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RPPR Final Report
as of 03-May-2023

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Proposal Number: 70202EM
Agreement Number: W911NF-17-1-0542

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Report Term: 0-Other
Submitted By: Keji Lai
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STEM Degrees: 4
STEM Participants: 4

Major Goals: The goal of this project is to locally probe the nanoscale conductivity distribution of quantum spin Hall and quantum anomalous Hall systems, with special focus on their topological edge channels, by near-field microwave impedance microscopy (MIM). The boundaries of these exotic quantum states can support dissipationless electrical transport and may serve as perfectly conducting channels in radio-frequency nanoelectronic and spintronic devices. While both effects have been successfully demonstrated by macroscopic measurements, much remains to be explored on the microscopic details of the edge states. Local probing of these exotic quantum states by the low-temperature MIM is there not only of great academic interest but also of technological importance to realize high-speed and low-power electronics for defense applications.

Accomplishments: See the uploaded PDF file for details.

Training Opportunities: The educational program focuses on the training of doctoral and undergraduate students. In total, two graduate students (one full time and the other part time) and two undergraduate students (both part time) have worked on this project. All four of them have received STEM degrees (one PhD, one Master, two Bachelors) under the support of this program. All participants in the program have the opportunity to learn the basic microwave network analysis, transport measurements, and scanning probe techniques in the laboratory. The skills will benefit them in their future endeavors in different scientific disciplines. The interpretation of the real-space conductivity images at low temperatures and under high magnetic fields requires a thorough understanding of near-field electromagnetic interactions and the relevant physics of quantum materials, which are of great interest to the DoD. The PhD student also had the opportunity to present his work in professional conferences like American Physical Society March Meeting.

Results Dissemination: 1. Three papers with the ARO support acknowledged have been published in peer-reviewed articles, including
1.1 Direct imaging of sketched conductive nanostructures at the LaAlO3/SrTiO3 interface, Applied Physics Letters 111, 233104 (2017).
1.2 Evidence for a higher-order topological insulator in a three-dimensional material built from van der Waals stacking of bismuth-halide chains, Nature Materials 20, 473–479 (2021).
1.3 Implementing Microwave Impedance Microscopy in a Dilution Refrigerator, Review of Scientific Instruments, 94, 053701 (2023).

2. Results supported by this program have been presented in the following conference talks. The ARO support was acknowledged in all presentations.
2.1 Two contributed talks (2018 and 2023) in the American Physical Society March Meeting, given by the PhD student.
2.2 One invited talk (2019) in the American Physical Society March Meeting, given by the PI.
2.3 One invited talk in the High Frequency Scanning Probe Microscopy Workshop, hosted by NIST, Boulder CO, in 2019.

3. Public outreach events.
3.1 The PI gave talks in the departmental “Alice in Wonderland” summer research experience program for high-school girls (2018 and 2019). The ARO support was acknowledged.

Honors and Awards: Trull Centennial Professorship in Physics, University of Texas at Austin

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: Keji Lai
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Zhanzhi Jiang
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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

Participant Type: Undergraduate Student
Participant: Shahin Jahanbani
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:

