Topological insulator thin film properties

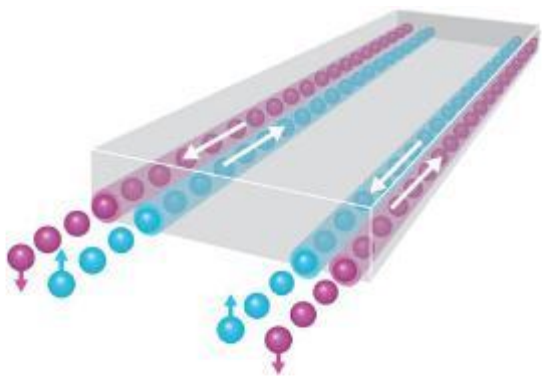
Ko-Hsuan Chen
Chun-Chia Chen

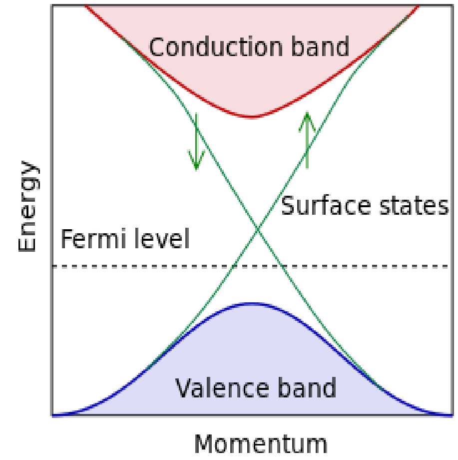
2019.11.14

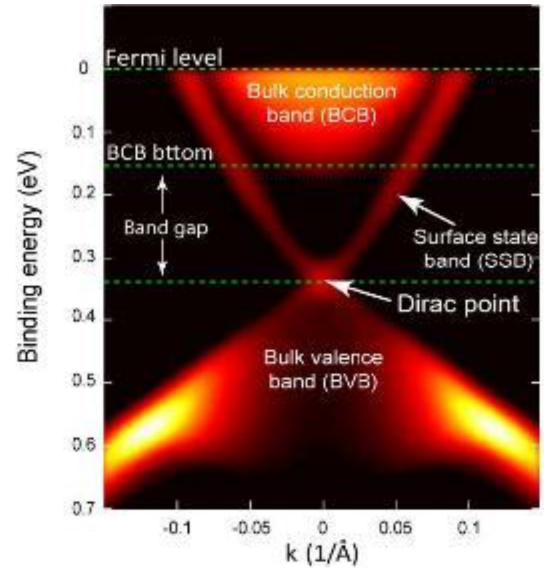
Outline

- Introduction to 3D topological insulator
- Common ways to make thin films and introduction to epitaxy
- Paper review: properties found in TI thin films
- Van der Waals epitaxy of topological insulator Bi_2Se_3 on single layer transition metal dichalcogenide MoS_2
- Crystal growth and electronic band structure of α-Sn thin films
- Thin film growth of topological insulators Bi_2Se_3 and $(Bi,Sb)_2Te_3$ toward bulk-insulating features and enhanced interfacial exchange coupling on rare earth iron garnets

Introduction of topological insulators (TIs)







Electronic band structure along the K-Γ-K direction of undoped Bi_2Se_3 by ARPES, Y. L. Chen et al, Science, (2010).

- Properties
  - Strong spin-orbital coupling
  - Spin momentum locked surface state protected by time reversal symmetry

- Applications
  - Spin momentum locked surface state - spintronics device
  - Interface of TI and superconductor - Majorana fermion, quantum computation

Tools for studying the properties of topological insulators

Material properties

- Crystal structure
  - X-ray diffraction
  - RHEED, LEED
  - TEM
  - …
- Chemical composition
  - XPS
  - EDX
  - …
- Surface morphology
  - AFM
  - STM

Topological insulators

Physical properties

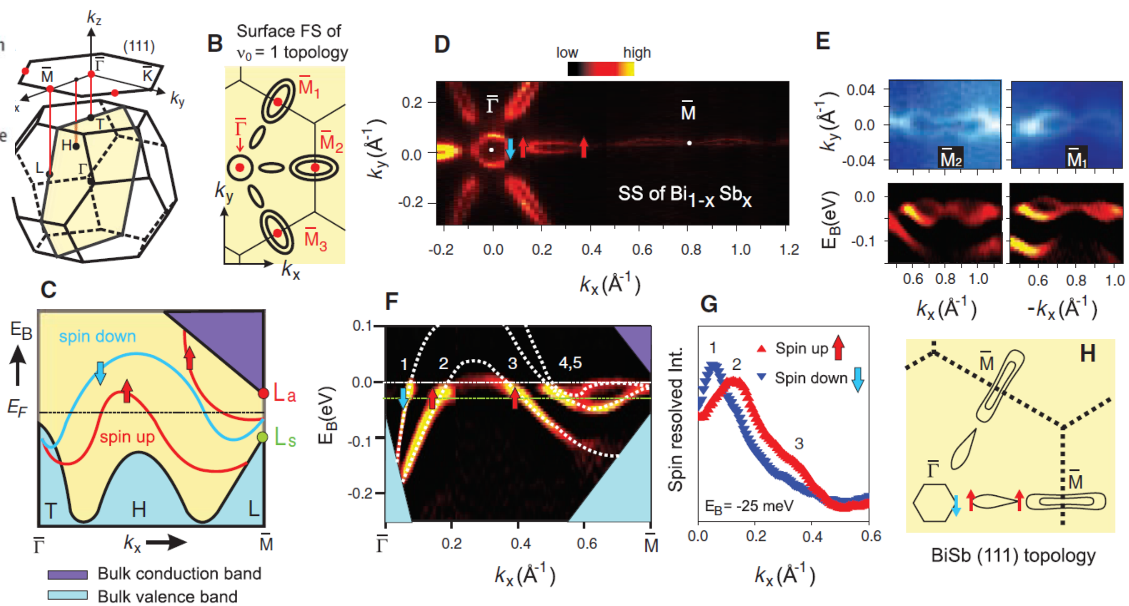
- Electrical transport properties
  - Hall measurement
    - High mobility
    - Weak antilocalization (spin-momentum locking)
- Electronic band structure
  - ARPES
    - Direct image of topological surface state in reciprocal space
  - STS

First 3D TI (first generation)

Observation of Unconventional Quantum Spin Textures in Topological Insulators

D. Hsieh,[1] Y. Xia,[1,2] L. Wray,[1,3] D. Qian,[1] A. Pal,[1] J. H. Dil,[4,5] J. Osterwalder,[5] F. Meier,[4,5] G. Bihlmayer,[6] C. L. Kane,[7] Y. S. Hor,[8] R. J. Cava,[8] M. Z. Hasan[1,2*]

A topologically ordered material is characterized by a rare quantum organization of electrons that evades the conventional spontaneously broken symmetry–based classification of condensed matter. Exotic spin-transport phenomena, such as the dissipationless quantum spin Hall effect, have been speculated to originate from a topological order whose identification requires a spin-sensitive measurement, which does not exist to this date in any system. Using Mott polarimetry, we probed the spin degrees of freedom and demonstrated that topological quantum numbers are completely determined from spin texture–imaging measurements. Applying this method to Sb and $Bi_{1-x}Sb_x$, we identified the origin of its topological order and unusual chiral properties. These results taken together constitute the first observation of surface electrons collectively carrying a topological quantum Berry's phase and definite spin chirality, which are the key electronic properties component for realizing topological quantum computing bits with intrinsic spin Hall–like topological phenomena.



Single crystal

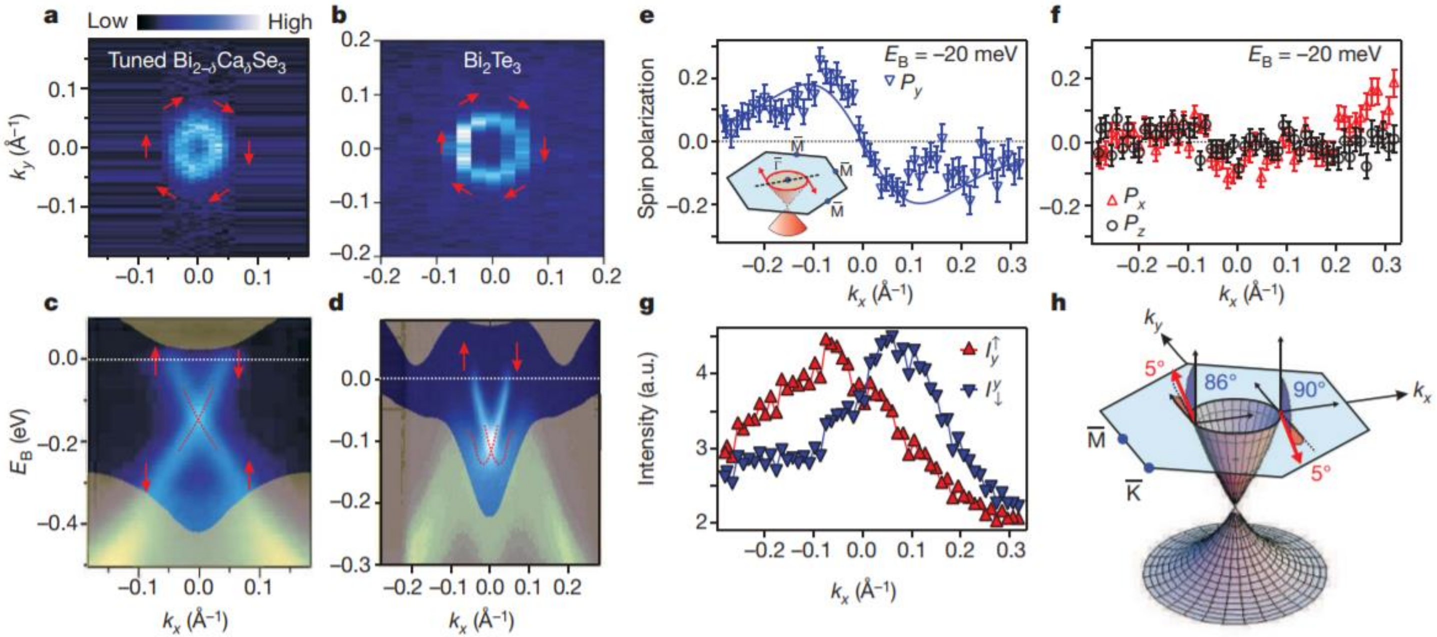
D. Hsieh et al., Science 323, 919 (2009).

Bi_2Se_3 family 3D TI (second generation)

A tunable topological insulator in the spin helical Dirac transport regime

Single crystal

D. Hsieh[1], Y. Xia[1], D. Qian[1,5], L. Wray[1], J. H. Dil[6,7], F. Meier[6,7], J. Osterwalder[7], L. Patthey[6], J. G. Checkelsky[1], N. P. Ong[1], A. V. Fedorov[8], H. Lin[9], A. Bansil[9], D. Grauer[2], Y. S. Hor[2], R. J. Cava[2] & M. Z. Hasan[1,3,4]

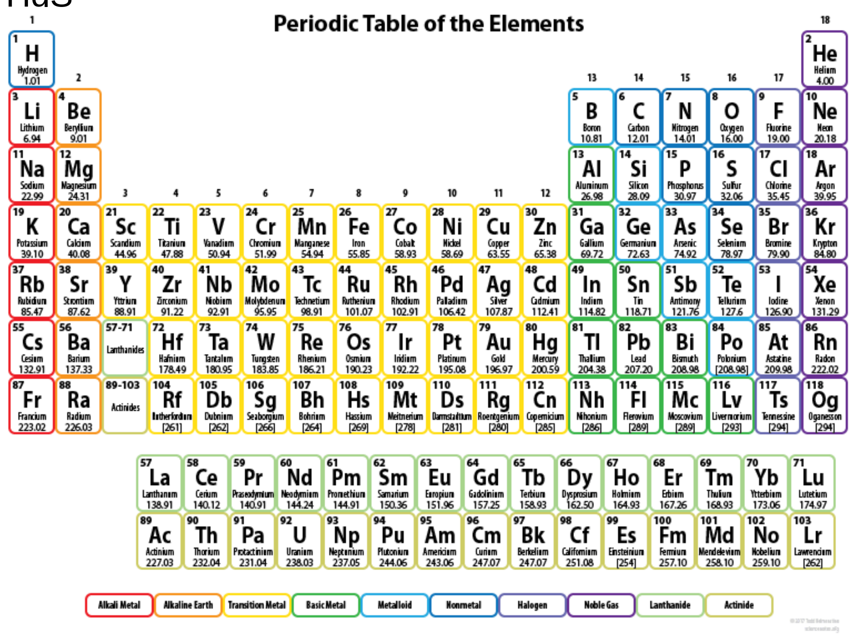


Topological insulators material

- Bi_2Se_3 family and related materials: Both conduction and valence band consist of *p* orbitals
  - Bi_2Se_3 family
  - $TlBiSe_2$
  - $LaBiTe_3$
  - $PbBi_2Se_4$

- HgTe and related materials: S-type Γ_6 band and p-type Γ_8 band
  - HgTe quantum wells and strained bulk HgTe, HgSe, β-HgS
  - AlSb/InAs/GaSb quantum wells
  - Heusler compounds
  - Chalcopyrite semiconductors
  - α- and β-Ag_2Te
  - Skutterudites and filled skutterudites

- Other materials
  - Bi_xSb_{1-x}
  - Graphene, silicene, and related material
  - PbTe, SnTe, and related material
  - Correlated materials with d or f orbitals



Bulk crystals versus thin films

Bulk crystals

Advantages:

- Usually easier to achieve
- Higher crystal quality and less defects
- Usually easier to prepare the fresh surface by cleaving for measurements such as ARPES, STM etc.

→ Faster in characterizing the topological phases/states of materials

Disadvantages:

- Hard to integrate with other materials for possible application for advance technologies
- Usually harder to study the properties in ultra thin regime

Thin film

Advantages:

- Usually easier to combine with other materials for the study of novel physical properties at the interface such as magnetic proximity effect in TI/magnetic material, majorana bound states in TI/SC etc.
- Easy to study the properties through a wide range of thickness, for example transport properties with electrical field effect
- Have chances to integrate with current Si technology or other industrial applications

Disadvantages:

- More complicated material issue need to be considered during growth, such as substrate selection, growth methods

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Common ways of making thin films

Physical Vapor Deposition (PVD)

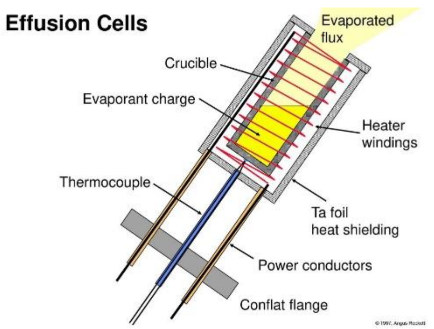
Evaporation

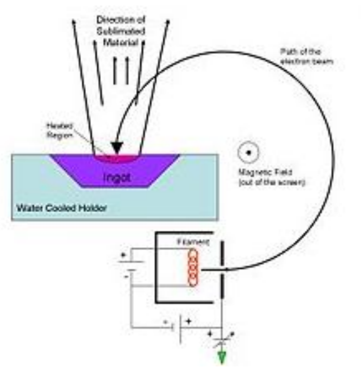
Source material (bulk) → atoms

Heating

- Resistance heated
- e-gun

⇨ MBE, evaporator





Sputtering

Target (Bulk) → atoms

Gas ion collision

Glowing/Plasma

Chemical Vapor Deposition (CVD)

one or more volatile precursors, which react and/or decompose on the substrate surface to produce the desired thin films

example

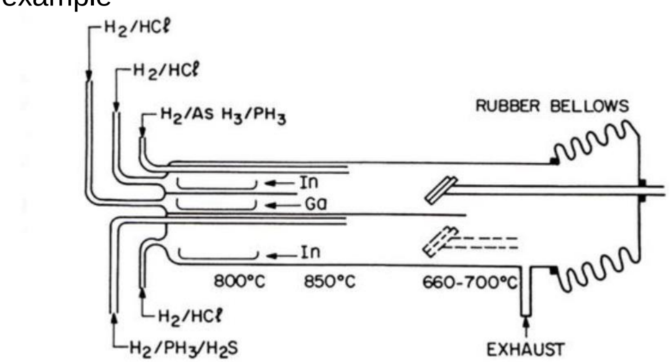
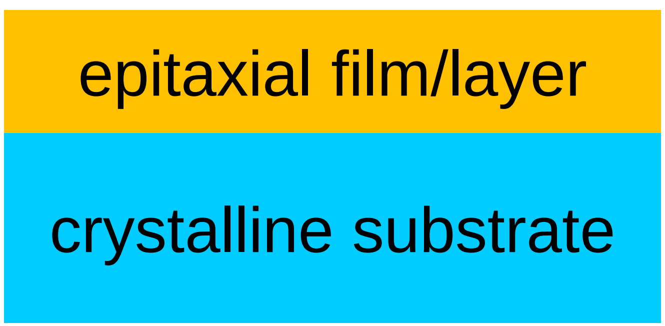


Figure 4-3. Schematic of atmospheric CVD reactor used to grow GaAs and other compound semiconductor films by the hydride process. (Reprinted with permission from Ref. 10).

