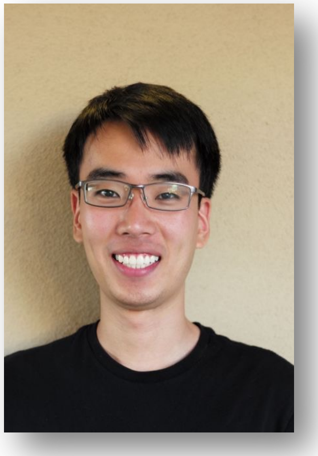
Topological States in Thin Films of Cadmium Arsenide

Susanne Stemmer
Materials Department, University of California, Santa Barbara

SPICE Workshop: “Non-Equilibrium Quantum Materials Design”
June 28, 2023

Acknowledgements



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Alex Lygo

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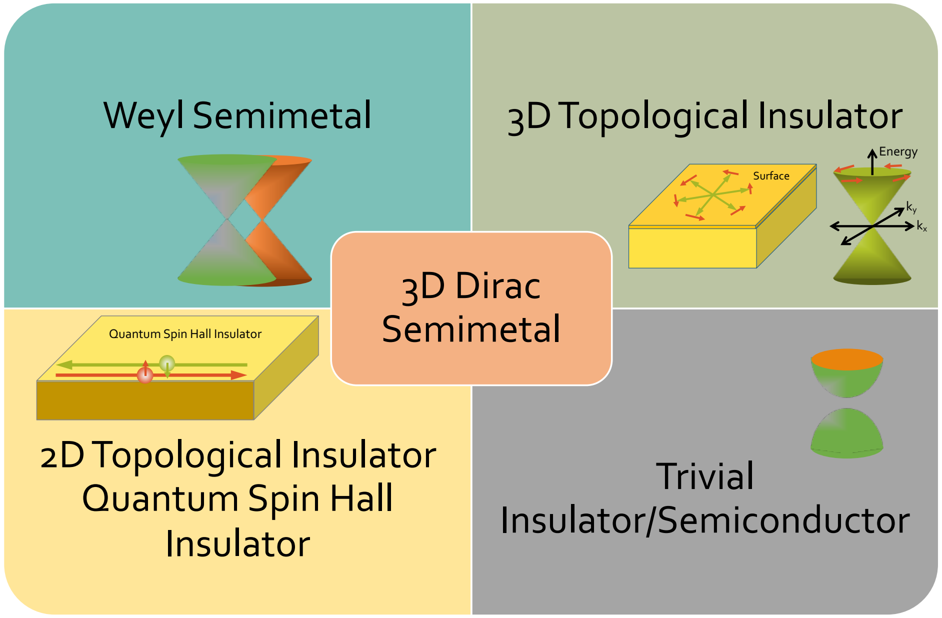
- Wangqian Miao (UCSB)

Air Force Office of Scientific Research
UCSB Quantum Foundry (NSF)
NSF Graduate Fellowships

Outline

- Cadmium arsenide films as a platform for creating different topological phases
- 2D TI state
- 2D Weyl semimetal
- Summary

Why topological semimetal thin films?



Weyl Semimetal

3D Topological Insulator

3D Dirac Semimetal

2D Topological Insulator Quantum Spin Hall Insulator

Trivial Insulator/Semiconductor

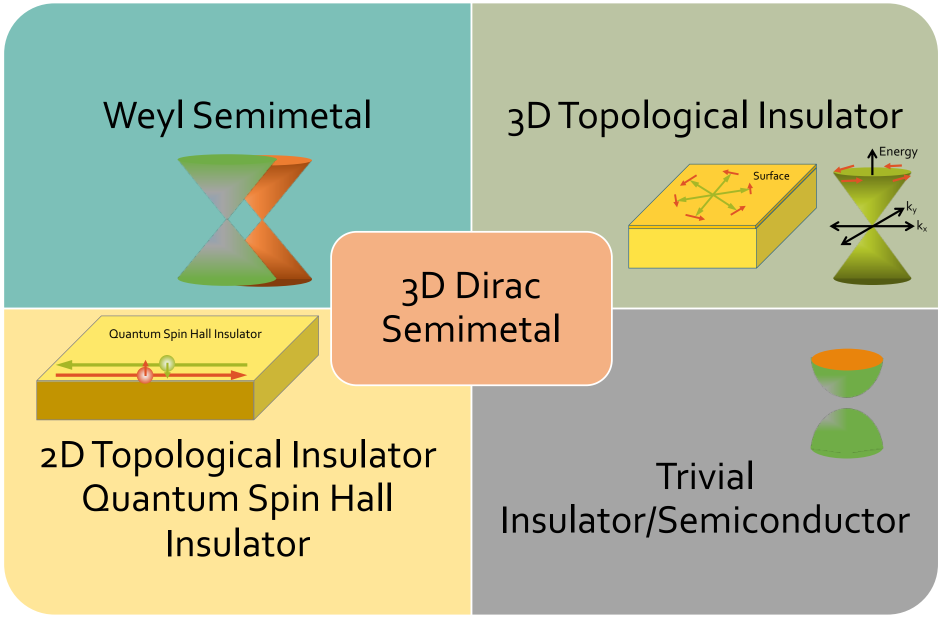
S. M. Young et al., Phys. Rev. Lett. 108, 140405 (2012).

3D Dirac semimetal thin films can be used to tune between topological phases, and manipulate and control their electronic states

Why topological semimetal thin films?

No "ideal" WSMs: many nodes, trivial bands crossing Fermi level obscure physics

Few good QSHI candidates & lots of open questions about the existing QSHI



Weyl Semimetal

3D Topological Insulator

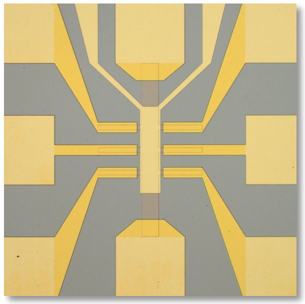
3D Dirac Semimetal

2D Topological Insulator
Quantum Spin Hall Insulator

Trivial
Insulator/Semiconductor

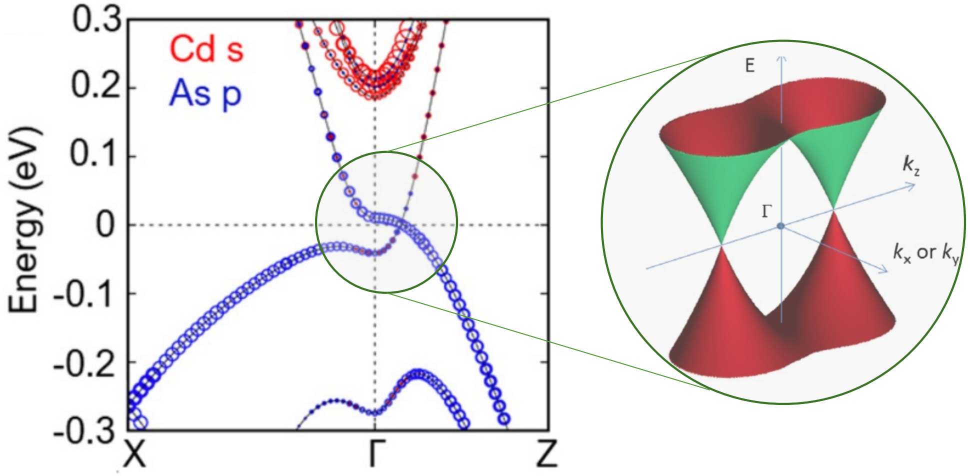
S. M. Young et al., Phys. Rev. Lett. 108, 140405 (2012).

