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RPPR Final Report
as of 20-Sep-2023

Agency Code: 21XD

Proposal Number: 76825EMYIP **Agreement Number: W911NF-20-1-0211**
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Report Date: 30-Sep-2023 Date Received: 20-Sep-2023
Final Report for Period Beginning 01-Jul-2020 and Ending 30-Jun-2023
Title: Band structure engineering of moiré van der Waals heterostructures with high pressure
Begin Performance Period: 01-Jul-2020 **End Performance Period:** 30-Jun-2023
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Submitted By: Matthew Yankowitz Email: myank@uw.edu
Phone: (206) 543-1685

Distribution Statement:

STEM Degrees: 1 **STEM Participants:** 3

Major Goals: This project aims to use mechanical deformations (e.g., high pressure) to control the electronic properties of moir van der Waals materials. One major goal of the project is to develop new experimental techniques for achieving such mechanical control, in particular the ability to apply very large pressures using diamond anvil cells. In parallel, the project aims to identify and understand the most exciting moir materials to control via these mechanical deformations. These structures are assembled from graphene and boron nitride building blocks, which are stacked upon one another with an interlayer twist angle in order to create a moir pattern. These moir materials can exhibit emergent strongly correlated and topological states of matter for specific combinations of the constituent crystals and specific twist angles between them. The properties of the moir pattern driving these interesting states can be tuned by applying mechanical deformations such as pressure to the structure.

Initially, the scope of the project was primarily limited to controlling correlated states in twisted bilayer graphene using diamond anvil pressure cells. Over the course of the project, the scope of the work has grown to include the development of additional forms of mechanical deformations (namely, uniaxial strain). It has also grown to include the discovery of correlated and topological states in novel moir vdW heterostructures. Combining these new moir materials with the newly developed experimental techniques enhances the overall impact of the research, but does not change the general aim of engineering new electronic properties in moir materials with mechanical deformations.

Accomplishments: See report in the attached PDF document.

Training Opportunities: This project supported three graduate students and one postdoctoral researcher, and provided research opportunities for two additional undergraduate researchers during the reporting period. All six junior participants received direct mentoring from the PI throughout their time working on the research, including weekly meetings and ad hoc discussions. All trainees learned nanofabrication techniques required to fabricate van der Waals heterostructure devices. The graduate students and postdoc additionally received training in cryogenic and high field transport measurements. This project has also provided professional development opportunities for the three graduate students (Minhao He, Xuetao Ma, and Manish Kumar), who presented research talks at international conferences such as the APS March Meeting.

RPPR Final Report
as of 20-Sep-2023

Results Dissemination: This work has been disseminated primarily in publications, talks at international conferences and invited colloquia and seminars delivered by the PI, as well as by a graduate students supported by the project (Minhao He, Xuetao Ma, Manish Kumar). Additionally, a poster of the results was presented at the 2023 MRS Spring Meeting by an undergraduate supported by the project (Alexander Sanchez).

A list of publications in which the research was disseminated is provided below:

1. M. He, J. Cai, Y.-H. Zhang, Y. Liu, Y. Li, T. Taniguchi, K. Watanabe, D. H. Cobden, M. Yankowitz, and X. Xu, "Symmetry-broken Chern insulators in twisted double bilayer graphene," arXiv:2109.08255 (under review at Nano Letters) (2023)

2. L. G. Pimenta Martins, R. Comin, M. J. S. Matos, M. S. C. Mazzoni, B. R. A. Neves, and M. Yankowitz, "High pressure studies of atomically-thin van der Waals materials," Applied Physics Reviews 10, 011313 (2023)

3. C.-C. Tseng, X. Ma, Z. Liu, T. Taniguchi, K. Watanabe, J.-H. Chu, and M. Yankowitz, "Anomalous Hall effect at half filling in twisted bilayer graphene," Nature Physics 18, 1038-1042 (2022)

4. M. He, Y. Zhang, Y. Li, Z. Fei, K. Watanabe, T. Taniguchi, X. Xu, and M. Yankowitz, "Competing correlated states and abundant orbital magnetism in twisted monolayer-bilayer graphene," Nature Communications 12, 4724 (2021)

5. S. Chen, M. He, Y. Zhang, V. Hsieh, Z. Fei, K. Watanabe, T. Taniguchi, D. H. Cobden, X. Xu, C. R. Dean, and M. Yankowitz, "Electrically tunable correlated and topological states in twisted monolayer-bilayer graphene," Nature Physics 17, 374-380 (2021)

6. M. He, Y. Li, J. Cai, Y. Liu, K. Watanabe, T. Taniguchi, X. Xu, and M. Yankowitz, "Symmetry breaking in twisted double bilayer graphene," Nature Physics 17, 26-30 (2021)

A list of talks in which the research was disseminated by the PI is provided below:

Invited Talks at International Conferences

1. Flat bands in twisted graphene (tutorial lecture);
Graphene Flagship Division 1 Study Day, Manchester, UK, June 2023

2. Correlated and topological states in moiré graphene multilayers;
29th International Conference on Low Temperature Physics, Sapporo, Japan – virtual, Aug. 2022

3. Strong correlations and topology in moiré structures approaching the graphitic limit;
International Conference on Frontier Materials 2022, Zhuhai, China – virtual, May 2022

4. Controlling the symmetry-broken states in twisted graphene multilayers;
IOP CMD 29, Manchester, UK – virtual, Oct. 2021

5. Strong correlations and topology in graphene-based moiré quantum materials;
PNWAVS 2021, Corvallis, OR – virtual, Sep. 2021

6. Moiré van der Waals materials;
APS March Meeting, (Lee Osheroff Richardson Prize Talk – virtual), Mar. 2021

7. Tunable correlated and topological states in twisted monolayer-bilayer graphene;
GrapheneforUS 2021, New York, NY – virtual, Feb. 2021

Invited Colloquia/Seminars

1. Moiré modification of graphitic thin films;
Condensed Matter Seminar (LMU Munich), Munich, Germany, June 2023

2. “Quantum alchemy” with two-dimensional materials;
Physics Colloquium (Oregon State University), Corvallis, OR, May 2023

3. Graphene: the carbon quantum chameleon;
Physics Colloquium (University of Washington, Bothell), Bothell, WA, Oct. 2022

4. Symmetry-broken states in twisted graphene multilayers;
Condensed Matter Theory Forum (University of Oxford – virtual), Oxford, UK, Nov. 2021

5. Symmetry-broken states in twisted graphene multilayers;
Condensed Matter Seminar (Washington University in St. Louis), St. Louis, MO, Nov. 2021

6. Tunable correlated and topological states in twisted graphene heterostructures;
Condensed Matter Seminar (UT Austin – virtual), Austin, TX, April 2021

7. Tunable correlated and topological states in twisted graphene heterostructures;
Physics Colloquium (George Mason University – virtual), Fairfax, VA, Feb. 2021

8. Tunable correlated and topological states in twisted graphene heterostructures;
Quantum Foundry Seminar (UC Santa Barbara – virtual), Santa Barbara, CA, Feb. 2021

9. Tunable correlated and topological states in twisted graphene heterostructures;
Condensed Matter Seminar (Case Western Reserve University – virtual), Cleveland, OH, Sept. 2020

10. Tunable correlated and topological states in twisted graphene heterostructures;
CAMP Seminar (Penn State University – virtual), State College, PA, Aug. 2020

A list of talks in which the research was disseminated by supported graduate students is provided below:

1. Technical advances in high pressure measurements of twisted graphene heterostructures;
APS March Meeting, Mar. 2023, Speaker: Manish Kumar

2. Uniaxial strain control of Bernal bilayer graphene;
APS March Meeting, Mar. 2023, Speaker: Xuetao Ma

3. Anomalous Hall effect near half-filling in twisted bilayer graphene;
APS March Meeting, Mar. 2022, Speaker: Xuetao Ma

4. Correlated and topological states in twisted graphene multilayers;
APS March Meeting, (virtual), Mar. 2021, Speaker: Minhao He

5. Strain tuning of 2D van der Waals heterostructures;
APS March Meeting, (virtual), Mar. 2021, Speaker: Xuetao Ma

Honors and Awards: 1. IUPAP Early Career Scientist Prize in Low Temperature Physics (2022)
2. NSF CAREER Award (2021)
3. Lee Osheroff Richardson Science Prize, Oxford Instruments (2021)

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PARTICIPANTS:

Participant Type: PD/PI
Participant: Matthew Yankowitz
Person Months Worked: 1.00 **Funding Support:**

