First-principles design of topological insulators

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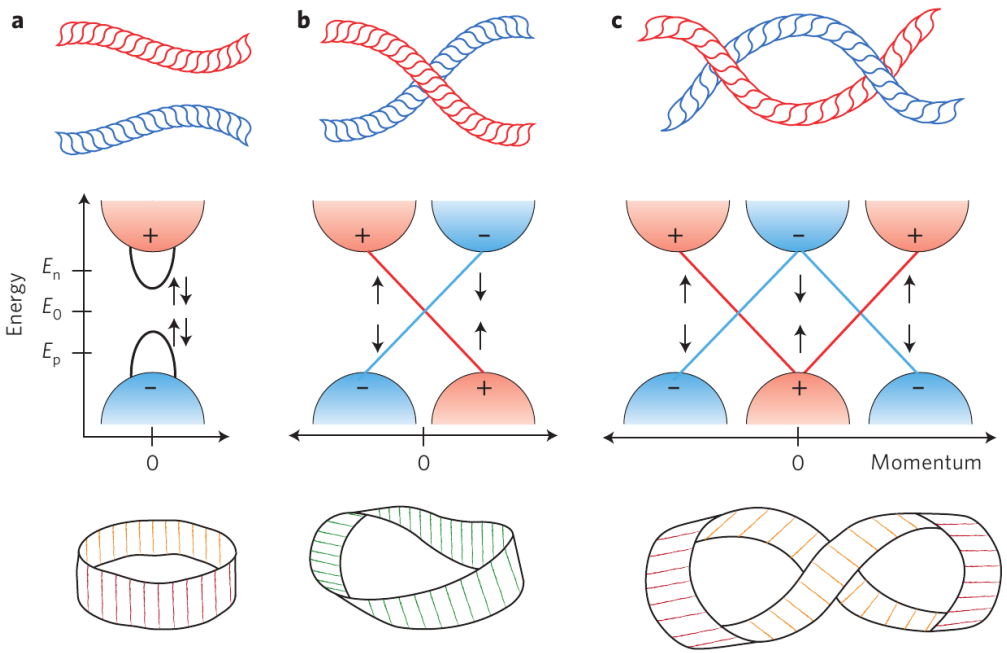
Topological insulators from first-principles

- **Motivation**
- **First-principles material design**
- **Topological insulators**
  - Design of new materials
  - Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$
  - Exchange interaction in binary diluted chalcogenides
  - Impact of electron-magnon interaction on the Dirac state
- **Summary & outlook**

Motivation



Schematic band structure of conventional and topological insulators

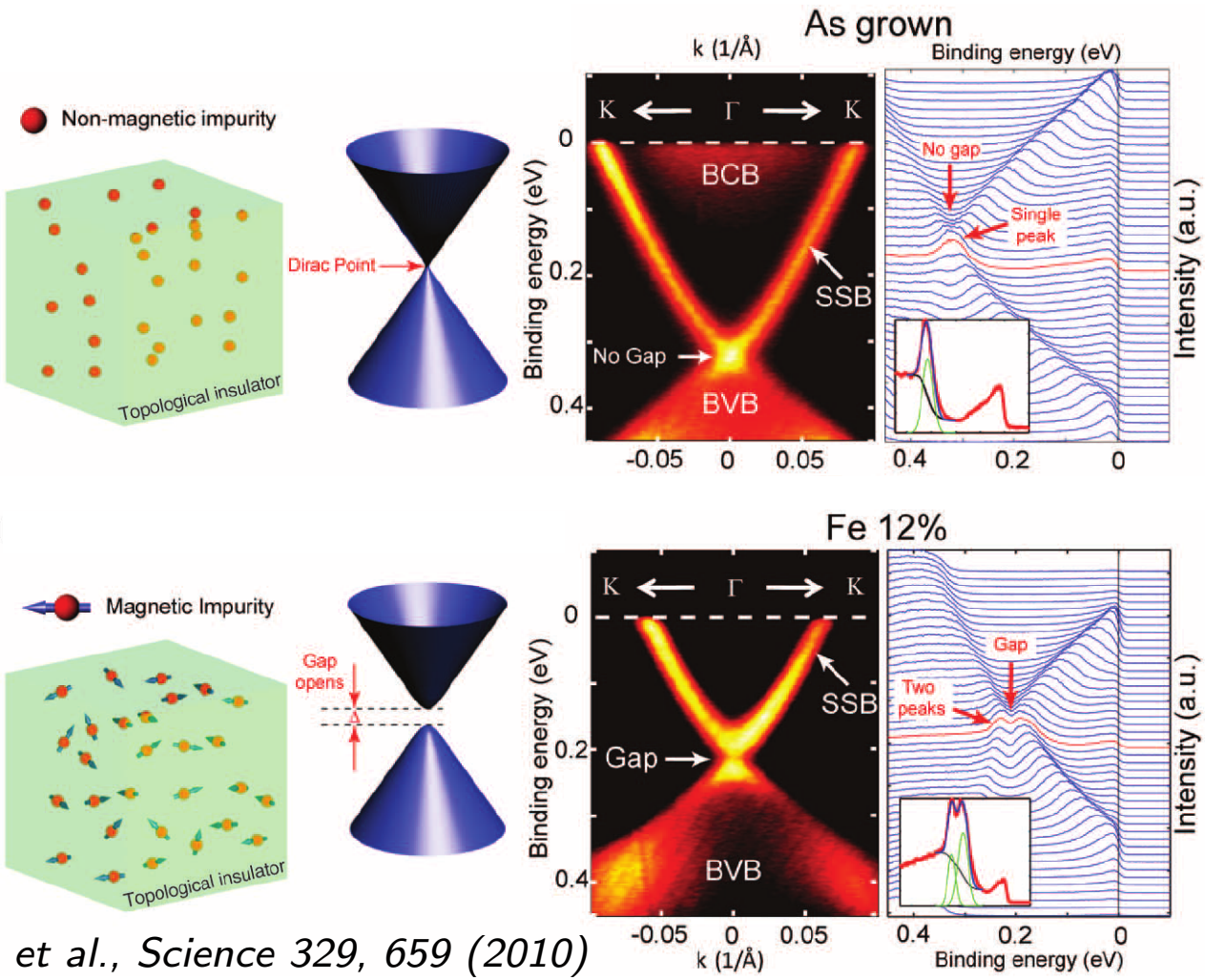
Electronic transport on the surface of a topological insulator

Main features of topological insulators

- Small band gap
- Strong spin-orbit coupling
- Dirac surface state

Motivation: Magnetic impurities in topological insulators

Bi_2Se_3: photoemission experiments



Y. L. Chen et al., Science 329, 659 (2010)

Goals

Search of new materials with desired properties

Motivation & Goals

- Spin momentum locked pure spin current
- Formation & spin texture of Dirac surface states
- Tuning the topological character
- Transport & spectroscopy
- Role of chemical & magnetic disorder
- Spin Hall effect
- Topological magneto-electric effects

Ab-initio Kohn-Sham approach

- **Wave function equation**

$$\left[\varepsilon + \frac{\hbar^2}{2m}\nabla^2 - V_{eff}(\mathbf{r})\right]\Psi(\mathbf{r};\varepsilon) = 0$$

- **Green function equation**

$$\left[\varepsilon + \frac{\hbar^2}{2m}\nabla^2 - V_{eff}(\mathbf{r})\right]G(\mathbf{r},\mathbf{r}';\varepsilon) = \delta(\mathbf{r}-\mathbf{r}')$$

Korringa (1947), Kohn & Rostoker (1954)

- **Dyson equation**

$$G = G_0 + G_0 \Delta V_{eff} G$$

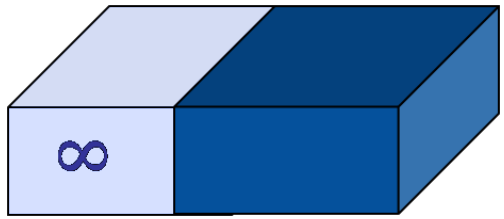
$$\Delta V_{eff} = V_{eff} - V^0_{eff}$$

- **Bulk**

$$G_{bulk} = G_{free} + G_{free} V_{eff} G_{bulk}$$

- **Surfaces & interfaces**

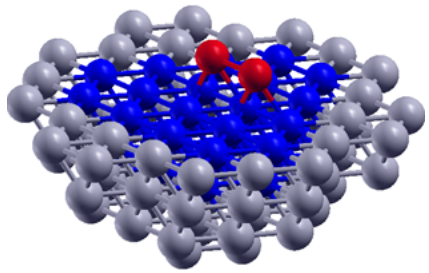
$$G_{surf} = G_{bulk} + G_{bulk} \Delta V_{eff} G_{surf}$$



Wildberger et al. (1997), Uiberacker et al. (1998)

- **Defects in bulk & surfaces**

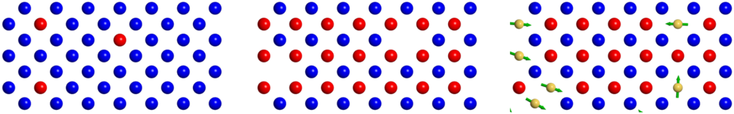
$$G_{cluster} = G_{host} + G_{host} \Delta V_{eff} G_{cluster}$$



Zeller & Dederichs (1979)

Method: Coherent potential approximation

Alloys and pseudo-alloys

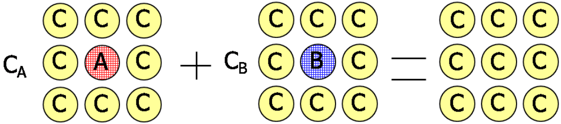


doping/alloys vacancies magnetic impurities

Coherent potential approximation

Soven (1967), Györffy (1972)

