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1. REPORT DATE (DD-MM-YYYY) 24-07-2018	2. REPORT TYPE Final Report	3. DATES COVERED (From - To) 22-Sep-2014 - 21-Sep-2017
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4. TITLE AND SUBTITLE Final Report: 4.1: Charge Density Wave in Mesoscopic 2-Dimensional Materials for Nanoelectronics	5a. CONTRACT NUMBER W911NF-14-1-0638
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER 611102

6. AUTHORS	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES Harvard University Office for Sponsored Programs 1033 Massachusetts Ave 5th Floor Cambridge, MA 02138 -5369	8. PERFORMING ORGANIZATION REPORT NUMBER
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9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	10. SPONSOR/MONITOR'S ACRONYM(S) ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S) 66399-EL.6

12. DISTRIBUTION AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.
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13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:	17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Philip Kim
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU	19b. TELEPHONE NUMBER 617-496-0714

RPPR Final Report

as of 25-Jul-2018

Agency Code:

Proposal Number: 66399EL

Agreement Number: W911NF-14-1-0638

INVESTIGATOR(S):

Name: Philip Kim
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Organization: **Harvard University**

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Country: USA

DUNS Number: 082359691

EIN: 042103580N

Report Date: 21-Dec-2017

Date Received: 24-Jul-2018

Final Report for Period Beginning 22-Sep-2014 and Ending 21-Sep-2017

Title: 4.1: Charge Density Wave in Mesoscopic 2-Dimensional Materials for Nanoelectronics

Begin Performance Period: 22-Sep-2014

End Performance Period: 21-Sep-2017

Report Term: 0-Other

Submitted By: Philip Kim

Email: pkim@physics.harvard.edu

Phone: (617) 496-0714

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

STEM Degrees: 2

STEM Participants: 1

Major Goals: In this project, we propose to investigate emergent science and novel electronic devices appeared 2-dimensional (2D) charge density waves (CDW) systems. By fabricating mesoscopic devices based on 2D CDW materials, various novel transport phenomena in new phases can be induced that were absent in bulk. The major goals of this projects are:

- Inducing electric-field dependent CDW phase transition to create nanometer scale of the embedded CDW phases to realize novel 2D transport.
- Investigation of the combined electron microscopy and in-situ transport measurement to understand mesoscopic CDW domain structures.
- Fabrication nanostructured TMDC materials and characterization of mesoscopic CDW phases in a confined space.
- Ionic liquid gating of CDW system for tuning their materials properties.
- Gate-tunable tunneling between 2D CDW systems and graphene across the van der Waals interface.

Accomplishments: Atomic lattice disorder in CDW phases in 2D limit crystalline TMD metals

Low-dimensional conductors developing charge density waves (CDW), such as 1T-TaS₂, permit unique phases that arise through electronic and structural reshaping known, respectively, as CDWs and periodic lattice distortions (PLDs). Determining the atomic structure of PLDs is critical toward understanding the origin of these charge-ordered phases and their effect on electronic properties. In this work, we revealed the microscopic nature of PLDs at cryogenic and room temperature in thin flakes of 1T-TaS₂ using atomic resolution scanning transmission electron microscopy. Real-space characterization of the local PLD structure across the phase diagram enables harnessing of emergent properties of thin transition metal dichalcogenides. This work was published in Proc. Nat. Acad. Sci. USA (PNAS) [1].

Atomic resolution electron microscopy study of periodic lattice distortions in 2D CDW near the commensuration-incommensuration transition

CDW and their concomitant periodic lattice distortions (PLD) govern the electronic properties in many layered transition-metal dichalcogenides. In particular, 1T-TaS₂ can undergo a PLD phase transition from a conducting to an insulating state as the PLD becomes commensurate with the crystal lattice. Here we directly image PLDs of the nearly-commensurate (NC) and commensurate (C) phases in thin exfoliated 1T-TaS₂ using atomic resolution cryogenic scanning transmission electron microscopy (cryo-STEM). Compared to scanning tunneling microscopy which allows mapping of CDWs at the material's surface, STEM provides direct information about the projected nuclear positions in thin films. We observed PLD superstructures in exfoliated 1T-TaS₂ samples, suggesting

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ordering of the CDWs in the out-of-plane direction. More importantly, PLDs exist in domains with different atomic lattice stacking order and their boundaries. Stacking faults in the atomic lattice should directly affect the relative alignment of the CDWs of adjacent layers and, thus, the electronic properties of 1T-TaS₂, especially in thin samples. This work was published in PNAS [2].

