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Towards Dissipation-less Conduction in Oxide Topological Insulators

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14. ABSTRACT

In this program, we address the development of heterostructures with emergent topological behavior based on complex oxide materials. More specifically, we have incorporated epitaxial spinel ferrite oxide thin films into heterostructures in order to stabilize (i) a magnetic topological insulator via magnetic proximity effect with a known topological insulator and (ii) topological spin textures via interfacial Dzyaloshinkii-Moriya interactions with a strongly spin-orbit coupled metal. Through this program, we have been able to enumerate the critical factors required for establishing a magnetic proximity effect between oxide and chalcogenide layers, and in turn, a magnetic topological insulator. In addition, we have also discovered signatures of topological spin textures in ferrite/Pt bilayers via Hall effect measurements.

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Report Abstract

In this program, we address the development of heterostructures with emergent topological behavior based on complex oxide materials. More specifically, we have incorporated epitaxial spinel ferrite oxide thin films into heterostructures in order to stabilize (i) a magnetic topological insulator via magnetic proximity effect with a known topological insulator and (ii) topological spin textures via interfacial Dzyaloshinkii-Moriya interactions with a strongly spin-orbit coupled metal. Through this program, we have been able to enumerate the critical factors required for establishing a magnetic proximity effect between oxide and chalcogenide layers, and in turn, a magnetic topological insulator. In addition, we have also discovered signatures of topological spin textures in ferrite/Pt bilayers via Hall effect measurements.

Accomplishments

Research Objectives. The main objective of the proposed program is (i) to develop a new class of emergent topological materials based on complex oxide thin films and interfaces and (ii) to fabricate dissipation-less channels and prototypical devices for information processing at elevated temperatures. The approach that we have taken is the following: (i) stabilizing a magnetic ground state in a topological insulator via the magnetic proximity effect; (ii) exploring topological spin textures in ultrathin ferrite thin films; (iii) low dimensional oxide interfaces as a possible route to a topological ground state.

Details of Accomplishments

Major Activities

The major research activities are comprised of the synthesis and characterization of complex oxide thin films and heterostructures that exhibit topological phenomena. The stabilization of topological ground states in complex oxides has been theoretically predicted but not experimentally verified. Since the stabilization of a bulk topological ground state in complex oxides appears to be elusive, we have taken the approach of exploiting emergent properties in oxide-based heterostructures as an alternative approach. More specifically, our approach has been threefold: (i) induce emergent topological states in known topological materials; (ii) stabilize topological textures and defects in oxide-based systems; (iii) identify low dimensional oxide interfaces with high spin-orbit coupling as candidates for emergent topological phenomena.

Educational aspects of the program include the training of graduate students as well as a continuing partnership with local high schools in San Mateo and San Jose (CA) in the form of short summer internship programs.

Specific Objectives

Our objective has been to explore the emergence of topological phenomena in different types of spinel ferrite based systems. More specifically, we explored heterostructures comprised of epitaxial ferrite films and known topological insulators and those comprised of epitaxial ferrite films and strongly spin-orbit coupled metals. The spinel ferrite layer induces magnetism in a topological insulator by proximity or interfacial Dzyaloshinkii-Moriya interactions in an adjacent heavy metal. In both cases, the role of the interface in mediating nearest and next-nearest neighbor magnetic interactions highlights the importance of the structural and chemical integrity at the interface.

Significant Results

Magnetic Proximity Effect in Topological Insulator/ Ferromagnetic Insulator Bilayers. A promising route to dissipation-less conduction is the stabilization of chiral edge states in magnetic topological insulators in the form of the quantum anomalous Hall effect (QAHE), where the longitudinal resistance drops to zero as the Hall resistance approaches the conductance quantum. Although dissipation-less chiral edge states associated with the QAHE have been observed in bulk topological materials, they are only stabilized at extremely low temperatures. The suppression of such behavior to low temperatures has been attributed to disorder induced by magnetically doping known topological insulators. More recently, magnetic van der Waal's materials have exhibited similar behavior but still only at extremely low temperatures. In an effort to elevate the temperature at which the QAHE can be observed, a number of researchers have tried to induce a magnetism in the surface states of known topological insulators (TIs) by a magnetic proximity effect from an adjacent ferromagnetic insulator with a magnetic ordering temperature well above room temperature. By making TIs magnetic, non-dissipative helical edge states in a TI are transformed to chiral edge states as time-reversal symmetry is broken and a gap is opened at the Dirac point.

