Unraveling Proximity and Topology at Interfaces with Next Generation Neutron Reflectometry

Alexander Grutter

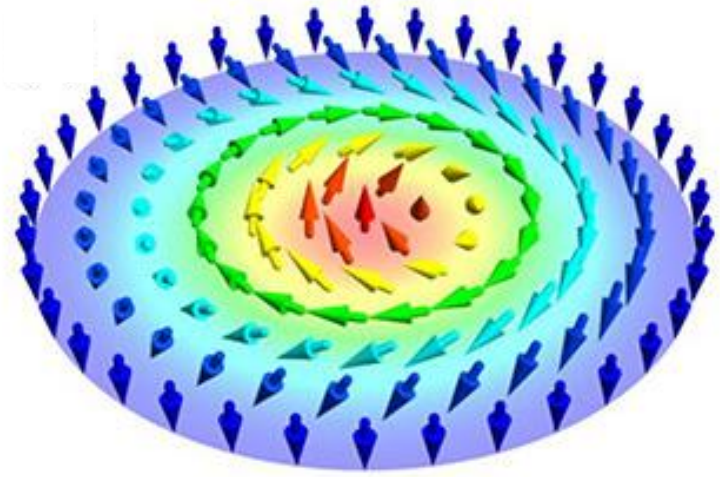
NIST Center for Neutron Research

CHRNS
CENTER FOR HIGH RESOLUTION NEUTRON SCATTERING
NSF
NIST

Topology, Magnetism, and Transport

Disclaimer: I am a neutron scatterer, not an expert on topology or electronic structure

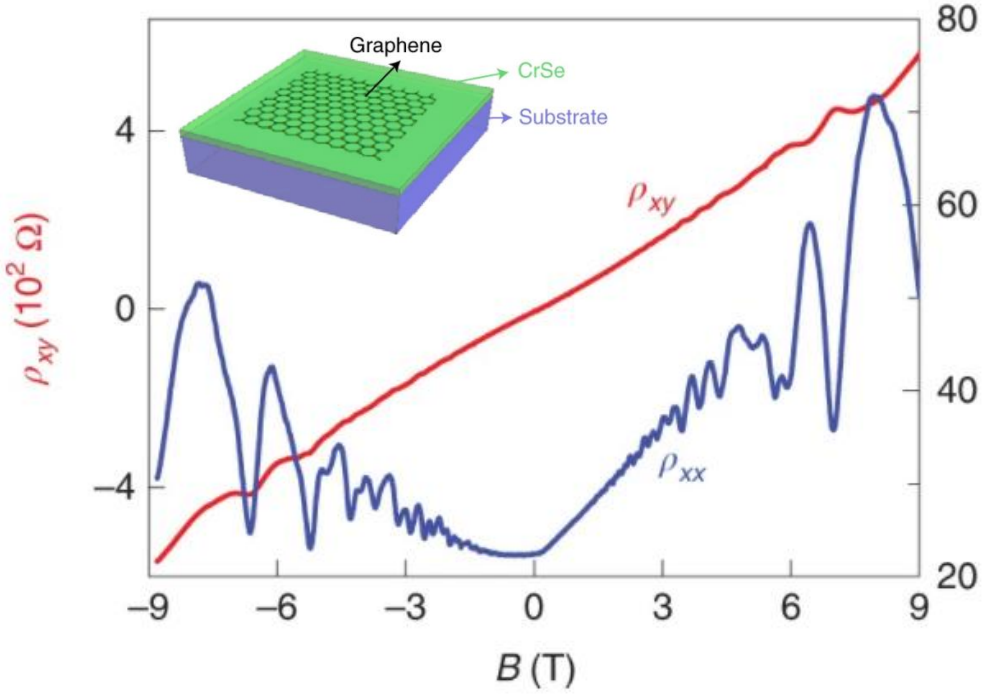
Nontrivial topology in real space – **Skyrmions**



P. K. Sivakumar et al., ACS Nano 14, 13463 (2020)

Winding affords some degree of protection against skyrmion annihilation

Magnetic field-induced topology in electronic states - **Quantum Hall Effect**



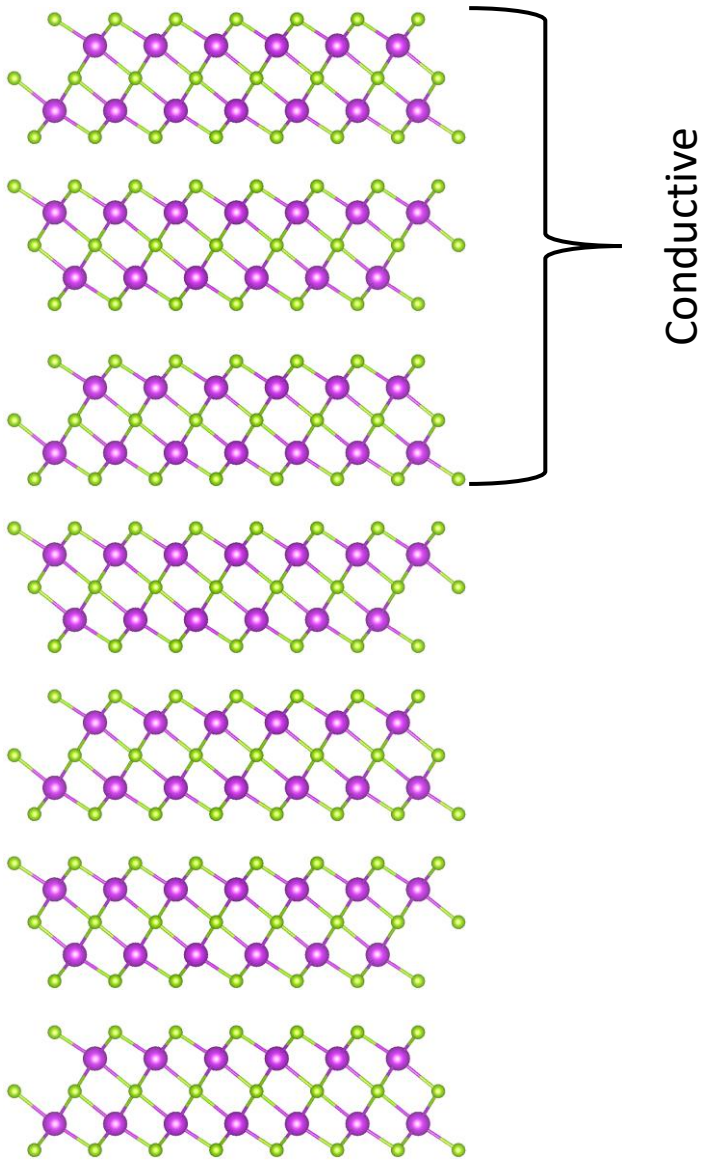
Y. Wu et al., Nat. Electronics 3, 604 (2020)

Coherent conduction along edge channel

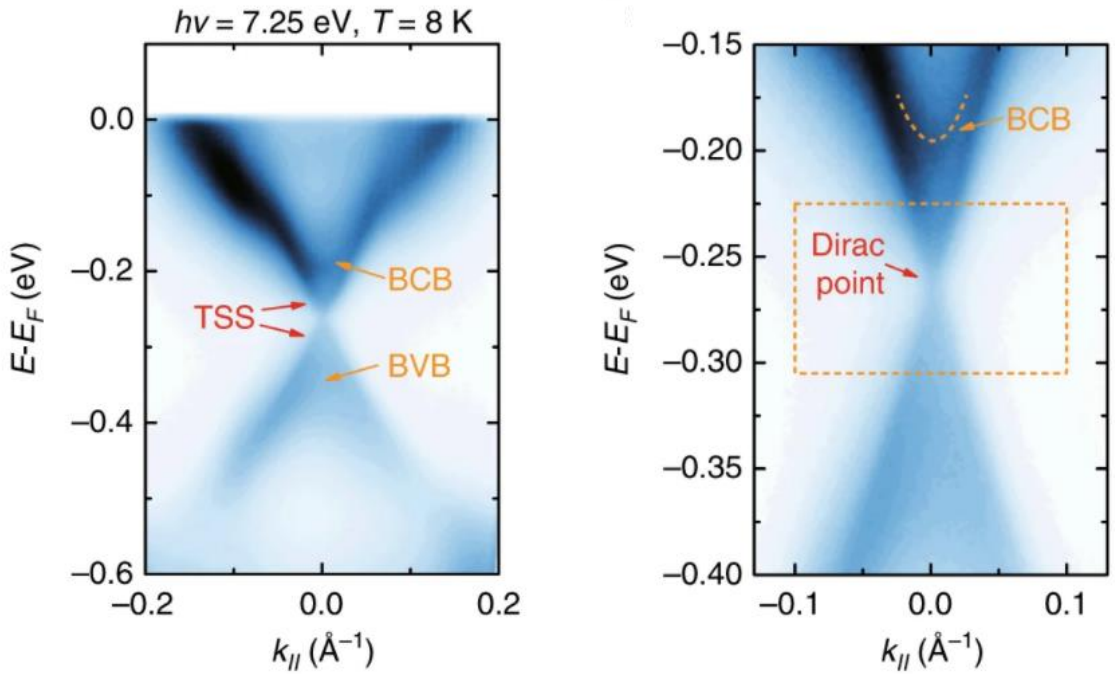
A Very Approximate Outline

- Why does anyone care about magnetic proximity effects (MPEs) in topological insulators?
- ***Are*** there MPEs in topological insulators?
- How do we ***currently*** characterize and understand MPEs (with examples)?
- How ***should*** we characterize MPEs in topologically nontrivial systems?

Topological Insulators (Bi_2Se_3/Bi_2Te_3/$(Bi,Sb)_2Te_3$)



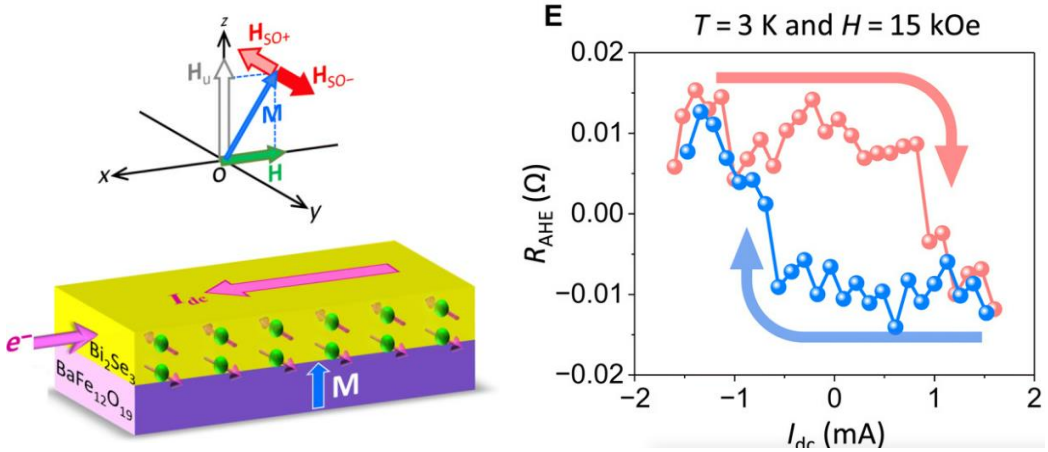
- Conductive surface state extends 3 nm from surface
- If top and bottom touch, hybridization…
- Dirac cone = massless Dirac fermions
- Strong spin-orbit interactions yield spin-momentum locking *i.e.* Spin-dependent conduction channels

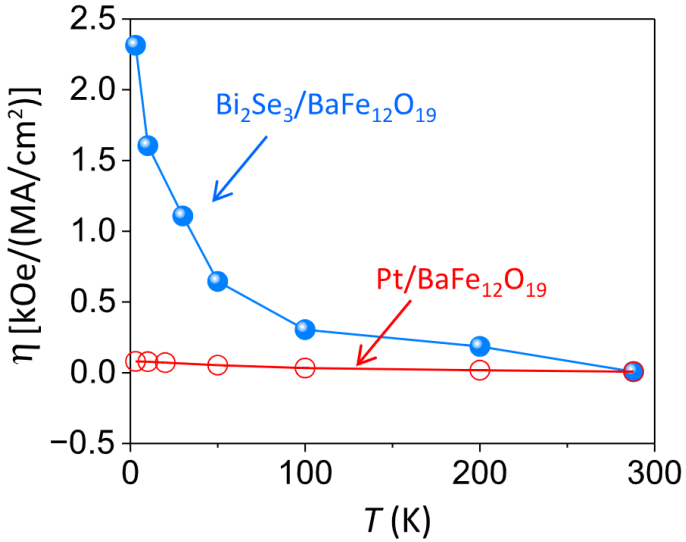


Chen, B., Fei, F., Zhang, D. *et al.* Intrinsic magnetic topological insulator phases in the Sb doped $MnBi_2Te_4$ bulks and thin flakes. *Nat Commun* **10,** 4469 (2019). https://doi.org/10.1038/s41467-019-12485-y

Topological Spintronics and Novel Quantum States

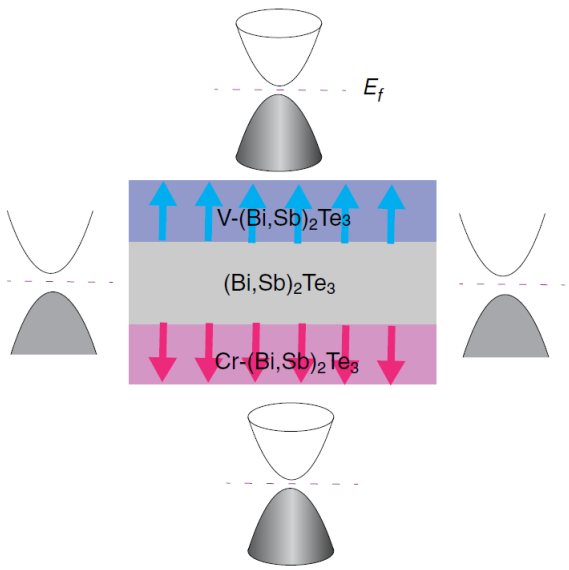
Extremely high spin-orbit torque efficiency

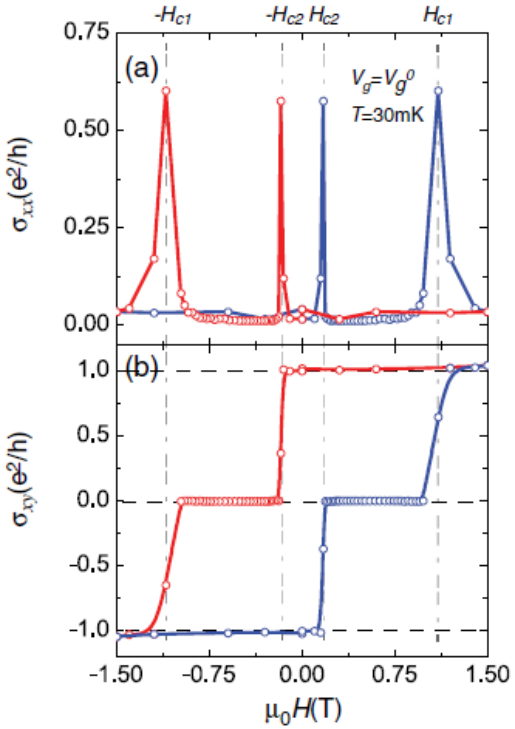




P. Li et al., Sci. Advances 5, eaaw3415 (2019)
DOI: 10.1126/sciadv.aaw3415

Axion Insulator State

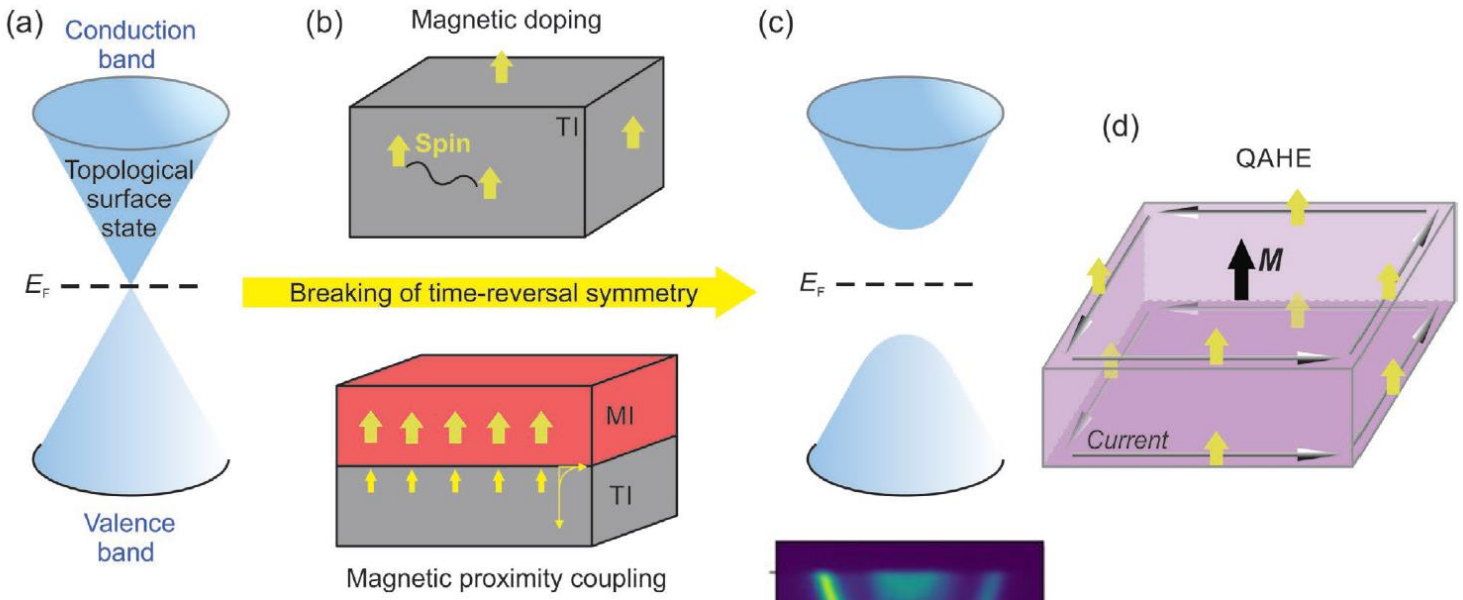




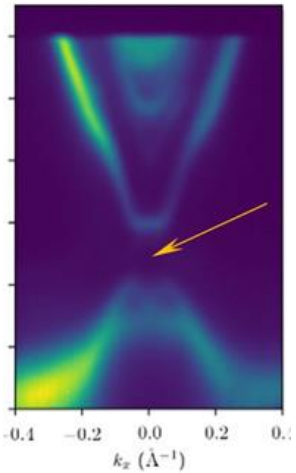
D. Xiao et al., Phys. Rev. Lett. 120, 056801 (2018)
Also see: M. Mogi et al., Nat. Materials 16, 516 (2017)

Magnetic TIs & Quantum Anomalous Hall Effect

- Adding magnetic impurities into the system opens a gap in the surface state
- Dirac fermions gain mass due to band curvature

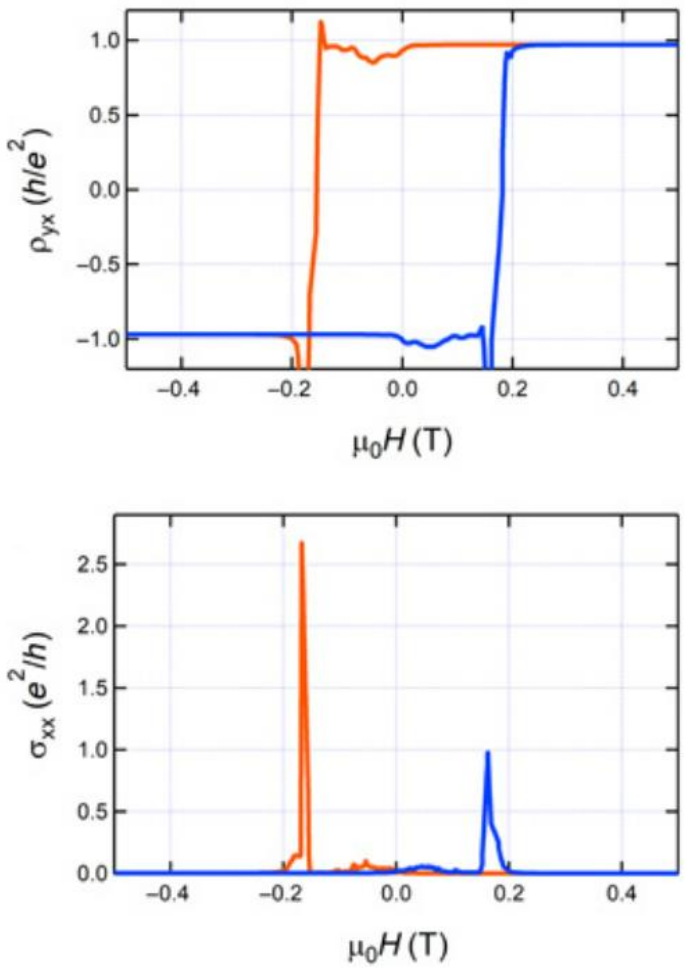


J. Liu and T. Hesjedal, Adv. Mater. 2102427 (2021)
https://doi.org/10.1002/adma.202102427



C. Hu et al., Sci. Advances 6, eaba4275 (2020)
DOI: 10.1126/sciadv.aba4275

Quantum Anomalous Hall Effect



L. Pan et al., Science Advances 6, eaaz3595 (2020)
C. Z. Chang et al., Science 340, 167 (2013)

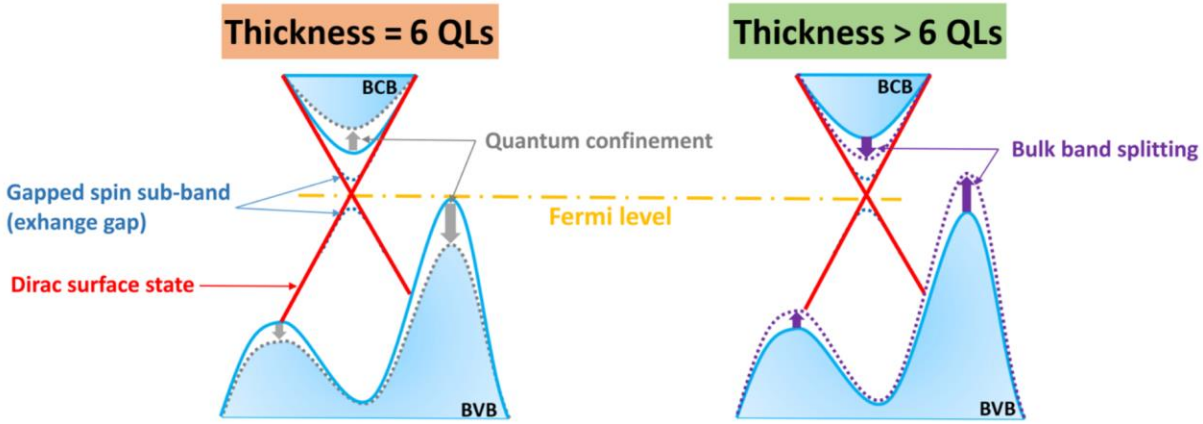
Homogeneity and Temperature Limitations

Doped $(Bi,Sb)_2Te_3$ structures

- has been limited to ≤ 2 K
- Possible explanations include
  - Dopant inhomogeneity (Cr, V, etc.)