Nature of the quantum metal in a two-dimensional crystalline superconductor

2D materials are not expected to be metals at low temperature owing to electron localization¹. Consistent with this, pioneering studies on thin films reported only superconducting and insulating ground states, with a direct transition between the two as a function of disorder or magnetic field. However, more recent works have revealed the presence of an intermediate quantum metallic state occupying a substantial region of the phase diagram, whose nature is intensely debated. In this work, we observed such a state in the disorder-free limit of a crystalline 2D superconductor, produced by mechanical co-lamination of NbSe₂ in an inert atmosphere. Under a small perpendicular magnetic field, we induce a transition from superconductor to the quantum metal. We find a unique power-law scaling with field in this phase, which is consistent with the Bose-metal model where metallic behavior arises from strong phase fluctuations caused by the magnetic field. This work was published in Nature Physics [3].

Observation of the breakdown of the Wiedemann-Franz law in 2D Dirac systems.

Interactions between particles in quantum many-body systems can lead to the collective behavior described by hydrodynamics. One such system is the electron-hole plasma in graphene near the charge neutrality point, which can form a strongly coupled Dirac fluid. This charge-neutral plasma of quasi-relativistic fermions is expected to exhibit a substantial enhancement of the thermal conductivity, thanks to the decoupling of charge and heat currents within hydrodynamics. For this study, we developed Johnson noise thermometry for measuring electronic contribution of thermal conductivity. Employing this novel high frequency experimental technique, we observed an order of magnitude increase in the thermal conductivity and the breakdown of the Wiedemann-Franz law in the thermally populated charge-neutral plasma in graphene. This result is a signature of the Dirac fluid and constitutes direct evidence of collective motion in a quantum electronic fluid. This work was published in Science [4]

Inducing Superconducting Correlation in Quantum Hall Edge States in 2D Materials

Creating a hybrid system of a superconductor (SC) and quantum Hall (QH) states have been a long-standing experimental and theoretical goal in condensed matter physics. Recently, this idea of hybrid SC/QH systems has been received intense attentions. We have realized hybrid system between chiral edge of quantum Hall state in 2D materials with superconductor to create electronic state with non-trivial topology. We have developed a novel device scheme for a SC/QH platform by employing high-mobility graphene with transparent superconducting contacts with a high critical magnetic field. If the SC electrode is narrower than the superconducting coherence length under a quantizing magnetic field, the incoming electron is correlated to the outgoing hole along the chiral QH edge state by the Andreev process across the SC electrode. In order to realize this crossed Andreev conversion (CAC), it is necessary to fabricate highly transparent and nanometer-scale superconducting junctions to the QH system. In this experiment we reported the observation of CAC in a graphene QH system contacted with a nanostructured NbN superconducting electrode. The chemical potential of the edge states across the SC electrode exhibited a sign reversal, providing direct evidence of CAC. The result was a hallmark of crossed Andreev conversion and constitutes direct evidence of coupling of counter propagating quantum Hall edge states via Cooper pairing. This work was published in Nature Physics [5]

Training Opportunities: The project activities have involved 3 postdocs and 3 graduate students, and 1 undergraduate students, trained for interdisciplinary research activities.

Dr. Adam Wei Tsen and Dr. Gil-Ho Lee completed their project and now becomes an Assistant Professors at the University of Waterloo, Canada and at POSTECH, Korea, respectively. Ms. Rebecca Engelke received her National Science Foundation Graduate Fellowship that related to the current project. One Harvard College student, Sebastian Wagner-Carena worked on the project as an undergraduate researcher.

