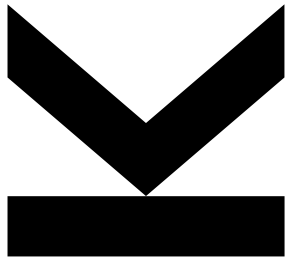


A stride down the quantum materials roadmap



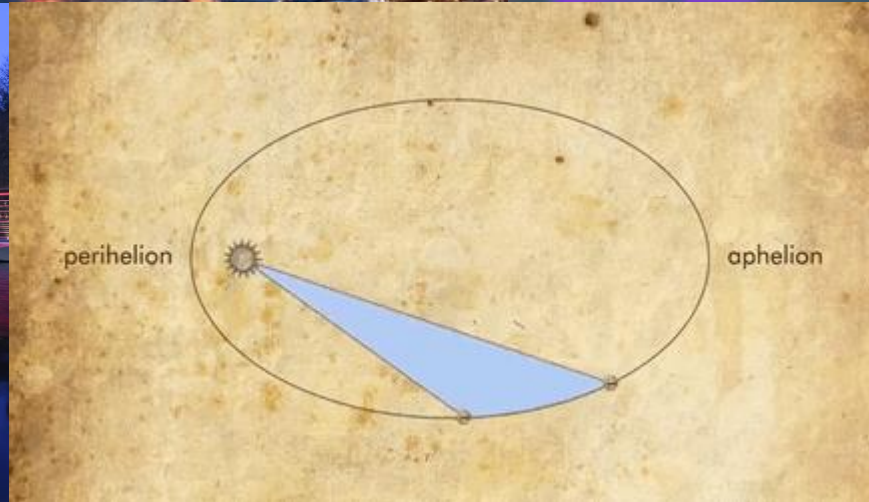
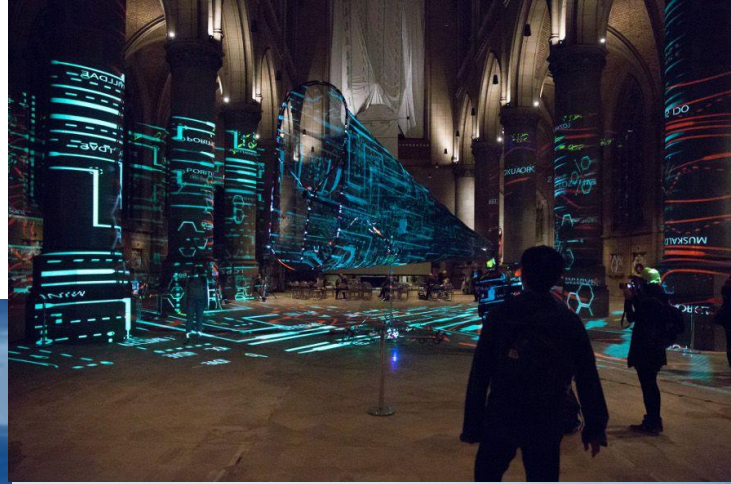
Alberta Bonanni

Institute for Semiconductor and Solid State Physics
Johannes Kepler University, Linz-Austria



Linz - Austria

JKU
JOHANNES KEPLER
UNIVERSITÄT LINZ



The logo for the IMAG Quantum Materials Group is located in the top right corner. It features a stylized white wireframe cat silhouette with a downward-pointing arrow on its left and an upward-pointing arrow on its right. To the right of the cat is the text "IMAG" in a large, bold, white sans-serif font. Below "IMAG" is the text "Quantum Materials Group" in a smaller, white sans-serif font.

IMAG
















Quantum Materials Group



Journal of Physics: Materials

ROADMAP

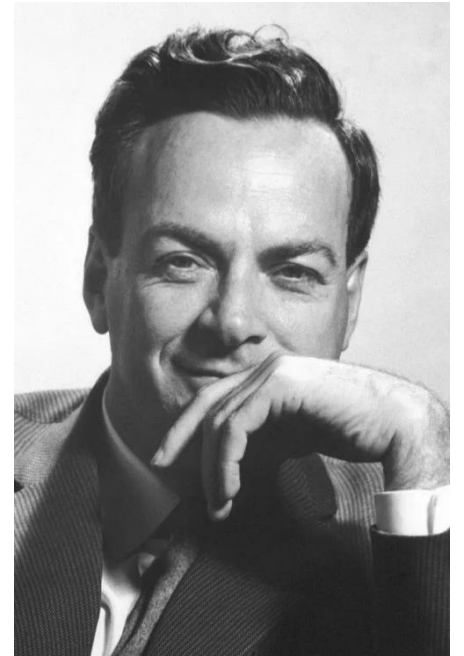
The 2021 (quantum) materials roadmap

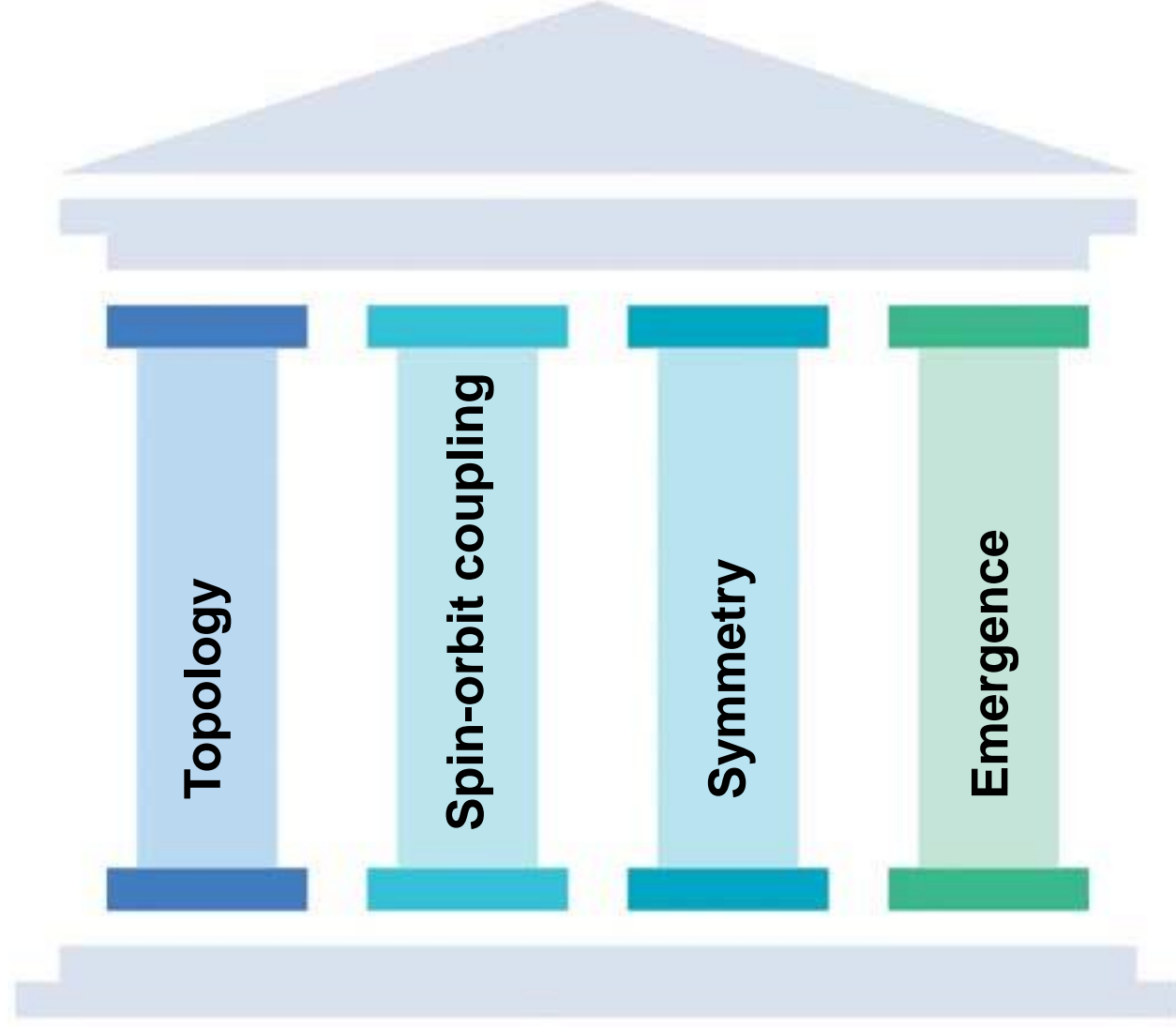
Feliciano Giustino^{1,2} , Jin Hong Lee³, Felix Trier³ , Manuel Bibes³ ,
Stephen M Winter⁴, Roser Valentí⁴ , Young-Woo Son⁵, Louis Taillefer^{6,7}, Christoph Heil⁸ ,
Adriana I Figueroa⁹ , Bernard Plaçais¹⁰ , QuanSheng Wu¹¹, Oleg V Yazyev¹¹ , Erik P A M Bakkers¹²,
Jesper Nygård¹³, Pol Forn-Díaz^{14,15}, Silvano De Franceschi¹⁶, J W McIver¹⁷ , L E F Foa Torres¹⁸ ,
Tony Low¹⁹, Anshuman Kumar²⁰, Regina Galceran⁹ , Sergio O Valenzuela^{9,21}, Marius V Costache⁹ ,
Aurélien Manchon²², Eun-Ah Kim²³ , Gabriel R Schleder^{24,25} , Adalberto Fazzio^{24,25} and
Stephan Roche^{9,21} 

What are quantum materials?

- Nature isn't classical and if you want to make a simulation of nature, you'd better make it quantum mechanical and it's a wonderful problem, because it doesn't look so easy

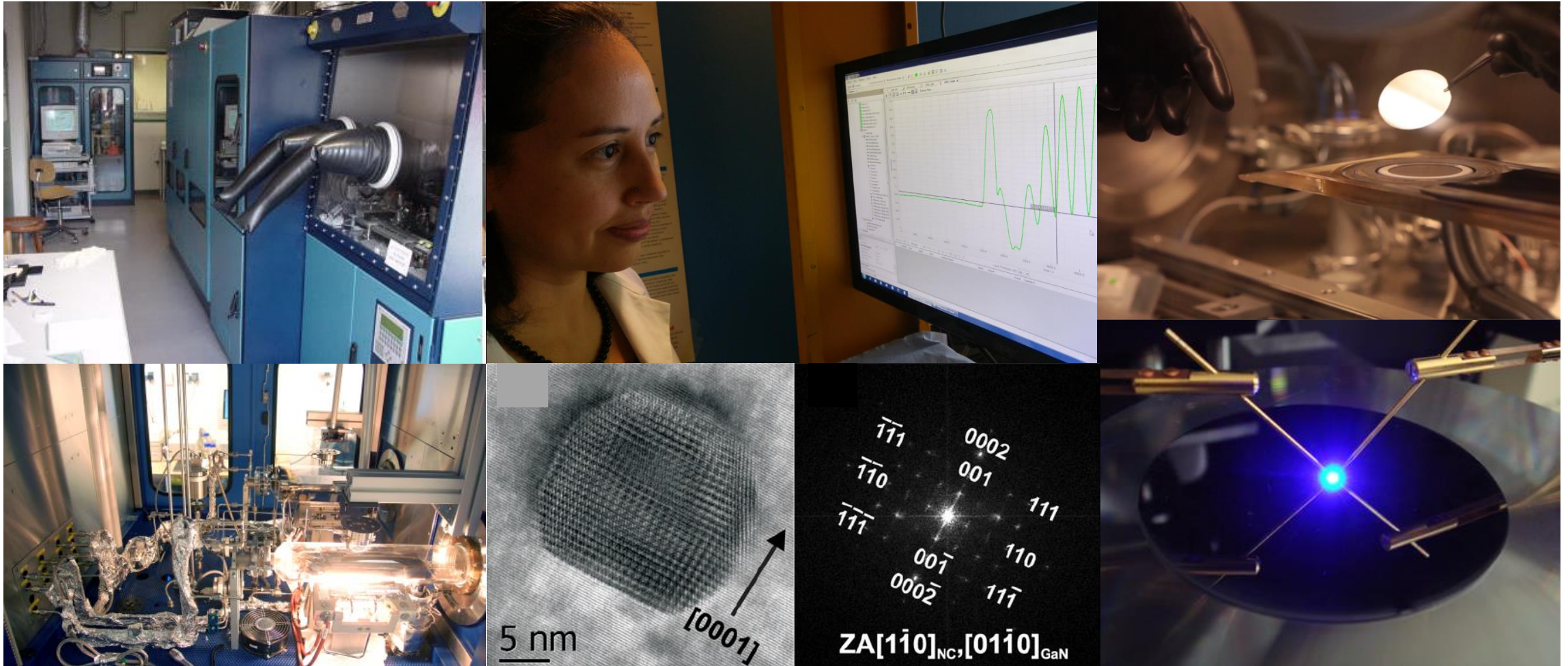
Richard Feynman 1981



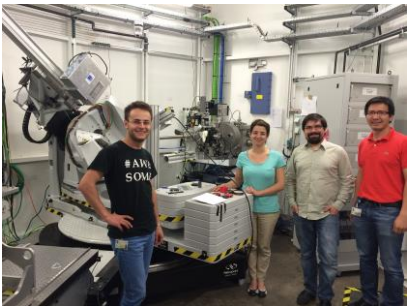
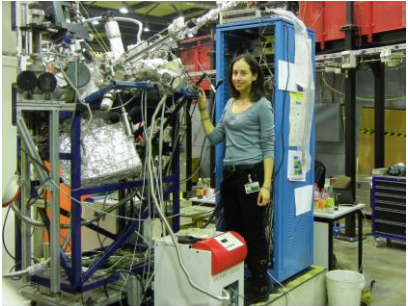
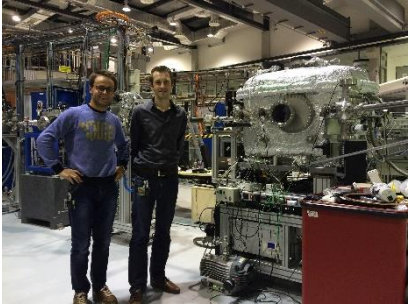




Long tradition: crystal growth and extended characterization



Characterization - synchrotrons



ESRF, Grenoble - France



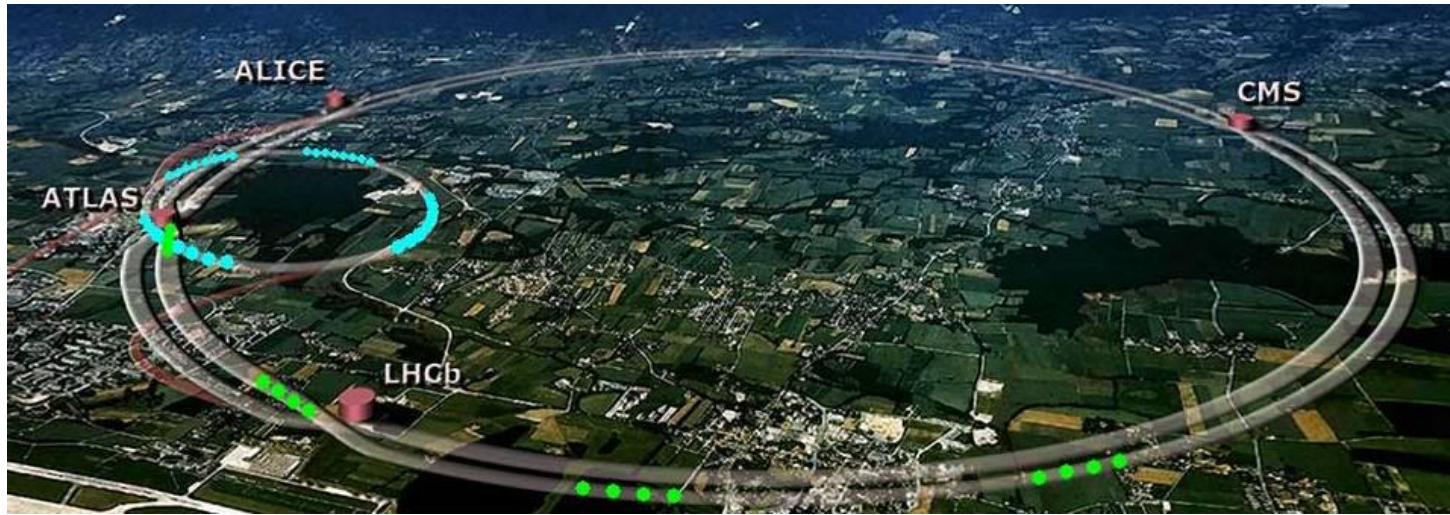
MaxLab, Lund - Sweden



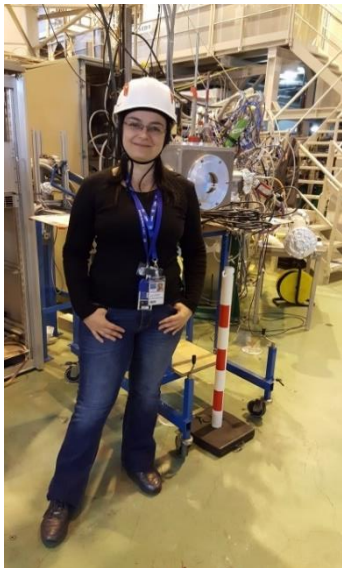
Elettra, Trieste - Italy



BESSY, Berlin - Germany



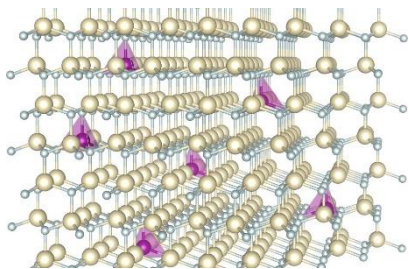
ISOLDE-CERN: Mössbauer, channeling



(Ga,Mn)N

[x up to 6% in MOVPE and up to 10% in MBE]

without band carriers



DMS → (Ga,Mn)N spin filters,
resonant structures

W. Stefanowicz,...AB, Phys.Rev.B **81**, 125210 [2010]

J. Suffczyński,...AB, Phys.Rev.B **83**, 235210 [2011]

M. Sawicki,...AB, Phys.Rev.B **85**, 205204 [2012]

G. Kunert,...AB,...Appl.Phys.Lett. **101**, 0224213 [2012]

S. Stefanowicz,...AB,... Phys.Rev.B **88**, 081201(R) [2013]

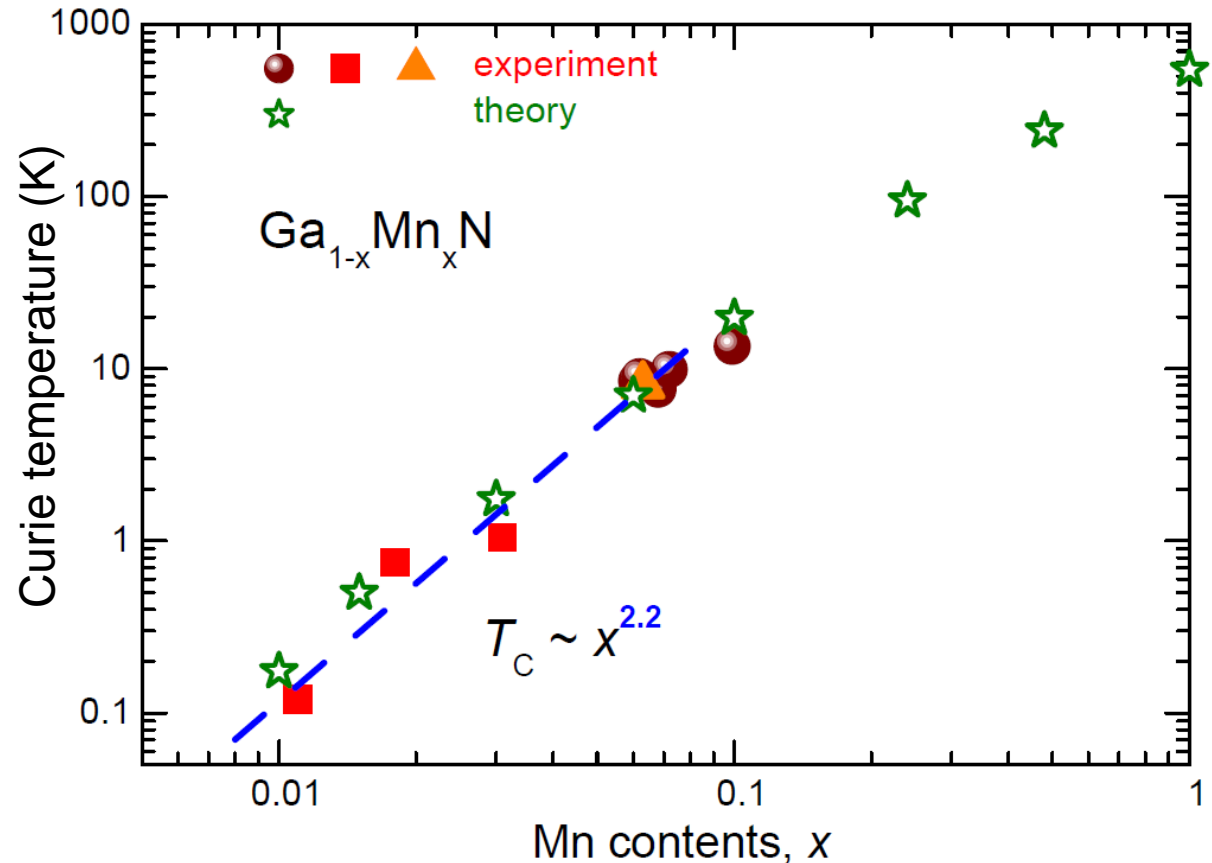
R. Adhikari,...AB, Phys.Rev.B **91**, 205204 [2015]

