New twists on magnetism in flatland

Leon Balents, KITP

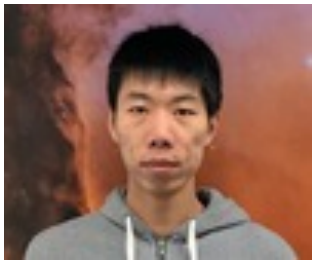
UMinn Colloquium, 4/1/21

Collaborators

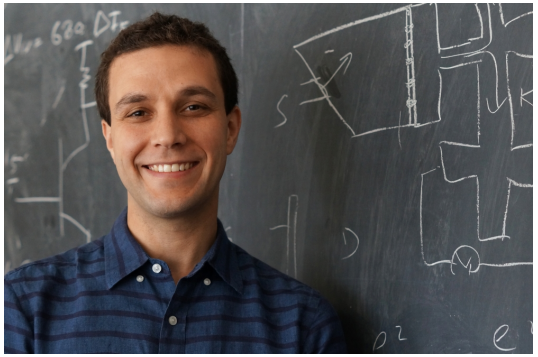
Kasra Hejazi

Zhu-Xi Luo

Mengxing Ye



Xuzhe Ying



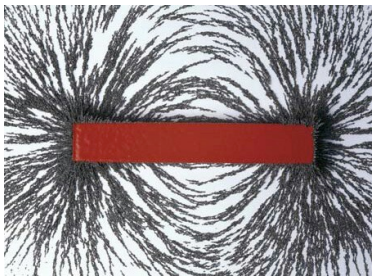
Andrea Young

Magnets

~500BC: Ferromagnetism documented in Greece, India, used in China

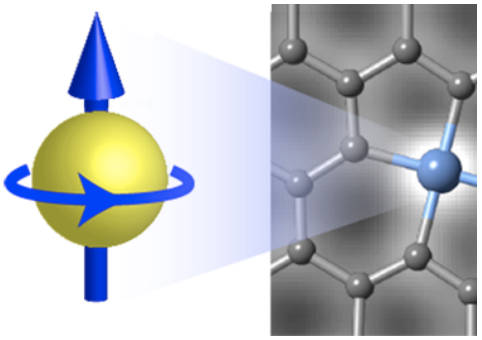
sinan, ~200BC

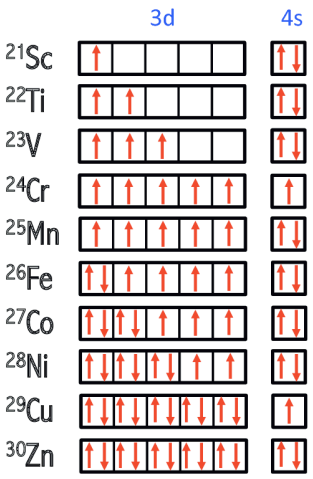
and in elementary school today



Spin

Almost all magnetism originates from unpaired d and f electrons in atoms





Friedrich Hund

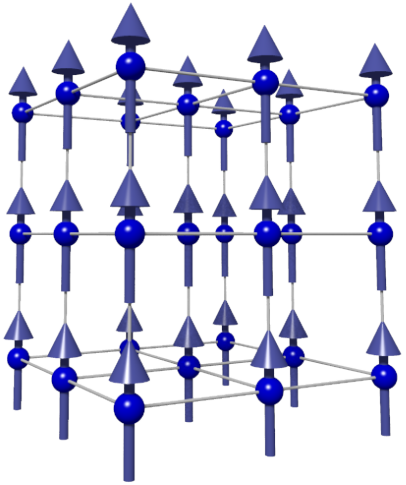
Hund's 1st rule: electrons avoid pairing within a shell

(this miminizes Coulomb repulsion within the same orbital)

Magnetism

- Atomic spins interact via *exchange* to favor an ordered arrangement

  - Aligned parallel: ferromagnets

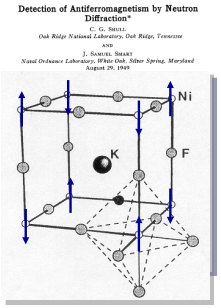


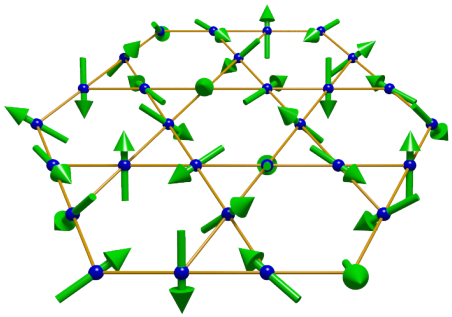
$$H = -J \sum_{\langle ij \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

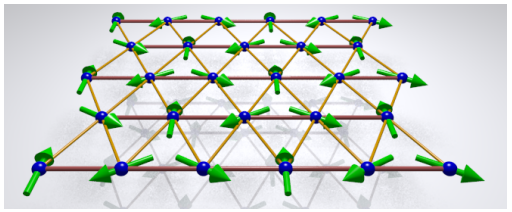
  - More complex arrangements: antiferromagnets

1949AD: antiferromagnetism proven experimentally

but there are 1000s of them, much more common than FMs







2d Ising model

- Onsager, 1943: Solved 2d Ising model exactly, proving that a phase transition exists with non-trivial critical behavior

Lars Onsager

$$H = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j$$

$$Z = \sum_{\{\sigma_i = \pm 1\}} e^{-\beta H}$$

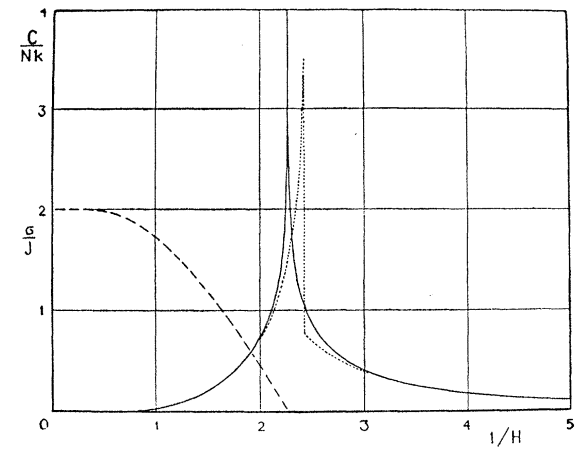


FIG. 6. Properties of quadratic crystal. — — — — — — Boundary tension σ between regions of opposite order. ———— Specific heat C. - - - - - - - - - - - - Approximate computation of C by Kramers and Wannier.

Mermin-Wagner-Hohenberg-Berezinskii theorem

- Heisenberg (vector) spins with finite interactions cannot order in two dimensions at any T>0

ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM
IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS*

N. D. Mermin† and H. Wagner‡
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York
(Received 17 October 1966)

It is rigorously proved that at any nonzero temperature, a one- or two-dimensional isotropic spin-S Heisenberg model with finite-range exchange interaction can be neither ferromagnetic nor antiferromagnetic. The method of proof is capable of excluding a variety of types of ordering in one and two dimensions.

Thermal fluctuations prevent order

- Rough idea: $\boldsymbol{S}_i = M\hat{\boldsymbol{z}} + \delta\boldsymbol{S}_i \qquad \delta\boldsymbol{S}_i \cdot \hat{\boldsymbol{z}} = 0$ suppose small fluctuation

$$E[\delta\boldsymbol{S}] \approx \int d^2\boldsymbol{x}\, \rho(\nabla\delta\boldsymbol{S})^2$$

rotational symmetry

$$\langle(\delta\boldsymbol{S})^2\rangle \approx \frac{k_B T}{\rho}\int \frac{d^2\boldsymbol{k}}{(2\pi)^2}\frac{1}{k^2} \sim \frac{k_B T}{2\pi\rho}\ln(L/a)$$

diverges no matter how low the temperature is

O(3) model

- At low temperature, non-linear sigma model applies

$$Z = \int_{|\boldsymbol{n}|=1} [d\boldsymbol{n}(\boldsymbol{x})] e^{-\beta\rho \int d^2\boldsymbol{x} (\nabla \boldsymbol{n})^2}$$

- Correlation length

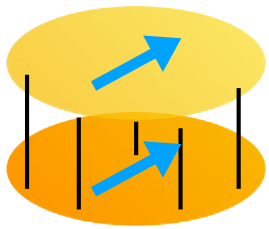
$$\langle \boldsymbol{n}(\boldsymbol{x}) \cdot \boldsymbol{n}(\boldsymbol{0}) \rangle \sim e^{-|x|/\xi}, \qquad \xi \sim e^{2\pi\rho/k_B T}$$

- When kT << ρ, correlation length becomes exponentially long and most properties become nearly indistinguishable from an ordered state

No phase transition but smooth evolution to almost ordered spins at low T

Quasi-2d systems

- Even most quasi-2d solids have some weak coupling between layers



$$E_{AF} - E_{FM} \sim J'\xi^2 \sim J' e^{4\pi\rho/k_B T}$$

E_{AF}-E_{FM}> kT when T<T_c, with

$$k_B T_c \sim 4\pi\rho/\ln(4\pi\rho/J')$$

❖ 3d ordering is induced by even very weak J'

Example: 2d square AF

- La_2CuO_4: parent material of high-T_c

Instantaneous spin correlations in La_2CuO_4

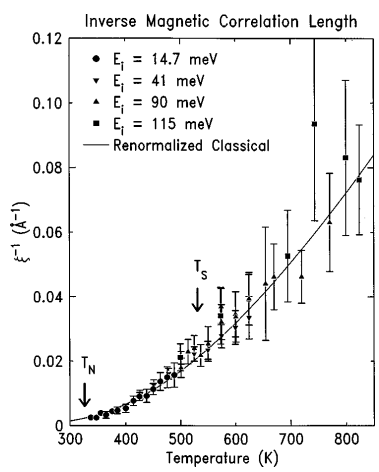
R. J. Birgeneau, M. Greven,* M. A. Kastner, Y. S. Lee, and B. O. Wells†
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

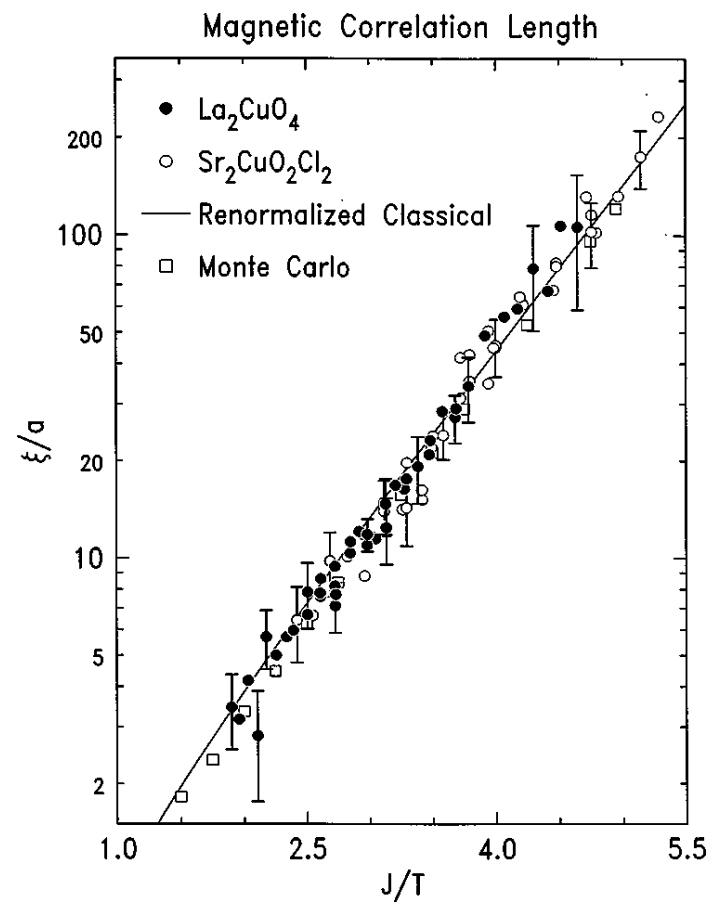
Y. Endoh and K. Yamada‡
Department of Physics, Tohoku University, Aramaki, Aoba-ku, Sendai 980, 980-77 Japan

G. Shirane
Brookhaven National Laboratory, Upton, New York 11973
(Received 23 October 1998)

TABLE I. Néel temperature, superexchange energy, and corrections to the 2D Heisenberg Hamiltonian for La_2CuO_4 (Ref. 9) and $Sr_2CuO_2Cl_2$ (Ref. 10). α_{DM} and α_{XY} are larger than the values quoted in Refs. 9 and 10 by factors by (Zc/Zg) and $(Zc/Zg)^2$, respectively. Here $Z_c(\frac{1}{2})\simeq 1.17$ and $Z_g(\frac{1}{2})\simeq 0.6$ are the quantum renormalization factors for the spin-wave velocity and spin-wave gap, respectively.

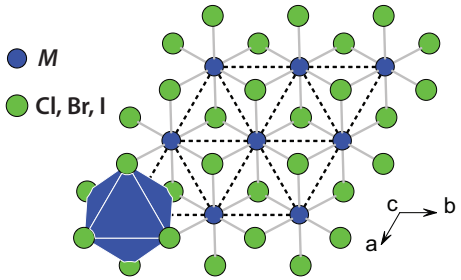
	La_2CuO_4	$Sr_2CuO_2Cl_2$
S	$\frac{1}{2}$	$\frac{1}{2}$
T_N (K)	325	256.5
J (meV)	135	125
α_{NNN}	~ 0.08	~ 0.08
α_{DM}	1.5×10^{-2}	–
α_{XY}	-5.7×10^{-4}	-5.3×10^{-4}
$\alpha_{\perp 1}-\alpha_{\perp 2}$	5×10^{-5}	$\sim 10^{-8}$

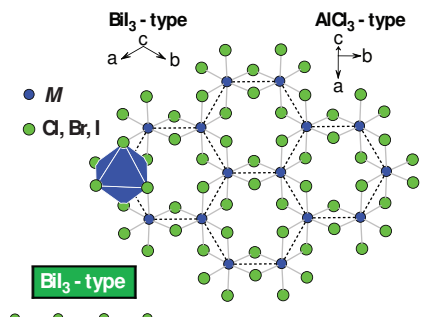




Truly 2d materials

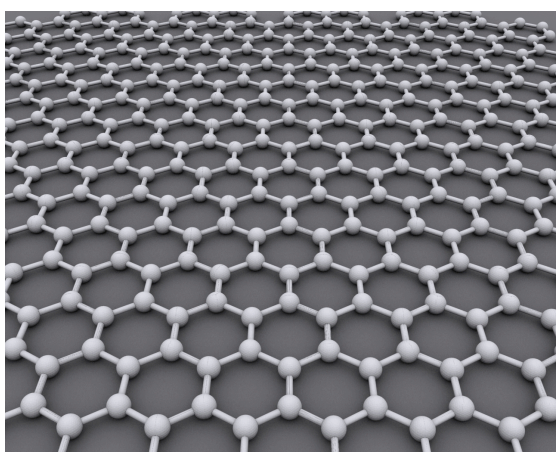
- 2d Van der Waals magnets





$MnPS_3$, $FePS_3$, $NiPS_3$, $CoPS_3$, $CrSiTe_3$...
CrI_3, $RuCl_3$, ...