RPPR Final Report
as of 20-Sep-2023

Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Manish Kumar
Person Months Worked: 4.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Xiaozhe Zhu
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Alexander Sanchez
Person Months Worked: 1.00 **Funding Support:**
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Date Submitted: 4/30/21 12:00AM Date Published: 9/14/20 2:00PM
Publication Location:
Article Title: Symmetry breaking in twisted double bilayer graphene
Authors: Minhao He, Yuhao Li, Jiaqi Cai, Yang Liu, K. Watanabe, T. Taniguchi, Xiaodong Xu, Matthew Yankowitz
Keywords: correlations, symmetry breaking, twisted double bilayer graphene
Abstract: The flat bands that appear in some twisted van der Waals heterostructures provide a setting in which strong interactions between electrons lead to a variety of correlated phases. In particular, heterostructures of twisted double bilayer graphene host correlated insulating states that can be tuned by both the twist angle and an external electric field. Here, we report transport measurements of twisted double bilayer graphene with which we examine the fundamental role of spontaneous symmetry breaking in its phase diagram. The metallic states near each of the correlated insulators exhibit abrupt drops in their resistivity as the temperature is lowered, along with associated nonlinear current–voltage characteristics. Despite qualitative similarities to superconductivity, the simultaneous reversals in the sign of the Hall coefficient point instead to spontaneous symmetry breaking as the origin of the abrupt resistivity drops, whereas Joule heating seems to underlie the nonlinear transport.
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Date Submitted: 4/30/21 12:00AM Date Published: 10/12/20 7:00AM
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Article Title: Electrically tunable correlated and topological states in twisted monolayer–bilayer graphene
Authors: Shaowen Chen, Minhao He, Ya-Hui Zhang, Valerie Hsieh, Zaiyao Fei, K. Watanabe, T. Taniguchi, Davic
Keywords: correlations, topology, orbital magnetism, twisted monolayer-bilayer graphene
Abstract: Twisted van der Waals heterostructures with flat bands have recently emerged as a platform for realizing correlated and topological states with a high degree of control and tunability. In graphene-based moiré heterostructures, the correlated phase diagram and band topology depend on the number of graphene layers and the external environment from the encapsulating crystals. Here, we report that the system of twisted monolayer–bilayer graphene (tMBG) hosts a variety of correlated metallic and insulating states, as well as topological magnetic states. Because of its low symmetry, the phase diagram of tMBG depends on the orientation of an external perpendicular electric field. In one direction, we observe correlated states that undergo an orbitally driven insulating transition above a critical perpendicular magnetic field. In the other, we observe the emergence of electrically tunable ferromagnetism at one-quarter filling of the conduction band, and an associated anomalous Hall effect.
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Article Title: Competing correlated states and abundant orbital magnetism in twisted monolayer-bilayer graphene
Authors: Minhao He, Ya-Hui Zhang, Yuhao Li, Zaiyao Fei, Kenji Watanabe, Takashi Taniguchi, Xiaodong Xu, Ma
Keywords: correlations, topology, orbital magnetism, twisted monolayer-bilayer graphene
Abstract: Flat band moiré superlattices have recently emerged as unique platforms for investigating the interplay between strong electronic correlations, nontrivial band topology, and multiple isospin 'flavor' symmetries. Twisted monolayer-bilayer graphene (tMBG) is an especially rich system owing to its low crystal symmetry and the tunability of its bandwidth and topology with an external electric field. Here, we find that orbital magnetism is abundant within the correlated phase diagram of tMBG, giving rise to the anomalous Hall effect (AHE) in correlated metallic states nearby most odd integer fillings of the flat conduction band, as well as correlated Chern insulator states at high field. The behavior of the states at zero field appears to be inconsistent with simple spin/valley polarization, and instead may result from an intervalley coherent state that breaks time reversal symmetry. The application of a magnetic field further tunes the competition between correlated states.
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Date Submitted: 9/24/21 12:00AM Date Published:
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Article Title: Chirality-dependent topological states in twisted double bilayer graphene
Authors: Minhao He, Jiaqi Cai, Ya-Hui Zhang, Yang Liu, Yuhao Li, Takashi Taniguchi, Kenji Watanabe, David Co
Keywords: correlations, topology, stacking chirality, twisted double bilayer graphene
Abstract: The properties of van der Waals (vdW) crystals and heterostructures depend sensitively on their layer stacking configuration. The twist angle and lattice mismatch between constituent vdW sheets have been shown to be crucial parameters influencing the strongly correlated and topological states of matter in moiré materials. Here, we demonstrate a new approach for controlling these emergent states by altering the stacking chirality of the moiré structure. We study twisted double bilayer graphene (tDBG) in an AB-BA stacking configuration (i.e., with the component Bernal bilayers rotated by nearly 60°) and observe topological and symmetry-broken states that are absent in AB-AB stacked tDBG. We observe an anomalous Hall effect at 3 electrons per moiré cell, and robust symmetry-broken Chern insulator states at 7/2 filling. Our results motivate future experiments in which the stacking chirality is employed as an important new degree of freedom for controlling correlated and topological states.
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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
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Volume: Issue: First Page #:
Date Submitted: 8/12/22 12:00AM Date Published: 8/11/22 7:00AM
Publication Location:
Article Title: Anomalous Hall effect at half filling in twisted bilayer graphene
Authors: Chun-Chih Tseng, Xuetao Ma, Zhaoyu Liu, Kenji Watanabe, Takashi Taniguchi, Jiun-Haw Chu, Matthev
Keywords: ferromagnetism, anomalous Hall effect, twisted bilayer graphene
Abstract: Magic-angle twisted bilayer graphene (tBLG) displays a variety of strongly correlated phenomena. In particular, the anomalous Hall effect has been observed when the bands are filled with an odd number of electrons per moiré unit cell, indicating the emergence of a zero-field orbital magnetic state with spontaneously broken time-reversal symmetry. Here we present measurements of two tBLG devices with twist angles slightly away from the magic angle and report the observation of the anomalous Hall effect at half filling of both the electron and hole moiré bands. We suggest that two factors—the increased band dispersion away from the magic angle, and substrate potentials from the encapsulating boron nitride—probably play critical roles in stabilizing a valley-polarized ground state at half filling. Our findings further expand the rich correlated phase diagram of tBLG, and indicate the need to develop a more complete understanding of its manifold of competing symmetry-breaking orders.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

