Lattice Reconstruction, Piezoelectricity, and Band Topology in Twisted $MoTe_2$

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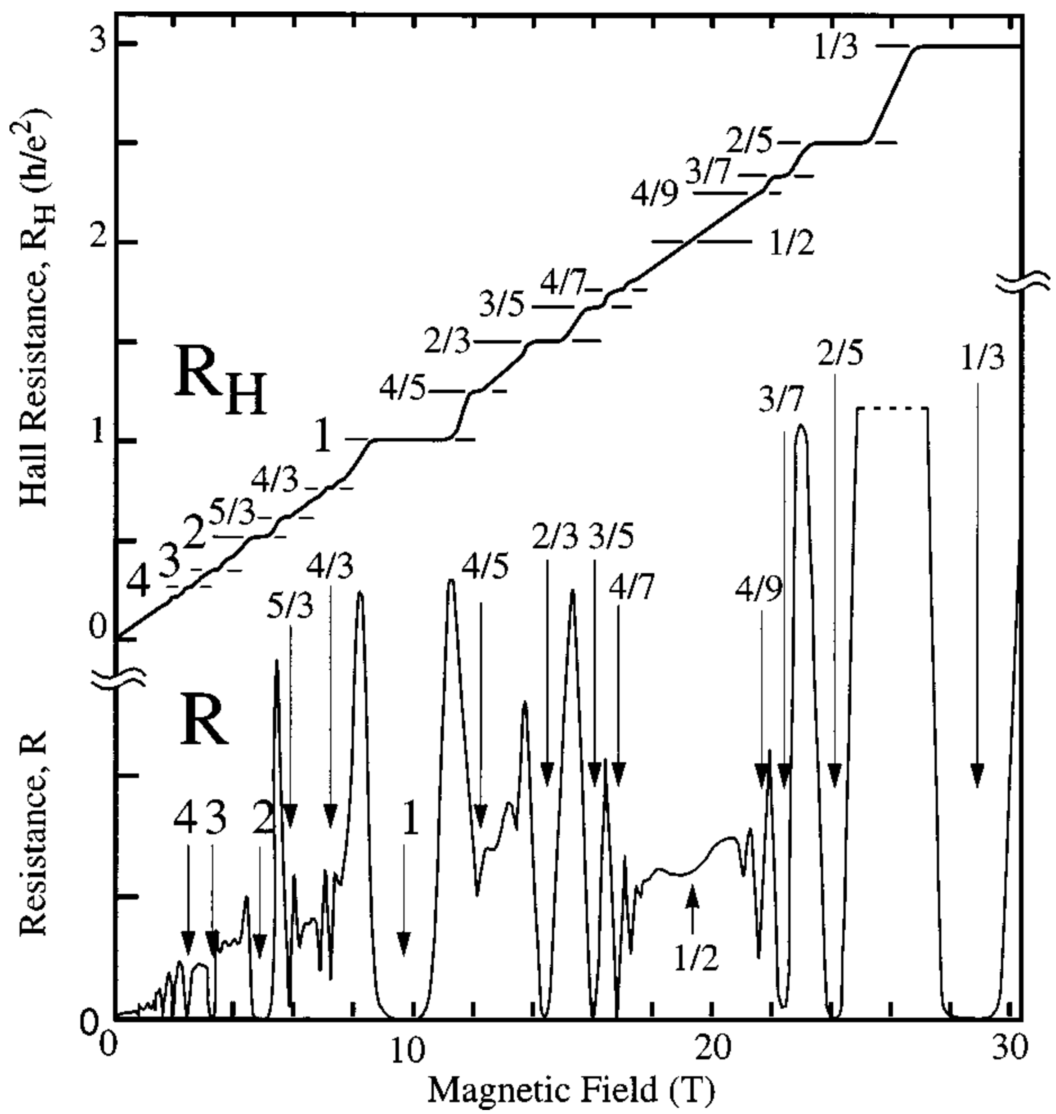
Outline

- The search for zero-field fractional quantum Hall effect
- Twisted transition metal dichalcogenides (TMD)
- Twist angle dependent topological moiré band structure
- Higher Landau level physics and non-Abelian states

Quantum Hall Effect



Stormer, Nobel Lecture

The requirement of a strong magnetic field is a challenge for conversion into technology

Quantized Hall Conductance in a Two-Dimensional Periodic Potential

D. J. Thouless, M. Kohmoto,[a] M. P. Nightingale, and M. den Nijs
Department of Physics, University of Washington, Seattle, Washington 98195
(Received 30 April 1982)

The Hall conductance of a two-dimensional electron gas has been studied in a uniform magnetic field and a periodic substrate potential U. The Kubo formula is written in a form that makes apparent the quantization when the Fermi energy lies in a gap. Explicit expressions have been obtained for the Hall conductance for both large and small $U/\hbar\omega_c$.

$$\sigma_{\mathrm{H}} = \frac{ie^2}{2\pi h}\sum\int d^2k\int d^2r\left(\frac{\partial u^*}{\partial k_1}\frac{\partial u}{\partial k_2} - \frac{\partial u^*}{\partial k_2}\frac{\partial u}{\partial k_1}\right)$$

$$= \frac{ie^2}{4\pi h}\sum\oint dk_j\int d^2r\left(u^*\frac{\partial u}{\partial k_j} - \frac{\partial u^*}{\partial k_j}u\right),$$

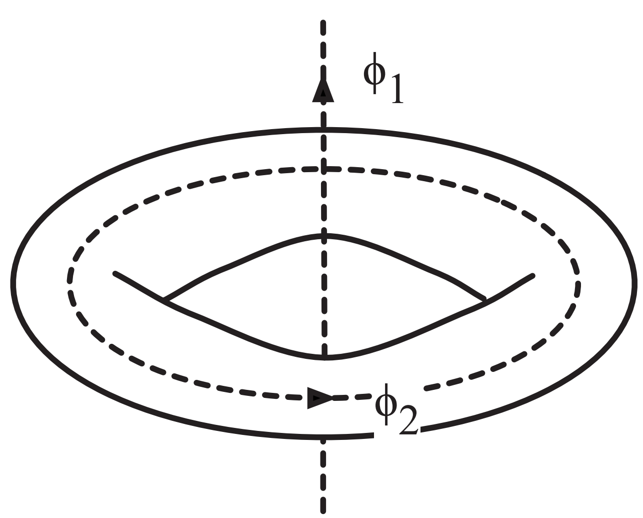
The Hall conductance can be written as a **topological number** (the Chern number) and must be quantized

PHYSICAL REVIEW B VOLUME 31, NUMBER 6 15 MARCH 1985

Quantized Hall conductance as a topological invariant

Qian Niu, D. J. Thouless,* and Yong-Shi Wu†
Department of Physics FM-15, University of Washington, Seattle, Washington 98195
(Received 21 September 1984)

Whenever the Fermi level lies in a gap (or mobility gap) the bulk Hall conductance can be expressed in a topologically invariant form showing the quantization explicitly. The new formulation generalizes the earlier result by Thouless, Kohmoto, Nightingale, and den Nijs to the situation where many-body interaction and substrate disorder are also present. When applying to the fractional quantized Hall effect, we draw the conclusion that there must be a symmetry breaking in the many-body ground state. The possibility of writing the fractionally quantized Hall conductance as a topological invariant is also discussed.

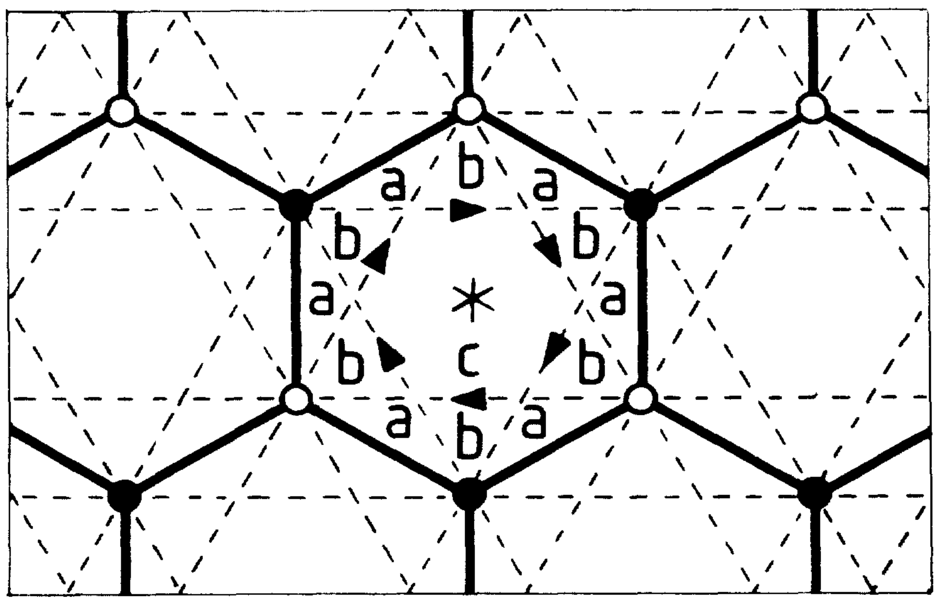


- The quantization of the Hall conductance is robust against disorder and interaction as long as there is a gap
- The only way for the Hall conductance to become **fractional** is that the ground state must be degenerate (on a torus)
- The presence of magnetic field is not essential to their derivation

VOLUME 61, NUMBER 18 PHYSICAL REVIEW LETTERS 31 OCTOBER 1988

Model for a Quantum Hall Effect without Landau Levels: Condensed-Matter Realization of the "Parity Anomaly"

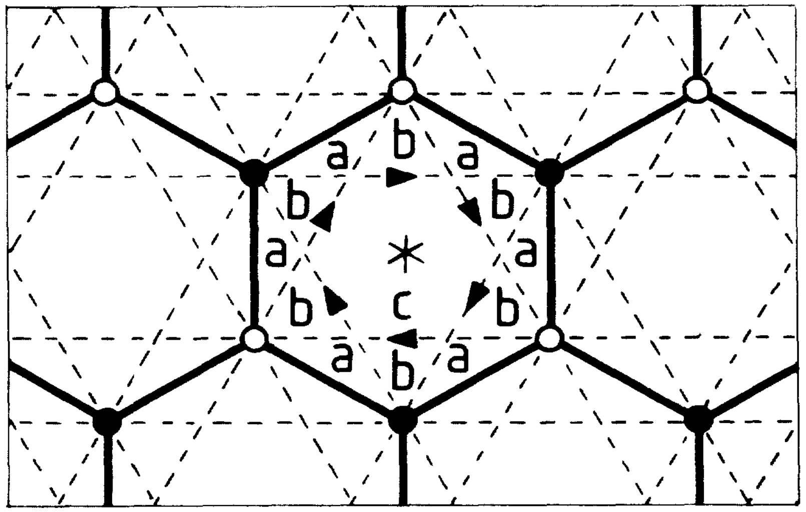
F. D. M. Haldane
Department of Physics, University of California, San Diego, La Jolla, California 92093
(Received 16 September 1987)



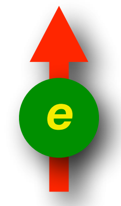
While the particular model presented here is unlikely to be directly physically realizable, it indicates that, at least in principle, the QHE can be placed in the wider context of phenomena associated with broken time-reversal invariance, and does not necessarily require external magnetic fields, but could occur as a consequence of magnetic ordering in a quasi-two-dimensional system.

First concrete model of the **quantum anomalous Hall effect** (Chern insulators)

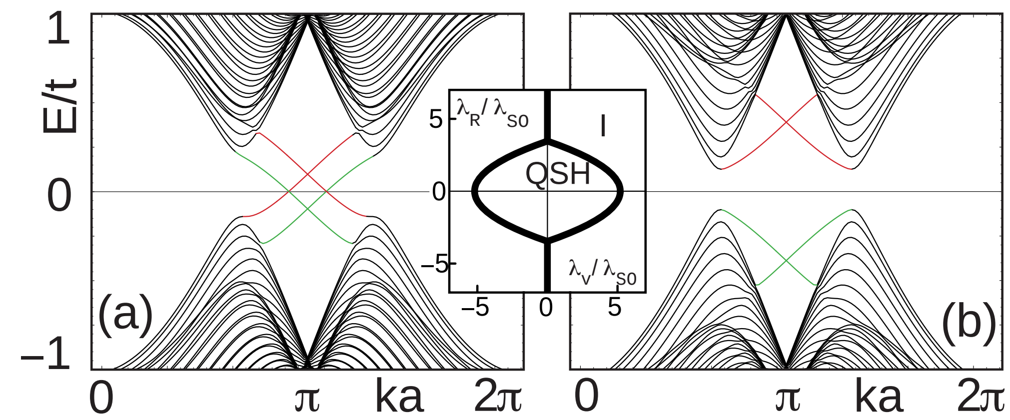
Bloch bands can also carry nonzero Chern numbers and exhibit the integer quantum Hall effect



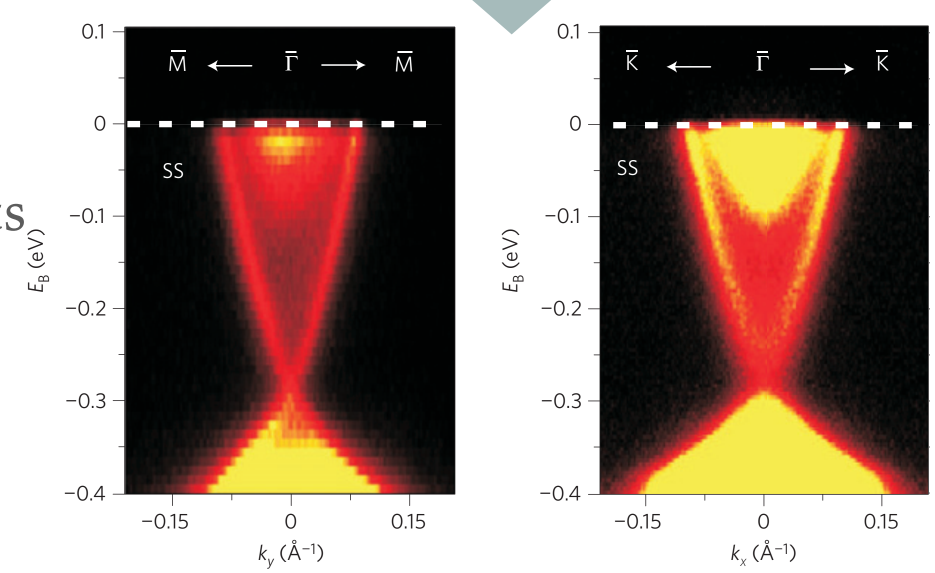
Haldane model of QAHE (1988)
broken T symmetry



Add electron spin

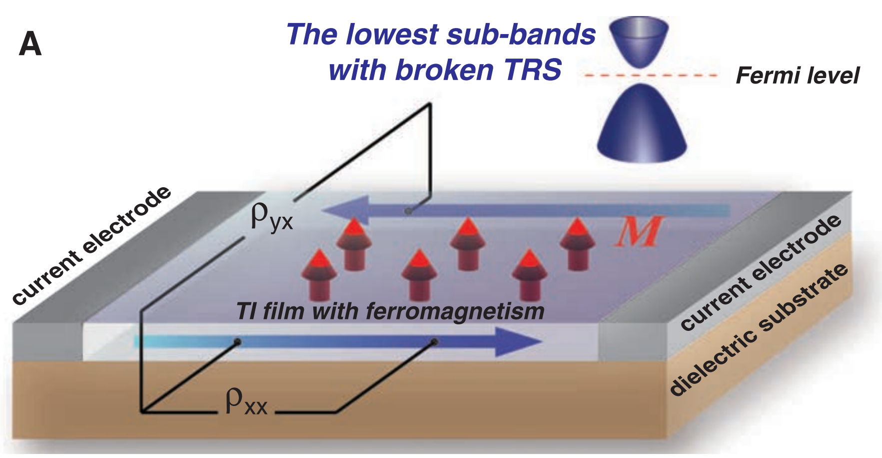


Kane-Mele model of QSHE (2005);
see also Bernevig & Zhang (2006)
with T-symmetry



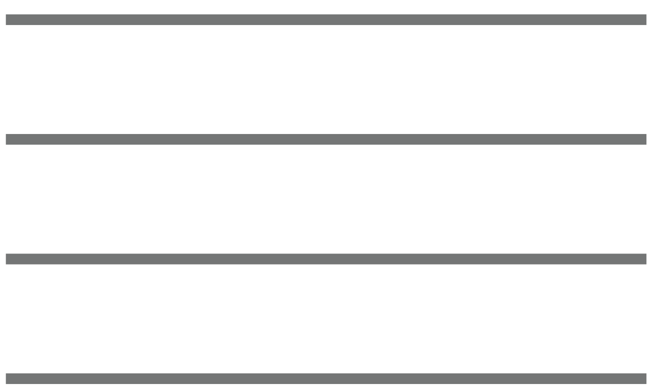
Topological insulators (2007)
with T-symmetry

Add magnetic dopants

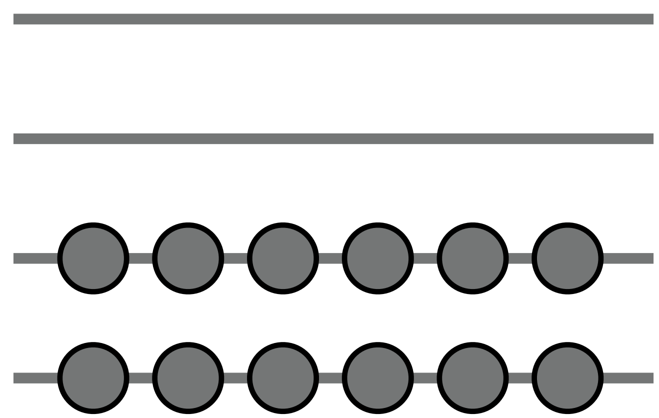


Magnetically doped TI (2013)
broken T-symmetry

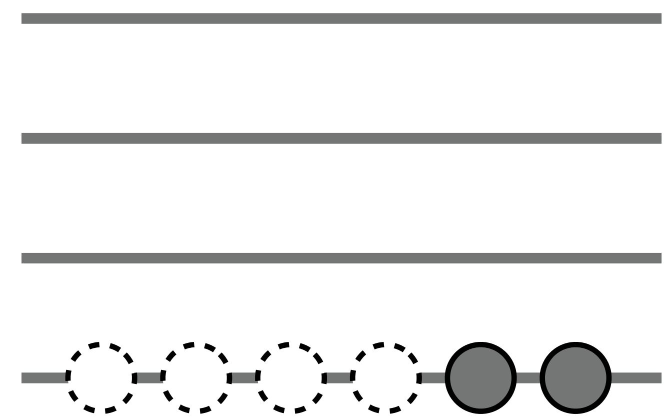
From Integer to Fractional



The Landau levels are the **oldest** **Chern** bands



Completely filled LLs give rise to the **integer quantum Hall effect**



Partially filled LLs give rise to the **fractional quantum Hall effect**

Even though the FQHE is an interacting effect, there is nothing special about the interaction. The physics is in fact dictated at the single-particle level by the guiding-center algebra, $[x, y] = i\ell^2$

A Quick Primer on Fractional Chern Insulators

- A **flat** Bloch band that has non-zero **Chern number** mimics the Landau level. When it is **partially** filled, a **fractional** quantum Hall effect can appear in the **absence** of magnetic field. This is called the fractional quantum anomalous Hall effect.