2D materials allow control via electric field



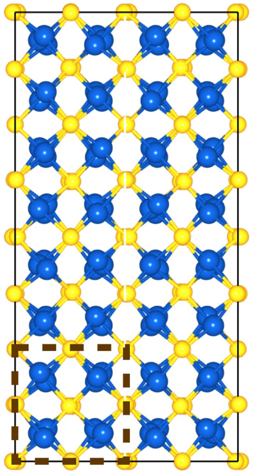
3D Dirac semimetal thin films can be used to tune between topological phases, and manipulate and control their electronic states

Three-dimensional Dirac Semimetal: Cadmium Arsenide (Cd_3As_2)

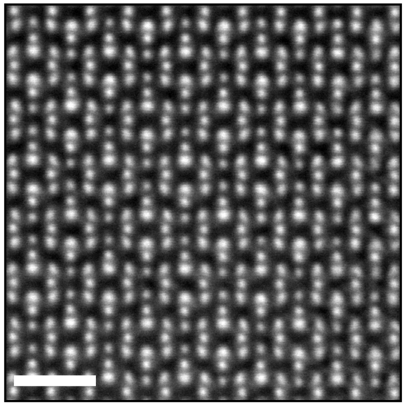


S. Baidya, D. Vanderbilt, Phys. Rev. B 102, 165115 (2020).

$I4_1/acd$



Antifluorite-related structure with
25% vacancies on the Cd sites

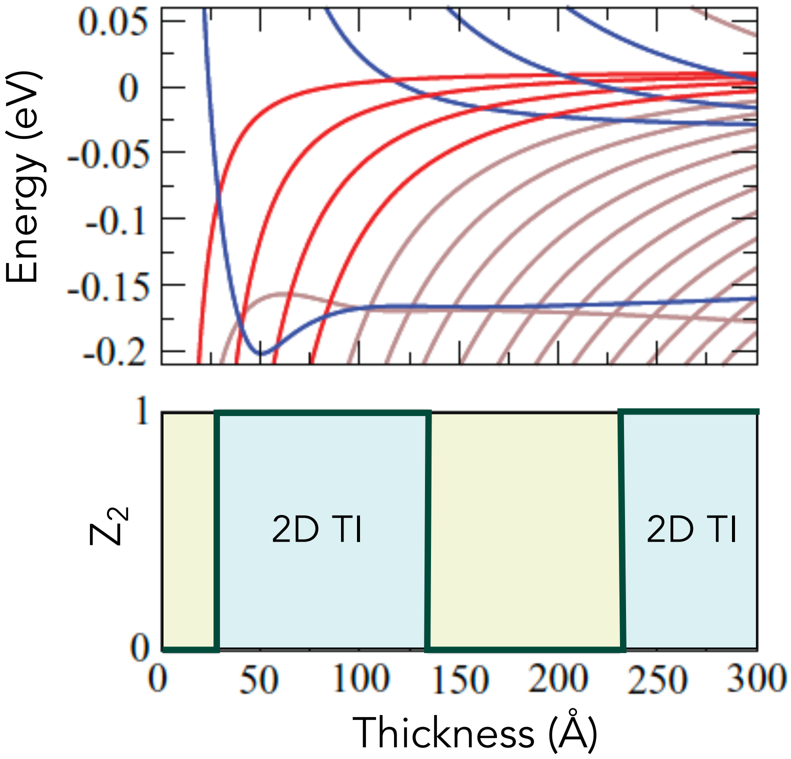


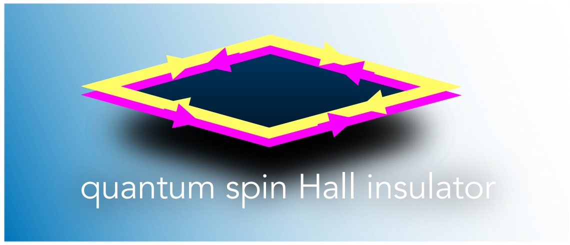
ADF-STEM

- Band inversion between Cd-5s and As-4p states at **Γ**
- Two crossings along **Γ**-Z protected by 4-fold symmetry
- High Fermi velocity
- High carrier mobilities

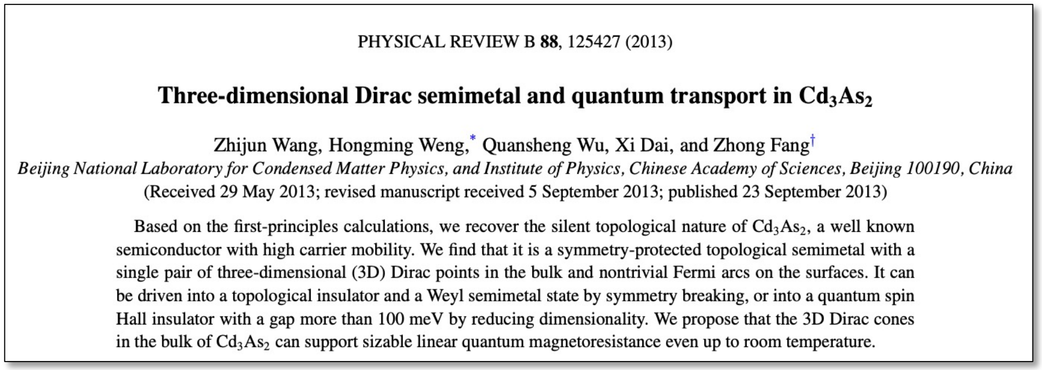
Wang et al., Phys. Rev. B 88, 125427 (2013).
Ali et al., Inorg. Chem. 53, 4062 (2014).
Borisenko, et al. Phys. Rev. Lett. 113, 165109 (2014).
Neupane et al., Nat. Commun. 5, 3786 (2014).
Liu et al., Nat. Mater. 13, 677 (2014).
Jeon et al., Nat. Mater. 13, 851 (2014).
Mosca Conte et al., Sci. Rep. 7, 45500 (2017).
A. J. Rosenburg, T. C. Harman, J. of Appl. Phys. 30, 1621 (1959).

Topological States of Cadmium Arsenide: Quantum Spin Hall Insulator





“A 5 nm thick Cd_3As_2 film is a good QSH insulator with a gap of more than 100 meV”


PHYSICAL REVIEW B **88**, 125427 (2013)

Three-dimensional Dirac semimetal and quantum transport in Cd_3As_2

Zhijun Wang, Hongming Weng,* Quansheng Wu, Xi Dai, and Zhong Fang†
Beijing National Laboratory for Condensed Matter Physics, and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

(Received 29 May 2013; revised manuscript received 5 September 2013; published 23 September 2013)

Based on the first-principles calculations, we recover the silent topological nature of Cd_3As_2, a well known semiconductor with high carrier mobility. We find that it is a symmetry-protected topological semimetal with a single pair of three-dimensional (3D) Dirac points in the bulk and nontrivial Fermi arcs on the surfaces. It can be driven into a topological insulator and a Weyl semimetal state by symmetry breaking, or into a quantum spin Hall insulator with a gap more than 100 meV by reducing dimensionality. We propose that the 3D Dirac cones in the bulk of Cd_3As_2 can support sizable linear quantum magnetoresistance even up to room temperature.

