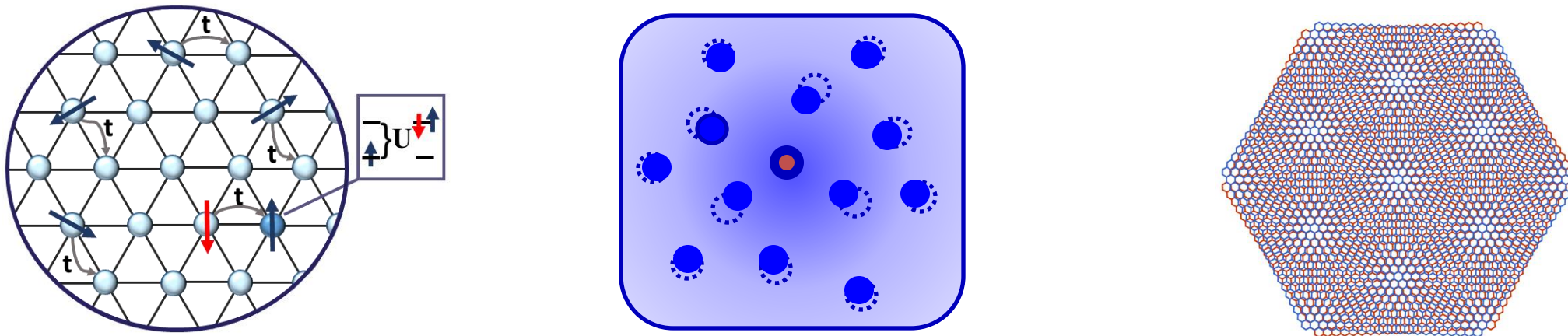


The interplay of field-tunable strongly correlated states in a multi-orbital moiré system



Mauro Brotons-Gisbert

*Institute of Photonics and Quantum Sciences, SUPA
Heriot-Watt University, Edinburgh, UK*

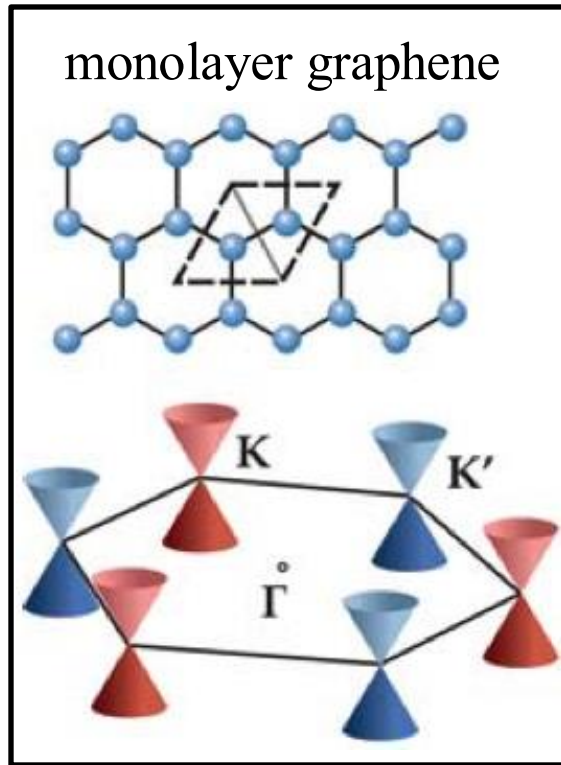
Young Research Leaders Group Workshop
Ingelheim, July 2025

Outline

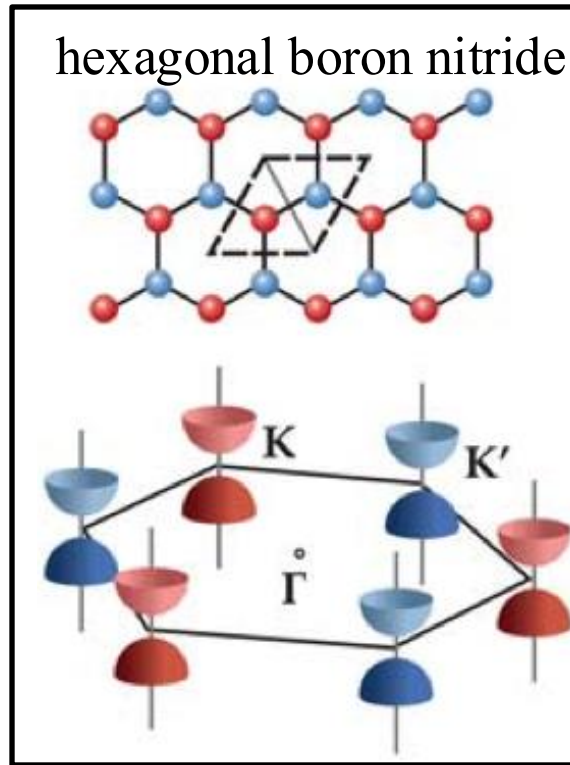
1. Intro: *moiré lattices* and *polaron spectroscopy*
2. Strongly correlated electronic states in moiré materials:
 - $\text{MoSe}_2/\text{WSe}_2$
 - $\text{MoSe}_2/$ Bilayer WSe_2 (multi-orbital moiré system)
3. Wigner crystallisation in monolayer WSe_2 & $\text{MoSe}_2/\text{WSe}_2$

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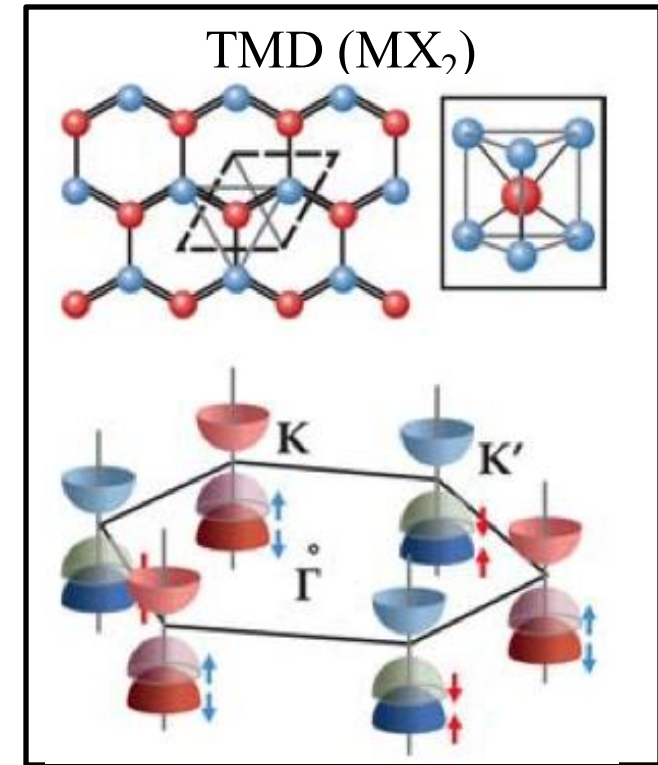
van der Waals crystals



- Dirac fermion
- No gap (symmetry protected)
- Extremely high mobility
- Excellent electric contact



- Insulator
- Good dielectric
- Excellent vdW substrate

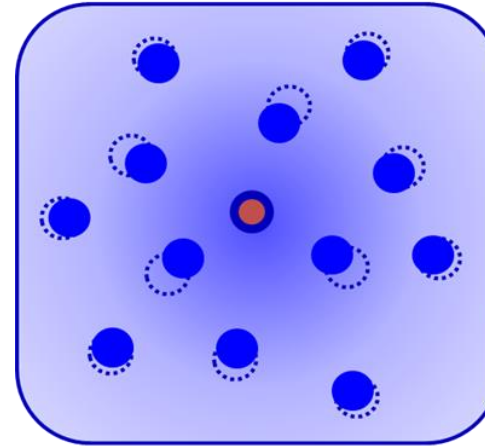


- Semiconductor
- Broken inversion symmetry
- Direct band-gap
- Strong Coulomb interactions
- Strong spin-orbit coupling
- Spin and valley locking

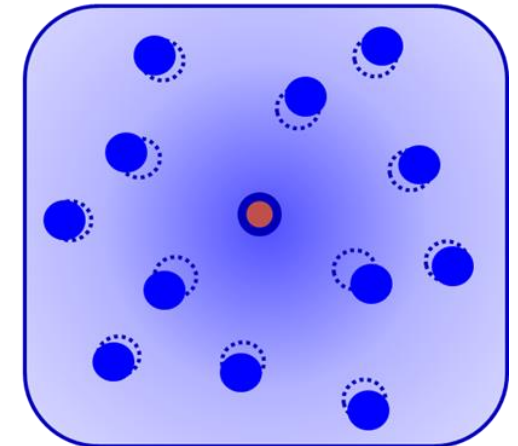
Excitons dressed by Fermi sea: Fermi - polarons

Exciton-electron interactions: Fermi-polarons

Attractive

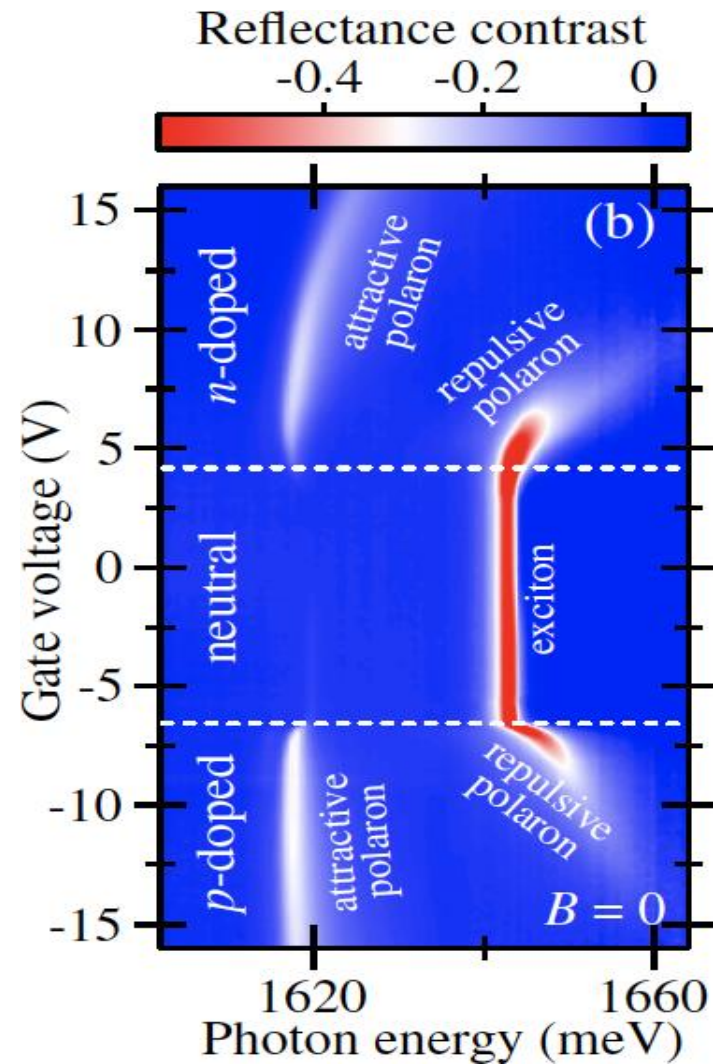


Repulsive



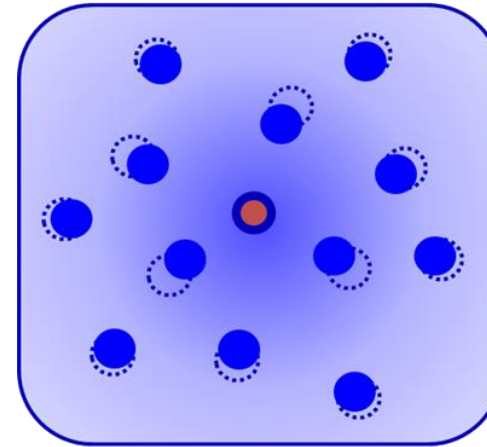
Excitons dressed by Fermi sea: Fermi - polarons

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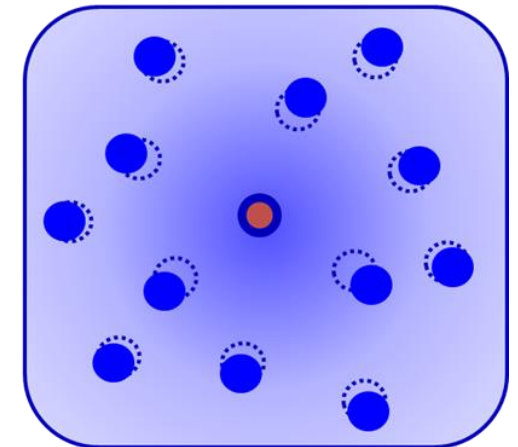


Smolenski et al., Phys. Rev. Lett. **123**, 097403 (2019)].

Attractive



Repulsive



Theory:

Suris *et al.*, Phys. Stat. Sol. (b) **227**, 343 (2001)

Chevy, Phys. Rev. A **74**, 063628 (2006)

Efimkin, MacDonald, Phys. Rev. B **95**, 035417 (2017)

Fey *et al.*, Phys. Rev. B **101**, 195417 (2020)

Glazov, J. Chem. Phys. **153**, 034703 (2020)

Experiment:

Sidler *et al.*, Nat. Phys. **13**, 255 (2017)

Back *et al.*, Phys. Rev. Lett. **118**, 237404 (2017)

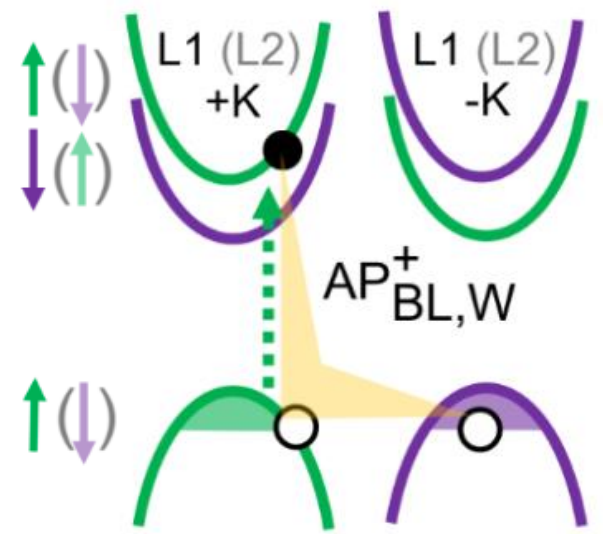
Roch *et al.*, Nat. Nano. **14**, 432 (2019);

Wang *et al.*, Phys. Rev. X **10**, 021024 (2020)

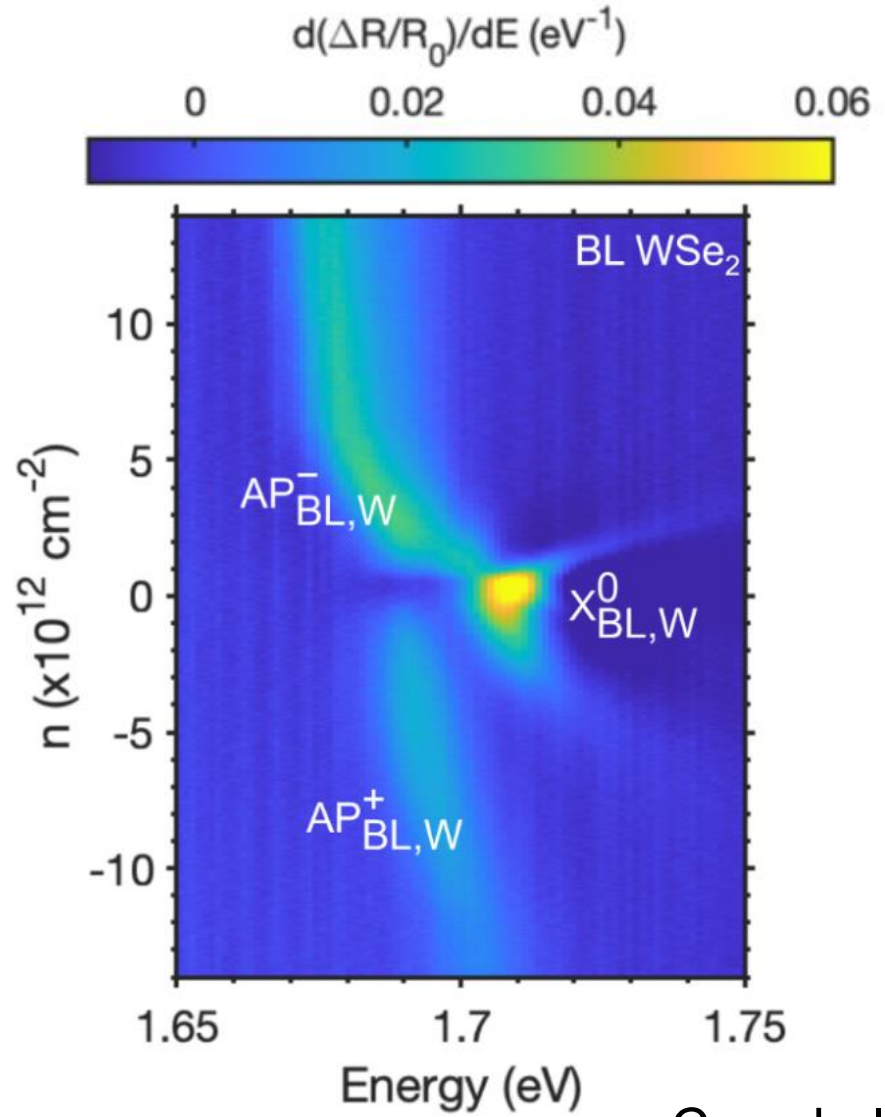
Fermi – polaron spectroscopy

Doping in $\pm K$

- Blue-shift of attractive polaron with increasing carrier concentration (**phase space filling**)
- Strong spin polarization of the exciton polarons

