Heavy Adatoms on Magnetic Surfaces: Chern insulator search

Kevin F. Garrity

and

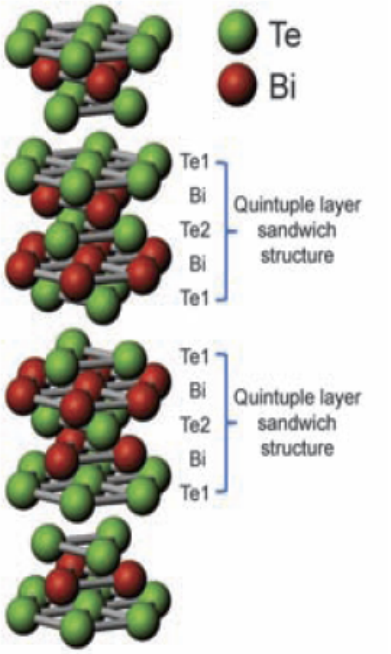
David Vanderbilt

Electronic Structure, 2013

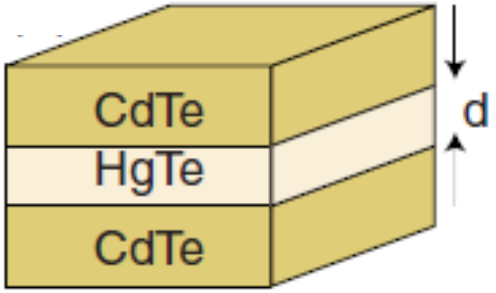
NSF DMR-10-05838

Topological Materials (TR-invariant)

3D: Strong Topological Insulator
Bi_2Se_3
Y.L. Chen et. al.
Science 325, 178 (2009)



2D: Quantum Spin Hall
HgTe-CdTe
M. Koning et. al.
Science 318, 766 (2007)



1988: QAH insulator (TR-broken)

VOLUME 61, NUMBER 18 PHYSICAL REVIEW LETTERS 31 OCTOBER 1988

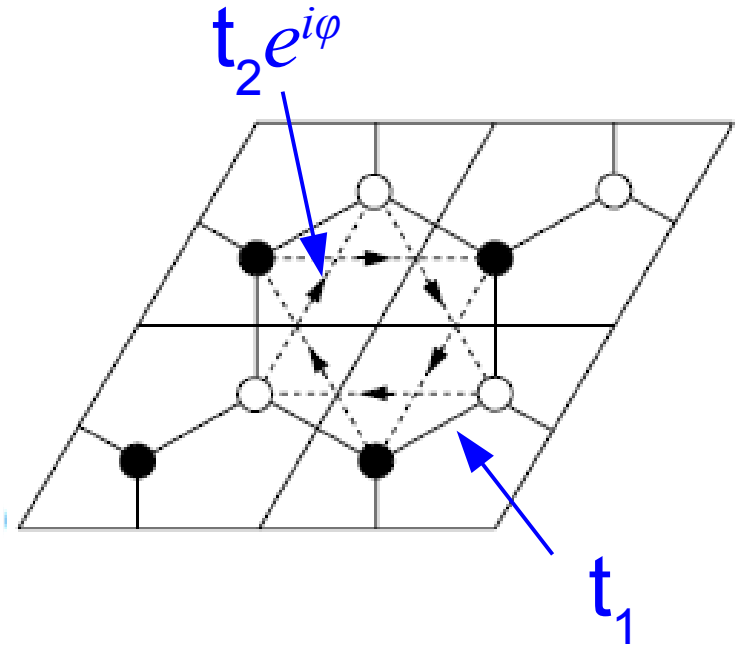
Model for a Quantum Hall Effect without Landau Levels: Condensed-Matter Realization of the "Parity Anomaly"

F. D. M. Haldane

Department of Physics, University of California, San Diego, La Jolla, California 92093

(Received 16 September 1987)

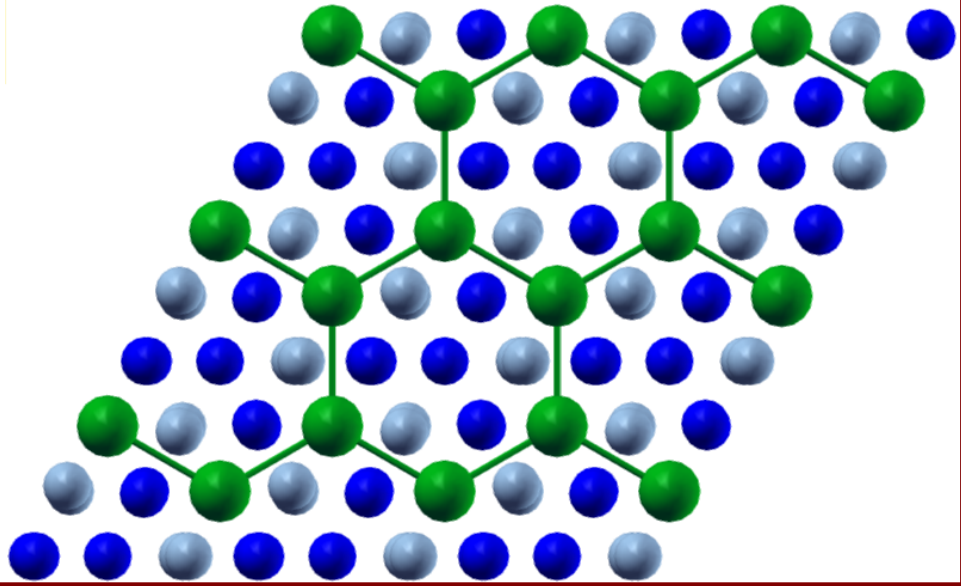
A two-dimensional condensed-matter lattice model is presented which exhibits a nonzero quantization of the Hall conductance σ^{xy} in the *absence* of an external magnetic field. Massless fermions *without spectral doubling* occur at critical values of the model parameters, and exhibit the so-called "parity anomaly" of (2+1)-dimensional field theories.



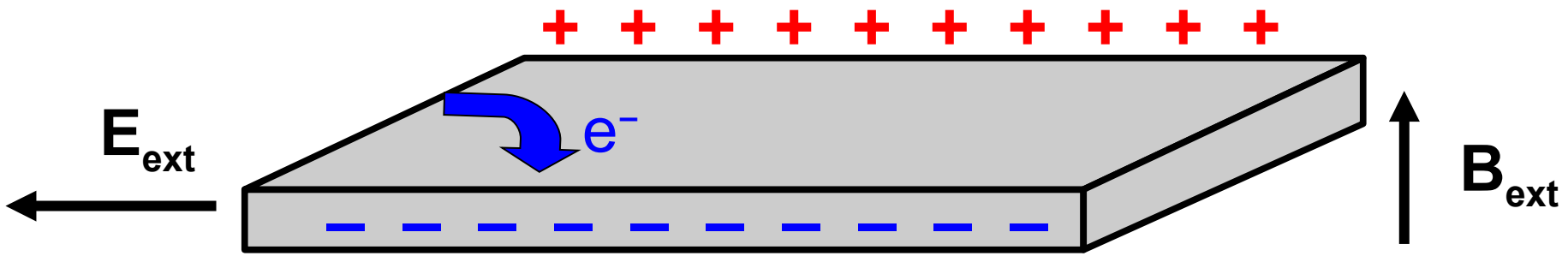
Quantum Anomalous Hall Insulator = Chern Insulator

Outline

- Introduction to Chern insulators
  - Berry curvature
  - Previous searches
- Our search strategy
  - Directly combine spin-orbit + magnetism
  - Produces many non-trivial band structures
- First principles verification
  - Several Chern insulators
- Conclusions



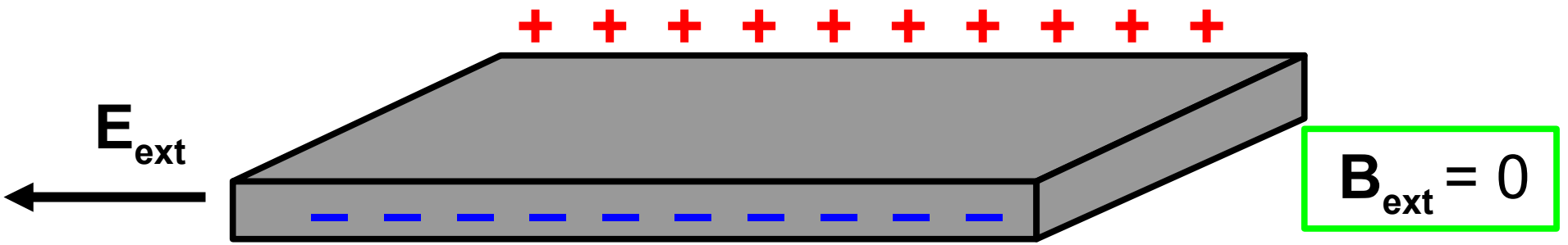
(Normal) Hall Effect



Normal Metal

- Electrons feel Lorentz force
- Charge builds up on sides

Anomalous Hall Effect



Ferromagnetic Metal

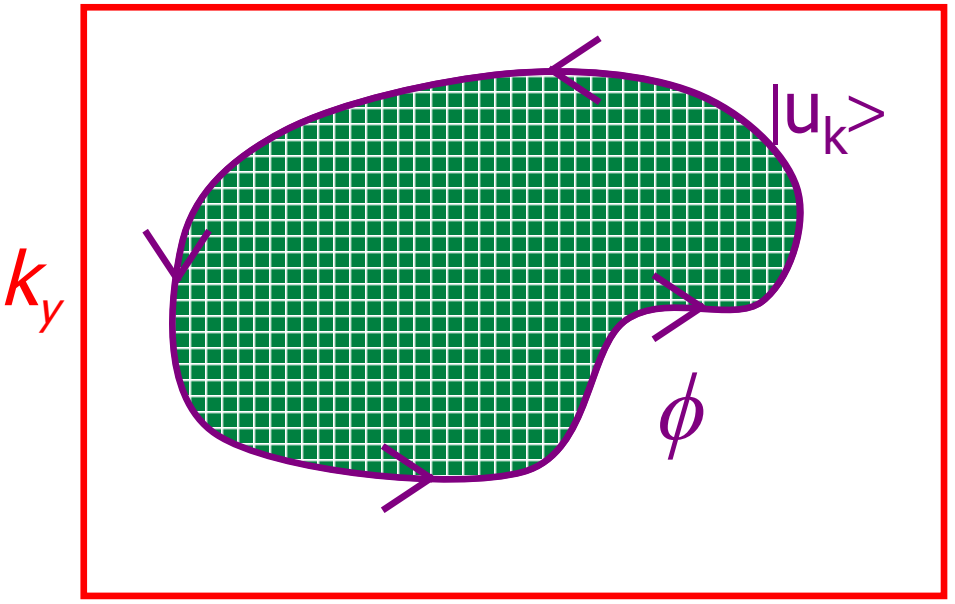
- No external magnetic field.
- Ferromagnetic metal (net **M**, breaks TR)
- Intrinsic contribution (single band):

Berry Curvature

$$\sigma_{xy} = -\frac{e^2}{2\pi h}\int d\mathbf{k} f(\epsilon_{\mathbf{k}})\Omega_z(\mathbf{k})$$

Karplus and Luttinger; Sundaram and Niu

Review: Berry phase and curvature



$$u_{\mathbf{k}}(\mathbf{r}) = e^{-i\mathbf{k}\cdot\mathbf{r}}\psi_{\mathbf{k}}(\mathbf{r})$$

Bloch function

Berry potential:

$$\mathbf{A}(\mathbf{k}) = -\mathrm{Im}\,\langle u_{\mathbf{k}}|\nabla_{\mathbf{k}}|u_{\mathbf{k}}\rangle$$

Berry phase:

$$\phi = \oint \mathbf{A}(\mathbf{k})\cdot d\mathbf{k}$$

mod 2π

Berry curvature:

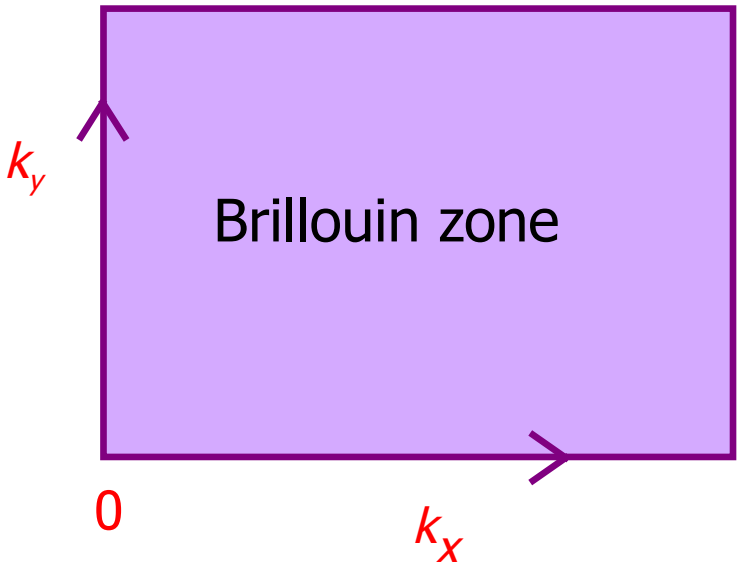
$$\Omega(\mathbf{k}) = \nabla \times \mathbf{A}$$

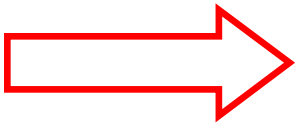
$$\Omega_z(\mathbf{k}) = -2\mathrm{Im}\left\langle \frac{du}{dk_x} \middle| \frac{du}{dk_y} \right\rangle$$

