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as of 19-Jan-2022

Agency Code: 21XD

Proposal Number: 70645ELYIP

Agreement Number: W911NF-17-1-0399

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Final Report for Period Beginning 11-Sep-2017 and Ending 31-Aug-2021

Title: Nanoscale Engineering of Superconducting Proximity Effect in Topological Insulator Thin Films

Begin Performance Period: 11-Sep-2017

End Performance Period: 31-Aug-2021

Report Term: 0-Other

Submitted By: Ilija Zeljkovic

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 4

STEM Participants: 12

Major Goals: The focus of the project is to create and investigate new topological phases, primarily focusing on topological superconducting states in heterostructures of topological materials and high-T_c superconductors.

Accomplishments: (1) Proximity-induced superconductivity in a TI by coupling to Fe(Te,Se).

We have successfully grown thin films of a topological insulator Bi₂Te₃ in a range of thicknesses on top of a high-temperature superconductor Fe(Te,Se) (superconducting T_c~14K). Using scanning tunneling microscopy and spectroscopy (STM/S), we detected a superconducting gap as high as ~3.5 meV on the surface of the TI. This is the largest induced gap by proximity to an s-wave superconductor, and the first time superconductivity in a TI was induced by proximity to an Fe-based high-T_c superconductor. By imaging the scattering and interference of surface state electrons, we also provide a microscopic visualization of the fully gaped Bi₂Te₃ surface state due to Cooper pairing [Zhao, ..., Zeljkovic, PRB 97, 224504 (2018)].

(2) Proximity-induced superconductivity in a topological crystalline insulator SnTe.

We used 1 quintuple layer (QL) thick Bi₂Te₃ grown on top of Fe(Te,Se) as a substrate for the MBE growth of topological crystalline insulator SnTe(111). SnTe is difficult to grow directly on commonly used superconducting substrates due to a large lattice mismatch, but it has a nearly perfect lattice match to Bi₂Te₃. Using STM/S, we demonstrated that the surface of SnTe also shows an induced superconducting gap. Interestingly, we were also able to evaporate off the buffer layer, creating direct interface between the SnTe and Fe(Te,Se), which also shows proximity induced superconductivity. It is worth noting that vortices at the surface of SnTe show a zero bias peak in dI/dV spectra, which does not split away from the center, similarly to the zero bias peaks observed on the surface of Bi₂Te₃/Fe(Te,Se). [Rachmilowitz, ..., Zeljkovic PRB 100, 241402 (2019)].

(3) Coulomb blockade effect in topological insulator/cuprate heterostructures

The evidence for proximity-induced superconductivity in heterostructures of topological insulators and high-T_c cuprates has been intensely debated in literature. We used molecular beam epitaxy to grow thin films of topological insulator Bi₂Te₃ on a cuprate Bi₂Sr₂CaCu₂O_{8+x}, and studied the surface of Bi₂Te₃ using low-temperature scanning tunneling microscopy and spectroscopy. In few unit-cell thick Bi₂Te₃ films, we found a V-shaped gap-like feature at the Fermi energy in dI/dV spectra. By reducing the coverage of Bi₂Te₃ films to create nanoscale islands, we discovered that this spectral feature dramatically evolves into a much larger hard gap, which can be understood as a Coulomb blockade gap. This conclusion is supported by the evolution of dI/dV spectra with the lateral size of Bi₂Te₃ islands, as well as by topographic measurements that show an additional barrier separating Bi₂Te₃ and Bi₂Sr₂CaCu₂O_{8+x}. [Rachmilowitz, ..., Zeljkovic, npj Quantum Materials 5, 72 (2020)].

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During this process, we have discovered that Bi-2212 gets extremely underdoped (T_c decreases) at the typical Bi₂Te₃ growth temperatures of ~300 °C because interstitial oxygen atoms, which hole-dope the system, escape from the surface during heating. As a result of this discovery, we lowered the Bi₂Te₃ growth temperatures to keep the Bi-2212 superconducting, but we could still not observe a clear superconducting gap on the surface of Bi₂Te₃, unlike what we observed in Bi₂Te₃/Fe(Te,Se) and SnTe(111)/ Bi₂Te₃/Fe(Te,Se) heterostructures. This demonstration of effective surface doping by Oxygen removal has spun into another experiment [Zhao, ..., Zeljkovic, Nature Materials 18, 103 (2019)], where we created and studied charge ordering in charge-transfer insulator state of Bi-2212.