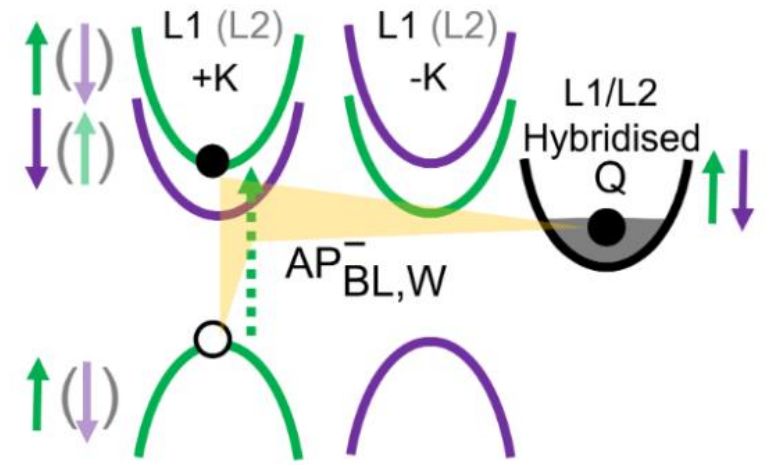


Natural 2H bilayer WSe₂



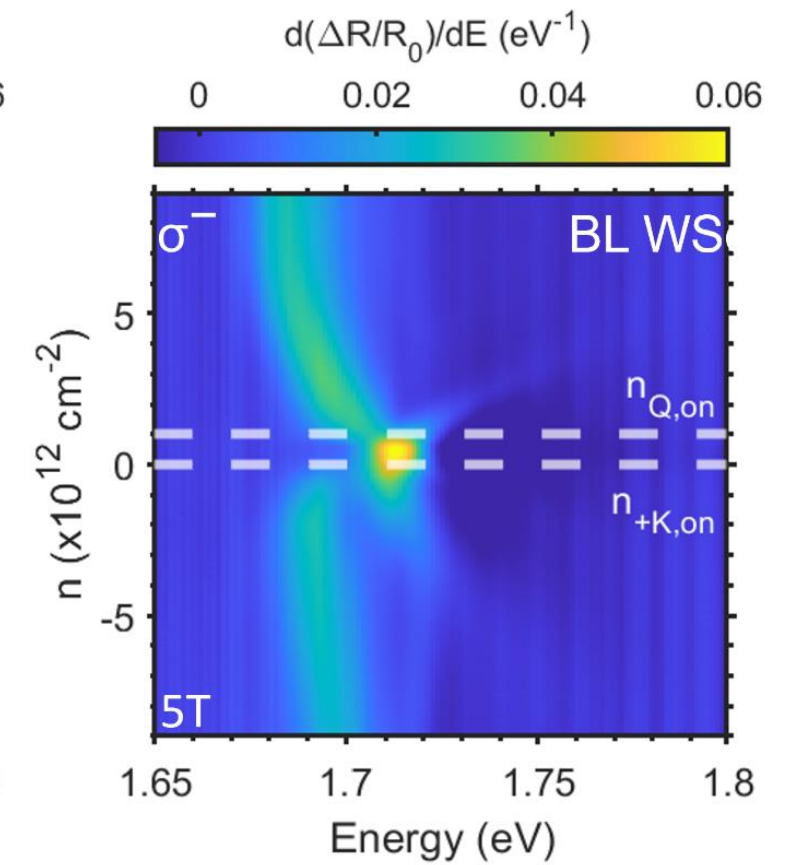
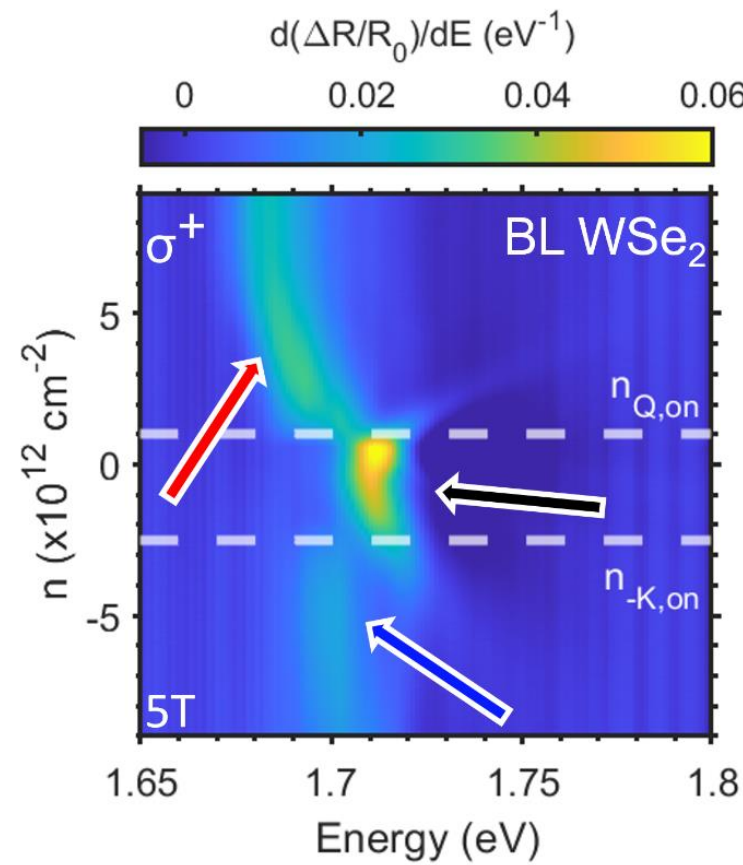
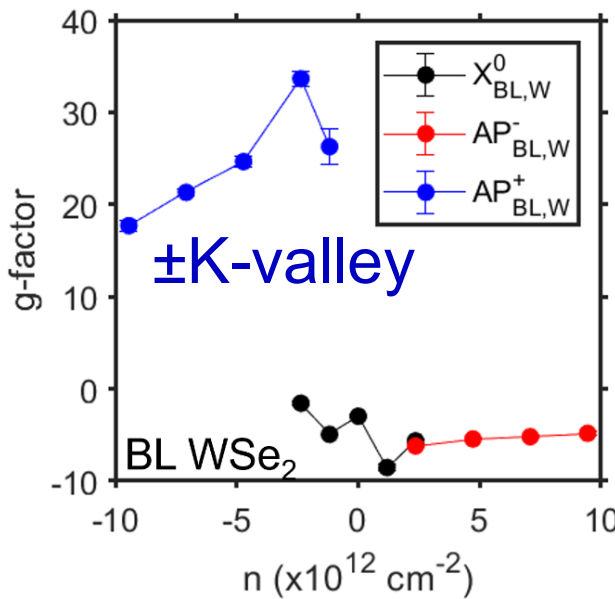
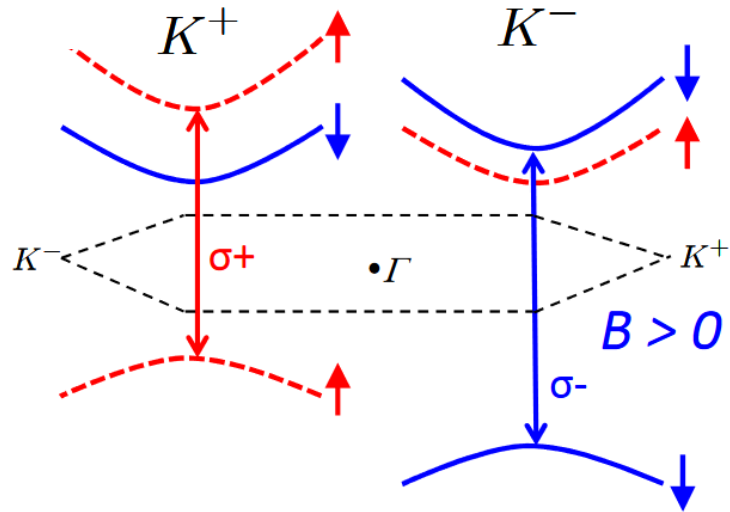
Doping in Q/ Γ

- Red-shift of attractive polaron with increasing carrier concentration (**no phase space filling**)
- Unpolarised exciton polarons

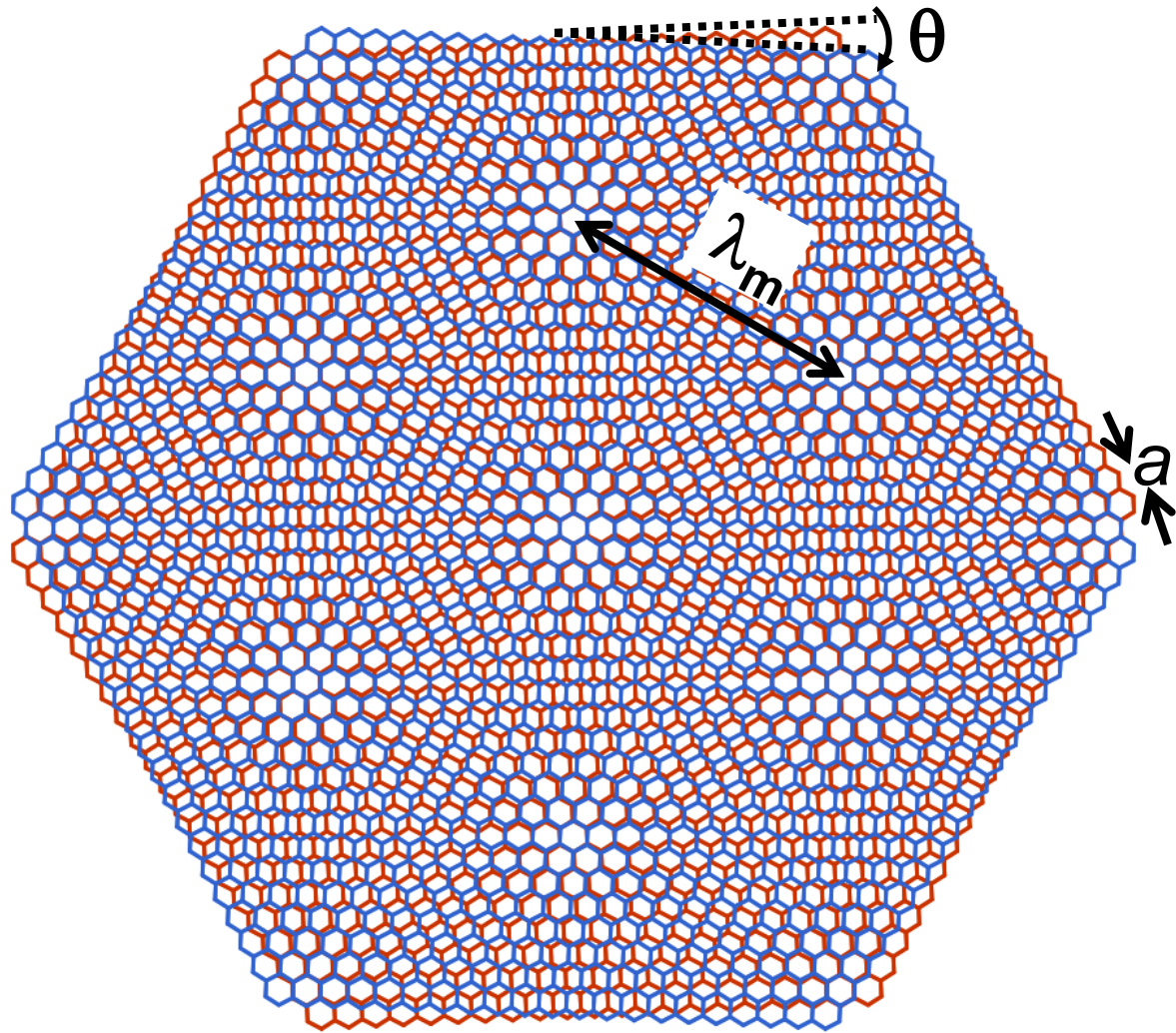


Fermi – polaron spectroscopy

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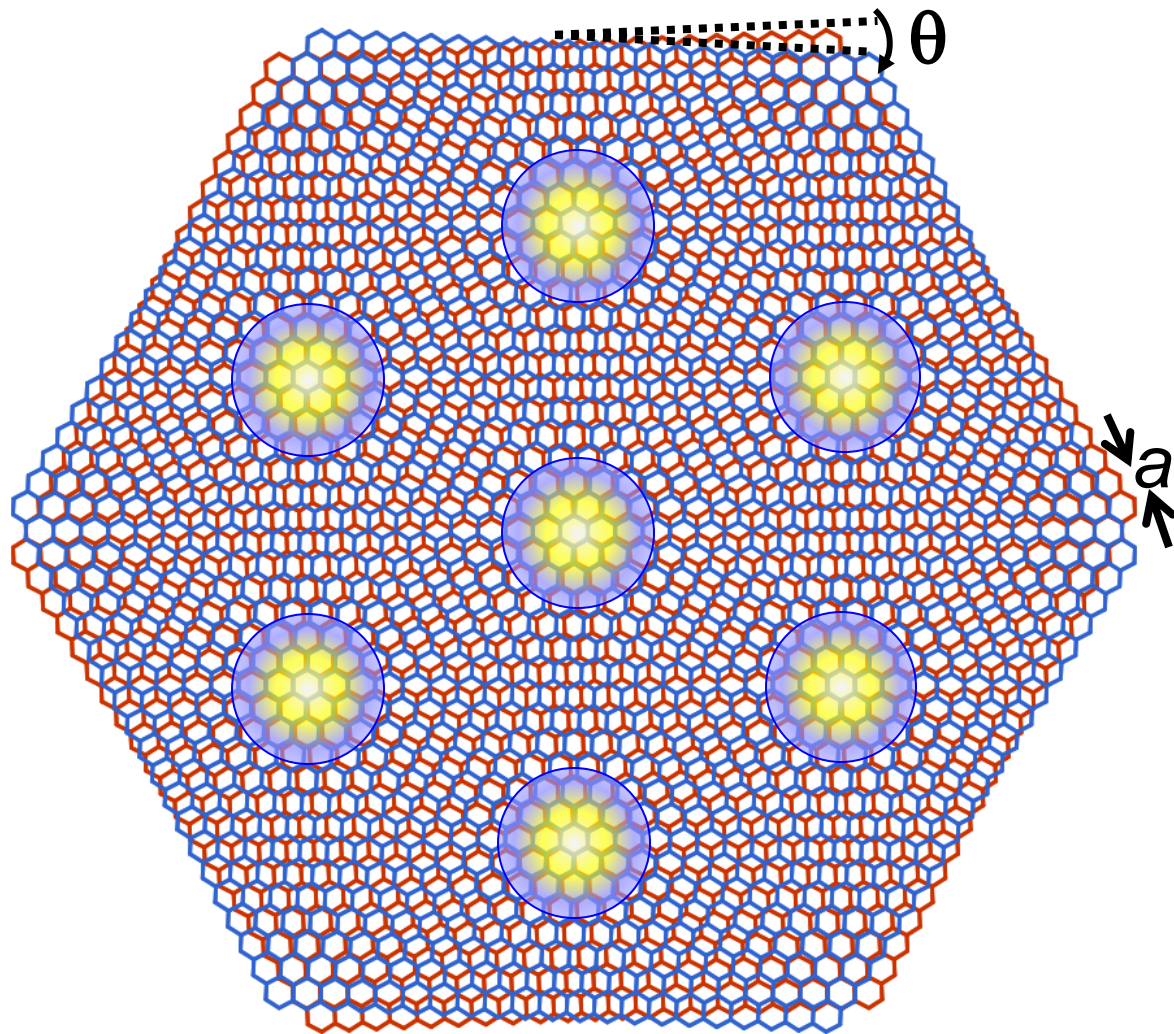


2D moiré superlattices

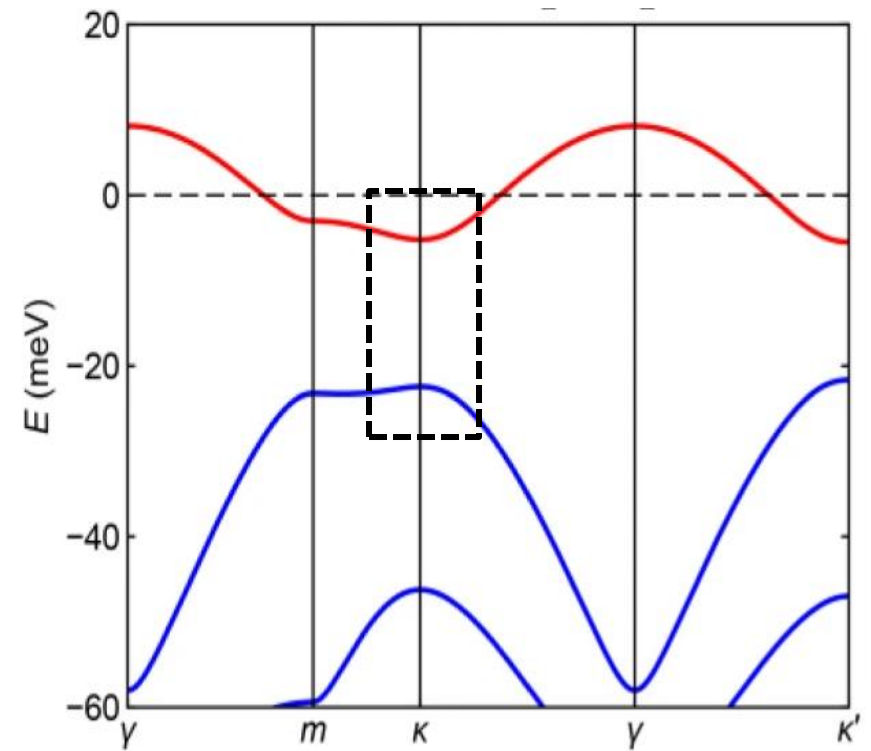


Moiré period: $\lambda_m \approx a/\sqrt{\delta\theta^2 + \delta^2}$

2D moiré superlattices



Flat moiré mini-band



- Carrier kinetic energy quenched
- Electronic interactions dominate

TMD moiré heterostructures

PHYSICAL REVIEW LETTERS **121**, 026402 (2018)

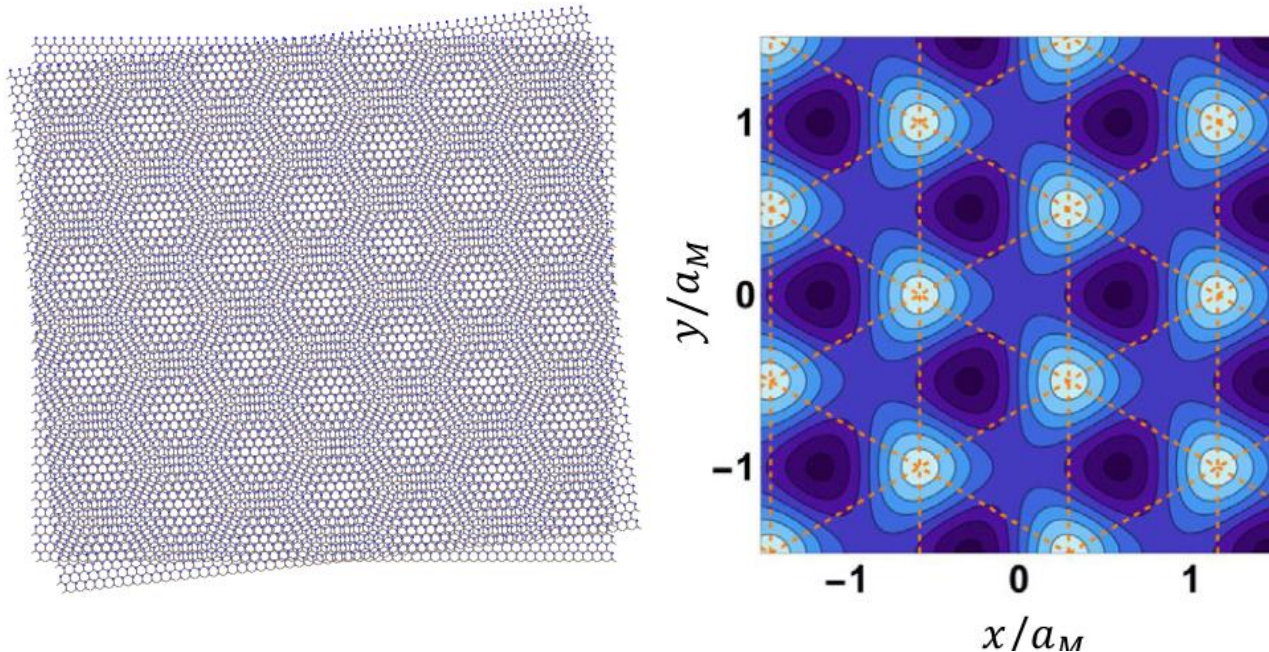
Hubbard Model Physics in Transition Metal Dichalcogenide Moiré Bands

Fengcheng Wu,¹ Timothy Lovorn,² Emanuel Tutuc,³ and A. H. MacDonald²

¹Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA

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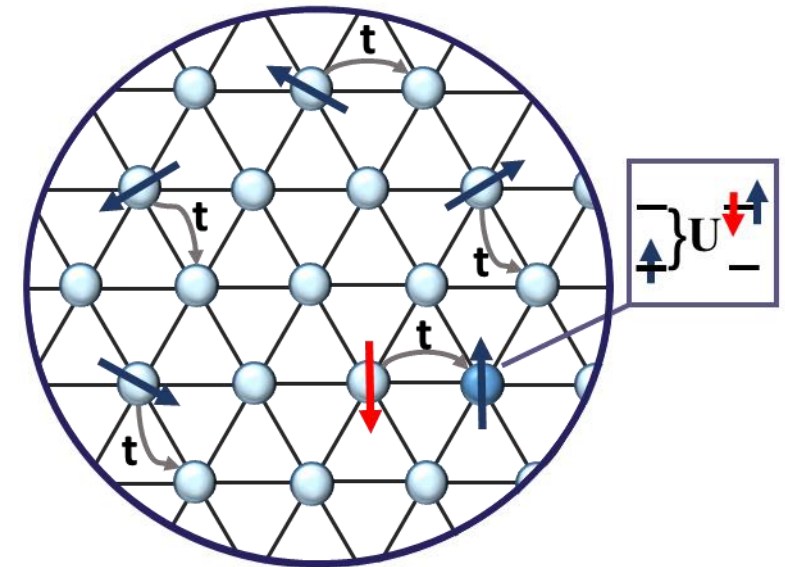