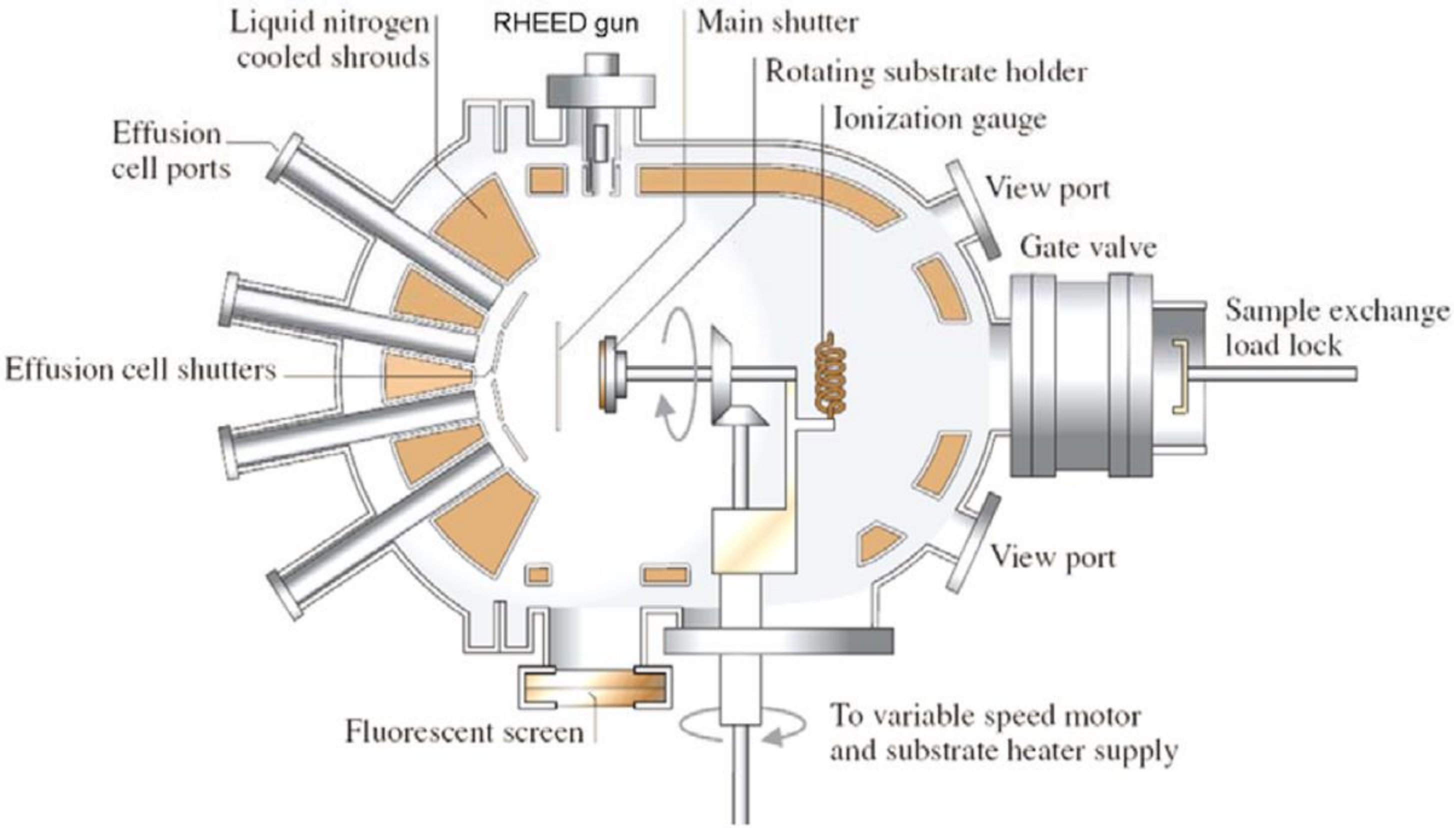
APCVD
LPCVD
MOCVD
ALD
….

Introduction to epitaxy

- The term epitaxy comes from the Greek roots epi, meaning "above", and taxis, meaning "in ordered manner".
- Homoepitaxy
  - Substrate and film are the same material.
- Heteroepitaxy
  - Substrate and film are different materials.



Molecular Beam Epitaxy



Advantages:

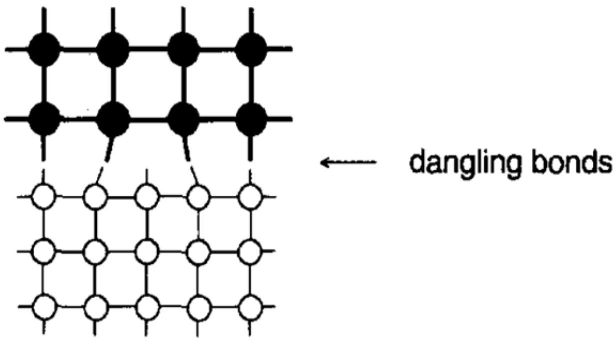
- Clean (UHV)
- Low and well- controlled growth rate
- Precise thickness control down to ML

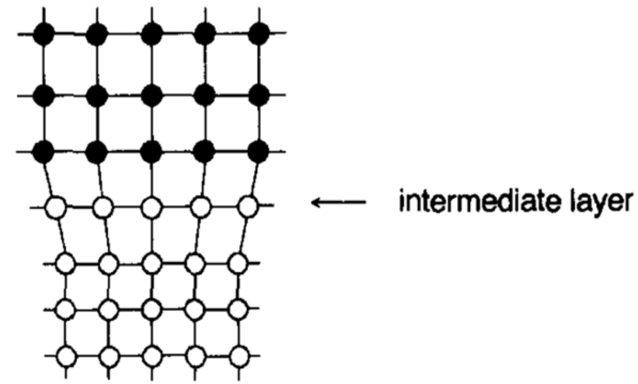
Heteroepitaxy

Covalent epitaxy

3D material

3D material

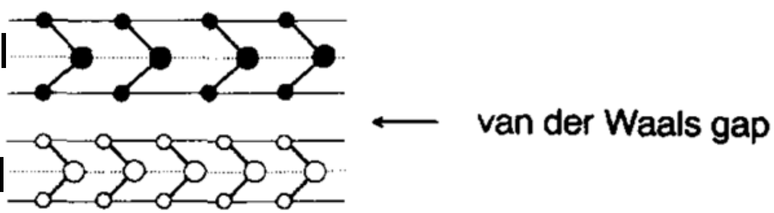




Van der Waals epitaxy

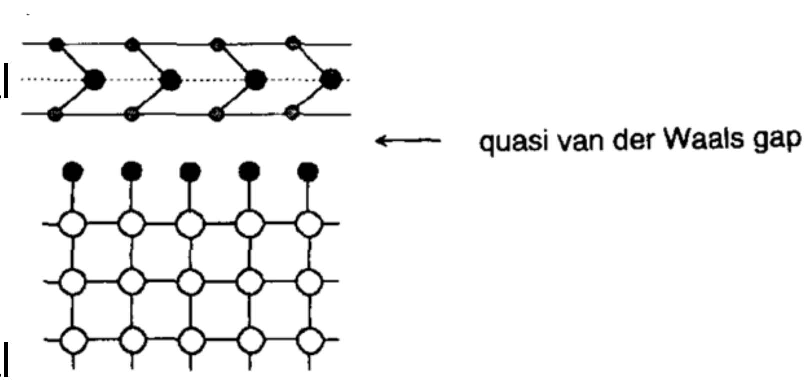
2D material

2D material



2D material

3D material



The lattice matching issue in epitaxy can be naturally overcome because of the layered structure of 2D materials.

A. Koma, Thin Solid Films 216, 72 (1992).

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Thickness limit of 3D-TIs

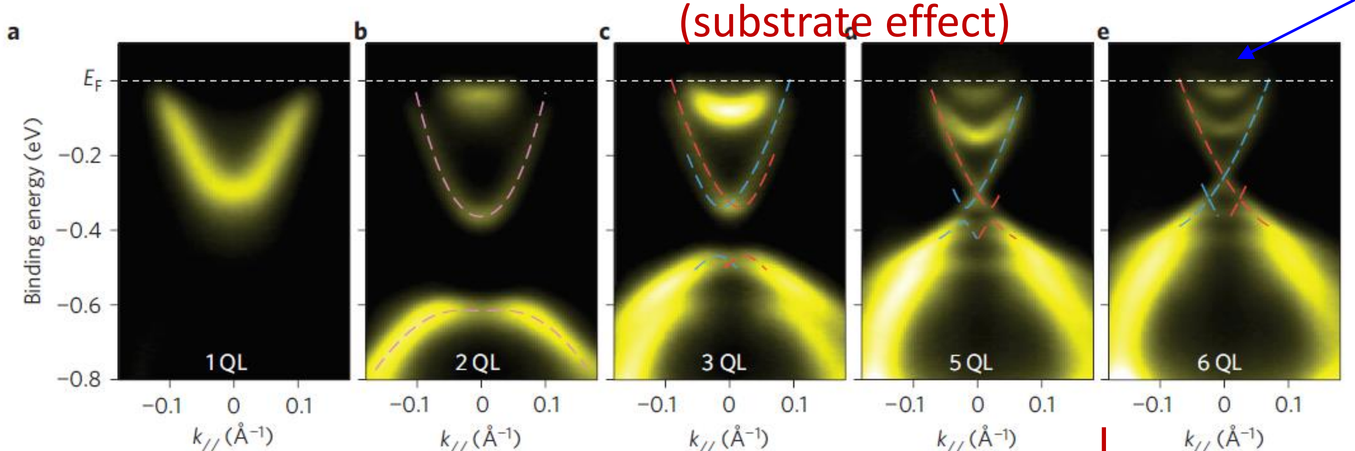
LETTERS
PUBLISHED ONLINE: 13 JUNE 2010 | DOI:10.1038/NPHYS1689
nature physics

Crossover of the three-dimensional topological insulator Bi_2Se_3 to the two-dimensional limit

Yi Zhang[1], Ke He[1]*, Cui-Zu Chang[1,2], Can-Li Song[1,2], Li-Li Wang[1], Xi Chen[2], Jin-Feng Jia[2], Zhong Fang[1], Xi Dai[1], Wen-Yu Shan[3], Shun-Qing Shen[3], Qian Niu[4], Xiao-Liang Qi[5], Shou-Cheng Zhang[5], Xu-Cun Ma[1] and Qi-Kun Xue[1,2]*

Rashba splitting (substrate effect)

Quantized subbands



3D TI

Trivial insulator

2D QSH insulator

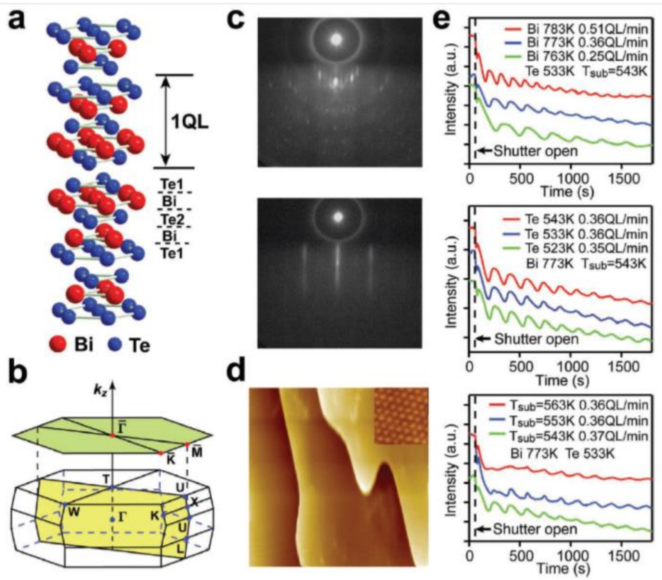
$$E_{\sigma\pm}(k_{//}) = E_0 - Dk_{//}^2 \pm \sqrt{(|\tilde{V}'| + \sigma v_F \hbar k_{//})^2 + \left(\frac{\Delta}{2} - Bk_{//}^2\right)^2}$$

Thickness limit of 3D-TIs

ADVANCED MATERIALS
www.advmat.de
Materials Views
www.MaterialsViews.com

Intrinsic Topological Insulator Bi_2Te_3 Thin Films on Si and Their Thickness Limit

By *Yao-Yi Li, Guang Wang, Xie-Gang Zhu, Min-Hao Liu, Cun Ye, Xi Chen, Ya-Yu Wang, Ke He, Li-Li Wang, Xu-Cun Ma, Hai-Jun Zhang, Xi Dai, Zhong Fang, Xin-Cheng Xie, Ying Liu, Xiao-Liang Qi, Jin-Feng Jia,* Shou-Cheng Zhang,* and *Qi-Kun Xue*