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Article Title: High-pressure studies of atomically thin van der Waals materials
Authors: Luiz G. Pimenta Martins, Riccardo Comin, Matheus J. S. Matos, Mário S. C. Mazzoni, Bernardo R. A. N
Keywords: van der Waals, pressure, two-dimensional materials
Abstract: Two-dimensional (2D) materials and their moiré superlattices represent a new frontier for quantum matter research due to the emergent properties associated with their reduced dimensionality and extreme tunability. The properties of these atomically thin van der Waals (vdW) materials have been extensively studied by tuning a number of external parameters such as temperature, electrostatic doping, magnetic field, and strain. However, so far pressure has been an under-explored tuning parameter in studies of these systems. The relative scarcity of high-pressure studies of atomically thin materials reflects the challenging nature of these experiments, but, concurrently, presents exciting opportunities for discovering a plethora of unexplored new phenomena. Here, we review ongoing efforts to study atomically thin vdW materials and heterostructures using a variety of high-pressure techniques, including diamond anvil cells, piston cylinder cells, and local scanning probes.
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Acknowledged Federal Support: **Y**

Partners

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I certify that the information in the report is complete and accurate:
Signature: Matthew Yankowitz
Signature Date: 9/20/23 12:30PM

Abstract

The ability to create designer heterostructures comprising various atomically-thin van der Waals (vdW) materials holds significant promise for realizing new electronic properties in a highly tunable condensed matter platform. The properties of these vdW heterostructures can be modified using a number of experimental knobs, including the twist angle between neighboring crystals and their interlayer hybridization strength. This project used mechanical deformations (e.g., high pressure and strain) to control the electronic properties of moiré van der Waals materials. One major goal of the project was to develop new experimental techniques for achieving such mechanical control, in particular the ability to apply very large pressures using diamond anvil cells, and to apply uniaxial strain with a home-built piezo-based device. In parallel, the project aimed to identify and understand the most exciting moiré materials to control via these mechanical deformations. These structures are assembled from graphene and boron nitride building blocks, which are stacked upon one another with an interlayer twist angle in order to create a moiré pattern. These moiré materials can exhibit emergent strongly correlated and topological states of matter for specific combinations of the constituent crystals and specific twist angles between them, and these states are highly sensitive to pressure and strain.

Objectives

Year 1

1. Develop diamond anvil cell pressure control of twisted bilayer graphene
2. Develop uniaxial strain control of twisted bilayer graphene
3. Develop pressure control of correlated states in twisted double bilayer graphene
4. Investigate the nature of isospin symmetry breaking in twisted double bilayer graphene
5. Investigate topological charge density wave states in twisted double bilayer graphene
6. Investigate correlated topological states in twisted monolayer-bilayer graphene

Year 2

1. Develop diamond anvil cell pressure control of twisted bilayer graphene
2. Develop uniaxial strain control of twisted bilayer graphene
3. Investigate topological charge density wave states in twisted double bilayer graphene
4. Investigate the anomalous Hall effect at half filling in twisted bilayer graphene

Year 3

1. Develop diamond anvil cell pressure control of twisted bilayer graphene
2. Develop uniaxial strain control of twisted bilayer graphene
3. Develop uniaxial strain control of Bernal bilayer graphene
4. Investigate topological charge density wave states in twisted double bilayer graphene

Findings

Develop diamond anvil cell pressure control of twisted bilayer graphene

We made significant progress in developing a new experimental technique allowing us to perform high-pressure measurements above 8 GPa on vdW heterostructure devices in a diamond anvil cell (DAC) pressure apparatus. This required developing new experimental techniques to fabricate our samples in the ultra-small sample space of the DAC, with typical diameter of only a few hundred micrometers. In particular, we deposited our samples directly onto the diamond surface and made in situ electrical

connections to create multiple sample electrodes and gates (see images in Figs 1a-b). Figure 1c shows a measurement of the resistance of a twisted bilayer graphene (tBLG) sample at the charge neutrality point as a function of pressure in a DAC. We observe a non-monotonic evolution of the resistance matching well with theoretical expectations, indicating that we are tuning the tBLG through a flat band condition. This will allow us to investigate the correlated and superconducting physics of tBLG in a regime of ultra-strong many-body interactions, which has so far been inaccessible under ambient conditions. These measurements were performed in collaboration with Prof. Wendy Mao's group at Stanford, who are experts in DAC operation. The ability to perform DAC measurements will be a major advance for this field.

