

一般セッション(口頭講演) | 4 JSAP-Optica Joint Symposia 2024 : 4.5 Nanocarbon and 2D Materials

📅 2024年9月17日(火) 9:00 ~ 12:00 🏢 A35 (朱鷺メッセ3F)

**[17a-A35-1~9] 4.5 Nanocarbon and 2D Materials**

北浦 良(物材機構)、松田 一成(京大)、宮内 雄平(京大)

## ◆ 英語発表

9:00 ~ 9:30

[17a-A35-1]

[JSAP-Optica Joint Symposia Invited Talk] Shift current photovoltaics in single domain ferroelectric SnS

○Kosuke Nagashio<sup>1</sup> (1.UTokyo)

## ◆ 英語発表

9:30 ~ 9:45

[17a-A35-2]

Absorption Enhancement of Excitons in WS<sub>2</sub> by Silicon Huygens' Metasurface○(D)Dingwei Chen<sup>1</sup>, Junichi Takahara<sup>1,2</sup> (1.GSE. Osaka Univ., 2.PC. Osaka Univ.)

## ◆ 英語発表

9:45 ~ 10:00

[17a-A35-3]

Magnetic brightening and its dynamics of defect-localized excitons in monolayer WSe<sub>2</sub>○(DC)Yubei Xiang<sup>1</sup>, Keisuke Shinokita<sup>1</sup>, Kenji Watanabe<sup>2</sup>, Takashi Taniguchi<sup>3</sup>, Kazunari Matsuda<sup>1</sup> (1.Institute of Advanced Energy, Kyoto Univ., 2.Research Center for Electronic and Optical Materials, NIMS, 3.Research Center for Materials Nanoarchitectonics, NIMS)

## ◆ 英語発表 ◆ 注目講演

10:00 ~ 10:30

[17a-A35-4]

[JSAP-Optica Joint Symposia Invited Talk] Electronic and excitonic properties of semiconductor bilayer moiré system revealed by optical spectroscopy

○Yuya Shimazaki<sup>1,2</sup> (1.RIKEN, CEMS, 2.Univ. of Tokyo, Eng.)

## ◆ 英語発表

10:30 ~ 10:45

[17a-A35-5]

Exciton-driven Floquet-Bloch States in 2D Semiconductors

Vivek Pareek<sup>1</sup>, David Bacon<sup>1</sup>, ○(DC)XING ZHU<sup>1</sup>, Yang-Hao Chan<sup>2</sup>, Fabio Bussolotti<sup>3</sup>, Nicholas S Chan<sup>1</sup>, Joel Perez Urquiza<sup>1</sup>, Kenji Watanabe<sup>4</sup>, Takashi Taniguchi<sup>4</sup>, Michael K. L. Man<sup>1</sup>, Julien Madeo<sup>1</sup>, Diana Qiu<sup>5</sup>, Kuan Eng Johnson Goh<sup>3,6,7</sup>, Felipe H. da Jornada<sup>8,9</sup>, Keshav M. Dani<sup>1</sup> (1.FSU, OIST, 2.IAMS, Academia Sinica, 3.IMRE, A\*STAR, 4.NIMS, 5.Yale Univ., 6.NUS, 7.NTU, 8.Stanford Univ., 9.SLAC)

## ◆ 英語発表

10:45 ~ 11:15

[17a-A35-6]

[JSAP-Optica Joint Symposia Invited Talk] Exciton transfer and interface excitons in mixed-dimensional heterostructures

○Nan Fang<sup>1</sup>, Yih-Ren Chang<sup>1</sup>, Shun Fujii<sup>1,2</sup>, Daiki Yamashita<sup>1,3</sup>, Mina Maruyama<sup>4</sup>, Yanlin Gao<sup>4</sup>,  
Chee Fai Fong<sup>1</sup>, Daichi Kozawa<sup>1,5</sup>, Keigo Otsuka<sup>1,6</sup>, Kosuke Nagashio<sup>6</sup>, Susumu Okada<sup>4</sup>, Yuichiro  
Kato<sup>1</sup> (1.RIKEN, 2.Keio Univ., 3.AIST, 4.Univ. of Tsukuba, 5.NIMS, 6.UTokyo)

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## ◆ 英語発表

11:15 ~ 11:30

[17a-A35-7]