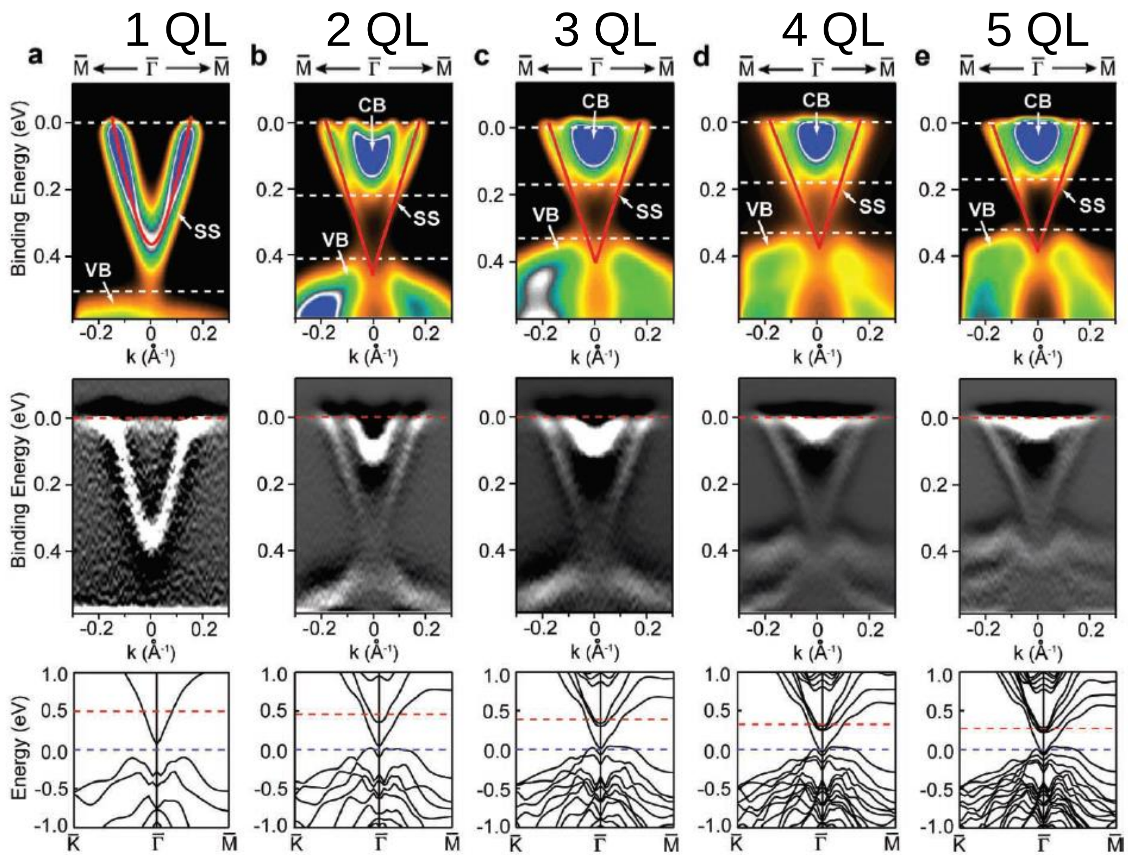


Figure 3. Band structure of ultrathin films of Bi_2Te_3. a) 1 QL. b) 2 QL. c) 3 QL. d) 4 QL. e) 5 QL. All the spectra were taken along the $\bar{\Gamma}-\bar{M}$ direction. Upper panels: ARPES intensity maps; Middle panels: differential ARPES intensity maps; Lower panels: band structures from first-principles calculations. If the Fermi level (blue dashed line) in the calculated band structure (lower panels) is shifted to higher energy (red dashed line), the major features seen in the middle panels are well reproduced.

Observation of quantum Hall effect in Bi_2Se_3 thin film

NANO LETTERS
Letter
pubs.acs.org/NanoLett

Record Surface State Mobility and Quantum Hall Effect in Topological Insulator Thin Films via Interface Engineering

Nikesh Koirala,[†] Matthew Brahlek,[†] Maryam Salehi,[‡] Liang Wu,[§] Jixia Dai,[†] Justin Waugh,[∥] Thomas Nummy,[∥] Myung-Geun Han,[⊥] Jisoo Moon,[†] Yimei Zhu,[⊥] Daniel Dessau,[∥] Weida Wu,[†] N. Peter Armitage,[§] and Seongshik Oh*[,†]

[†]Department of Physics and Astronomy and [‡]Department of Materials Science and Engineering, Rutgers, The State University of New Jersey, Piscataway, New Jersey 08854, United States

[§]Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218, United States

[∥]Department of Physics, University of Colorado, Boulder, Colorado 80309, United States

[⊥]Condensed Matter Physics and Materials Science, Brookhaven National Lab, Upton, New York 11973, United States

Supporting Information

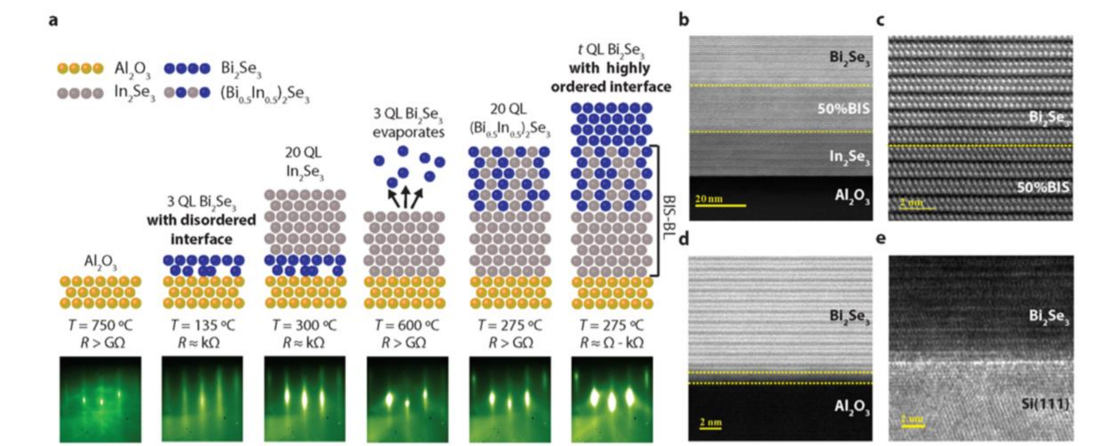
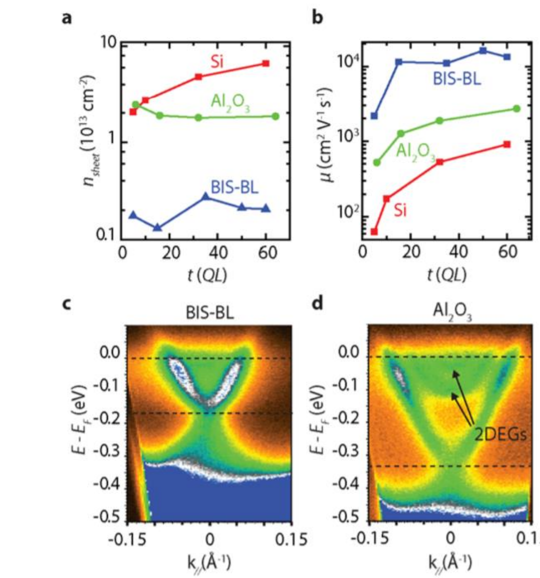


Figure 1. Growth process of Bi_2Se_3 films on the 20 QL In_2Se_3/20 QL $(Bi_{0.5}In_{0.5})_2Se_3$ buffer layer (BIS-BL) and comparison with films grown on Al_2O_3(0001) and Si(111). (a) Cartoon showing each stage of film growth along with the corresponding growth temperature (T), sheet resistance (R) and RHEED images. HAADF-STEM image of Bi_2Se_3 grown on (b) BIS-BL, which (c) shows an atomically sharp interface between Bi_2Se_3 and BIS-BL, while (d) Bi_2Se_3 grown directly on Al_2O_3(0001) has clearly disordered interface. (e) TEM image of Bi_2Se_3 grown on Si(111) (from ref 13). In (b,c), $(Bi_{0.5}In_{0.5})_2Se_3$ is written as 50%BIS.



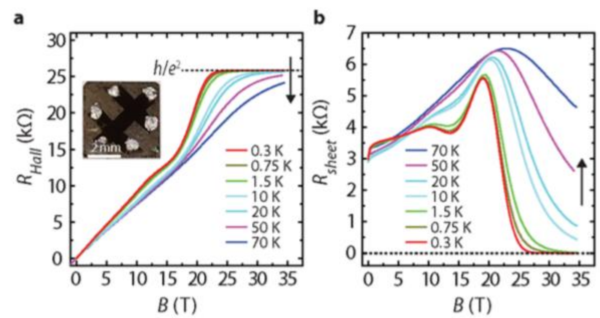


Figure 4. QHE in an 8 QL thick Bi_2Se_3 film grown on BIS-BL and capped by both MoO_3 and Se. (a) Hall resistance at different temperatures in magnetic field up to 34.5 T, which quantizes to $(1.00000 \pm 0.00004)h/e^2$ $(25813 \pm 1\ \Omega)$ at low temperatures. The inset shows the Hall-bar pattern of the measured film. (b) Corresponding longitudinal sheet resistance, which drops to zero $(0.0 \pm 0.5\ \Omega)$ when Hall resistance quantizes to h/e^2. The vertical arrows indicate the direction of increasing temperature.

Observation of quantum Hall effect in $(Bi_{0.53}Sb_{0.47})Te_3$ thin film

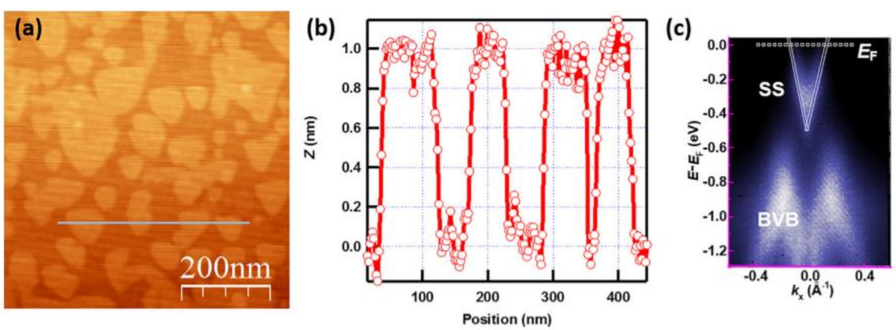
APPLIED PHYSICS LETTERS **110**, 212401 (2017)

Observation of Quantum Hall effect in an ultra-thin $(Bi_{0.53}Sb_{0.47})_2Te_3$ film

Wenqin Zou,[1,a)] Wei Wang,[2,a)] Xufeng Kou,[3] Murong Lang,[3] Yabin Fan,[3] Eun Sang Choi,[4] Alexei V. Fedorov,[5] Kejie Wang,[2] Liang He,[2,3,b)] Yongbing Xu,[2,b)] and Kang. L. Wang[3,b)]
[1]*National Lab of Solid State Microstructures, Department of Physics, Nanjing University, Nanjing 210093, People's Republic of China*
[2]*York-Nanjing Joint Center (YNJC) for Spintronics and Nano Engineering, School of Electronics Science and Engineering, Nanjing University, Nanjing 210093, China*
[3]*Department of Electrical Engineering, University of California, Los Angeles, Los Angeles, California 90095, USA*
[4]*National High Magnetic Field Laboratory, Tallahassee, Florida 32310, USA*
[5]*Advanced Light Source Division, Lawrence Berkeley national laboratory, 1 Cyclotron Road, Berkeley, California 94720, USA*

(Received 5 November 2016; accepted 2 April 2017; published online 22 May 2017)

We report the observation of the Quantum Hall effect from the topological surface states in both the Dirac electron and Dirac hole regions in a 4 quintuple layer $(Bi_{0.53}Sb_{0.47})_2Te_3$ film grown on GaAs (111)B substrates. The Fermi level is sitting within the enlarged bulk band gap due to the quantum confinement of the ultra-thin film and can be tuned through the Dirac point by gate biases. Furthermore, the Hall resistance R_{xy} shows even denominator plateaus, which could be fractional Quantum Hall states. This may be due to the hybridization between the top and bottom surface states and suggests the possible way to manipulate the interaction of two surfaces for potential spintronic devices. *Published by AIP Publishing.* [http://dx.doi.org/10.1063/1.4983684]



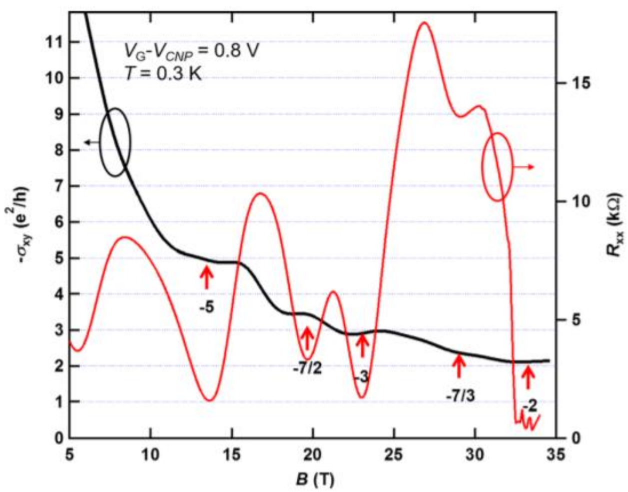


FIG. 3. Fractional quantum hall effect of $(Bi_{0.53}Sb_{0.47})_2Te_3$ at a high field. (Left) The Hall conductivity σ_{xy} at $V_G - V_{CNP} = 0.8$ V; the plateaus in σ_{xy} can be observed clearly. The Landau filling factors are also marked. (Right) The oscillations of the longitudinal resistance R_{xx}.

p-n junction

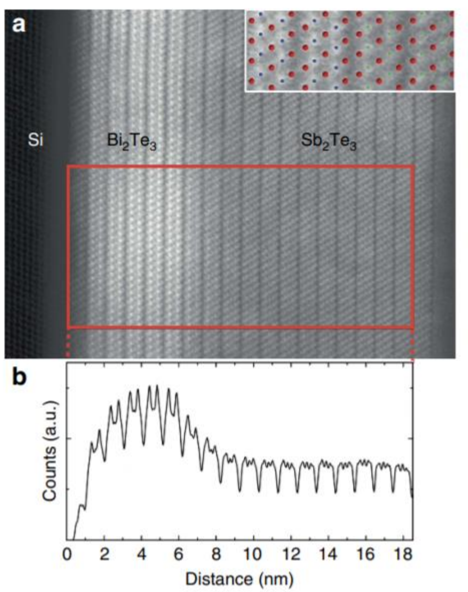
nature COMMUNICATIONS

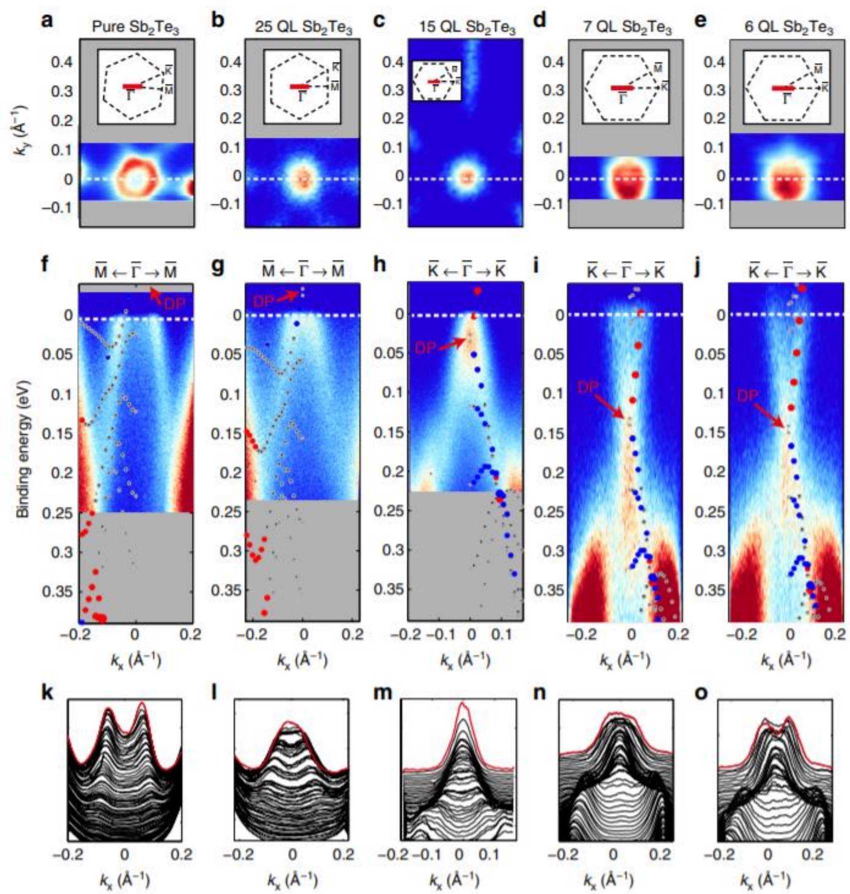
ARTICLE

Received 8 May 2015 | Accepted 6 Oct 2015 | Published 17 Nov 2015 DOI: 10.1038/ncomms9816 OPEN