  Tang, Mei & Wen, PRL (2011); Neupert, Santos, Chamon & Mudry, PRL (2011); Sun, Gu, Katsura & Dạs Sarma (2011); Sheng, Gu, Sun & Sheng, Nature Comm. (2011); Regault & Bernevig, PRX (2011);

- What does flat mean? Flat in both **energy dispersion** and **band geometry** (Berry curvature and quantum metric)

- The band-projected position operators do not commute,

$$[x, y] = i\Omega(k) \qquad \Leftrightarrow \qquad [x, y] = i\ell^2$$

  For unit Berry curvature, $B_{\text{eff}} = 2\pi \cdot 625$ Tesla/(unit cell nm^2). If lattice constant is **5 nm**, then the effective B field is **157 Tesla**!

ARTICLE

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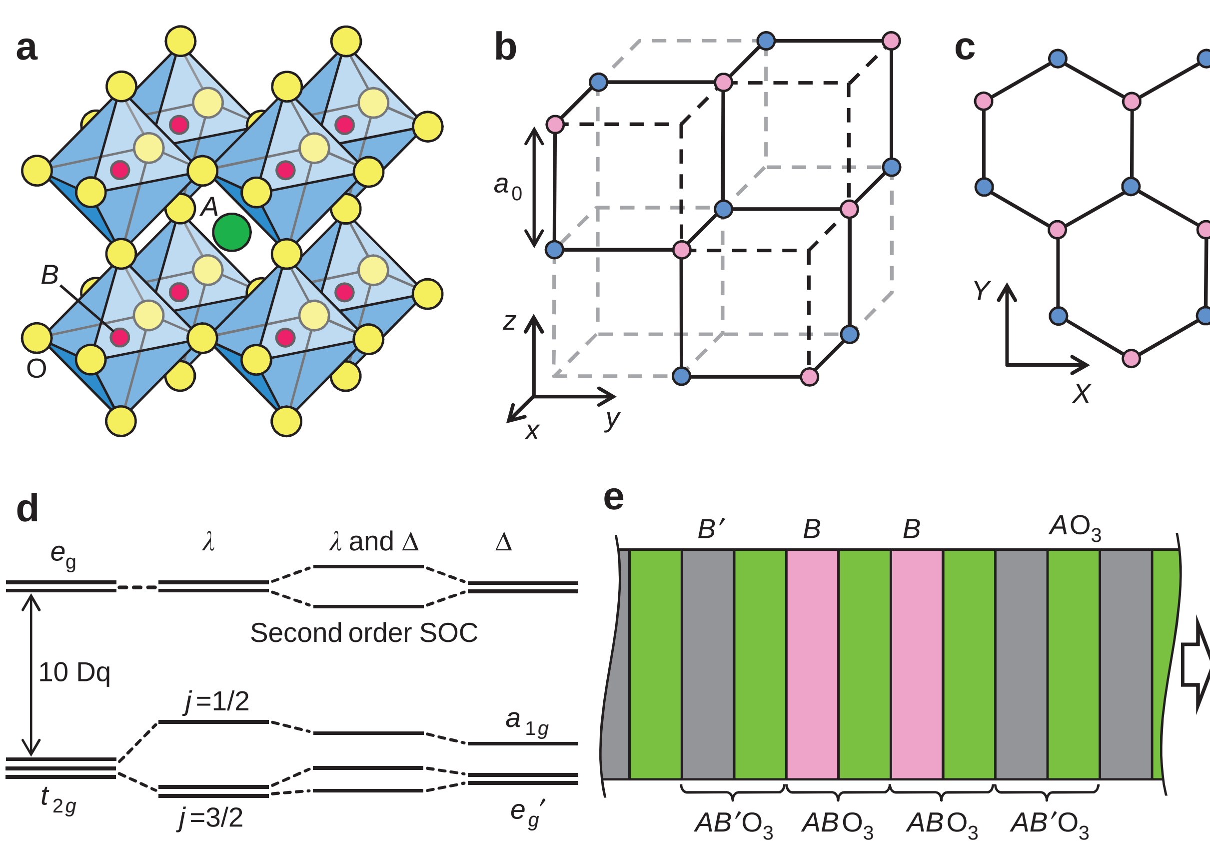
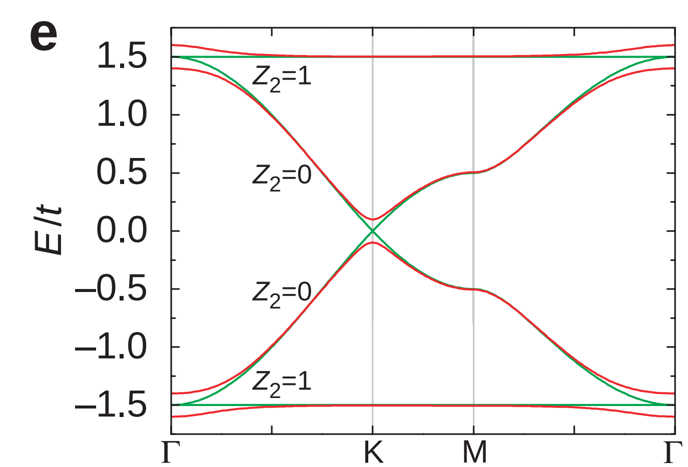
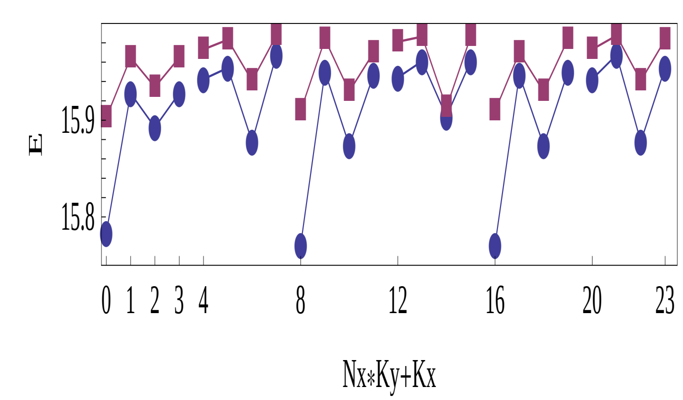
Interface engineering of quantum Hall effects in digital transition metal oxide heterostructures

Di Xiao[1], Wenguang Zhu[1,2], Ying Ran[3], Naoto Nagaosa[4,5] & Satoshi Okamoto[1]





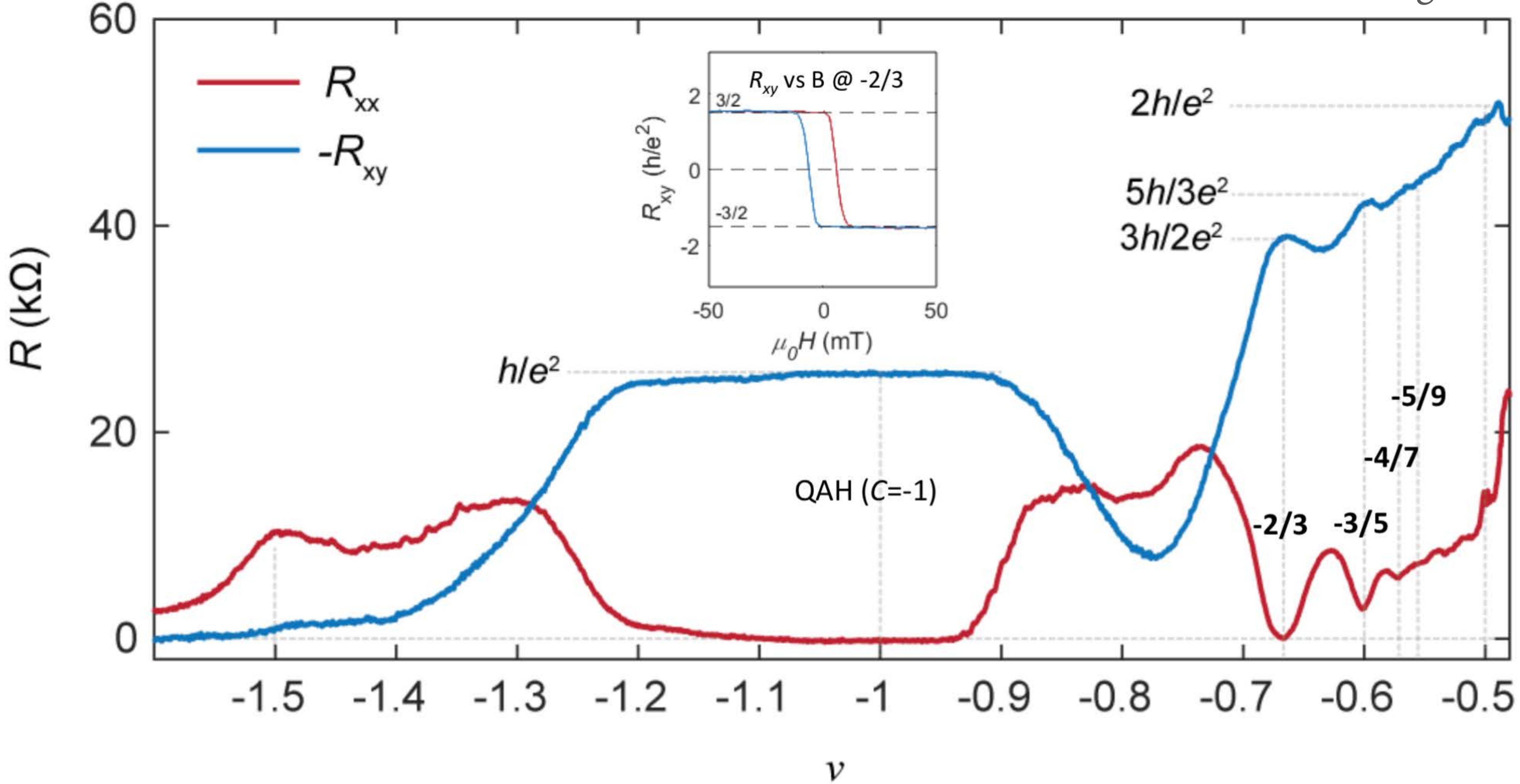


A material-based spinful model to realize FCI

Where else can we find flat Chern bands?

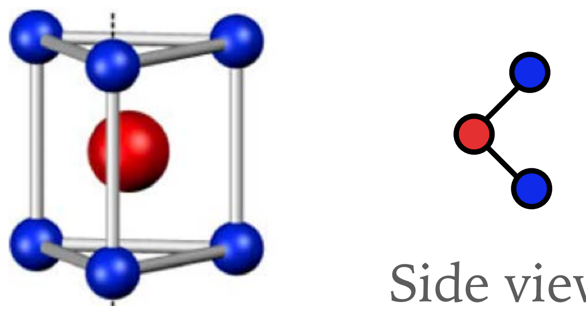
Fractional Chern Insulators in tMoTe2

From Xiaodong Xu

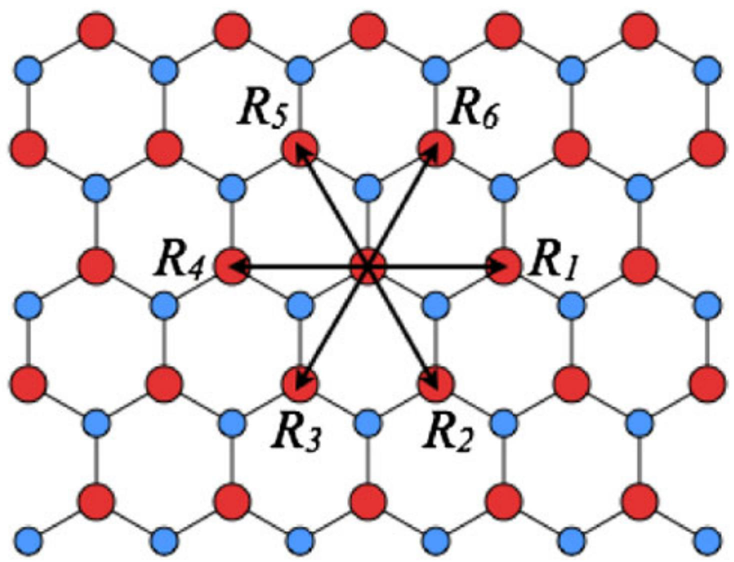


Cai et al, Nature (2023); Park et al, Nature (2023); Zeng et al, Nature (2023); Xu et al, PRX (2023);
For graphene, see Lu et al, Nature (2024);

Transition Metal Dichalcogenides (TMD)

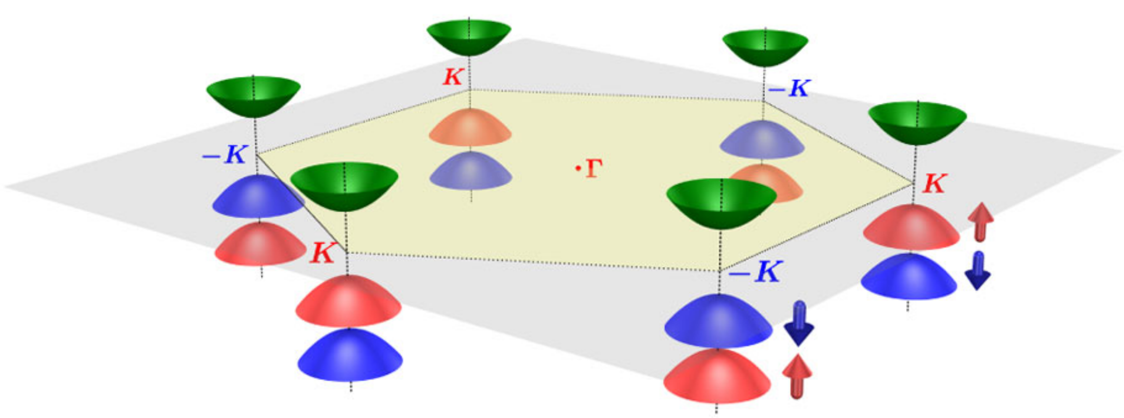


Side view



Top view

MX_2 (M = Mo,W, X = S, Se, Te)



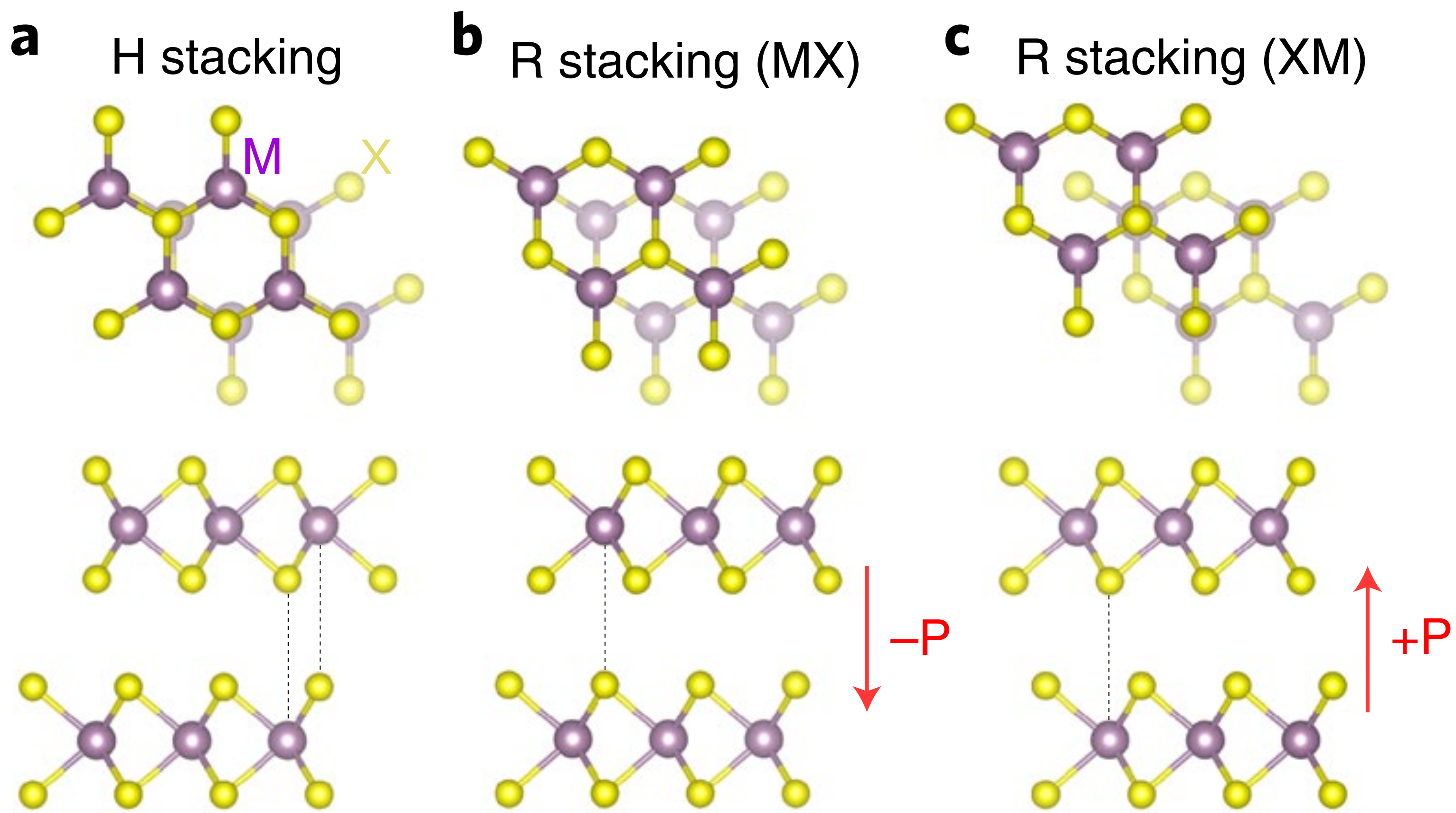
Band structure

Monolayer TMD **breaks inversion symmetry**, with a large **spin splitting** at the band edge.
Spin and valley are **locked**.

