

Hidden Interfaces and High-Temperature Magnetism in Intrinsic Topological Insulator - Ferromagnetic Insulator Heterostructures

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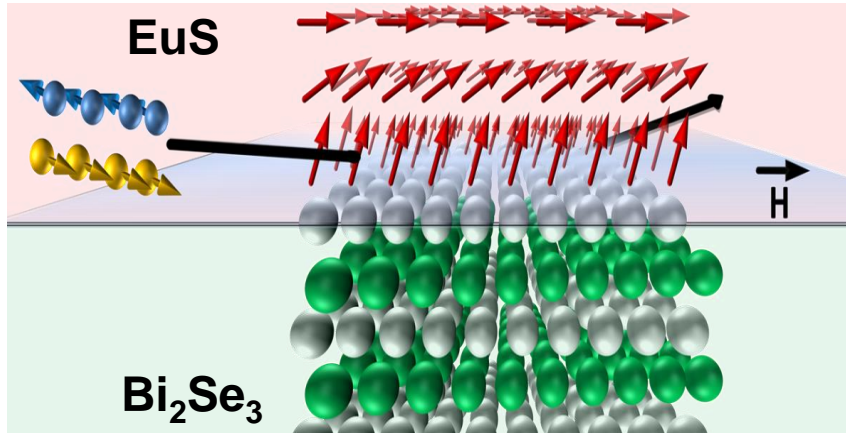
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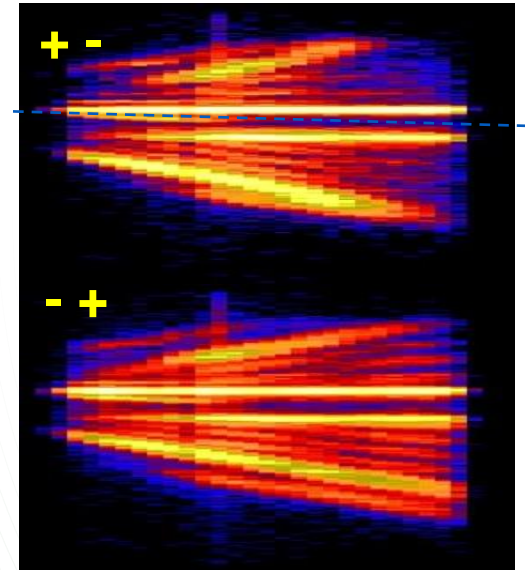
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Outline



- **Topological Insulators - a new phase of matter with TRS**
- **Symmetry breaking in TI via *magnetic proximity***
- **Induce ferromagnetism in Topological Insulator via exchange coupling in TI-FMI**

- **Polarized Neutron Reflectometry: *depth resolved vector magnetometry* for TI – FMI heterostructures**



Interface ferromagnetism in TI via magnetic proximity

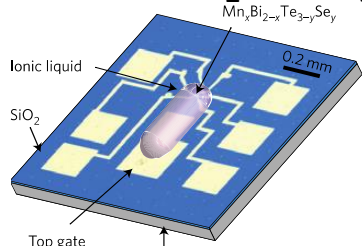


Introducing ferromagnetic order in TI:

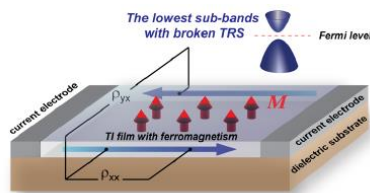
- **by doping with specific elements:**
 - hard to separate the surface and the bulk phases.
 - introduces crystal defects, magnetic scattering centers, impurity states in the insulating gap are detrimental to mobility and the transport of spin-momentum locked surface electrons in TIs.
- **by uniformly depositing magnetic atoms (Fe) over the TI surface:**
 - the transport properties of a TI are influenced by the metallic ferromagnetic overlayer or atoms.
- **by magnetic proximity with FI:**
 - the spin-momentum locked helical electronic states in TIs and topological magneto-electric effect

Topological insulator materials: Magnetic?

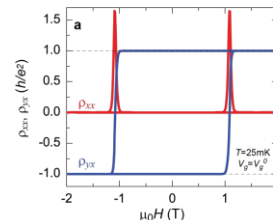
Mn-doped $\text{Bi}_2(\text{TeSe})_3$



Cr-doped $(\text{BiSb})_2\text{Te}_3$



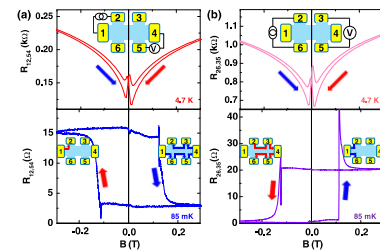
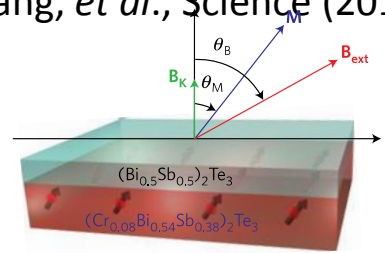
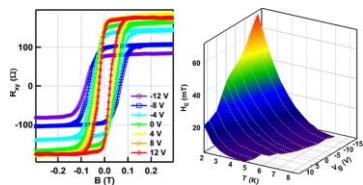
V-doped $(\text{BiSb})_2\text{Te}_3$



Checkelsky, *et al.*, Nat. Phys. (2012)

Chang, *et al.*, Science (2013)

Chang, *et al.*, Nat. Mat. (2015)

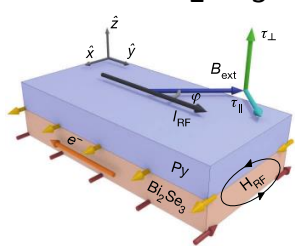


Kou, *et al.*, Nano Lett. (2013)

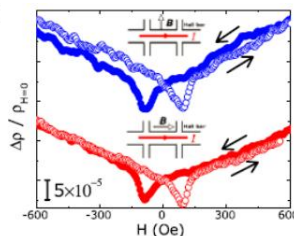
Fan, *et al.*, Nat. Mat. (2014)

Kou, *et al.*, PRL (2014)

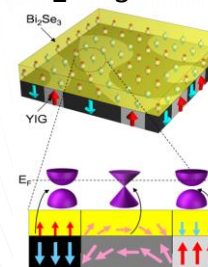
NiFe/ Bi_2Se_3



EuS/ Bi_2Se_3



$\text{Bi}_2\text{Se}_3/\text{YIG}$



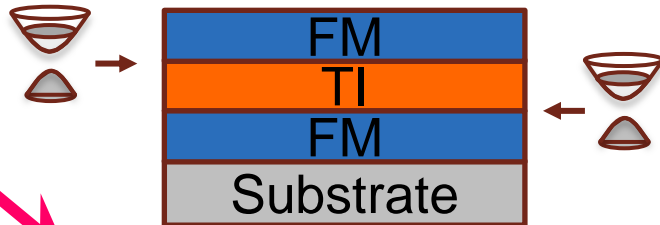
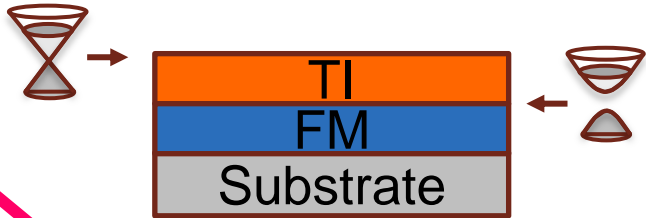
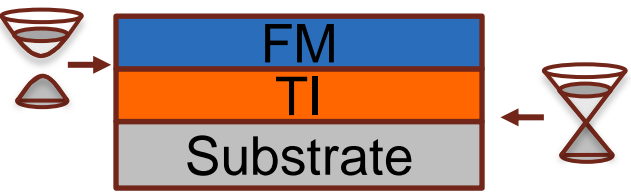
Melnik, *et al.*, Nature (2014)

Wei, *et al.*, PRL (2013)

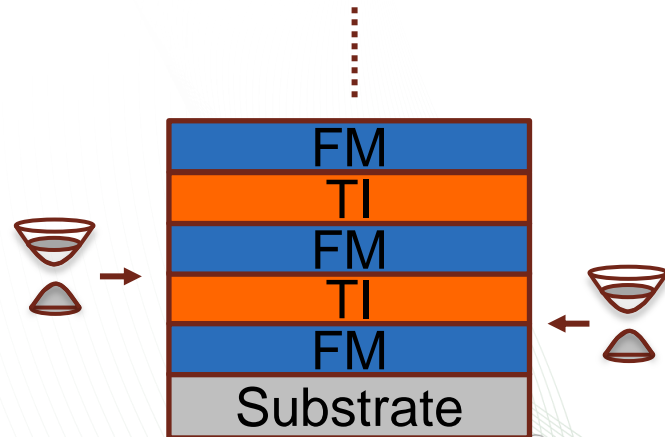
Lang, *et al.*, Nano Lett. (2014)