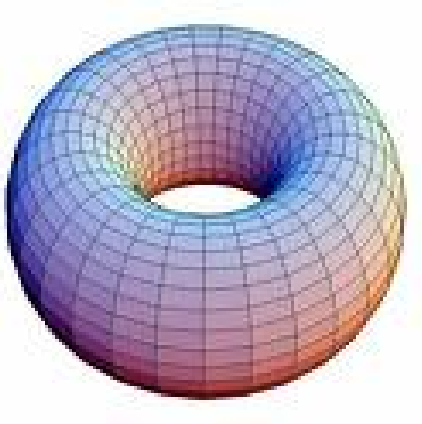
Stokes' theorem:

$$\phi = \int \Omega_z(\mathbf{k})\, d^2k$$

Review: Chern Theorem







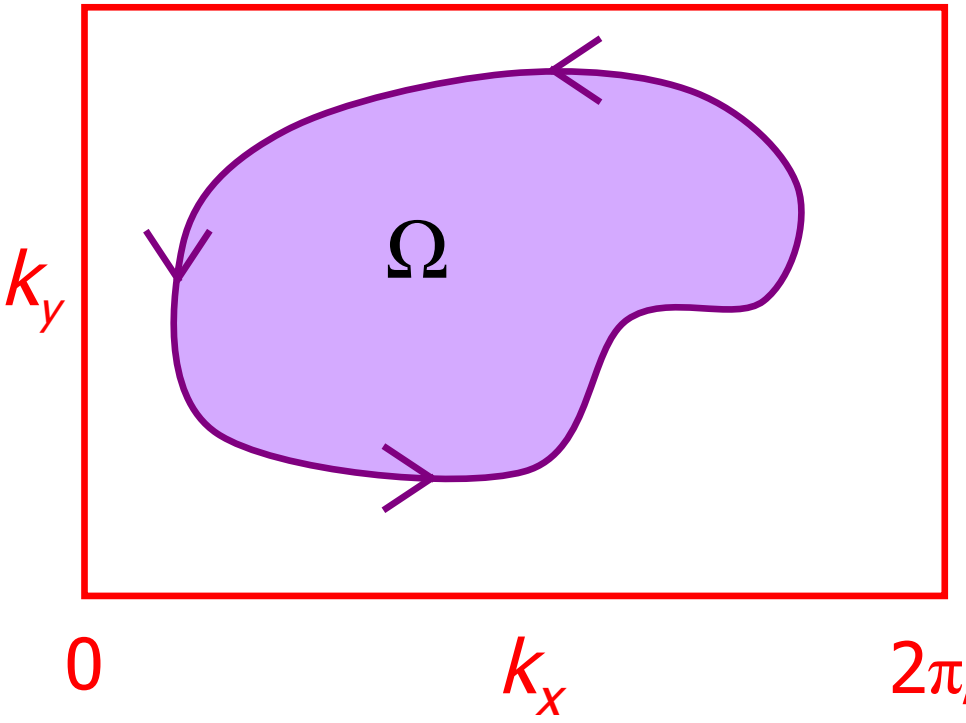
Brillouin zone is closed manifold

Chern theorem: $$\oint_{BZ} \Omega_z(\mathbf{k})\, d^2k = 2\pi\, C$$

Chern Number

Berry curvature and AHC

For Metals



$$\Omega_z(\mathbf{k}) = -2\mathrm{Im}\left\langle \frac{du}{dk_x} \middle| \frac{du}{dk_y} \right\rangle$$

$$\phi = \int_{\text{Fermi Sea}} \Omega_z(\mathbf{k})\, d^2k$$

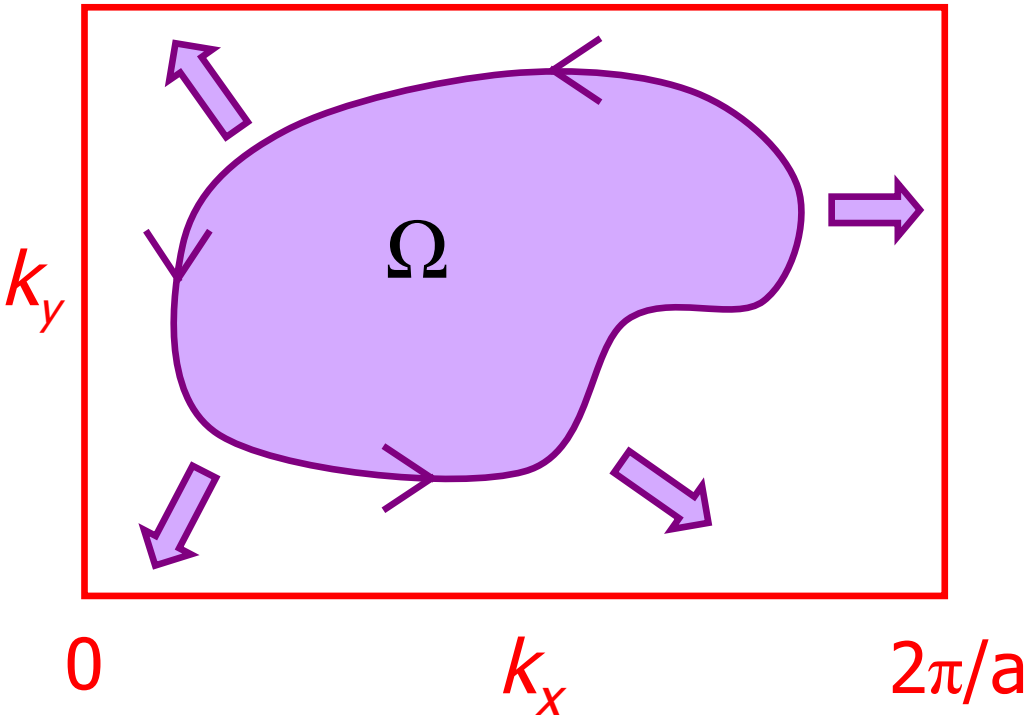
Metal

Anomalous Hall conductivity:

$$\sigma_{xy} = \frac{-e^2}{2\pi h}\,\phi$$

Karplus and Luttinger; Sundaram and Niu

Berry curvature and AHC



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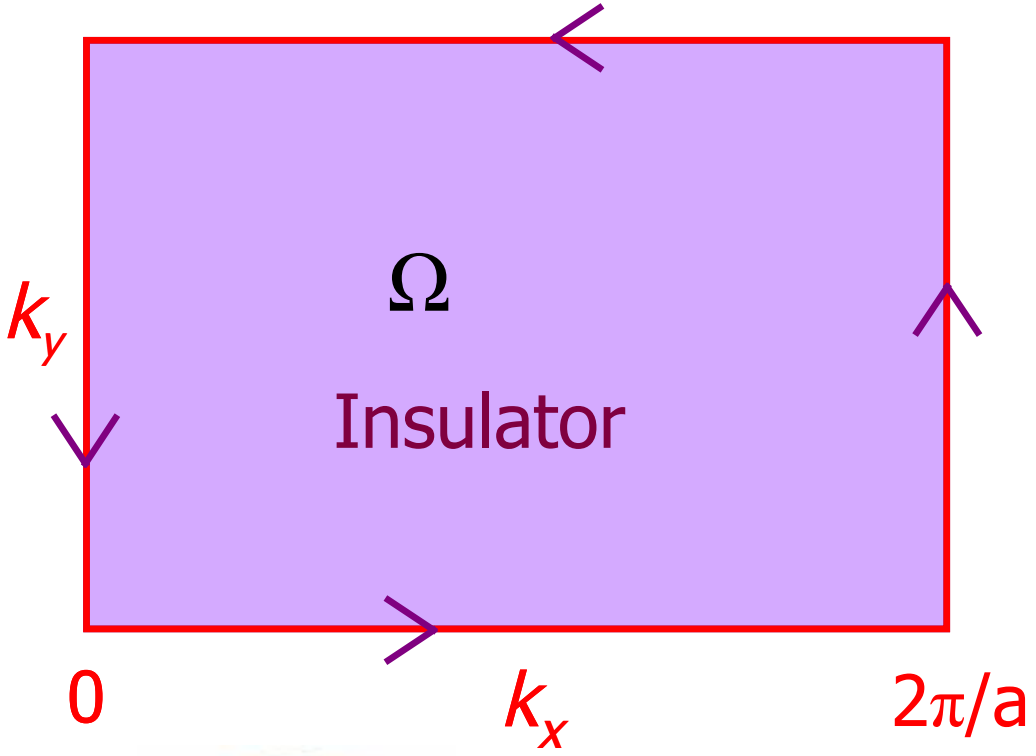

$$\Omega_z(\mathbf{k}) = -2\mathrm{Im}\left\langle \frac{du}{dk_x} \middle| \frac{du}{dk_y} \right\rangle$$

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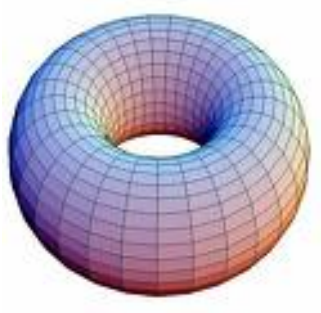


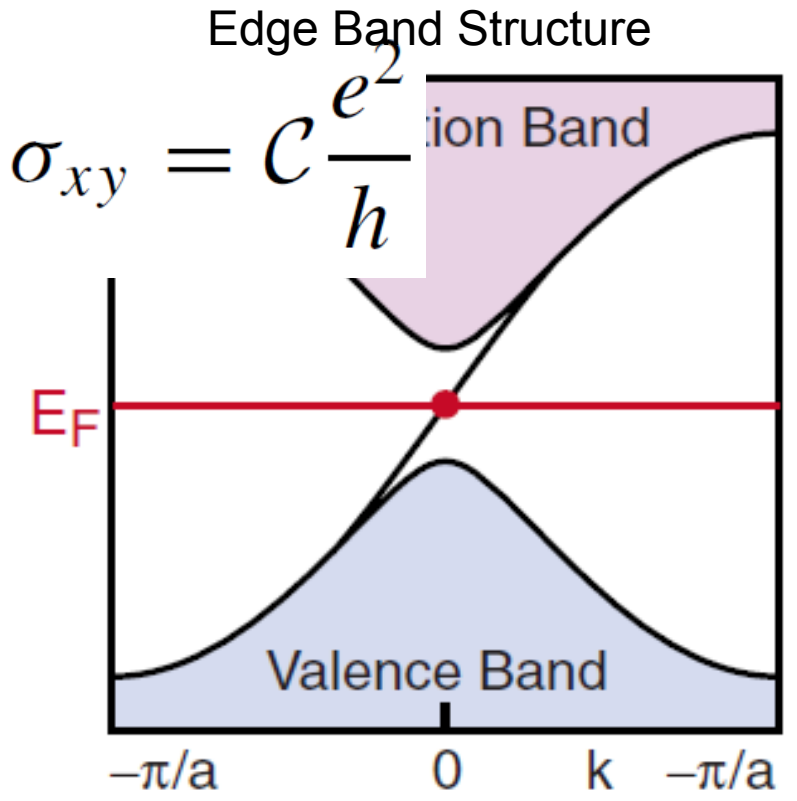
Quantum Anomalous Hall:

$$\sigma_{xy} = \frac{-e^2}{h} C$$

"Chern number" or "TKNN invariant"

2D Chern Insulators

- **Requirements:**
  - Bulk band gap
  - Broken time-reversal (TR)
  - Spin-orbit coupling
- **Features:**
  - Spin-polarized edge state
    - Dissipationless transport
  - Magnetoelectric effects
- **Questions:**
  - How to construct?
  - Large gap? (room temperature?)