Identification and manipulation of valley coherence in monolayer WSe<sub>2</sub>

○(D)Wang Haonan<sup>1</sup>, Kenji Watanabe<sup>2</sup>, Takashi Taniguchi<sup>2</sup>, Kazunari Matsuda<sup>1</sup> (1.IAE, Kyoto  
Univ., 2.NIMS)

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## ◆ 英語発表

11:30 ~ 11:45

[17a-A35-8]

Dry transfer and optical properties of CVD-grown transition metal dichalcogenides

○Wenjin Zhang<sup>1</sup>, Tomoya Ogawa<sup>1</sup>, Takahiko Endo<sup>1</sup>, Kenji Watanabe<sup>2</sup>, Takashi Taniguchi<sup>2</sup>,  
Takumi Sannomiya<sup>3</sup>, Kazunari Matsuda<sup>4</sup>, Yasumitsu Miyata<sup>1</sup> (1.Tokyo Metropolitan Univ.,  
2.NIMS, 3.Tokyo Tech., 4.Kyoto Univ.)

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## ◆ 英語発表

11:45 ~ 12:00

[17a-A35-9]

Structural and Electrical Properties of Millimeter Scale CVD Graphene

○(P)Sengottaiyan Chinnasamy<sup>1</sup>, Kazunori Hirose<sup>1</sup>, Yuta Kurachi<sup>1</sup>, Masanori Hara<sup>1</sup>,  
Masamichi Yoshimura<sup>1</sup> (1.Toyota Tech. Inst.)

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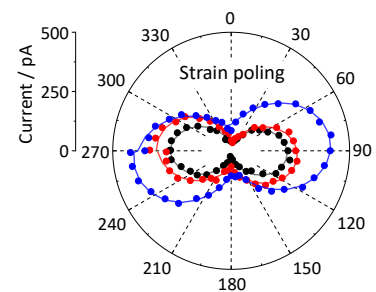
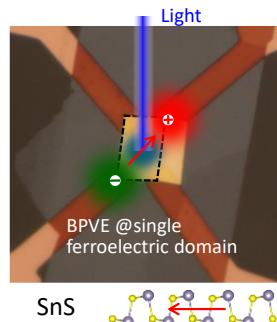
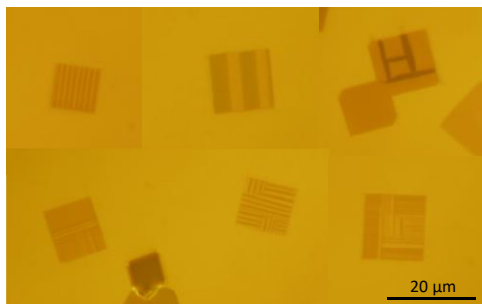
## Shift current photovoltaics in single domain ferroelectric SnS

Kosuke Nagashio

Department of Materials Engineering, The University of Tokyo, Japan

nagashio@material.t.u-tokyo.ac.jp

The bulk photovoltaic effect (BPVE) in ferroelectrics, wherein spontaneous polarization can be reversed within crystals lacking centrosymmetry, encompasses the significant contribution of ferroelectric domain walls (DWs), known as DW-PVE. Nevertheless, the separation between intrinsic BPVE within the domain and DW-PVE remains unexplored in two-dimensional (2D) ferroelectrics, notwithstanding its significant importance. In this study, we have successfully grown sizable crystals of 2D ferroelectric SnS, facilitating comprehensive yet intricate examination of domain configurations utilizing polarized optical microscopy and piezoresponse force microscopy. By properly selecting the large ferroelectric single domain within SnS crystals, uniform intrinsic BPVE across the domain was unequivocally demonstrated. Furthermore, to further enhance intrinsic BPVE, manipulation of strain poling increased photocurrent, suggesting that locally distributed polarizations due to imperfection introduced in SnS crystals are aligned by strain. These results will offer a new avenue for rigorous comprehension of DW-PVE in 2D ferroelectrics.



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- [6] Ferroelectricity: Nature commun. 2020, 11, 2428.
- [7] Electrical transport: Nanoscale 2018, 10, 22474.
- [8] Mechanical exfoliation: MRS Advances 2018, 3, 2809.