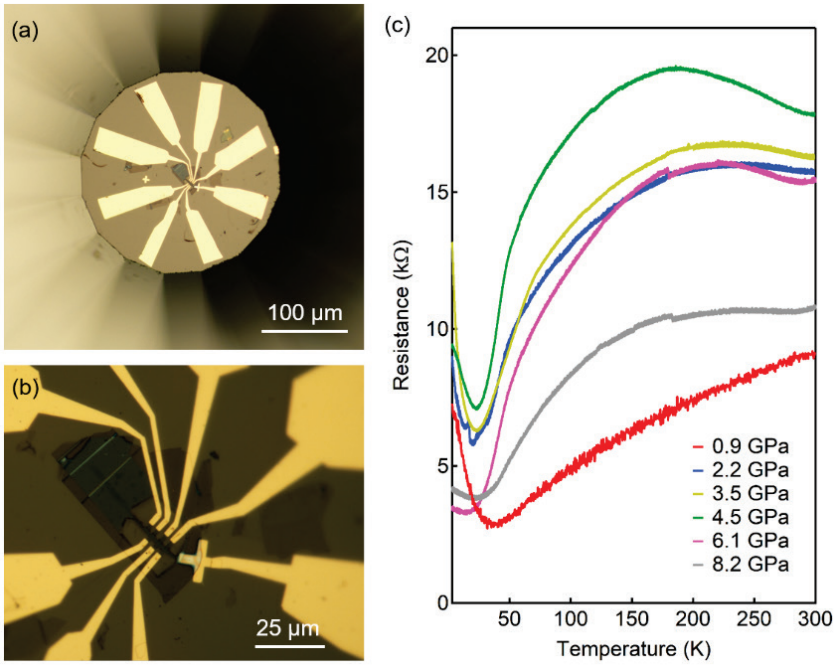

Figure 1. Tuning the flat bands of tBLG with pressure in a DAC. (a) Optical micrograph of a tBLG device fabricated onto a diamond culet. (b) Zoomed-in image of the device. (c) Resistance of a tBLG device with a twist angle of ~1.5° as a function of temperature acquired at the charge neutrality point measured at different pressures in a DAC. The resistance exhibits a non-monotonic dependence on pressure as anticipated theoretically, indicating that pressure tunes through a maximally-flat band hosting strongly correlated states.

Develop pressure control of correlated states in twisted double bilayer graphene

We used a piston-cylinder pressure cell in order to apply hydrostatic pressure up to 2.5 GPa to twisted double bilayer graphene (tDBG), comprising two slightly rotated sheets of Bernal-stacked bilayer graphene. We studied devices near the optimal twist angle, in which the bands are maximally flat and host robust strongly correlated states. We found that we are able to carefully control the strength of the correlated states upon applying pressure. For sufficiently high pressure, we demonstrated the ability to suppress the correlated states entirely (Fig. 2). These findings established our ability to control the correlated states of new graphene-based moiré materials with pressure.

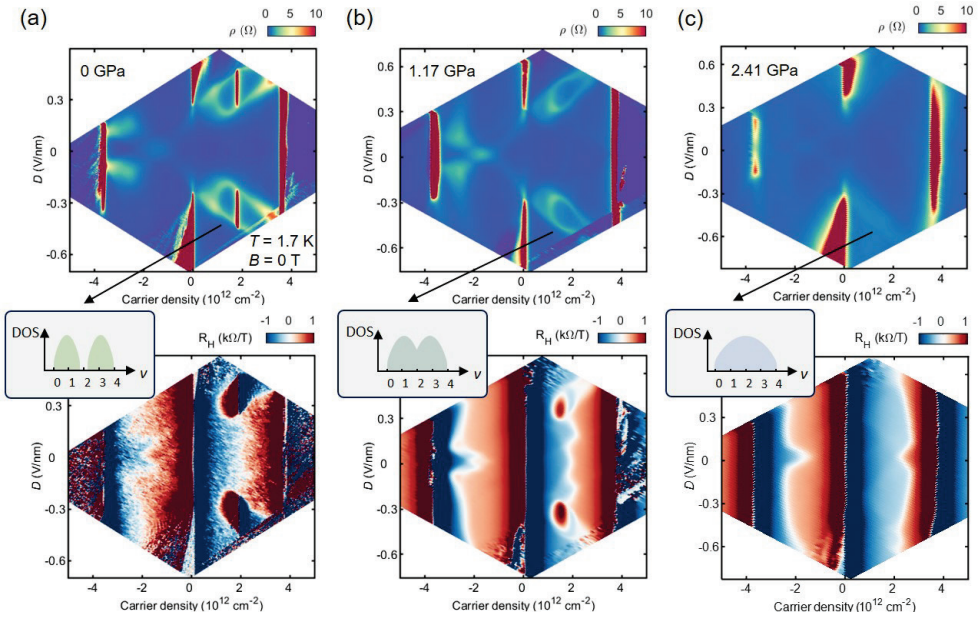

Figure 2. Tuning correlation strength in tDBG with pressure. The top row shows maps of the device resistivity, at (a) 0 GPa, (b) 1.2 GPa, and (c) 2.4 GPa, and the bottom row shows corresponding maps of the Hall resistance, for a device with θ=1.25°. Halo-like features in the conduction band (i.e., carrier densities greater than zero) at 0 GPa indicate symmetry broken correlated metallic states, enclosing a correlated insulating state at half-filling, along with a corresponding sign change in the Hall resistance. These features weaken and disappear as the pressure is raised. Cartoon insets denote the destruction of the correlated state with pressure.