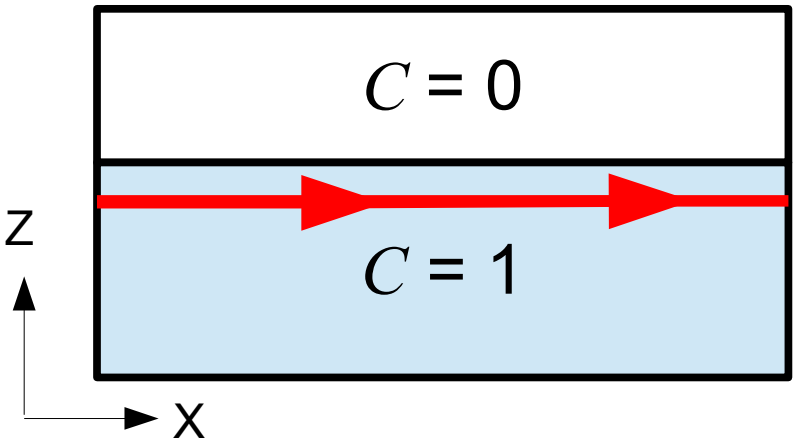


Image: Hasan et. al. RMP **80**, 3245 (2010)

Previous Proposals

- Haldane Model
- Magnetically dope known topological system:[1]
  - Mn in HgTe[2-3], Cr/Fe in Bi_2Se_3[4-7], Fe on graphene[8], Bi-bilayers[9]
- Stoichiometric compounds:
  - $GdBiTe_3$[10], $HgCr_2Se_4$[11]
- Difficult to achieve experimentally
  - Leaky, small gaps
  - Align spins

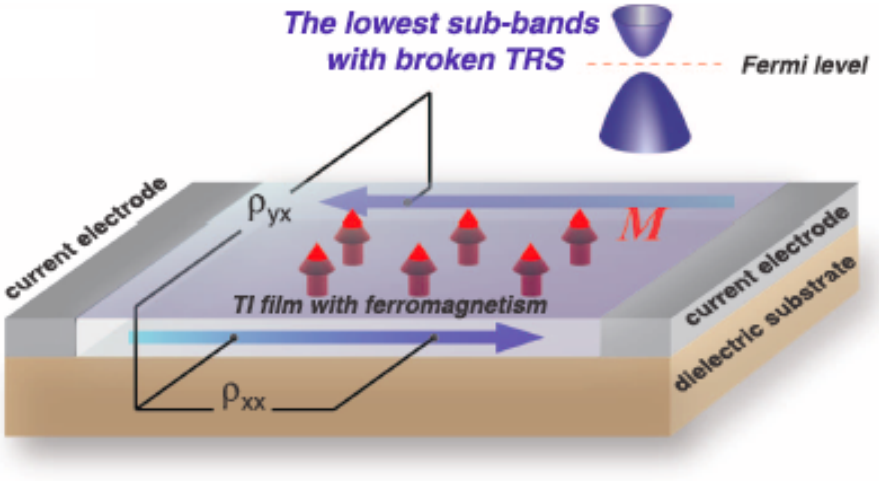
[1]Jiang *et. al. PRB* 85 045445 (2012)
[2]Liu et. al. *PRL* 101 146802 (2008)
[3]Buhmann. *March Meeting* P27.1 (2012)
[4]Yu *et. al. Science* 32961 (2010)
[5]Kou *et. al. J. Appl. Phys.* 112 063912 (2012)
[6]Xu *et. al. Nat. Phys.* 8 (2012)
[7]Niu *et. al. APL* 99 142502 (2011)
[8]Qiao *et. al. PRB* 82 161414 (2010)
[9]Zhang *et. al.* arXiv:13014426 (2013)
[10]Zhang *et. al.* ArXiv:1108.4857 (2011)
[11]Xu *et. al. PRL* 186806 (2011)

Magnetic doping: Claim for QAH

www.sciencemag.org SCIENCE VOL 340 12 APRIL 2013

Experimental Observation of the Quantum Anomalous Hall Effect in a Magnetic Topological Insulator

Cui-Zu Chang,[1,2*] Jinsong Zhang,[1*] Xiao Feng,[1,2*] Jie Shen,[2*] Zuocheng Zhang,[1] Minghua Guo,[1] Kang Li,[2] Yunbo Ou,[2] Pang Wei,[2] Li-Li Wang,[2] Zhong-Qing Ji,[2] Yang Feng,[1] Shuaihua Ji,[1] Xi Chen,[1] Jinfeng Jia,[1] Xi Dai,[2] Zhong Fang,[2] Shou-Cheng Zhang,[3] Ke He,[2†] Yayu Wang,[1†] Li Lu,[2] Xu-Cun Ma,[2] Qi-Kun Xue[1†]

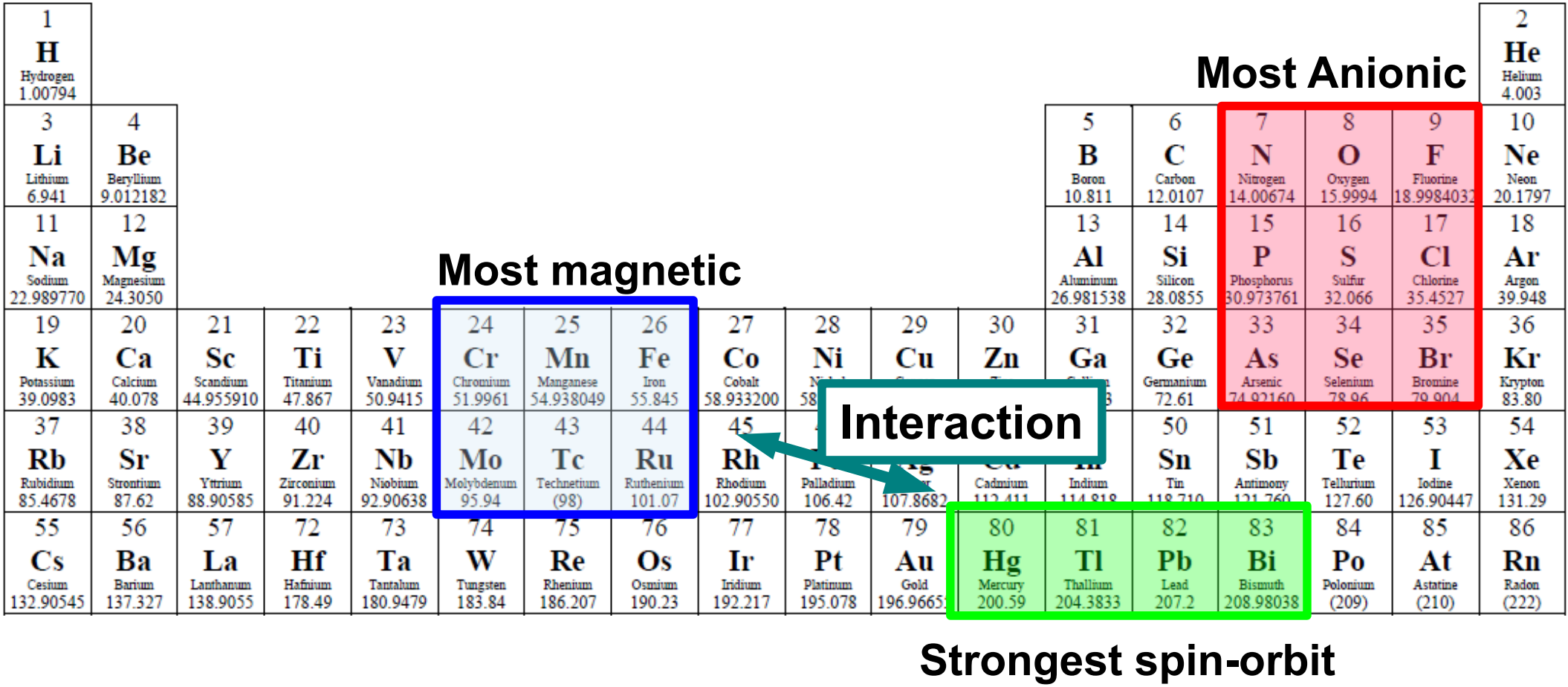


Observed below ~1K

$(Bi,Sb)_2Te_3$ doped with Cr

Aside on chemistry

- Need strong magnetism, spin-orbit, insulating
- Hard to find all three together.
- We combine on surface.



Our Strategy

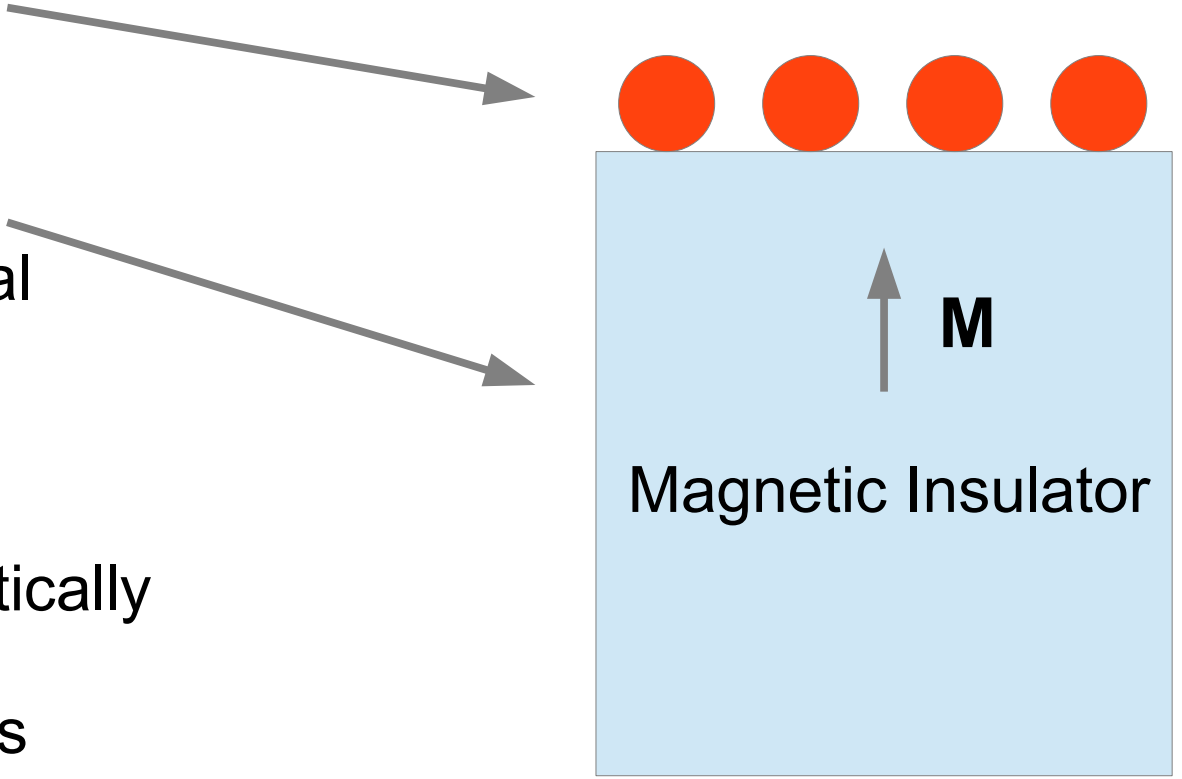
Combine: heavy atoms + magnetic insulator

Heavy Atoms
- Au to Bi
- Large spin-orbit

Magnetic Insulator
- Breaks time reversal
- FM or A-type AFM
- Topologically trivial

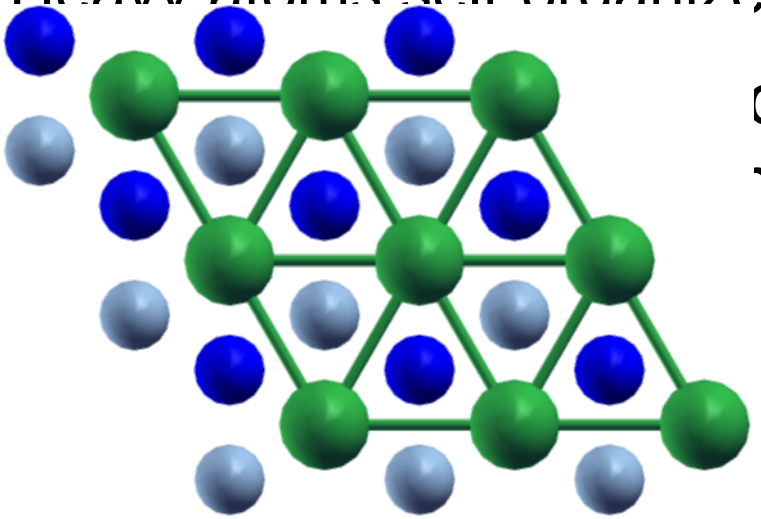
Advantages:
- Spins align automatically
- No doping
- Large gap insulators
- Large spin-orbit

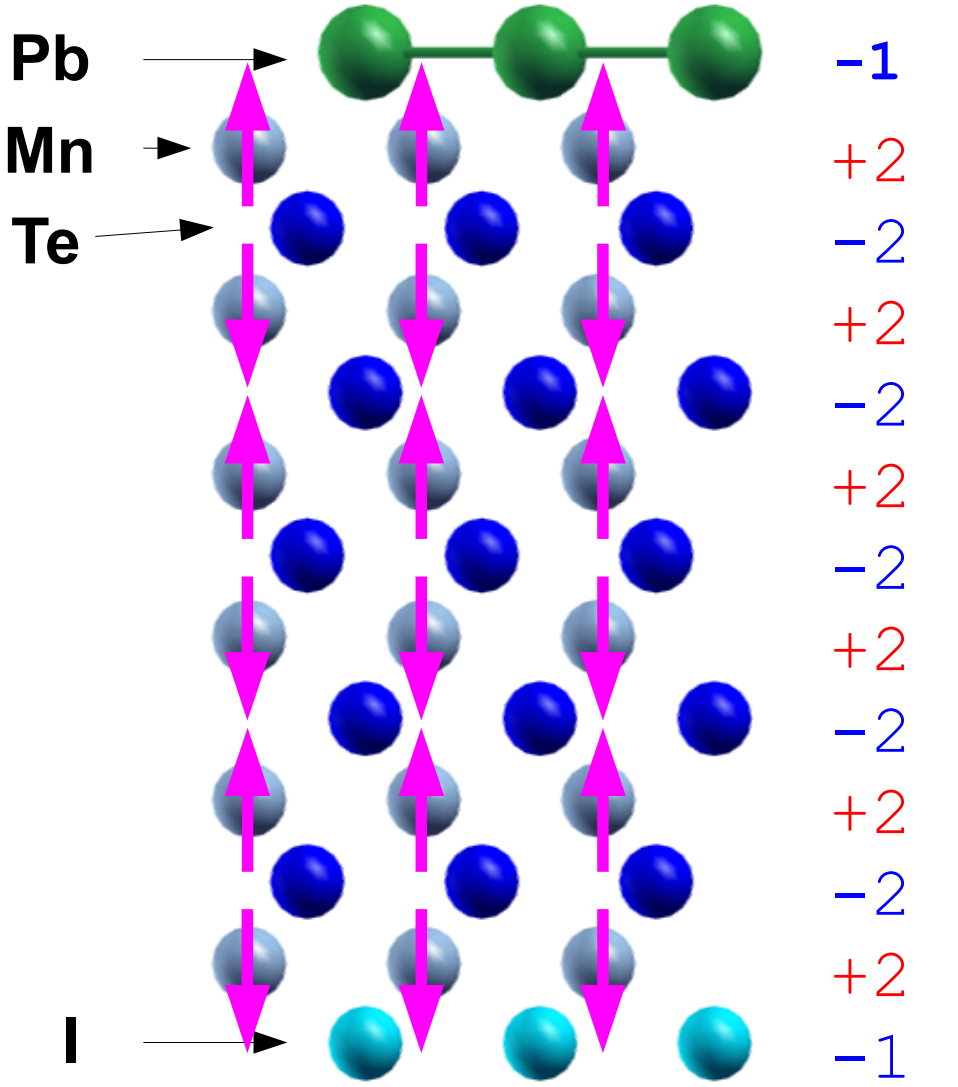
Disadvantages:
- Hard to prepare surface