T. Dietl,...AB,..., Rev.Mod.Phys **87**, 1311 [2015]

D. Sztenkiel, ...AB,..., Nat.Comm. **7**, 13232 [2016]

(Ga,Mn)N – T_C – Experiment vs. TBA + MonteCarlo

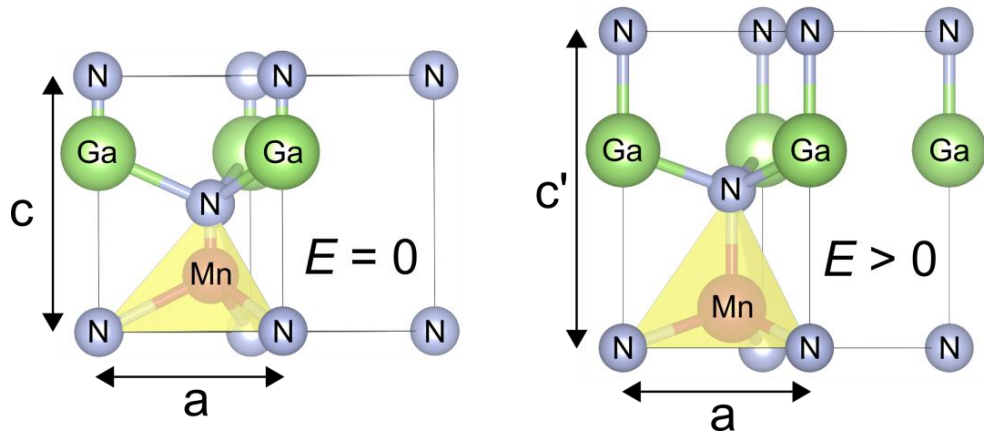
- Ferromagnetic interaction
- Character of $sp-d$ hybridization
→ scaling dependence
- Monte Carlo with exchange integrals from tight-binding
- FM superexchange



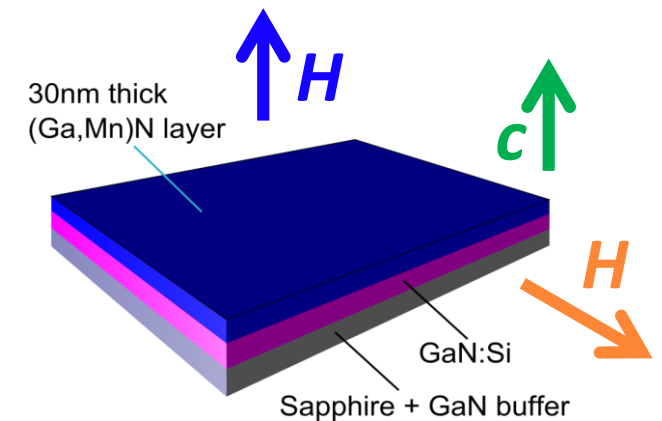
M. Sawicki,...AB, Phys.Rev.B **85**, 205204 [2012]

S. Stefanowicz,...AB,... Phys.Rev.B **88**, 081201(R) [2013]

Electric field modulation of magnetism in (Ga,Mn)N



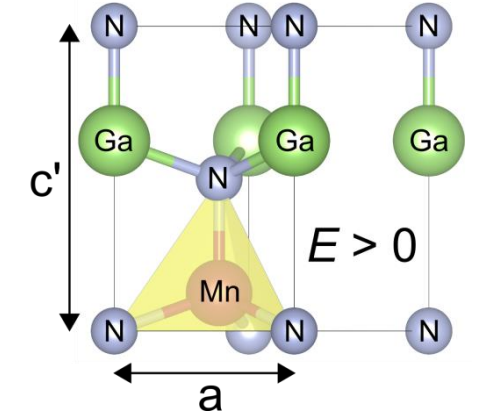
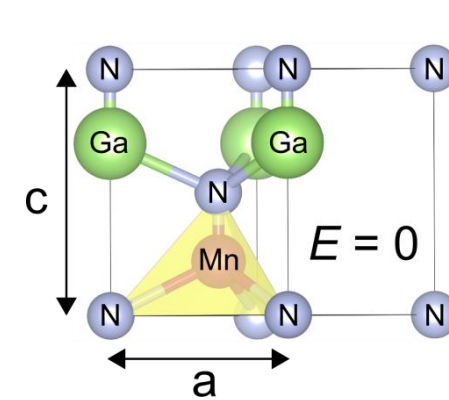
D. Sztenkiel, ...AB,... Nat. Commun. **7**, 13232 [2016]



Magnetic anisotropy in (Ga,Mn)N

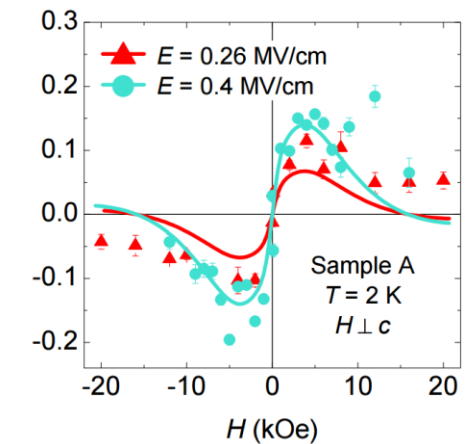
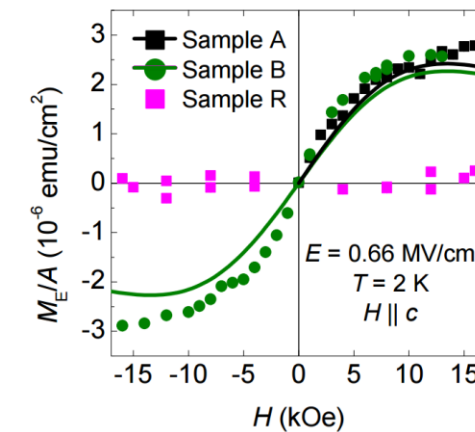
$$H = H_{CF} + H_{JT} + H_{TR} + H_{SO} + H_Z$$

↓ Tetrahedral crystal field ↓ Zeeman splitting
↓ Jahn-Teller splitting ↓ Spin-orbit coupling
↓ Trigonal distortion



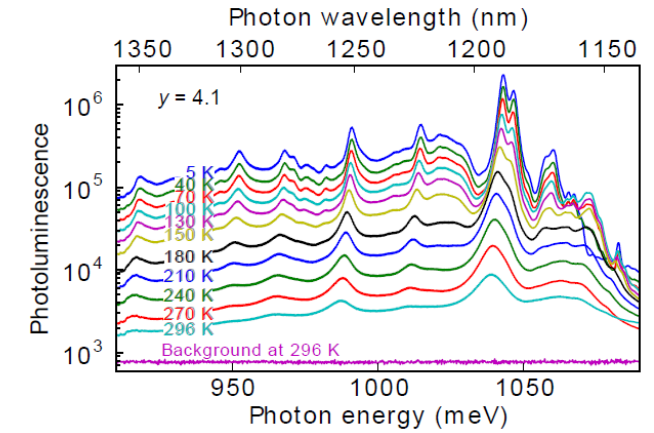
$$H_{TR} \propto \xi = \frac{c}{a} - \sqrt{\frac{8}{3}}$$

$$\text{In (Ga,Mn)N} \rightarrow \xi < 0 \rightarrow M(H \perp c) > M(H \parallel c)$$



W. Stefanowicz, ...AB, Phys.Rev.B **81**, 125210 [2010]

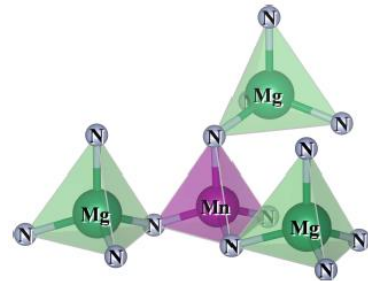
(Ga,Mn)N:Mg



complexes → Centers for solotronics and quantum computing

complexes → Photonics:
[(Al)Ga,Mn]N:Mg-based infrared lasers

Optimized DUV LEDs



T. Devillers,...,AB, Sci. Rep. **2**, 722 [2012]

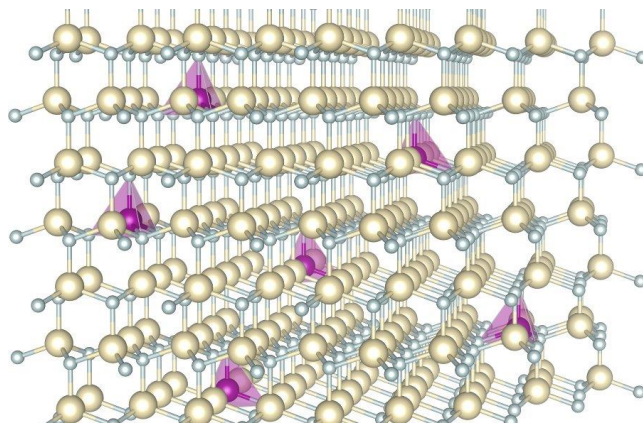
T. Devillers,...,AB, Appl. Phys. Lett. **103**, 211909 [2013]

G. Capuzzo,...,AB, Sci. Rep. **7**, 426972 [2017]

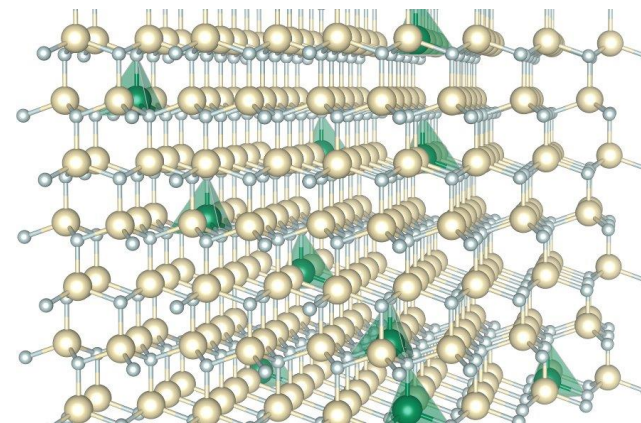
D. Kysylychyn,...,AB, Phys. Rev. B **97**, 245311 [2018]

A. Nikolenko,...,AB, Crystals **9**, 235 [2019]

(Ga,Mn)N:Mg – MnMg_k complexes

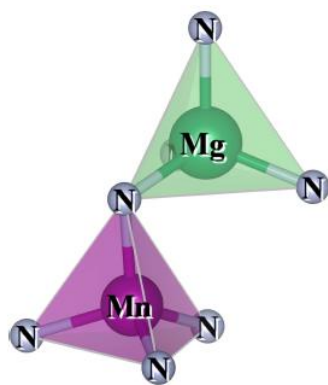


+

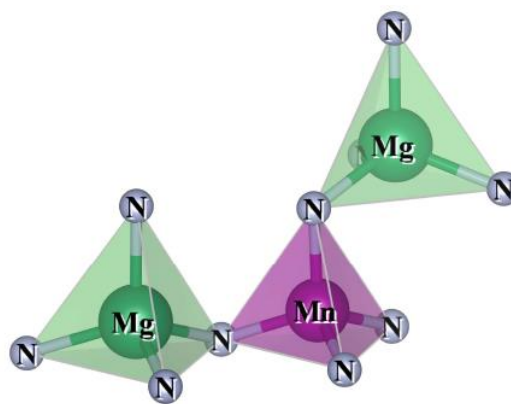


(Ga,Mn)N

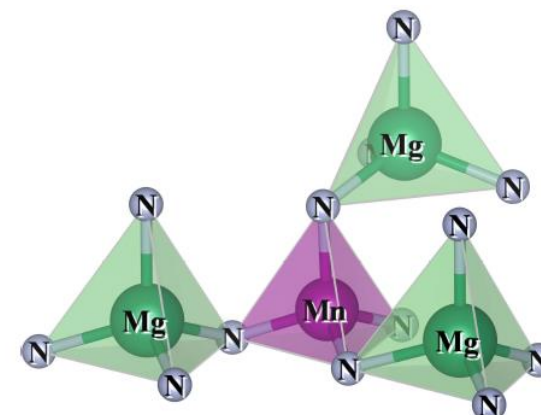
(Ga,Mg)N



MnMg (-0.51eV)

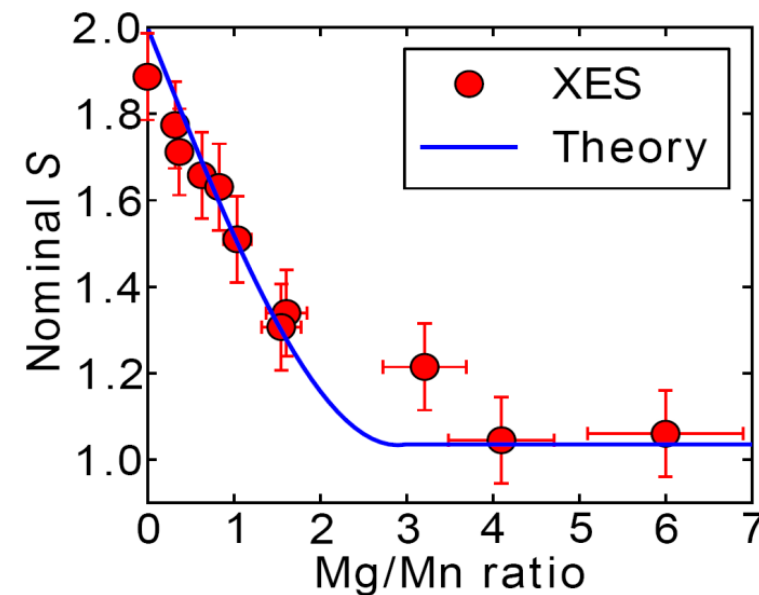
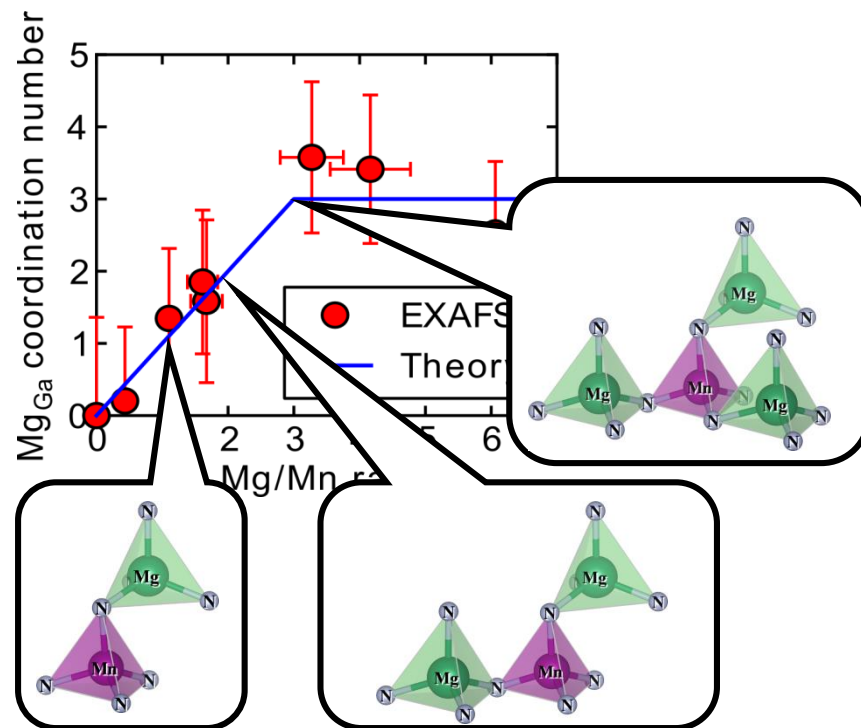


MnMg₂ (-0.58eV)



MnMg₃ (-0.21eV)

(Ga,Mn)N:Mg – control over charge and spin state

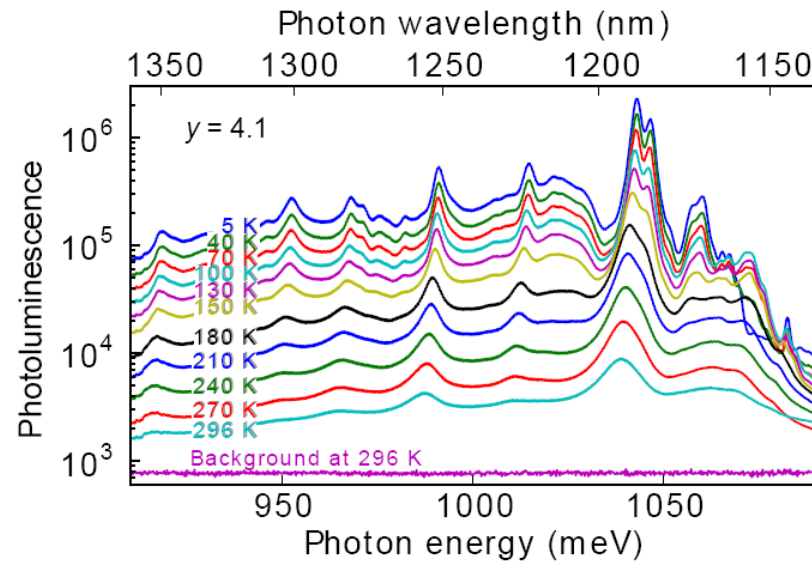
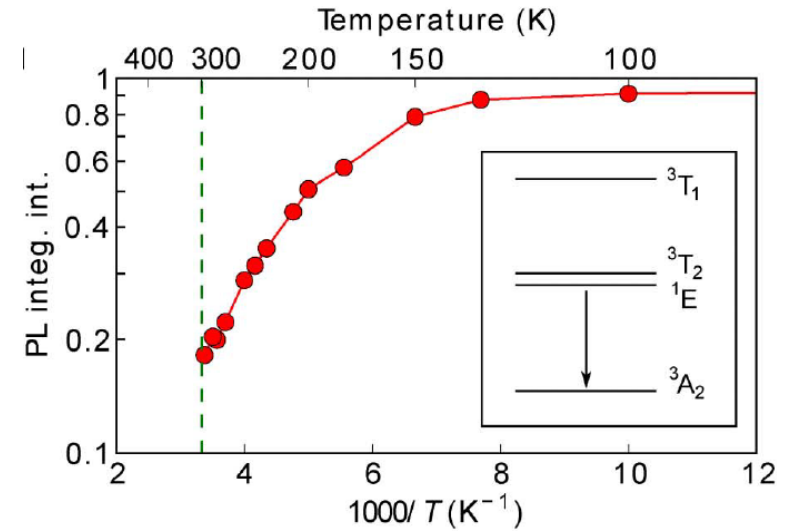
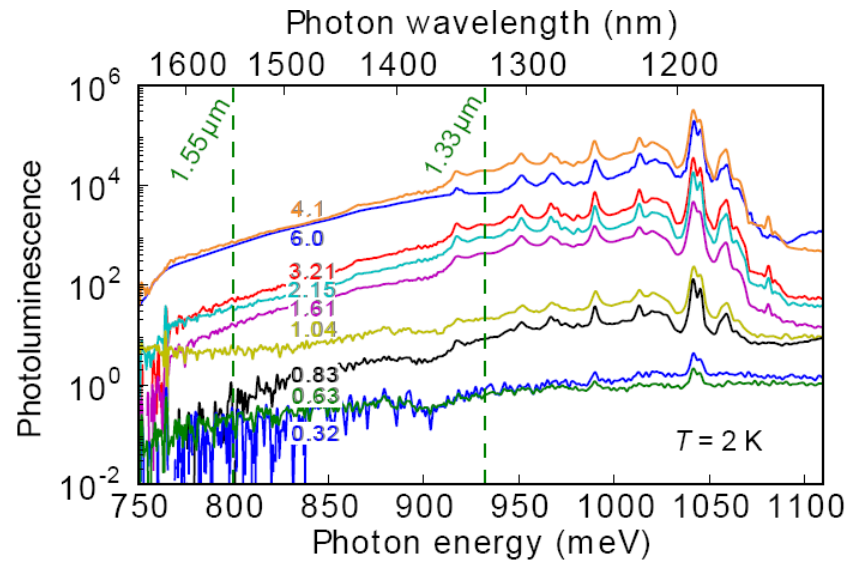