# Absorption Enhancement of Excitons in WS<sub>2</sub> by Silicon Huygens' Metasurface

Dingwei Chen<sup>1</sup> and Junichi Takahara<sup>1,2</sup>

<sup>1</sup> Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

<sup>2</sup> Photonics Center, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

E-mail: takahara@ap.eng.osaka-u.ac.jp

## 1. Introduction

Strong visible absorption of light in semiconducting two-dimensional transition-metal dichalcogenides (TMDCs) is essential for improving the photocarrier extraction efficiency in optoelectronic devices [1,2]. However, their ultrathin thickness leads to limited optical absorption [3]. To accomplish adequate light absorption, a dielectric-based Silicon (Si) Huygens' metasurface (HMS) based on degenerate critical coupling (DCC) can realize superimposed modes enhancement effects on WS<sub>2</sub> [4, 5].

In this study, we design and fabricate Si HMS to enhance the absorption of WS<sub>2</sub>. The enhancement is as high as 0.56 in simulation. Meanwhile, we demonstrate that small monolayer WS<sub>2</sub> fragment is successfully transferred to the metasurface utilizing poly (propylene) carbonate (PPC) films.

## 2. Results and Discussion

First, we designed and fabricated Si metasurface. In our design, the degenerate of Electric Dipole (ED) and Magnetic Dipole (MD) modes was realized (namely, the so-called HMS). Figure 1a shows the designed structure and Scanning Ion Microscope (SIM) image of the sample. The Si metasurface was comprised by cylindrical resonator arrays with period  $d = 334$  nm, radius  $r = 112$  nm, and thickness  $h = 89$  nm.

As shown in Fig. 1b, we simulated and measured the absorption spectra of the metasurface. Because the height and radius of prepared sample was different from designed, the degenerate was not completely achieved. However, we partially realized DCC with ED/MD modes shown as the fitting curve 1 and 2.

Without background, the simulated absorption of WS<sub>2</sub> on metasurface is increased by about 0.52 (15.5 times) under DCC condition compared with WS<sub>2</sub> on Si substrate as shown in Fig. 1c. The inset shows the raw absorption spectrum of WS<sub>2</sub> on metasurface and the corresponding fitting curve.

Furthermore, we tried to transfer monolayer WS<sub>2</sub> on the metasurface by dry release transfer method with PPC films. The optical microscope photo is shown in Fig. 1d. Due to the thermoplastic properties of PPC

film, the adhesion between WS<sub>2</sub> and PPC decreases at about 70 °C. With temperature control, WS<sub>2</sub> flakes on PPC can be easily dry-transferred.

Our observations undeniably establish the groundwork for a material platform enabling high-efficiency photodetection, energy harvesting, and thermal emission based on TMDCs.

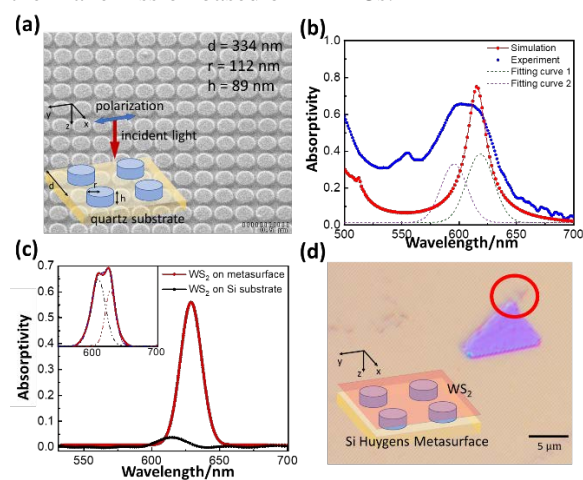


Figure 1. (a) The SIM image of the sample and the schematic illustration of Si HMS, which was constituted by Mie resonator arrays with  $h = 89$  nm,  $d = 334$  nm and  $r = 112$  nm, and (b) experimental and simulated [6] absorptivity for the metasurface, and MD /ED mode fitting curves with the experimental spectrum. (c) Simulated absorptivity subtracting background for WS<sub>2</sub> on Si substrate and HMS. (d) Optical microscope image of monolayer WS<sub>2</sub> transferred on the metasurface. The scale bar is 5  $\mu$ m.

## Acknowledgements

This work was supported by JSPS KAKENHI 23H00274. DW. C. was supported by CSC (No. 202204910108).

