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University of Waterloo
200 University Avenue West

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a. REPORT	b. ABSTRACT	c. THIS PAGE			Adam Tsen
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RPPR Final Report
as of 25-May-2021

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Proposal Number: 75117ELII
Agreement Number: W911NF-19-1-0267

INVESTIGATOR(S):

Name: Adam Wei Tsen
Email: awtsen@uwaterloo.ca
Phone Number: 5198884567x30375
Principal: Y

Organization: **University of Waterloo**
Address: 200 University Avenue West, Waterloo, Ontario, N2L 3G1
Country: CAN

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Report Date: 05-Apr-2021
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Final Report for Period Beginning 06-May-2019 and Ending 05-Jan-2021
Title: Route to Quantum Anomalous Hall Effect Using Magnetic Van der Waals Heterostructures
Begin Performance Period: 06-May-2019
End Performance Period: 05-Jan-2021
Report Term: 0-Other
Submitted By: Adam Tsen
Email: awtsen@uwaterloo.ca
Phone: (519) 888-4567x30375
Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 0 **STEM Participants:** 4

Major Goals: The goals of this project are to 1) understand the nature of magnetism in the chromium trihalide family of layered magnetic insulators (CrX3, X = I, Br, Cl), and 2) interface them with intrinsically doped topological insulators in order to realize the quantum anomalous Hall effect, potentially at elevated temperatures.

Accomplishments: A number of significant discoveries were made shedding light on the nature of 2D magnetism in the chromium trihalides and demonstrating novel spintronic behavior in their device heterostructures.

1) We demonstrated a giant tunnel magnetoresistance in graphene/CrX3/graphene heterostructures that is tunable with sample thickness, bias, and temperature [Nano Letters 19, 5739-5745 (2019)].

2) At high bias, we demonstrated robust memristive swtiching in graphene/CrI3/graphene heterostructures with the unique property of being tunable with magnetic field [Advanced Materials 32, 1905433 (2020)].

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Additional findings were made on other 2D systems showing novel electronic and/or spintronic behavior.

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Training Opportunities: Four graduate students (Archana Tiwari, Tarun Patel, Shazhou Zhong, and Fangzhou Yin) were trained in the following areas during this project: nanofabrication, cryogenics, magnetotransport measurements, and optoelectronic measurements.

Results Dissemination: The following technical talks relevant to the project were delivered.

"Tunneling Probe of Two-Dimensional Magnetism," Graphene 2020, Oct. 22, 2020

"Tunneling Probe of Two-Dimensional Magnetism," Department of Physics Colloquium, California State University, Long Beach, Oct. 5, 2020

"Giant Nonlinear Anomalous Hall Effect," Spin Phenomena Interdisciplinary Center: 2D van der Waals Spin Systems Conference, Mainz, Germany, Aug. 5, 2020

"Tunneling Probe of Two-Dimensional Magnetism," Regroupement Quebecois sur les Materiaux de Point Seminar, Ecole Polytechnique de Montreal, Mar. 9, 2020

"Tunneling Probe of Two-Dimensional Magnetism," Department of Physics and Astronomy Condensed Matter Seminar, University of California, Riverside, Feb. 6, 2020

"Tunneling Probe of Two-Dimensional Magnetism," Department of Physics and Astronomy Condensed Matter Seminar, University of California, Irvine, Feb. 5, 2020

"Tunneling Probe of Two-Dimensional Magnetism," School of Mathematics and Physics Seminar, Queen's University UK, Jan. 22, 2020

"Tunneling Probe of Two-Dimensional Magnetism," Stewart Blusson Quantum Matter Institute Condensed Matter Seminar, University of British Columbia, Oct. 24, 2019

"Tunneling Probe of Two-Dimensional Magnetism," Material Research Science and Engineering Center Seminar, Columbia University, Sept 17, 2019

"Two-Dimensional Magnetism and Spintronics," UW Quantum Materials Workshop, University of Waterloo, July 26, 2019

"Two-Dimensional Magnetism and Spintronics," UW-HKUST Workshop: Emerging Quantum Technologies in Solid-State and Atomic Systems, University of Waterloo, July 23, 2019

"Two-Dimensional Magnetism and Spintronics," Department of Physics Condensed Matter Seminar, University of Virginia, May 30, 2019

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Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: Adam Wei Tsen
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