charge: **Mn³⁺** → **Mn⁵⁺**

spin: **S=2** → **S=1**

T. Devillers, ..., AB, Sci. Rep. **2**, 722 [2012]

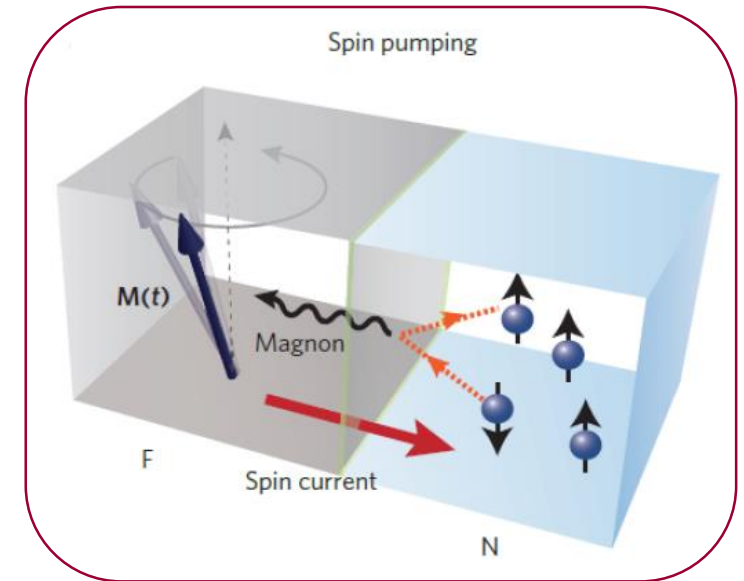
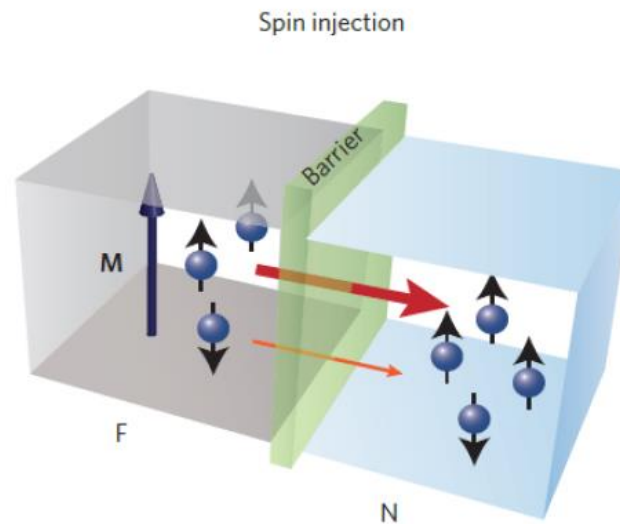
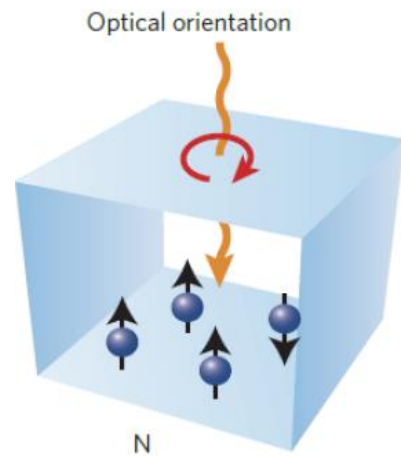
(Ga,Mn)N:Mg – and emitting in the IR



\ IR emission

\ up to room temperature

Generation of spin currents



J. Sinova, ... T. Jungwirth, *Rev. Mod. Phys.* **87**, 1213 [2015]

I. Žutić and H. Dery, *Nat. Mater.* **10**, 647 [2011]

Y. Tserkovnyak *et al.*,
Phys. Rev. Lett. **88**, 117601 [2002]

E. Saitoh *et al.*,
Appl. Phys. Lett. **88**, 182509 [2006]

Estimation of spin Hall angle θ_{HA} in $n\text{-GaN:Si}$

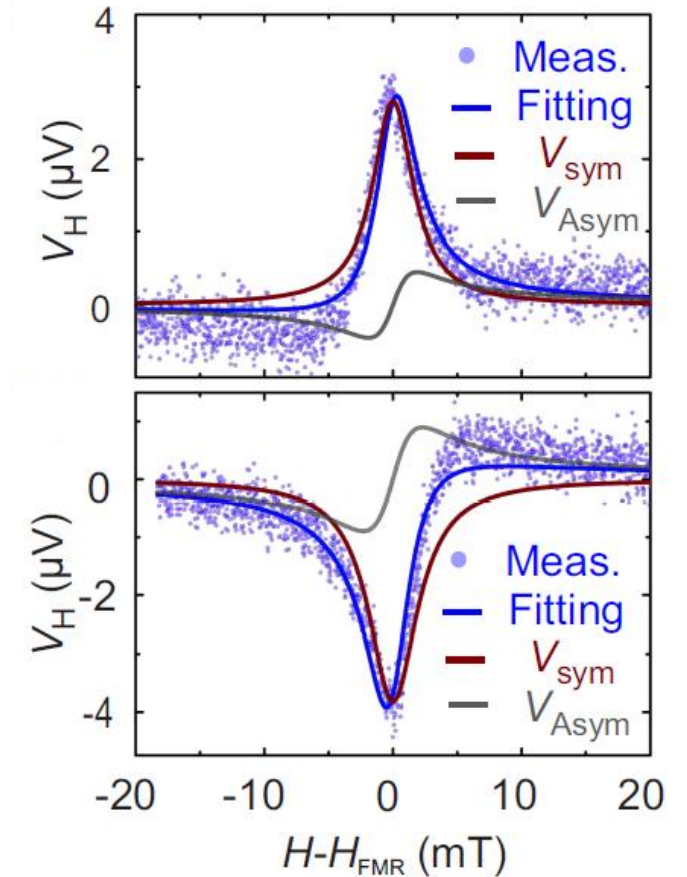
$$\theta_{\text{SH}} = \left(\frac{\hbar}{2e} \right) \frac{V_{\text{ISHE}} (d_N \sigma_N + d_F \sigma_F)}{w \sigma_N \tanh \left(\frac{d_N}{2\lambda_N} \right) j_s^0}$$

■ Function of:

- spin current density j_s^0
- inverse spin Hall voltage V_{ISHE}

■ $\theta_{\text{HA}} = 3.03 \times 10^{-3}$ for $n\text{-GaN:Si}$

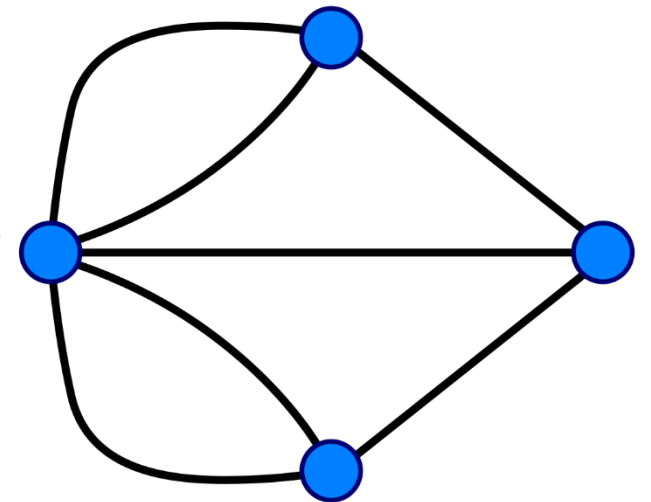
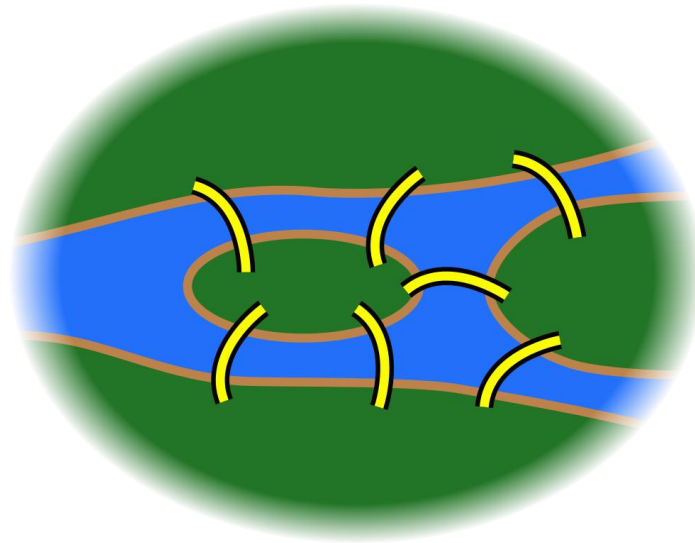
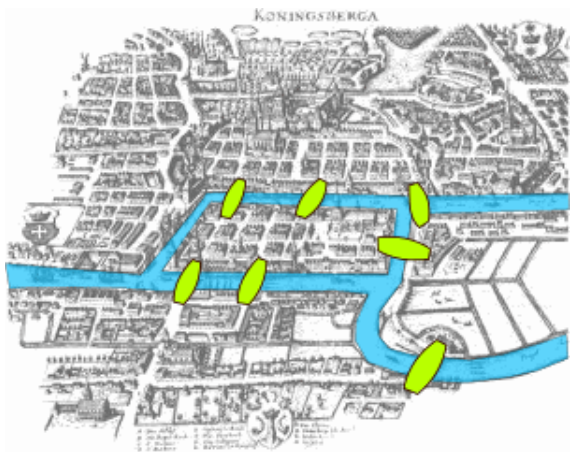
■ 10 \times than other semiconductors like e.g.: Si, Ge, ZnO, $n\text{-GaAs}$



R. Adhikari,.....,AB, Phys. Rev. B **94**, 085205 [2016]

$\text{Sn}_{(1-x)}[\text{Mn}_{(x)}]\text{Te}$
a crystalline topological insulator

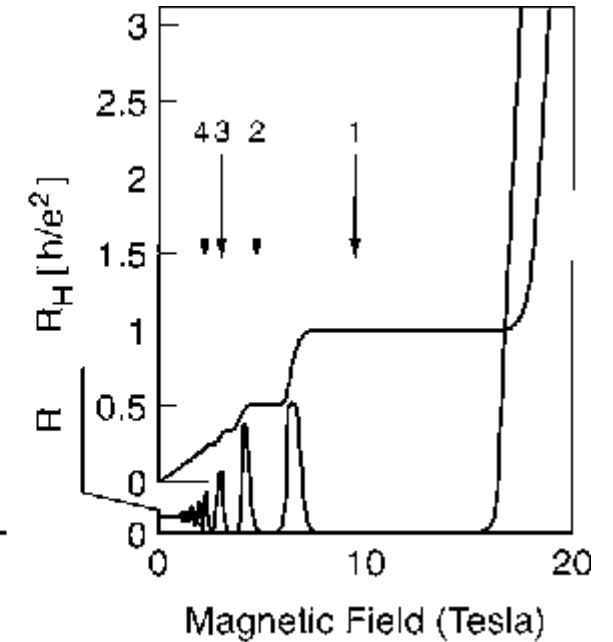
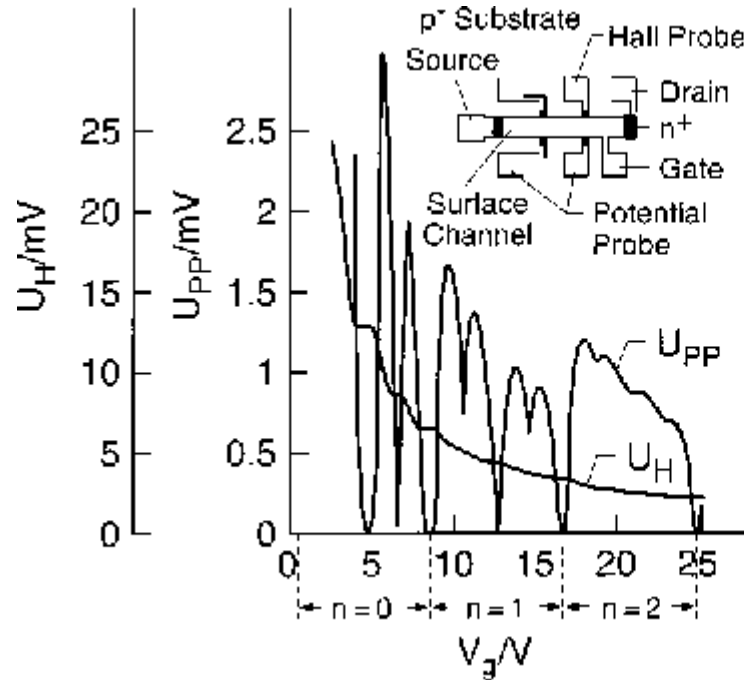
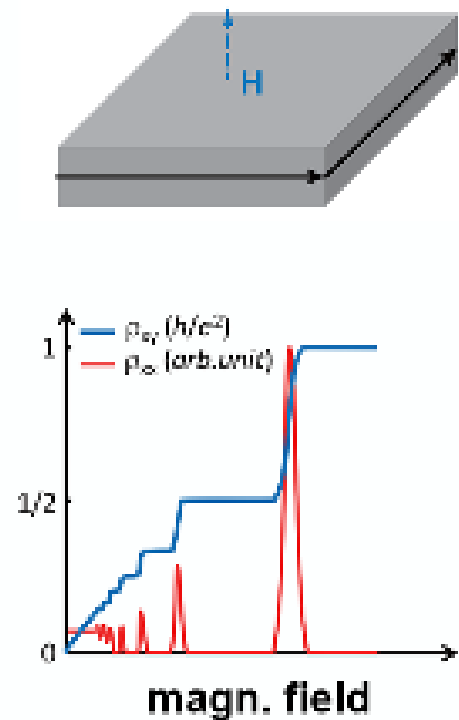
Königsberg bridges: the birth of topology



■ Leonhard Euler [1736] → solution with network diagrams [vertices and arcs]

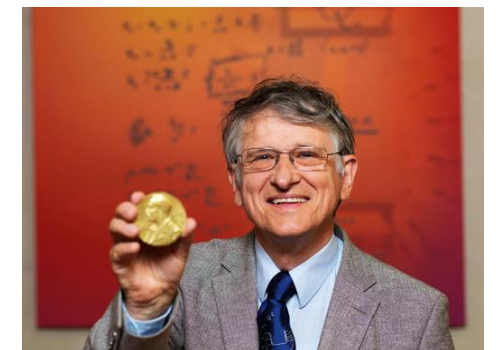
Topology in condensed matter physics

[integer] quantum Hall effect



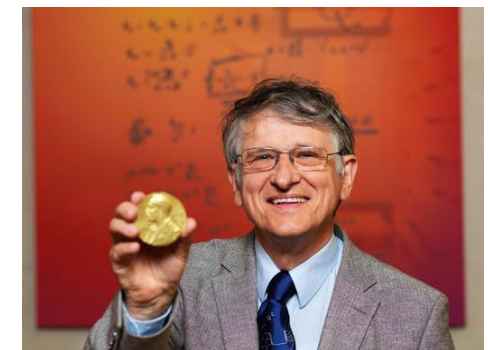
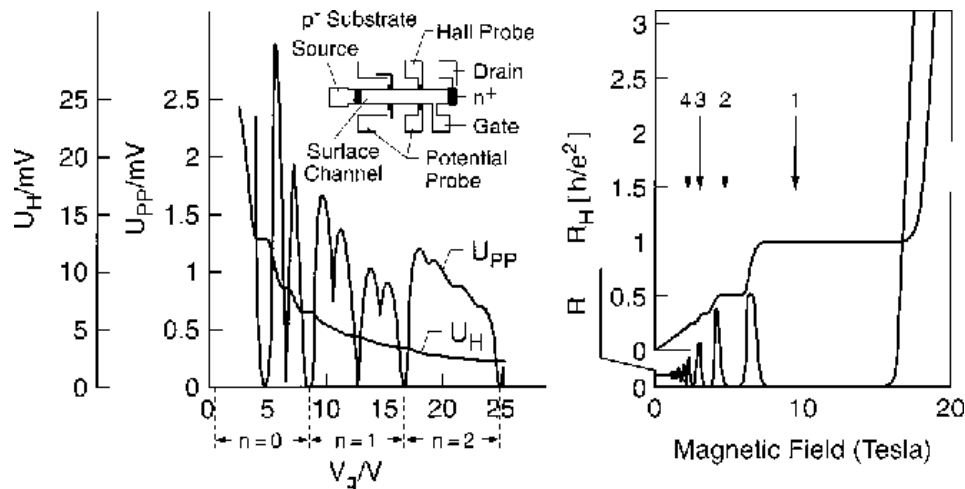
K. von Klitzing *et al.*, Phys. Rev. Lett. **45**, 494 [1980]

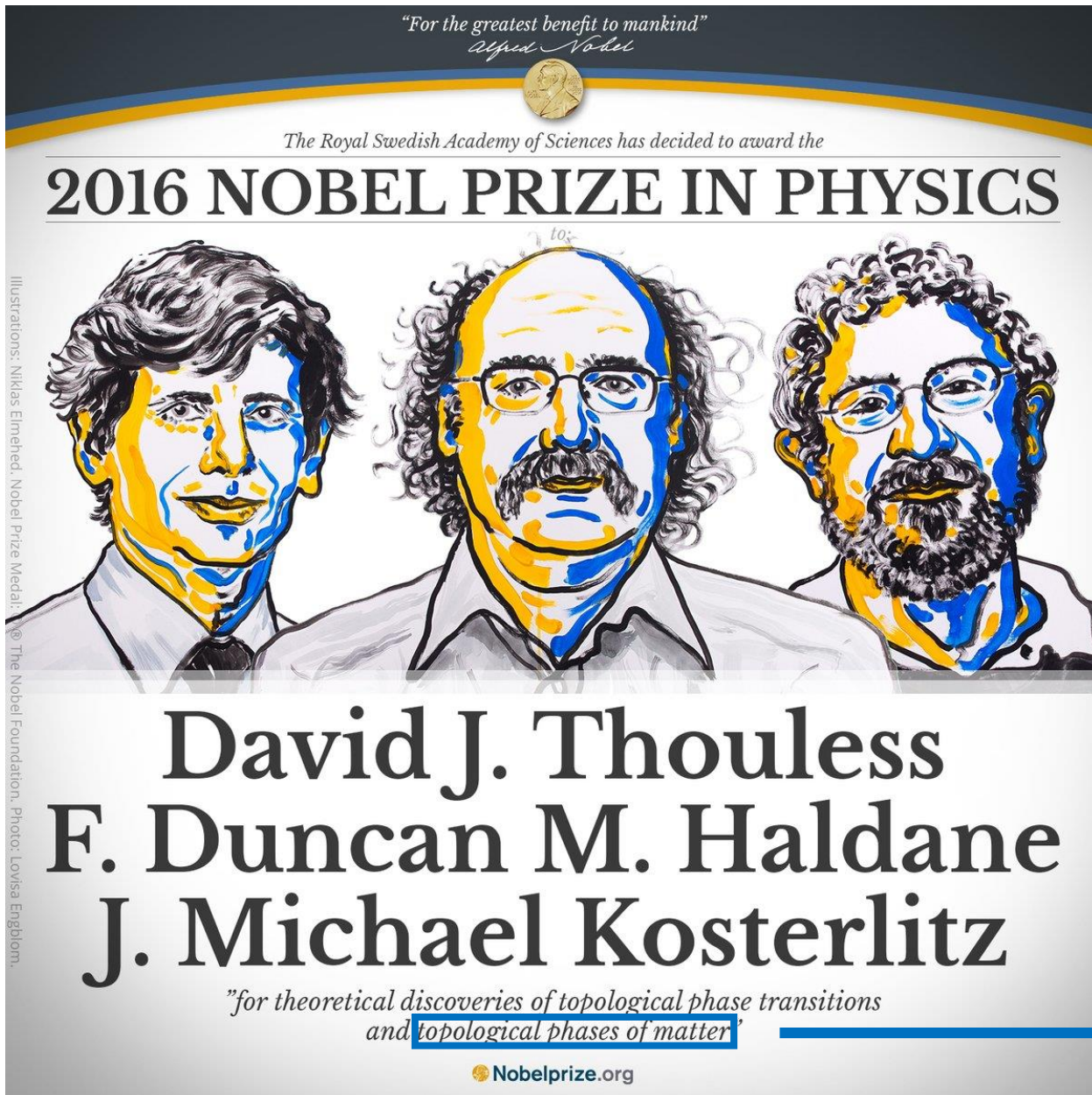
K. von Klitzing *et al.*, Annu. Rev. Condens. Mater. **8**, 13 [2017]