## References

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# Magnetic brightening and its dynamics of defect-localized excitons in monolayer WSe<sub>2</sub>

Yubei Xiang<sup>1</sup>, Keisuke Shinokita<sup>1</sup>,  
Kenji Watanabe<sup>2</sup>, Takashi Taniguchi<sup>3</sup> and Kazunari Matsuda<sup>1</sup>

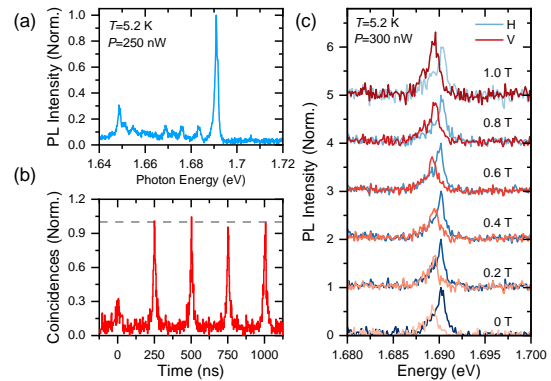
<sup>1</sup> Institute of Advanced Energy, Kyoto University, <sup>2</sup> Research Center for Electronic and Optical Materials, NIMS, <sup>3</sup> Research Center for Materials Nanoarchitectonics, NIMS  
E-mail: xiang.yubei.86h@st.kyoto-u.ac.jp

## 1. Introduction

Quantum light sources, in particular solid-states single-photon emitters have attracted considerable attention due to their crucial roles in the field of quantum information technologies. Recently, defects in two-dimensional monolayer transition metal dichalcogenides, such as tungsten diselenide (WSe<sub>2</sub>), have been demonstrated to be promising candidates for stable and bright quantum light sources [1,2]. However, the external controllability of single-photon emission has not been fully understood.

## 2. Results

In this study, we have investigated the nature and dynamics of defect-localized exciton emissions in monolayer WSe<sub>2</sub> under magnetic fields for novel single-photon emitters with external tunability. Figure 1(a) depicts the typical photoluminescence (PL) spectra of monolayer WSe<sub>2</sub> at low temperature. The strong and sharp PL peaks are clearly observed and can be attributed to defect-localized exciton emissions. Figure 1(b) shows the photon statistics obtained from defect-localized exciton emissions using the Hanbury-Brown and Twiss experimental setup, demonstrating photon antibunching behavior. This indicates that the observed defect-localized exciton emission functions as a single-photon emitter. Moreover, the polarization-resolved PL spectra with an in-plane magnetic field are shown in Figure 1(c), where each PL spectrum is normalized by the peak intensity of the higher energy states for each magnetic field. The PL intensity of the lower energy states significantly



**Figure 1** (a) Typical low temperature PL spectra of monolayer WSe<sub>2</sub>. (b) Second-order correlation measurement of the localized exciton emission. The cross-talk signals in the histogram are removed. (c) Polarization-resolve PL spectra with the increasing in-plane magnetic fields.

increases with increasing magnetic field within a small range below 1 T. We discuss the dynamics of the magnetic brightening from the defect-localized exciton emission in monolayer WSe<sub>2</sub>.

## 3. Conclusions

Our findings on the physical nature of defect-localized exciton states in monolayer WSe<sub>2</sub> and the dynamics of the magnetic brightening offer a novel approach to manipulating single-photon emitters using an external magnetic field in the field of quantum optics application.

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## Electronic and excitonic properties of semiconductor bilayer moiré system revealed by optical spectroscopy

RIKEN CEMS<sup>1</sup>, Univ. of Tokyo, Eng.<sup>2</sup>, °Yuya Shimazaki<sup>1, 2</sup>

E-mail: yuya.shimazaki@riken.jp

The recent discovery of many-body physics such as strongly correlated electrons, superconductivity and magnetism in precisely twist angle-controlled bilayer graphene at a magic angle revived enormous interest on moiré lattice system. Many-body physics in bilayer moiré system is not limited to graphene, but rather robustly appears in 2D semiconductor materials such as transition metal dichalcogenides (TMDs). Monolayer semiconductor TMDs have conduction and valence bands with relatively large effective mass which enhances the influence of Coulomb interactions, also resulting in formation of strongly bound excitons with optical excitations. In twisted bilayer semiconductor TMDs, the superlattice effect due to the formation of moiré lattice further enhances the influence of Coulomb interaction for electrons and expected to show many-body electronic phases. On the other hand, it has been a long-standing issue that semiconductor TMDs have poor electrical contact properties due to the formation of Schottky barriers, which has been hindering the observation of those many-body electronic phases in moiré system via transport measurement. Instead, we performed optical microscopic spectroscopy experiments of a bilayer TMD moiré system which revealed many-body electronic phase, quantum coupled excitonic states, and novel quantum mixture of exciton – hole states.

