

Quantum Materials Science Unit (Yoshinori Okada)

# FY2022 Annual Report

Quantum Materials Science Unit

Assistant Professor Yoshinori Okada



## Abstract

In FY2022, we investigated the electronic properties of epitaxial thin films grown by multiple deposition systems, such as molecular beam epitaxy and pulsed laser deposition. Furthermore, our strength of combining spectroscopic and macroscopic transport studies enabled us to deepen our understanding of the magnetic semimetal material  $\text{Cr}_{1+\delta}\text{Te}_2$ , which simultaneously hosts a large Berry curvature in momentum space and antiferromagnetic fluctuations in real space. These studies unambiguously demonstrate our unique capability of pursuing condensed matter physics research. Followed by these accomplishments and several experimental milestones, we are currently accelerating our research activity and productivity.

## 1. Member

## **1.1 Staff**

- Dr. Yoshinori Okada, PI
- Dr. Yuita Fujisawa, Postdoctoral scholar
- Dr. Sheng Liu, Postdoctoral scholar
- Dr. Yukiko Obata, Postdoctoral scholar
- Dr. Gopi Nath Daptry, Postdoctoral scholar

## **1.2 Graduate Students**

- Mr. Mohmed Atwa
- Mr. Barnaby Smith
- Mr. Markel Pardo Almanza
- Ms. Anjana Krishnadas
- Mr. Takatsugu Onishi (Narita Unit)
- Mr. Dyon van Dinter
- Mr. Monika Eggenberger
- Ms. Nanami Tomoda (Rotation Student)
- Mr. Thomas Johannes Hasiweder (Rotation student)

## **1.3 Visiting Scientist/student**

- Dr. Tessui Nakagawa (Ryukyu University, Japan)
- Mr. Bruno Saika Kenichi (The University of Tokyo, Japan)

## **1.4 Intern Student**

- Mr. Kavin Ketan Bhatt (Imperial Collage London, UK)
- Ms. Amna Salah Mahmoud Ahmed (Assiut University, Egypt)
- Mr. Leonel Angel Cabrera Loo (Yachay Tech University, Ecuador)
- Mr. Vadim Samardak (Far Eastern Federal University, Russia)

# **2. Collaborations**

## **2.1 Oxide electronics**

- Dr. Takayuki Harada (NIMS, Japan)

## **2.2 Magnetic Imaging**

- Dr. Anjan Soumyanarayanan (A-Star, Singapore)

## **2.3 Device Fabrication**

- Prof. Yasuhiro Niimi (Osaka University, Japan)

## **2.4 Photoemission Experiments**

- Prof. Takeshi Kondo (The University of Tokyo, Japan)
- Prof. Masaki Kobayashi (The University of Tokyo, Japan)

- Prof. Kenta Kuroda (The University of Tokyo, Japan)

## 2.5 First Principle Calculations

- Prof. Feng-Chuan Chuang (National Sun Yet-sen University, Taiwan)
- Dr. Hsin Lin (Academia Sinica, Taiwan)
- Dr. Khoong Hong Khoo (A-Star, Singapore)

## 2.6 Scanning Probe Microscopy Experiments

- Prof. Yasuyuki Yoshida (Kanazawa University, Japan)

## 2.7 Thermal Management Project (JST-CREST)

- Prof. Tsunehiro Takeuchi (Toyota Technological Institute, Japan)
- Prof. Katsumi Tanigaki (Tohoku University, Japan)
- Prof. Fuyuki Shimojo (Kumamoto University, Japan)
- Prof. Kazunori Sato (Osaka University, Japan)

## 2.8 X-ray absorption spectroscopic Experiments

- Prof. Chang-Yang-Kuo (National Chiao Tung University, Taiwan)

## 2.9 Raman spectroscopic Experiments

- Prof. B.J.Kim (Pohang University of Science and Technology, Korea)

# 3. Activities and Findings

## 3.1 Widely tunable magnetic semimetal $\text{Cr}_{1+\delta}\text{Te}_2$

We have previously published a paper reporting on a unique deposition process enabling systematic compositional control in a ferromagnetic  $\text{Cr}_{1+\delta}\text{Te}_2$  in 2020. Using this unique magnetic system, we further investigated the magnetotransport properties, the electronic states using ARPES, and thermodynamical properties. These results are summarized below.