Integer quantum Hall effect – topologically protected phase

- Response function characterized by topological invariant $n \in \mathbb{Z}$
- Hall conductance $\sigma_{xy} = \left(\frac{ne^2}{h}\right)$
- $n \rightarrow$ number of Landau levels under Fermi level



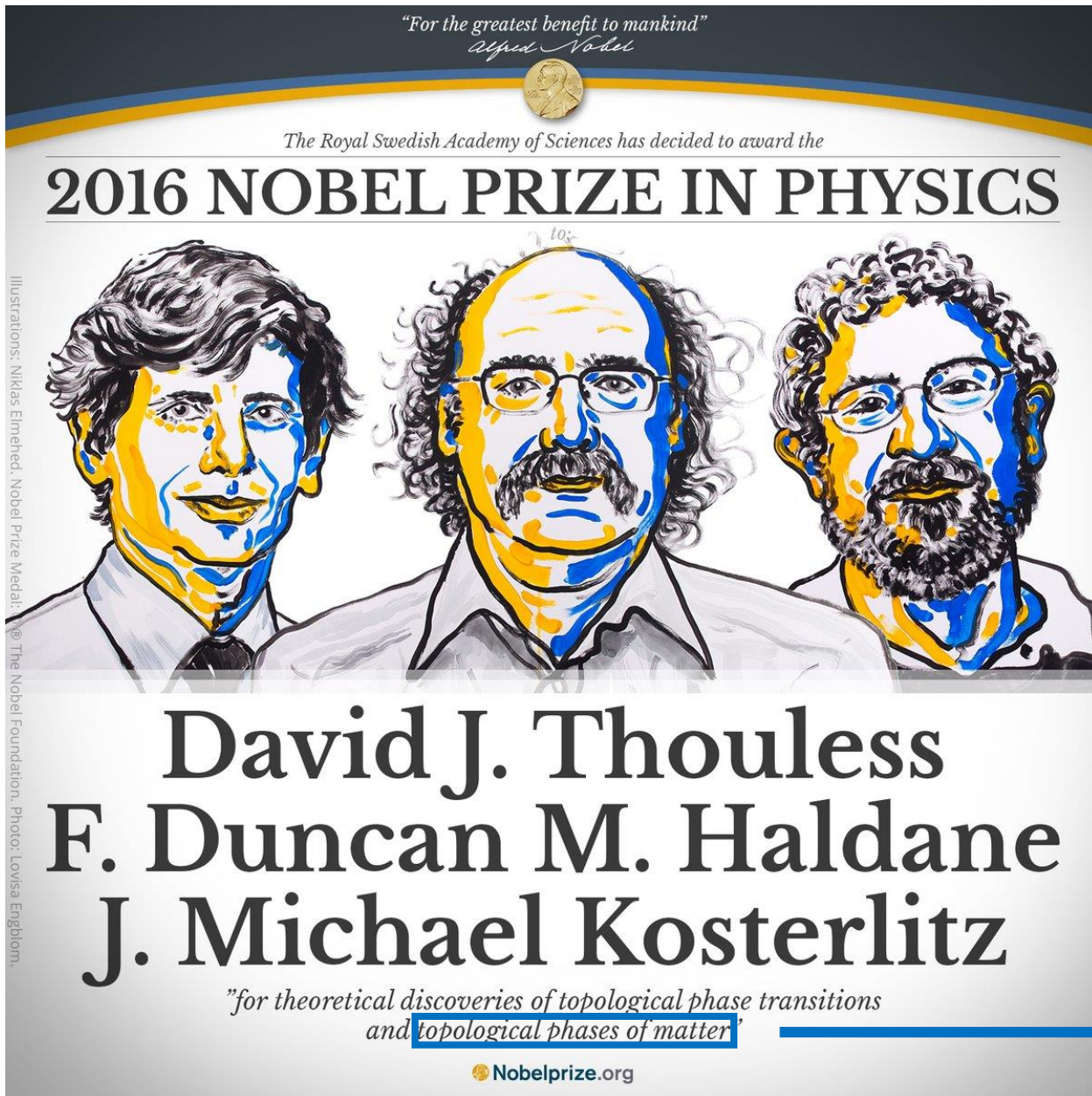


$$C = \frac{1}{2\pi} \int d^2 \mathbf{k} \mathcal{F}_b$$

- Quantization of Hall conductance
- Odd under time-reversal symmetry \mathcal{T}
 - symmetry breaking \rightarrow non-trivial states
- High magnetic fields required

Symmetry protected
topological phases
of matter

D.J. Thouless *et al.*, Phys. Rev. Lett. **49**, 405 [1982]



$$C = \frac{1}{2\pi} \int d^2 \mathbf{k} \mathcal{F}_b$$

- Quantization of Hall conductance
- Odd under time-reversal symmetry \mathcal{T}
 - symmetry breaking \rightarrow non-trivial states
- SOC \rightarrow topological insulators with preserved \mathcal{T}

M.Z. Hasan and C.L. Kane, Rev. Mod. Phys. **82**, 041004 [2010]
M. König *et al.*, Science **318**, 766 [2007]

Symmetry protected topological phases of matter

D.J. Thouless *et al.*, Phys. Rev. Lett. **49**, 405 [1982]

Chiral edge states as dissipationless channels

- Chern number C → number of dissipationless channels
- Topological insulators → $C = \pm 1$ → only time-reversal \mathcal{T} protects the topology
- Challenge → $|C| > 1$

PRL **106**, 106802 (2011)

PHYSICAL REVIEW LETTERS

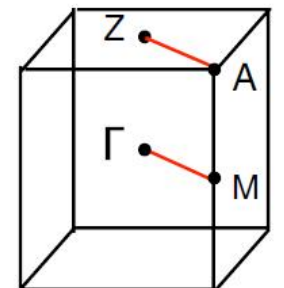
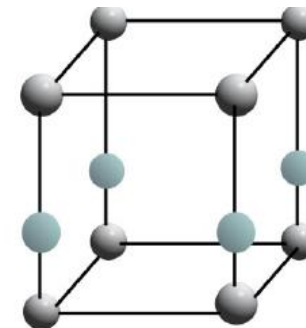
week ending
11 MARCH 2011

Topological Crystalline Insulators

Liang Fu

Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA

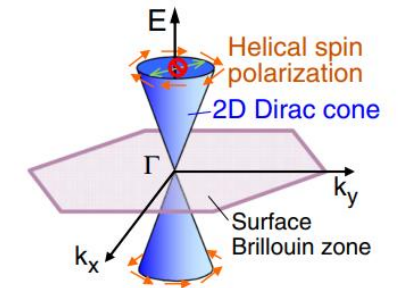
Topology protected by mirror symmetry \mathcal{M}



Topological crystalline insulators

- Crystal point group symmetries accounted for
- Presence of surfaces \rightarrow crystal symmetry breaking
- TCI \rightarrow counterpart of TI in systems without SOC
 - e^- orbital degree of freedom \rightarrow role similar to spin

- TI \rightarrow linear dispersion of Dirac surface states
 - protection: time-reversal invariant
 - single Dirac cone



Y. Ando *et al.*, J. Phys. Soc. Jap. **82**, 102001 [2013]

- TCI \rightarrow quadratic band degeneracy
 - protection: crystal [point-group] symmetry
 - mirror \mathcal{M} symmetry \rightarrow two independent topological invariants C_t and $C_{\mathcal{M}}$
 - four Dirac cones

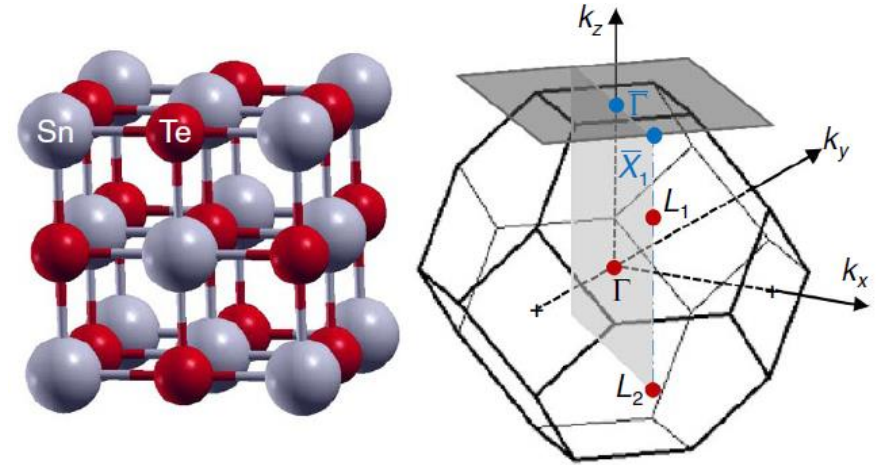
ARTICLE

Received 23 Feb 2012 | Accepted 21 Jun 2012 | Published 31 Jul 2012

DOI: 10.1038/ncomms1969

Topological crystalline insulators in the SnTe material class

Timothy H. Hsieh¹, Hsin Lin², Junwei Liu^{1,3}, Wenhui Duan³, Arun Bansil² & Liang Fu¹



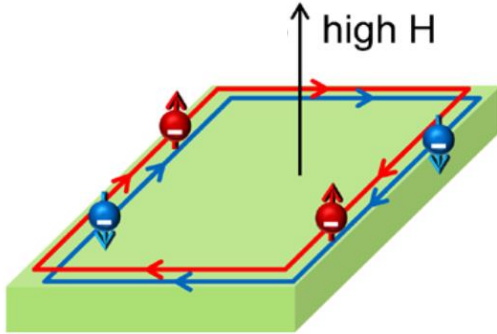
- $|C_{\mathcal{M}}| \neq 0 = 2$

- [one isotropic Dirac surface state $\bar{\Gamma}$] + [three anisotropic at \bar{M}]

- Majorana-like excitations stabilized at surface steps

P. Sessi *et al.*, Science **354**, 1269 [2016]

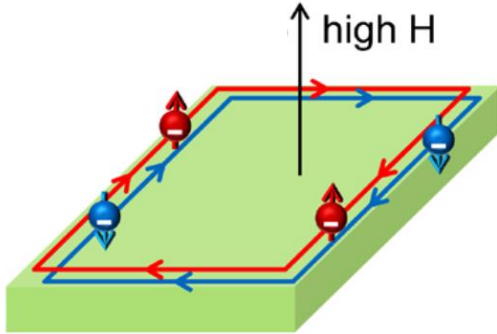
Chiral edges as dissipationless channels



QHE

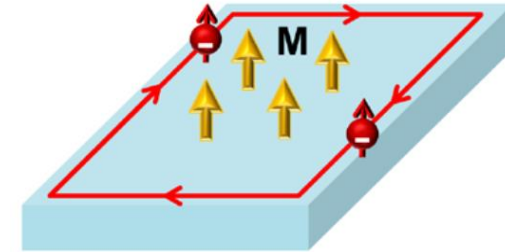
- 2D electron gas
- high magnetic field required

By adding magnetic elements



QHE

- 2D electron gas
- high magnetic field required



QAHE

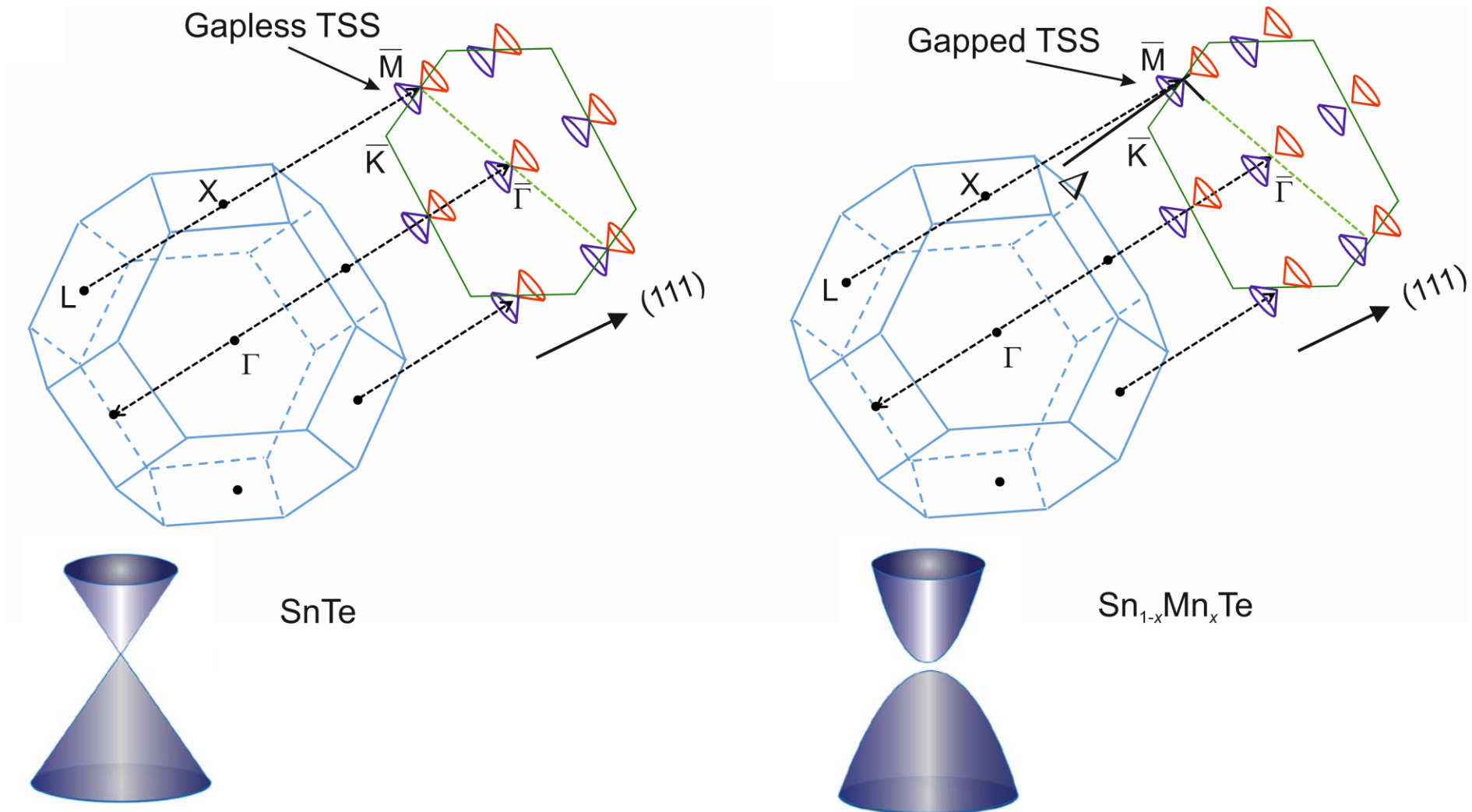
- magnetic topological insulator
- intrinsic M
- no magnetic field required

C.Z. Chang *et al.*, Science **340**, 6129 [2013]

Ferromagnetic insulator with $|C| \neq 0$:

- [intrinsic anomalous Hall effect and QHE] & [geometric Berry phase] → QAH states
- quantization of Hall conductance σ_{xy} in the absence of external field

Effect of Mn doping on SnTe band structure



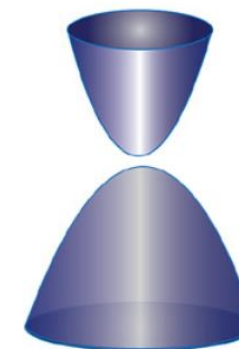
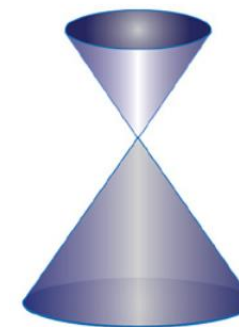
SnTe vs. $\text{Sn}_{1-x}\text{Mn}_x\text{Te}$

■ SnTe

- narrow band-gap [direct, ~ 0.18 eV]
- thermoelectric material
- p -type conductivity, $p \sim 10^{20} \text{ cm}^{-3}$

■ $\text{Sn}_{1-x}\text{Mn}_x\text{Te}$

- dilute magnetic semiconductor
- carrier-mediated ferromagnetism [RKKY]



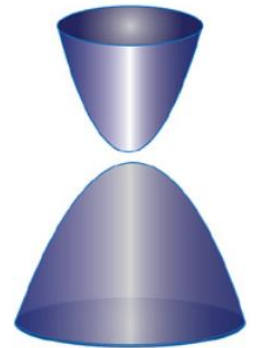
Mn doping and topology in SnTe

■ $\text{Sn}_{1-x}\text{Mn}_x\text{Te}$

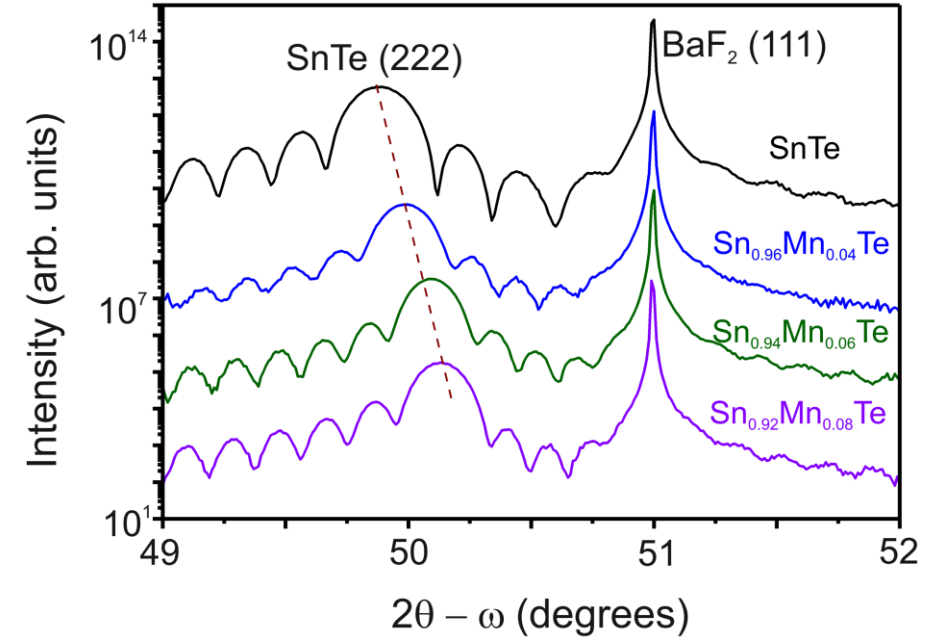
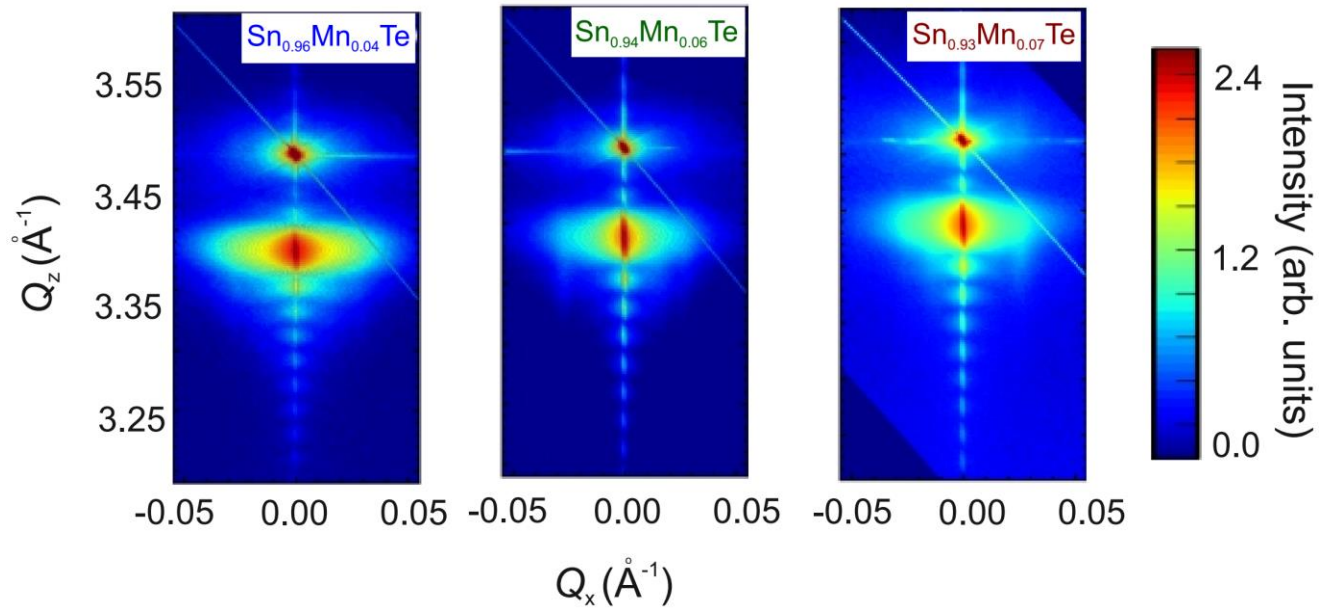
- gapped Dirac cones at the time-reversal invariant momenta [TRIM] points $\bar{\Gamma}$ and \bar{M}
- total Chern number $\rightarrow C_{\text{tot}} = C_{\text{T}} + C_{\text{M}}$
- QAHE in ferromagnetic SnTe [predicted]
- 4 dissipationless chiral edge channels

