



Perspective: Probing 2-D magnetic structures in a 3-D world

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Perspective: Probing 2-D magnetic structures in a 3-D world

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Magnetic interfaces have been identified as promising systems upon which to base next-generation spintronic devices. In these nearly two-dimensional systems, deviations from bulk electronic structure and competition between nearly degenerate magnetic ground states allow the stabilization of widely tunable emergent properties. However, ever smaller length scales pose new challenges which must be overcome in order to understand and control magnetic properties at the atomic level. Using recent examples in oxide heterostructures and topological insulators, we discuss how combining techniques such as neutron scattering, X-ray scattering, X-ray spectroscopy, and transmission electron microscopy enables the probing of magnetism on the Angstrom scale. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [<http://dx.doi.org/10.1063/1.4944630>]

Among the most important goals of nanoscale magnetism research is the development of new materials and structures to achieve reversible, non-volatile control of magnetism while minimizing energy expenditure. Meeting this goal will enable the emergence of a new class of spintronic devices, which use the magnetic moments of electrons to store and transmit information rather than relying on the transport of electronic charge. These spintronic devices have the potential to revolutionize computing and data storage, and in fact are already responsible for enormous increases in information storage capacity. However, the development of many next generation systems is reliant on transitioning to new, non-traditional mechanisms which will enable precise control of nanoscale magnetic interactions.

In particular, interfaces within magnetic heterostructures give rise to competing interactions and complex energy landscapes useful for engineering and controlling magnetism. However, the characterization of magnetic interfaces poses unique challenges not present in the study of either bulk materials or thin films. Sub-nanometer length scales require probes with significantly higher spatial resolution, while the buried nature of these structures favors techniques which can penetrate capping layers. Furthermore, the underlying cause of a given effect is often very difficult to isolate, as the signatures of roughness, intermixing, and even charge transfer may be very similar. Finally, the measurement of magnetic moments originating within layers less than a nanometer thick is fraught with potential for contamination by extrinsic magnetic signals. Given the importance of separating intrinsic effects from extrinsic artifacts, element-specific and depth-resolved techniques must play an increasingly important role in these studies.

Examples of depth resolved probes include polarized neutron reflectometry (PNR) and X-ray resonant magnetic scattering (XRMS), which exploit the magnetic sensitivity of reflected neutrons and X-rays to probe the structural and magnetic depth profile of heterostructures.¹ PNR is highly signal limited but sensitive exclusively to the chemical composition and net magnetization.²⁻⁸ Combination and co-refinement of PNR data with XRMS measurements, which is sensitive to the electronic structure and may cover a wider angular range due to high synchrotron photon flux, can provide improved constraints for interpreting both data sets.⁹⁻²¹ Depth or surface sensitivity may also be achieved through magneto-optic Kerr effect (MOKE) microscopy, which allows magnetization mapping across a film surface, and through electron transport measurements which exploit the



anomalous Hall effect to probe magnetization near the conducting region of a film.^{22–25} Hall effect measurements are particularly useful in cases where only a specific region of the film, such as the interface, is conductive.

Element specific techniques include X-ray absorption spectroscopy (XAS), and X-ray magnetic circular dichroism (XMCD), which probe the electronic structure by gathering X-ray absorption spectra at elemental absorption edges.^{26–30} XAS and XMCD can detect slight variations in electronic and magnetic structure, but achieve the highest sensitivity and resolution when probing only the top few nanometers of a structure.²⁷ Combining XAS/XMCD with highly penetrative probes such as PNR or XRMS preserves the chemical sensitivity, provides additional constraints for model refinement, and probes the entire structure. Performing transmission electron microscopy (STEM) with electron energy loss spectroscopy (EELS) allows the gathering of high-resolution, element specific information in real space, but is typically limited to examining one nanoscale region at a time.^{31–37} Thus, the careful and synergistic application of several specialized techniques can overcome individual shortcomings and allow us to formulate a complete picture of the interface in question. Below, we will discuss how several recent advances in magnetic materials have used a combination of X-ray, neutron, and TEM techniques to demonstrate unprecedented control of magnetism at interfaces, focusing on examples based on complex oxides, magnetoelectric oxide heterostructures, and magnetic topological insulators.

The wide range of ground states and tunable properties in perovskite oxide heterostructures goes hand in hand with interfacial physics which is exceedingly difficult to unravel.^{38–44} Perovskite interfaces routinely exhibit discontinuities in oxidation state, band structure, magnetic order, surface polarity, and structure, each of which may readily influence the interfacial magnetism to stabilize highly tunable emergent states not found in the bulk.^{38–53} However, these interfaces can appear very similar and separating intermixing from charge transfer or exchange interactions from structural distortions crossing the interface is extremely challenging. In $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{Eu}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ superlattices, Moon *et al.* combined synchrotron X-ray diffraction of structural distortion peaks and polarized neutron reflectometry to show that the tilt and rotation of oxygen octahedra propagate 1-2 unit cells across the interface to stabilize a new magnetic state distinct from that of an alloy.⁵⁴ In superlattices composed of CaMnO_3 , an antiferromagnetic insulator, and the paramagnetic metals CaRuO_3 or LaNiO_3 , an emergent ferromagnetic moment is attributed to interfacial charge transfer.^{55,56} However, these materials form a ferromagnetic metal when alloyed, and separating the two effects proved particularly challenging. As illustrated in Figure 1, element specific information from X-ray absorption spectroscopy on the magnetic properties of Ru, Ni, and Mn was crucial to the refinement of resonant X-ray scattering and polarized neutron reflectometry models which showed the effect to be interfacial, while EELS measurements showed a corresponding change in electronic structure at the interface.^{57–59}

