## Physics of 2D Systems (Phys2D)

***External User Publications (Phys2D)***

H. Zhang, C.Q. Xu, **S.H. Lee**, **Z. Q. Mao**, X. Ke, "Thermal and Thermoelectric Properties of an Antiferromagnetic Topological Insulator $MnBi_2Te_4$," *Physical Review B* 105, 184411 (2022). 10.1103/PhysRevB.105.184411.

In this paper, electronic, thermal, and thermoelectric transport studies of $MnBi_2Te_4$ are reported The temperature and magnetic field dependence of its resistivity, thermal conductivity, and Seebeck coefficient indicate strong coupling between charge, lattice, and spin degrees of freedom in this system. Furthermore, $MnBi_2Te_4$ exhibits a large anomalous Nernst signal, which is associated with nonzero Berry curvature of the field-induced canted antiferromagnetic state. Bulk Crystal are grown using a 2DCC muffle furnace.

- External User Project S0074

Y. Lin, M. Huber, S. Rajpurohit, Y. Zhu, K.M. Siddiqui, D.H. Eilbott, L. Moreschini, P. Ai, J.D. Denlinger, **Z. Mao**, L.Z. Tan, A. Lanzara, "Evidence of Nested Quasi-one-dimensional Fermi Surface and Decoupled Charge-lattice Orders in Layered $TaTe_2$," *Physical Review Research* 4 (2), L022009 (2022). 10.1103/PhysRevResearch.4.L022009.

Results suggest that TaTe2 manifests intrinsic mixed dimensionality between its electronic and lattice structure and that the CDW-like phase transition is likely governed by multiple mechanisms. This work provides routes for forging unconventional CDW phases and charge-lattice entanglement that would otherwise not be available in materials with fixed dimensionality.

- External User Project S0076 (National Lab)

L.J. Riddiford, A.J. Grutter, T. Pillsbury, M. Stanley, **D.R. Hickey**, P. Li, **N. Alem**, **N. Samarth**, Y. Suzuki, "Understanding Signatures of Emergent Magnetism in Topological Insulator/Ferrite Bilayers," *Physical Review Letters* 128 (12), 126802 (2022). 10.1103/PhysRevLett.128.126802.

Magnetic insulator-topological insulator heterostructures have been studied in search of chiral edge states via proximity induced magnetism in the topological insulator, but these states have been elusive. This study identified $MgAl_{0.5}Fe_{1.5}O_4$=$Bi_2Se_3$ bilayers for a possible magnetic proximity effect. The results provide a strategy via correlation of microstructure with magnetic data to confirm a magnetic proximity effect. Materials were grown using 2DCC equipment MBE1.

- External User Project S0030

Z. Pan, **S.H. Lee**, K. Wang, **Z. Mao**, D. Li, "Elastic stiffening induces one-dimensional phonons in thin $Ta_2Se_3$ nanowires," *Applied Physics Letters* 120, 062201 (2022). 10.1063/5.0083980.

Demonstration of $Ta_2Se_3$ exfoliation into nanowires that indicates strong anisotropy in the bonding strength within the basal place. Systematic thermal property measurements disclose signatures of one-dimensional phonons as the nanowire hydraulic diameter reduces below 19.2 nm with linearly escalating thermal conductivity as temperature increases and size dependence inconsistent with the classical size effect. The nanowires of $Ta_2Se_3$ used for this study were obtained from microexfoliation of bulk $Ta_2Se_3$ crystals grown using a chemical vapor transport method at the 2DCC Bulk Growth facility

- External User Project S0061

C. Yan, E. Green, R. Fukumori, N. Protic, **S.H. Lee**, S. Fernandez-Mulligan, R. Raja, R. Erdakos, **Z. Mao**, S. Yang, "An integrated quantum material testbed with multi-resolution photoemission spectroscopy," *Review of Scientific Instruments* 92 (11), 113907 (2021). 10.1063/5.0072979.

Development of a novel quantum material testbed using $FeSe/SrTiO_3$ thin films and bulk $MnBi_4Te_7$ magnetic topological insulators (Tis). The 2DCC grew the magnetic TIs for the user to use in the testbed.

- External User Project S0085

K.M. Siddiqui, D.B. Durham, F. Cropp, C. Ophus, S. Rajpurohit, Y. Zhu, J.D. Carlstrom, C. Stravrakas, **Z. Mao**, A. Raja, P. Musumeci, L.Z. Tan, A.M. Minor, D. Filippetto, R.A. Kaindl, "Ultrafast optical melting of trimer superstructure in layered 1T'-$TaTe_2$," *Communications Physics* 4 (1), 152 (2021). 10.1038/s42005-021-00650-z.

Quasi-two-dimensional transition-metal dichalcogenides are a key platform for exploring emergent nanoscale phenomena arising from complex interactions. Access to the underlying degrees-of-freedom on their natural time scales motivates the use of advanced ultrafast probes sensitive to self-organized atomic-scale patterns. This study reported the ultrafast investigation of $TaTe_2$, which exhibits unique charge and lattice trimer order characterized by a transition upon cooling from stripe-like chains into a $(3 \times 3)$ superstructure of trimer clusters. The work paves the way for further exploration and ultimately rapid optical and electronic manipulation of trimer superstructures. The 2DCC synthesized bulk materials used in this study using non-MIP equipment.

- External User Project S0076 (National Lab)

Y. Tian, Y. Zhu, R. Li, **Z. Mao**, J.H. Ross, "NMR determination of Van Hove singularity and Lifshitz transitions in the nodal-line semimetal ZrSiTe," *Physical Review B* 104 (4), L041105 (2021). PhysRevB.104.L041105.

This study focuses on the Dirac semimetal ZrSiTe. Low-T behavior is dominated by a symmetry-protected nodal line, with NMR providing a sensitive probe of the diamagnetic response of the associate carriers. A Van Hove singularity is identified that is closely connected to this nodal line, and an associated T -induced Lifshitz transition. A disconnect in the NMR shift and linewidth at this temperature indicates the change in electronic behavior associated with this topological change. These features have an orientation-dependent behavior indicating a field-dependent scaling of the associated band energies. The 2DCC synthesized bulk crystals for this study using non-MIP equipment.