First Principles Unit Cell

- Substrates:
  - MnTe, MnSe (A-type AFM)
  - EuS (FM)
- 0-1 ML heavy atom
  - Au to Bi
- Polar surface
  - Cleave surface in vacuum
  - Heavy atoms self-organize
- St on
  a netry





Calculation Details

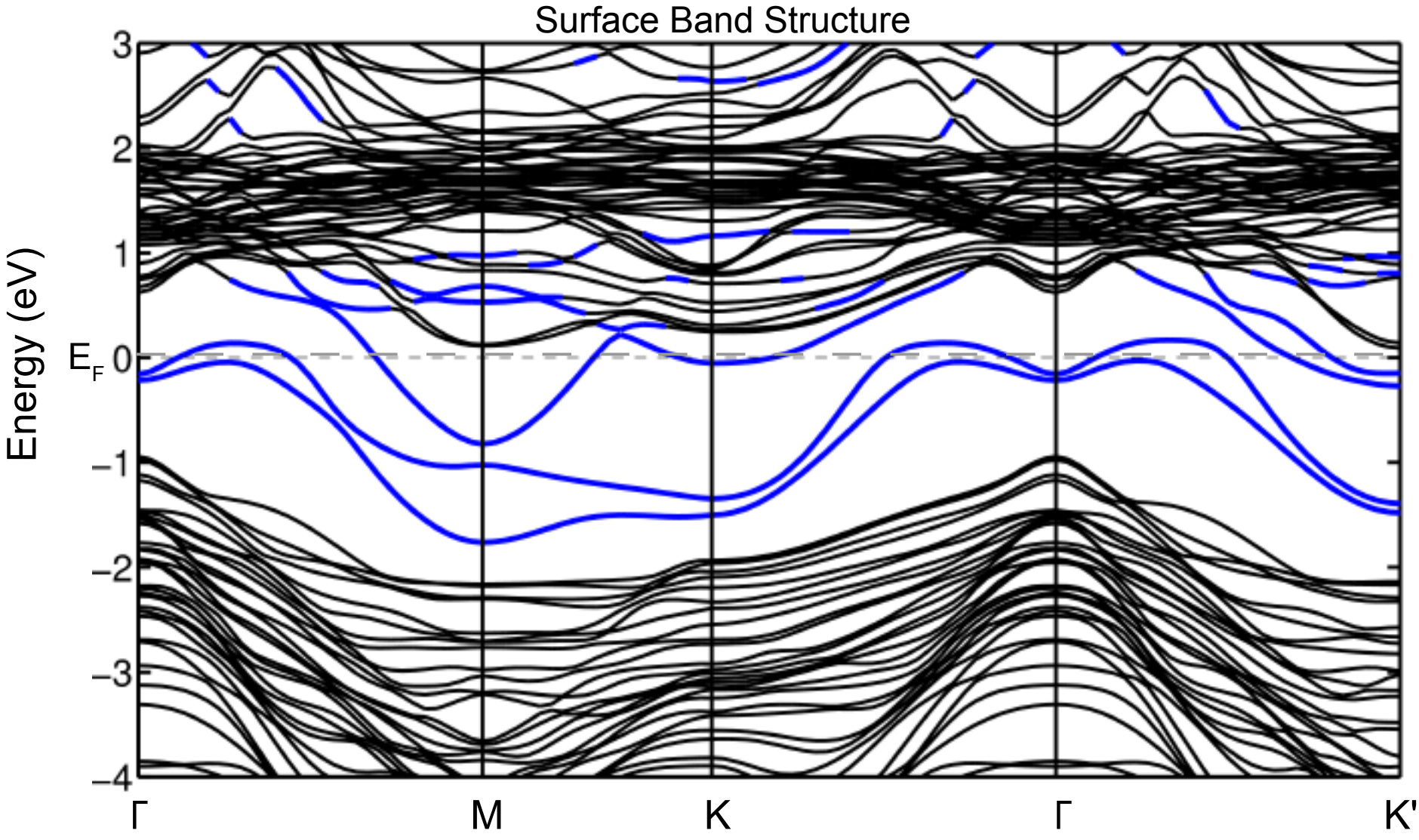
- Quantum Espresso with OPIUM norm-conserving potentials
- VASP PAW's
- LDA + U
  - U=5 eV for Mn, 6 eV for Eu
- wannier90 → Interpolation to compute Chern numbers

$$C = \frac{1}{2\pi}\int_{\mathrm{BZ}} d\mathbf{k}\Omega(\mathbf{k}) = \frac{1}{2\pi}\oint_{\mathrm{BZ}} d\mathbf{k}\cdot\mathbf{A}(\mathbf{k})$$

- wannier90 post-processing code AHC[1-2]

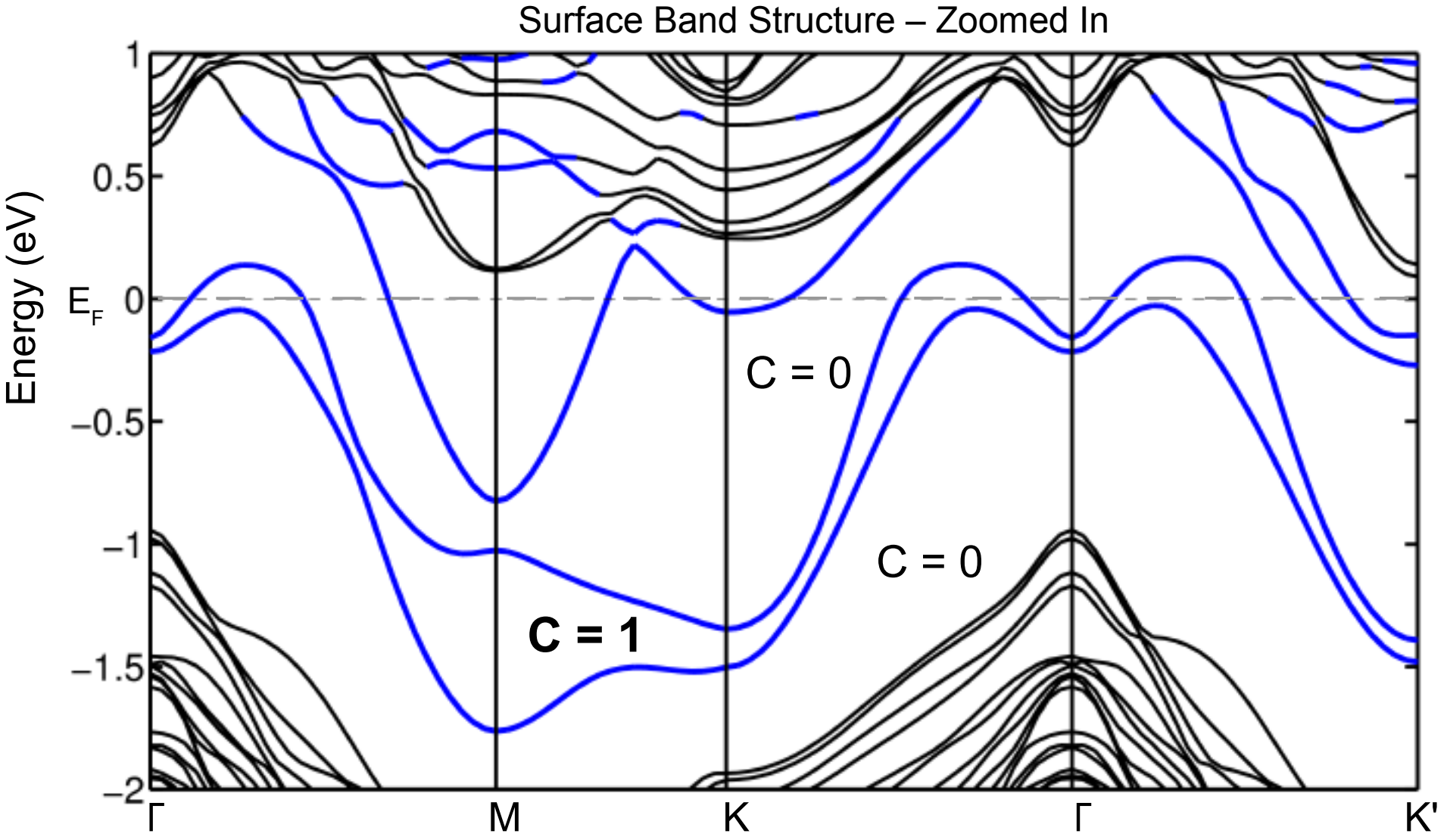
[1]Wang *et. al.* PRB 74 195118 (2006)
[2]Wang *et. al.* PRB 76 195109 (2007)

1 ML Tl on MnTe



- Isolated surface bands

1 ML Tl on MnTe



- Isolated surface bands
- Non-trivial Chern numbers
- Metallic

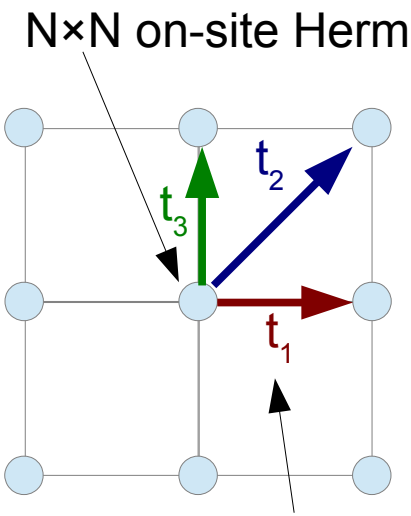
Observation 1 – No degeneracies

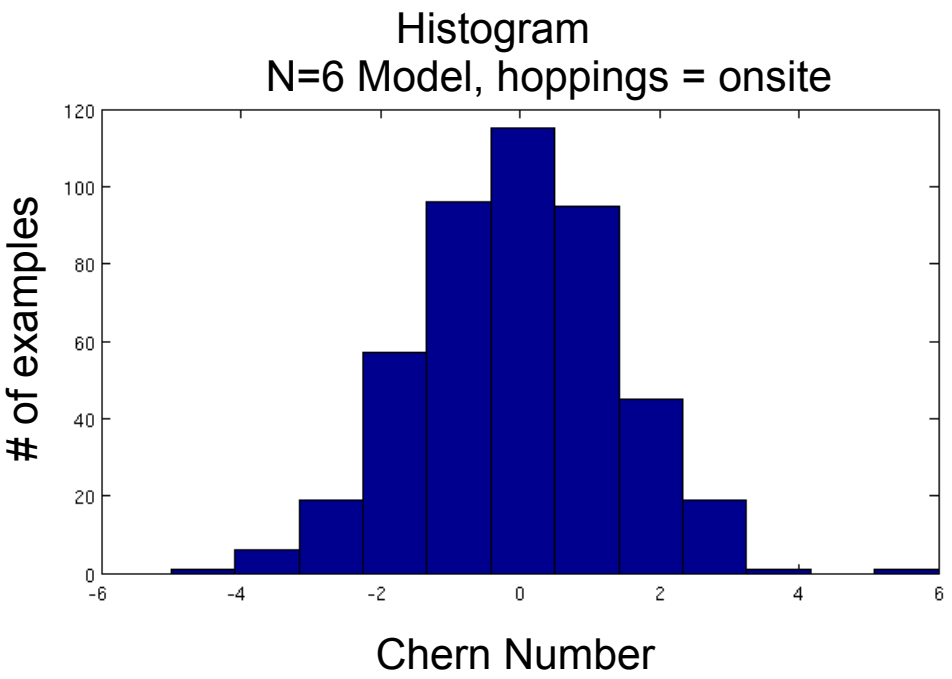
2D → Isolated bands

- SO + broken TR = no high symmetry degeneracies
- No accidental degeneracies:
  - Near Crossing $H = \vec{h} \cdot \vec{\sigma}$ ← Pauli matrices
  - Degeneracy → $h_x = h_y = h_z = 0$
  - Only vary (k_x, k_y)