    E. O. Lachman et al., Science Advances 1, e1500740 (2015)
  - TI crystal inhomogeneity (Bi, Sb)

    S. Qi et al., Phys. Rev. B 101, 241407(R) (2020)
  - Topologically trivial conduction pathways (defects)



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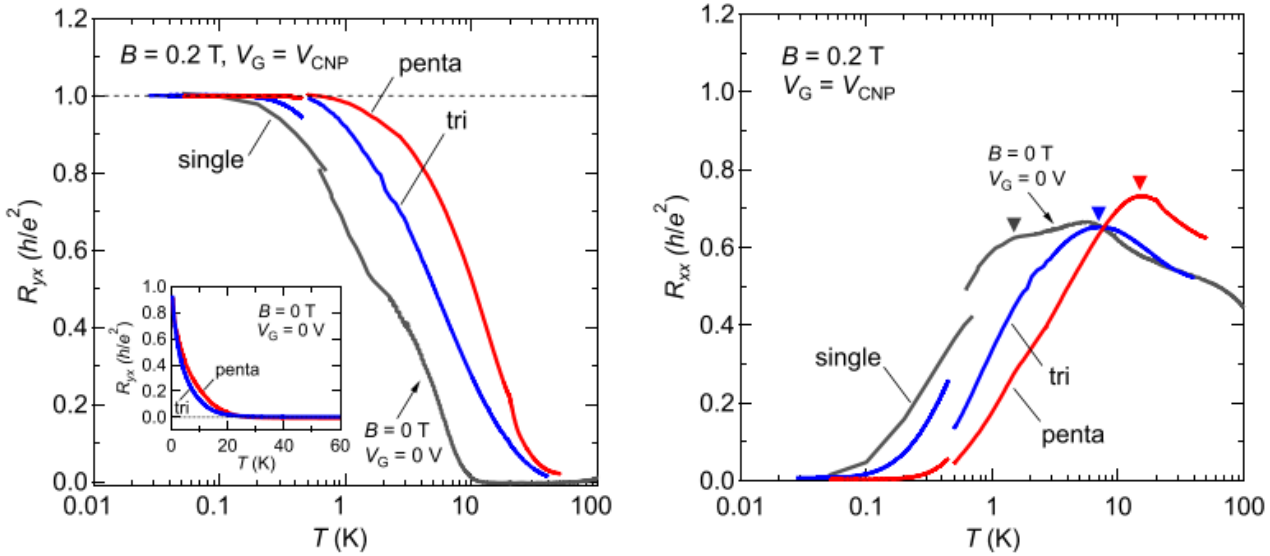
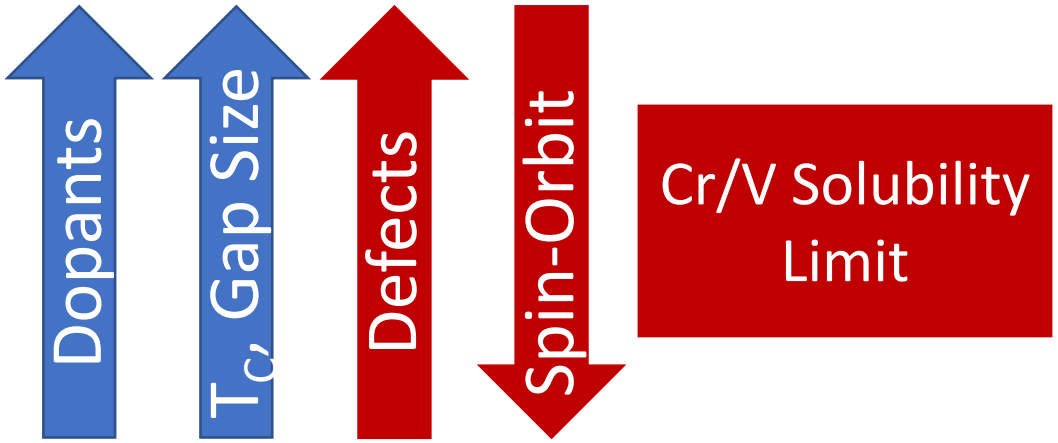


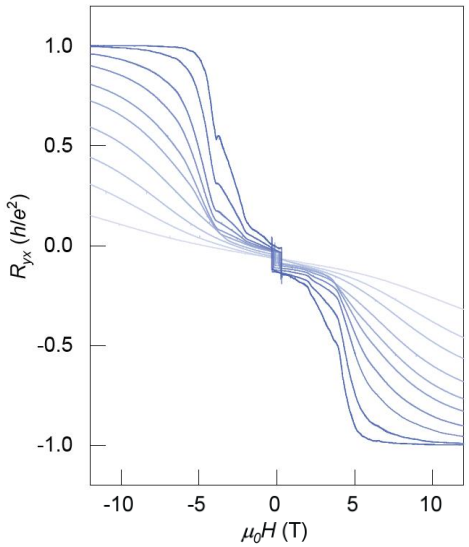
M. Mogi et al., Appl. Phys. Lett. 107, 182401 (2015)

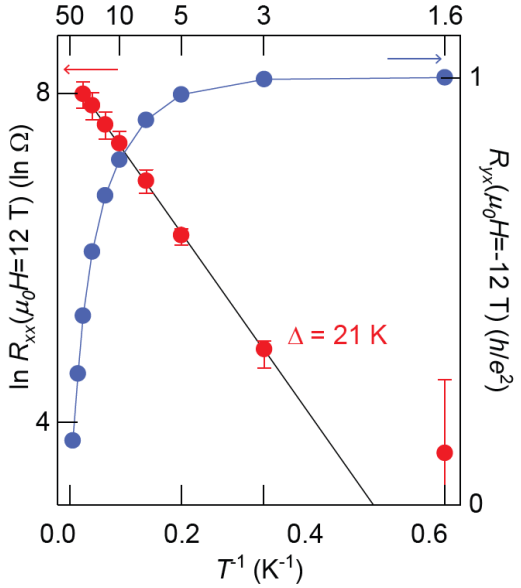


Intrinsically Magnetic TIs

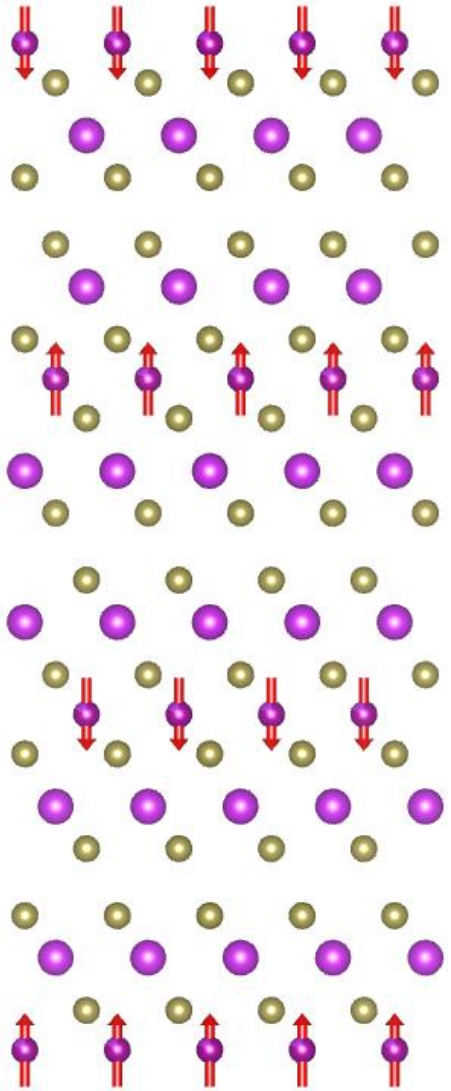
$MnBi_2Te_4$: An antiferromagnetic TI

- Zero-field QAH effect at 1.4 K
- In high magnetic field, 6.5 K
- Why is the temperature still low?
  - Exchange gap expected to be large
  - Magnetic inhomogeneity/defects?
  - Trivial conduction pathways?

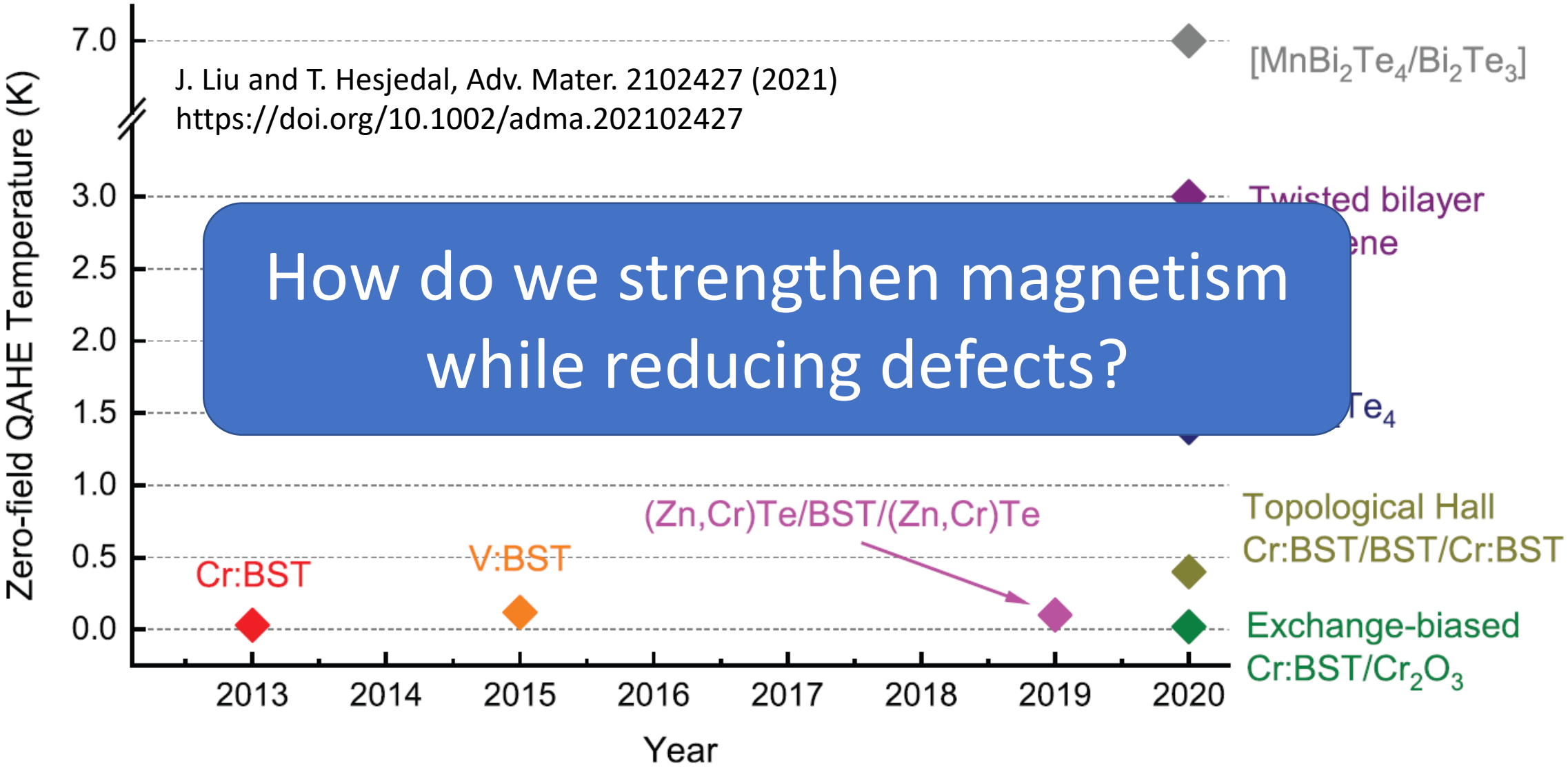




Y. Deng et al., arXiv:1904.11468v1 (2019)



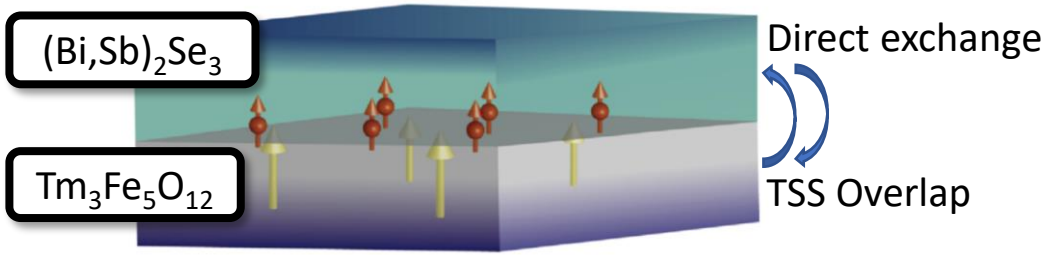
Progress and Temperature Limitations

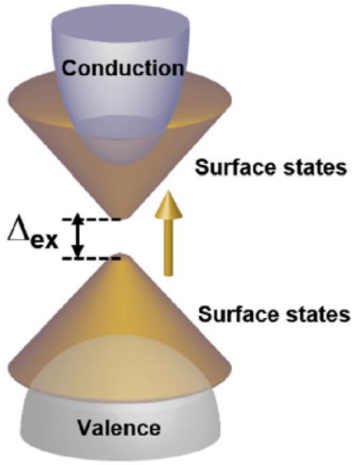


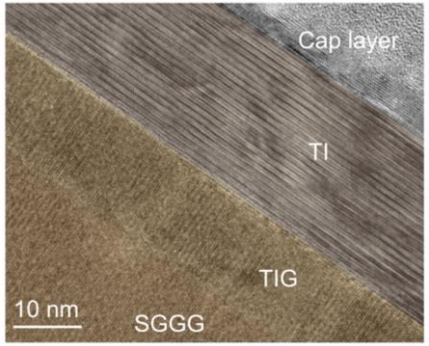
How do we strengthen magnetism while reducing defects?

Magnetic Proximity Effects

Interfacial Proximity Effects:



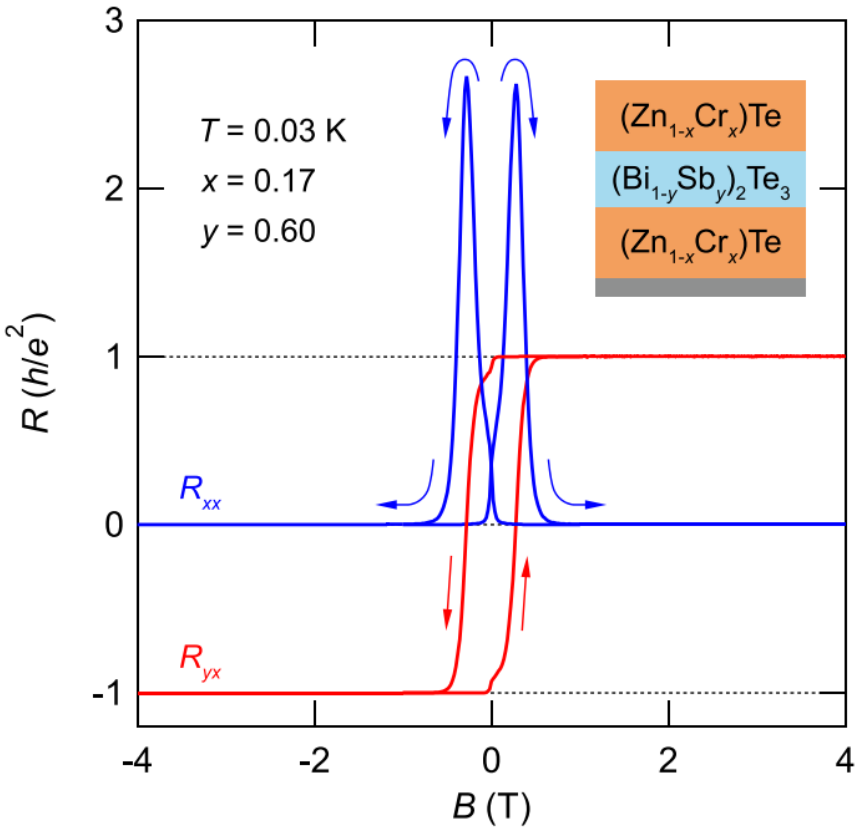


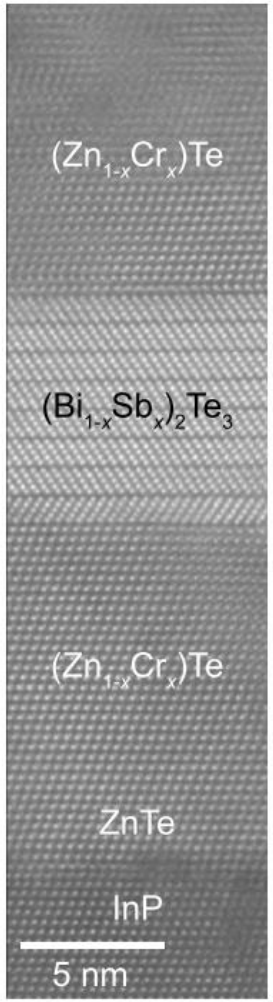


C. Tang et al., Science Advances 3, e1700307 (2017)

- $Y_3Fe_5O_{12}/Bi_2Se_3$ — *M. Lang et al., Nano Lett. 14, 3459 (2014)*
- $EuS/Sb_{2-x}V_xTe_3$ — *M. Li et al., PRL 115, 087201 (2015)*
- EuS/Bi_2Se_3 — *F. Katmis et al., Nature 533, 513 (2016)*
- $CrSb/(Bi,Sb)_{2-x}Cr_xTe_3$ — *Q. L. He et al., Nat. Mat. 16, 94 (2017)*

BST/$(Zn_{1-x}Cr_x)Te$ Interface:





R. Watanabe et al., *Applied Phys. Lett.* **115**, 102403 (2019)

Magnetic Proximity Effects

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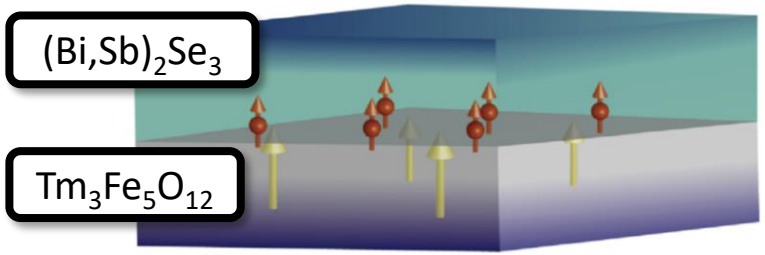

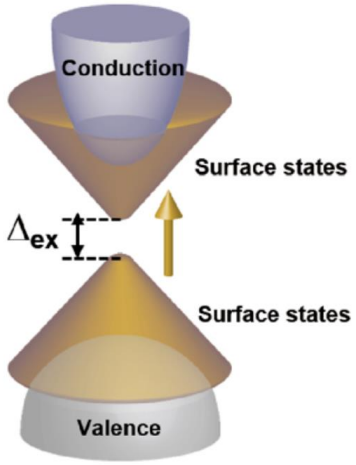

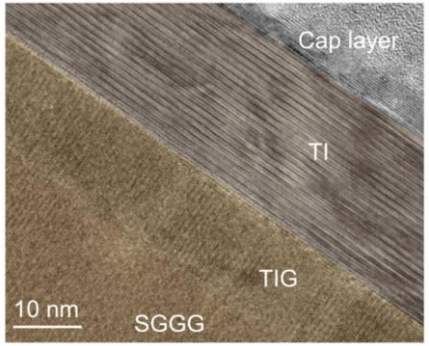

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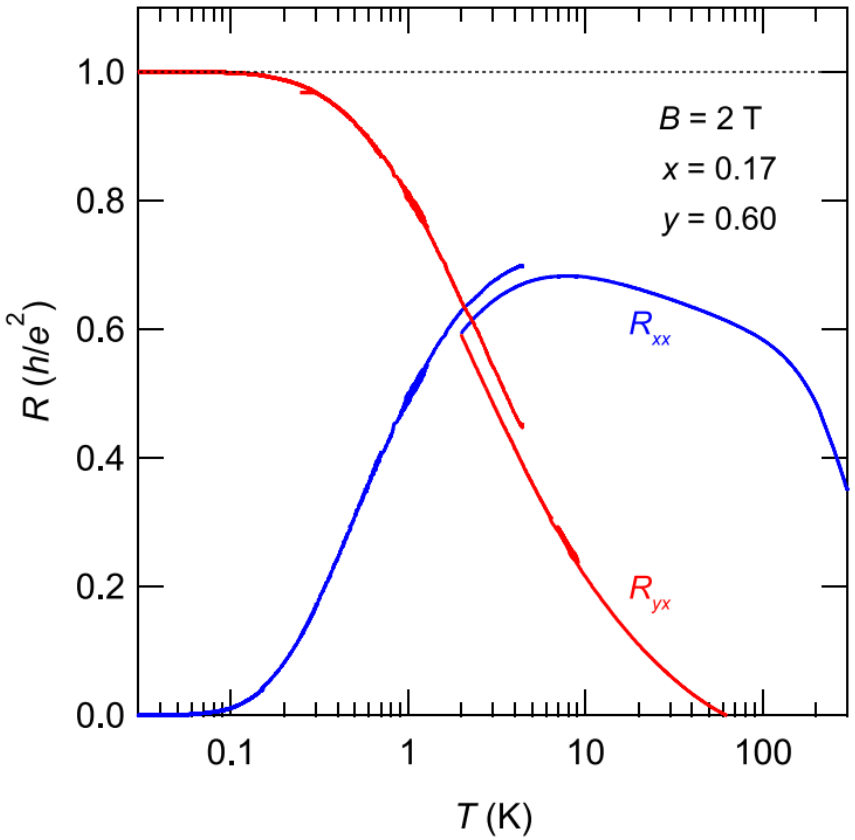
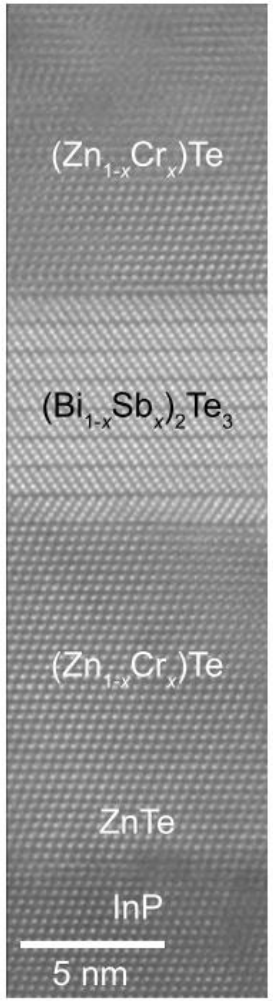


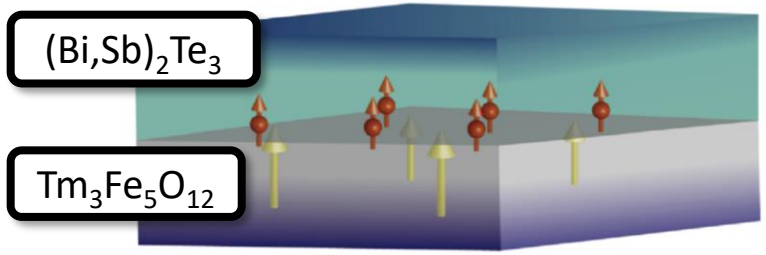


Still have dopant-related inhomogeneity?

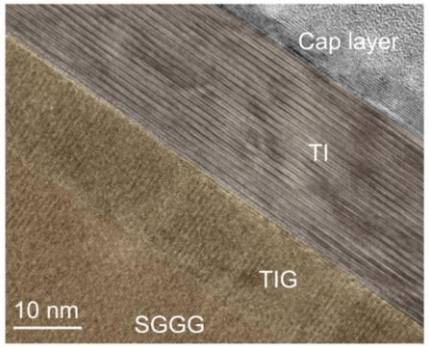
R. Watanabe et al., *Applied Phys. Lett.* **115**, 102403 (2019)

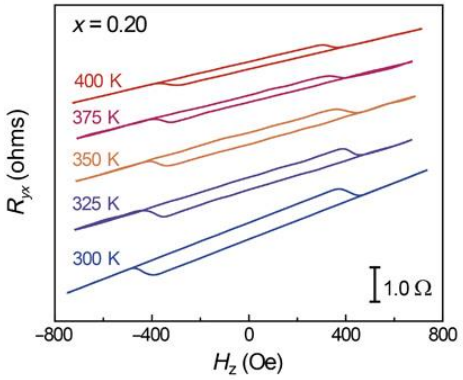
Uniform Proximity Reservoirs and Transport Data

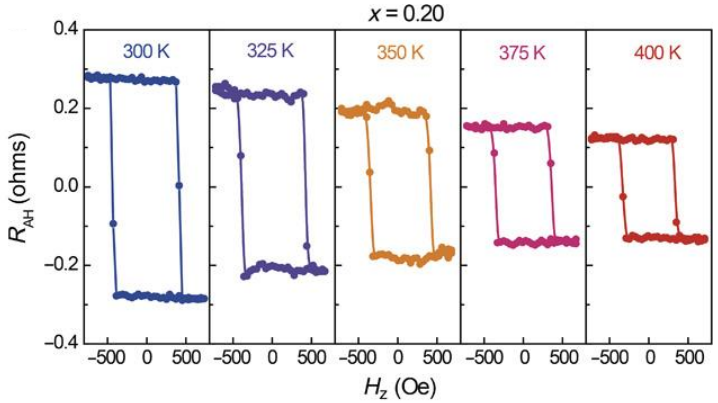
One Conductive Layer: Transport



C. Tang et al., *Science Advances* 3, e1700307 (2017)





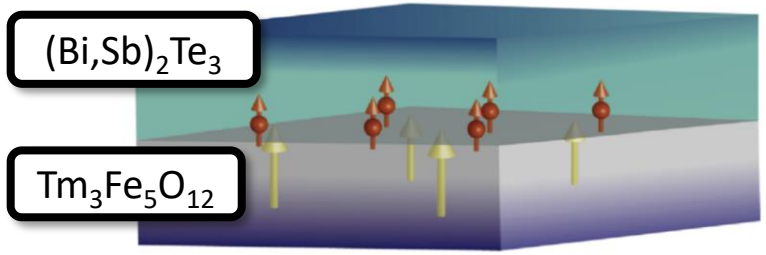


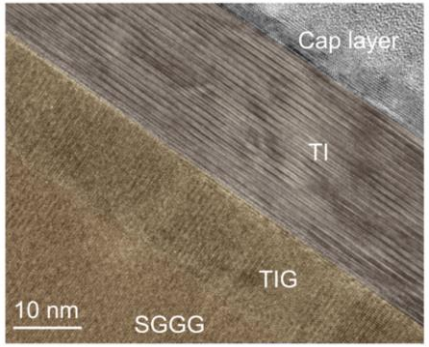
- Uniform magnet/TI heterointerfaces have led to the highest-temperature proximity effects reported
- BUT have yet to realize the QAHE
- $Cr_2Ge_2Te_6/(Bi,Sb)_2Te_3$ might be close *(M. Mogi et al., Phys. Rev. Lett 123, 016804)*

More direct probes?