QSHIs are rare

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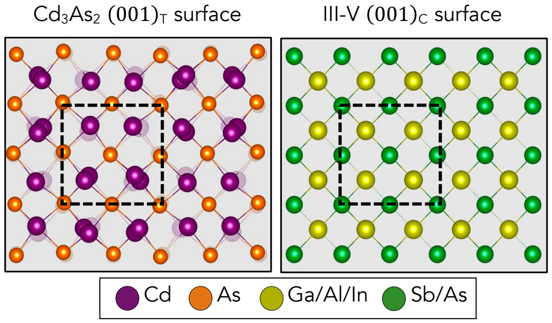
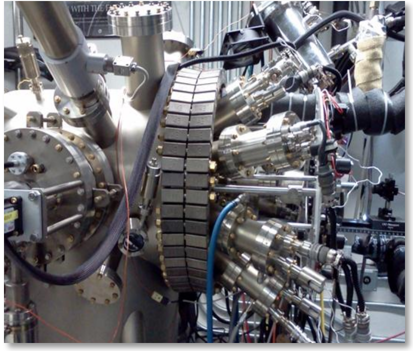
Molecular Beam Epitaxy of (001)Cd$_3$As$_2$



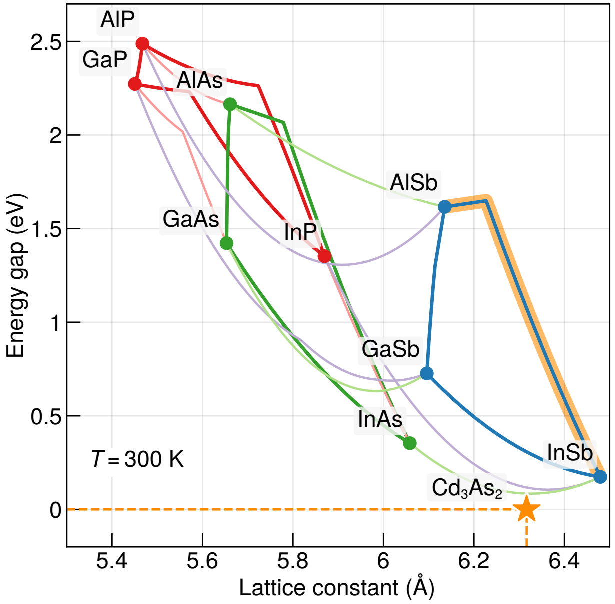
Applied Epi Modified GEN 2

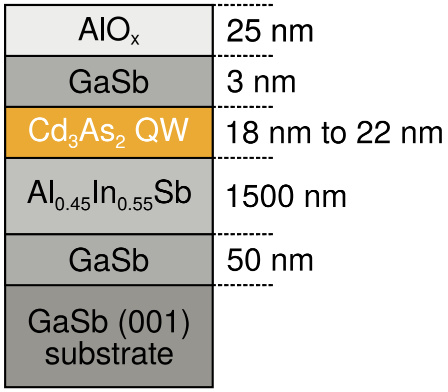


T. Schumann et al., APL Mater. 4, 126110 (2016).
T. Schumann et al., Phys. Rev. B 95, 241113(R) (2017).
D. A. Kealhofer et al. Phys. Rev. Mater. 3, 031201(R) (2019).
M. Goyal et al., APL Mater., 8, 051106 (2020).

High quality epitaxy requires:

- Symmetry matched + chemically similar substrate
- GaSb substrate + buffer layer: Al$_{0.45}$In$_{0.55}$Sb ternary alloy
- GaSb cap

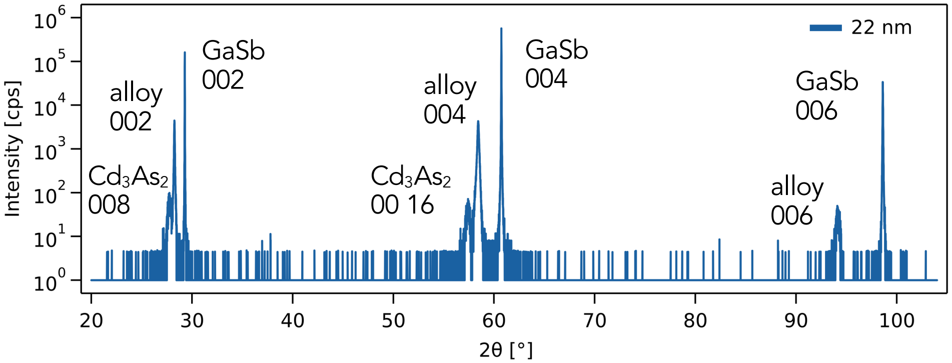




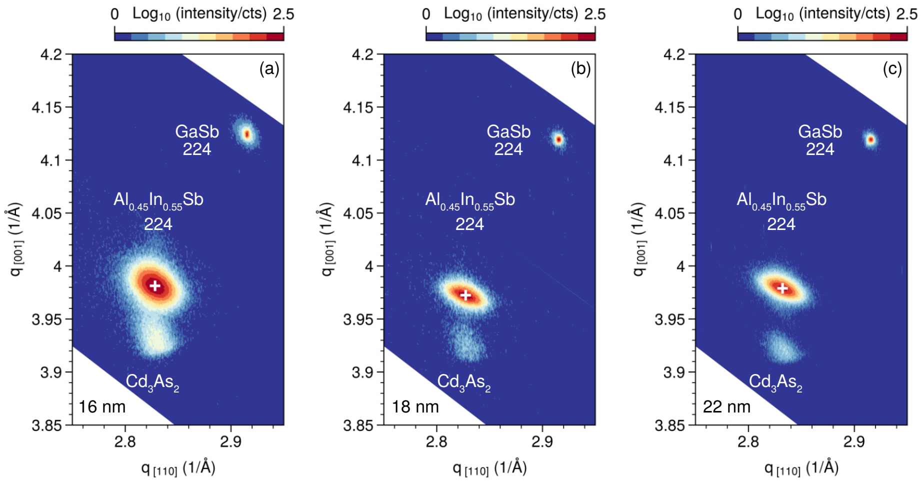
AlO$_x$	25 nm
GaSb	3 nm
Cd$_3$As$_2$ QW	18 nm to 22 nm
Al$_{0.45}$In$_{0.55}$Sb	1500 nm
GaSb	50 nm
GaSb (001) substrate	

Molecular Beam Epitaxy of (001)Cd_3As_2

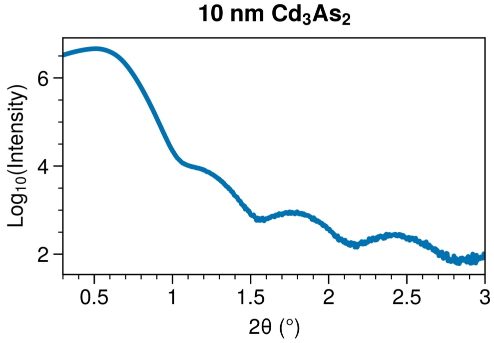
Wide-angle, out-of-plane 2**θ**-**ω** scan