Topological insulator materials: Magnetism via proximity

Increasing Complexity

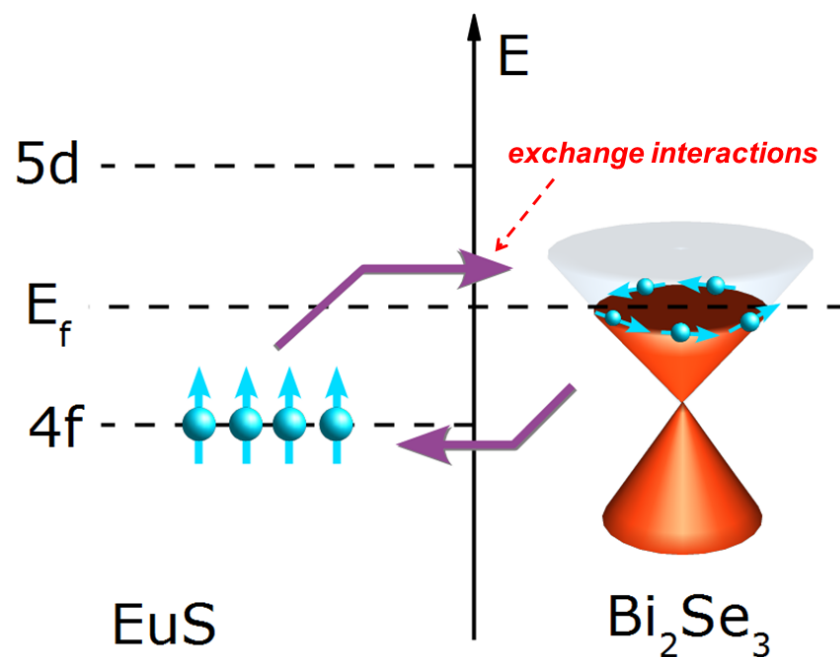


Increasing Magnetization



Interface ferromagnetism in TI via magnetic proximity

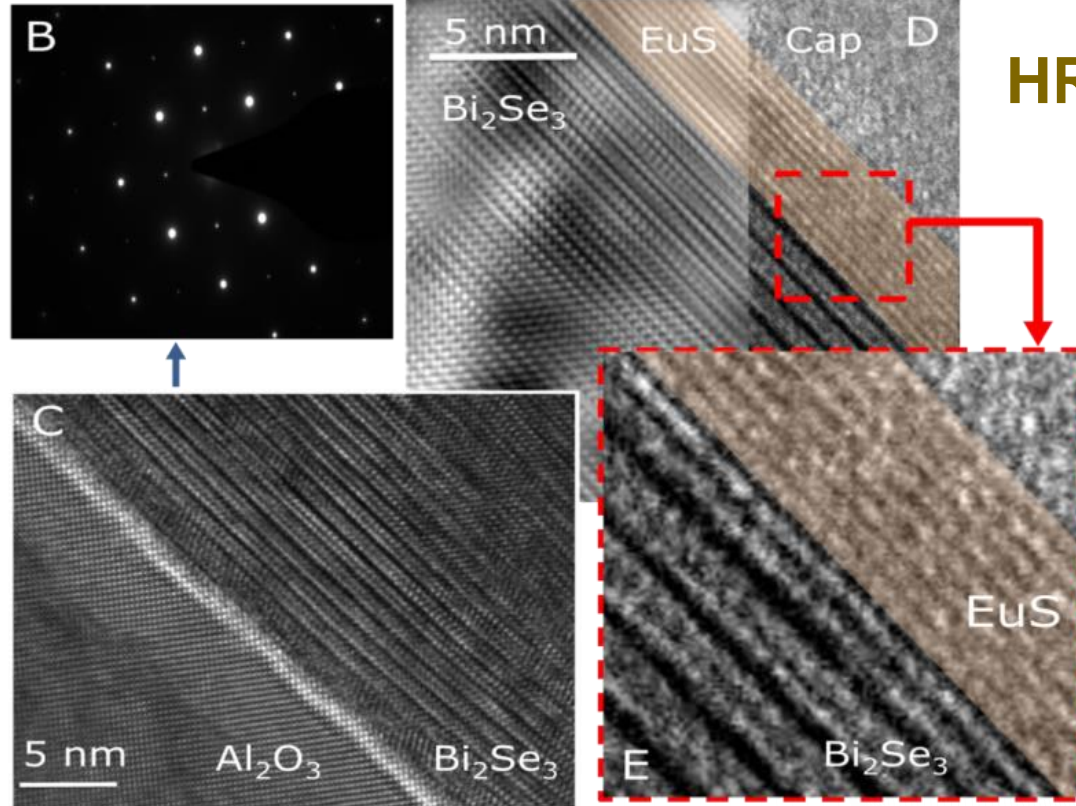
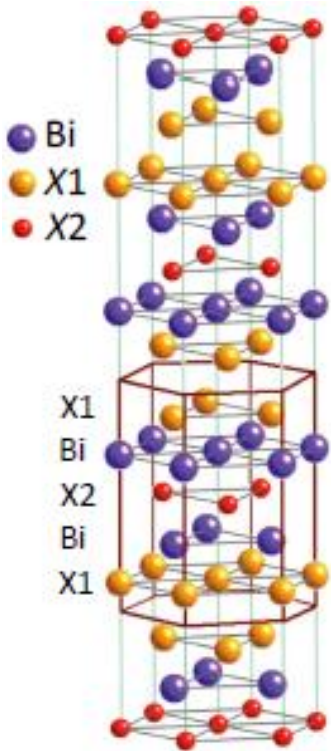
- The particular type of *interface* - between a *topological insulator* and a *ferromagnet* – might become key to the computer industry's future ability.
- The goal is the ability to manipulate surface electron states.
- We introduce ferromagnetic order onto the surface of TI Bi_2Se_3 thin films by using FI EuS.
- EuS 4f-5d energy gap 1.64 eV
Fermi level inside the gap



Characterization Bi₂Se₃/EuS bilayers

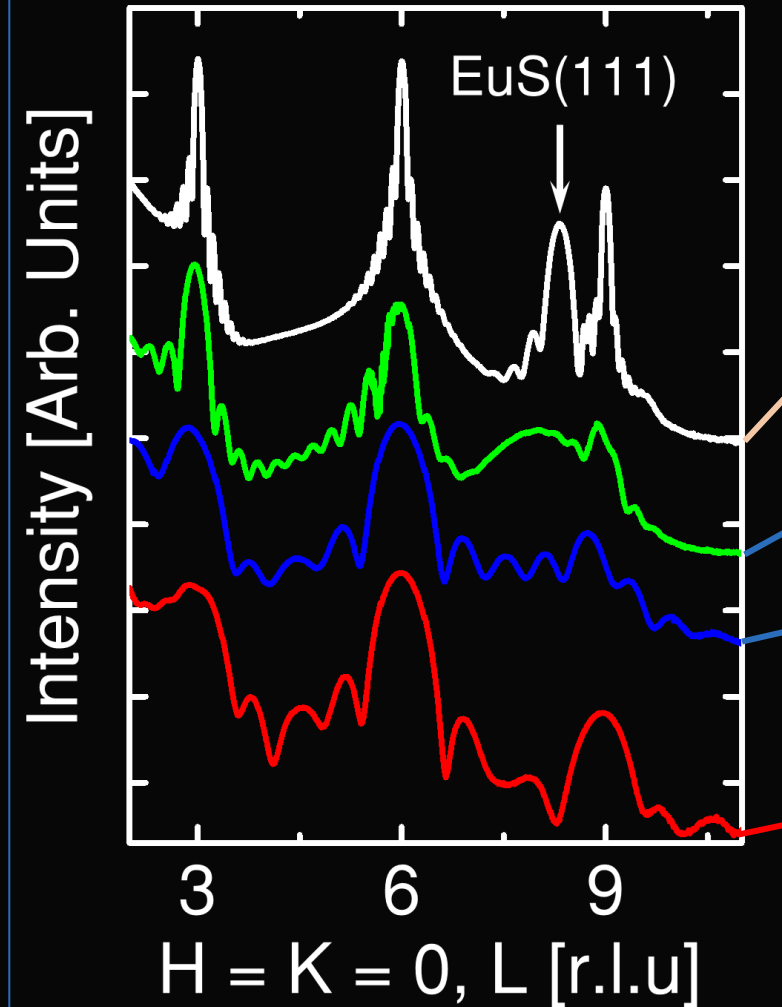
Bi₂Se₃

HRTEM



- (B) electron diffraction image with an hexagonal symmetry of Bi₂Se₃
- (C) HRTEM image for substrate and Bi₂Se₃
- (D) HRTEM images for EuS and Bi₂Se₃ interface