Realization of a vertical topological p–n junction in epitaxial Sb_2Te_3/Bi_2Te_3 heterostructures

Markus Eschbach[1], Ewa Młyńczak[1,2], Jens Kellner[3], Jörn Kampmeier[4], Martin Lanius[4], Elmar Neumann[5], Christian Weyrich[4], Mathias Gehlmann[1], Pika Gospodarič[1], Sven Döring[1], Gregor Mussler[4], Nataliya Demarina[6], Martina Luysberg[5,7], Gustav Bihlmayer[8], Thomas Schäpers[4], Lukasz Plucinski[1], Stefan Blügel[8], Markus Morgenstern[3], Claus M. Schneider[1] & Detlev Grützmacher[4]





Magnetizing topological insulator via proximity effect

SCIENCE ADVANCES | RESEARCH ARTICLE

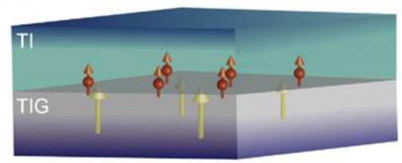
MATERIALS SCIENCE

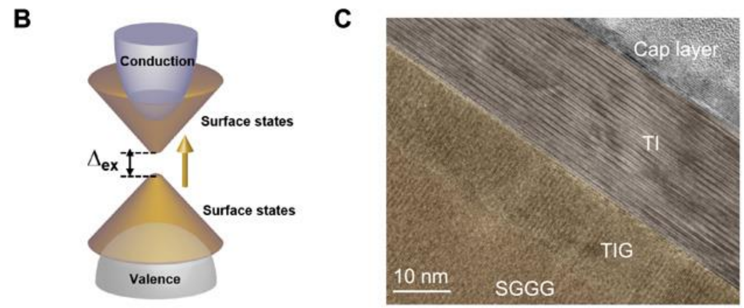
Above 400-K robust perpendicular ferromagnetic phase in a topological insulator

Chi Tang,[1]* Cui-Zu Chang,[2,3]* Gejian Zhao,[4] Yawen Liu,[1] Zilong Jiang,[1] Chao-Xing Liu,[3] Martha R. McCartney,[4] David J. Smith,[4] Tingyong Chen,[4] Jagadeesh S. Moodera,[2,5] Jing Shi[1†]

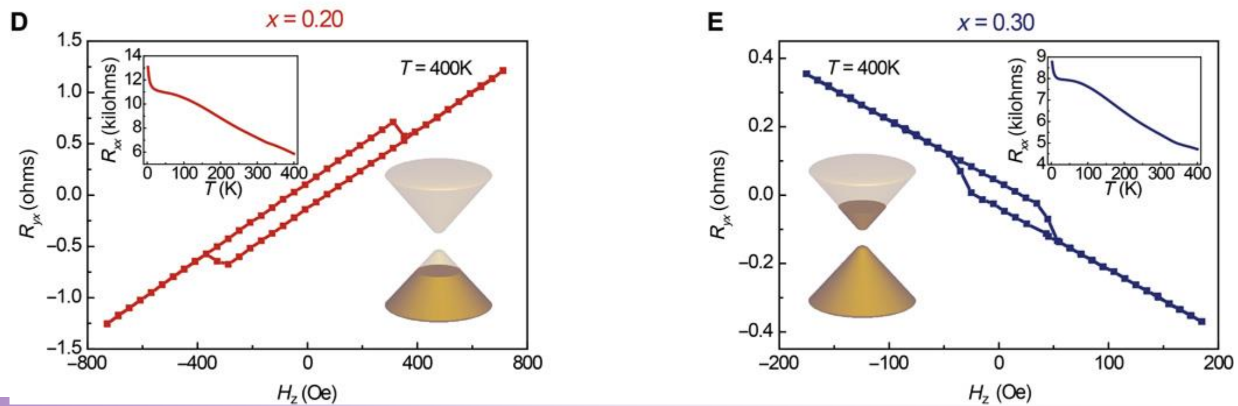
The quantum anomalous Hall effect (QAHE) that emerges under broken time-reversal symmetry in topological insulators (TIs) exhibits many fascinating physical properties for potential applications in nanoelectronics and spintronics. However, in transition metal–doped TIs, the only experimentally demonstrated QAHE system to date, the QAHE is lost at practically relevant temperatures. This constraint is imposed by the relatively low Curie temperature (T_c) and inherent spin disorder associated with the random magnetic dopants. We demonstrate drastically enhanced T_c by exchange coupling TIs to $Tm_3Fe_5O_{12}$, a high-T_c magnetic insulator with perpendicular magnetic anisotropy. Signatures showing that the TI surface states acquire robust ferromagnetism are revealed by distinct squared anomalous Hall hysteresis loops at 400 K. Point-contact Andreev reflection spectroscopy confirms that the TI surface is spin-polarized. The greatly enhanced T_c, absence of spin disorder, and perpendicular anisotropy are all essential to the occurrence of the QAHE at high temperatures.

Breaking time reversal symmetry in TI





Anomalous Hall effect in TI



Van der Waals epitaxy of topological insulator Bi_2Se_3 on single layer transition metal dichalcogenide MoS_2

K. H. M. Chen *et al., Appl. Phys. Lett.* **111**, 083106 (2017).

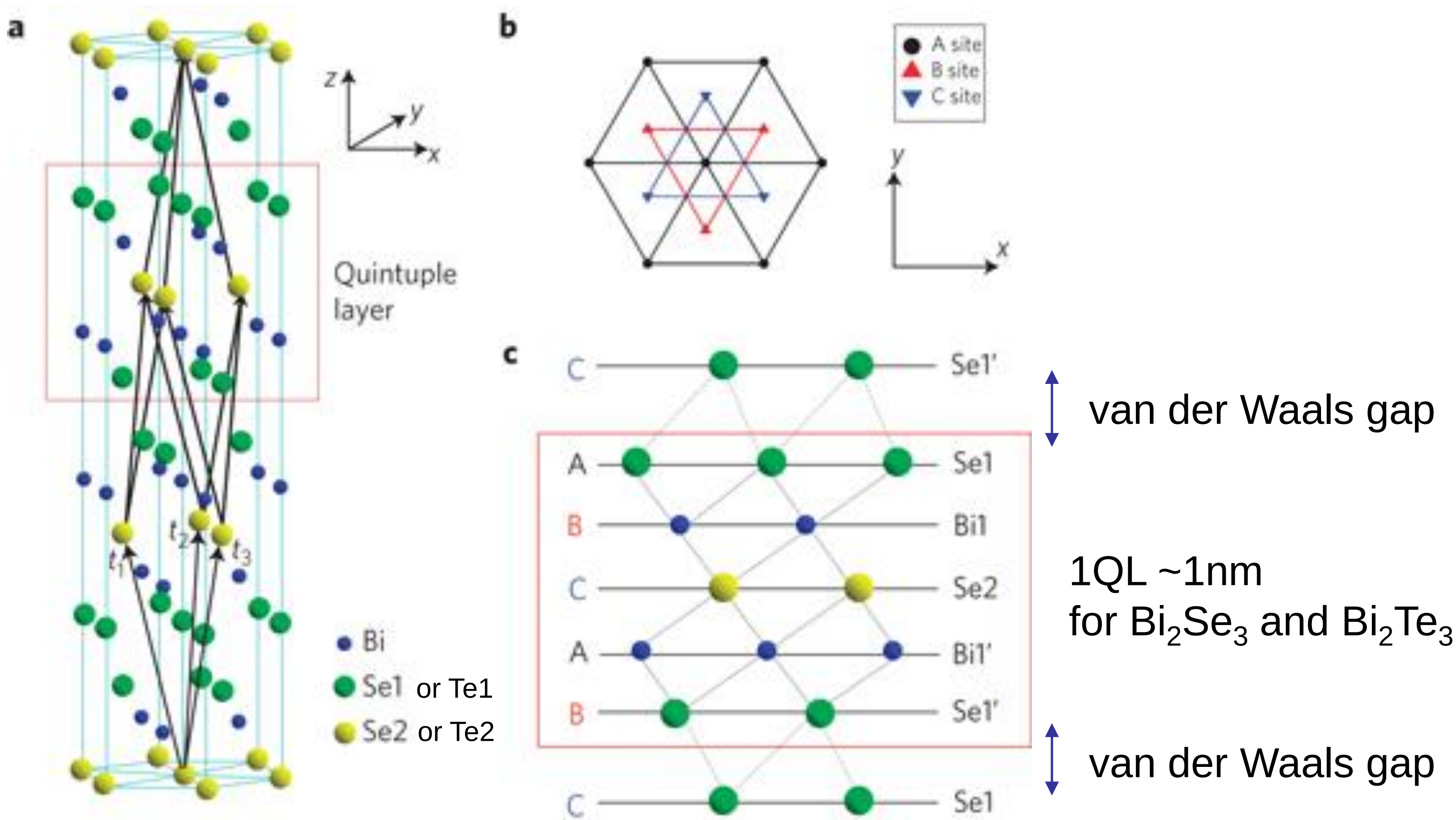
Ko-Hsuan Mandy Chen, H. Y. Lin, S. R. Yang, C. Y. Wang, and J. Kwo
Dept. of Phys., National Tsing Hua Univ., Hsinchu, Taiwan

X. Q. Zhang, and Y. H. Lee
Dept. of Mat. Sci. and Eng., National Tsing Hua Univ., Hsinchu, Taiwan

C. K. Cheng, and M. Hong
Grad. Inst. of Appl. Phys., National Taiwan Univ., Taipei, Taiwan

C. M. Cheng, and C. H. Hsu
National Synchrotron Radiation Research Center, Hsinchu, Taiwan

Crystal structure of Bi_2Se_3 and Bi_2Te_3



trigonal

H. Zhang et. al., Nat. Phys. 5 (6), 438 (2009)

Motivation

- Challenge in thin film
  - High defect density such as Se vacancies

    ⇨ Fermi level locates at conduction band easily

- Other's work in Bi_2Se_3 thin film on different substrates

  (refer to Prof. M. H. Xie's work in Chin. Phys. B, 22, 6, 068101 (2013))
  - **non van der Waals type**

How about inserting TMD material such as MoS_2?

✓ **2D layered structure**

✓ **hexagonal symmetry**

✓ **van der Waals type surface**

✓ **can be grown on diverse substrates**

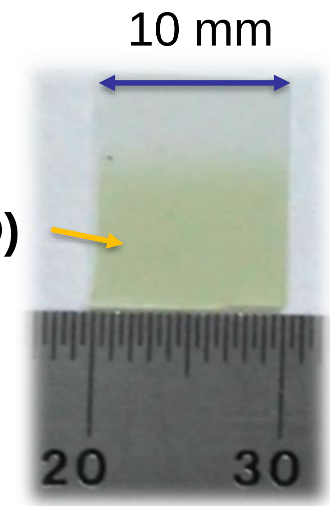
  - **van der Waals type**

    graphene

Sample Growth

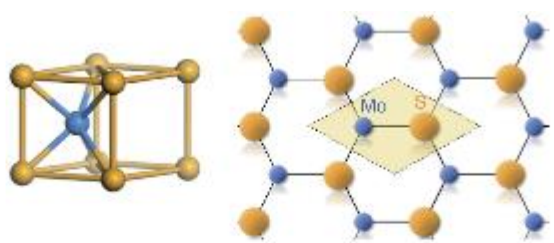
- **clean Al_2O_3(0001) substrate**
  surface roughness: 0.16nm

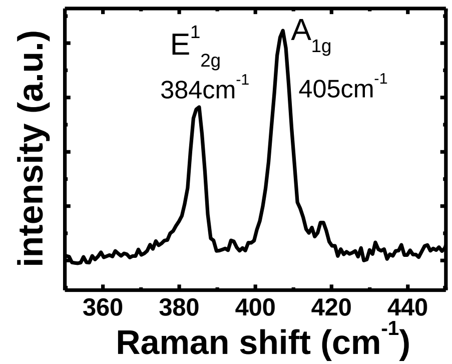
1. H_2SO_4:H_2O_2=1:1 30 min
2. acetone 5min in ultrasonic bath
3. alcohol 5min in ultrasonic bath



- **grow MoS_2 monolayer by chemical vapor deposition (CVD)**

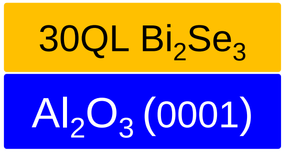
crystal structure: hexagonal P63/mmc
lattice constant: a = 0.31 nm, *c* = 1.28 nm
surface roughness: 0.15 nm
typical triangular domain size: 20-30 μm
large and continuous area up to 10 mm x 8 mm
provided by Prof. Y. H. Lee's group in NTHU



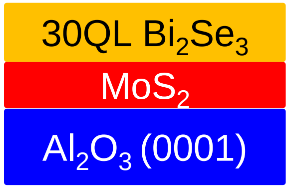


Nature Comm., 10, 1038 (2012)
Nano Lett., 12 (3), 1538-1544 (2012)

- **grow Bi_2Se_3 film**
  by molecular beam epitaxy (MBE)
  rate: ~1 QL/min, Se/Bi flux ratio: 20



two step growth
Ts = 180℃/280 ℃

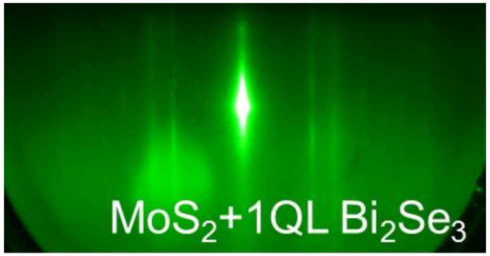


one step growth
Ts = 280 ℃

lattice mismatch: 15.7% (Al_2O_3), -24.9% (MoS_2)

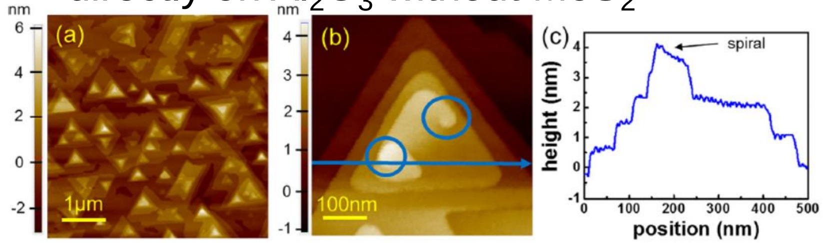
High quality Bi_2Se_3 thin films using a MoS_2 template