Reciprocal space maps show that Cd_3As_2 is coherently strained to buffer alloy



X-ray reflectivity: smooth Cd_3As_2 layers even at 10 nm



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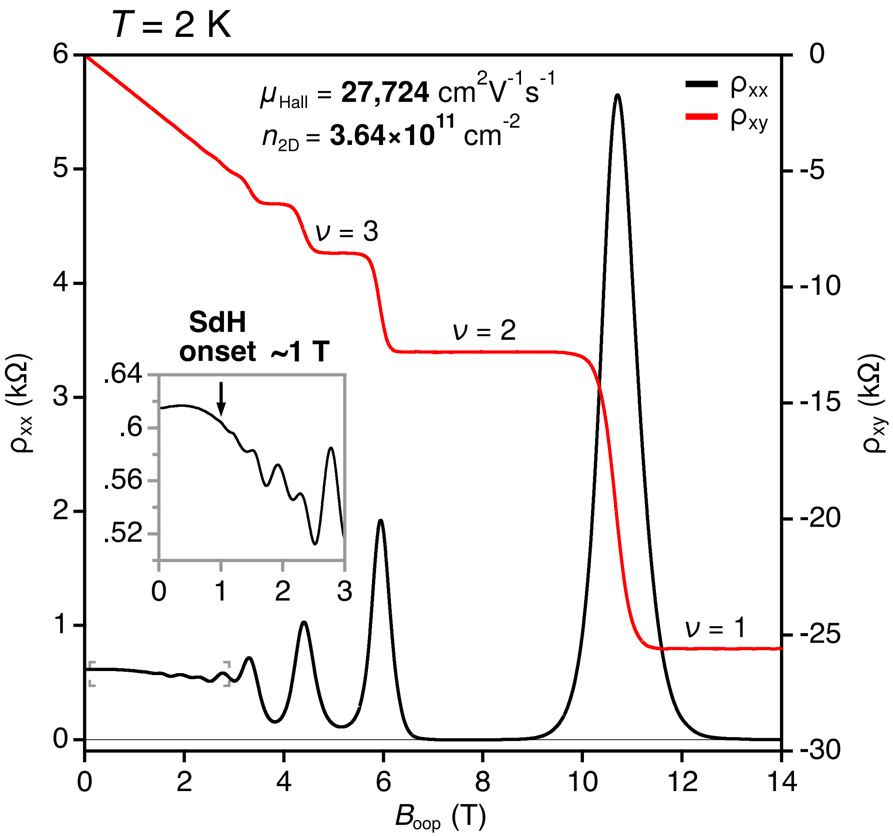
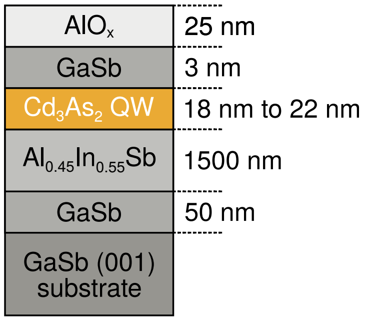
B. Guo et al., unpublished

Molecular Beam Epitaxy of (001)Cd_3As_2





AlO_x	25 nm
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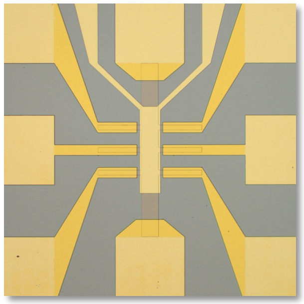
GaSb (001) substrate	

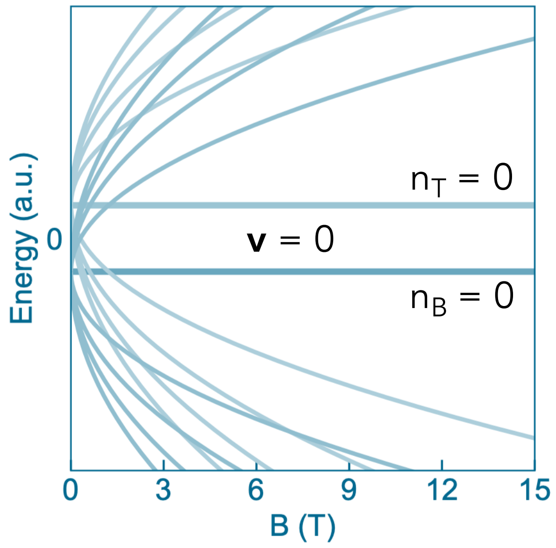


- Lithographically defined Hall bar
- Around zero field: Hall mobility, area density
- Low field: estimate quantum mobility from onset of Shubnikov-de Haas oscillations at 1 T
- High field: formation of Landau levels + quantized plateaus in Hall resistivity, with simultaneously vanishing longitudinal resistivity

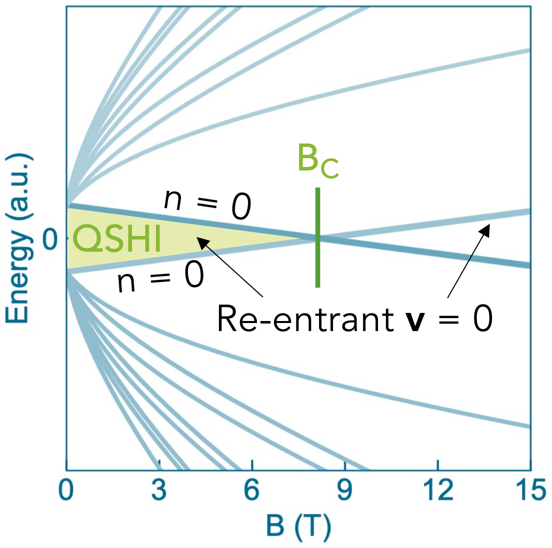
B. Guo et al., unpublished

Topological Insulators in a Perpendicular Magnetic Field

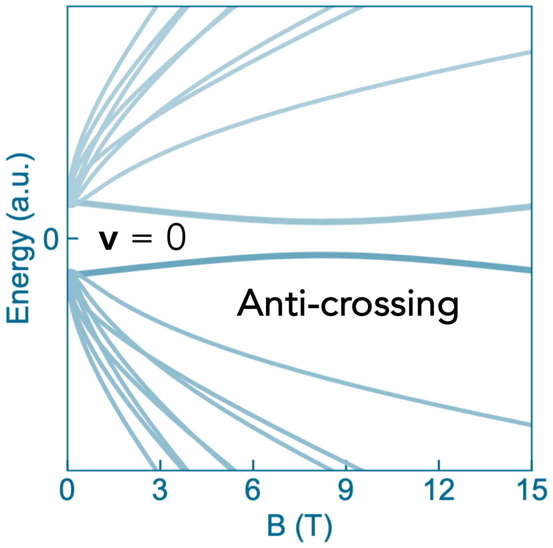
3D Topological Insulator
with energy offset
between the top and
bottom surfaces



2D Topological Insulator
no SIA



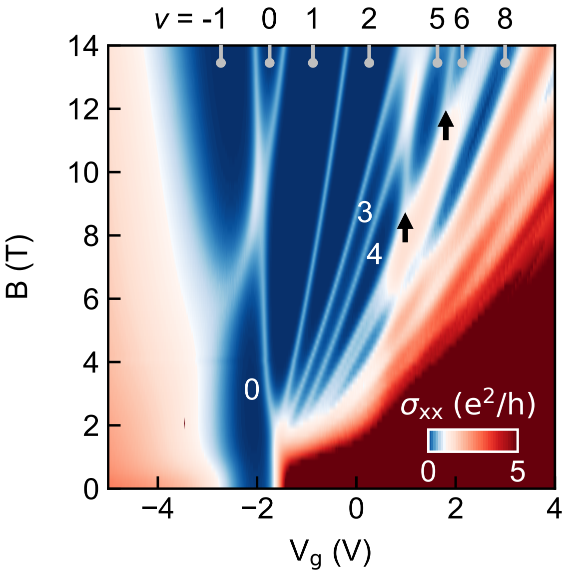
2D Insulator
(un-inverted due to SIA)



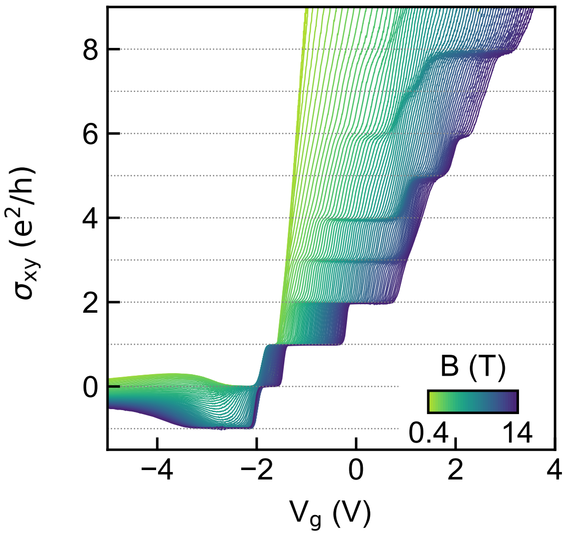
C.-X. Liu et al., Phys. Rev. B 81, 041307R (2010); H.-Z. Lu et al., Phys. Rev. B 81, 115407 (2010); J. Linder et al., Phys. Rev. B 80, 205401 (2009).

