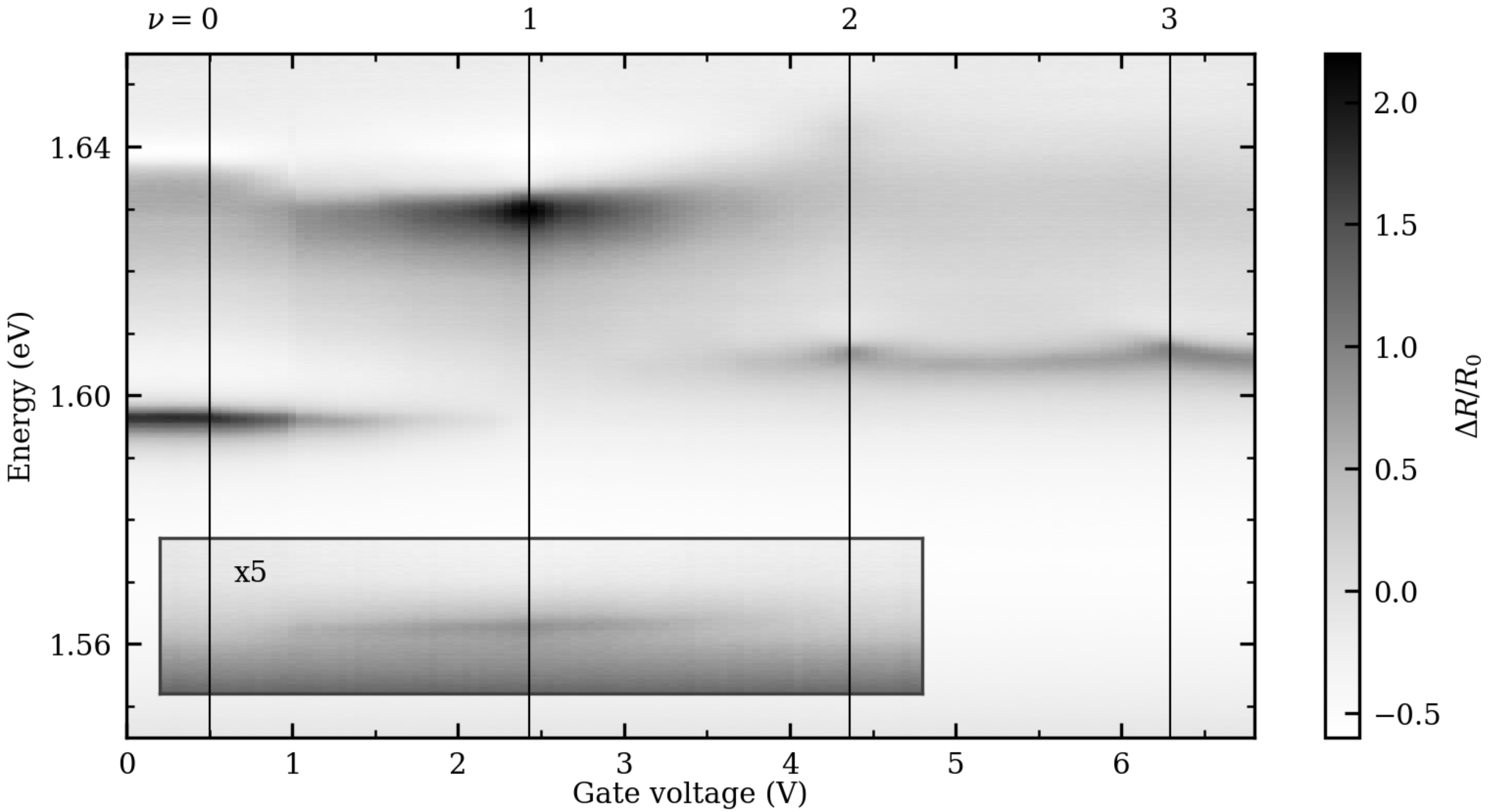






Attractive polaron: dressing of lowest energy moire exciton by electrons strongly localized on moire lattice sites

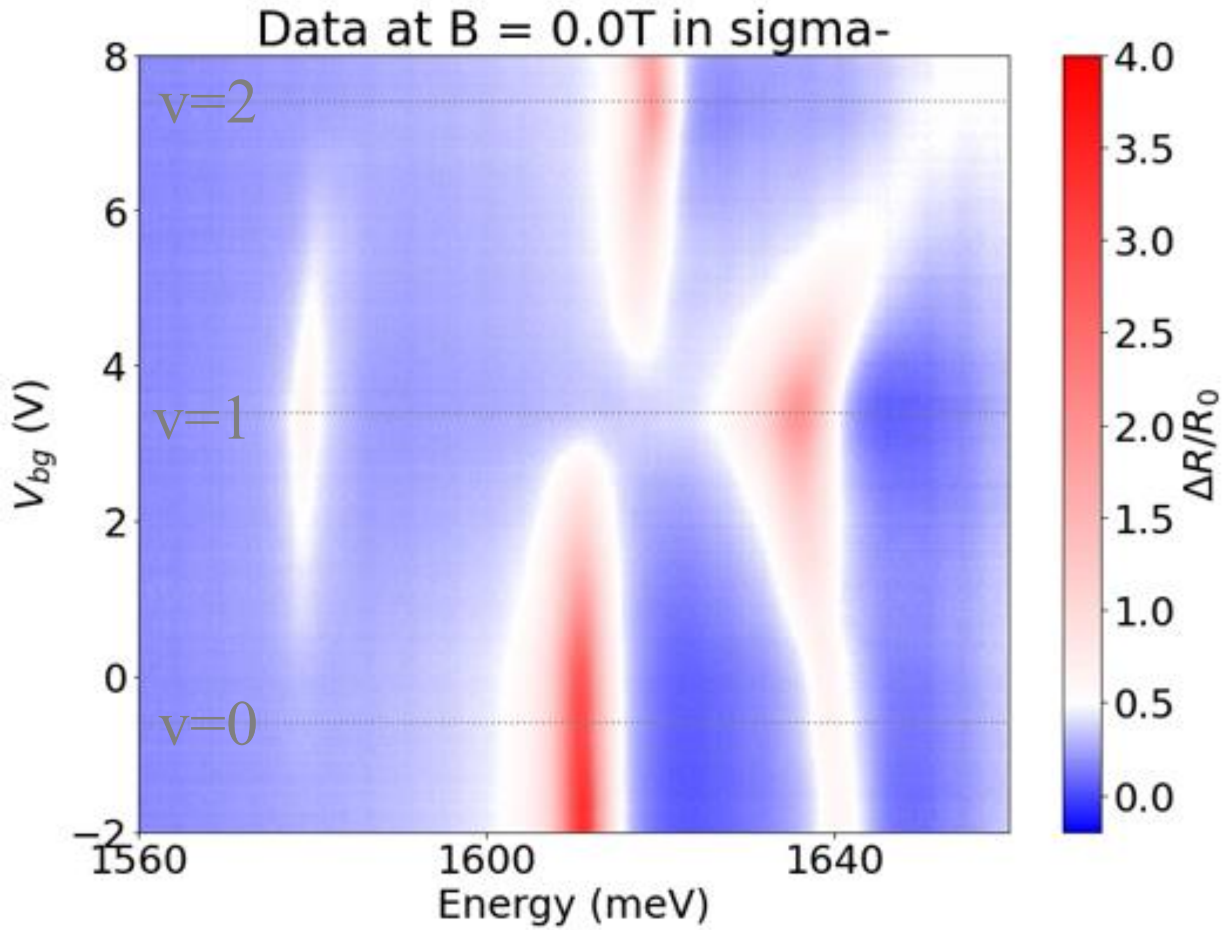
Semiconductor moire system: $MoSe_2/WS_2$: Second device