Epitaxial relationship between EuS & Bi₂Se₃



- Heterostructures grown on Al₂O₃ (0001)

30 QL Bi₂Se₃ with 10 nm EuS

10 QL Bi₂Se₃ with 3 nm EuS

5 QL Bi₂Se₃ with 1 nm EuS

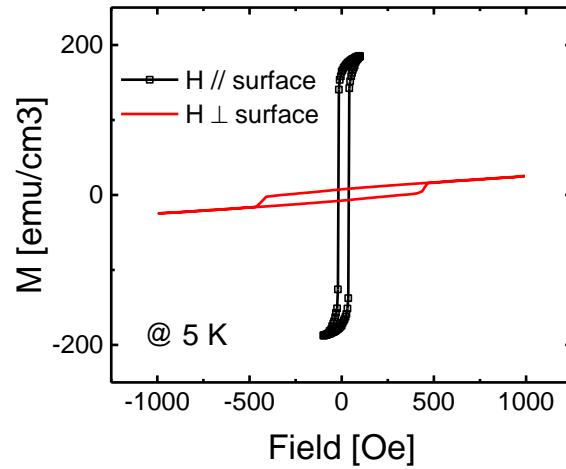
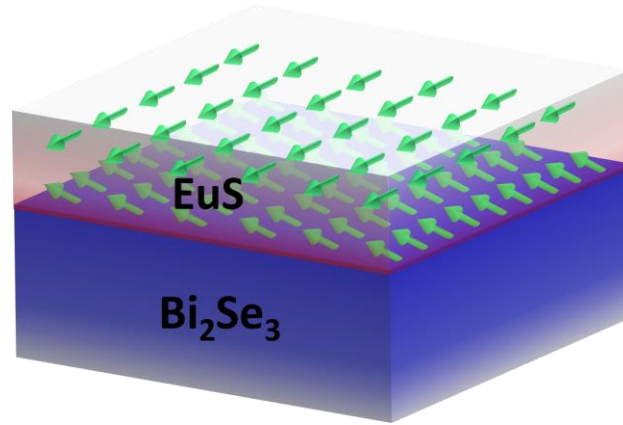
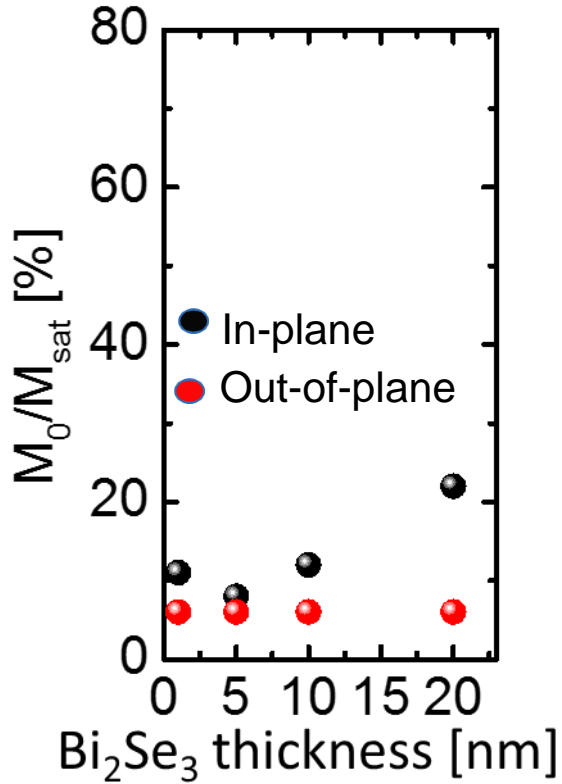
5 QL Bi₂Se₃

- Bi₂Se₃ (0001) // Al₂O₃ (0001)
- EuS (111) // Bi₂Se₃ (0001)

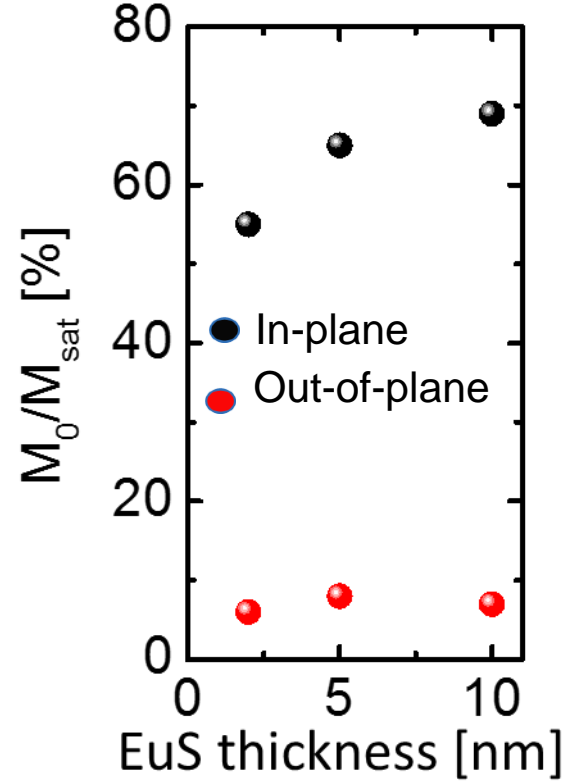
Strong interface magnetization

SQUID

EuS = 1 nm



Bi₂Se₃ = 20 nm



- All samples display ferromagnetism
- Out-of-plane remanance ratio does not depend on thicknesses ----->
-----> evidence that the out-of-plane component is at the interface

Samples for PNR experiments



$\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$

MBE with a base pressure of 10^{-10} Torr on **substrates Al_2O_3**

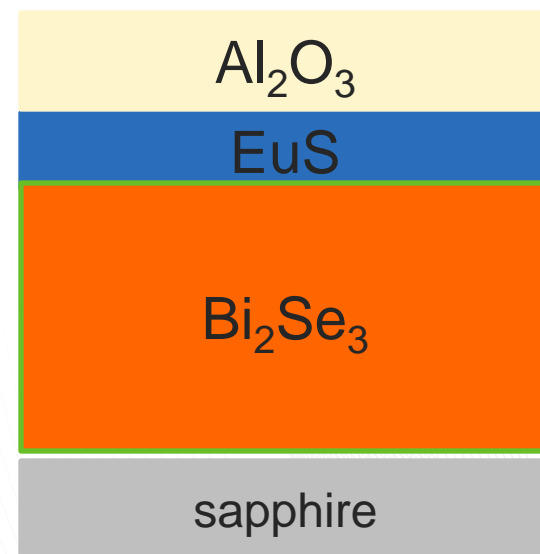
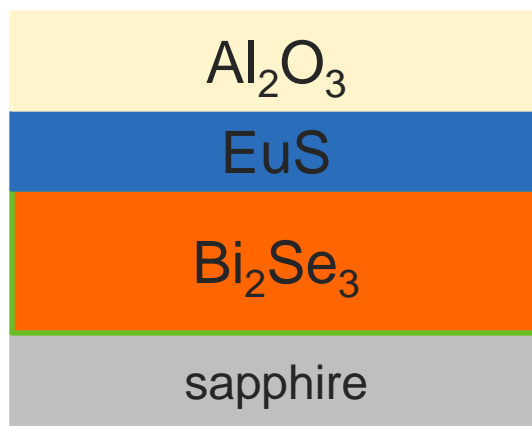
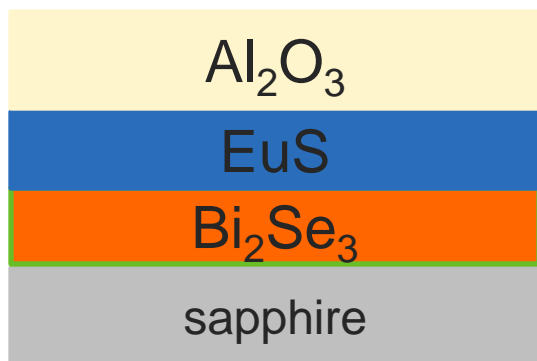
EuS (5 nm)Bi₂Se₃ (5nm)

EuS (5 nm)Bi₂Se₃ (10nm)

EuS (5 nm)Bi₂Se₃ (20nm)