RPPR Final Report
as of 25-May-2021

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

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

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

Participant Type: Graduate Student (research assistant)
Participant: Fangzhou Yin
Person Months Worked: 3.00 **Funding Support:**
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Date Submitted: 4/20/21 12:00AM Date Published: 4/1/21 8:00AM
Publication Location:
Article Title: Giant c-axis nonlinear anomalous Hall effect in Td-MoTe2 and WTe2
Authors: Archana Tiwari, Fangchu Chen, Shazhou Zhong, Elizabeth Drueke, Jahyun Koo, Austin Kaczmarek, Co
Keywords: anomalous Hall effect, Weyl semimetal
Abstract: While the anomalous Hall effect can manifest even without an external magnetic field, time reversal symmetry is nonetheless still broken by the internal magnetization of the sample. Recently, it has been shown that certain materials without an inversion center allow for a nonlinear type of anomalous Hall effect whilst retaining time reversal symmetry. The effect may arise from either Berry curvature or through various asymmetric scattering mechanisms. Here, we report the observation of an extremely large c-axis nonlinear anomalous Hall effect in the non-centrosymmetric Td phase of MoTe2 and WTe2 without intrinsic magnetic order. We find that the effect is dominated by skew-scattering at higher temperatures combined with another scattering process active at low temperatures. Application of higher bias yields an extremely large Hall ratio of E?/E||?=?2.47 and corresponding anomalous Hall conductivity of order 8?×?10^7?S/m.
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Publication Location:
Article Title: Photocurrent Imaging of Multi-Memristive Charge Density Wave Switching in Two-Dimensional 1T-TaS
Authors: Tarun Patel, Junichi Okamoto, Tina Dekker, Bowen Yang, Jingjing Gao, Xuan Luo, Wenjian Lu, Yuping
Keywords: charge density waves, memristors, scanning photocurrent microscopy
Abstract: Transport studies of atomically thin 1T-TaS2 have demonstrated the presence of intermediate resistance states across the nearly commensurate (NC) to commensurate (C) charge density wave (CDW) transition, which can be further switched electrically. While this presents exciting opportunities for memristor applications, the switching mechanism could be potentially attributed to the formation of inhomogeneous C and NC domains. Here, we present combined electrical driving and photocurrent imaging of ultrathin 1T-TaS2 in a heterostructure geometry. While micron-sized CDW domains are seen upon cooling, electrically driven transitions are largely uniform, indicating that the latter likely induces true metastable CDW states, which we then explain by a free energy analysis. Additionally, we are able to perform repeatable and bidirectional switching across the intermediate states without changing sample temperature, demonstrating that atomically thin 1T-TaS2 can be further used as a robust and re
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Date Submitted: 4/20/21 12:00AM Date Published: 10/1/20 4:00AM
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Article Title: Tunable layered-magnetism–assisted magneto-Raman effect in a two-dimensional magnet CrI
Authors: Wencan Jin, Zhipeng Ye, Xiangpeng Luo, Bowen Yang, Gaihua Ye, Fangzhou Yin, Hyun Ho Kim, Laura
Keywords: two-dimensional layered magnetism, magneto-Raman effect, Raman spectroscopy
Abstract: We used a combination of polarized Raman spectroscopy experiment and model magnetism–phonon coupling calculations to study the rich magneto-Raman effect in the two-dimensional (2D) magnet CrI3. We reveal a layered-magnetism–assisted phonon scattering mechanism below the magnetic onset temperature, whose Raman excitation breaks time-reversal symmetry, has an antisymmetric Raman tensor, and follows the magnetic phase transitions across critical magnetic fields, on top of the presence of the conventional phonon scattering with symmetric Raman tensors in N-layer CrI3. We resolve in data and by calculations that the first-order Ag phonon of the monolayer splits into an N-fold multiplet in N-layer CrI3 due to the interlayer coupling (N?2) and that the phonons within the multiplet show distinct magnetic field dependence because of their different layered-magnetism–phonon coupling. We further find that such a layered-magnetism–phonon coupled Raman scattering mechanism extends beyond first-orde
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Volume: 11 Issue: 1 First Page #:
Date Submitted: 4/20/21 12:00AM Date Published: 9/1/20 4:00AM
Publication Location:
Article Title: Observation of the polaronic character of excitons in a two-dimensional semiconducting magnet CrI3
Authors: Wencan Jin, Hyun Ho Kim, Zhipeng Ye, Gaihua Ye, Laura Rojas, Xiangpeng Luo, Bowen Yang, Fangzh
Keywords: excitons, two-dimensional magnetism
Abstract: Exciton dynamics can be strongly affected by lattice vibrations through electron-phonon coupling. This is rarely explored in two-dimensional magnetic semiconductors. Focusing on bilayer CrI3, we first show the presence of strong electron-phonon coupling through temperature-dependent photoluminescence and absorption spectroscopy. We then report the observation of periodic broad modes up to the 8th order in Raman spectra, attributed to the polaronic character of excitons. We establish that this polaronic character is dominated by the coupling between the charge-transfer exciton at 1.96?eV and a longitudinal optical phonon at 120.6?cm?1. We further show that the emergence of long-range magnetic order enhances the electron-phonon coupling strength by ~50% and that the transition from layered antiferromagnetic to ferromagnetic order tunes the spectral intensity of the periodic broad modes, suggesting a strong coupling among the lattice, charge and spin in two-dimensional CrI3. Our study ope
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Date Submitted: 4/20/21 12:00AM Date Published: 3/1/20 5:00AM
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Article Title: Magnetic-Field-Induced Quantum Phase Transitions in a van der Waals Magnet
Authors: Siwen Li, Zhipeng Ye, Xiangpeng Luo, Gaihua Ye, Hyun Ho Kim, Bowen Yang, Shangjie Tian, Chenghe
Keywords: magnetic order, quantum phase transitions, spin dynamics
Abstract: Exploring new parameter regimes to realize and control novel phases of matter has been a main theme in modern condensed matter physics research. The recent discovery of two-dimensional (2D) magnetism in nearly freestanding monolayer atomic crystals has already led to observations of a number of novel magnetic phenomena absent in bulk counterparts. Such intricate interplays between magnetism and crystalline structures provide ample opportunities for exploring quantum phase transitions in this new 2D parameter regime. Here, using magnetic field- and temperature-dependent circularly polarized Raman spectroscopy of phonons and magnons, we map out the phase diagram of chromium triiodide (CrI3) that has been known to be a layered antiferromagnet (AFM) in its 2D films and a ferromagnet (FM) in its three-dimensional (3D) bulk. However, we reveal a novel mixed state of layered AFM and FM in 3D CrI3 bulk crystals where the layered AFM survives in the surface layers, and the FM appears in deeper
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Volume: 32 Issue: 2 First Page #: 1905433
Date Submitted: 4/20/21 12:00AM Date Published: 10/1/19 4:00AM
Publication Location:
Article Title: Magneto-Memristive Switching in a 2D Layer Antiferromagnet
Authors: Hyun Ho Kim, Shengwei Jiang, Bowen Yang, Shazhou Zhong, Shangjie Tian, Chenghe Li, Hechang Lei
Keywords: 2D magnetism, chromium triiodide, memristive switching, van der Waals heterostructures
Abstract: Memristive devices whose resistance can be hysteretically switched by electric field or current are intensely pursued both for fundamental interest as well as potential applications in neuromorphic computing and phase?change memory. When the underlying material exhibits additional charge or spin order, the resistive states can be directly coupled, further allowing electrical control of the collective phases. The observation of abrupt, memristive switching of tunneling current in nanoscale junctions of ultrathin CrI3, a natural layer antiferromagnet, is reported here. The coupling to spin order enables both tuning of the resistance hysteresis by magnetic field and electric?field switching of magnetization even in multilayer samples.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

