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The Research Grants Council of Hong Kong
NSFC/RGC Joint Research Scheme
Joint Completion Report

(Please attach a copy of the completion report submitted to the NSFC by the Mainland researcher)

Part A: The Project and Investigator(s)

1. Project Title

Investigations on Two-dimensional Layered Materials with Strong Interlayer Coupling

2. Investigator(s) and Academic Department/Units Involved

	Hong Kong Team	Mainland Team
Name of Principal Investigator *(with title)*	Dr. Yang Chai	Prof. Wei Ji
Post	Associate Professor	Professor
Unit / Department / Institution	Applied Physics / The Hong Kong Polytechnic University	Department of Physics/ Renmin University of China
Contact Information	Email: ychai@polyu.edu.hk	Email: wji@ruc.edu.cn
Co-investigator(s) *(with title and institution)*	Prof. Shu Ping Lau	

3. Project Duration

	Original	Revised	Date of RGC/ Institution Approval *(must be quoted)*
Project Start date	1 Jan 2018	N/A	1 Jan 2018
Project Completion date	31 Dec 2021	N/A	31 Dec 2021
Duration *(in month)*	48	N/A	48
Deadline for Submission of Completion Report	31 Dec 2022	N/A	31 Dec 2022

Part B: The Completion Report

5. Project Objectives

5.1 Objectives as per original application

1. To theoretically predict two-dimensional (2D) layered materials with strong interlayer coupling, tunable direct bandgap, and high carrier mobility.
2. To experimentally prepare 2D layered materials and characterize electrical, photoelectric, vibrational, thermal and thermoelectric properties of those 2D layered materials.

3. To theoretically understand the underlying mechanisms governing these experimental phenomena, and experimentally validate the physical properties predicted by theoretical calculations.

5.2 Revised Objectives

N.A.

6. Research Outcome

Major findings and research outcome

(maximum 1 page; please make reference to Part C where necessary)

It is generally considered that the interlayer coupling of two-dimensional materials is a weak van der Waals effect, and the effect on its properties is insignificant. Recently, some two-dimensional materials (such as black phosphorus, PtS_2, $PtSe_2$, Te and some oxide materials) have new properties of strong interlayer electron coupling. This property is reflected in its various properties with strong layer dependency characteristics. This is a new degree of freedom that can be used to control material properties, design and build new devices. However, at present, the essential factors affecting the coupling between layers, the properties of the single-layer, multi-layer, heterojunction they constitute, and the electrical, optical, thermal and other physical properties of the construction device are still poorly understood. The project predicts several emerging layered materials with strong interlayer and inter-chain coupling. For example, tellurium exhibits the unique characteristics, such as high mobility, adjustable bandgap, approximate direct bandgap, and efficient photoelectric conversion.

Specifically, the project explored in depth the layer thickness-related band gap regulation, high carrier mobility, anisotropic mechanics and transport, optical absorption rate and environmental stability of multilayer α phase tellurium elemental crystals (*Science Bulletin* 2018, 63 (3):159~168; *Nanoscale*, 2018, 10(47), 22263-22269; *Chinese Physics B*, 2020, 29(9), 097103). It was found that the mutual transformation of different phases of Te can be regulated by the method of charge doping, and the energy gap regulation from 1.17 eV to 0.33 eV can be realized at the same time as the phase transition, and even the transition from semiconductors to metals. At the same time, characteristics such as chiral mixed phase, anisotropy and central inversion symmetry can be introduced in the few layers of Te. Two exceptionally stable new structures in the presence of a small layer of Te were predicted, namely the Epsilon phase and the Zeta phase. We also show strong inter-chain interaction in quasi 1D gold selenide (AuSe), which possesses highly anisotropic crystal structure, excellent electrical conductivity, giant magnetoresistance, and unusual reentrant metallic behavior (*Science Bulletin*, 2020, 65, 17, 1451-1459). We also investigate the interaction between single atoms and PtS_2 for their electrochemical activity (*ACS Appl. Mater. Interfaces* 2020, 12, 18, 20448–20455).