EuS (5 nm) – reference sample

**Sample size
20 X 20 mm²**

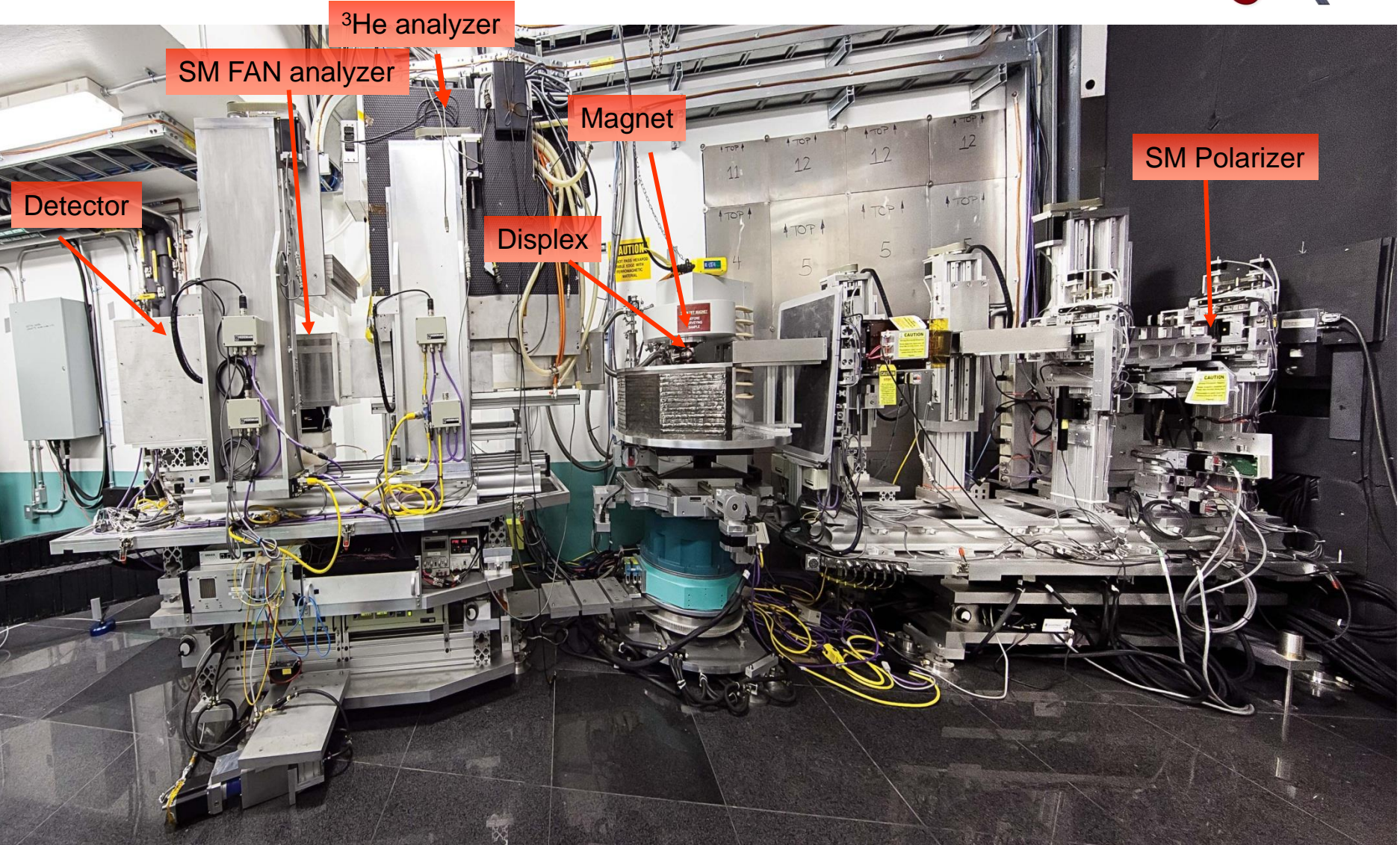


Spallation Neutron Source, ORNL, TN



07-P04234/arm

Magnetism Reflectometer at SNS



Magnetism Reflectometer at SNS



High intensity sample size 5x5 mm²
Low background 10⁻⁸
High polarization 98.5%
Polarization analysis

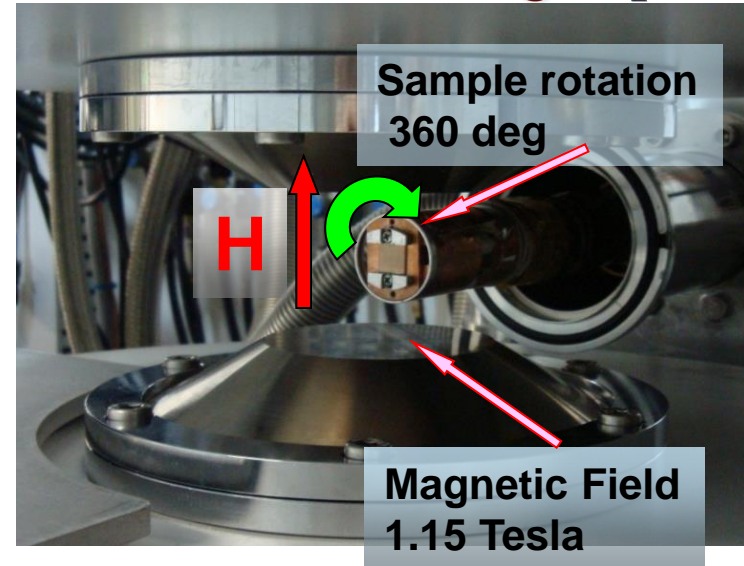
Polarized and unpolarized beam
Fast laser pre-alignment
Efficient thermal cycling (5K – 750K)

Sample environment new features:

Displex:

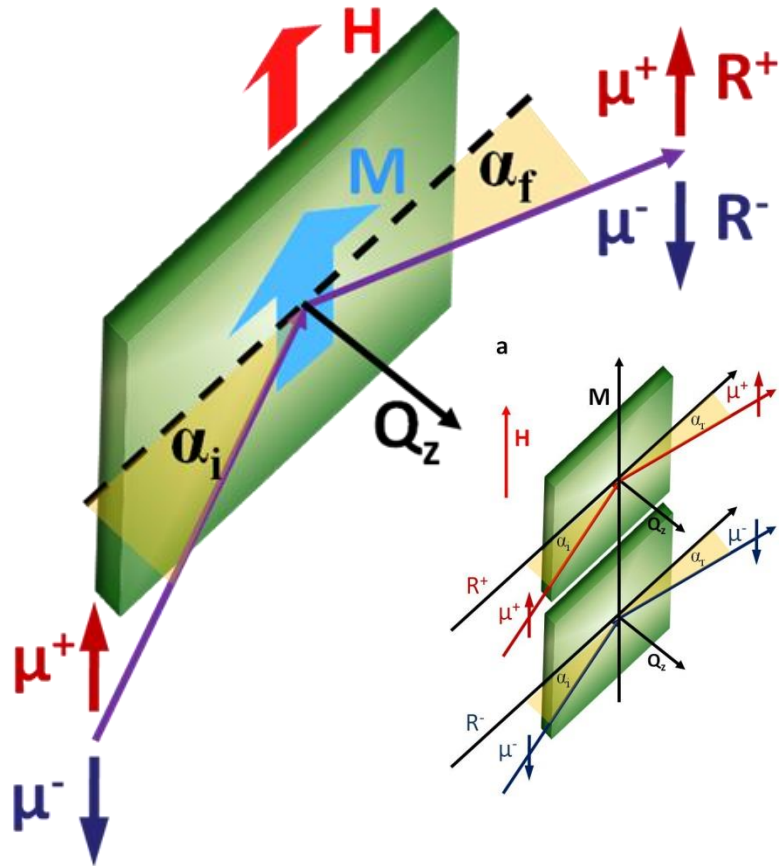
In-situ annealing
Sample rotation
Bias voltage

Electromagnet 1.15 Tesla (50 mm gap)
1.24 Tesla (46 mm gap)
2.40 Tesla (15 mm gap)



Sample Temperature
from 5K to 750K

Polarized Neutron Reflectometry experiment on $\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$



Fermi pseudopotential:

$$V_{\pm} = 2\pi\hbar/m N(b_n \pm b_m)$$

Momentum transfer

$$Q = 4\pi \sin \alpha_i / \lambda$$

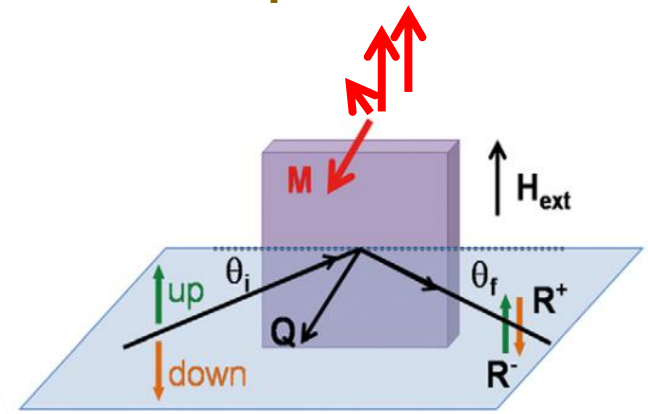
Nb_n – structural composition

Nb_m – *absolute* magnetization vector profile \vec{M} !