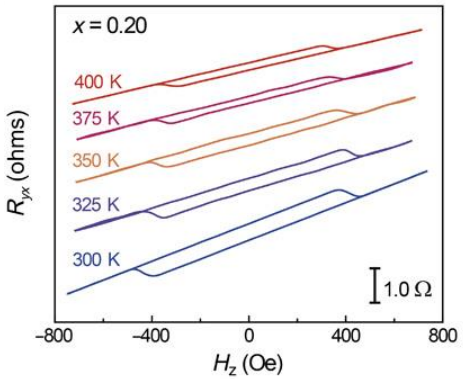
X-ray spectroscopy or neutron reflectometry

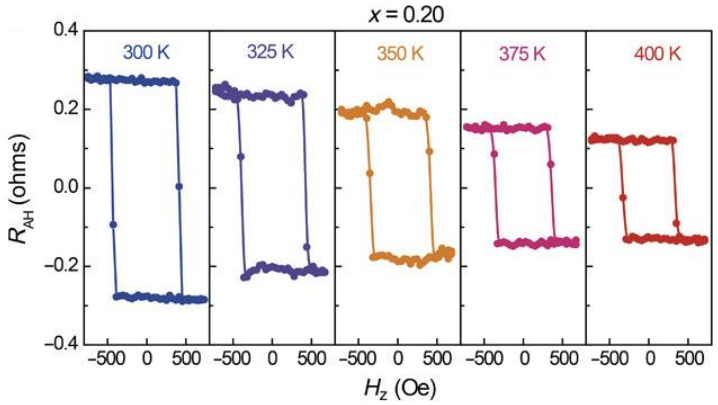
X-ray Spectroscopy: Element-Specific Information

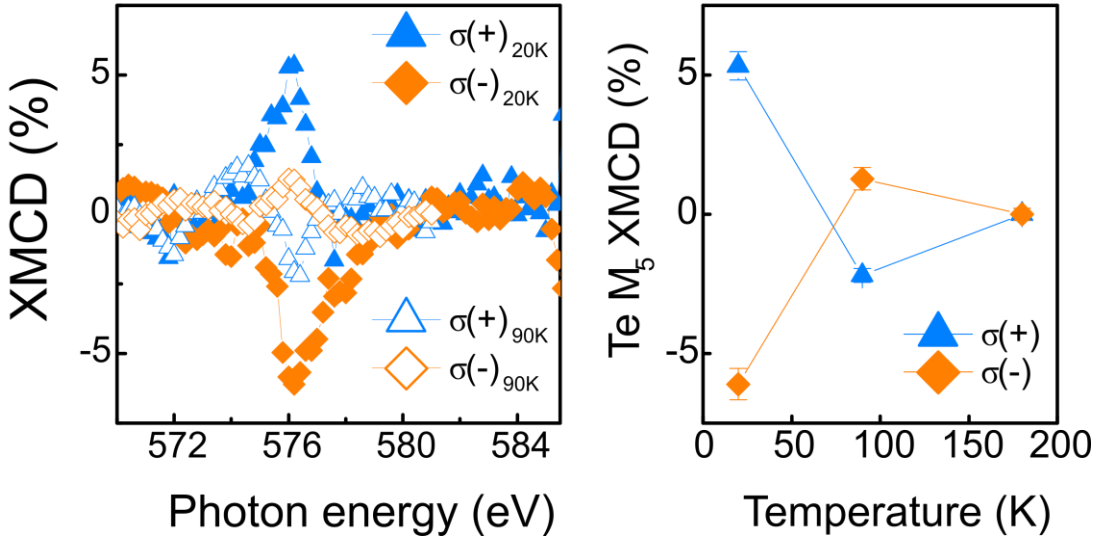




C. Tang et al., *Science Advances* 3, e1700307 (2017)



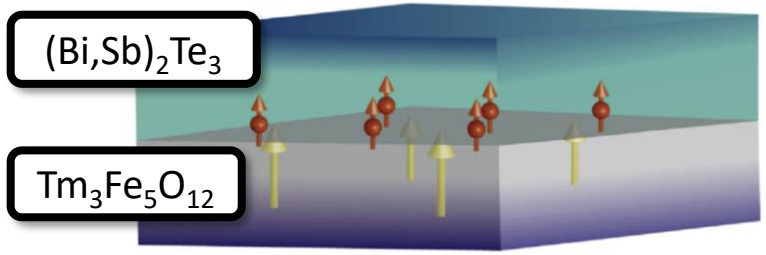


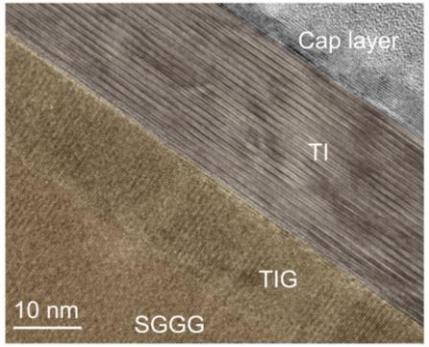


C.-Y. Yang, Appl. Phys. Lett. **114**, 082403 (2019)

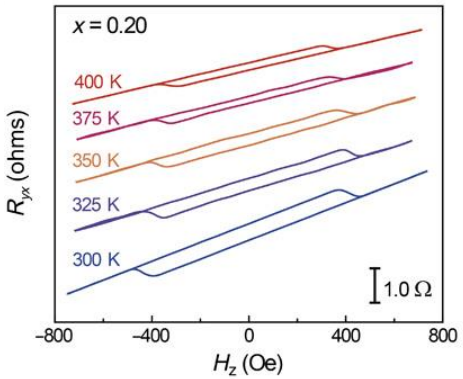
Transport shows hysteresis > 400 K, Te XMCD is gone by 200 K. Proves a proximity effect, but temperature-dependence contradictory.

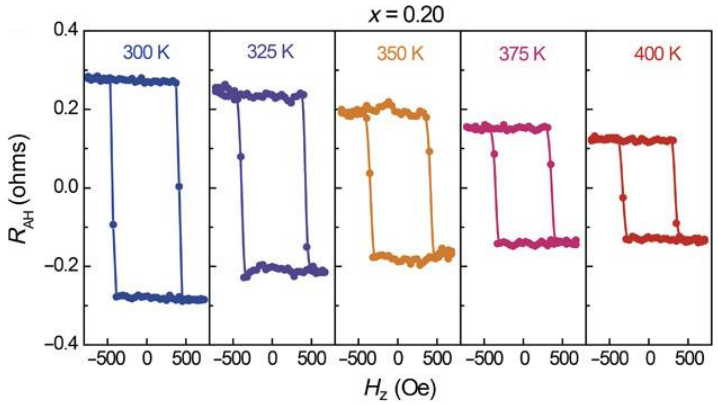
X-ray Spectroscopy: Element-Specific Information



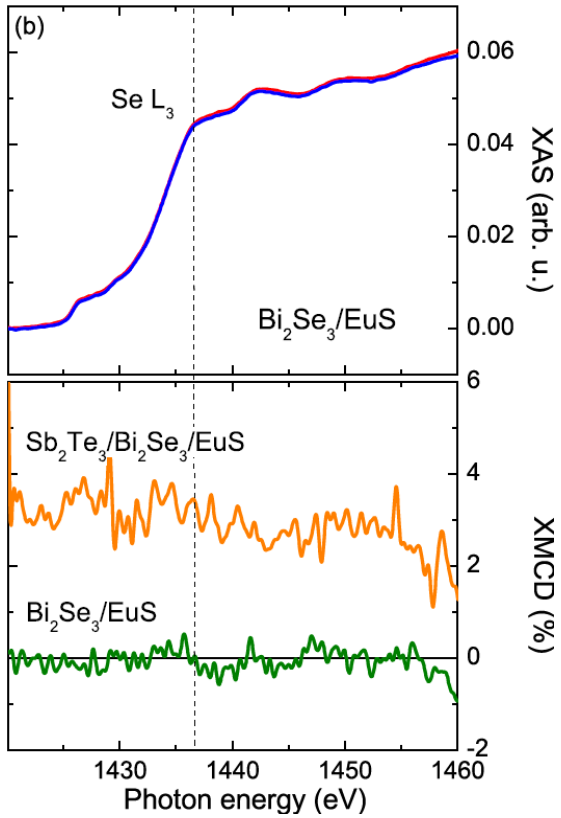


C. Tang et al., *Science Advances* 3, e1700307 (2017)



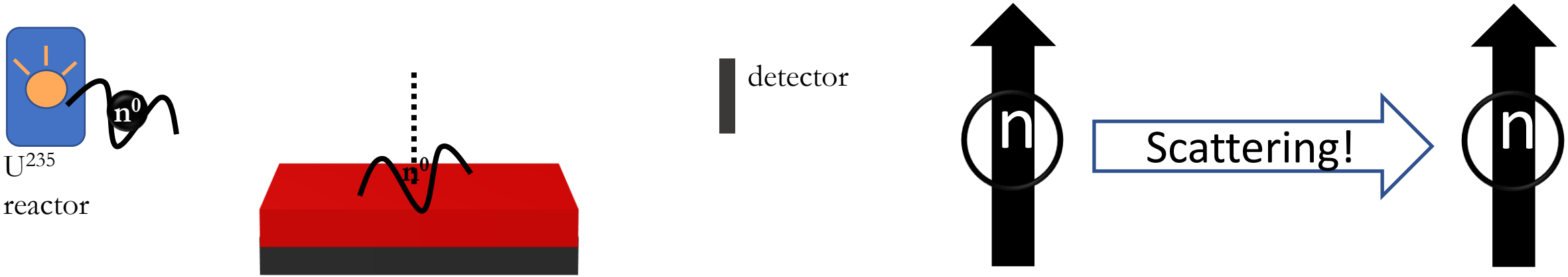


XMCD: No proximity effect in EuS/Bi_2Se_3 (Previously reported at room temperature)



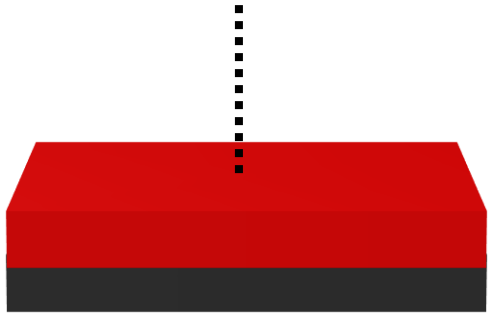
A. I. Figueroa et al., PRL **125,** 226801 (2020)

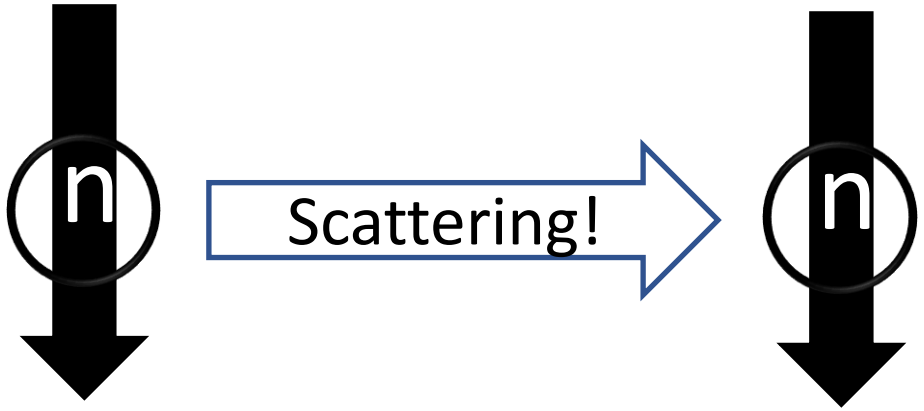
Polarized Neutron Reflectometry

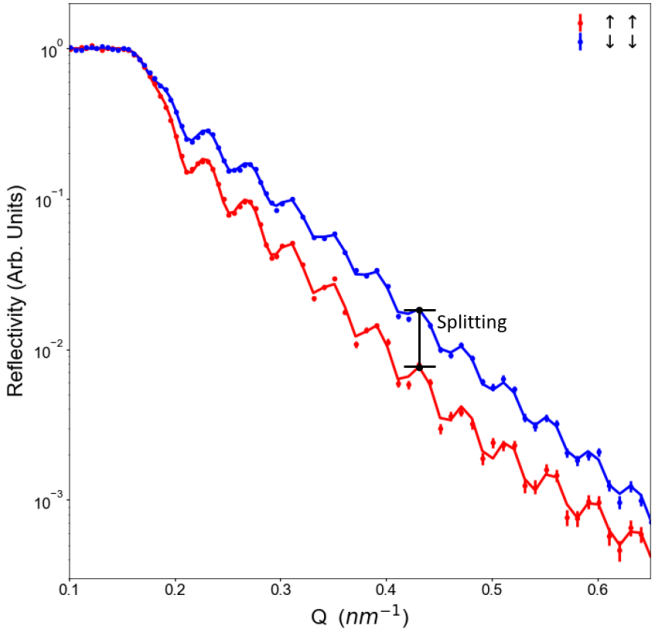


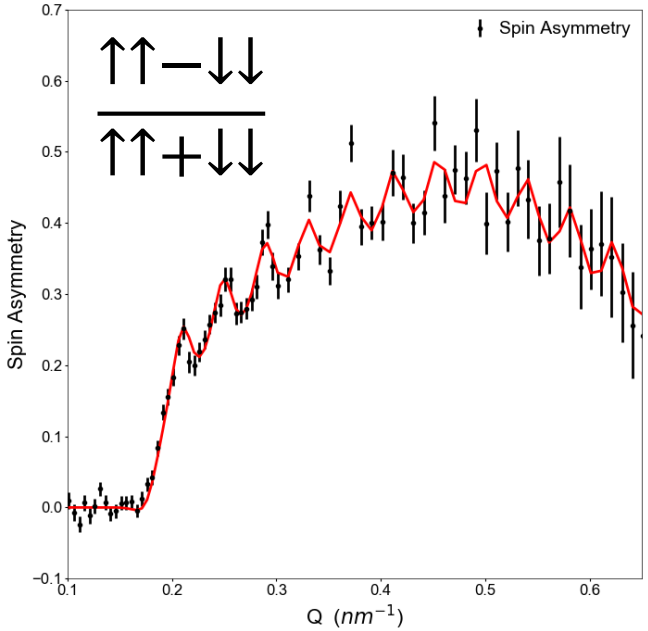
Polarized Neutron Reflectometry

U^{235} reactor

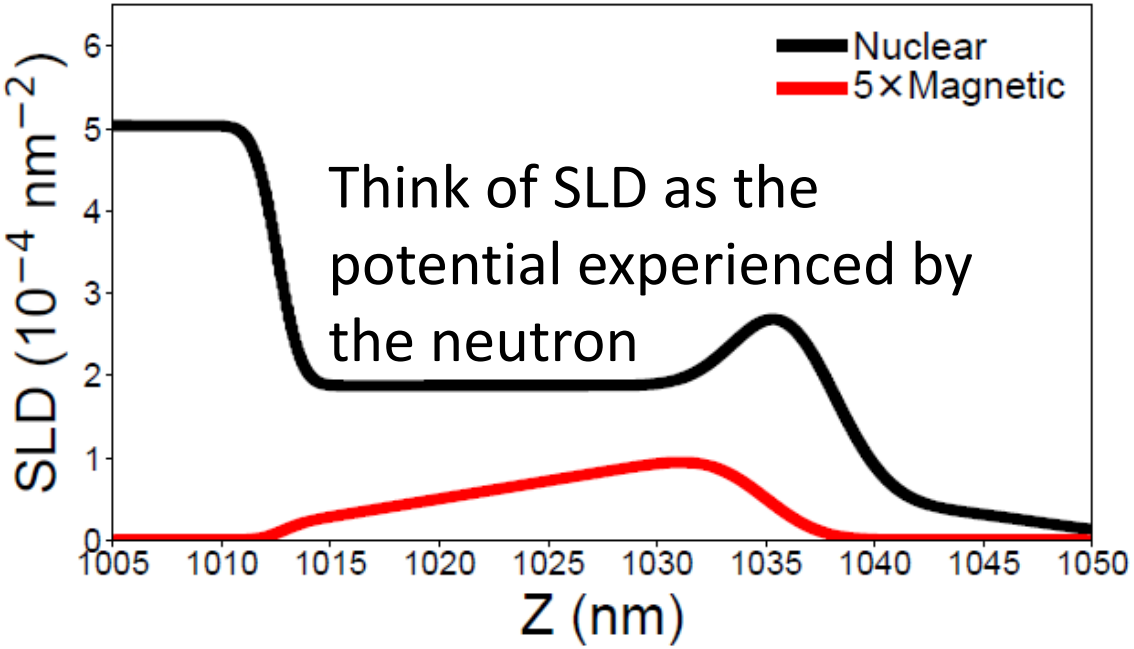








L. Pan et al., Advanced Materials 32, 2001460 (2020)



Example:
$MgAl_{0.5}Fe_{1.5}O_4/Bi_2Se_3$ Bilayer

Stanford University
Lauren Riddiford, Yuri Suzuki

Auburn University
Peng Li

Pennsylvania State University
Timothy Pillsbury, Max Stanley, Danielle Reifsnyder Hickey, Nasim Alem Nitin Samarth

NIST
Alexander Grutter

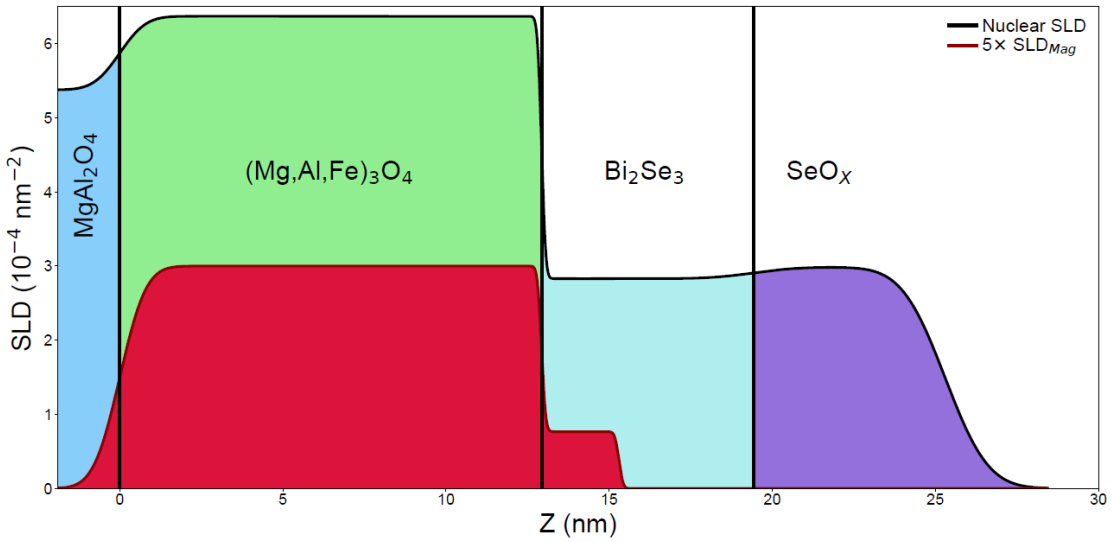
L. Riddiford et al., *Phys. Rev. Lett.* (In Press.)

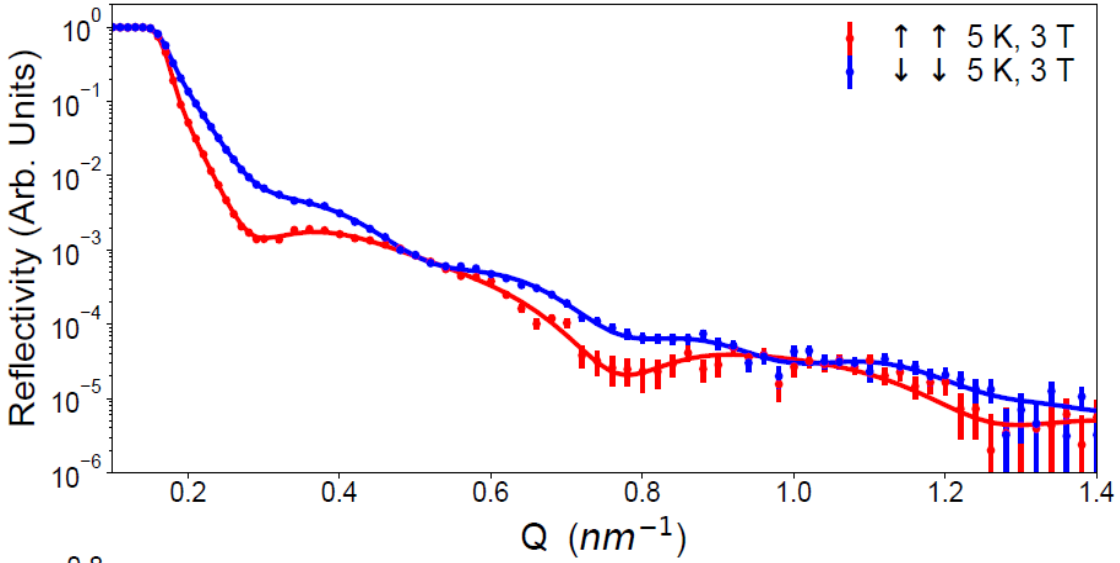
Proximity-Induced Interfacial Magnetization

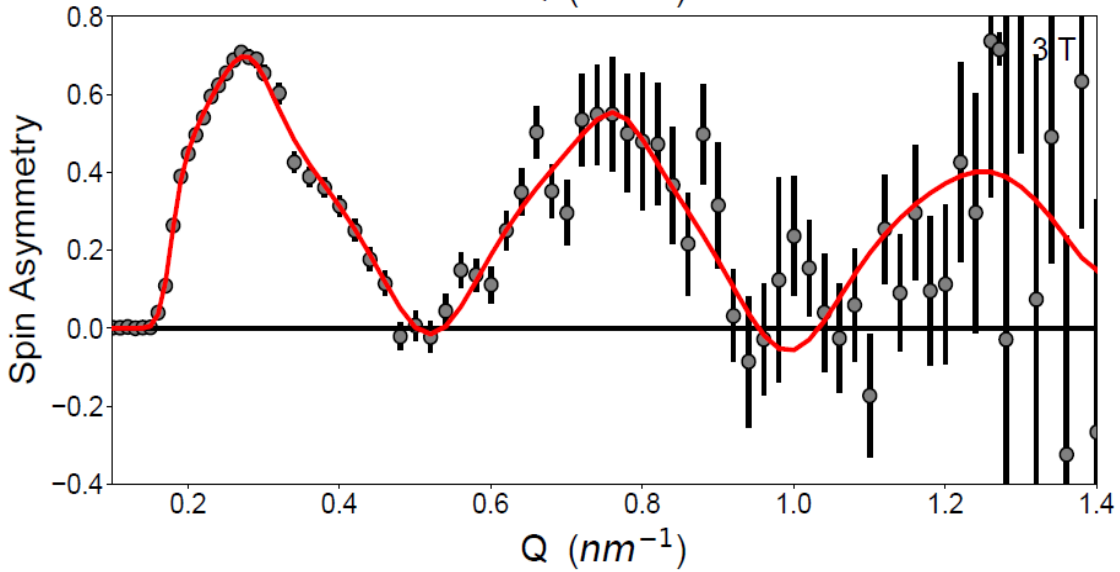
$MgAl_{0.5}Fe_{1.5}O_4$ is a new and promising magnetic insulator with ultralow damping and little to no magnetic dead layer at the surface

Ideal candidate for introducing a MPE in adjacent TI, and for integration into low-power spintronics

First look at this PNR suggests there is a strong MPE







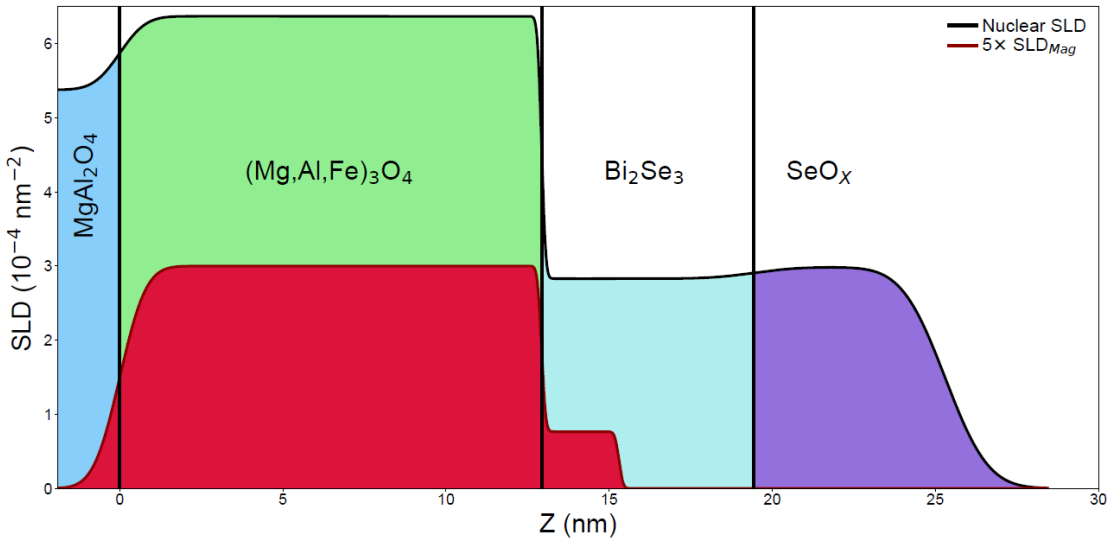
L. Riddiford et al., *Phys. Rev. Lett.* (In Press.)