■ Criteria for QAH states

- ferromagnetic ordering
- emergence of AHE
- perpendicular magnetic anisotropy
- sample thickness vs. decay length of topological surface states



Sn_{1-x}Mn_xTe(111) – structural characterization

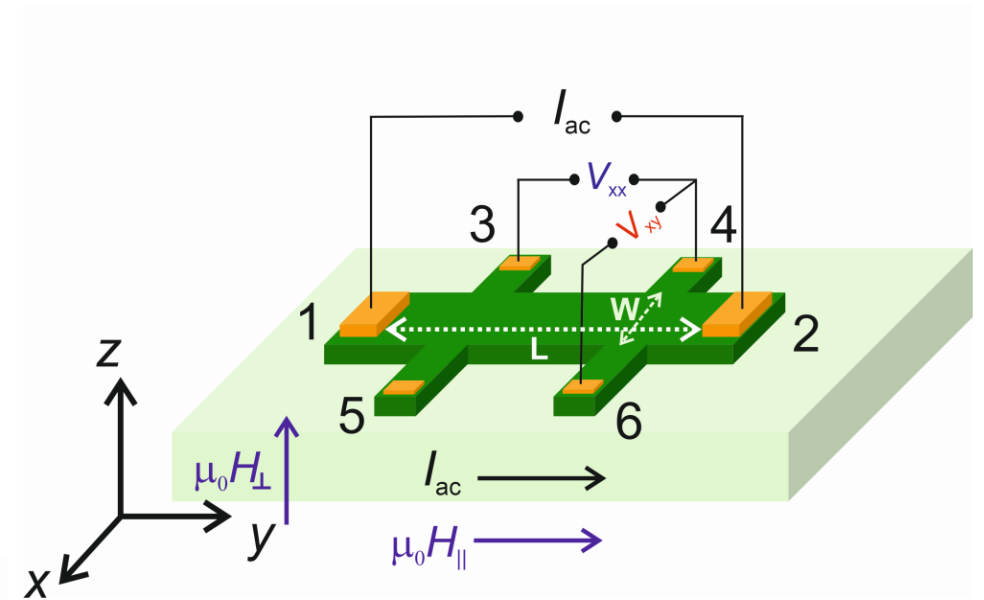
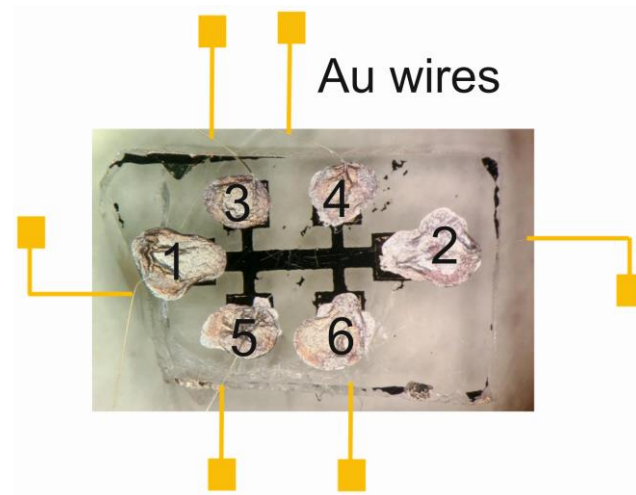
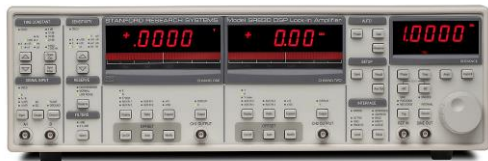


- Sn_{1-x}Mn_xTe(111) strained on BaF₂(111) substrate
- Mn incorporated into SnTe lattice
- no evidence of secondary phases
- Mn concentration follows Vegard's law

$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – electronic properties

Magnetotransport

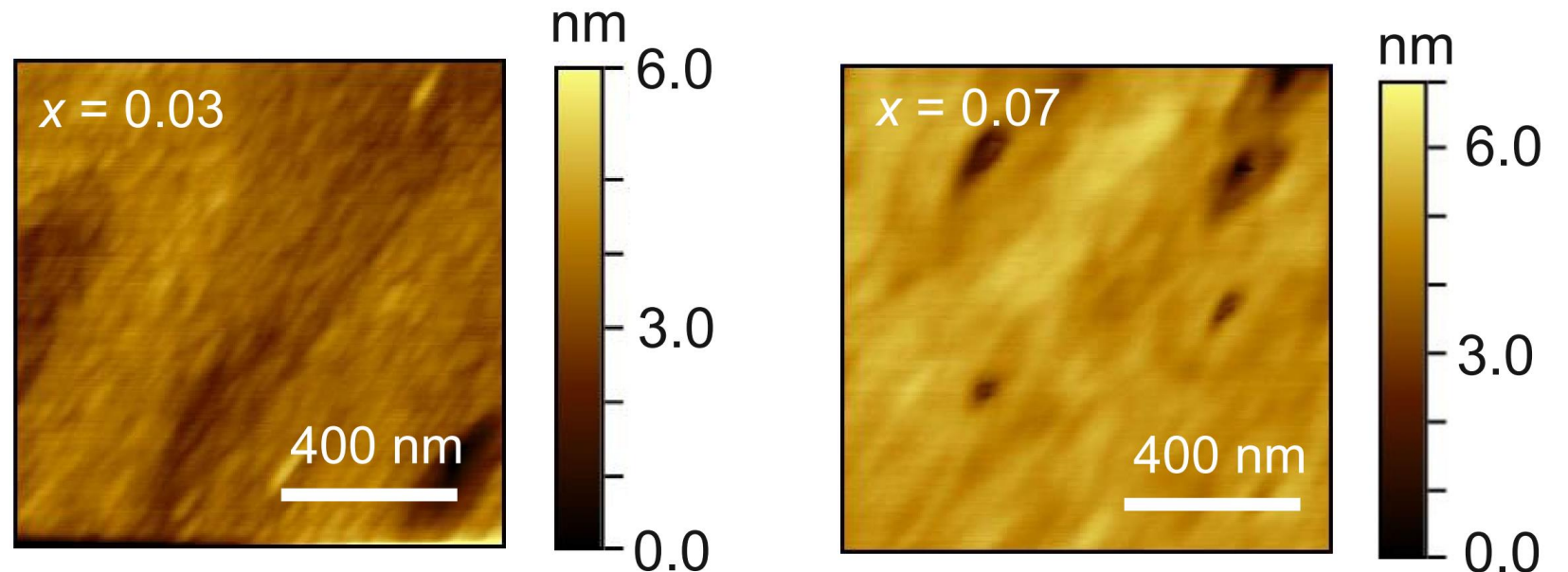
- lithographically designed Hall bars
- ac lock-in
- $1.8 \text{ K} \leq T \leq 300 \text{ K}$
- $-7 \text{ T} \leq \mu_0 H \leq +7 \text{ T}$



$\text{Sn}_{1-x}\text{Mn}_x\text{Te}$ samples

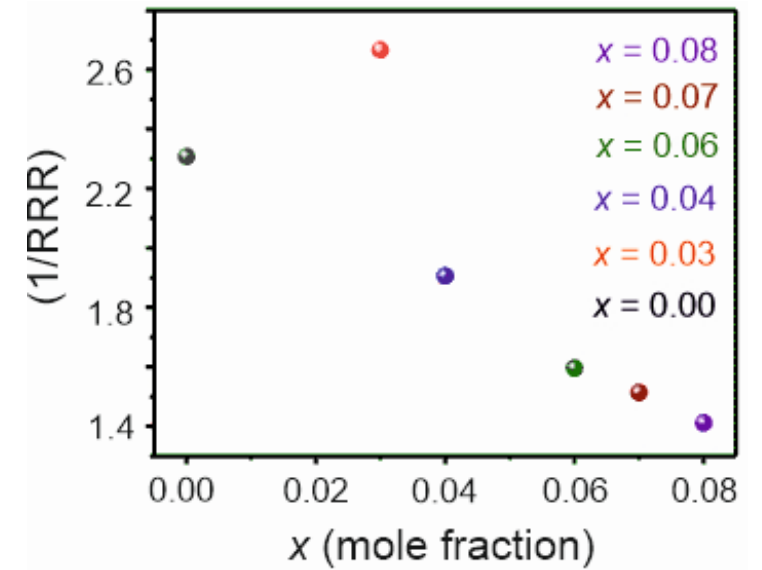
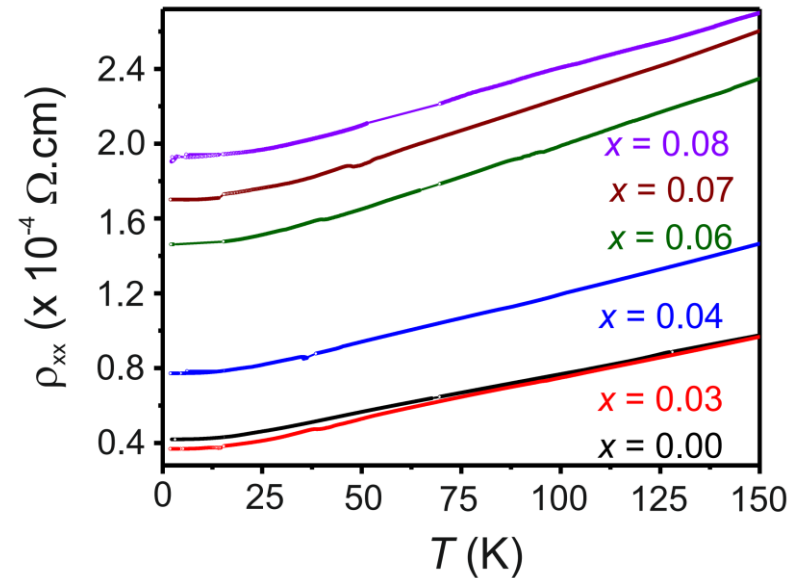
■ Epitaxial $\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$

- molecular beam epitaxy
- substrates: $\text{BaF}_2(111)$ → \mathcal{M} symmetry and \mathcal{T} invariance
- thickness: 30 nm → adjusted to decay length of surface states; high crystallinity
- $x = 0.00; 0.03; 0.04; 0.06; 0.07$ and 0.08



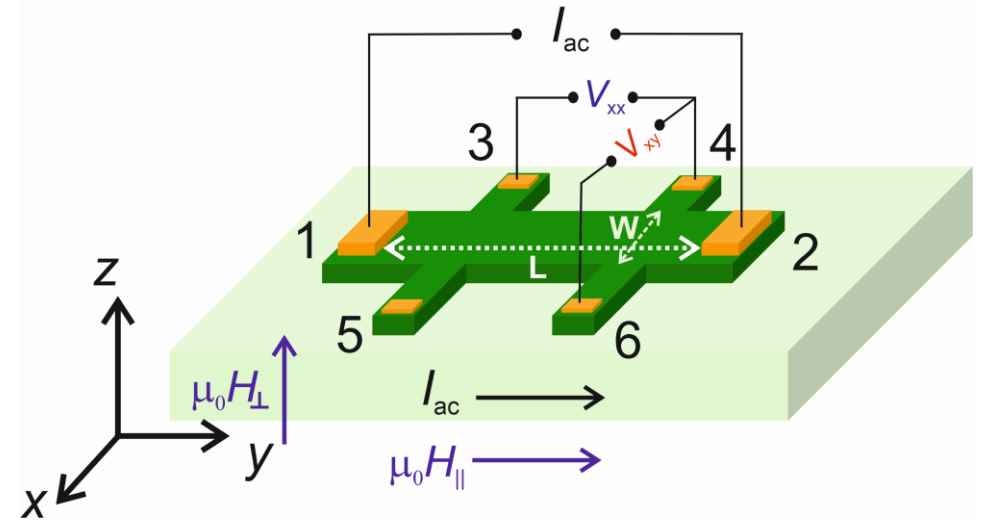
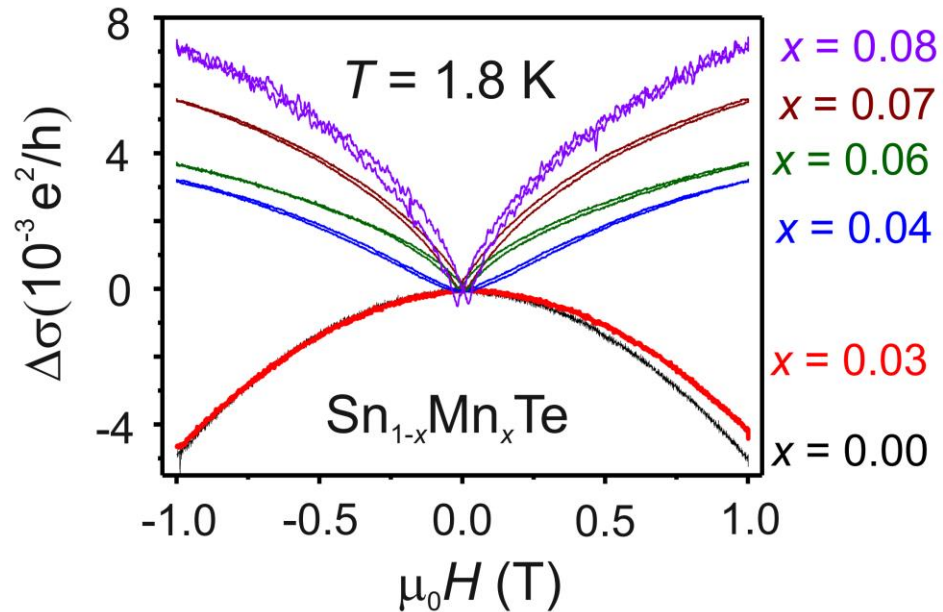
$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – resistivity

- semi-metallic behaviour
- residual resistivity ratio [RRR]
- disorder increases with Mn content



Sn_{1-x}Mn_xTe(111) – magnetoconductance

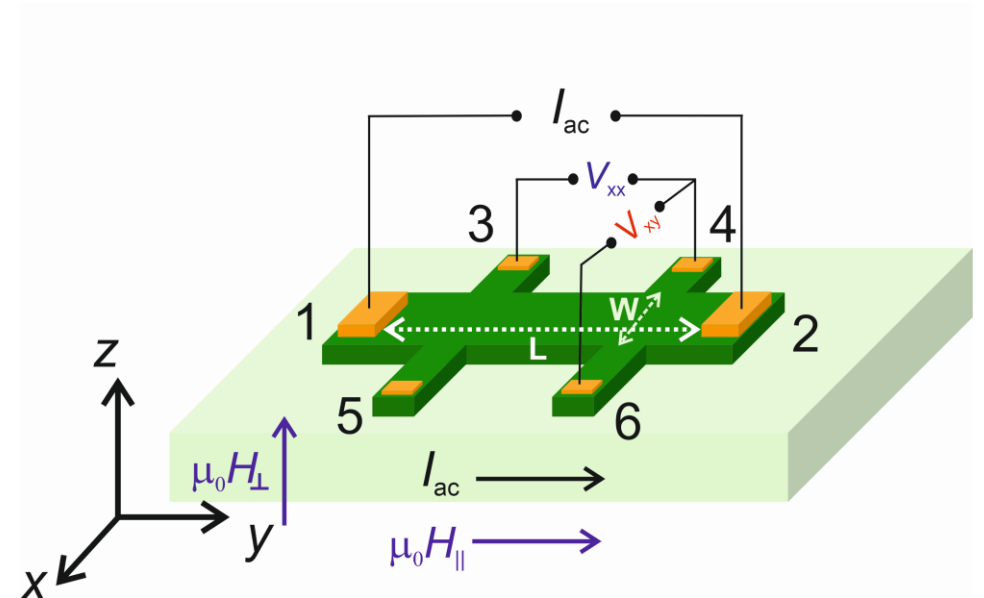
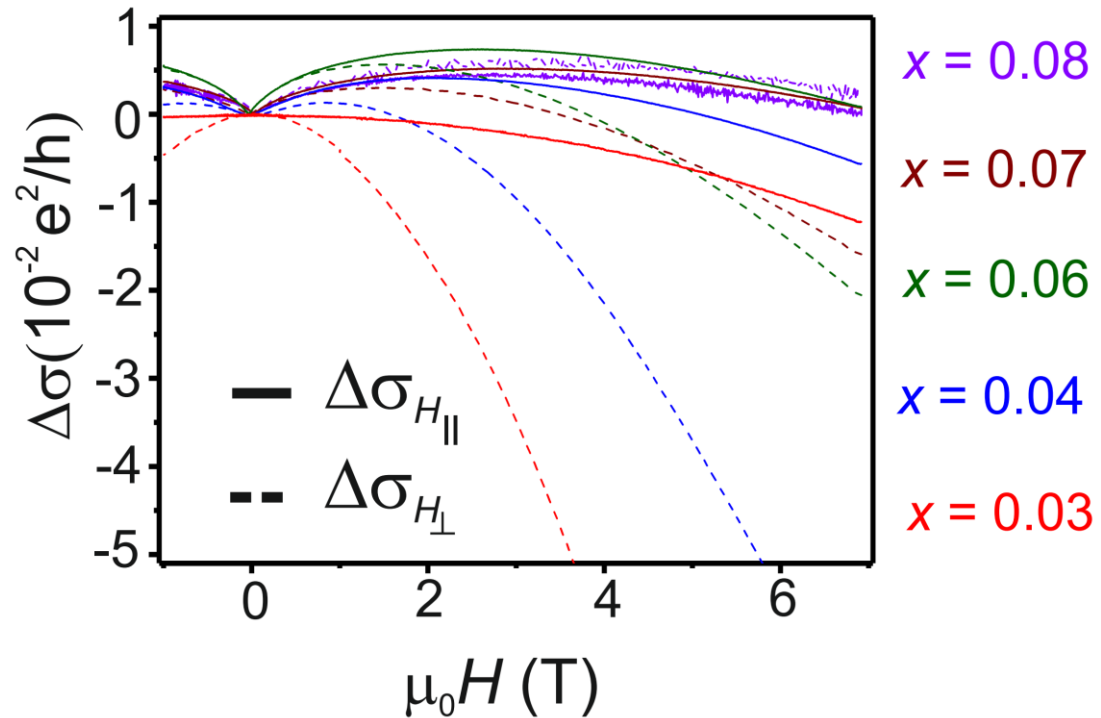
$$\sigma_{xx} = \left(\frac{L}{W}\right) \left[R_{xx} \left(\frac{e^2}{h}\right) \right]$$



$$\Delta\sigma = [\sigma_{xx}(H) - \sigma_{xx}(0)] \left(\frac{e^2}{h}\right)$$

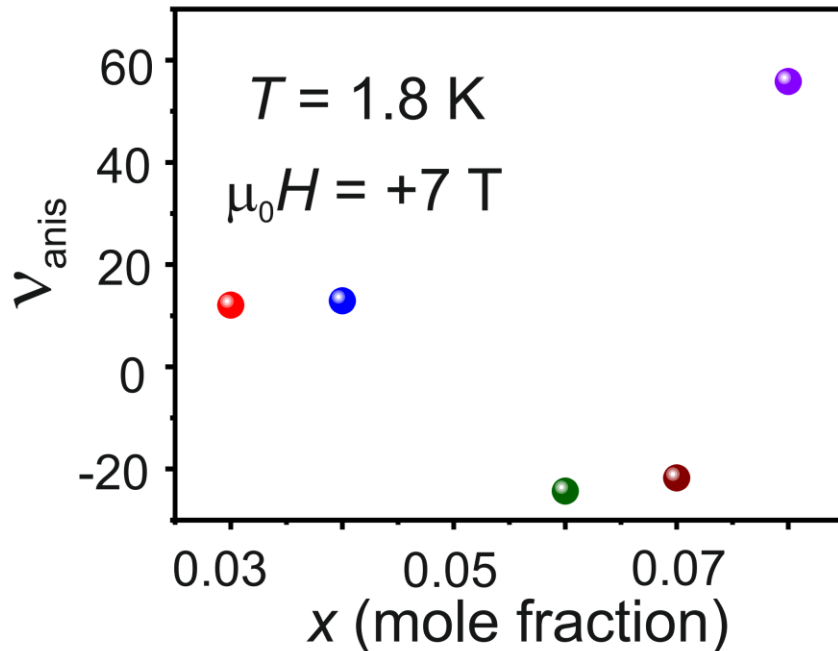
$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – magnetoconductance \rightarrow PMA