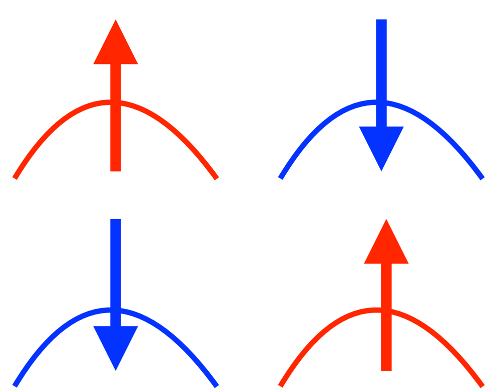
DX, Zhu, Liu, Xu, & Yao PRL (2012)

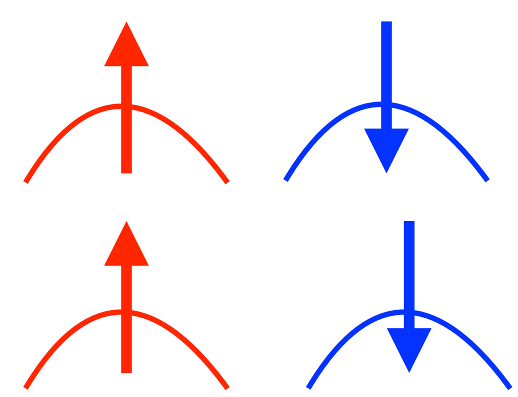
Two Types of Bilayers

Wang et al, Nature Nanotech (2022)



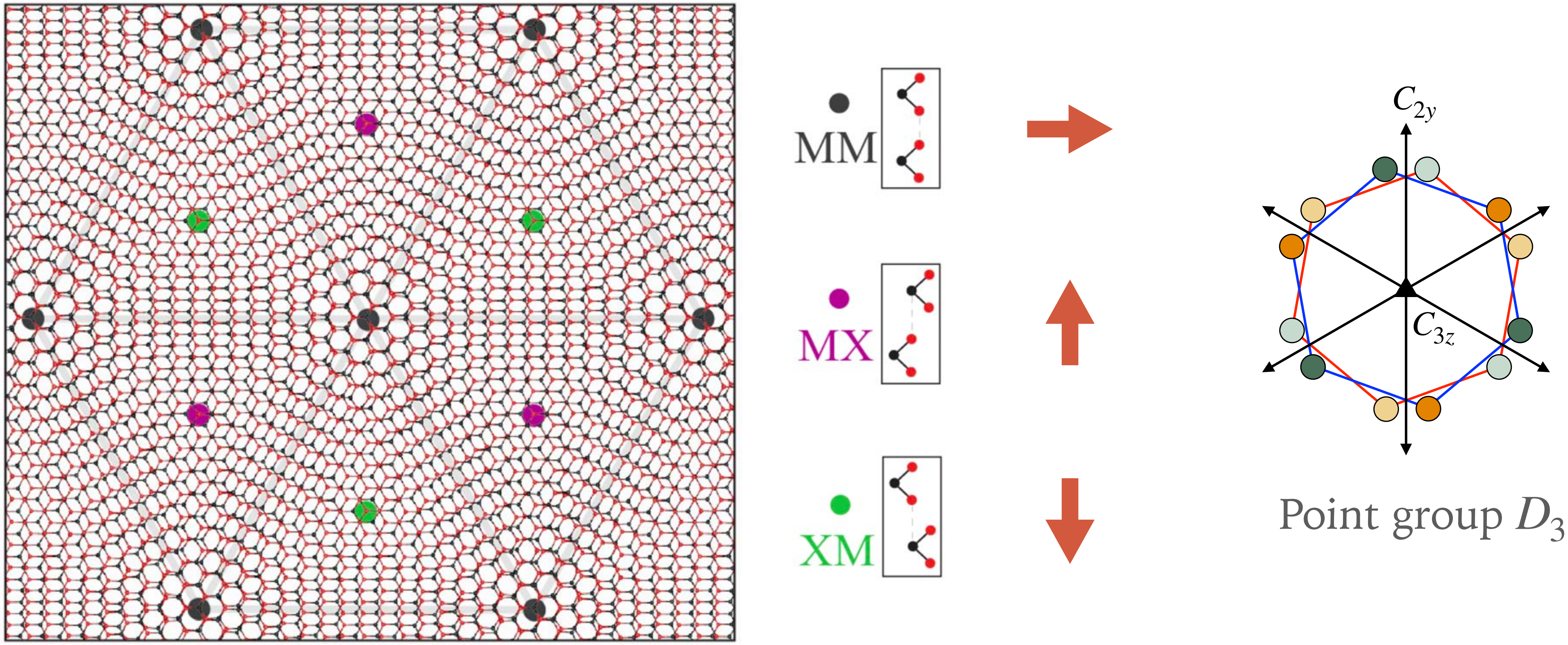
H: Interlayer tunneling forbidden





R: Interlayer tunneling allowed

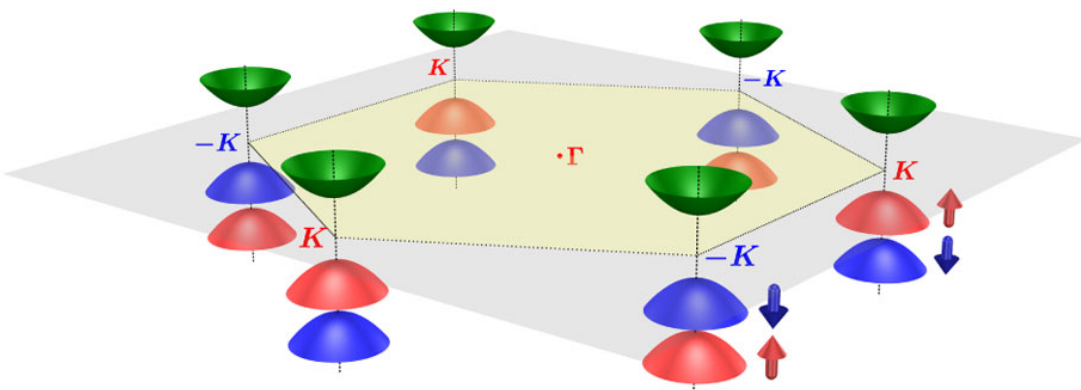
Twisted R-stacked TMD Homobilayer

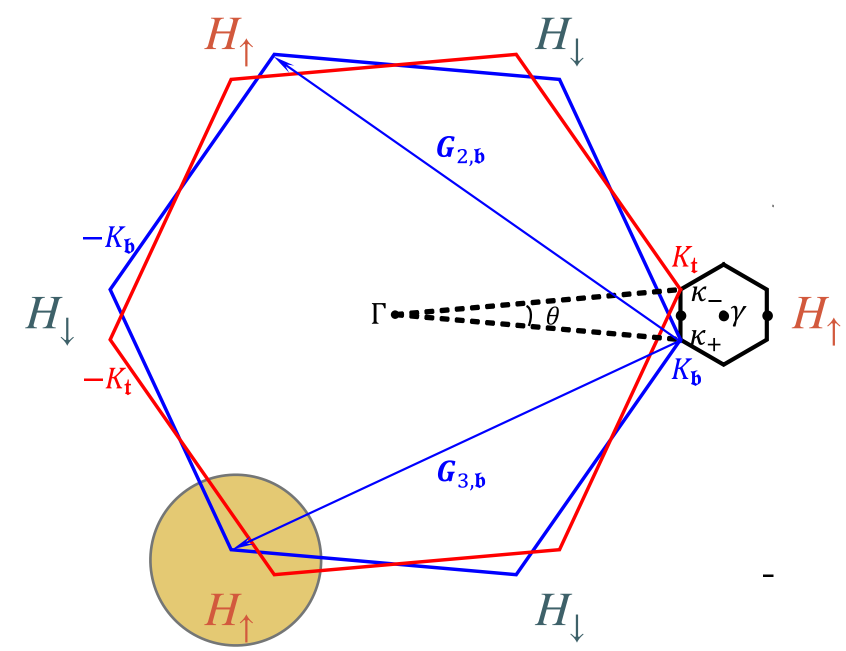


Point group D_3

The variation of the local stacking will lead to **alternating** out-of-plane **electric dipoles** in the MX and XM region, called moire ferroelectricity

Continuum Hamiltonian





$$\mathcal{H}_\uparrow = \begin{pmatrix} -\frac{\hbar^2(\boldsymbol{k}-\boldsymbol{\kappa}_+)^2}{2m^*} + \Delta_{\mathfrak{b}}(\boldsymbol{r}) & \Delta_T(\boldsymbol{r}) \\ \Delta_T^\dagger(\boldsymbol{r}) & -\frac{\hbar^2(\boldsymbol{k}-\boldsymbol{\kappa}_-)^2}{2m^*} + \Delta_{\mathfrak{t}}(\boldsymbol{r}) \end{pmatrix}$$

Top layer Bottom layer

$$\Delta_{b/t}(r) = 2v \sum_{j=1,3,5} \cos(G_j \cdot r \pm \psi)$$

$$\Delta_T(r) = w(1 + e^{-iG_2\cdot r} + e^{-iG_3\cdot r})$$

First harmonic expansion

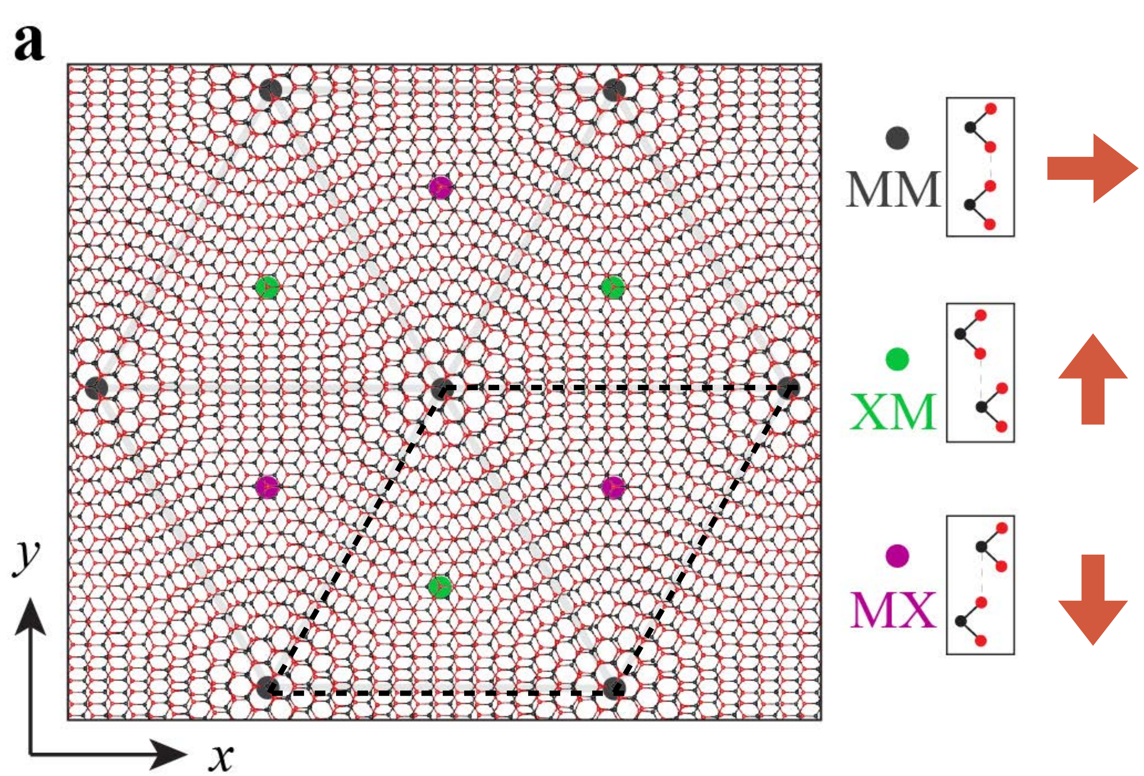
Wu, Lovorn, Tutuc, Martin & MacDonald, PRL (2019)

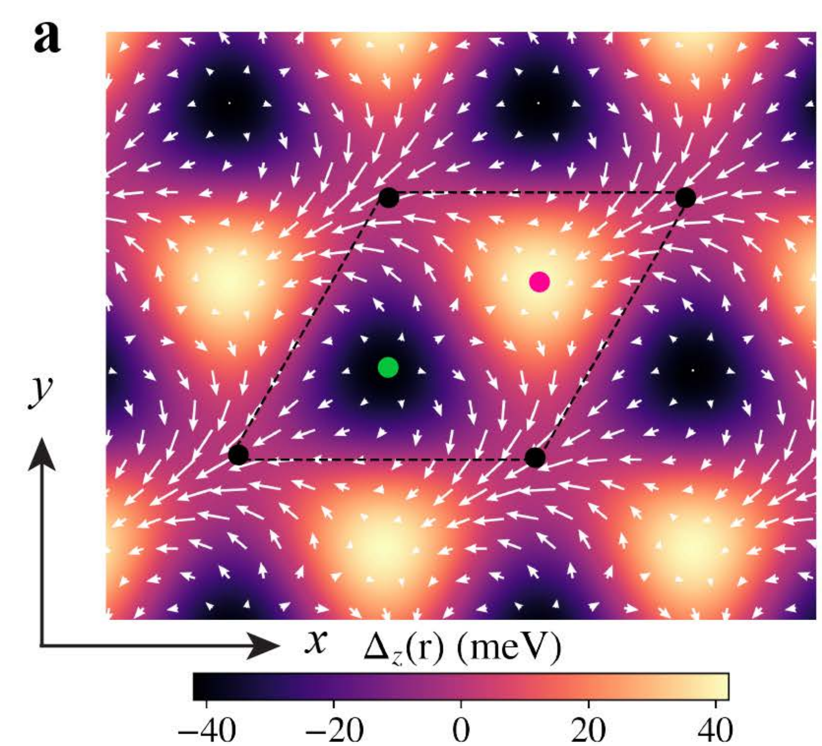
Layer Pseudospin Skyrmions

$$\mathcal{H}_\uparrow = \begin{pmatrix} -\frac{\hbar^2(\boldsymbol{k}-\boldsymbol{\kappa}_+)^2}{2m^*} + \Delta_{\mathfrak{b}}(\boldsymbol{r}) & \Delta_T(\boldsymbol{r}) \\ \Delta_T^\dagger(\boldsymbol{r}) & -\frac{\hbar^2(\boldsymbol{k}-\boldsymbol{\kappa}_-)^2}{2m^*} + \Delta_{\mathfrak{t}}(\boldsymbol{r}) \end{pmatrix}$$

$$\boldsymbol{\Delta} = \left(\mathrm{Re}\Delta_T, \mathrm{Im}\Delta_T, \frac{1}{2}(\Delta_b - \Delta_t)\right)$$

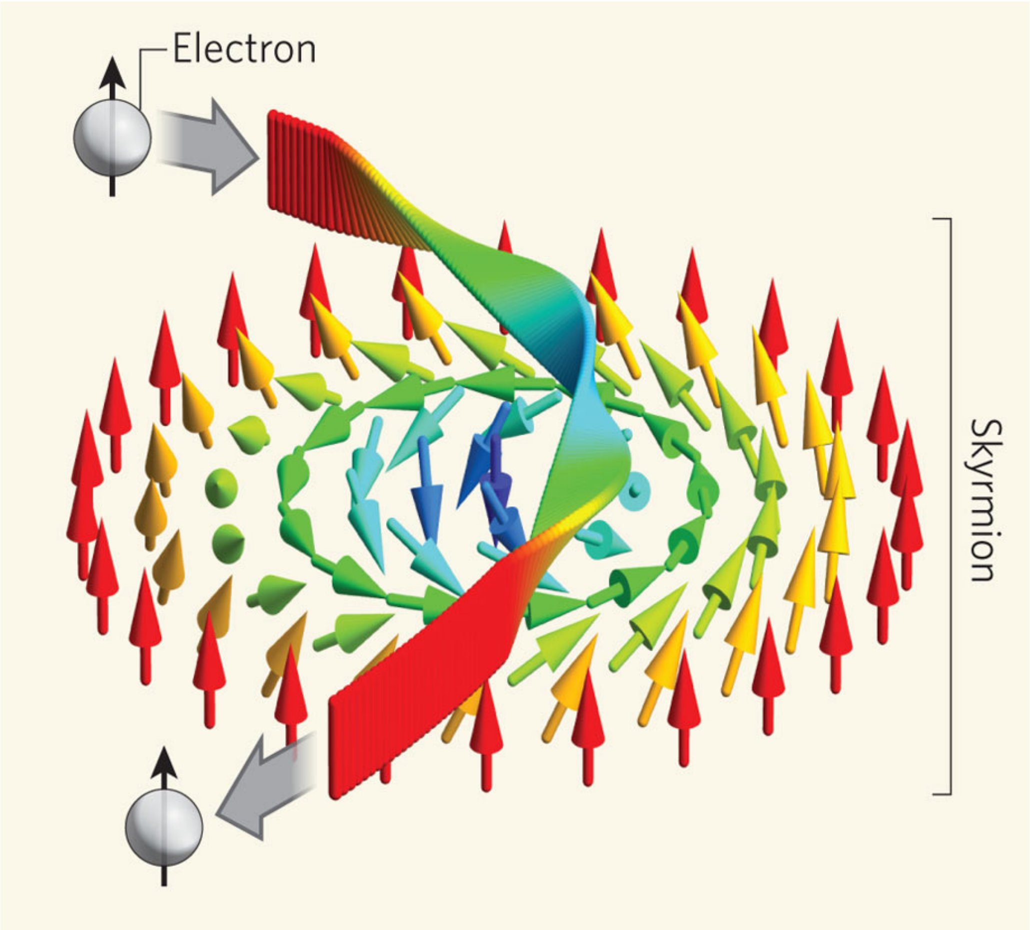
The continuum Hamiltonian describes electrons moving in a pseudospin (layer index) skyrmion texture!