Here we studied a twisted bilayer MoSe<sub>2</sub> system with monolayer hBN tunnel barrier (Figure 1). The combination of the top and bottom gates allows to control the chemical potential and the perpendicular electric field independently. The energy shifts of excitonic resonances (exciton polarons) probe the carrier densities of the top and bottom layers independently, which revealed the formation of moiré sub bands in the system. By changing the energy detuning between layers via control of the perpendicular electric field, we observed an abrupt charge transfer at  $\nu = 1$  (1 electron per moiré lattice) and a stabilized charge transfer plateau at  $\nu = 2$  around zero detuning point, which evidences the existence of strongly correlated electrons [1]. We further observed the existence of charge order at these fillings from the Umklapp scattering of excitons [2]. Last but not least, we figured out that the tunnel coupling of holes through the monolayer hBN barrier results in formation of hybrid exciton states [1] and electric field-controlled exciton – hole Feshbach resonances [3].

### References

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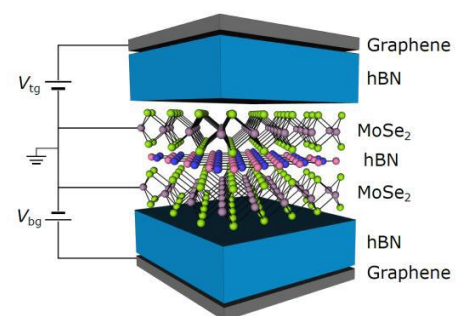


Figure 1: Schematic sketch of MoSe<sub>2</sub> / hBN / MoSe<sub>2</sub> heterostructure



## Exciton-driven Floquet-Bloch States in 2D Semiconductors

Vivek Pareek<sup>1†</sup>, David Bacon<sup>1†</sup>, Xing Zhu<sup>1†</sup>, Yang-Hao Chan<sup>2</sup>, Fabio Bussolotti<sup>3</sup>, Nicholas S Chan<sup>1</sup>, Joel Pérez Urquiza<sup>1</sup>, Kenji Watanabe<sup>4</sup>, Takashi Taniguchi<sup>5</sup>, Michael K. L. Man<sup>1</sup>, Julien Madéo<sup>1</sup>, Diana Qiu<sup>6</sup>, Kuan Eng Johnson Goh<sup>3,7,8</sup>, Felipe H. da Jornada<sup>9,10\*</sup>, Keshav M. Dani<sup>1\*</sup>

<sup>1</sup> Femtosecond Spectroscopy Unit, Okinawa Institute of Science and Technology Graduate University; Onna, Okinawa, Japan 904-0495, <sup>2</sup> Institute of Atomic and Molecular Sciences, Academia Sinica, and Physics Division, National Center of Theoretical Sciences; Taipei, Taiwan, <sup>3</sup> Institute of Materials Research and Engineering (IMRE), Agency for Science, Technology and Research (A\*STAR); 2 Fusionopolis Way, Singapore, 138634 Singapore, <sup>4</sup> Research Center for Functional Materials, National Institute for Materials Science; 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan, <sup>5</sup> International Center for Materials Nanoarchitectonics, National Institute for Materials Science; 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan, <sup>6</sup> Department of Mechanical Engineering and Materials Science, Yale University; New Haven, CT, USA, <sup>7</sup> Department of Physics, National University of Singapore; 2 Science Drive 3, Singapore, 117551 Singapore, <sup>8</sup> Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University; 50 Nanyang Avenue, Singapore 639798, Singapore, <sup>9</sup> Department of Materials Science and Engineering, Stanford University; Stanford, CA, USA <sup>10</sup> Stanford PULSE Institute, SLAC National Accelerator Laboratory; Menlo Park, CA, USA († equal contribution)

\*Email address: jornada@stanford.edu, kmdani@oist.jp

### 1. Introduction

Floquet engineering, in which a temporal periodic drive breaks the continuous temporal symmetry and dynamically engineers the electronic structure, has attracted enormous attention in condensed matter physics. However, only a handful of studies have experimentally demonstrated Floquet effects driven by optical fields [1-4], which are limited by weak light-matter interactions. Meanwhile, theoretically it is predicted that an internal oscillating field, such as phonons and excitons, could also drive giant Floquet effects without the undesirable aspects of the optical case [5,6]. In this talk, we will discuss the experimental observation of the Floquet-Bloch states induced by the excitons in 2D semiconductors.

and angle-resolved photoemission spectroscopy (Tr-ARPES). The experimental setup, as employed in our previous studies [7,8], consists of a home-built table-top beamline generating 21.7-eV XUV probe to photoemit the electrons from the sample. The electrons are further analyzed by a momentum microscope to map the 3D band-structure of the material ( $E$ ,  $k_x$ ,  $k_y$ ).