Emergence of perpendicular magnetic anisotropy [PMA]



$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – magnetoconductance \rightarrow PMA

Emergence of perpendicular magnetic anisotropy [PMA]



$$V_{\text{anis}} = \frac{\Delta\sigma_{H\perp}}{\Delta\sigma_{H\parallel}}$$

$x = 0.08$

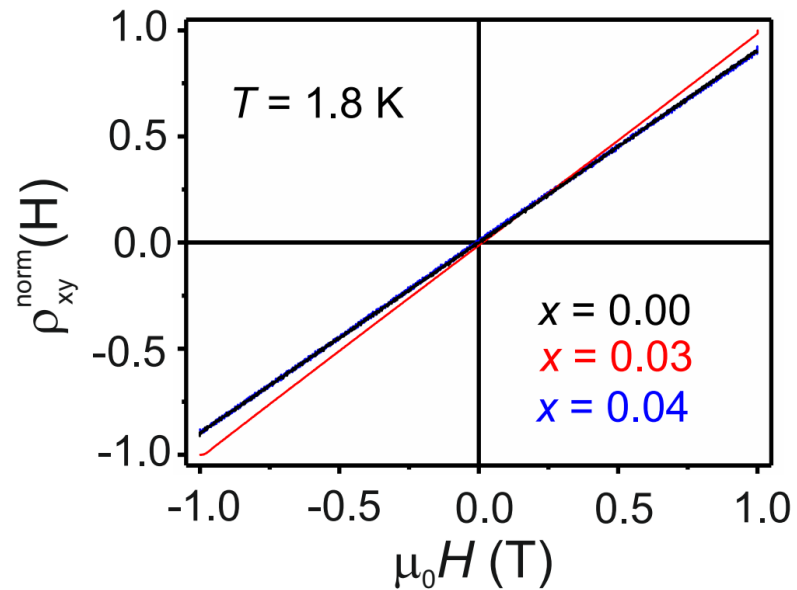
- contribution: surface states
- immune to disorder

$0.03 \leq x \leq 0.07$

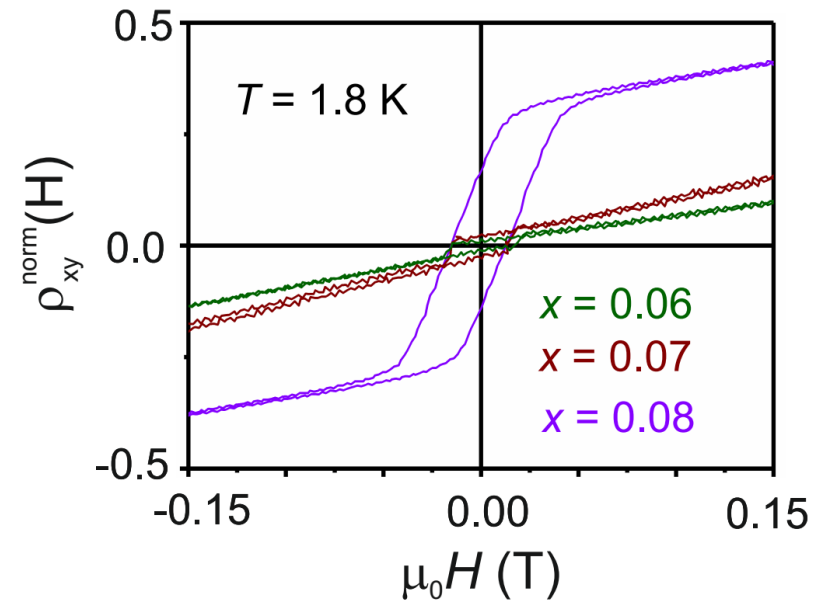
- contribution: (surface + bulk) states

$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – Hall resistivity \rightarrow AHE

ordinary Hall effect

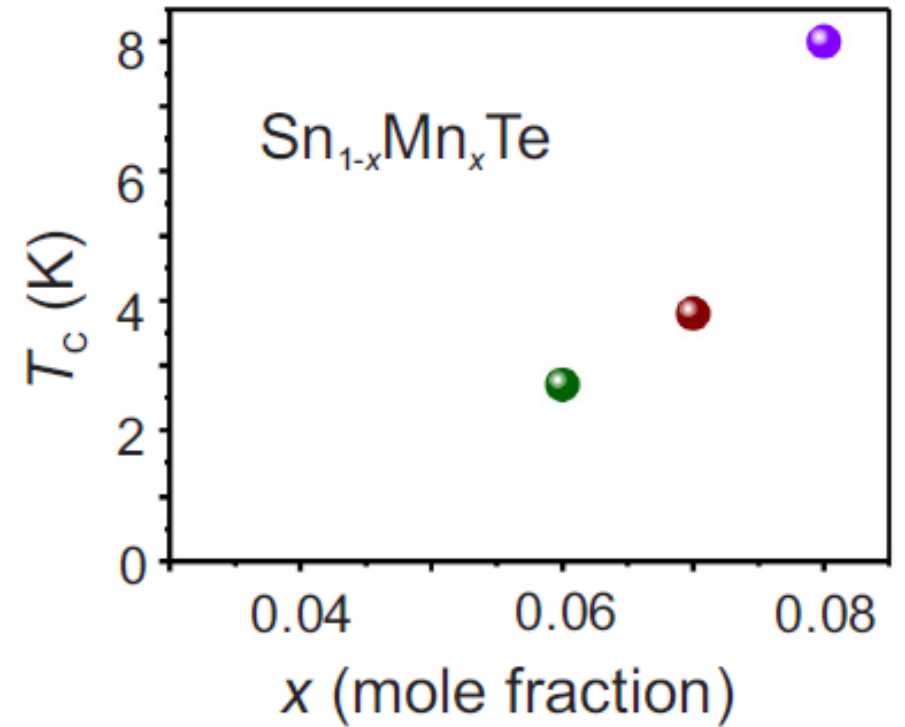
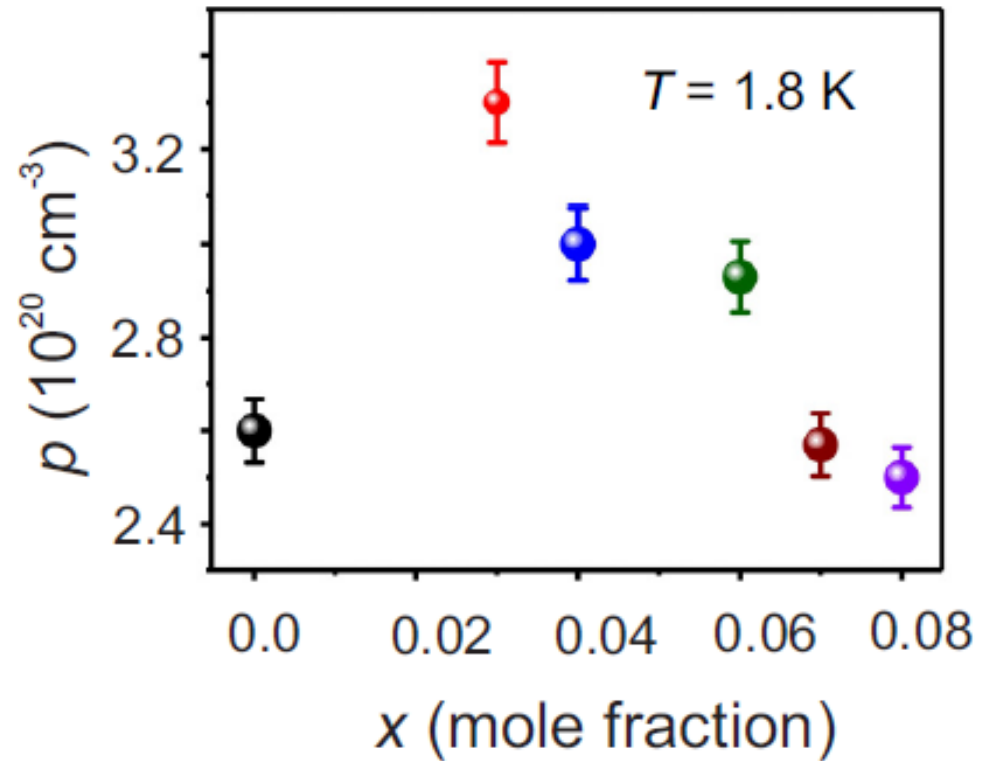


anomalous Hall effect

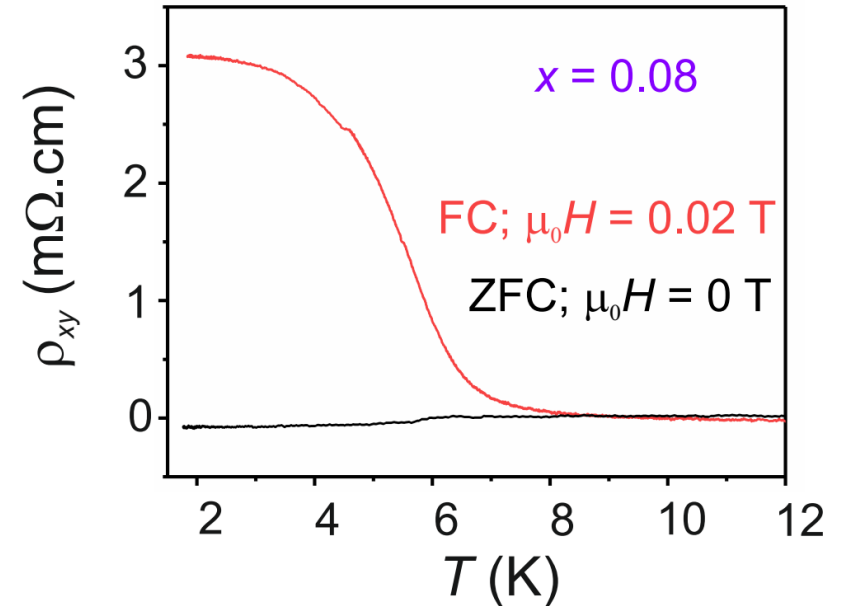
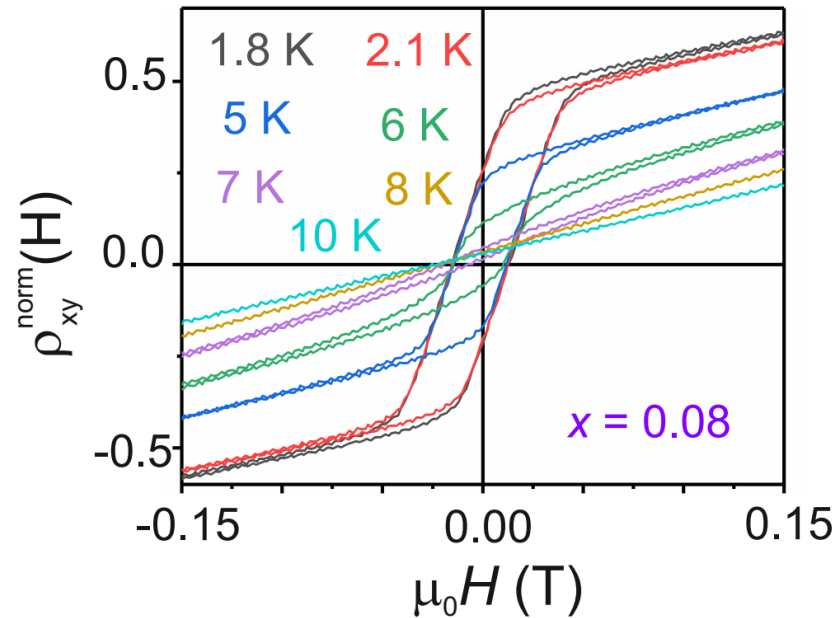


$$\rho_{xy}(H) = R_H H + \mu_0 R_{AH} M.$$

$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111) - T_c$

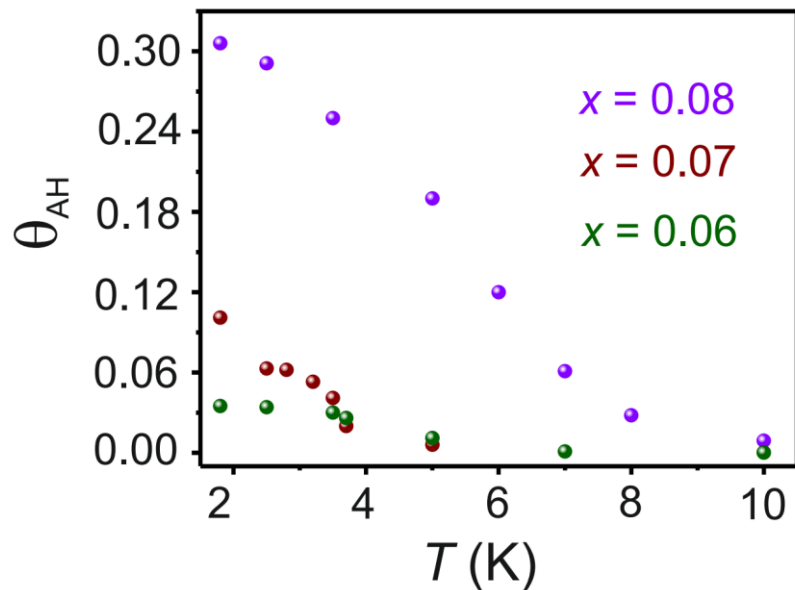


$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – Hall resistivity

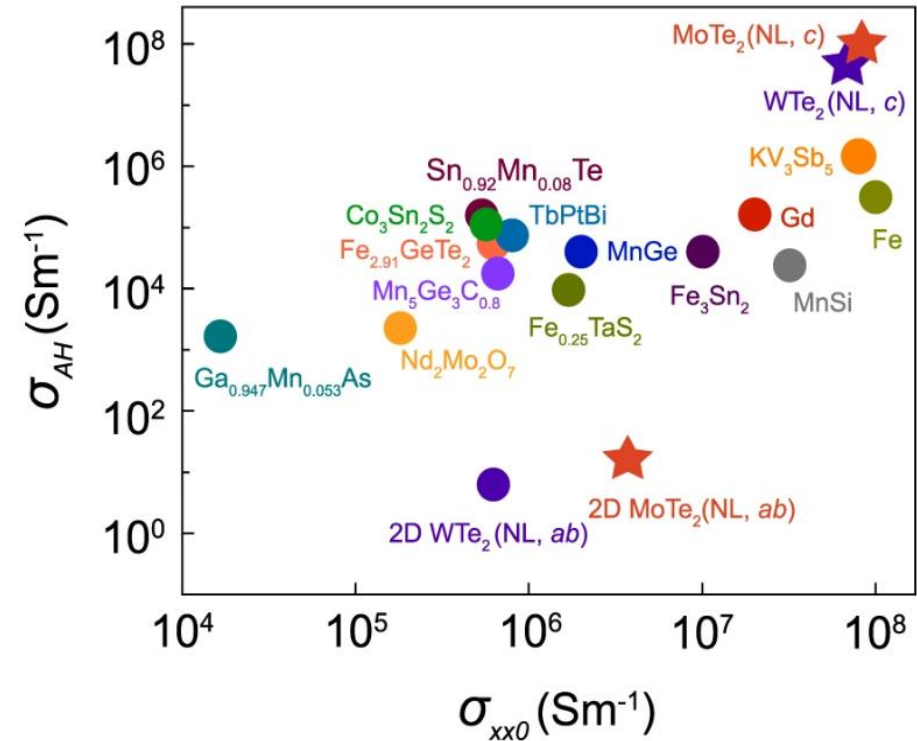


- hysteretic anomalous Hall effect
- ferromagnetic order for $x \geq 0.06$
- carrier mediated FM
- highest $T_C \sim 7.5$ K for $x = 0.08$

Sn_{1-x}Mn_xTe(111) – anomalous Hall angle



$$\theta_{\text{AH}} = \frac{\sigma_{\text{xy}}^{\text{AH}}}{\sigma_{\text{xx}}}$$



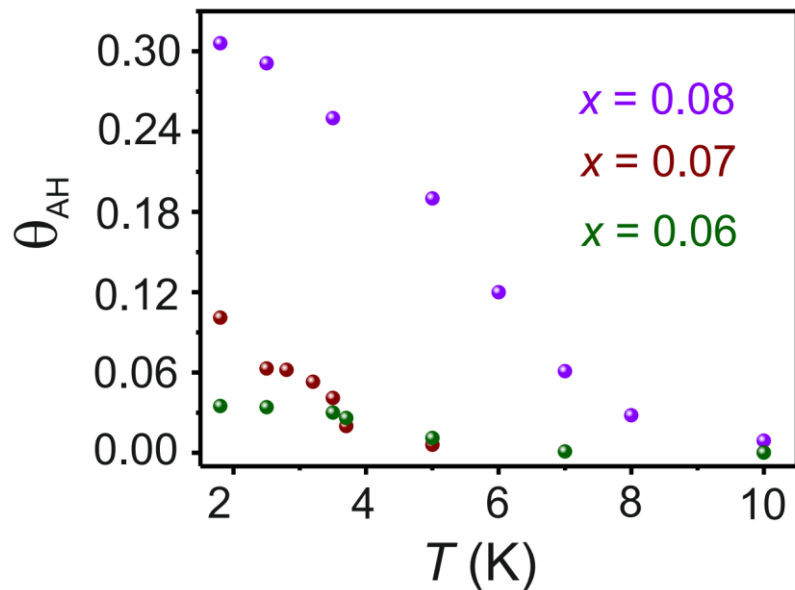
A. Tiwari *et al.* Nat. Commun. **12**, 2049 [2021]

□ $\theta_{\text{AH}} = 0.3$ for Sn_{0.92}Mn_{0.08}Te(111)

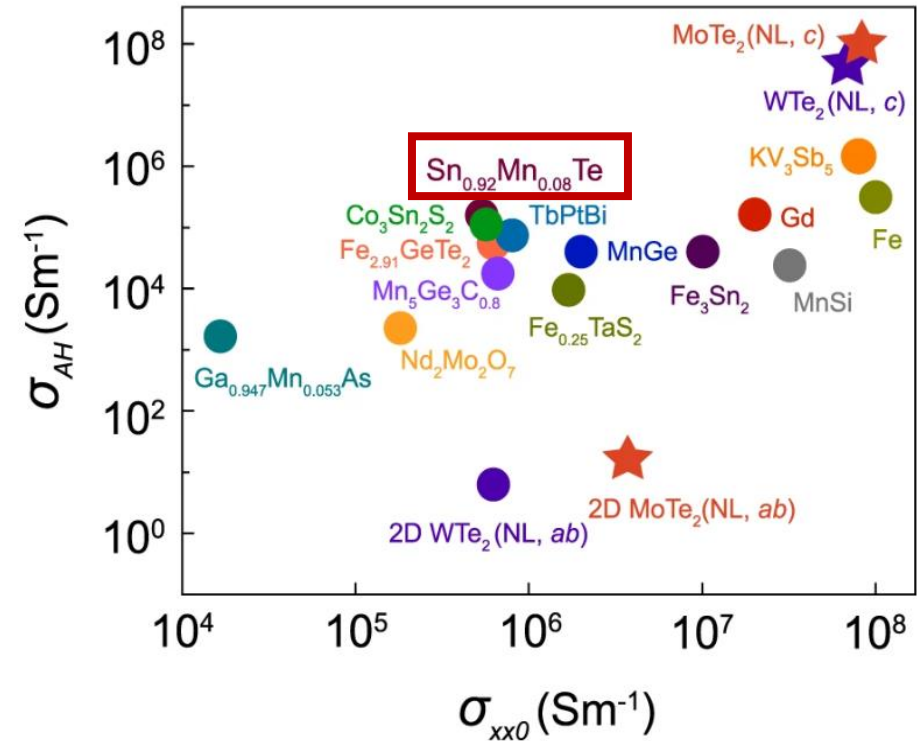
● $T = 1.8$ K, $\mu_0 H = +1$ T

Among the highest reported for a magnetic topological insulator

Sn_{1-x}Mn_xTe(111) – anomalous Hall angle



$$\theta_{\text{AH}} = \frac{\sigma_{\text{xy}}^{\text{AH}}}{\sigma_{\text{xx}}}$$