Develop uniaxial strain control of twisted bilayer graphene and Bernal bilayer graphene

We developed a new experimental platform to apply uniaxial strain to moiré vdW devices, working in collaboration with Prof. Jiun-Haw's group at UW (see pictures in Fig. 3). This technique enabled us to stretch and compress vdW heterostructure devices in-plane, in contrast to the out-of-plane deformations induced by high pressure. The development of this technique gave us critical additional control over the moiré pattern: whereas pressure controls the amplitude of the moiré potential, uniaxial strain continuously tunes the shape of the moiré pattern. Importantly, the strain can be tuned continuously even down to millikelvin temperatures. In our preliminary results, we demonstrated the ability to control superconductivity and orbital magnetism in magic-angle tBLG with strain. We also identified possible signatures of quantum criticality as evidenced by an inverse temperature scaling of the elastoresistance.

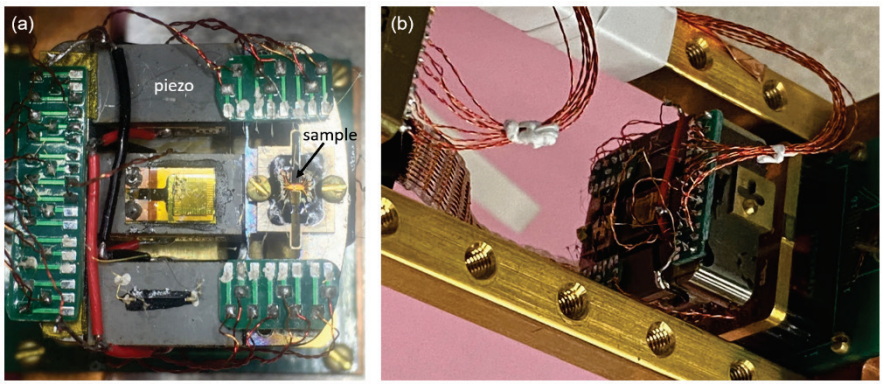

Figure 3. Pictures of the experimental strain cell setup. (a) Home-built strain cell, assembled from three piezoelectric stacks and custom titanium end-plates. The sample is affixed over a gap with a width that is tuned by the piezo expansion and contraction. The sample and strain gauges are wired to custom PCBs. (b) Strain cell mounted onto a probe compatible with dilution refrigerators.

As described in more detail later, we discovered an unexpected anomalous Hall effect (AHE) at half-filling of the flat bands of tBLG in this project. In order to further probe the origin of the AHE, we performed measurements in which we applied tunable uniaxial strain to the sample under cryogenic conditions. We found that the amplitude of the AHE could be tuned by strain, changing by over 300% (Fig. 6c). Our results may indicate that the flatness of the tBLG bands (determining the strength of interactions) is tunable by strain, leading to the observed modification of the AHE. However, we also cannot rule out an alternative explanation in which the applied strain modifies structural disorder in the sample. Further work will be necessary to determine the working mechanism of this effect.

We also studied the properties of Bernal-stacked bilayer graphene as a function of strain. Bernal-stacked bilayer graphene has been investigated for almost 20 years, but also holds new surprises. It was recently appreciated that in very high-quality devices, the flat band edge near charge neutrality can host strongly-correlated symmetry broken orders and spin-triplet superconductivity. We performed preliminary studies of the effect of uniaxial strain on the symmetry-broken states. We observed some promising strain-tuned signatures in bilayer graphene that we will investigate in more detail in the future (Fig. 4).