Excellent crystallinity

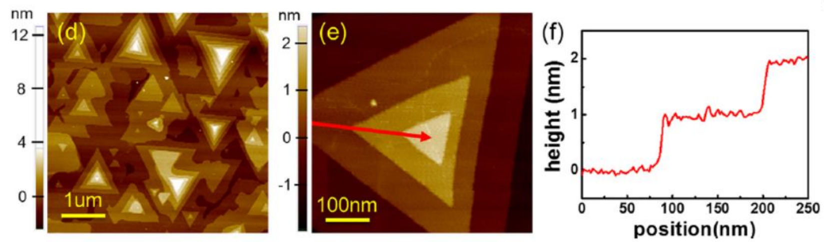


Large triangular shaped domain

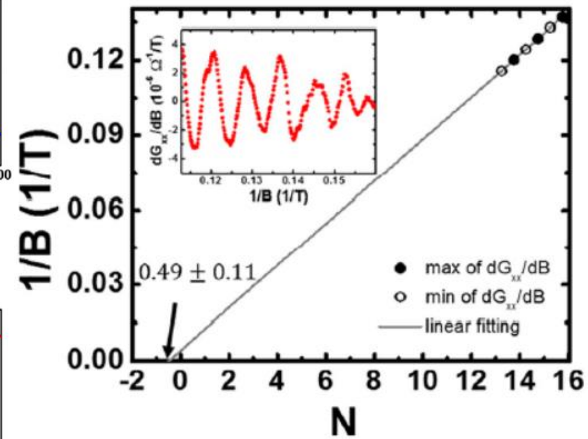
directly on Al_2O_3 without MoS_2



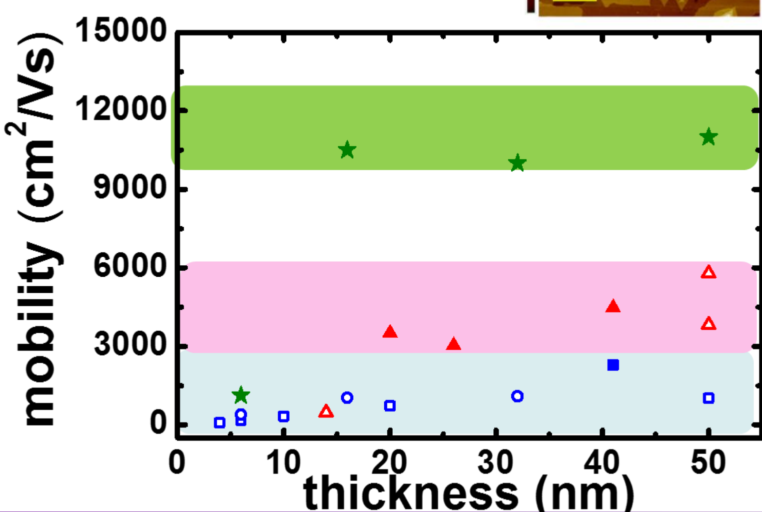
with MoS_2 template



SdH oscillation from topological surface state



High mobility



- van der Waals type surface
- good interface
- good in-plane domain order

- van der Waals type surface
- good interface
- poor in-plane domain order

- non van der Waals type surface
- poor interface
- good in-plane order

○ on Al_2O_3 (no capping)

★ on $(Bi_xIn_{1-x})_2Se_3/Al_2O_3$(no capping)

S. Oh et al, Nano Lett., **15** (12), 8245 (2015).

△ on MoS_2/Al_2O_3 (no capping) our data

▲ on MoS_2/Al_2O_3 (30nm Se capping) our

□ on Al_2O_3 (no capping) our data

■ on Al_2O_3 (30nm Se capping) our data

Crystal growth and electronic band structure of α-Sn thin films

Ko-Hsuan (Mandy) Chen, H. Y. Lin, S. W. Huang, and J. Kwo*
Dept. of Phys., National Tsing Hua Univ., Hsinchu, Taiwan

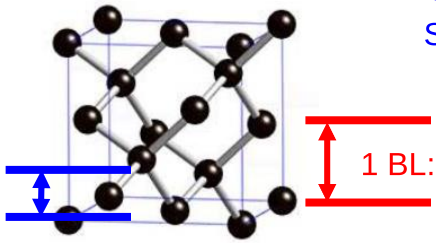
C. K. Cheng, K. Y. Lin, and M. Hong*
Grad. Inst. of Appl. Phys., National Taiwan Univ., Taipei, Taiwan

S. W. Lien, and T. R. Chang
Dept. of Phys., National Cheng Kung Univ., Taipei, Taiwan

C. M. Cheng, and C. H. Hsu
National Synchrotron Radiation Research Center, Hsinchu, Taiwan

Introduction: crystal structure of Sn

α–Sn
Gray tin
face centered cubic (diamond)



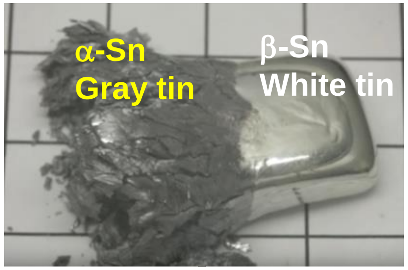
1 BL: 6.49 Å/2 = 3.245 Å

1 ML: 6.49 Å/4 = 1.62 Å

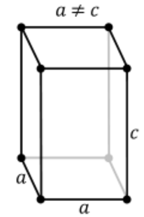
a = 6.49 Å

Low temperature phase

Same as Si and Ge



β–Sn
White tin
tetragonal



a = 5.83 Å
c = 3.18 Å

High temperature phase

below ⇦ **Phase transition temperature** ⇨ **above**

bulk crystal: 13.2 ℃

thin film: 70 - 170 ℃

- on nearly lattice matched substrates (InSb, CdTe)
- depend on film thickness and orientation

R. F. C. Farrow, Mat. Res. Soc. Symp. Proc. **37**, 275. (1984).
T. Osaka *et al.*, Phys. Rev. B **50**, 7567 (1994).

Introduction: discovery of α-Sn

2008
First 3D-TI observed

2014
First 3D-TDS observed

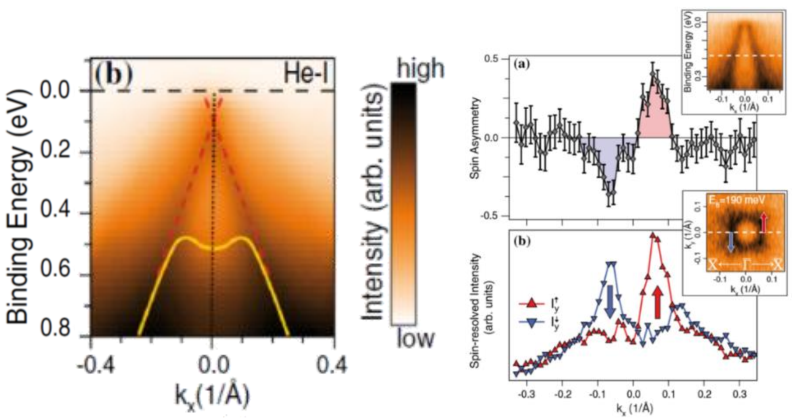
1851
- first report of α-Sn by Erdmann

1950s and 1960s
- Band structure calculation of α-Sn
- Zero gap semiconductor

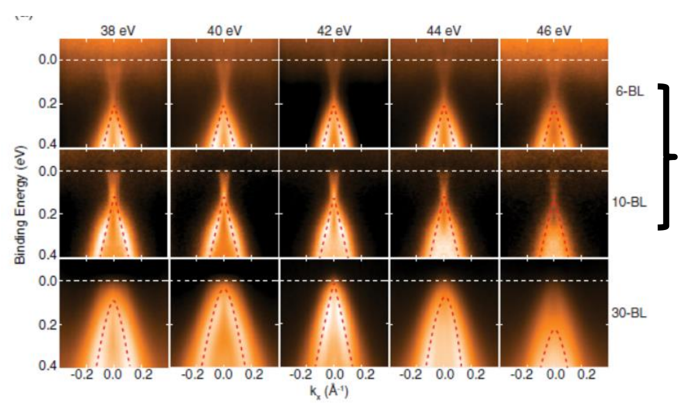
1980s
- MBE grown α-Sn thin films

2010s
- α-Sn(001) thin film in TI phase (2013)
  A. Barfuss *et al*., Phys. Rev. Lett. **111**, 157205 (2013).
  Y. Ohtsubo *et al*., PRL **111**, 216401 (2013)
- α-Sn(111) thin film in TDS phase (2017) Cai-Zhi Xu *et al*., PRL **118**, 146402 (2017).

α-Sn(001) thin film in 3D-TI phase (2013)



α-Sn(111) thin film in topological Dirac semimetal (TDS) phase (2017)



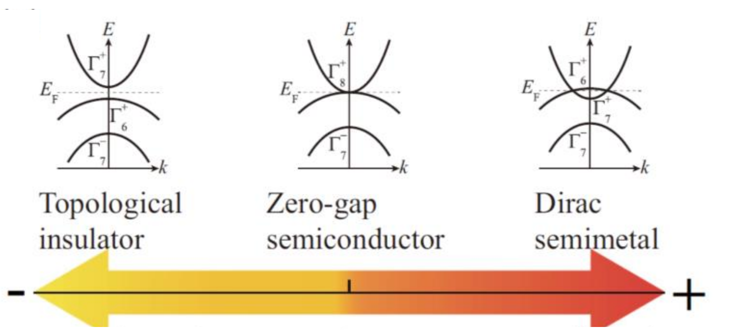
2D TDS

3D TDS

Very broadened spectrum

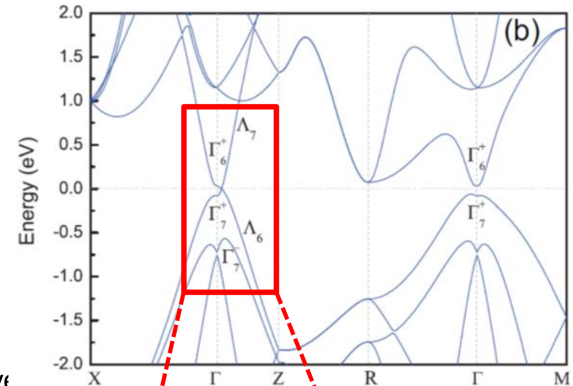
Introduction: discovery of α-Sn

Predictions of α-Sn(001) as a TDS under in-plane compressive strain

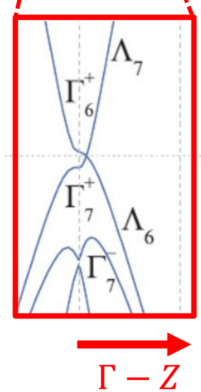


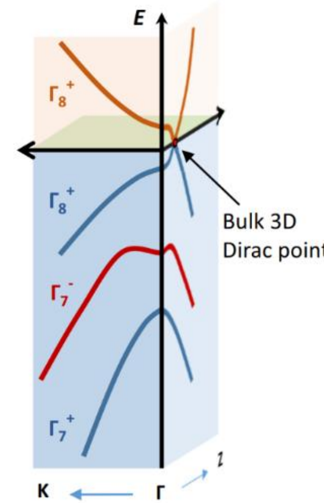
Out-of-plane: compressive
In-plane: tensile

Out-of-plane: tensile
In-plane: compressive



Huaqing Huang and Feng Liu, *Phys. Rev. B* **95**, 201101(R) (2017).



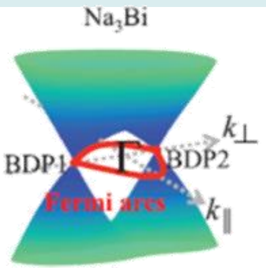


M. R. Scholz *et al.*, *Phys. Rev. B* **97**, 075101 (2018).

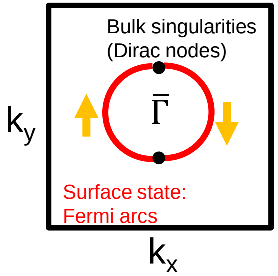
- Band crossing along $\Gamma - Z$
- Still lack of experimental prove

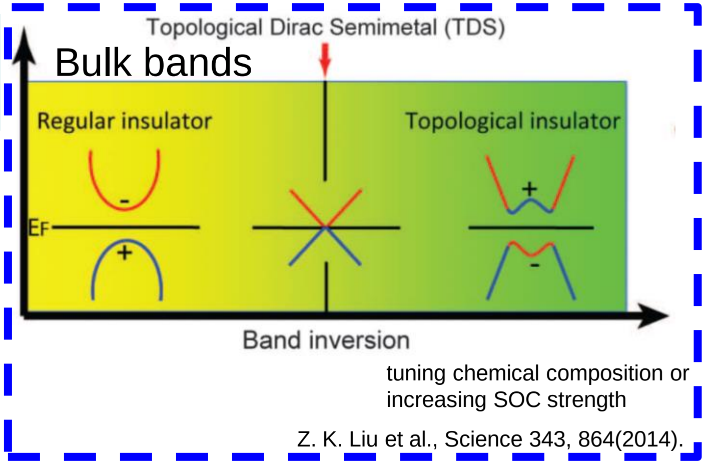
Introduction: topological insulator and topological Dirac semimetal

Topological Dirac semimetal (TDS)

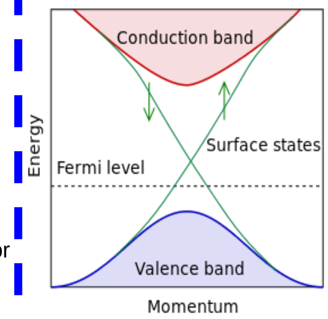


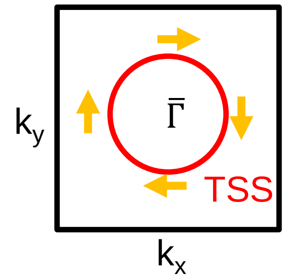
Su-Yang Xu et al., Science 347, 294 (2015).





Topological insulator (TI)





2D TDS: graphene
3D TDS (3D analog of graphene):
Na_3Bi (C3 symmetry), Cd_3As_2 (C4 symmetry)

- Gap closed at Dirac node
- Spin-polarized Fermi arcs connecting Dirac nodes

2D TI:
HgTe/CdTe quantum well
3D TI:
Bi_xSb_{1-x}, Bi_2Se_3, Bi_2Te_3,…

- Fully gapped bulk state
- A chiral edge/surface state connecting bulk conduction and valence band

Motivation

- Expected to be an ideal elemental topological material (TM) with less material defects compared with binary or ternary compound TM

- Non-toxic group IV material

  → 1. easy to tackle
  2. greater potential in combining with current semiconductor technology

- Large spin-to-charge conversion efficiency (λ_{IEE} ~ 2.1 nm) at room temperature compared to other conventional TI hetero-structure (λ_{IEE} ~0.009-0.43 nm)

  J.-C. Rojas-Sánchez *et al.*, PRL **116**, 096602 (2016).

  → novel spintronic devices

- Phase transition from topological insulator to topological Dirac semimetal by strain manipulation

  → a fascinating material for studying topological phase transition

Cai-Zhi Xu *et al.*, PRL **118**, 146402 (2017).
Huaqing Huang and Feng Liu, PRB **95**, 201101(R) (2017).
Dongqin Zhang et al., PRB **97**, 195139 (2018).