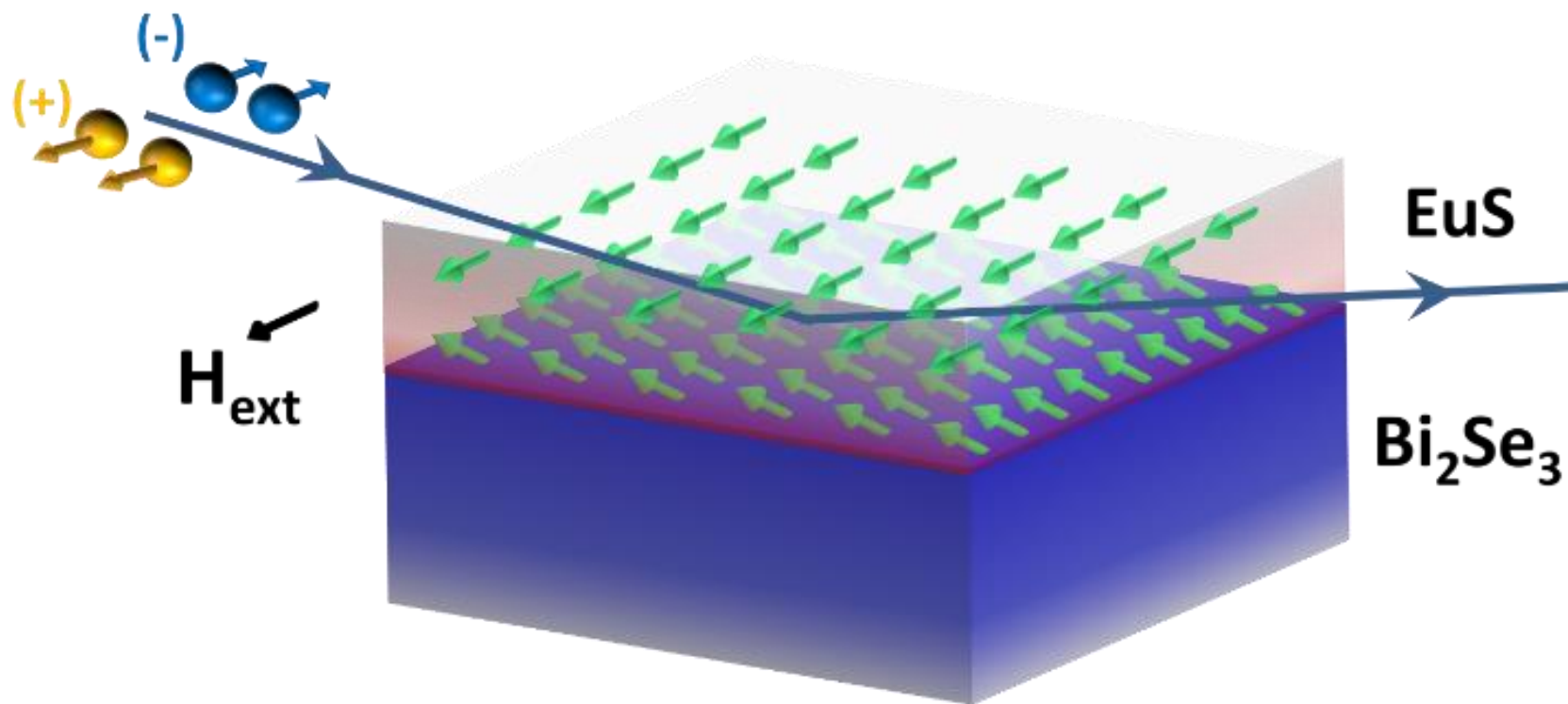
NIm_b - absorption profile

- $\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$ – structure profile
- $\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$ – magnetization profile
- $\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$ – absorption profile

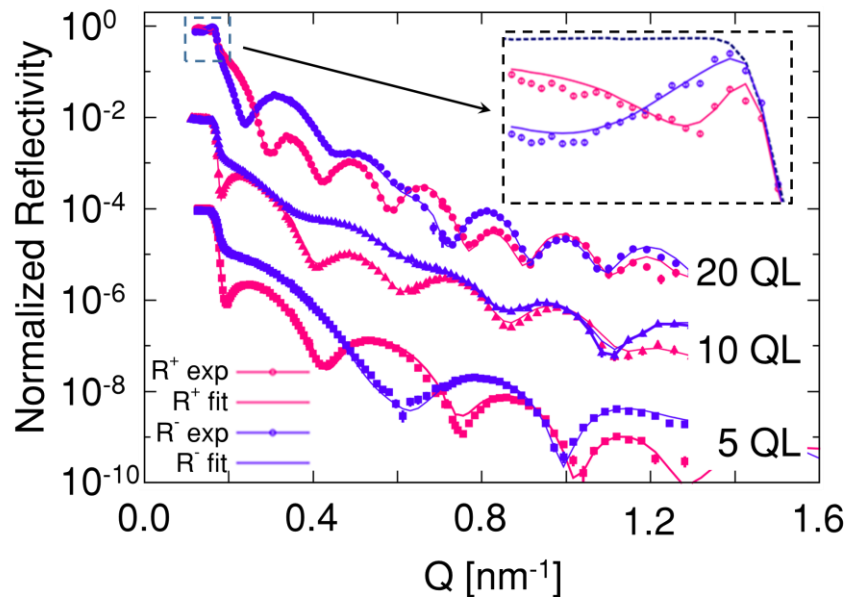
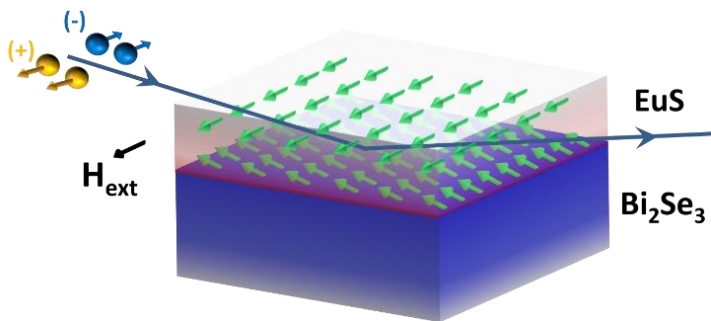
Perpendicular \vec{M}



Configuration of PNR experiment probing the magnetic moment distribution *inside* Bi₂Se₃/EuS interface

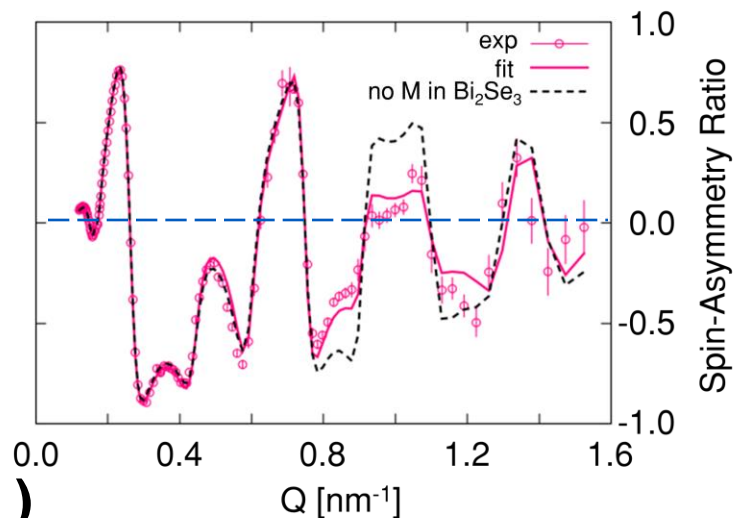
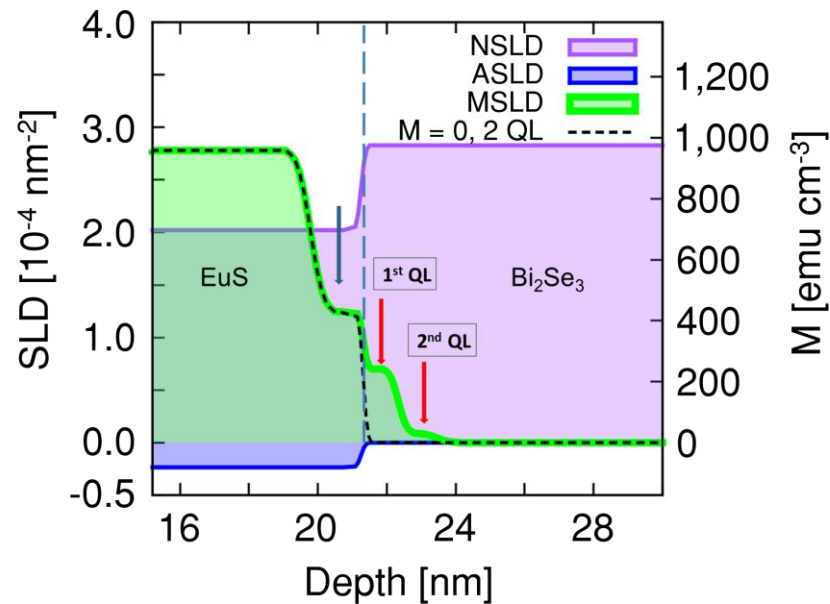