To study Floquet effects driven by excitons, a pump pulse resonant with the  $\text{WS}_2$  A exciton (2.1 eV) is used to excite the sample. As shown in figure 1(b), we observe the striking change of dispersion around the center of valence band top, transforming from ground-state parabolic dispersion to Mexican-hat dispersion. This change results from the hybridization of the valence band and the conduction band replica dressed by excitons. Similar change is also observed in the replica of valence band at 2.1 eV.

### 3. Conclusions

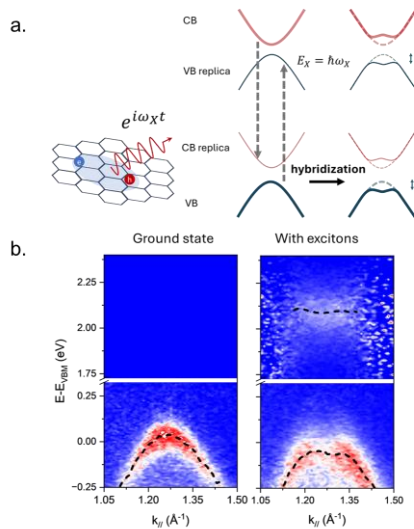
Using Tr-ARPES, we studied the Floquet-Bloch states in 2D semiconductors, driven by the excitonic field internally.

### Acknowledgements

This work was supported in part by JSPS Kakenhi grant number 21H01020. Funding was also provided in part by the Femtosecond Spectroscopy Unit of the Okinawa Institute of Science and Technology Graduate University. We thank the OIST engineering support section for their support.

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**Figure 1.** a. Schematics of Floquet effects driven by excitons. The bands are replicated by energy of exciton. The original valence band hybridize with the conduction band replica, transforming to Mexican-hat dispersion. b. ARPES spectrum of (left) ground state of valence band, (right) the valence band and its replica with excitons ( $N_X = 3 \times 10^{12} \text{ cm}^{-2}$ ).

### 2. Experiment

We perform experiments on monolayer  $\text{WS}_2$  using time-

## Exciton transfer and interface excitons in mixed-dimensional heterostructures

○N. Fang<sup>1,2</sup>, Y. R. Chang<sup>1</sup>, S. Fujii<sup>2,3</sup>, D. Yamashita<sup>2,4</sup>, M. Maruyama<sup>5</sup>, Y. Gao<sup>5</sup>, C. F. Fong<sup>1</sup>,  
D. Kozawa<sup>1,2,6</sup>, K. Otsuka<sup>1,7</sup>, K. Nagashio<sup>8</sup>, S. Okada<sup>5</sup>, Y. K. Kato<sup>1,2</sup>

<sup>1</sup> *Nanoscale Quantum Photonics Laboratory, RIKEN Cluster for Pioneering Research, Saitama, Japan*

<sup>2</sup> *Quantum Optoelectronics Research Team, RIKEN Center for Advanced Photonics, Saitama, Japan*

<sup>3</sup> *Department of Physics, Keio University, Kanagawa, Japan*

<sup>4</sup> *Platform Photonics Research Center, National Institute of Advanced Industrial Science and Technology (AIST), Ibaraki, Japan*

<sup>5</sup> *Department of Physics, University of Tsukuba, Ibaraki, Japan*

<sup>6</sup> *Research Center for Materials Nanoarchitectonics, National Institute for Materials Science, Ibaraki, Japan*

<sup>7</sup> *Department of Mechanical Engineering, The University of Tokyo, Tokyo, Japan*

<sup>8</sup> *Department of Materials Engineering, The University of Tokyo, Tokyo, Japan*