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Results Dissemination: 1. A. W. Tsen, R. Hovden, D. Z. Wang, Y. D. Kim, J. Okamoto, K. A. Spoth, Y. Liu, W. J. Lu, Y. P. Sun, J. Hone, L. F. Kourkoutis, P. Kim, and A. N. Pasupathy, "Structure and Control of Charge Density Waves in Two-Dimensional 1T-TaS₂," Proc. Nat. Acad. Sci. USA 112, 15054–15059 (2015)
2. R. Hovden, A. W. Tsen, P. Liu, B. H. Savitzky, I. E. Baggari, Y. Liu, W. Lu, Y. Sun, P. Kim, A. N. Pasupathy, and L. F. Kourkoutis, "Atomic lattice disorder in charge-density-wave phases of exfoliated dichalcogenides (1T-TaS₂)," Proc. Nat. Acad. Sci. USA 113, 11420–11424 (2016).
3. A. W. Tsen, B. Hunt, Y. D. Kim, Z. J. Yuan, S. Jia, R. J. Cava, J. Hone, P. Kim, C. R. Dean, and A. N. Pasupathy, "Evidence for a Bose Metal in a Two-Dimensional Crystalline Superconductor," Nature Physics 12, 208–212 (2016).
4. J. Crossno, J. K. Shi, K. Wang, X. Liu, A. Harzheim, A. Lucas, S. Sachdev, P. Kim, T. Taniguchi, K. Watanabe, T. A. Ohki, K. C. Fong, "Observation of the Dirac fluid and the breakdown of the Wiedemann-Franz law in graphene," Science 351, 1058-1061 (2016).
5. G.-H. Lee, K.-F. Huang, D. K. Efetov, D. S. Wei, S. Hart, T. Taniguchi, K. Watanabe, A. Yacoby, P. Kim, "Inducing Superconducting Correlation in Quantum Hall Edge States," Nature Physics 13, 693–698 (2017).

Honors and Awards: During the project time period, the PI received following honors:
Thomson Reuters: 2015-17 Most Highly Cited Researchers
Abigail and John Van Vleck Lecture, University of Minnesota (2017);
Robert Meservey Memorial Lecture, MIT (2016);
Rustgi Lecture, SUNY Buffalo (2015);

Protocol Activity Status:

Technology Transfer: Disseminated high mobility and low disorder graphene device and Josephson junction devices for noise measurement at BBN Raytheon.

PARTICIPANTS:

Participant Type: PD/PI

Participant: Philip Kim

Person Months Worked: 1.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Luis Jauregui

Person Months Worked: 12.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Hyobin Yoo

Person Months Worked: 6.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

RPPR Final Report
as of 25-Jul-2018

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Gil-Ho Lee

Person Months Worked: 3.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Adam Wei Tsen

Person Months Worked: 3.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Jing Shi

Person Months Worked: 12.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Artem Talanov

Person Months Worked: 3.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Rebecca Engelke

Person Months Worked: 6.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

RPPR Final Report
as of 25-Jul-2018

REPORT DOCUMENTATION PAGE

*Form Approved
OMB No. 0704-0188*

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1. REPORT DATE (DD-MM-YYYY) 07/23/2018	2. REPORT TYPE FINAL Technical	3. DATES COVERED (From - To) 09/22/2014-09/21/2017
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4. TITLE AND SUBTITLE Charge Density Waves in Mesoscopic 2-Dimensional Materials for Nanoelectronics	5a. CONTRACT NUMBER
	5b. GRANT NUMBER W911NF-14-1-0638
	5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S) Kim, Philip, Prof.	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) President and Fellows of Harvard College 1033 Massachusetts Avenue, 5th Floor Cambridge, MA 02138	8. PERFORMING ORGANIZATION REPORT NUMBER
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9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) ONRRO BOSTON 495 Summer Street, Room 627 Boston, MA 02210-2109	10. SPONSOR/MONITOR'S ACRONYM(S) ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT PUBLIC-No restrictions
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13. SUPPLEMENTARY NOTES

14. ABSTRACT In this project, we investigated various emergent physical phenomena based on correlated electronic states in 2-dimensional (2D) materials that can be utilized for novel electronic devices, such as charge density waves (CDW) and 2D superconductivity (SC). We observed effect of atomic lattice disorder in CDW phases of 2D limit transition metal dichalcogenide (TMD) crystallites, studying periodic lattice distortions in 2D CDW system. We measured electron transport in 2D crystalline superconductor under magnetic fields, revealing the nature of the low dimensional quantum metals. We also measured the electronic thermal conductivity measurement and observed the breakdown of the Wiedemann-Franz law in 2D Dirac systems. We fabricated of SC/2D quantum Hall hybrid system to demonstrate induced SC correlation in QH edge states.
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15. SUBJECT TERMS Low dimensional electronic system, charge density waves, 2D superconductivity, transition metal dichalcogenide
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16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	18. NUMBER OF PAGES 1	19a. NAME OF RESPONSIBLE PERSON Adam Ackerman
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU			19b. TELEPHONE NUMBER (Include area code) 617-495-4475