$U/t \gg 1$: strong correlations

Fully tunable triangular Hubbard lattice

- Wide choice of heterobilayer materials
- Precise control of fractional filling (ν)
- No ‘magic-angle’: full twist angle range
- More robust correlation physics

Triangular Hubbard lattice



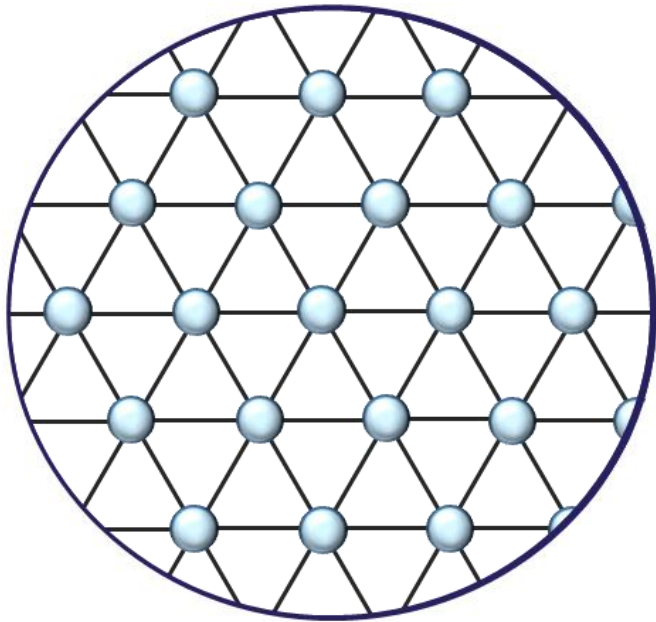
Hopping amplitude: $-t$

On-site Coulomb interaction: U

Strongly correlated electronic states

Electron-electron interactions: Fermi-Hubbard Model

Insulator



$$\nu = 0$$



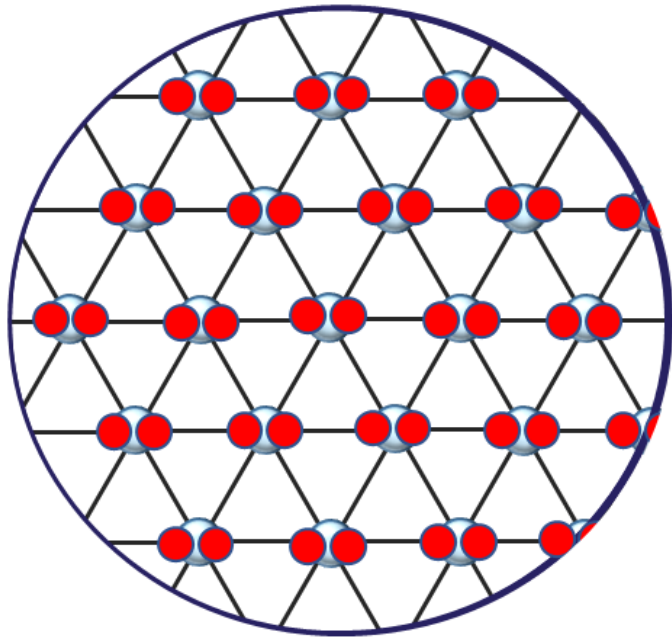
0

electrons per site (ν)

Strongly correlated electronic states

Electron-electron interactions: Fermi-Hubbard Model

Insulator



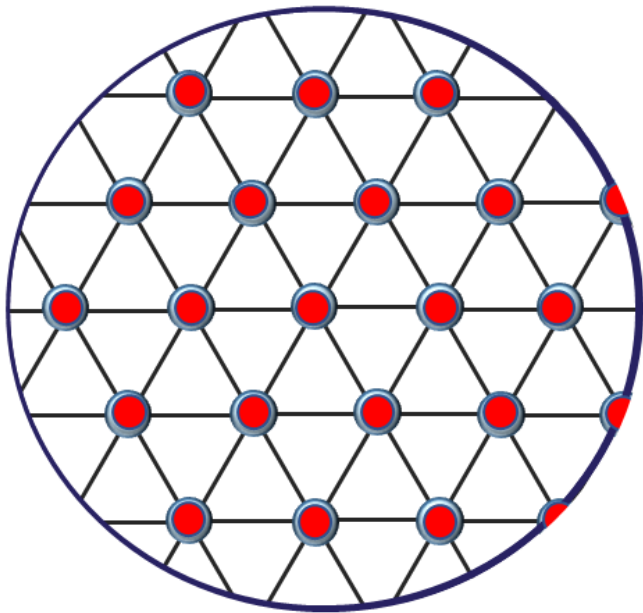
$$\nu = 2$$



electrons per site (ν)

Strongly correlated electronic states

Mott Insulator



$$\nu = 1$$

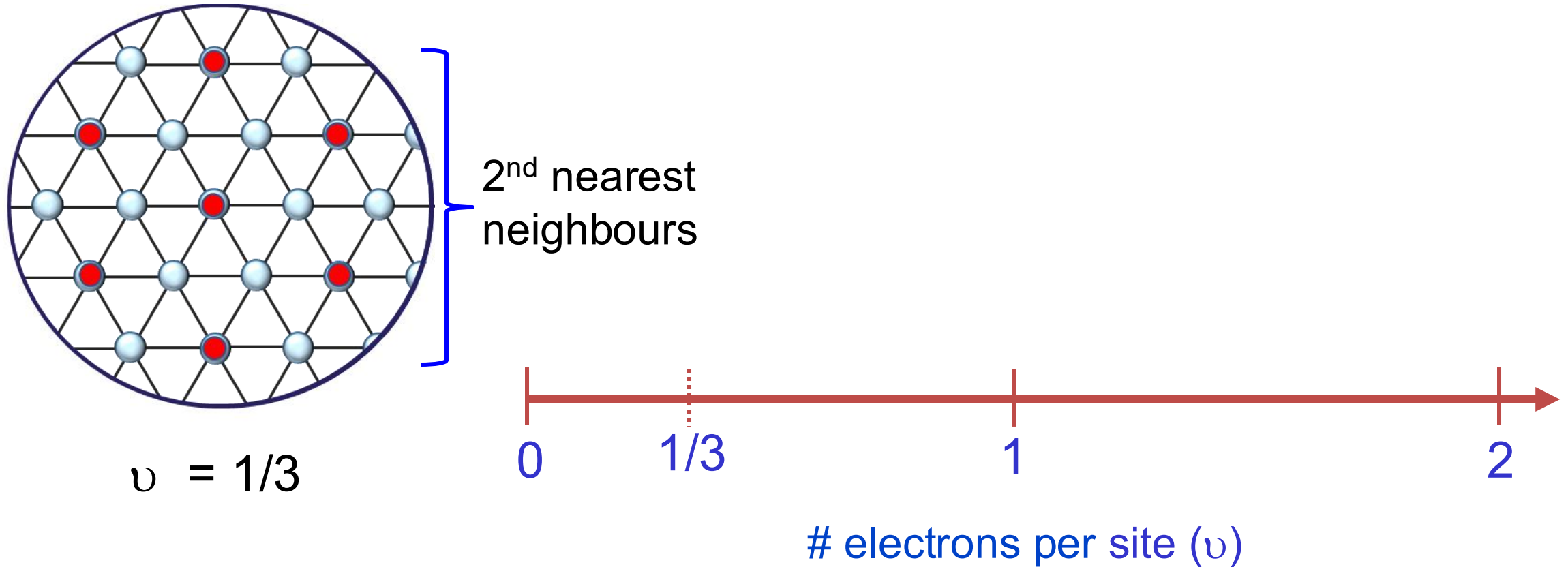


electrons per site (ν)

Strongly correlated electronic states

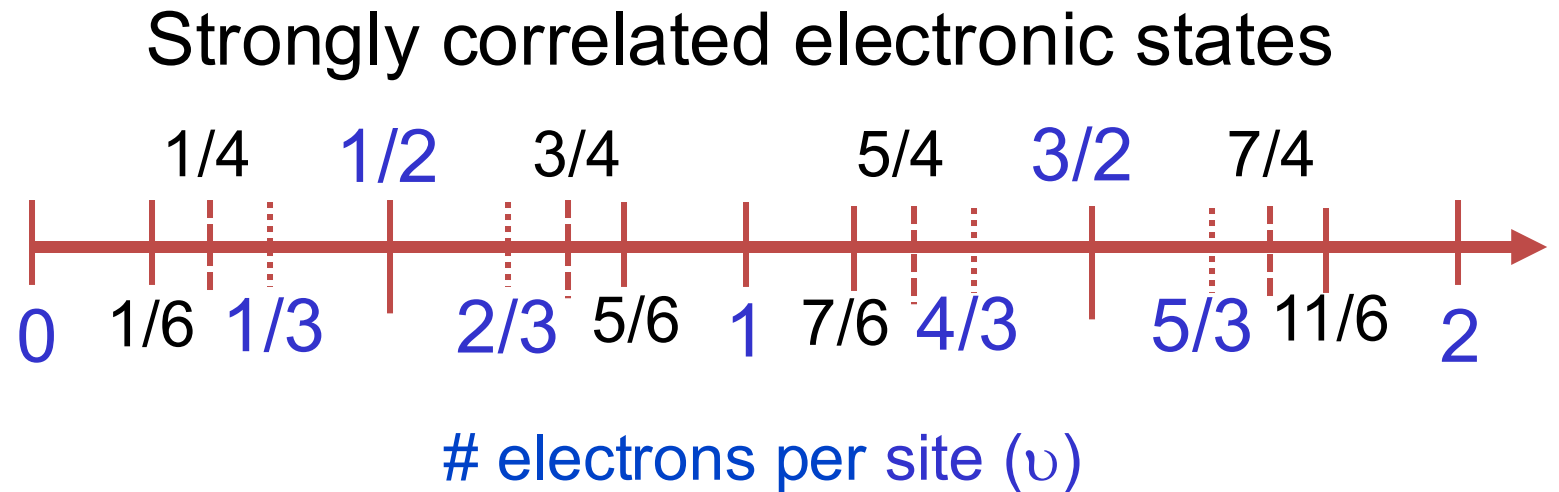
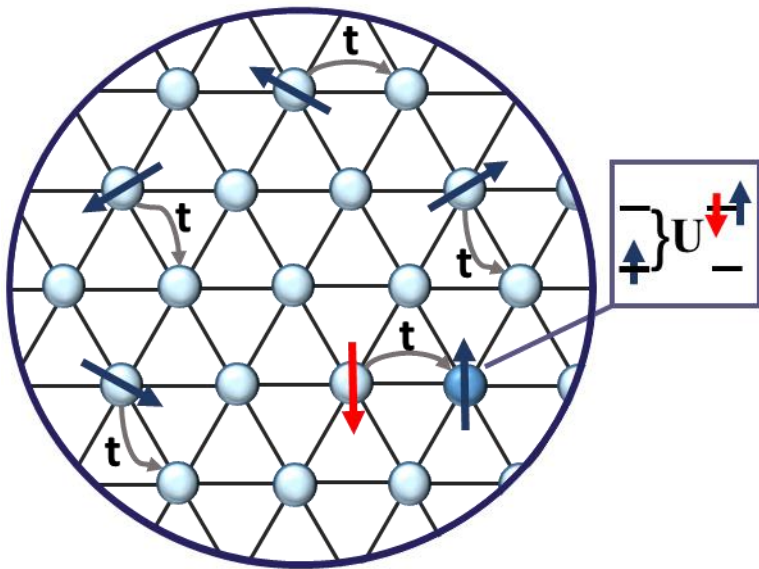
Electron-electron interactions: Fermi-Hubbard Model

Wigner Crystal



Strongly correlated electronic states

Electron-electron interactions: Fermi-Hubbard Model



PHYSICAL REVIEW LETTERS **121**, 026402 (2018)

Hubbard Model Physics in Transition Metal Dichalcogenide Moiré Bands

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Theory:

F. Wu et al. Phys. Rev. Lett. 122, 086402 (2019).

H. Pan et al. Phys. Rev. B. 102, 201104(R) (2020).

Y. Zhang et al. Phys. Rev. B. 102, 201115(R) (2020).

K. Slagel and L. Fu, Phys. Rev. B 102, 235423 (2020).

L. Xian et al. Nature Commun. 12, 5644 (2021).

H. Pan and S. Das Sarma, Phys. Rev. Lett. 127, 096802 (2021).

B. Padhi et al. Phys. Rev. B 103, 125146 (2021).

Y. Zhang et al. Phys. Rev. B 103, 155142 (2021).

N. C. Hu and A. MacDonald. Phys. Rev. B 104, 214403 (2021).

TMD moiré heterostructures

PHYSICAL REVIEW LETTERS **121**, 026402 (2018)

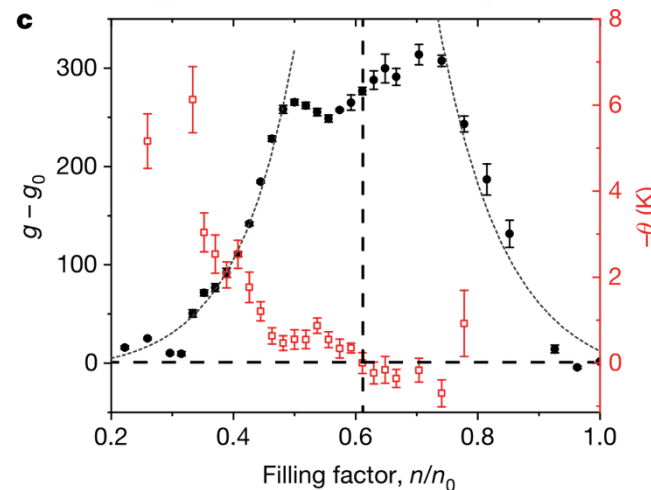
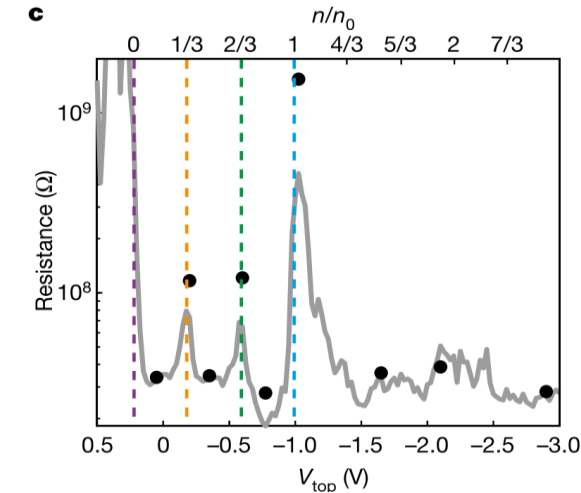
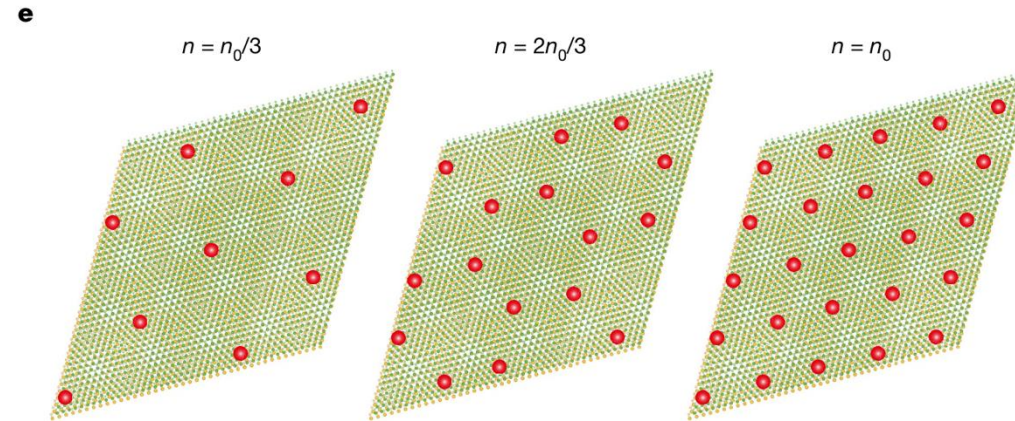
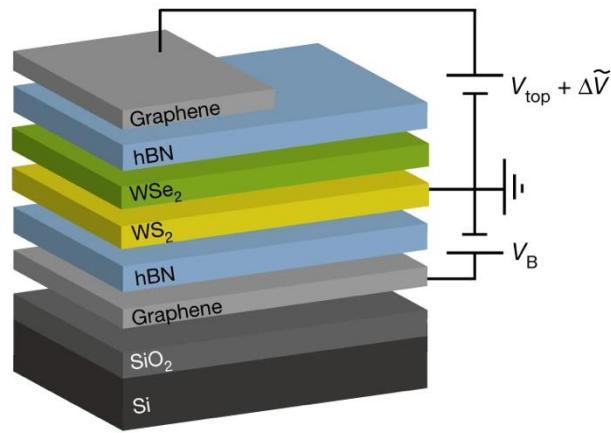
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Angle aligned WS₂ / WSe₂



Magnetic susceptibility $\chi \sim g - g_0$
 Weiss constant θ

- Regan *et al.*, Nature **579**, 359 (2020)
 Tang *et al.*, Nature **579**, 353 (2020)
 Shimazaki *et al.*, Nature **580**, 472 (2020)
 Xu *et al.*, Nature **587**, 214 (2020)
 Huang *et al.*, Nature Physics **17**, 715 (2021)
 Ghiotto *et al.*, Nature **597**, 345 (2021)

PHYSICAL REVIEW LETTERS **121**, 026402 (2018)

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- **WSe₂/WS₂** [1-4]
- **WSe₂/WSe₂** [5, 6]
- **MoSe₂/MoSe₂** [7]
- **MoSe₂/WS₂** [8, 9]
- **WSe₂/MoTe₂** [10, 11]
- **MoTe₂/MoTe₂** [12]
- **WSe₂/MoSe₂** [13]

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[1] Regan *et al.*, Nature **579**, 359 (2020)

[2] Tang *et al.*, Nature **579**, 353 (2020)

[3] Xu *et al.*, Nature **587**, 214 (2020)

[4] Huang *et al.*, Nature Physics **17**, 715 (2021)

[5] Wang *et al.*, Nature Materials **19**, 861 (2020)

[6] Ghiotto *et al.*, Nature **597**, 345 (2021)

[7] Shimazaki *et al.*, Nature **580**, 472 (2020)

[8] Li *et al.*, Nature Nanotechnology **16**, 1068–1072 (2021)

[9] Ciorciaro *et al.*, arXiv:2305.02150 (2023)

[10] Li *et al.*, Nature **597**, 350 (2021)

[11] Li *et al.*, Nature **600**, 641 (2021)

[12] Anderson *et al.*, Science, eadg4268 (2023).