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National Academy Member: N

Participant Type: Undergraduate Student
Participant: Dishan Abeysinghe
Person Months Worked: 1.00 **Funding Support:**
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Date Submitted: 7/28/21 12:00AM Date Published: 12/2/17 12:00AM
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Article Title: Direct imaging of sketched conductive nanostructures at the LaAlO
Authors: Zhanzhi Jiang, Xiaoyu Wu, Hyungwoo Lee, Jung-Woo Lee, Jianan Li, Guanglei Cheng, Chang-Beom E
Keywords: microwave impedance microscopy, electron gas, oxide interface
Abstract: Low-dimensional van der Waals materials have been extensively studied as a platform with which to generate quantum effects. Advancing this research, topological quantum materials with van der Waals structures are currently receiving a great deal of attention. Here, we use the concept of designing topological materials by the van der Waals stacking of quantum spin Hall insulators. Most interestingly, we find that a slight shift of inversion centre in the unit cell caused by a modification of stacking induces a transition from a trivial insulator to a higher-order topological insulator. Based on this, we present angle-resolved photoemission spectroscopy results showing that the real three-dimensional material Bi4Br4 is a higher-order topological insulator. Our demonstration that various topological states can be selected by stacking chains differently, combined with the advantages of van der Waals materials, offers a playground for engineering topologically non-trivial edge states toward
Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
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Article Title: Evidence for a higher-order topological insulator in a three-dimensional material built from van der Waals stacking of bismuth-halide chains
Authors: Ryo Noguchi, Masaru Kobayashi, Zhanzhi Jiang, Kenta Kuroda, Takanari Takahashi, Zifan Xu, Daehun
Keywords: higher-order topological insulator, van der Waals, angle-resolved photoemission spectroscopy, scanning probe microscopy
Abstract: Low-dimensional van der Waals materials have been extensively studied as a platform with which to generate quantum effects. Advancing this research, topological quantum materials with van der Waals structures are currently receiving a great deal of attention. Here, we use the concept of designing topological materials by the van der Waals stacking of quantum spin Hall insulators. Most interestingly, we find that a slight shift of inversion centre in the unit cell caused by a modification of stacking induces a transition from a trivial insulator to a higher-order topological insulator. Based on this, we present angle-resolved photoemission spectroscopy results showing that the real three-dimensional material Bi_4Br_4 is a higher-order topological insulator. Our demonstration that various topological states can be selected by stacking chains differently, combined with the advantages of van der Waals materials, offers a playground for engineering topologically non-trivial edge states toward
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Publication Location:
Article Title: Implementing microwave impedance microscopy in a dilution refrigerator
Authors: Zhanzhi Jiang, Su Kong Chong, Peng Zhang, Peng Deng, Shizai Chu, Shahin Jahanbani, Kang L. Wan
Keywords: Microwave impedance microscopy, dilution refrigerator, quantum anomalous Hall effect, topological edge state
Abstract: We report the implementation of a dilution refrigerator-based scanning microwave impedance microscope with a base temperature of ?100 mK. The vibration noise of our apparatus with tuning-fork feedback control is as low as 1 nm. Using this setup, we have demonstrated the imaging of quantum anomalous Hall states in magnetically (Cr and V) doped $(Bi, Sb)_2Te_3$ thin films grown on mica substrates. Both the conductive edge modes and topological phase transitions near the coercive fields of Cr- and V-doped layers are visualized in the field-dependent results. Our study establishes the experimental platform for investigating nanoscale quantum phenomena at ultralow temperatures.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

Partners

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I certify that the information in the report is complete and accurate:
Signature: Keji Lai
Signature Date: 5/1/23 4:15PM

Final Report on DOD-Army W911NF-17-1-0542

Near-field Microwave Probing of Nontrivial Topological Boundary States

PI: Prof. Keji Lai, Department of Physics, University of Texas at Austin

Reporting Period: 09/15/2017 – 12/31/2022

Abstract: This ARO project studies the microscopic conductivity distribution of topological edge channels by near-field microwave impedance microscopy (MIM). The program is significant because it aims at the fundamental understanding of nanoscale electronic structures of topologically ordered quantum states. The spatially resolved conductivity maps provides critical feedback for material scientists to improve the sample quality and increase the onset temperatures of quantum spin Hall and quantum anomalous Hall states towards practical device applications. Moreover, the electrical properties of the edge and bulk states will be studied at the frequency spectrum of 0.1 – 10 GHz, which is the regime of interest for many defense applications.

Objectives: The goal of this project is to locally probe the nanoscale conductivity distribution of quantum spin Hall and quantum anomalous Hall systems, with special focus on their topological edge channels, by near-field microwave impedance microscopy (MIM). The specific objectives include the following. Year 1: Testing of the MIM technique and room-temperature imaging capability. Year 2 – 3: Imaging conductive edge states at the room temperature. Year 2 – 5: Implementing a dilution-refrigerator-based MIM. Year 4 – 5: Imaging conductive edge states at milli-Kelvin temperatures.

Findings:

1. Testing the room-temperature imaging capability

In Year 1 of the program, we tested the capability of imaging local conductivity at the two-dimensional gas confined in the $LaAlO_3/SrTiO_3$ interface by room-temperature MIM. The $LaAlO_3/SrTiO_3$ (LAO/STO) interface has been in the limelight of material research in the past decade. When a tip voltage is applied on 3 unit-cell of LAO grown on STO substrates, a two-dimensional electron gas (2DES) forms spontaneously at the interface. As shown in Fig. 1(a), we demonstrated the writing and imaging of 2DES by the microwave probe. The ARO support allowed us to perform the data analysis in Fig. 1(b). This work, titled "Direct imaging of sketched conductive nanostructures at the $LaAlO_3/SrTiO_3$ interface", was published as *Applied Physics Letters* **111**, 233104 (2017).