CPA equation for a binary alloy: $c_A G_A + c_B G_B = G_C$



Nonlocal CPA: Charge and Spin-Fluctuations

D. A. Rowlands, A. Ernst, J. B. Staunton, B. L. Györffy, PRB 73, 165122 (2006)

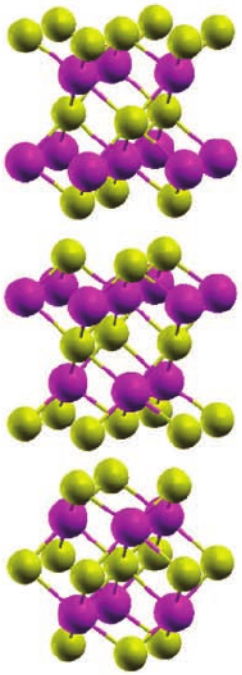
Method: short summary

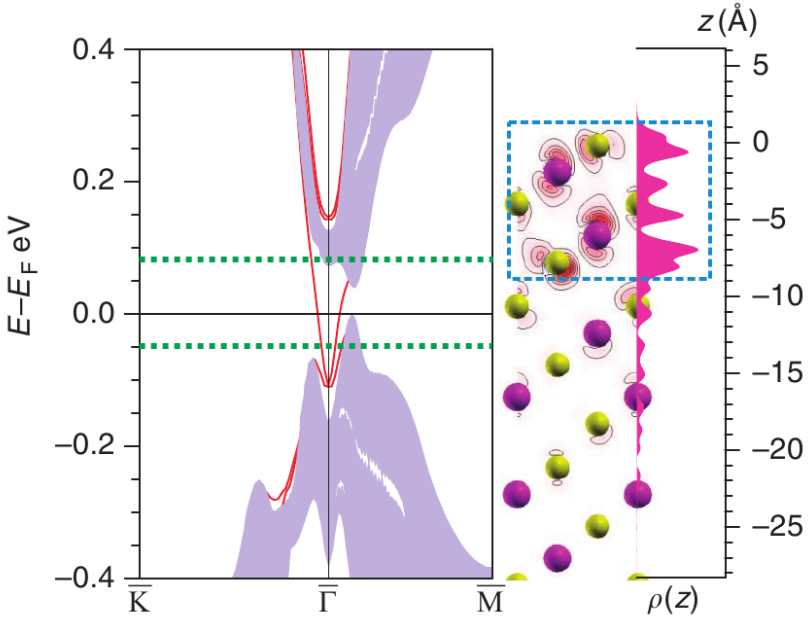
Green-Funktion-Methode

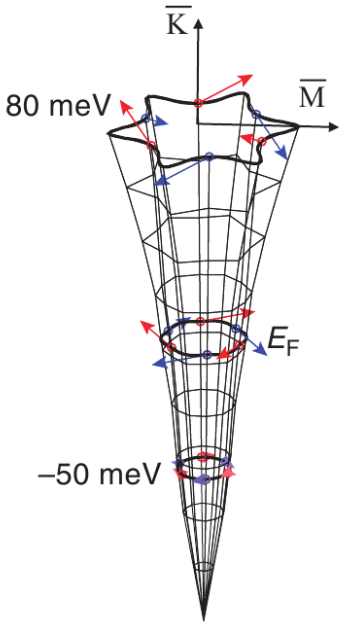
- Explicit Green Function
- Dimensions: 1D, 2D, 3D & Cluster
- $\mathcal{O}(N)$ method
- CPA for disordered alloys
- *multi-code approach*: crystalline structure from VASP or experiments

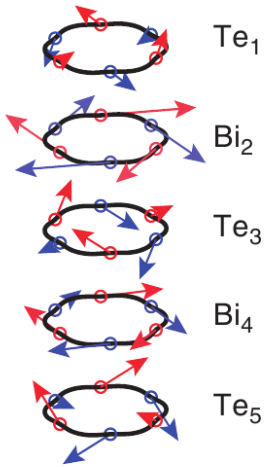
Design of new materials: Topological insulators

Conventional 3D topological insulator Bi_2Te_3









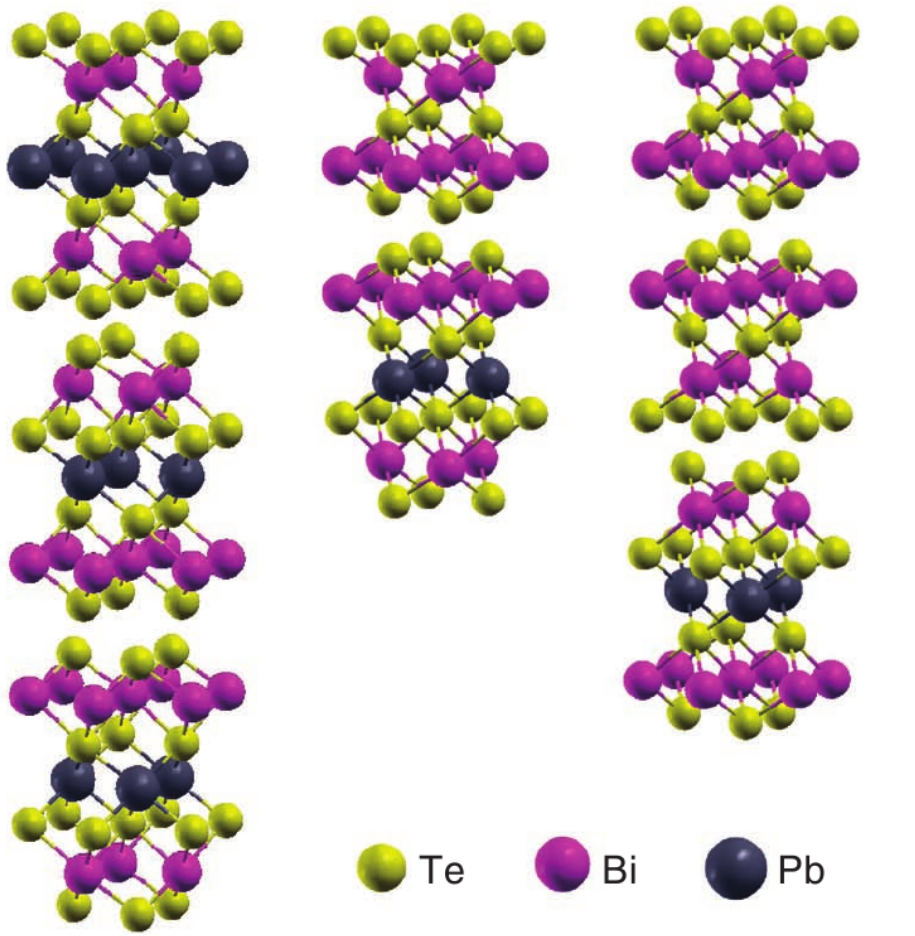
Crystalline structure

Band structure & spin texture

Can we tune specifically topological properties of these materials?