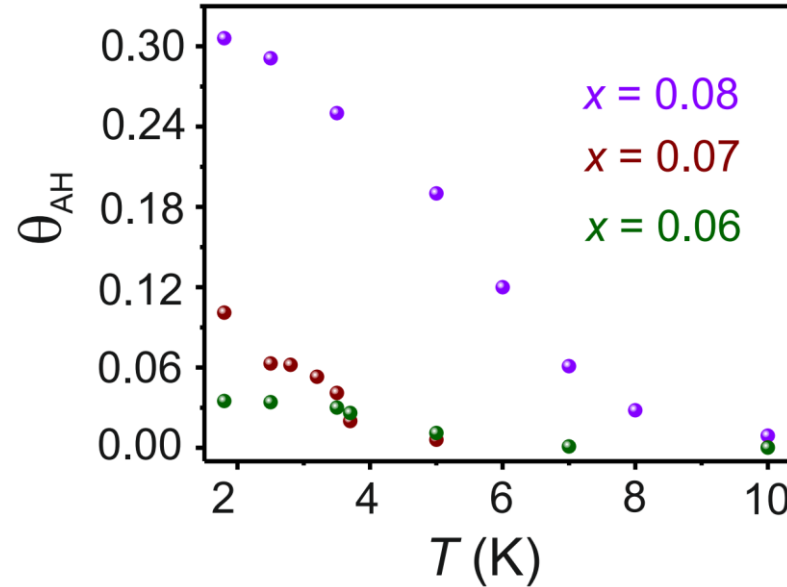
A. Tiwari *et al.* Nat. Commun. **12**, 2049 [2021]

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Among the highest reported for a magnetic topological insulator

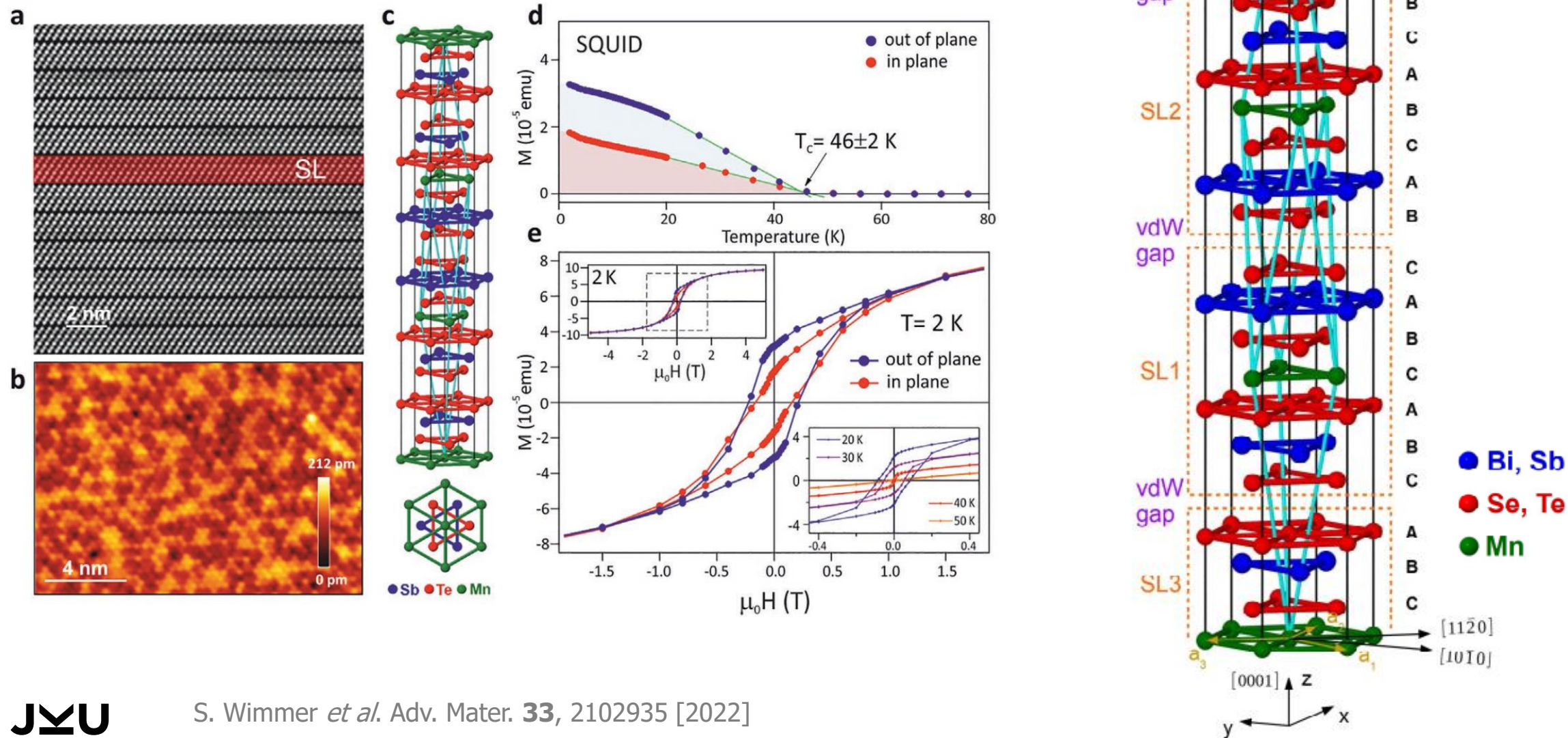
$\text{Sn}_{1-x}\text{Mn}_x\text{Te}(111)$ – synopsis



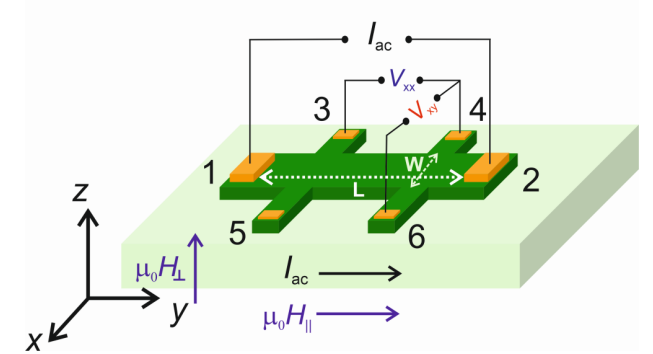
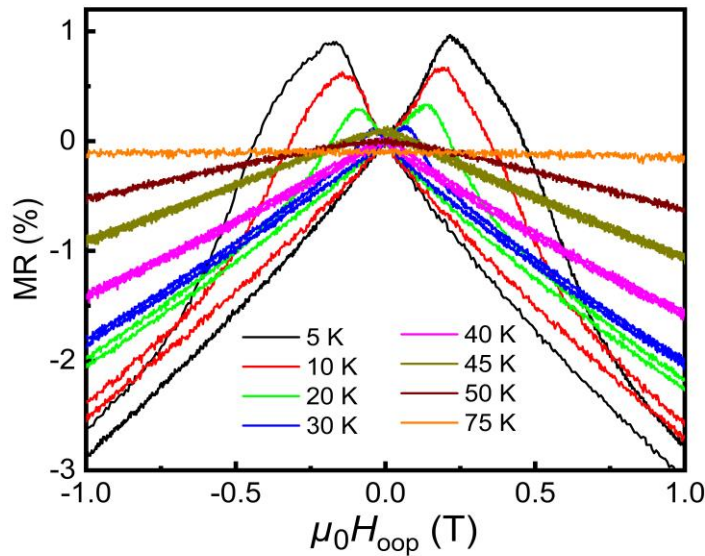
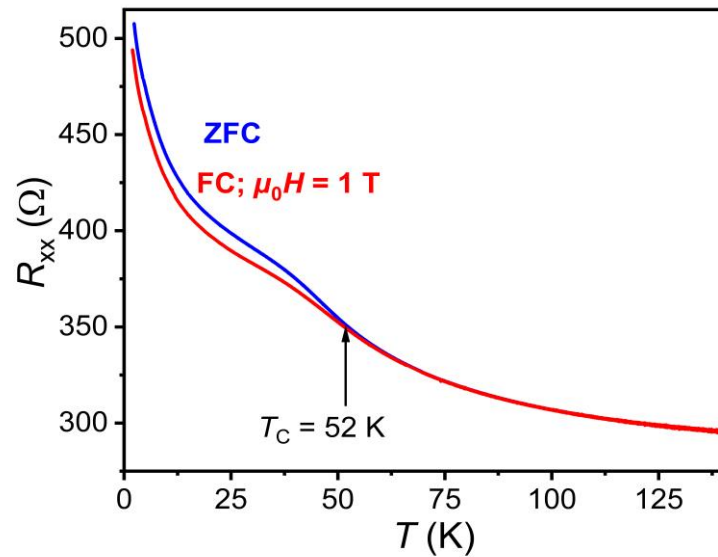
$$\theta_{\text{AH}} = \frac{\sigma_{\text{xy}}^{\text{AH}}}{\sigma_{\text{xx}}}$$

- [carrier mediated] ferromagnetic ordering
- emergence of AHE
- PMA
- thickness adjusted to the decay length of topological surface states

MnSb₂Te₄ – intrinsic MTI

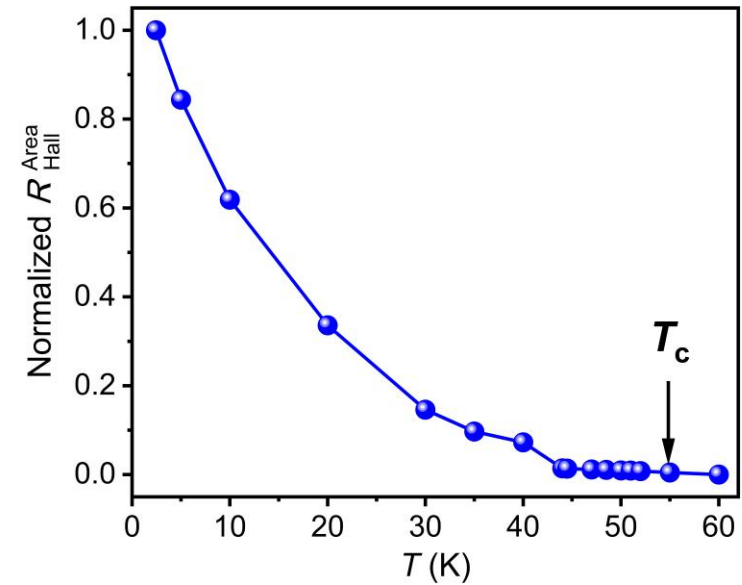
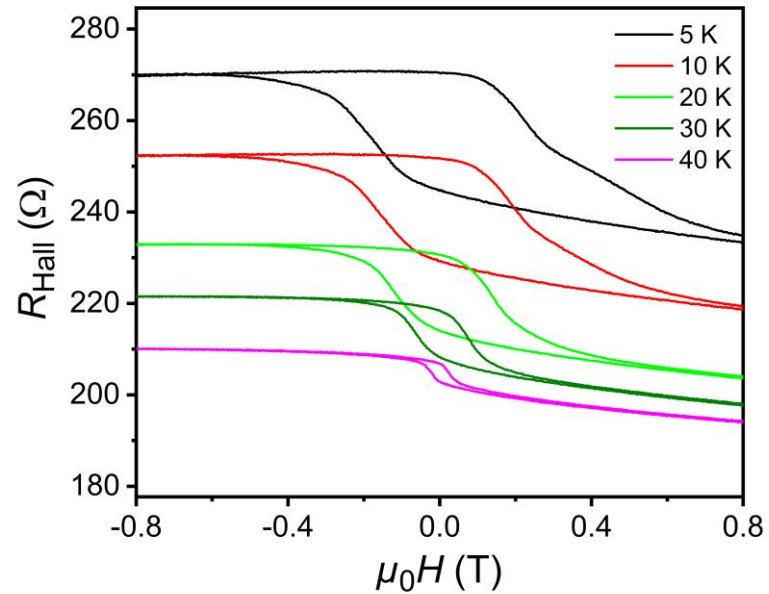


MnSb₂Te₄ – magnetotransport



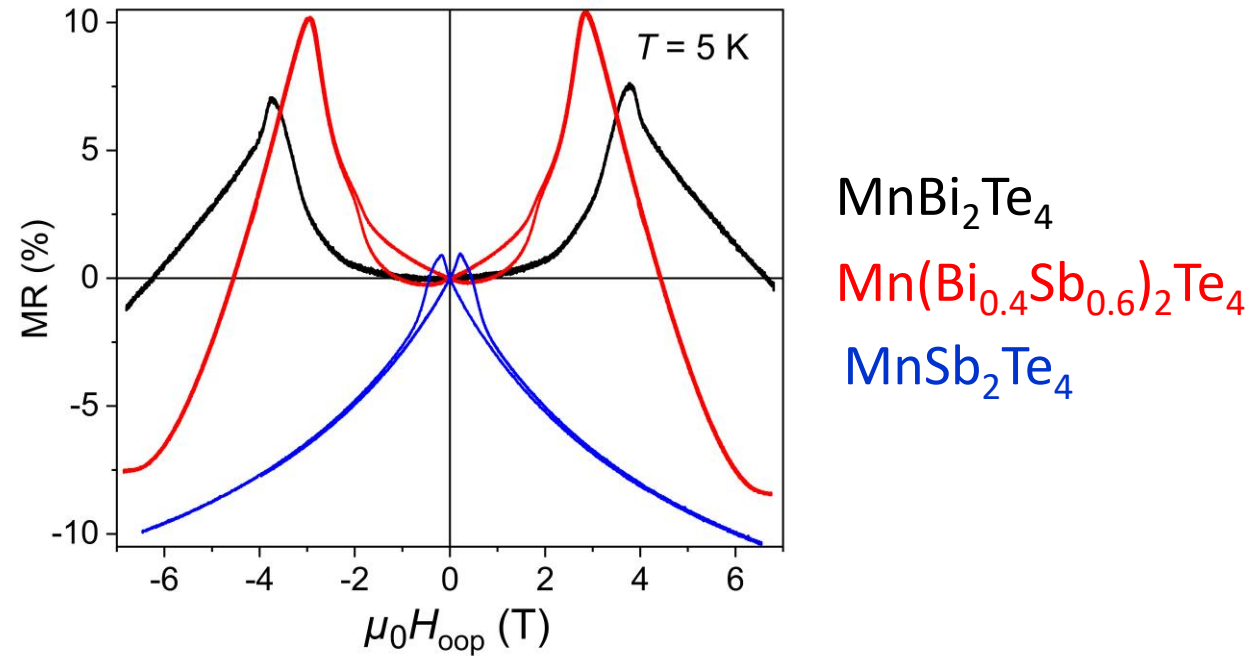
- Ferromagnetic ordering temperature $T_C = 55$ K
- Hysteretic butterfly magnetoresistance \rightarrow spin-dependent transport

MnSb₂Te₄ – magnetotransport AHE



- AHE \rightarrow pre-requisite for QAH in MTI
- $T_c = 55$ K \rightarrow QAHE expected $T > 4.2$ K

Mn(Bi,Sb₂)Te₄ – evolution of interactions



- MnBi₂Te₄ → antiferromagnetism
- Mn(Bi_{0.04},Sb_{0.06})₂Te₄ → canted antiferromagnetism
- MnSb₂Te₄ → ferromagnetism

Synopsis

■ $\text{Sn}_{0.92}\text{Mn}_{0.08}\text{Te}$

□ Remarkable $\theta_{\text{AH}} = 0.3$

■ MnSb_2Te_4

□ Intrinsic ferromagnetic TI $\rightarrow T_{\text{C}} = 55 \text{ K}$

Topology in condensed matter physics

■ Weyl equation for relativistic particles

$$H_{\pm} = \pm c \begin{bmatrix} p_z & p_x - ip_y \\ p_x + ip_y & -p_z \end{bmatrix}$$

- Solution → massless fermions with definite handedness or chirality
- Intended as model for elementary particles
 - 90 years → no Weyl fermions in high energy physics



H. Weyl [1929]



AUGUST 15, 1937

PHYSICAL REVIEW

VOLUME 52

Accidental Degeneracy in the Energy Bands of Crystals

CONYERS HERRING

Princeton University, Princeton, New Jersey

(Received June 16, 1937)

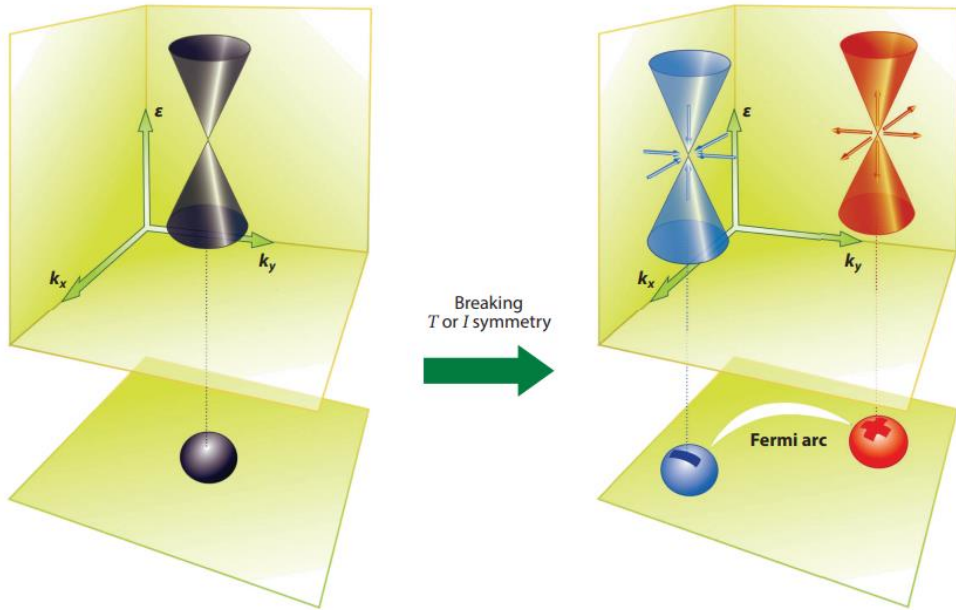
- Conditions for energy bands in solids to have the same energy
- Weyl equation in solid state physics

JYU → will Weyl fermions emerge in low energy physics?