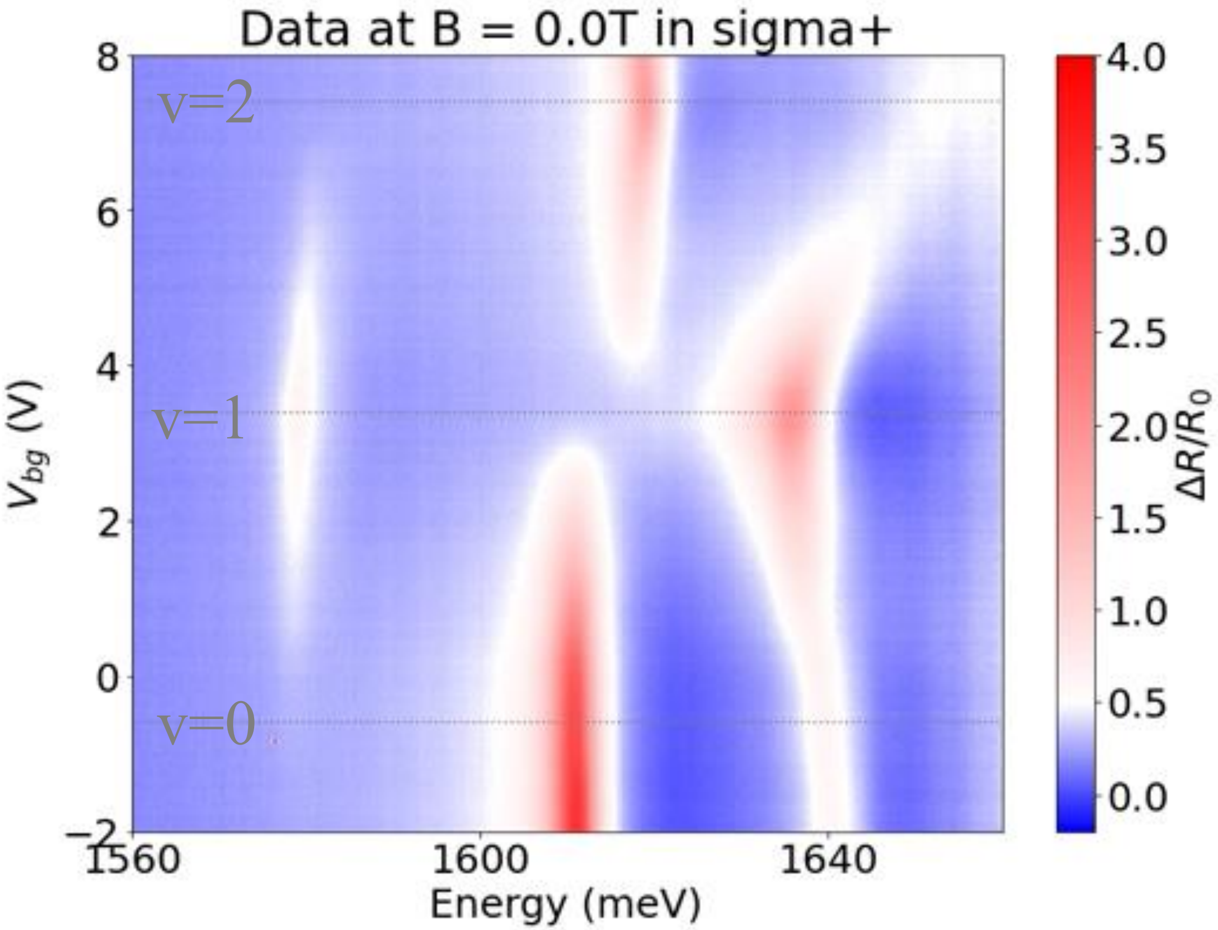


Cusps in reflection for ν = 2 and ν = 3 signaling incompressible electronic states even at high doping

Magnetic field response at T=4 K

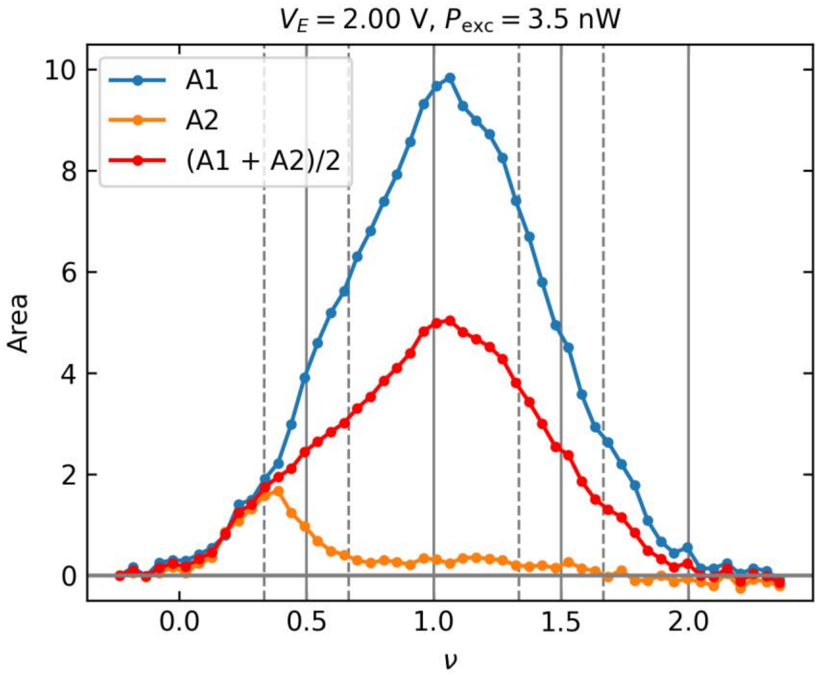
To explore magnetic properties, we determine magnetization as a function of temperature and magnetic field