Design of new materials: Topological insulators

Idea: Tuning the spin-orbit coupling using Pb

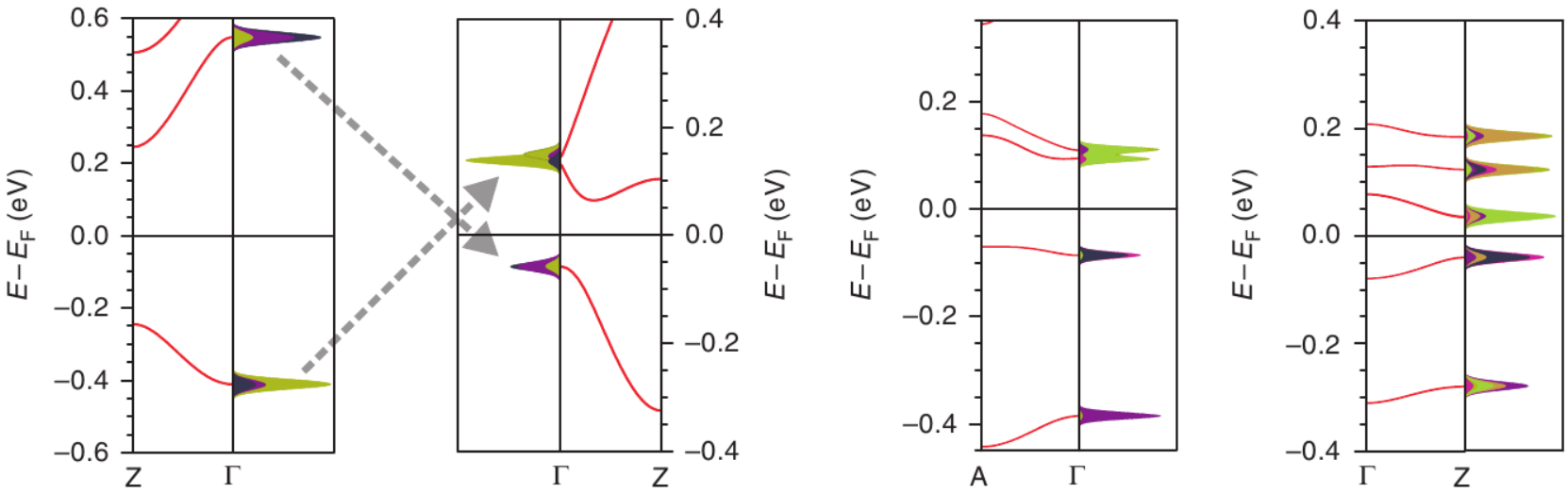


$PbBi_2Te_4$ $PbBi_4Te_7$ $PbBi_6Te_{10}$

Design of new materials: Topological insulators

Idea: Tuning the spin-orbit coupling using Pb

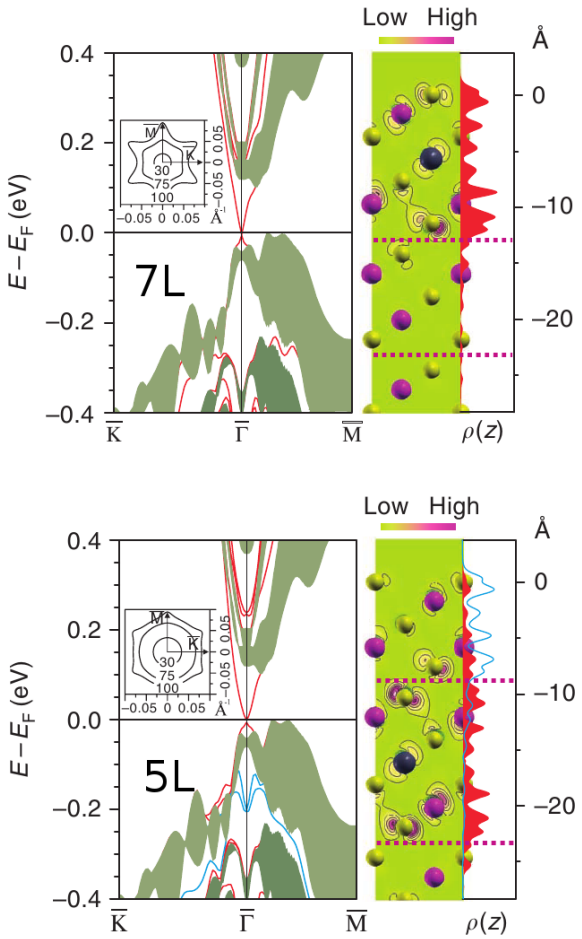
Band structure of bulk

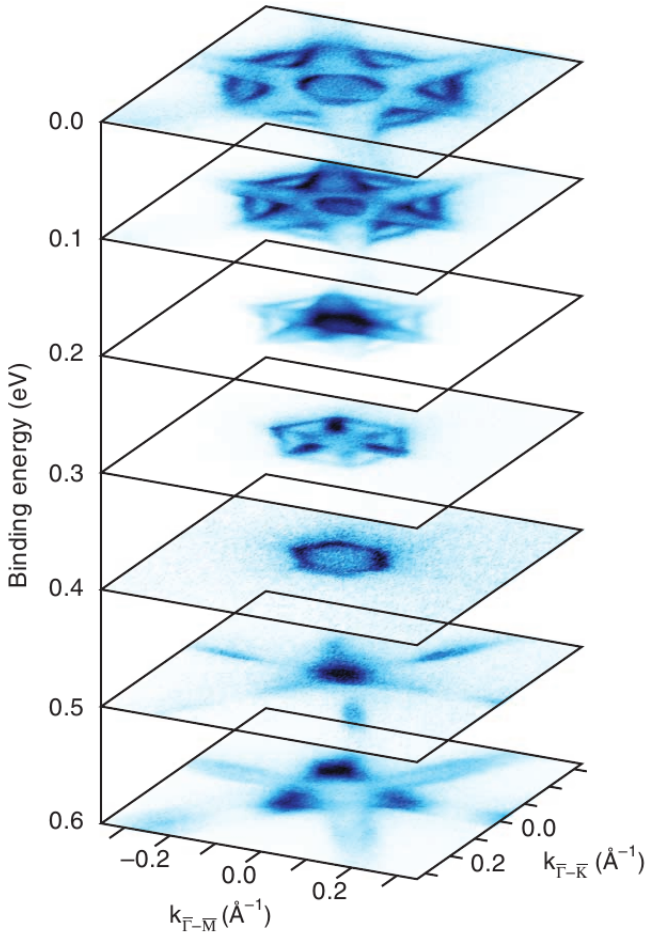


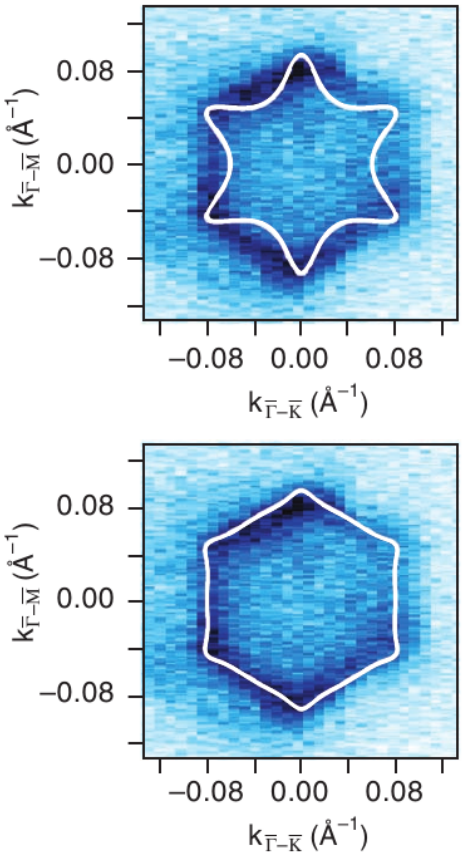
Band inversion in $PbBi_2Te_4$ $PbBi_4Te_7$ $PbBi_6Te_{10}$

Design of new materials: Topological insulators

$PbBi_4Te_7$: Theory & Experiment (ARPES)







S. V. Eremeev, G. Landolt, T. V. Menshchikova, B. Slomski, Y. M. Koroteev, Z. S. Aliev, M. B. Babanly, J. Henk, A. Ernst, L. Patthey, A. Eich, A. A. Khajetoorians, J. Hagemeister, O. Pietzsch, J. Wiebe, R. Wiesendanger, P. M. Echenique, S. S. Tsirkin, I. R. Amiraslanov, J. H. Dil, & E. V. Chulkov, Nature communications (2012)

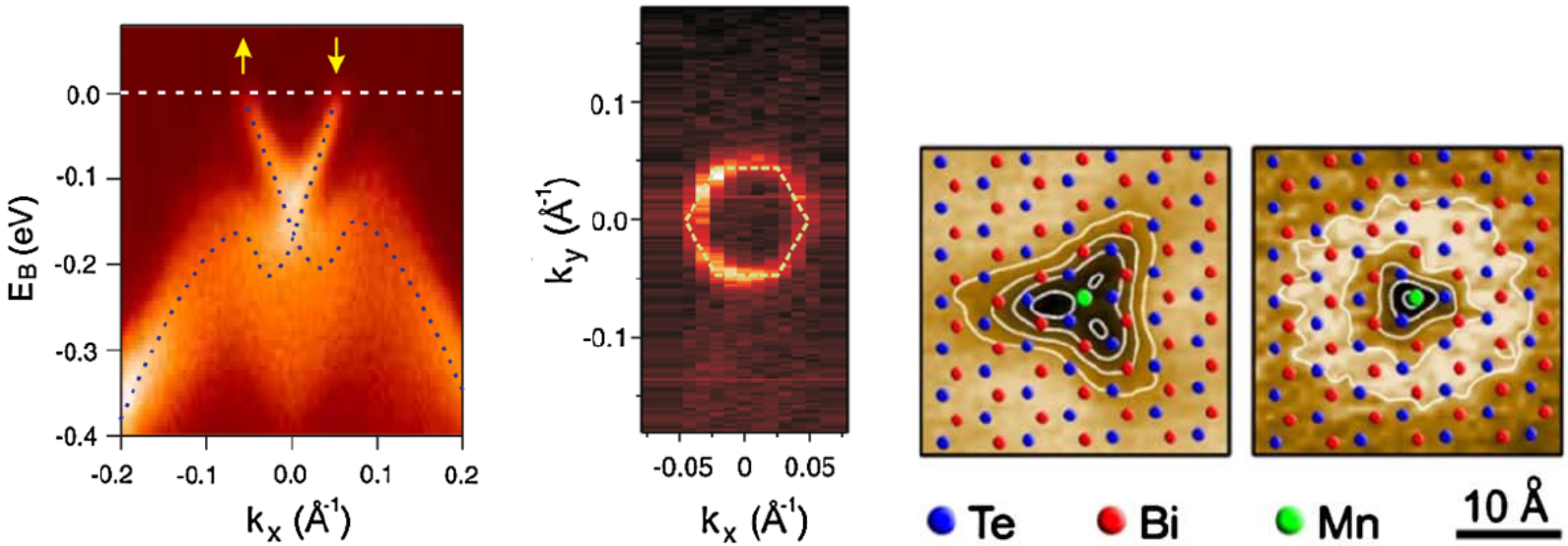
Design of new materials: Topological insulators

Short summary

- Dirac surface state can be tuned in Bi_2Te_3 and Bi_2Se_3 with the addition of a third element of the group IV
- Part of new systems represent naturally grown superlattices composed of 5L and 7L blocks
- New materials exhibit much richer physics rather the parent compounds
- First theory then experiment (not otherwise)

Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$

Photoemission and STM experiments on $Bi_{2-x}Mn_xTe_3$

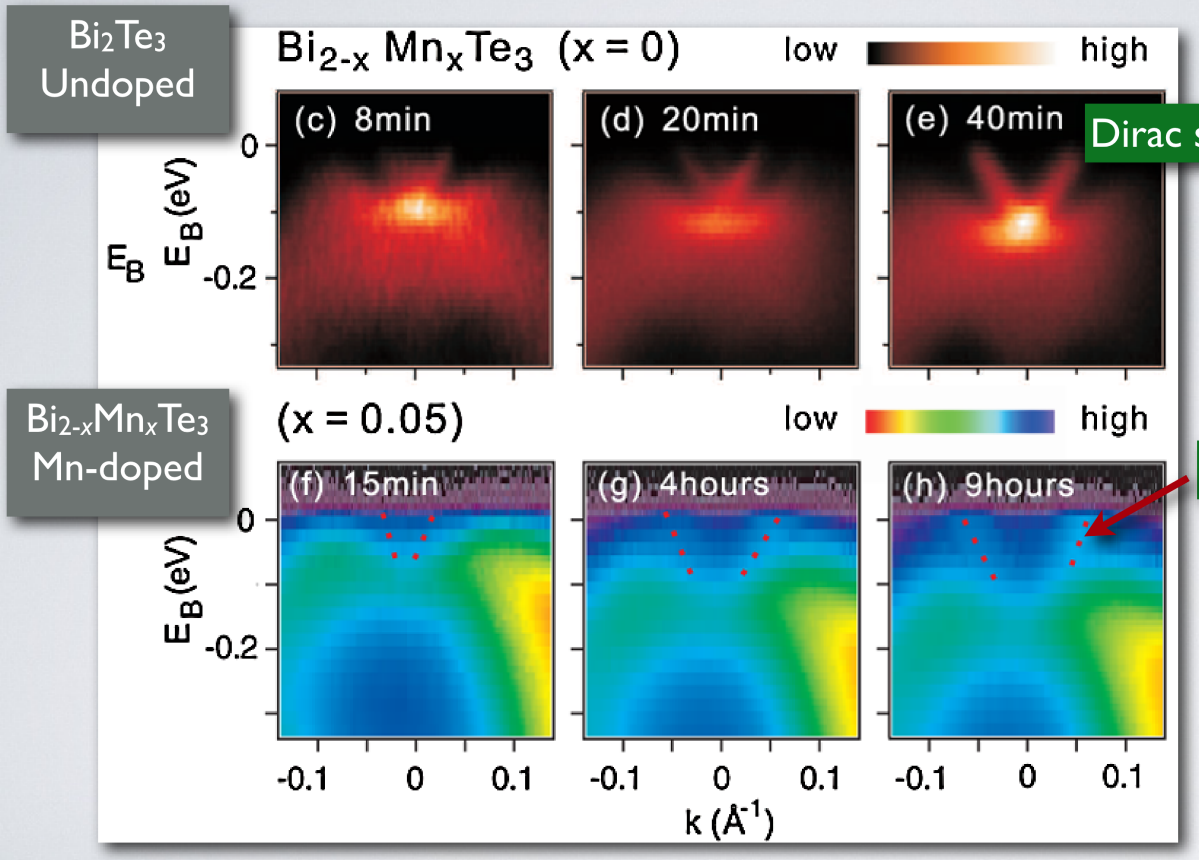


Hsieh et al, PRL (2009)

Hor et al., PRB (2010)

Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$

Photoemission experiments on $Bi_{2-x}Mn_xTe_3$



`Dirac state´

- survives
- blurred
- weak intensity
- dispersion

➠ Model and *ab initio* calculations

Hsieh et al, PRL (2009)