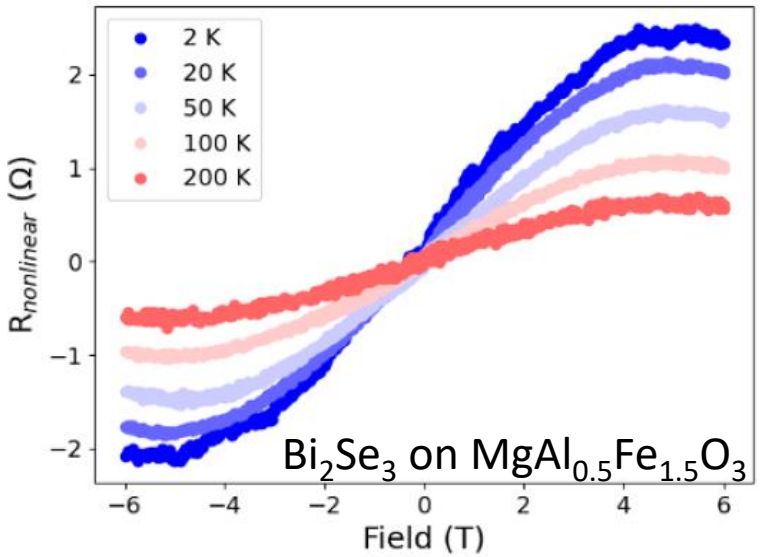
Proximity-Induced Interfacial Magnetization

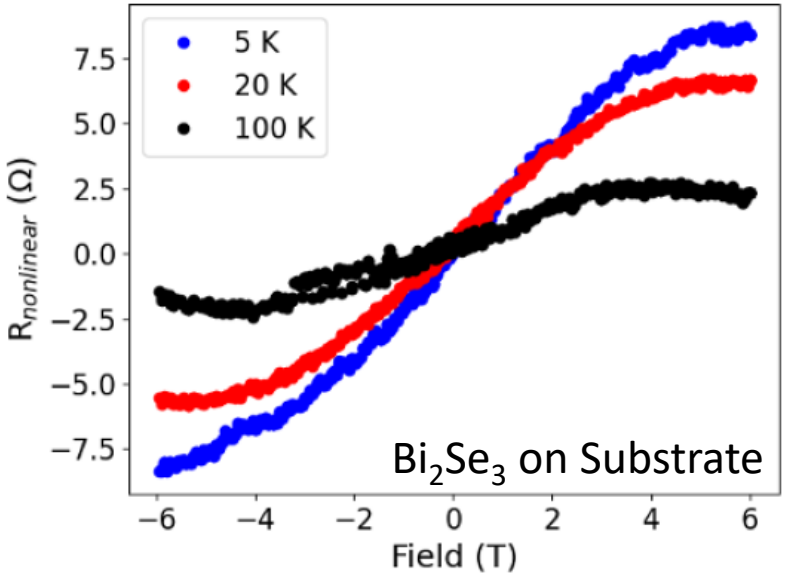
$MgAl_{0.5}Fe_{1.5}O_4$ is a new and promising magnetic insulator with ultralow damping and little to no magnetic dead layer at the surface

Ideal candidate for introducing a MPE in adjacent TI, and for integration into low-power spintronics

First look at this PNR suggests there is a strong MPE







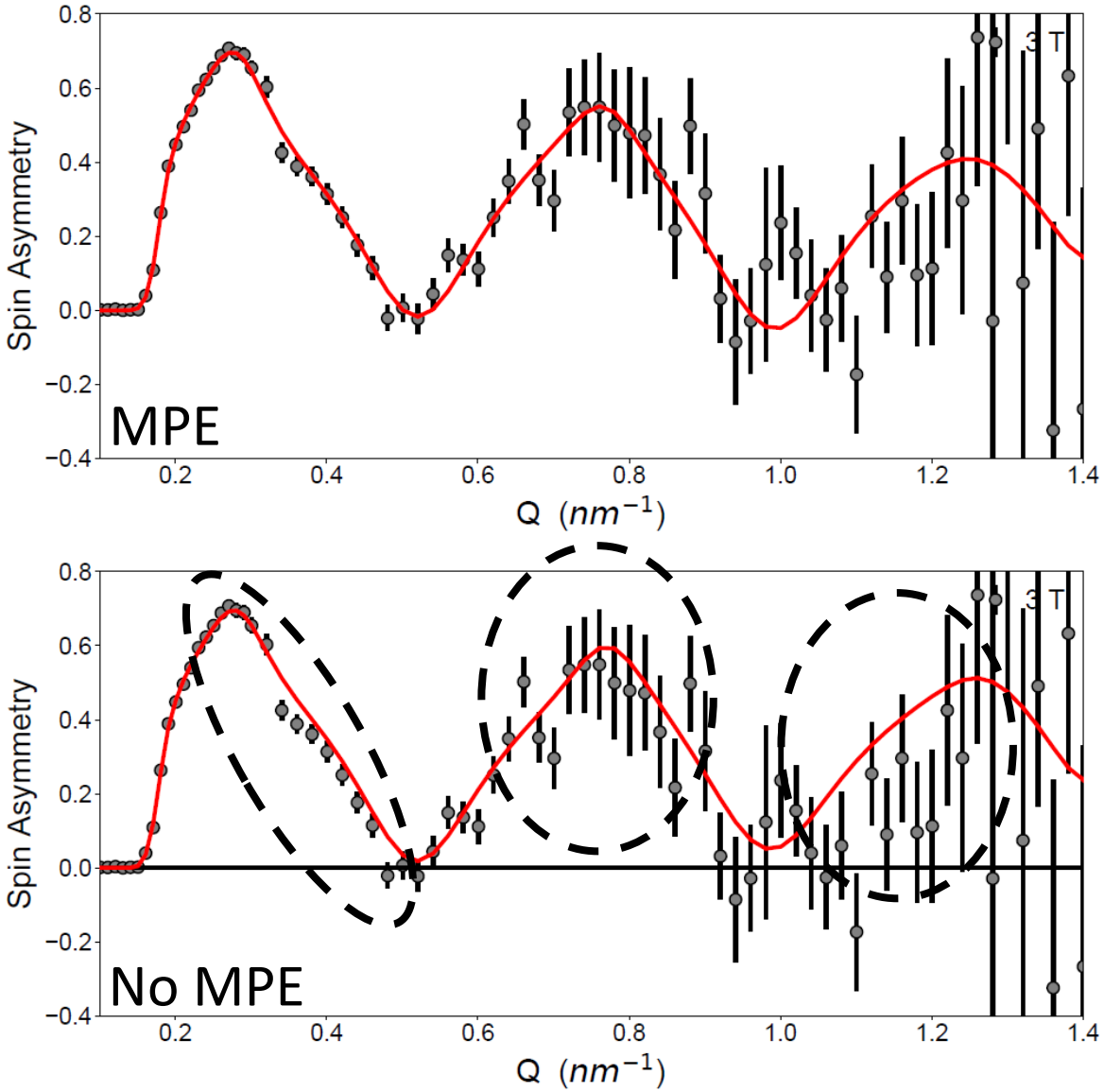
L. Riddiford et al., *Phys. Rev. Lett.* (In Press.)

How Much Does the Proximity Effect Matter?

- Compare simulated curves from two different models
- Identical except that one (top) allows MPE in Bi_2Se_3 while the other (bottom) does not
- Model without a MPE misses in several key places

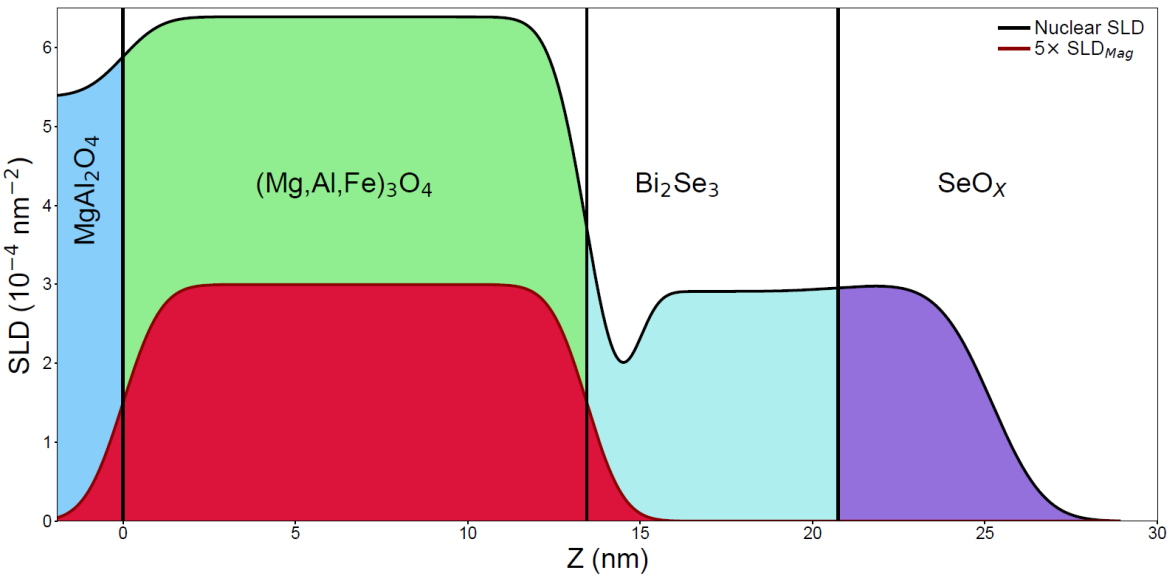
<u>So what do we learn here?</u>

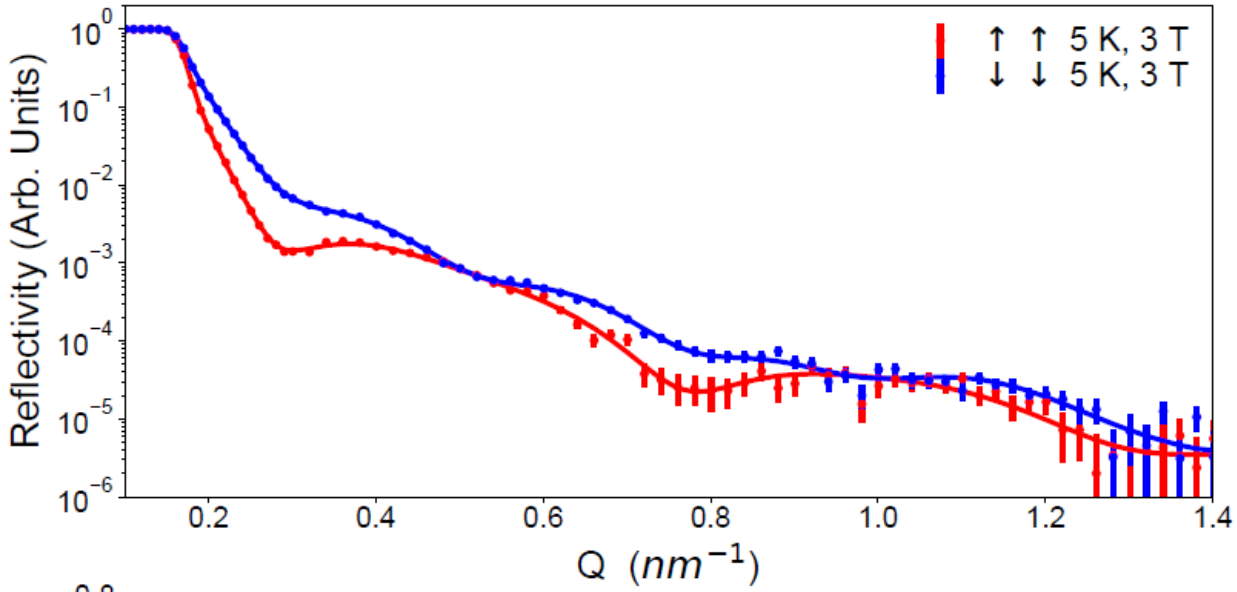
- Expected MPE effects are ***SUBTLE***
- Fits dominated by the ferrimagnetic layer
- Seems, to indicate a magnetic proximity effect

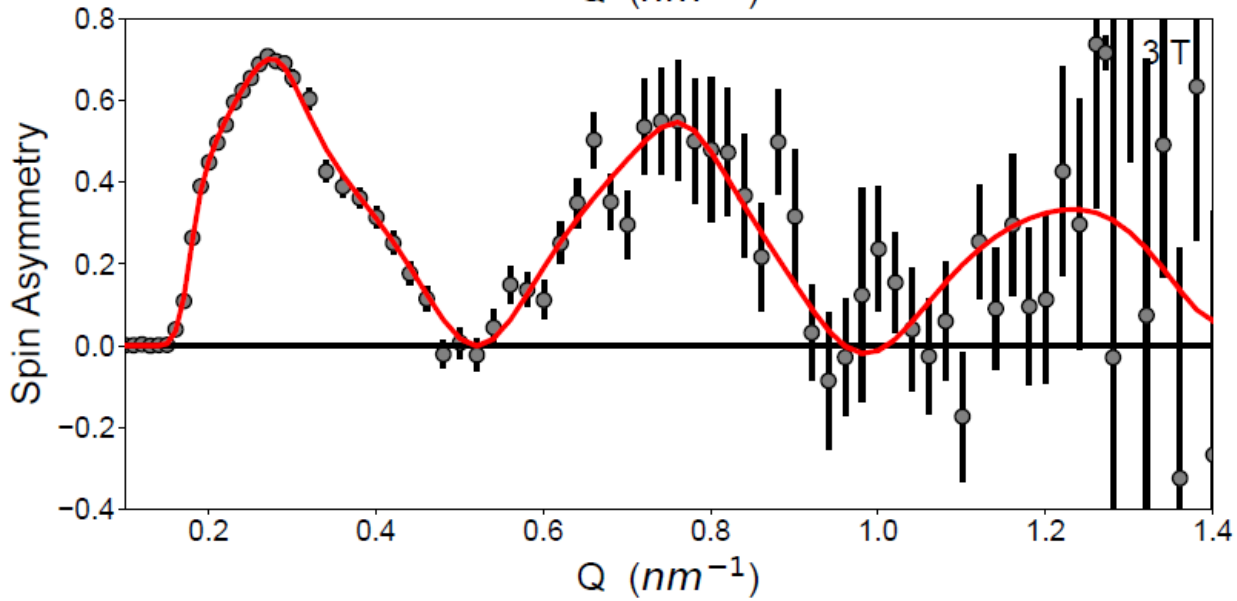


L. Riddiford et al., *Phys. Rev. Lett.* (In Press.)

Alternative Models







So clearly transitional growth regions or interface structures *can* spoof a proximity effect…

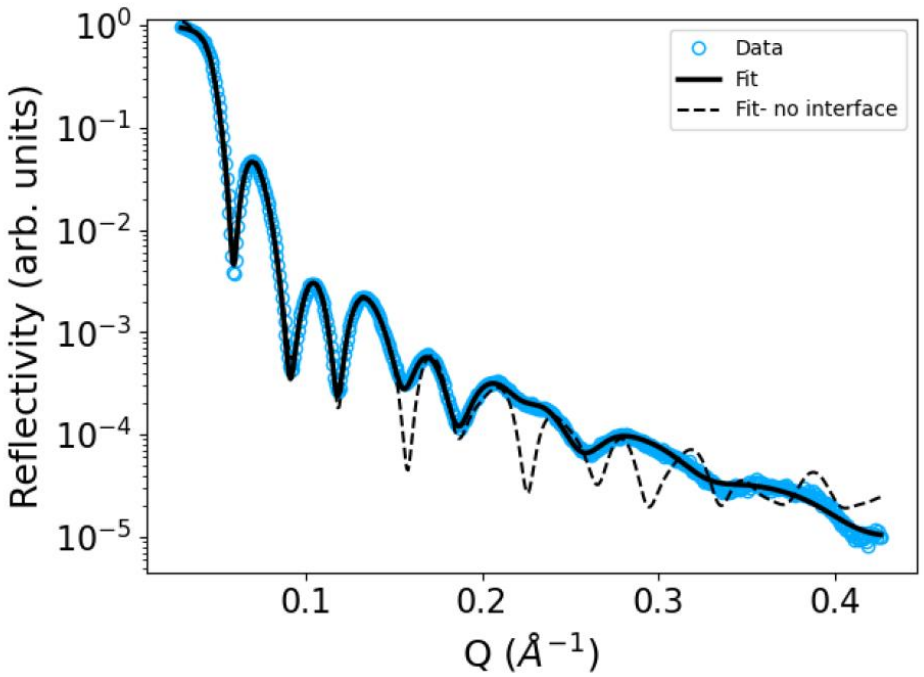
So what do we do about this?

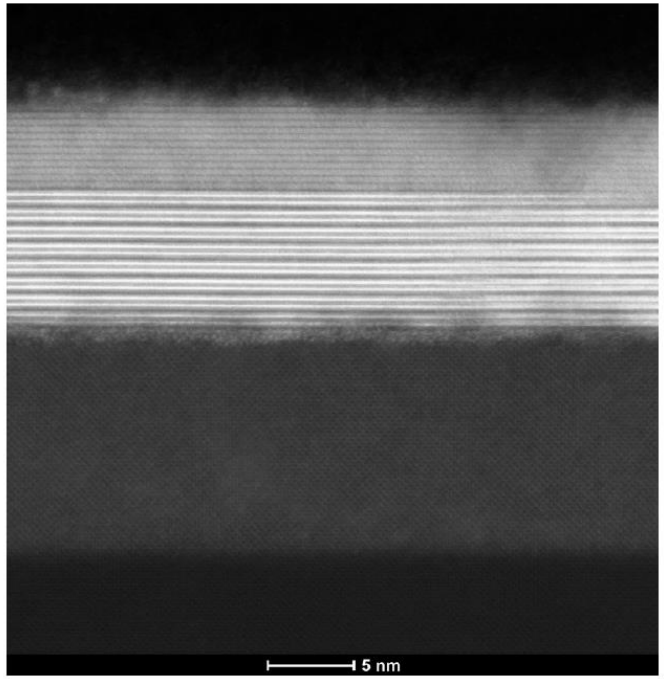
We need to bring in other techniques that can separate the two competing models

L. Riddiford et al., *Phys. Rev. Lett.* (In Press.)

Alternative Models

- XRR and STEM confirm a transitional growth region at the interface
- No proximity effect





<u>What do we do about this?</u>

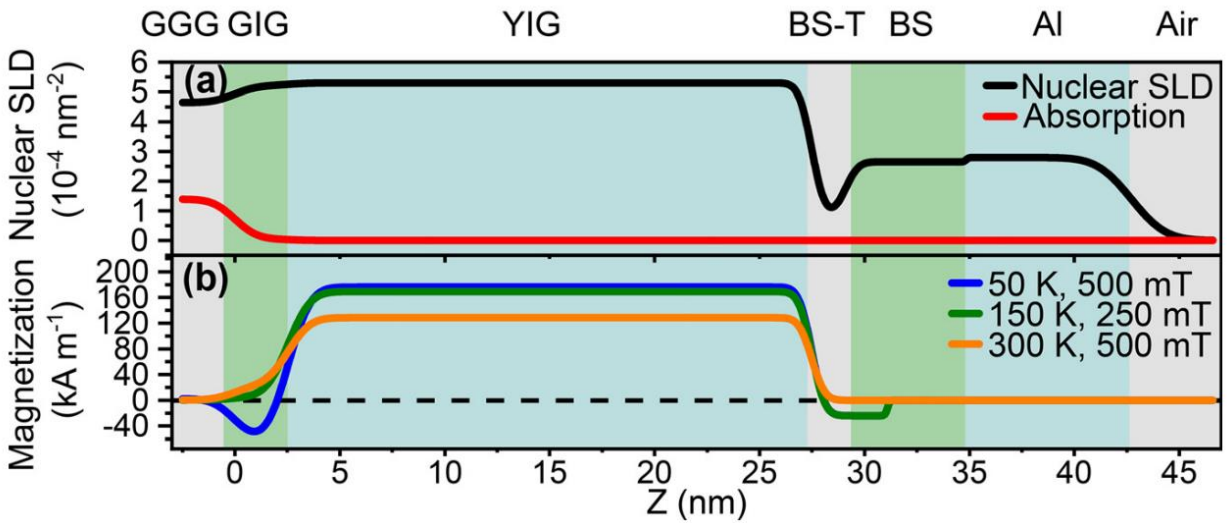
(A) Complementary measurements allowing model selection
(B) Find systems with high sensitivity to the magnetic interface
(C) Take higher-precision PNR measurements over a wider Q-range

L. Riddiford et al., *Phys. Rev. Lett.* (In Press.)

How General is This?