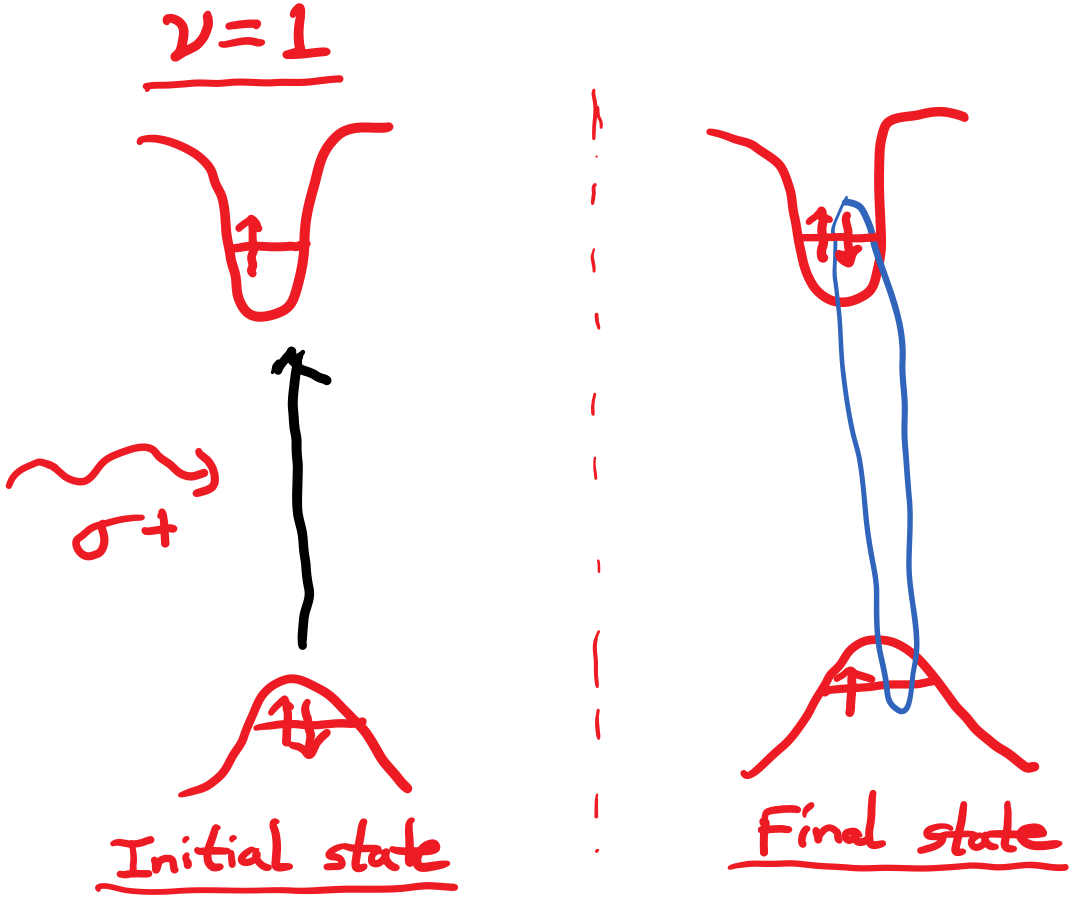
Evidence for the (extended) single-band Hubbard model

- For B=4T, electrons are spin-valley polarized in K'-valley; the strength of K-valley AP increases linearly till ν=1 and then decreases linearly; for ν=2, all electronic states in the lowest moire flat band are filled and they cannot contribute to AP/trion formation.

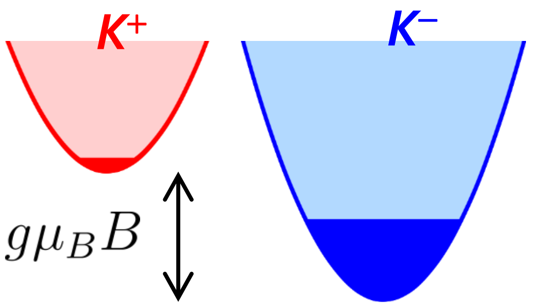


A1: strength of K-valley (σ+ polarized) AP

A2: strength of K'-valley (σ- polarized) AP



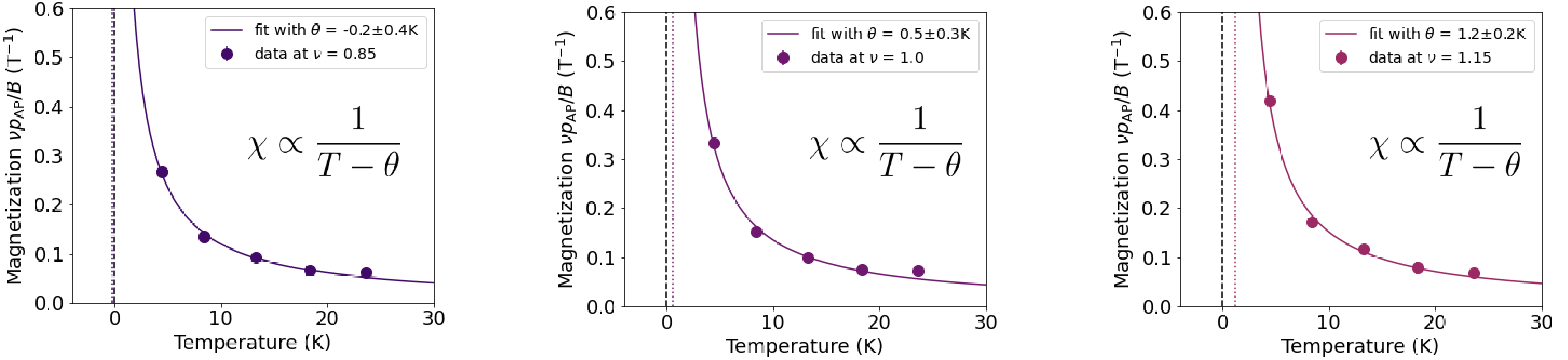
Direct measurement of magnetization (p_{AP}) using attractive polarons



$$n_+ = n_- \exp\left(-\frac{g\mu_B B}{k_B T}\right)$$

$$p_{AP} = \frac{n_- - n_+}{n_- + n_+}$$

Spin susceptibility: $\chi \propto \lim_{B\to 0} \nu \frac{p_{AP}}{B}$

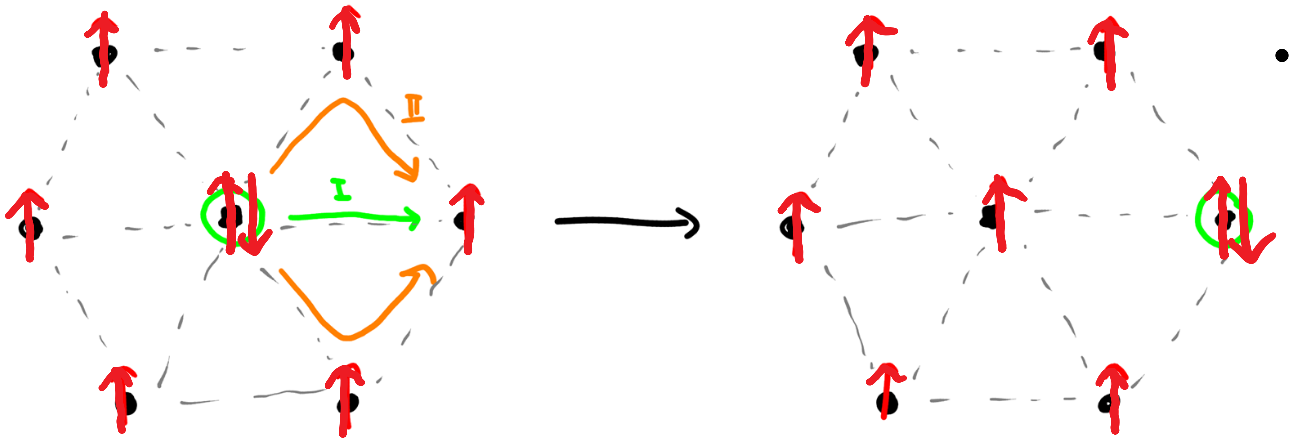


Electron moire magnetism for v ~ 1: determined by interactions or kinetic energy?