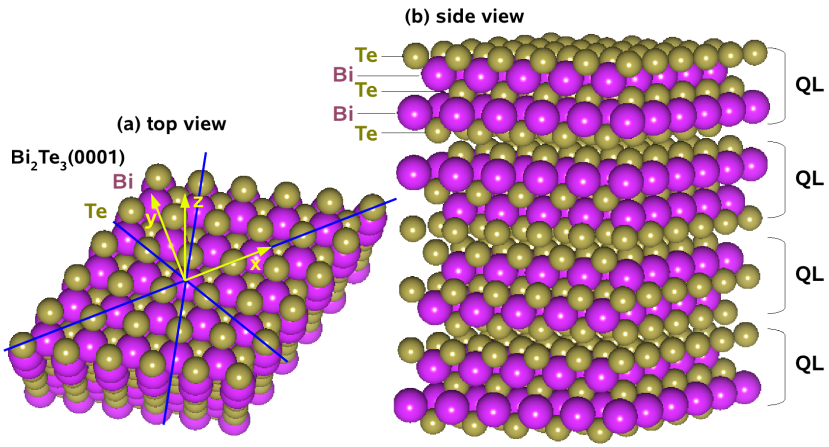
First-principles Green function calculations

Bloch spectral density

$$N_{i\alpha}(E,\mathbf{k}_{\parallel}) = -\frac{1}{\pi}\mathrm{Im}\ G^{+}_{ii}(E,\mathbf{k}_{\parallel})\Big|_{\alpha}$$

Spin structure as spin projections

$$S_{i\mu}(E,\mathbf{k}_{\parallel}) = N_{i\mu\uparrow}(E,\mathbf{k}_{\parallel}) - N_{i\mu\downarrow}(E,\mathbf{k}_{\parallel})$$



Model

L. Fu, PRL **103** (2009) 266801

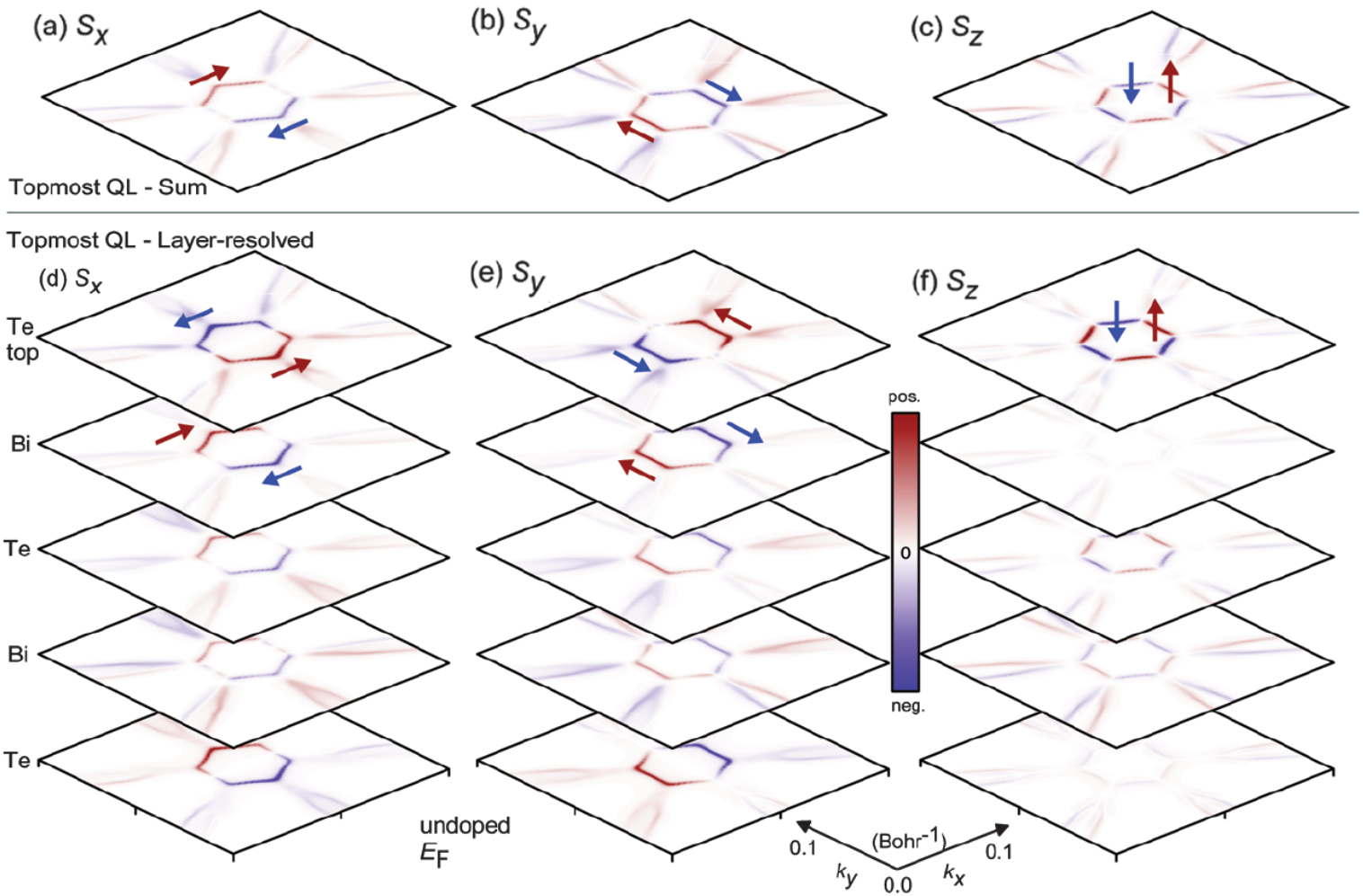
$$H(\vec{k}) = \boxed{E_0(\vec{k})} + \boxed{v_k\left(k_x\sigma_y - k_y\sigma_x\right)} + \boxed{\frac{\lambda}{2}\left(k_+^3 + k_-^3\right)\sigma_z} + \boxed{g\mu_B\vec{B}\cdot\vec{\sigma}}$$

$$k_{\pm} = k_x \pm 1k_y$$

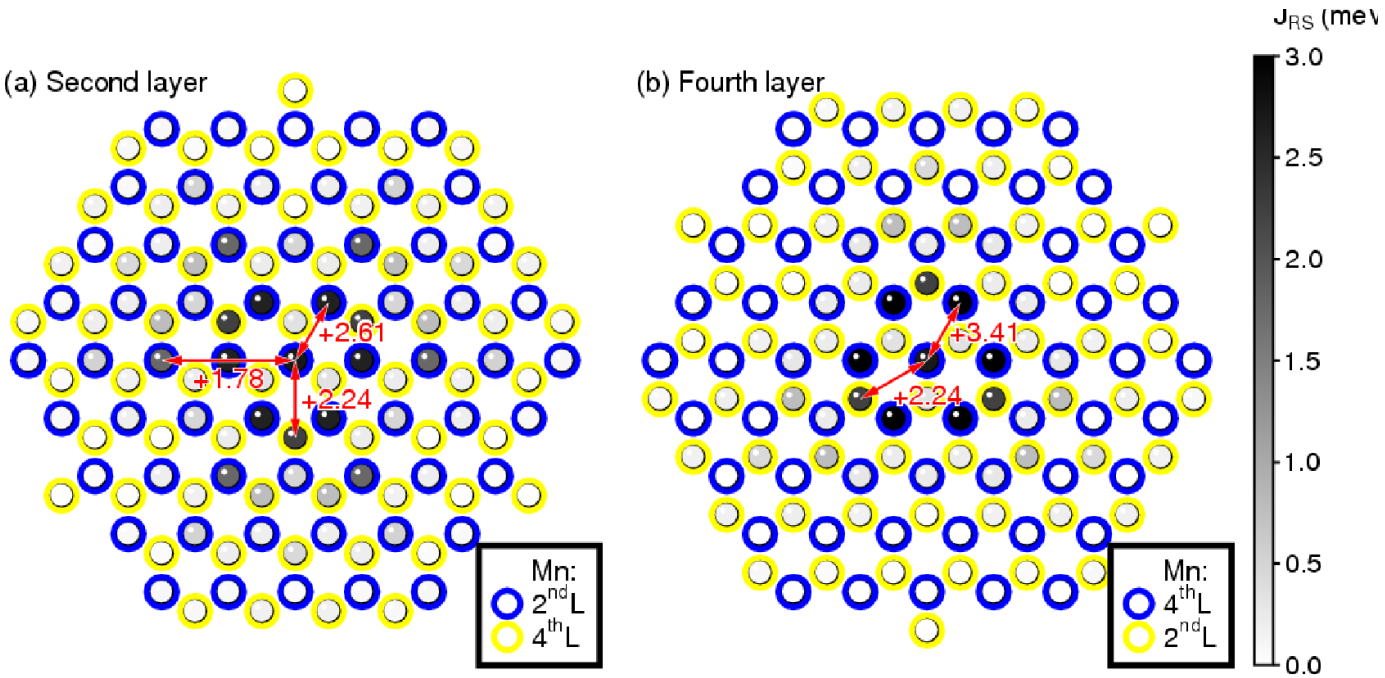
1. Particle-hole asymmetry
2. Rashba spin-orbit coupling
3. Warping
4. Magnetism

Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$

Layer-resolved spin texture of the Dirac state in the topmost QL of undoped Bi_2Te_3 at E_F