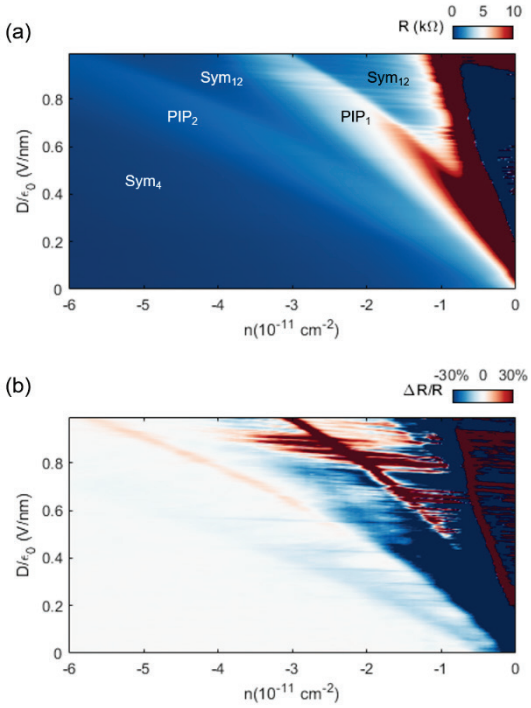

Figure 4. Strain dependence of symmetry-broken states in Bernal bilayer graphene. (a) Measurement of the resistance of hole-doped bilayer graphene as a function of doping and displacement field. Symmetry-broken phases, labeled as PIP_1 and PIP_2, indicate regions of spontaneous partial flavor polarization and correspond to an enhanced resistance. (b) Measurement of the elastoresistance of the device, which is large at the PIP_1/Sym_{12} and PIP_2/Sym_{12} boundaries, demonstrating the strain-tunability of these correlated phases.

Investigate the nature of isospin symmetry breaking in twisted double bilayer graphene

Prior work on tDBG by a number of research groups around the world identified strongly correlated states with associated transport features resembling spin-triplet superconductivity. However, the existence of superconductivity was not clear, and a number of other features of the correlated states of tDBG were poorly understood. In our work, we investigated tDBG devices with various twist angles in order to clarify its correlated phase diagram. We found that the abrupt drops in resistivity observed as the temperature is lowered are not signatures of superconductivity, but rather arise owing to spontaneous symmetry breaking due to the effects of strong correlations. Our results were the key to understanding similar transport features observed by other groups studying various moiré materials, and our understanding was

quickly adopted by the community for differentiating superconductivity from other effects of correlations. We reported these results in a publication in Nature Physics (M. He *et al.*, Nature Physics 17, 26 (2021)).

Investigate topological charge density wave states in twisted double bilayer graphene

We further pushed the frontiers of understanding and controlling the properties of tDBG by identifying a new form of symmetry breaking in this material: the formation of topological symmetry-broken Chern insulator (SBCI) states that spontaneously enlarge the moiré unit cell. Specifically, we observed SBCI states at band filling factors of 7/2 and 11/3, indicating the formation of states with spontaneously doubled and tripled areas of the moiré unit cell (Fig. 5). The former is likely a stripe phase that breaks translational and rotational symmetry, whereas the latter likely preserves the original symmetries of the moiré. A manuscript reporting these results is currently under review at Nano Letters (M. He *et al.*, arXiv:2109.08255 (2023)).

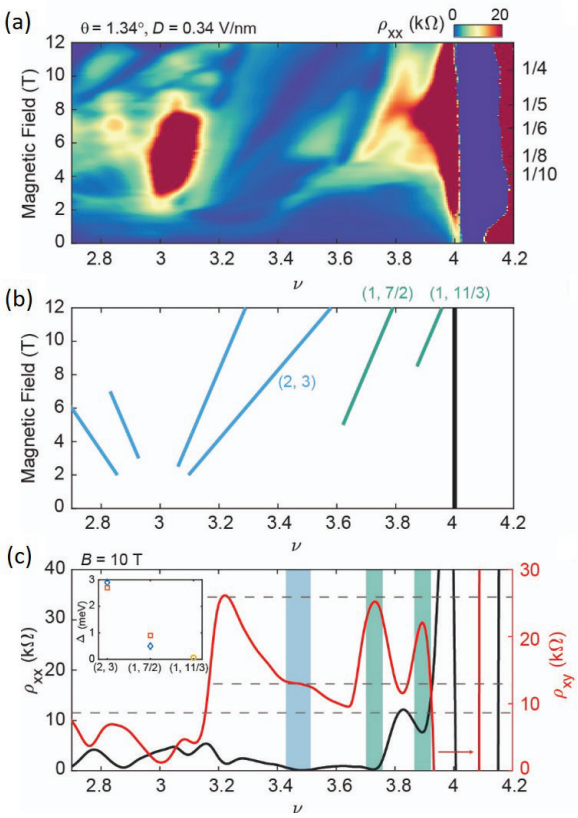

Figure 5. Symmetry-broken Chern insulator states in tDBG. (a) Landau fan diagram of the longitudinal resistivity of a tDBG device with a twist angle of 1.34° around three-quarters filling of the flat conduction band. (b) Schematic denoting the observed gapped topological states. The states denoted by the green lines are symmetry-broken Chern insulators with Chern number of 1. The 7/2 state doubles the moiré unit cell, whereas the 11/3 state triples its area. (c) Measurements of the longitudinal (black) and Hall (red) resistivities at B = 10 T showing the development of the SBCI. (inset) Measurements of the activation gap of each state, showing that the SBCI states are weak compared to the parent symmetry-broken order (2,3).