Wu, Lovorn, Tutuc, Martin & MacDonald, PRL (2019); see also Yu, Chen & Yao, Nat. Sci. Rev. (2020)

Topological Hall effect

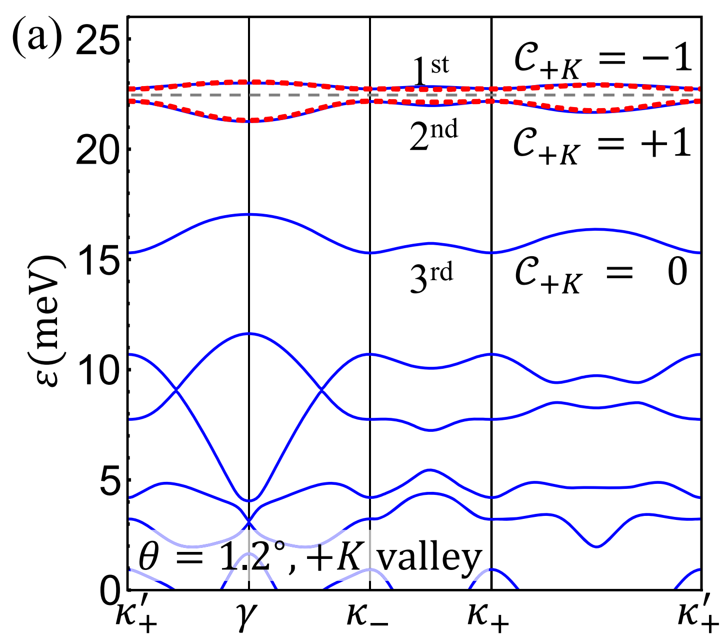


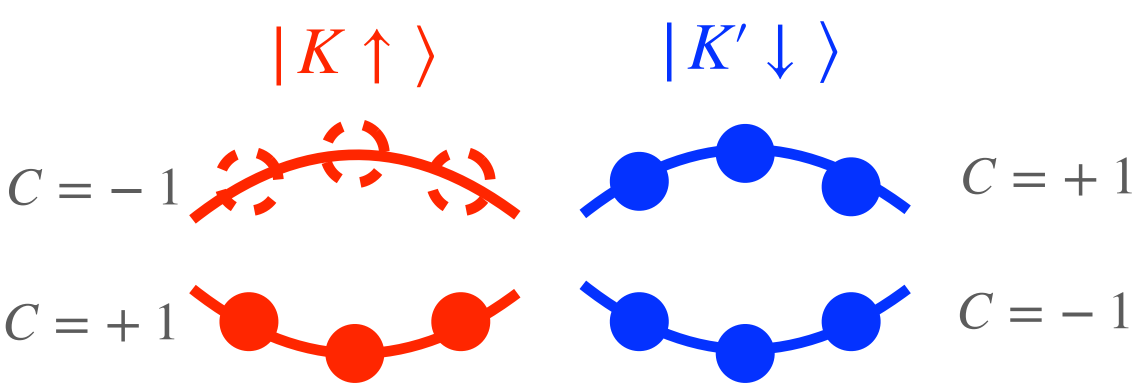
Electrons travel in a spin texture experiences an effective magnetic field, which can result in Landau-level like Bloch bands

$$B_{\mathrm{eff}} = \mathbf{\Delta} \cdot (\partial_x \mathbf{\Delta} \times \partial_y \mathbf{\Delta})$$

Topological Moire Bands

- Non-zero Chern number comes from layer pseudo-spin skyrmions
- Two time-reversal copies with opposite spins and opposite Chern numbers originating from the two valleys (K and K')
- Interaction can then drive the system into various symmetry breaking/topological states





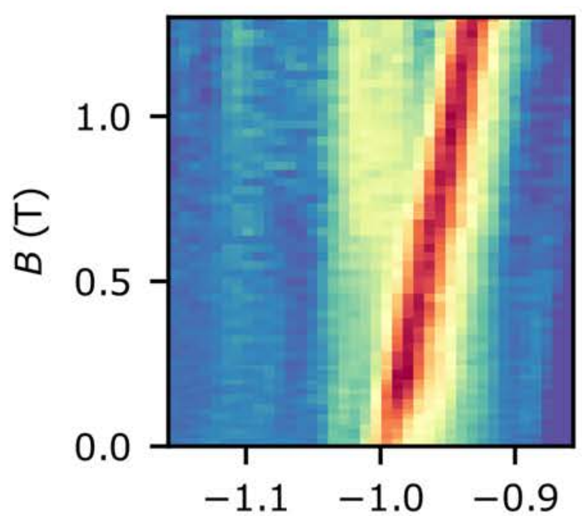
Theory says there should be flat Chern band and fractional quantum anomalous Hall effect

Experiment found it…Where is the problem?

Puzzle I

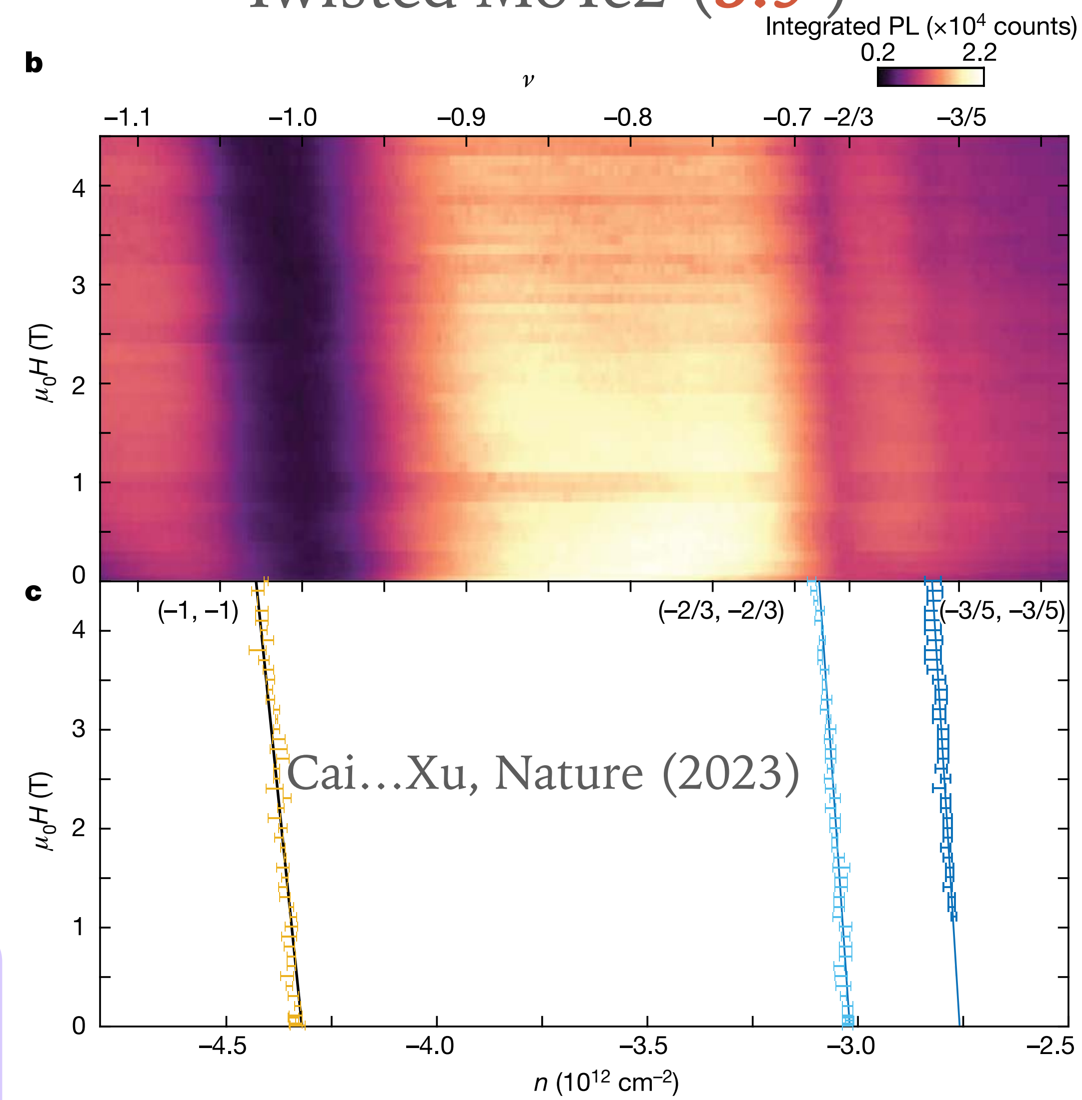
At $\nu = -1$, the Chern numbers in 3.9 degree tMoTe2 and 1.2 degree tWSe2 have opposite sign

Twisted WSe2 (**1.23°**)



Foutty…Feldman et al, Science (2024)

Twisted MoTe2 (**3.9°**)



Cai…Xu, Nature (2023)

The difference probably comes from angle dependence, not material difference

Puzzle II

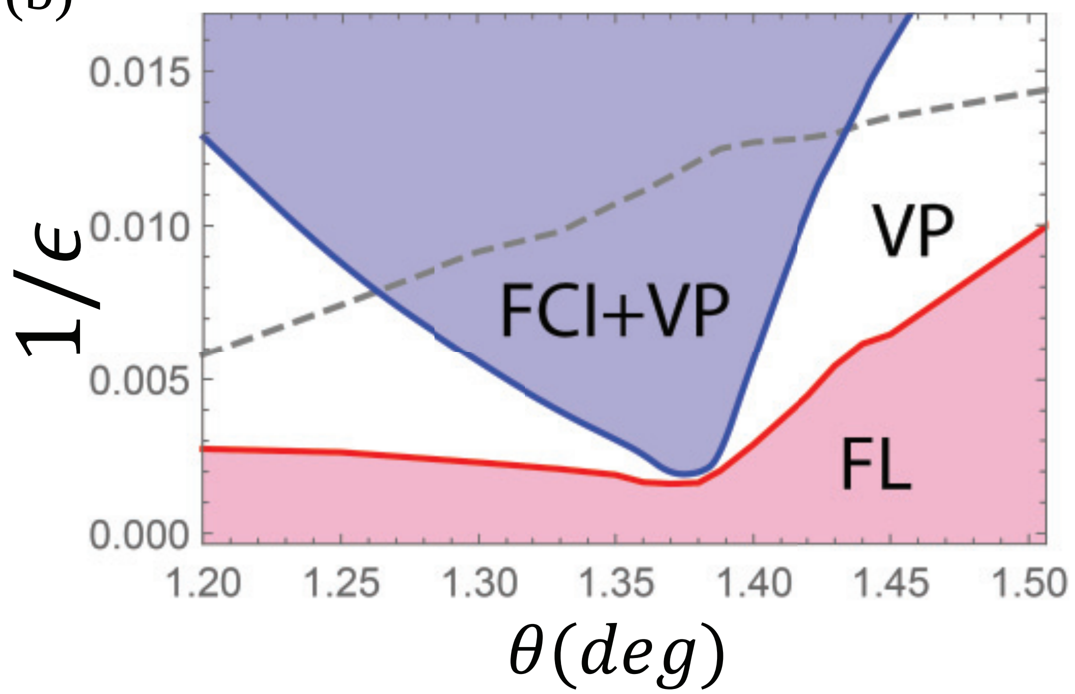
PHYSICAL REVIEW RESEARCH **3**, L032070 (2021)

Letter

Spontaneous fractional Chern insulators in transition metal dichalcogenide moiré superlattices

Heqiu Li,[1] Umesh Kumar,[2] Kai Sun,[1] and Shi-Zeng Lin[3]

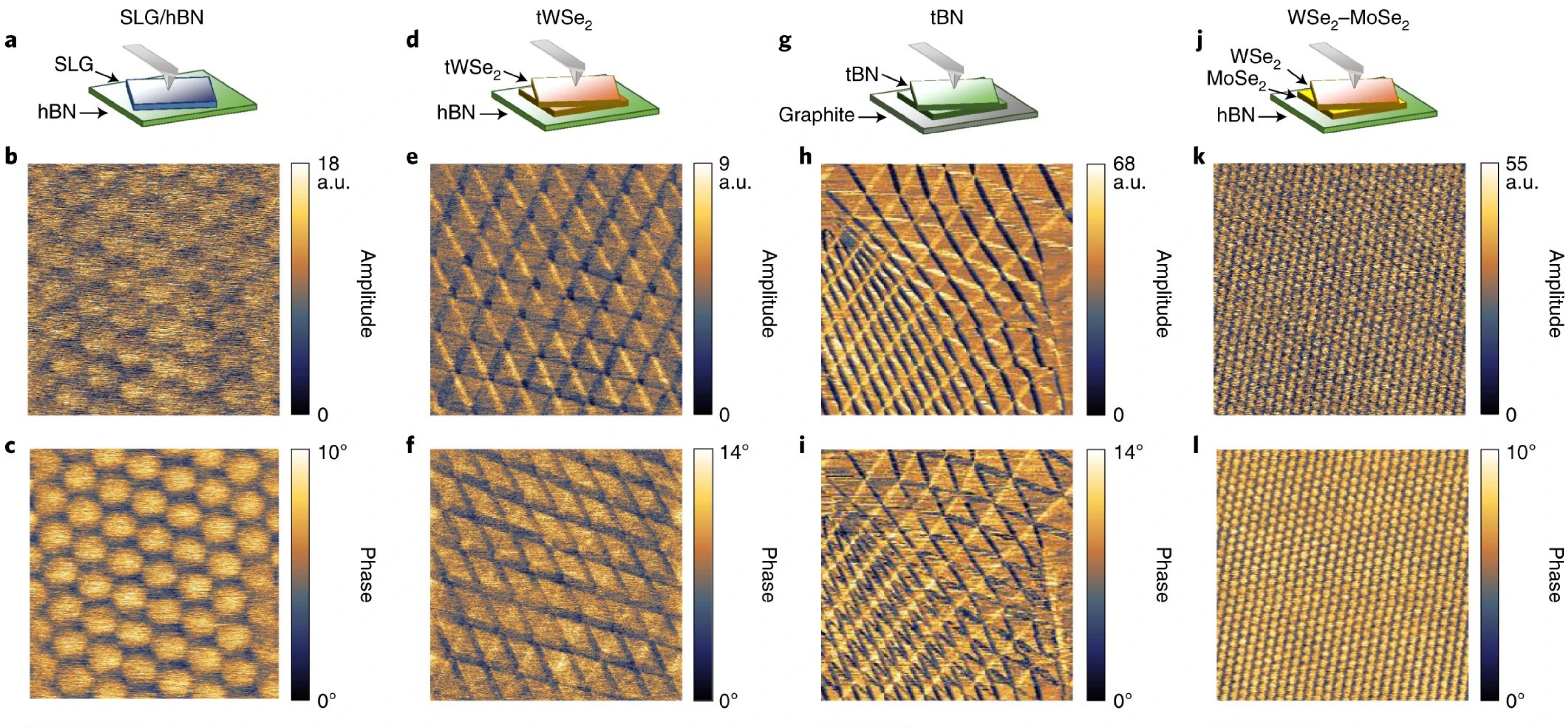
(b)



Using the parameters from MacDonald, the optimal twist angle is around **1.4 degree**, but the experimental twist angle is **3.9 degree**

Lattice Reconstruction

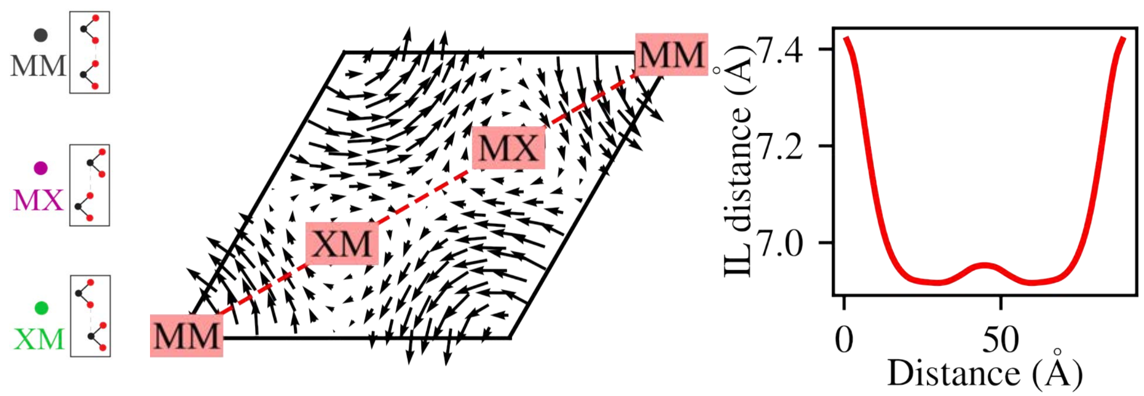
Previous calculations didn't include **lattice reconstruction** effect



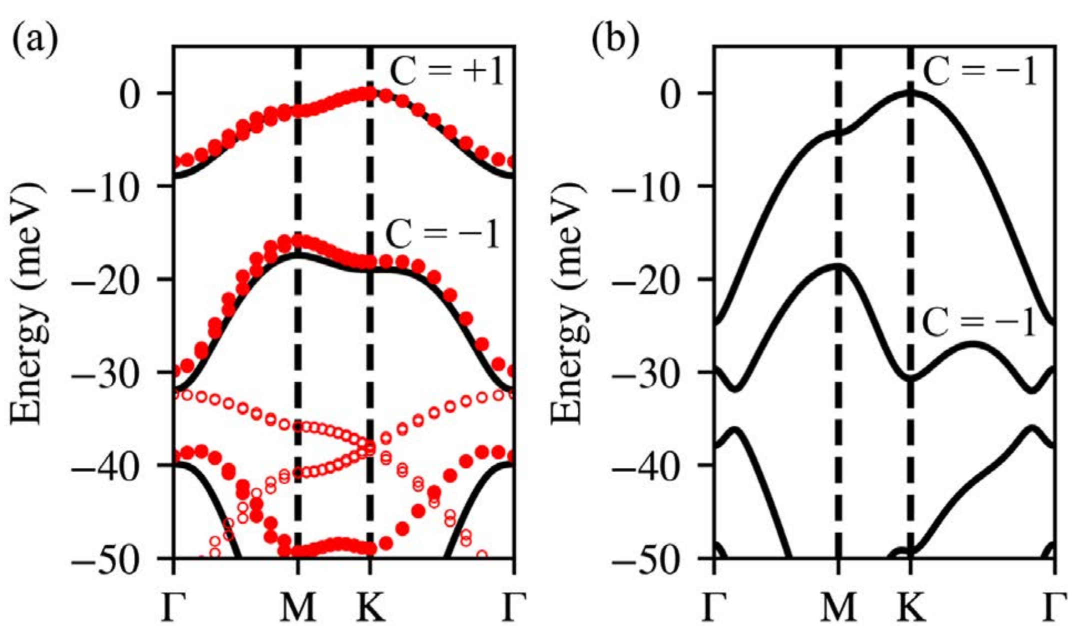
McGilly et al, Nature Nanotech (2020)

Effect of Lattice Reconstruction

In-plane relaxation   Out-of-plane relaxation



$\theta = 3.9°$



Large-scale DFT
w/ lattice relaxation

Local-stacking approx.
w/o lattice relaxation

$$H = \begin{pmatrix} -\frac{(k-K_b)^2}{2m^*} + \Delta_b(r) & \Delta_T(r) \\ \Delta_T^\dagger(r) & -\frac{(k-K_t)^2}{2m^*} + \Delta_t(r) \end{pmatrix}$$

$$\Delta_{b/t}(r) = 2v \sum_{j=1,3,5} \cos(G_j \cdot r \pm \psi)$$

$$\Delta_T(r) = w(1 + e^{-iG_2 \cdot r} + e^{-iG_3 \cdot r})$$

First harmonic approximation

	v (meV)	ψ (°)	w (meV)
Local-stacking approx. [28]	8.0	-89.6	-8.5
Large-scale DFT	20.8	+107.7	-23.8