- **For ν=1**, $\theta_{CW} \sim J = t^2/U \sim 0$, since U= 50 meV and t=1 meV

Electron moire magnetism for ν ~ 1: determined by interactions or kinetic energy?

- **For ν=1**, $\theta_{CW} \sim J = t^2/U \sim 0$, since U= 50 meV and t=1 meV
- **For ν>1**, $\theta_{CW} > 0$: Nagaoka ferromagnetism (FM) with $\theta_{CW} \sim t$



- Constructive interference of different paths if all spins are aligned

Electron moire magnetism for ν ~ 1: determined by interactions or kinetic energy?

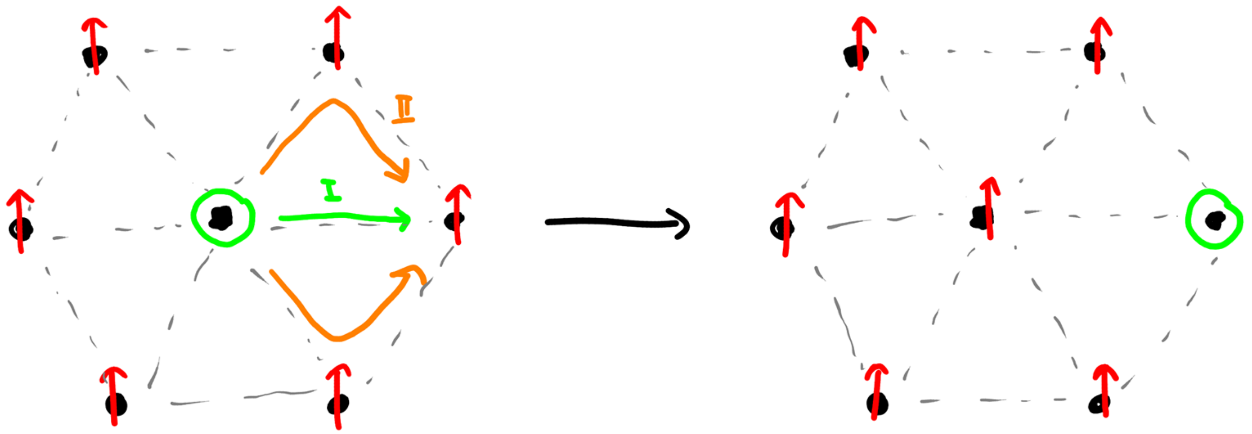
- **For ν=1**, $\theta_{CW} \sim J = t^2/U \sim 0$, since U= 50 meV and t=1 meV

- **For ν>1**, $\theta_{CW} > 0$: Nagaoka ferromagnetism (FM) with $\theta_{CW} \sim t$

- **For ν<1**, $\theta_{CW} < 0$: kinetic Antiferromagnetism (AF) with $\theta_{CW} \sim t$, due to kinetic frustration in a triangular lattice

Antiferromagnetic correlations due to kinetic frustration

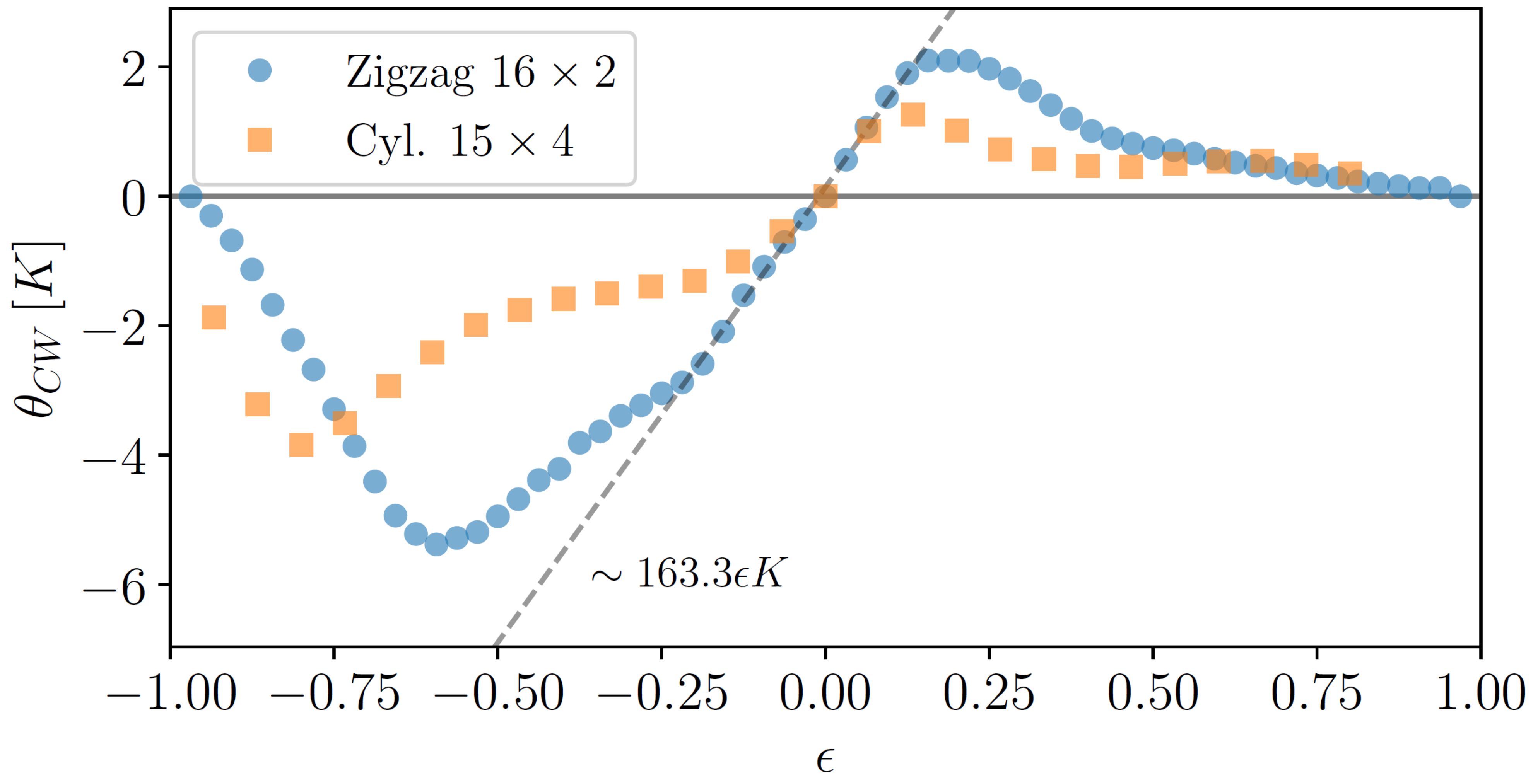
Fermi-Hubbard model for **v<1**



→ Indistinguishable paths interfere destructively due to negative hopping t

→ Spin-polaron formation (hole bound to a spin-flip) allows for KE gain
(Haerter&Shastry,Fu,Demler)