Observation 2 – Many Chern #s

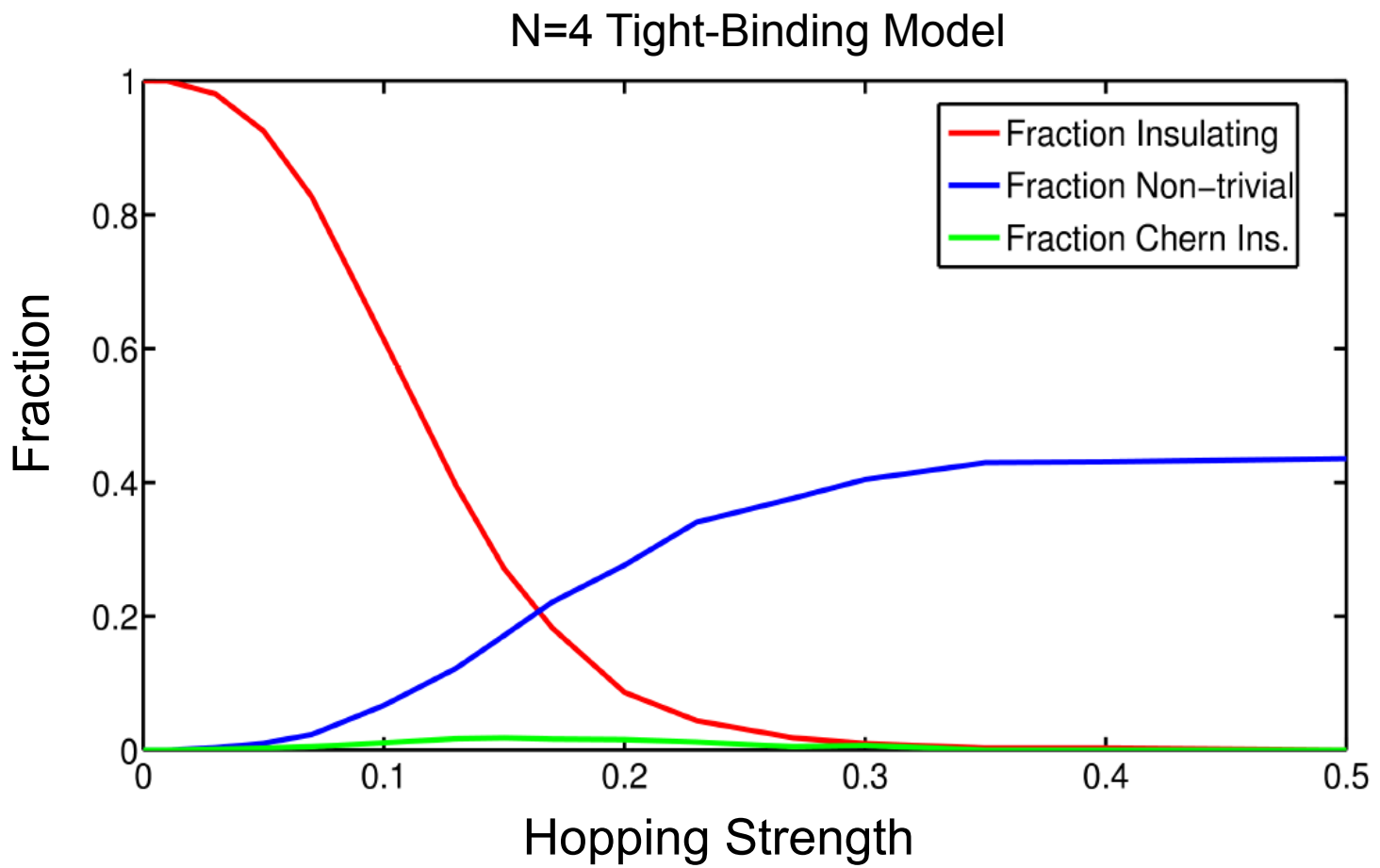
- If spin-orbit, magnetic exchange, hoppings similar strength
- Then: many non-trivial Chern numbers
- Example – random N-orbital tight-binding model
  - Random complex on-site & hopping parameters
  - 1st n.n., diagonal hoppings





Need Chern # + Gap across BZ

- Competition between Chern numbers and gaps



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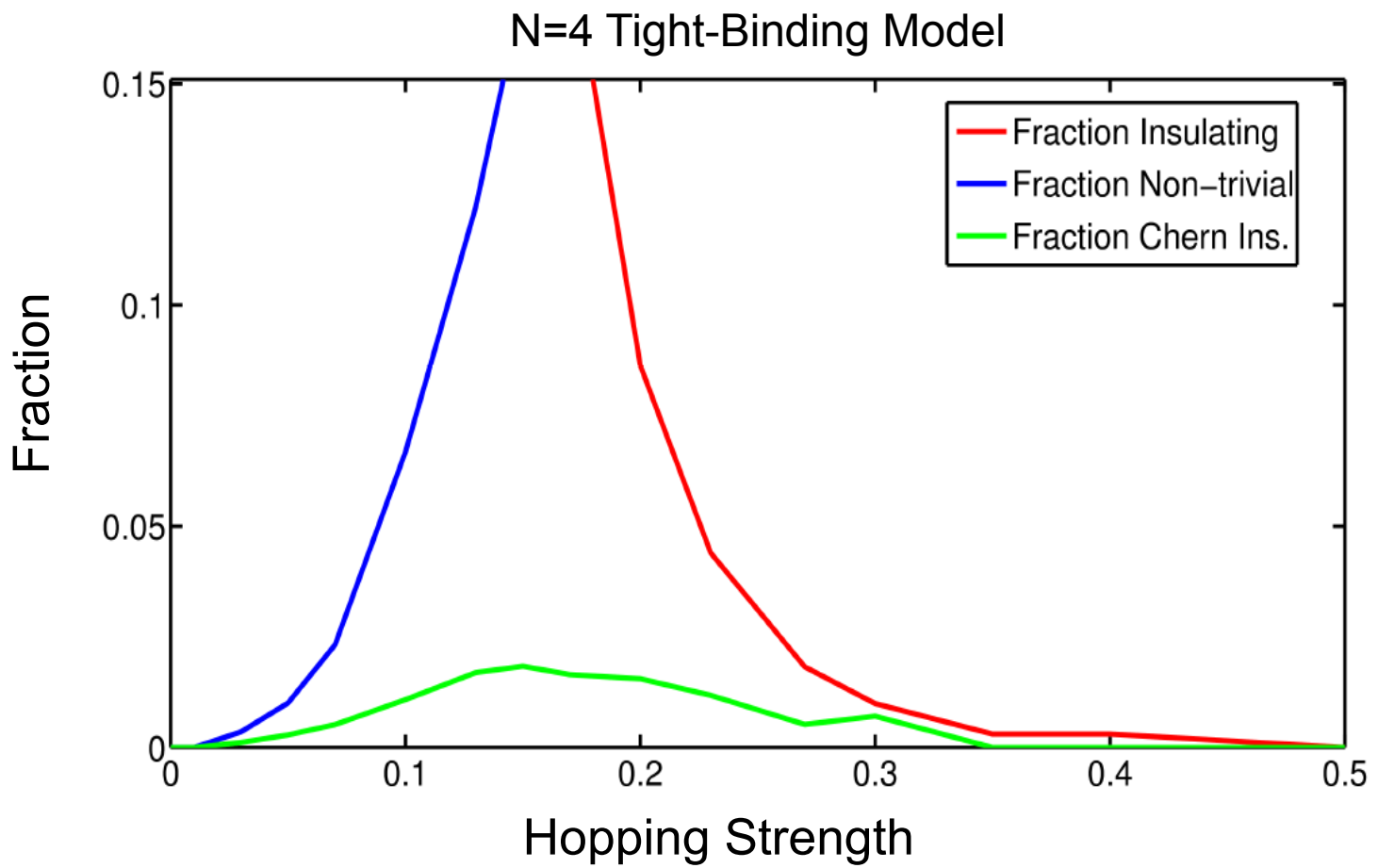
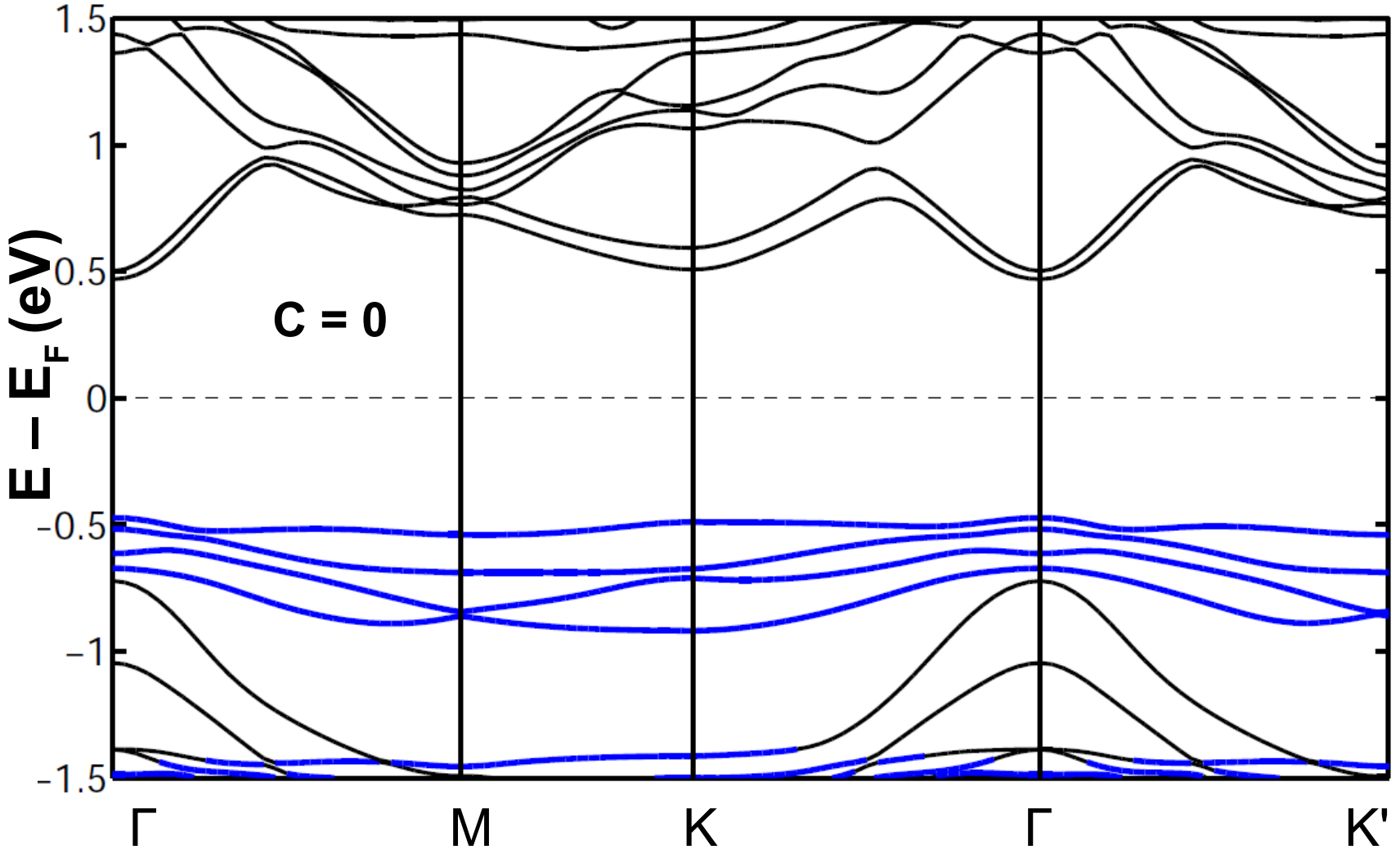


- Need to reduce band dispersion.
  - Idea: lower adatom coverage

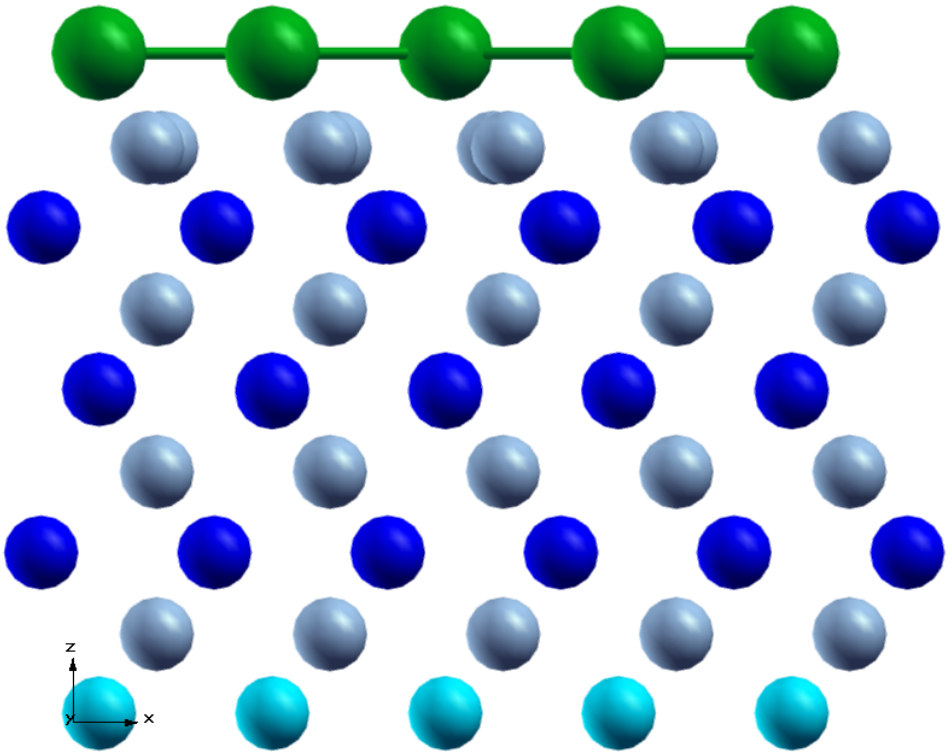
Attempt II: 1/3 ML Bi on MnSe

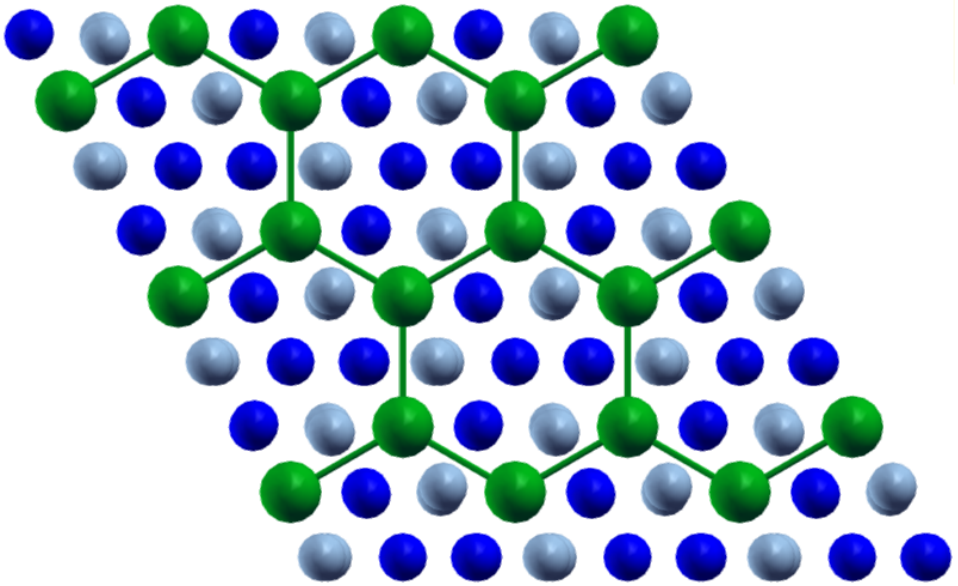


- Too little hopping, no non-trival Chern numbers.