- External User Project S0078

C. Yan, S. Fernandez-Mulligan, R. Mei, **S.H. Lee**, N. Protic, R. Fukumori, B. Yan, **C.-X. Liu**, **Z. Mao**, S. Yang, "Origins of electronic bands in the antiferromagnetic topological insulator $MnBi_2Te_4$," *Physical Review B* 104 (4), L041102 (2021). 10.1103/PhysRevB.104.L041102.

Despite the rapid progress in understanding the first intrinsic magnetic topological insulator $MnBi_2Te_4$, its electronic structure remains a topic under debate. This study performs a thorough spectroscopic investigation into the electronic structure of $MnBi_2Te_4$ via laser-based angle-

resolved photoemission spectroscopy. The results represent a solid step forward in reconciling the existing controversies in the electronic structure of $MnBi_2Te_4$ and provides an important framework to understand the electronic structures of other relevant topological materials $MnBi_{2n}Te_{3n+1}$.The 2DCC synthesized bulk crystals for this study using MIP equipment. 2DCC theory personnel also contributed to the development of the theoretical model.

- External User Project S0085
- Also science driver AdvCM

A.S. McLeod, A. Wieteska, G. Chiriaco, B. Foutty, Y. Wang, Y. Yuan, F. Xue, **V. Gopalan**, L.-Q. Chen, **Z.Q. Mao**, A.J. Millis, A.N. Pasupathy, D.N. Basov, "Nano-imaging of a strain-tuned stripe textures in a Mott crystal," *npj Quantum Materials* 6, 46 (2021). 10.1038/s41535-021-00339-0.

This work reveals a spontaneous striped texture of coexisting insulating and metallic domains in single crystals of the quasi-2D, bilayer ruthenate $Ca_3(Ti_xRu_{1-x})_2O_7$ across its first-order Mott transition at T≈95K through multi-messenger low-temperature nano-imaging. The sample used in this study was synthesized by the 2DCC researchers using non-MIP equipment.

- External User Project S0059

L. Yang, Y. Tao, Y. Zhu, M. Akter, K. Wang, Z. Pan, Y. Zhao, Q. Zhang, Y.-Q. Xu, R. Chen, T.T. Xu, Y. Chen, **Z. Mao**, D. Li, "Observation of superdiffusive phonon transport in aligned atomic chains," *Nature Nanotechnology* 16, 764-768 (2021). 10.1038/s41565-021-00884-6.

This work reports the experimental observation of divergence of thermal conductivity ($\kappa$) at room temperature in ultrathin van der Waals crystal $NbSe_3$ nanowires. The $\kappa$ of $NbSe_3$ nanowires was also found to follow a 1/3 power law with wire length, consistent with the superdiffusive phonon transport model. These results not only demonstrate the divergent trend of the observed thermal conductivity with sample length in 1-D atomic chain system, but also unveil a possible way of creating novel 1-D van der Waals crystal-based thermal superconductors with exceptionally high κ values. The 2DCC researchers not only synthesized high-quality $NbSe_3$ single crystals using MIP equipment for this work, but also demonstrated $NbSe_3$ nanowires are stable in air using transmission electron microscopy. These combined synthesis and characterization efforts at the 2DCC enable this achievement.

- External User Projects S0049 and S0061

P. Li, J. Ding, S.-L. Zhang, J. Kally, T. Pillsbury, O.G. Heinonen, G. Rimal, C. Bi, A. DeMann, S.B. Field, W. Wang, J. Tang, J.S. Jiang, A. Hoffmann, **N. Samarth**, M. Wu, "Topological Hall Effect in a Topological Insulator Interfaced with a Magnetic Insulator," *Nano Letters* 21 (1), 84-90 (2021). 10.1021/acs.nanolett.0c03195.

In this paper, we used the 2DCC MBE facility to grow thin films of a topological insulator (TI) ($Bi_2Se_3$) on magnetic insulator (MI) substrates ($BaFe_{12}O_{19}$) provided by the user (Professor Wu, Colorado State). Measurements of the Hall effect made by the user revealed evidence of a genuine topological effect in the temperature range of T = 2–3 K and an anomalous Hall effect at T = 80–300 K. Over T = 3–80 K, the two effects coexist but show opposite temperature dependencies. Control measurements, calculations, and simulations together suggest that the

observed topological Hall effect originates from skyrmions formed due to a Dzyaloshinskii–Moriya interaction at the interface. The strength of this interaction based on fitting the data is estimated to be substantially higher than that in the more extensively studied skyrmion systems derived from heavy metal-based systems. The 2DCC synthesized materials for this study using the MIP MBE1 system.

- External User Project S0057

H. Zhang, Y. L. Zhu, Y. Qiu, W. Tian, H. B. Cao, **Z. Q. Mao**, X. Ke, "Field-induced magnetic phase transitions and the resultant giant anomalous Hall effect in the antiferromagentic half-Heusler compound DyPtBi," *Physical Review B* 102 (9), 094424 (2020). 10.1103/PhysRevB.102.094424.

Systematic Neutron scattering and transport studies of a half-Heusler compound DyPtBi were performed using single crystals grown by the 2DCC Bulk Growth facility. This study shows that DyPtBi hosts a delicate balance between two different magnetic ground states, which can be controlled by moderate magnetic fields. One of the magnetic states hosts a giant anomalous Hall effect. These results indicate that DyPtBi is a potential material for realizing the anomalous Hall effect in an antiferromagnet with a face-centered-cubic lattice.

- External User Project S0074

Z. Pan, L. Yang, Y. Tao, Y. Zhu, Y-Q. Xu, **Z. Mao**, D. Li, "Net negative contributions of free electrons to the thermal conductivity of NbSe3 nanowires," *Physical Chemistry Chemical Physics* 22, 21131-21138 (2020). 10.1039/D0CP03484C.