[13] Campbell *et al.* npj 2D Mat. & Appl. **6**, 79 (2022)

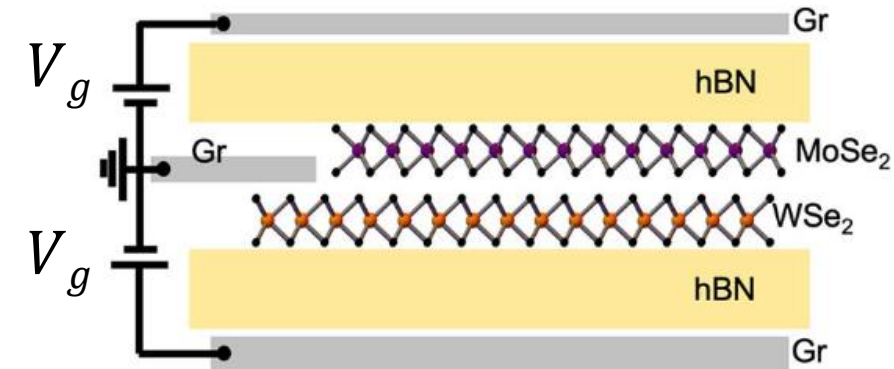
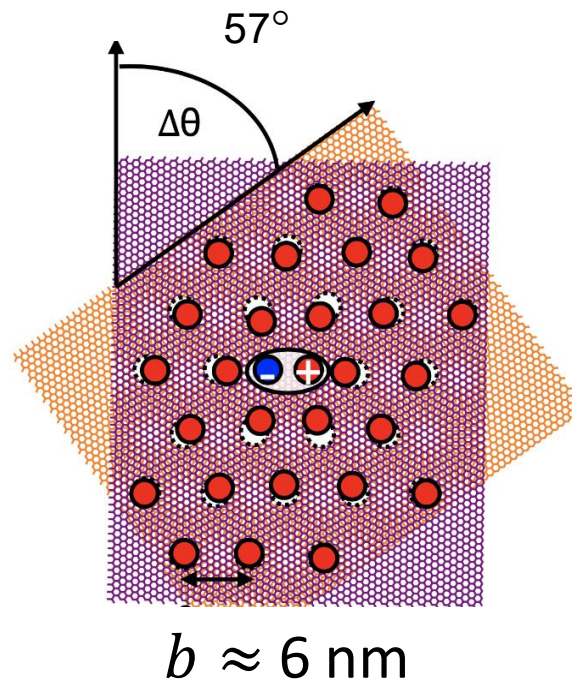
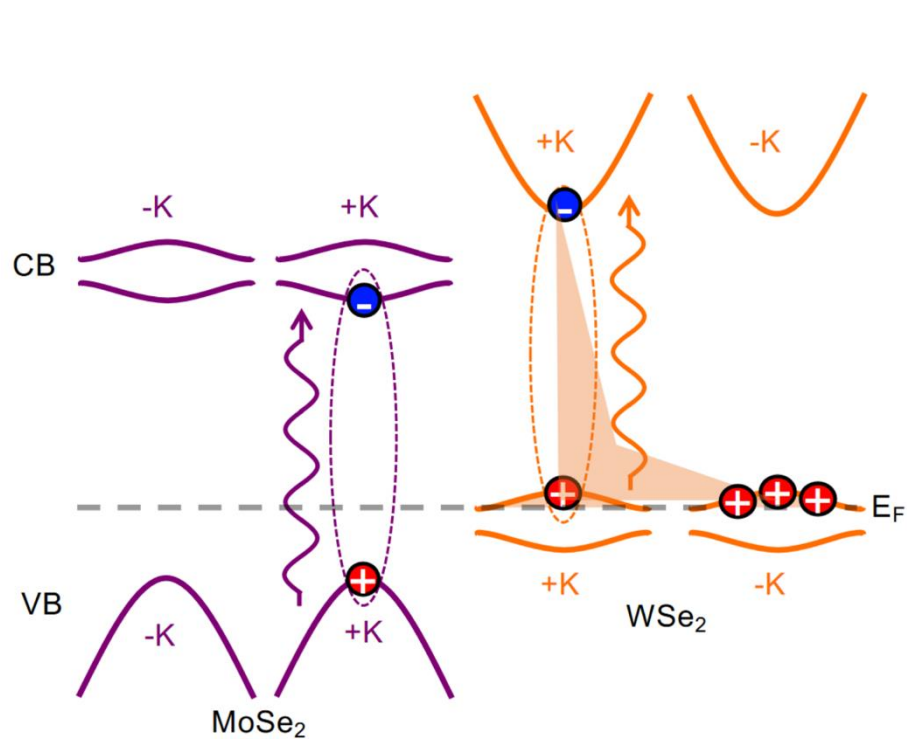
Outline

1. Intro: *moiré lattices* and *polaron spectroscopy*
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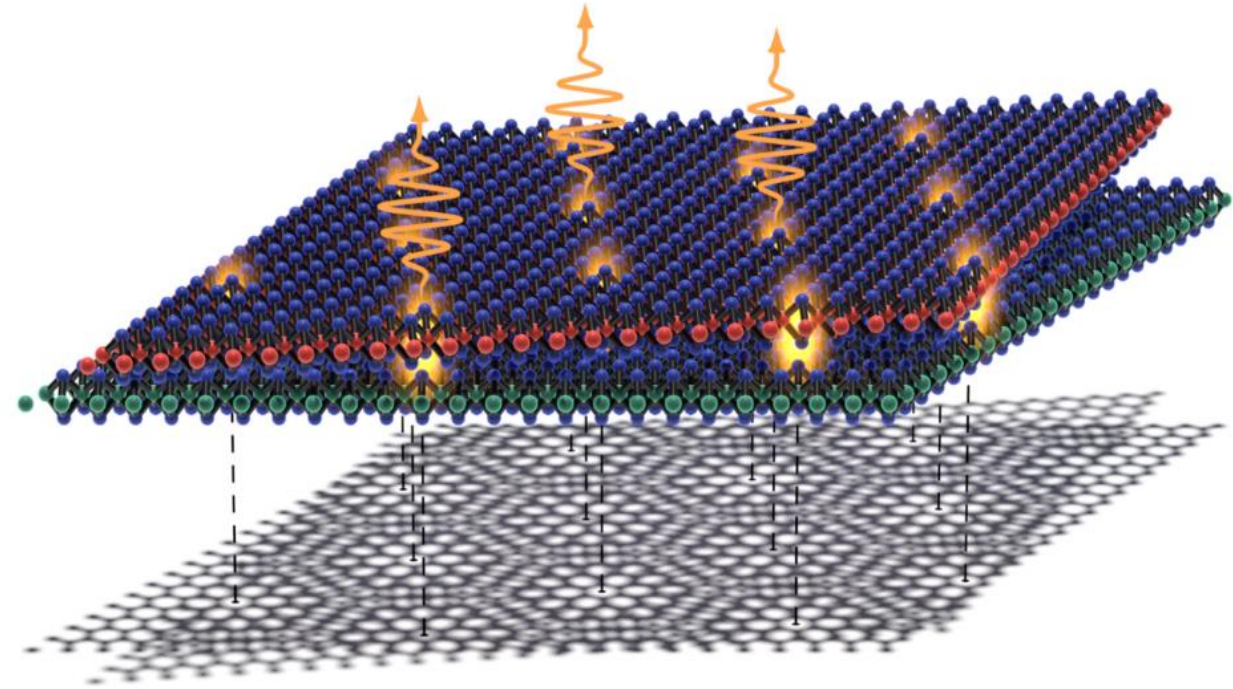
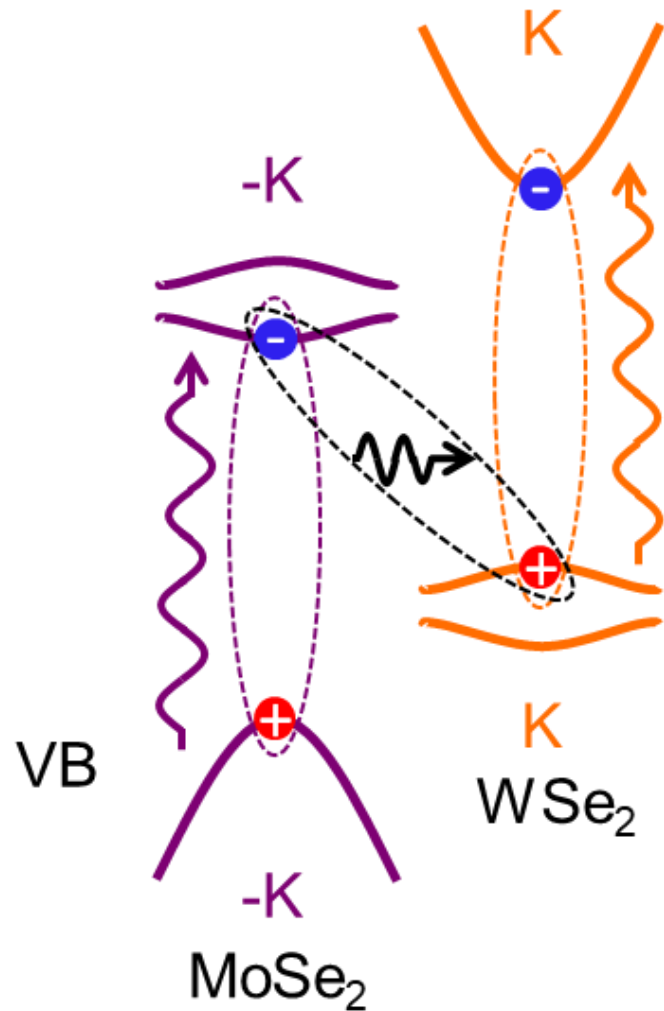
MoSe₂ / WSe₂ strongly correlated states

Hetero-bilayer MoSe₂/WSe₂: ±K-triangular Hubbard system

Type-II band alignment



Moiré-confined interlayer excitons



M. Brotons-Gisbert, H. Baek, *et al.* Nature Materials **19**, 630 (2020)

H. Baek, M. Brotons-Gisbert, *et al.*, Science Advances **6**, eaba8526 (2020)

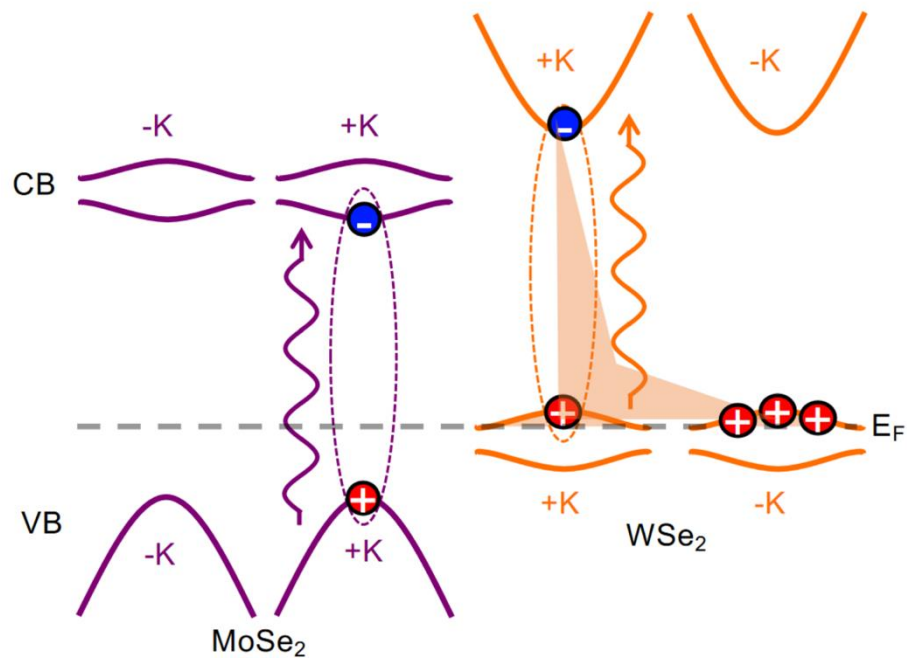
M. Brotons-Gisbert, H. Baek, A. Campbell. *et al.* Phys. Rev. X **11**, 031033 (2021)

H. Baek, M. Brotons-Gisbert, A. Campbell. *et al.* Nature Nanotech. **16**, 1237 (2021)

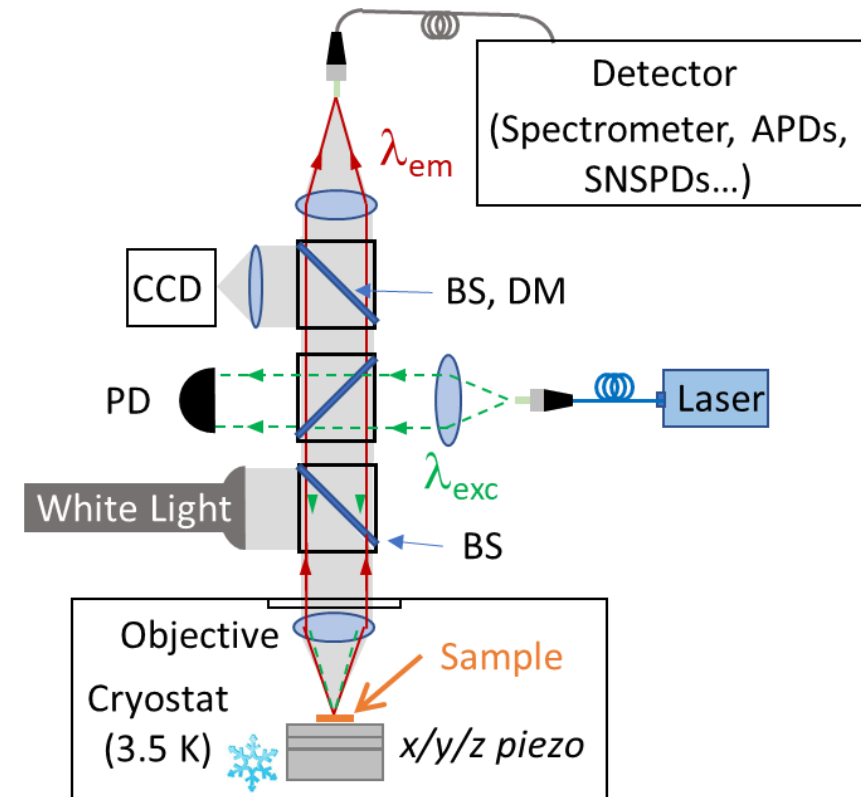
MoSe₂ / WSe₂ strongly correlated states

Hetero-bilayer MoSe₂/WSe₂: $\pm K$ -triangular Hubbard system

Type-II band alignment



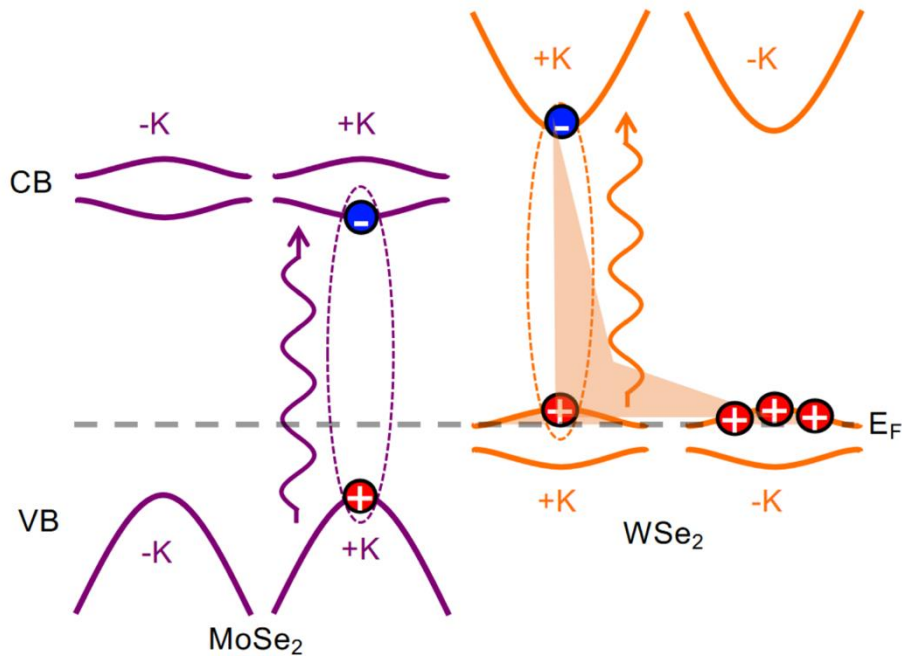
Experimental technique: differential reflectivity



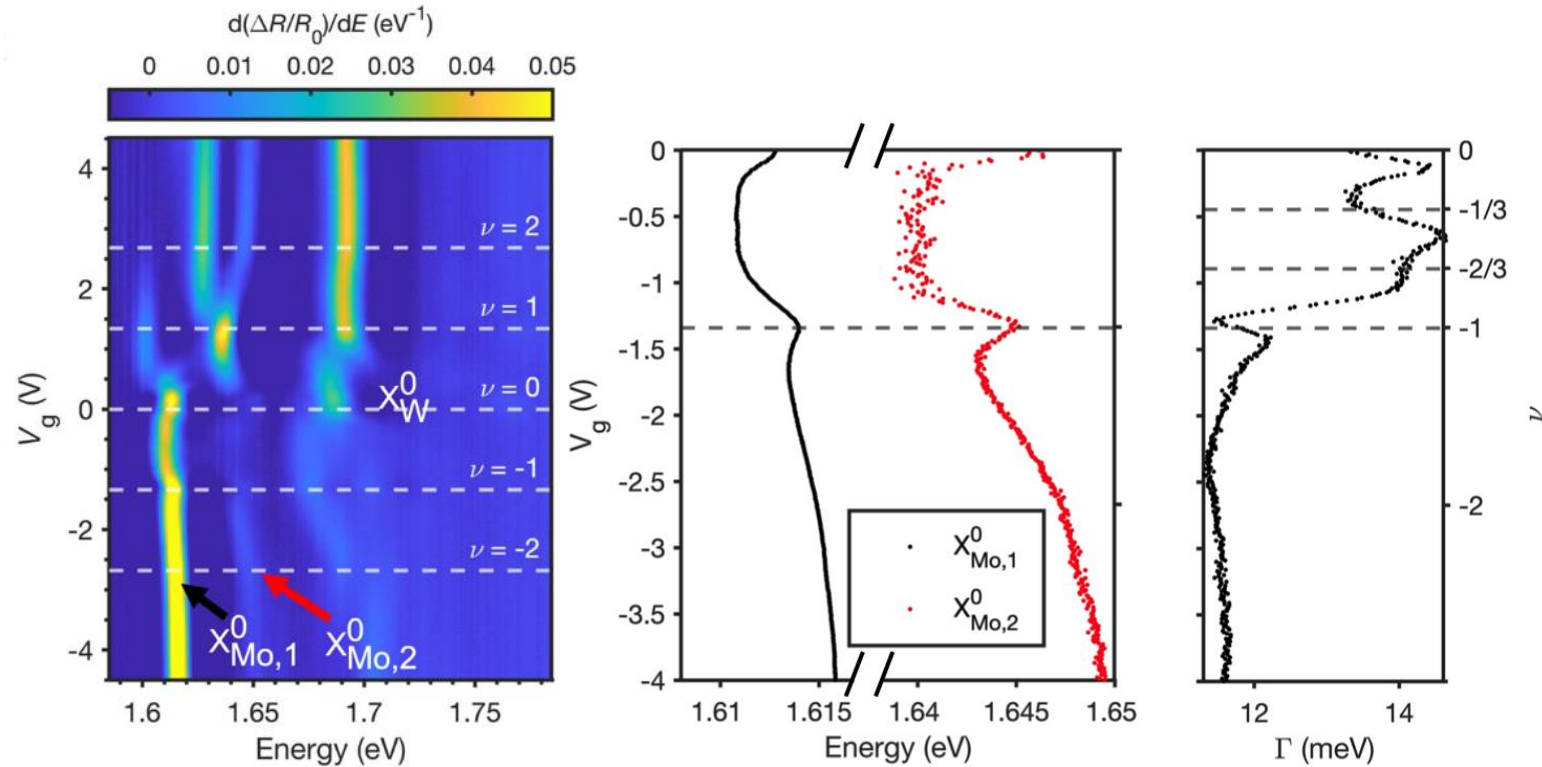
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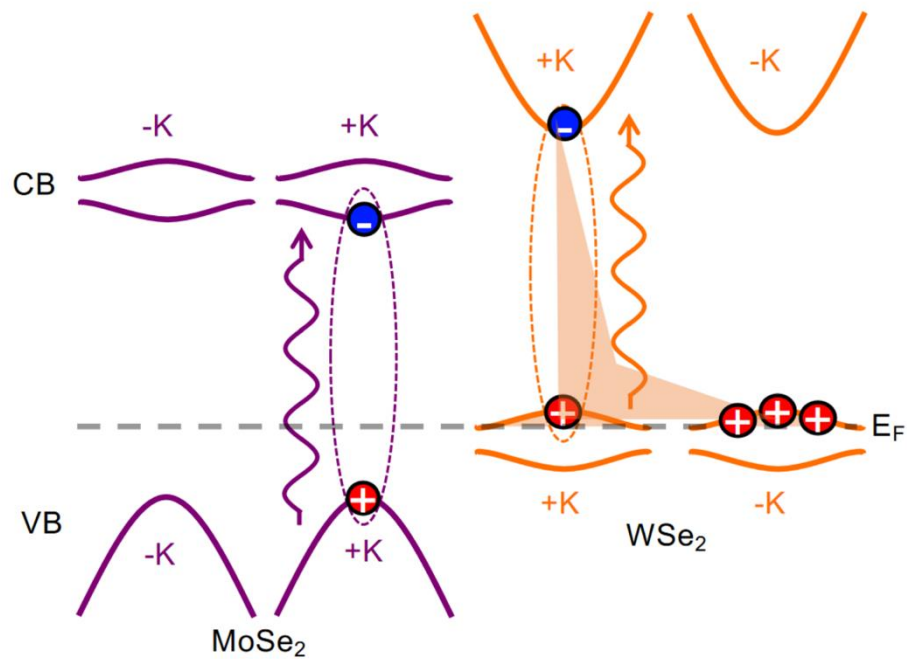
Strongly Correlated States:
Mott insulating and Wigner crystals