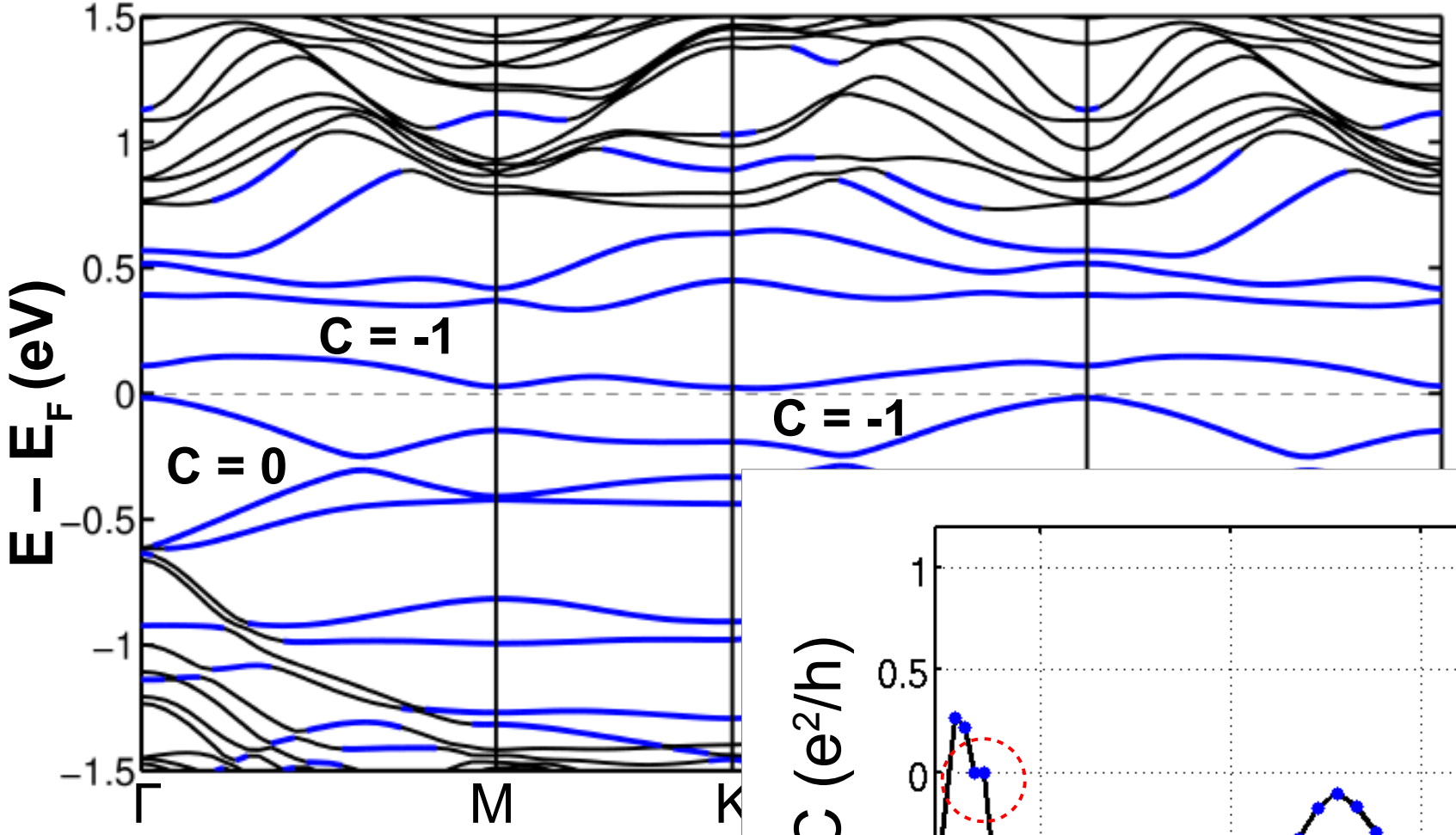
Attempt III - Honeycomb

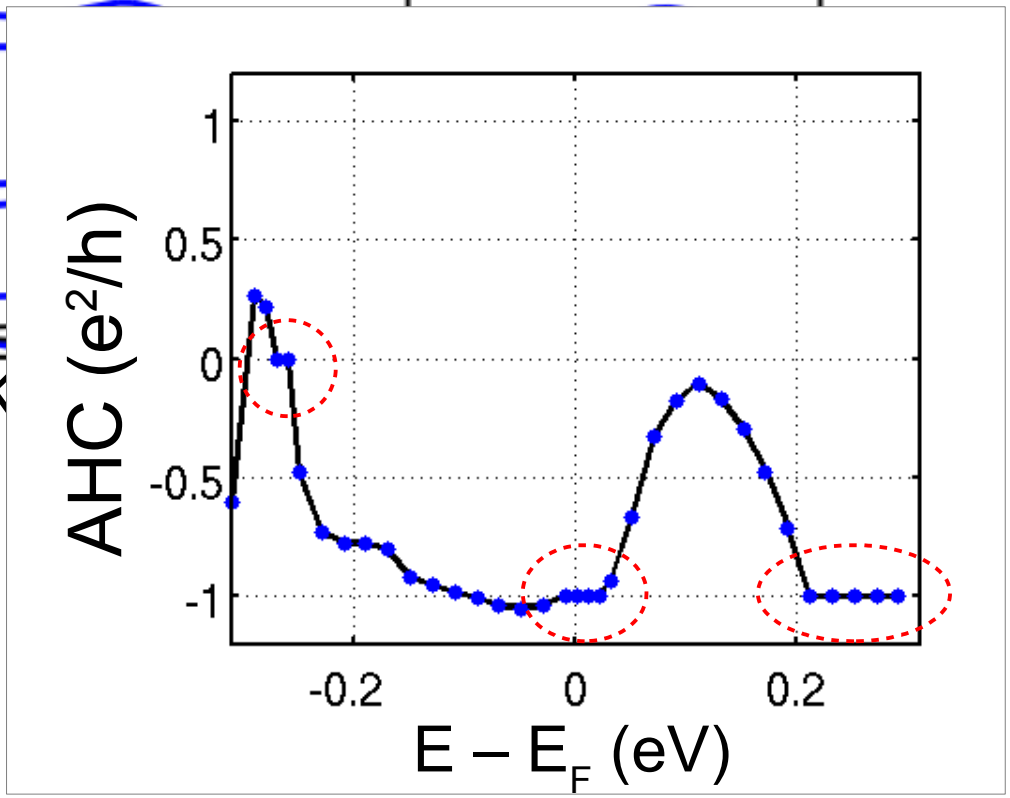
- Idea: tune surface hopping
- 2/3 ML honeycomb
  - Triples u.c.
  - Doubles # adatom bands





