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2D magnetic topological materials: insights from DFT

28. November 2022 | G. Bihlmayer, Peter Grünberg Institut and
Institute for Advanced Simulation
Forschungszentrum Jülich, Germany

Thanks to...

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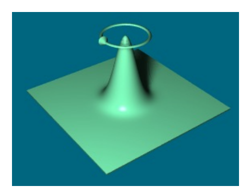
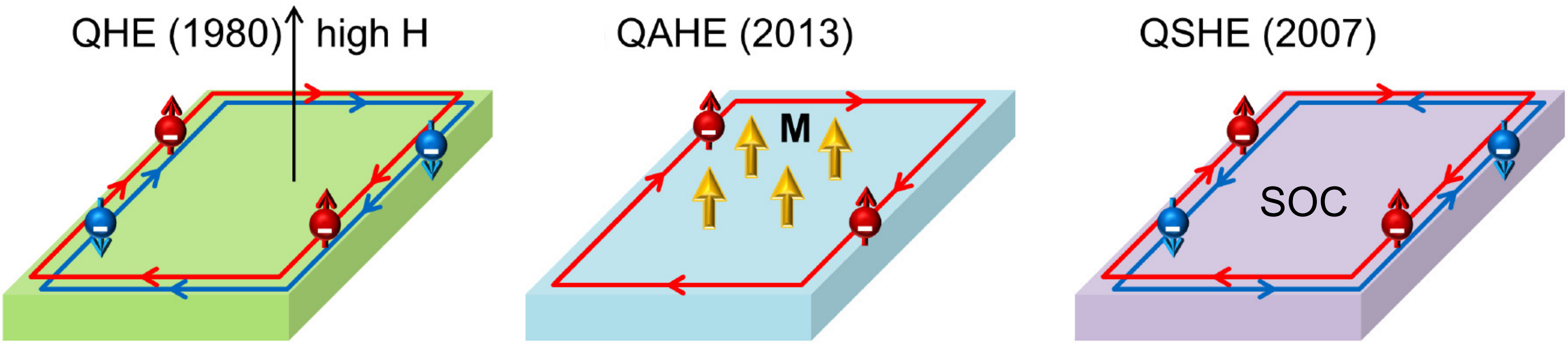
- Shuichi Murakami, Masaki Wada,
  Tokyo Institute of Technology, Japan



SPP1666 (DFG) **VITI (HGF)** **VH-NG-513**

Quantum Hall effects (QHE) in 2D materials

with quantum anomalous Hall effect (QAHE) and quantum spin Hall effect (QSHE)



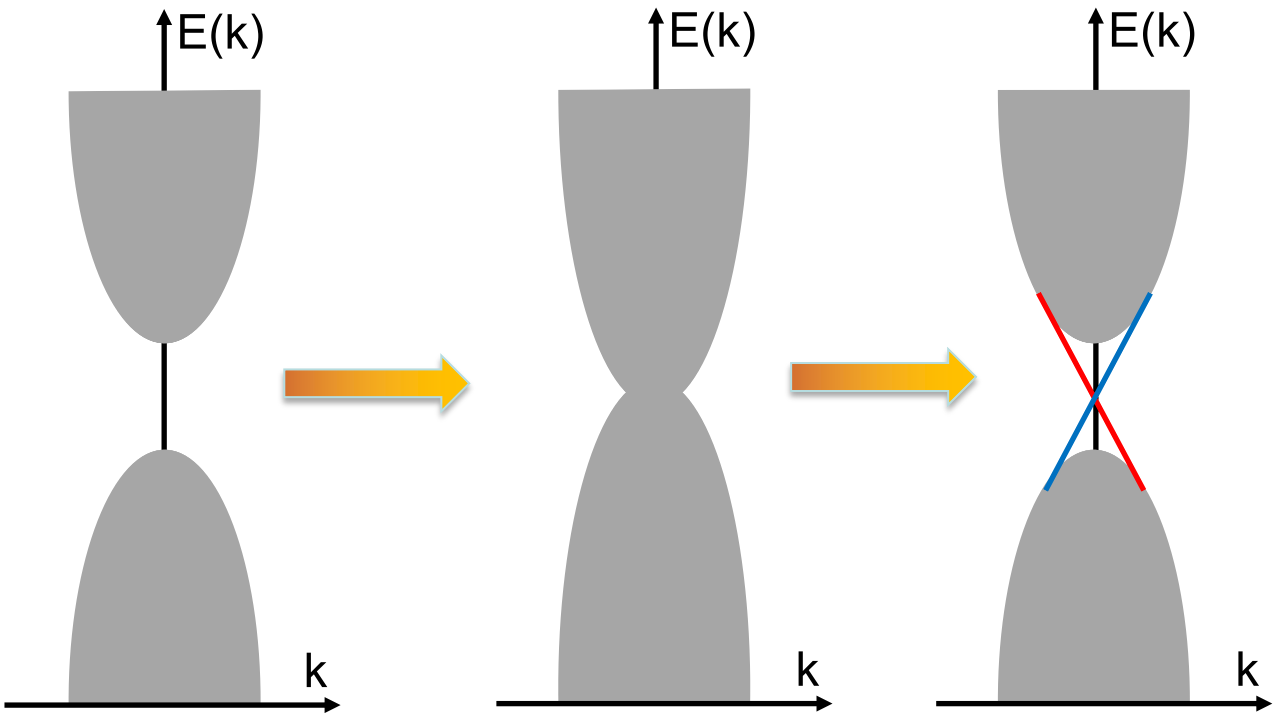
C.-Z. Chang & M. Li, J. Phys.: Condens. Matter **28** 123002 (2016)

common features:

- Two-dimensional insulating “bulk”
- Magnetic fields (external, internal, SOC)
- Dissipationless edge currents (charge or spin)

Topological phase transitions

normal insulator (NI) to topological insulator (TI) transition:

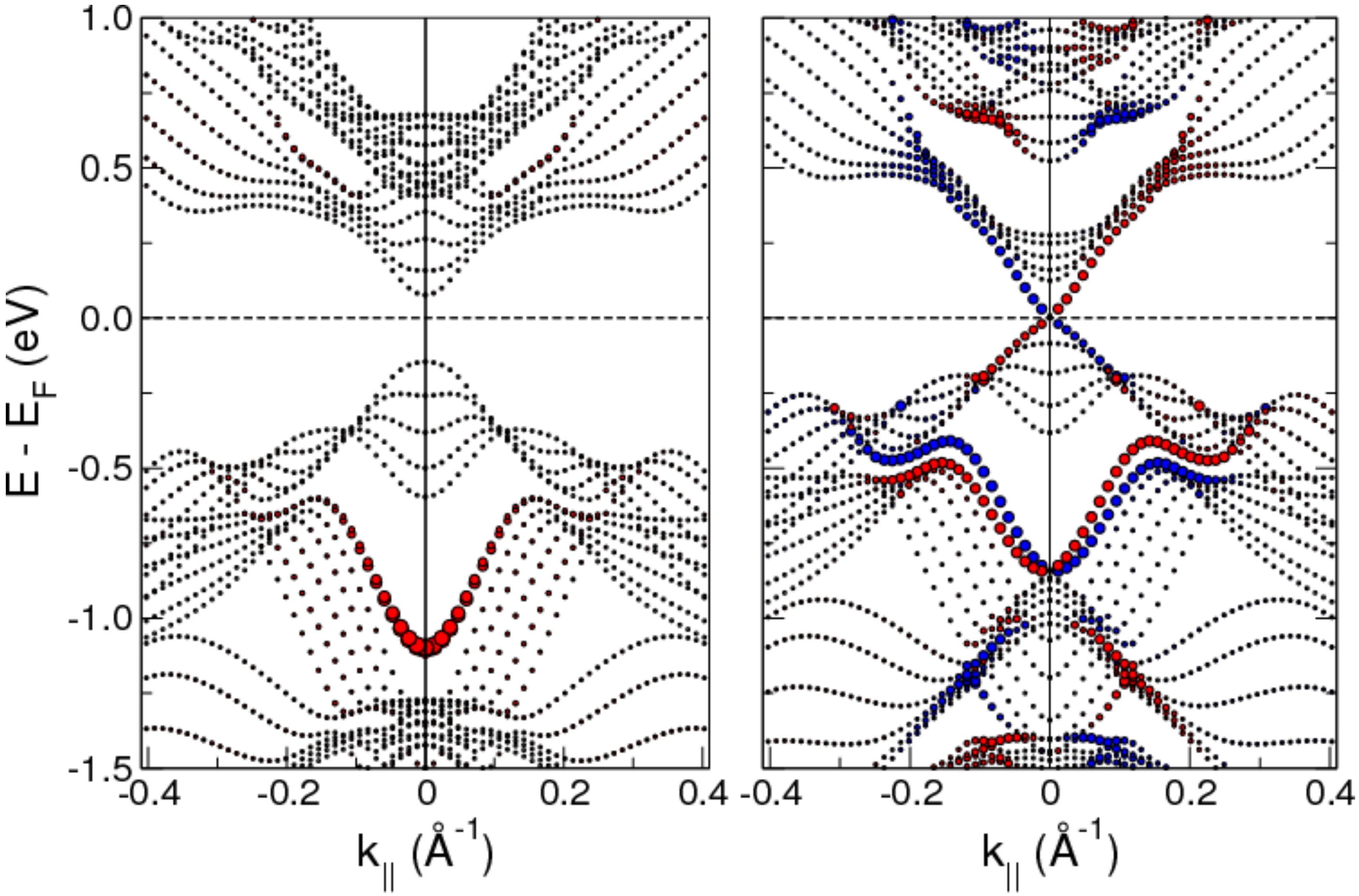


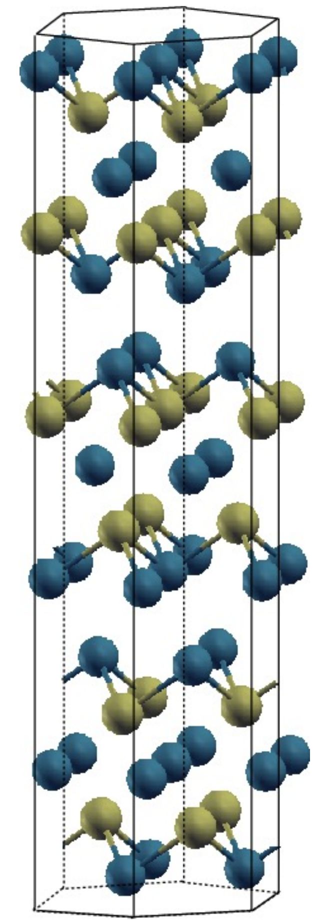
NI (C_s=0) metal TI (C_s=1)

driving parameter: spin-orbit coupling strength, strain, …

band-inversion in Sb_2Te_3

surface without SOC → and with SOC:





- red/blue: spin-polarization at surface, black: "bulk"

cf. Sánchez-Barriga et al., Phys. Rev. B **98** 235110 (2018)

Berry curvature & Chern numbers

$$\Omega_z(\mathbf{k}) = 2\mathrm{Im} \sum_{n,\ m \neq n} \frac{\langle m\mathbf{k}|\nabla_x|n\mathbf{k}\rangle\langle n\mathbf{k}|\nabla_y|m\mathbf{k}\rangle}{(\varepsilon_n(\mathbf{k}) - \varepsilon_m(\mathbf{k}))^2}$$

calculate with DFT:

www.flapw.de

Fleur

WANNIER90

Integer Chern number and Quantum Hall effect (Thouless et al. 1982)

$$\boxed{\mathcal{C} = 1/2\pi \int \Omega_z(\mathbf{k})d\mathbf{k}}$$

QSHE: consider separately spin-up and spin-down:

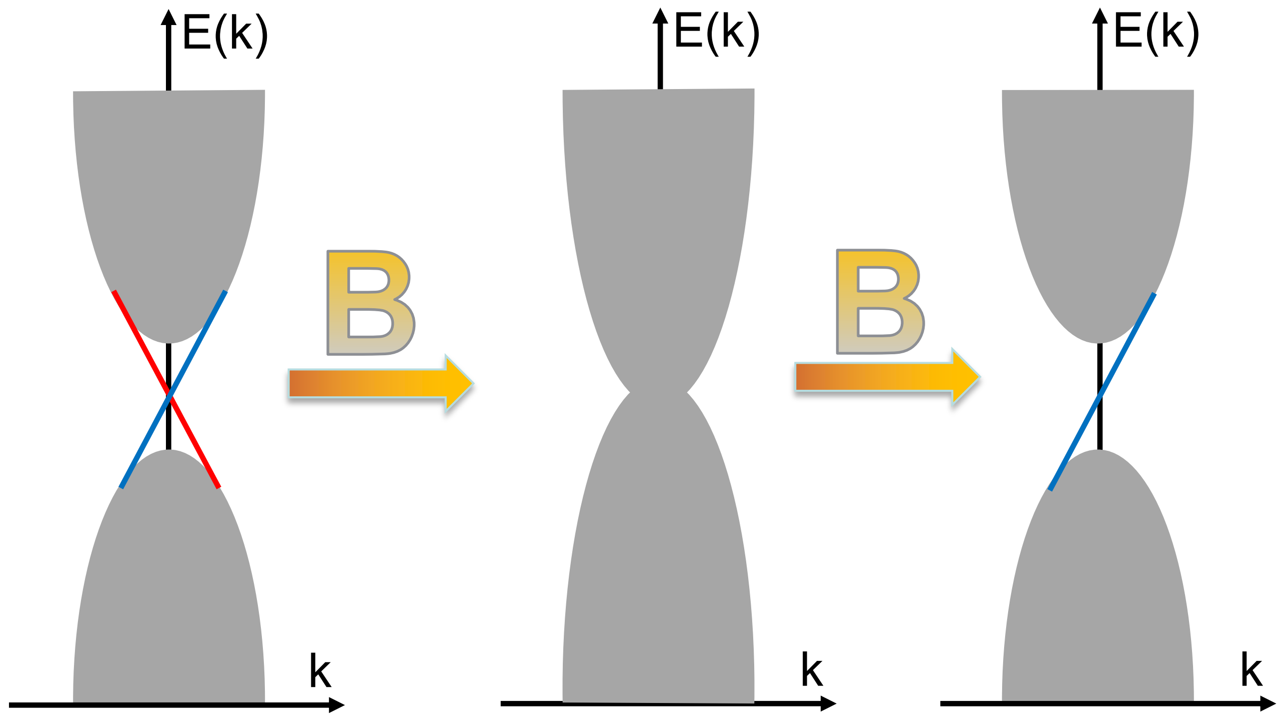
$$\mathcal{C}^{\uparrow} = 1/2\pi \int \Omega_z^{\uparrow}(\mathbf{k})d\mathbf{k}$$
$$\mathcal{C}^{\downarrow} = 1/2\pi \int \Omega_z^{\downarrow}(\mathbf{k})d\mathbf{k}$$

$$\boxed{\mathcal{C}_s \sim \mathcal{C}^{\uparrow} - \mathcal{C}^{\downarrow} \quad \sim \mathbb{Z}_2 \quad \sim \sigma_H^s}$$

Spin Chern Number

Topological phase transitions (2)

topological insulator (TI) in magnetic field (B):