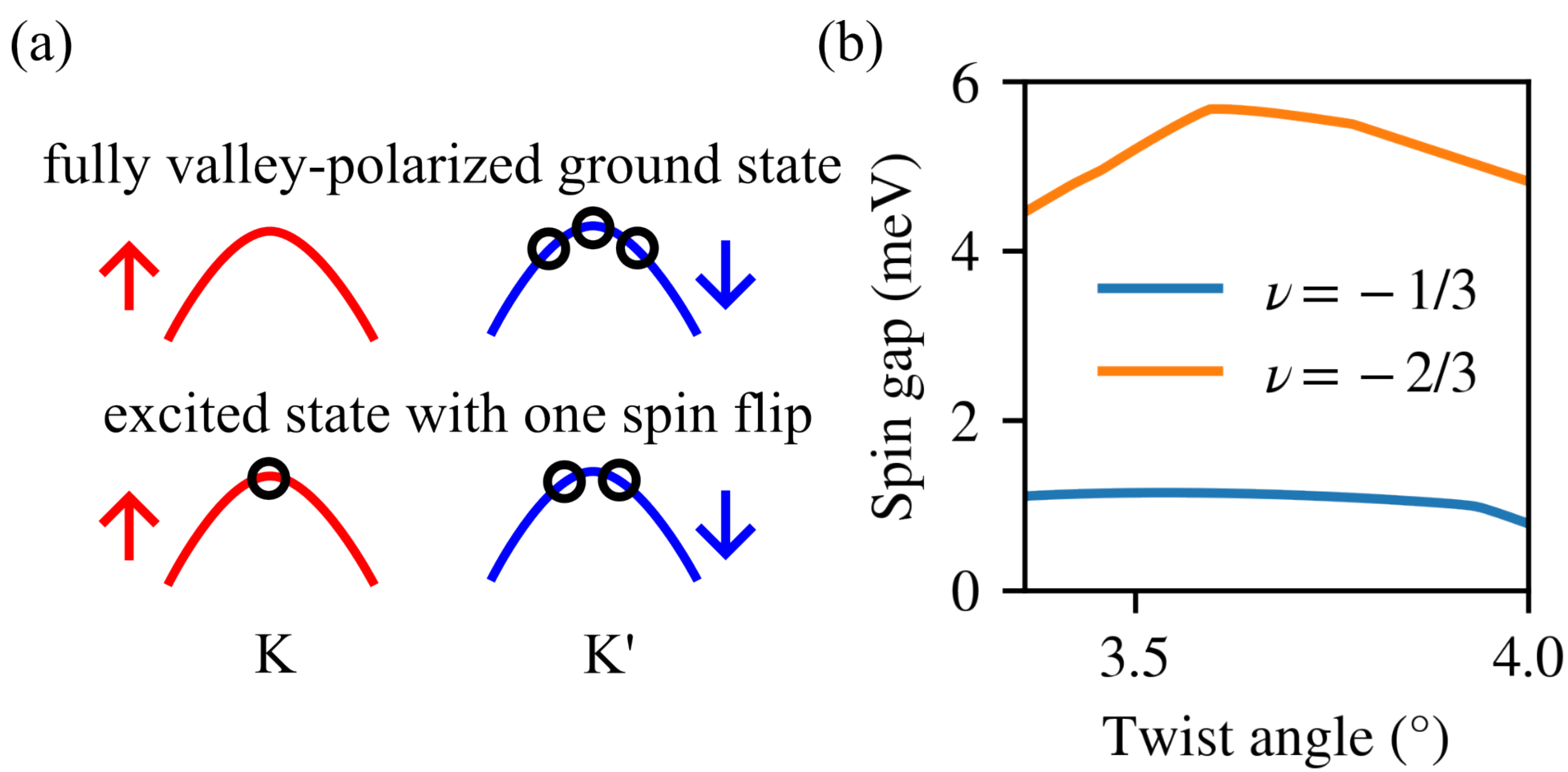
Wang, … Cao & DX, PRL (2024)

Valley Polarization

We now add interaction

$$H_{\text{int}} = \frac{1}{2A} \sum_{l,l',\tau,\tau',\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} V(\boldsymbol{q}) c^{\dagger}_{l\tau\boldsymbol{k}+\boldsymbol{q}} c^{\dagger}_{l'\tau'\boldsymbol{k}'-\boldsymbol{q}} c_{l'\tau'\boldsymbol{k}'} c_{l\tau\boldsymbol{k}}$$

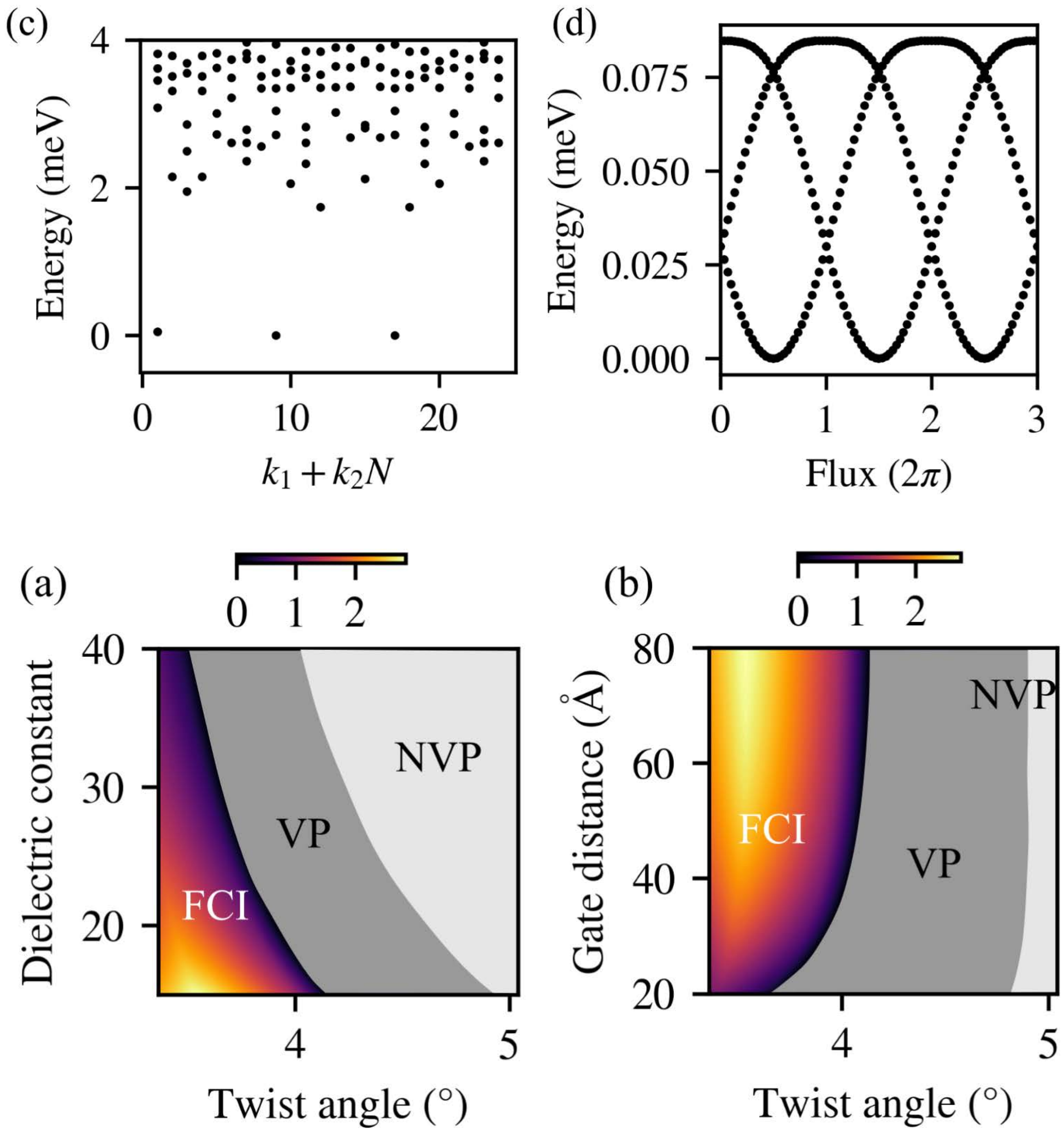
$$V(\boldsymbol{q}) = e^2 \tanh(|q|d)/2\epsilon_0\epsilon|q|$$



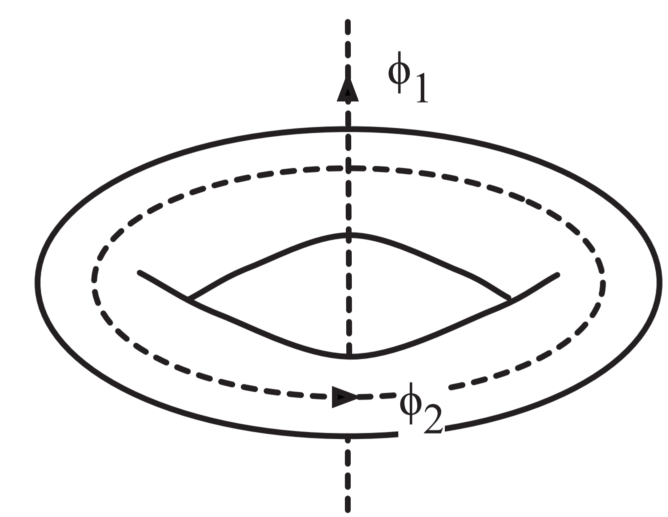
Strong electron-electron interaction lifts valley degeneracy for $\nu \leq 1$

All the dynamics occur within a single valley

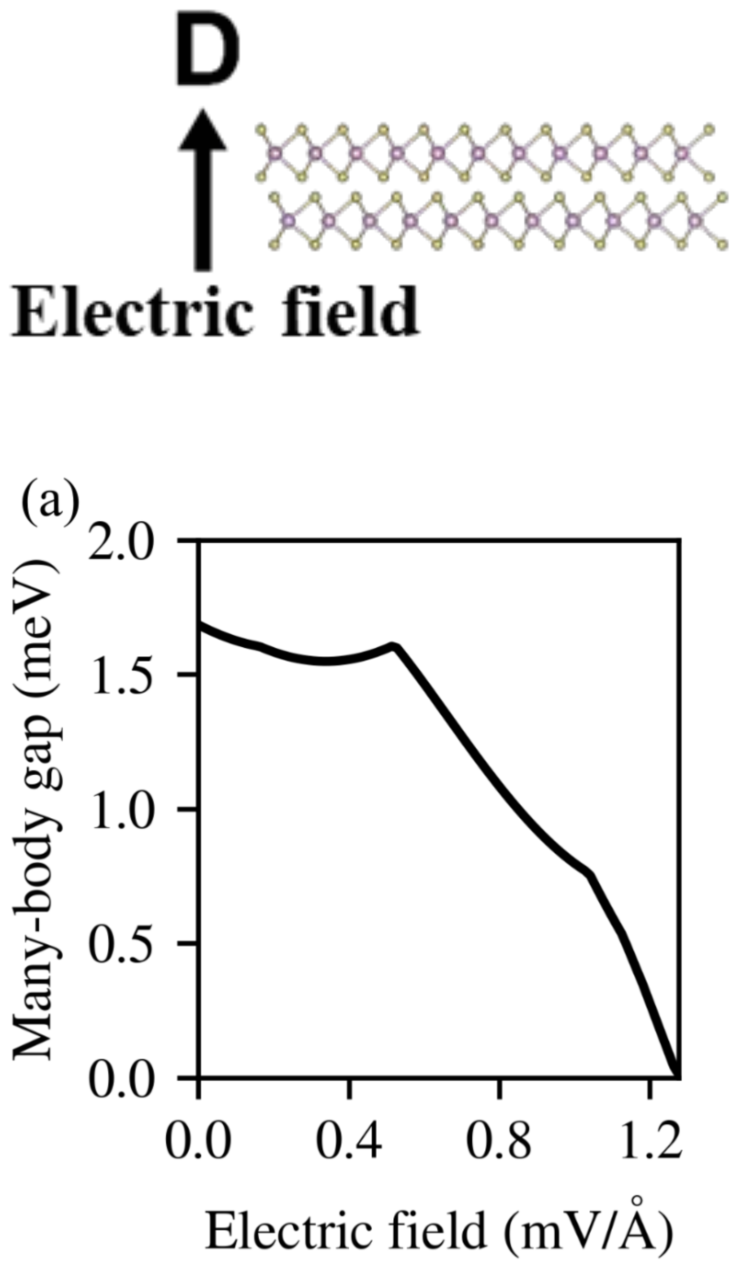
Fractional Chern Insulators in tMoTe2

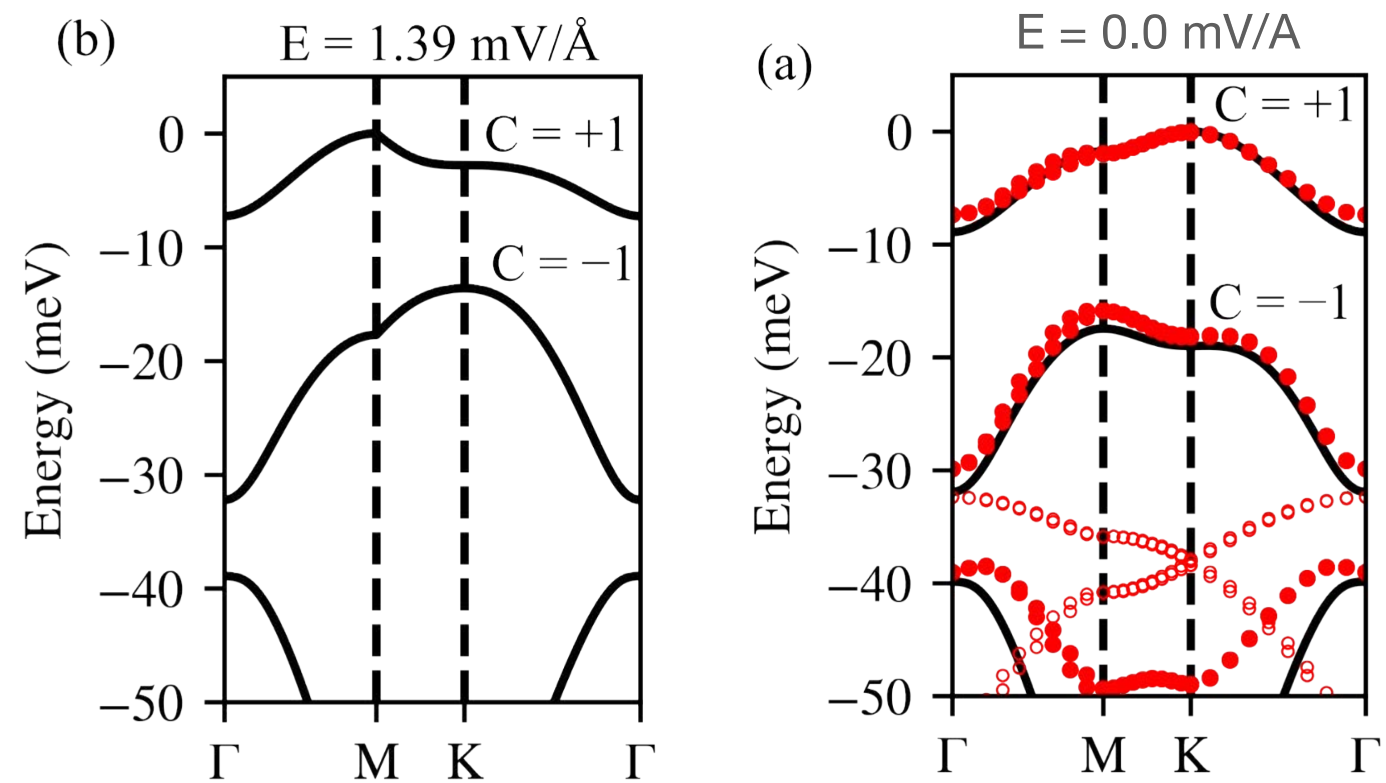


- Exact diagonalization on a torus (4 x 6)
- Single-band projection
- Remote band effects are important (Yu et al., arXiv:2309.14429, Abouelkomsan et al, arXiv:2309.16548)



Filed Tuning of the Fractional Chern Insulator State





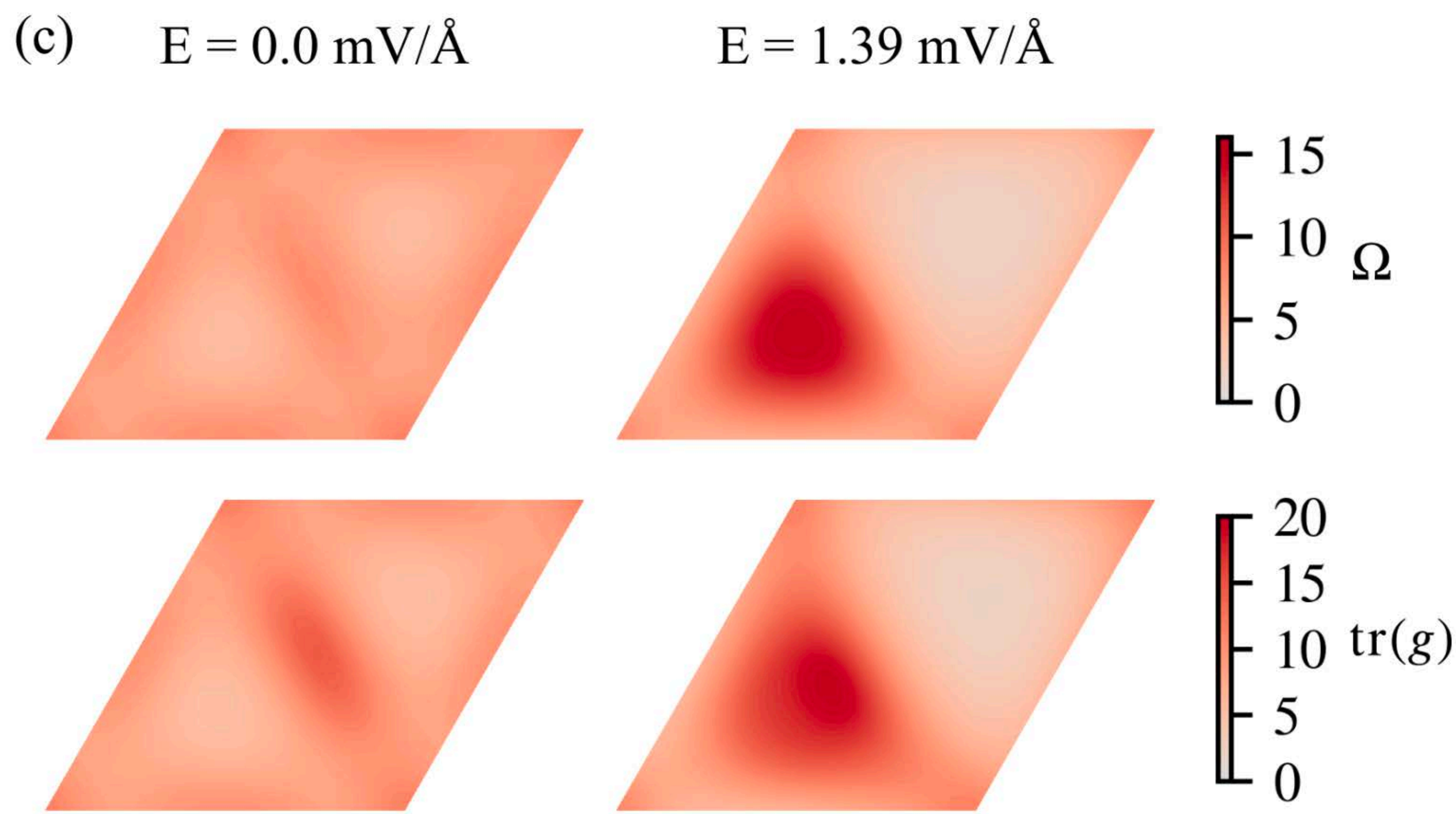
The band structures below and above the critical electric field are very similar to each other, in terms of both their band width and Chern numbers.