This paper reports comprehensive experimental studies of the thermal transport properties of $NbSe_3$ nanowires, exfoliated from the bulk $NbSe_3$ crystals grown by the 2DCC Bulk Growth facility. This work reveals that the electron-phonon scattering in the $NbSe_3$ nanowire is enhanced as the free electrons are condensed during the charge density wave transition, thus resulting in the decrease of overall thermal conductivity. This result not only reveals a net negative contribution of the free electrons due to the escalated electron-phonon scattering, but also provides insight into the competing roles of free electrons, which could lead to unexpected trends in thermal conductivity.

- External User Project Collaboration between S0061 and S0049

A. Rossi, G. Resta, **S.H. Lee**, **R.D. Redwing**, C. Jozwiak, A. Bostwick, E. Rotenberg, S.Y. Savrasov, I.M. Vishik, "Two phase transitions driven by surface electron doping in $WTe_2$," *Physical Review B* 102, 121110(R) (2020). 10.1103/PhysRevB.102.121110.
This work identified phase transitions that occur in $WTe_2$ as a result of electron doping with potassium. A postdoctoral scholar from UC-Davis (external user) received training from 2DCC personnel in the Bulk Growth Facility on CVT synthesis and worked on-site to prepare their samples. The external user also used nano-ARPES equipment at LBNL (national lab) to characterize the surface electronic structure of the samples.

- External User Project R0017

T. Liu, J. Kally, T. Pillsbury, C. Liu, H. Chang, J. Ding, Y. Cheng, M. Hilse, **R. Engel-Herbert**, **A. Richardella**, **N. Samarth**, M. Wu, "Changes of Magnetism in a Magnetic Insulator due to

Proximity to a Topological Insulator," *Physical Review Letters* 125, 017204 (2020). 10.1103/PhysRevLett.125.017204.

Thin films of the topological insulator Bi2Se3 were grown by MBE on a magnetic insulator $Y_3Fe_5O_{12}$ thin film. Ferromagnetic resonance measurements show that the topological surface state in $Bi_2Se_3$ produces a perpendicular magnetic anisotropy, results in a decrease in the gyromagnetic ratio, and enhances the damping in $Y_3Fe_5O_{12}$. These topological surface state-induced changes become more pronounced as the temperature decreases from 300 to 50 K. These results suggest a completely new approach for control of magnetism in magnetic thin films. Control measurements using $(Bi,In)_2Se_3$, a trivial insulator rule out possible artifacts.

- External User Project S0057

Y. Shao, A.N. Rudenko, J. Hu, Z. Sun, Y. Zhu, S. Moon, A.J. Millis, S. Yuan, A.I. Lichtenstein, D. Smirnov, **Z.Q. Mao**, M.I. Katsnelson, D.N. Basov, "Electronic correlations in nodal-line semimetals," *Nature Physics* 16, 636-641 (2020). 10.1038/s41567-020-0859-z.

Spectroscopic hallmarks of electronic correlations (i.e. strong reduction of the Drude weight and the Fermi velocity) are observed in a topological nodal-line semimetal ZrSiSe. This work establishes the first platform to explore correlation of relativistic fermions in low dimension. Some of the crystals used in this work were grown using the 2DCC Bulk Growth facility.

- External User Project S0082

S.M. Oliver, J.J. Fox, A. Hashemi, A. Singh, R.L. Cavalero, S. Yee, **D.W. Snyder**, R. Jaramillo, H.-P. Komsa, P.M. Vora, "Phonons and excitons in $ZrSe_2$-$ZrS_2$ alloys," *Journal of Materials Chemistry C* (2020) in press. 10.1039/D0TC00731E.

A comprehensive analysis of photons and excitons in $Z(S,Se)_2$ alloy crystals (synthesized in the 2DCC Bulk Growth facility) was carried out using Raman spectroscopy and spectroscopic ellipsometry. The Raman spectrum was found to be dominated by nominally IR phonons due to the large ionicity of bonding.

- External User Projects R0014 and R0016

A. Gangshettiwar, Y. Zhu, Z. Jiang, J. Peng, Y. Wang, J. He, J. Zhou, **Z. Mao**, K. Lai, "Emergence of a competing stripe phase near the Mott transition in Ti-doped bilayer calcium ruthenates," *Physical Review B*, 101(20), 201106(R) (2020). 10.1103/PhysRevB.101.201106.

This work reveals a new exotic phenomenon of correlated electrons: a competing stripe phase at a Mott transition. The sample used in this study was synthesized using non-MIP equipment.

- External User Project S0060

F. Lupke, D. Waters, S.C. de la Barrera, M. Widom, D.G. Mandrus, J.Q. Yan, R.M. Feenstra, B.M. Hunt, "Proximity-induced superconducting gap in the quantum spin Hall edge state of monolayer $WTe_2$," *Nature Physics* 16, 526-530 (2020). 10.1038/s41567-020-0816-x.

This study used scanning tunneling spectroscopy of 2DCC-grown $WTe_2$ monolayer samples in contact with $NbSe_2$ to study proximity-induced superconductivity in the quantum spin Hall phase. This is an important advance toward establishing a 1D topological superconductor and Majorana zero modes in condensed matter.

- Also science driver NGDev
- External User Project S0027

P. Li, J. Kally, S.-L. Zhang, T. Pillsbury, J. Ding, G. Csaba, J. Ding, J.S. Jiang, Y. Liu, R. Sinclair, C. Bi, A. DeMann, G. Rimal, W. Zhang, S.B. Field, J. Tang, W. Wang, O.G. Heinonen, V. Novosad, A. Hoffman, **N. Samarth**, M. Wu, "Magnetization switching using topological surface states," *Science Advances,* 5 (8), eaaw3415 (2019). 10.1126/sciadv.aaw3415

This project used 2DCC MBE-grown topological insulator/ferromagnet insulator bilayers ($Bi_2Se_3$/$BaFe_{12}O_{19}$) to fabricate spintronic devices that showed current-induced magnetization switching. The pronounced increase in switching efficiency at cryogenic temperatures led to the conclusion that this process is dominated by the spin-momentum locking of topological surface states that have enhanced surface conductivity at low temperatures where bulk conductivity freezes out.