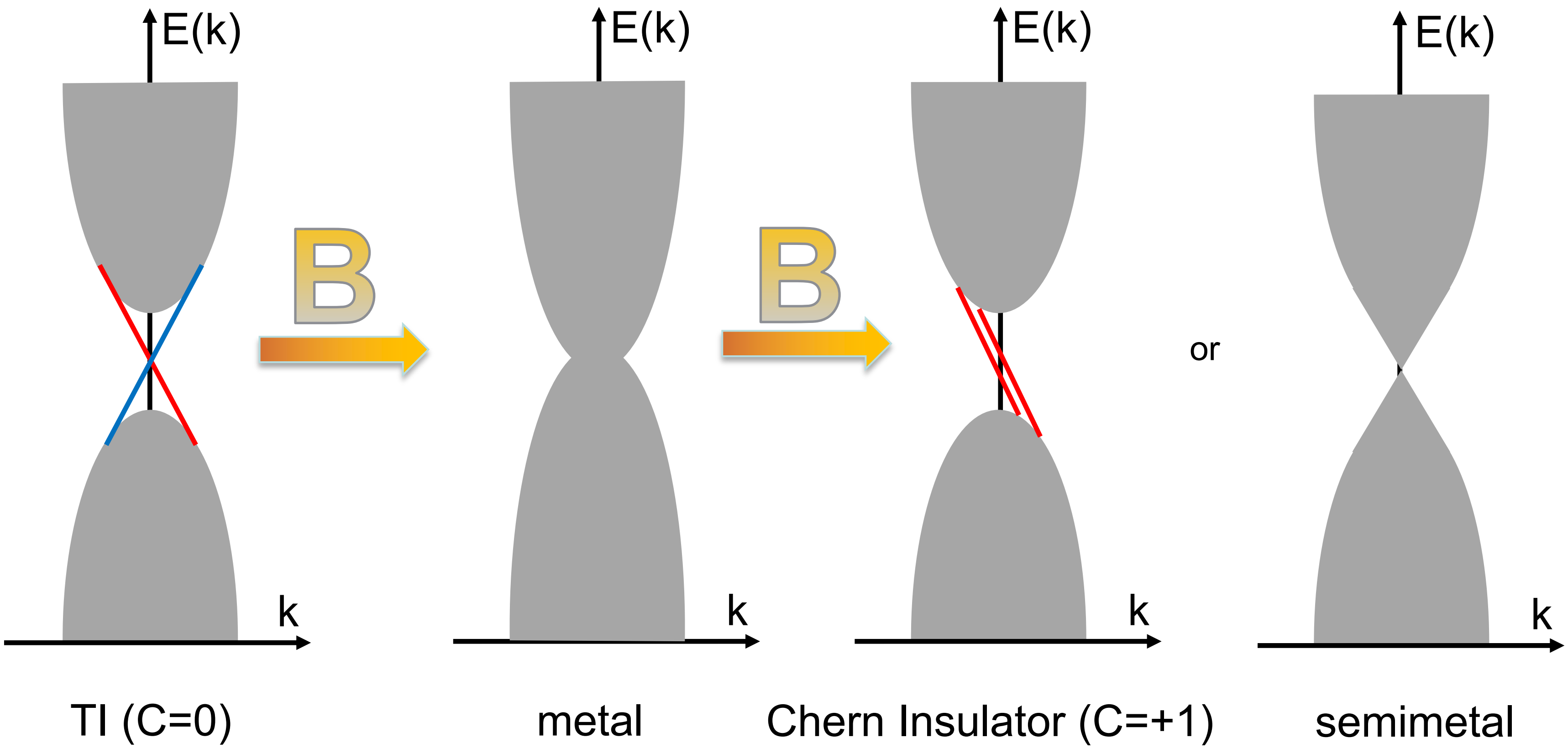


TI (C=0) metal Chern Insulator (C=+1)

driving parameter: magnetic field (strength, direction)

Topological phase transitions (2)

topological insulator (TI) in magnetic field (B):

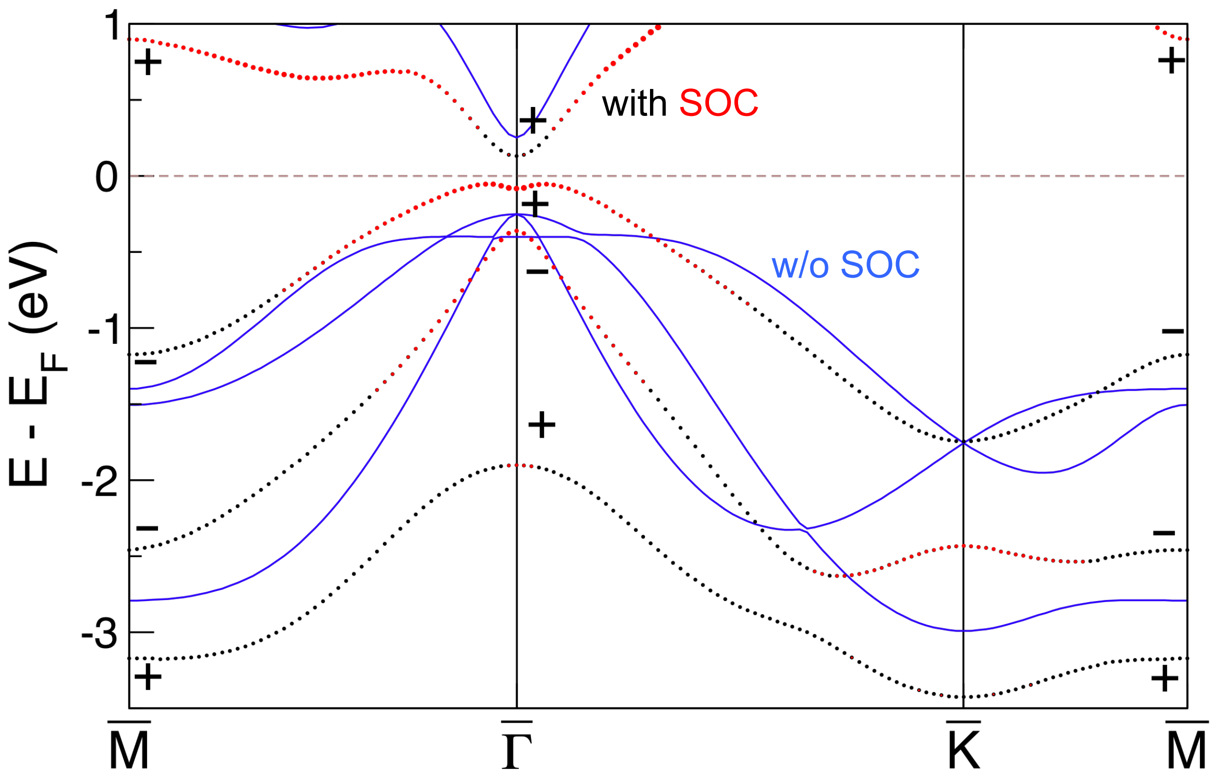


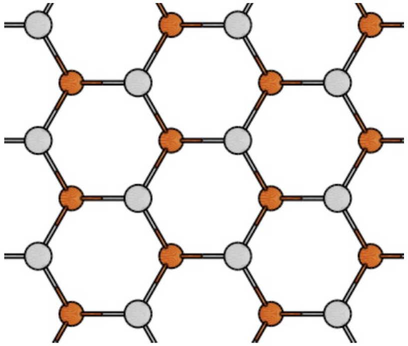
driving parameter: magnetic field (strength, direction)

2D TIs in magnetic fields:

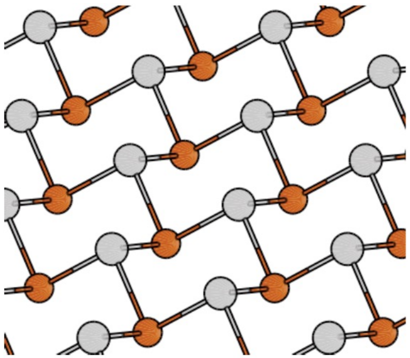
QSH – QAH transition

Buckled honeycomb lattice: Bi(111) bilayer





Top view

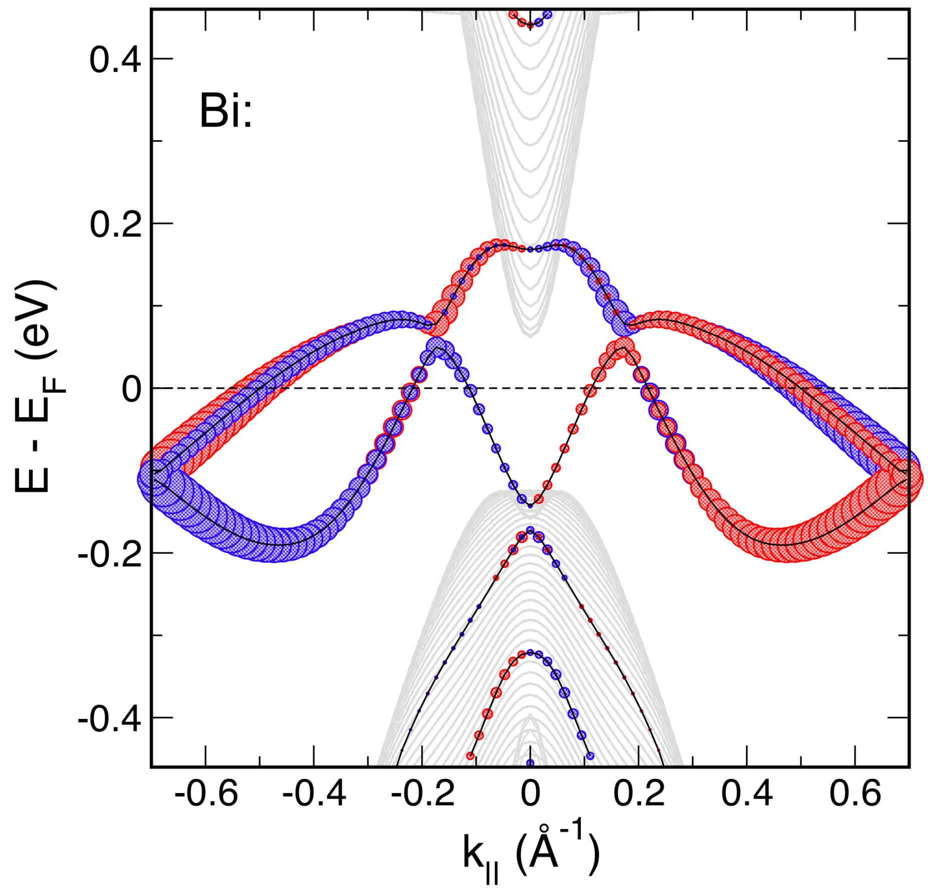


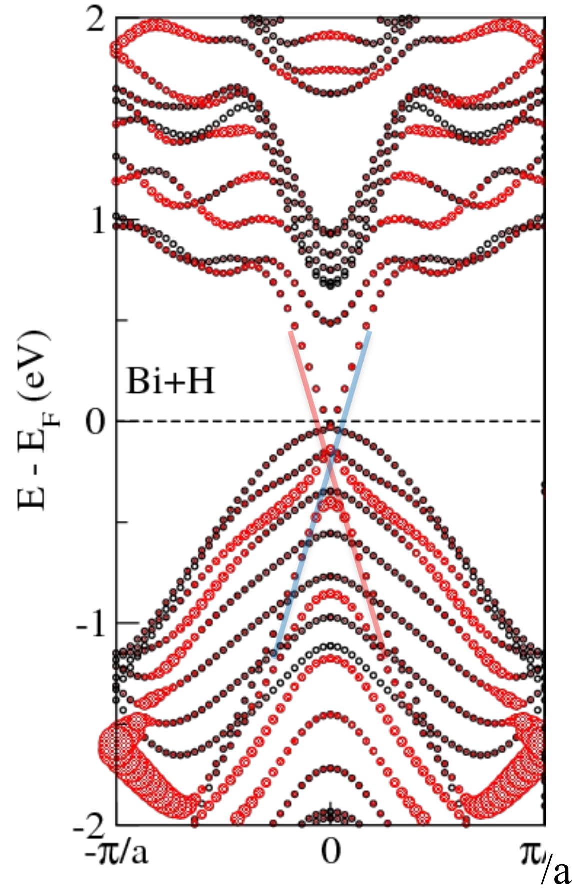
Oblique view

Band structure of Bi(111) bilayer: $\mathbb{Z}_2$=1; 2D TI

S. Murakami, Phys. Rev. Lett. **97**, 236805 (2006)
Yu. M. Koroteev et al., Phys. Rev. B **77**, 045428 (2008)
M. Wada et al., Phys. Rev. B **83**, 121310 (2011)

Edge states of a Bi-bilayer nanoribbon:



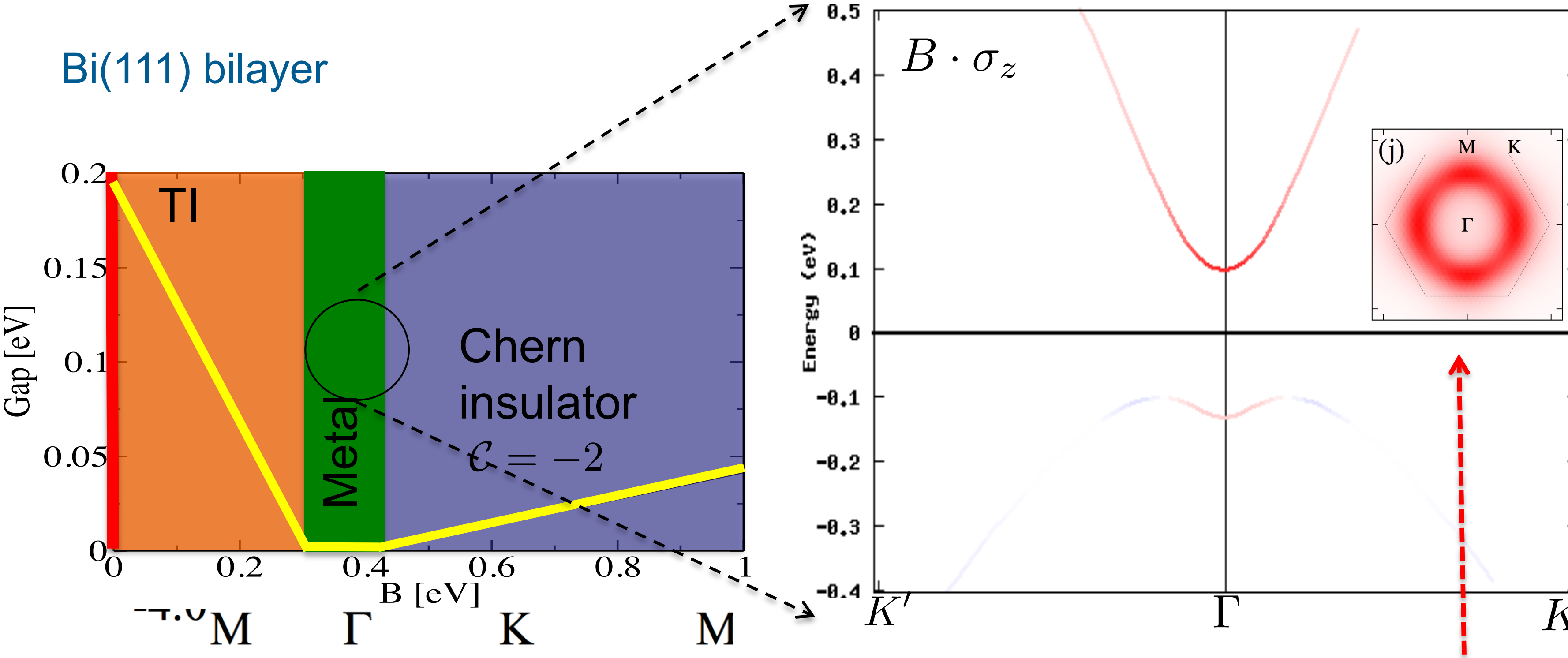


Zig-zag edge: coexistence of dangling bond + "topological edge state"

cf. Jeong et al., Phys. Rev. B **98** 075402 (2018)

2D TI in magnetic (exchange) field

Bi(111) bilayer



H. Zhang et al., Phys. Rev. Lett. **106**, 117202 (2011) **hot loop**

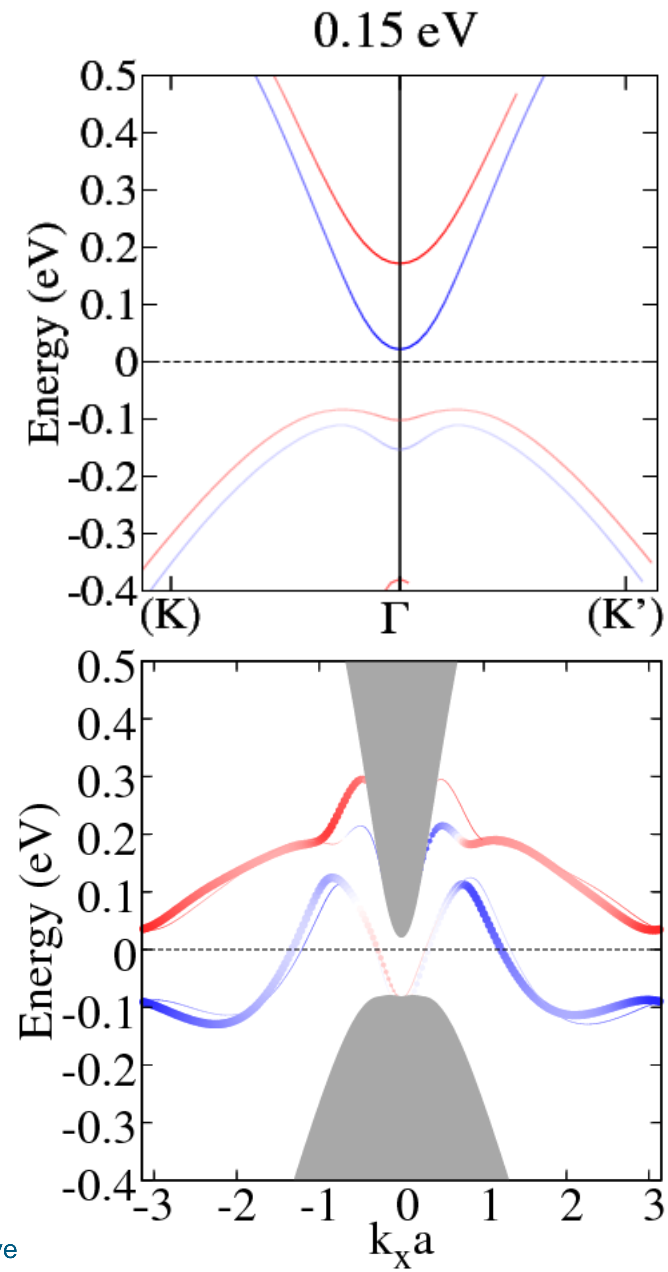
Exchange of **spin Chern number** along the **hot loops**