Electric field dependence of FQAH

FQAHE | No FQAHE

Berry curvature

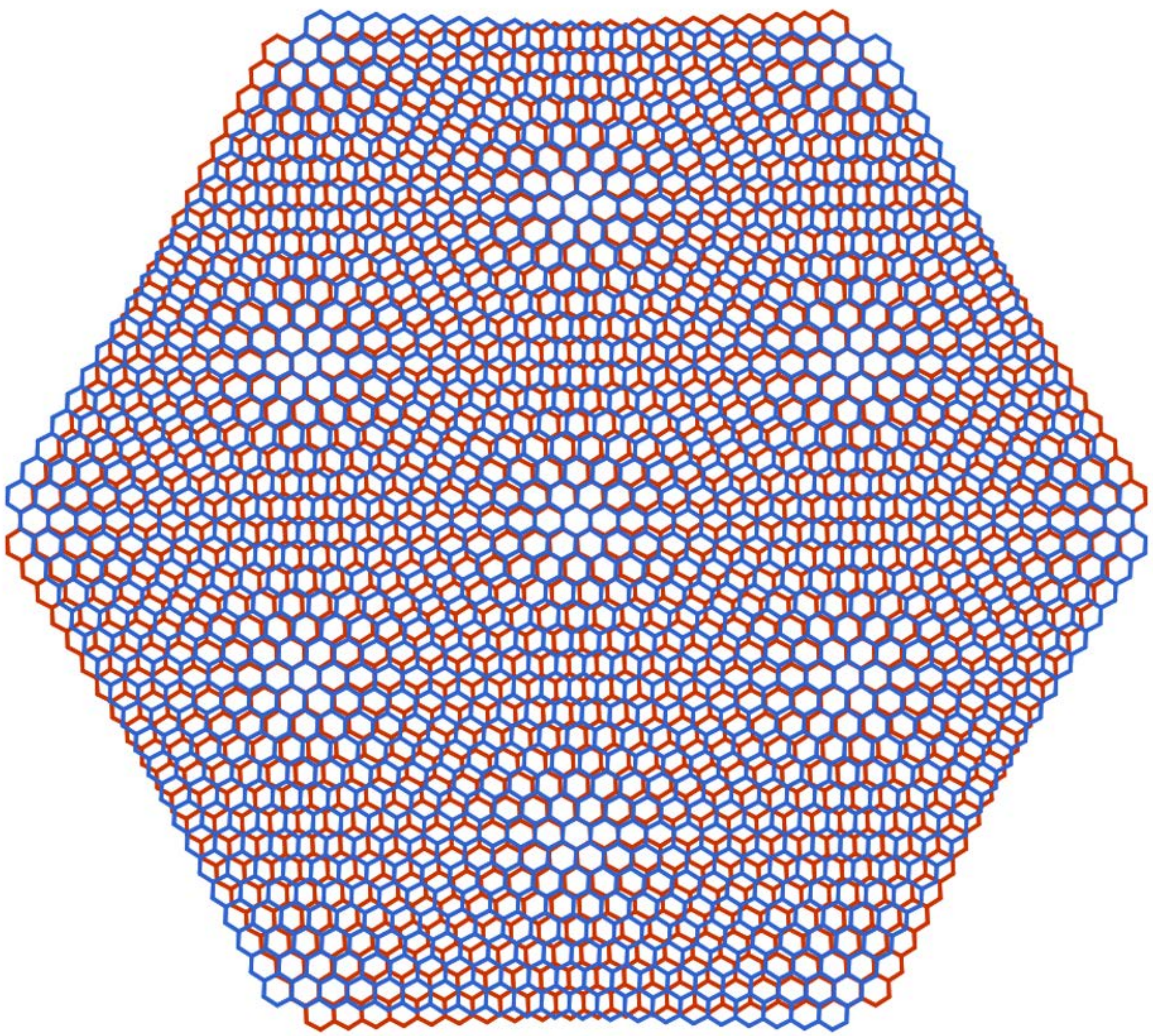
Quantum metric



The Berry curvature Ω and and quantum metric tensor g are constant for Landau levels. The flatness of these two quantities in the k-space is heuristically viewed as a promising indicator for the emergence of FCIs.

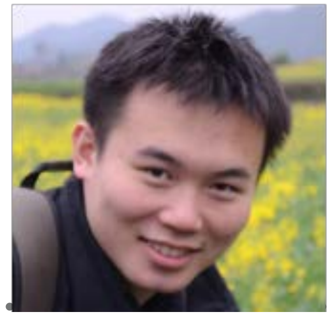
Lattice reconstruction fundamentally reshape the electronic structure

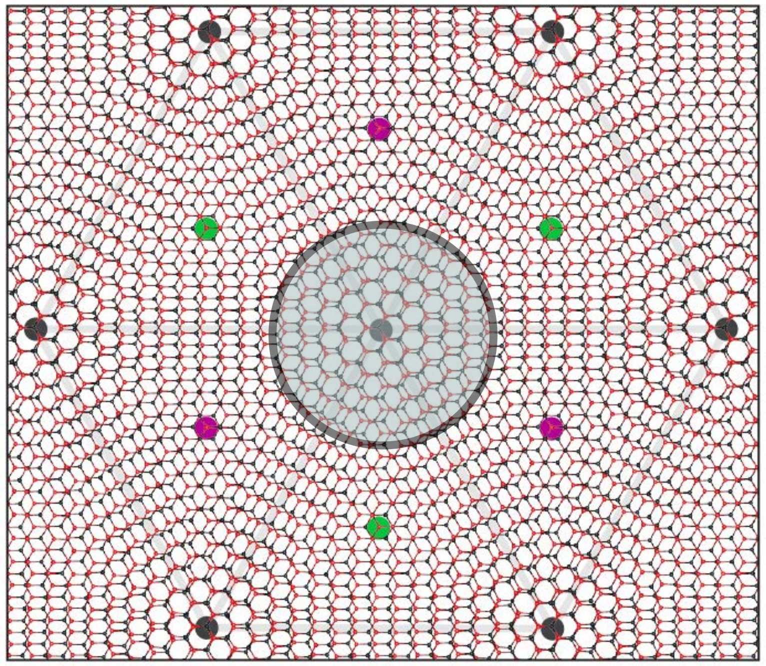
What about small twist angles?



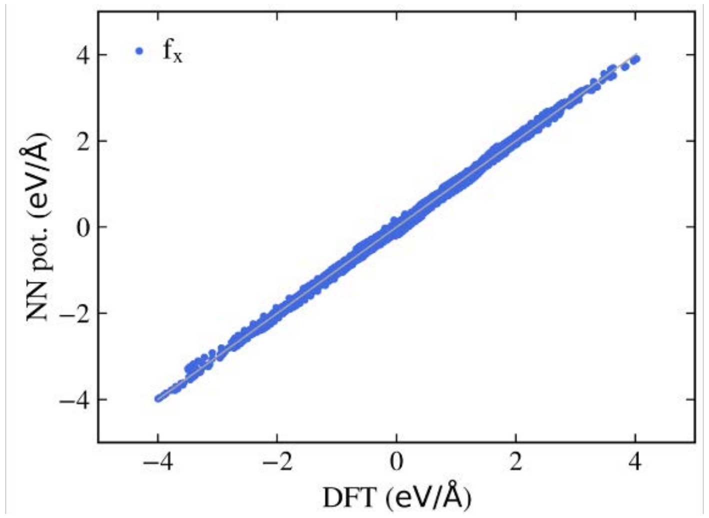
At 1.5 degree twist angle, the moire period is ∼ 10 nm, and the moire unit cell contains more than 10,000 atoms. Direct DFT relaxation is not possible!

Machine Learning to the Rescue


Ting Cao

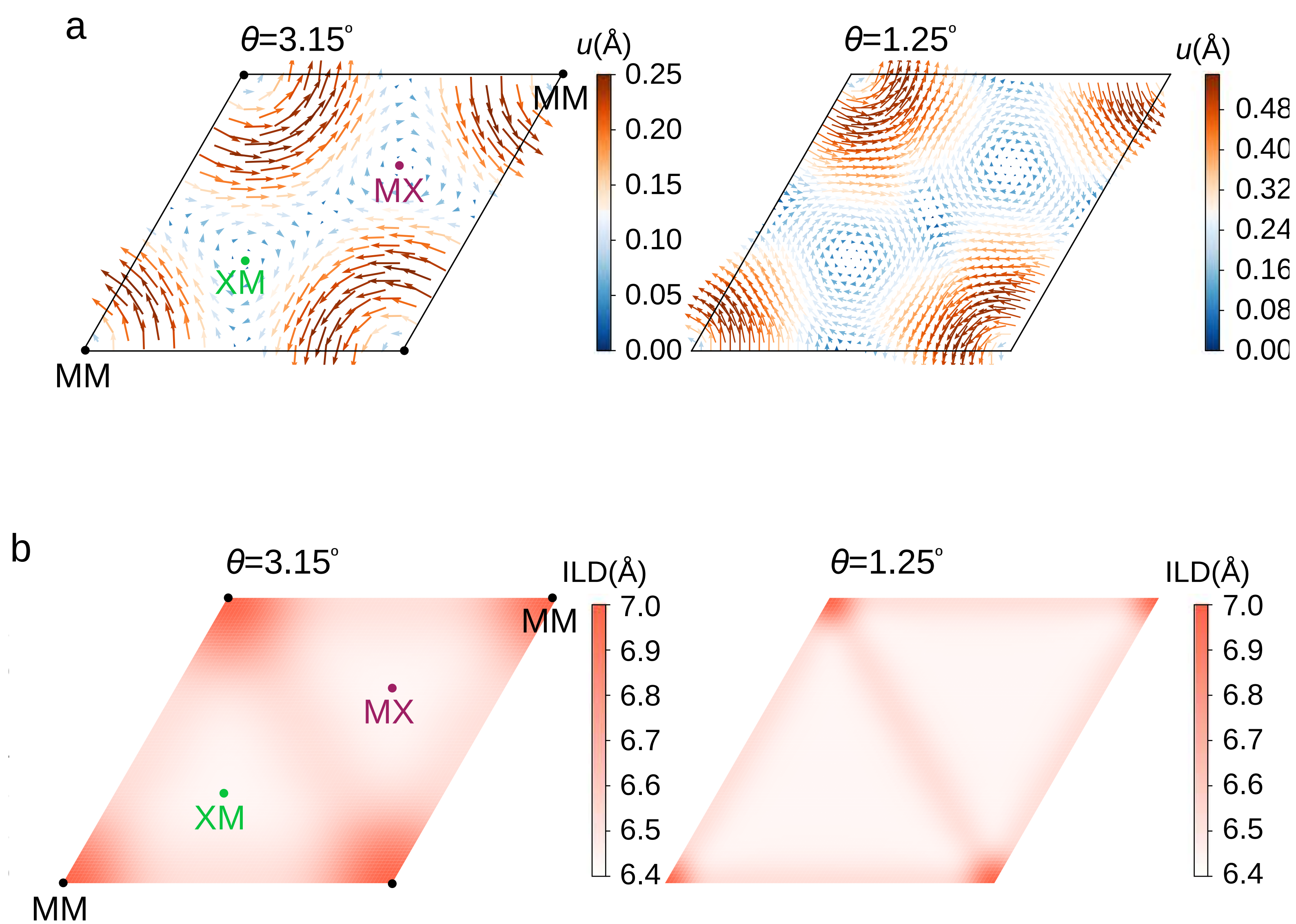


- The total energy is a function of all atom positions $E(r_1, r_2, \cdots r_{10,000})$. Assuming the energy is local, we choose a cut-off of 10 Å
- Training data is obtained at 6° twist angle, 5,000 MD steps at 500 K
- Verified at 5° twist angle



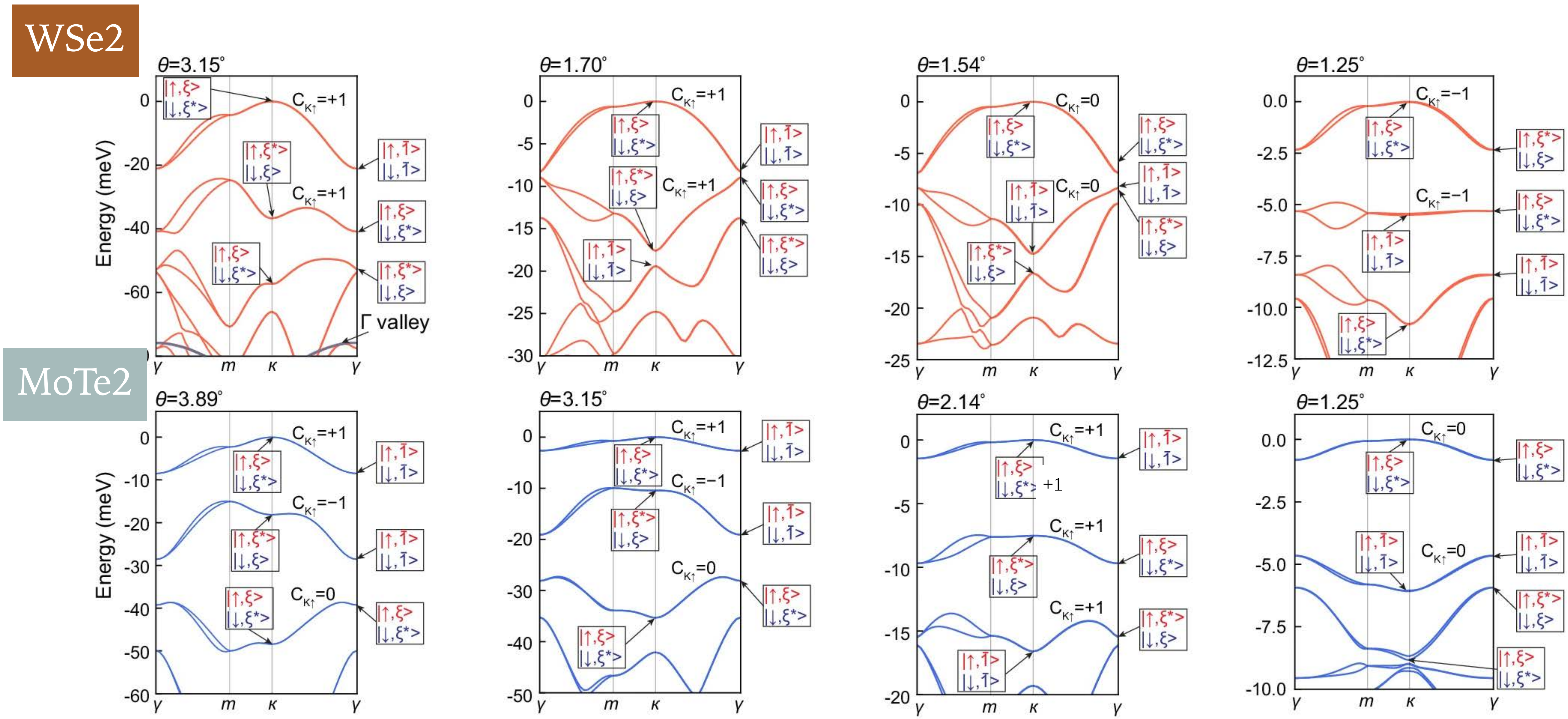
Data+Training takes ~2 weeks, direct DFT relaxation would take years (may not even converge to global minimum)!

Lattice Reconstruction



Twist-Angle Dependent Moiré Band Structure

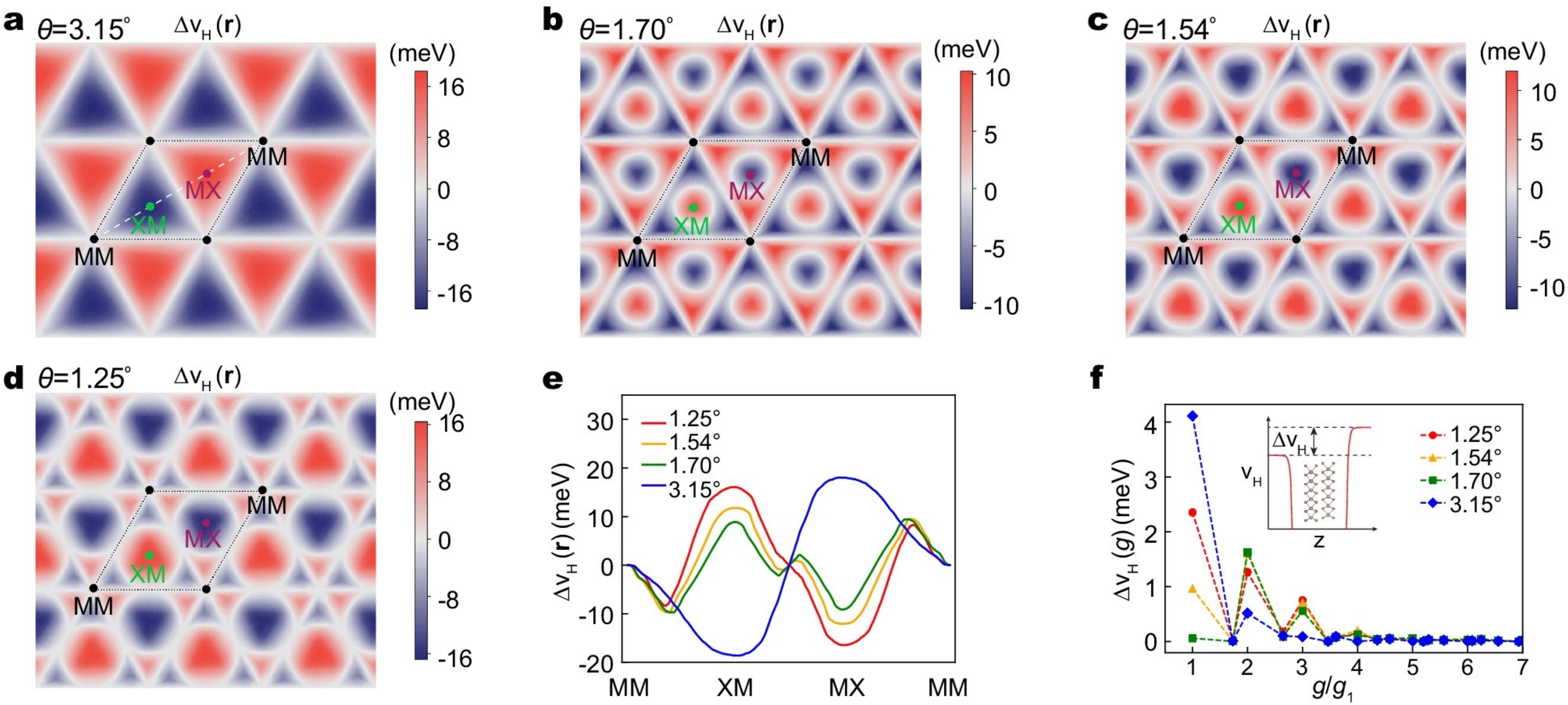
$$\exp(i\frac{2\pi}{3}C) = -\,\xi_\gamma \xi_\kappa \xi_{\kappa'}$$



Zhang, Wang, Liu, Fan, Cao & DX, Nature Comm. (2024)

Back To Real Space: Visualizing Moiré Potential

The moiré potential flips sign at the MX and XM points!