- Also science driver NGDev
- External User Project S0025

***Local User Publications (Phys2D)***

H. Padmanabhan, M. Poore, P.K. Kim, N.Z. Koocher, **V.A. Stoica**, D. Puggioni, H. Wang, X. Shen, A.H. Reid, M. Gu, M. Wetherington, **S.H. Lee**, R.D. Schaller, **Z. Mao**, A.M. Lindenberg, X. Wang, J.M. Rondinelli, R.D. Averitt, *V. Gopalan*, "Interlayer magnetophononic coupling in $MnBi_2Te_4$," *Nature Communications* 13, 1929 (2022). 10.1038/s41467-022-29545-5.
This study presents evidence for interlayer magnetophononic coupling in the layered magnetic topological insulator $MnBi_2Te_4$. Anomalies in phonon scattering intensities across magnetic field-driven phase transitions are observed, despite the absence of discernible static structural changes. This behavior is a consequence of a magnetophononic wave-mixing process that allows for the excitation of zone-boundary phonons that are otherwise 'forbidden' by momentum conservation. The microscopic model based on density functional theory calculations reveals that this phenomenon can be attributed to phonons modulating the interlayer exchange coupling. In light of the intimate connection between magnetism and topology in $MnBi_2Te_4$, the magnetophononic coupling represents an important step towards coherent on-demand manipulation of magnetic topological phases. Materials grown in this project done with 2DCC muffle furnace equipment.

- Local user project S0062 collaboration with in-house research
- Also science driver AC&M

***In-house Research Publications (Phys2D)***

J. He, **S.H. Lee**, F. Naccarato, G. Brunin, R. Zu, **Y. Wang**, L. Miao, H. Wang, **N. Alem**, G. Hautier, G.-M. Rignanese, **Z. Mao**, **V. Gopalan**, "$SnP_2S_6$: A Promising Infrared Nonlinear Optical Crystal with Strong Nonresonant Second Harmonic Generation and Phase-Matchability," *ACS Photonics* 9 (5), 1724-1732 (2022). 10.1021/acsphotonics.2c00131.
High-power infrared laser systems with broad-band tunability are of great importance due to their wide range of applications in spectroscopy and free-space communications. These systems require nonlinear optical (NLO) crystals for wavelength up/down conversion using sum/difference frequency generation, respectively. NLO crystals need to satisfy many competing criteria, including large nonlinear optical susceptibility, large laser-induced damage threshold (LIDT), wide transparency range, and phase-matchability. This study reveals that $SnP_2S_6$ is an outstanding candidate. Bulk crystals of $SnP_2S_6$ were grown by 2DCC CVT equipment.

Y. Lv, J. Kally, T. Liu, P. Quarterman, T. Pillsbury, B.J. Kirby, A.J. Grutter, P. Sahu, J.A. Borchers, M. Wu, **N. Samarth**, J.-P. Wang, "Large unidirectional spin Hall and Rashba-Edelstein magnetoresistance in topological insulator/magnetic insulator heterostructures," *Applied Physics Reviews* 9 (1), 011406 (2022). 10.1063/5.0073976.

The unidirectional spin Hall and Rashba−Edelstein magnetoresistance is of great fundamental and practical interest, particularly in the context of reading magnetization states in two-terminal spin–orbit torque memory and logic devices due to its unique symmetry. Here, we report large unidirectional spin Hall and Rashba−Edelstein magnetoresistance in a new material family—magnetic insulator/topological insulator $Y_3Fe_5O_{12}/Bi_2Se_3$ bilayers. Ee demonstrate a prototype memory device based on a magnetic insulator/topological insulator bilayer, using unidirectional spin Hall and Rashba−Edelstein magnetoresistance for electrical readout of current-induced magnetization switching aided by a small Oersted field. The materials in this study were synthesized by the 2DCC MIP MBE1 system.

- In-house collaboration with external user S0057

W. Yanez, **Y. Ou**, R. Xiao, J. Koo, J.Y. Held, S. Ghosh, J. Rable, T. Pillsbury, E. Gonzlaez Delgado, K. Yang, J. Chamorro, A.J. Grutter, P. Quarterman, **A. Richardella**, A. Sengupta, T. McQueen, J.A. Borchers, K.A. Mkhoyan, B. Yan, **N. Samarth**, "Spin and Charge Interconversion in Dirac-Semimetal Thin Films," *Physical Review Applied* 16 (5), 054031 (2021). 10.1103/PhysRevApplied.16.054031.

This study uses spin torque ferromagnetic resonance and ferromagnetic-resonance-driven spin pumping to detect spin-charge interconversion at room temperature in heterostructure devices that interface an archetypal Dirac semimetal, $Cd_3As_2$, with a metallic ferromagnet, $Ni_{0.80}Fe_{0.20}$ (permalloy). Angle-resolved photoemission directly reveals the Dirac-semimetal nature of the samples prior to device fabrication and high-resolution transmission electron microscopy is used to characterize the crystalline structure and the relevant heterointerfaces. We find that the spin-charge interconversion efficiency in $Cd_3As_2$/permalloy heterostructures is comparable to that in heavy metals and that it is enhanced by the presence of an interfacial oxide. Spin torque ferromagnetic resonance measurements reveal an in-plane spin polarization regardless of an oxidized or pristine interface. The 2DCC MIP facility was used for ARPES measurements of the $Cd_3As_2$ thin films via *in vacuo* transfer within the 2DCC highly integrated vacuum environment (HIVE).