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INSTRUCTIONS FOR COMPLETING SF 298

1. REPORT DATE. Full publication date, including day, month, if available. Must cite at least the year and be Year 2000 compliant, e.g. 30-06-1998; xx-06-1998; xx-xx-1998.

2. REPORT TYPE. State the type of report, such as final, technical, interim, memorandum, master's thesis, progress, quarterly, research, special, group study, etc.

3. DATES COVERED. Indicate the time during which the work was performed and the report was written, e.g., Jun 1997 - Jun 1998; 1-10 Jun 1996; May - Nov 1998; Nov 1998.

4. TITLE. Enter title and subtitle with volume number and part number, if applicable. On classified documents, enter the title classification in parentheses.

5a. CONTRACT NUMBER. Enter all contract numbers as they appear in the report, e.g. F33615-86-C-5169.

5b. GRANT NUMBER. Enter all grant numbers as they appear in the report, e.g. AFOSR-82-1234.

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5e. TASK NUMBER. Enter all task numbers as they appear in the report, e.g. 05; RF0330201; T4112.

5f. WORK UNIT NUMBER. Enter all work unit numbers as they appear in the report, e.g. 001; AFAPL30480105.

6. AUTHOR(S). Enter name(s) of person(s) responsible for writing the report, performing the research, or credited with the content of the report. The form of entry is the last name, first name, middle initial, and additional qualifiers separated by commas, e.g. Smith, Richard, J, Jr.

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES). Self-explanatory.

8. PERFORMING ORGANIZATION REPORT NUMBER. Enter all unique alphanumeric report numbers assigned by the performing organization, e.g. BRL-1234; AFWL-TR-85-4017-Vol-21-PT-2.

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES). Enter the name and address of the organization(s) financially responsible for and monitoring the work.

10. SPONSOR/MONITOR'S ACRONYM(S). Enter, if available, e.g. BRL, ARDEC, NADC.

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14. ABSTRACT. A brief (approximately 200 words) factual summary of the most significant information.

15. SUBJECT TERMS. Key words or phrases identifying major concepts in the report.

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REPORT OF INVENTIONS AND SUBCONTRACTS
(Pursuant to "Patent Rights" Contract Clause) (See Instructions on back)

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 OMB No. 9000-0095
 Expires Jan 31, 2008*

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1.a. NAME OF CONTRACTOR/SUBCONTRACTOR		c. CONTRACT NUMBER		2.a. NAME OF GOVERNMENT PRIME CONTRACTOR		c. CONTRACT NUMBER		3. TYPE OF REPORT <i>(X one)</i>	
								a. INTERIM	b. FINAL
b. ADDRESS <i>(Include ZIP Code)</i>			d. AWARD DATE <i>(YYYYMMDD)</i>	b. ADDRESS <i>(Include ZIP Code)</i>			d. AWARD DATE <i>(YYYYMMDD)</i>	4. REPORTING PERIOD <i>(YYYYMMDD)</i>	
								a. FROM	
								b. TO	

SECTION I - SUBJECT INVENTIONS

5. "SUBJECT INVENTIONS" REQUIRED TO BE REPORTED BY CONTRACTOR/SUBCONTRACTOR *(If "None," so state)*

a. NAME(S) OF INVENTOR(S) <i>(Last, First, Middle Initial)</i>	b. TITLE OF INVENTION(S)	c. DISCLOSURE NUMBER, PATENT APPLICATION SERIAL NUMBER OR PATENT NUMBER	d. ELECTION TO FILE PATENT APPLICATIONS <i>(X)</i>				e. CONFIRMATORY INSTRUMENT OR ASSIGNMENT FORWARDED TO CONTRACTING OFFICER <i>(X)</i>	
			(1) UNITED STATES		(2) FOREIGN			
			(a) YES	(b) NO	(a) YES	(b) NO	(a) YES	(b) NO