Band structure of α-Sn(001) studied by ARPES

α-Sn on InSb(001)

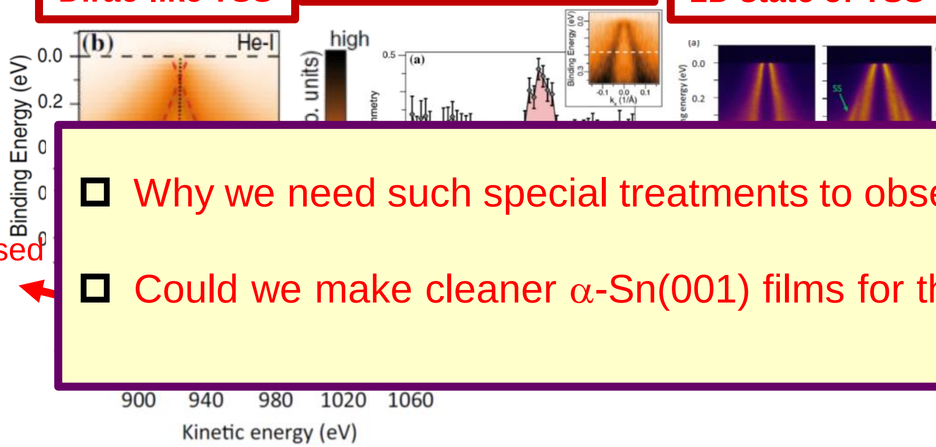
Prof. J. Schafer and R. Claessen's group in Germany

A. Barfuss *et al.*, PRL **111**, 157205 (2013)
Victor A. Rogalev *et al.*, PRB **95**, 161117(R) (2017)
M. R. Scholz *et al.*, PRB **97**, 075101 (2018)

Dirac-like TSS | **Spin-momentum locked surface state** | **2D state of TSS**

Te doped α-Sn
InSb(001)-c(8x2)

Te peaks increased at 60° off normal

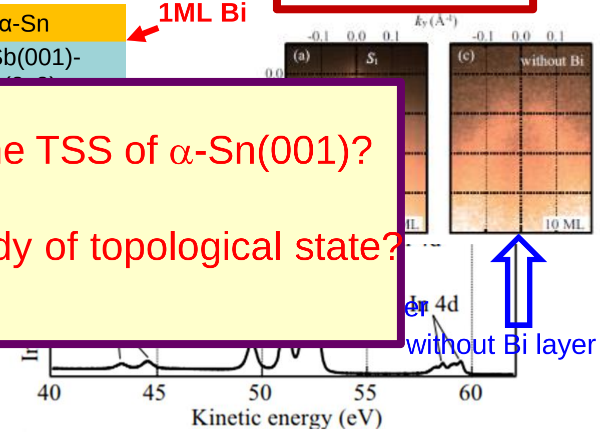


Dr. Amina Taleb-Ibrahimi and Prof. A. Fert's group in France

Y. Ohtsubo *et al.*, PRL **111**, 216401 (2013)
J.-C. Rojas-Sánchez *et al.*, PRL **116**, 096602 (2016)
Q. Barbedienne *et al.*, arXiv:1807.11377 (2018)

Dirac-like TSS

α-Sn
InSb(001)-
1ML Bi

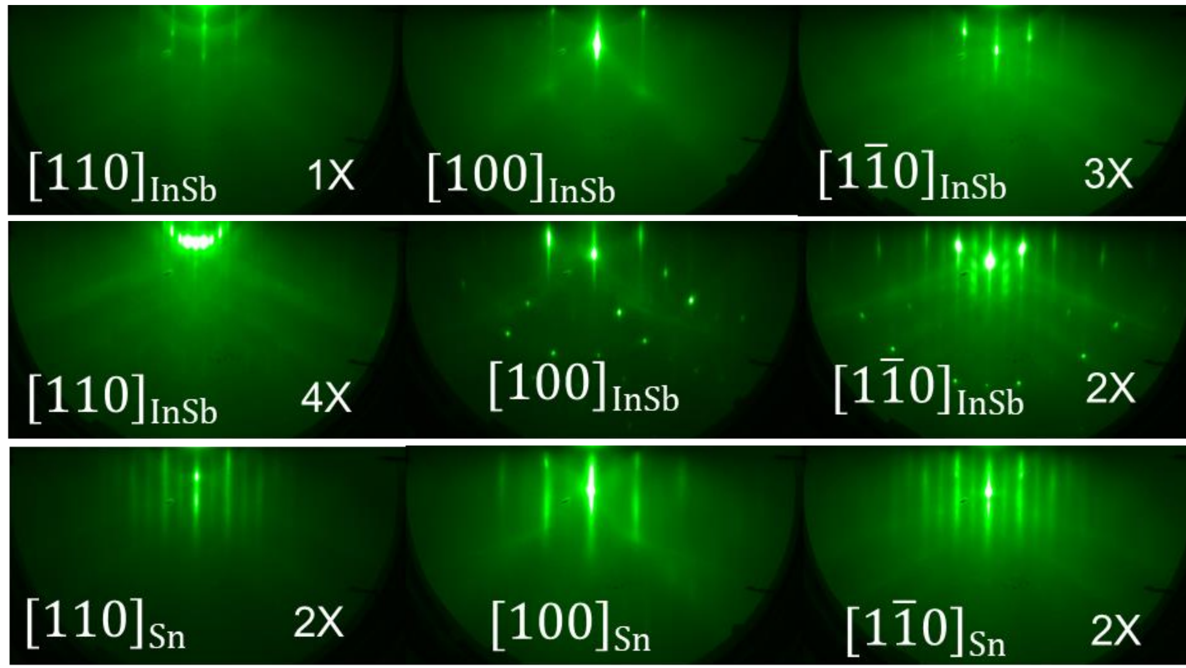


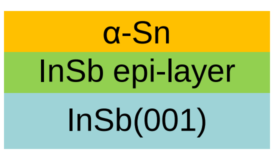
- ☐ Why we need such special treatments to observe the TSS of α-Sn(001)?
- ☐ Could we make cleaner α-Sn(001) films for the study of topological state?

- ☐ InSb wafer cleaned by sputtering and annealing
- ☐ No TSS observed without resorting to Te atoms or Bi buffer layer
- ☐ No clear evidence of TDS phase found in α-Sn(001) as predicted in calculation
- ☐ Severe In diffusion problem ⟹ p-type pristine surface
- ☐ Te or Bi atoms segregated to the top of Sn films

Sample preparation

- **InSb(001) substrate cleaning using CP4 solution**
- **Native oxides desorption under Sb flux** at T_s ~450 ℃
- **Growth of InSb epilayer** at T_s of 380 to 400 ℃
- **Growth of α-Sn film** at T_s of ~ 0 to 100 ℃

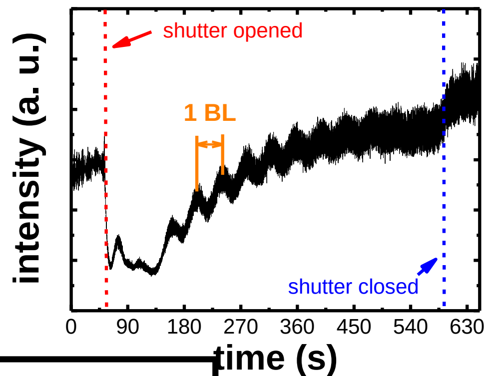




- ***In-situ* measurements: LEED, ARPES, XPS, STM**
- ***Ex-situ* measurements: XRD, AFM**

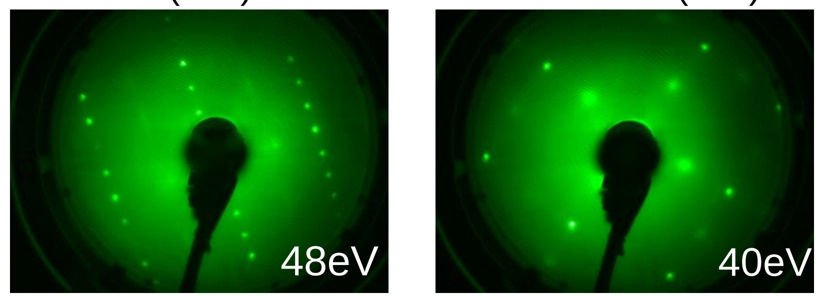
Crystal structure study

RHEED oscillation during Sn growth



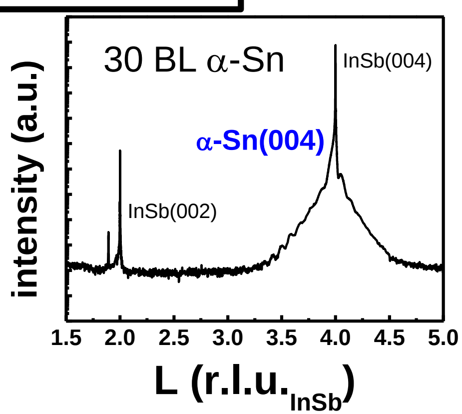
LEED patterns

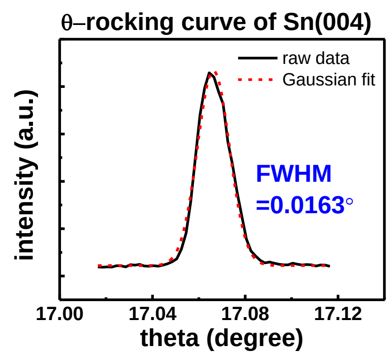
InSb(001)-4x2 Two-domain Sn(001)-2x1



Four fold symmetry

X-ray diffraction



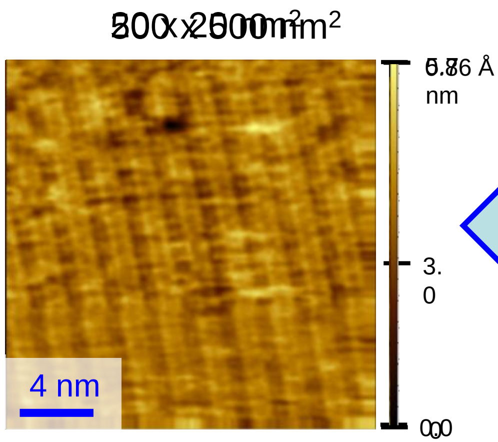


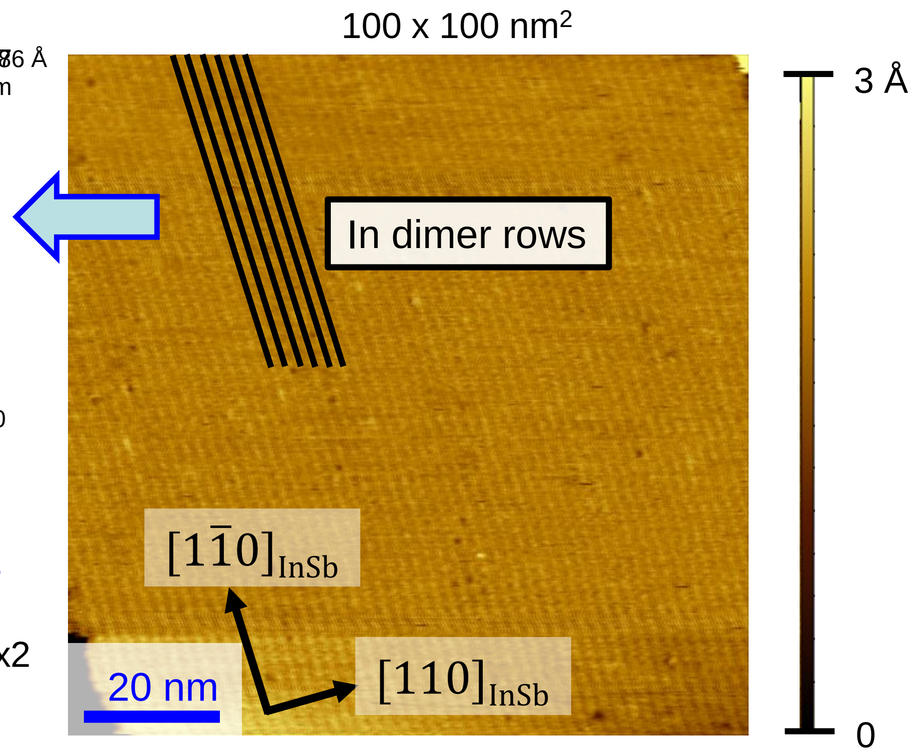
InSb(004) FWHM: 0.01455°

- Layer-by-layer growth of Sn with RHEED oscillation observed in every sample
- Smooth surface
- Sharp RHEED and LEED patterns with two-domain 2x1 surface reconstruction
- Pure α phase Sn with excellent crystallinity

Surface morphology: InSb(001)-4x2

Using *in-situ* STM





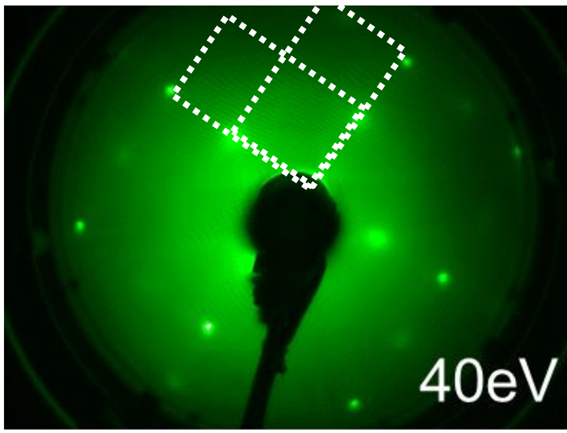
- Terraces up to 100 nm
- Smooth surface R_q: 0.16 Å
- Real space image of 4x2 surface reconstruction

Surface morphology: α-Sn(001)-2x1

Two-domain Sn(001)-2x1

spacing of Sn dimer rows

500 x 500 nm^2



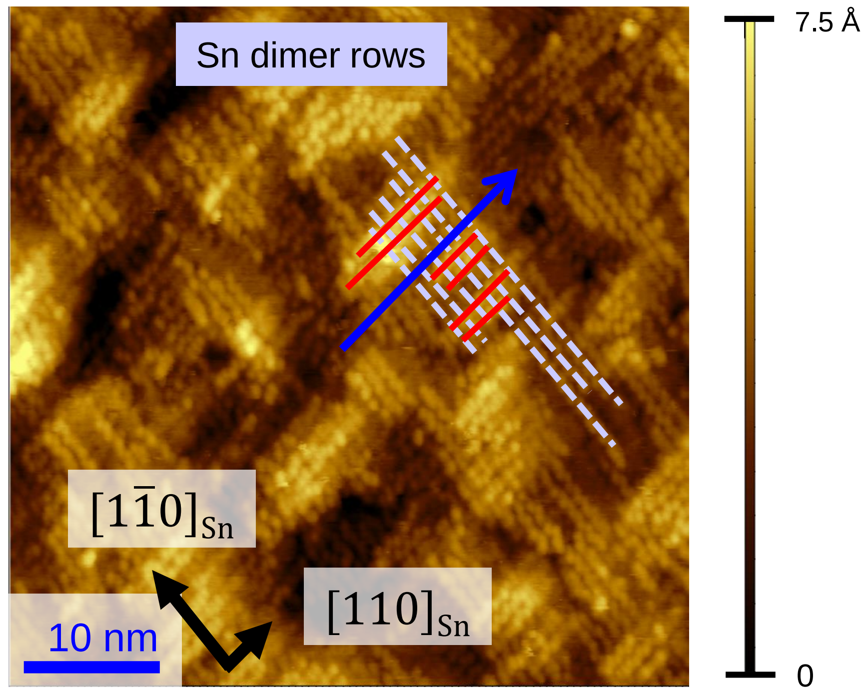
nm

ML

1.6 Å

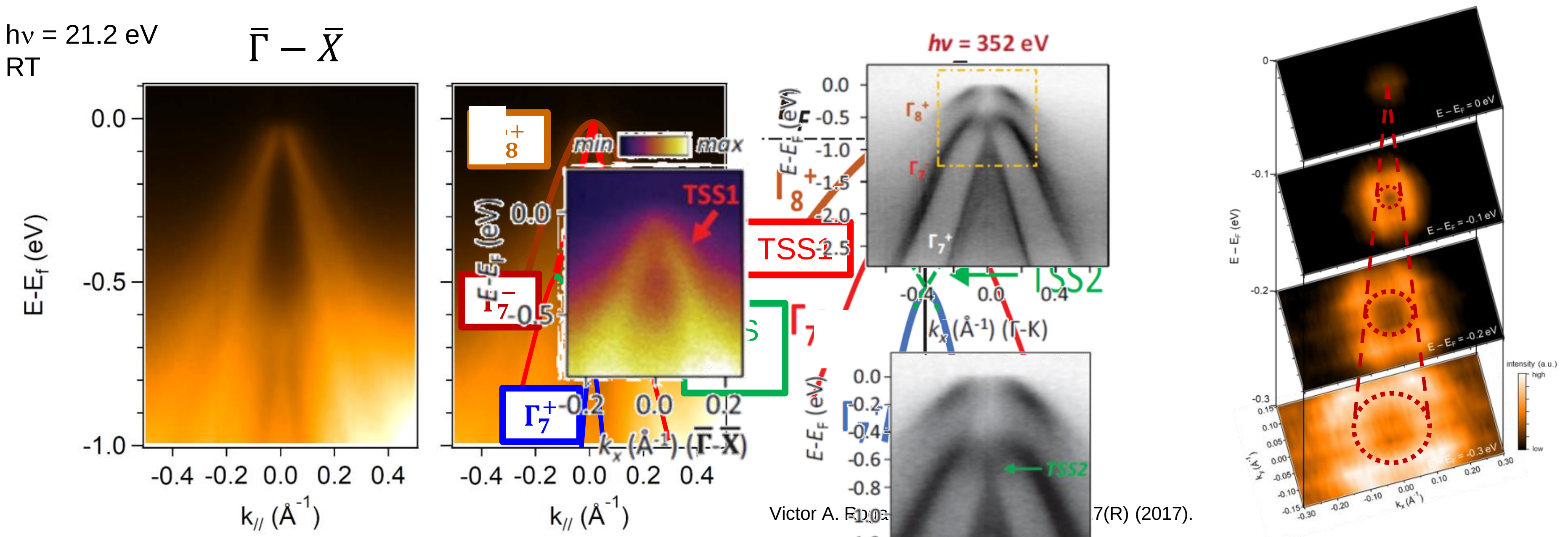
Using *in-situ* STM

50 x 50 nm^2



- Smooth surface R_q: 1.3 Å
- Real space image of 2x1 surface reconstruction
- Consistent with the observation in LEED