- The parent of 2d materials: graphene



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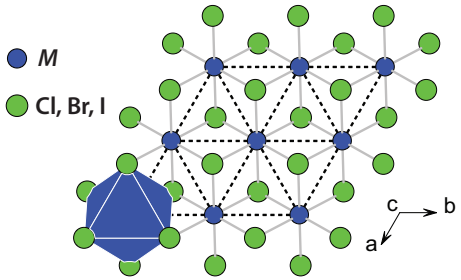
extraordinarily defect-free material, highly conducting, described by 2d Dirac equation

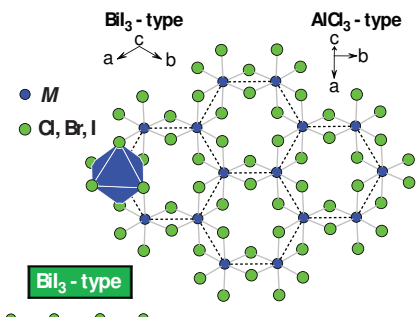
Entirely composed of C atoms, with electrons in bonded s+p orbitals. No local spins.

but it still shows magnetism!!

Truly 2d materials

- **2d Van der Waals magnets**





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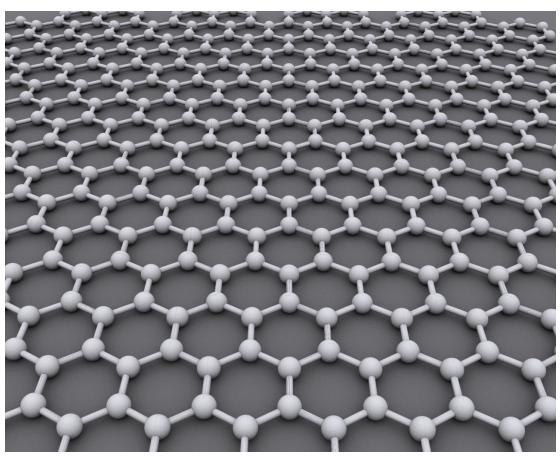

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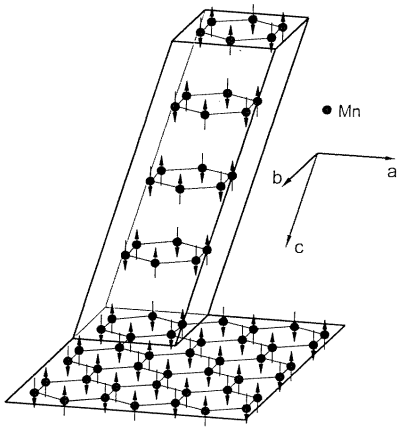
2d VdW magnets

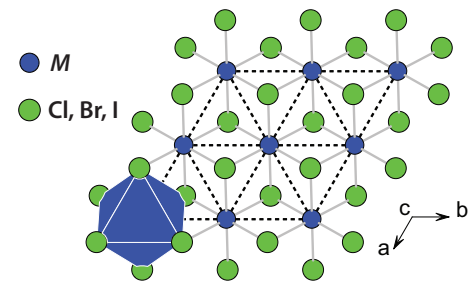


$MnPS_3$, $FePS_3$, $NiPS_3$, $CoPS_3$, $CrSiTe_3$...

***M*Cl$_2$**

Ti	V	Cr	Mn	Fe	Co	Ni
Zr	Nb	Mo	Tc	Ru	Rh	Pd
Hf	Ta	W	Re	Os	Ir	Pt

***M*Br$_2$**



***M*Cl$_3$**

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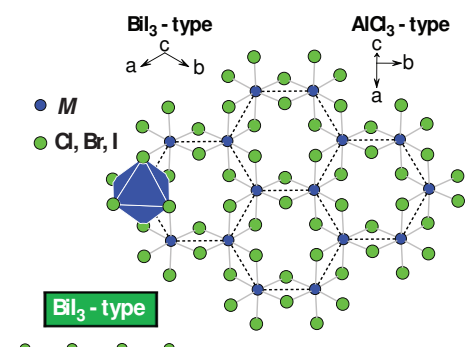
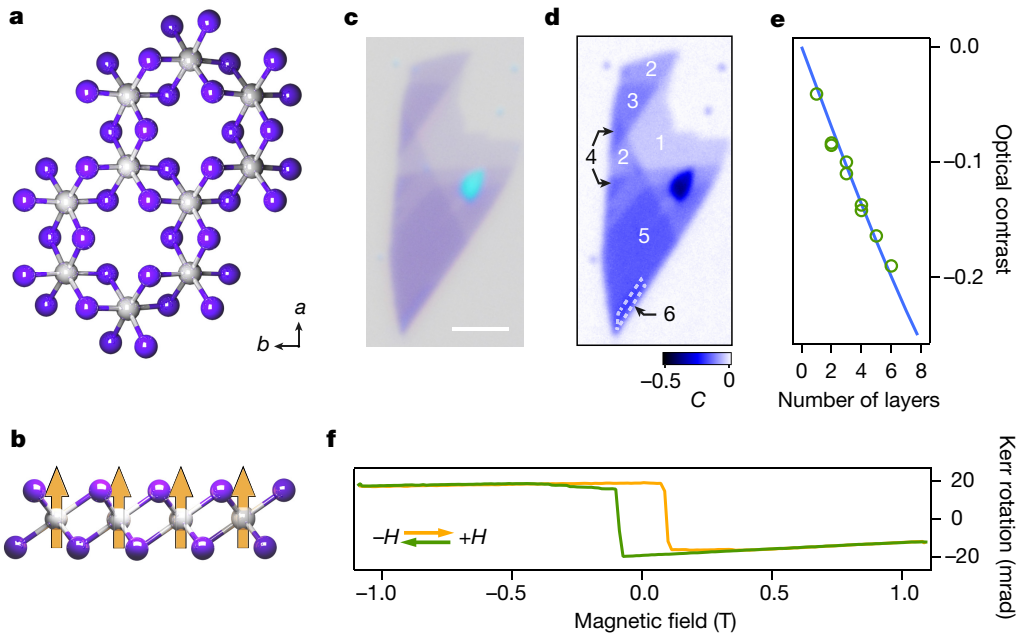


CrI_3, $RuCl_3$,...