**S.H. Lee**, D. Graf, L. Min, Y. Zhu, H. Yi, S. Ciocys, **Y. Wang**, E.S. Choi, R. Basnet, A. Fereidouni, **V. Gopalan**, H.O.H. Churchill, A Lanzara, **N. Samarth**, **C.-Z. Chang**, J. Hu, **Z. Mao**, "Evidence for a Magnetic-Field-Induced Ideal Type-II Weyl State in Antiferromagnetic Topological Insulator $Mn(Bi_{1-x}Sb_x)_2Te_4$," *Physical Review X* 11 (3), 031032 (2021). 10.1103/PhysRevX.11.031032.

In this article, we report transport evidence for a TRS-breaking type-II WSM observed in the intrinsic antiferromagnetic topological insulator $Mn(Bi_{1-x}Sb_x)_2Te_4$ under magnetic fields. This state is manifested by the electronic structure transition caused by the spin-flop transition. The transition results in an intrinsic anomalous Hall effect and negative c-axis longitudinal magnetoresistance attributable to the chiral anomaly in the ferromagnetic phases of lightly hole-doped samples. Our results establish a promising platform for exploring the underlying physics of the long-sought, ideal TRS-breaking type-II WSM. The 2DCC synthesized bulk crystals for this study using MIP equipment.

- Also science driver AdvCM

J.Y. Liu, J. Yu, J.L. Ning, H.M. Yi, L. Miao, L.J. Min, Y.F. Zhao, W. Ning, K.A. Lopez, Y.L. Zhu, T. Pillsbury, Y.B. Zhang, **Y. Wang**, J. Hu, H.B. Cao, B.C. Chakoumakos, F. Balakirev, F. Weickert, M. Jaime, Y. Lai, L. Yang, J.W. Sun, **N. Alem**, **V. Gopalan**, **C.Z. Chang**, **N. Samarth**, **C.X. Liu**, R.D. McDonald, **Z.Q. Mao**, "Spin-valley locking and bulk quantum Hall effect in a noncentrosymmetric Dirac semimetal $BaMnSb_2$," *Nature Communications* 12 (1), 4062 (2021). 10.1038/s41467-021-24369-1.

Spin-valley locking in monolayer transition metal dichalcogenides has attracted enormous interest, since it offers potential for valleytronic and optoelectronic applications. Such an exotic electronic state has sparsely been seen in bulk materials. Here, we report spin-valley locking in a Dirac semimetal $BaMnSb_2$. This is revealed by comprehensive studies using first principles calculations, tight-binding and effective model analyses, angle-resolved photoemission spectroscopy measurements. The 2DCC synthesized bulk crystals for this study using MIP equipment and contributed first-principles calculations from 2DCC personnel.

- Also science driver AdvCM

N.P. de Leon, K.M. Itoh, D. Kim, K.H. Mehta, T.E. Northup, H. Pak, B.S. Palmer, **N. Samarth**, S. Sangtawesin, D.W. Steuerman, "Materials challenges and opportunities for quantum computing hardware," *Science* 372 (6539), eabb2823 (2021). 10.1126/science.abb2823.

This is a review paper that provides a sweeping overview of the field of quantum computing from the perspective of a materials scientist, describing the key challenges faced by the various principal platforms for building computing hardware (superconducting Josephson junction, semiconductor quantum dots, single spin defects, ion traps, and topological superconductors). Most of the paper discussed materials beyond the scope of 2DCC, but some materials of relevance to 2DCC that were commented on include topological superconductors, graphene, and 2D van der Waals materials.

- Also science driver NGDev

W. Ge, P.M. Sass, J. Yan, **S.H. Lee**, **Z. Mao**, W. Wu, "Direct Evidence of Ferromagnetism in $MnSb_2Te_4$," *Physical Review B* 103 (13), 134403 (2021). 10.1103/PhysRevB.103.134403.

This work reveals direct evidence of ferromagnetism in $MnSb_2Te_4$ using cryogenic magnetic force microscopy. A part of the materials used in this study was synthesized using the 2DCC bulk growth facility. While $MnSb_2Te_4$ was previously reported to be antiferromagnetic, the 2DCC researchers succeeded in growing the ferromagnetic phase which was not predicted in theory through tuning growth conditions. The FM $MnSb_2Te_4$ offers a new platform to explore new exotic quantum states in 2D magnetic materials. The 2DCC synthesized bulk crystals for this study using MIP equipment.

Y. Li, Z. Wang, R. Xiao, Q. Li, K. Wang, **A. Richardella**, J. Wang, **N. Samarth**, "Capping layer influence and isotropic in-plane upper critical field of the superconductivity at the $FeSe/SrTiO_3$ interface," *Physical Review Materials* 5 (3), 034802 (2021). 10.1103/PhysRevMaterials.5.034802.

In this in house project, we used the 2DCC multimodule MBE system to grow and characterize ultrathin FeSe films on $SrTiO_2$. Understanding the superconductivity at the interface

of $FeSe/SrTiO_3$ is a problem of great contemporary interest due to the significant increase in critical temperature (Tc) compared to that of bulk FeSe, as well as the possibility of an unconventional pairing mechanism and topological superconductivity. We studied the influence of a capping layer on superconductivity in thin films of FeSe grown on $SrTiO_3$ using molecular beam epitaxy. We used the 2DCC *in vacuo* four-probe electrical resistance measurement facility (LT-Nanoprobe) and *ex situ* magnetotransport measurements to examine the effect of three capping layers that provide distinct charge transfer into FeSe: insulating FeTe, nonmetallic Te, and metallic Zr. Our results show that FeTe provides an optimal cap that barely influences the inherent Tc found in pristine $FeSe/SrTiO_3$, while the transfer of holes from a nonmetallic Te cap completely suppresses superconductivity and leads to insulating behavior. Finally, we used *ex situ* magnetoresistance measurements in FeTe capped FeSe films to extract the angular dependence of the in-plane upper critical magnetic field. Our observations reveal an almost isotropic in-plane upper critical field, providing insight into the symmetry and pairing mechanism of high-temperature superconductivity in FeSe. The 2DCC synthesized materials in this study using the MIP MBE1 and in vacuo STM.