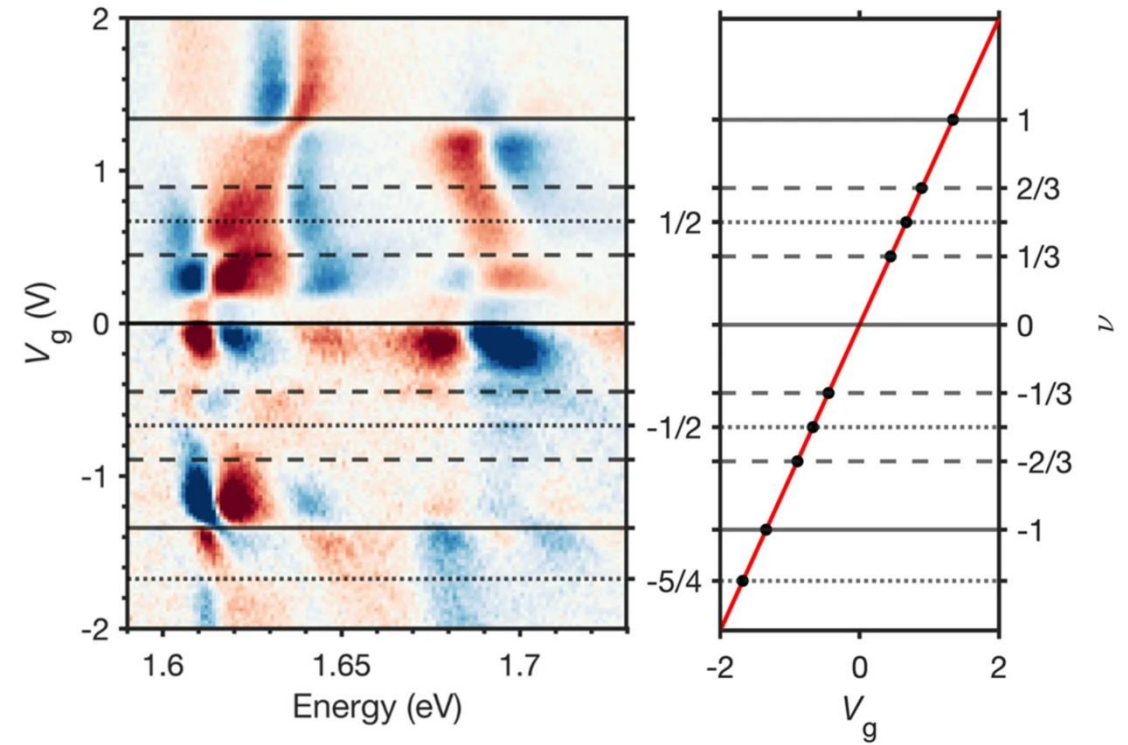
MoSe₂ / WSe₂ strongly correlated states

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Type-II band alignment



Strongly Correlated States:
Mott insulating and Wigner crystals



Magnetic interactions: spin Heisenberg model

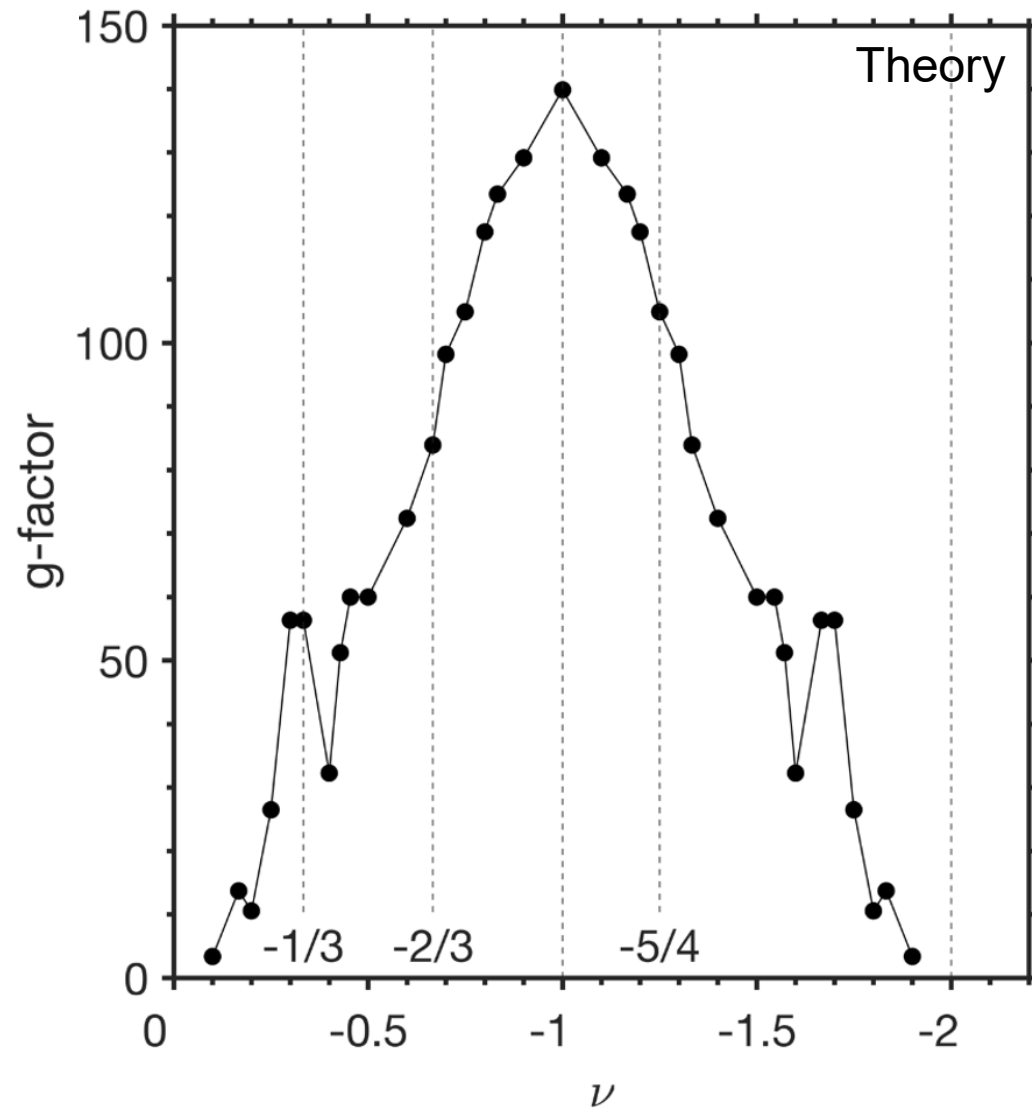
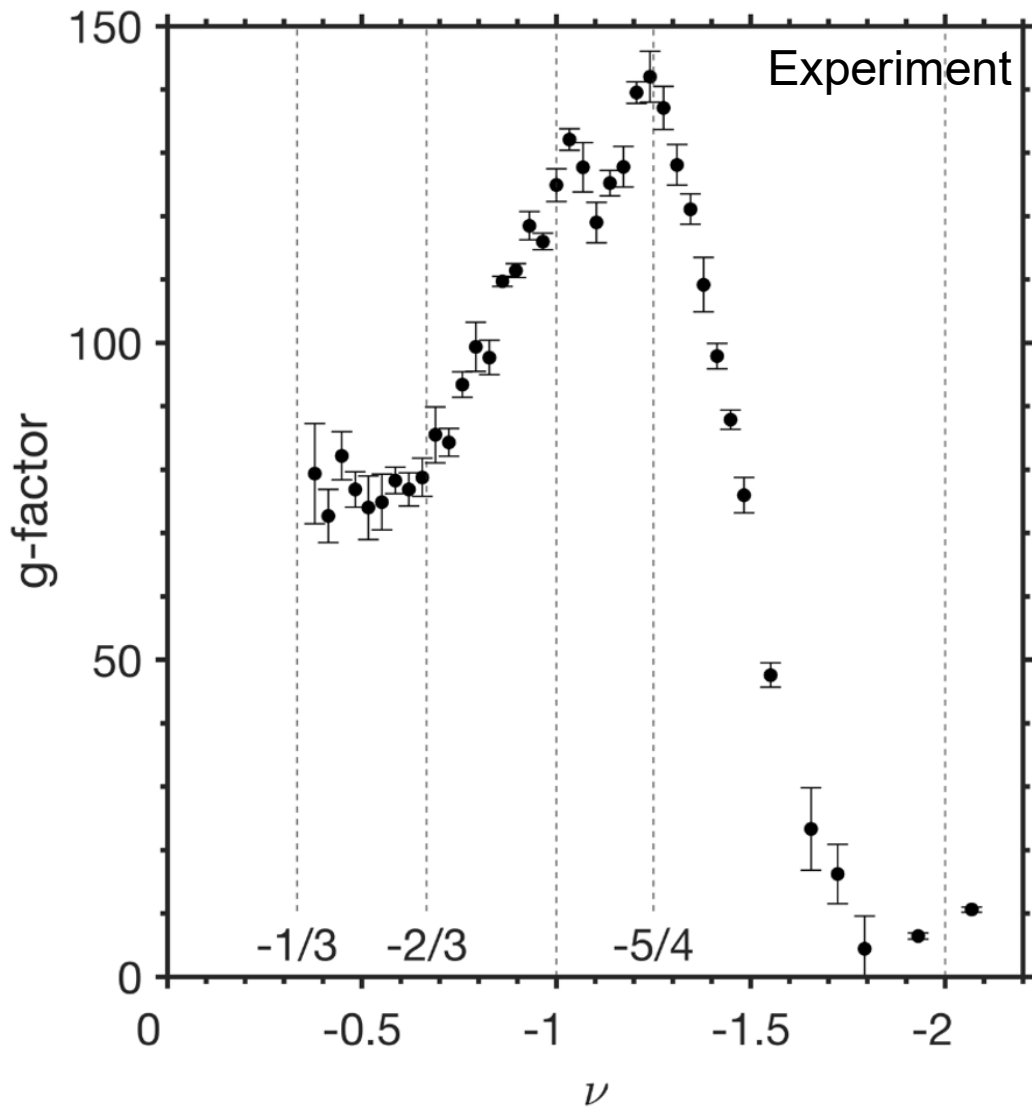
Imperial College
London



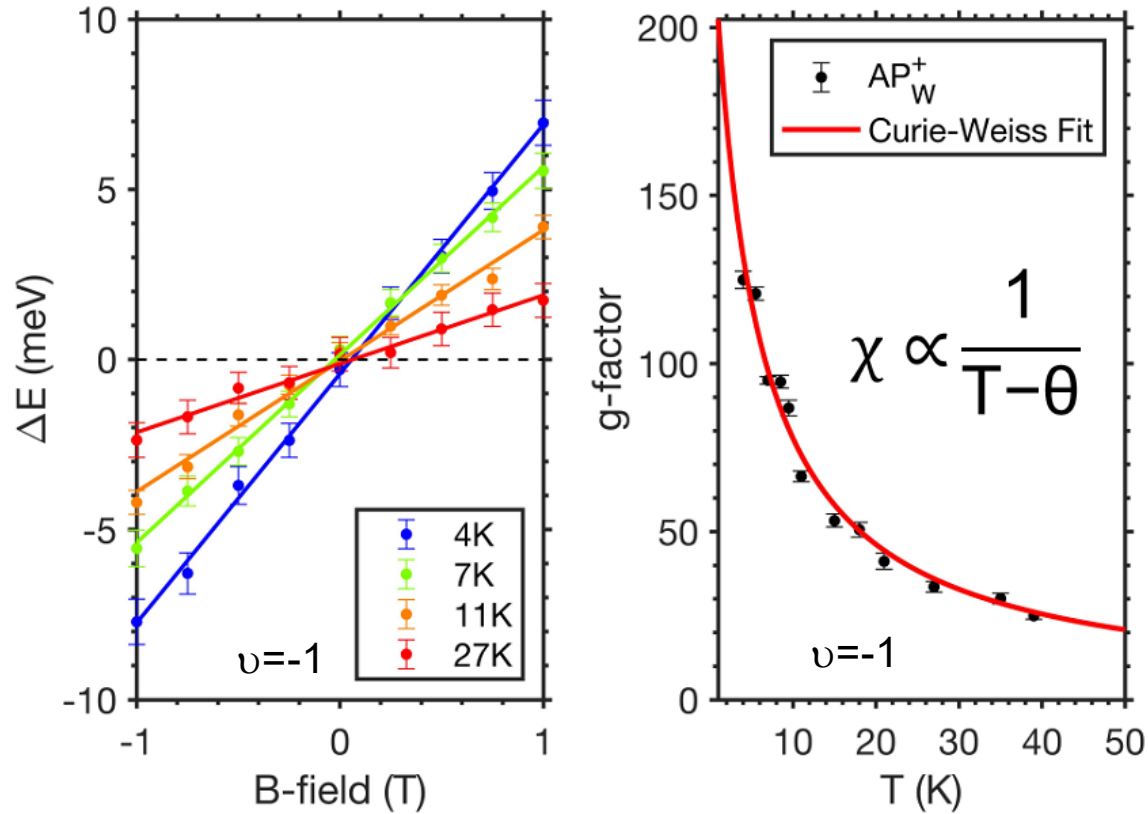
Valerio
Vitale



Johannes
Lischner

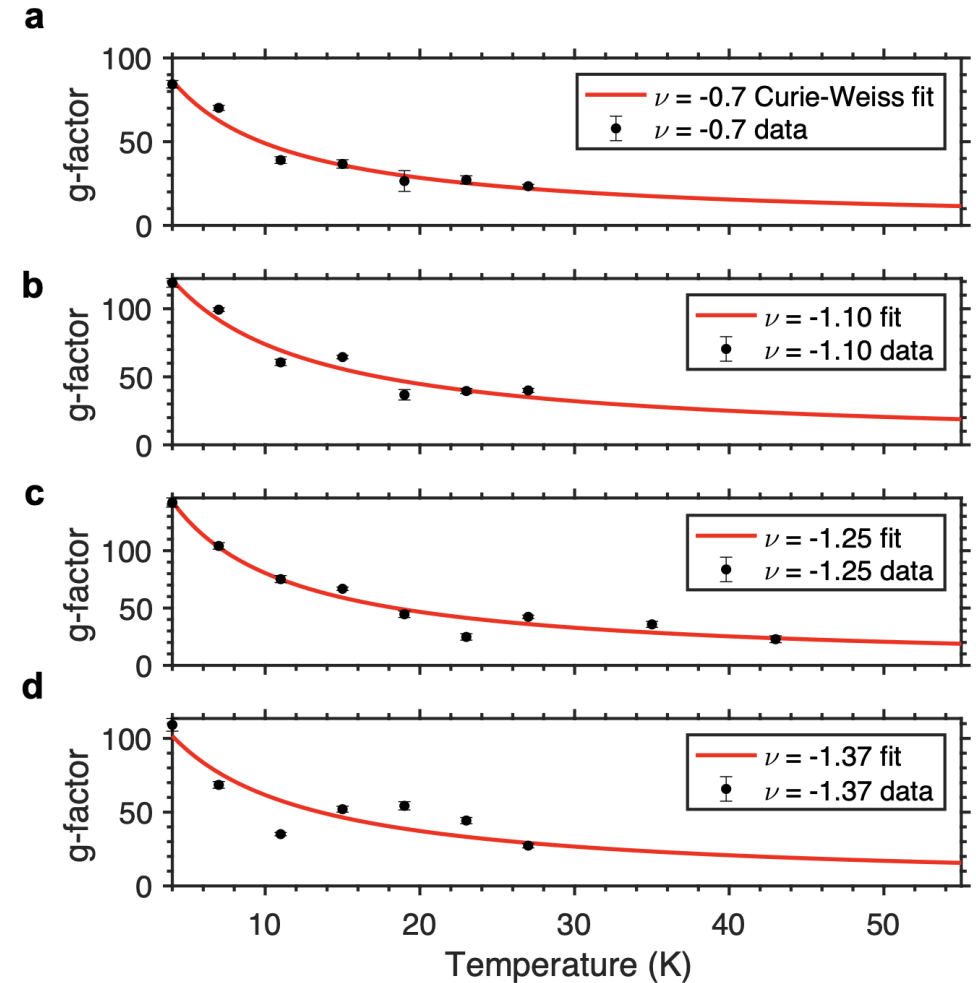


Temperature dependence of magnetic interactions



$$\theta = -4.6 \pm 0.9 \text{ K}$$

Suggests antiferromagnetic ordering at one hole per site



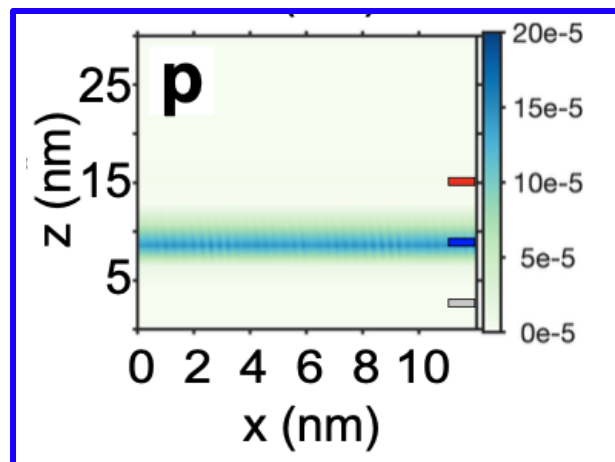
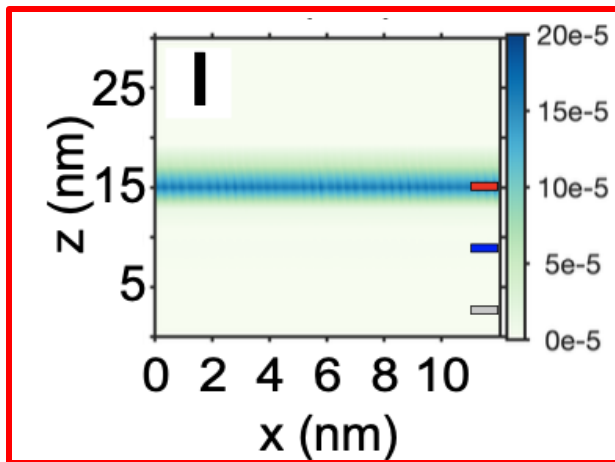
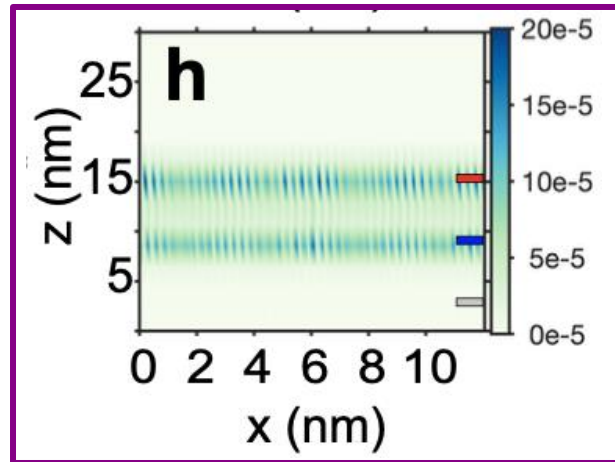
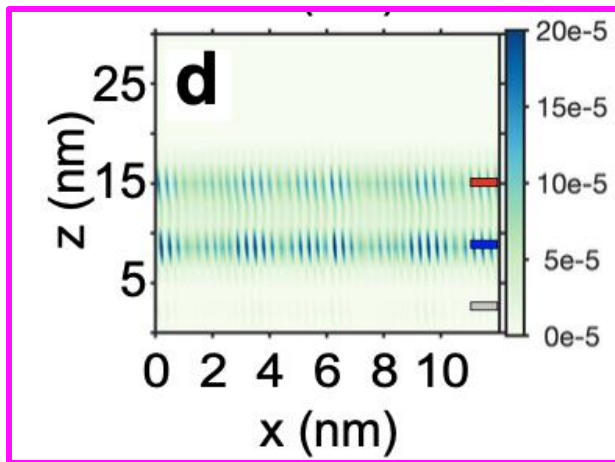
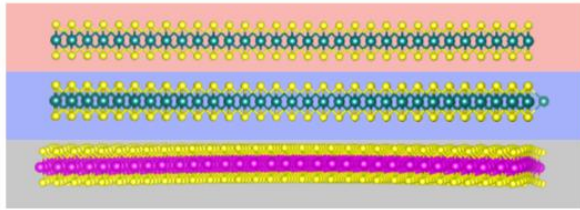
$$\theta = [-3.8 \ -5.4] \text{ K}$$