Magnetization – PNR @ 5 K

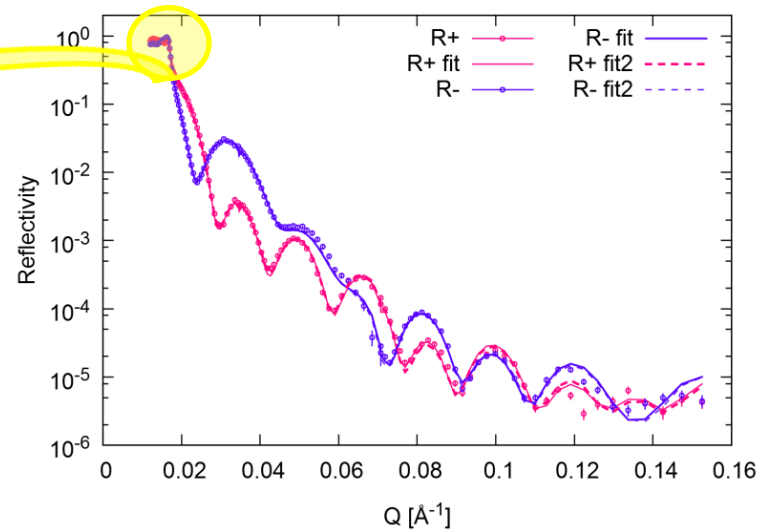
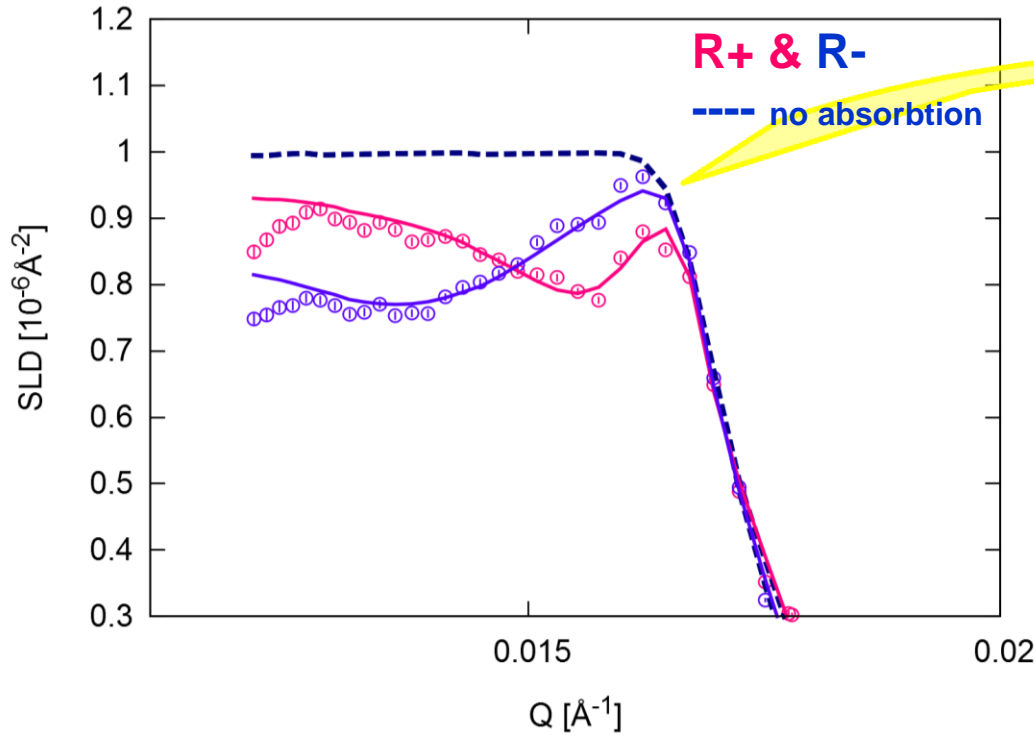


$$Q = 4\pi \sin \alpha_i / \lambda$$

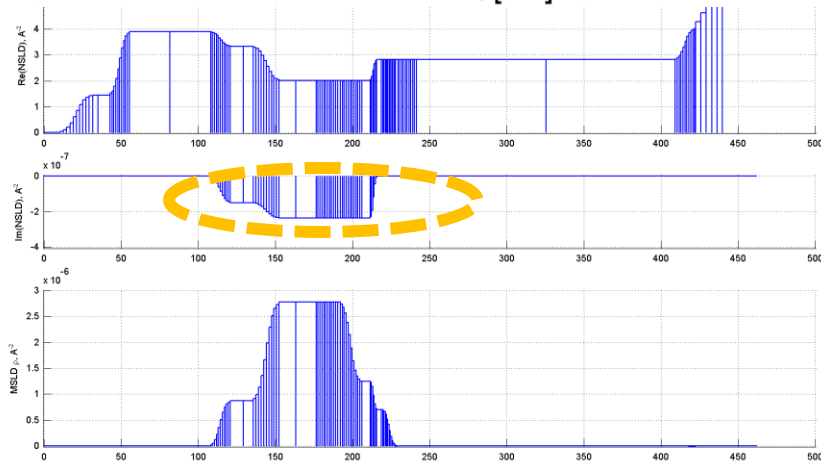
$$\text{Spin-Asymmetry : SA} = (R^+ - R^-) / (R^+ + R^-)$$



Absorption effect

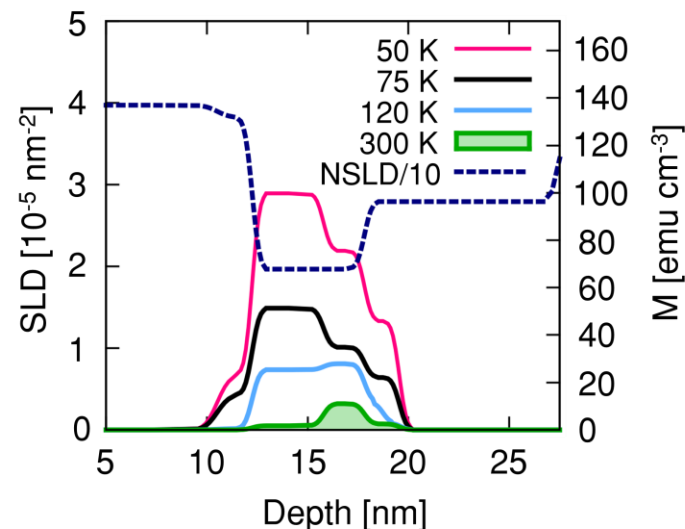
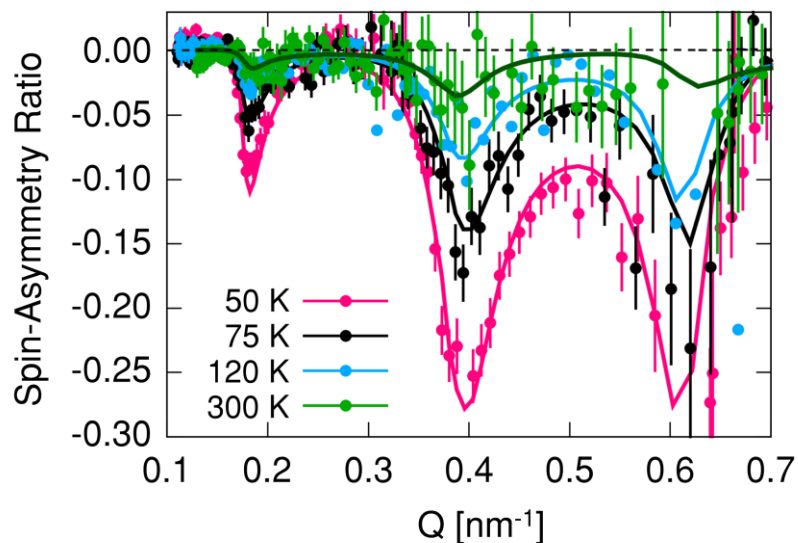


- Absorption: Im part of the SLD provides additional information about Eu distribution
- No Eu atoms are determined in Bi_2Se_3