E-mail: nan.fang@riken.jp

Two-dimensional van der Waals heterostructures have introduced unconventional phenomena that emerge at atomically precise interfaces, and further development is expected in mixed-dimensional heterostructures. Here we discuss exciton physics in 1D-2D heterostructures consisting of one-dimensional carbon nanotubes and two-dimensional tungsten diselenide. Both the chirality and the layer number are identified before assembling the clean and free-standing heterostructures, allowing for investigation of the band alignment effects. For small band gap nanotubes corresponding to type I band alignment, exciton transfer is observed [1]. The mixed-dimensional heterostructures display a remarkable exciton reservoir effect where the longer-lifetime excitons within the two-dimensional semiconductor are funneled into carbon nanotubes through diffusion. With increasing the nanotube band gap, the transfer efficiency shows a pronounced enhancement indicating a resonance in the band alignment. For large band gap nanotubes corresponding to type II band alignment, exciton transfer diminishes whereas bright emission peaks originating from the interface are identified [2]. We assign the peaks to interface excitons as they only appear in type-II heterostructures. Localization of low-energy interface excitons is indicated by extended lifetimes as well as small excitation saturation powers, and photon correlation measurements confirm room-temperature quantum emission. With mixed-dimensional van der Waals heterostructures where band alignment can be engineered, new opportunities for quantum photonics are envisioned.

This work is supported by JSPS (KAKENHI JP22K14624, JP22K14625, JP21K14484, JP22K14623, JP22H01893, JP21H05233, JP22F22350, JP23H00262, JP20H02558, JP24K08296) and MEXT (ARIM JPMXP1222UT1135). Y.R.C. is supported by JSPS (International Research Fellow). N.F. and C.F.F. are supported by the RIKEN Special Postdoctoral Researcher Program. We thank the Advanced Manufacturing Support Team at RIKEN for technical assistance.

### References

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- [2] N. Fang, Y. R. Chang, S. Fujii, D. Yamashita, M. Maruyama, Y. Gao, C. F. Fong, D. Kozawa, K. Otsuka, K. Nagashio, S. Okada, and Y. K. Kato, [Nat. Commun. 15, 2871 \(2024\)](#).



# Identification and manipulation of valley coherence in monolayer WSe<sub>2</sub>

Haonan Wang<sup>1</sup>, Kenji Watanabe<sup>2</sup>, Takashi Taniguchi<sup>3</sup>, and Kazunari Matsuda<sup>1</sup>

<sup>1</sup>Institute of Advanced Energy, Kyoto University, <sup>2</sup>Research Center for Electronic and Optical Materials, NIMS, <sup>3</sup>Research Center for Materials Nanoarchitectonics, NIMS

E-mail: wang.haonan.36t@st.kyoto-u.ac.jp

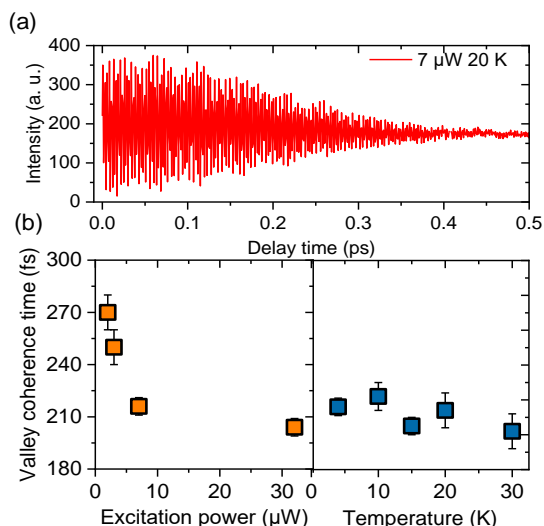
## 1. Introduction

The monolayer transition metal dichalcogenide (TMDs) are granted with valley degree of freedom due to broken inversion symmetry, and strong spin-orbit coupling. The degenerated states at band-edges of K(K') valley possess information of valley pseudospin, which experiences intervalley decoherence process during emission [1]. With valley decoherence not coupling to any radiative dipole, direct probing or manipulation of valley coherence in the time domain has remain a challenge. Here we propose a method of optically exploring the valley coherence time in the time-domain measurement.