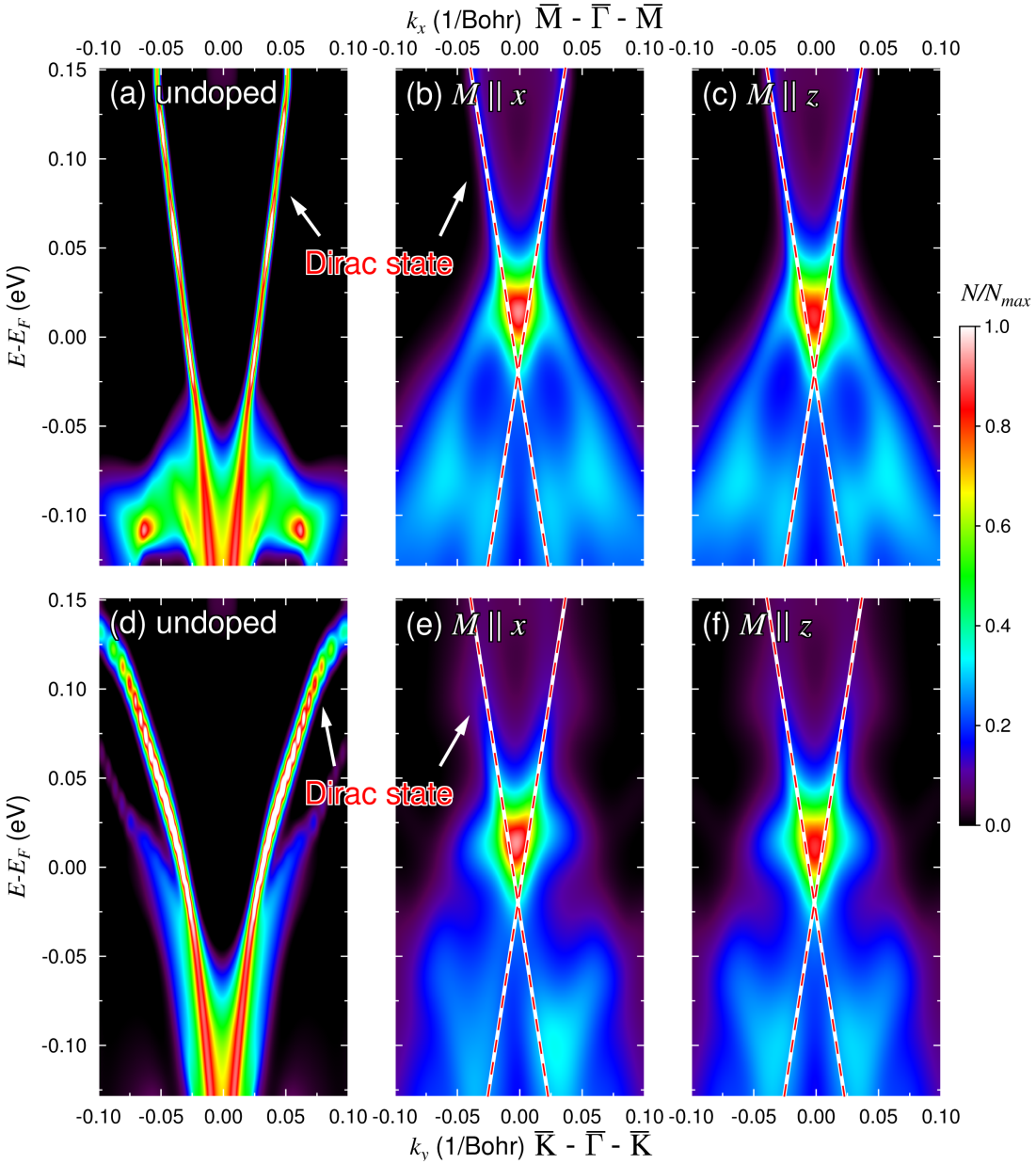


Exchange parameters J_{ij} in $Bi_{1.9}Mn_{0.1}Te_3$



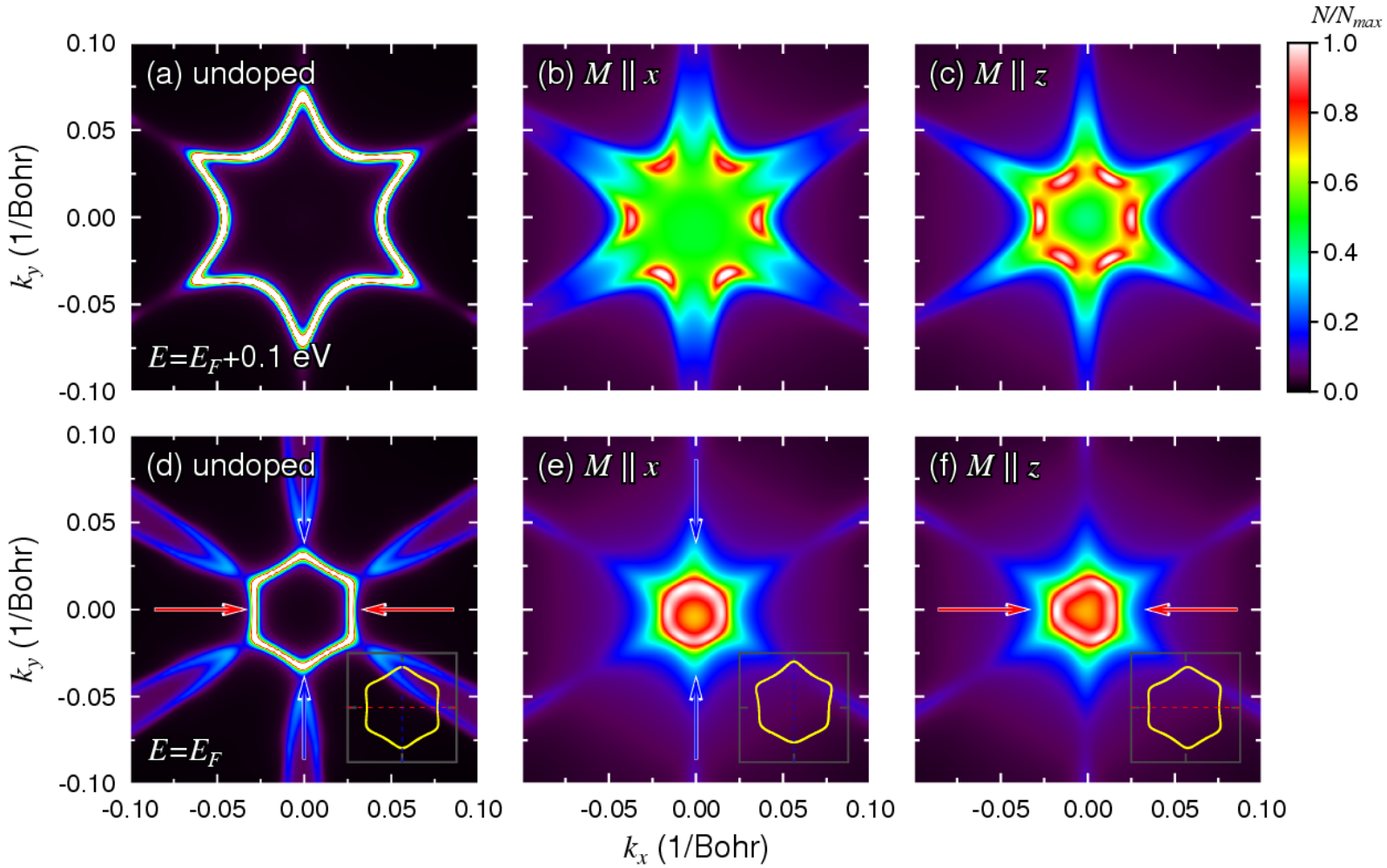
Estimated T_C=12 K, T_C^{exp}=15 K

Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$



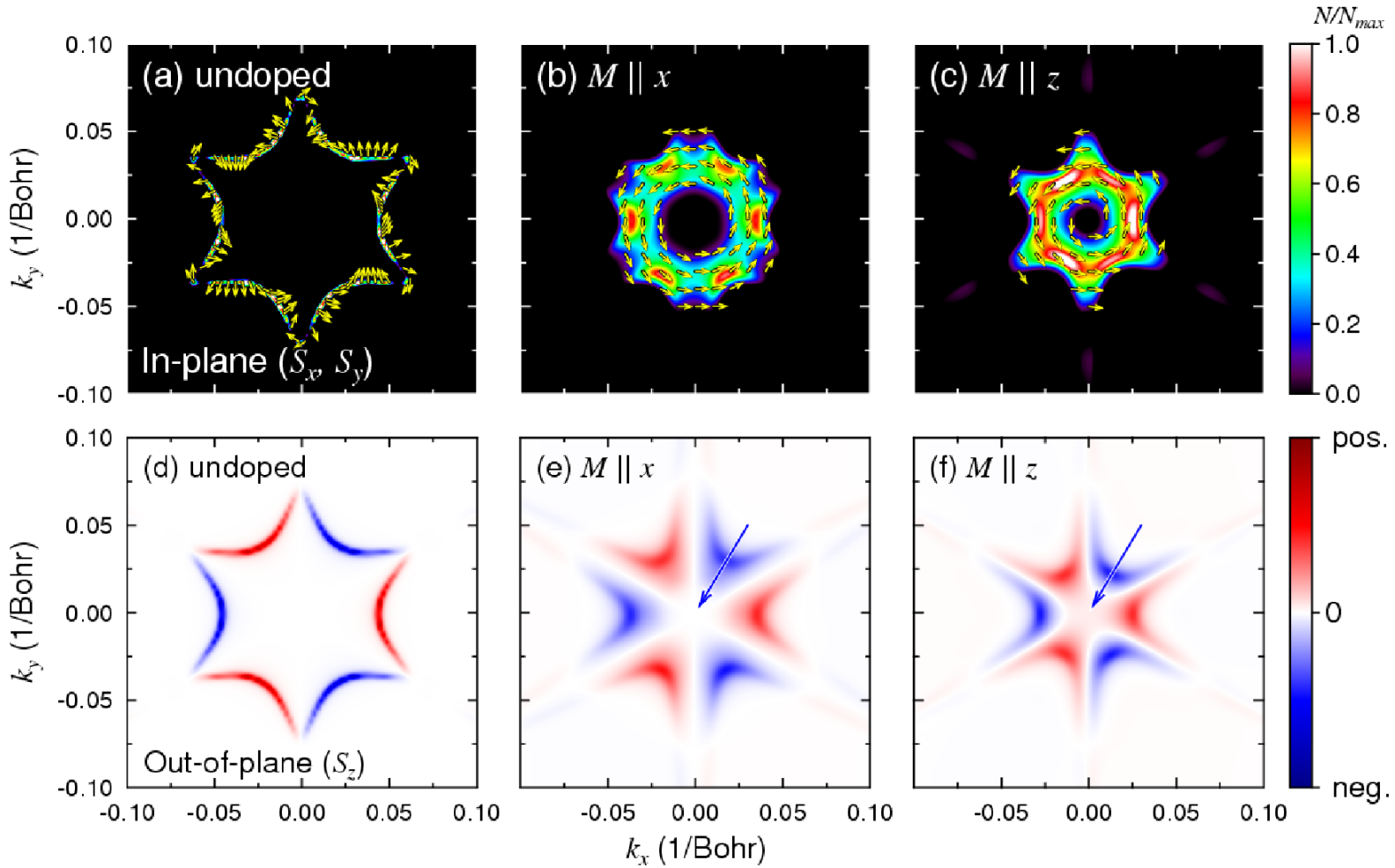
Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$