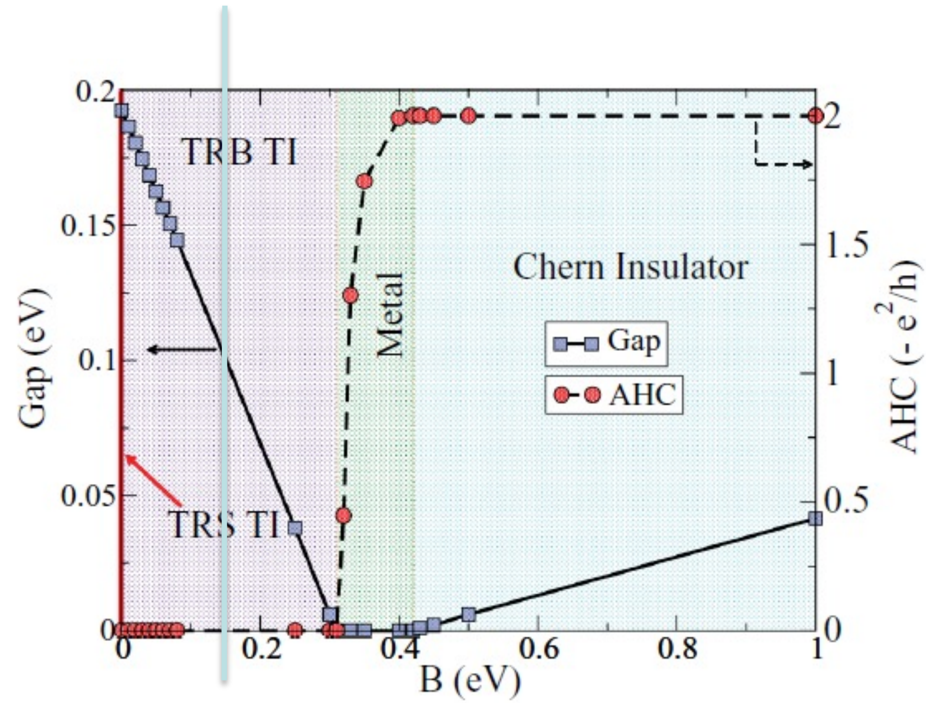
spin-to-charge "conversion"

$$\mathbb{Z}_2 \sim \mathcal{C}^{\uparrow} - \mathcal{C}^{\downarrow} \longleftrightarrow \mathcal{C} \sim \mathcal{C}^{\uparrow} + \mathcal{C}^{\downarrow}$$

$$\boxed{\mathcal{C}^{\uparrow}_{unocc} \longleftrightarrow \mathcal{C}^{\downarrow}_{occ}}$$

Evolution of Bi(111) film bandstructure

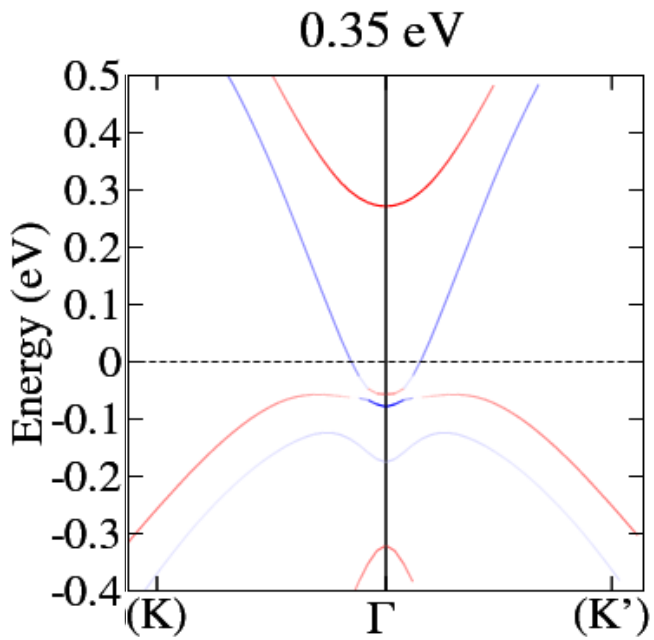


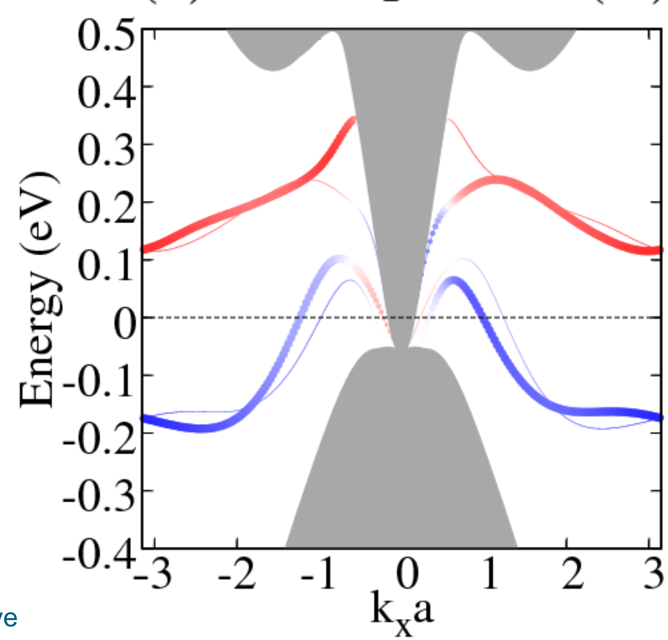


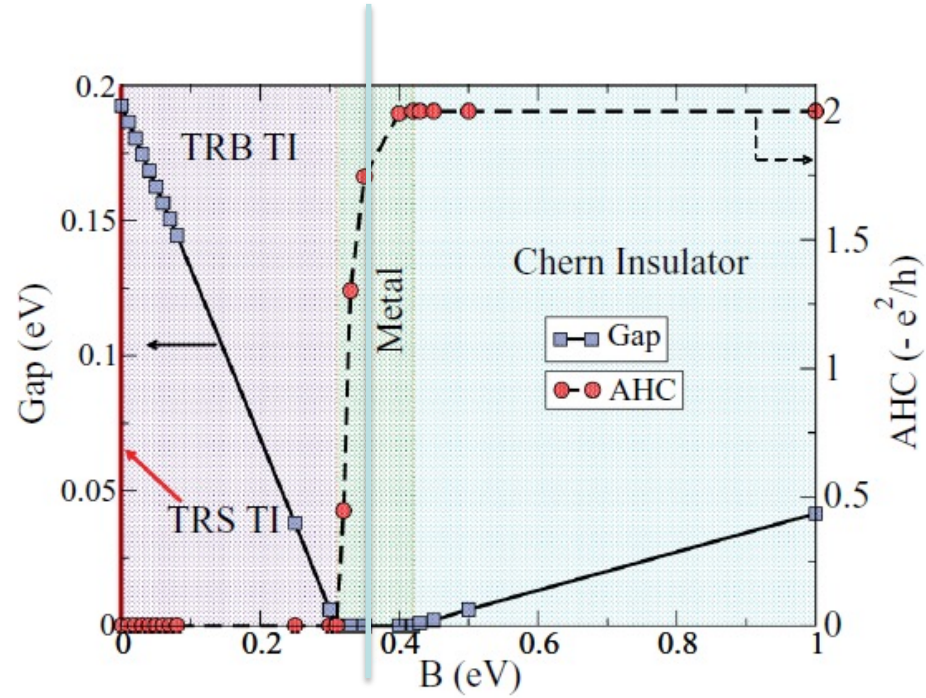
H. Zhang et al., Phys. Rev. B **86**, 035104 (2012)

➔ huge magnetic fields required
(0.2 eV = 3454 T/μ_B)
… use exchange fields

Evolution of Bi(111) film bandstructure







H. Zhang et al., Phys. Rev. B **86**, 035104 (2012)

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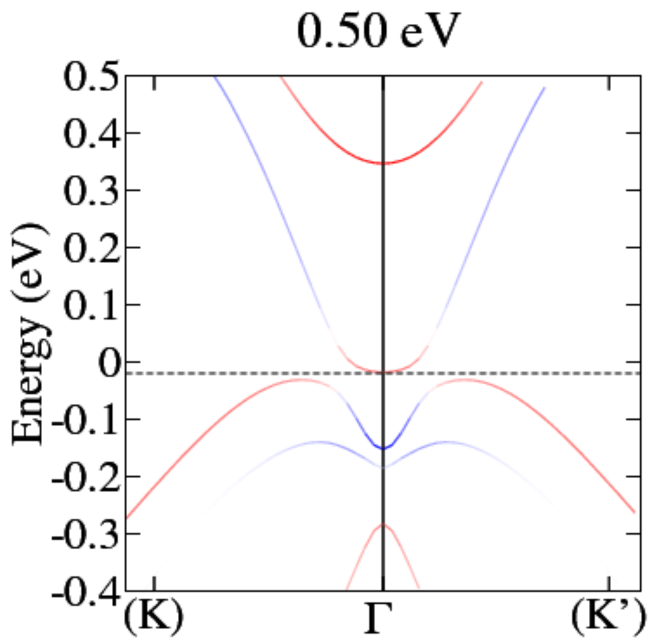