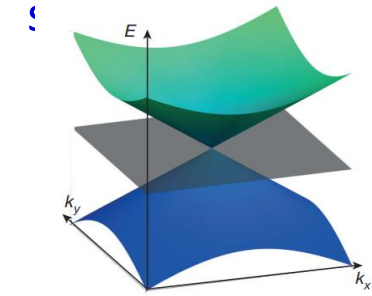


C. Herring [1937]

Weyl semimetals

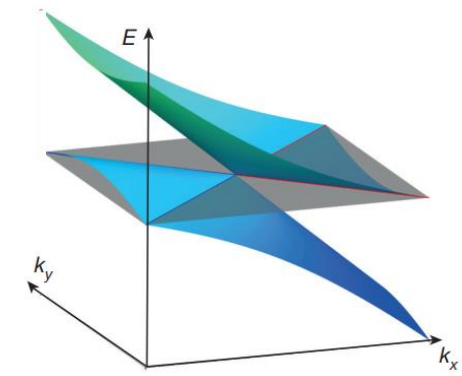


Type-I Weyl

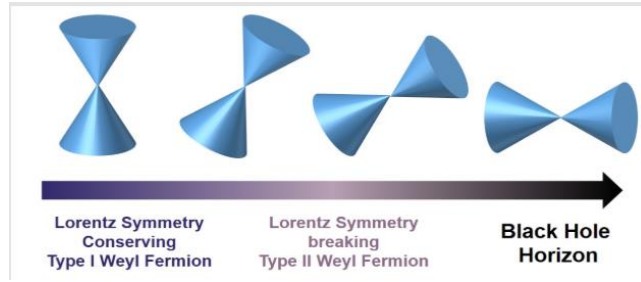


TaAs, NbAs, NbP

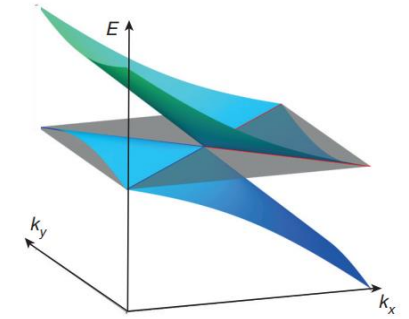
Type-II Weyl semimetals



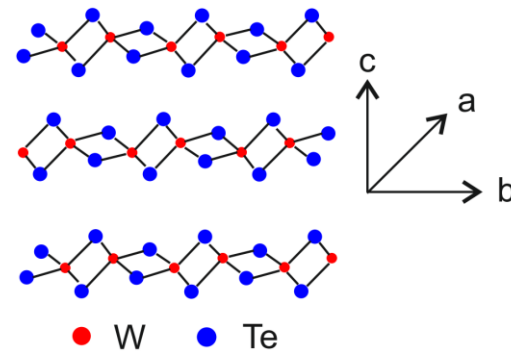
T_d -MoTe₂, T_d -WTe₂



Type II Weyl semimetal – T_d -WTe₂

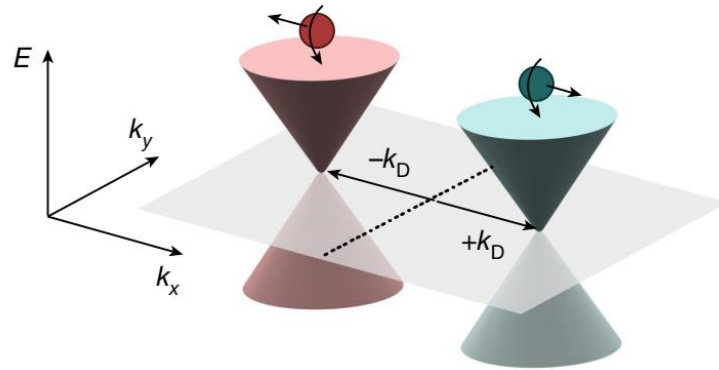


- Tilted electronic band structure
- Anisotropic magnetotransport
- Quantum anomalies – Adler-Bell-Jackiw or chiral anomaly
 - classical conservation laws broken at quantum level

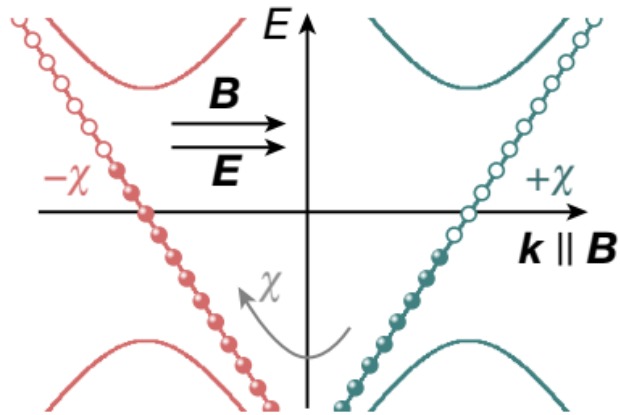


- van der Waals structure
- Insight into Weyl quasiparticles – massless, high mobility
- High-speed devices, THz photonics, emergent phenomena in 2D

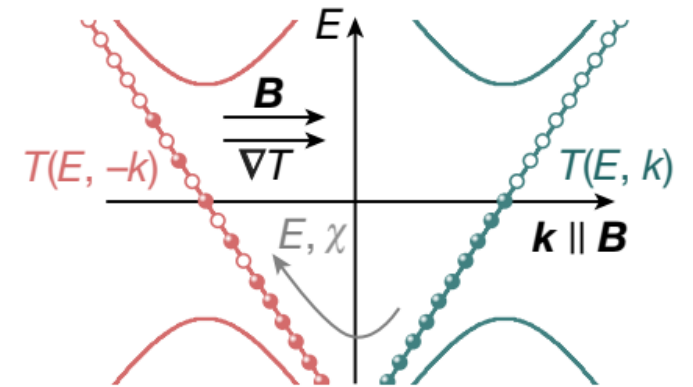
Weyl semimetal – quantum anomalies



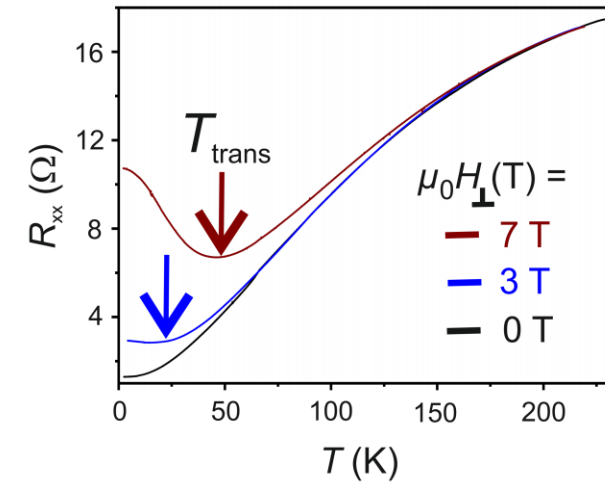
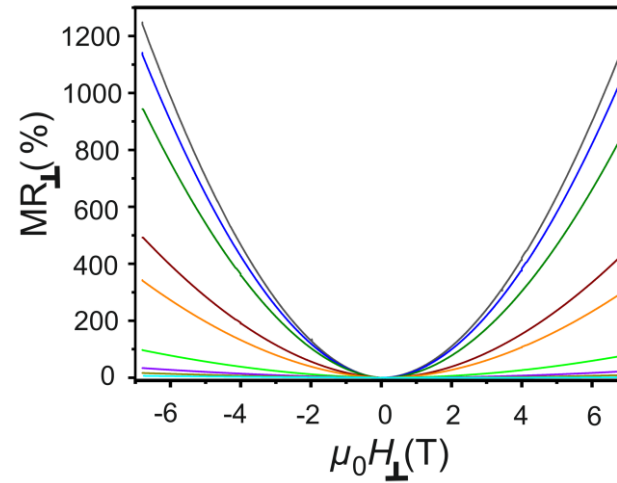
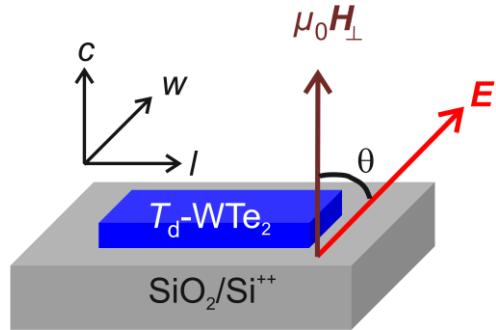
Chiral anomaly



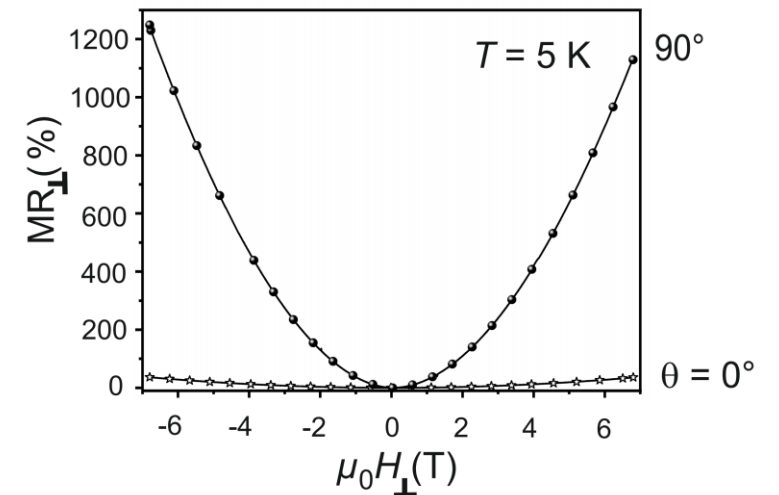
Axial-gravitational anomaly



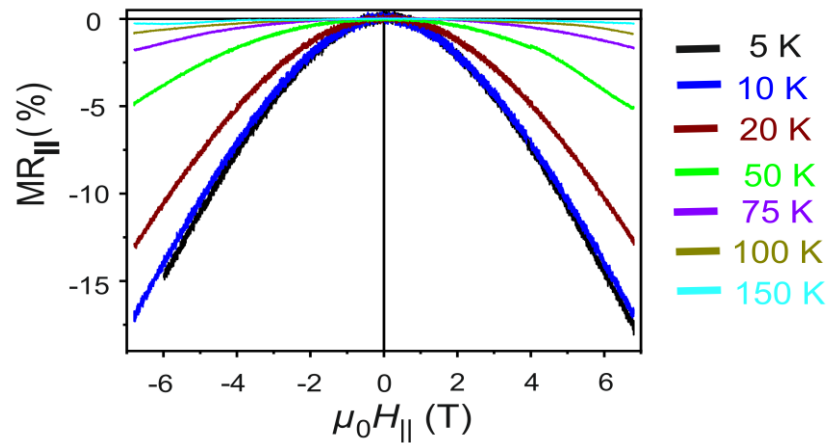
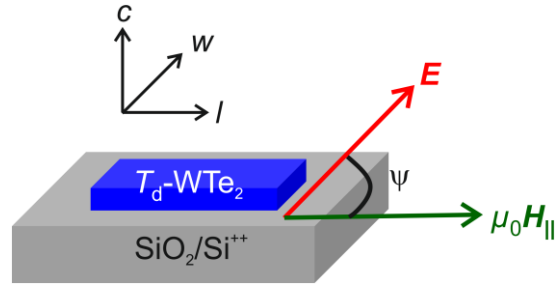
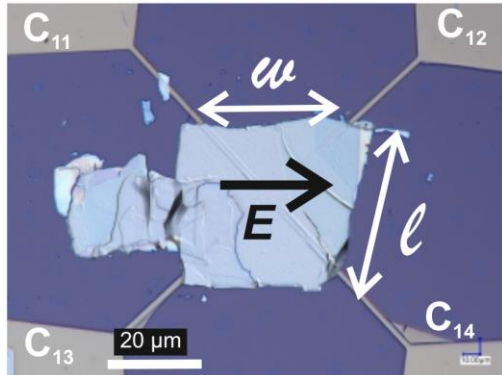
T_d -WTe₂ – perpendicular magnetic fields



- Fermi liquid below 50 K
- Significant positive MR
 - ~1200%, due to charge compensation
 - → electron and hole pockets
- Anisotropic electronic properties

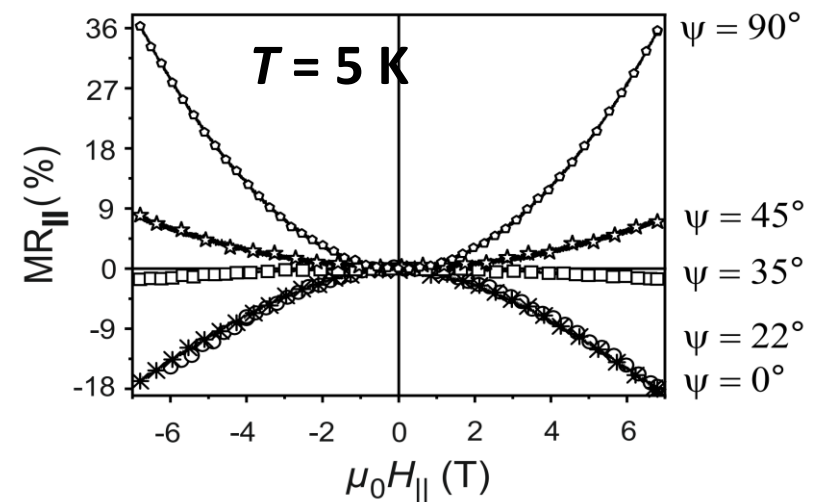
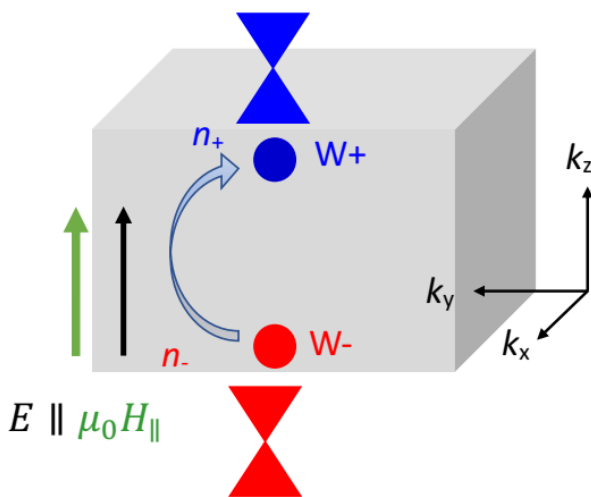
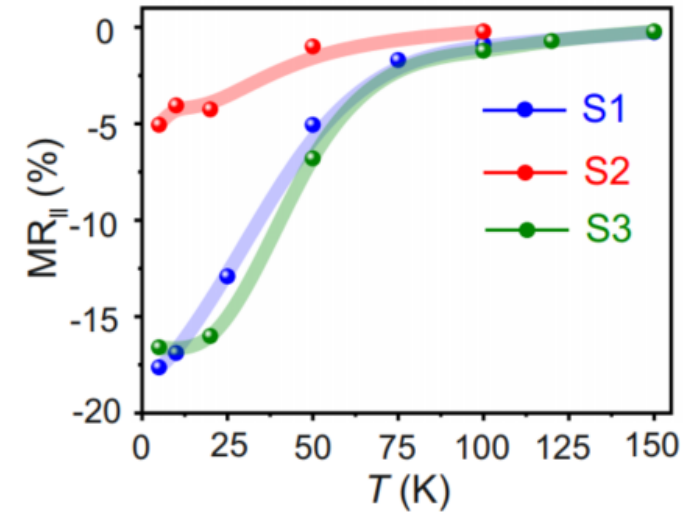


T_d -WTe₂ – parallel magnetic fields and quantum anomaly



$(E \parallel \mu_0 H_{\parallel}) \parallel b$

Chiral anomaly up to 100 K



Synopsis

■ T_d -WTe₂

- Large non saturating MR ~1200% at 5 K, 7 T
- Charge compensation
- Electron – hole pockets
- Chiral anomaly up to 100 K

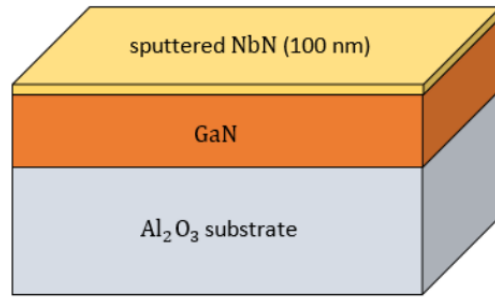
Back to superconductivity

- Perfect diamagnetism and zero resistivity
- Type I and type II superconductors
- Conventional and unconventional superconductors
- NBN: conventional, s-wave, BCS superconductor

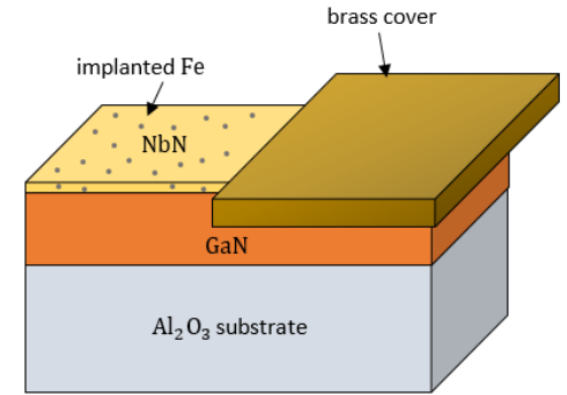
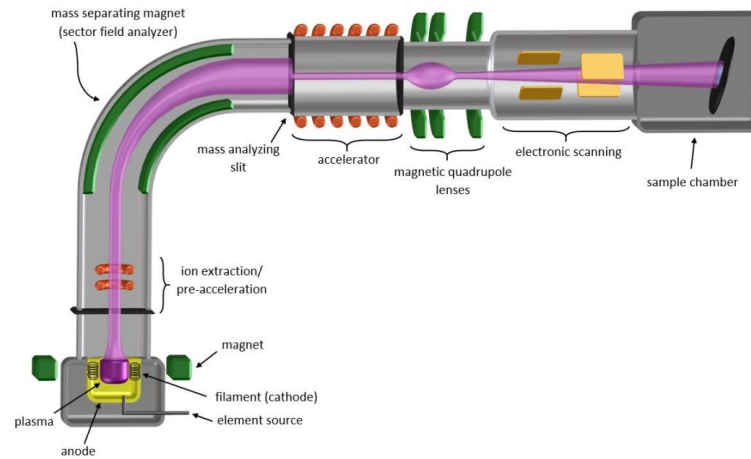
■ Why?

- Proximity effects and emergence
- Anderson-Higgs mechanism
- Yu-Shiba-Rusinov states
- Spin-triplet Cooper pairing
- Topological superconductivity
- Odd frequency superconductivity

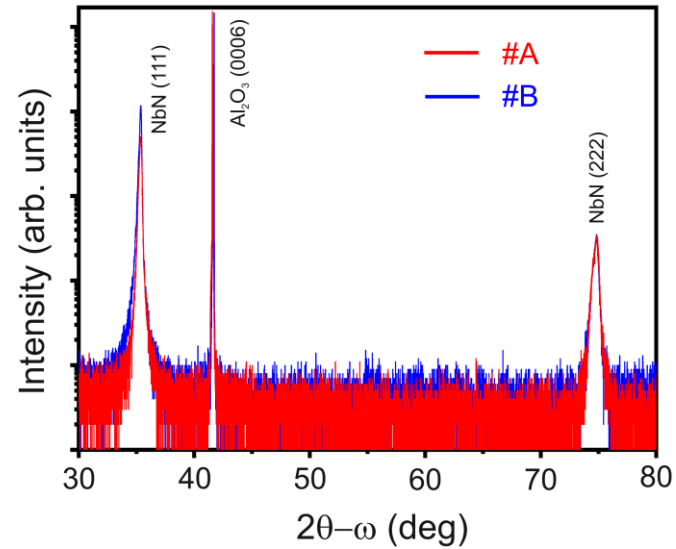
Fe implanted NbN



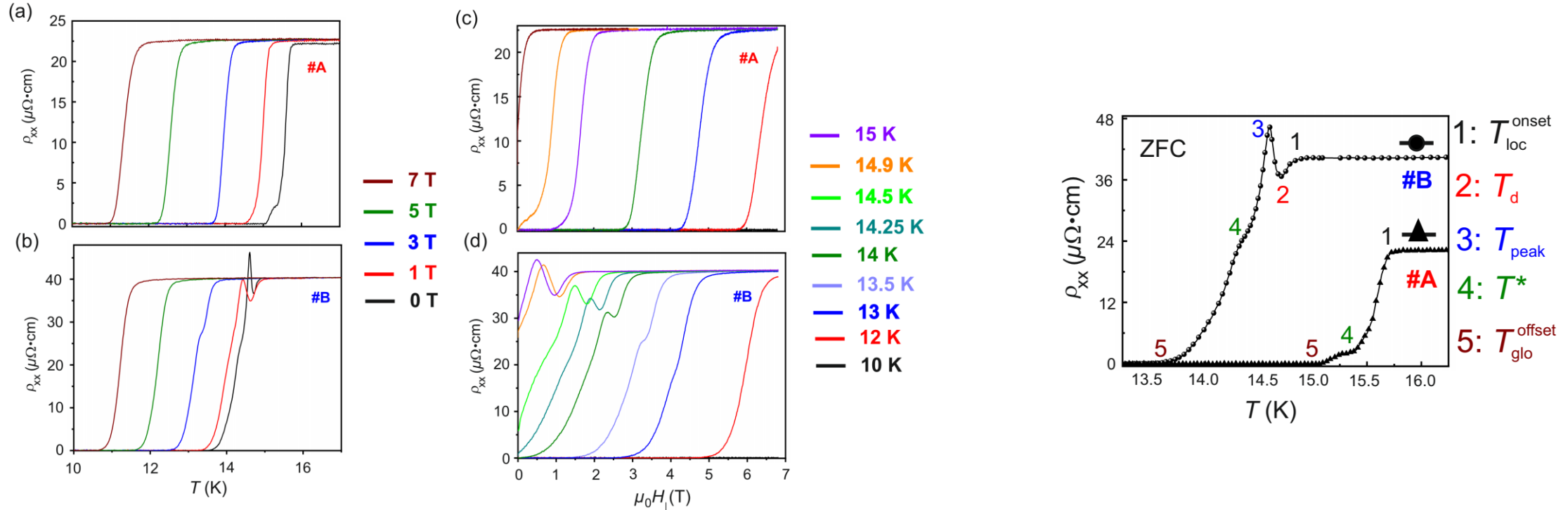
#A NbN



#B Fe:NbN



Fe implanted NbN - Magnetotransport



- $T_C \sim 15$ K
- Superconductivity persisting in Fe:NbN
- Re-entrant resistive behaviour in fe:NbN
- Bosonic islands percolation

Synopsis

■ Fe:NbN

- Superconductivity
- Emergence of bosonic insulator state
- N-shaped re-entrant resistive character of metal-to-superconductor transition