Constant-energy contours of the Dirac-state dispersion



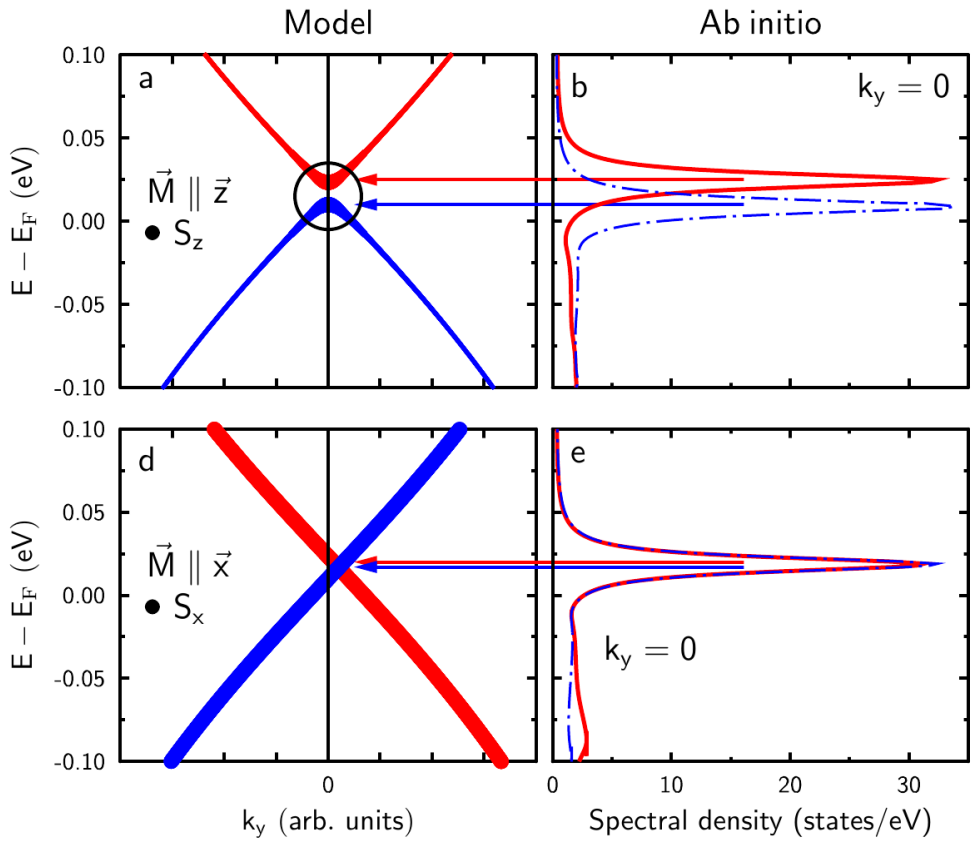
Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$

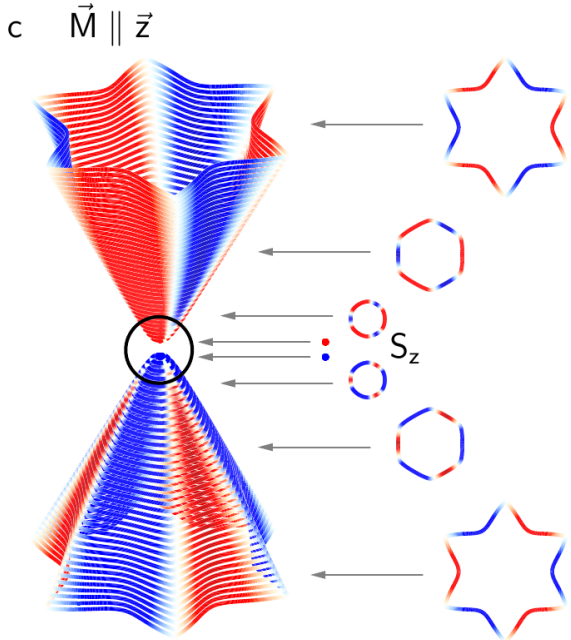
Spin texture of the Dirac state in a topmost Te site at $E_F + 0.1eV$



Opening the bang gap

Model of Fu, PRL (2009): $\hat{H} \equiv \hat{H}_{ph} + \hat{H}_{soc} + \hat{H}_{warp} + \hat{H}_{magn}$





Topological character of the Dirac state in $Bi_{2-x}Mn_xTe_3$

Short summary

Undoped Bi_2Te_3

- Spin reversal in the Te top layer
- Spin vortices at the cusps of the constant-energy contours
- Out-of-plane spin only in the Te top layer

$Bi_{2-x}Mn_xTe_3$

- Weak or blurred spectral weight
- Pronounced dispersion minimum (Dirac point)
- Agreement with photoemission experiments
- Band gap opens for $\mathbf{M} \parallel \mathbf{z}$

J. Henk, A. Ernst, S. V. Eremeev, E. V. Chulkov, I. V. Maznichenko, and I. Mertig, PRL 108, 206801 (2012)

J. Henk, M. Flieger, I. V. Maznichenko, I. Mertig, A. Ernst, S. V. Eremeev, and E. V. Chulkov, PRL 109, 076801 (2012)

Exchange interaction in binary diluted chalcogenides

Motivation

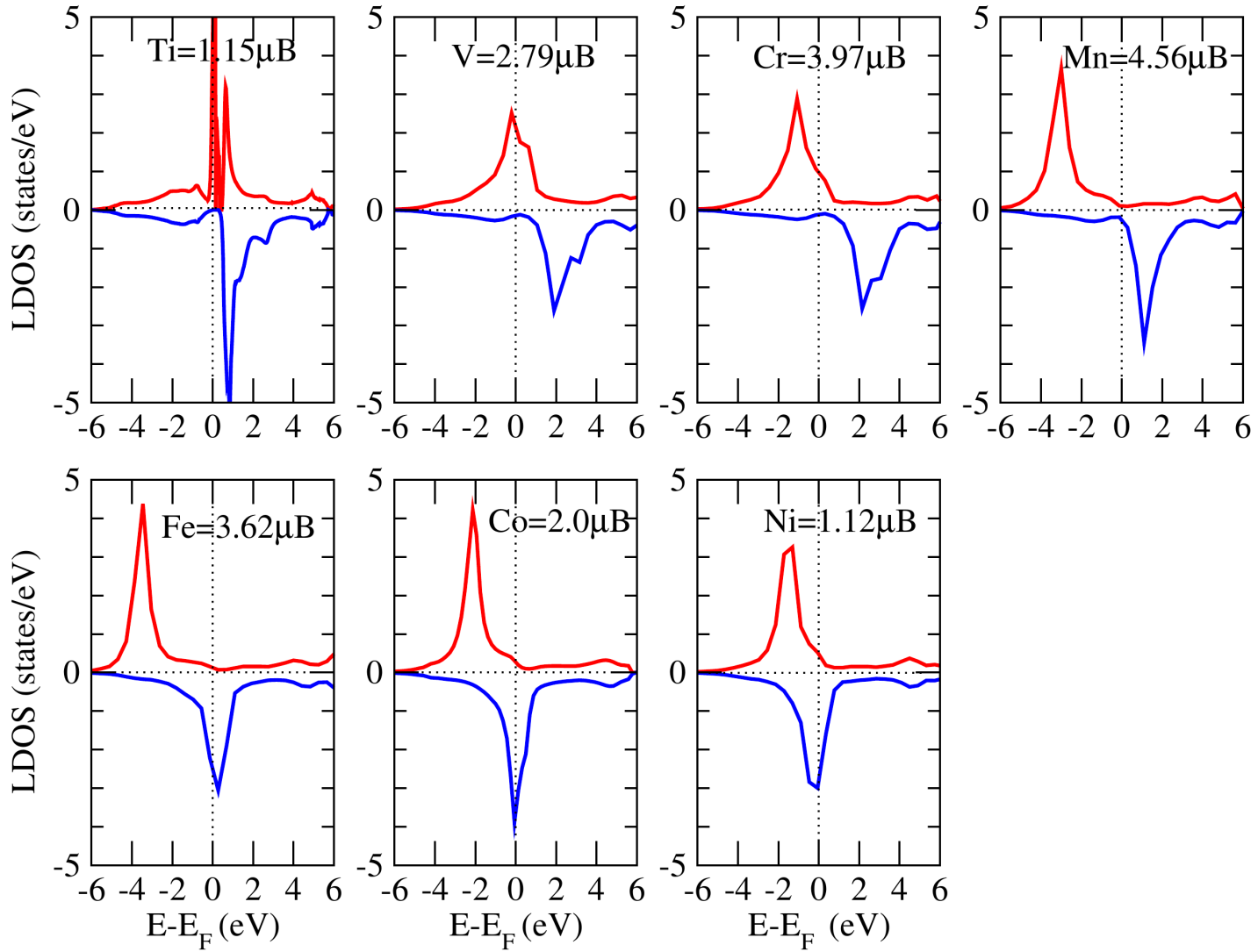
- Types of magnetic interaction binary diluted chalcogenides
- Impact of doping
- Magnetic order
- Critical temperature
- Tuning the exchange interaction

Experiment

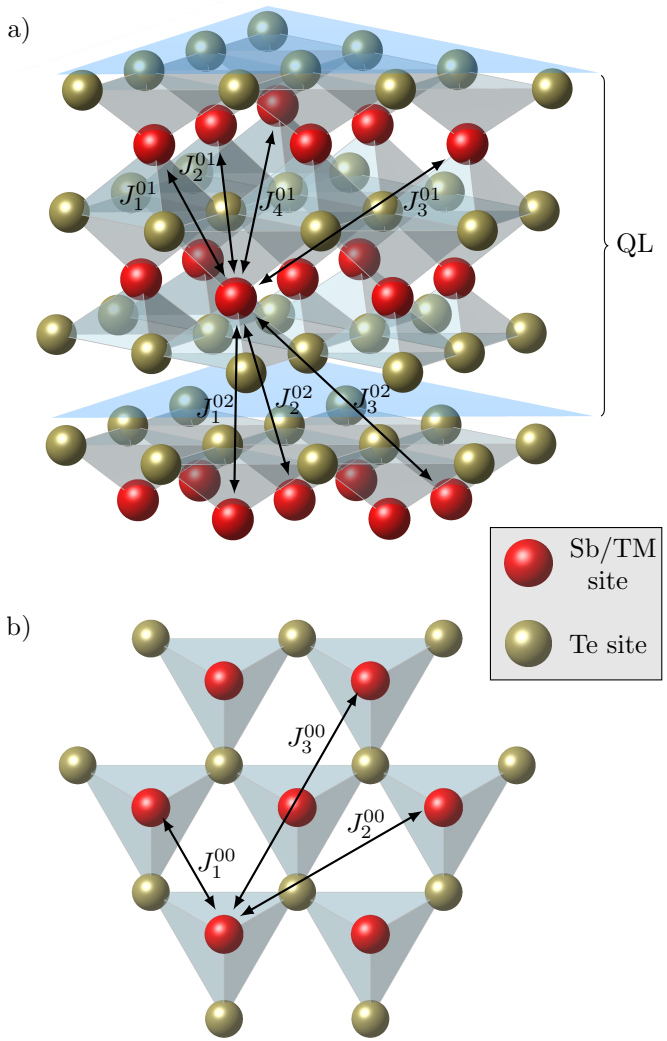
- Single crystals: $x < 0.1$
- MBE films: $x < 0.6$
- Mn doping : ferromagnetic with a very low T_C or spin glass
- Fe, Co doping : antiferromagnetic or paramagnetic
- Cr, V : ferromagnetic
- MBE $Sb_{2-x}Cr_xTe_3$ films: T_C=190 K at $x = 0.6$

