Neutrons and Muons for Topological Quantum Materials: Exploring Magnetic Order

Emily Heppell[1,2,3], Christy J. Kinane[2], Andrew J. Caruana[2], Nina-J. Steinke[4], Xinqi Liu[5], Xufeng Kou[5], Gerrit van der Laan[3], Dirk Backes[3], Sean Langridge[2], and Thorsten Hesjedal[1,2,3]

[1]Clarendon Laboratory, Department of Physics, University of Oxford, Oxford OX1 3PU, United Kingdom
[2]ISIS Neutron and Muon Source, Harwell Science and Innovation Campus, Didcot OX11 0QX, United Kingdom
[3]Diamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, United Kingdom
[4]Institut Laue-Langevin, 71 Avenue des Martyrs, 38042 Grenoble Cedex 9, France
[5]ShanghaiTech Laboratory for Topological Physics, ShanghaiTech University, Shanghai 201210, China

Topological insulators become functional magnetic quantum materials once time-reversal symmetry is broken, enabling phenomena such as the quantum anomalous Hall effect and related chiral transport states. Over the past decade, we have pursued a systematic program to understand how magnetic order can be introduced, controlled, and quantitatively characterized in topological materials grown by molecular beam epitaxy. This contribution reviews our work on magnetic doping and magnetic proximity effects in $(Bi,Sb)_2Te_3$-based systems, with a particular emphasis on depth-resolved and local probes of magnetism. Using a combination of polarized neutron reflectometry, muon spin spectroscopy, and element-specific x-ray techniques, we have established where magnetic order resides, how homogeneous it is, and how it couples across interfaces. We show that magnetic doping often leads to intrinsically inhomogeneous magnetic states, while carefully engineered heterostructures can imprint or enhance magnetism in a controlled manner. Recent results on $CrTe_2/Bi_2Te_3$ heterostructures provide direct evidence for proximity-induced magnetism in a topological insulator without chemical doping. Together, these studies demonstrate how neutrons and muons provide essential insight into magnetic topological materials and guide the design of platforms for quantum and spintronic devices.

***Index Terms*—Topological insulators, magnetic doping, polarized neutron reflectometry, muon-spin rotation.**

I. Introduction

Topological insulators (TIs) become functional magnetic quantum materials once time-reversal symmetry is broken, enabling, e.g., the quantum anomalous Hall effect (QAHE), which has been predicted and experimentally demonstrated in magnetically doped thin films [1-5]. While this approach has enabled both fundamental studies and emerging metrological applications [6], magnetic disorder and spatial inhomogeneity strongly limit device performance [7-9].

An alternative route is to induce magnetism via interfacial exchange coupling in epitaxial heterostructures, avoiding chemical modification of the topological layer. However, distinguishing genuine proximity-induced magnetism from intermixing, roughness, or extrinsic effects remains experimentally challenging and requires probes with depth sensitivity and microscopic resolution [10,11]. All material systems discussed here are realized as epitaxial thin films and heterostructures grown by molecular beam epitaxy [12], which provides the level of control required to achieve atomically sharp interfaces and well-defined layer thicknesses.

The central focus of this contribution is on how neutrons and muons provide decisive insight into these materials. Polarized neutron reflectometry (PNR) yields quantitative magnetic depth profiles across buried interfaces [13], while muon spin rotation and relaxation (μSR) probes local magnetic fields, volume fractions, and fluctuations that are inaccessible to conventional magnetometry [14]. Together, these techniques address where magnetic order resides, how homogeneous it is, and how it evolves with temperature and depth, forming a key part of a broader multi-technique approach that also includes element-specific x-ray spectroscopy and real-space magnetic imaging [5,8-11,15,16].

II. Neutrons and Muons as Quantitative Probes of Magnetic Topological Materials

A central challenge in magnetic topological materials is to determine where magnetic order resides, how homogeneous it is, and how it couples across interfaces, particularly when the relevant magnetism is buried beneath the surface or intrinsically inhomogeneous. These questions cannot be addressed reliably by macroscopic magnetometry alone and often remain ambiguous when using surface-sensitive probes.