It still remains a great challenge to controllably grow 2D tellurene (Te) with good repeatability, uniformity and highly aligned orientation using vapor growth method. We design a Cu foil-assisted alloy-buffer-controlled growth method to epitaxially grow aligned single-crystalline 2D Te on an insulating mica substrate, providing a method to grow aligned high-quality 2D Te in a controllable manner. We also fabricate the field-effect transistors with high carrier mobility. Through the magneto-transport studies, we reveal giant magnetoresistance, and unusual reentrant metallic behavior in AuSe and demonstrate field-effect chirality devices with $PtSe_2$ (*Adv. Func. Mater*., 2021, 31, 2104192). In addition, we also study their interaction for highly efficient electrochemical reactions, including tullurene (*Mater., Today, Ene*., 2021, 21, 100720) and boron doping effect on the selenide (*ACS Nano*, 2019, 13(10), 11469; *ACS Nano*, 2021, 15(5), 8537). We study the lattice oxygen redox chemistry in the strongly correlated oxides and characterize their electrochemical properties (*Energy Environ. Sci.*, 2021, 14, 4647-4671). We assembled layered MnO_2 on the surface of holey rGO. The strong layer interaction between MnO2 and rGO enables it to be an excellent cathode materials of supercapacitor (*Adv. Ene. Mater.*, 2019, 1900037).

Potential for further development of the research and the proposed course of action *(maximum half a page)*

Our findings show that the strong interlayer interaction can give rise to unique bandstructures in these 2D materials. For example, the two spin-polarized bands can cross in the conduction/valence bands, forming Dirac/Weyl points. The transport in Weyl/Dirac materials is theoretically predicted to be nearly non-dissipative over long distances. Thick $PtSe_2$ has been verified as a Dirac semimetal and Te was recently discovered as a Weyl semiconductor. It still lacks experimental ways to efficiently control its transport. In the future, we can investigate field-effect chirality or valley devices with Dirac semimetal $PtSe_2$ or Weyl semiconducting Te.

The chiral anomaly can be further corroborated with planar Hall effect and nonlocal valley transport measurement, which can also be effectively modulated by external fields, showing robust nonlocal valley transport with micrometer diffusion length. Similar to charge-based FETs, the chiral conductivity in $PtSe_2$ devices can be modulated by electrostatic gating with an ON/OFF ratio more than 10^3. We can also deisgn low-loss topological phase change transistors (TPCTs) based on tellurium (Te), a Weyl semiconductor. By modulating the energy separation between the Fermi level and the Weyl point of Te through electrostatic gate modulation, the device exhibits topological phase change between Weyl (Chern number $\neq 0$) and conventional (Chern number = 0) semiconductors. In the Weyl ON-state, the device possesses low-loss transport characteristics due to the global topology of gauge fields against external perturbations; the OFF-state exhibits trivial charge transport in the conventional phase by moving the Fermi level into the band gap.

7. The Layman's Summary

(describe in layman's language the nature, significance and value of the research project, in no more than 200 words)

Two-dimensional (2D) materials possess unique physical properties that are absent in bulk counterpart. The interlayer coupling of 2D materials is generally regarded as weak Van der Waals interaction, and has negligible influence on their physical properties. Recently, it was found that some 2D materials, *e.g.,* black phosphorus, PtS_2 and $PtSe_2$, Te, and some oxides, have exceptional strong interlayer coupling. It has been shown with strong layer-dependent physical properties, such as, bandstructures, lattice constant, and vibration properties. This strong layer-dependence is a new dimension to tune the properties of materials, and can be employed to design new devices structures. However, the key factors for determining the interlayer coupling are so far yet to be clearly revealed. It is still unclear any other 2D materials beyond BP, PtS2, PtSe2, Te belong to this category with strong interlayer coupling. It is a fundamental interest to investigate the physical properties of monolayers, few-layers and heterojunctions and the device performance constructed with these 2D materials. This project aims to carry out experimental and theoretical investigations on these 2D materials with strong interlayer coupling. We start from revealing the key factors governing interlayer couplings of 2D materials, and find parameters to classify them. Based on these understandings, new members of the materials with strong interlayer coupling will be predicted in theory and synthesized by various experimental methods. Our ultimate goals is to identify a few specially advanced novel 2D materials and their device structures.