$(Bi,Sb)_2Te_3$ on $Y_3Fe_5O_{12}$

Yang Lv,[1] James Kally,[2] Tao Liu,[3] Patrick Quarterman,[4] Timothy Pillsbury,[2] Brian J. Kirby,[4] Alexander J. Grutter,[4] Protyush Sahu,[5] Julie A. Borchers,[4] Mingzhong Wu,[3] Nitin Samarth,[2] and Jian-Ping Wang[1,5,a)]

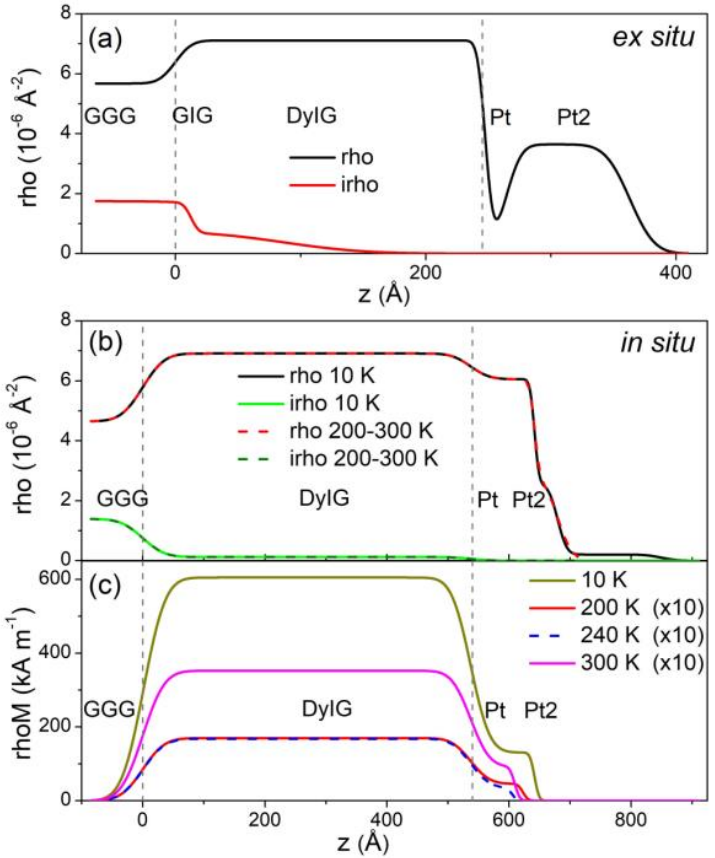


This interface reconstruction appears to pop up in many different systems.

Y. Lv, Applied Physics Reviews **9**, 011406 (2022)

Pt on $Dy_3Fe_5O_{12}$ with and without vacuum break between layers

Jackson J. Bauer,[1] Patrick Quarterman,[2] Alexander J. Grutter,[2] Bharat Khurana,[1] Subhajit Kundu,[3] K. Andre Mkhoyan,[3] Julie A. Borchers,[2] and Caroline A. Ross[1,*]



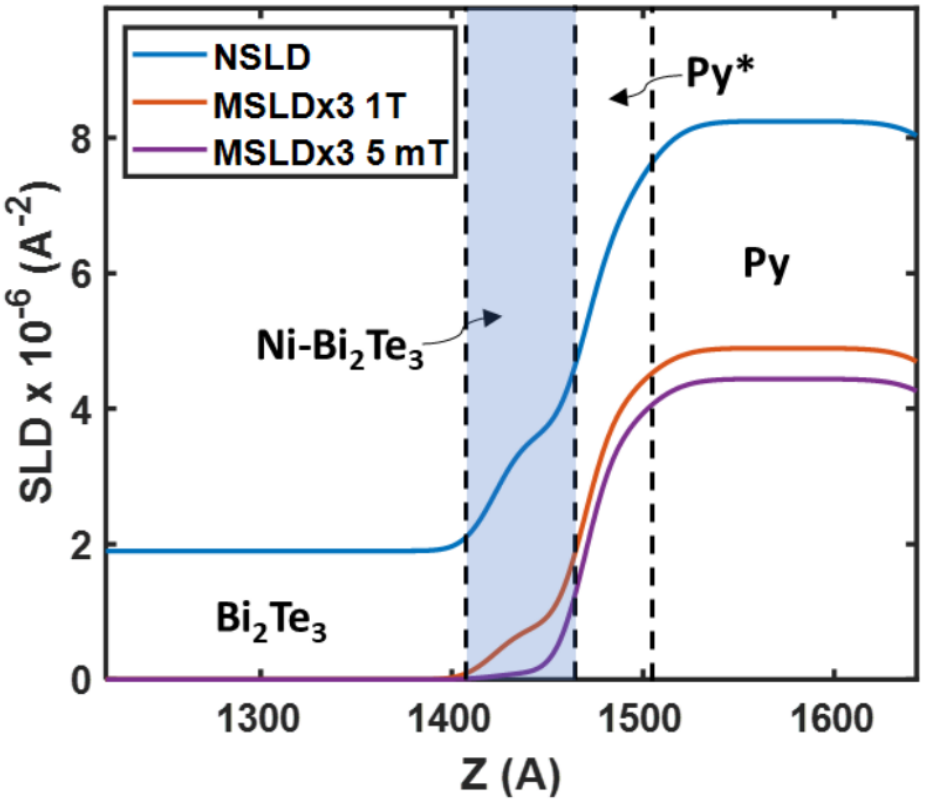
J. J. Bauer, *Phys. Rev. B* **104**, 094403 (2021)

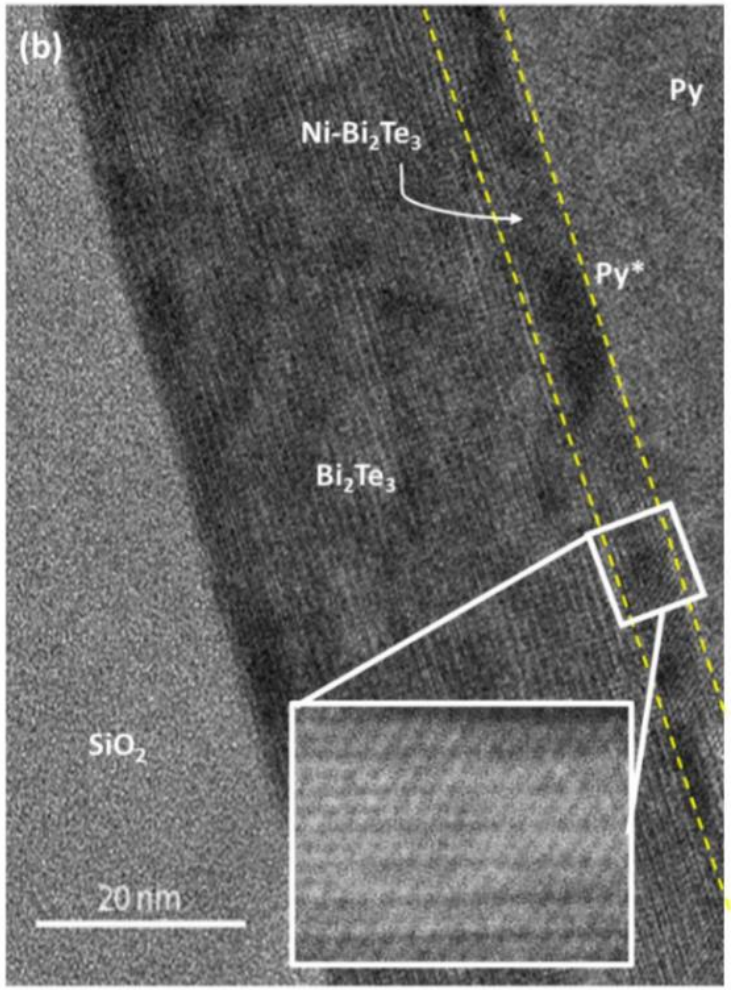
Sputtered Bi_2Te_3/Permalloy Interfaces

- Even *in-situ* sputtered heterostructures of TI/Metal systems can be complicated
- Intermixing can lead to new phases, blamed in some EuS systems
- Emergent antiferromagnetic order and exchange bias
- Topological Antiferromagnet $NiBi_2Te_4$?

Nirjhar Bhattacharjee[1], Krishnamurthy Mahalingam[3], Adrian Fedorko[2], Valeria Lauter[4], Matthew Matzelle[2], Bahadur Singh[6], Alexander Grutter[5], Alexandria Will-Cole[1], Michael Page[3], Michael McConney[3], Robert Markiewicz[2], Arun Bansil[2], Donald Heiman[2], Nian Xiang Sun[1*]

Advanced Materials, Just Accepted:
https://doi.org/10.1002/adma.202108790





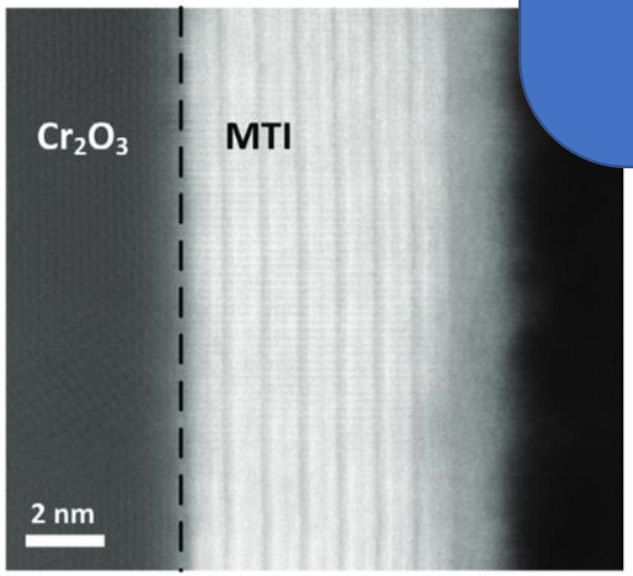
Clean Oxide/TI Interfaces are Possible

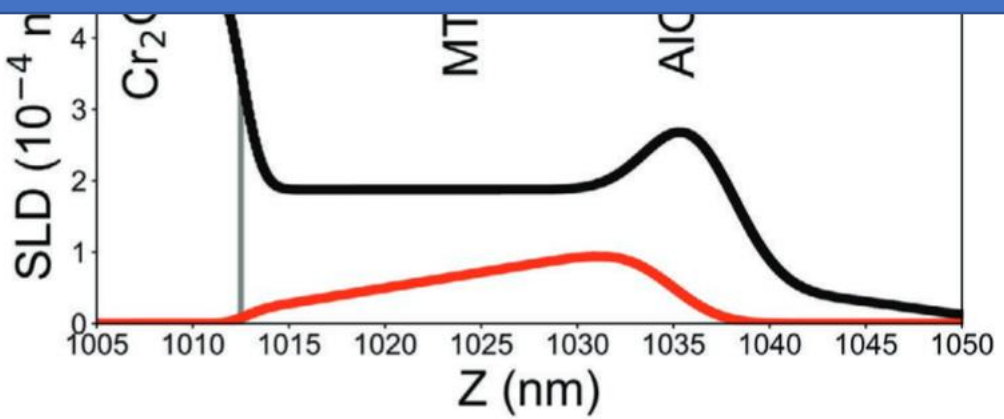
*Lei Pan, Alexander Grutter, Peng Zhang, Xiaoyu Che, Tomohiro Nozaki, Alex Stern, Mike Street, Bing Zhang, Brian Casas, Qing Lin He, Eun Sang Choi, Steven M. Disseler, Dustin A. Gilbert, Gen Yin, Qi Xufeng Kou, Sahashi Masas and Kang L. Wang**

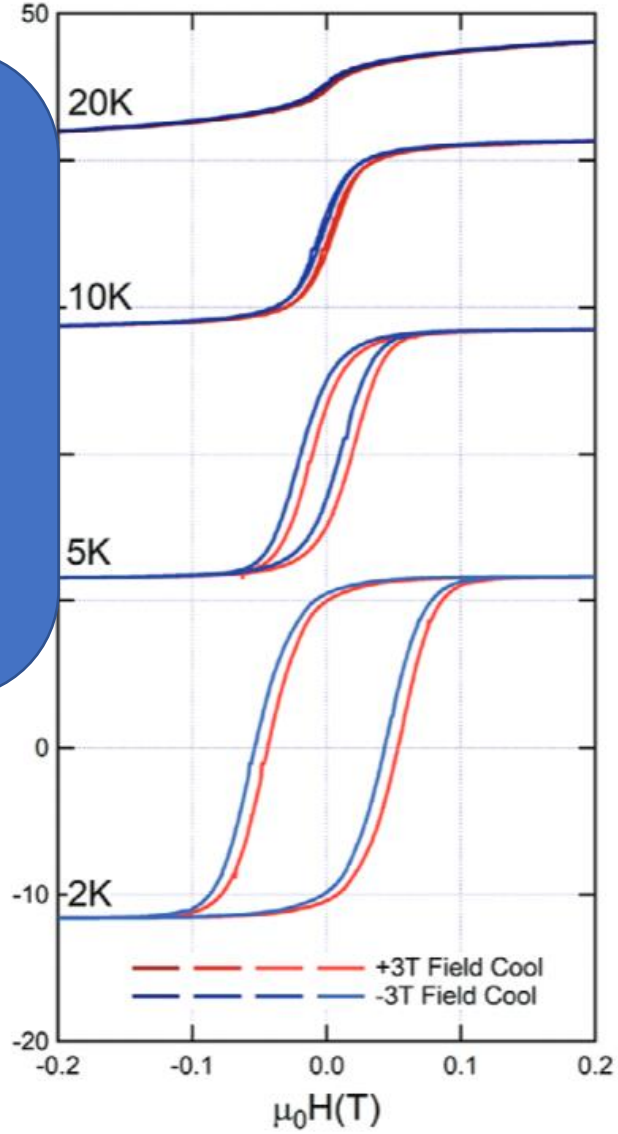
L. Pan et al., Advanced Mat
See also: F. Wang et al., Nar

Is the heat treatn
the Cr_2O_3 surface

- Interfaces are complicated
- Being sure about things is very difficult
- How do we maximize our chances of a definitive answer?







Cr-$(Bi,Sb)_2Te_3$/SnTe: Topological Insulator and Topological Crystalline Insulator

A simple case of "No detectable MPE"

UCLA
Peng Deng, Peng Zhang, Shuaihang Pan, Kang K. Wang

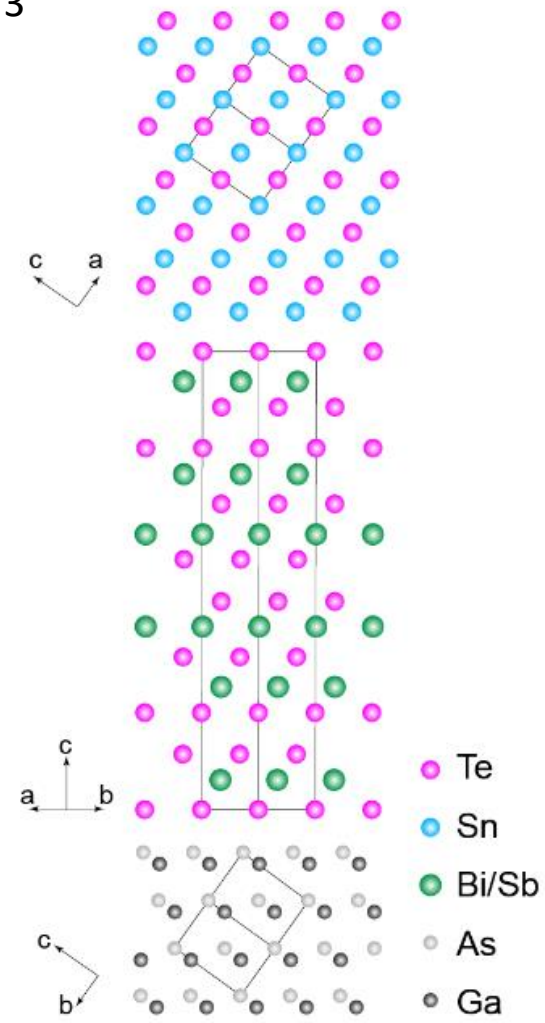
NIST
Megan Holtz, Patrick Quarterman

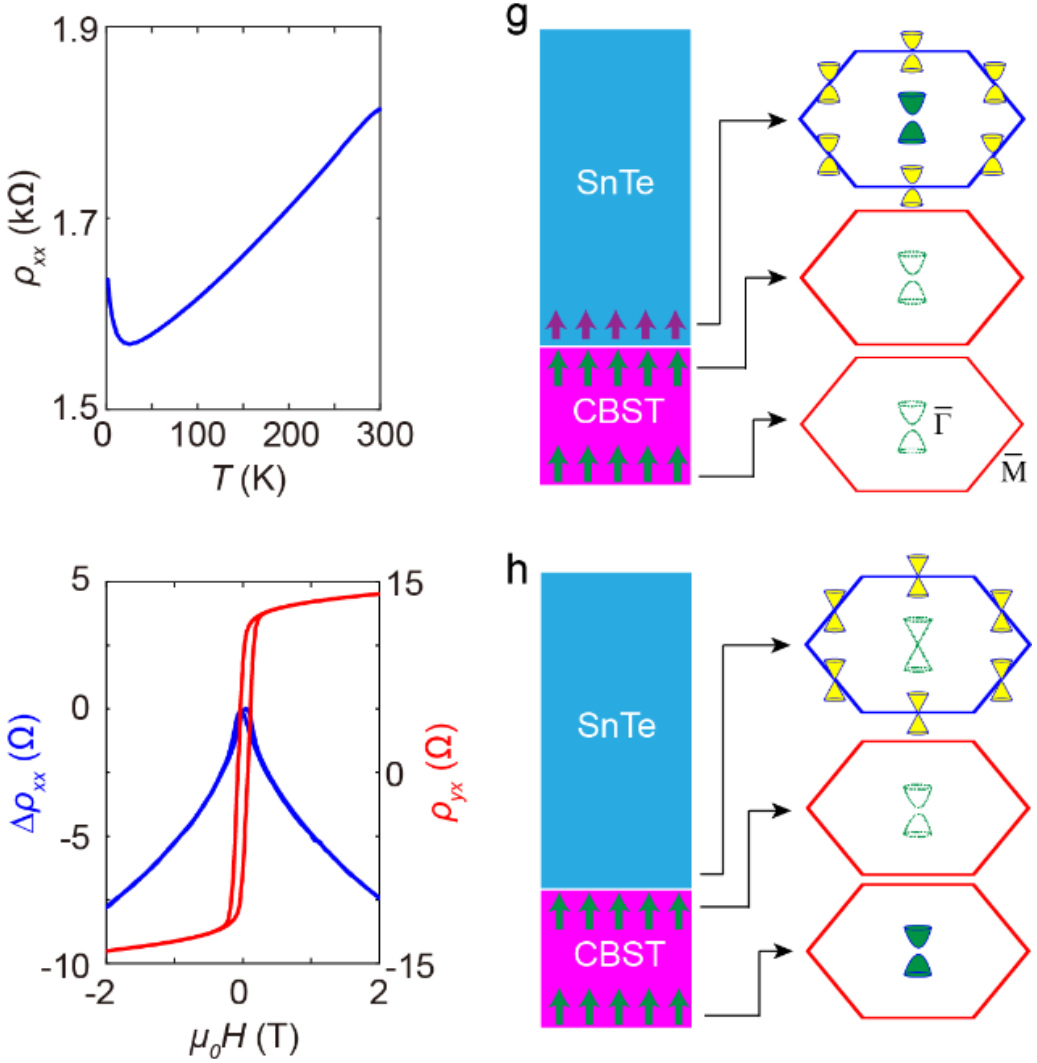
Topological Insulator/Topological Crystalline Insulator Interface

SnTe and Cr-doped $(Bi,Sb)_2Te_3$

- Epitaxial
- Excellent Crystallinity
- Extremely Sharp Interfaces

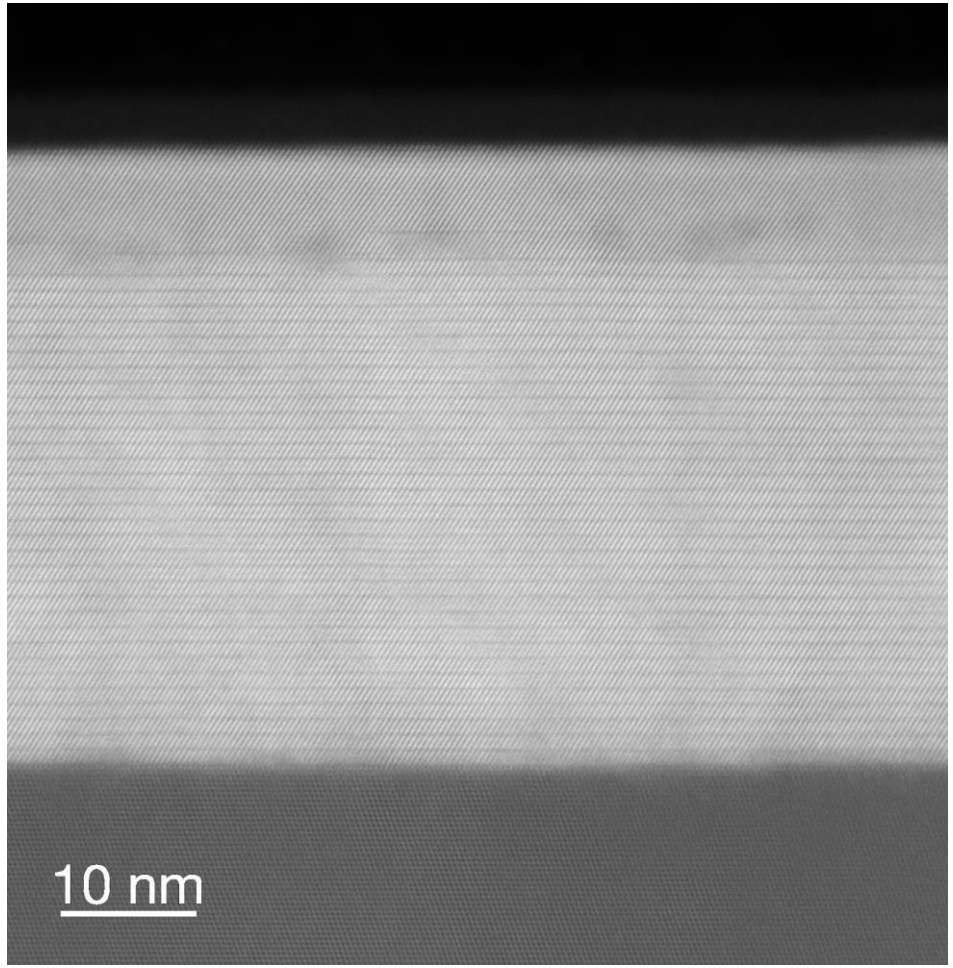
- 2 nm Cr:BST / SnTe yields an anomalous Hall effect
- 2 nm Cr:BST should be insulating
- Cr:BST induces proximity magnetism in SnTe?