Exchange interaction in binary diluted chalcogenides

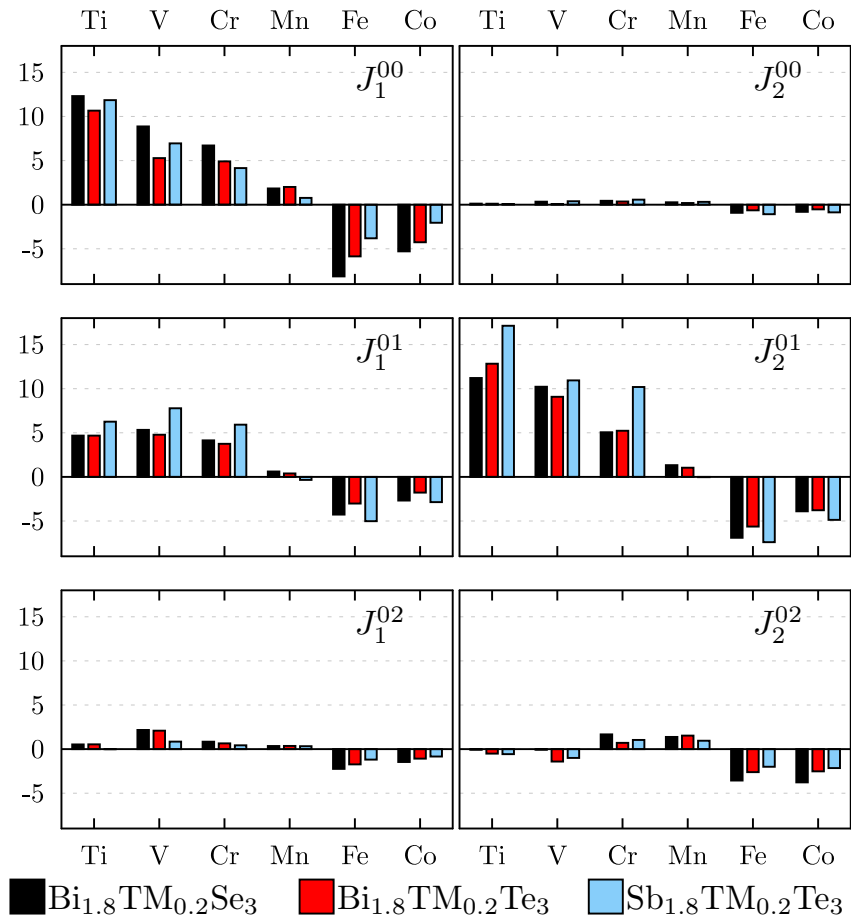
Density of states of $Sb_{1.9}M_{0.1}Te_3$ (M = Ti, V, Cr, Mn, Fe, Co, Ni)



Exchange interaction in binary diluted chalcogenides



Scheme of the exchange coupling in $Sb_{2-x}M_xTe_3$



Exchange constants of various magnetic chalcogenides

Exchange interaction in binary diluted chalcogenides

Identified interactions

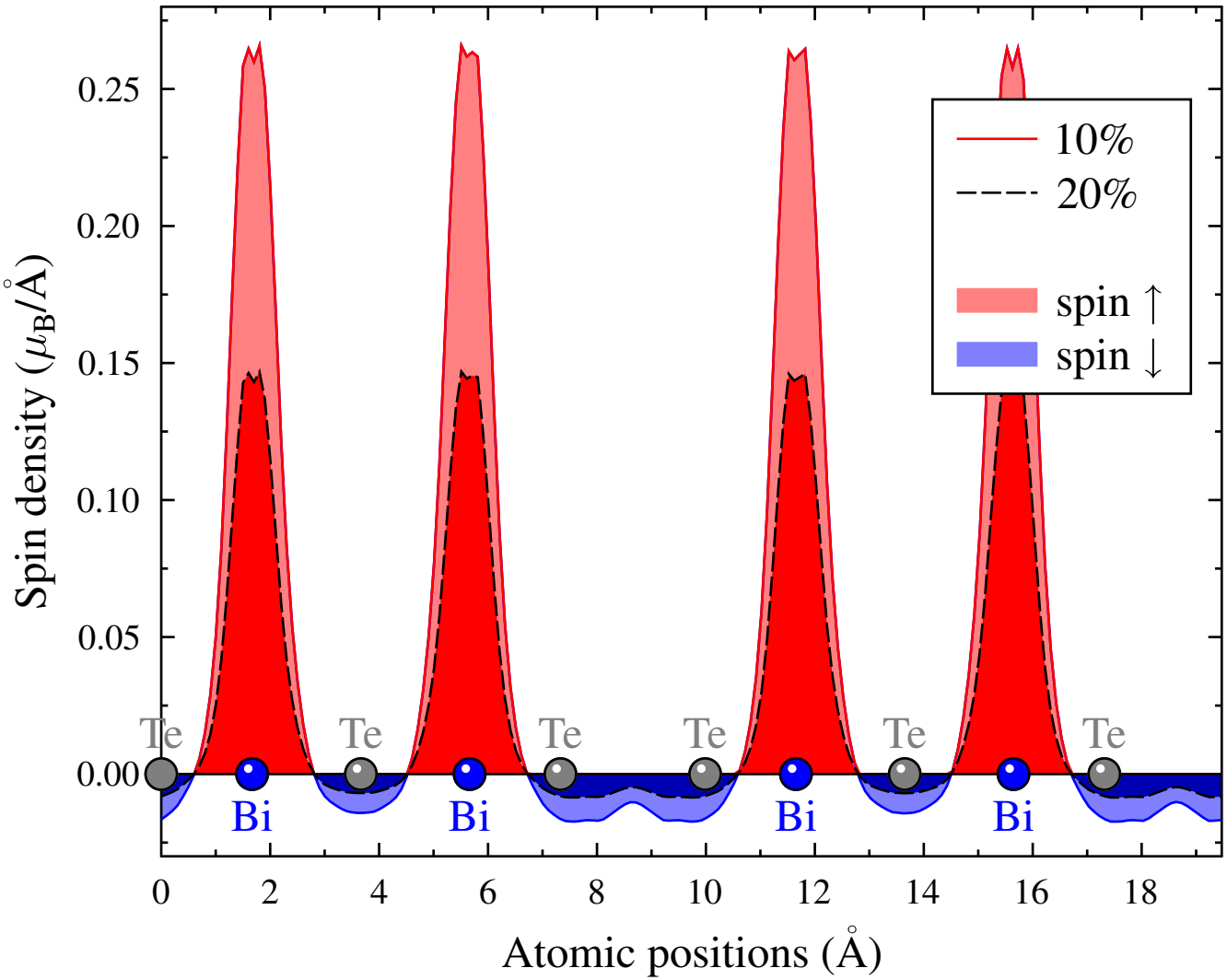
- Indirect exchange interaction of Zener type within the cation layers
- Double or super exchange over the anion layers

Main features:

- Decreases with the increase of number of $3d$ electrons
- Long-range
- There is interaction over van der Waals gap

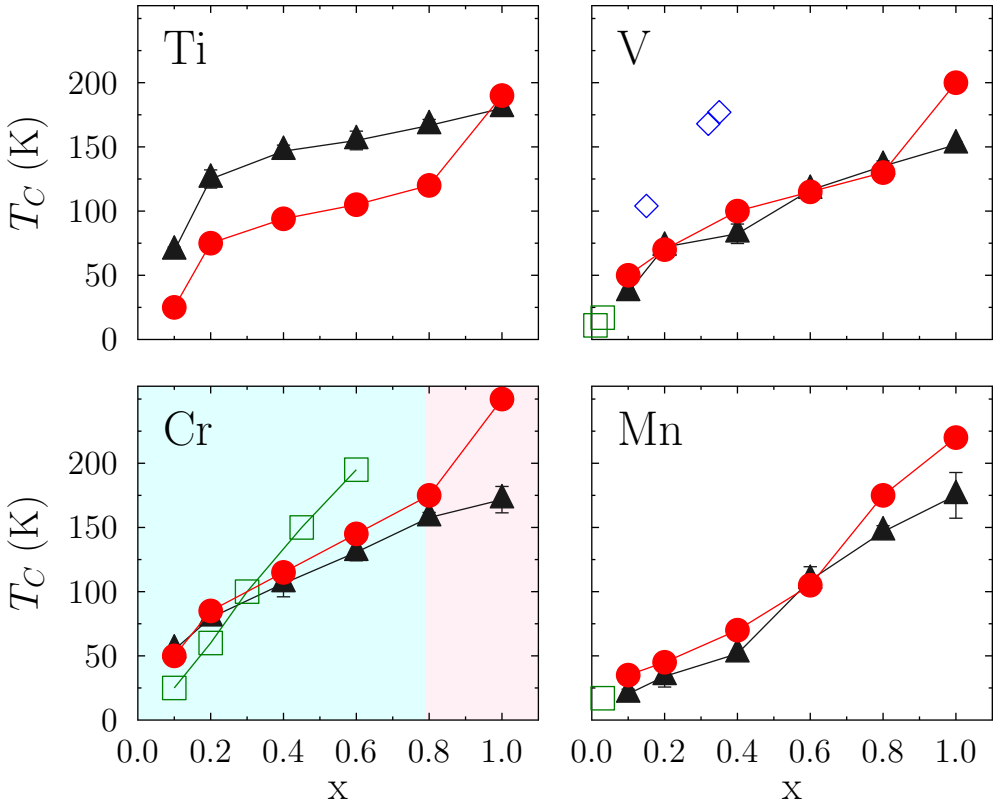
Exchange interaction in binary diluted chalcogenides