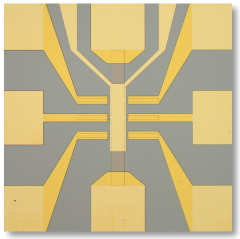
Landau levels and quantum Hall effect in a 20 nm film

Color map of $\boldsymbol{\sigma}_{xx}$ shows the
evolution of Landau levels
with field and gate voltage



Quantum Hall plateaus





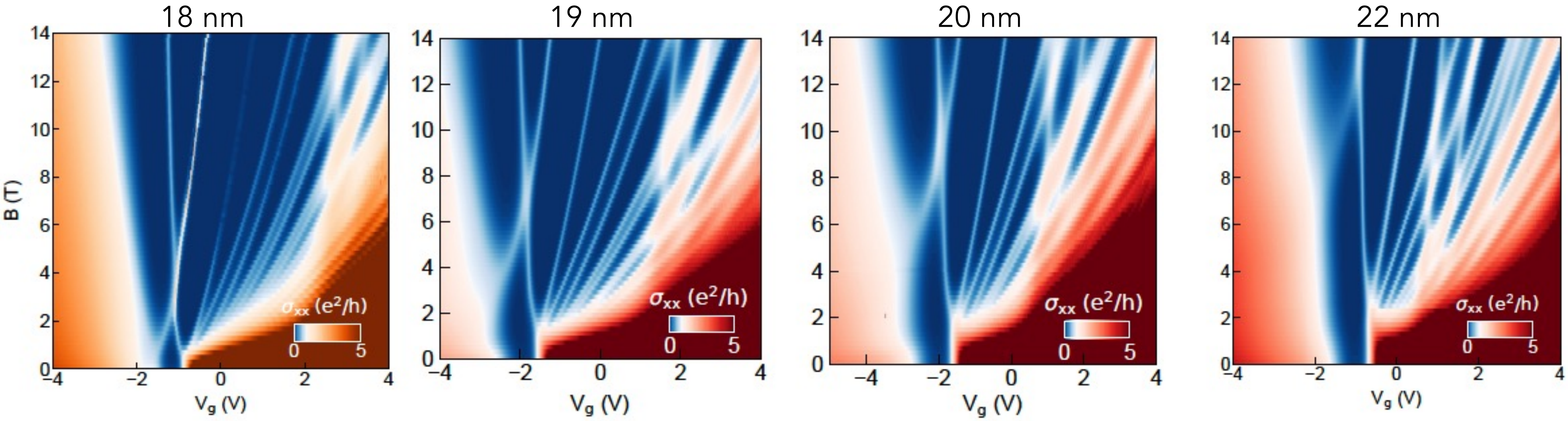
Gated Hall bar

- Signature of inverted spectrum: crossing of n = 0 Landau levels at a critical field
- Re-entrant **v** = 0 plateau due to crossing of zeroth Landau levels
- Additional higher energy subband

A. C. Lygo et al., Phys. Rev. Lett. 130, 046201 (2023).

Landau Level Spectrum: Thickness Dependence

Color maps of $\boldsymbol{\sigma}_{xx}$ show the evolution of Landau levels



- Signature of inverted spectrum: crossing of n = 0 Landau levels at a critical field
- Re-entrant **ν** = 0 plateau due to crossing of zeroth Landau levels
- Systematic behavior with thickness
- Second conduction subband moves out to higher energies with decreasing thickness

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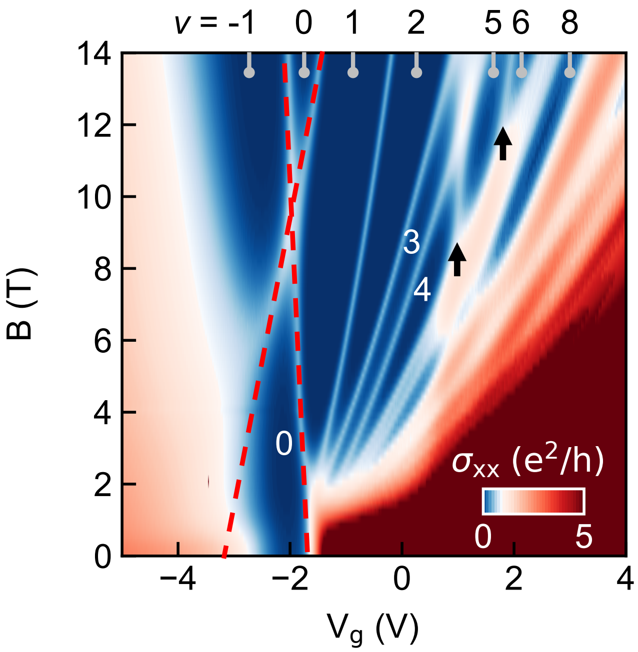
A. C. Lygo et al., Phys. Rev. Lett. 130, 046201 (2023).

$$B_c = \frac{M}{2\pi B/\Phi_0 - (g_e + g_h)\mu_B/4}$$

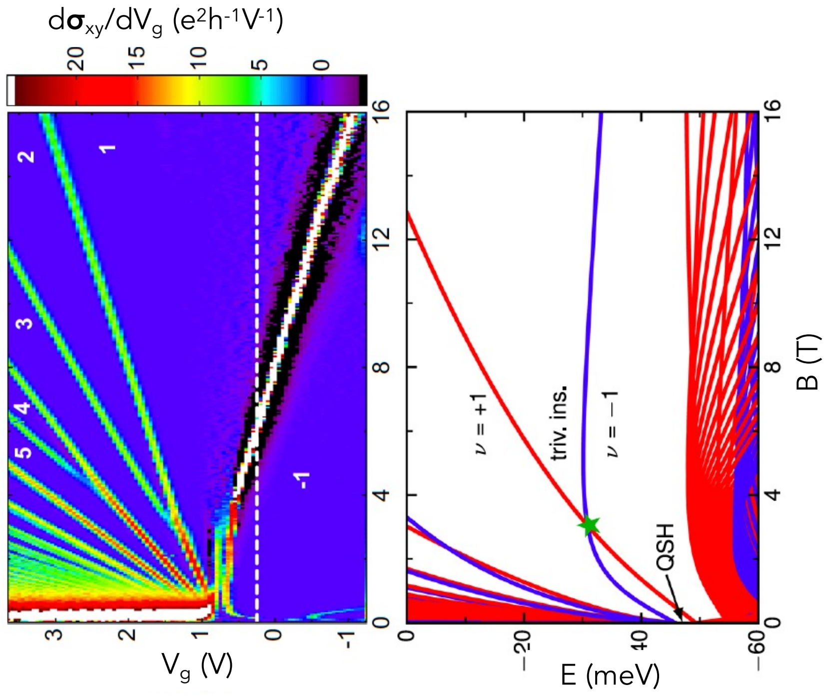
Scharf et al., PRB 86, 075418 (2012)