Y. Zhu, J. Hu, D. Graf, X. Gui, W. Xe, **Z. Mao**, "Quasi-two-dimensional relativistic fermions probed by de Haas-van Aplhen quantum oscillations in $LuSn_2$," *Physical Review B* 103 (12), 125109 (2021). 10.1103/PhysRevB.103.125109.

This work revealed the distorted Sn-square net layer in the layered compound $LuSn_2$ generates relativistic fermions. The two-dimensionality of the relativistic band is found to be significantly enhanced due to the suppressed corrugation of the Sn square net layer as compared to the previously reported topological semimetal $YSn_2$. These results suggest that the dimensionality of the relativistic band in $RESn_2$ (RE=rare earth) can be tuned by the electronegativity of RE atoms. Some samples used in this study were synthesized using MIP equipment in the 2DCC bulk synthesis facility.

F. Wang, X. Wang, Y.-F. Zhao, D. Xiao, L.-J. Zhou, W. Liu, Z. Zhang, W. Zhao, M.H.W. Chan, **N. Samarth**, **C. Liu**, H. Zhang, **C.-Z. Chang**, "Interface-induced sign reversal of the anomalous Hall effect in magnetic topological insulator heterostructures," *Nature Communications* 12, 79 (2021). 10.1038/s41467-020-20349-z.

In this in house research, we used both a faculty MBE chamber (Chang) and the 2DCC MIP equipment (MBE1) to grow quantum anomalous Hall insulator samples derived from V- and Cr-doped $Sb_2Te_3$ and $(Bi,Sb)_2Te_3$. These samples were then studied using low temperature magnetotransport measurements to understand the intrinsic anomalous Hall effect in terms of non-zero Berry curvature in momentum space. We find that the sign of the anomalous Hall effect in the magnetic chalcogenide topological insulator layer can be changed from being positive to negative by varying the heterostructure details (e.g. layer thickness). First-principles calculations by 2DCC theorists (Liu) show that the built-in electric fields at heterointerfaces influence the band structure of the magnetically doped layers, and thus lead to a reconstruction of the Berry curvature in the heterostructure samples. This enabled the design and demonstration of an artificial "topological Hall effect"-like feature.

- Also science driver AdvCM

W. Ning, **Z. Mao**, "Recent advancements in the study of intrinsic magnetic topological insulators and magnetic Weyl semimetals," *APL Materials* 8 (9), 090701 (2020). 10.1063/5.0015328.

The breaking of time-reversal symmetry in topological materials has been extensively studied as a platform to generate quantum effects, such as the quantum anomalous Hall effect. In this research review, the recent research progress in magnetic topological materials, including intrinsic magnetic topological insulators and magnetic Weyl semimetals, are briefly overviewed.

S.-W. Wang, D. Xiao, Z. Dou, M. Cao, Y.-F. Zhao, **N. Samarth**, **C.-Z. Chang**, M.R. Connolly, C. G. Smith, "Demonstration of Dissipative Quasihelical Edge Transport in Quantum Anomalous Hall Insulators," *Physical Review Letters* 125 (12), 126801 (2020). 10.1103/PhysRevLett.125.126801.

Thin heterostructure films of magnetically-doped topological insulators (TIs), specifically Cr-doped $(Bi,Sb)_2Te_3$, were grown by molecular beam epitaxy on $SrTiO_3$ substrates. Using an electrostatic back gate, the films could be tuned into the quantum anomalous Hall (QAH) insulator state. Collaborators at the University of Cambridge (UK) then studied the temperature- and magnetic-field-dependence of the magnetoresistance of a magnetic TI sandwich heterostructure device. The measurements demonstrated that the predominant dissipation mechanism in thick QAH insulators can switch between nonchiral edge states and residual bulk states in different magnetic-field regimes. The paper provides a way to distinguish between the dissipation arising from the residual bulk states and nonchiral edge states, which is crucial for achieving true dissipationless transport in QAH insulators and for providing deeper insights into QAH-related phenomena.

K. Nisi, S. Subramanian, W. He, K.A. Ulman, H. El-Sherif, F. Sigger, M. Lassauniere, M.T. Wetherington, N. Briggs, J. Gray, A.W. Holleitner, N. Bassim, S.Y. Quek, **J.A. Robinson**, U. Wurstbauer, "Light-Matter Interaction in Quantum Confined 2D Polar Metals," *Advanced Functional Materials*, 2005977 (2020). 10.1002/adfm.202005977.

In this study, we explore the linear optical response of 2D Ga and 2D In. The fundamental light–matter interaction which is described by the complex dielectric functions. We determine the dielectric functions of 2D Ga and 2D In via a combination of spectroscopic ellipsometry (SE) and density functional theory (DFT) in a large spectral range from NIR to UV. The MIP provided the 2D metals for the study.

M. Kopf, J. Ebad-allah, **S.H. Lee**, **Z.Q. Mao**, C.A. Kuntscher, "Influence of magnetic ordering on the optical response of the antiferromagentic topological insulator $MnBi_2Te_4$," *Physical Review B* 102 (16), 165139 (2020). 10.1103/PhysRevB.102.165139.

Comprehensive temperature-dependent optical conductivity studies were performed on $MnBi_2Te_4$ that were grown using the 2DCC Bulk Growth facility. The observations of strong changes in the optical conductivity at Neel temperature confirms the impact of magnetic ordering on the bulk electronic properties of $MnBi_2Te_4$.

M.A. Steves, **Y. Wang**, N. Briggs, T. Zhao, H. Elh-Sherif, B.M. Betrsch, S. Subramanian, C. Dong, T. Bowen, A. De La Fuente Duran, K. Nisi, M. Lassauniere, U. Wurstbauer, N.D. Bassim, J. Fonseca, J.T. Robinson, **V.H. Crespi**, **J.A. Robinson**, K.L. Knappenberger Jr., "Unexpected

Near-Infrared to Visible Nonlinear Optical Properties from 2-D Polar Metals," *Nano Letters* 20 (11), 8312-8318 (2020). 10.1021/acs.nanolett.0c03481.