To this end, we synthesized heterostructures comprised of $Mg(Al,Fe)_2O_4$ and Bi_2Se_3 films where the chalcogenide films were grown by molecular beam epitaxy at low temperatures on top of epitaxial ferrite films (grown by pulsed laser deposition). We discovered evidence that suggested a magnetic proximity effect was induced at this $Mg(Al,Fe)_2O_4/Bi_2Se_3$ interface. Hall effect measurements showed a nonlinear magnetic field dependence that could be attributed to proximity -induced ferromagentism. Magnetic profiling by polarized neutron reflectometry (PNR) measurements indicated an asymmetry in the non-spin-flip reflectivity spectra, suggesting a magnetic proximity effect with a sharp interface. Many previous studies of ferromagnetic insulators with known topological insulator have asserted the presence of a magentic proximity effect with one or both of these pieces of evidence. However both non-linear Hall effect and PNR measurments can be interpreted in alternative ways. Non-linear Hall effect measurements may be attributed to multi-band conduction and, in fact, a single Bi_2Se_3 layer can also show similar nonlinear magnetic field dependence. Asymmetry in the non-spin-flip reflectivity PNR spectra can be modeled as a magnetic proximity effect with a sharp interface but also as no magnetic proximity effect with a rough surface or no magnetic proximity effect with an interfacial layer. We performed both x-ray reflectivity (XRR) and transmission electron microscopy(TEM)/ x-ray dispersive spectroscopy (XDS) to distinguish between these interpretations and found that XRR indicates the presence of an interfacial layer and TEM/XDS indicates an interdiffused layer (Figure 1). Therefore a comprehensive set of structural, magnetic and transport measurements are critical to determining whether there is truly a magnetic proximity effect at a given interface.

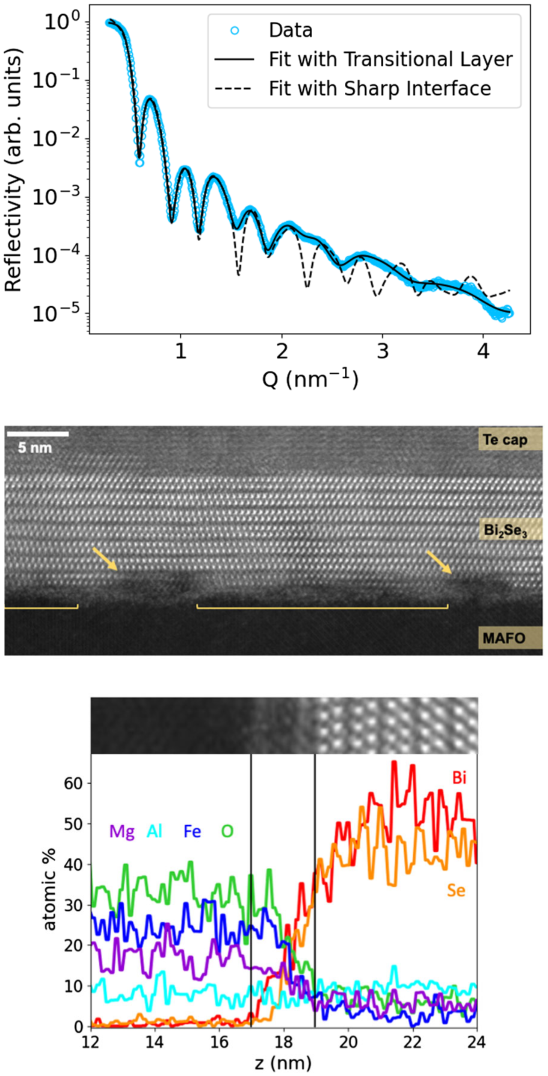

Figure 1. (top) X-ray reflectivity of a $Mg(Al,Fe)_2O_4/Bi_2Se_3$ bilayer. The solid line indicates the best fit including an interfacial layer between the two materials, while the dashed line shows the best fit assuming no interfacial layer; (middle) Annular dark field scanning transmission electron microscopy image showing disorder at the ferrite/TI interface. Brackets and arrows indicate disordered interfacial regions with height of ~1 and ~2 nm, respectively; (bottom) EDX elemental analysis, showing an image of the region analyzed and an elemental line profile.