Comparison with HgTe Quantum Wells

20 nm Cd_3As_2



11 nm Mn:HgTe QW



Experiment

Calculation

S. Shamim, …., L. W. Molenkamp, Nat. Comm. 13, 2682 (2022)

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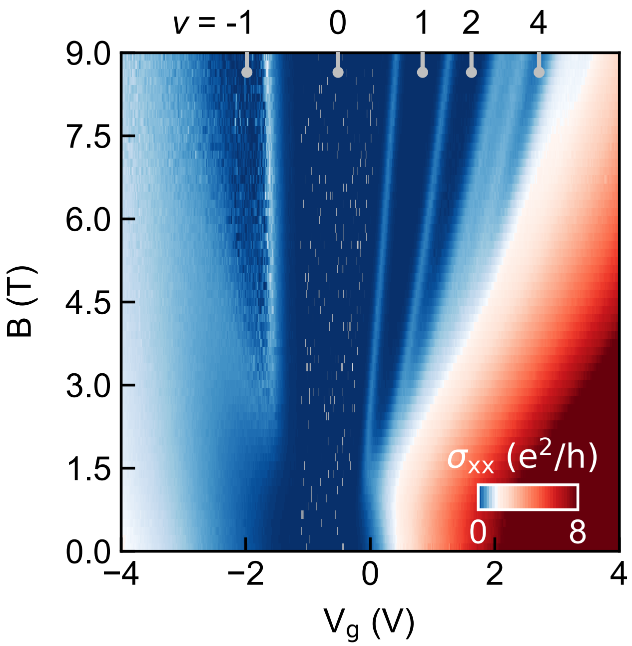
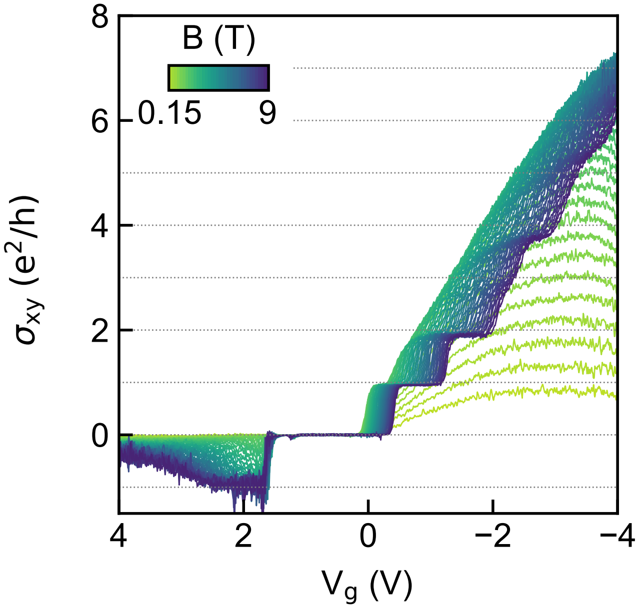
Trivial 2D Insulator: 14 nm film





Electron and hole-like zeroth Landau levels are “un-inverted” and do not cross

Film thickness can be used to tune between topological and trivial states, just as predicted

A. C. Lygo, et al., Phys. Rev. Lett. 130, 046201 (2023).

Cadmium arsenide as a novel two-dimensional topological insulator

- “Textbook” 2D TI seen in LL spectroscopy
- Should host helical edge states
- Mobilities are high for a near surface electronic system
- Facilitates hybrid structures with superconductors
- Tunability by strain and film thickness
- Good platform for novel quantum devices

Two-dimensional topological insulator in an in-plane magnetic field

- Out-of-plane field is responsible for the orbital effect and Landau quantization
- In-plane magnetic field: main effect on energy bands is the Zeeman energy:

$$\Delta E_Z \sim g\mu_B B_{ip}$$

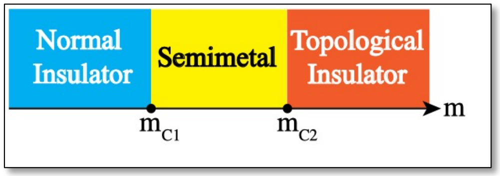
- The g-factor is sample/material specific
- In narrow-gap semiconductors, like a 2D TI, the Zeeman field may be sufficient to close the band gap

S. Sun,… X. Dai, Phys. Rev. B 106, L241105 (2022).
F. Dominguez, … E. M. Hankiewicz, SciPost Phys. Core 5, 024 (2022).
O. E. Raichev, Phys. Rev. B 85, 045310 (2012).
M. Kubisa, K. Ryczko, Phys. Rev. B 104, L161406 (2021).
A. A. Zyuzin, … A. A. Burkov, Phys. Rev. B 83, 245428 (2011).

Symmetry considerations for a gapless phase in Cd_3As_2:

- Zero-field crystallographic point group: 4/mmm
- With in-plane field, magnetic point group: 2′/m′

2D WSM for C_2T symmetry

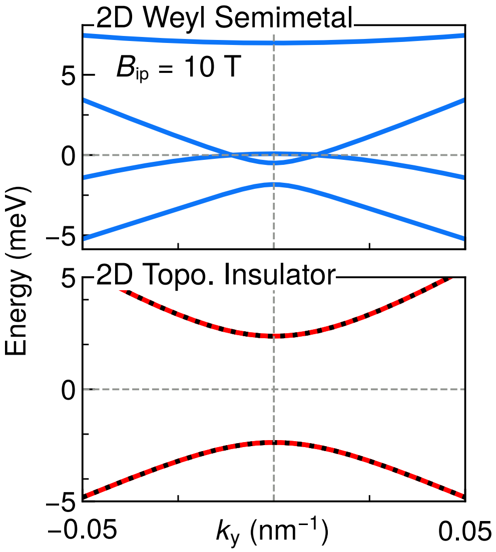


J. Ahn, B. J. Yang, PRL 118, 156401 (2017).

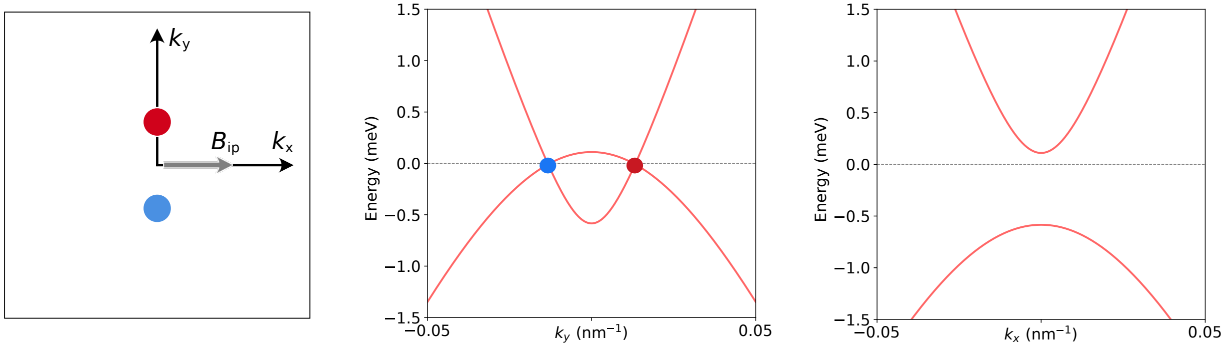
C_2T symmetry guarantees that the Berry curvature to be zero everywhere except at the Weyl points.