Part C: Research Output

8. Peer-reviewed journal publication(s) arising directly from this research project

(Please attach a copy of each publication and/or the letter of acceptance if not yet submitted in the previous progress report(s). All listed publications must acknowledge RGC's funding support by quoting the specific grant reference.)

The Latest Status of Publications				Author(s) (**bold** *the authors belonging to the project teams and denote the corresponding author with an asterisk**)	Title and Journal/ Book *(with the volume, pages and other necessary publishing details specified)*	Submitted to RGC *(indicate the year ending of the relevant progress report)*	Attached to this report *(Yes or No)*	Acknowledged the support of this Joint Research Scheme *(Yes or No)*	Accessible from the institutional repository *(Yes or No)*
Year of publication	Year of Acceptance *(For paper accepted but not yet published)*	Under Review	Under Preparation *(optional)*						
2018				**Qiao Jingsi, Pan Yuhao, Yang Feng, Wang Cong, Chai Yang, Ji Wei***	Few-layer Tellurium: one-dimensional-like layered elementary semiconductor with striking physical properties, Science Bulletin, 2018, 63 (3): 159~168		Yes	Yes	2018
2018				**Wang Cong, Zhou Xieyu, Qiao Jingsi, Zhou Linwei, Kong Xianghua, Pan Yuhao,** Cheng Zhihai, **Chai Yang, Ji Wei ***	Charge-governed phase manipulation of few-layer tellurium, Nanoscale, 2018, 10 (47): 22263~22269	2019	No	Yes	2018

2019				**Yunpeng Zuo**, Dewei Rao, Sainan Ma, Tingting Li, Yuen Hong Tsang, Stepan Kment, and **Yang Chai***	Valence Engineering via Dual-Cation and Boron Doping in Pyrite Selenide for Highly Efficient Oxygen Evolution, ACS Nano, 2019, 13(10), 11469-11476	2019	No	Yes	2019
2019				Yang Wang, Songyang Su, **Lejuan Cai**, Bocheng Qiu, Ni Wang, Jie Xiong, Cheng Yang, Xiaoming Tao,* and **Yang Chai***	Monolithic Integration of All-in-One Supercapacitor for 3D Electronics, Advanced Energy Materials, 2019, 1900037	2019	No	Yes	2019
2019				Peng Lang, **Qiao Jingsi**, Xian Jing Jing, **Pan Yuhao**, **Ji Wei***, Zhang Wenhao*, Fu Ying Shuang*	Unusual Electronic States and Superconducting Proximity Effect of Bi Films Modulated by a $NbSe_2$ Substrate, ACS Nano, 2019, 2, 13 (2): 1885~1892		Yes	Yes	2019

2020				**Changlin Yan, Cong Wang**, Linwei Zhou, Pengjie Guo, Kai Liu, Zhong-Yi Lu, Zhihai Cheng, **Yang Chai,** Anlian Pan and **Wei Ji***	Two ultra-stable novel allotropes of tellurium few-layers, ***Chinese Phys. B, 2020,*** 29 **097103**		Yes	Yes	2019
2020				**Jingli Wang, Jingsi Qiao, Kang Xu, Jiewei Chen, Yuda Zhao, Bocheng Qiu, Ziyuan Lin, Wei Ji,** and **Yang Chai***	Quasi one-dimensional van der Waals gold selenide with strong interchain interaction and giant magnetoresistance, Science Bulletin, 2020, 65, 17, 1451-1459.		Yes	Yes	2020