CrI_3

B. Huang *et al*, 2017



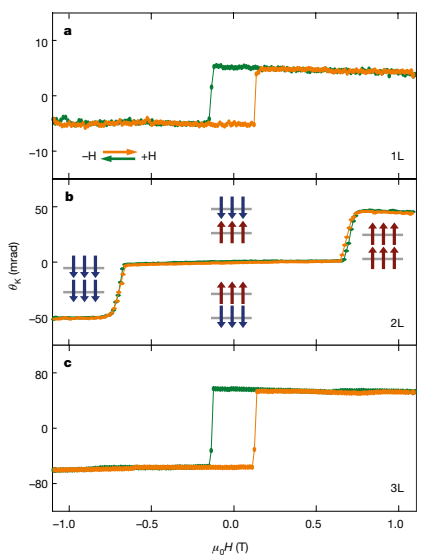
Ferromagnetic 2d honeycomb layers

Optical measurement of magnetization

1L

2L

3L



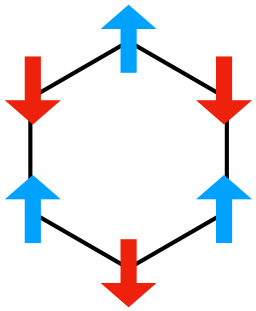
Still ferromagnetic in single layer

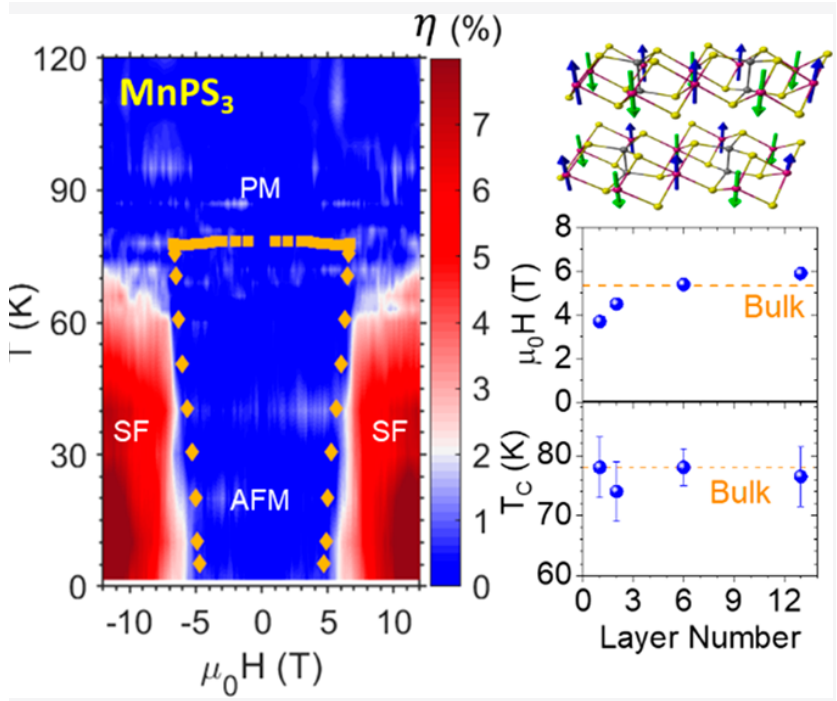
Surprise: bilayer is anti-ferromagnetic

$MnPS_3$

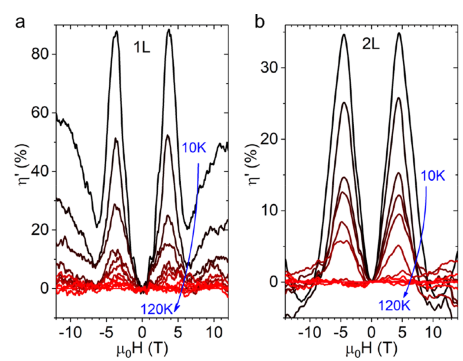
G. Long *et al*, 2020

antiferromagnetic honeycomb



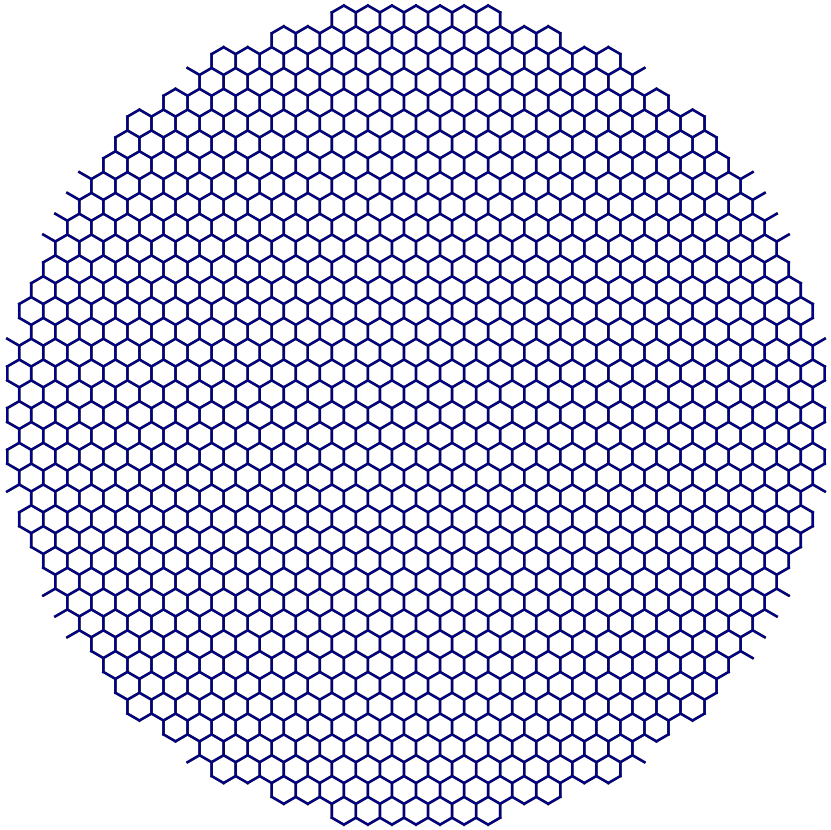


order persists to single layer



tunneling MR

Twisting and moiré

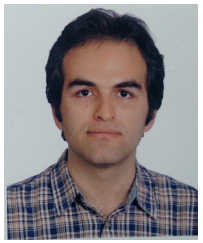


Moiré

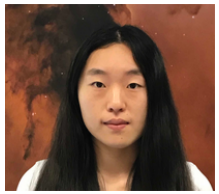
mohair

6°

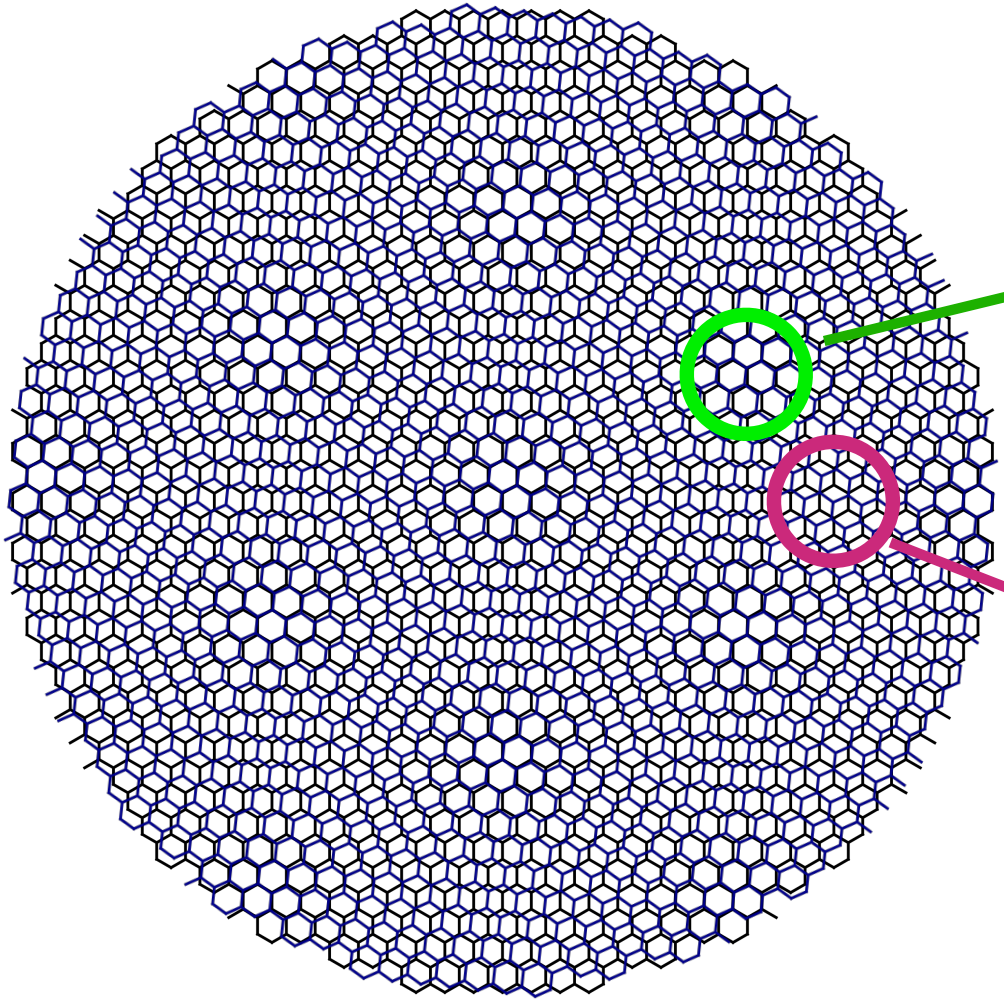
Twisted AF

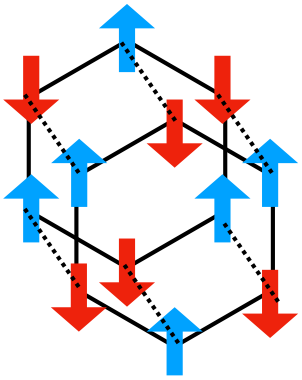


Kasra Hejazi



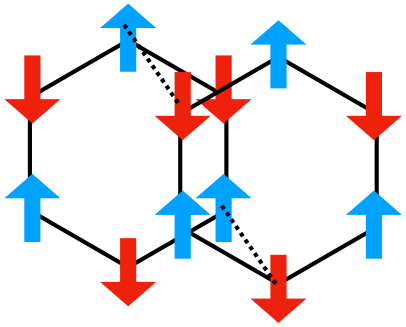
Zhu-Xi Luo





AA

$$\boldsymbol{N}_1 = -\boldsymbol{N}_2$$

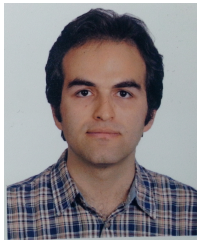


AB

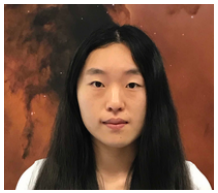
$$\boldsymbol{N}_1 = \boldsymbol{N}_2$$

Frustration: Neél vectors must rotate

Continuum model(s)



Kasra Hejazi



Zhu-Xi Luo

- Basic assumptions:
  - Inter-layer coupling weak J′ << J
  - Rotation angle is small (can also treat general strains)
- Example: $MnPS_3$: excellent Heisenberg AF

$$\mathcal{L} = \sum_l \frac{\rho}{2v^2} \left(\partial_t \boldsymbol{N}_l\right)^2 - \mathcal{H}_{\mathrm{cl}}$$

$$\mathcal{H}_{\mathrm{cl}} = \sum_l \left[\frac{\rho}{2} \left(\nabla \boldsymbol{N}_l\right)^2 - d\left(N_l^z\right)^2\right] - J' \Phi(\boldsymbol{x}) \boldsymbol{N}_1 \cdot \boldsymbol{N}_2$$

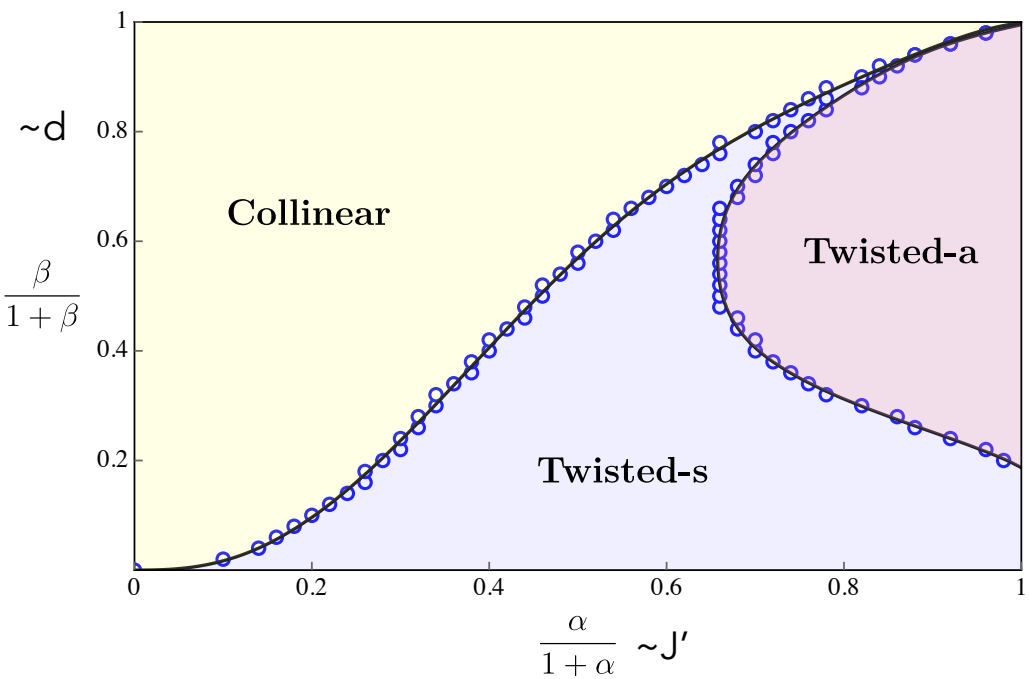
$$\Phi(\boldsymbol{x}) = \sum_{a=1}^{3} \cos(\boldsymbol{q}_a \cdot \boldsymbol{x})$$

Can predict spin textures, magnon subbands, etc.

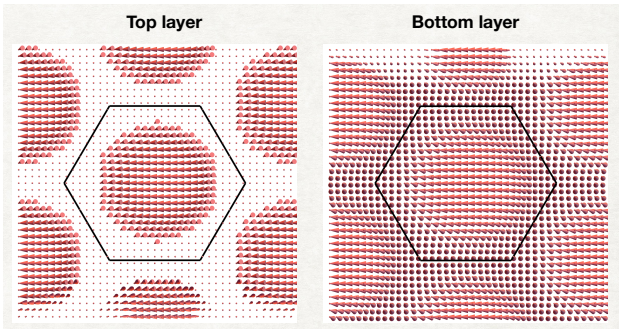
Twisted AF

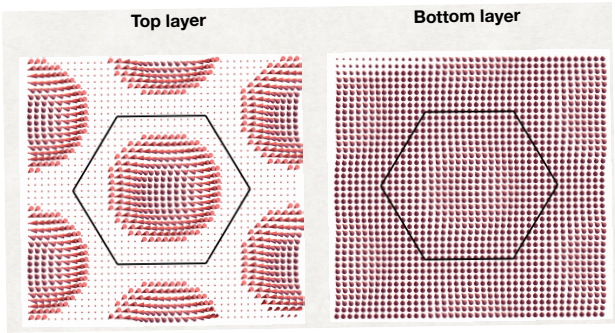
$$\mathcal{H}_{\rm cl} = \sum_l \left[\frac{\rho}{2} \left(\nabla \boldsymbol{N}_l \right)^2 - d \left(N_l^z \right)^2 \right] - J' \Phi(\boldsymbol{x}) \boldsymbol{N}_1 \cdot \boldsymbol{N}_2$$

Dimensionless parameter $\alpha = \dfrac{2J'}{\rho q_m^2} \sim \dfrac{J'}{J\theta^2}$



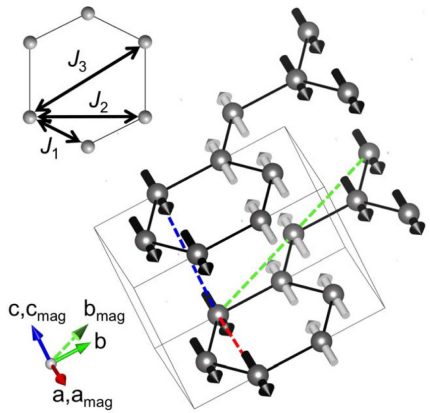
Coplanar spin textures





Transitions should be tunable by applied field

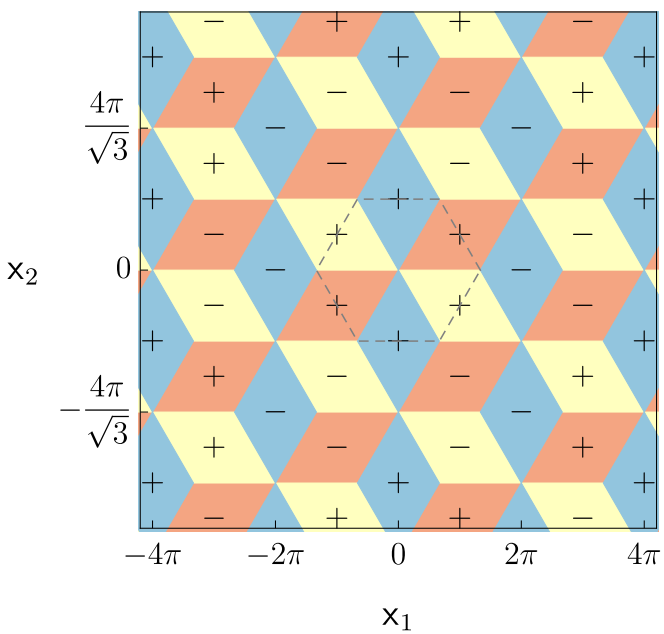
Zig-Zag antiferromagnets



$$\mathcal{H}_{cl} = \sum_{a,l} \frac{\rho}{2} (\boldsymbol{\nabla} \boldsymbol{N}_{a,l})^2 - \frac{J'}{2} \sum_a \boldsymbol{N}_{a,1} \cdot \boldsymbol{N}_{a,2} \cos\left(\frac{\boldsymbol{q}_a \cdot \boldsymbol{x}}{2}\right)$$

3 distinct q_a domains

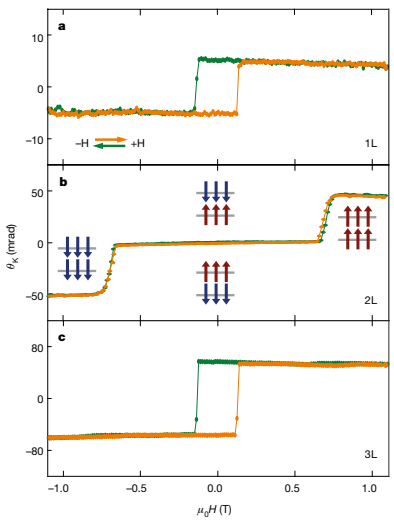
$NiPS_3$,$FePS_3$, $CoPS_3$,$RuCl_3$…



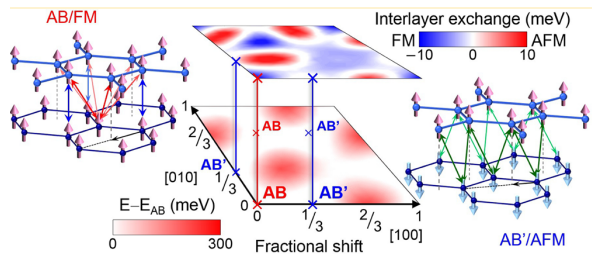
Strong-coupling domains have structure of "dice lattice"