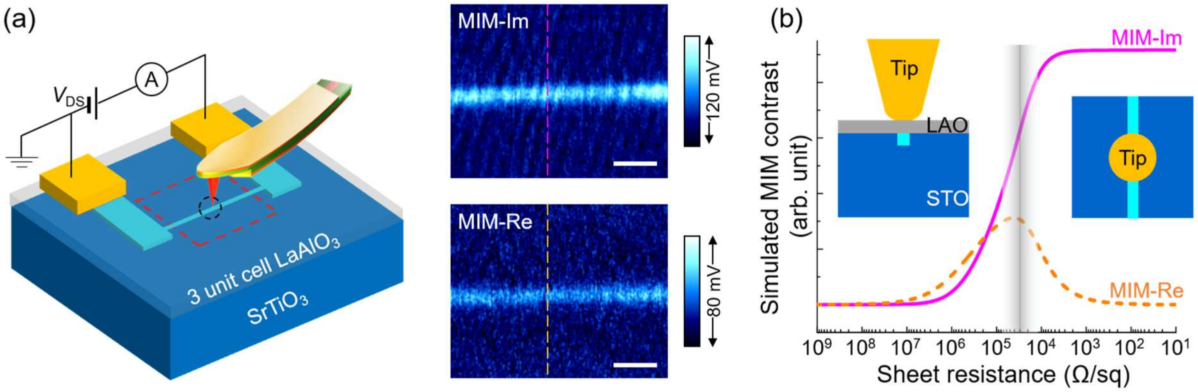

Figure 1. (a) Left: Schematic setup of the sketched device on LAO/STO interface. Right: MIM images of the written nanowire. (b) Simulated MIM signals. The shaded area indicates the effective sheet resistance of the 2DES.

2. *Imaging conductive edges at the room temperature*

Higher-order topological insulators are expected to be built from stacking quantum spin Hall insulators. In order to obtain spatially resolved information of the topological hinge states, we performed local conductivity imaging on Bi_4Br_4 by MIM. Here, the 1 GHz microwave is sent to the shielded cantilever probe, and the reflected signal is detected by the MIM electronics. Fig. 2 shows the room-temperature AFM and MIM images for ribbon-like flakes. On one side of the sample (lower edge), the MIM signal first increases as the tip approaches the flake and decreases when the tip climbs up the step, which is a clear indication of topographic artifact. On the other side (upper edge), however, the MIM map only displays strong peaks at the edges, as seen in Fig. 2b. The same line profiles in Fig. 2c are obtained in both forward and reverse raster scans, i.e., the effect due to tip twisting is minimal. The phenomenon cannot be ascribed to simple cross-talk and can be taken as signatures of the topological hinge states in Bi_4Br_4. The results are published as "Evidence for a higher-order topological insulator in a three-dimensional material built from van der Waals stacking of bismuth-halide chains", *Nature Materials* **20**, 473–479 (2021).

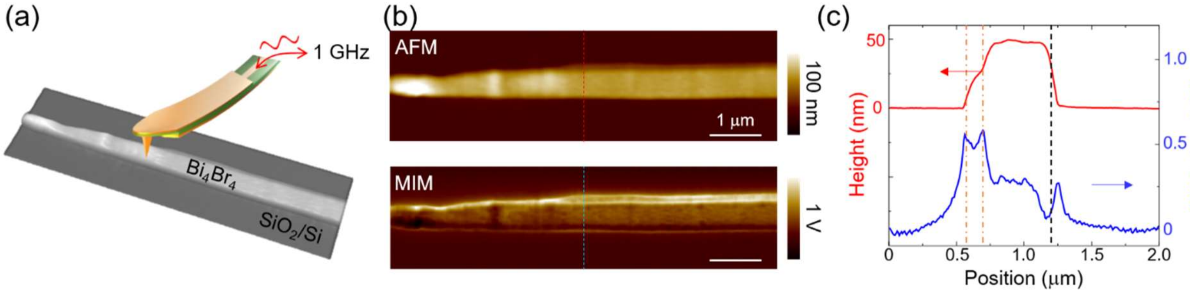

Figure 2. (**a**) Schematics of the MIM setup and Bi_4Br_4 sample on SiO_2/Si. (**b**) AFM (top) and MIM (bottom) images of an exfoliated Bi_4Br_4 thin flake. (**c**) Line profiles across the Bi_4Br_4 flake, as indicated in the surface topography and MIM images in (b). The two orange dash-dotted lines mark the edges with high MIM signals. The black dashed line marks the edge with ambipolar MIM signals due to topographic artifact.