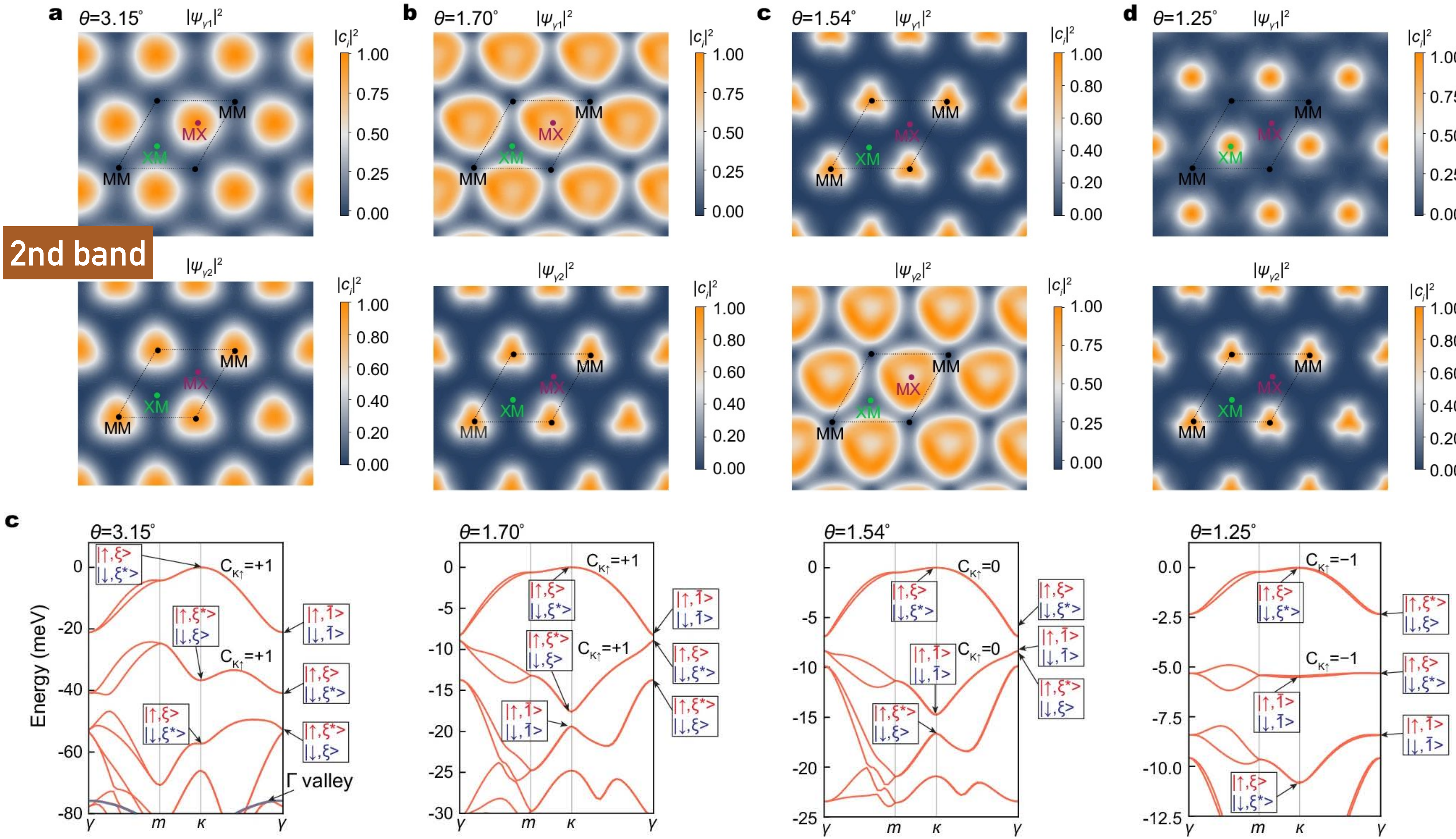


First harmonic approximation is not enough, higher G expansion is necessary.
See Jia, … Bernevig, & Wu, PRB (2024).

Wave Function (tWSe2)

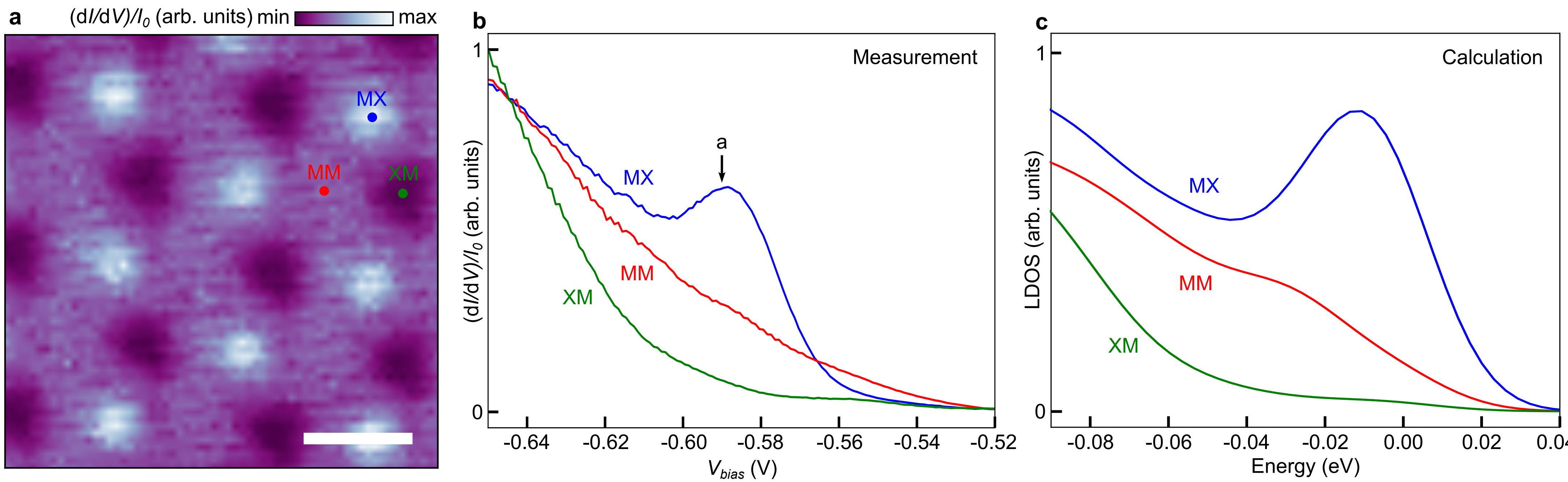
1st band

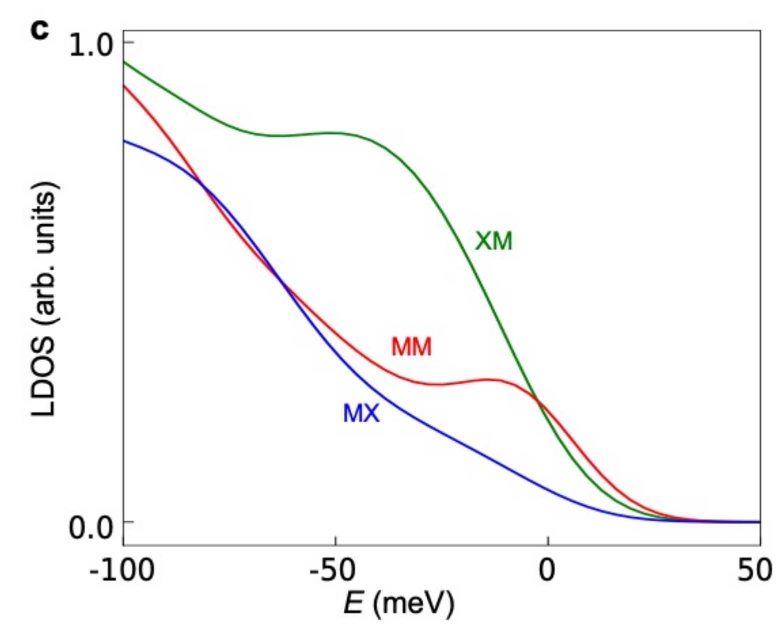
2nd band



Experimental Evidence

STM probe of layer localization of band edge





Without in-plane relaxation the position of MX and MX switch

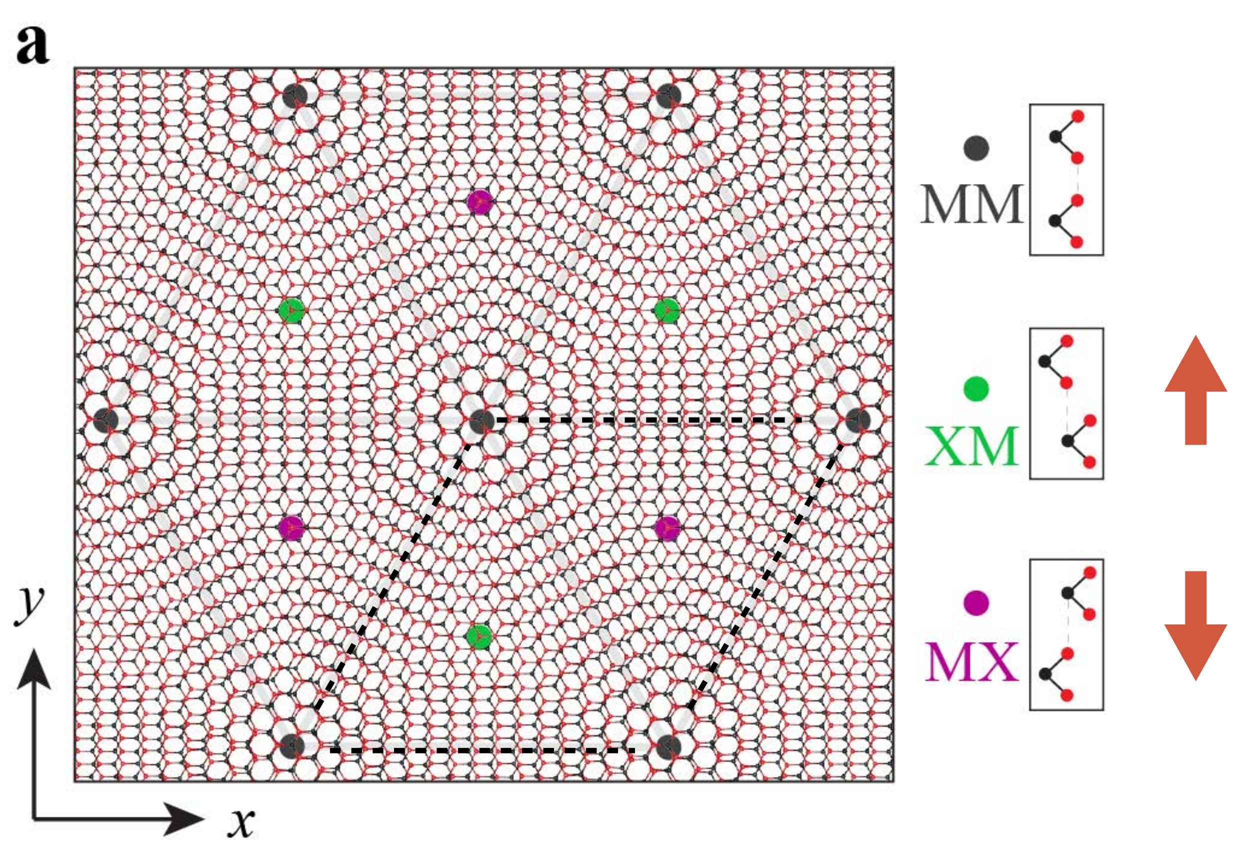
Thompson, DX, Yankowitz, Nat. Phys. (Accepted)

Excluding in-plane relaxation


But
Why?!

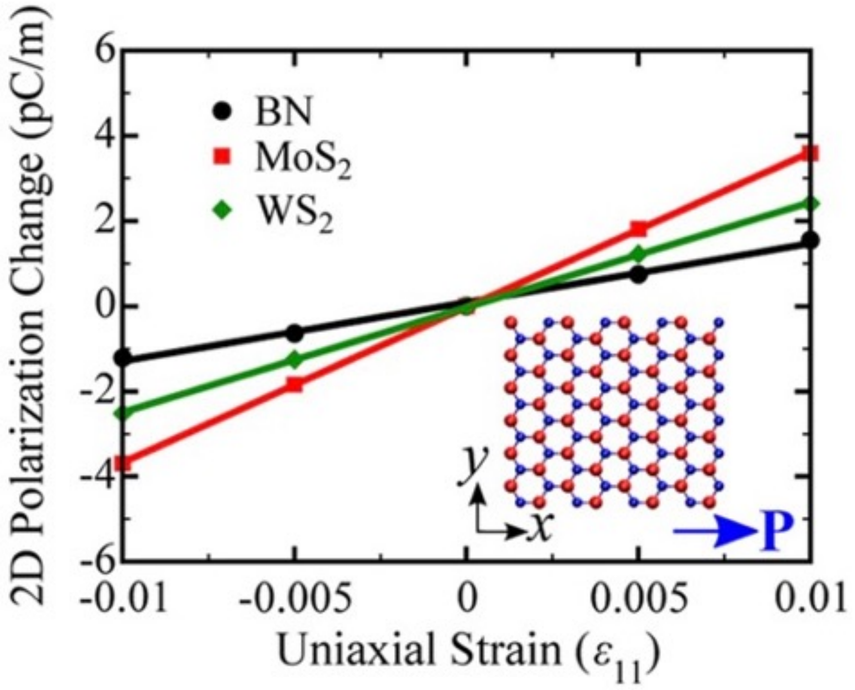
Interlayer Polarization

- In a charge neutral system, the potential difference between the top and bottom surface must comes from interlayer dipoles
- Where does the interlayer dipole come from?



If the dipole only comes from stacking caused ferroelectricity, it will never flip!

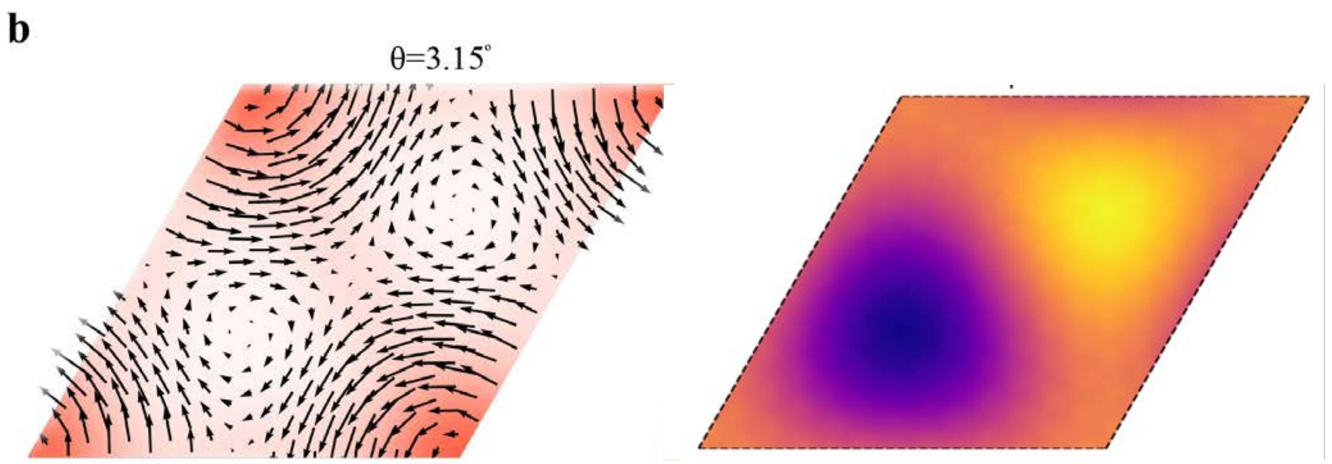
Piezoelectricity

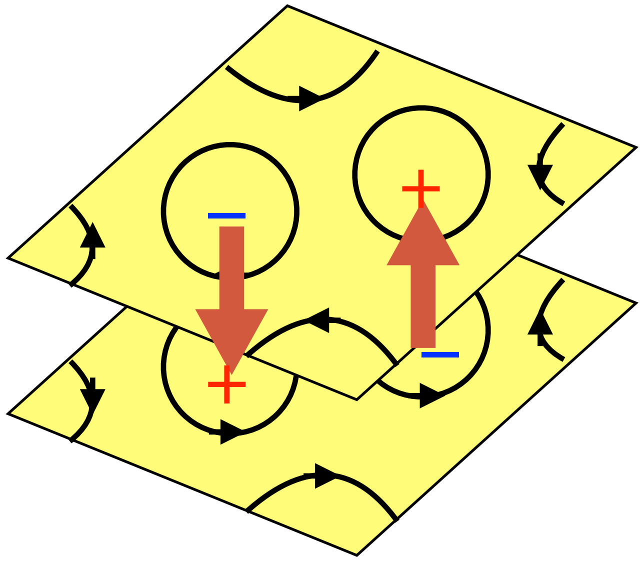


Monolayer TMD breaks inversion symmetry, therefore it is piezoelectric active

$$\rho_{\text{piezo}} = e_{11}[2\partial_x u_{xy} + \partial_y(u_{xx} - u_{yy})]$$

Duerloo, Ong, Reed, JPCL (2012)





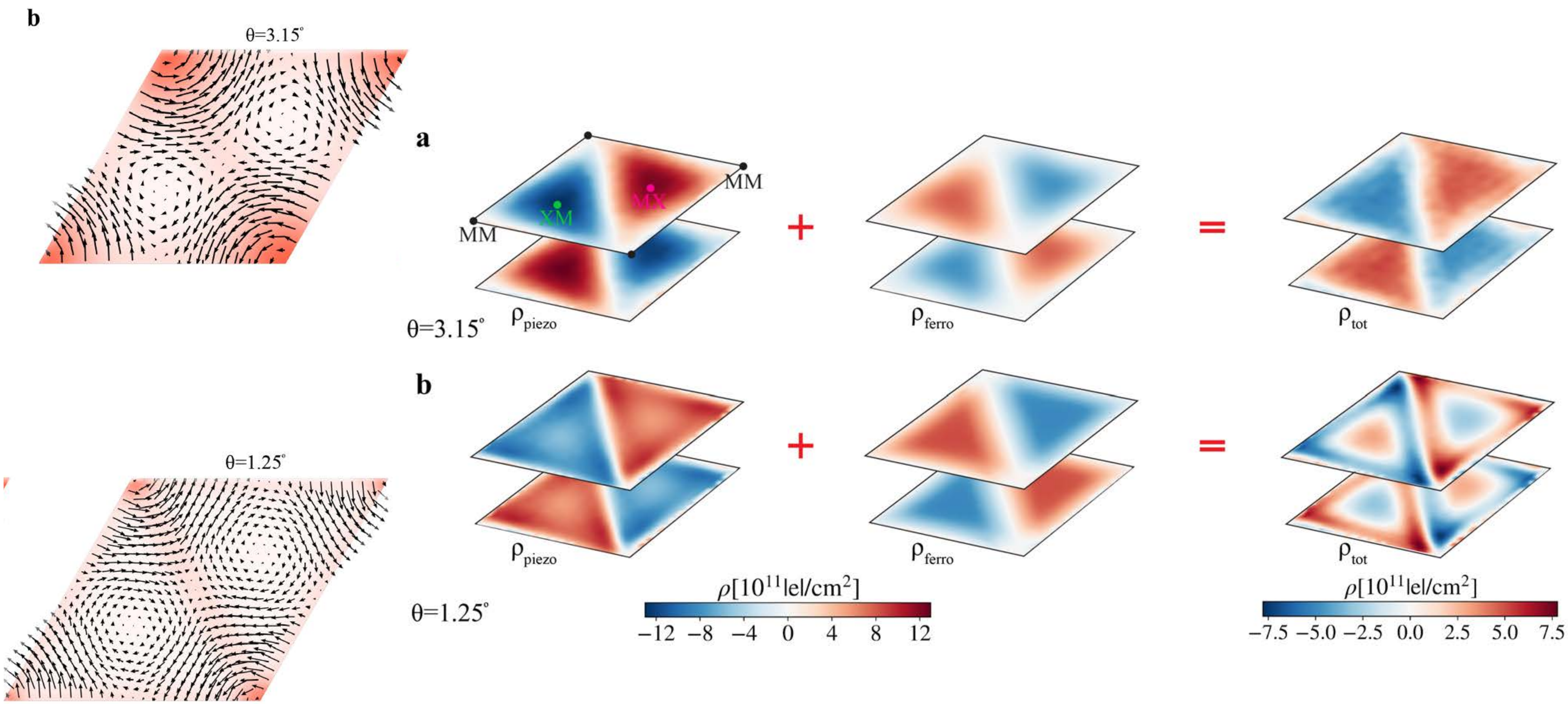
Displacement field

Piezoelectric charge

The competition between ferroelectric charge and piezoelectric charge was first discussed in Enaldiev … Falko, PRL (2020)