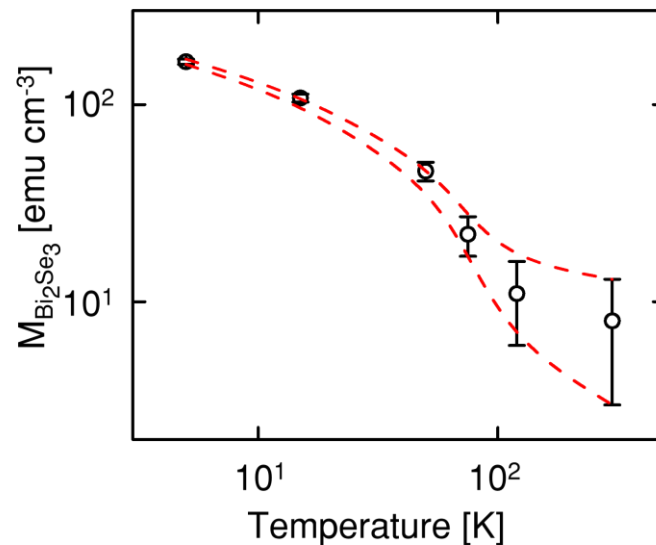


$\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$ – structure profile
 $\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$ – magnetization profile
 $\text{Al}_2\text{O}_3/\text{EuS}/\text{Bi}_2\text{Se}_3//\text{Al}_2\text{O}_3$ – absorption profile

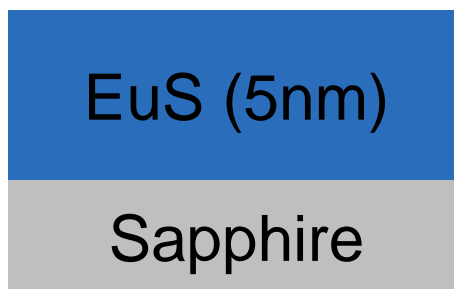
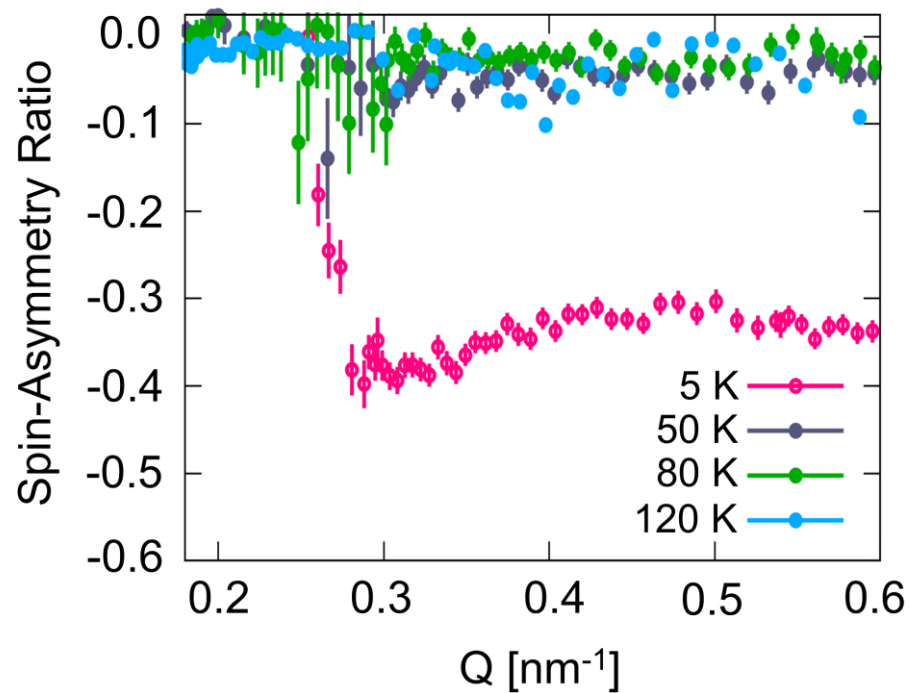
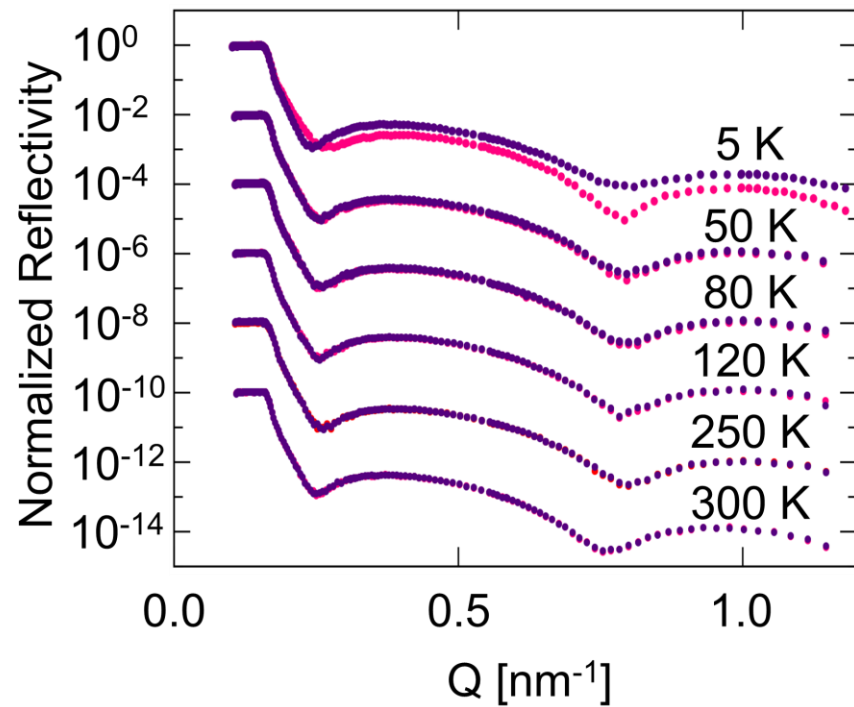
Magnetization – PNR @ 50,75,120,300 K



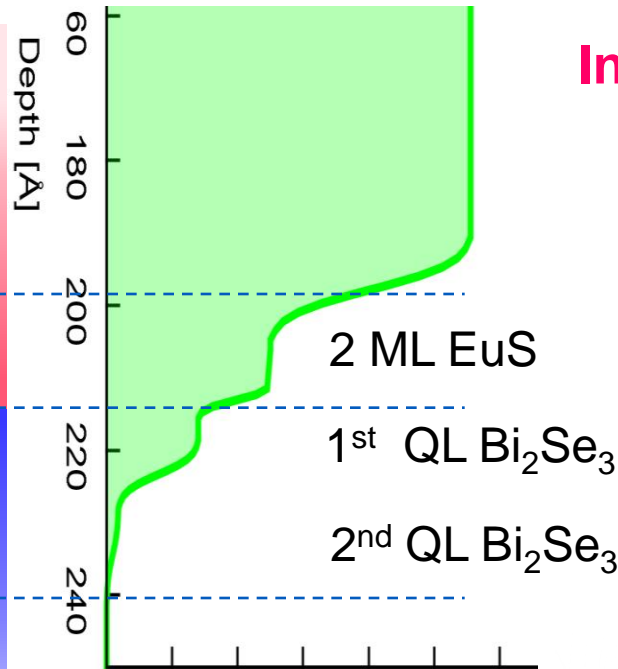
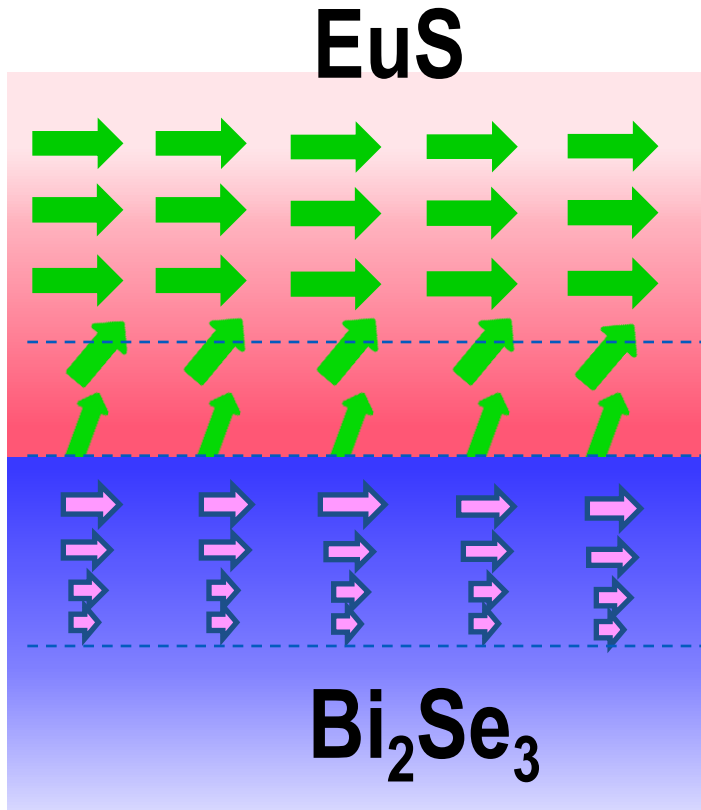
- Magnetization behavior of bilayers (10 QL/ 5 nm)
- Non-zero magnetization present in the 2 QL Bi_2Se_3 interfacial layer also penetrates into the EuS layer
- Magnetization reduced by an order of magnitude at higher temperatures
- No magnetization was detected above ~ 50 K in the pure EuS film.