3. *Implementing a dilution-refrigerator-based MIM*

Before this work, most cryo-MIM experiments have been performed in helium-4 (He4) cryostats with a base temperature (T) of 2 – 4 K. The only sub-Kelvin MIM work was carried out in a He3 refrigerator with a limited hold time and a base T of ~ 450 mK. In contrast, a He3/He4 dilution refrigerator (DR) will enable continuous operation at 100 mK or lower temperatures. In this ARO program, we have implemented the milli-Kelvin MIM inside a dilution refrigerator inside a liquid helium dewar with an 8 T superconducting magnet, as schematically shown in Fig. 3a. The MIM module (Fig. 3b) is mounted at the bottom of the mixing chamber of the DR. The module consists of the positioning and scanning stages, sample and tip holders, and a few microwave components. Fig. 3c shows a close-up view of the tip assembly. A quartz tuning fork is used for controlling tip-sample distance and performing topography imaging. A tungsten wire with a diameter of 25 μm is etched to a sharp tip with a radius of curvature around 100 nm at the apex. With this design, we have successfully demonstrated the simultaneous AFM and MIM imaging at a base temperature of ~ 100 mK, topographic noise of ~ 1 nm, and impedance sensitivity of ~ 0.1 aF at 1 GHz. The instrumentation results are published as “Implementing microwave impedance microscopy in a dilution refrigerator”, *Review of Scientific Instruments* **94**, 053701 (2023).

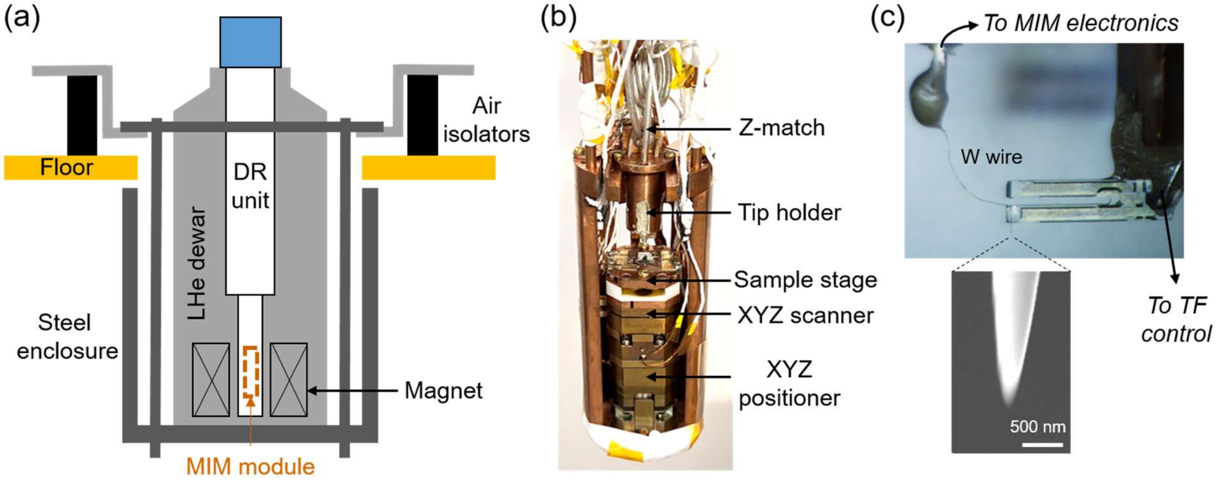

Figure 3. (a) Schematic of the dilution refrigerator and vibration isolation. (b) Picture of the MIM module with various components. (c) Close-up view of the quartz tuning fork and a tungsten wire glued to one prong. The inset shows an SEM image of the etched tip apex.

4. *Imaging conductive edge states at milli-Kelvin temperatures*

We demonstrate the milli-Kelvin MIM experiment by imaging a magnetically doped topological insulator (Bi, Sb)$_2$Te$_3$ (BST) sample grown on mica by molecular beam epitaxy (MBE). Figs. 4a and 4b show the B-field dependence of Hall (σ_{xy}) and longitudinal (σ_{xx}) conductance at 120 mK and back-gate voltage V_g = -30 V, respectively. The quantum anomalous Hall effect (QAHE) states beyond ±1 T are clearly seen from the transport data. The prominent σ_{xx} peaks are associated with the sequential magnetization flipping of Cr- and V-doped BST layers, whose coercive fields are substantially different.