CrI_3

Might not expect much from a *ferro*-magnet, but…



B. Huang *et al*, 2017

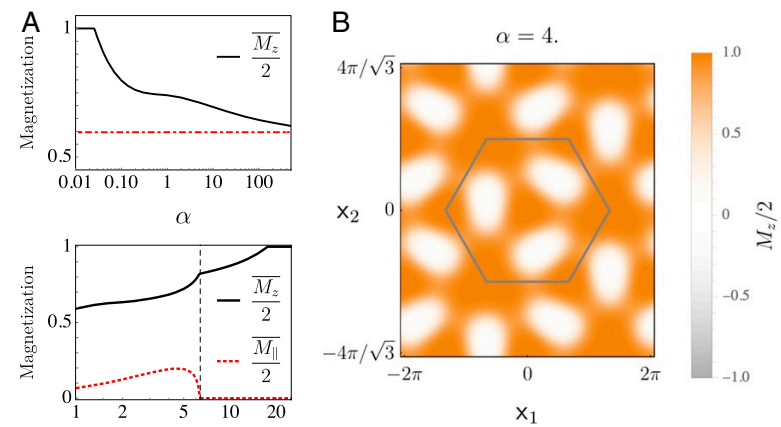


Sivadas *et al*, 2018

Sign-changing stacking-dependent interactions

$$\mathcal{H}_{\mathrm{cl}} = \sum_l \left[\frac{\rho}{2} \left(\nabla \boldsymbol{M}_l \right)^2 - d \left(M_l^z \right)^2 \right] - J' \tilde{\Phi}(\boldsymbol{x}) \boldsymbol{M}_1 \cdot \boldsymbol{M}_2$$

from DFT theory



mixed $\theta(^\circ)$

non-collinear

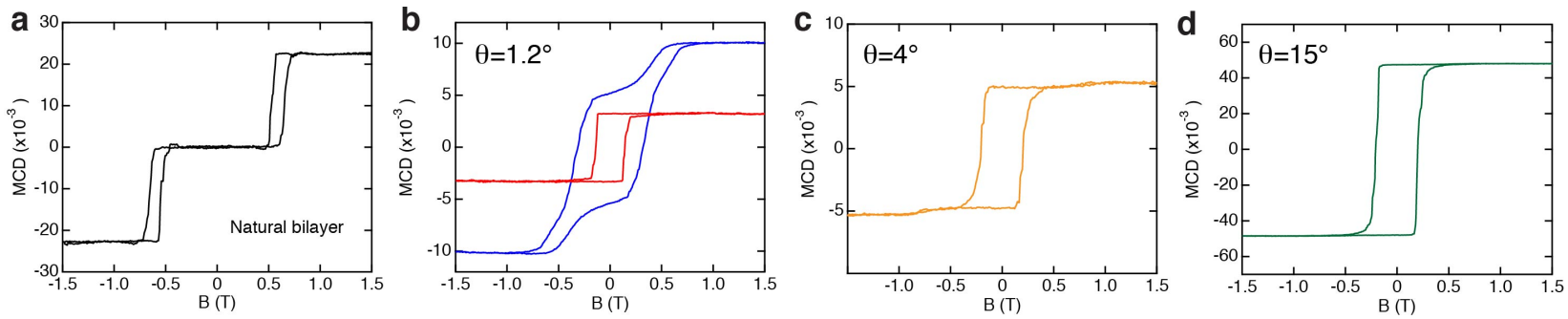
FM

CrI_3

Emergence of a noncollinear magnetic state in twisted bilayer CrI_3

Yang Xu[1,2], Ariana Ray[1], Yu-Tsun Shao[1], Shengwei Jiang[1], Daniel Weber[3], Joshua E. Goldberger[3], Kenji Watanabe[4], Takashi Taniguchi[4], David A. Muller[1,5], Kin Fai Mak[1,5,6*], Jie Shan[1,5,6*]

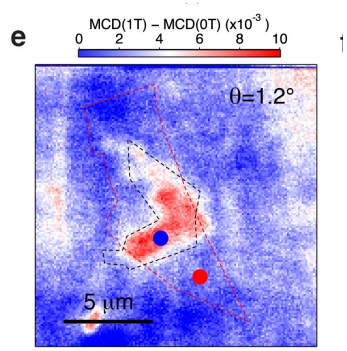
arXiv:2103.09850



bilayer: AF

small twist: mixed

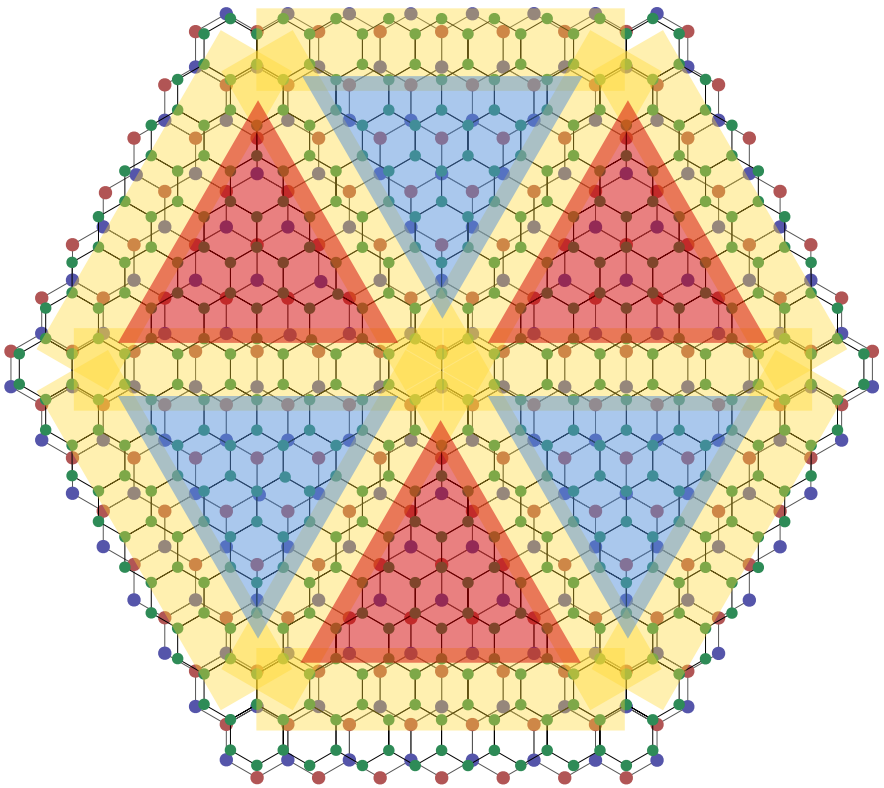
large twist: FM

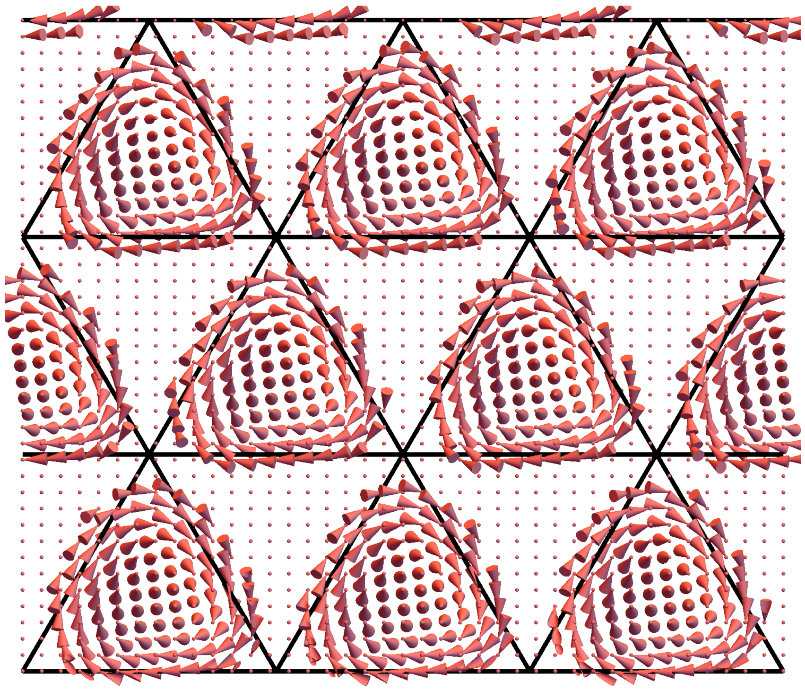


n.b. Also recent twisted double bilayer arXiv:2103.13573 claims evidence of non-collinear state

Moiré skyrmions

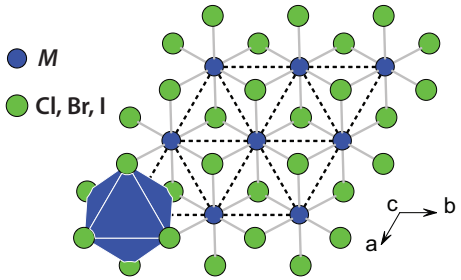
FM+AF heterobilayer

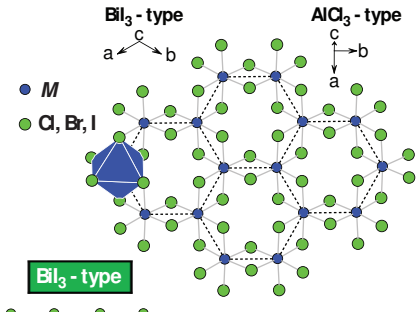




Truly 2d materials

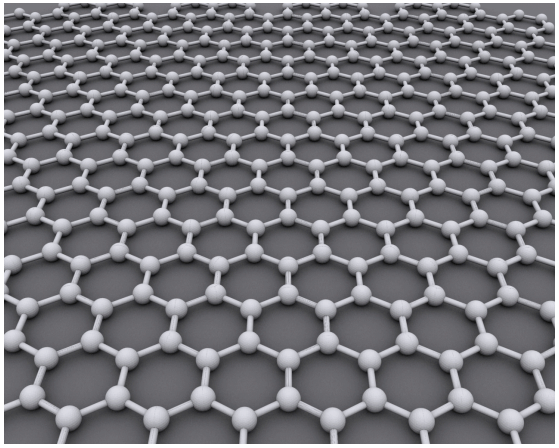
- 2d Van der Waals magnets





$MnPS_3$, $FePS_3$, $NiPS_3$, $CoPS_3$, $CrSiTe_3$...
CrI_3, $RuCl_3$, ...

- **The parent of 2d materials: graphene**



By AlexanderAlUS - Own work, CC BY-SA 3.0, https://commons.wikimedia.org/w/index.php?curid=11294534

extraordinarily defect-free material, highly conducting, described by 2d Dirac equation

Entirely composed of C atoms, with electrons in bonded s+p orbitals. No local spins.

but it still shows magnetism!!