One of the main findings is the discovery of a large Berry curvature in the band structure of this material [48]. While recent studies have explored real-space Berry curvature effects, we shed light on the momentum-space counterpart. The ARPES (angle resolved photoemission spectroscopy) has revealed the presence of a characteristic semi-metallic band region, which shows a rigid-like energy shift with  $\delta$ . The DFT calculation suggests that the semi metallic band region possesses a large Berry curvature which may affect the magneto-transport properties, such as the Hall effect. Indeed, our transport experiments show that the intrinsic component of the anomalous Hall effect (AHE) is sizable and undergoes a sign flip across  $\delta$ . These findings underscore the increasing relevance of momentum-space Berry curvature in magnetic TMDs and provide a unique platform for intertwining topological physics in real and momentum space.

Another finding is the existence of large antiferromagnetic fluctuation which is enhanced at higher  $\delta$ , leading to prominent enhancement of the magneto-Seebeck effect. We showed the temperature and magnetic field dependence of the longitudinal thermopower  $S_{xx}$  for different  $\text{Cr}_{1+\delta}\text{Te}_2$  compositions [45]. As doping increases, the sign of  $S_{xx}$  changes from positive to negative at the critical doping level of  $\delta \sim 0.5$ . This

observed doping-dependent trend in the thermopower is consistent with the evolution of the semi metallic band structure observed in ARPES. Importantly, an anomalous enhancement of the thermoelectric response near TC is also observed around  $\delta \sim 0.5$ . Combining information from magnetometry and ARPES measurements, the existence of the critical nature of the doping level  $\delta_c$  ( $\sim 0.5$ ) is unveiled in magnetic semimetal  $\text{Cr}_{1+\delta}\text{Te}_2$ , where antiferromagnetic fluctuation and near-Fermi-energy pseudo gap formation play a potential vital role in enhancing thermoelectric energy conversion.

These studies involving unique synthesis techniques, electronic states investigation by spectroscopy and transport experiments synergistically point out that  $\text{Cr}_{1+\delta}\text{Te}_2$  is a unique magnetic material hosting a large tunability in magnetism, transport properties associated with the band structure and Berry curvature. We are conducting scanning tunneling spectroscopy study to further reveal the magnetic field effect on the large Berry curvature in momentum space as well as the possible existence of skyrmions, the real space counterpart as a source of Berry curvature.

### 3.2 Observation of robust metallic surface states in a delafossite epitaxial film

Epitaxial thin films of metallic delafossites are a recent topic of intense investigation due to their intriguing electronic states, such as high carrier mobility and conductivity. Using in situ angle-resolved photoemission spectroscopy, we investigated the electronic states of epitaxial  $\text{PdCoO}_2$  thin films with high crystalline quality [43,47]. On this characteristic surface, we observed a surprisingly prominent bulk-like single hexagonal large Fermi surface. The observation of a sharp Fermi surface relies on the minimized atomic scale disorder in our high-quality film surface. Additionally, the predominantly two-dimensional bulk electronic state with the Fermi group velocity parallel to the plane of  $\text{PdCoO}_2$  is expected to make the near-surface bulk-like state less sensitive to the scattering by longer length scale random bipolar surface potentials. These findings are invaluable for accelerating the search for exotic functionalities in epitaxial ultrathin films and heterostructures of metallic delafossites.

## 4. Publications

### 4.1 Journals

1. Y. Fujisawa, M. Pardo-Almanza, C. H. Hsu, A. Mohamed, K. Yamagami, A. Krishnadas, F. C. Chuang, K. H. Khoo, J. Zang, A. Soumyanarayanan, Y. Okada, *Widely tunable Berry curvature in the Magnetic Semimetal  $\text{Cr}_{1+\delta}\text{Te}_2$* , Advanced Materials 2207121(2023)
2. Tomoki Kawamoto, Anjana Krishnadas, Chia-Hsiu Hsu, Markel Pardo-Almanza, Yuita Fujisawa, Guoqing Chang, Takayuki Harada, Y. Okada, *Visualization of robust two-dimensional bulk states with suppressed surface state on epitaxial  $\text{PdCoO}_2$  thin films with bipolar surfaces*, Phys.Rev.Materials 7,024001(2023)
3. Margarita G. Dronova, Feng Ye, Scott E. Cooper, Anjana Krishnadas, Christina M. Hoffmann, Yuita Fujisawa, Y. Okada, D. I. Khomskii, Y. Feng, *Controlling inversion disorder in a stoichiometric spinel magnet*, PNAS 119, e2208748119(2022)
4. M. Atwa, Y. Fujisawa, T. Onishi, M. Pardo-Almanza, M. Couillard, K. Harada, T. Takeuchi, Y. Okada, *Large antiferromagnetic fluctuation enhancement of the thermopower at a critical doping in magnetic semimetal  $\text{Cr}_{1+\delta}\text{Te}_2$* , Phys.Rev.B106, 155112(2022)
5. D. Ueta, R. Kobayashi, H. Sawada, Y. Iwata, S. Yano, S. Kuniyoshi, Y. Fujisawa, T. Masuda, Y. Okada, and S. Itoh, *Anomalous Magnetic Moment Direction under Magnetic Anisotropy Originated from Crystalline Electric Field in van der Waals Compounds  $\text{CeTe}_3$  and  $\text{CeTe}_2\text{Se}$* ,