Near-infrared-to-visible second harmonic generation from air-stable two-dimensional polar gallium and indium metals is described. The photonic properties of 2D metals -including the largest second-order susceptibilities reported for metals (approaching 10nm2/V) –are determined by the atomic-level structure and bonding of two-to-three-atom-thick crystalline films. The MIP played a key role in developing the 2D metals and providing theory on the origin of the optical response.

- Collaboration with External User R0024 (National Lab)

P. Li, J. Koo, W. Ning, J. Li, L. Miao, L. Min, Y. Zhu, Y. Wang, N. Alem, **C.-X. Liu**, **Z. Mao**, B. Yan, "Giant room temperature anomalous Hall effect and tunable topology in a ferromagnetic topological semimetal $Co_2MnAl$," *Nature Communications* 11 (1), 3476 (2020). 10.1038/s41467-020-17174-9.

This work not only reveals a giant room temperature anomalous Hall effect in a Heusler alloy $Co_2MnAl$, but also demonstrates its band topology can be tuned by the rotation of magnetization driven by small magnetic fields. These results pay a way for potential applications of 2D thin film of this material in spintronic devices.

X. Wu, D. Xiao, C.-Z. Chen, J. Sun, L. Zhang, M.H.W. Chan, N. Samarth, X.C. Xie, X. Lin, C.-Z. Chang, "Scaling Behavior of the Quantum Phase Transition from a Quantum Anomalous Hall Insulator to an Axion Insulator," *Nature Communications* (2020). 10.1038/s41467-020-18312-z.

Heterostructures of magnetically-doped topological insulators were grown by MBE and used to study the phase transition from the quantum anomalous Hall phase to an axion insulator phase. We find that the transition follows a universal scaling behavior when we analyze the temperature dependence of the derivative of the longitudinal resistance on magnetic field at the transition point. This behavior follows a characteristic power-law that indicates a universal scaling behavior that can be understood by the Chalker-Coddington network model with a critical exponent which agrees with recent high-precision numerical results.

Y. Chen, Y.-W. Chuang, S.H. Lee, Y. Zhu, K. Honz, Y. Guan, Y. Wang, K. Wang, Z. Mao, J. Zhu, C. Heikes, P. Quarterman, P. Zajdel, J.A. Borchers, W. Ratcliff II, "Ferromagnetism in van der Waals compound $MnSb_{1.8}Bi_{0.2}Te_4$," *Phys. Rev. Mater.*, 4, 064411 (2020). 10.1103/PhysRevMaterials.4.064411

A new ferromagnetic phase showing unusual anomalous Hall effect was synthesized through the control of disorders. This material offers opportunity to explore new topological quantum states in 2D. This involves collaboration with a minority researcher at NIST.

Y. Zhu, B. Singh, Y. Wang, C.-Y. Huang, W.-C. Chiu, B. Wang, D. Graf, Y. Zhang, H. Lin, J. Sun, A. Bansil, **Z. Mao**, "Exceptionally large anomalous Hall effect due to anticrossing of spin-split bands in the antiferromagnetic half-Heusler compound TbPtBi," *Physical Review B*, 101, 161105 (2020). 10.1103/PhysRevB.101.161105.

This work reveals a large intrinsic anomalous Hall effect with a record value of the Hall angle in a half Heusler compound. This phenomenon arises from the anticrossing of spin-split bands near the Fermi level. The physics revealed in this work can be extended to 2D systems.

J. Jiang, D. Xiao, F. Wang, J.H. Shin, D. Andreoli, J. Zhang, R. Xiao, Y.-F. Zhao, M. Kayyalha, L. Zhang, K. Wang, J. Zang, **C. Liu**, **N. Samarth**, M.H.W. Chang, **C.Z. Chang**, "Concurrence of

quantum anomalous Hall and topological Hall effects in magnetic topological insulator sandwich structures," *Nature Materials* 19, 732-737 (2020). 10.1038/s41563-020-0605-z.

MBE-grown magnetically doped topological insulator heterostructures were used to demonstrate the voltage tuned transition between and concurrence of Berry phase spin texture (characterized by the quantum anomalous Hall effect) and real space spin texture (characterized by the topological Hall effect).

- Also science driver NGDev

M. Kayyalha, D. Xiao, R. Zhang, J. Shin, J. Jiang, F. Wang, Y.-F. Zhao, R. Xiao, L. Zhang, K.M. Fijalkowski, P. Mandal, M. Winnerlein, C. Gould, Q. Li, L.W. Molenkamp, M.H.W. Chan, **N. Samarth**, **C.-Z. Chang**, "Absence of evidence for chiral Majorana modes in quantum anomalous Hall-superconductor devices," *Science*, 367 (6473), 64-67, (2020). 10.1126/science.aax6361

This study used MBE-grown magnetically doped topological insulator heterostructures with highly transparent superconducting contacts to show that the half-quantized conductance of a quantum anomalous Hall insulator channel with proximitized superconductivity is not a signature of chiral Majorana fermions as predicted by theory.

S. Islam, S. Bhattacharya, H. Nhalil, M. Banerjee, **A. Richardella**, A. Kandala, D. Sen, **N. Samarth**, S. Elizabeth, A. Ghosh, "Low-temperature saturation of phase coherence length in topological insulators," *Physical Review B* 99, 245407 (2019). 10.1103/PhysRevB.99.245407

This collaborative paper used 2DCC MBE-grown samples to study the magnetoresistance and conductance fluctuations measurements in topological insulator thin films. The studies indicated the need to identify an alternative source of dephasing that dominates at low temperature in topological insulators, causing saturation in the phase breaking length and time.