Spin density in z direction integrated over the xy plane in $Sb_{2-x}Cr_xTe_3$



Exchange interaction in binary diluted chalcogenides

Theoretical and experimental critical temperatures in Sb2-x$M_x$$Te_3$

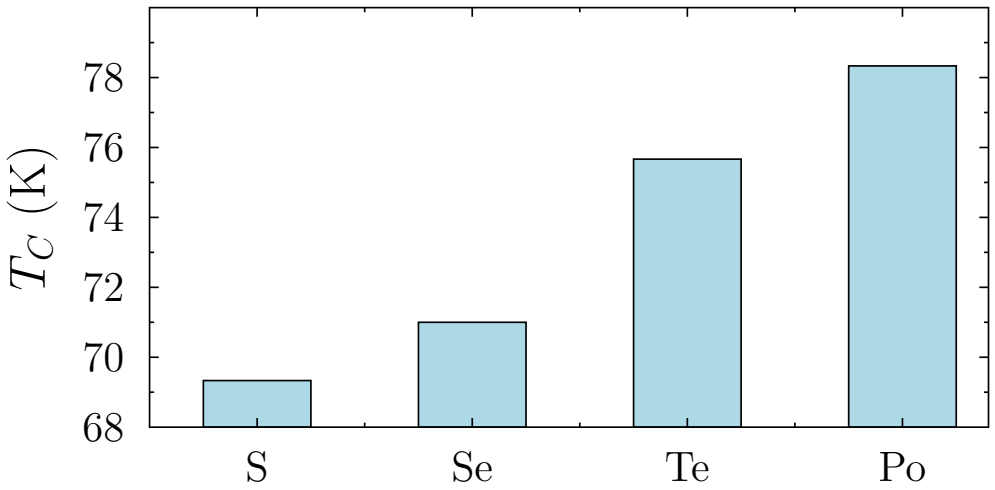


Exchange interaction in binary diluted chalcogenides

Tuning the exchange interactions

- Replacing cations: tuning the Zener exchange interaction
- Replacing anions: tuning the double exchange interaction
- Doping van der Waals gap

Example: Replacing the middle anion layer in $Sb_{1.9}Cr_{0.1}Te_3$



Exchange interaction in binary diluted chalcogenides

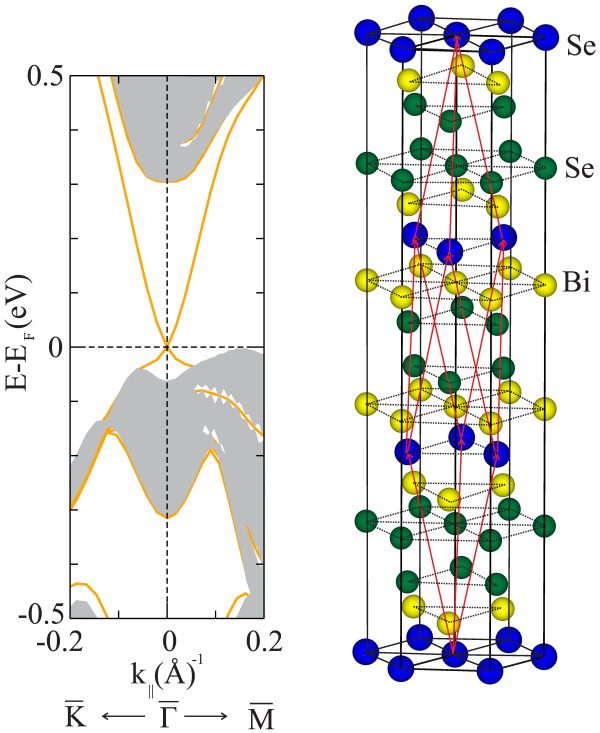
Short summary

- Two type of the interaction are identified
  - Zener exchange interaction via free electrons within a cation layer
  - Double or super exchange via a anion layer
- The interaction are long-range
- T_C is in a good agreement with experiment
- The interaction can be tuned

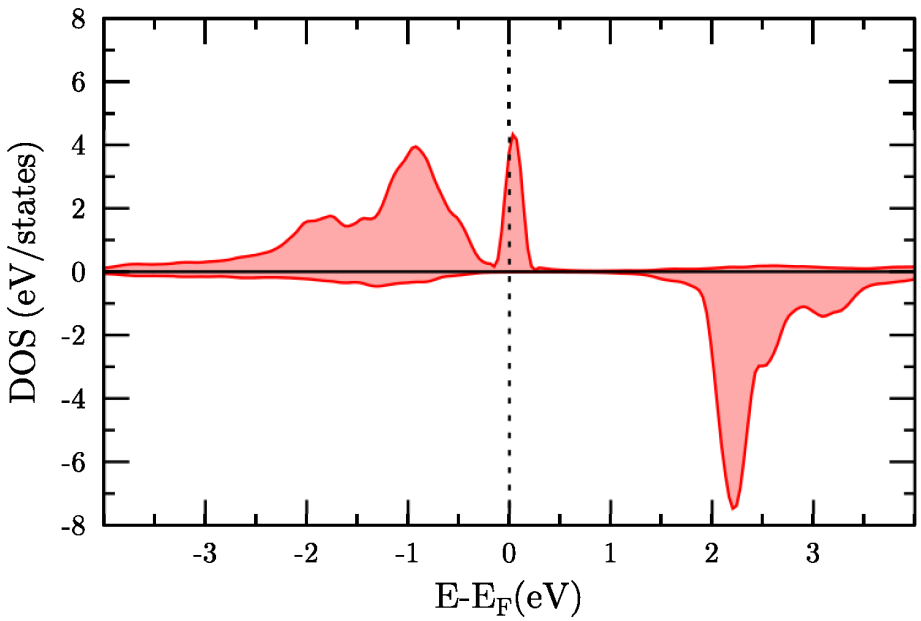
Impact of electron-magnon interaction on the Dirac state

Motivation

- Magnetic impurities on TI surfaces: low concentration limit
- Surface magnons
- Change of the band structure due to electron-magnon interaction



pure Bi_2Se_3 surface



Cr impurity in Bi_2Se_3 surface

Impact of electron-magnon interaction on the Dirac state

Model

- Hamiltonian of free electrons

$$\mathcal{H}_0 = -iv\left(\sigma_x\partial_x + \sigma_y\partial_y\right)$$

- Coupling of surface electrons to the spin density distribution $S(\mathbf{r})$

$$\mathcal{H}_{int} = \frac{\lambda_I}{2}\left[\sigma_+ S_-(\mathbf{r}) + \sigma_- S_+(\mathbf{r})\right] + \lambda_t \sigma_z S_z(\mathbf{r}),$$

- Magnonic Green function

$$D_{+-}(\mathbf{q},\omega) = \frac{S_z}{\omega + S_z[V(\mathbf{q}) - V(0)]},$$

Impact of electron-magnon interaction on the Dirac state

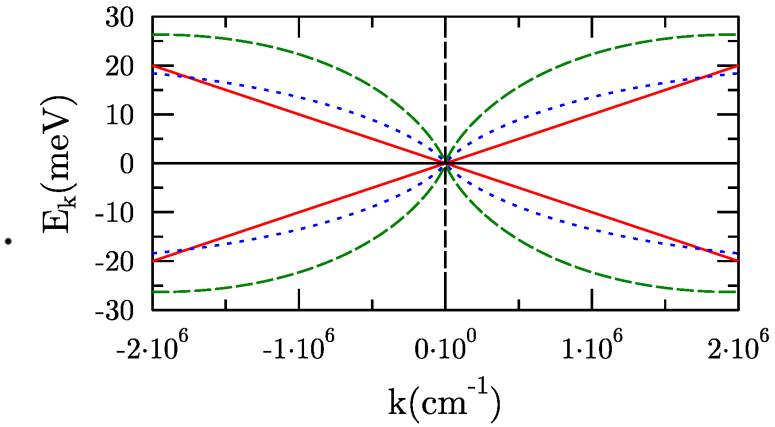
Model

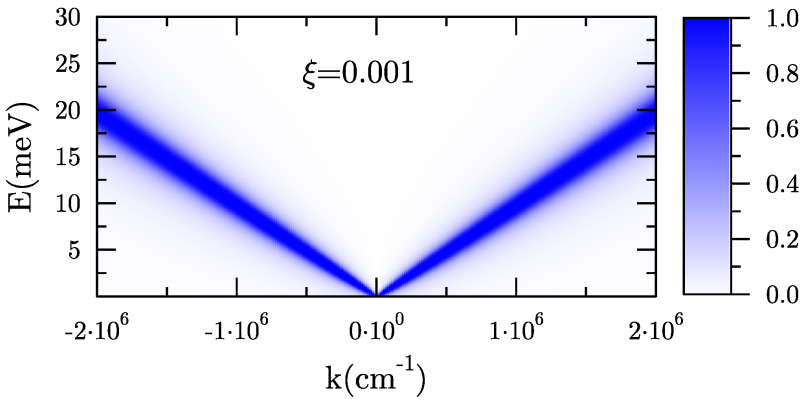
- Energy of the interaction between two moments

$$E_{ij}^{int}(\mathbf{r}-\mathbf{r}') = S_i(\mathbf{r})\, S_j(\mathbf{r}')\, \chi_{ij}(\mathbf{r}-\mathbf{r}'),$$

- Electron self-energy related to the coupling to magnons

$$\Sigma(\mathbf{k},\varepsilon) = i\lambda_t^2 \int \frac{d\omega}{2\pi} \frac{d^2\mathbf{q}}{(2\pi)^2}\, \sigma_-\, G_0(\mathbf{k}-\mathbf{q},\varepsilon-\omega)\, \sigma_+ D_{+-}(\mathbf{q},\omega)$$





Band structure and spectral function for various interaction strengths

Impact of electron-magnon interaction on the Dirac state

Short summary

- Indirect coupling of the magnetic impurities results in a ferromagnetic ordering
- Magnetization is out-of-plane
- Interaction between magnons and the surface state renormalizes the electron energy spectrum
- Electron velocity near the Dirac point depends on the electron-magnon coupling

First-principles design of topological insulators

Summary

- We are able to describe electronic structure of TI
- Computational design of TI is possible
- Dirac surface state can survey under an applied magnetic field
- TI can be ferromagnetic using a specific doping
- The interaction between the magnons and electrons can modify the band structure

Outlook

- Further investigations of defects and impurities in TI
- Study of transport properties
- Study of TI under an applied electric field

Team

MPI Halle

- Maia Vergniory
- Matthias Geilhufe
- Martin Hoffmann
- Xabi Zubizarreta

University of Halle

- Jürgen Henk
- Igor Maznichenko
- Markus Flieger
- Ingrid Mertig
- Levan Chotorlishvili
- Vitalii Dugaev

DIPC, San-Sebastian

- Sergey Eremeev
- Mikhail Otrokov
- Eugenie Chulkov