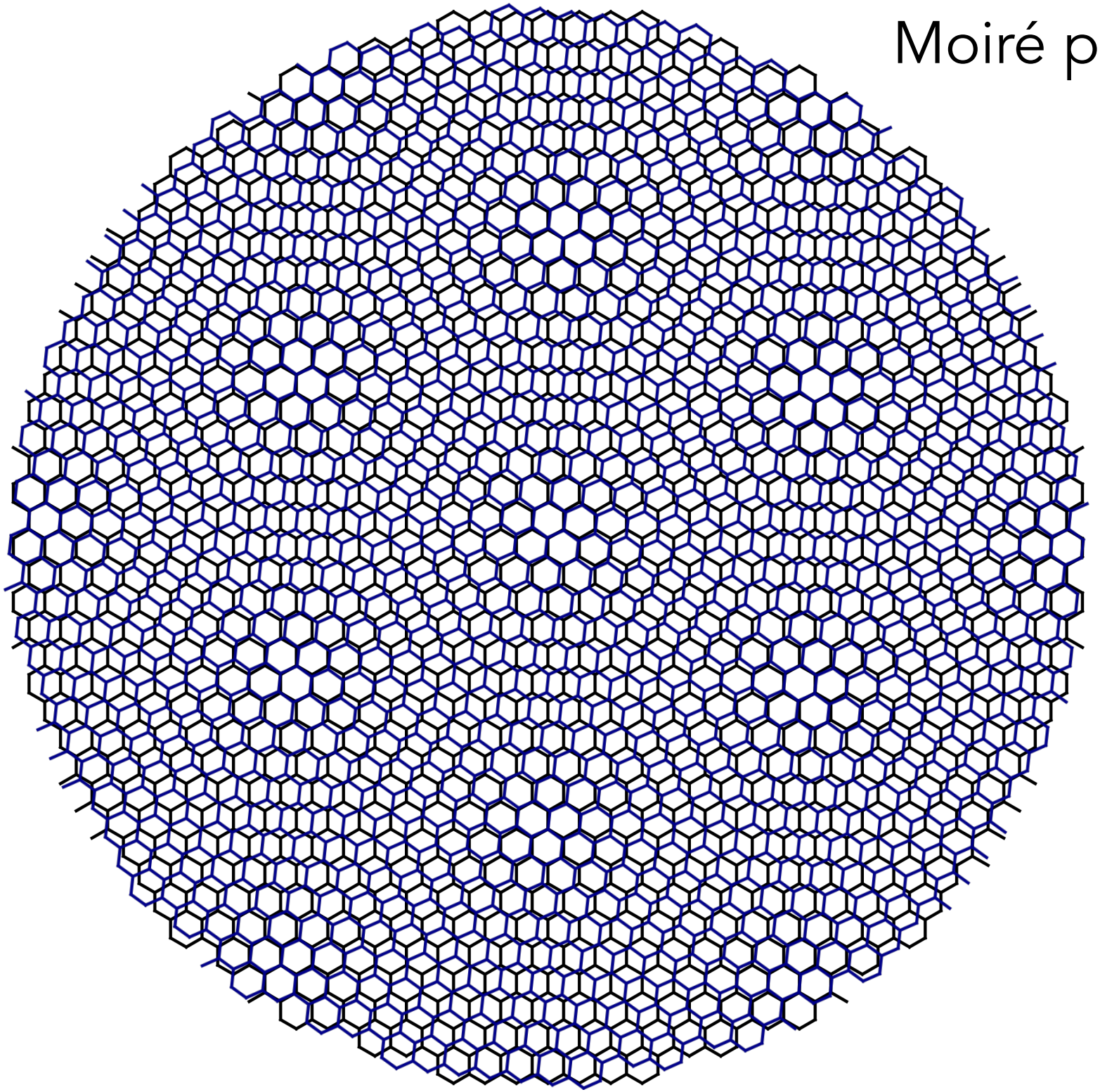
How to make graphene magnetic

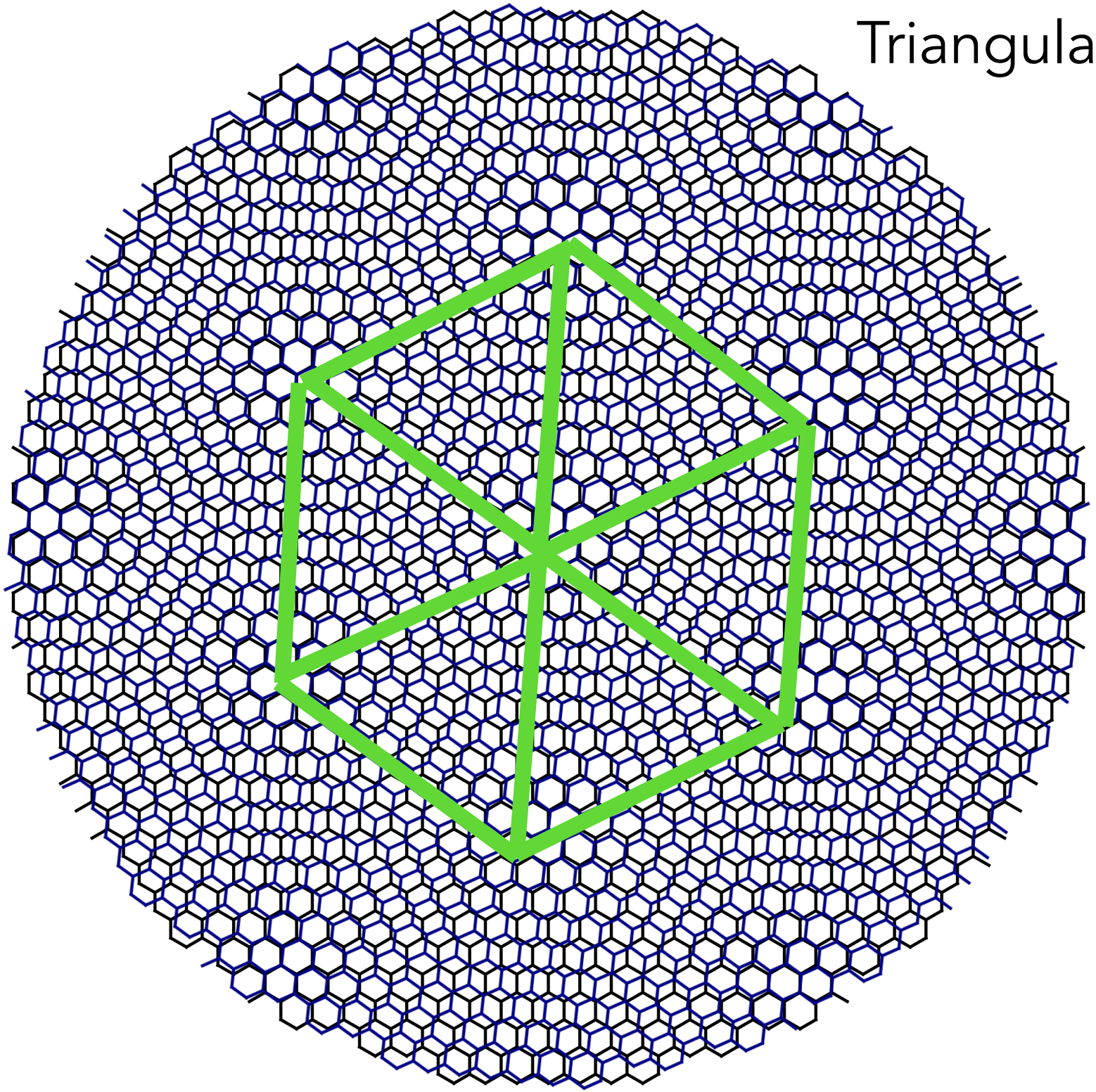
- Actually lots of ideas in the literature (but no good evidence that any of them work)

- What works: *twisting*

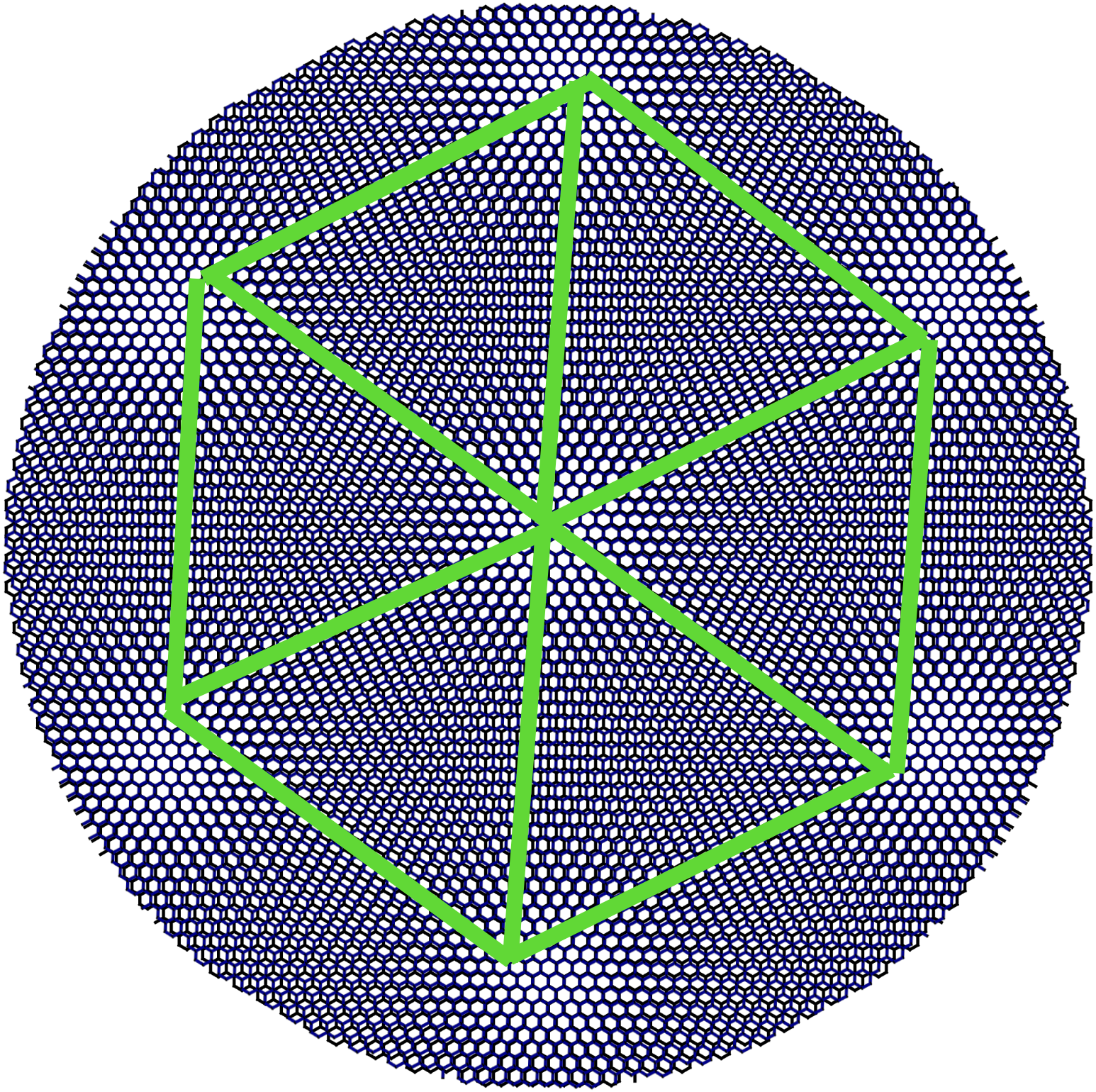
Moiré pattern

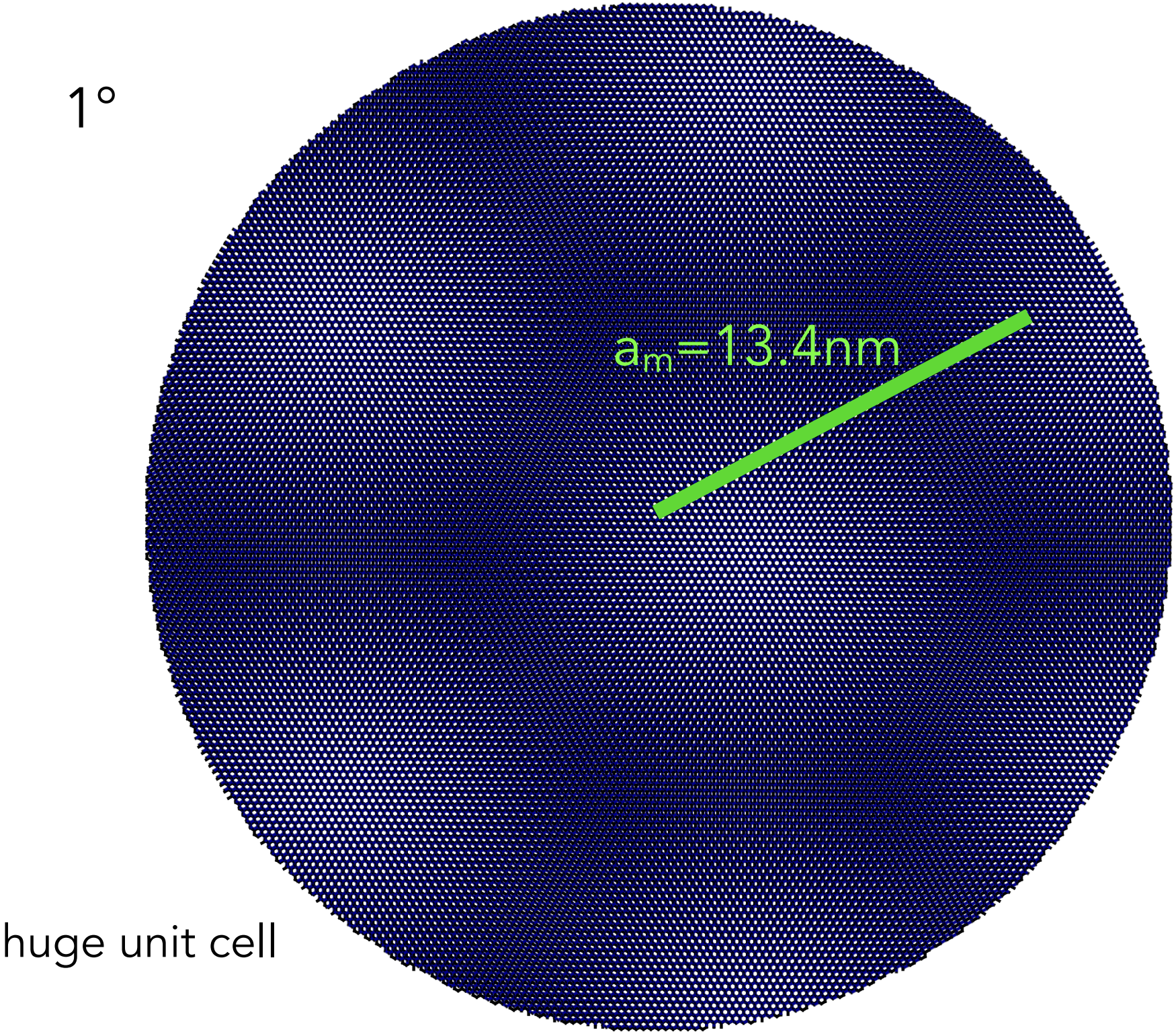
6°




6°
Triangular lattice

2°

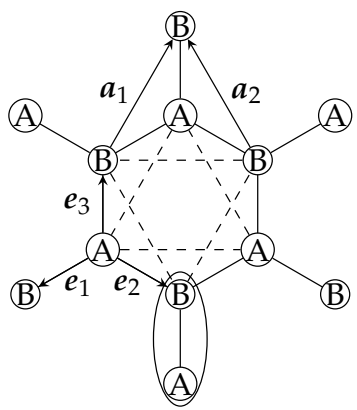



1°
$a_m = 13.4nm$
huge unit cell

Moiré Bands

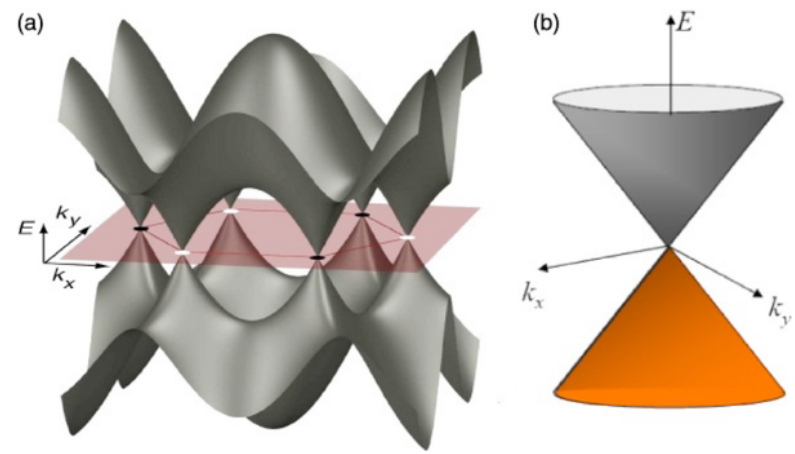
- What you need to know:
  - Moiré pattern creates an effective “artificial lattice” with huge lattice spacing ✅
  - Corresponding moiré bands occupy a tiny region of momentum space, separated into two “valleys” originating from the underlying graphene Dirac points
  - The important moiré bands (near the Fermi energy) become exceptionally narrow near the “magic angle” ~1degree
  - The moiré bands are topological, and in particular become *Chern bands* when the graphene is aligned with its hBN substrate