DMRG results (Morera & Demler)

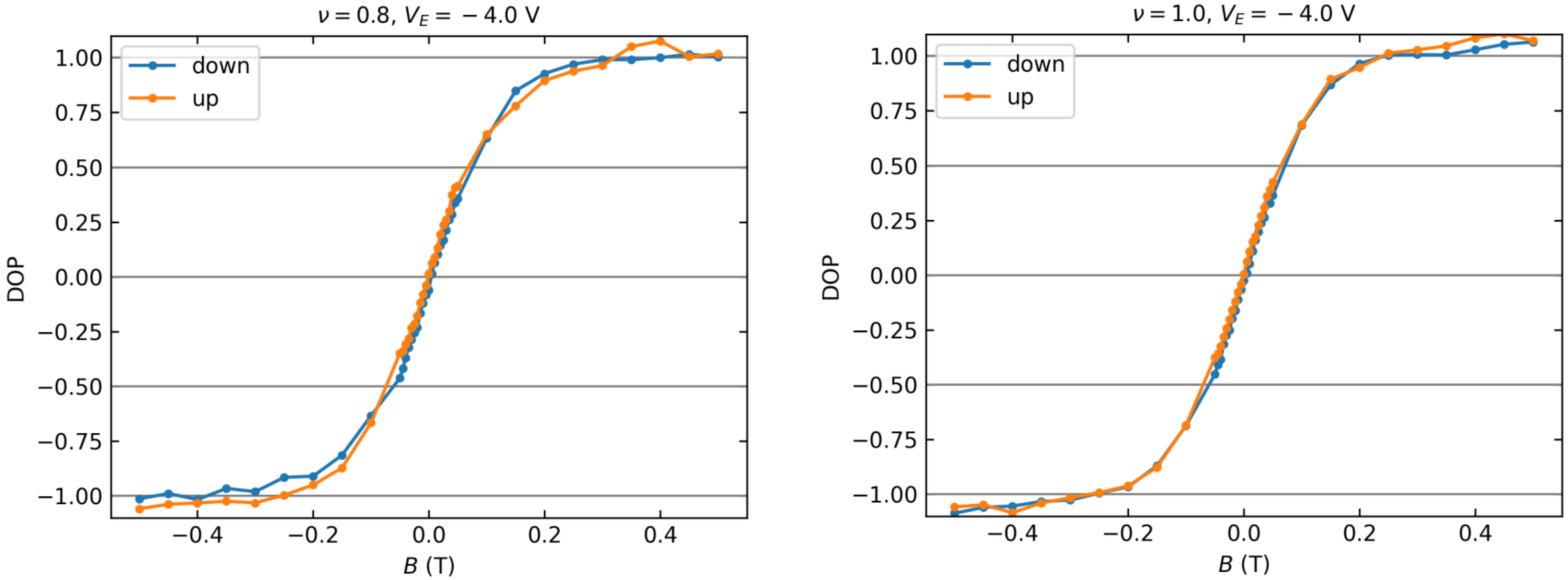


When Coulomb is replaced by infinite strength on-site repulsion (contact interaction), DMRG predicts a switch from AF to F limits

Experimental magnetization at T < 500 mK

Paramagnetic response for ν ≤ 1

~ 1 photon every 100 ns

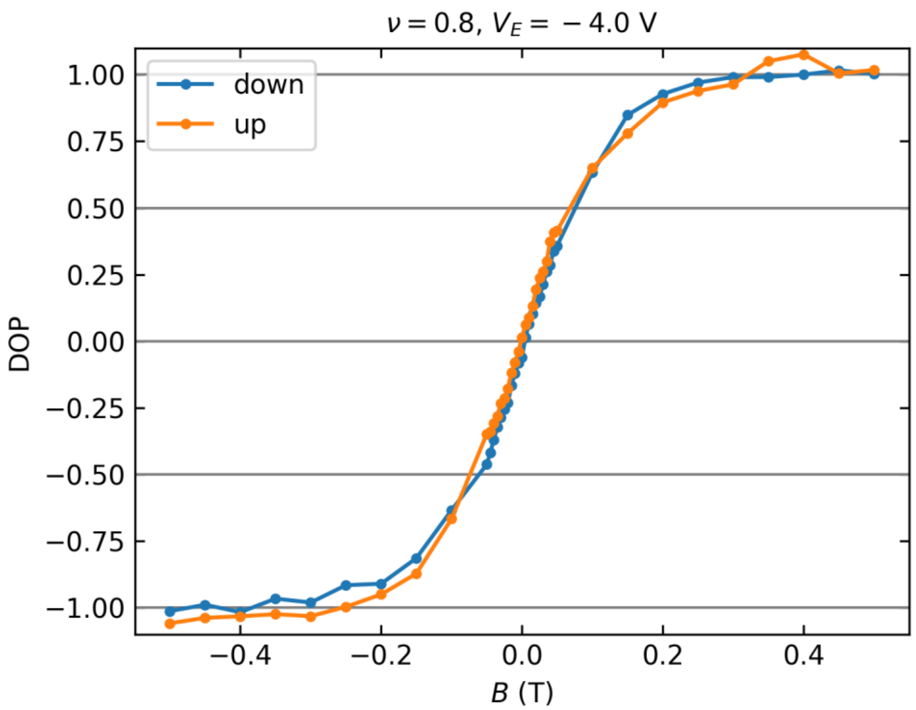


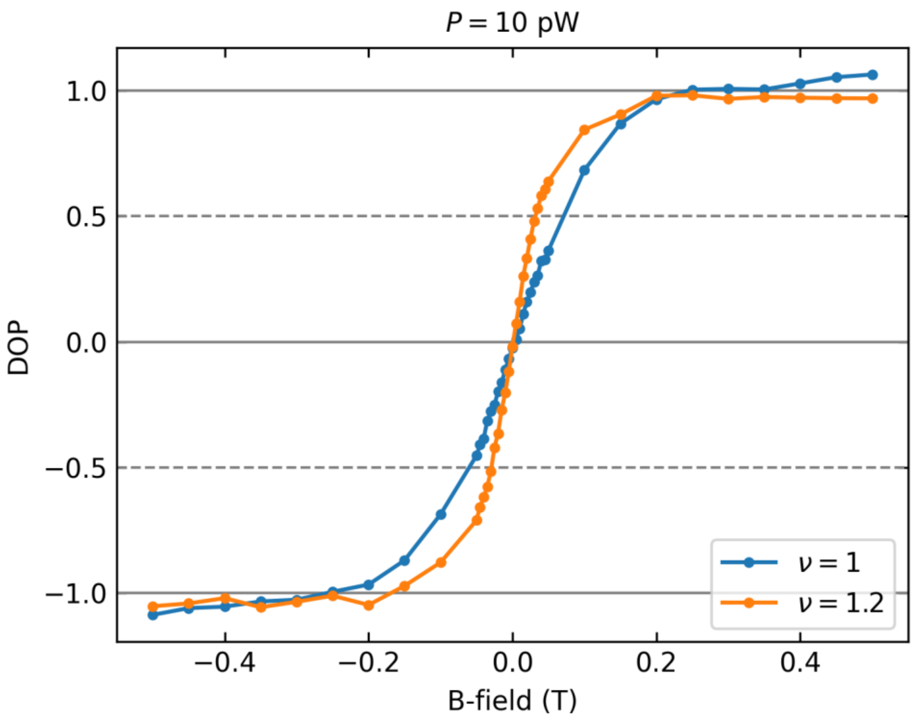
→ Degree of polarization (DOP) as a function of B identical for $\nu = 0.8$ and $\nu = 1$