(4) Nematic instability and suppression of superconductivity in Fe(Te,Se)

We also carefully investigated the surface of Fe(Te,Se) (Se:Te~0.45:0.55) used as the substrate in our proximity studies, as emergent experimental evidence suggested it may be an intrinsic topological superconductor. Fe(Te, Se) undergoes both a topological transition, linked to the electronic band inversion, and an electronic nematic phase transition, associated with rotation symmetry breaking, around the same critical composition x_c where superconducting T_c peaks. At this regime, nematic fluctuations and symmetry-breaking strain could have an enormous impact. Using low-temperature spectroscopic-imaging scanning tunneling microscopy, we visualized the electronic nematic transition in Fe(Te,Se). At x_c , we discovered the emergence of nanoscale regions hosting electronic nematicity with suppressed superconductivity, embedded within the non-nematic superconducting matrix. While we found that ~1% uniaxial strain can significantly weaken superconducting coherence and enhance static electronic nematicity near x_c , strongly nematic regions with destroyed superconductivity emerge even in the absence of substantial strain. The underlying physics of these regions could be understood in terms of nematic fluctuations, generally believed to enhance superconductivity, becoming static in localized regions where superconductivity is suppressed. Our experiments revealed a strong tendency of Fe(Te,Se) near critical composition to phase separate, and demonstrated a dramatic effect of external strain, which could considerably impact exotic topological phenomena reported in this system. [Zhao, ..., Zeljkovic, Nature Physics 17, 903 (2021); cover of Nature Physics, August 2021].

(5) Imaging Dirac band curvature manipulation by magnetic field in YMn₆Sn₆

We used low-temperature spectroscopic-imaging scanning tunneling microscopy to visualize the low-energy electronic band structure of an antiferromagnetic kagome metal YMn₆Sn₆. We pinpointed several distinct features in the electronic band structure, including: (A) a sharp spectral peak, interpreted as the van Hove singularity located only ~7 meV below the Fermi level, and (B) gapped Dirac cone. Interestingly, magnetic field measurements unveil an anomalous evolution of these features, which do not split, and shift to higher energy, regardless of the relative direction of the magnetic field applied. This unusual behavior cannot be explained by spin Zeeman coupling, and may be consistent with exotic orbital magnetism expected in Chern insulators.

Training Opportunities: The project has provided a part-time support for 7 graduate students: 4 executing STM/S experiments and data analysis, and 3 performing MBE synthesis. It also provided opportunities for 5 undergraduate students (primarily supported by internal BC funds) to get involved in this project, by learning how to for example etch STM tips and analyze STM data.

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Results Dissemination: Two notable papers selected for the cover of Nature Physics (December 2019 and August 2021)

All papers published or submitted that were at least in part supported by this grant:

- (1) He Zhao, Bryan Rachmilowitz, Zheng Ren, Ruobin Han, J. Schneeloch, Ruidan Zhong, Genda Gu, Ziqiang Wang and Ilija Zeljkovic, Superconducting Proximity Effect in a Topological Insulator using Fe(Te,Se), PRB 97, 224504 (2018)
- (2) He Zhao, Zheng Ren, Bryan Rachmilowitz, John Schneeloch, Ruidan Zhong, Genda Gu, Ziqiang Wang and Ilija Zeljkovic, Charge-stripe crystal phase in an insulating cuprate, Nature Materials 18, 103 (2019)
- (3) Bryan Rachmilowitz, He Zhao, Hong Li, Alexander LaFleur, J Schneeloch, Ruidan Zhong, Genda Gu, Ilija Zeljkovic, "Proximity-induced superconductivity in a topological crystalline insulator", Physical Review B 100, 241402 (2019)
- (4) He Zhao, Sujit Manna, Zach Porter, Xiang Chen, Andrew Uzdejczyk, Jagadeesh Moodera, Ziqiang Wang, Stephen D. Wilson and Ilija Zeljkovic, "Atomic-scale fragmentation and collapse of antiferromagnetic order in a doped Mott insulator", Nature Physics 15, 1267 (2019) [Cover article, December 2019]
- (5) Lianyang Dong, He Zhao, Ilija Zeljkovic, Stephen D. Wilson and John W. Harter, "Bulk Superconductivity in FeTe_{1-x}Se_x via physiochemical pumping of excess iron", Physical Review Materials 3, 114801 (2019)
- (6) Bryan Rachmilowitz, He Zhao, Zheng Ren, Hong Li, Shang Gao, John Schneeloch, Ruidan Zhong, Genda Gu, Ilija Zeljkovic, Coulomb Blockade Effects in a Topological Insulator Grown on a High-Tc Cuprate Superconductor, npj Quantum Materials 5, 72 (2020)
- (7) He Zhao, Hong Li, Lianyang Dong, Binjie Xu, John Schneeloch, Ruidan Zhong, Minghu Fang, Genda Gu, John Harter, Stephen D. Wilson, Ziqiang Wang and Ilija Zeljkovic, Visualizing nematic instability and dramatic suppression of superconductivity in Fe(Te,Se), Nature Physics 17, 903 (2021) [Cover article, August 2021]
- (8) Hong Li, He Zhao, Kai Liu, Ziqiang Wang, Hechang Lei and Ilija Zeljkovic, Imaging orbital magnetism in a kagome antiferromagnet YMn₆Sn₆, under review

The PI gave a CIQM colloquium at Harvard University in March, 2018.