Fig. 4c displays the evolution of MIM-Im signals as the B-field is swept from 2 T to −2 T along a single line across the Hall bar. The characteristic line profiles at five different fields are shown in Fig. 4d. When the system is in the C = 1 QAHE state (2 T > B > 0 T), the MIM-Im profile exhibits two prominent peaks near the physical boundaries, consistent with the existence of chiral edge modes under this condition. Around B = −0.08 T, the edge state disappears, and the sample is uniformly conductive. The MIM signal in the bulk drops slightly at B = −0.12 T and then increases again around B = −0.24 T. Beyond −1 T, the edge state reappears, as seen in the B = −2 T (C = −1 QAHE) data. The same evolution can be observed by plotting the field-dependent MIM-Im signals through the center of the sample in Fig. 4e. Here the narrow and broad peaks correspond to the magnetization flipping of Cr- and V-doped BST layers, respectively, and the dip between them represents the axion insulator state. Finally, the apparent edge width of 0.6 ~ 1 μm is likely due to the finite tip size and residual bulk conductance. Future experiments using sharper tips and better samples are expected to provide more accurate information on the dimension of the edge states. Improved spatial resolution may also allow us to visualize the internal conductive edge modes along magnetic domain walls during the topological phase transitions. The results are published in the same *Rev. Sci. Instrum*. paper discussed above.

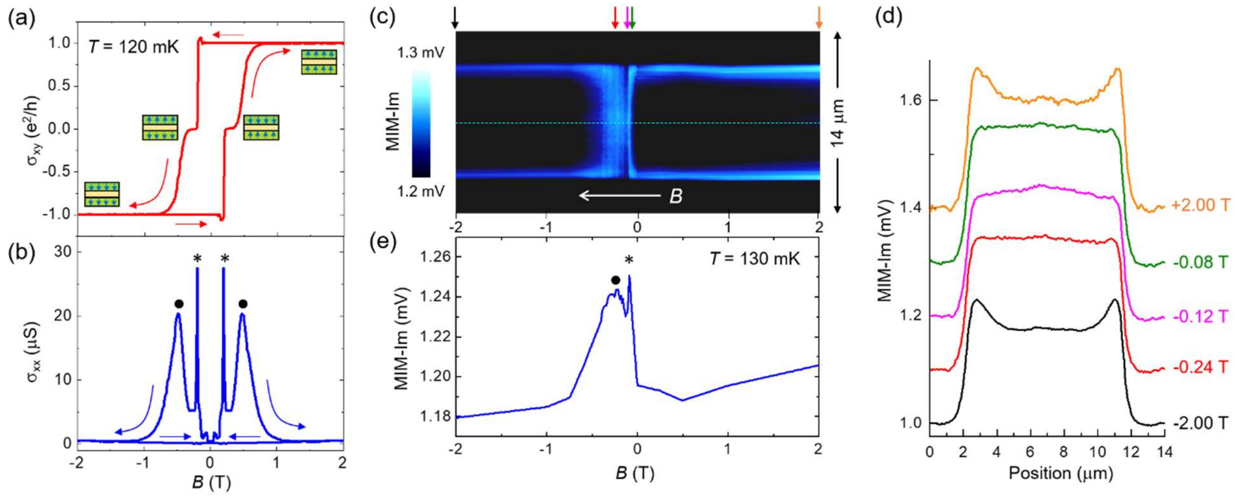

Figure 4. (a) Magnetic field dependence of σ_{xy} and (b) σ_{xx} at T = 120 mK and V_g = –30V in this sample. Arrows indicate the field sweep direction. The asterisks and dots denote the σ_{xx} peaks associated with topological phase transitions in the Cr-doped and V-doped BST layers, respectively. (c) MIM-Im line profile along the dashed line in Figure 4b as the magnetic field is swept from 2T to –2T at 130 mK. (d) Selected profiles at 5 representative fields. (e) Field-dependent MIM-Im signals at the center of the Hall bar and on the substrate. The asterisk and dot again denote the peaks associated with the switching of Cr-doped and V-doped layers.

As a final remark, thanks to the continuous support from this ARO grant, the PI's group has fully implemented the highly sophisticated DR-MIM and routinely obtained results at milli-Kelvin temperatures. We will be continuing the ultralow-temperature MIM experiments on various topological edge states, including fractional quantum Hall effect in graphene and GaAs, quantum spin Hall effect (QSHE) in WTe_2 and InAs/GaInSb, and quantum anomalous Hall effect (QAHE) in $MnBi_2Te_4$. We are confident that many publication-quality results will be achieved from this unique setup.