Dirac bands in graphene



2 sublattice spinor

$$\psi = \begin{pmatrix} \psi_A \\ \psi_B \end{pmatrix}$$

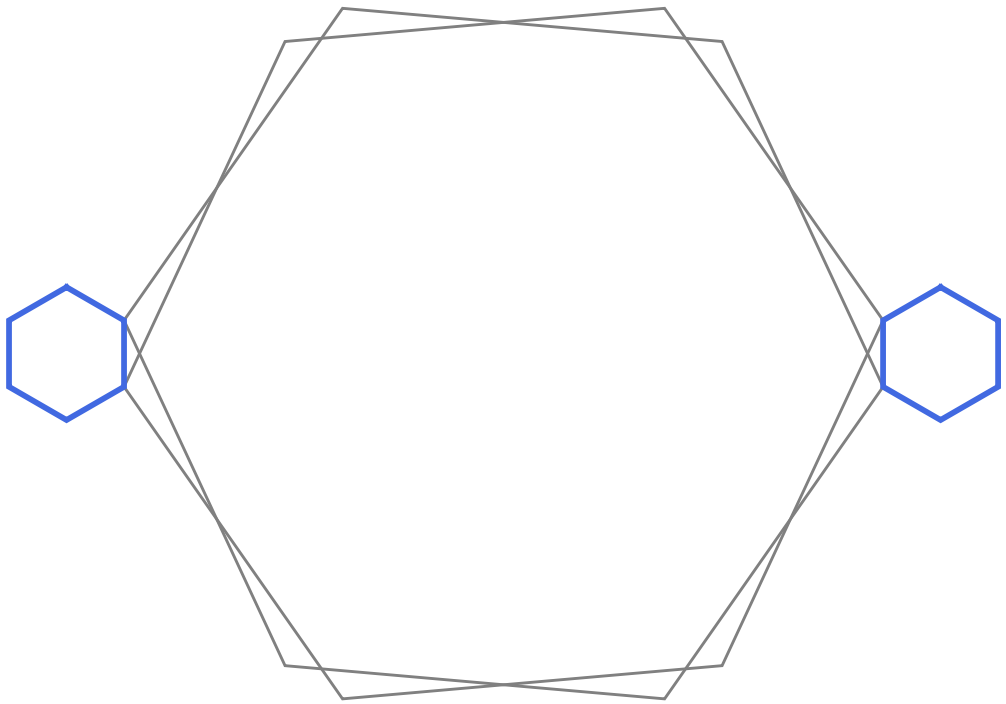


$$H_K = v\,\vec{k}\cdot\vec{\sigma}$$

$$H_{K'} = [H_K(-\vec{k})]^*$$

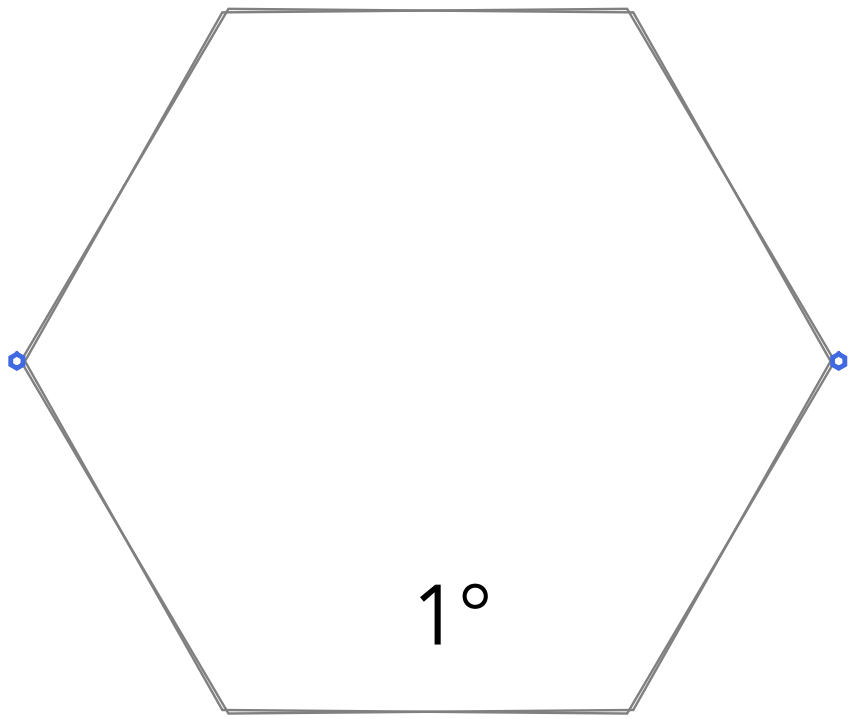
K,K′ “valleys”

Moiré bands



Two layers define two slightly shifted Dirac points for each valley

Moiré bands



Tiny moiré bands at each valley are well separated - act as two “flavors” of electrons

Moiré Bands

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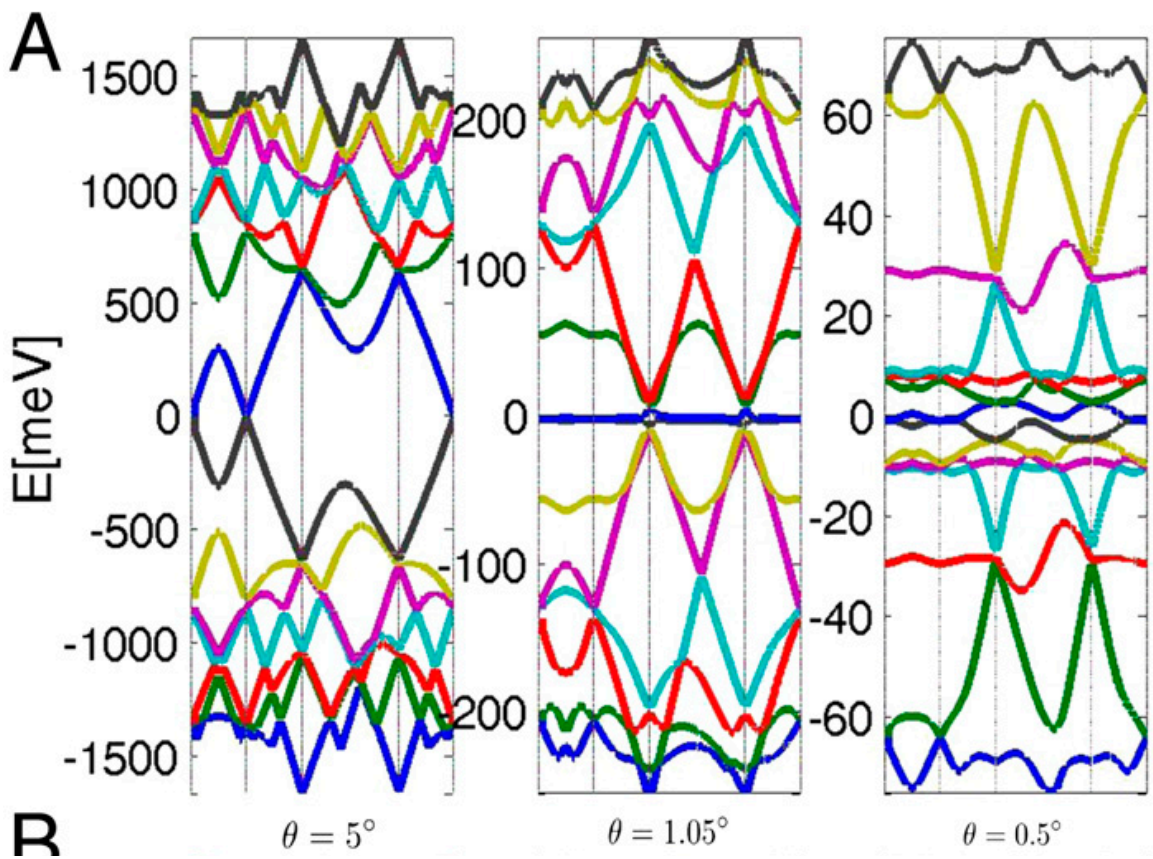
Magic angle bands

Bistritzer+Macdonald, 2011: moiré pattern behaves like an artificial lattice with its own bands, which becomes especially flat at the "magic" angle ~1degree

These bands retain their Dirac points, i.e. Berry phase



narrow band = small kinetic energy

Moiré Bands

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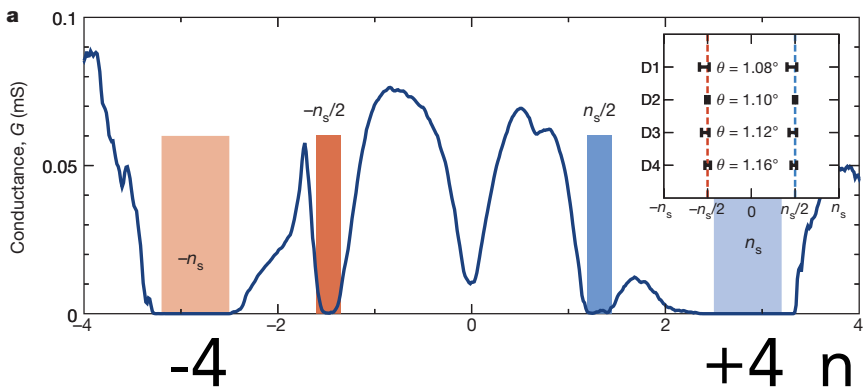
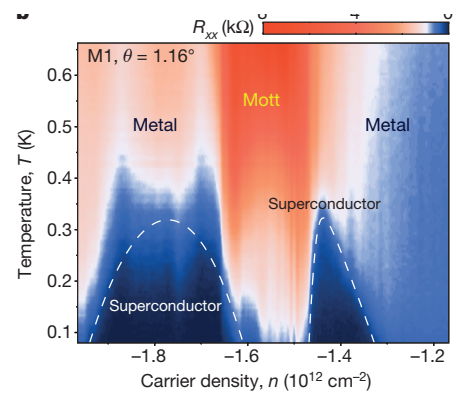
Correlation effects

- A quantum correlated electron simulator
- Tunable electron density
- Tunable correlation
- (Somewhat) Tunable topology
- 1/100 energy scale of usual solids



(e's/m.u.c.)



Jarillo-Herrero group

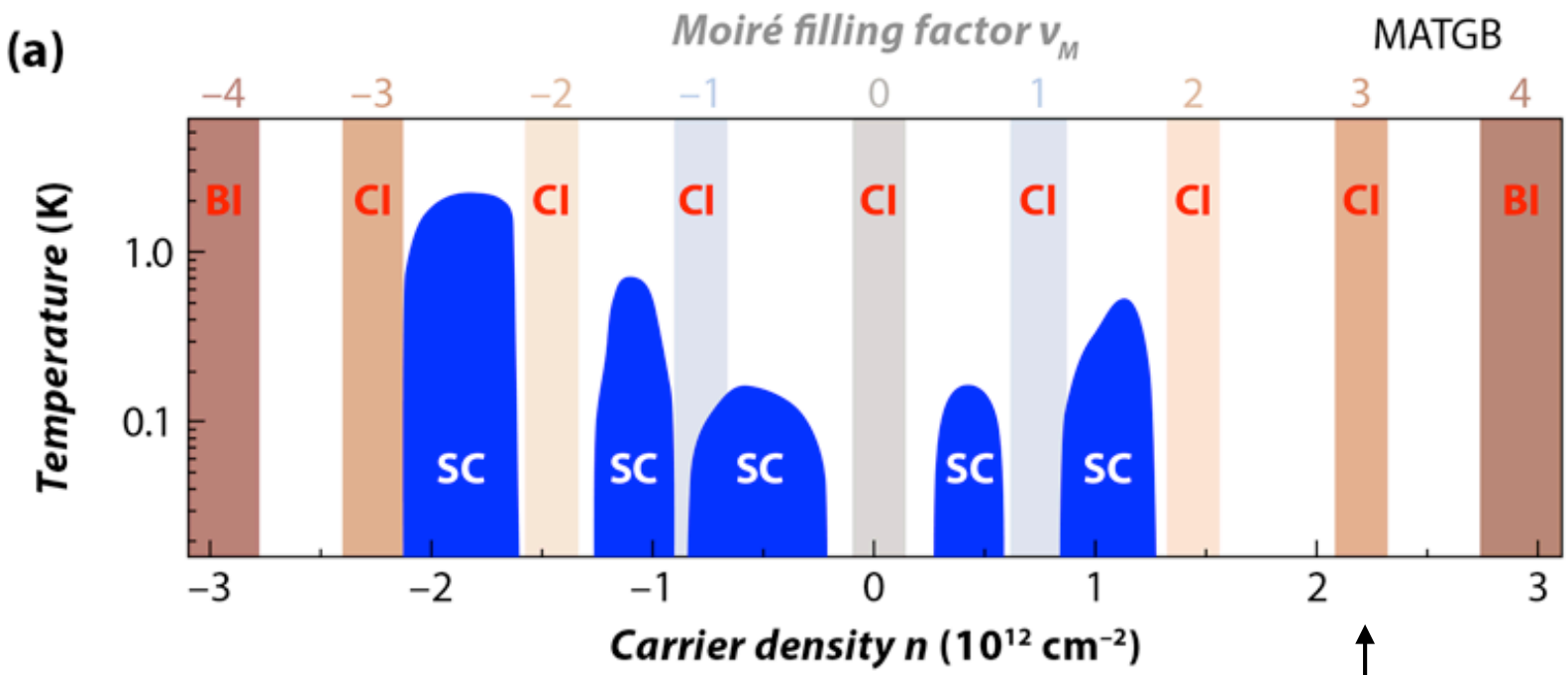
Nature 2018

2021:

- 235 talks at APS mtg with "twisted", 113 w/ "magic"
- ~300 papers on arXiv in past year

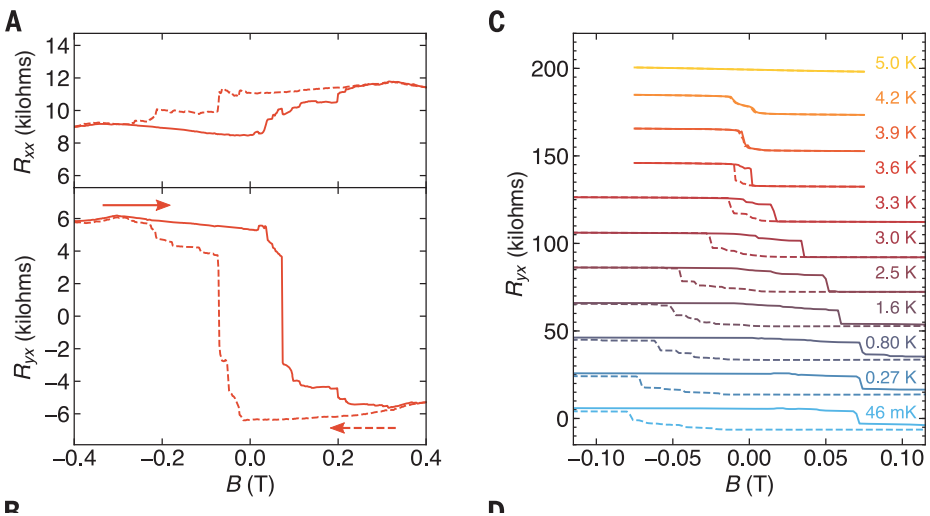
Correlation effects

from A. Macdonald, Physics today

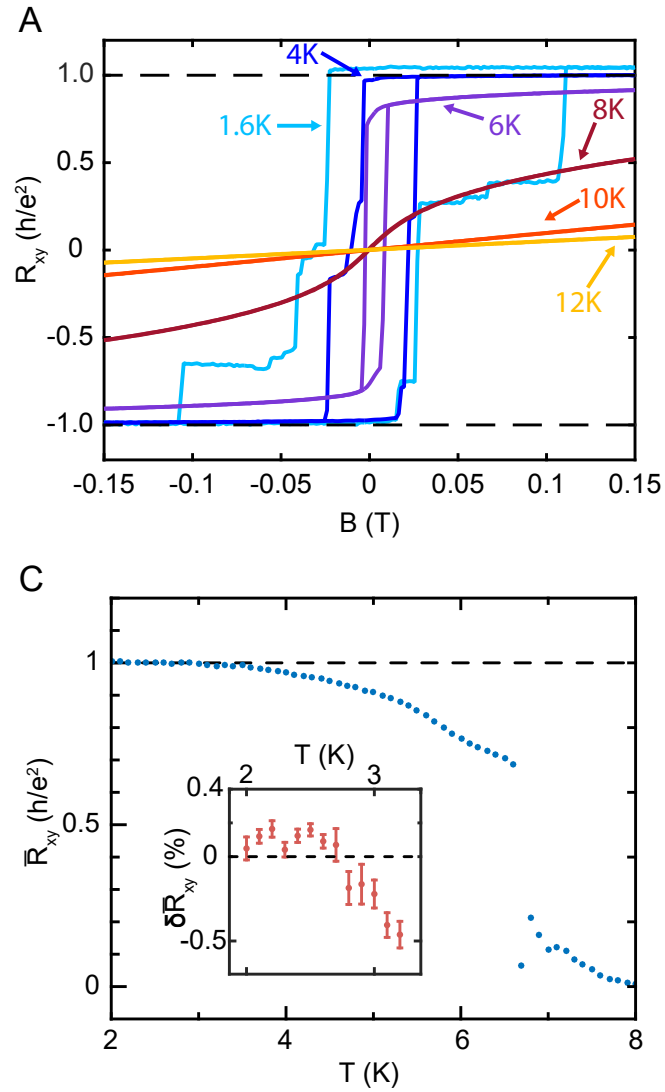


Today: magnetism

Ferromagnetism in TBG



A. Sharpe *et al*, 2019



M. Serlin *et al*, 2019

Moiré Bands

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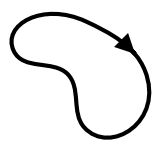
  - **The moiré bands are topological, and in particular become *Chern bands* when the graphene is aligned with its hBN substrate**