ARPES spectra of 30 BL α-Sn/InSb(001)



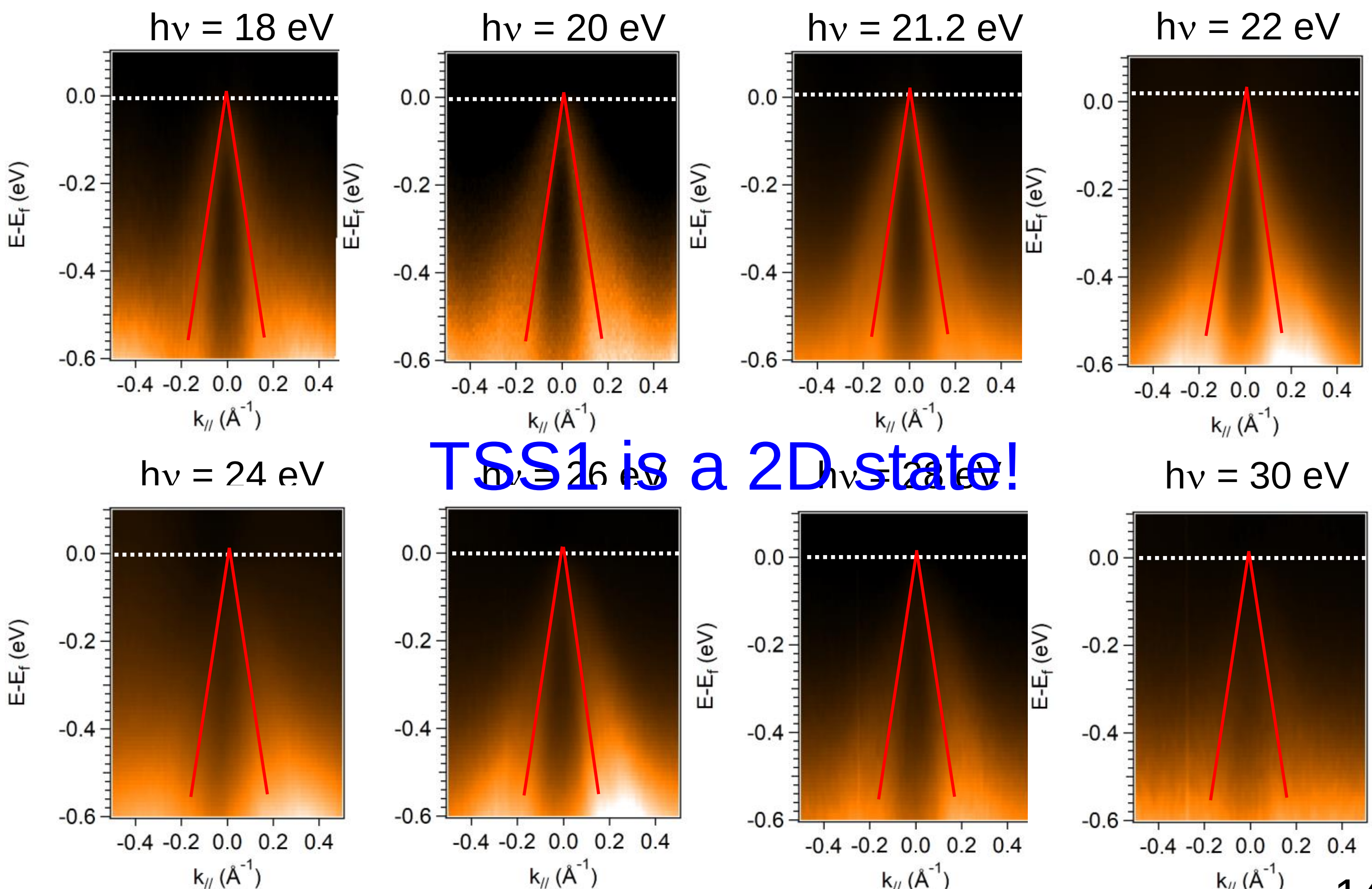
Victor A. Rogalev et al., Phys. Rev. B 95, 161117(R) (2017).

- Dirac-like state and three bulk bands were observed in our α-Sn/InSb(001) without adding Te in Sn or using Bi buffer layer.
- The Fermi velocity (v_F) of TSS1 is $(6.5-7)\times10^{5}\ m/s$, which is similar to reported value ($7.3\times10^{5}\ m/s$), larger than the one of Bi_2Se_3 ($2.9\times10^{5}\ m/s$) and smaller than the one of graphene ($1\times10^{6}\ m/s$) .

Victor A. Rogalev *et al.*, *Phys. Rev. B* **95**, 161117(R) (2017).

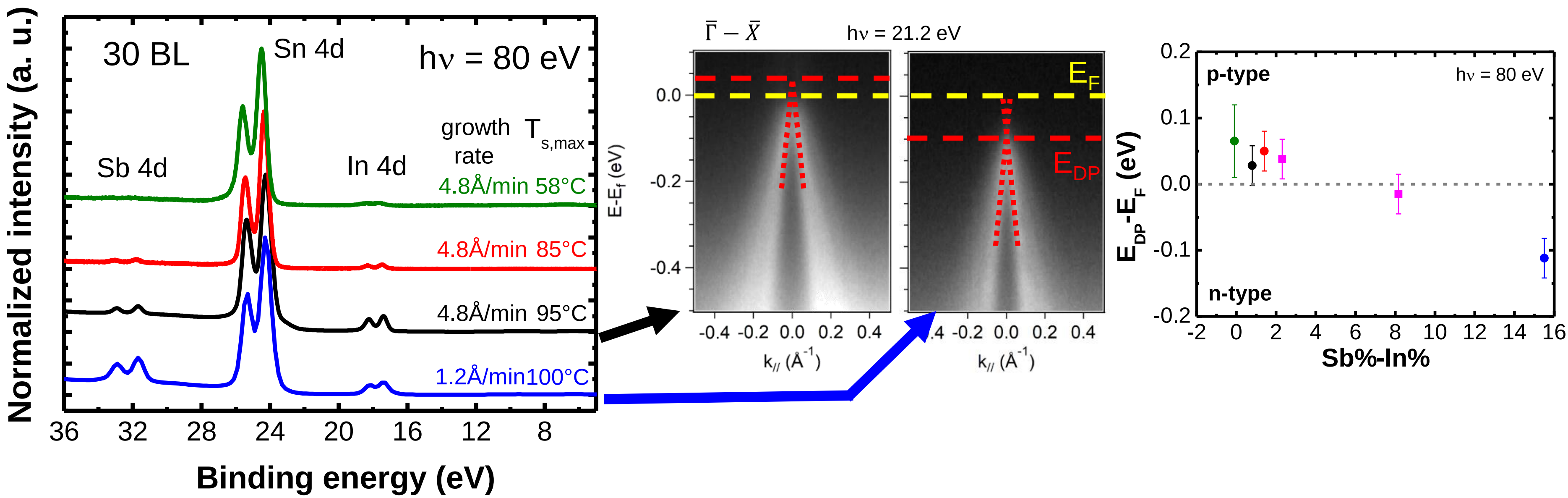
Y. Ohtsubo *et al.*, *Phys. Rev. Lett.* **111**, 216401 (2013).
K. Kuroda et al., *Phys. Rev. Lett.* **105**, 146801 (2010).
C. Berger et al., *Science* **312**, 1191 (2006).

Energy dependent ARPES spectra of α-Sn



TSS1 is a 2D state!

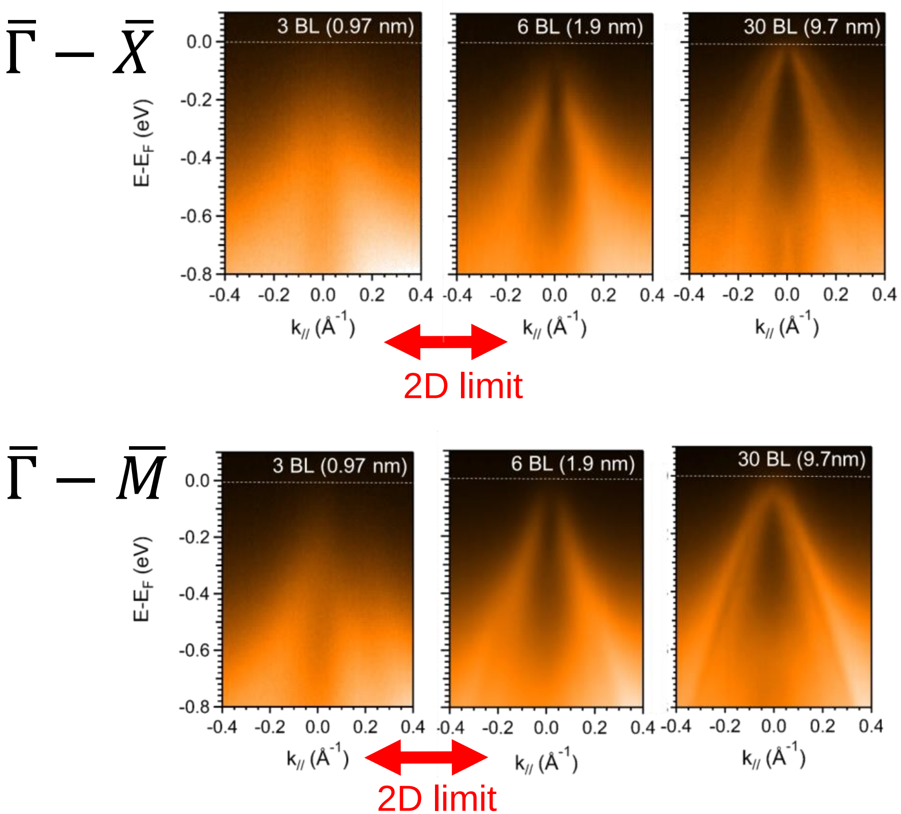
X-ray photoemission study

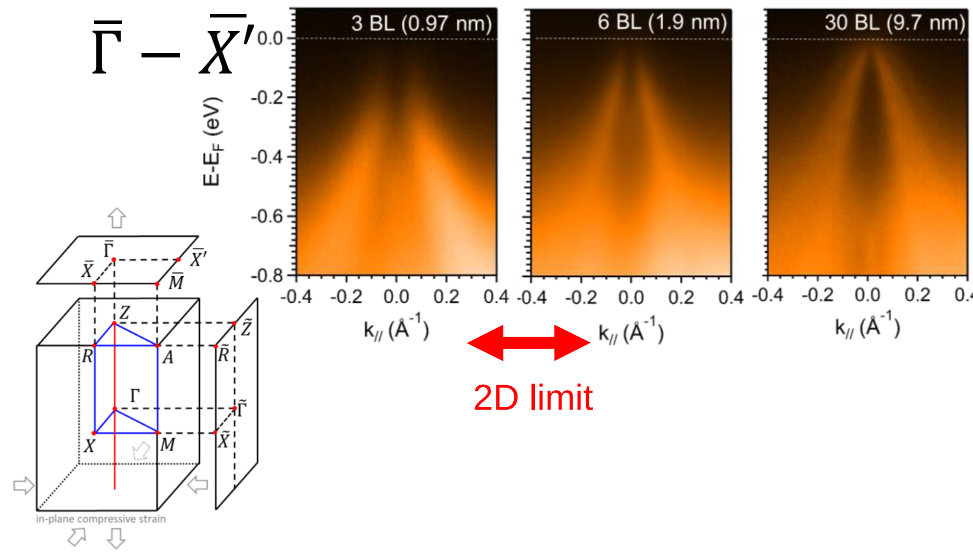


- Diffusion problem minimized by lowering substrate temperature
- Fermi level of α-Sn varied from p-type to n-type by controlling the inter-diffusion without severe degradation of TSS

Thickness dependent ARPES spectra of α-Sn(001): 3-30 BL

hν = 21.2 eV   RT



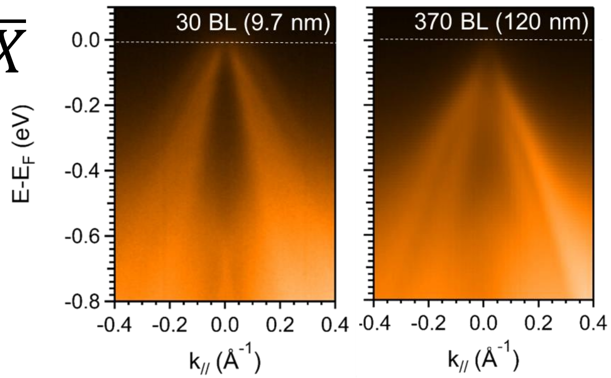


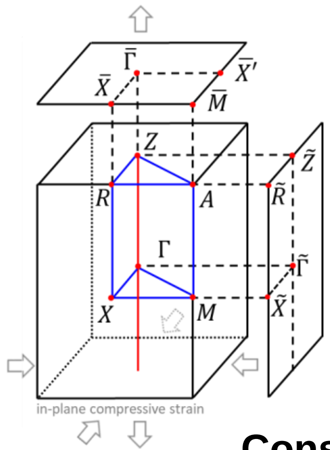
- TSS state becomes very weak and broadened in 6 BL sample
- No TSS observed in 3 BL sample
- 2D limit between 4-6 BL