The PI gave an invited talk at the APS March Meeting in 2019 (X09.00005).

Two graduate students presented talks at the APS March Meeting in 2018 on: (1) proximity induced superconductivity in a topological insulator Bi₂Te₃ using Fe(Te,Se) and (2) the absence of proximity induced superconductivity in a topological insulator Bi₂Te₃ using high-Tc superconductor Bi₂Sr₂CaCu₂O_{8+x}. Two more graduate students presented talks at the APS March Meeting in 2019 (H33.00007 and V05.00014).

Our work (PRB 97, 224504 (2018)) was covered by Superconductor Week journal, 08/10/2018 issue.

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Ilija Zeljkovic

Person Months Worked: 3.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Shang Gao

Person Months Worked: 2.00

Project Contribution:

National Academy Member: N

Funding Support:

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Participant Type: Graduate Student (research assistant)
Participant: Alexander LaFleur
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Hong Li
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Bryan Rachmilowitz
Person Months Worked: 11.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Zheng Ren
Person Months Worked: 10.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: He Zhao
Person Months Worked: 9.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Shrinkhala Sharma
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

ARTICLES:

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Volume: 97

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First Page #: 224504

Date Submitted: 6/13/18 12:00AM

Date Published: 6/1/18 4:00AM

Publication Location:

Article Title: Superconducting proximity effect in a topological insulator using Fe(Te, Se)

Authors: He Zhao, Bryan Rachmilowitz, Zheng Ren, Ruobin Han, J. Schneeloch, Ruidan Zhong, Genda Gu, Ziqiang

Keywords: topological superconductivity, scanning tunneling microscopy, molecular beam epitaxy

Abstract: Interest in the superconducting proximity effect has recently been reignited by theoretical predictions that it could be used to achieve topological superconductivity. Low-Tc superconductors have predominantly been used in this effort, but small energy scales of ~ 1 meV have hindered the characterization of the emergent electronic phase, limiting it to extremely low temperatures. In this work, we use molecular beam epitaxy to grow topological insulator Bi₂Te₃ in a range of thicknesses on top of a high-Tc superconductor Fe(Te,Se). Using scanning tunneling microscopy and spectroscopy, we detect Δ_i as high as ~ 3.5 meV, which is the largest reported gap induced by proximity to an s-wave superconductor to date. We find that Δ_i decays with Bi₂Te₃ thickness, but remains finite even after the topological surface states have been formed. Finally, by imaging the scattering and interference of surface state electrons, we provide a microscopic visualization of the fully gapped Bi₂Te₃ SS.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nature Materials

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41563-018-0243-x

Volume: 18

Issue:

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Date Published: 12/17/18 5:00AM

Publication Location:

Article Title: Charge-Stripe Crystal Phase in an Insulating Cuprate

Authors: He Zhao, Zheng Ren, Bryan Rachmilowitz, John Schneeloch, Ruidan Zhong, Genda Gu, Ziqiang Wang

Keywords: high-Tc superconductivity, scanning tunneling microscopy

Abstract: High-temperature (high-Tc) superconductivity in cuprates arises from carrier doping of an antiferromagnetic Mott insulator. This carrier doping leads to the formation of electronic liquid-crystal phases¹. The insulating charge-stripe crystal phase is predicted to form when a small density of holes is doped into the charge-transfer insulator state¹⁻³, but this phase is yet to be observed experimentally. Here, we use surface annealing to extend the accessible doping range in Bi-based cuprates and realize the lightly doped charge-transfer insulating state of the cuprate Bi₂Sr₂CaCu₂O_{8+x}. In this insulating state with a charge transfer gap on the order of ~ 1 eV, our spectroscopic imaging scanning tunnelling microscopy measurements provide strong evidence for a unidirectional charge-stripe order with a commensurate $4a_0$ period along the Cu-O-Cu bond. Notably, this insulating charge-stripe crystal phase develops before the onset of the pseudogap and formation of the Fermi surface.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

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Volume: Issue:

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Article Title: Atomic-scale fragmentation and collapse of antiferromagnetic order in a doped Mott insulator

Authors: He Zhao, Sujit Manna, Zach Porter, Xiang Chen, Andrew Uzdejczyk, Jagadeesh Moodera, Ziqiang Wan