Creating MI-TI heterostructures with a high-quality interface is a big challenge, but we have established that there are several possible solutions. First, in situ heterostructure growth with all-chalcogenide films have shown atomically sharp interfaces, due to matching crystal structures and avoiding hydrocarbon contamination. Ex-situ growth can be successful with magnetic films that are robust to high temperature annealing and can grow epitaxially on standard substrates for TIs like Al_2O_3. In the absence of these options, exfoliating and transferring thin films of TIs may be preferable to attempting direct growth for a high-quality

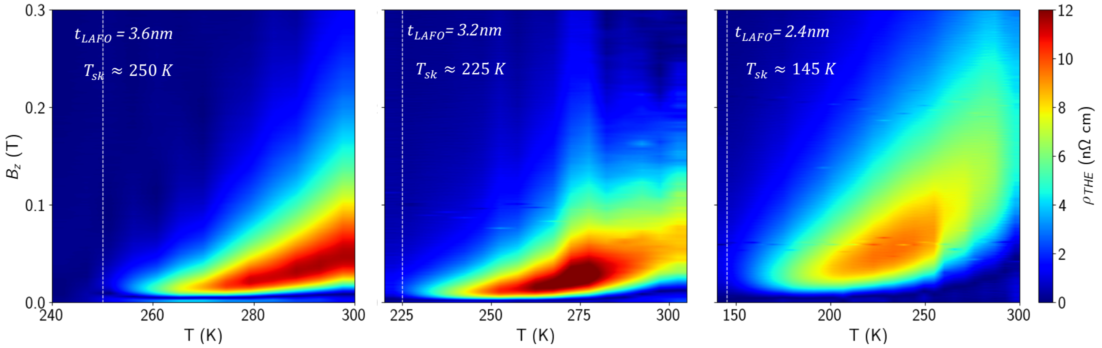

Figure 2. Topological Hall effect measurements of $Li_{0.5}(Al,Fe)_{2.5}O_4$/Pt bilayers show a peak in Hall resistivity at low magnetic fields. For bilayers with 2nm Pt and varying thicknesses, and hence magnetic anisotropy, of $Li_{0.5}(Al,Fe)_{2.5}O_4$, we find that the topological Hall effect shifts to higher temperatures

interface. Notably, the sole observation of a proximity induced QAHE was in an in-situ grown chalcogenide trilayer where an atomically sharp interface was verified with TEM.

Topological Spin Textures in Epitaxial Ferrite Heterostructures. We have discovered evidence for topological spin textures at oxide-based interfaces. Materials with bulk Dzyaloshinkii-Moriya interactions due to crystal symmetry breaking or with interfacial Dzyaloshinkii-Moriya interactions due to crystal symmetry breaking at interfaces have been identified as candidates for the stabilization of topological spin textures. We have taken the approach of putting together a high spin orbit coupling material with a perpendicular magnetic anisotropy material. We have combined low loss $Li_{0.5}AlFe_{1.5}O_4$ films, grown on $MgGa_2O_4$ substrates, with perpedicular magnetic anisotropy– a recent discovery in our group– and ultra-thin Pt layers. Hall effect measurements of these bilayers show a contribution to the transverse voltage from fields emerging from topological spin textures. This topological Hall effect (THE) feature manifests itself at low magnetic fields and can be plotted as a function of magnetic field and temperature as shown in Figure 2. The onset temperature of the THE shifts to higher temperatures with a reduction in perpendicular magnetic anisotropy.

Quantum Oscillations in Oxide Interfaces. Interfaces of complex oxides host a rich variety of phenomena and exotic phases not found in their bulk counterparts. The $LaTiO_3/SrTiO_3$ (LTO/STO) system is particularly interesting due to the presence of a high mobility interfacial 2DEG as well as a conductive LTO thin film layer on STO. Moreover reduction of STO during the growth process may also contribute to conduction. To date, the extent of the

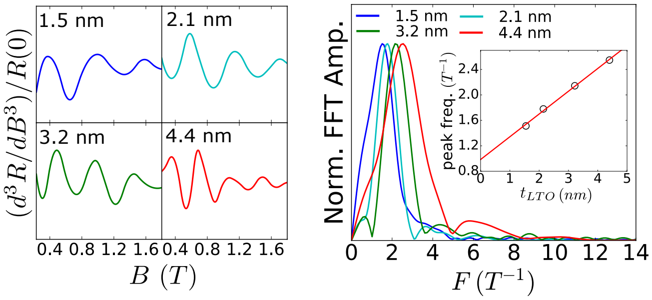

Figure 3. The third derivative of the longitudinal resistance for different LTO thicknesses and their fast Fourier transform allows us to estimate the mean free path of charge carriers parallel to the magnetic field.

conducting region has not been quantified. We have quantified the 2DEG thickness in LTO/STO through Sondheimer oscillations in the longitudinal magnetoresistace. By analyzing the LTO thickness dependence of these oscillations we estimate that the 2DEG penetrates into the STO substrate by approximately 2.8 nm, indicating that the metallic bulk LTO is not the sole contributor to metallicity. We have provided the first quantitative estimate of the STO conduction depth in LTO/STO using Sondheimer oscillations and paves the way for future studies of oxide heterostructures with multiple channels of conduction.