2020				**Lejuan Cai, Ning Zhang, Bocheng Qiu, and Yang Chai***	Computational Design of Transition Metal Single-Atom Electrocatalysts on PtS_2 for Efficient Nitrogen Reduction, *ACS Appl. Mater. Interfaces* 2020, 12, 18, 20448–20455		Yes	Yes	2020
2021				**Ning Zhang** and **Yang Chai***	Lattice oxygen redox chemistry in solid-state electrocatalysts for water oxidation, *Energy Environ. Sci.*, 2021, **14**, 4647-4671		Yes	Yes	2021

2021				**Ning Zhang, Cong Wang, Jiewei Chen,** Canyu Hu, Jun Ma, Xi Deng, **Bocheng Qiu, Lejuan Cai**, Yujie Xiong*, and **Yang Chai***	Metal Substitution Steering Electron Correlations in Pyrochlore Ruthenates for Efficient Acidic Water Oxidation, *ACS Nano* 2021, 15, 5, 8537–8548		Yes	Yes	2021
2021				**Bocheng Qiu, Cong Wang, Jingli Wang, Ziyuan Lin, Ning Zhang, Lejuan Cai**, Xiaoming Tao, **Yang Chai***	Metal-free tellurene cocatalyst with tunable bandgap for enhanced photocatalytic hydrogen production, Materials Today Energy, 2021, 21, 100720		Yes	Yes	2021
2021				**Jiewei Chen**, Ting Zhang, **Jingli Wang, Ning Zhang, Wei Ji**, Shuyun Zhou, and **Yang Chai***	Field-Effect Chiral Anomaly Devices with Dirac Semimetal, Adv. Funct. Mater.2021, 31, 2104192		Yes	Yes	2021

9. Recognized international conference(s) in which paper(s) related to this research project was/were delivered *(Please attach a copy of each delivered paper. All listed papers must acknowledge RGC's funding support by quoting the specific grant reference.)*

Month/Year/ Place	Title	Conference Name	Submitted to RGC *(indicate the year ending of the relevant progress report)*	Attached to this report *(Yes or No)*	Acknowledged the support of this Joint Research Scheme *(Yes or No)*	Accessible from the institutional repository *(Yes or No)*
04/2019 Taiwan	TWO-DIMENSIONAL MATERIALS ELECTRON DEVICES: CONTACT AND DOPING	The 2019 International Symposium on VLSI Technology, Systems and Applications	2019	No	No	04/2019 Taiwan
10/2019 Suzhou/China	Quasi One-dimensional AuSe with Strong Interchain Interaction and Giant Magnetoresistance	The 5th International Conference on 2D Materials and Technology	2019	No	No	10/2019 Suzhou/China

10. Student(s) trained *(Please attach a copy of the title page of the thesis.)*

Name	Degree registered for	Date of registration	Date of thesis submission/ graduation
Cong WANG	PhD	13/Jan/2020	Not yet
Qiao Jingsi	PhD	1 September 2013	12 June 2018
Yang Feng	PhD	1 September 2014	18 December 2019

11. Other impact *(e.g. award of patents or prizes, collaboration with other research institutions, technology transfer, etc.)*

Dr. Yang Chai received PolyU FAST Faculty Award in Research and Scholar Activities in 2018/2019, Young Scientist Award of ICON-2DMAT in 2019, PolyU President's Award in Research and Scholar Activities in 2019/2020, NR45 Young Innovators Award in 2021, and Young Scientist of World Laureate Forum in 2021. He was elected as a member of The Hong Kong Young Academy of Sciences in 2019.

12. Statistics on Research Outputs *(Please ensure the summary statistics below are consistent with the information presented in other parts of this report.)*

	Peer-reviewed journal publications	Conference papers	Scholarly books, monographs and chapters	Patents awarded	Other research outputs (Please specify)
No. of outputs arising directly from this research project [or conference]	12	2	0	0	0