J.Phys.Soc.Jpn.91,094706(2022)

6. T. Harada and Y. Okada, *Metallic delafossite thin films for unique device applications*, APL Materials 10,070902
7. K. Yamagami, Y. Fujisawa, M. Almanza, B. R. M. Smith, K. Sumida, Y. Takeda, Y. Okada, *Enhanced  $d-p$  hybridization intertwined with anomalous ground state formation in the van der Waals itinerant magnet  $\text{Fe}_5\text{GeTe}_2$* , Phys.Rev.B106,045137(2022)
8. Z. Wang, C. Y. Huang, C. H. Hsu, H. Namiki, T. R. Chang, F. C. Chuang, H. Lin, T. Sasagawa, V. Madhavan, and Y. Okada, *Observation of a van Hove singularity of a surface Fermi arc with prominent coupling to phonons in a Weyl semimetal*, Phys.Rev.B105,075110(2022)
9. Z. Wang, J. Olivares, H. Namiki, T. Sasagawa, V. Madhavan and Y. Okada, *Visualizing superconductivity in a doped Weyl semimetal with broken inversion symmetry*, Phys.Rev.B104,115102(2021)
10. R. Okuma, C. Ritter, G. J. Nilsen, Y. Okada, *Magnetic frustration in a van der Waals metal  $\text{CeSi}$* , Phys.Rev.Materials(L)5,L121401(2021)

## 4.2 Books and other one-time publications

Nothing to report.

## 4.3 Oral and Poster Presentations

1. Barnaby Smith, *Real Space Observation of Structural Phase Segregation in  $\text{Fe}_5\text{GeTe}_2$  using STM/STS*American Physical Society, March Meeting,Las Vegas,Nevada,USA, 5-10 March, 2023 (Poster)
2. Yuita Fujisawa, *Laboratory-based in-situ photoemission spectroscopies of quantum material epitaxial films*, The 27th Hiroshima International Symposium on Synchrotron Radiation, Hiroshima University Faculty Club, Japan, 9-10 March,2023
3. Anjana Krishnadas, *Evolution of Electronic States in Epitaxial YBCO Thin Films with Calcium Doping by Angle-Resolved Photoemission Spectroscopy*, The 27th Hiroshima International Symposium on Synchrotron Radiation, Hiroshima University Faculty Club, Japan 9-10 March,2023 (Poster)
4. Yuita Fujisawa, *スピネル超伝導体 $\text{LiTi}_2\text{O}_4$ の新奇電子状態イメージング非自明な電子状態で発現する超電導現象の新しい潮流*, 京都大学基礎物理研究会, Yukawa Institute for Theoretical Physics, Kyoto University,21-23 December,2022
5. Mohamed Atwa, *Magnetic Fluctuation Enhancement of the Thermoelectric Energy Conversion at a Critical Doping in Magnetic Semimetal  $\text{Cr}_{1+\delta}\text{Te}_2$* , Materials Research Society (MRS) Boston, Massachusetts, USA, 27 November-02 Decmber,2022
6. Yuita Fujisawa, Mohamed Atwa, Markel Pardo-Almanza, Kohei Yamagami, Yoshinori Okada, *Tunable  $\text{Cr}_{1+\delta}\text{Te}_2$  films hosting skyrmion from frustrated ferrimagnetic background  $\text{Cr}_{1+\delta}\text{Te}_2$  薄膜における変調可能なフラストレートフェリ磁性とスカーミオン形成*第83回応用物理学会秋季学術講演会(online) 22a-C202-1,22 September,2022 (講演奨励賞受賞記念講演)
7. Markel Pardo-Almanza, Yuita Fujisawa, Yoshinori Okada, *Vector field STM study of  $\text{Cr}_{1.6}\text{Te}_2$  Epitaxial Films*, JPS Autumn Meeting 13pW631-7, Tokyo Institute of Technology 12-15 September,2022
8. Barnaby R. M. Smith, Yuita Fujisawa, Yukiko Obata, Nanami Tomoda, Daichi Ueta, Yoshinori Okada, *Investigation of Novel Low Temperature Phases in  $\text{CeTe}_3$  by STM/STS*, JPS Autumn Meeting 12pW541-13, Tokyo Institute of Technology, 12-15 September,2022
9. Yukiko Obata, Yuita Fujisawa, Yuto Hasuo, Takahiro Urata, Hiroshi Ikuta, Yoshinori Okada, *2次元トポロジカル絶縁体候補物質 $\text{HfZrTe}_4$ のSTM/STS*, JPS Autumn Meeting 13aW521-8, Tokyo Institute of Technology, 12-15 September,2022
10. Kohei Yamagami, Yoshinori Okada, *遷移金属リントリカルコゲナイド $\text{MPS}_3$  ( $M = \text{Mn}, \text{Fe}, \text{Co}, \text{Ni}$ )のCr K $\alpha$  硬X線光電子分光*, JPS Autumn Meeting 14pW241-6, Tokyo Institute of Technology, 12-15 September,2022