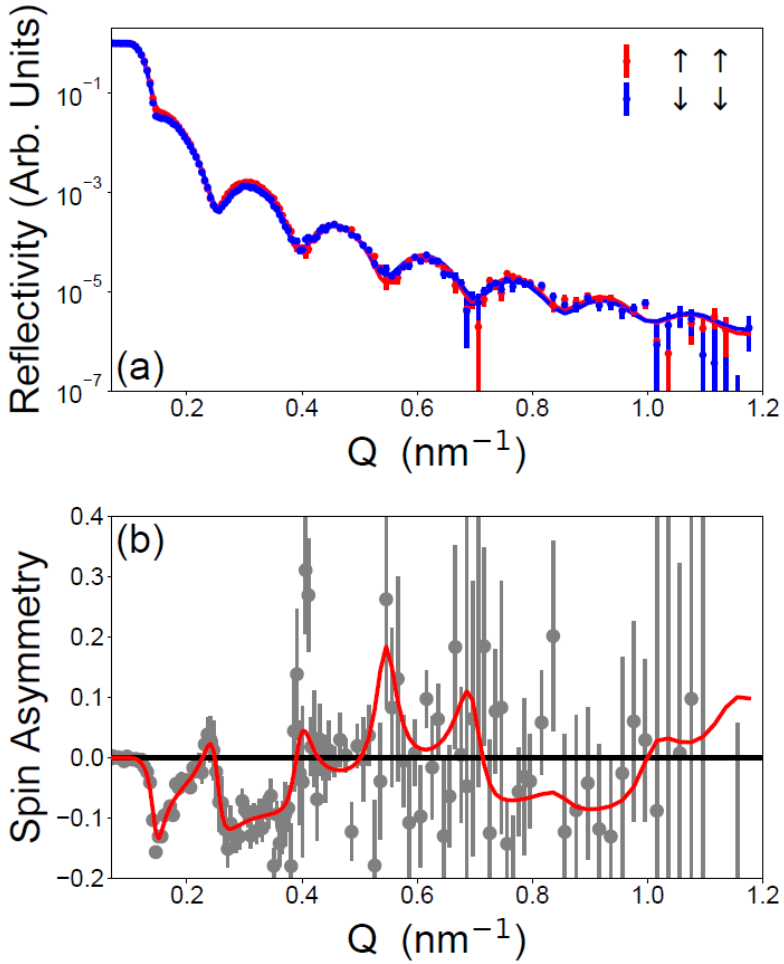


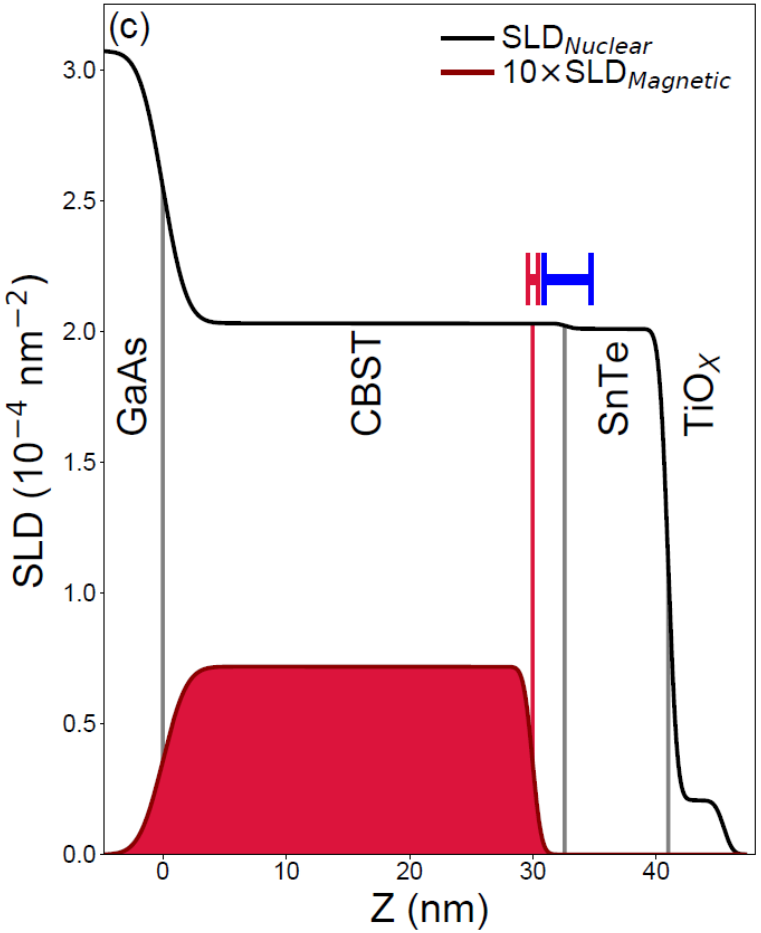


P. Deng et al., *Under Review* (2021)

Modeling *without* a Proximity Effect



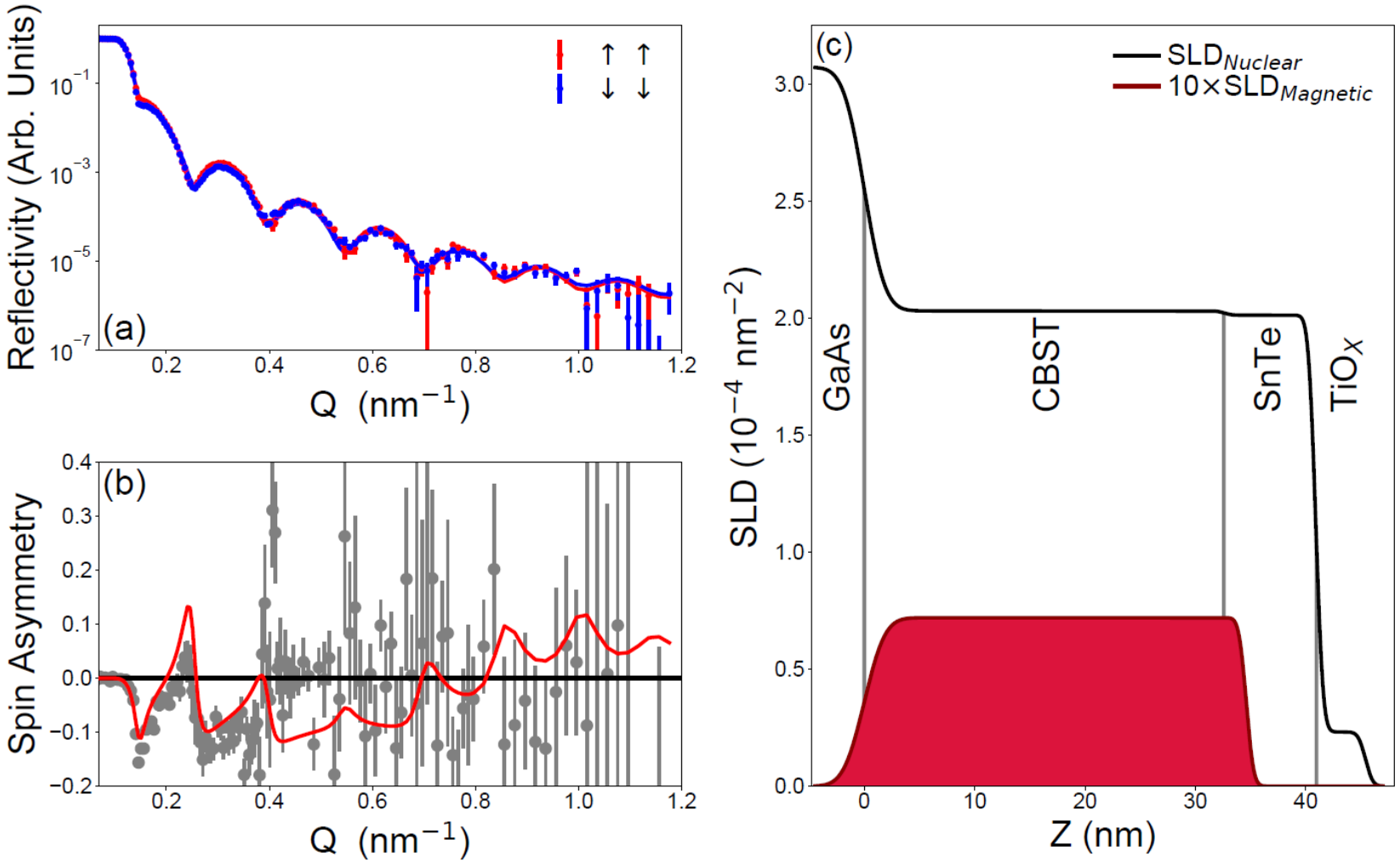




- Combining STEM imagine with PNR allows us to compare the structural thickness with the magnetic thickness, overcoming the weak nuclear contrast between the CBST and SnTe layers
- Magnetic thickness < CBST Thickness

P. Deng et al., *Under Review* (2021)

Modeling *with* a Proximity Effect



Forcing a magnetic proximity effect in the SnTe clearly fails to fit the data. No proximity effect (that we can detect)

P. Deng et al., *Under Review* (2021)

$CrSe/(Bi,Sb)_2Te_3$: A Proximity Effect Dependent on Interface Symmetry

The clearest MPE I've personally seen

UCLA

Chao-Yao Yang, Lei Pan, Xiaoyu Che, Qing Lin He, Yingying Wu, Hao Wu, Gen Yin, Peng Deng, Kang K. Wang

Henan Normal University

Haiyang Wang

NIST

Dustin Gilbert (*Now at UT Knoxville*), Julie Borchers, William Ratcliff II

Advanced Light Source

Padraic Shafer, Elke Arenholz

PNR: CrSe and TI

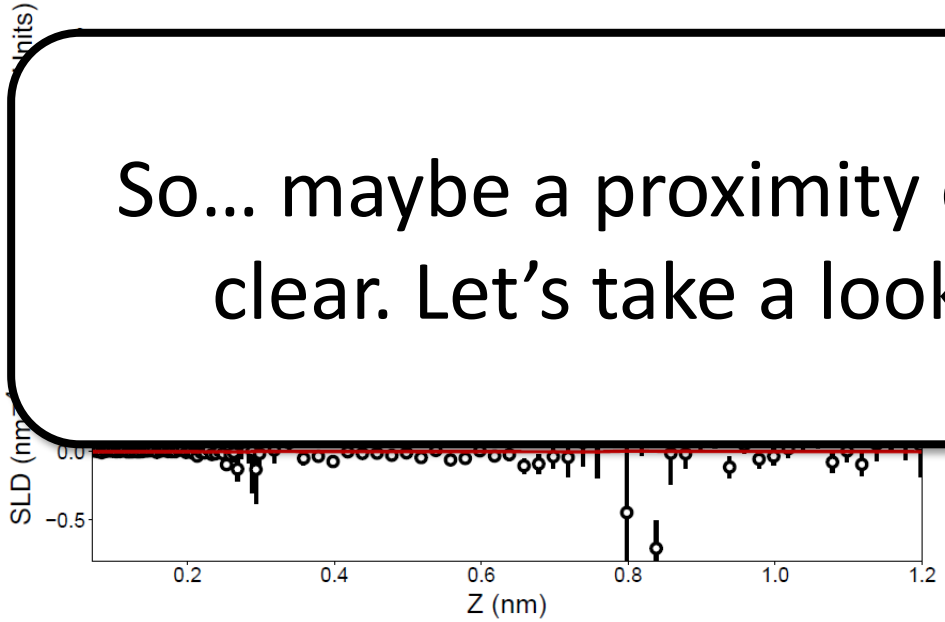
CrSe Film

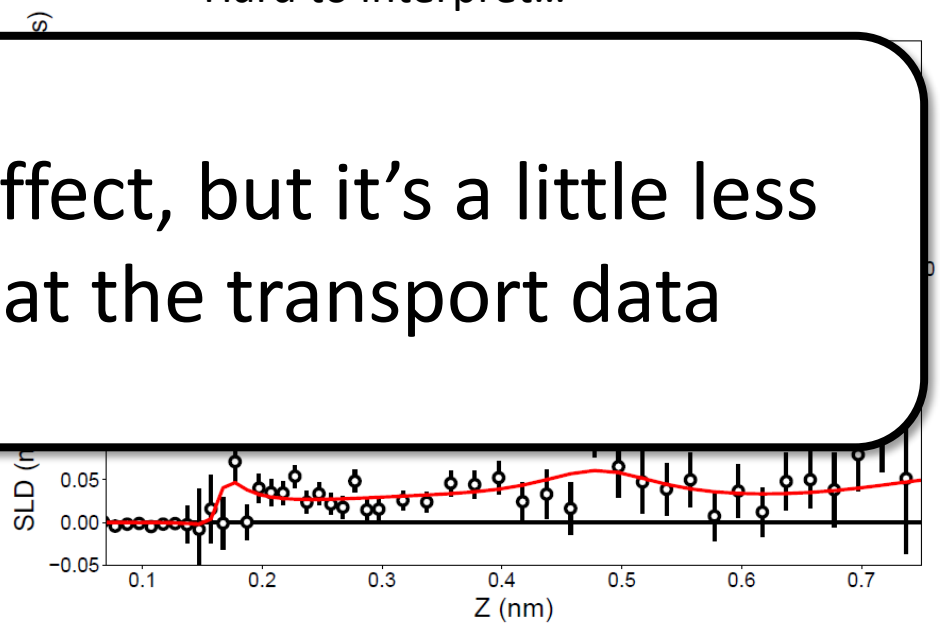
No magnetization, as expected

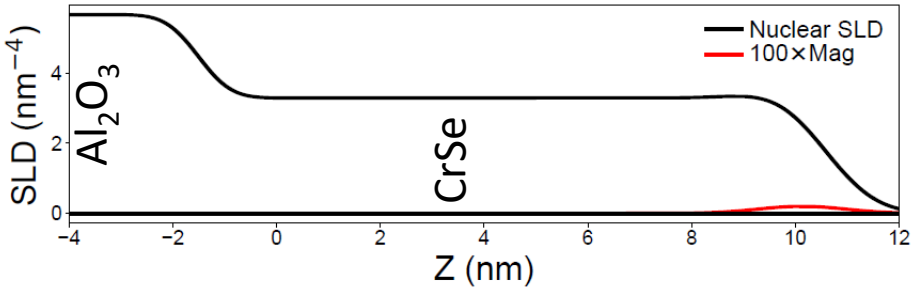
CrSe/TI Bilayer

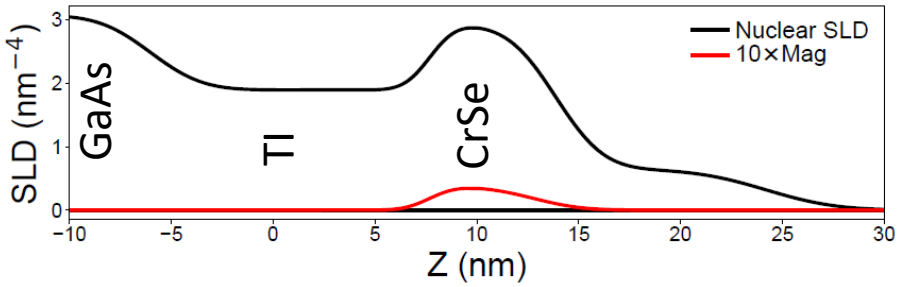
Magnetic, but rough interface.
Hard to interpret…

So… maybe a proximity effect, but it's a little less clear. Let's take a look at the transport data

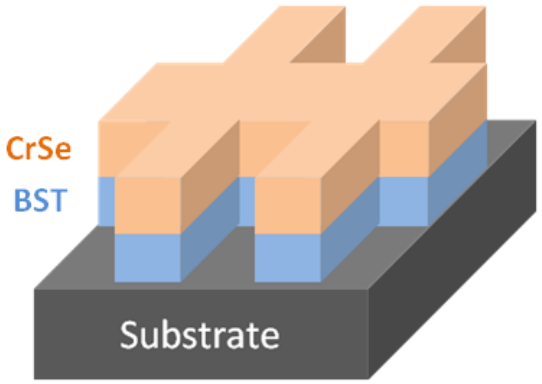


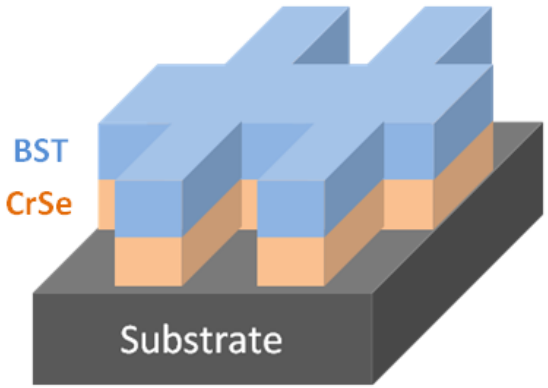




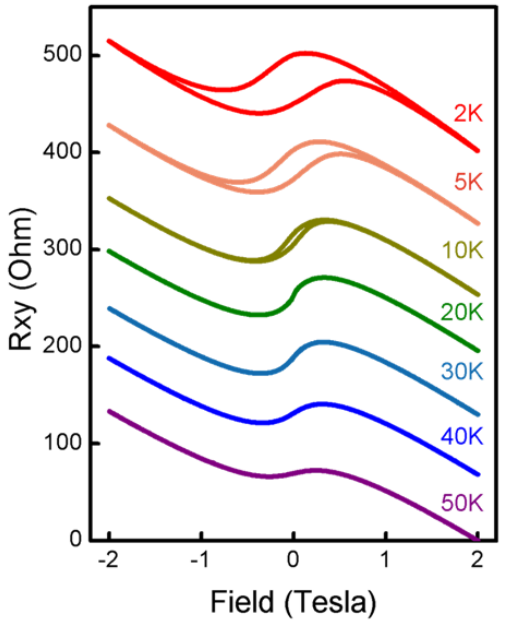


Transport: CrSe/BST vs. BST/CrSe

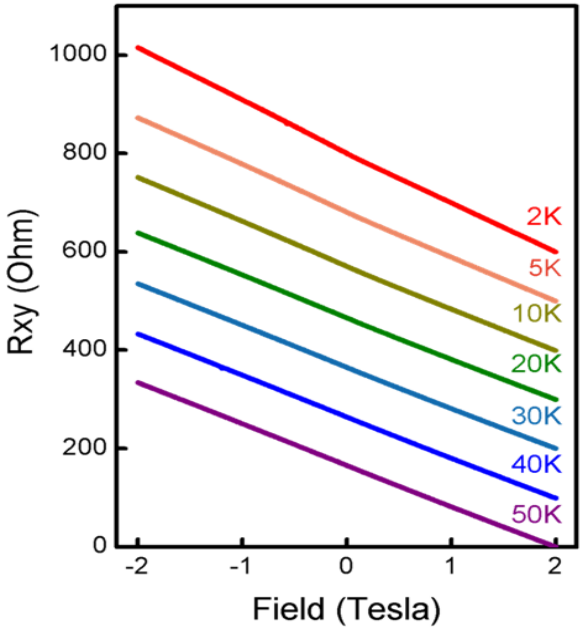




Hall Resistance



Hall Resistance



Order Matters

BST/CrSe shows an AHE indicating net magnetization

CrSe/BST has no net magnetization

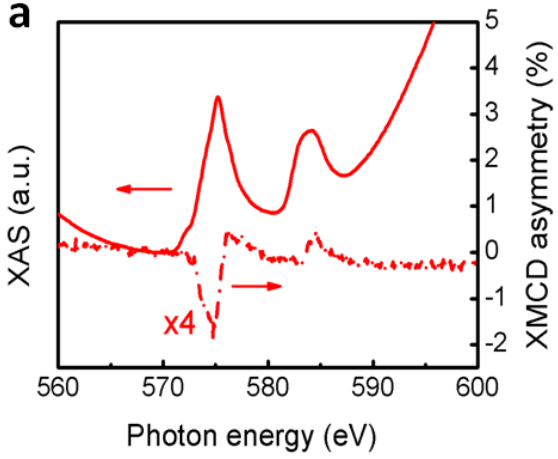
Ordering temperature too high for intermixing

C.-Y. Yang et al., *Science Advances* **6**, eaaz8463 (2020)

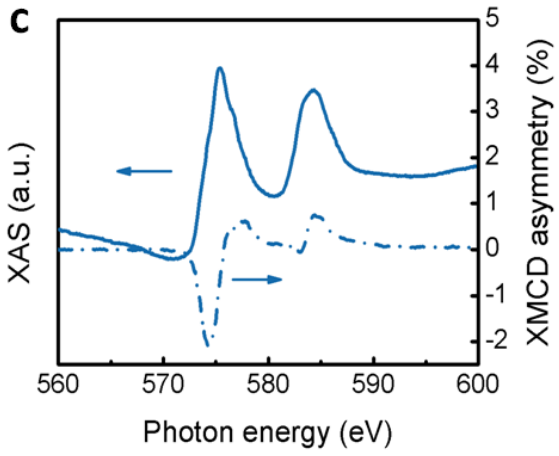
XMCD: Element Specific Magnetism

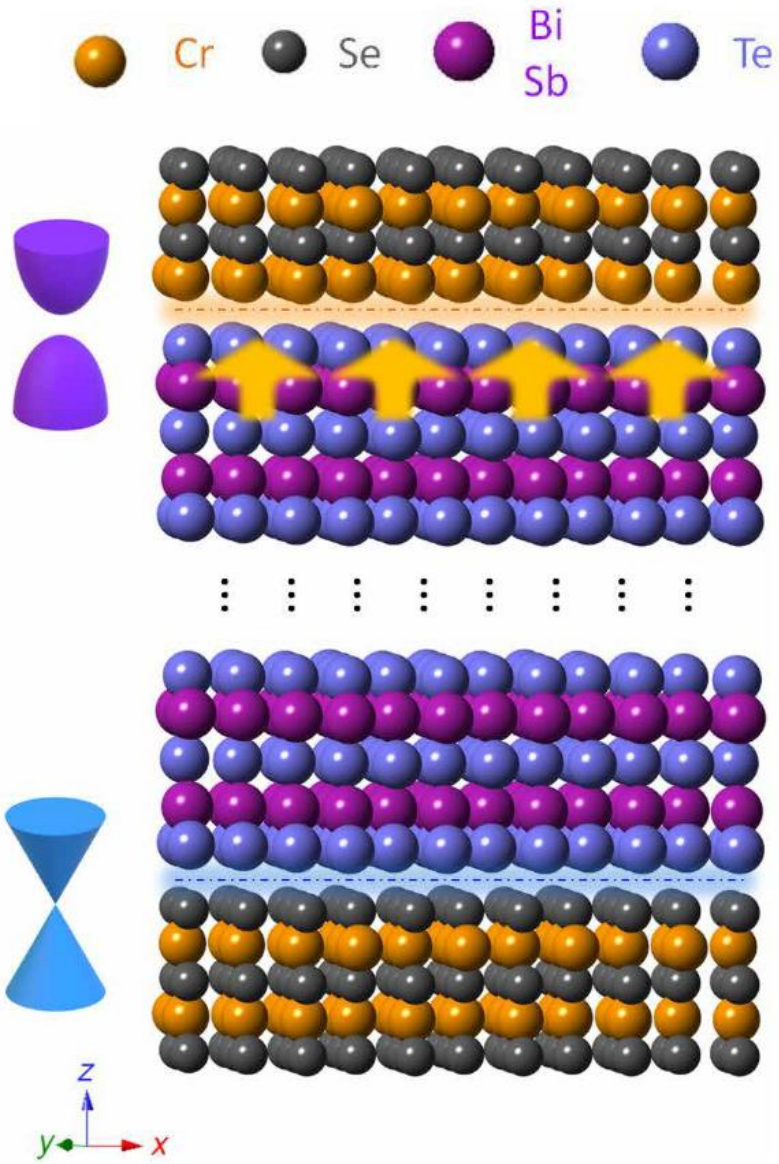
Cr-edge

$(Bi, Sb)_2Te_3$ on CrSe

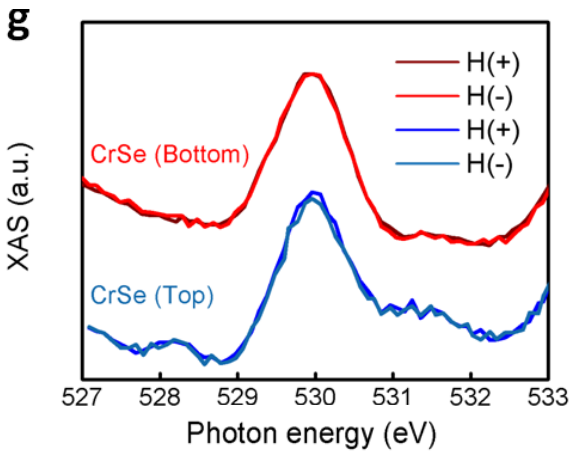


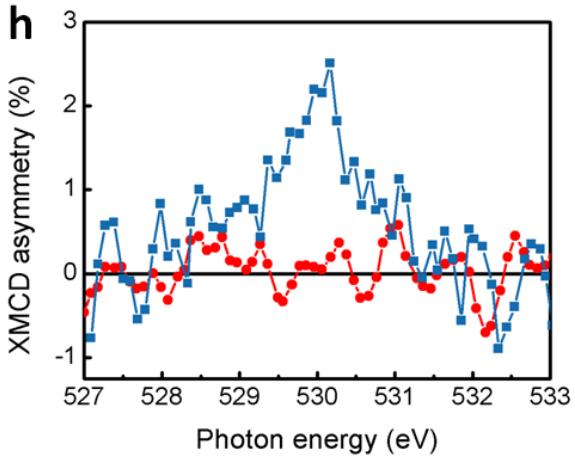
CrSe on $(Bi, Sb)_2Te_3$





Sb-edge

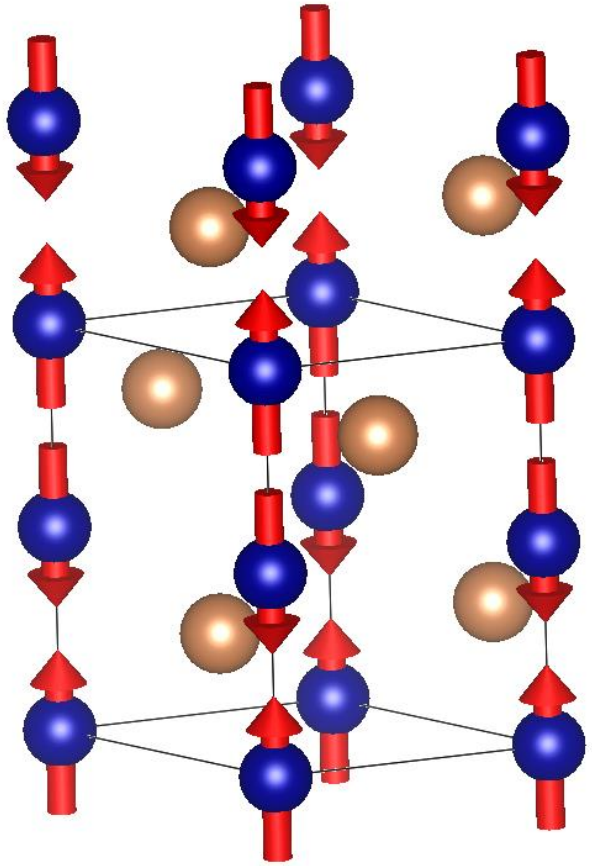




C.-Y. Yang et al., *Science Advances* **6**, eaaz8463 (2020)