Investigate correlated topological states in twisted monolayer-bilayer graphene

We discovered twisted monolayer-bilayer graphene (tMBG) to be a new correlated moiré material. Because of its low crystal symmetry, the correlated phase diagram differs depending on whether the electric field points towards the monolayer or the bilayer graphene. We found that tMBG hosts an

emergent quantum anomalous Hall effect in a device with a twist angle of 0.89° at one-quarter filling of the moiré conduction band over a certain range of displacement field. Underlying this effect is an orbital magnetic state, in which the system becomes spontaneously magnetized owing to the coordinated orbital motion of the electrons. This state differs from conventional magnets, which are typically dominated by the electron spin. Although orbital magnetism has been seen recently in twisted bilayer graphene, it required either high magnetic field or additional alignment with a boron nitride crystal. In contrast, we found that this topological magnetic state is intrinsic in tMBG. Furthermore, we found that this topological state was destroyed in devices with slightly larger twist angles, and a novel correlated ground state characterized by intervalley coherence with broken time-reversal symmetry emerged in its place. These states are expected to host counterflow valley-exciton superfluidity, and have never been identified experimentally before. We reported these results in publications in Nature Physics (S. Chen *et al*., Nature Physics 17, 374 (2021)) and Nature Communications (M. He *et al*., Nature Communications 12, 4727 (2021)).

Investigate the anomalous Hall effect in twisted bilayer graphene

Twisted bilayer graphene has been the subject of intense research attention over the past few years, but still has the capacity to surprise. In the course of our efforts to perform pressure and strain characterization of tBLG as part of this project, we made the surprising discovery of orbital magnetism at half-filling of the flat bands (Fig. 6a-b). This ferromagnetism manifests as an anomalous Hall effect in the absence of an external magnetic field. The AHE has been reported previously in a small number of tBLG devices, but only when the flat bands were filled to either quarter- or three-quarters filling. Although at first seemingly just a small difference, observing the AHE at half-filling is actually quite unexpected based upon current theoretical modeling of tBLG. The AHE is most easily understood to arise as a consequence of spontaneous valley polarization of the charge carriers. However, competing ground states without AHE are expected to have lower energy at half-filling, including spin-polarized and intervalley-coherent orders, in contrast to quarter- and three-quarters fillings which are naturally expected to support valley-polarized states. Thus, our observation of the AHE at half-filling necessitates developing a more detailed theoretical understanding of tBLG, and likely relates to the unique properties of our devices including close rotational alignment with the substrate boron nitride and a detuning of the graphene-graphene twist slightly away from the magic angle. Our observations also carry potentially profound consequences for understanding the origin of superconductivity in the material. We notably do not observe superconductivity in our devices upon doping away from half-filling, in sharp contrast with typical tBLG devices of similar twist angles. Our observation of the AHE implies that it is not favorable to Cooper pairs from a valley-polarized state, potentially owing to the large center-of-mass momentum the pairs would carry. Our work was published in Nature Physics (C.-C. Tseng *et al*., Nature Physics 18, 1038 (2022)).

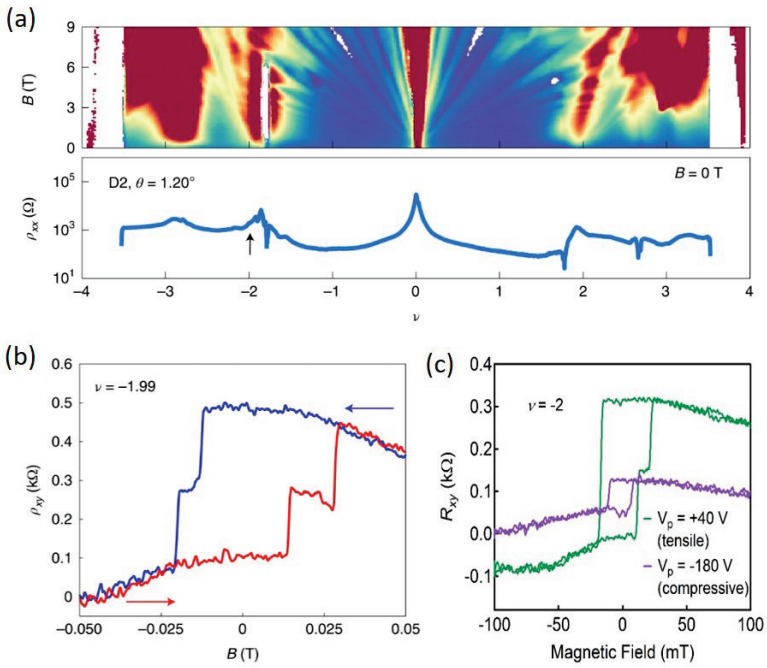

Figure 6. Anomalous Hall effect at half filling in tBLG. (a) Landau fan diagram of the longitudinal resistivity of a tBLG device with a twist angle of 1.20°. (b) Hall resistance measured as the magnetic field is swept back and forth at half filling of the flat valence band (position of the arrow in the bottom panel of (a)). The hysteresis loop indicates the AHE at half filling. (c) Hall resistance measured at different values of applied uniaxial strain. The amplitude of the AHE can by tuned by over 300% with strain.