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Date Submitted: 4/20/21 12:00AM Date Published: 7/1/19 4:00AM
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Article Title: Tailored Tunnel Magnetoresistance Response in Three Ultrathin Chromium Trihalides
Authors: Hyun Ho Kim, Bowen Yang, Shangjie Tian, Chenghe Li, Guo-Xing Miao, Hechang Lei, Adam W. Tsen
Keywords: 2D magnetism, chromium trihalides, tunnel magnetoresistance, van der Waals heterostructures
Abstract: Materials that demonstrate large magnetoresistance have attracted significant interest for many decades. Extremely large tunnel magnetoresistance (TMR) has been reported by several groups across ultrathin CrI3 by exploiting the weak antiferromagnetic coupling between adjacent layers. Here, we report a comparative study of TMR in all three chromium trihalides (CrX3, X = Cl, Br, or I) in the two-dimensional limit. As the materials exhibit different transition temperatures and interlayer magnetic ordering in the ground state, tunneling measurements allow for an easy determination of the field–temperature phase diagram for the three systems. By changing sample thickness and biasing conditions, we then demonstrate how to maximize and further tailor the TMR response at different temperatures for each material. In particular, near the magnetic transition temperature, TMR is nonsaturating up to the highest fields measured for all three compounds owing to the large, field-induced exchange coupl
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Partners

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Prof. Guoxing Miao, University of Waterloo, Canada Prof. Liuyan Zhao, University of Michigan--Ann Arbor, USAProf

I certify that the information in the report is complete and accurate:
Signature: Adam Wei Tsen
Signature Date: 4/20/21 5:52PM

Major Goals:

The goals of this project are to 1) understand the nature of magnetism in the chromium trihalide family of layered magnetic insulators (CrX_3, X = I, Br, Cl), and 2) interface them with intrinsically doped topological insulators in order to realize the quantum anomalous Hall effect, potentially at elevated temperatures.

Accomplished under Goals:

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Other Collaborators:

We collaborated with the following PI's during the course of this project.

Prof. Guoxing Miao, University of Waterloo, Canada
Prof. Liuyan Zhao, University of Michigan--Ann Arbor, USA
Prof. Rui He, Texas Tech University, USA
Prof. Kin Fai Mak, Cornell University, USA
Prof. Jie Shan, Cornell University, USA
Prof. Binghai Yan, Weizmann Institute of Science, Israel
Prof. Hechang Lei, Renmin University of China, China
Prof. Yuping Sun, Chinese Academy of Sciences, China