Outline

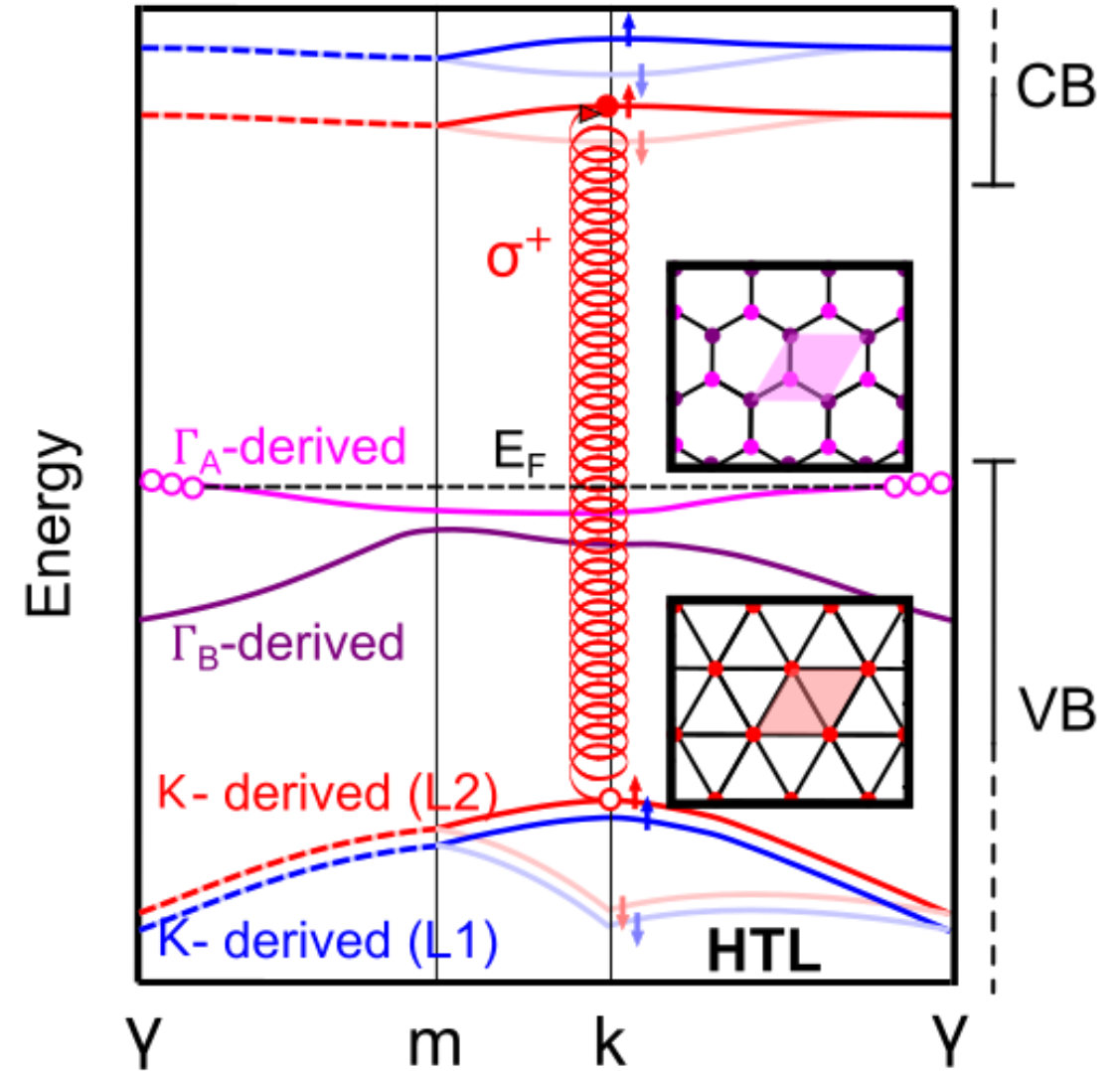
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Moiré band engineering

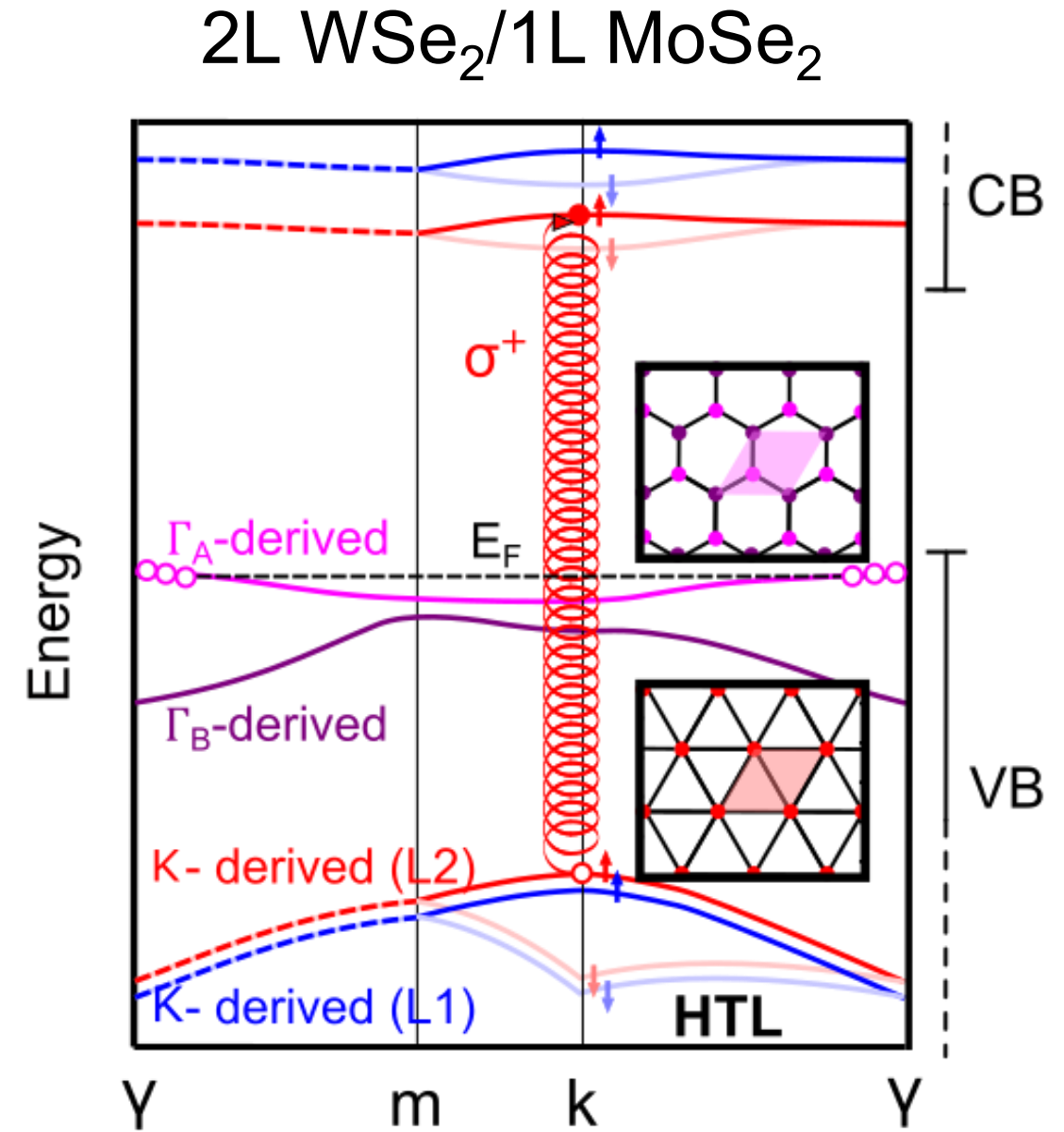
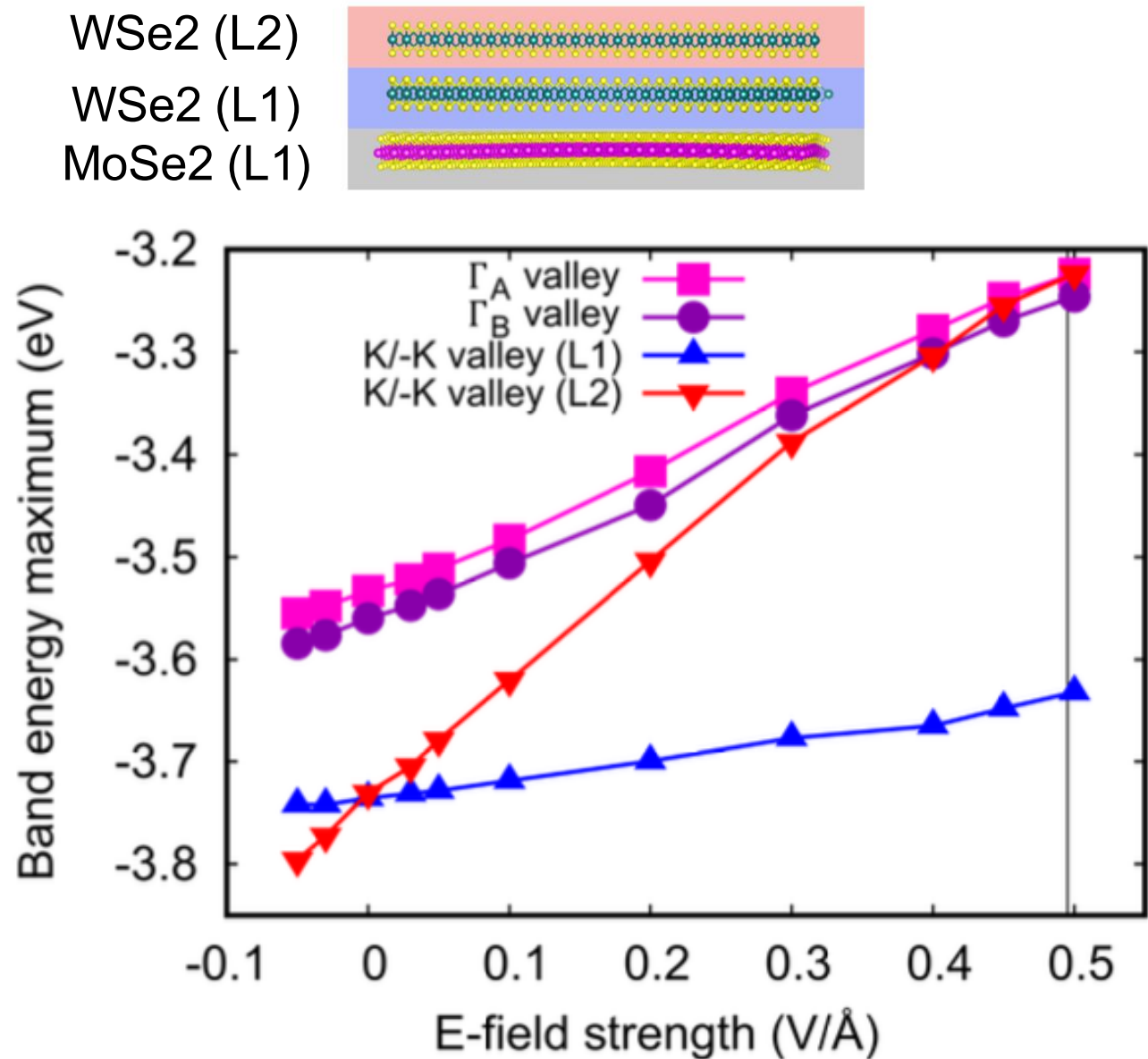
WSe₂ (L2)
 WSe₂ (L1)
 MoSe₂ (L1)