Keywords: spin-polarized STM, Mott insulator

Abstract: Disentangling the relationship between the insulating state with a charge gap and the magnetic order in an antiferromagnetic Mott insulator remains difficult due to inherent phase separation as the Mott state is perturbed 1–7. Measuring magnetic and electronic properties at the atomic length scales would provide crucial insight, but this is yet to be experimentally achieved. Here we use spin-polarized scanning tunneling microscopy (SP-STM) to visualize the periodic spin-resolved modulations originating from the antiferromagnetic order in a relativistic Mott insulator Sr₂IrO₄ 8,9, and how they change as a function of doping. We find that near insulator-to-metal transition (IMT), the long-range antiferromagnetic order melts into a fragmented state with short-range correlations. Crucially, we discover that this static short-range antiferromagnetic order is locally uncorrelated with the observed spectral gap magnitude. This suggests that short range antiferromagnetic correlations are unlikely.

Distribution Statement: 1-Approved for public release; distribution is unlimited.

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Publication Location:

Article Title: Proximity-induced superconductivity in a topological crystalline insulator

Authors: Bryan Rachmilowitz, He Zhao, Hong Li, Alexander LaFleur, J. Schneeloch, Ruidan Zhong, Genda Gu, Il

Keywords: MBE, topological crystalline insulator heterostructure, STM

Abstract: Superconducting topological crystalline insulators (TCIs) are predicted to host new topological phases protected by crystalline symmetries, but available materials are insufficiently suitable for surface studies. To induce superconductivity at the surface of a prototypical TCI SnTe, we use molecular beam epitaxy to grow a heterostructure of SnTe and a high-T_c superconductor Fe(Te,Se), utilizing a “buffer” layer to bridge the large lattice mismatch between SnTe and Fe(Te,Se). Using low-temperature scanning tunneling microscopy and spectroscopy, we measure a prominent spectral gap on the surface of SnTe, and demonstrate its superconducting origin by its dependence on temperature and magnetic field. Our work provides a platform for atomic-scale investigations of emergent topological phenomena in superconducting TCIs.

Distribution Statement: 1-Approved for public release; distribution is unlimited.

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Journal: Nature Physics

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Article Title: Nematic transition and nanoscale suppression of superconductivity in Fe(Te,Se)

Authors: He Zhao, Hong Li, Lianyang Dong, Binjie Xu, John Schneeloch, Ruidan Zhong, Minghu Fang, Genda G

Keywords: Fe(Te,Se), strain, electronic nematicity

Abstract: In this work, we report the experimental discovery of an unprecedented, strongly momentum-dependent g-factor in a kagome magnet YMn₆Sn₆. Using spectroscopic-imaging scanning tunneling microscopy, we map the evolution of a massive Dirac band in the vicinity of the Fermi level as a function of magnetic field. We find that electronic states at different lattice momenta exhibit markedly different Zeeman energy shifts, giving rise to an anomalous g-factor that peaks around the Dirac point. This strongly suggests a direct observation of the magnetic field coupling to the large orbital magnetization near gapped Dirac points induced by the non-trivial Berry curvature of the Bloch electron wave functions. The k-dependent renormalization of the g-factor enables fine control of the electronic structure by magnetic field, which should be highly relevant to topological systems with pronounced Berry curvature variations.

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Acknowledged Federal Support: Y

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Journal: Nature

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Article Title: Momentum-dependent g-factor and orbital magnetization in a kagome magnet YMn₆Sn₆

Authors: Hong Li, He Zhao, Kun Jiang, Qi Wang, Qiangwei Yin, Ning-Ning Zhao, Kai Liu, Ziqiang Wang, Hechang

Keywords: kagome magnet, orbital magnetization, Berry curvature

Abstract: In this work, we report the experimental discovery of an unprecedented, strongly momentum-dependent g-factor in a kagome magnet YMn₆Sn₆. Using spectroscopic-imaging scanning tunneling microscopy, we map the evolution of a massive Dirac band in the vicinity of the Fermi level as a function of magnetic field. We find that electronic states at different lattice momenta exhibit markedly different Zeeman energy shifts, giving rise to an anomalous g-factor that peaks around the Dirac point. This strongly suggests a direct observation of the magnetic field coupling to the large orbital magnetization near gapped Dirac points induced by the non-trivial Berry curvature of the Bloch electron wave functions. The k-dependent renormalization of the g-factor enables fine control of the electronic structure by magnetic field, which should be highly relevant to topological systems with pronounced Berry curvature variations.

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Acknowledged Federal Support: Y

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Partners

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Genda Gu, BNL: provided Fe(Te,Se) and Bi-2212 bulk single crystals for our heterostructure growth.

I certify that the information in the report is complete and accurate:

Signature: Ilija Zeljkovic

Signature Date: 12/3/21 1:40PM

The main accomplishments and publications are listed within the body of the report.