PNR and μSR provide complementary, quantitative access to these issues by combining depth resolution with sensitivity to local magnetic order [13,14]. PNR yields the magnetic depth profile across the entire film stack, including buried interfaces, allowing interfacial magnetization, magnetic homogeneity, and proximity-induced moments to be determined directly [11]. In early studies of rare-earth- and transition-metal-doped $(Bi,Sb)_2Te_3$ systems, PNR established that magnetic order can extend uniformly across entire layers rather than being confined to interface regions, highlighting the importance of heterostructure design over simplistic proximity arguments [11]. For example, in heterostructures combining high-moment rare-earth-doped Bi_2Te_3 with Cr-doped $(Sb,Bi)_2Te_3$, PNR revealed homogeneous magnetization profiles across the individual layers, despite strong interfacial magnetic coupling inferred from x-ray spectroscopy. This demonstrated that proximity effects can modify the magnetic ground state of an entire layer rather than producing a narrow interfacial magnetization confined to a few atomic planes. In contrast, PNR has revealed cases where magnetic moments are strongly depth dependent, but only when combined with stringent structural constraints, highlighting both the sensitivity of the method and its vulnerability to artefacts arising from intermixing, roughness, or model bias [10].

Muon spin spectroscopy offers complementary sensitivity to local magnetic fields and magnetic volume fractions on microscopic length scales. In particular, the use of low-energy (slow) muons is essential for thin films and heterostructures, as it enables controlled implantation at defined depths from a few nanometers below the surface into the film interior, providing true depth selectivity [14]. In magnetically doped topological insulators, μSR has demonstrated that the magnetic state is often intrinsically inhomogeneous, consisting of magnetically ordered regions embedded in a paramagnetic background, with slow fluctuations persisting well below the nominal ordering temperature [8,9]. Depth-resolved μSR measurements further showed that this inhomogeneous magnetic state extends throughout the film thickness, with no enhancement at the surface, ruling out simple surface-driven magnetism. These results provided the first direct, microscopic evidence that magnetic percolation and slow fluctuations are intrinsic to magnetically doped $(Bi,Sb)_2Te_3$ thin films and directly impact their transport properties [8,9].

Together, PNR and μSR link depth-resolved magnetic profiles to local magnetic volume fractions and dynamics, enabling a quantitative description of magnetic order in topological materials. In particular, combining PNR and μSR made it possible to disentangle depth-dependent magnetization profiles from volume-averaged magnetic responses, resolving apparent contradictions between magnetometry, transport, and spectroscopic measurements [10,11,16].

These neutron and muon measurements are complemented by element-specific x-ray magnetic spectroscopy and real-space imaging techniques [3-5,10], enabling a consistent interpretation of magnetic order in topological materials. When combined with precise epitaxial growth, this methodology provides a robust framework for identifying and optimizing interfacial magnetism in a wide range of quantum and spintronic heterostructures.

III. Case Study: Proximity-Induced Magnetism

A particularly instructive example is provided by epitaxial $CrTe_2/Bi_2Te_3$ heterostructures [15], which combine a van der Waals ferromagnet with a TI in a structurally compatible geometry [12]. In this system, the ability to achieve atomically sharp interfaces with suppressed interdiffusion is critical, as even minimal structural disorder can mimic or obscure proximity-induced magnetic signals.

The structural integrity of the heterostructures was established using a combination of high-resolution scanning transmission electron microscopy and energy-dispersive x-ray spectroscopy, which reveal atomically abrupt $CrTe_2/Bi_2Te_3$ interfaces with no detectable intermixing across the interface. X-ray reflectometry confirms low interface roughness, below one $CrTe_2$ trilayer, and well-defined layer thicknesses, providing the structural constraints required for quantitative magnetic modelling.