Experimental magnetization at T < 500 mK

Enhanced susceptibility for ν > 1

~ 1 photon every 100 ns

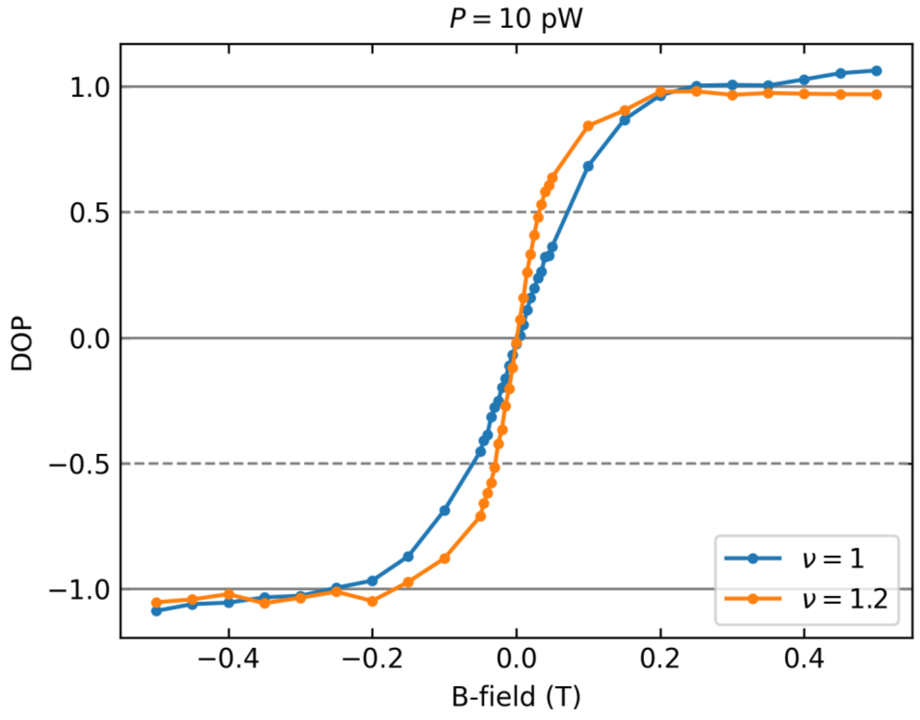


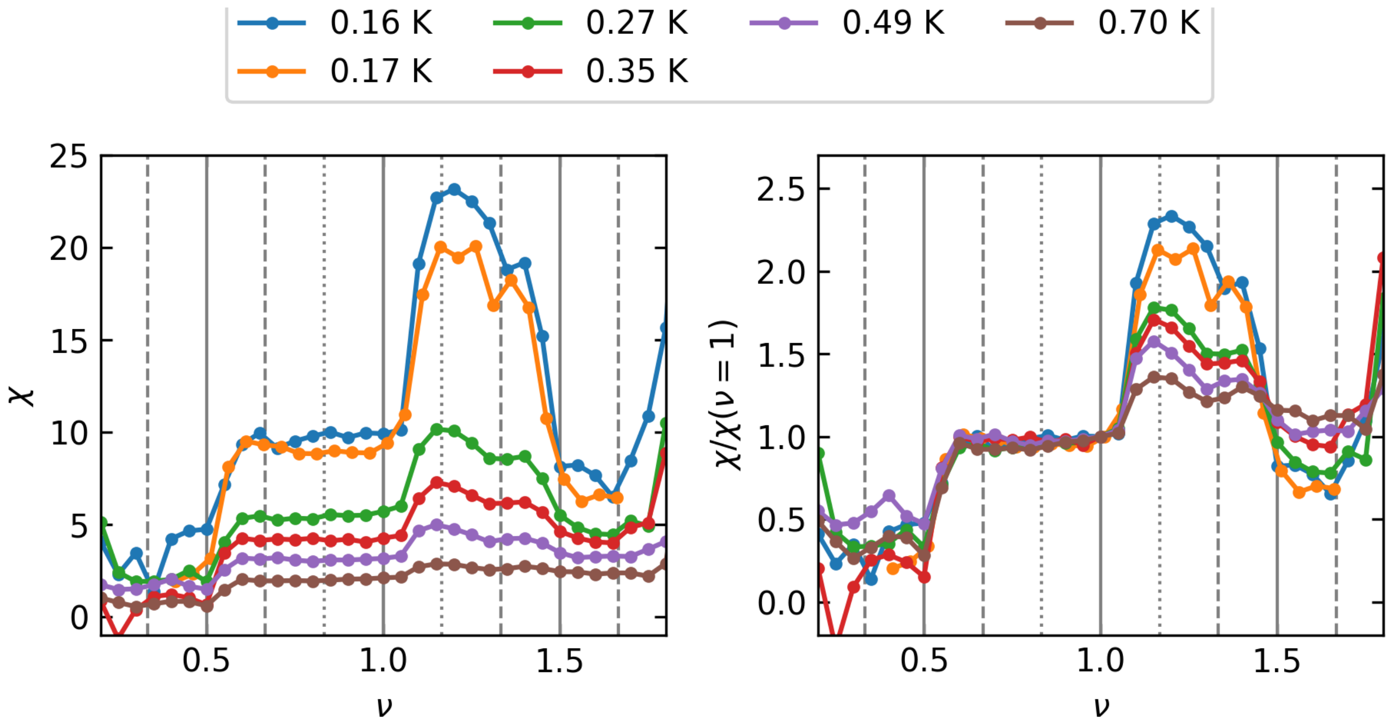


→ Degree of polarization (DOP) as a function of B identical for ν = 0.8 and ν = 1 <u>but not</u> for ν = 1.2