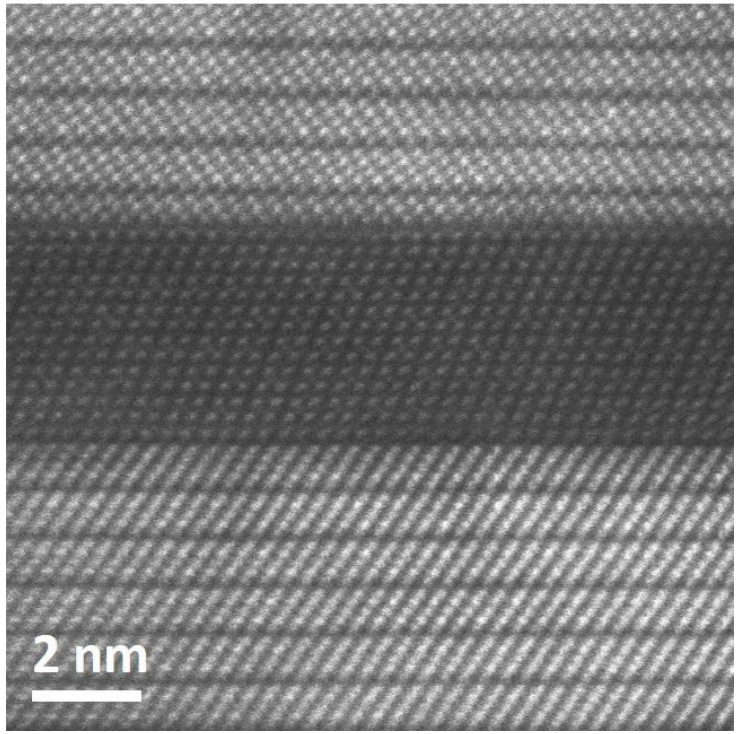
$CrSb/(Bi,Sb)_2Te_3$

Qing Lin He,[1,2,*] Gen Yin,[1] Luyan Yu,[1] Alexander J. Grutter,[3] Lei Pan,[1] Chui-Zhen Chen,[4] Xiaoyu Che,[1] Guoqiang Yu,[1] Bin Zhang,[5] Qiming Shao,[1] Alexander L. Stern,[6] Brian Casas,[6] Jing Xia,[6] Xiaodong Han,[5] Brian J. Kirby,[3] Roger K. Lake,[7] K. T. Law,[4] and Kang L. Wang[1,†]



Reduce the confounding effects of the magnetization from the magnetically ordered reservoir layer

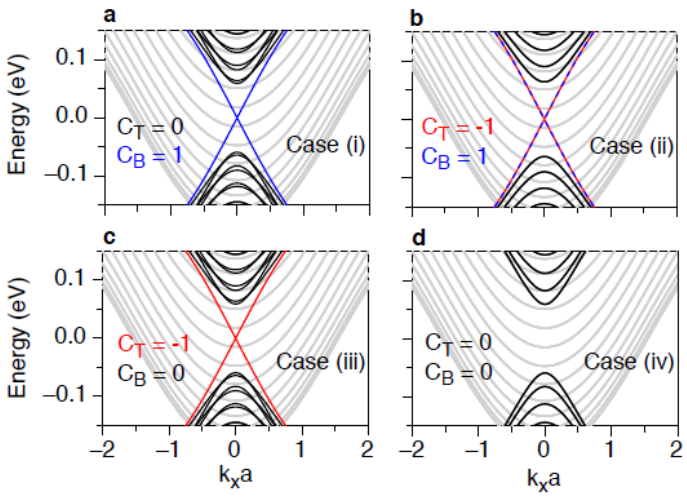
Pushing the limits of what we can detect

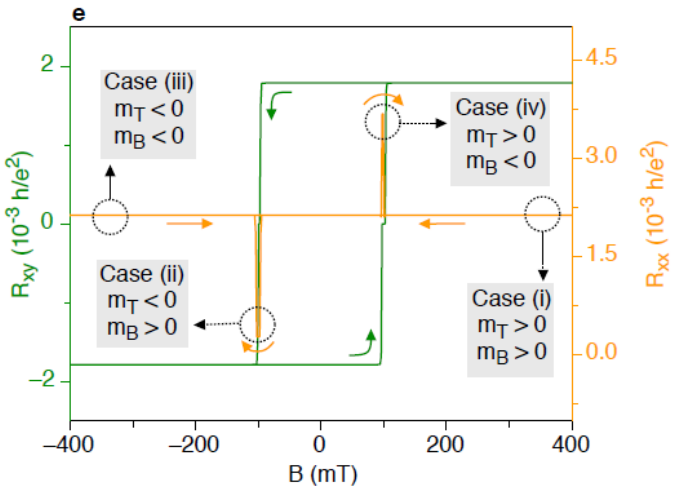


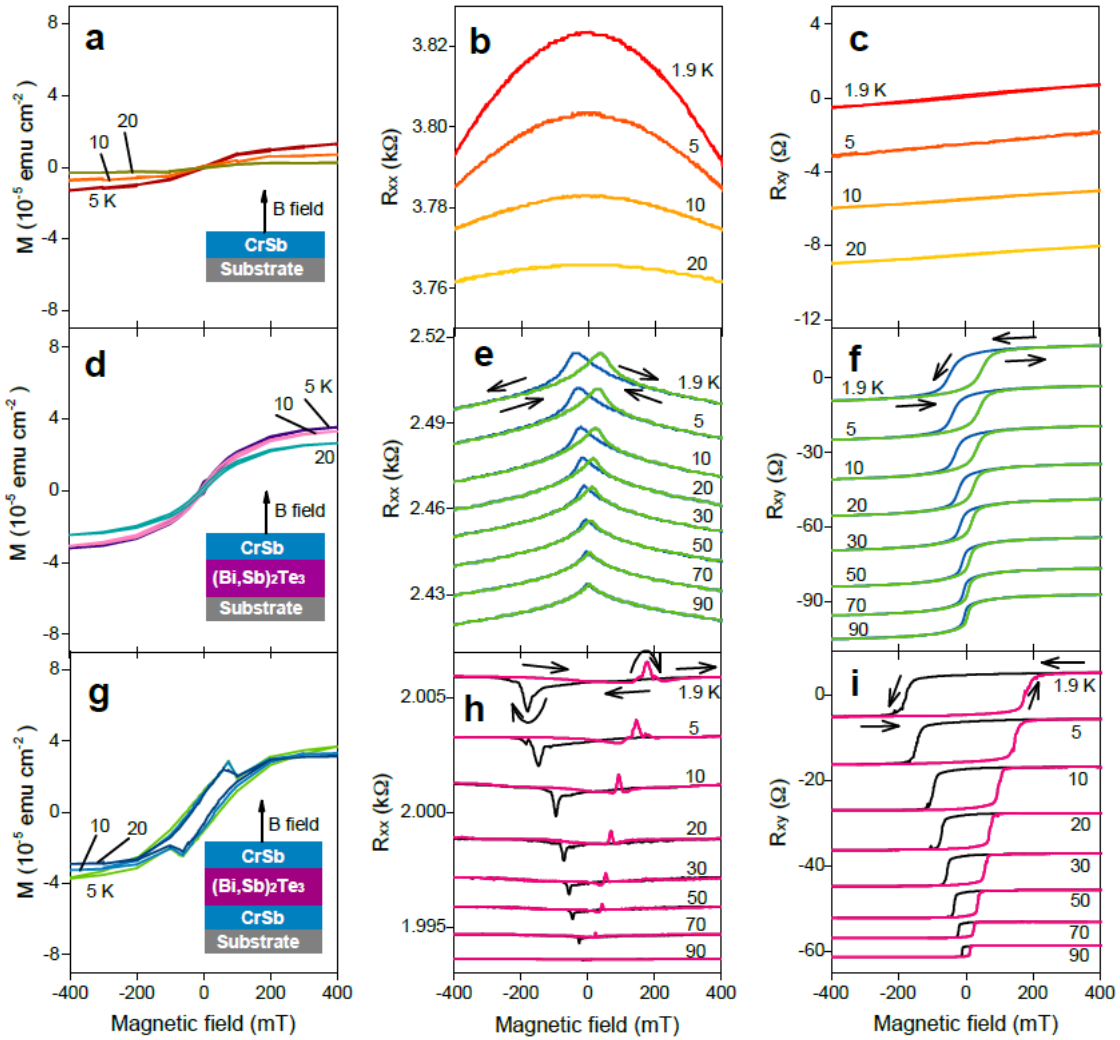
Physical Review Letters **121**, 096802 (2018)

$CrSb/(Bi,Sb)_2Te_3$: Electrical Transport

- Bilayers and trilayers show indications of switchable magnetization
- Unusual emergent lineshapes in the magnetoresistance of trilayers
- Consistent with theory showing a topological phase transition induced by magnetic proximity



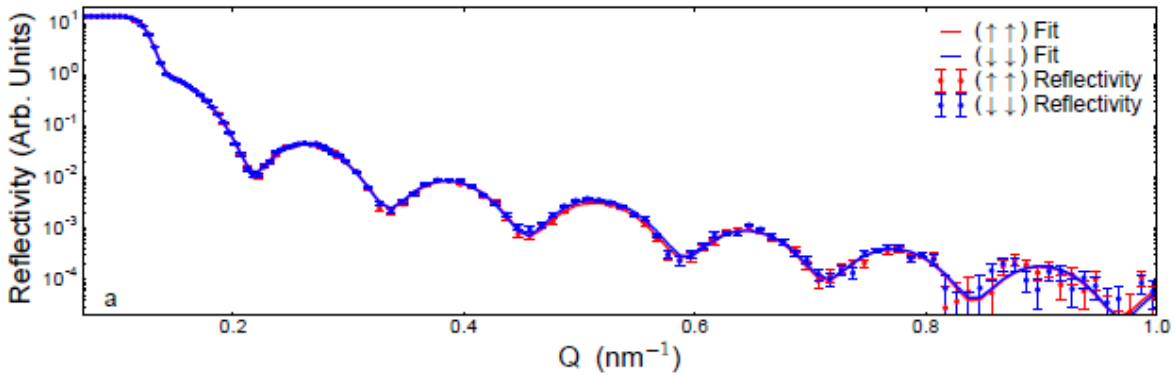


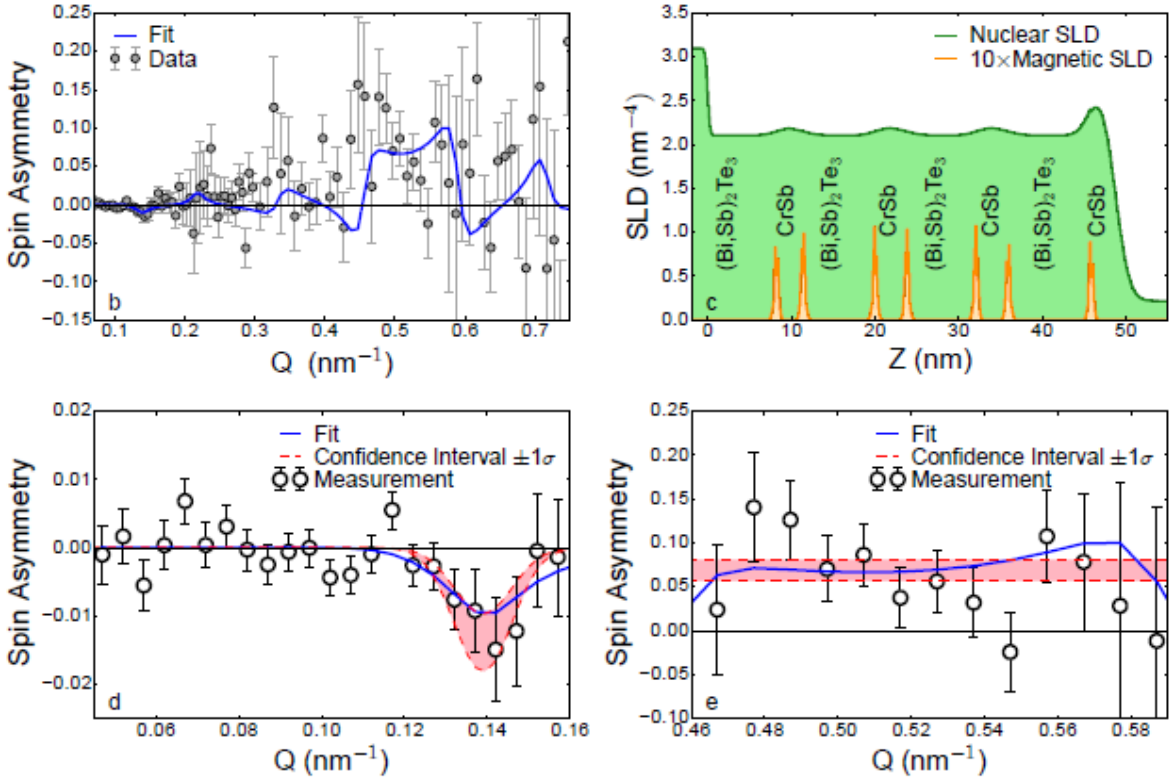


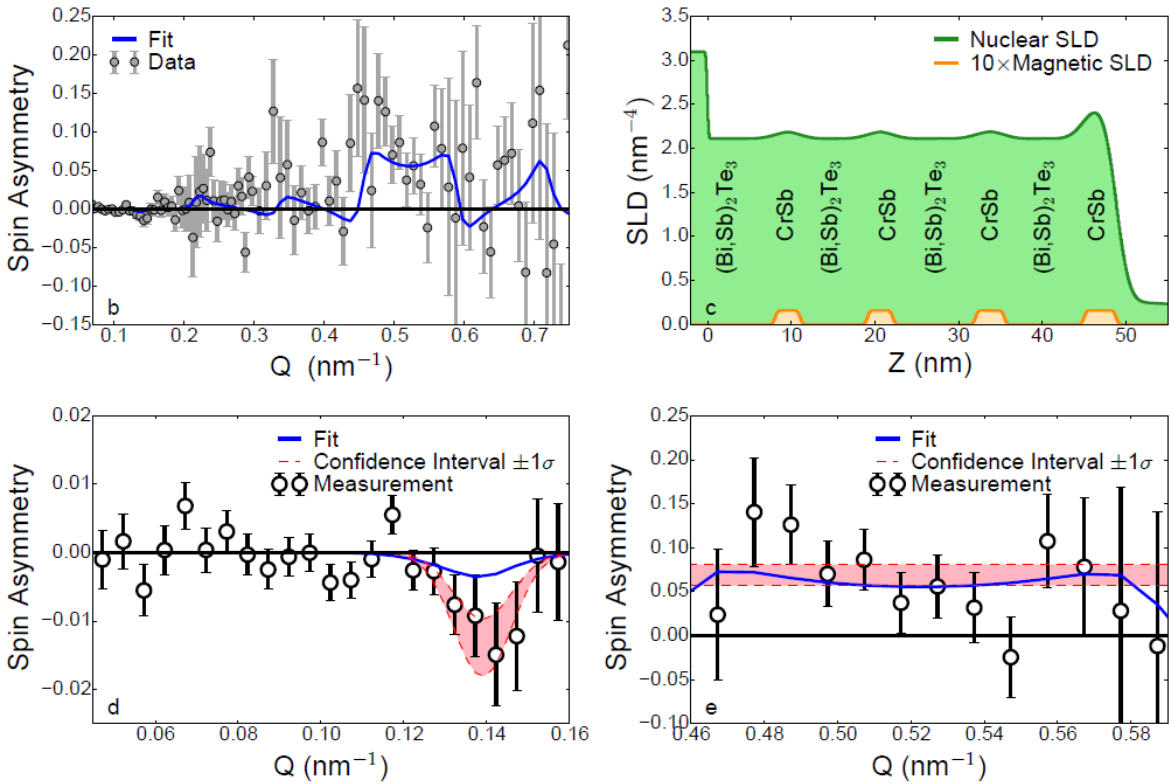
Physical Review Letters **121**, 096802 (2018)

$CrSb/(Bi,Sb)_2Te_3$: Neutron Reflectometry

- PNR from a $CrSb/(Bi,Sb)_2Te_3$ superlattice shows extremely weak magnetism
- Most consistent with interface magnetism
  - Proximity?
  - Intermixing?



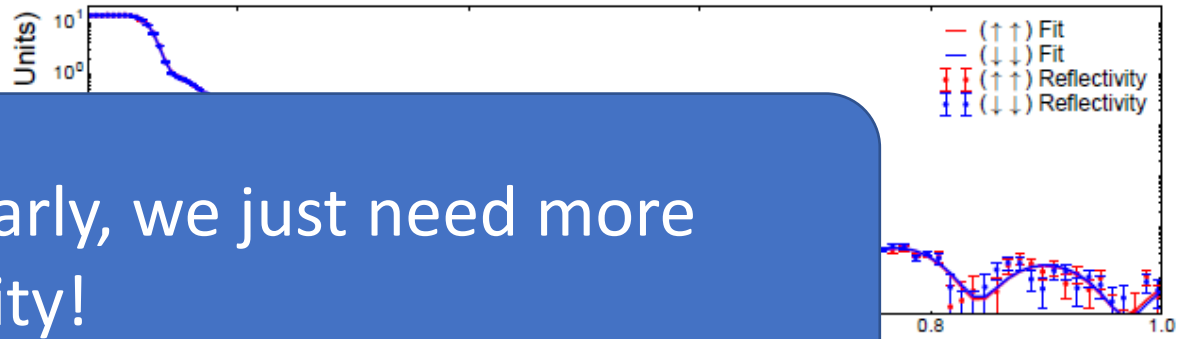


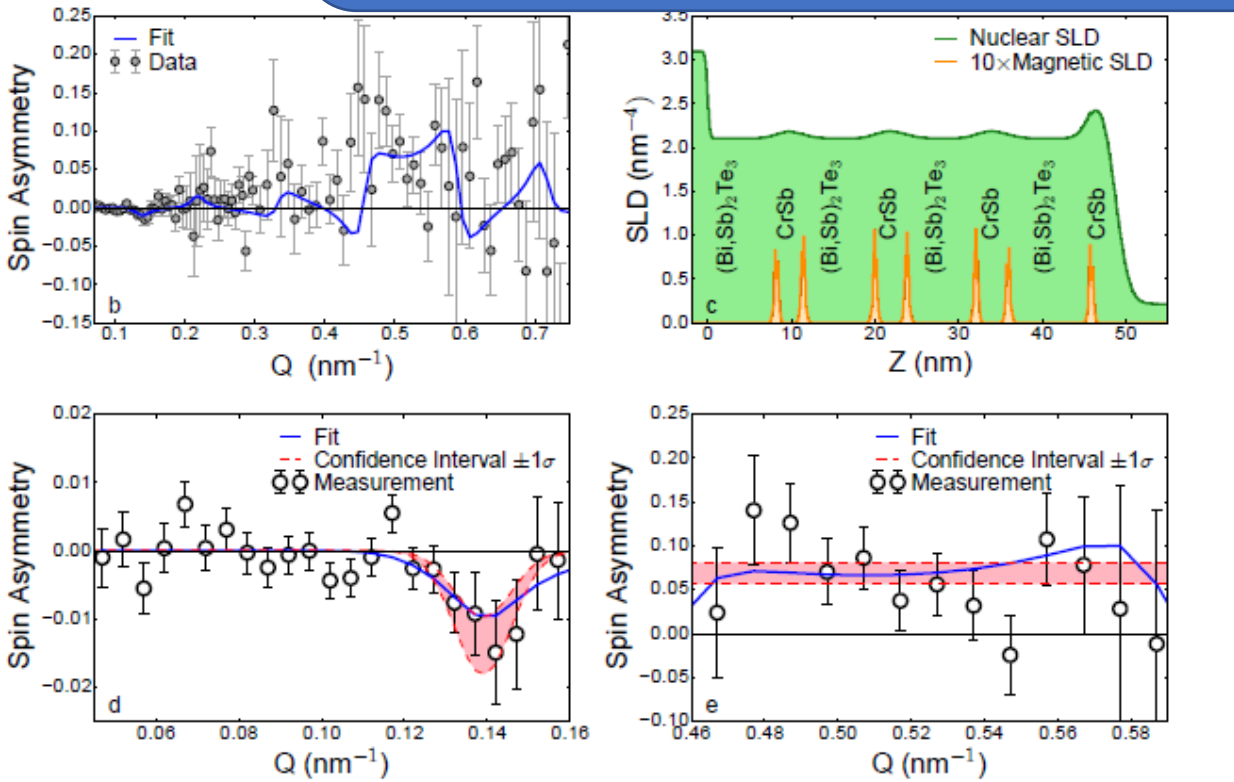


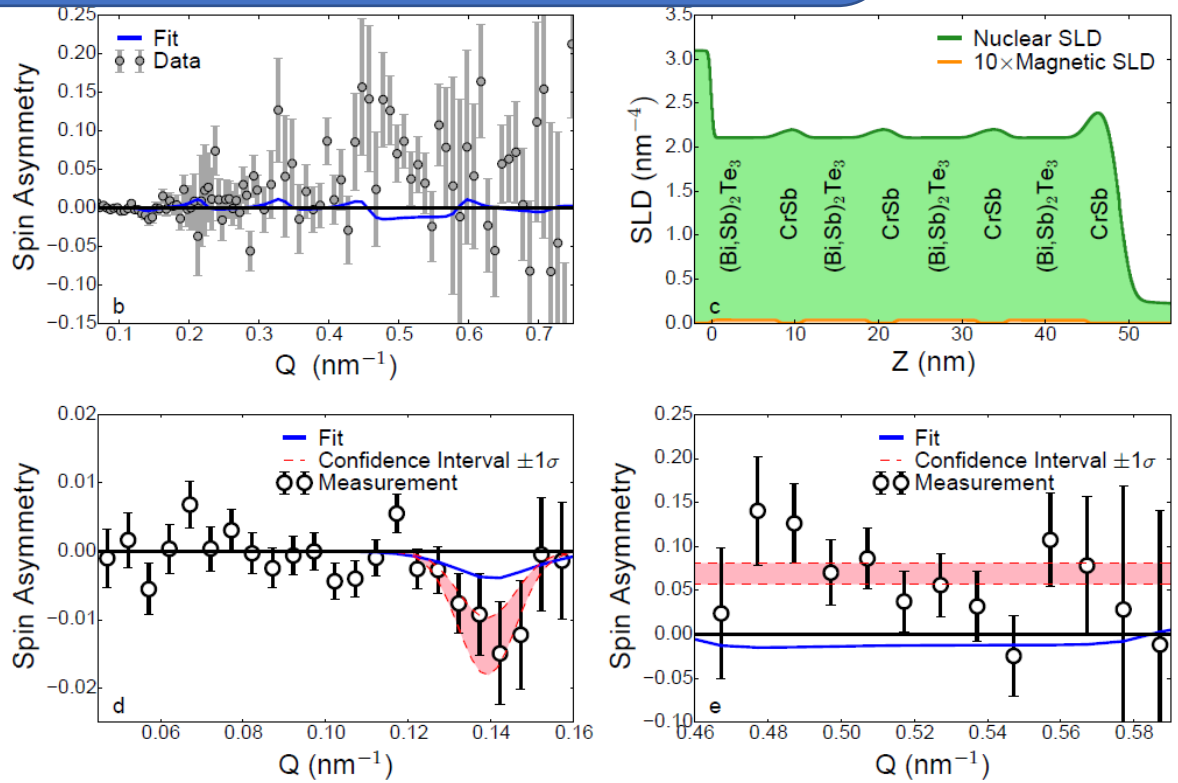
$CrSb/(Bi,Sb)_2Te_3$: Neutron Reflectometry

- PNR from a $CrSb/(Bi,Sb)_2Te_3$ superlattice shows extreme
- Most co
  - Pro
  - Inte

It worked, but… barely Clearly, we just need more intensity!