**S.H. Lee, Y. Zhu,** Y. Wang, L. Miao, T. Pillsbury, H. Yi, S. Kempinger, J. Hu, C.A. Heikes, P. Quaterman, W. Ratcliff, J.A. Borchers, H. Zhang, X. Ke, D. Graf, N. Alem, **C-Z. Chang**, **N. Samarth**, and **Z. Mao**, "Spin scattering and noncollinear spin structure-induced intrinsic anomalous Hall effect in antiferromagnetic topological insulator $MnBi_2Te_4$," *Phys. Rev. Res.*, 1, 012011R (2019). 10.1103/PhysRevResearch.1.012011

This study used the 2DCC Bulk Growth facility and the 2DCC ARPES facility to study the antiferromagnetic (AFM) topological insulator $MnBi_2Te_4$. The key findings included the discovery of a magnetic field-driven non-collinear spin structure with an intrinsic anomalous Hall effect and a large intrinsic gap in the surface states created by strong spin fluctuations.

F. Wang, D. Xiao, W. Yuan, J. Jiang, Y.-F. Zhao, L. Zhang, Y. Yao, W. Liu, Z. Zhang, **C. Liu**, J. Shi, W. Han, M. H. W. Chan, **N. Samarth**, and **C.-Z. Chang**, "Observation of Interfacial Antiferromagnetic Coupling between Magnetic Topological Insulator and Antiferromagnetic Insulator," *Nano Letters* 19(5) 2945-2952 (2019). 10.1021/acs.nanolett.9b00027

This study used MBE-grown ferromagnetic topological insulator/antiferromagnetic insulator heterostructures ($(Cr,Sb)_2Te_3/Cr_2O_3$) to demonstration rich temperature-tuned interfacial antiferromagnetic exchange coupling and an exchange-enhanced Curie temperature in the ferromagnetic topological insulator.

L.-H. Hu, **C.-X. Liu**, F.-C. Zhang, "Topological Larkin-Ovchinnikov phase and Majorana zero mode chain in bilayer superconducting topological insulator films," *Commun. Physics*, 2 (1), 1-7 (2019). 10.1038/s42005-019-0126-8

This theoretical paper predicts the emergence of a magnetic field-induced topological Larkin-Ovchinnikov superconducting phase with a finite momentum pairing in bilayer superconducting

topological insulator films. The theoretical model can be naturally realized in superconductor/topological insulator sandwich structure or in a Fe(Te, Se) film.

D. Xiao, J. Jiang, J.H. Shin, W. Wang, F. Wang, Y.F. Zhao, **C.X. Liu**, W.D Wu, M. H. W. Chan, **N. Samarth**, and **C.Z. Chang**, "Realization of the Axion Insulator State in Quantum Anomalous Hall Sandwich Heterostructures," *Phys. Rev. Lett.*, 120, 056801 (2018). 10.1103/PhysRevLett.120.056801

This study used 2DCC MBE-grown magnetic topological insulator heterostructures to realize a new quantum state of matter known as the axion insulator, wherein both the longitudinal and Hall conductivity vanish when the opposite surfaces of a topological insulator are oppositely gapped.

- Also science driver NGDev

S. Islam, S. Bhattacharyya, **A. Richardella**, **N. Samarth**, and A. Ghosh, "Bulk-impurity Induced Noise in Large-area Epitaxial Thin Films of Topological Insulators", *Appl. Phys. Lett* **2017**, *111*, 062107. 10.1063/1.4998464

This collaborative paper used 2DCC MBE-grown samples to study the low frequency electrical noise in topological insulator thin films. The studies showed that even in very thin films, defect states within a bulk impurity band are the source of significant electrical noise in surface electrical transport.

Y. Pan, Q-Z. Wang, A. Yeats, T. Pillsbury, T. Flanagan, **A. Richardella**, H. Zhang, D. Awschalom, **C-X. Liu**, **N. Samarth**, "Helicity Dependent Photocurrent in Electrically Gated $(Bi_{1-x}Sb_x)_2Te_3$ Thin Films", *Nature Commun.* **2017**, *8*, 1037; 10.1038/s41467-017-00711-4

This study used 2DCC MBE-grown samples to study the circular photogalvanic effect in topological insulator thin films as a function of chemical potential. The key result shows that even when photocurrents are excited using photon energies well above the bulk band gap, the transitions are still influenced by the spin-momentum correlation present in the Dirac states leading to direction control of photocurrents by the circular polarization of the optical excitation.

- Also science driver NGDev

A. Yeats, P. Mintun, Y. Pan, **A. Richardella**, B. Buckley, **N. Samarth**, and D. Awschalom, "Local Optical Control of Ferromagnetism and Chemical Potential in a Topological Insulator", *PNAS* **2017**, *114* (9), 10379-10383. 10.1073/pnas.1713458114

This collaborative paper demonstrates micron-scale persistent optical patterning of ferromagnetism and chemical potential landscape in magnetically doped topological insulators grown in 2DCC Thin Films facility.

- Also science driver NGDev

W. Dai, **A. Richardella**, R. Du, W. Zhao, X. Liu, C-X. Liu, S-H. Huang, R. Sankar, F. Chou, **N. Samarth**, and Q. Li, "Proximity-effect-induced Superconducting Gap in Topological Surface States - A Point Contact Spectroscopy Study of $NbSe_2/Bi_2Se_3$ Superconductor-Topological Insulator Heterostructures", *Scientific Reports* **2017**, *7*. 10.1038/s41598-017-07990-3

Point-contact study of the proximity-induced superconductivity in a topological insulator/superconductor bilayer ($Bi_2Se_3/NbSe_2$) grown using the 2DCC Thin Films facility.

- Also science driver NGDev

**N. Samarth**, "Quantum Materials Discovery From a Synthesis Perspective," *Nature Materials* **2017**, *16*, 1068-1076. 10.1038/NMAT5010

Review article on status and opportunities in materials synthesis of quantum materials including those of central interest to the 2DCC Thin Films facility.

- Also science driver NGDev

J. P. Heremans, R.J. Cava, and **N. Samarth**, "Tetradymites as Thermoelectrics and Topological Insulators", *Nat. Rev. Mater.* **2017**, *2*, 17049. 10.1038/natrevmats.2017.49

Review article on the synthesis and properties of chalcogenide crystals (tetradymites) that are of central interest to the 2DCC Thin Films facility.

- Also science driver NGDev