Berry phase and curvature

- Hamiltonian depending on parameters

$$H = H(\{\lambda_i\})$$

- Under adiabatic evolution $\lambda_i = \lambda_i(t)$
  - Wavefunction accumulates a geometric (Berry) phase over a closed loop



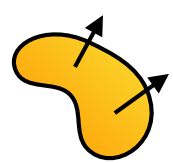
$$\gamma = i \oint d\lambda_i \langle\psi| \frac{\partial}{\partial \lambda_i} |\psi\rangle$$

~ Aharonov-Bohm phase

  - Using Stokes' theorem

$$\gamma = \iint d\lambda_{\mu\nu} F_{\mu\nu}$$

$$F_{\mu\nu} = i\left(\langle\partial_\mu\psi|\partial_\nu\psi\rangle - \langle\partial_\nu\psi|\partial_\mu\psi\rangle\right)$$



Berry curvature

~ magnetic field

Berry curvature in 2d solids

- Hamiltonian depends on k_x,k_y

$$\Omega = i\left(\langle \partial_{k_x}\psi | \partial_{k_y}\psi\rangle - \langle \partial_{k_y}\psi | \partial_{k_x}\psi\rangle\right)$$

- Controls several physical effects
  - Hall conductivity $\sigma_{xy} = \frac{e^2}{\hbar}\sum_n \int \frac{d^2\boldsymbol{k}}{(2\pi)^2}\Omega_n(\boldsymbol{k}) f(\epsilon_n(\boldsymbol{k}))$
  - Orbital magnetic moment $m_n^z(\boldsymbol{k}) \sim e\Omega_n(\boldsymbol{k})$
- T=0, insulator: quantization

$$\sigma_{xy} = \frac{e^2}{\hbar}\sum_{n \text{ occupied}} \int \frac{d^2\boldsymbol{k}}{(2\pi)^2}\Omega_n(\boldsymbol{k}) = \frac{e^2}{h}\sum_{n \text{ occupied}} C_n$$

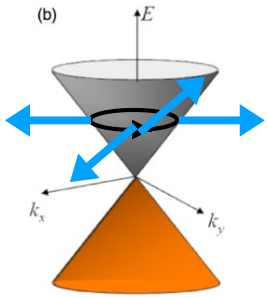
Chern number
= integer

Dirac and topology

- Dirac electrons are topological

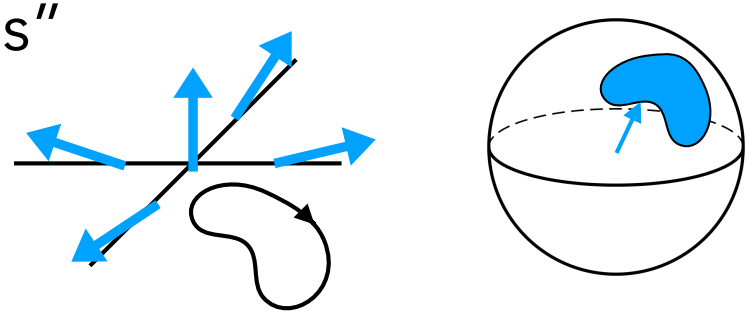
$$H_K = v\,\vec{k}\cdot\vec{\sigma}$$

$$\gamma = \pm\pi$$



- When symmetry relating the two sublattices is broken, the Dirac fermions acquire a "mass"

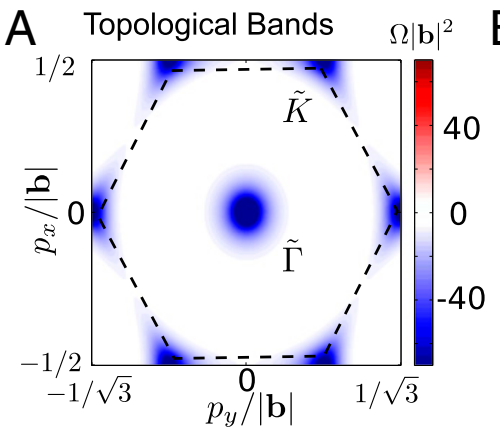
$$H_K \to v\,\vec{k}\cdot\vec{\sigma} + m\sigma^z$$



  - Berry curvature = 1/2 (area swept on sphere/unit area in k space). Large near Dirac points.

Moiré Chern bands

- Small coupling to aligned hBN substrate readily creates such Berry curvature



Topological Bloch bands in graphene superlattices

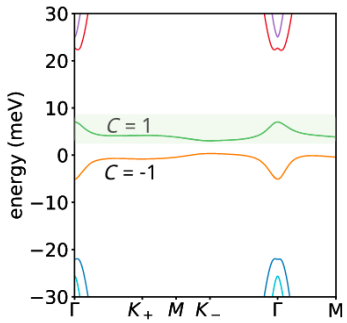
Justin C. W. Song[a,b,c,1], Polnop Samutpraphoot[c], and Leonid S. Levitov[c,1]

[a]Walter Burke Institute for Theoretical Physics, California Institute of Technology, CA 91125; [b]Institute for Quantum Information and Matter, and Department of Physics, California Institute of Technology, CA 91125; and [c]Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139

Edited by Subir Sachdev, Harvard University, Cambridge, MA, and approved June 24, 2015 (received for review December 30, 2014)

- The flat moiré bands then become Chern bands

e.g.



N. Bultinck *et al*, 2020

States in each valley carry Hall current and large orbital magnetic moment

Moiré Bands

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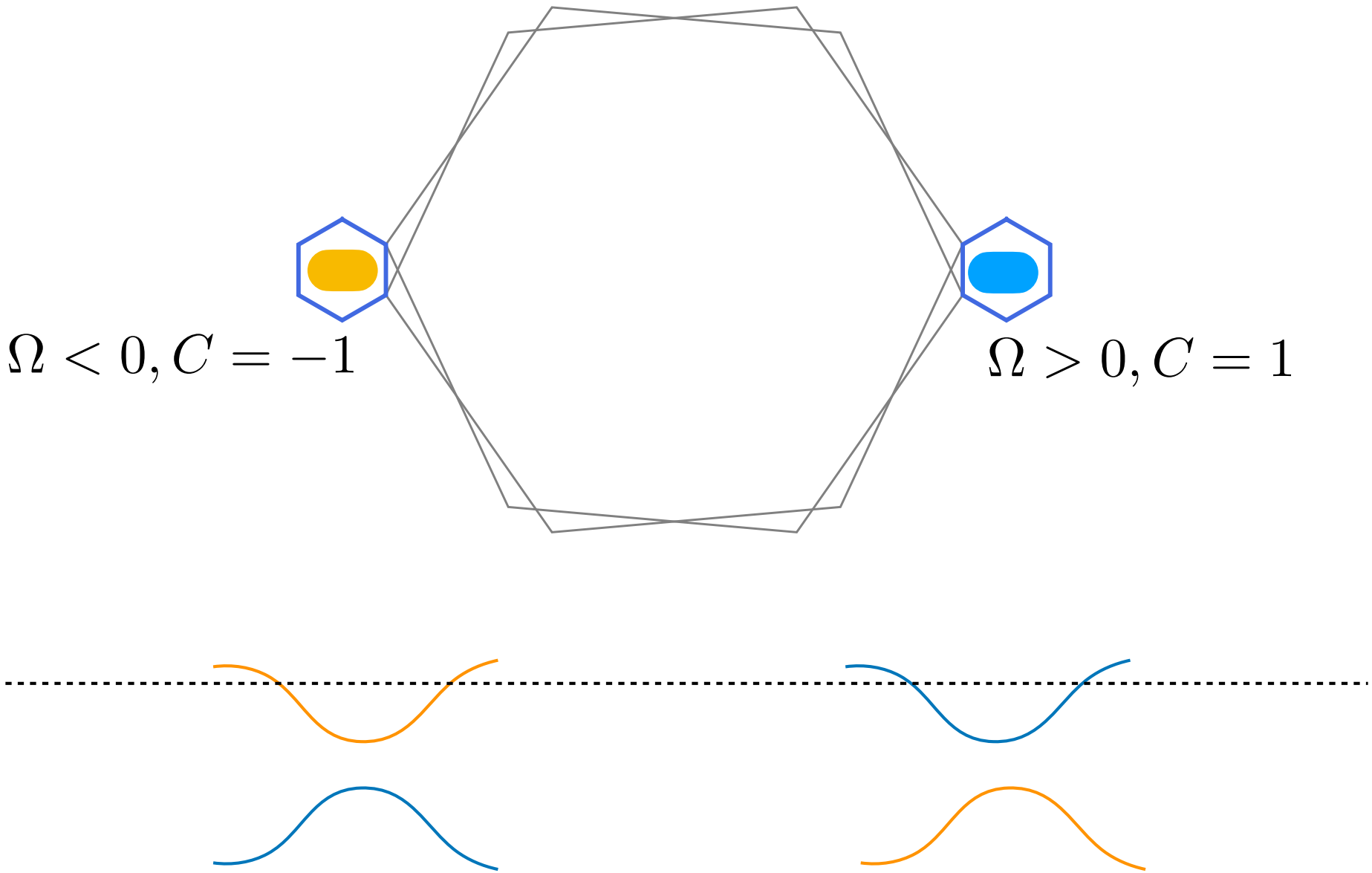
  - The moiré bands are topological, and in particular become *Chern bands* when the graphene is aligned with its hBN substrate ✅

Valley Chern number



Stoner ferromagnetism

- Coulomb repulsion favors polarization of spin and valley, at cost of kinetic (band) energy

  Due to narrow magic angle bands, Coulomb wins

$$H_{\text{int}} = \sum_{a<b} U_{ab} n_a n_b$$

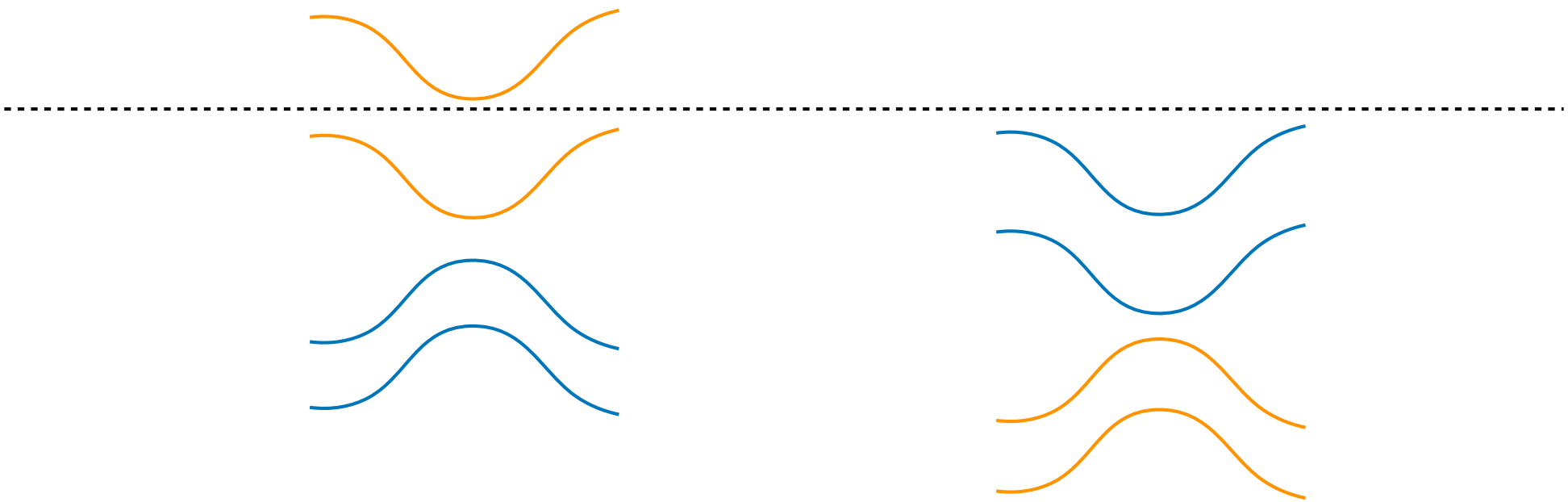
(a=spin+valley)

Stoner ferromagnetism

- Coulomb repulsion favors polarization of spin and valley, at cost of kinetic (band) energy

$$H_{\text{int}} = \sum_{a<b} U_{ab} n_a n_b$$

(a=spin+valley)