f. EMPLOYER OF INVENTOR(S) NOT EMPLOYED BY CONTRACTOR/SUBCONTRACTOR			g. ELECTED FOREIGN COUNTRIES IN WHICH A PATENT APPLICATION WILL BE FILED		
(1) (a) NAME OF INVENTOR <i>(Last, First, Middle Initial)</i>	(2) (a) NAME OF INVENTOR <i>(Last, First, Middle Initial)</i>	(1) TITLE OF INVENTION		(2) FOREIGN COUNTRIES OF PATENT APPLICATION	
(b) NAME OF EMPLOYER	(b) NAME OF EMPLOYER				
(c) ADDRESS OF EMPLOYER <i>(Include ZIP Code)</i>	(c) ADDRESS OF EMPLOYER <i>(Include ZIP Code)</i>				

SECTION II - SUBCONTRACTS *(Containing a "Patent Rights" clause)*

6. SUBCONTRACTS AWARDED BY CONTRACTOR/SUBCONTRACTOR *(If "None," so state)*

a. NAME OF SUBCONTRACTOR(S)	b. ADDRESS <i>(Include ZIP Code)</i>	c. SUBCONTRACT NUMBER(S)	d. FAR "PATENT RIGHTS"		e. DESCRIPTION OF WORK TO BE PERFORMED UNDER SUBCONTRACT(S)	f. SUBCONTRACT DATES <i>(YYYYMMDD)</i>	
			(1) CLAUSE NUMBER	(2) DATE <i>(YYYYMM)</i>		(1) AWARD	(2) ESTIMATED COMPLETION

SECTION III - CERTIFICATION

7. CERTIFICATION OF REPORT BY CONTRACTOR/SUBCONTRACTOR *(Not required if: (X as appropriate))*

I certify that the reporting party has procedures for prompt identification and timely disclosure of "Subject Inventions," that such procedures have been followed and that all "Subject Inventions" have been reported.

a. NAME OF AUTHORIZED CONTRACTOR/SUBCONTRACTOR OFFICIAL <i>(Last, First, Middle Initial)</i>	b. TITLE	c. SIGNATURE	d. DATE SIGNED
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DD FORM 882 INSTRUCTIONS

GENERAL

This form is for use in submitting INTERIM and FINAL invention reports to the Contracting Officer and for use in reporting the award of subcontracts containing a "Patent Rights" clause. If the form does not afford sufficient space, multiple forms may be used or plain sheets of paper with proper identification of information by item number may be attached.

An INTERIM report is due at least every 12 months from the date of contract award and shall include (a) a listing of "Subject Inventions" during the reporting period, (b) a certification of compliance with required invention identification and disclosure procedures together with a certification of reporting of all "Subject Inventions," and (c) any required information not previously reported on subcontracts containing a "Patent Rights" clause.

A FINAL report is due within 6 months if contractor is a small business firm or domestic nonprofit organization and within 3 months for all others after completion of the contract work and shall include (a) a listing of all "Subject Inventions" required by the contract to be reported, and (b) any required information not previously reported on subcontracts awarded during the course of or under the contract and containing a "Patent Rights" clause.

While the form may be used for simultaneously reporting inventions and subcontracts, it may also be used for reporting, promptly after award, subcontracts containing a "Patent Rights" clause.

Dates shall be entered where indicated in certain items on this form and shall be entered in six or eight digit numbers in the order of year and month (YYYYMM) or year, month and day (YYYYMMDD). Example: April 2005 should be entered as 200504 and April 15, 2005 should be entered as 20050415.

1.a. Self-explanatory.

1.b. Self-explanatory.

1.c. If "same" as Item 2.c., so state.

1.d. Self-explanatory.

2.a. If "same" as Item 1.a., so state.

2.b. Self-explanatory.

2.c. Procurement Instrument Identification (PII) number of contract (DFARS 204.7003).

2.d. through 5.e. Self-explanatory.

5.f. The name and address of the employer of each inventor not employed by the contractor or subcontractor is needed because the Government's rights in a reported invention may not be determined solely by the terms of the "Patent Rights" clause in the contract.