Experimental magnetization at T < 500 mK

Enhanced susceptibility for ν > 1 – Nagaoka mechanism

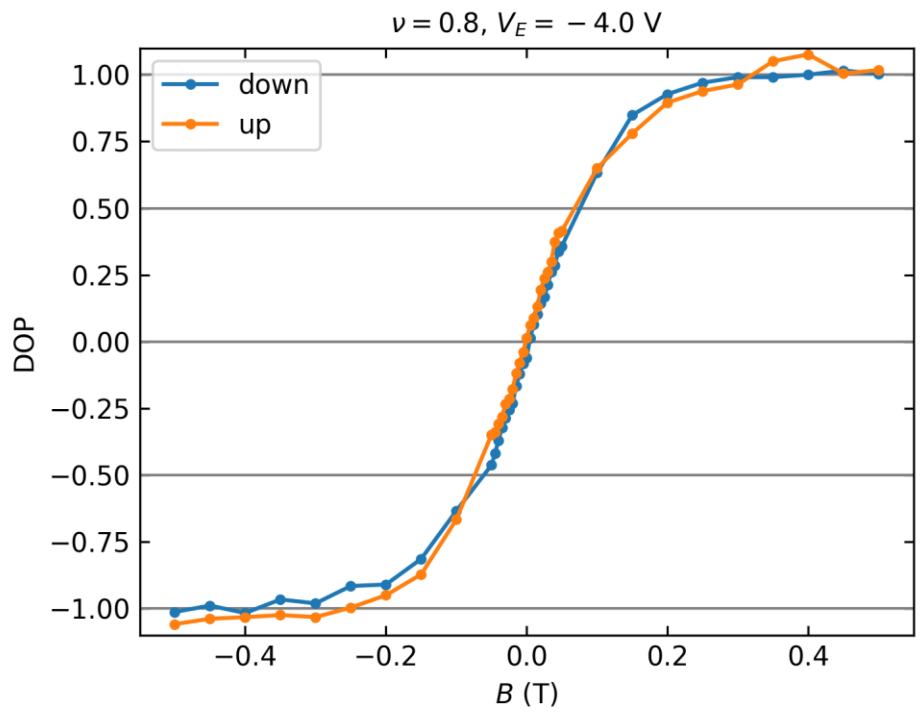




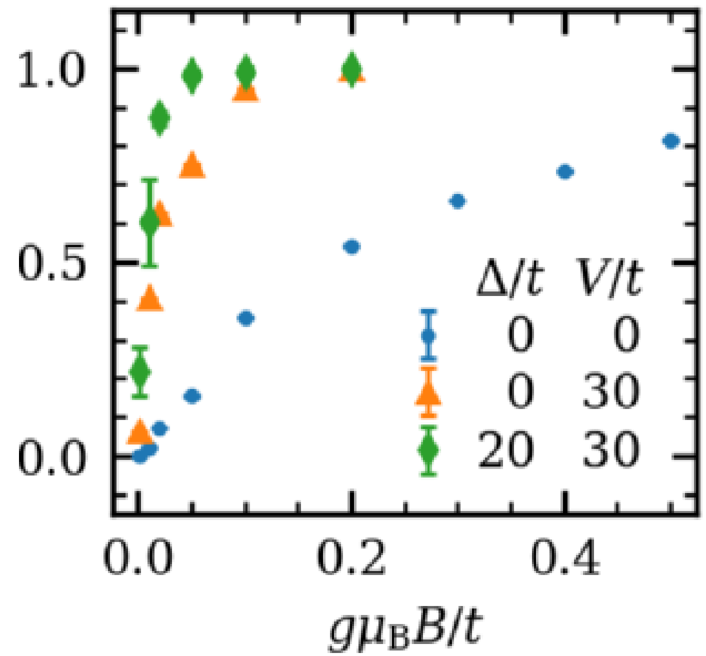
Sharp increase in spin susceptibility χ for ν>1

Experimental magnetization at T < 500 mK

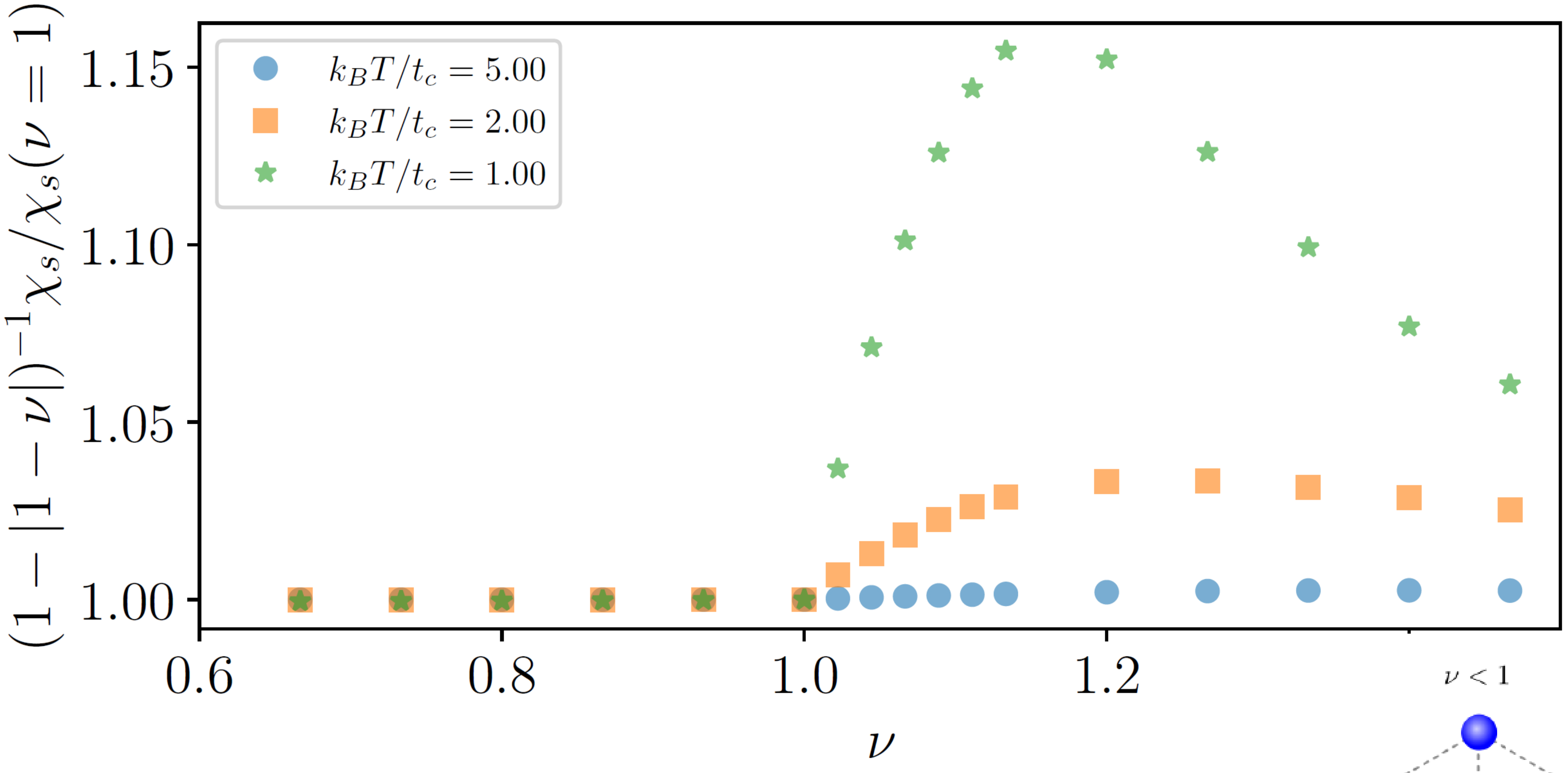
Paramagnetic response for ν ≤ 1 –Are long-range interactions the culprit?