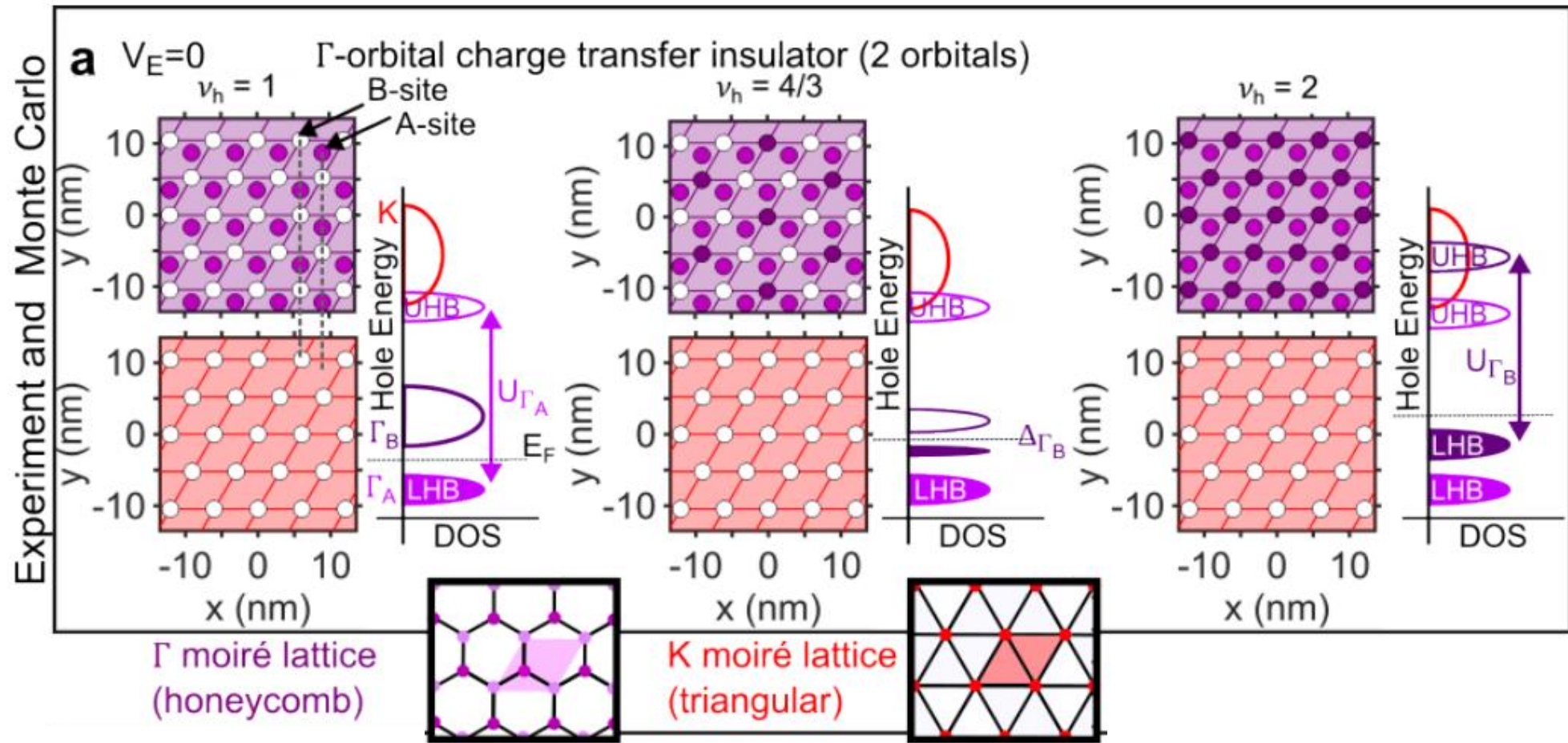
2L WSe₂/1L MoSe₂



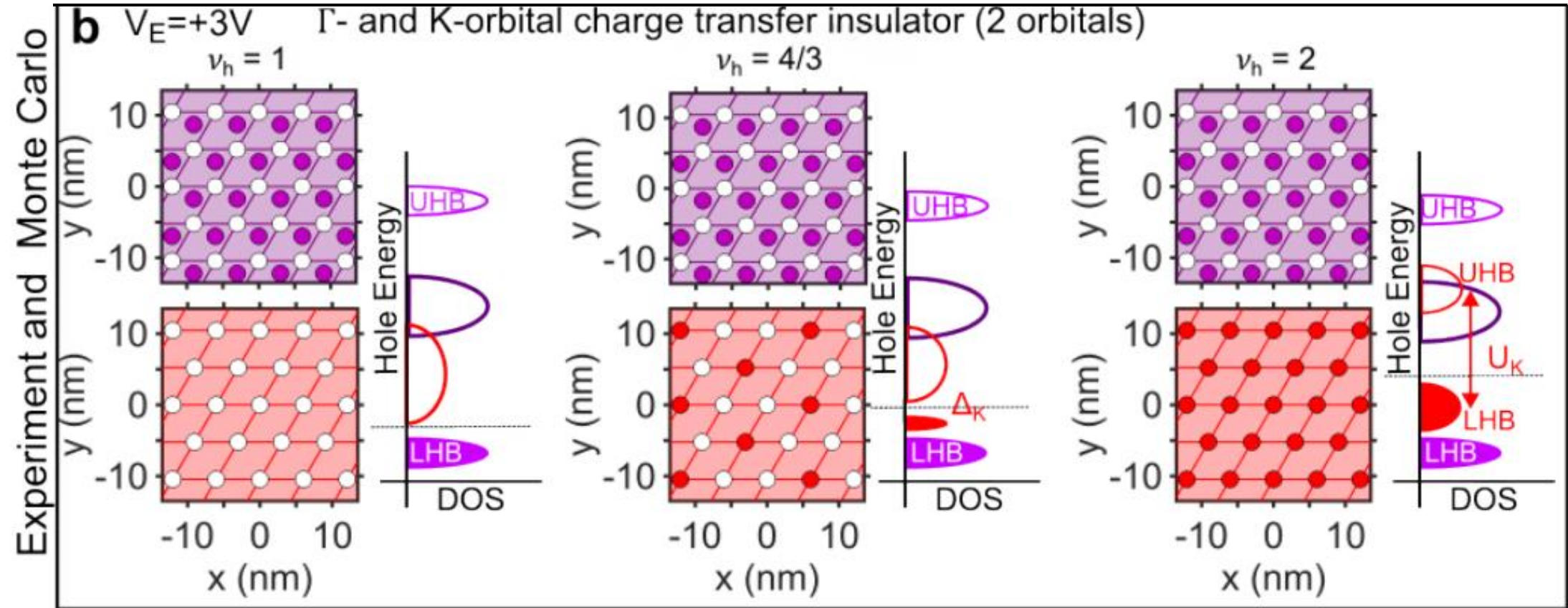
Moiré band engineering



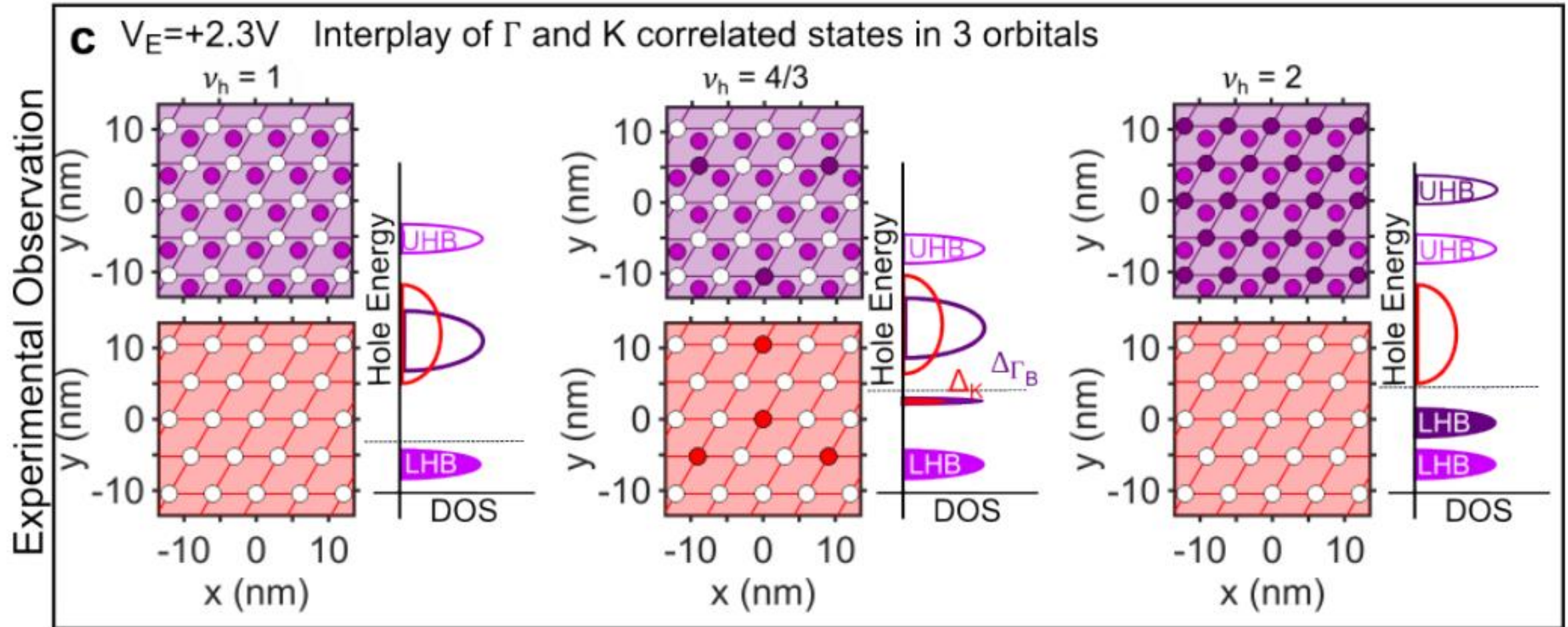
Γ -valley correlated states in heterotrilayer



Interplay between Γ - & K-valley correlated states



Interplay between Γ - & K-valley correlated states

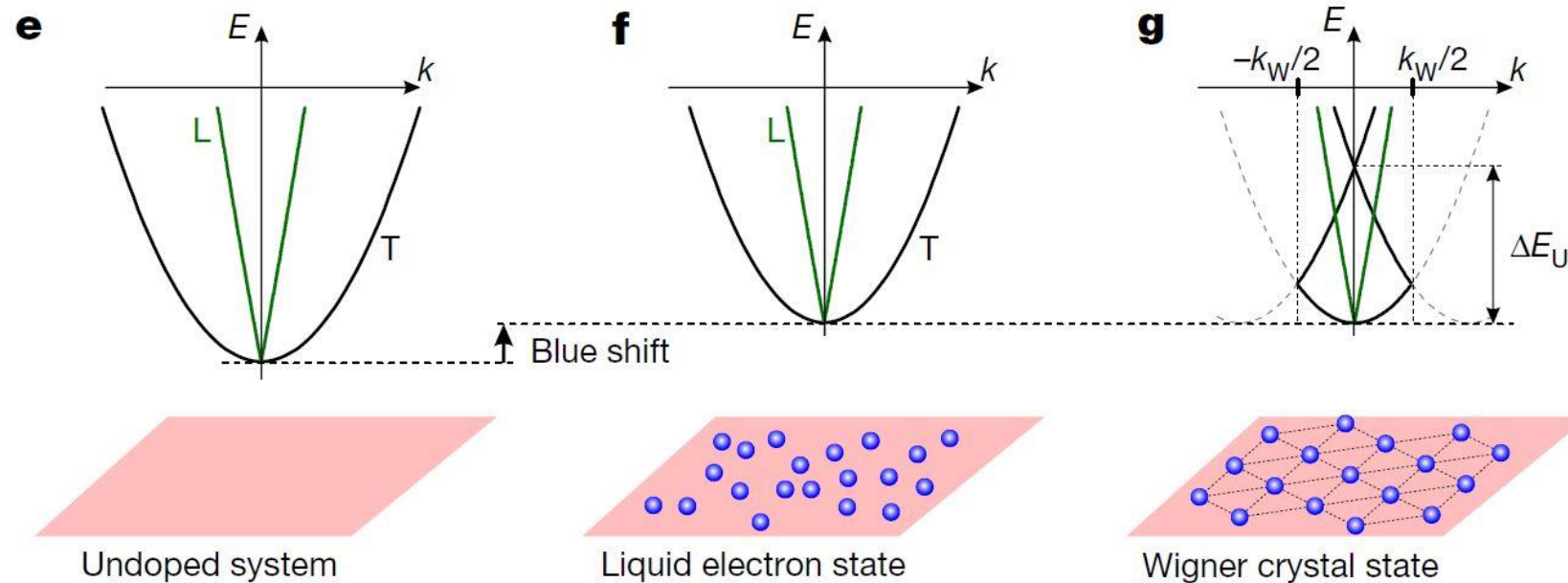


Outline

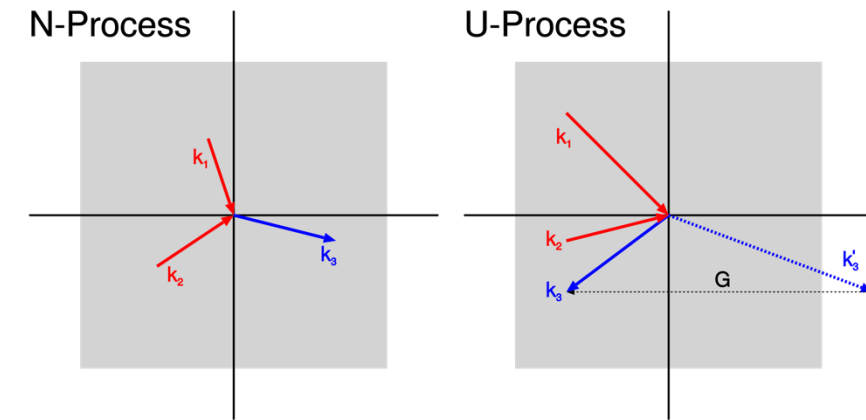
1. Intro: *moiré lattices* and *polaron spectroscopy*
2. Strongly correlated electronic states in moiré materials:
 - MoSe₂/WSe₂
 - MoSe₂/ Bilayer WSe₂ (multi-orbital moiré system)
3. Wigner crystallisation in monolayer WSe₂ & MoSe₂/WSe₂

Umklapp Process: Signature of Wigner Crystallisation

Exciton dispersion in a monolayer semiconductor



Umklapp scattering mechanism

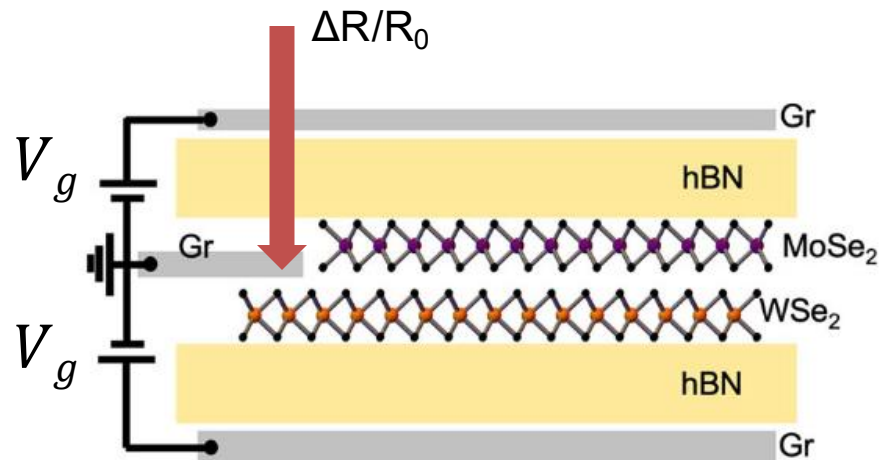


Exciton KE: $\Delta E \approx \hbar^2 k_W^2 / 2m_x^2$,

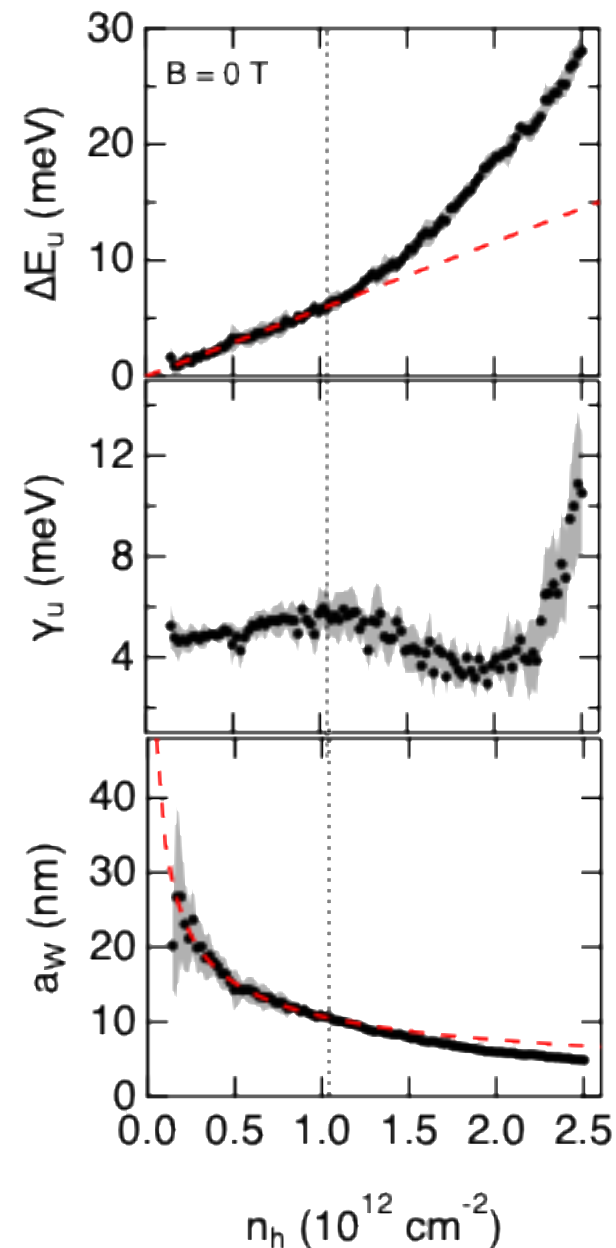
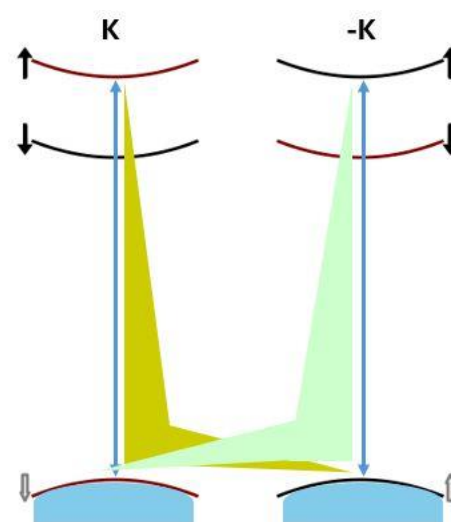
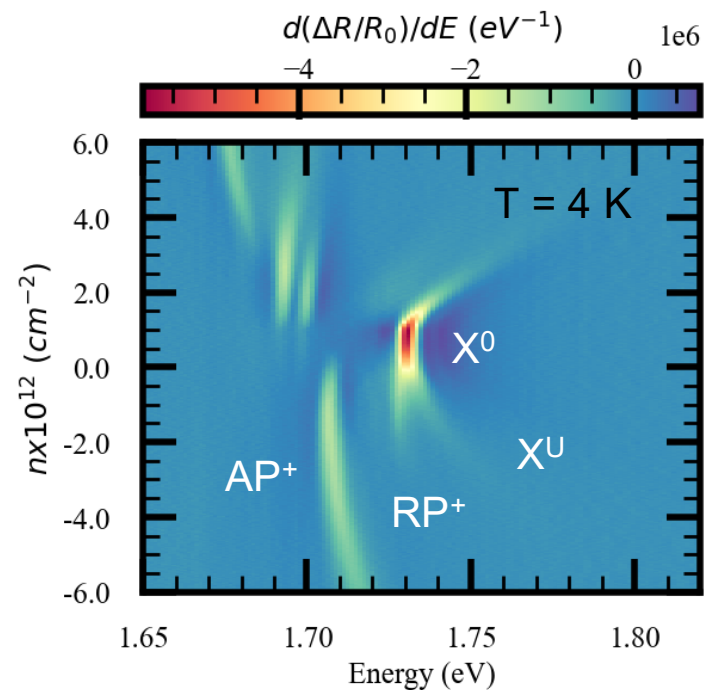
where m_x is the exciton mass and k_W is the reciprocal lattice vector of Wigner crystal

For a triangular lattice, $\Delta E = \hbar^2 n / \sqrt{3} m_x$

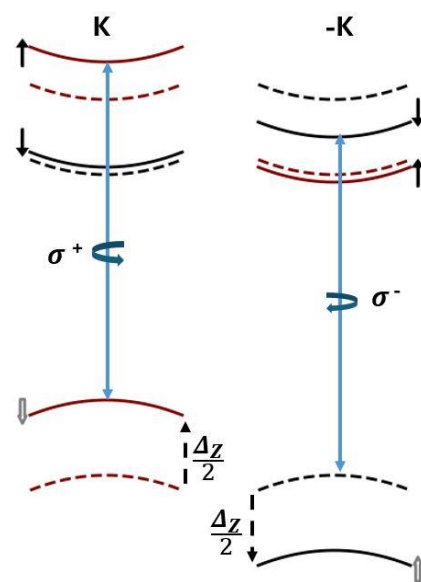
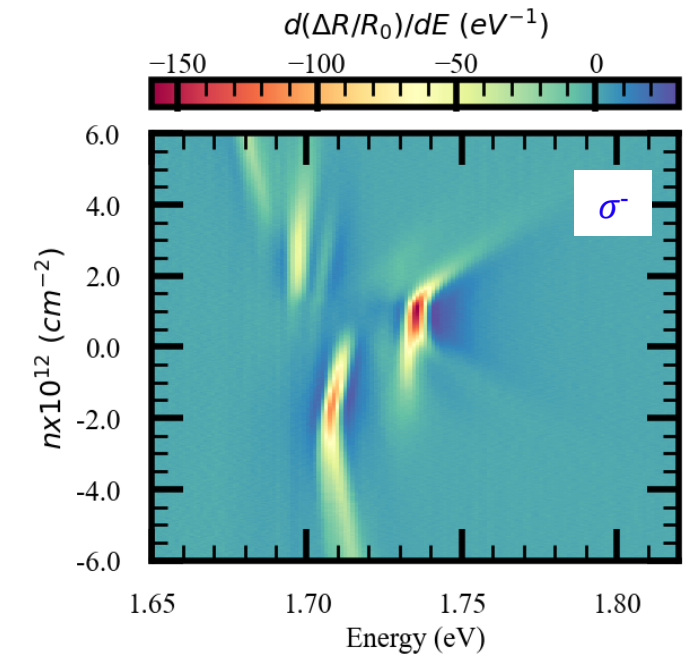
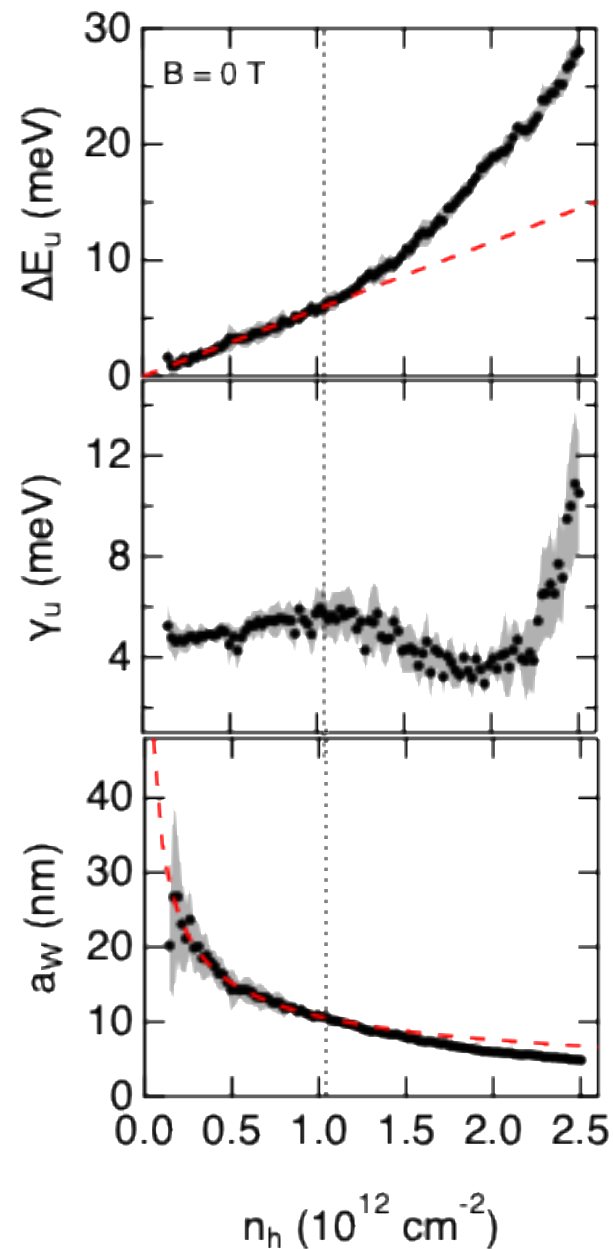
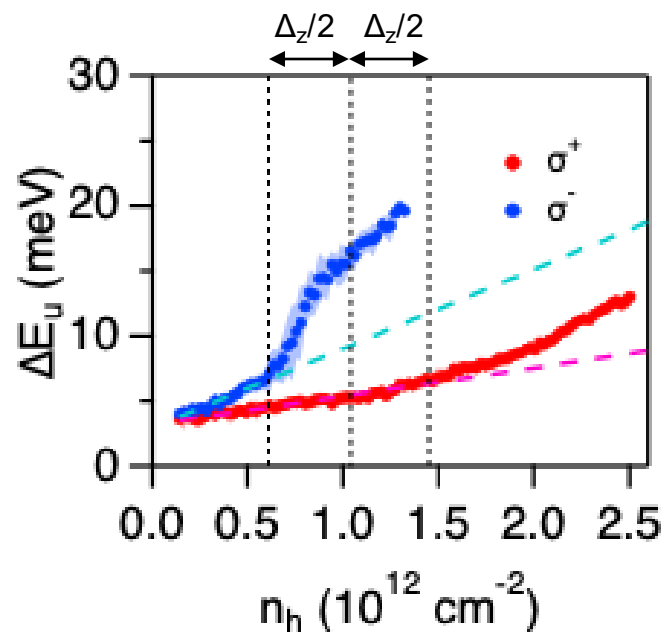
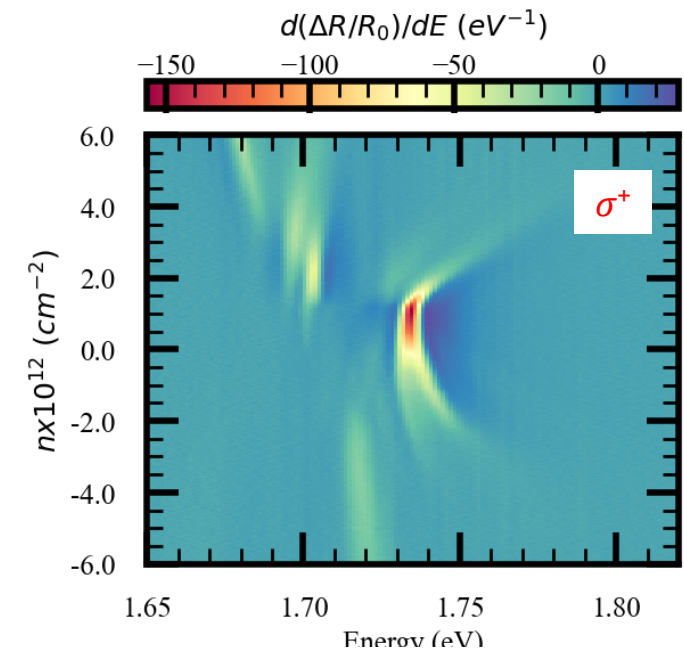
Wigner crystallisation in monolayer WSe₂ (B = 0 T)