Example 1: If an invention is made by a Government employee assigned to work with a contractor, the Government rights in such an invention will be determined under Executive Order 10096.

Example 2: If an invention is made under a contract by joint inventors and one of the inventors is a Government employee, the Government's rights in such an inventor's interest in the invention will also be determined under Executive Order 10096, except where the contractor is a small business or nonprofit organization, in which case the provisions of 35 U.S.C. 202(e) will apply.

5.g.(1) Self-explanatory.

5.g.(2) Self-explanatory with the exception that the contractor or subcontractor shall indicate, if known at the time of this report, whether applications will be filed under either the Patent Cooperation Treaty (PCT) or the European Patent Convention (EPC). If such is known, the letters PCT or EPC shall be entered after each listed country.

6.a. Self-explanatory.

6.b. Self-explanatory.

6.c. Self-explanatory.

6.d. Patent Rights Clauses are located in FAR 52.227.

6.e. Self-explanatory.

6.f. Self-explanatory.

7. Certification not required by small business firms and domestic nonprofit organizations.

7.a. through 7.d. Self-explanatory.

“Charge Density Waves in Mesoscopic 2-Dimensional Materials for Nanoelectronics”

7/17/2018

Name of Principal Investigators (PI): Philip Kim (PI)

- e-mail address : pkim@physics.harvard.edu
- Institution : Harvard University
- Mailing Address : 11 Oxford Street, Cambridge, MA 02138
- Phone : 617-496-0714
- Fax : 617- 495-0416

Period of Performance: September 22, 2014 – September 21, 2017

Foreword:

In this project, we investigated various emergent physical phenomena based on correlated electronic states in 2-dimensional (2D) materials that can be utilized for novel electronic devices, such as charge density waves (CDW) and 2D superconductivity (SC). We observed effect of atomic lattice disorder in CDW phases of 2D limit transition metal dichalcogenide (TMD) crystallites, studying periodic lattice distortions in 2D CDW system. We measured electron transport in 2D crystalline superconductor under magnetic fields, revealing the nature of the low dimensional quantum metals. We also measured the electronic thermal conductivity measurement and observed the breakdown of the Wiedemann-Franz law in 2D Dirac systems. We fabricated of SC/2D quantum Hall hybrid system to demonstrate induced SC correlation in QH edge states.

Statement of the problem studied:

On the mesoscopic scale, which bridges the atomistic or microscopic scale to macroscopic scales in the bulk, quantum confinement provides a rich playground for the study of emergent phenomena. Many-body electron interaction, commensuration versus incommensuration with the lattice interactions, and phase coherence in mesoscopic systems are some of the key elements to understand new collective phenomena that might be relevant for novel nanoelectronics applications. To study these topics, we fabricated mesoscopic devices based on 2D CDW/ SC materials, and studied novel transport phenomena in new quantum phases can be induced that were absent in bulk. In particular, we studied:

- Effect of atomic lattice disorder in CDW phases of 2D limit transition metal dichalcogenide (TMD) crystallites.
- Mapping periodic lattice distortions in 2D CDW system with atomic resolution electron microscopy.
- Electron transport in 2D crystalline superconductor to understand the nature of the low dimensional quantum metals.
- Development noise thermometry for electronic thermal conductivity measurement and observation of the breakdown of the Wiedemann-Franz law in 2D Dirac systems.
- Fabrication of SC/2D quantum Hall (QH) hybrid system and demonstration of induced SC correlation in QH edge states.

Summary of the most important results:

Atomic lattice disorder in CDW phases in 2D limit crystalline TMD metals

Low-dimensional conductors developing charge density waves (CDW), such as 1T-TaS₂, permit unique phases that arise through electronic and structural reshaping known, respectively, as CDWs and periodic lattice distortions (PLDs). Determining the atomic structure of PLDs is critical toward understanding the origin of these charge-ordered phases and their effect on electronic properties. In this work, we revealed the microscopic nature of PLDs at cryogenic and room temperature in thin flakes of 1T-TaS₂ using atomic resolution scanning transmission electron microscopy. Real-space characterization of the local PLD structure across the phase diagram enables harnessing of emergent properties of thin transition metal dichalcogenides. This work was published in Proc. Nat. Acad. Sci. USA (PNAS) [1].