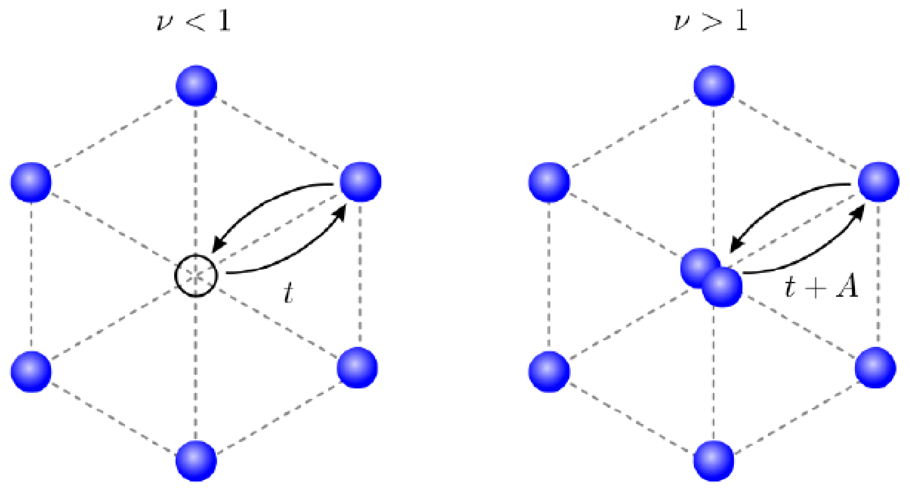


DMRG calculation of magnetization with NN interactions (V) and disorder (Δ)



DMRG with Coulomb assisted hopping

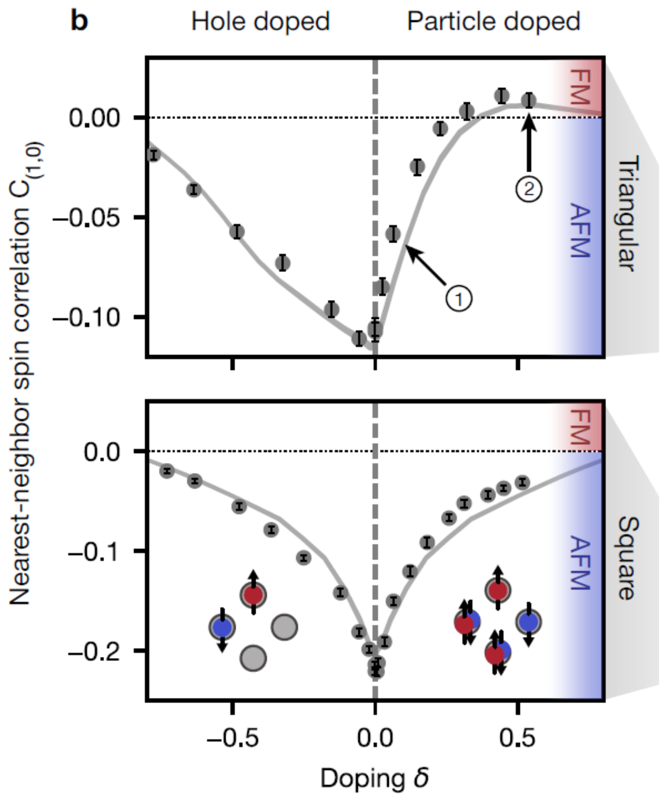




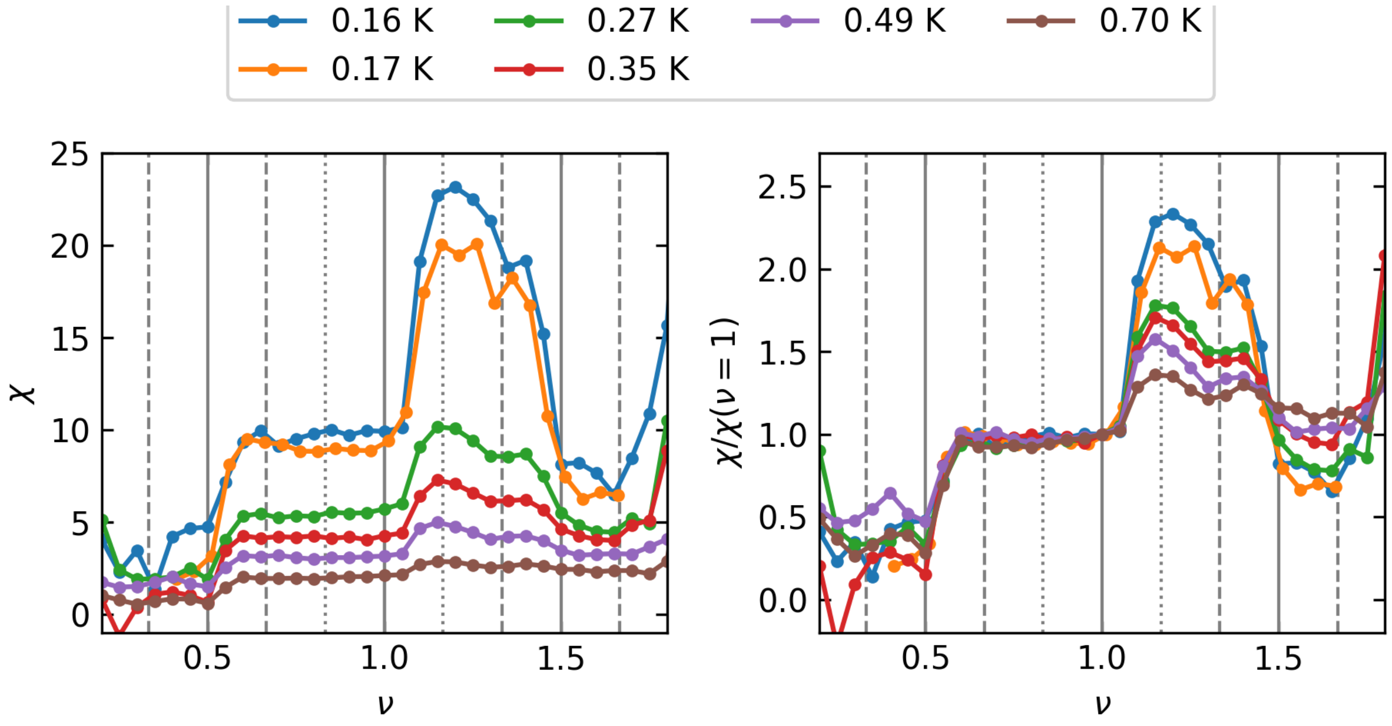
Long-range Coulomb interactions suppress AF correlations while enhancing FM through Coulomb assisted hopping

Quantum simulators of the Fermi-Hubbard model

- Kinetic magnetism in a triangular&square optical lattice of ultracold fermions (Greiner group)



- Kinetic magnetism in triangular MoSe2/WS2 moire heterostructure



- Strong AF correlations at ν=1
- FM (AF) correlations for ν >1 (ν <1)
- Fully consistent with theoretical predictions

- Paramagnetic spin susceptibility for ν ≤ 1
- Nagaoka FM only for ν >1
- Long range Coulomb + disorder suppresses AF for ν <1

Summary and outlook

- Evidence for magnetism that originates from itinerant electrons minimizing kinetic energy – in stark contrast to usual magnetism which stem from electron-electron interactions.

- Future directions:
  - Chiral spin liquids using layer pseudo-spin
  - Kondo lattice