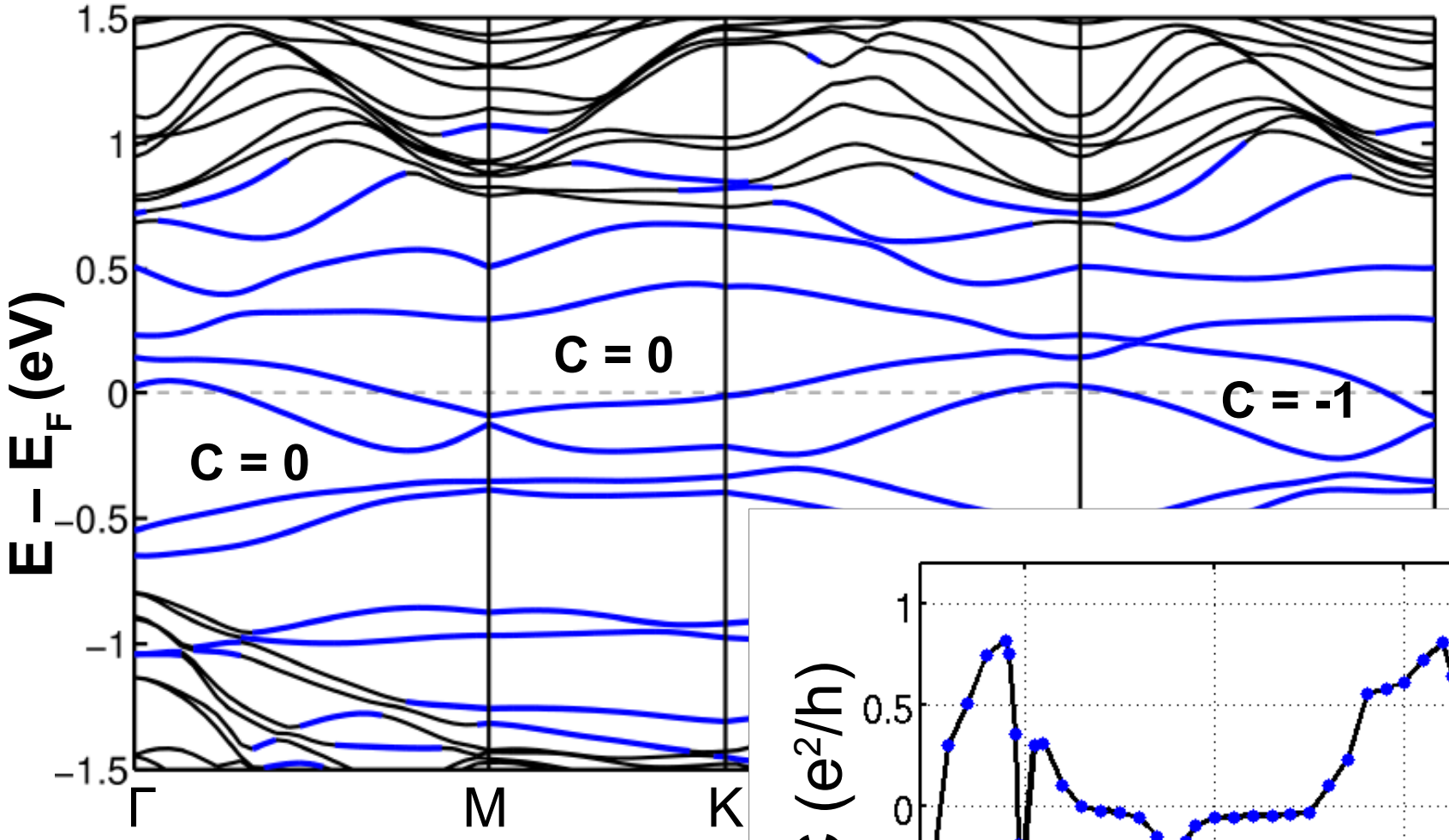
2/3 ML Pb on MnTe – *z* spins

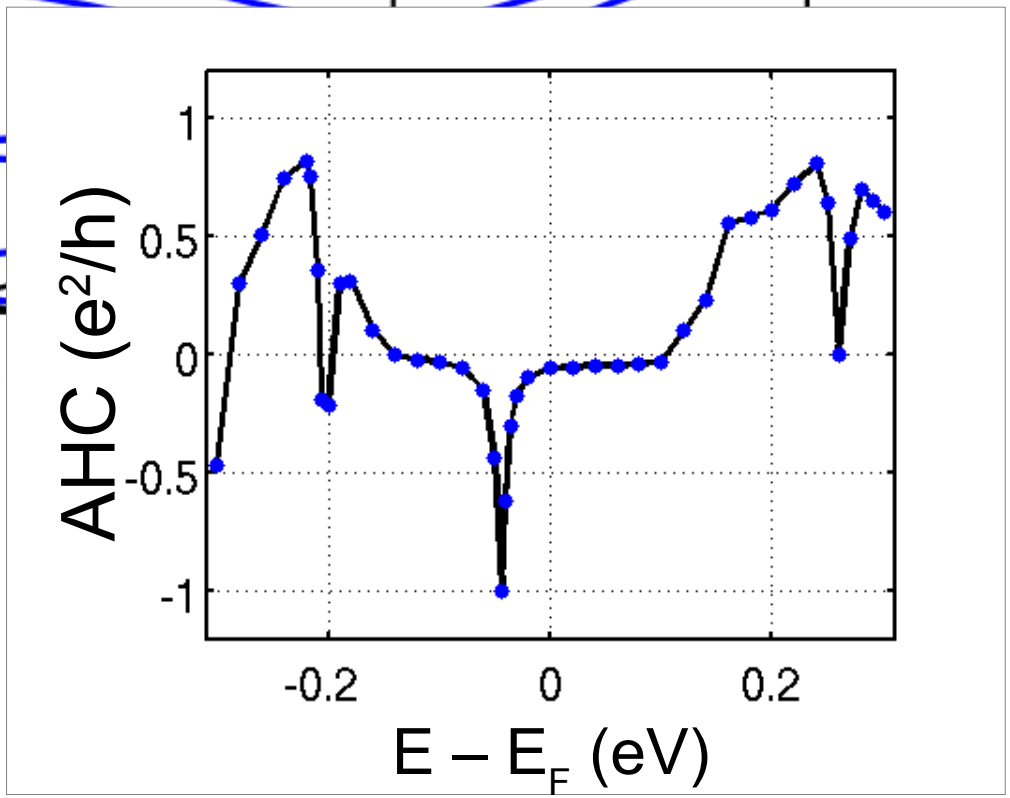




- Isolated surface bands
- Non-trivial Chern numbers
- Insulating

2/3 ML Pb on MnTe – *x* spins

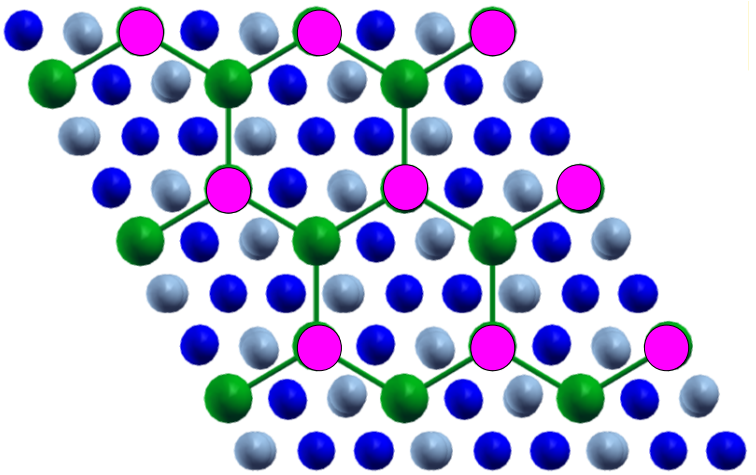




- Isolated surface bands
- Non-trivial Chern numbers
- Metallic

Search for Chern Insulators

- **Ingredients:**
  - Substrates:
    - MnTe, EuS
    - MnSe (-2% epitaxial strain to align spins along **z**)
  - 1-2 Heavy adatoms per tripled u.c.
  - 0-2 Anion:
    - Br, I, Se, Te
    - Adjust Fermi level
- **Screening Procedure:**
  - Initial calculations with 2 ML substrate.
    - ~50% non-trivial, ~20% Chern insulators
  - If interesting, final calculations 4 ML.

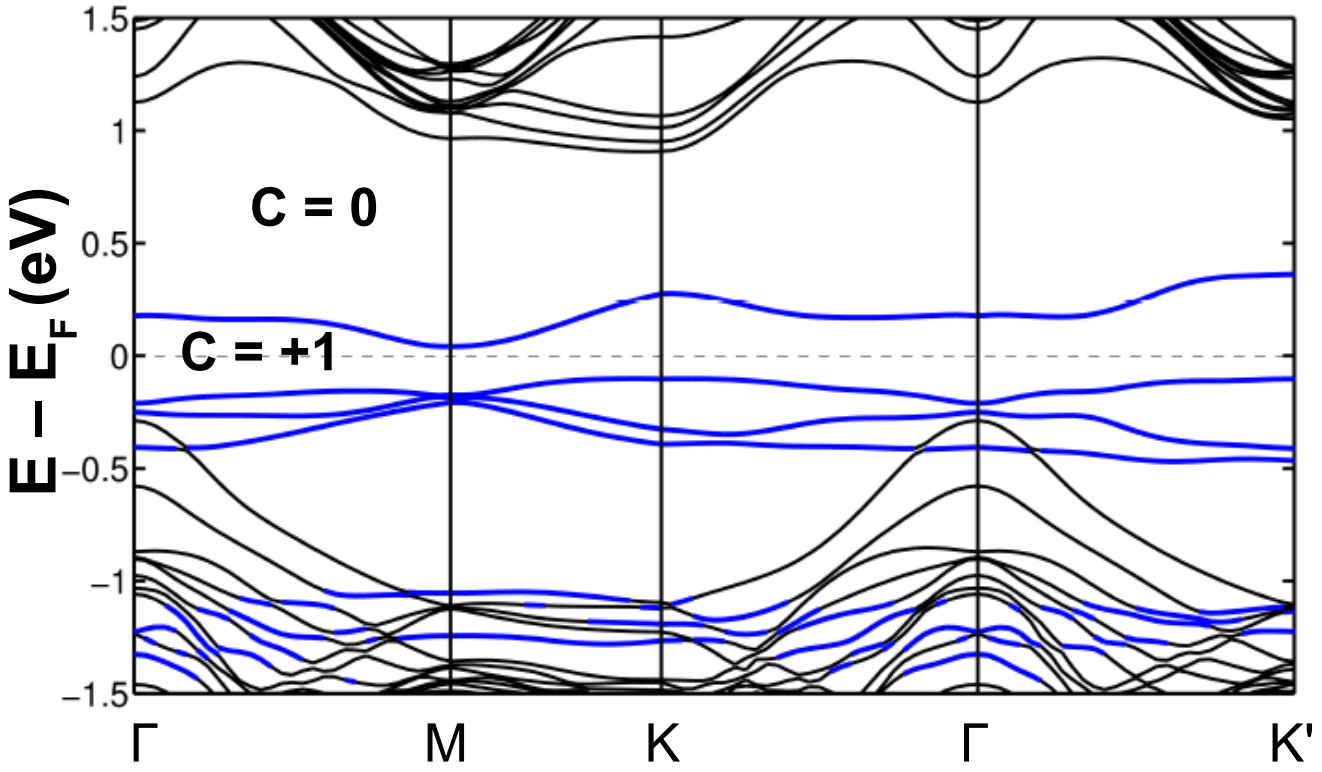


Search for Chern Insulators

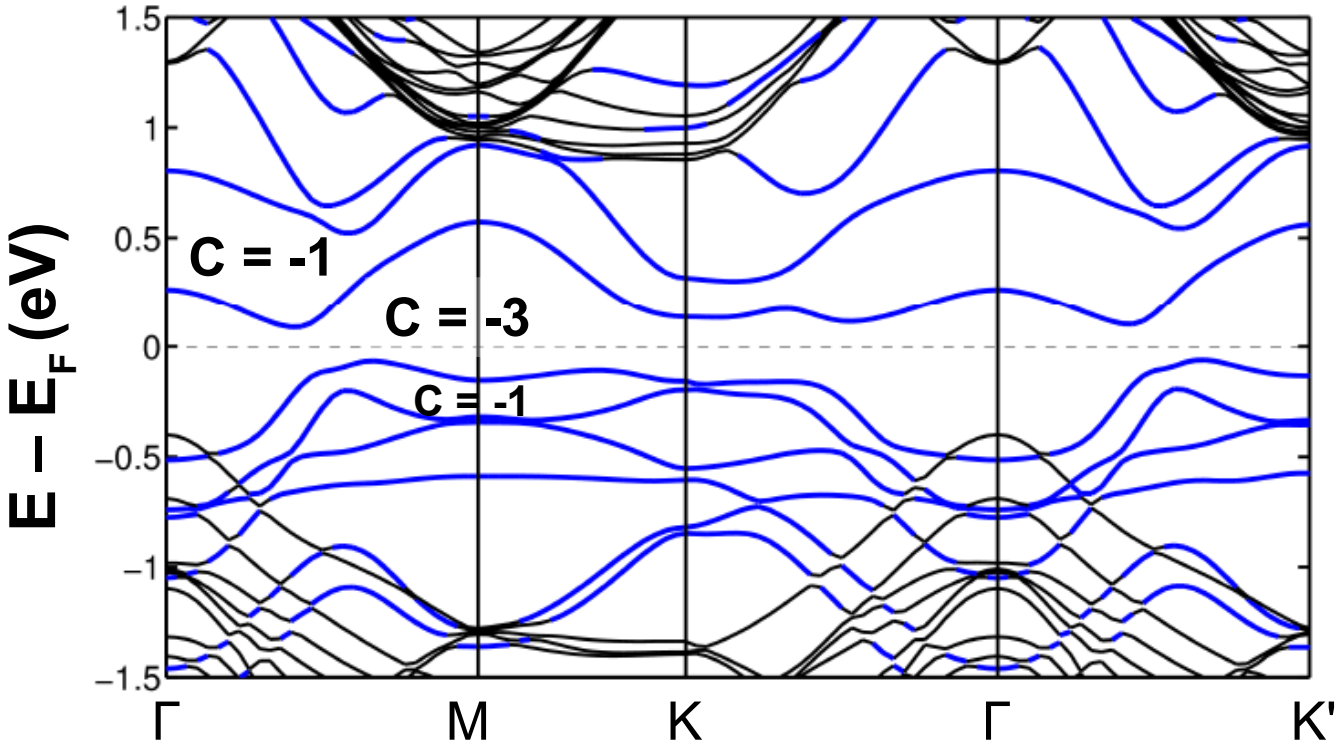
Substrate	Surface	Spin direction	C	E_g^{dir} (meV)	E_g^{indir} (meV)
MnTe	**AuAu**	z	1	141	36
MnTe	AuAu	x	m	m	m
MnTe	HgHg	z	0	31	−341
MnTe	TlTl	z	m	m	m
MnTe	**PbPb**	z	−1	126	36
MnTe	PbPb	x	−1	12	−156
MnTe	BiBi	z	m	m	m
MnSe	Pb	z	0	314	123
MnSe	AuAu	z	1	64	−731
MnSe	**PbPb**	z	−1	213	1
MnSe	PbPb	x	−1	12	−103
MnSe	PbBi	z	−2	31	−9
MnSe	**PbPbI**	z	−3	84	56
MnSe	**BiI**	z	1	302	41
MnSe	**BiBr**	z	1	213	142
MnSe	TlI	z	0	5	−53
MnSe	HgSe	z	−1	22	−23
EuS	PbPb	z	−1	91	−48
EuS	AuAu	z	0	188	−251

Strained
-2%

BiBr on MnSe
142 meV gap

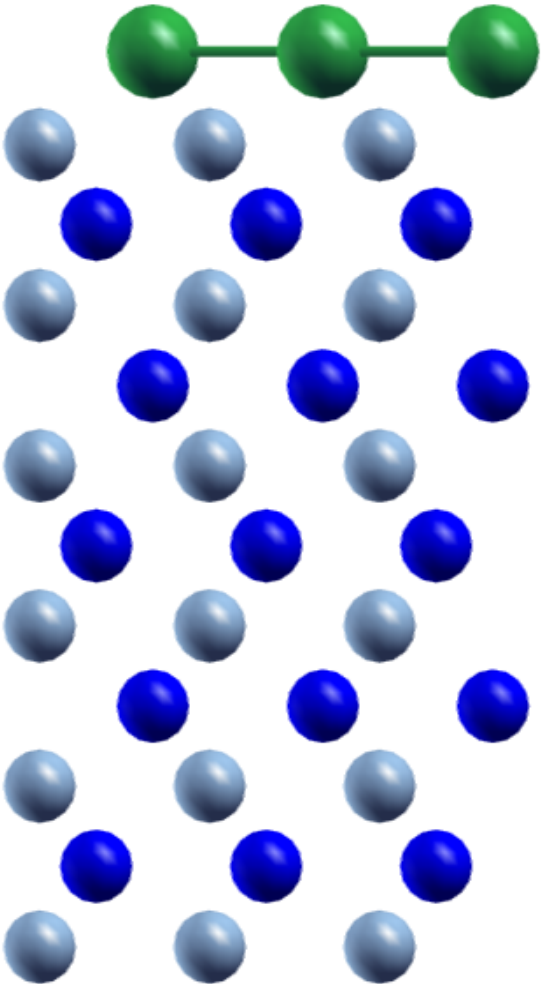


PbPbI on MnSe
56 meV gap



Future Work: Experimental/theoretical search

- Heavy atoms + Magnetic Insulator = Many CI's
- These examples for computational convenience
- Which surfaces can be prepared?
  - Many magnetic insulators
  - Deposit low coverages heavy atoms
    - Characterize surface
    - Look for large AHC
  - Theoretical input to modify surface
    - Make insulating / non-trivial



Conclusions

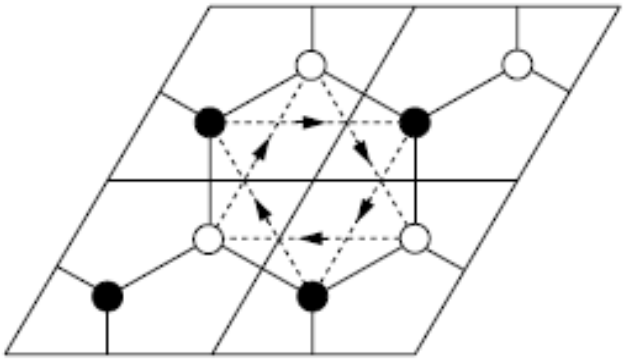
Phys. Rev. Lett.110, 116802 (2013)