Atomic resolution electron microscopy study of periodic lattice distortions in 2D CDW near the commensuration-incommensuration transition

CDW and their concomitant periodic lattice distortions (PLD) govern the electronic properties in many layered transition-metal dichalcogenides. In particular, 1T-TaS₂ can undergo a PLD phase transition from a conducting to an insulating state as the PLD becomes commensurate with the crystal lattice. Here we directly image PLDs of the nearly-commensurate (NC) and commensurate (C) phases in thin exfoliated 1T-TaS₂ using atomic resolution cryogenic scanning transmission electron microscopy (cryo-STEM). Compared to scanning tunneling microscopy which allows mapping of CDWs at the material's surface, STEM provides direct information about the projected nuclear positions in thin films. We observed PLD superstructures in exfoliated 1T-TaS₂ samples, suggesting ordering of the CDWs in the out-of-plane direction. More importantly, PLDs exist in domains with different atomic lattice stacking order and their boundaries. Stacking faults in the atomic lattice should directly affect the relative alignment of the CDWs of adjacent layers and, thus, the electronic properties of 1T-TaS₂, especially in thin samples. This work was published in PNAS [2].

Nature of the quantum metal in a two-dimensional crystalline superconductor

2D materials are not expected to be metals at low temperature owing to electron localization. Consistent with this, pioneering studies on thin films reported only superconducting and insulating ground states, with a direct transition between the two as a function of disorder or magnetic field. However, more recent works have revealed the presence of an intermediate quantum metallic state occupying a substantial region of the phase diagram, whose nature is intensely debated. In this work, we observed such a state in the disorder-free limit of a crystalline 2D superconductor, produced by mechanical co-lamination of NbSe₂ in an inert atmosphere. Under a small perpendicular magnetic field, we induce a transition from superconductor to the quantum metal. We find a unique power-law scaling with field in this phase, which is consistent with the Bose-metal model where metallic behavior arises from strong phase fluctuations caused by the magnetic field. This work was published in Nature Physics [3].

Observation of the breakdown of the Wiedemann-Franz law in 2D Dirac systems.

Interactions between particles in quantum many-body systems can lead to the collective behavior described by hydrodynamics. One such system is the electron-hole plasma in graphene near the charge neutrality point, which can form a strongly coupled Dirac fluid. This charge-neutral plasma of quasi-relativistic fermions is expected to exhibit a substantial enhancement of the thermal conductivity, thanks to the decoupling of charge and heat currents within hydrodynamics. For this study, we developed Johnson noise thermometry for measuring electronic contribution of thermal conductivity. Employing this novel high

frequency experimental technique, we observed an order of magnitude increase in the thermal conductivity and the breakdown of the Wiedemann-Franz law in the thermally populated charge-neutral plasma in graphene. This result is a signature of the Dirac fluid and constitutes direct evidence of collective motion in a quantum electronic fluid. This work was published in Science [4]

Inducing Superconducting Correlation in Quantum Hall Edge States in 2D Materials

Creating a hybrid system of a superconductor (SC) and quantum Hall (QH) states have been a long-standing experimental and theoretical goal in condensed matter physics. Recently, this idea of hybrid SC/QH systems has been received intense attentions. We have realized hybrid system between chiral edge of quantum Hall state in 2D materials with superconductor to create electronic state with non-trivial topology. We have developed a novel device scheme for a SC/QH platform by employing high-mobility graphene with transparent superconducting contacts with a high critical magnetic field. If the SC electrode is narrower than the superconducting coherence length under a quantizing magnetic field, the incoming electron is correlated to the outgoing hole along the chiral QH edge state by the Andreev process across the SC electrode. In order to realize this crossed Andreev conversion (CAC), it is necessary to fabricate highly transparent and nanometer-scale superconducting junctions to the QH system. In this experiment we reported the observation of CAC in a graphene QH system contacted with a nanostructured NbN superconducting electrode. The chemical potential of the edge states across the SC electrode exhibited a sign reversal, providing direct evidence of CAC. The result was a hallmark of crossed Andreev conversion and constitutes direct evidence of coupling of counter propagating quantum Hall edge states via Cooper pairing. This work was published in Nature Physics [5]