Two-dimensional topological insulator in an in-plane magnetic field

Ideal 2D Weyl semimetal, with two nodes at the charge neutrality level



B. Guo et al., submitted



Calculations by Wangqian Miao and Xi Dai for an 18 nm (001) Cd_3As_2 film

Requires accurate *g*-factor calculations that consider the combined effects of remote bands and quantum confinement*

* Z. Song,… X. Dai, arXiv:1512.05084 (2019).
S. Sun,… X. Dai, Phys. Rev. B 101, 125118 (2020).

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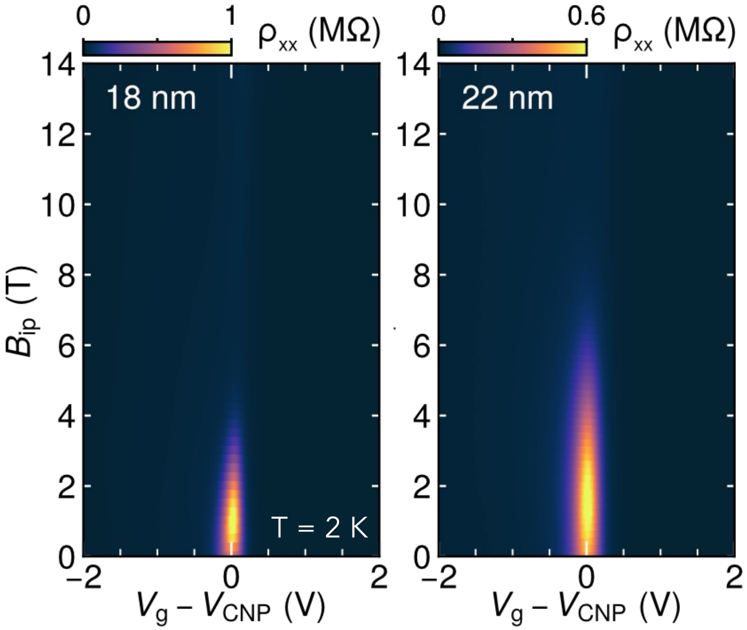
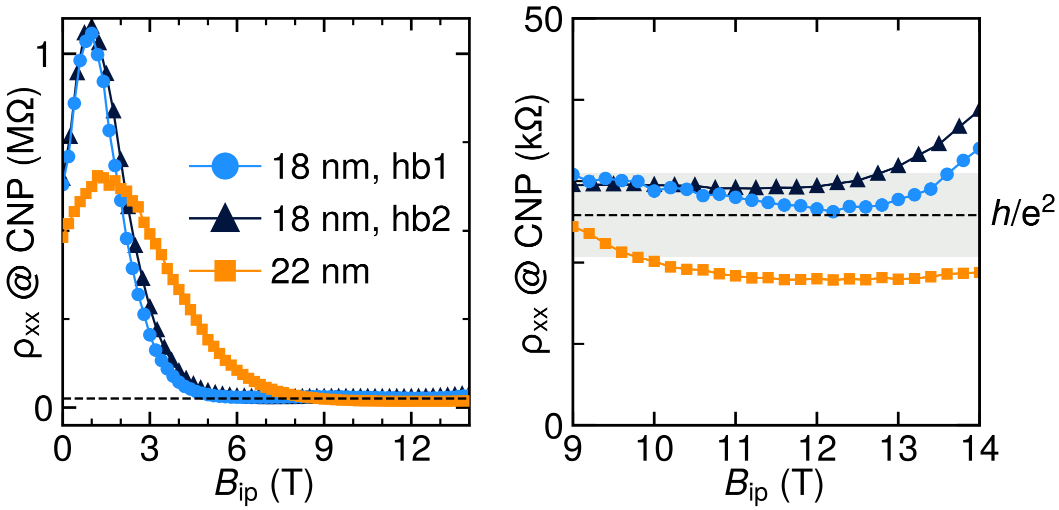
Zeeman field-induced 2D Weyl Semimetal: Experiment

Gap closing and metallic phase

- Non-monotonic evolution of peak resistivity with in-plane field
- Band gap closing: sharp drop past 1 T



- Initial increase and peak possibly related to current-carrying edge states
- Resistivities on the order of ~h/e^2 in a wide range of B-fields
- Evidence for the symmetry-protection of the metallic phase

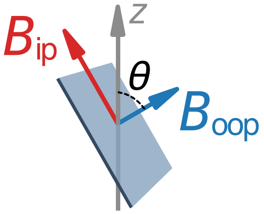


Peak resistivities at the charge neutrality point

B. Guo et al., submitted

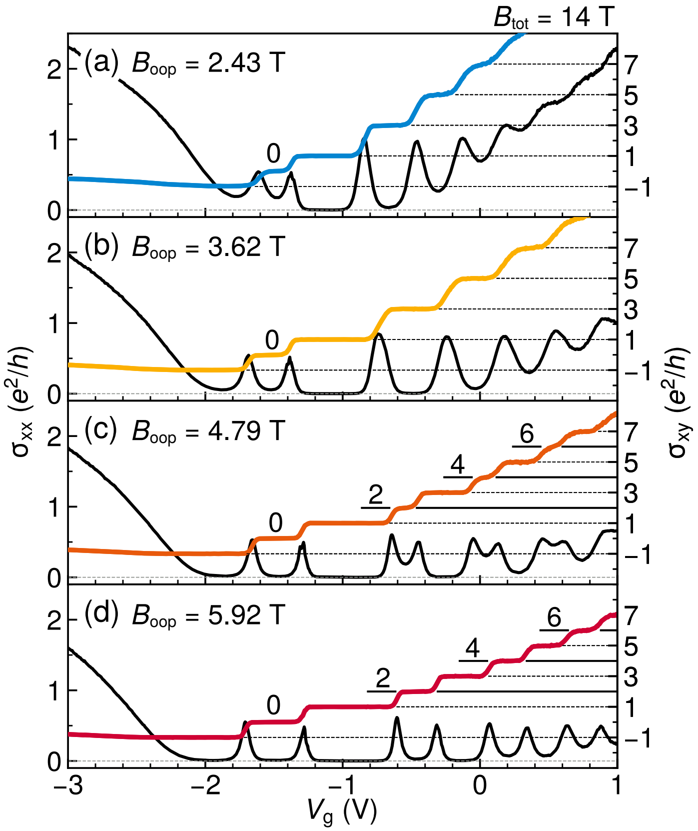
Zeeman field-induced 2D Weyl Semimetal: Experiment

Odd-integer quantum Hall effect



Sample:

- 18 nm QW
- T = 2 K



- The two n = 0 Landau levels are spin resolved
- Connect the ν = 0 and ν = ± 1 plateaus
- $|n|$ > 1 Landau levels are doubly degenerate and produce an odd integer quantum Hall effect
- Odd-integer QHE implies the existence of a pair of massive Weyl fermions: **ν = (n + ½) per node**
- Valley degeneracy is lifted at higher out-of-plane fields; requires further investigation