Magnetic characterization combines polarized neutron reflectometry with element-specific x-ray magnetic circular dichroism (XMCD). PNR measurements acquired above and below the Curie temperature were simultaneously fitted using Bayesian analysis. The nuclear scattering length density profile was constrained to be common across temperatures, while the magnetic profile was allowed to vary, thereby reducing parameter uncertainties and model degeneracy. This approach, in combination with constraints from complementary techniques, significantly reduces model ambiguity and allows statistically robust discrimination between competing magnetic depth profiles. The resulting magnetic depth profile reveals an enhanced magnetic moment on the $CrTe_2$ side of the interface, increasing from significantly less than 1 μ_B per Cr in the bulk to >1 μ_B at the interface, accompanied by a finite, anti-aligned magnetic moment extending ~1 nm into the Bi_2Te_3 layer. This induced moment cannot be explained by roughness, diffusion, or model artefacts. XMCD measurements independently confirm robust magnetization in the $CrTe_2$ layer and support the interfacial origin of the observed magnetic enhancement.

This study demonstrates that, through the combination of exceptional epitaxial quality and rigorous statistical analysis, PNR can provide unambiguous, depth-resolved evidence for proximity-induced magnetism in TI heterostructures, resolving ambiguities that have persisted in earlier studies.

References

[1] R. Yu *et al.*, "Quantized anomalous Hall effect in magnetic topological insulators," *Science*, vol. 329, pp. 61-64, 2010.

[2] C.-Z. Chang *et al.*, "Experimental observation of the quantum anomalous Hall effect in a magnetic topological insulator," *Science*, vol. 340, pp. 167-170, 2013.

[3] C.-Z. Chang, C. Liu, and A. H. MacDonald, "Colloquium: Quantum anomalous Hall effect," *Rev. Mod. Phys.*, vol. 95, 011002, 2023.

[4] Y. Tokura, K. Yasuda, and A. Tsukazaki, "Magnetic topological insulators," *Nat. Rev. Phys.*, vol. 1, pp. 126-143, 2019.

[5] J. Liu and T. Hesjedal, "Magnetic topological insulator heterostructures: A review," *Adv. Mater.*, vol. 35, 2102427, 2023.

[6] N. J. Huang *et al.*, "Quantum anomalous Hall effect for metrology," *Appl. Phys. Lett.*, vol. 126, 040501, 2025.

[7] T. Hesjedal, "Rare earth doping of topological insulators: A brief review of thin film and heterostructure systems," *Phys. Status Solidi A*, vol. 216, 1800726, 2019.

[8] L. B. Duffy *et al.*, "Microscopic effects of Dy doping in the topological insulator Bi_2Te_3," *Phys. Rev. B*, vol. 97, 174427, 2018.

[9] N.-J. Steinke *et al.*, "Magnetic correlations in the magnetic topological insulator $(Cr,Sb)_2Te_3$," *Phys. Rev. B*, vol. 106, 224425, 2022.

[10] G. Awana *et al.*, "Critical analysis of proximity-induced magnetism in $MnTe/Bi_2Te_3$ heterostructures," *Phys. Rev. Mater.*, vol. 6, 053402, 2022.

[11] L. B. Duffy *et al.*, "Magnetic profile of proximity-coupled $(Dy,Bi)_2Te_3$/$(Cr,Sb)_2Te_3$ topological insulator heterostructures," *Phys. Rev. B*, vol. 100, 054402, 2019.

[12] L. He, X. Kou, K. L. Wang, "Review of 3D topological insulator thin-film growth by molecular beam epitaxy and potential applications," *Phys. Status Solidi RRL*, vol. 7, pp. 50-63, 2013.

[13] G. L. Causer, L. Guasco, O. Paull, D. Cortie, "Topical Review of Quantum Materials and Heterostructures Studied by Polarized Neutron Reflectometry," *Phys. Status Solidi RRL*, vol. 17, 2200421, 2023.

[14] A. D. Hillier, S. J. Blundell, I. McKenzie *et al.*, "Muon spin spectroscopy," *Nat. Rev. Methods Primers* **2**, 4, 2022.

[15] X. Liu *et al.*, "Wafer-scale epitaxial growth of the thickness-controllable van der Waals ferromagnet $CrTe_2$ for reliable magnetic memory applications," *Adv. Funct. Mater.*, vol. 33, 2304454, 2023.

[16] A. I. Figueroa *et al.*, "Magnetic order in 3D topological insulators—Wishful thinking or gateway to emergent quantum effects?," *Appl. Phys. Lett.*, vol. 117, 150502, 2020.