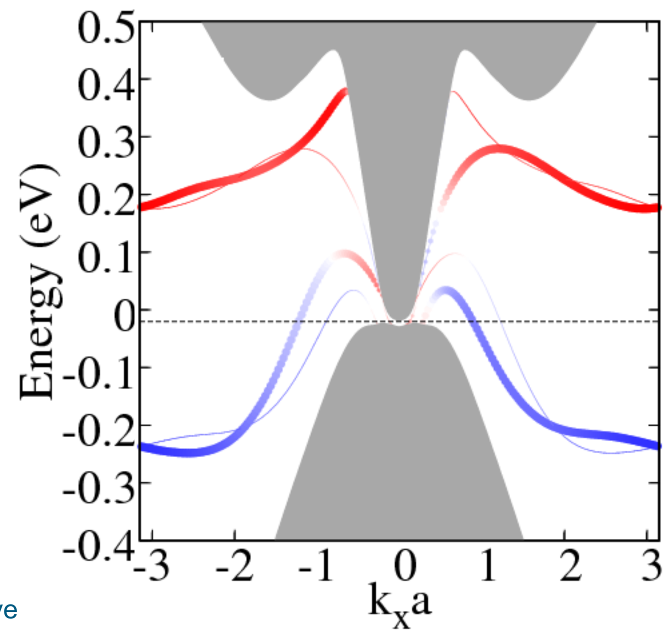

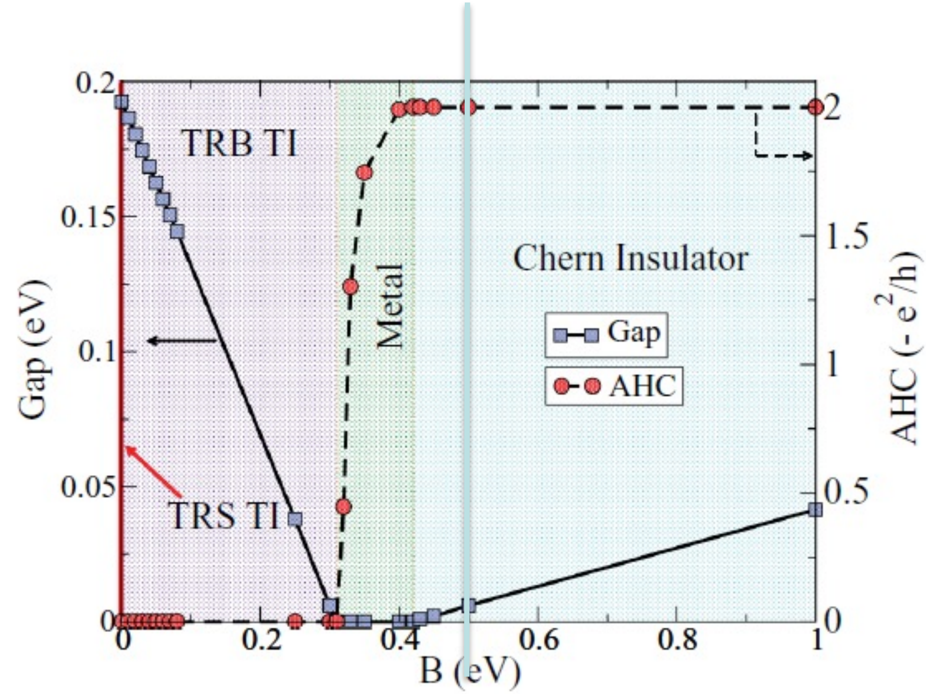

H. Zhang et al., Phys. Rev. B **86**, 035104 (2012)

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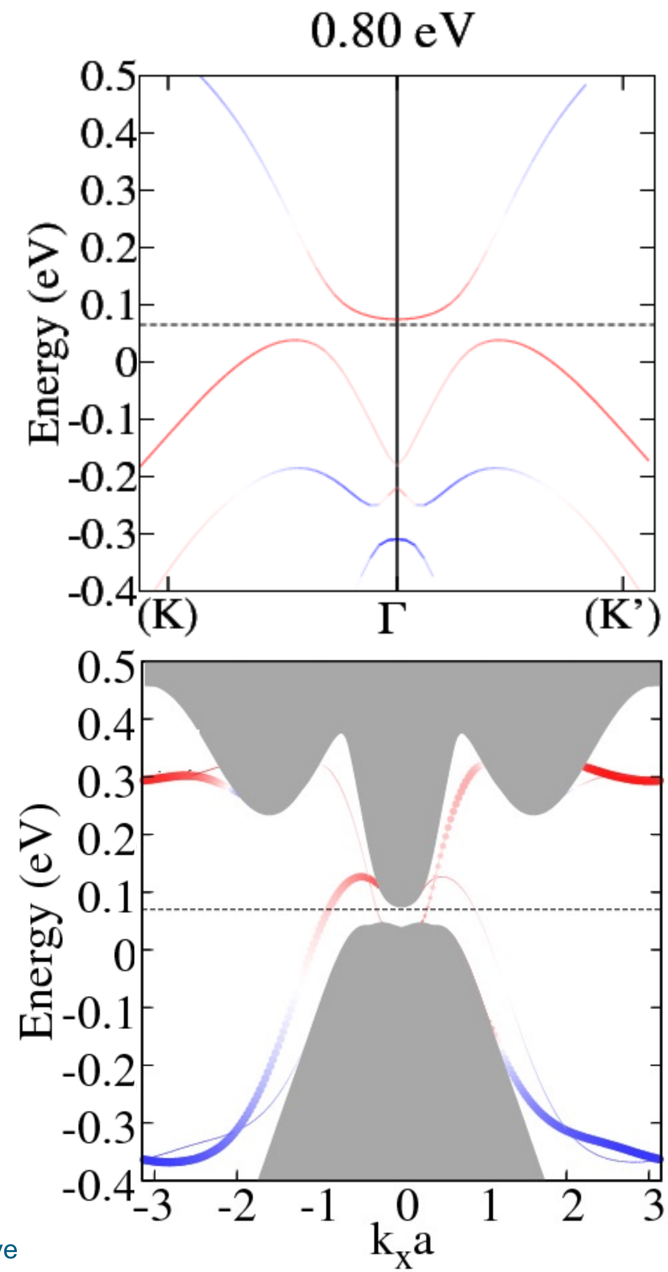
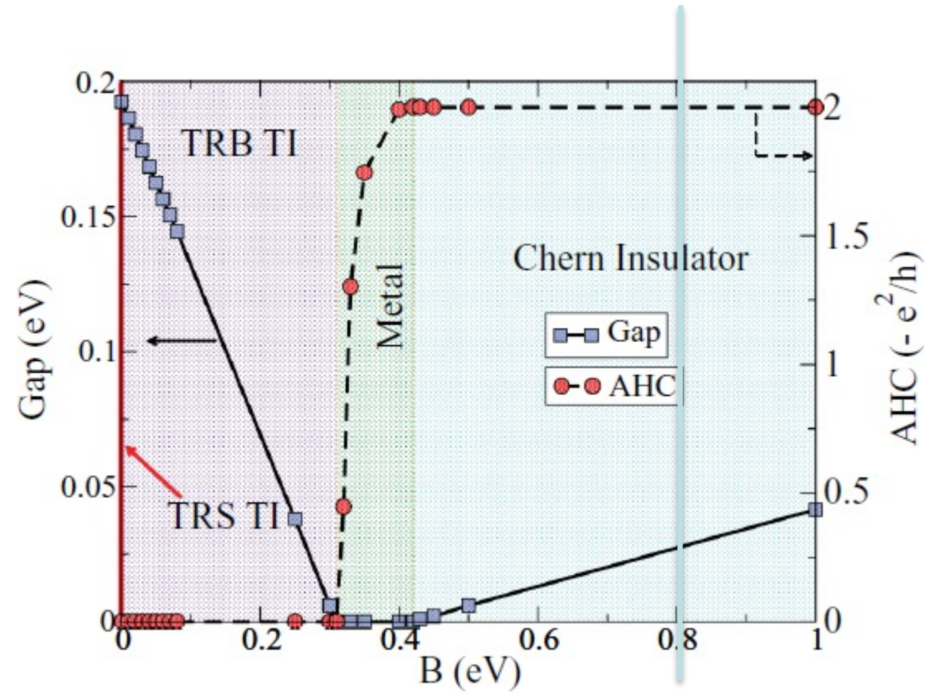
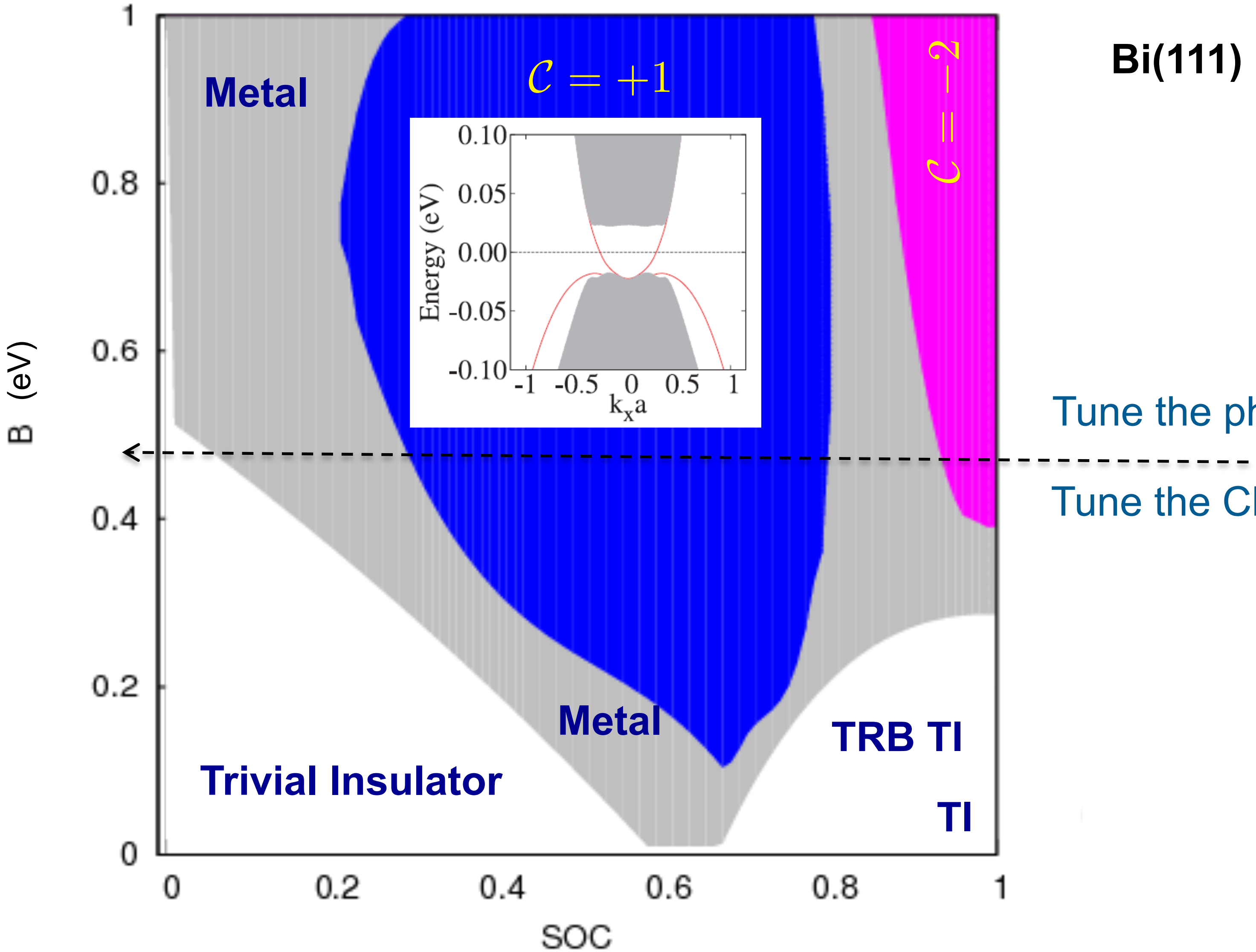




H. Zhang et al., Phys. Rev. B **86**, 035104 (2012)

➔ huge magnetic fields required
(0.2 eV = 3454 T/μ_B)
… use exchange fields

Phase Diagram with varying SOC stength

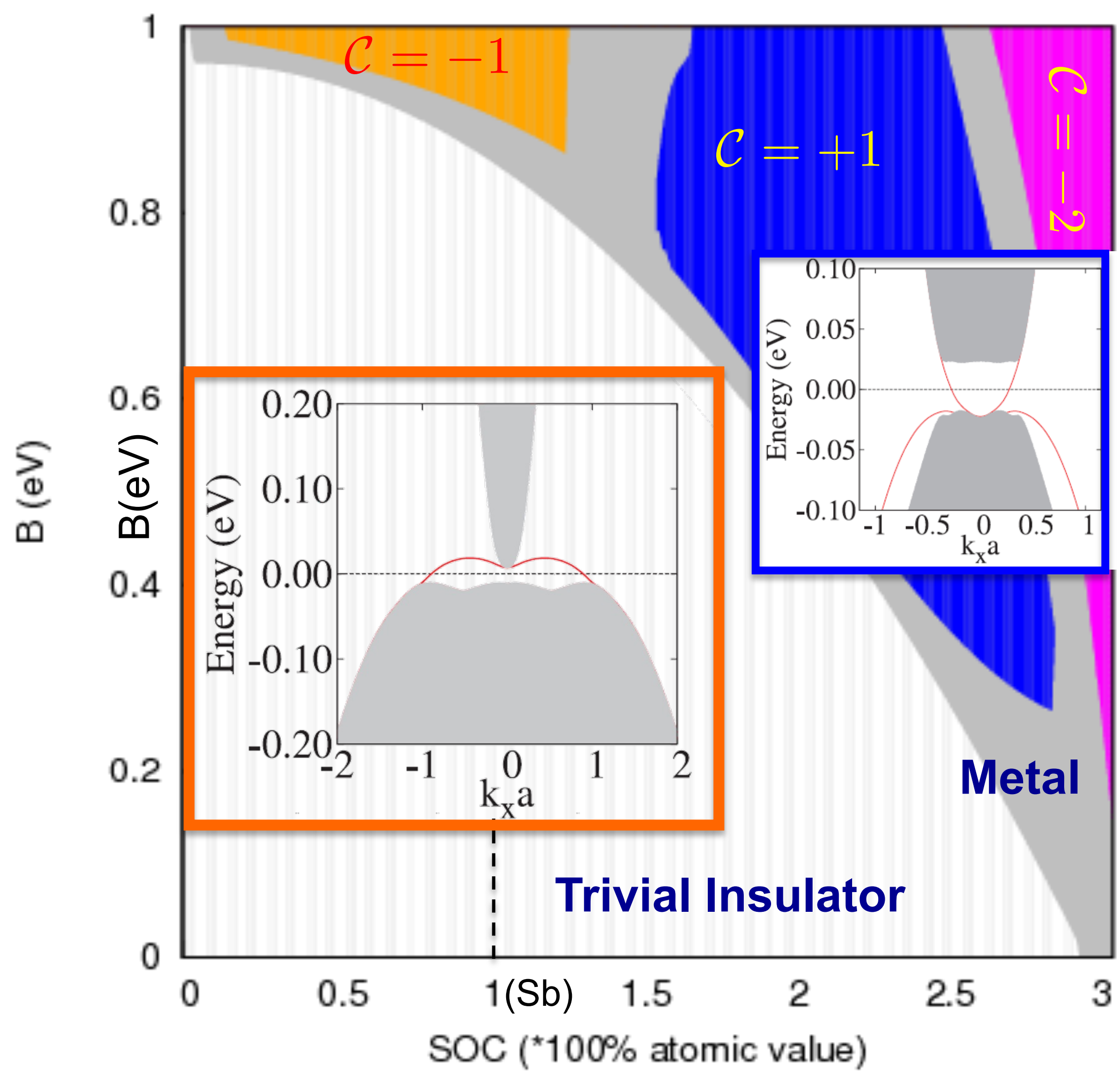


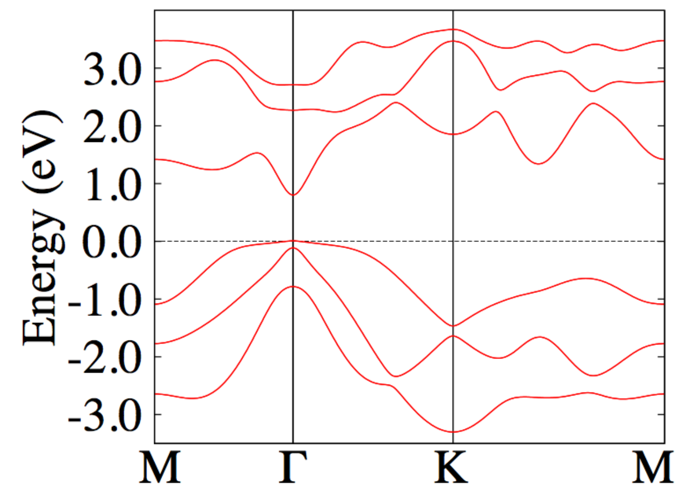
Bi(111) bilayer

Tune the phase

Tune the Chern number

Scaling SOC in Sb(111) BL





Sb(111) bilayer, B = 0

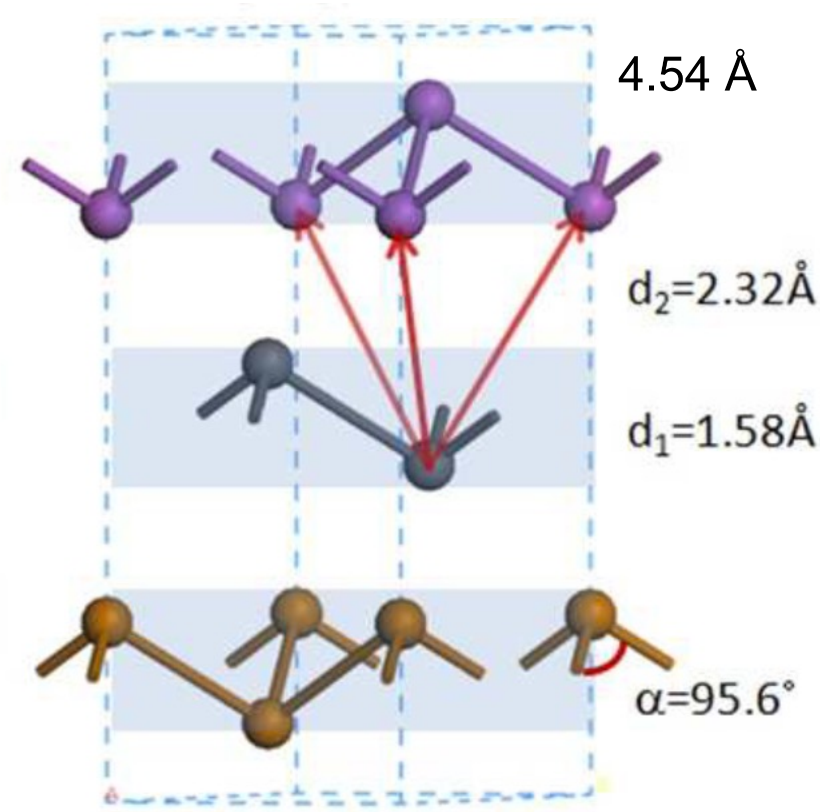
C = -1: edge state propagates oppositely to C = +1 (cf. Bi(111) BL)

H. Zhang, F. Freimuth, G. Bihlmayer, M. Lezaic, S. Blügel, and Y. Mokrosuov, Phys. Rev. B **87**, 205132 (2013)

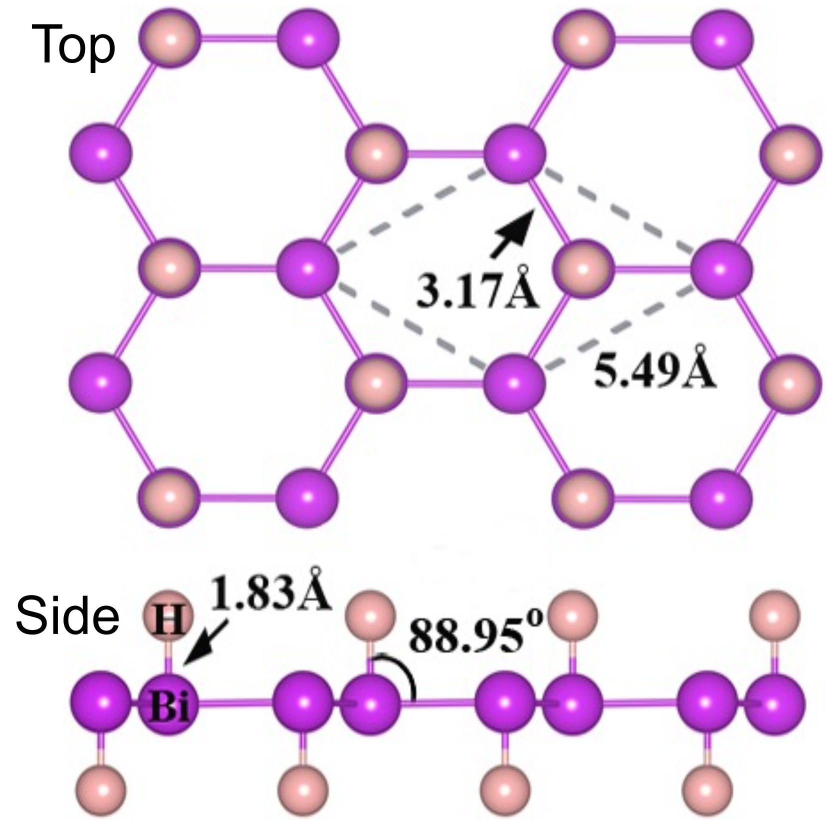
Introducing exchange splittings by chemical modification

Crystal structure of H-Bi(111)

single-crystal Bi

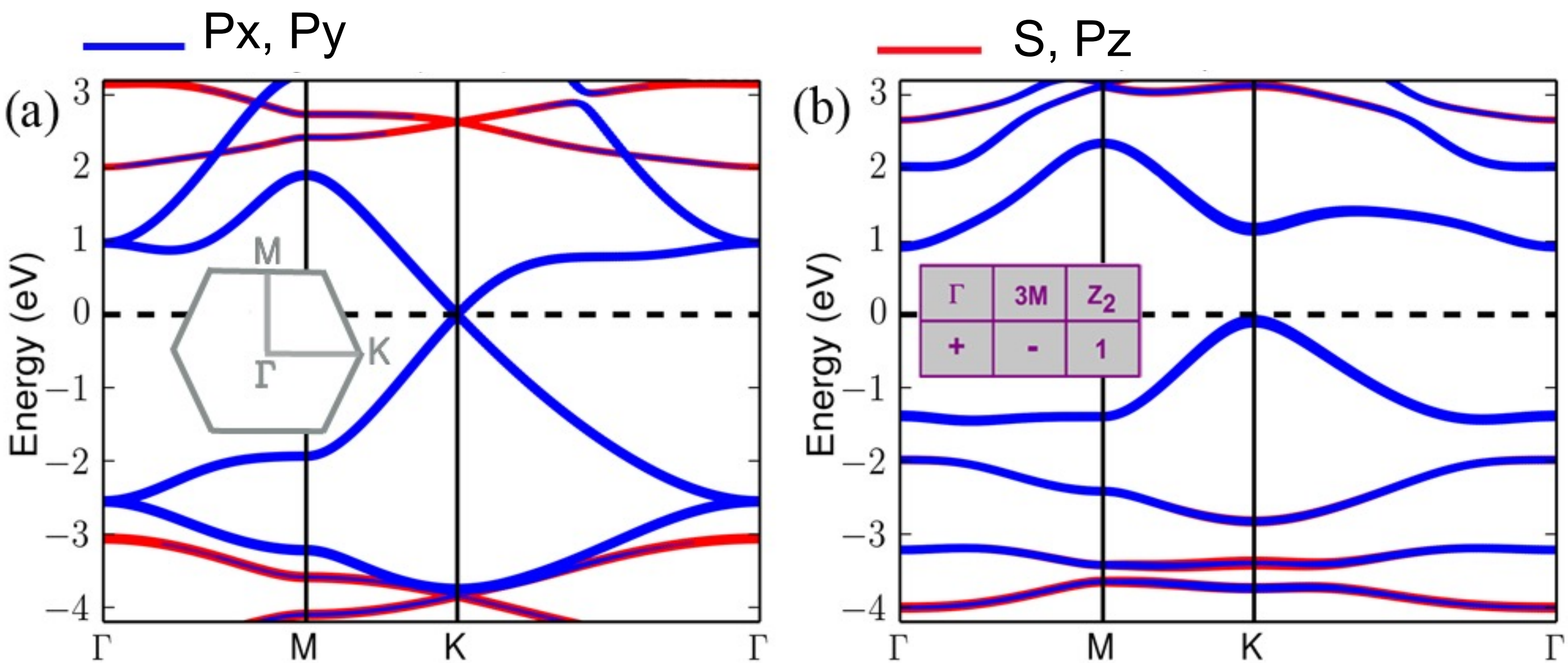


H-decorated Bi(111)



from :
C. Niu, et al., Phys. Rev. B **91**, 041303(R) (2015)