Reference Sample - EuS//Sapphire

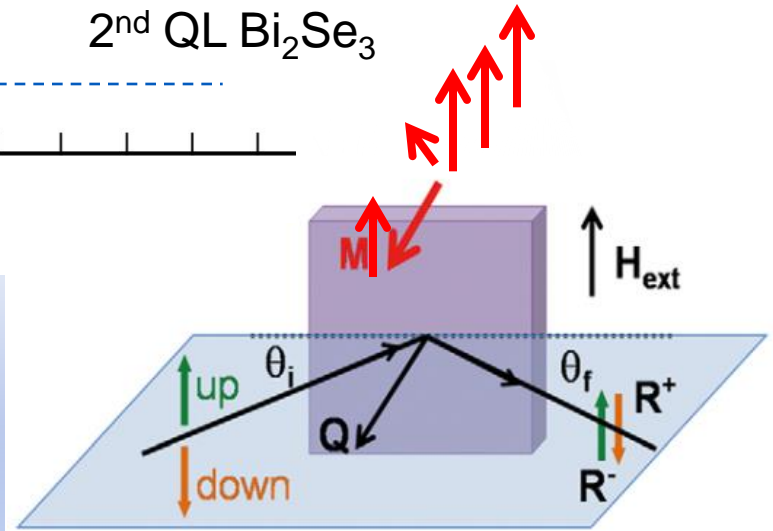


PNR: Magnetic moment distribution



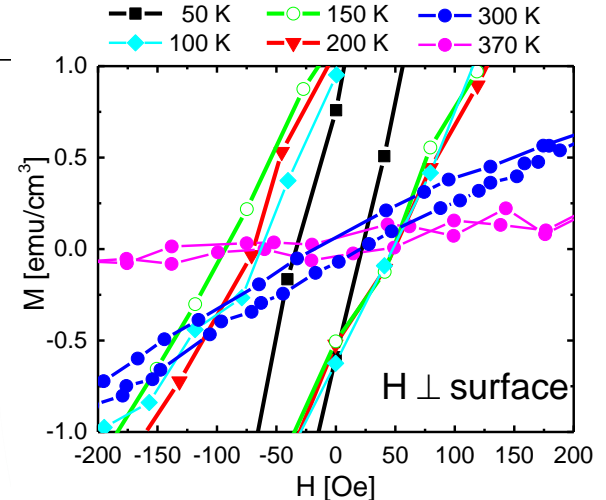
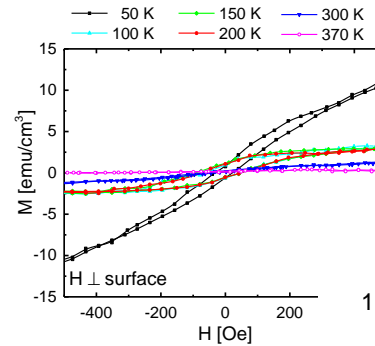
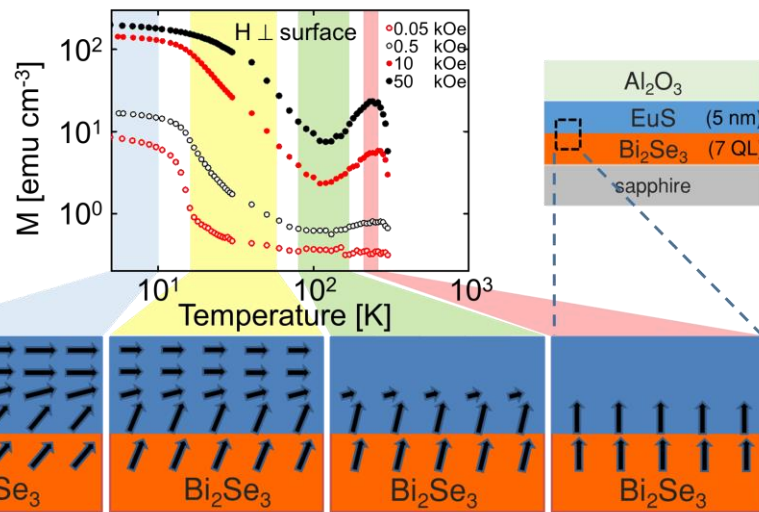
PNR measures
In-plane projection
of **M**

At the interface EuS/Bi₂Se₃, the strong spin-orbit coupling affects anisotropy direction, leading to an out-of-plane magnetic moment and results in generating a gap in the TI surface state



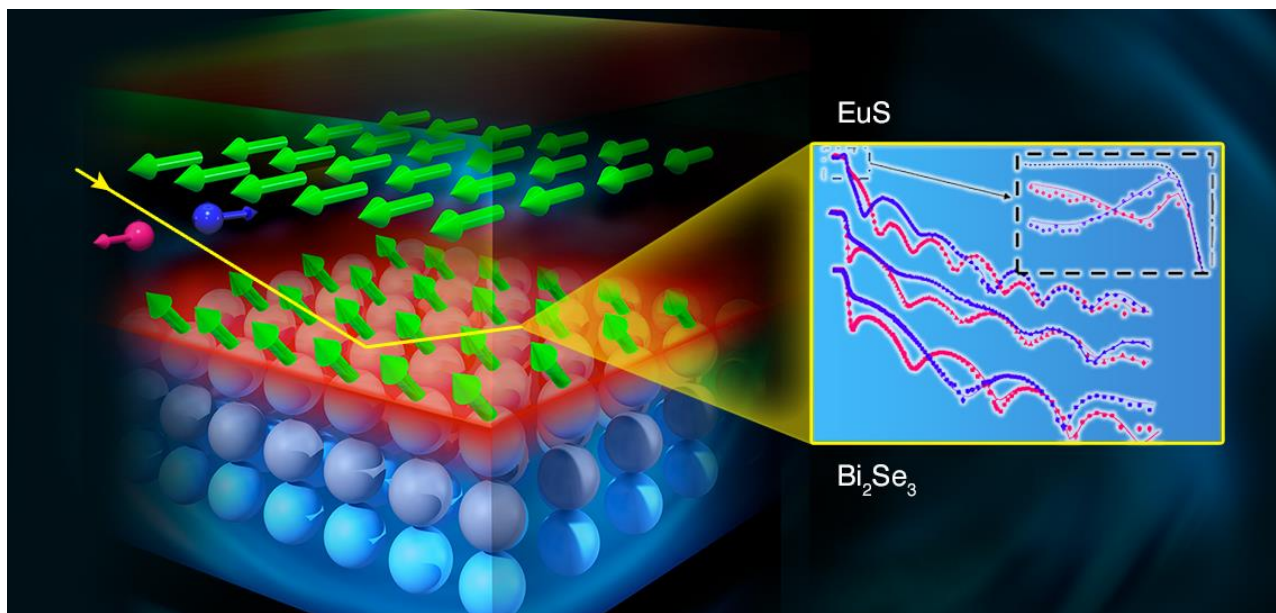
SQUID magnetometry measurement

Magnetization versus Temperature at various perpendicular applied fields



At the interface

- Large S-O interaction, the spin-momentum locking at Dirac surface state creates strong anisotropy and stabilizes the ferromagnetic state!



LETTER *Nature* 17635, May 9, 2016 /doi:10.1038

A high-temperature ferromagnetic topological insulating phase by proximity coupling

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* *Authors contributed equally to this work*

Results

- **PNR - depth resolved vector magnetometry- measures the spatial distribution of magnetization at the buried interfaces of $\text{Bi}_2\text{Se}_3/\text{EuS}$ bilayers and the detailed chemical composition of the heterostructure**
- **The magnetization in the interfacial 2 ML EuS layer has an out-of-plane component.**
- **PNR provides evidence that $\text{Bi}_2\text{Se}_3/\text{EuS}$ heterostructures exhibit proximity-induced interfacial magnetization in 3QL layer of Bi_2Se_3**
- **This effect originates through exchange interaction without structural perturbation at the interface**
- **Magnetism persists up to high T above the T_c of EuS**

Acknowledgements

The work at SNS was supported by the Scientific User Facilities Division, Office of Basic Energy Sciences and DOE