Papers

1. Di Yi, Houari Amari, Purnima Balakrishnan, Christoph Klewe, Alpha N'Diaye, Padraic Shafer, Nigel Browning, Yuri Suzuki, "Enhanced Interface-Driven Perpendicular Magnetic Anisotropy by Symmetry Control in Oxide Superlattices," *Physical Review Applied* **15** 024001 (2020).
2. Peng Li, Chong Bi, Jacob J, Wisser, Lauren J. Riddiford, Xiao-Qi Sun, Arturas Vaillionis, Michael J. Veit, Aaron Altman, Xiang Li, Mahendra DC, Shan X. Wang, Y. Suzuki, Satoru Emori, "Charge-to-Spin Conversion Mechanisms in Epitaxial Pt probed by Spin-Orbit Torques in a Magnetic Insulator," *Physical Review Materials* **5** 064404 (2021).
3. Margaret Kane, Arturas Vailionis, Lauren Riddiford, Apurva Mehta, Alpha N'Diaye, Elke Arenholz, Yuri Suzuki, "Emergent Long Range Magnetic Order in (111)-Oriented Ultrathin Lanthanum Nickelate Films," *npj Quantum Materials* **6** 44 (2021).
4. Xiang Li, Peng Li, Vincent D.-H. Hou, Mahendra DC, Chih-Hung Nien, Fen Xue, Di Yi, Chong Bi, Chien-Min Lee, Shy-Jay Lin, Wilman Tsai, Yuri Suzuki, Shan X, Wang, "Large and Robust Charge-to-Spin Conversion in Sputtered Conductive WTe_x with Disorder," *Matter* **4** 1639 (2021).
5. Lauren Riddiford, Alexander Grutter, Timothy Pillsbury, Max Stanley, Nitin Samarth, Yuri Suzuki, "Understanding Signatures of Emergent Magnetism in Topological Insulator/Ferrite Bilayers," *Physical Review Letters* **128** 126802 (2022).
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7. C. Klewe, P. Shafer, J.E. Shoup, C. Kons, Y. Pogoryelov, R. Knut, B.A. Gray, H.-M. Jeon, B.M. Howe, O. Karis, Y. Suzuki, E. Arenholz, D.A. Arena, S. Emori, "Observation of Coherently Coupled Cation Spin Dynamics in an Insulating Ferrimagnetic Oxide," *Applied Physics Letters* **122** 132401 (2023).
8. R. Li, L. J. Riddiford, Y. Chai, M. Dai, H. Zhong, B. Li, P. Li, D. Yi, D. A. Broadway, A. Dubois, P. Maletinsky, J. Hu Y. Suzuki, D. C. Ralph, and T. Nan, "A puzzling insensitivity of magnon spin diffusion to the presence of 180° domain walls," *Nature Comm.* **14** 2393 (2023).
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10. Xin Yu Zheng, Sanyum Channa, Lauren Riddiford, Jacob J. Wisser, Alpha T. N'Diaye, Egecan Cogulu, Haowen Ren, Zbigniew Galazka, Andrew D. Kent, Yuri Suzuki, "Ultra-thin lithium

aluminate spinel ferrite films with perpendicular magnetic anisotropy and low damping," *Nature Communications* **14** 4918 (2023).

11. Daisy O'Mahoney, Sanyum Channa, Xin Yi Zheng, Arturas Vailionis, Padraic Shafer, Alpha N'Diaye, Chriistoph Klewe, Yuri Suzuki, "Aluminum substitution in low damping epitaxial lithium ferrite films," *Applied Physics Letters* **123** 172405 (2023).
12. Xin Yu Zheng, Michael J. Veit, Yuri Suzuki, "Depth of the Two Dimensional Electron Gas in $LaTiO_3/SrTiO_3$ Heterostructures Determines with Sondheimer Oscillations," in preparation (2023).

Theses/Dissertations

- Jacob Wisser, "Magnetization and spin dynamics in complex oxide heterostructures," Ph.D. dissertation, Stanford University (2022).
- Lauren Riddiford, "Interfacial Spin Wave Phenomena in Thin Film Oxide Heterostructures," Ph.D. dissertation, Stanford University (2022).