- Heavy atoms + magnetic substrates
  - Isolated bands
  - Typically have Chern numbers
  - Find global gap
- First principles verification
  - Gaps at least 0.14 eV
- Future work
  - Theory / experimental collaboration
  - What surfaces are achievable in practice

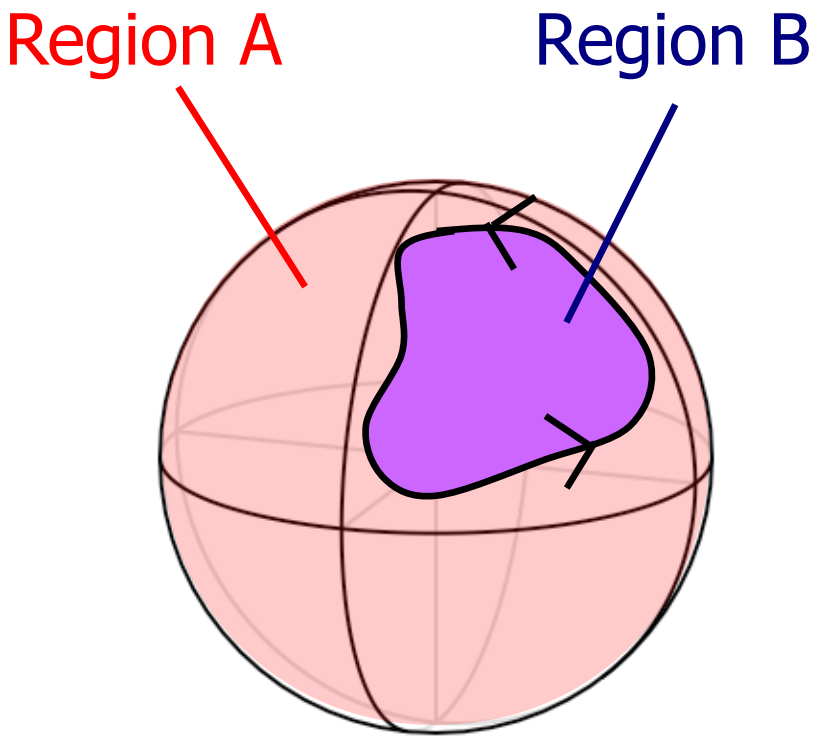
2D Chern Insulators

- Integer quantum Hall effect (1980s)
  - Strong magnetic field
  - Quantized conductance
- Quantum anomalous Hall (QAH)
  - No external field
  - Haldane model (1988)
  - QAH requirements:
    - Broken time reversal
    - Spin-orbit coupling

$$\sigma_{xy} = C\frac{e^2}{h}$$



Chern Theorem



Stokes applied to A:

$$\phi_A = \int_A \Omega_z(\mathbf{k})\, d^2k$$

Stokes applied to B:

$$\phi_B = \int_B \Omega_z(\mathbf{k})\, d^2k$$

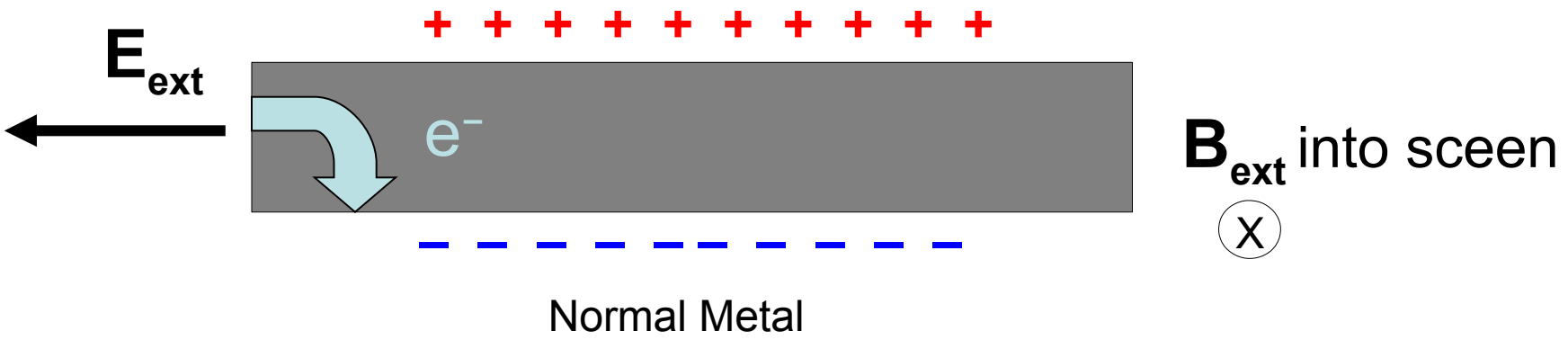
Uniqueness of ϕ mod 2π:

$$\phi_A = -\phi_B + 2\pi C$$

Chern theorem:

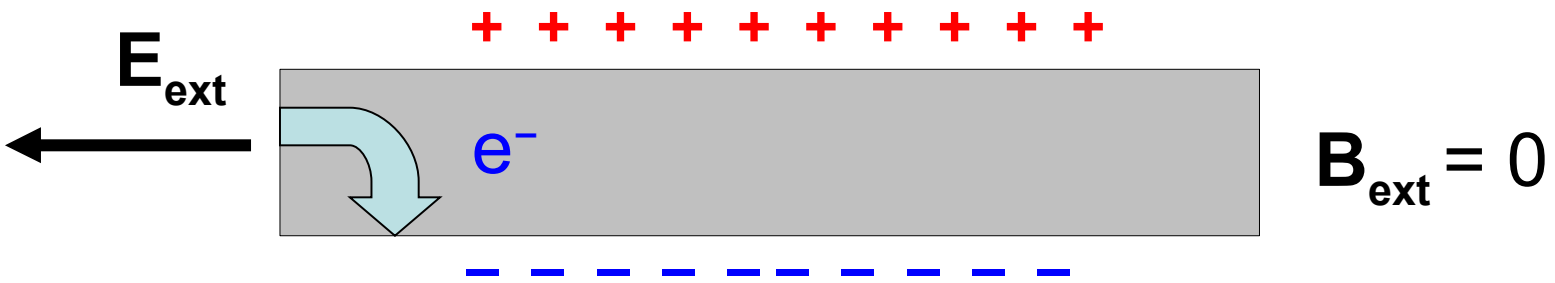
$$\oint_{BZ} \Omega_z(\mathbf{k})\, d^2k = 2\pi C$$

(Normal) Hall Effect



- Electrons feel Lorentz force
- Charge builds on top/bottom

Anomalous Hall Effect



- No external magnetic field.
- Ferromagnetic metal (breaks TR)
- Intrinsic contribution (single band):

Berry Curvature

$$\sigma_{xy} = -\frac{e^2}{2\pi h}\int d\mathbf{k} f(\epsilon_{\mathbf{k}})\Omega_z(\mathbf{k})$$

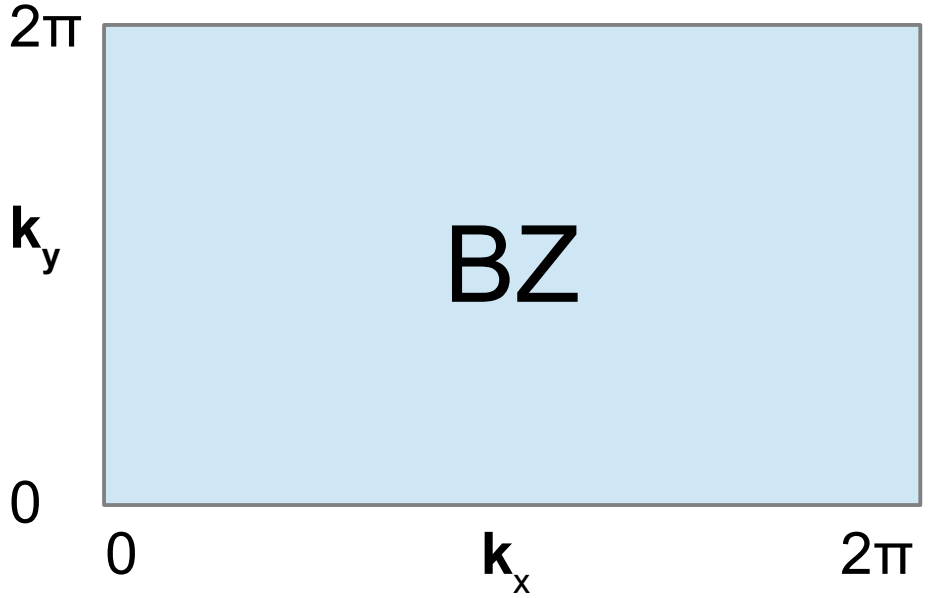
Karplus and Luttinger; Sundaram and Niu

Computing Chern

- Continuous-**k:** $$C = \frac{1}{2\pi}\int_{BZ} d\mathbf{k}\Omega(\mathbf{k}) = \frac{1}{2\pi}\oint_{BZ} d\mathbf{k}\cdot\mathbf{A}(\mathbf{k})$$

where $$\mathbf{A}(\mathbf{k}) = i\langle u_{\mathbf{k}}|\partial_{\mathbf{k}}|u_{\mathbf{k}}\rangle$$ Berry Connection

$$\Omega(\mathbf{k}) = \nabla_{\mathbf{k}} \times \mathbf{A}(\mathbf{k})$$ Berry Curvature



Computing Chern

- Continuous-**k:** $$C = \frac{1}{2\pi}\int_{\mathrm{BZ}} d\mathbf{k}\Omega(\mathbf{k}) = \frac{1}{2\pi}\oint_{\mathrm{BZ}} d\mathbf{k}\cdot\mathbf{A}(\mathbf{k})$$
- Discrete-**k:**

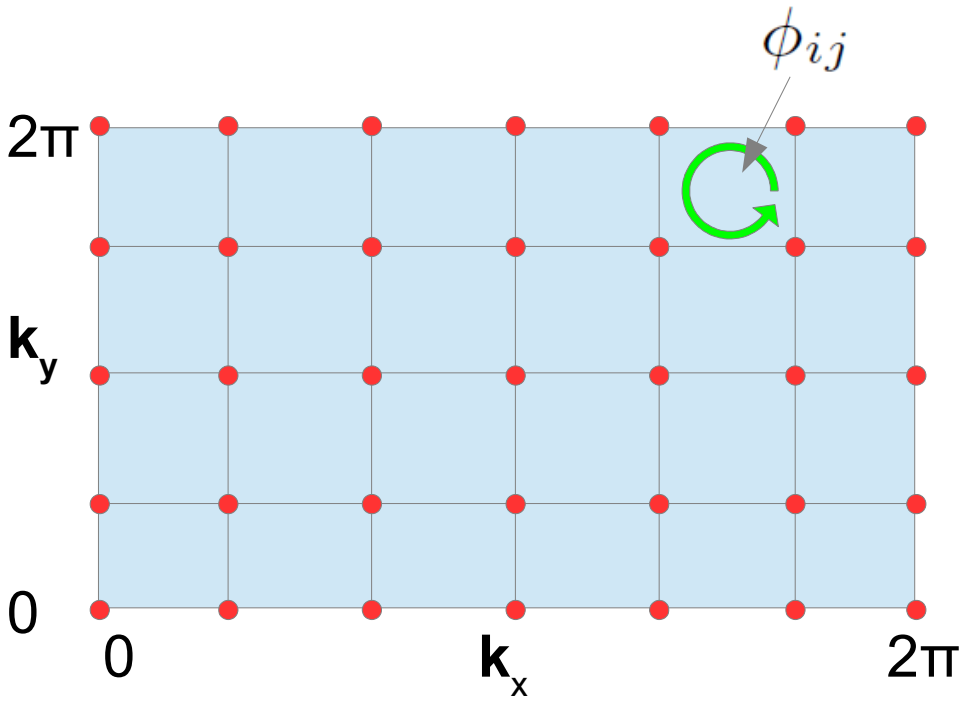
$$\mathbf{A}(\mathbf{k}) = i\langle u_{\mathbf{k}}|\partial_{\mathbf{k}}|u_{\mathbf{k}}\rangle$$

  - Need Berry's Phase around each discrete loop.

$$\begin{aligned}\phi_{ij} &= \mathrm{Im}\oint_{loop_{ij}} dk\langle u_{nk} \mid \partial_k \mid u_{nk}\rangle \\ &= \mathrm{Im}\ln\prod_{l=loop_{ij}} \langle u_{n,l} \mid u_{n,l+1}\rangle\end{aligned}$$

$$C = \frac{1}{2\pi}\sum_{ij}\phi_{ij}$$

- Works with random phase eigenvectors



Use Wannier Functions

$$|w_{n\mathbf{R}}\rangle = \frac{\Omega}{(2\pi)^3} \int d\mathbf{k}\, \mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}}\, |\psi_{n\mathbf{k}}\rangle$$

- Gives real-space tight-binding Hamiltonian
  - Cheap k-space interpolation
  - Easy to calculate overlaps

- wannier90 – maximally-localized Wannier functions
  - Disentangle conduction band states

- Also calculate anomalous hall conductivity (AHC) for metals. (Wang *et. al.* Phys. Rev. B 74, 195118 (2006))