Piezoelectricity vs Ferroelectricity

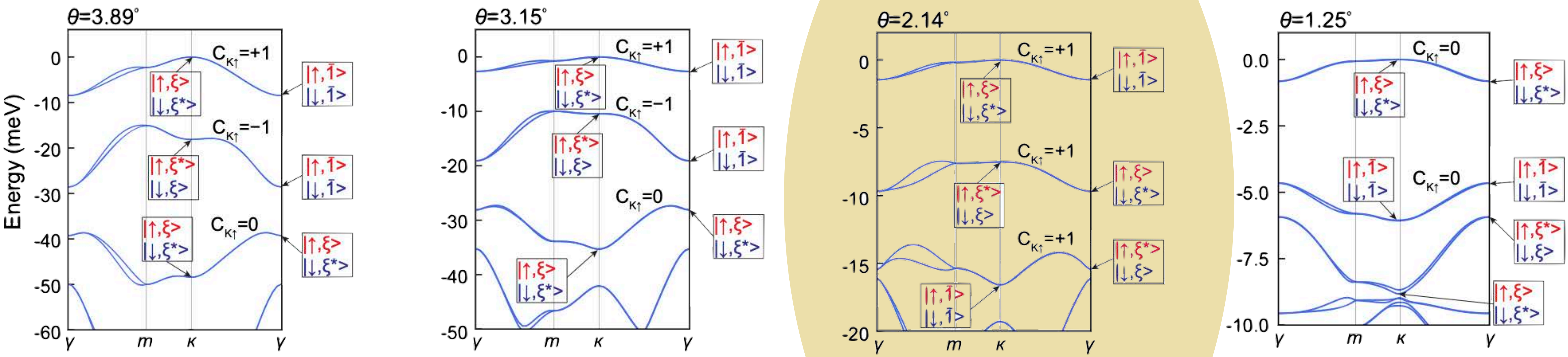
Zhang… Cao, DX, Nature Comm (2024)



Note that since graphene is inversion symmetric, the physics discussed here does not appear in twisted graphene systems

Large θ Small θ

Magic continuum: FCI everywhere

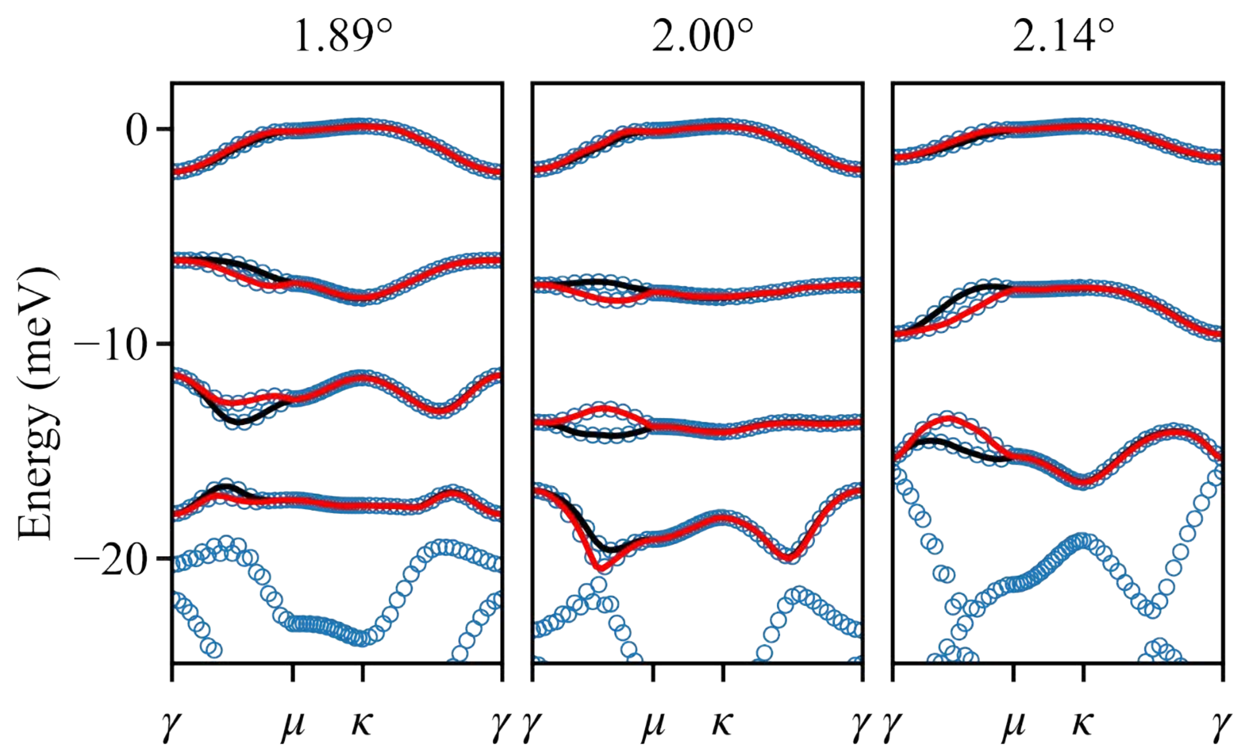


Agrees with the conductance measurement at $\nu = -2, -4, -6$ by Kang et al Nature (2024)

Are these bands analogues of a set of Landau levels?

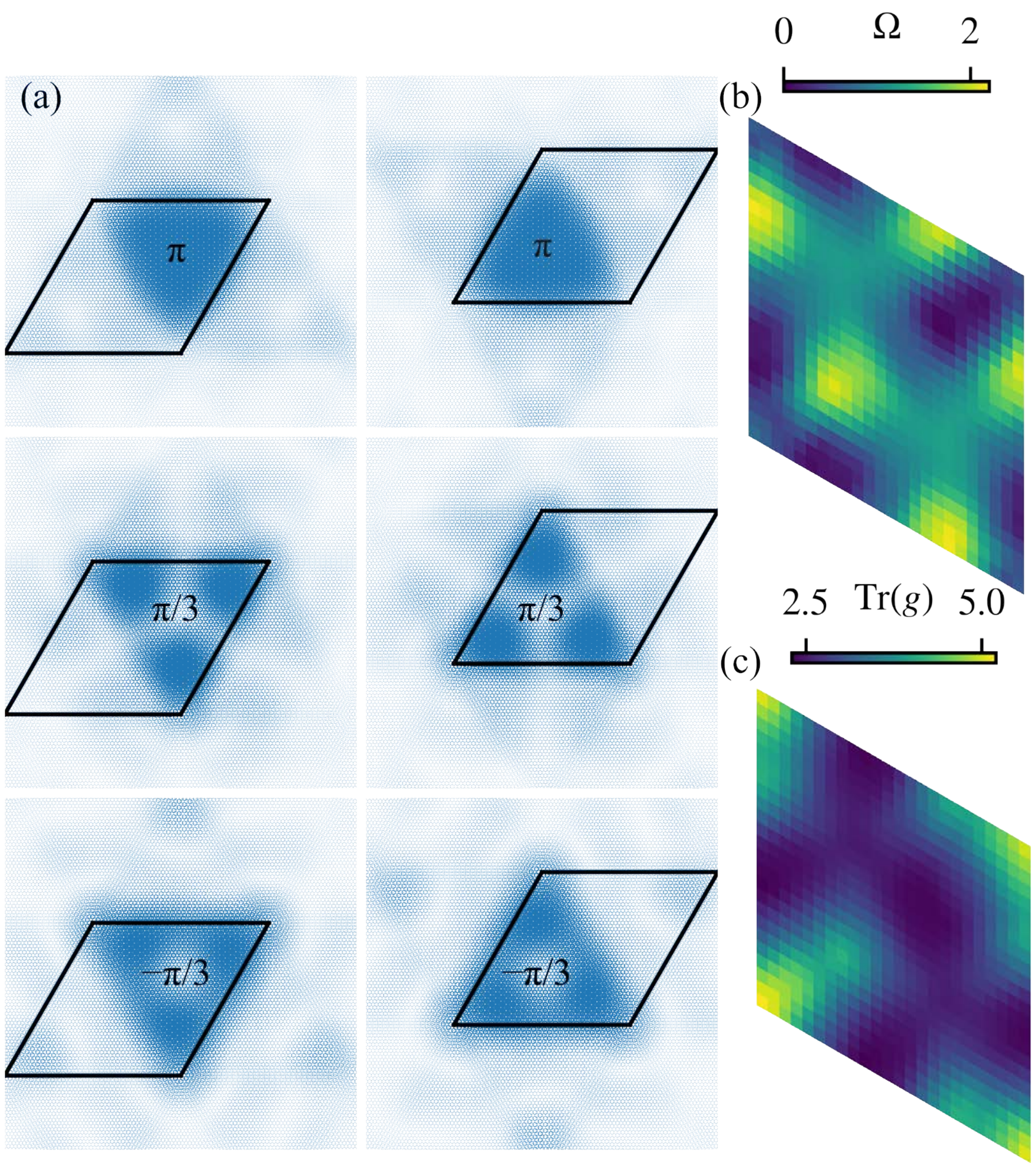
Higher Landau Level Physics

- There is a small twist angle range in which multiple flat C=1 bands appear.
- Are these bands analogous to a Landau level set? If so, is it possible to realize higher LL physics?



Ideal Landau levels has C = 1, and $\chi = \frac{1}{2\pi}\int d\boldsymbol{k}\,\mathrm{Trg}(\boldsymbol{k}) = 2n + 1$

Quantum Geometry from Wannier Function

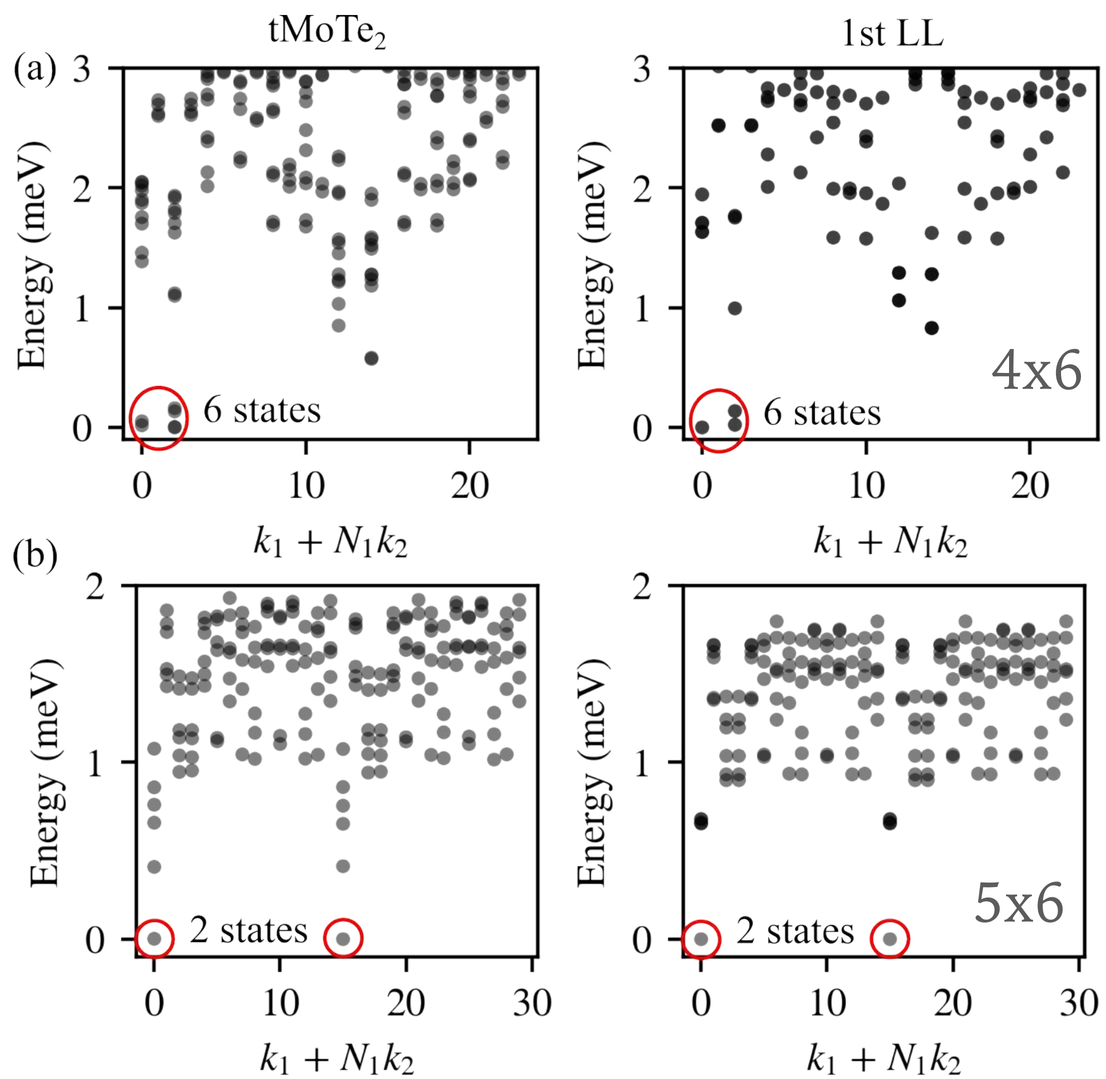


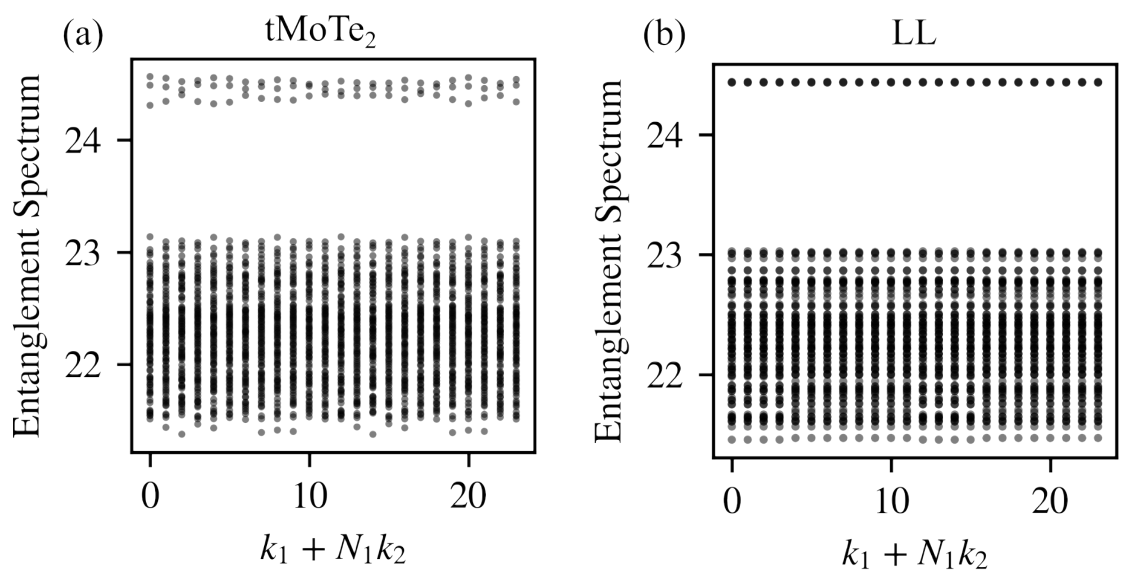
- Fitting to continuum model is a messy business (higher G coefficients are needed!). Instead we construct Wannier functions directly.
- For twist angle $\theta = 2^{\circ}$, we have $\chi = 1.04, 3.09, 5.11, 7.53$.

Wang et al. arXiv:2404.05697

For a plane wave projection fitting, see Zhang et al, arXiv:2411.08108

Many-Body Spectrum for half-filled second Moire band





Particle-cut entanglement spectrum

Wang et al. arXiv:2404.05697.

See also Reddy et al, arXiv:2403.00059; Ahn et al, arXiv:2403.19155; Xu et al, arXiv:2403.17003

Are multiple LL-like bands bound to happen?

- The skyrmion model of MoTe2 indicates that in the strong coupling limit (when the local spin splitting is large) we can view electrons as moving in an effective magnetic field $B = \mathbf{\Delta} \cdot (\partial_x \mathbf{\Delta} \times \partial_y \mathbf{\Delta})$. If B is sufficiently uniform, then we should have multiple LL-like bands. [see Morales-Duran et al, PRL (2024); Reddy et al, PRL (2024)]
- But does it bound to happen as twist angle changes?

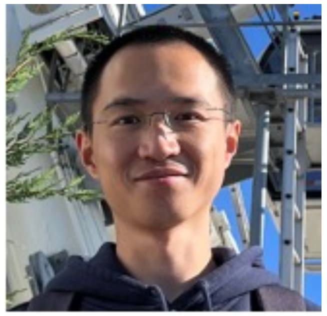
$$H = H_{\text{local}} + H_{\text{long range}}$$

H_{local}: Effective mass, inter layer tunneling. Parameters here are θ independent

$H_{\text{long range}}$: Polarization charges, which can be computed from the lattice relaxation pattern, and its associated potential can be obtained by solving the Poisson equation. This term is strongly θ dependent. Since it is long range, ML packages such as DeepH cannot accurately capture this part.

Summary

- Machine-learning based approach provides a powerful tool to study moiré superlattices
- The competition between stacking ferroelectricity and piezoelectricity determines the band topology
- Strain could potentially be an effective tuning knob
- Higher-level physics is possible around $\theta = 2^{\circ}$



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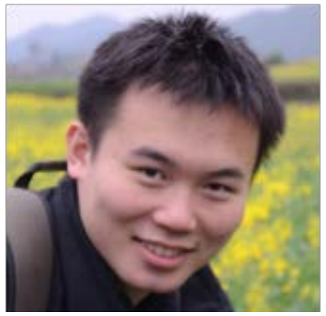

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