Talks

- "Disentangling the magnetic proximity effect in topological insulators with $Mg(Al,Fe)_2O_4/Bi_2Se_3$ Thin Films," Lauren Riddiford, Alexander Grutter, Timothy S. Pillsbury, Nitin Samarth, Yuri Suzuki, 2021 American Physical Society March Meeting, March 2021, Denver, CO.
- "Spin Current Generation and Spin-Orbit Torque Switching in Perpendicularly Magnetized Insulating $Ni_{0.65}Zn_{0.35}Fe_{1.2}Al_{0.8}O_4$ Films," Sanyum Channa, Zbigniew Galazka, Satoru Emori, Matthew Gray, Yuri Suzuki, 2021 American Physical Society March Meeting, March 2021, Denver, CO.
- "Room Temperature Topological Hall Effect in Insulating Ferrite/Heavy Metal Bilayers," Sanyum Channa, Xin Yu Zheng, Zbigniew Galazka, Lauren Riddiford, Egecan Cogolu, Haowen Ren, Andrew Kent, Yuri Suzuki, 2022 APS March Meeting, Chicago, IL (2022).
- "Anisotropic spin-orbit torques in thin film bilayers of $Li_{0.5}(Al,Fe)_{2.5}O_4$ and Pt studied with spin-torque ferromagnetic resonance," Lauren Riddiford, Charles Zheng, Sanyum Channa, Zbigniew Galazka, and Yuri Suzuki, 2022 APS March Meeting, Chicago, IL (2022).
- "Temperature Dependence of Spin-Orbit Torques Exerted by a 2DEG in $CoFeB/LaTiO_3/SrTiO_3$ Thin-Film Heterostructures," Lauren Riddiford, Charles Zheng, Fen Xue, Shan X. Wang, Yuri Suzuki, 2022 Spring Materials Research Society Meeting, Honolulu, HI (2022).
- "Measuring spin torque efficiencies in Pt/ferromagnetic insulator bilayers with spin-torque ferromagnetic resonance," Sanyum Channa, Xin Yu Zheng, Zbigniew Galazka, Haowen Ren, Andrew Kent, Yuri Suzuki, 2023 APS March Meeting, Las Vegas (2023).

How were the results disseminated to communities of interest?

We have disseminated our research results to the scientific community via presentations at national conferences as well as in seminars and publications. We have had summer internship programs with two local high schools in San Mateo and San Jose, CA. The graduate students, funded with this program, have actively been involved in the internship program and have been important role models for the high school internship students who hail from an all-girls high school in San Jose and a co-educational high school in San Mateo.

Impacts

Development of the principal discipline(s) of the project

Our efforts have focused on developing complex oxide materials with topological ground states or properties. More specifically the stabilization of a magnetic topological insulator via proximity induced magnetism in a known chalcogenide topological insulator and topological magnetic texture in a system with interfacial Dzyaloshinkii-Moriya interactions have been the focus. These efforts have significant impact in the areas of materials science and condensed matter physics but also promising for memory and sensor applications in spin-wave spintronics. They demonstrate the power of spin waves in developing a new platform for energy efficient microelectronics.

Development of other discipline(s) of the project

Complex oxide materials with topological properties provide advanced materials for a new type of spin-wave-based microelectronics.

Describe the impact in this reporting period on the development of human resources

This program has provided opportunities for training and education of graduate students from Stanford during the school year plus summer as well as high school students from nearby schools during the summer.

Describe the impact on teaching and educational experiences

The project has provided training of one full graduate student as well as part time graduate students. Our project results have provided considerable impact on the development of young scientists in materials and physics research. In addition, the grant has provided the resources to develop a high school internship program with local high schools (San Jose and San Mateo, CA). This high school internship program is a follow-on to my previous successful effort involving over thirty local high school students (Oakland, CA).

Describe the impact in this reporting period on physical, institutional, and information resources that form infrastructure.

In terms of physical resources, a significant part of this research program is the continued development and improvement of physical vapor deposition systems for thin film growth in my laboratory. In terms of institutional resources, a significant part of this research program is the development of processing and fabrication techniques for complex oxide materials at the Stanford facilities that include my laboratory as well as the shared facilities. Stanford provides the program with user characterization facilities, including x-ray diffraction, electron microscopy and electron microprobe etc. as well as a nanofabrication facility. In terms of information resources, this program has provided research results through our publications, presentations and website.

Impact on society beyond science and technology:
On a more global scale, this project has provided the infrastructure for me to formulate a magnetic materials program that has reached high school students.

Changes

None