- Wigner crystallization in ML WSe₂ at 4 K!
- Linear $\Delta E_U \Leftrightarrow m_X \sim 3m_0$



Wigner crystallisation in monolayer WSe₂ (B = 5 T)



Wigner crystallisation in monolayer MoSe₂

Article

Signatures of Wigner crystal of electrons in a monolayer semiconductor

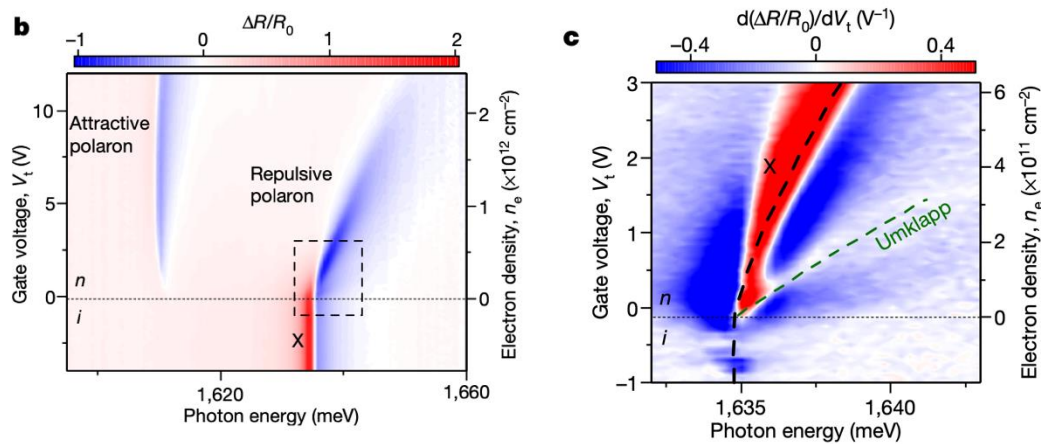
<https://doi.org/10.1038/s41586-021-03590-4>

Received: 6 October 2020

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Tomaz Smolerski^{1,2}, Pavel E. Dolgirev², Clemens Kuhlenskamp^{1,3,4}, Alexander Popert¹, Yuya Shimazaki¹, Patrick Back¹, Xiaobo Lu¹, Martin Kroner¹, Kenji Watanabe⁵, Takashi Taniguchi⁶, Ilya Esterlis², Eugene Demler^{2,7,8} & Ataç Imamoğlu^{1,5}

T = 80 mK



nature physics

Article

<https://doi.org/10.1038/s41567-024-02759-8>

An electronic microemulsion phase emerging from a quantum crystal-to-liquid transition

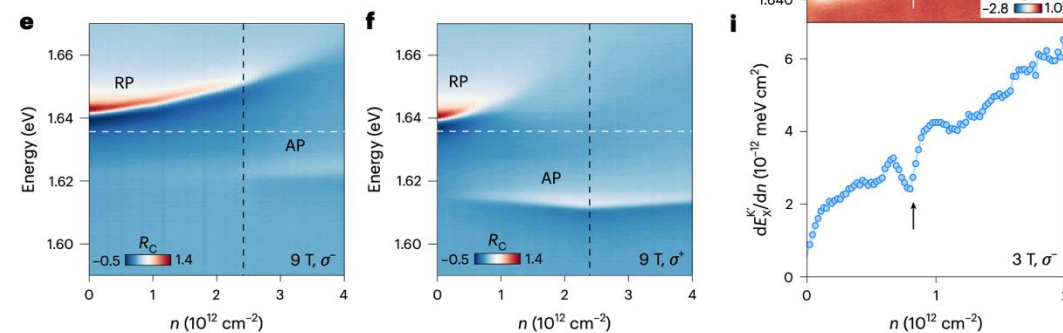
Received: 19 February 2024

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Jiho Sung^{1,2,12}, Jue Wang^{1,2,12}, Ilya Esterlis^{2,3,12}, Pavel A. Volkov^{2,4,12}, Giovanni Scuri^{1,2,5}, You Zhou⁶, Elise Brutschea¹, Takashi Taniguchi⁷, Kenji Watanabe⁷, Yubo Yang⁸, Miguel A. Morales⁵, Shiwei Zhang⁹, Andrew J. Millis^{8,9}, Mikhail D. Lukin², Philip Kim^{2,10}, Eugene Demler¹¹ & Hongkun Park^{1,2}

T = 16 mK



RESEARCH ARTICLES

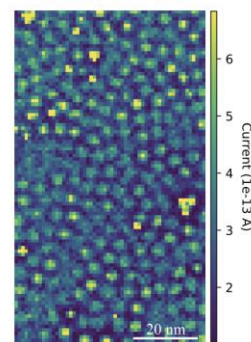
Science 15 MAY 2025

2D MATERIALS

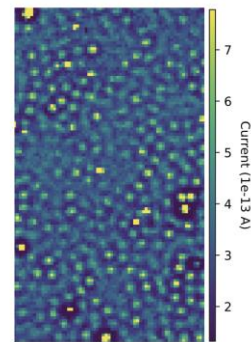
Imaging quantum melting in a disordered 2D Wigner solid

Ziyu Xiang^{1,2,3,†}, Hongyuan Li^{1,2,3,†}, Jiangnan Xiao^{1,2,3,†}, Mit H. Naik^{1,3}, Zhehao Ge¹, Zehao He¹, Sudi Chen^{1,3,4}, Jiahui Nie¹, Shiyu Li¹, Yifan Jiang¹, Renee Sailus⁵, Rounak Banerjee⁵, Takashi Taniguchi⁶, Kenji Watanabe⁷, Sefaattin Tongay⁵, Steven G. Louie^{1,3}, Michael F. Crommie^{1,3,4*}, Feng Wang^{1,3,4*}

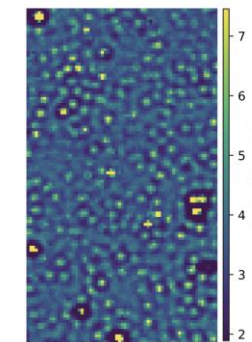
G $n \sim 4.2 \times 10^{12}$ cm⁻²



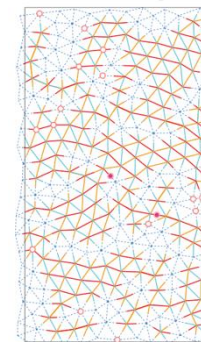
H $n \sim 6.1 \times 10^{12}$ cm⁻²



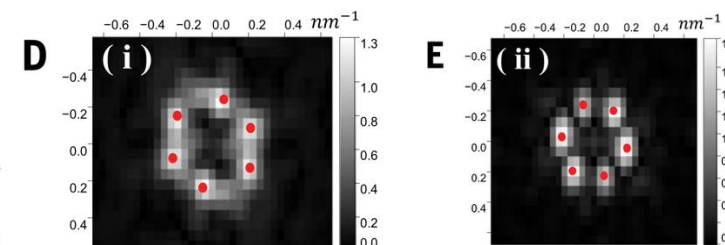
I $n \sim 7.0 \times 10^{12}$ cm⁻²



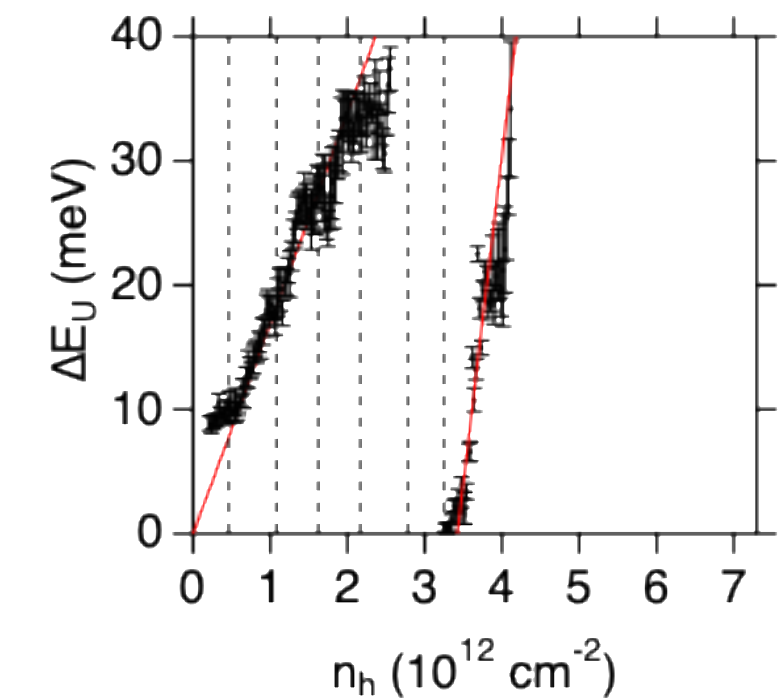
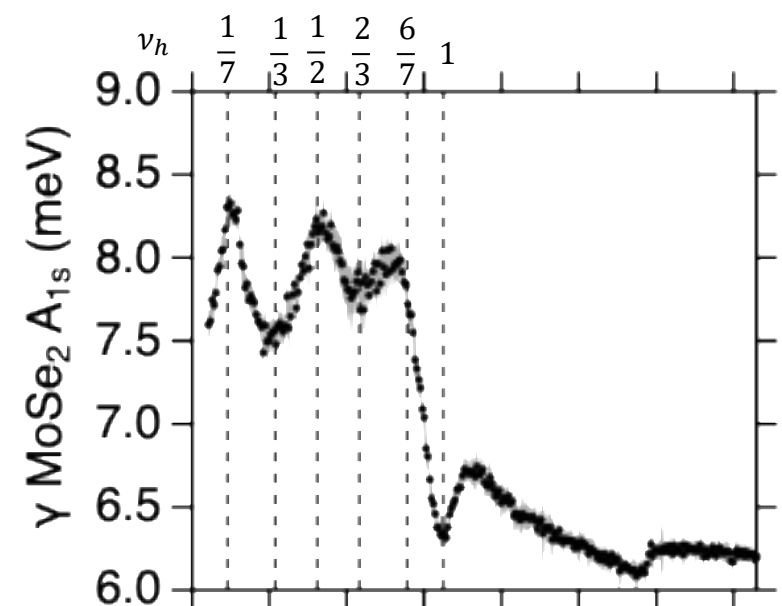
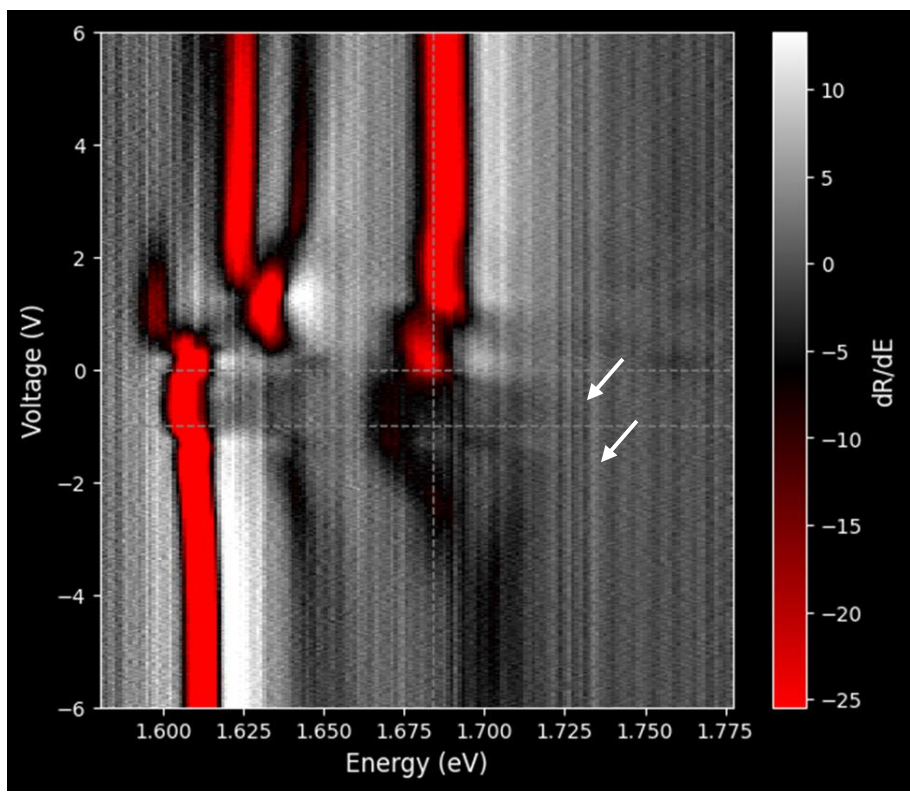
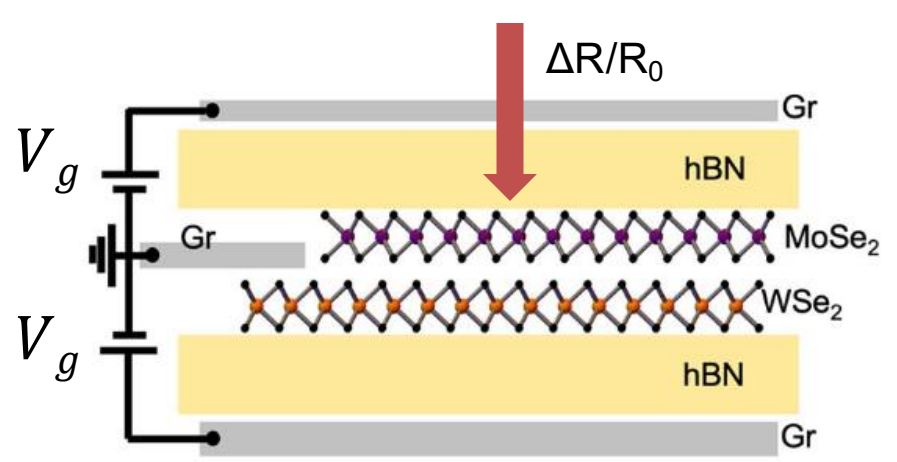
C Voronoi diagram



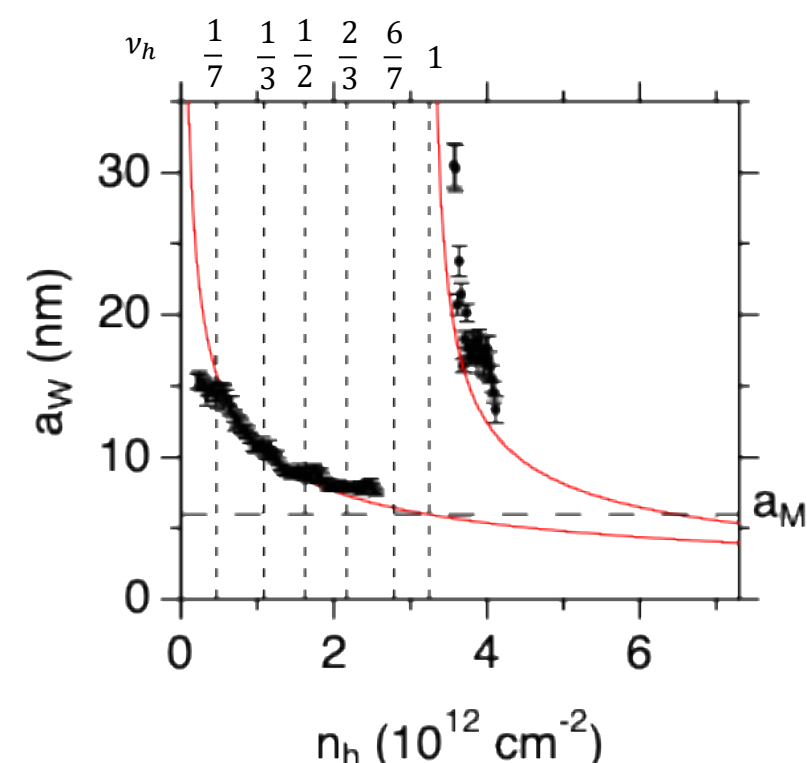
T = 5.4 K (bilayer MoSe₂)



Umklapp scattering in MoSe₂/WSe₂



From ΔE_U vs n :
 $m_x \approx m_0$
 $m_x \approx 0.5 m_0$



Summary

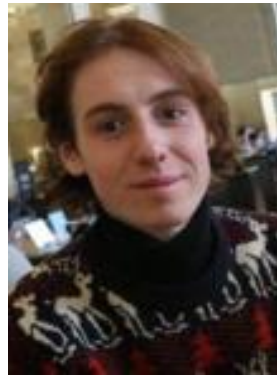
1. Strongly correlated states for a variety of fractional lattice fillings in $\text{MoSe}_2/\text{WSe}_2$
2. Excitons at $\pm K$ -valleys are excellent probes of correlated states in all valleys (but insensitive to spin-ordering in Γ -valleys)
3. Observation of Wigner crystallisation in monolayer WSe_2 and moiré $\text{MoSe}_2/\text{WSe}_2$ via exciton Umklapp scattering



Brian. D.
Gerardot



Hyeonjun
Baek



Aidan
Campbell



Bibi Mary
Francis



Kenji
Watanabe

Imperial College
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Valerio
Vitale



Johannes
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Takashi
Taniguchi