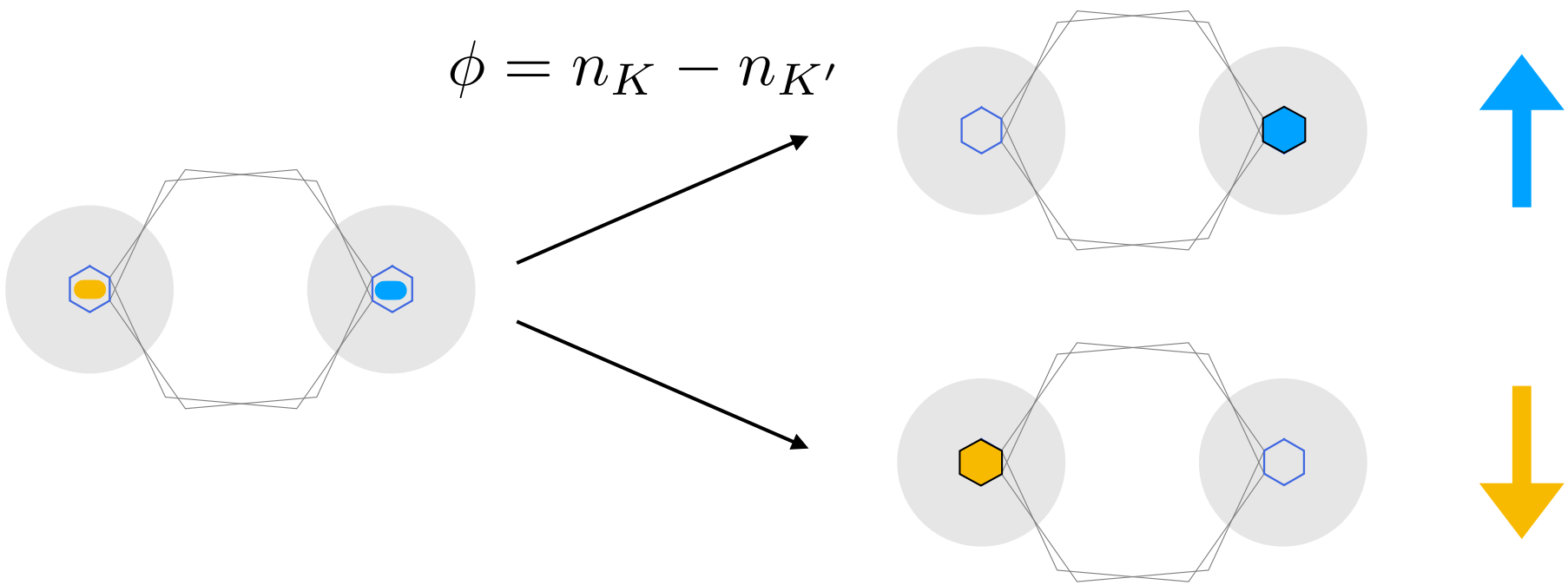


Valley ferromagnetism

- Recall Mermin-Wagner: is 2d ordering possible?

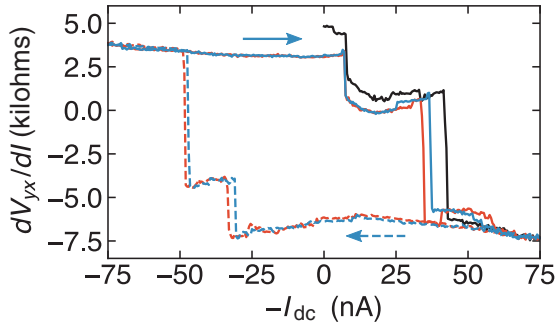
A1: ordering of the *valley* is possible, because this is an *Ising* symmetry breaking ✅

$$\phi = n_K - n_{K'}$$

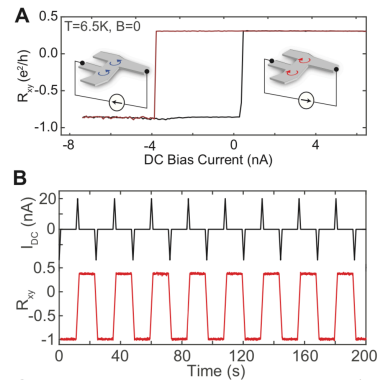


A2: We expect no additional transition associated with spin ✅

Current control

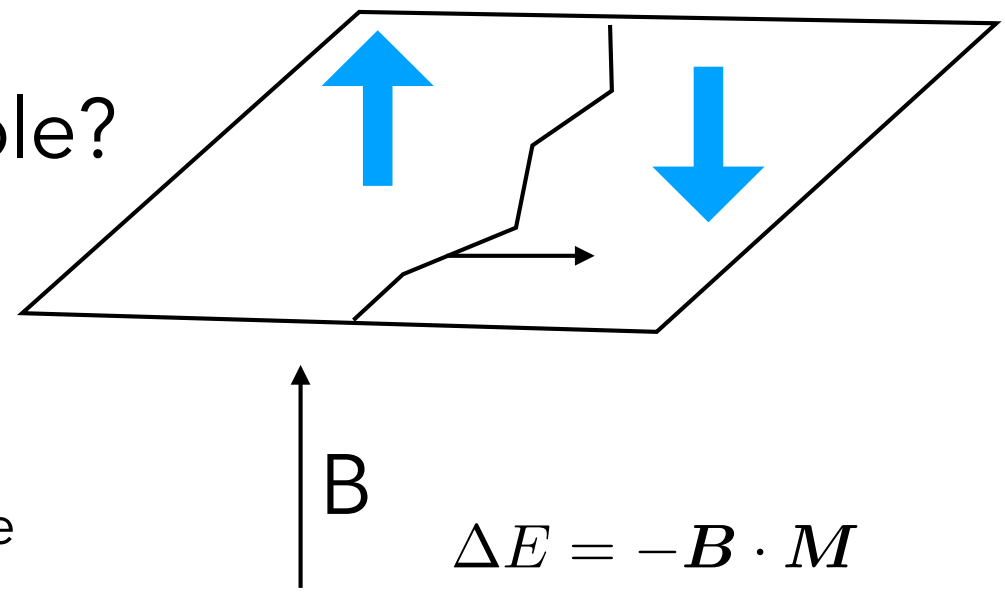


A. Sharpe *et al*, 2019



M. Serlin *et al*, 2019

How does the current couple?



For magnetic fields it is simple

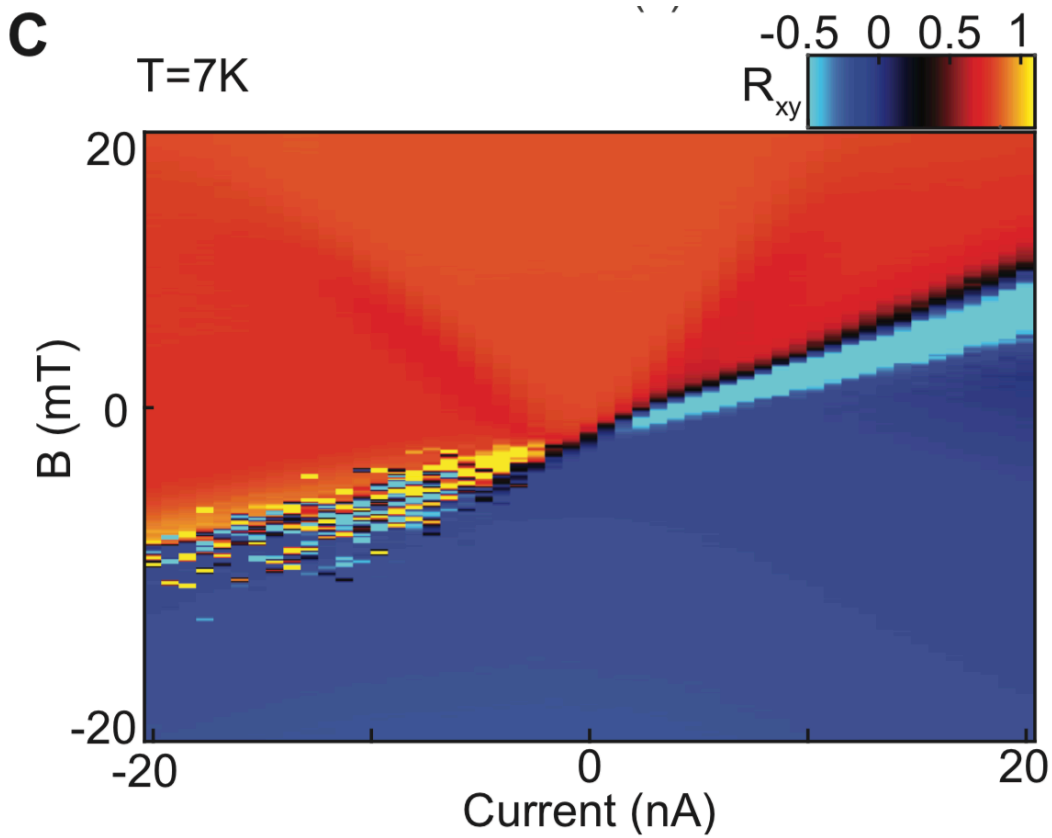
$$\Delta E = -\boldsymbol{B} \cdot \boldsymbol{M}$$

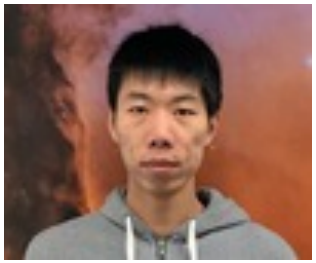
Dissipative Regime

Close to Curie temperature

Small Hall angle

Presumably no spin polarization



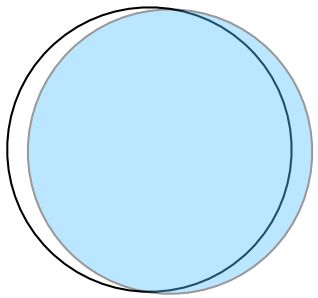


Xuzhe Ying

Mengxing Ye

A fully non-equilibrium problem, bulk 2d physics

Can current induce M?



Liouville theorem: semi-classical dynamics preserves phase space volume: Valley polarization is **not** induced by equations of motion

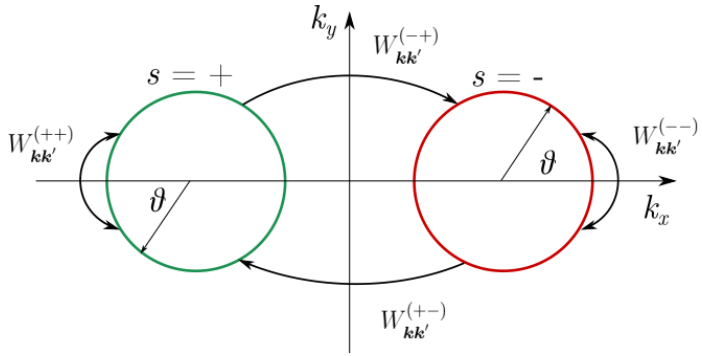
$$\partial_t f_{\boldsymbol{k}}^{(s)} + \boldsymbol{v}_{\boldsymbol{k}}^{(s)} \cdot \partial_{\boldsymbol{x}} f_{\boldsymbol{k}}^{(s)} + e\boldsymbol{E} \cdot \partial_{\boldsymbol{k}} f_{\boldsymbol{k}}^{(s)} = \sum_{s'=\pm} \int \mathrm{d}\Gamma' W_{\boldsymbol{k}\boldsymbol{k}'}^{(ss')} \left(f_{\boldsymbol{k}'}^{(s')} - f_{\boldsymbol{k}}^{(s)} \right) \delta(\epsilon_{\boldsymbol{k}'}^{(s')} - \epsilon_{\boldsymbol{k}}^{(s)})$$

In relaxation time approximation:

$$M = \alpha E \sim \tau E \sim j$$

$\phi = 0$ Valley polarization is <u>primary</u> order parameter, but not induced by ME mechanism (not in equilibrium)

Inter-valley motion



anisotropic scattering between valleys can induce polarization

$$\partial_t f^{(s)}_{\boldsymbol{k}} + \boldsymbol{v}^{(s)}_{\boldsymbol{k}} \cdot \partial_{\boldsymbol{x}} f^{(s)}_{\boldsymbol{k}} + e\boldsymbol{E} \cdot \partial_{\boldsymbol{k}} f^{(s)}_{\boldsymbol{k}} = \sum_{s'=\pm} \int \mathrm{d}\Gamma' W^{(ss')}_{\boldsymbol{k}\boldsymbol{k}'} \left(f^{(s')}_{\boldsymbol{k}'} - f^{(s)}_{\boldsymbol{k}} \right) \delta(\epsilon^{(s')}_{\boldsymbol{k}'} - \epsilon^{(s)}_{\boldsymbol{k}})$$

e.g.

$$W^{(-+)}_{\boldsymbol{k}\boldsymbol{k}'} = W^{(+-)}_{\boldsymbol{k}'\boldsymbol{k}} = \frac{1}{\nu\tau'} \left(1 + a_1 \cos\theta_{\boldsymbol{k}} + b_1 \sin\theta_{\boldsymbol{k}} + a'_1 \cos\theta_{\boldsymbol{k}'} + b'_1 \sin\theta_{\boldsymbol{k}'}\right)$$

$$\Delta n_0 = n^{(+)} - n^{(-)} = \tfrac{1}{2}\nu v_{\mathrm{F}} \tau \left[eE_x \left(a_1 + a'_1\right) + eE_y \left(b_1 + b'_1\right)\right] \propto \frac{\tau}{\tau'}$$

$$\Delta n_0 = \frac{1}{2hv_{\mathrm{F}}} \frac{h}{e^2} \left[ej_x \left(a_1 + a'_1\right) + ej_y \left(b_1 + b'_1\right)\right]$$

Result

Very rough estimates $\Delta n_0 \simeq \frac{1}{ev_{\mathrm{F}}} \boldsymbol{j} \cdot \boldsymbol{\delta}_\epsilon \propto \frac{\epsilon}{\theta_w} \frac{1}{hv_F} \frac{h}{e^2} ej$

This is the "bare" response just from quasiparticle physics. Should be included in a TDGL-like formulation as a force, to take into account both quasiparticle physics and interactions.

This gives $\alpha_4 \phi^3 + (r - r_c)\phi = \alpha_0 \Delta n_0$
describes hysteresis etc.

Quantitative estimates consistent with the observed effect

X. Ying, M. Ye, LB, PRB, in press

Thanks

Exciting new opportunities to study truly 2d magnetism in Van der Waals moiré materials

Twisted magnets: K. Hejazi, Z.-X. Luo, LB, PNAS **117**, 10721 (2020); ibid, arXiv:2009.00860.
QAHE and FM: M. Serlin *et al*, Science **367**, 900 (2019).
Current drive: X. Ying, M. Ye, LB, arXiv:2101.01790

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