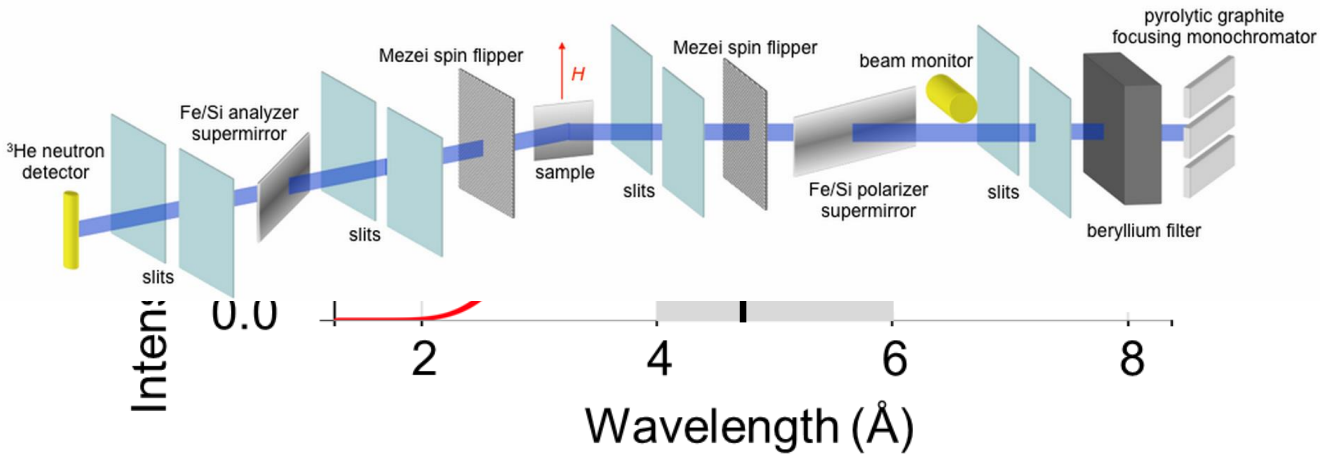


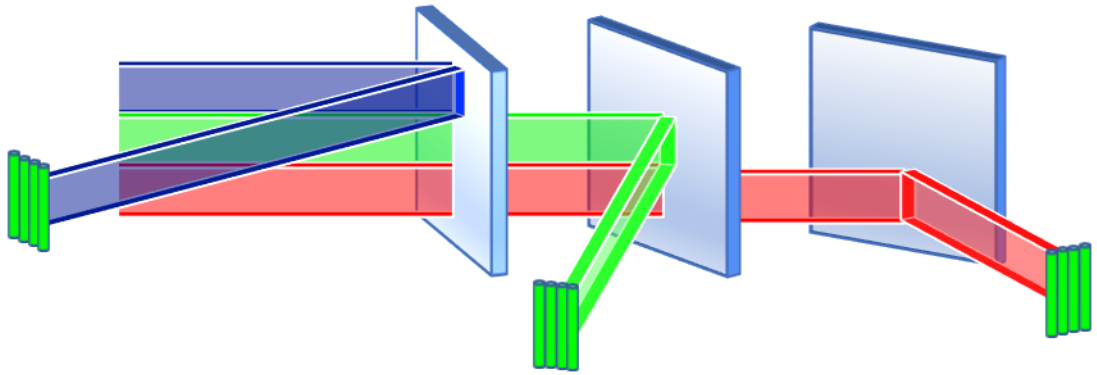




CANDOR: Using More Neutrons

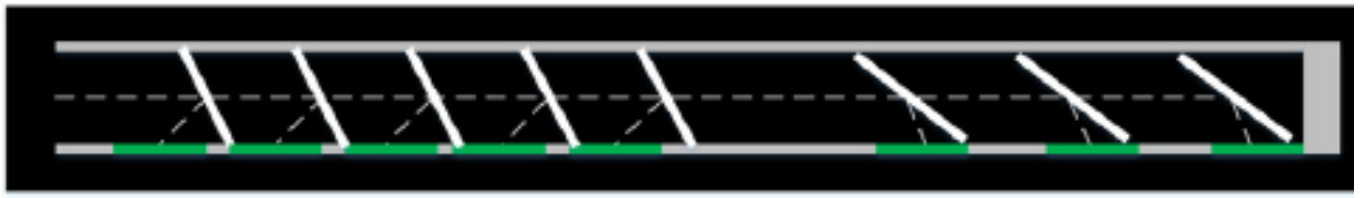
- We already have a ton of unused neutrons!
- Traditional continuous beam (reactor) reflectometers have a monochromator to select a single wavelength
- What if we instead took all the wavelengths and sorted them out on the other side?



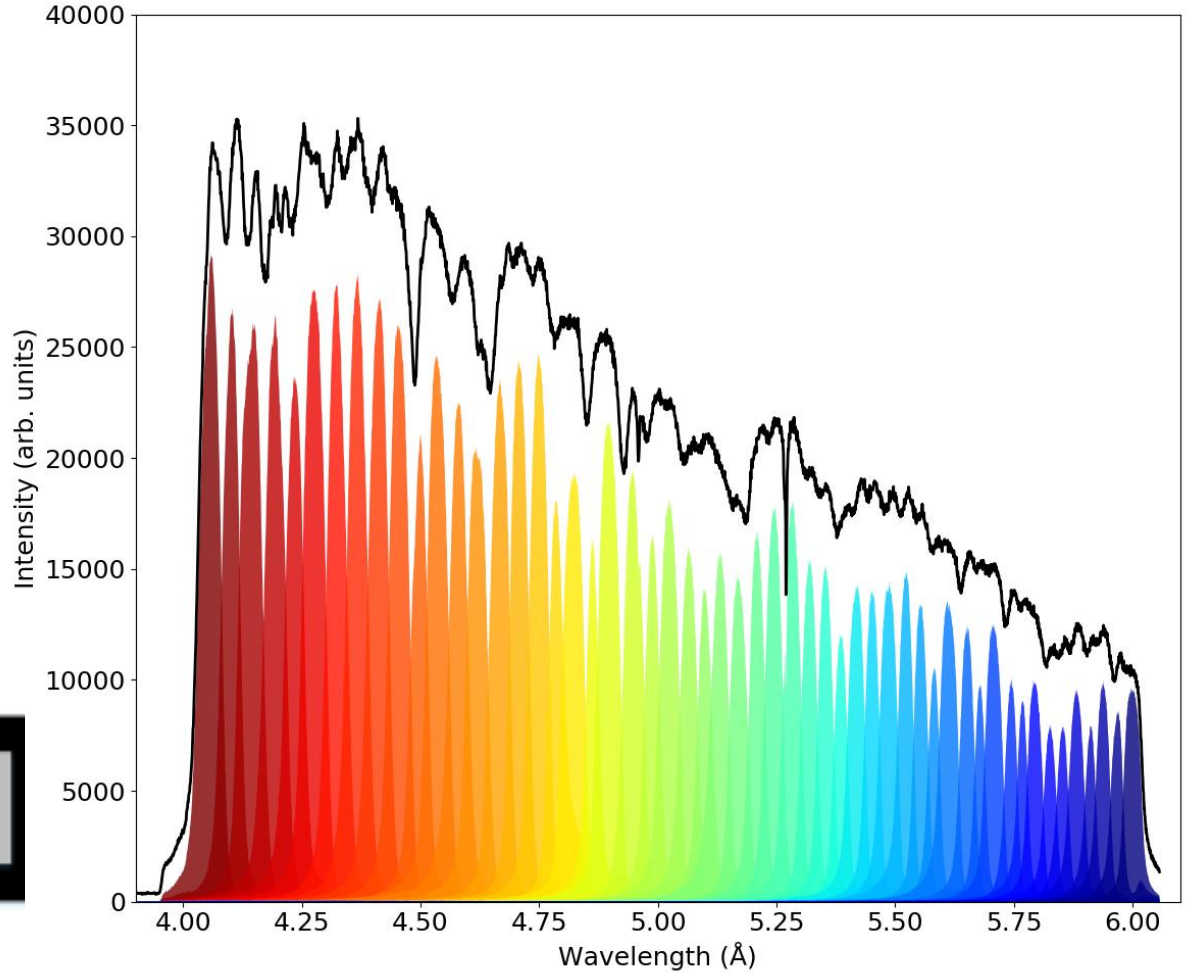


Polychromatic Detector Array

- Each detector array consists of 54 graphite crystals directing neutrons into detectors
- Arranged in two banks of 54 detectors each (eventually more than 20 banks)
- Cryocooled to reduce thermal diffuse scattering and improve background

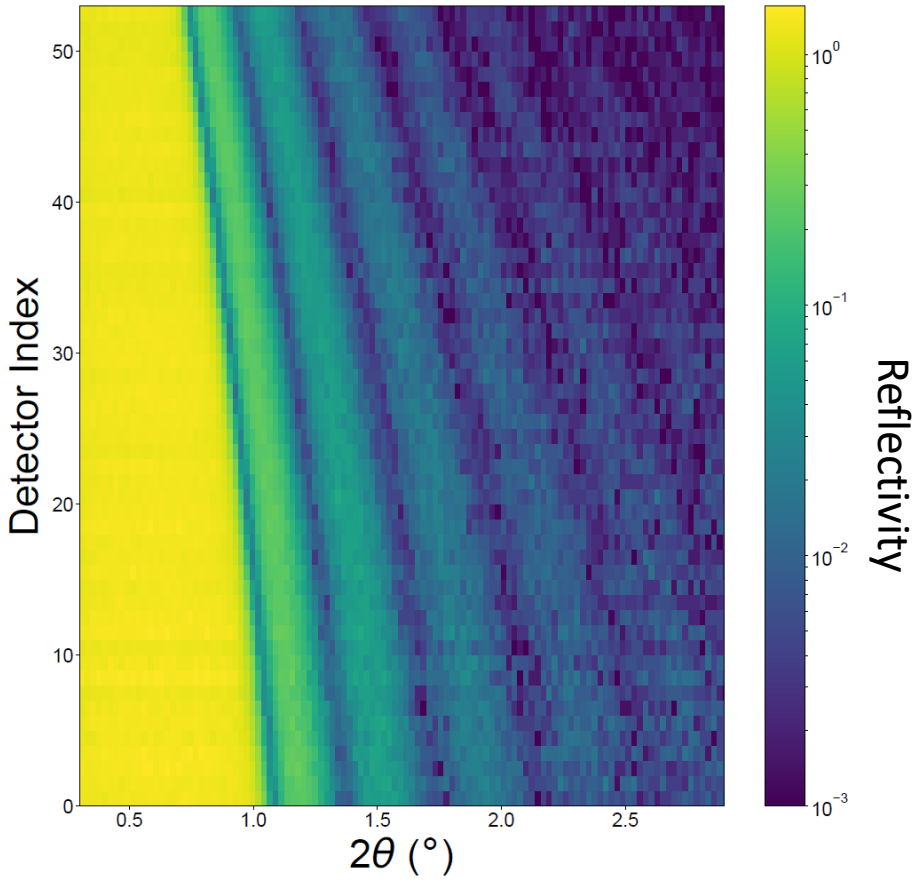


λ (Å)	5.98	5.96	5.93	5.90	5.87	•••	4.09	4.05	4.00
θ (°)	63.1	62.6	62.1	61.6	61.1		37.6	37.1	36.6

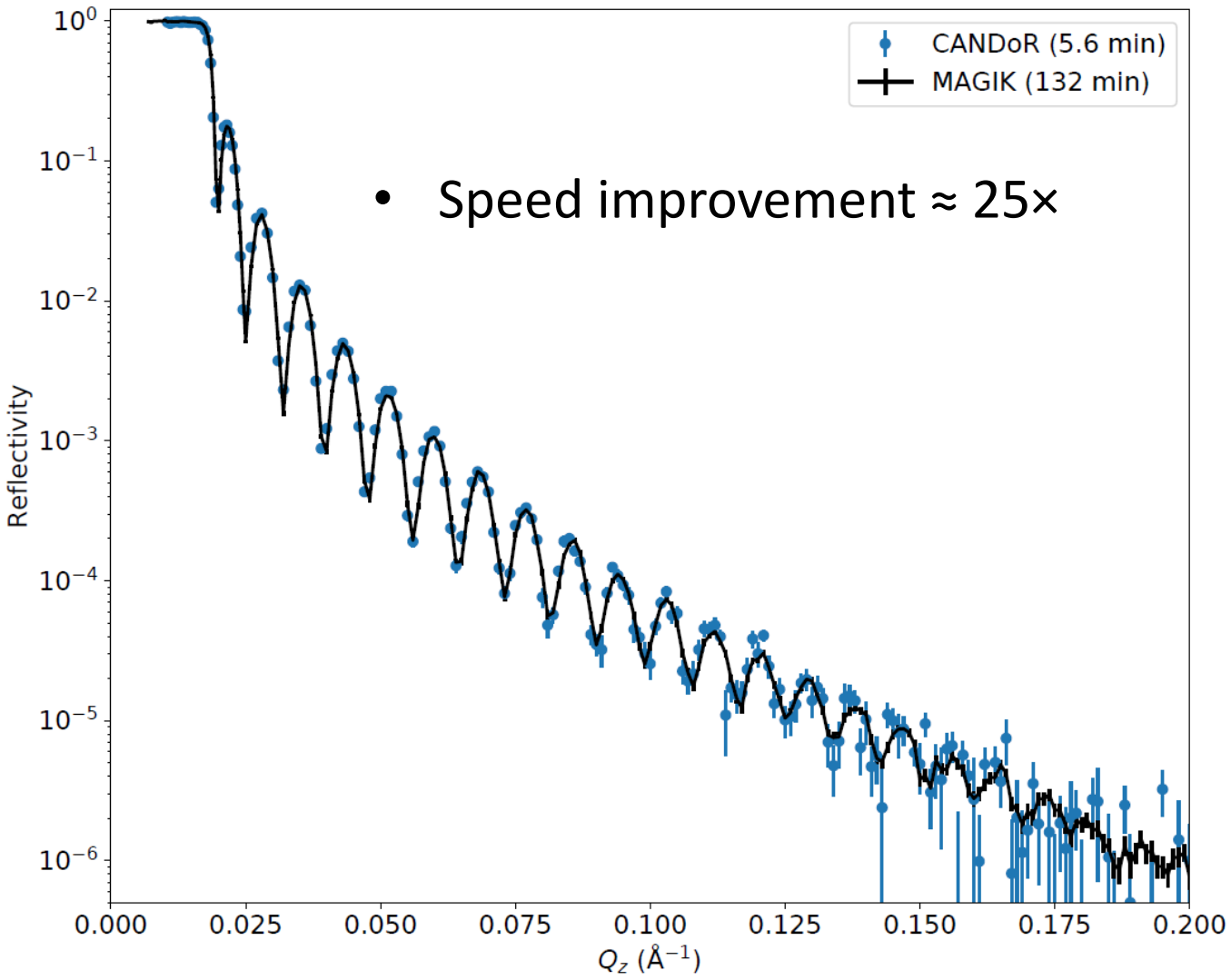


N. Maliszewskyj *et al.* 2018, *Nucl. Inst. Meth. Phys. Res. A*, **907**, 90

First Measurements: Beam Intensity



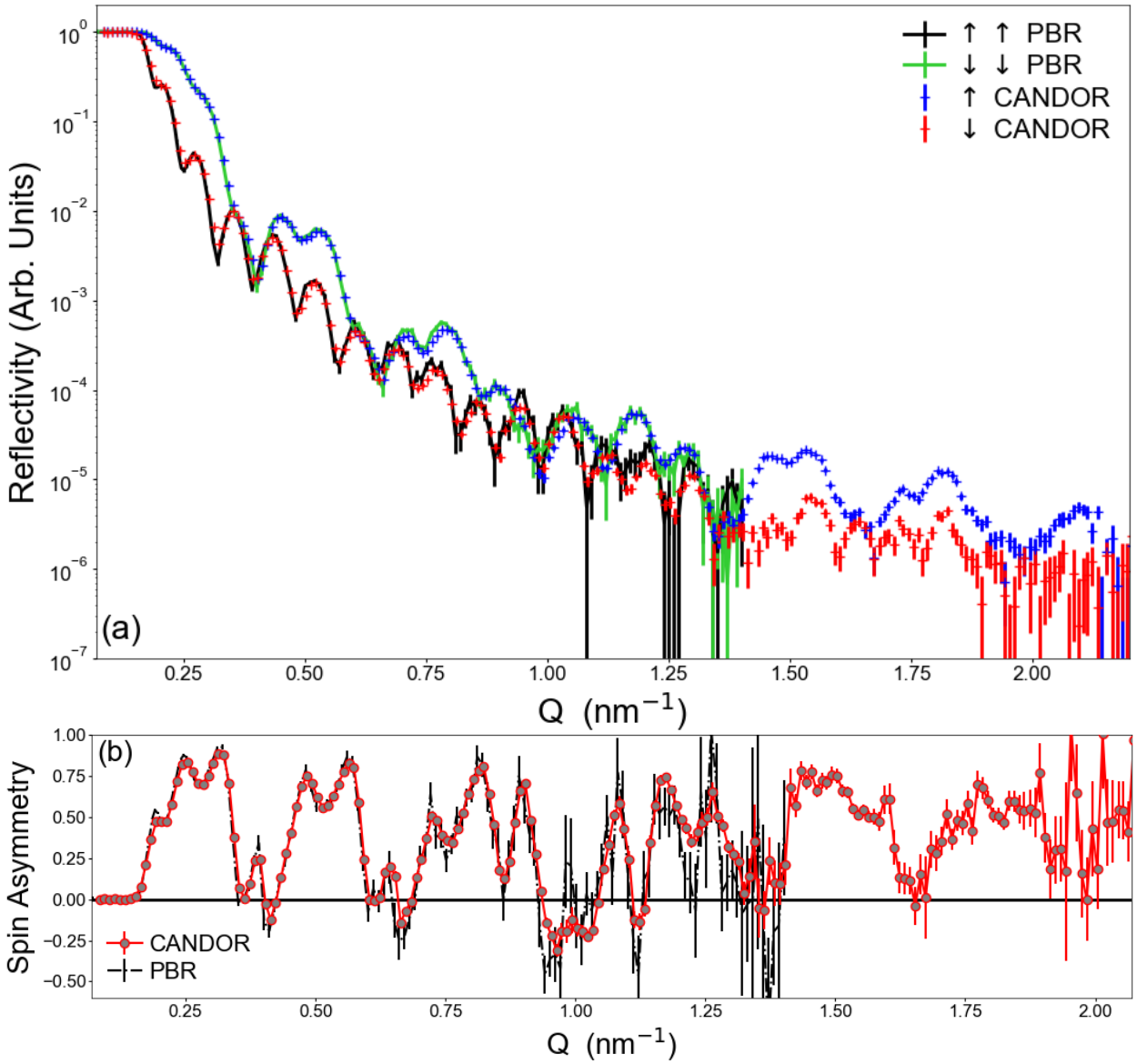
Standard thick film sample (J. Dura, NIST)



- Speed improvement ≈ 25×

First Measurements: Magnetic Materials

- Beam polarization essentially perfect, with *uncorrected* CANDOR data matching corrected PBR data
- Fitting data gives the same answer
- Much wider useful Q-range

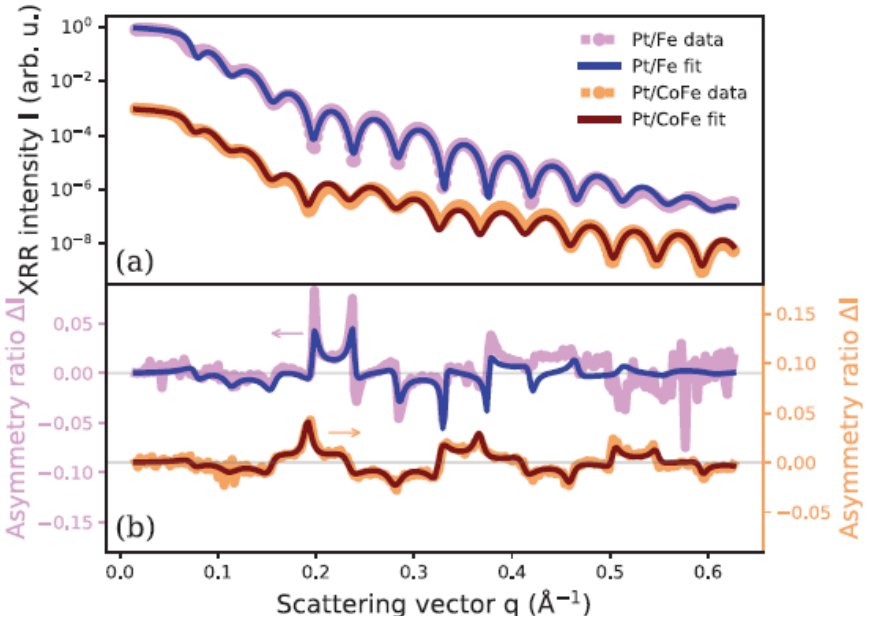


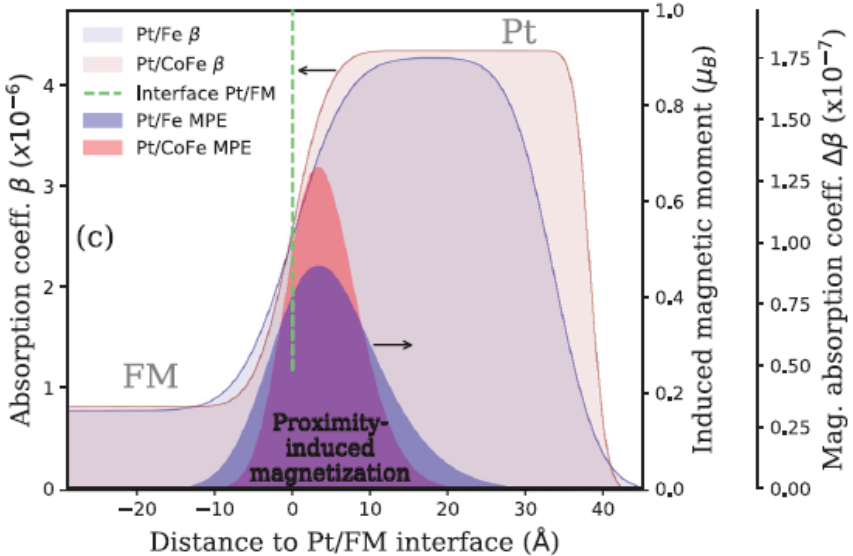
X-ray Resonant Magnetic Reflectivity

Quantitative comparison of the magnetic proximity effect in Pt detected by XRMR and XMCD

Dominik Graulich,[1,a)] Jan Krieft,[1] Anastasiia Moskaltsova,[1] Johannes Demir,[1] Tobias Peters,[1] Tobias Pohlmann,[2,3] Florian Bertram,[3] Joachim Wollschläger,[2] Jose R. L. Mardegan,[3] Sonia Francoual,[3] and Timo Kuschel[1]

Appl. Phys. Lett. **118**, 012407 (2021); doi: 10.1063/5.0032584





- XRMR has, in principle, everything one needs to characterize magnetic proximity effects and interface coupling with phenomenal precision
- Huge strides in analysis recently
- Relatively underutilized technique in topologically nontrivial matter

Conclusions

- Depth and element resolved probes are critical components for probing magnetic proximity effects
- Extreme care must be taken about interpretation, in particular when confounding structural effects may be present
- Combine as many independent techniques as possible
- Looking forward to new instrumentation and techniques like multiplexing neutron reflectometers and the application of X-ray resonant magnetic reflectivity

<u>Further Reading:</u>

S. Bhattacharyya et al., Adv. Mater. 33, 2007795 (2021)
J. Liu and T. Hesjedal, Adv. Mater. 2102427 (2021)
A. J. Grutter and Q. L. He, Phys. Rev. Mater. 5, 090301 (2021)

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- Yuri Suzuki

Samarth Group, Penn State
- Timothy Pillsbury
- Wilson Yanez
- Arpita Mitra
- Max Stanley
- Anthony Richardella
- Nitin Samarth

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alexander.grutter@nist.gov
ncnr.nist.gov