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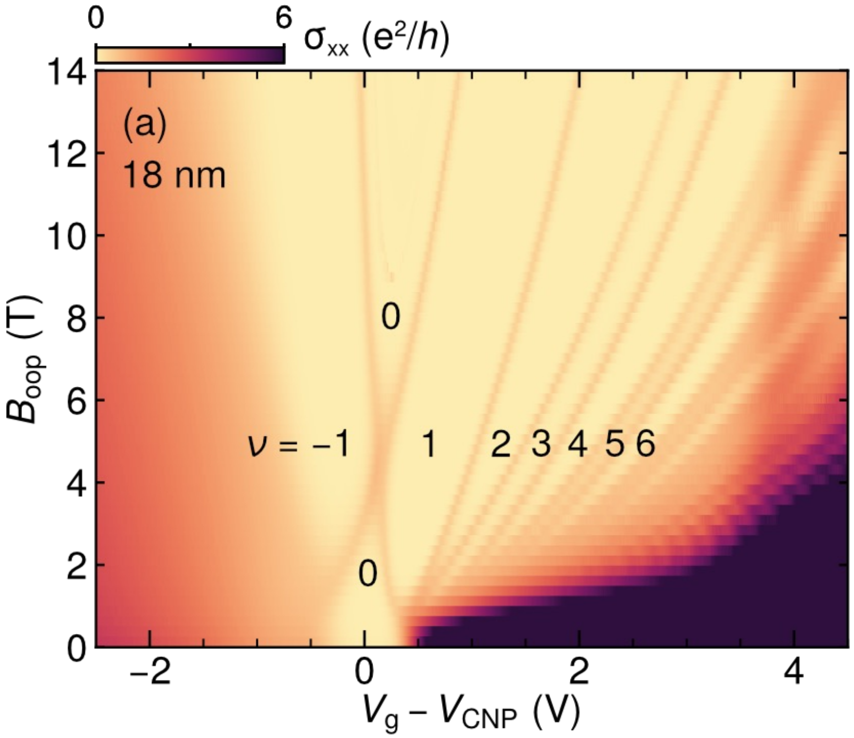
B. Guo et al., submitted

Comparison with oop field experiments

Out-of-plane magnetic field, no in-plane field: 2D TI phase
QHE with all degeneracies are lifted

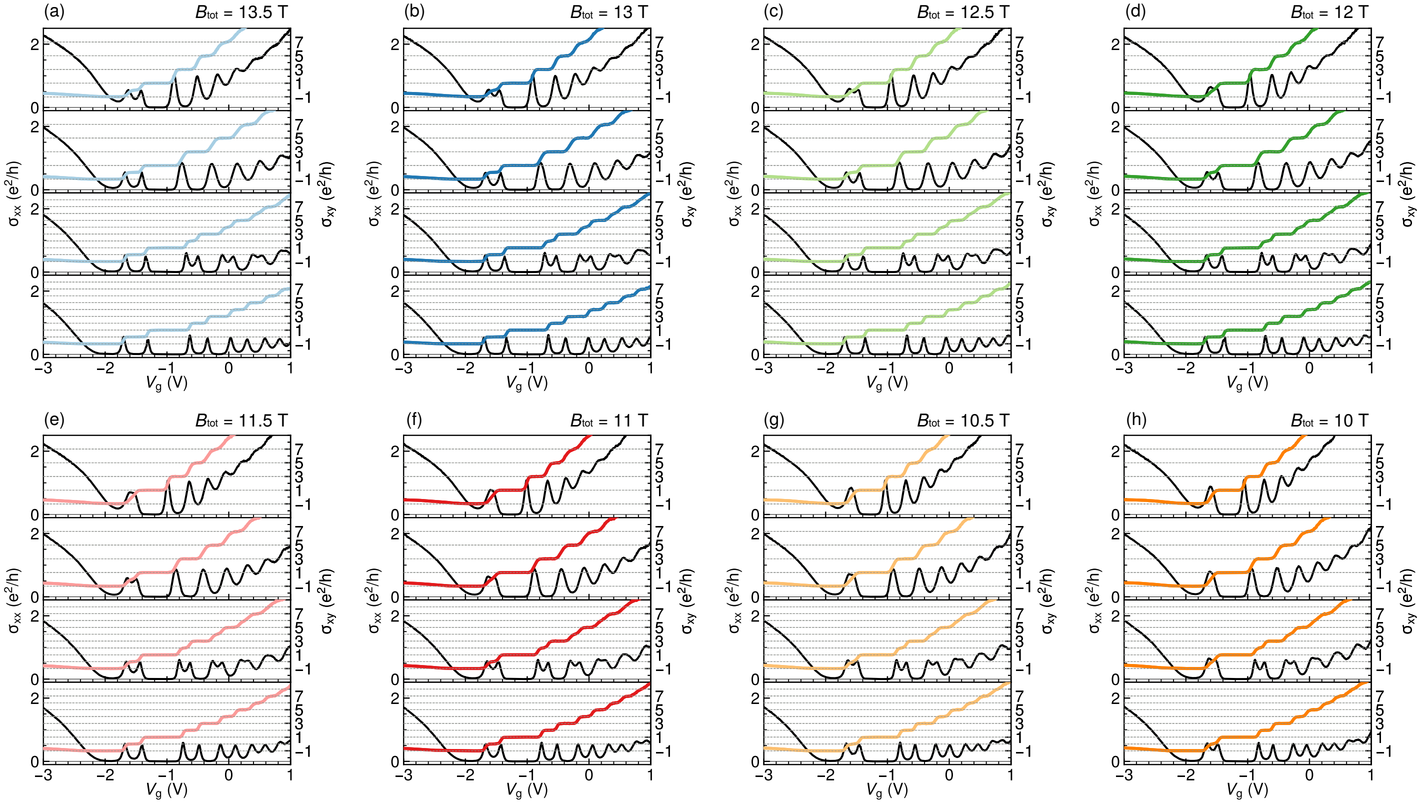


- The filling factor **v** = 0 state appears twice because of crossing Landau levels
- All degeneracies are lifted: even and odd integer filling factors
- Spin degeneracy is immediately lifted

B. Guo et al., submitted

Zeeman field-induced 2D Weyl Semimetal: Experiment

Odd-integer quantum Hall effect



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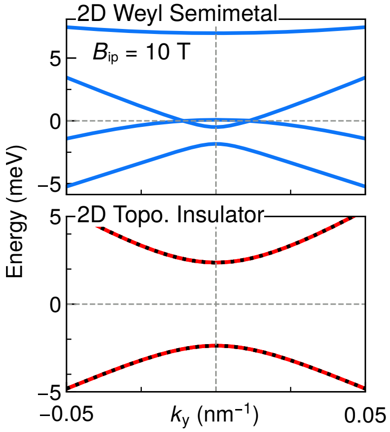
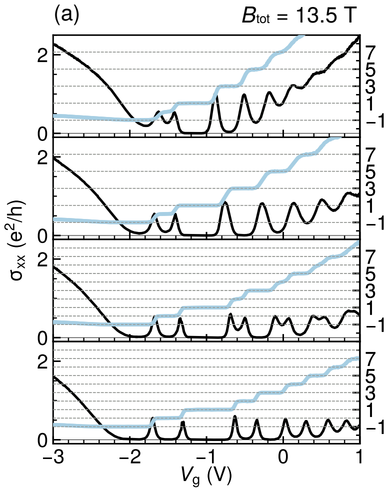
B. Guo et al., submitted

Two-dimensional Weyl semimetal phase

- Strong in-plane Zeeman field converts the 2D TI into a 2D WSM
- Ideal WSM
  - Only two Weyl nodes
  - Nodes at the charge neutrality point
  - No other bands
- Odd-integer quantum Hall effect
- Symmetry protected
- Host for other topological states, e.g. combination with superconductors





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Summary

- Cd_3As_2 is a platform for creating, and controlling different topological phases
- "Ideal" 2D TI
- "Ideal" 2D Weyl semimetal
- Tunability by strain, film thickness, magnetic fields
- Excellent transport properties
- Promising platform for novel quantum devices

Thank you!