Thickness dependent ARPES spectra of α-Sn(001): 30 BL and 370 BL

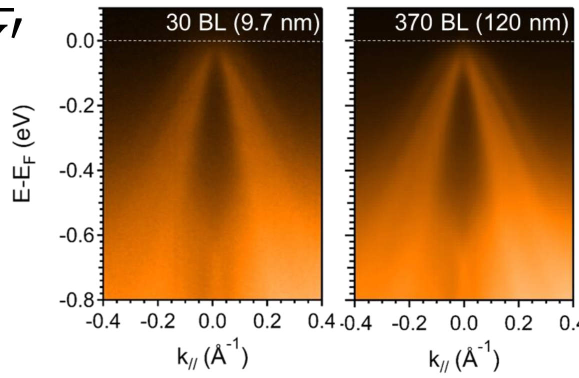
$h\nu$ = 21.2 eV RT

$\bar{\Gamma} - \bar{X}$

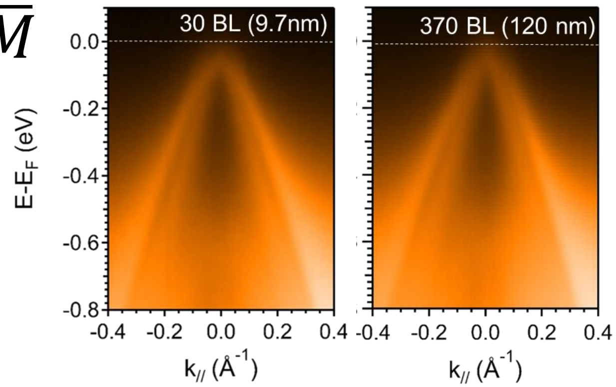




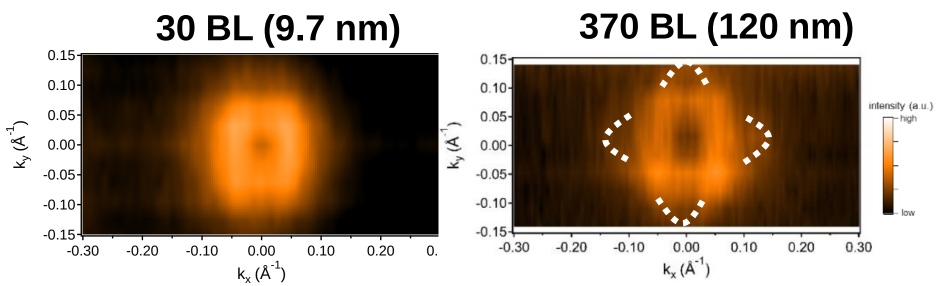
$\bar{\Gamma} - \bar{X}'$



$\bar{\Gamma} - \bar{M}$

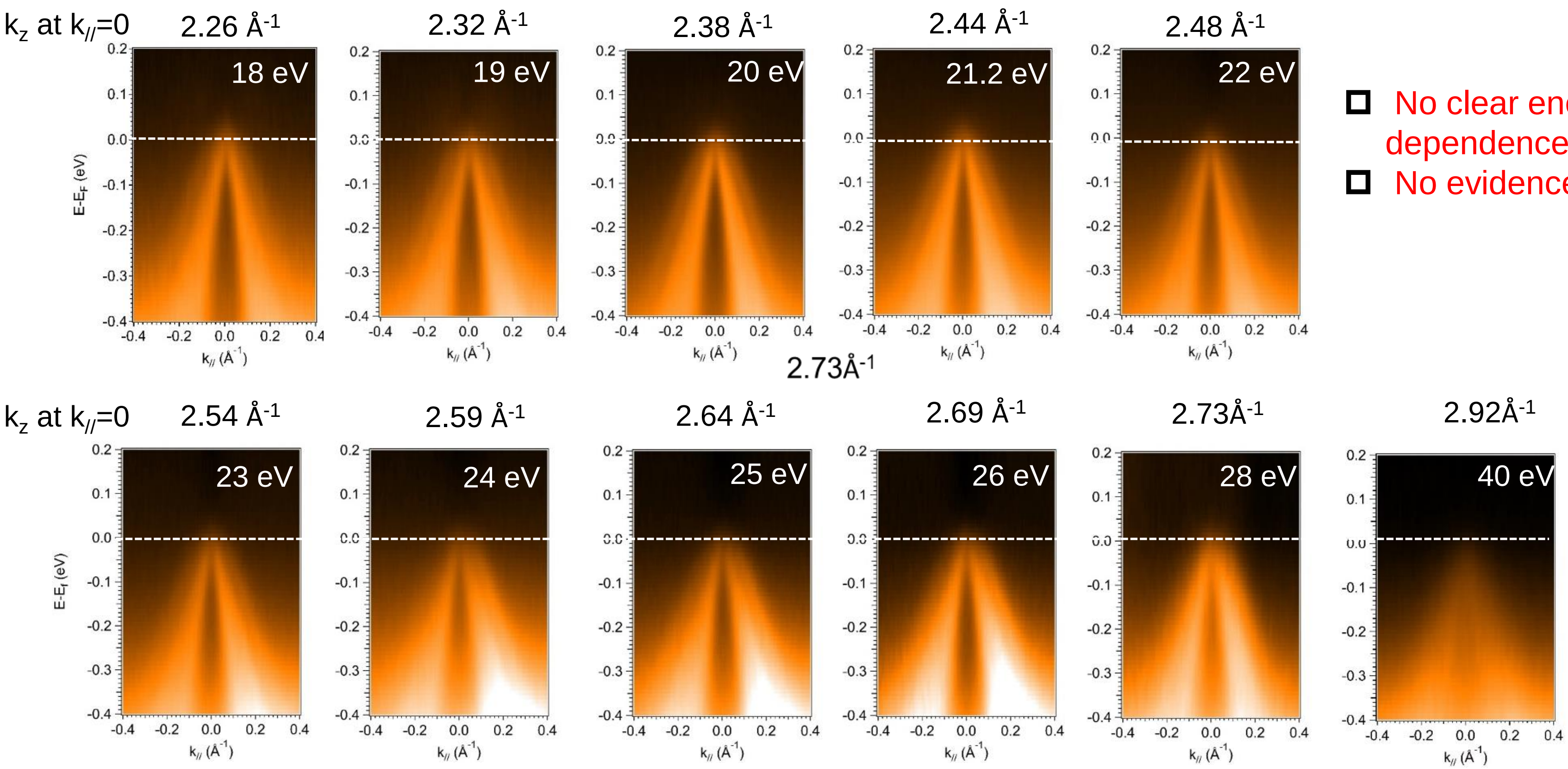


Constant energy contour (E - E$_F$ = -100 meV)



- More distinct bands

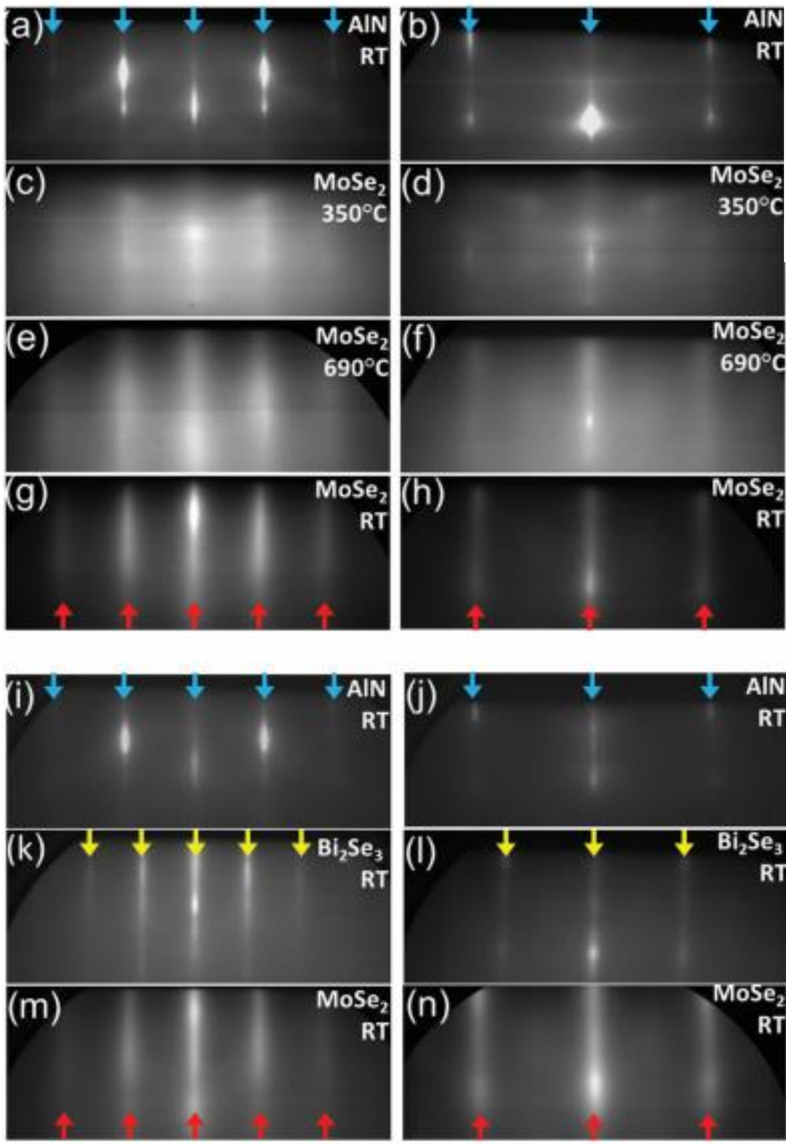
Energy-dependent analysis of 370 BL (120 nm) α-Sn/InSb(001)



- ☐ No clear energy dependence of TSS1
- ☐ No evidence of 3D-TDS

Thank you for your attention!

$[11\bar{2}0]$ $[1\bar{1}00]$



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High-quality, large-area $MoSe_2$ and $MoSe_2/Bi_2Se_3$ heterostructures on AlN(0001)/Si(111) substrates by molecular beam epitaxy†

E. Xenogiannopoulou,[a] P. Tsipas,[a] K. E. Aretouli,[a] D. Tsoutsou,[a] S. A. Giamini,[a] C. Bazioti,[b] G. P. Dimitrakopulos,[b] Ph. Komninou,[b] S. Brems,[c] C. Huyghebaert,[c] I. P. Radu[c] and A. Dimoulas*[a]

Fig. 1 RHEED patterns of: (a–h) 1 ML $MoSe_2$ deposited on AlN(0001) by the 2 step-process, and (i–n) 2 ML $MoSe_2$ deposited on a 5 QL Bi_2Se_3 buffer layer, epitaxially grown on AlN(0001). (a) and (i) show the bare AlN (0001) pattern along the [11−20] azimuth and (b and j) bare AlN(0001) along the [1−100] azimuth. (c) and (d) are recordings at 350 °C after the deposition of $MoSe_2$, (e) and (f) are recordings at 690 °C after the post-deposition annealing step, while (g) and (h) present the $MoSe_2$ film on AlN after final cooling down to RT. In (e) and (f) $MoSe_2$ becomes highly crystalline with the respective crystal orientations aligned to those of AlN, which is clearly observed after cooling down in (g) and (h). (k) and (l) show a 5 QL Bi_2Se_3 buffer layer grown epitaxially on AlN(0001) at 300 °C with high crystalline quality, (m) and (n) show that $MoSe_2$ deposited on the Bi_2Se_3 buffer layer at 300 °C grows epitaxially, with $[11-20]_{MoSe_2}//[11-20]_{Bi_2Se_3}$ and $[1-100]_{MoSe_2}//[1-100]_{Bi_2Se_3}$. The blue and yellow downward arrows show AlN and Bi_2Se_3 streaks, respectively. The red upward arrows show $MoSe_2$ streaks.

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Topological insulator Bi_2Se_3 thin films grown on double-layer graphene by molecular beam epitaxy

Can-Li Song,[1,2] Yi-Lin Wang,[1] Ye-Ping Jiang,[1,2] Yi Zhang,[1] Cui-Zu Chang,[1,2] Lili Wang,[1] Ke He,[1] Xi Chen,[2] Jin-Feng Jia,[2] Yayu Wang,[2] Zhong Fang,[1] Xi Dai,[1] Xin-Cheng Xie,[1] Xiao-Liang Qi,[3] Shou-Cheng Zhang,[3] Qi-Kun Xue,[1,2,a)] and Xucun Ma[1]

[1]*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People's Republic of China*
[2]*Department of Physics, Tsinghua University, Beijing 100084, People's Republic of China*
[3]*Department of Physics, Stanford University, Stanford, California 94305-4045, USA*

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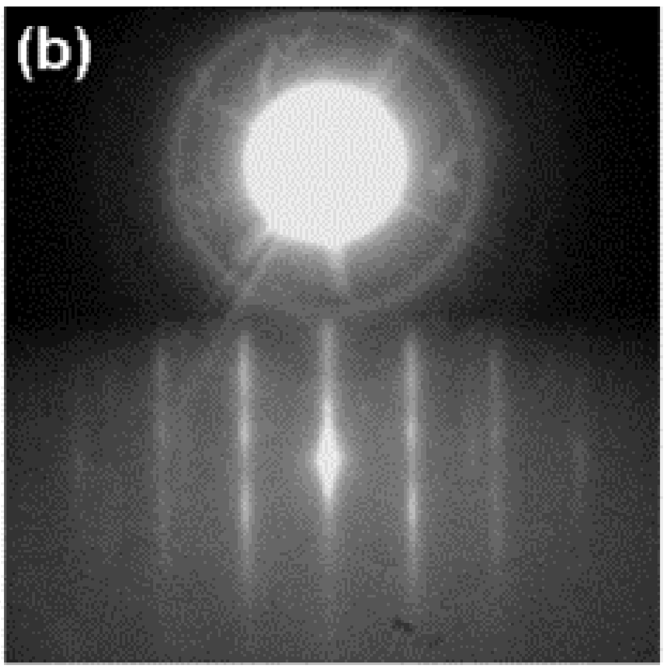

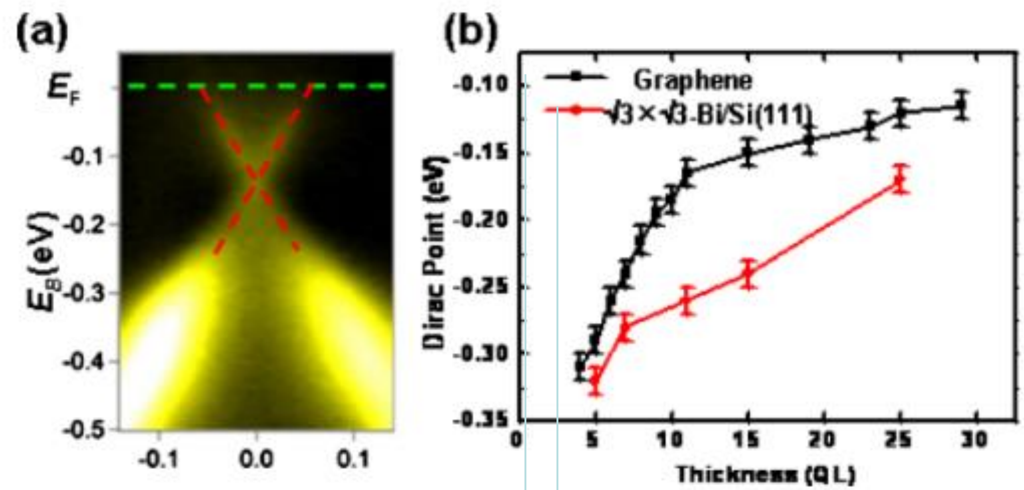

FIG. 2. (Color online) (a) ARPES intensity map of the 26 QL film along the Γ-K direction. The dotted line indicates the Fermi level, and the topological surface states, respectively. (b) Thickness-dependent surface Dirac point of Bi_2Se_3 films on graphene and Si substrates.