Band structure for H-Bi(111)



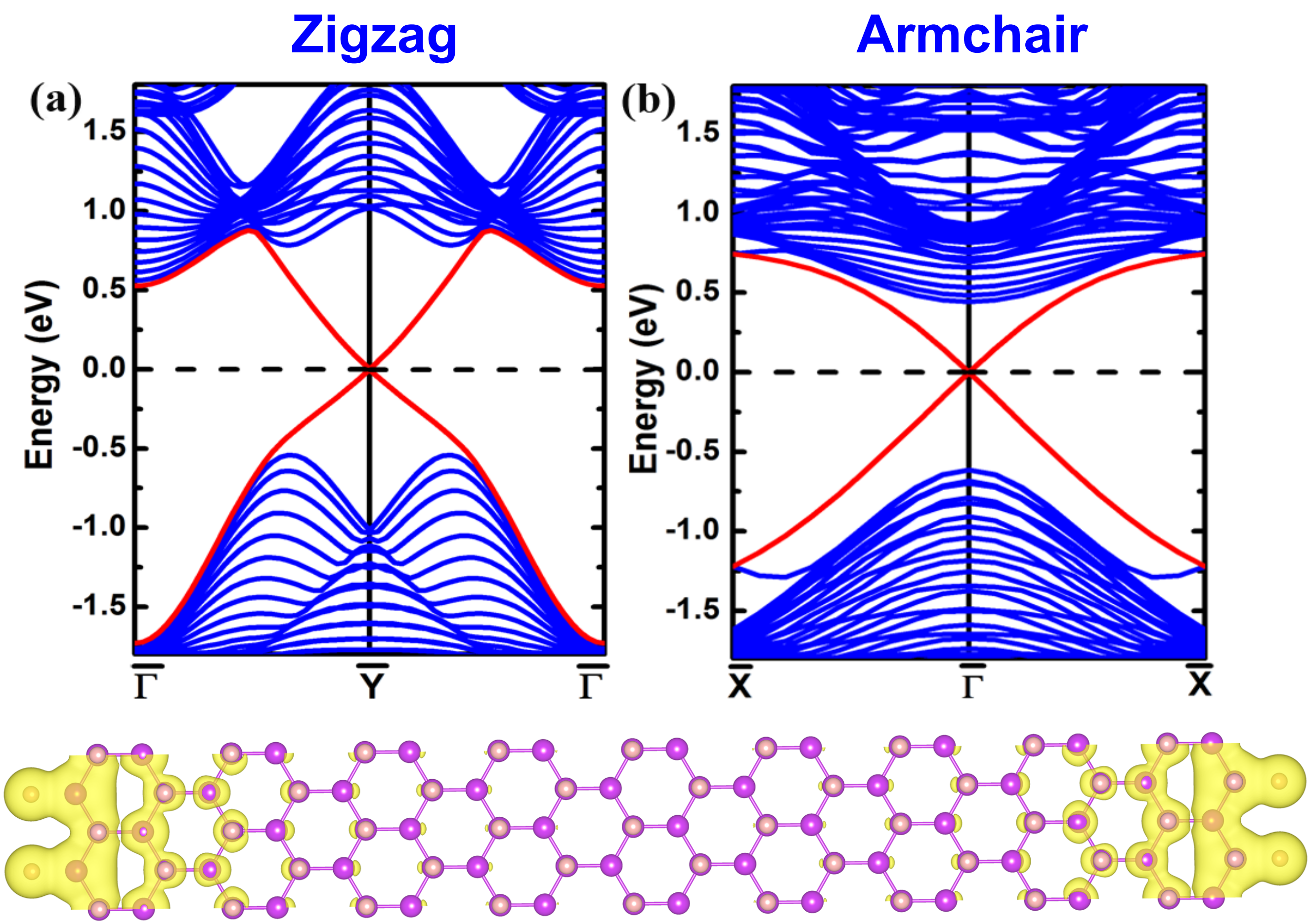
without SOC

gapless, graphene-like band crossing at K (but p_x,p_y)

with SOC

ν=1; indirect band gap of 1.01 eV

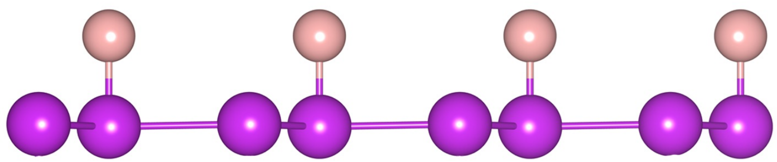
Edge states of H-Bi(111)

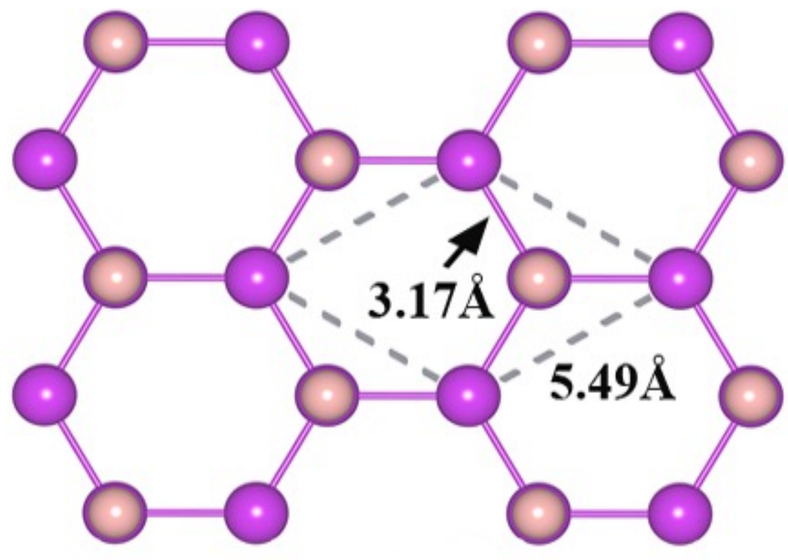


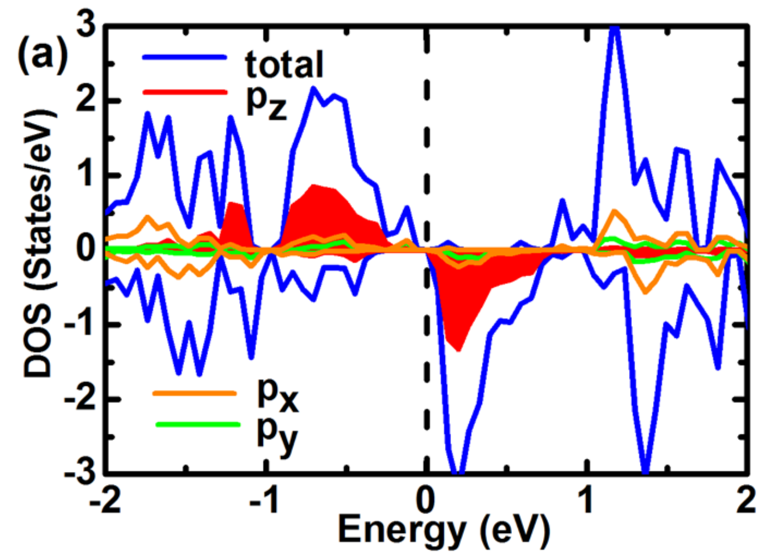
Edge states demonstrate 2D TI character

Ferromagnetism in half H-decorated Bi(111)

Crystal structure of half H-decorated Bi(111)





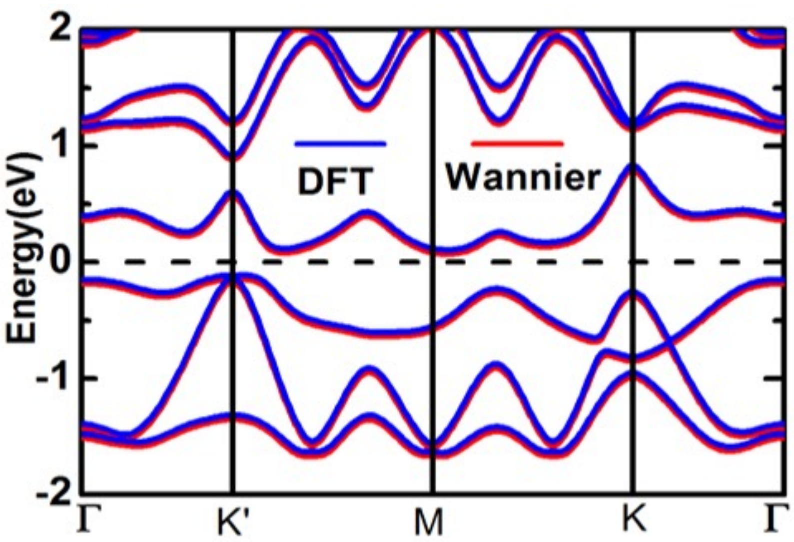


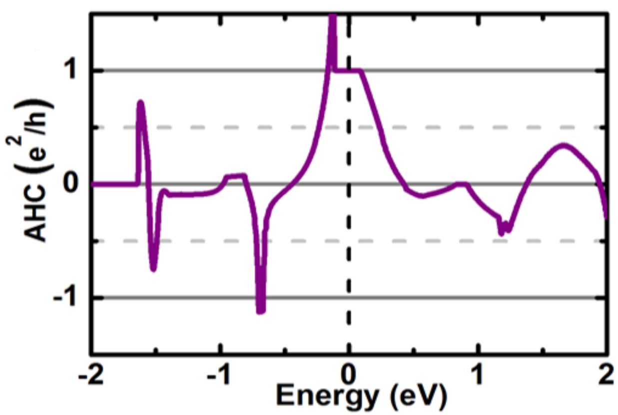
- Spin-polarization is mainly carried by p_z states of the unhydrogenated Bi
- Magnetic moment is $1.0\mu_B$ with out-of-plane spin orientation
- $\Delta E = E_{AFM} - E_{FM} = 21.79$ meV

C. Niu et al., Phys. Rev. B **91**, 041303(R) (2015)
see also: C.-C. Liu et al., Phys. Rev. B **91,** 165430 (2015)

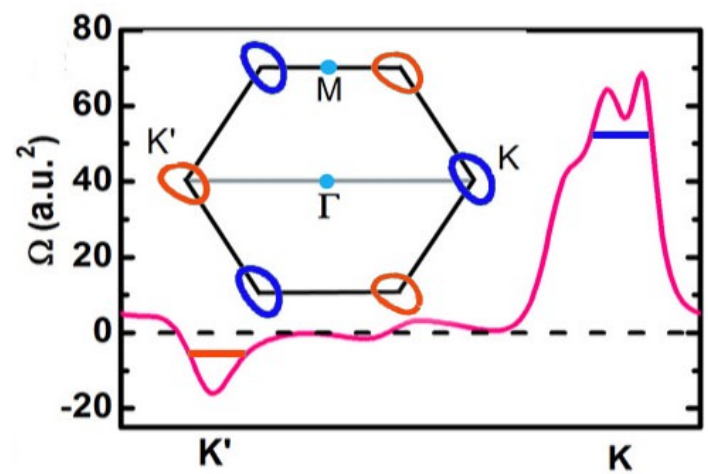
Valley-polarized QAH states

Band structures





Anomalous Hall conductivity



Berry curvature distribution

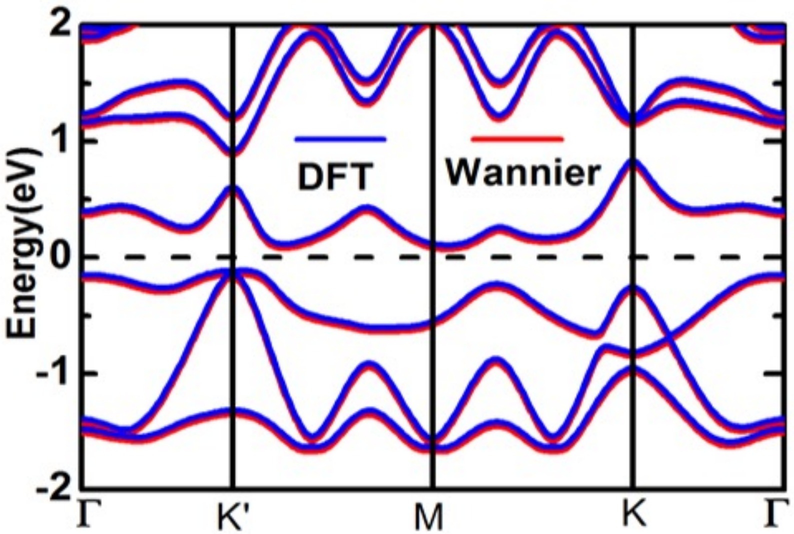
- Insulating, magnetic ground state
- Quantum anomalous Hall effect

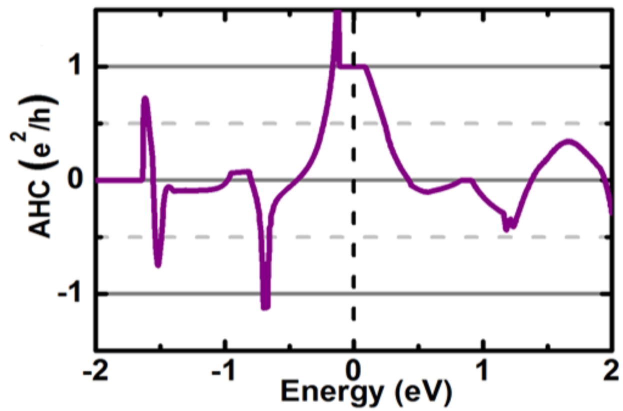
  $C = 1$
- Quantum-valley Hall effect

  $C_K = 1 \quad C_{K'} = 0$

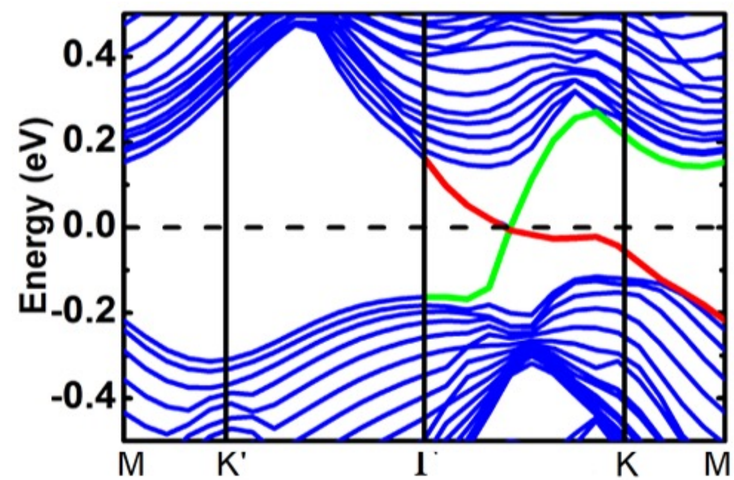
Valley-polarized QAH states

Band structures





Anomalous Hall conductivity



Edge states

- Insulating, magnetic ground state
- Quantum anomalous Hall effect

  $C = 1$
- Quantum-valley Hall effect

  $C_K = 1 \quad C_{K'} = 0$

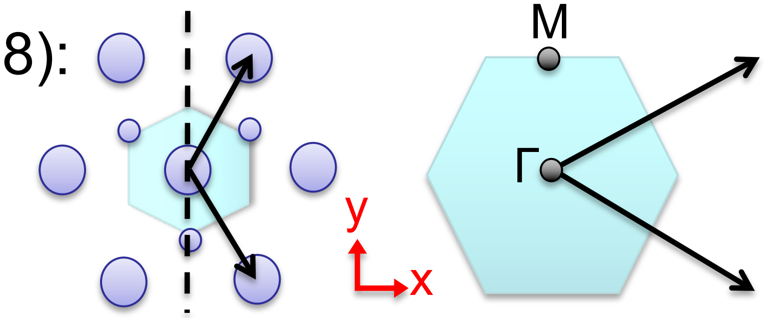
2D Topological Crystalline Insulators:

TCI-QAHI transitions

TIs and mirror Chern numbers

Teo, Fu & Kane PRB **78** 045426 (2008):

Surface states of Bi_xSb_{1-x} along ΓM



Mirror operation: $M = P \cdot C_2^x$

Spin rotation matrix: $U(\alpha,\beta,\gamma) = \begin{pmatrix} e^{-\frac{i(\alpha+\gamma)}{2}} \cos\left(\frac{\beta}{2}\right) & -e^{-\frac{i(\alpha-\gamma)}{2}} \sin\left(\frac{\beta}{2}\right) \\ e^{\frac{i(\alpha-\gamma)}{2}} \sin\left(\frac{\beta}{2}\right) & e^{\frac{i(\alpha+\gamma)}{2}} \cos\left(\frac{\beta}{2}\right) \end{pmatrix}$ e.g. $U(0,0,\pi) = \begin{pmatrix} -i & 0 \\ 0 & i \end{pmatrix}$

Mirror eigenvalues: $M\Psi_{\pm S} = \pm i\Psi_{\pm S}$

Berry curvature & Chern numbers

$$\Omega_z(\mathbf{k}) = 2\mathrm{Im} \sum_{n,m\neq n} \frac{\langle m\mathbf{k}|\nabla_x|n\mathbf{k}\rangle\langle n\mathbf{k}|\nabla_y|m\mathbf{k}\rangle}{(\varepsilon_n(\mathbf{k}) - \varepsilon_m(\mathbf{k}))^2}$$

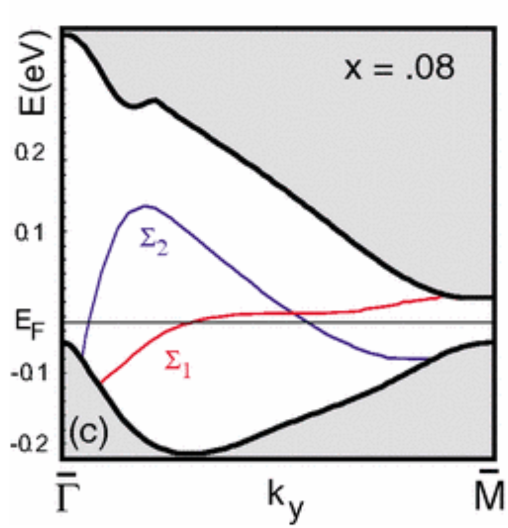
Consider separately the +i and –i sector: **Mirror Chern Number**

$$n_{+i} = \frac{1}{2\pi}\int \Omega_z^{+i}(\mathbf{k})d\mathbf{k}$$

$$n_{-i} = \frac{1}{2\pi}\int \Omega_z^{-i}(\mathbf{k})d\mathbf{k}$$

$$\boxed{n_{\mathcal{M}} = \frac{1}{2}(n_{+i} - n_{-i})}$$

determines connectivity of edge states in TIs:

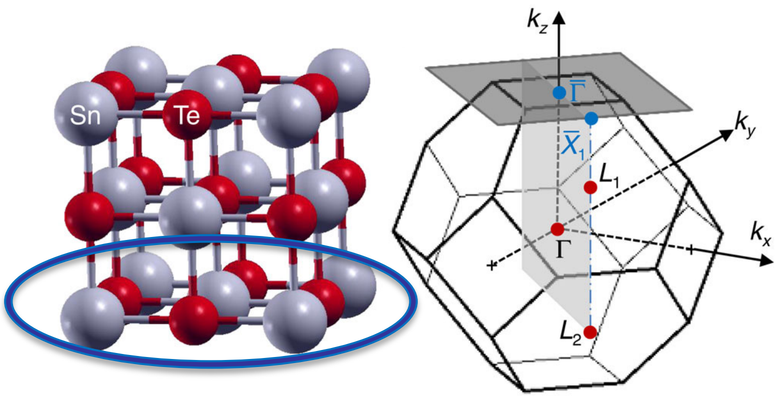


Topological Crystalline Insulators

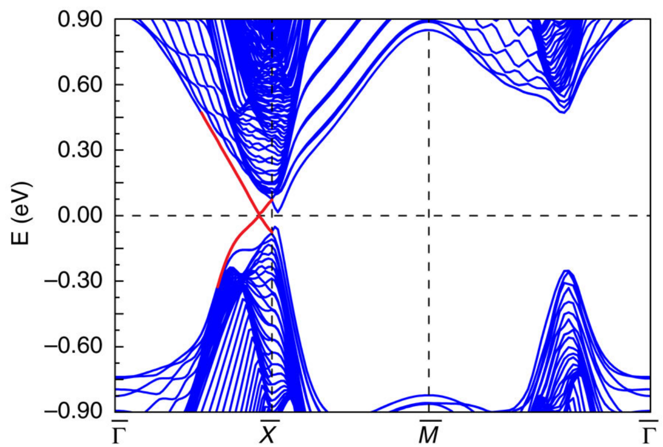
L. Fu [PRL **106**, 106802 (2011)] :

Systems with finite mirror Chern number possess gapless surface states even if they are no TIs!

Example: SnTe:

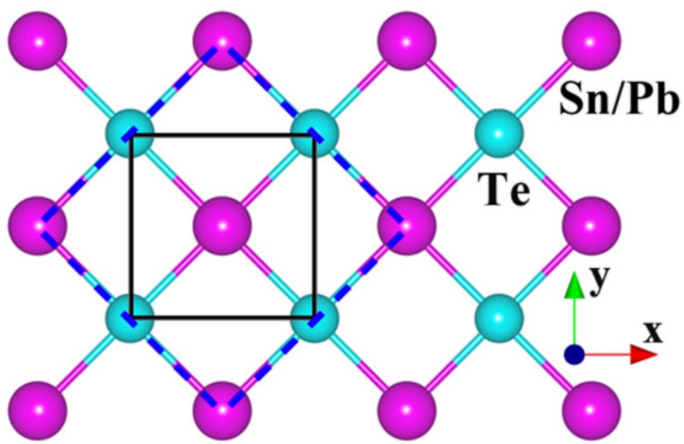


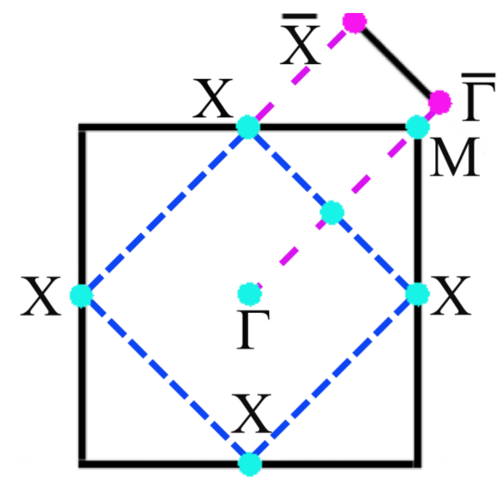
consider
one layer (2D)

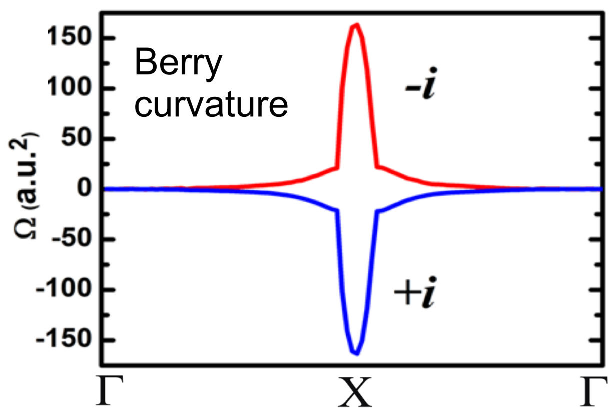


Hsieh et al., Nature Comms. **3**, 982 (2012)

2D topological crystalline insulator (TCI)

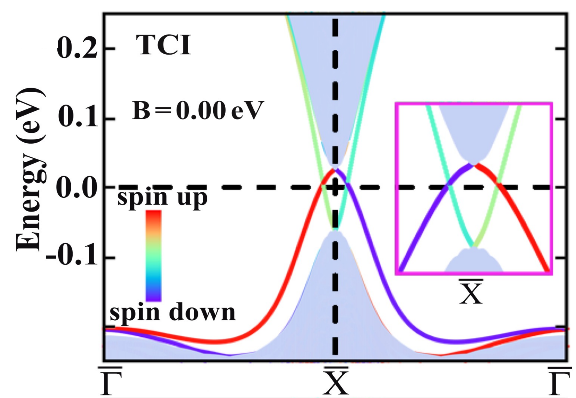






$$n_{+i} = -2 \quad n_{-i} = 2$$

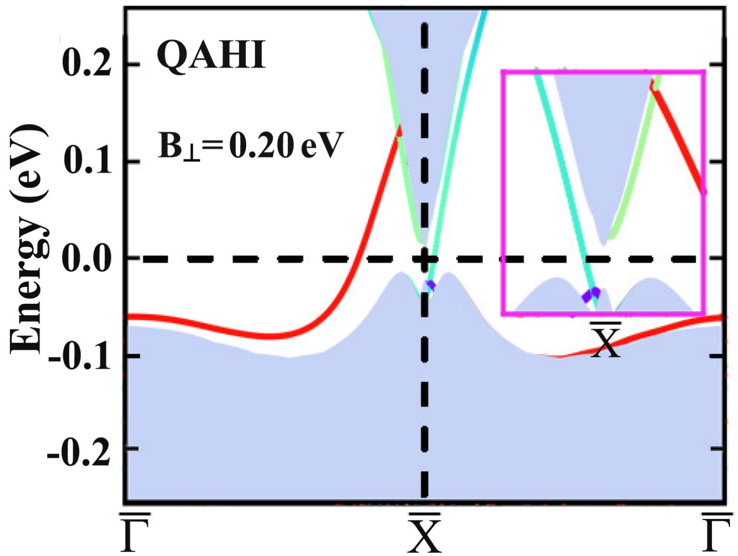
$$n_M = -2$$

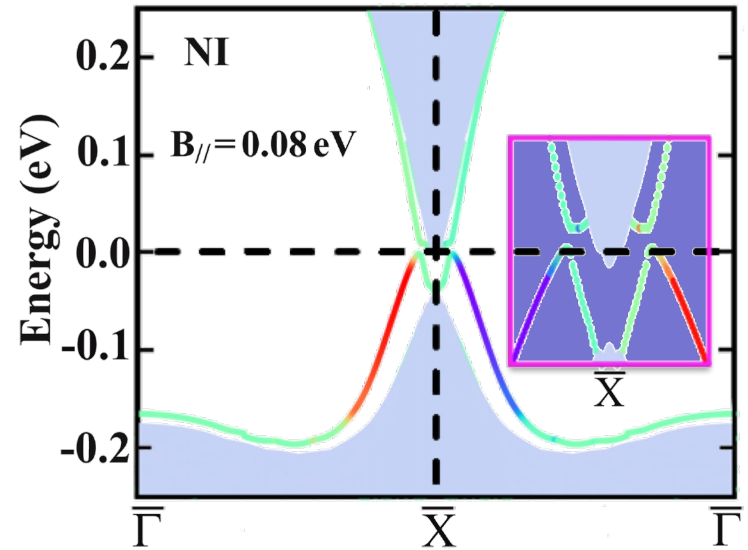


Two pairs of gapless edge states

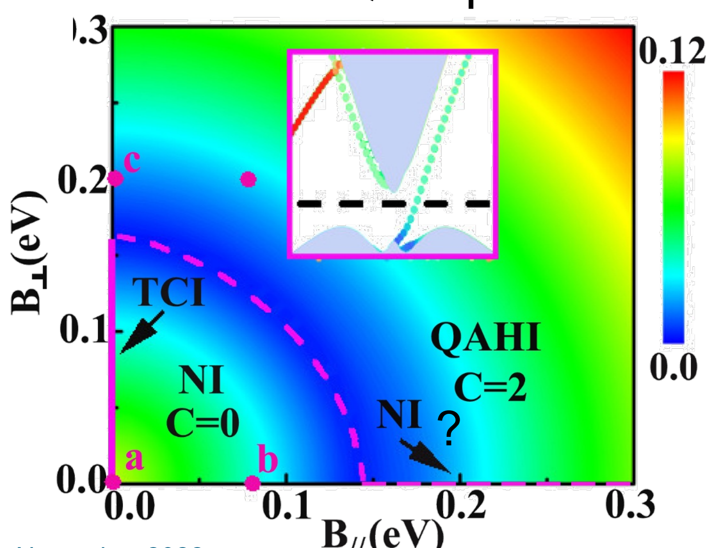
C. Niu, et al., Phys. Rev. B 91, 201401(R) (2015)

2D TCI – QAHE transition



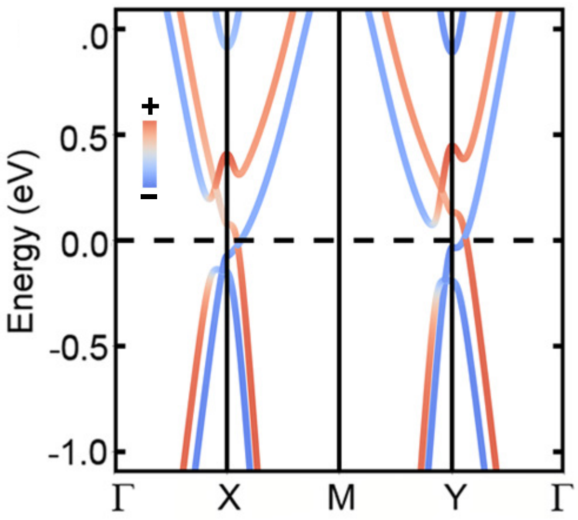


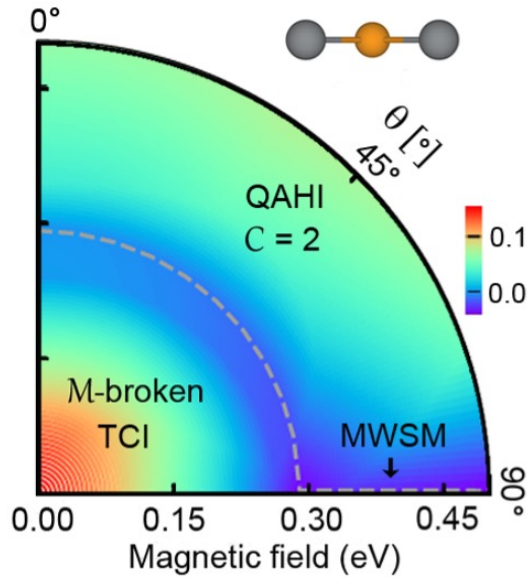
- Out of plane exchange field: QAH phase



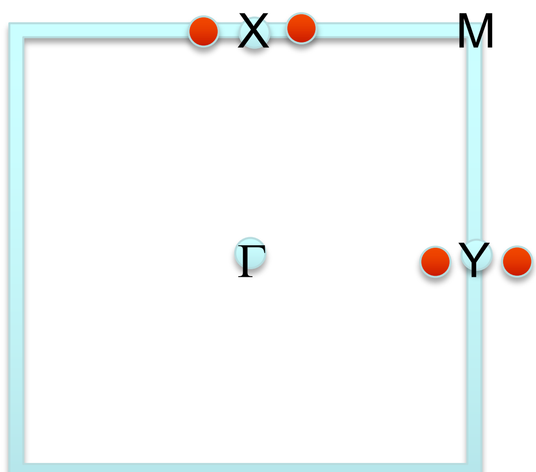
- In-plane exchange field: Gap
- QAH phase robust to the direction magnetization of adatoms or the substrate

Other example: TlSe monolayer





in-plane field
$B_{||}$ = 0.5 eV
nodal points

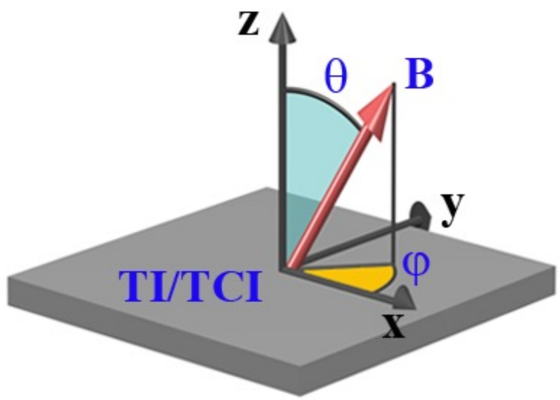


Mixed topological semimetals:

Weyl points and nodal lines in (B,k) space

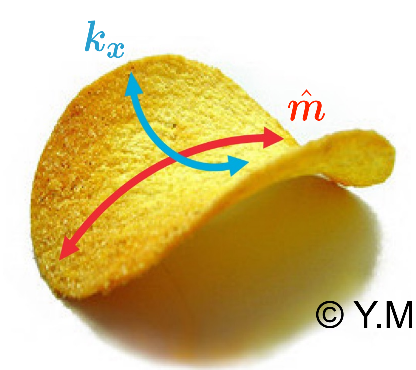
Berry curvature & Chern numbers

$$\Omega_{xy}^{\mathbf{kk}} = 2\mathrm{Im}\sum_{n}^{\mathrm{occ}} \langle \partial_{k_x} u_{\mathbf{k}n} | \partial_{k_y} u_{\mathbf{k}n} \rangle$$