11. Yuita Fujisawa, Anjana Krishnadas, Barnaby R. M. Smith, Markel Pardo-Almanza, Yukiko Obata, Yoshinori Okada, *STM/STSによるスピネル超伝導体LiTi<sub>2</sub>O<sub>4</sub>の渦糸配列の可視化*, JPS Autumn Meeting 15aW541-8, Tokyo Institute of Technology, 12-15 September, 2022
12. M. G. Dronova, Feng Ye, S. E. Cooper, Anjana Krishnadas, C. M. Hoffmann, Yuita Fujisawa, Yoshinori Okada, D. I. Khomskii, Yejun Feng, *Defects control in magnetic spinel oxide ZnFe<sub>2</sub>O<sub>4</sub>*, JPS Autumn Meeting 13pW631-11, Tokyo Institute of Technology, 12-15 September, 2022
13. Anjana Krishnadas, Yuita Fujisawa, T. Takeda, Markel Pardo-Almanza, Kohei Yamagami, M. Kobayashi, Yoshinori Okada, *ARPES study of the band structure in the normal state of a spinel oxide superconductor LiTi<sub>2</sub>O<sub>4</sub>*, JPS Autumn Meeting 15aW541-7, Tokyo Institute of Technology, 12-15 September, 2022

#### 4.4 Invited Talk

1. Title : スピネル超伝導体の電子状態イメージング  
 Invitation : Center for Spintronics Research Network, Osaka University  
 Date : 02 December, 2022  
 Speaker : Yoshinori Okada
2. Title : Imaging Quantum Materials and heterostructures  
 Invitation : National Yang Ming Chiao Tung University, Taiwan  
 Date : 06 December, 2022  
 Speaker : Yoshinori Okada

### 5. Intellectual Property Rights and Other Specific Achievements

Nothing to report.

### 6. Meetings and Events

1. 2nd IUVESTA workshop on Advanced Spectroscopy and Transport for 2D Materials at Surfaces and The 4th Asia-Pacific Symposium on Solid Surfaces (APSSS-4)  
 Date : 18-21 September, 2022  
 Venue : OIST Auditorium  
 Organizer : The Japan Society of Vacuum and Surface Science  
 Sponsored by The International Union for Vacuum Science, Technique and Applications (for IUVESTA Workshop), The Japan Society for the Promotion of Science (for APSSS-4) and OIST
2. Lecture  
 Dr. Tadashi Machida, RIKEN  
 Title : Detecting Majorana QPs topological Superconductors by STM, Yu-Shiba-Rusinov (YSR) STM tip for high spin resolution STM  
 Date : 1-2 November, 2022
3. Lecture  
 Prof. Satoru Tanaka, Kyushu University  
 Title : graphene on SiC  
 Date : 12 December, 2022

## 7. Other

Nothing to report.