Once the charge transfer mechanism was confirmed, a highly tunable interface was revealed and magnetoelectricity has recently been demonstrated through modulation of the charge transfer magnitude.⁶⁰ In such interfacial magnetoelectric systems, tracking small changes in interfacial

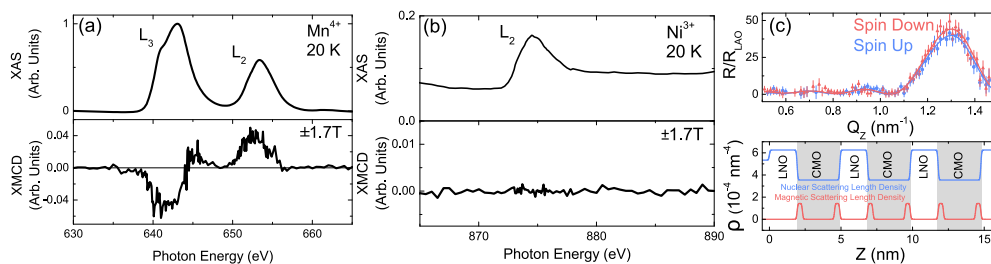


FIG. 1. XAS and XMCD of the (a) Mn and (b) Ni L-edges alongside (c) PNR of $\text{CaMnO}_3/\text{LaNiO}_3$ superlattices. Knowing from the XMCD that the Mn was magnetic, while the Ni was not, allowed the exclusion of all models with magnetic Ni when fitting the PNR data.⁵⁹ Thus, a unique fit emerged in which the CaMnO_3 magnetization was confined to a single unit cell at the interface. Adapted with permission from Grutter *et al.*, Phys. Rev. Lett. **111**, 087202 (2013). Copyright 2013 American Physical Society.

valence state or oxygen content with the application of an electric field is as challenging as it is critical. In $\text{CaRuO}_3/\text{CaMnO}_3$ heterostructures, PNR measurements enabled the detection of extremely small changes in magnetization at the interface. Specifically, with the application of a modest electric field, a change in magnetization from $1 \mu_B/\text{Mn}$ to $3 \mu_B/\text{Mn}$ was induced within a single unit cell of CaMnO_3 at the interface.⁶⁰ Such a variation is well within the accepted range for contamination effects or magnetometry artifacts, but may be reliably extracted thanks to the insensitivity of PNR to contamination or electric fields. As probes that are insensitive to electric fields and surface contamination, PNR and XRMS in particular are ideally placed to sidestep the artifacts that plague attempts to detect subtle magnetoelectric coupling effects at interfaces. However, X-ray absorption techniques are often equally valuable. In the BaTiO_3/Fe system, Radaelli *et al.* relied on element specific XMCD and EELS techniques to demonstrate control of the interfacial Fe magnetization through switching of the BaTiO_3 ferroelectric polarization.⁶¹ Outside of perovskite systems, novel magnetoelectric interfaces have also been recently demonstrated at oxide/metal interfaces such as between GdO_x and Co.^{62–67} Both Bauer *et al.* and Bi *et al.* have reported reversible voltage control of interfacial oxygen ion migration from GdO_x , to an ultrathin layer of Co.^{62–64} In this case, changes in oxidation state and magnetism with electric field are tracked using MOKE microscopy in concert with XAS and XMCD.

Magnetic topological insulators are a new class of two-dimensional magnetic materials currently pushing the limits of magnetic characterization capability.^{69–73} Motivated by the opportunity to realize novel quantum states and devices based on topological insulators, much recent effort has been focused on inducing ferromagnetic states through either doping or interfacial proximity effects.^{68,74–81} Interest has been further stimulated by recent predictions for electric field control of magnetic surface states in systems such as Cr or V-doped $(\text{Bi},\text{Sb})_2\text{Te}_3$.^{82,83} Due to the surface nature of these predicted effects, extremely sensitive spatially resolved magnetic characterization must be performed.^{74,83} For example, proximity induced ferromagnetism was observed by Li *et al.* at the interface between ferromagnetic EuS and the topological insulator $\text{Sb}_{2-x}\text{V}_x\text{Te}_3$. An enhanced magnetization in the interfacial $\text{Sb}_{2-x}\text{V}_x\text{Te}_3$ was detected primarily through extremely subtle variations in polarized neutron reflectometry spectra.⁷⁴ In this case, the demonstration of a small interfacial magnetic moment in the topological insulator relied on a complete picture of the system which enabled highly constrained modeling.⁷⁴

Whether or not electric fields are applied, the ability to explicitly separate surface and interface effects from bulk behaviors will remain critical to the study of 2-D magnetic systems. With continual improvements in XRMS modeling and enormous imminent increases in neutron flux for PNR, experiments which are currently out of reach will soon be possible. Higher signal to noise ratios will enable the extension of reflectivity techniques to higher scattering angles, improving real space resolution. Just as significant, however, is the recent emergence of electron magnetic circular dichroism techniques (EMCD) based on TEM/EELS measurements.^{84–87} EMCD provides high-resolution, real space, element specific chemical, electronic, and magnetic information simultaneously. With ever increasing resolution and the potential for integration with STEM techniques, we may hope for element specific magnetic characterization with angstrom-level real space resolution in the near future.^{84–87}

As the search for new spintronic device materials continues to shift to single unit cell length scales we can expect that these issues will become ever more important, driving further innovation in nanoscale magnetic characterization. By approaching the study of interfacial magnetic phenomena with a wide variety of nontraditional characterization techniques, we may reduce our sensitivity to magnetic contamination and achieve a complete picture of the magnetism and chemistry at heterostructure interfaces, enabling us to isolate key control variables with which to tune emergent interfacial magnetic properties.

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