Quantity in (x,y), include direction of B-field to expand space by θ,φ:

$$\Omega_{\theta i}^{\hat{\mathbf{m}}\mathbf{k}} = 2\mathrm{Im}\sum_{n}^{\mathrm{occ}} \langle \partial_{\theta} u_{\mathbf{k}n}^{\theta} | \partial_{k_i} u_{\mathbf{k}n}^{\theta} \rangle$$



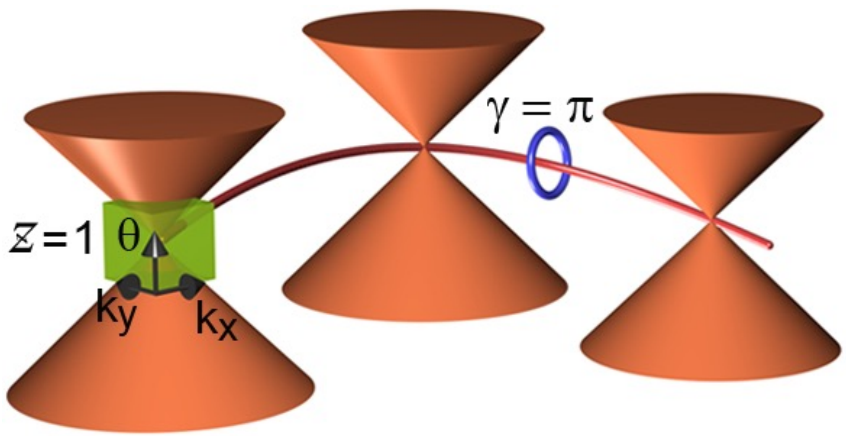
Define new topological charge:

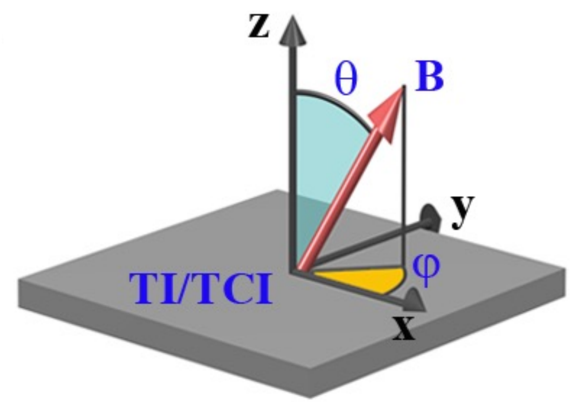
$$\mathbf{\Omega} = \left(-\Omega_{\theta y}^{\hat{\mathbf{m}}\mathbf{k}}, \Omega_{\theta x}^{\hat{\mathbf{m}}\mathbf{k}}, \Omega_{xy}^{\mathbf{kk}}\right)$$

$$\mathcal{Z} = \frac{1}{2\pi}\int_{S} \mathbf{\Omega} \cdot d\mathbf{S}$$

Hanke et al., Nat. Comm. **8**, 1479 (2017)

Nodal lines and points in (B,k)-space





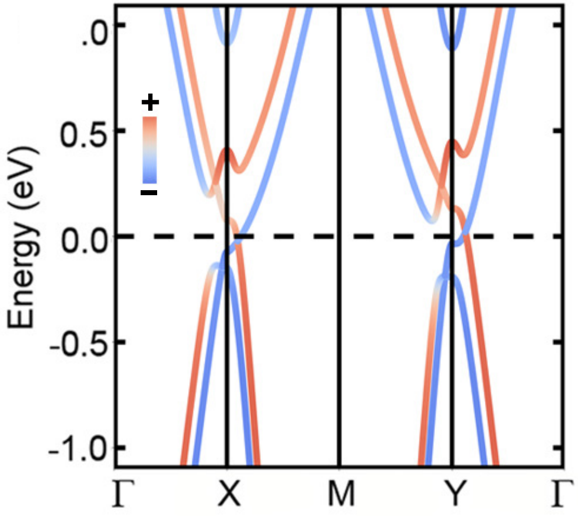
mixed Chern number characterizes *nodal points and lines in ($k_{||}$, θ) space*

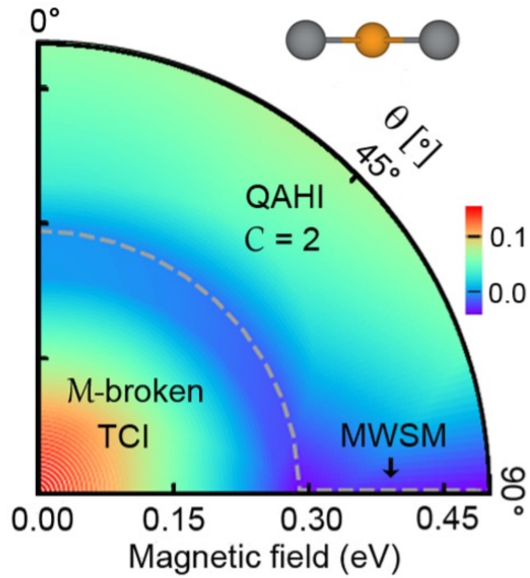
- anti-damping SO-torque mediated by *mixed* phase-space topology

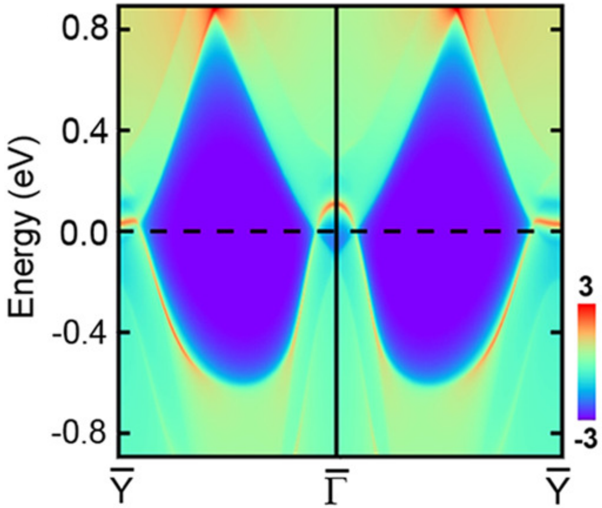
$$\tau_{ij} \sim 2\mathrm{Im} \sum_n \hat{e}_i \cdot \left\langle \frac{\partial u_{\boldsymbol{k}n}}{\partial \hat{\boldsymbol{m}}} \middle| \frac{\partial u_{\boldsymbol{k}n}}{\partial k_j} \right\rangle$$

Freimuth, et al., Phys. Rev. B '13 – '17,
J. Phys.: Cond. Matter **26**, 104202 (2014),
Hanke et al., Nat. Comm. **8**, 1479 (2017)

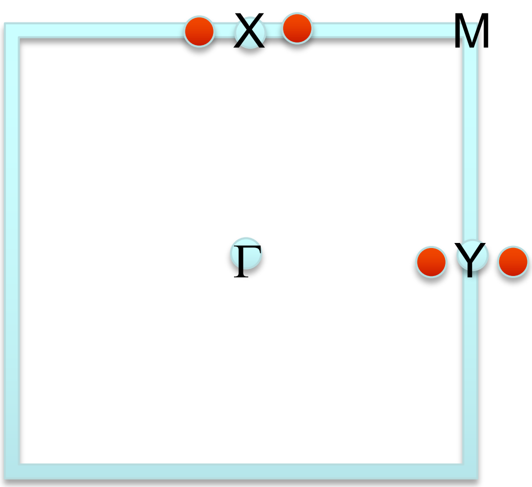
Example: TlSe monolayer







in-plane field
$B_{||}$ = 0.5 eV
nodal points



Fermi arcs between the nodal points

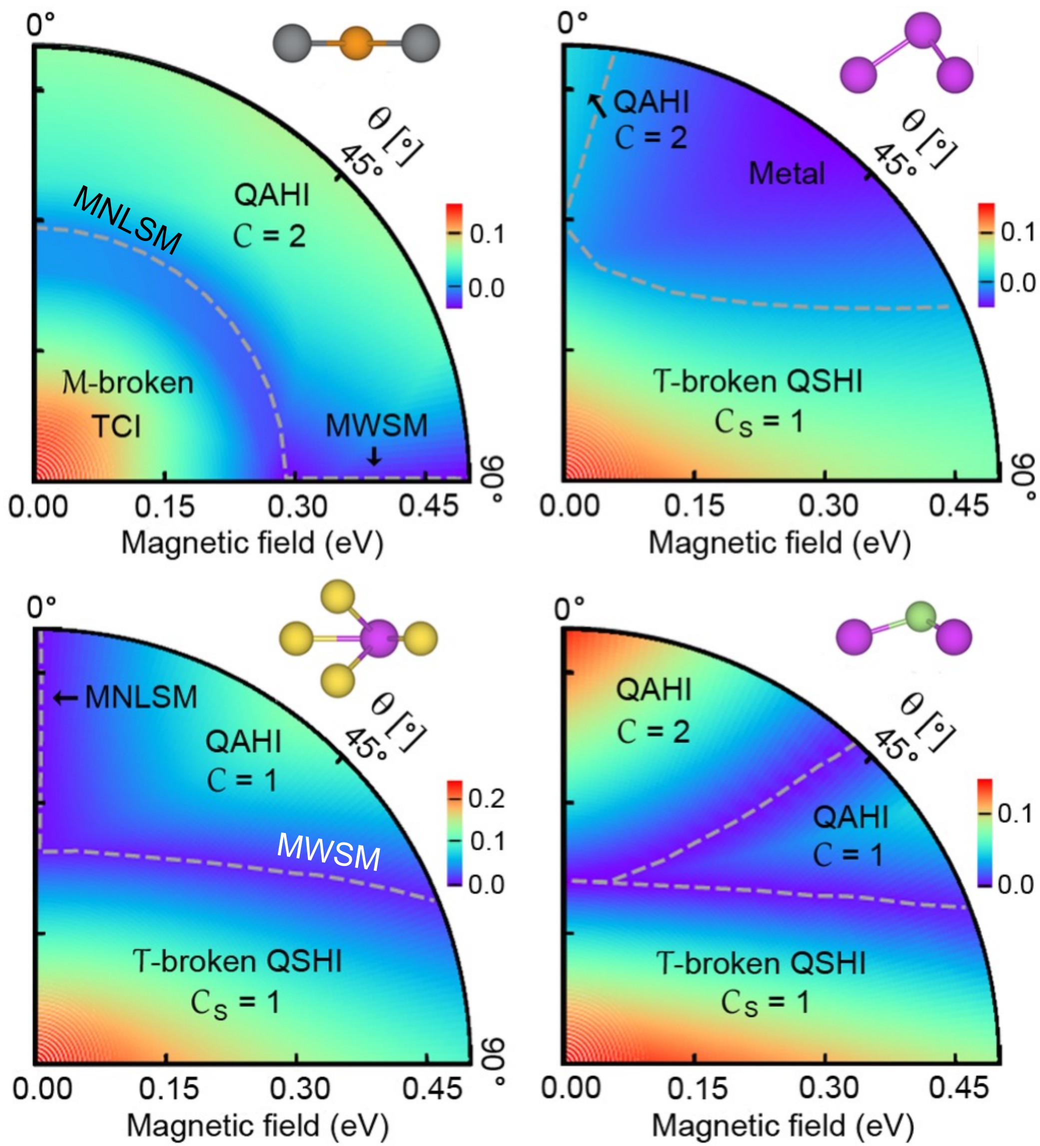
Band gap as function of B-field orientation

Mirror symmetry:

TlSe

Mixed nodal line and Weyl semimetal phases:

Na_3Bi



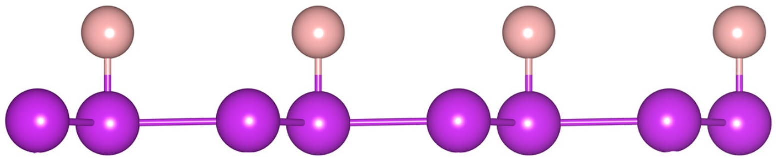
No semimetal phase:

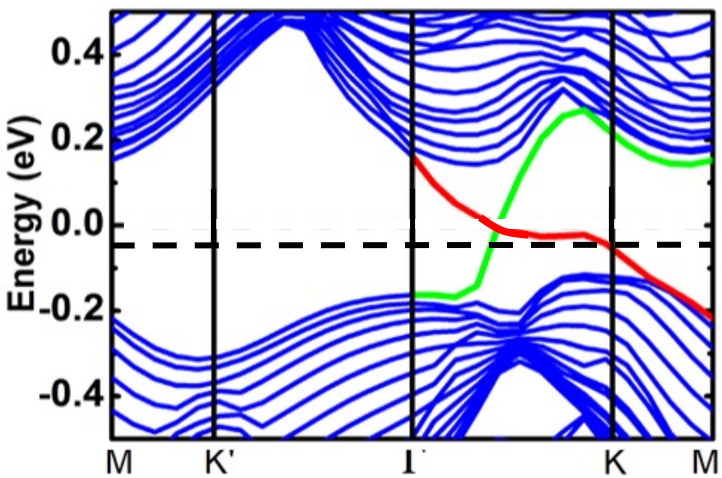
Bi bilayer

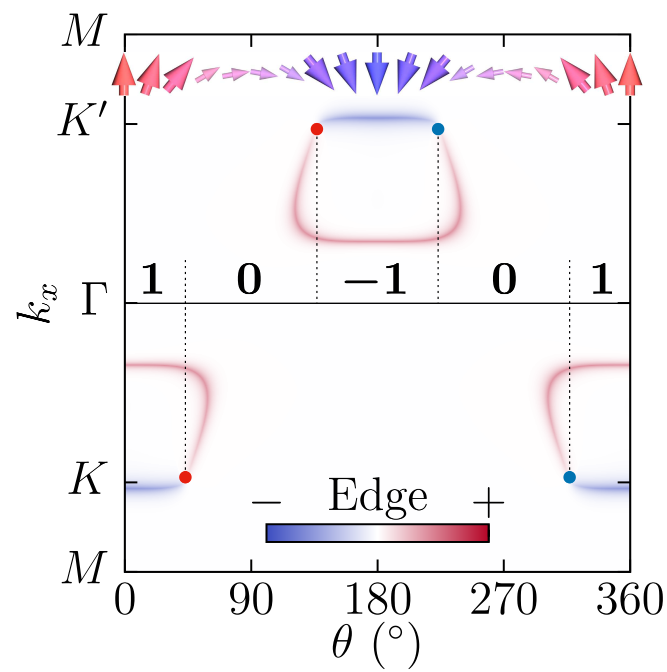
only MWSM:

GaBi bilayer

semi-hydrogenated Bi bilayer:



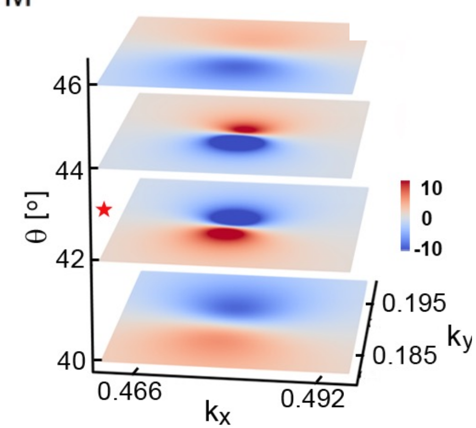




red/green: opposite edges

huge orbital moment change at the Weyl point:

- ➢ large spin-orbit torques
- ➢ jump in magnetoelectric response

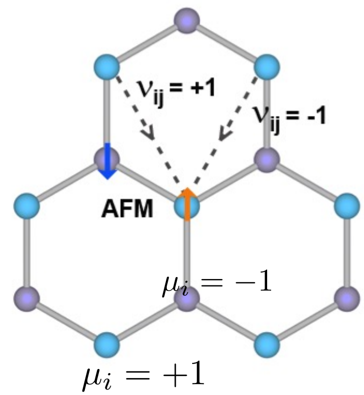


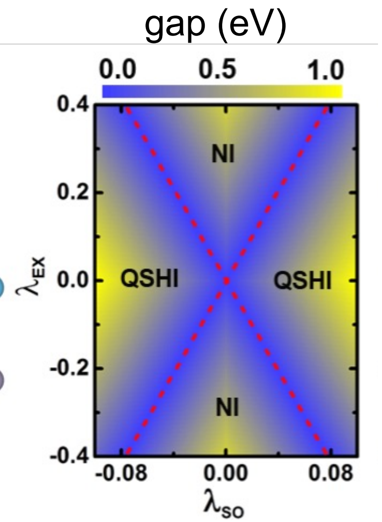
Antiferromagnetic spin-Hall insulators:

Sn-halogenides

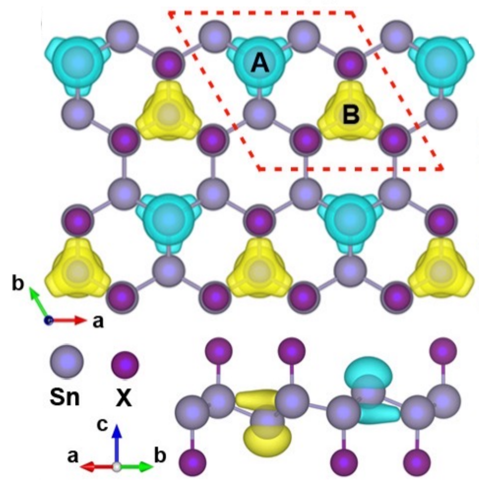
Antiferromagnetic order + QAHE = QSHE

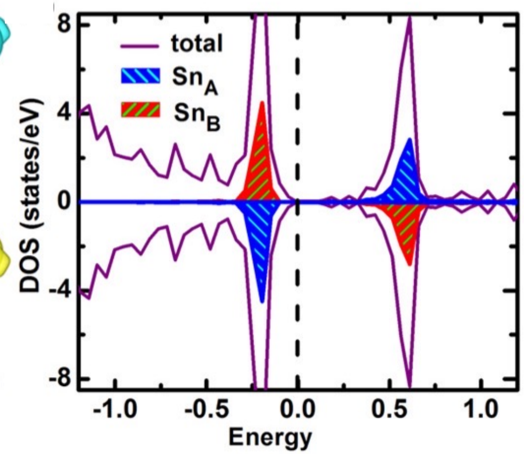
$$
\begin{aligned}
H = &-t \sum_{\langle ij\rangle,\alpha} c^{\dagger}_{i,\alpha} c_{j,\alpha} \\
&+ i\lambda_{\mathrm{SO}} \sum_{\langle\langle ij\rangle\rangle,\alpha\beta} v_{i,j}\sigma^{z}_{\alpha,\beta} c^{\dagger}_{i,\alpha} c_{j,\beta} \\
&+ \lambda_{\mathrm{EX}} \sum_{i,\alpha} \mu_i \sigma^{z}_{\alpha\alpha} c^{\dagger}_{i,\alpha} c_{i,\alpha}
\end{aligned}
$$





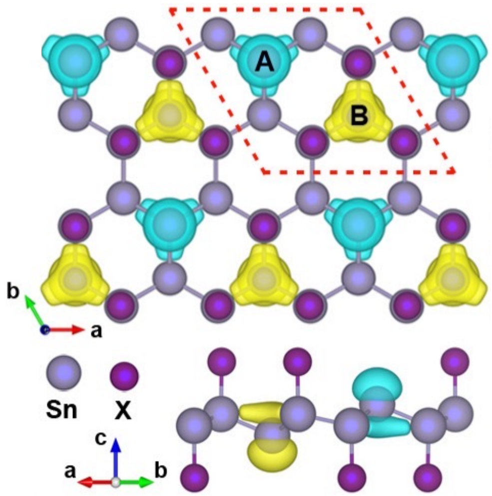
material realization: halogenated Sn bilayer (Sn_4I_3)

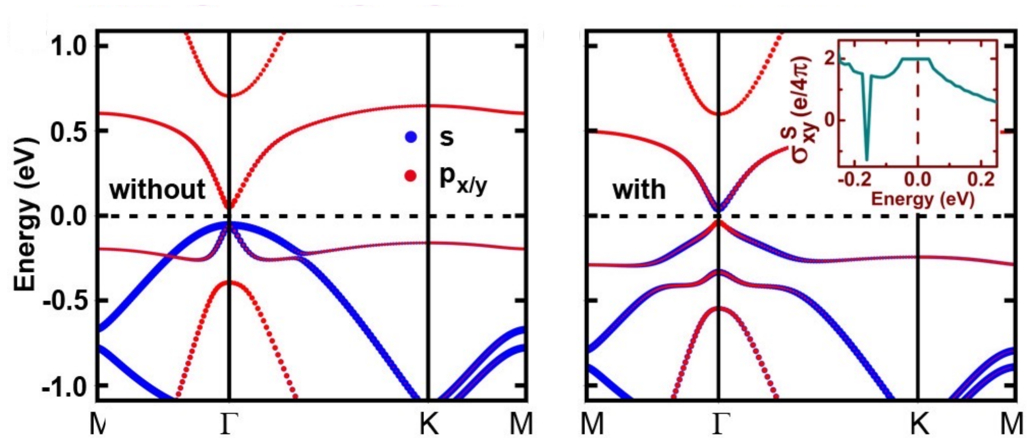




C. Niu et al.
arXiv:1705.07035
and Phys. Rev. Lett. **124,** 066401 (2020)

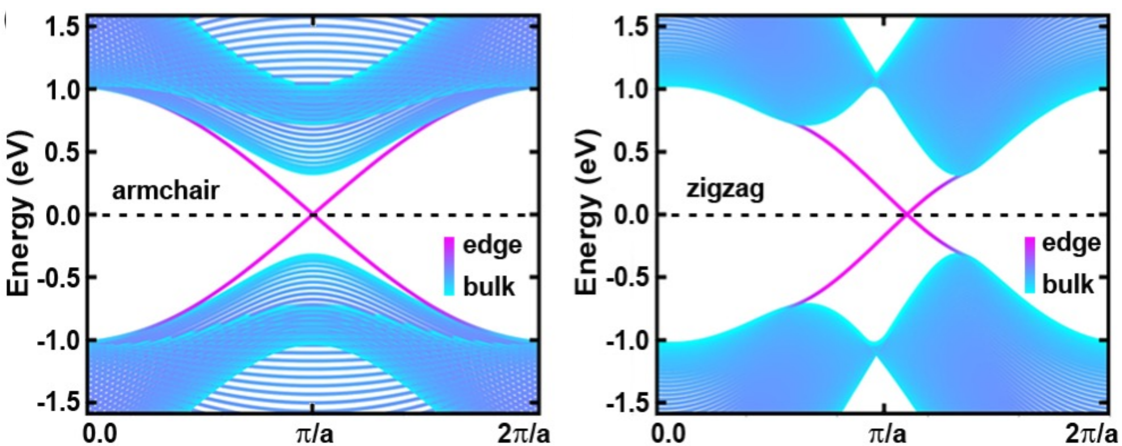
QSH effect in antiferromagnetic TIs





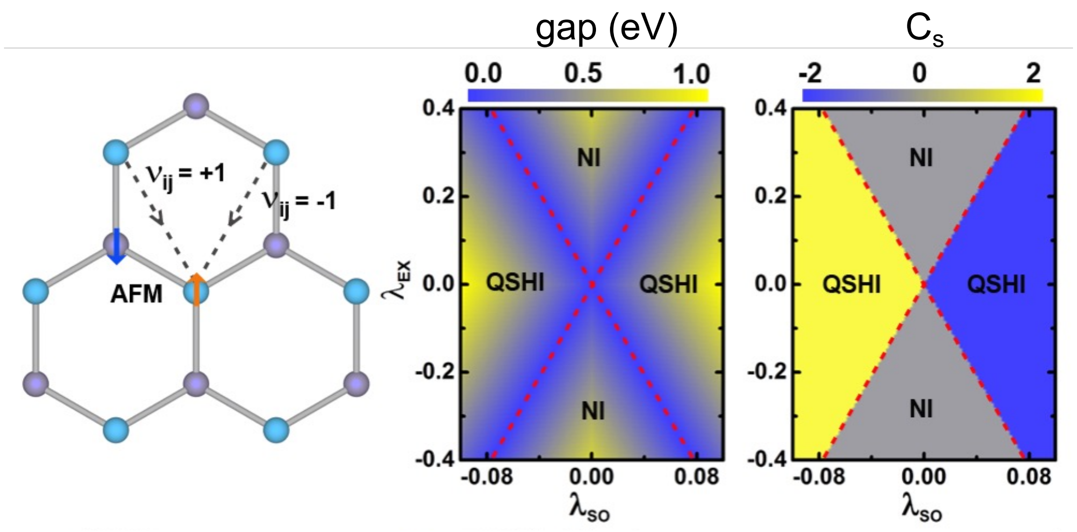
band inversion creates quantized SH current

additional valley-polarization (model result):

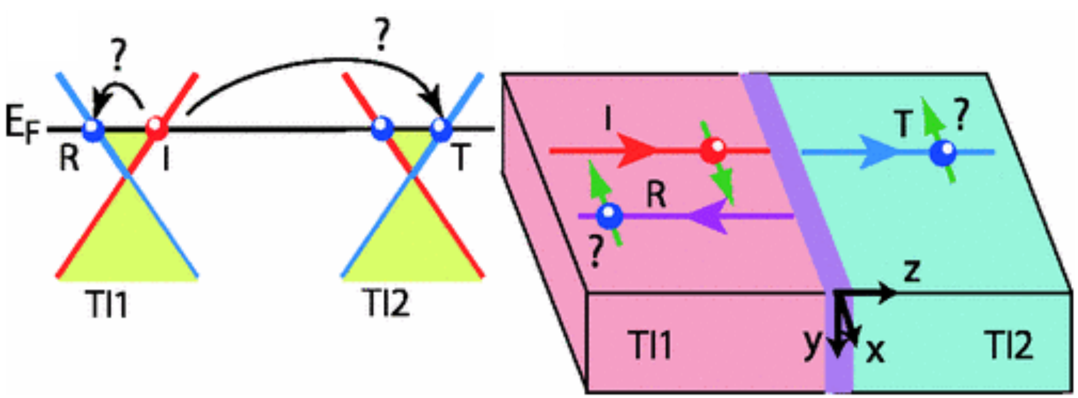


Switchable QSH states

$$H = -t \sum_{\langle ij \rangle, \alpha} c^{\dagger}_{i,\alpha} c_{j,\alpha}$$
$$+ i\lambda_{\mathrm{SO}} \sum_{\langle\langle ij \rangle\rangle \alpha\beta} v_{i,j} \sigma^z_{\alpha,\beta} c^{\dagger}_{i,\alpha} c_{j,\alpha}$$
$$+ \lambda_{\mathrm{EX}} \sum_{i,\alpha} \mu_i \sigma^z_{\alpha\alpha} c^{\dagger}_{i,\alpha} c_{i,\alpha}$$



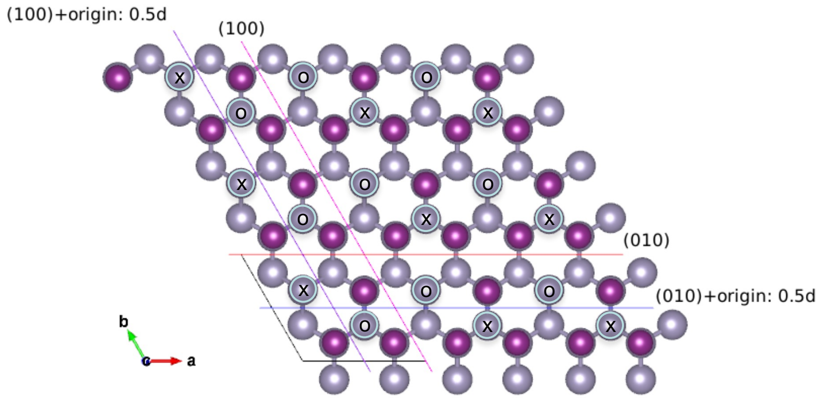
control of the spin-current direction via sublattice magnetization



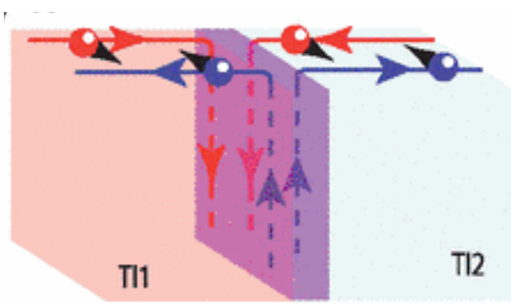
Gapless interface state appears between two TIs with opposite Dirac velocities

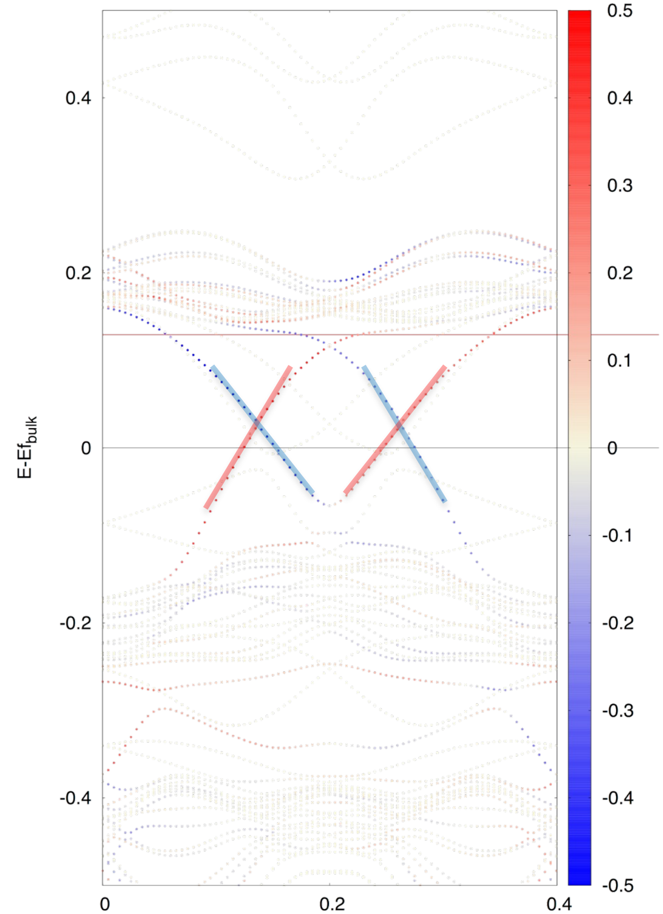
[Takahashi & Murakami, Phys. Rev. Lett. **107**, 166805 (2011)]

A-B domain wall in Sn_4I_3



zig-zag domain wall between two
AFM domains hosts four edge states:





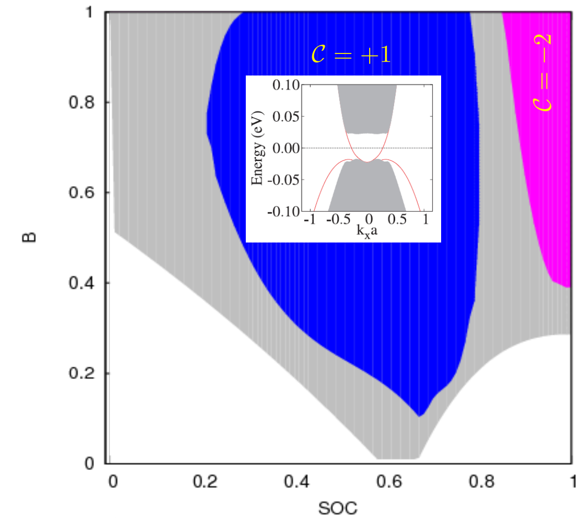
G. Wójtowicz, I. Aguilera (2018)

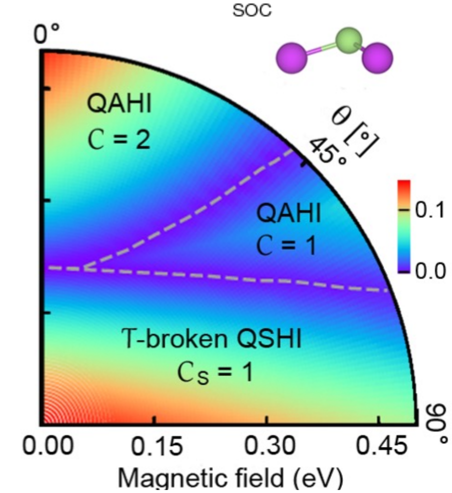
Takahashi & Murakami, PRL **107**, 166805 (2011)

Topological AFM spintronics: Nat. Phys. **14**, 242 (2018)

Summary:

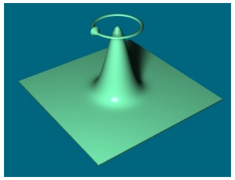
- QSHI – QAHI transition
  - large B-fields needed
  - rich phase diagrams
  - chemical modification: H-Bi
- TCI– QAHI transition
  - small **B**: directional dependence
  - large **B**: robust QAH phase
- Mixed Weyl semimetals
  - magnetic moment direction as new variable
  - nodal lines/points in (**B**,**k**) space
- AFM order in TIs
  - handle on spin current direction





Acknowledgement

- SPP 1666 of the DFG
- VH-NG-513 of the HGF



- VITI

Thank you for your attention