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III. SCIENTIFIC ACCOMPLISHMENTS

Charge Density Waves in Mesoscopic 2-Dimensional Materials for Nanoelectronics

Professor Philip Kim, Harvard University, Single Investigator Award

The objective of this project is to investigate emergent science and novel electronic devices appeared 2-dimensional (2D) correlated electronic systems. In this period, we have realized hybrid system between chiral edge of quantum Hall state in 2D materials with superconductor to create electronic state with non-trivial topology. The quantum Hall (QH) effect supports a set of chiral edge states at the boundary of a 2D electron gas system. A superconductor (SC) contacting these states can induce correlations of the quasiparticles in the dissipationless 1D chiral QH edge states. Creating a hybrid system of a superconductor (SC) and quantum Hall (QH) states has been a long-standing experimental goal in condensed matter physics. Recently, this idea of hybrid SC/QH systems has received high attention. For example, there has been a theoretical proposal that SC/QH system provides a novel route to realizing non-Abelian zero-energy modes including Majorana modes, which allow bona fide universal topological quantum computation. However, the realization of combined SC/QH devices has been a challenging task due to the formation of large contact barriers at the SC/QH interfaces. In this funding period, we have developed a novel device scheme for a SC/QH platform by employing high-mobility graphene with transparent superconducting contacts with a high critical magnetic field. For this experiment, we fabricated highly transparent NbN superconducting electrodes on a hexagonal boron nitride (hBN) encapsulated graphene samples (Fig. 1A). If the SC electrode is narrower than the superconducting coherence length under a quantizing magnetic field, the incoming electron is correlated to the outgoing hole along the chiral QH edge state by the Andreev process across the SC electrode. In order to realize this crossed Andreev conversion (CAC), it is necessary to fabricate highly transparent and nanometer-scale superconducting junctions to the QH system. In this experiment we reported the observation of CAC in a graphene QH system contacted with a nanostructured NbN superconducting electrode. The chemical potential of the edge states across the SC electrode exhibited a sign reversal, providing direct evidence of CAC. The result was a hallmark of crossed Andreev conversion and constitutes direct evidence of coupling of counter propagating quantum Hall edge states via Cooper pairing. This hybrid SC/QH system can also enable a novel route to create isolated non-Abelian anyonic zero modes in resonance with the chiral QH edge, which will be discussed in the later part of this proposal.

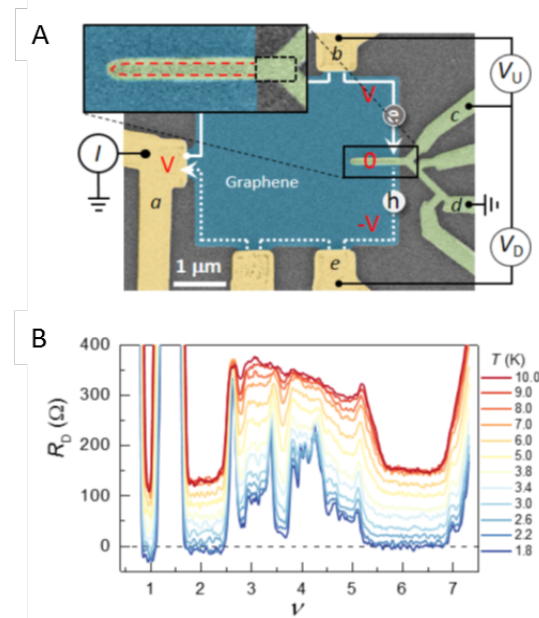


Fig. 1. (A) False color scanning electron microscopy (SEM) image of the device with measurement configurations. Ti/Au normal electrodes (yellow) and a NbN superconducting electrode (green) contacts the graphene Hall bar (blue). Inset, one-dimensional NbN contact to the graphene edge is highlighted with a dotted red line. Note that due to the finite slope of the etching profile of h-BN, the NbN contact is positioned slightly more inwards than the boundary shown in SEM image. A black dotted rectangle guides the NbN segment contributing to the downstream resistance. (B) The filling fraction (ν) dependence of the downstream edge resistance (R_D) at different temperatures with $B = 14$ T.