## 2. Results

We have developed a method of direct measuring valley coherence time of the free exciton in monolayer WSe<sub>2</sub>. By employing polarized interferometer, the decoherence process between K and K' valley excitons under various temperature and excitation power are directly measured. **Figure 1(a)** shows the typical interferogram arising from valley coherence. The exacted valley coherence times from the interferogram are plotted in **Figure 1(b)**. It can be seen that the values of valley coherence time remain stable under temperature from 4 to 30 K, which is consistent with the previous result [2]. Moreover, the valley coherence time gradually decreases due to increased exciton-exciton collision with increasing excitation power condition. We also explored the valley coherence of monolayer (1L) WSe<sub>2</sub> device with changing carrier density in the spectral and time domain. A wide tuning range of degree of linear polarization

(DOLP) is observed, with the values ranging from nearly 0% under heavily positive doping, to 50% under heavily negative doping conditions. The measured valley coherence time shows the similar trend as the DOLP, which will be further discussed in detail.



**Figure 1** (a) Interferogram of intervalley decoherence process of free exciton in 1L-WSe<sub>2</sub> under 7  $\mu$ W and 20 K. (b) Extracted valley coherence time of free exciton in 1L-WSe<sub>2</sub> under various temperature and excitation power

## 3. Conclusions

A new direct method of exciton valley coherence has been applied to 1L-WSe<sub>2</sub> in the time-domain. The detail of intervalley decoherence process is explored under various temperature, excitation power and carrier doping condition, which will facilitate further understanding of valleytronics in 1L-TMDs.

## References

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## Dry transfer and optical properties of CVD-grown transition metal dichalcogenides

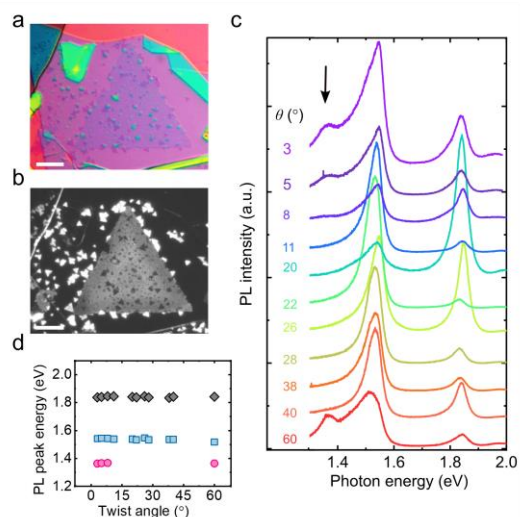
Wenjin Zhang<sup>1</sup>, Tomoya Ogawa<sup>1</sup>, Takahiko Endo<sup>1</sup>, Kenji Watanabe<sup>2</sup>, Takashi Taniguchi<sup>2</sup>, Takumi Sannomiya<sup>3</sup>, Kazunari Matsuda<sup>4</sup>, Yasumitsu Miyata<sup>1</sup>

<sup>1</sup> Tokyo Metropolitan University, <sup>2</sup> National Institute for Materials Science, <sup>3</sup> Tokyo Institute of Technology, <sup>4</sup> Institute of Advanced Energy, Kyoto University

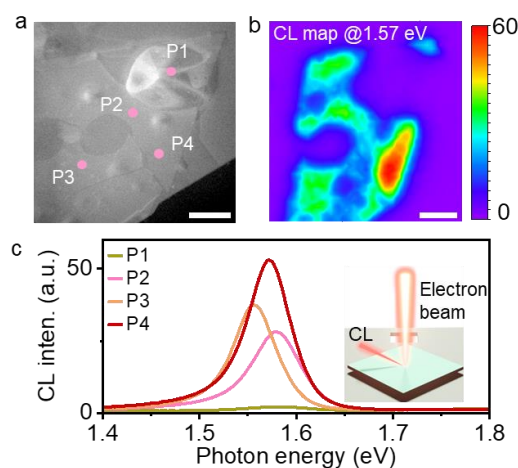
E-mail: ymiyata@tmu.ac.jp; wjzhang@tmu.ac.jp

Improving the transfer technique of 2D materials is an important issue for their fundamental study and future applications. Even though much progress has been made, the search for exfoliated thin and sufficiently large flakes of 2D materials is still very time-consuming. In contrast, the use of triangular single crystals of transition metal dichalcogenides (TMDCs) allows for easy determination of crystal orientation and thus easy control of twist angles. However, there have been only a few reports on dry transfer and its effect on the optical properties of CVD-grown TMDCs. One of the major technical difficulties is the efficient peeling of CVD-grown monolayers from growth substrates. In general, CVD-grown monolayers are known to adhere strongly to growth substrates compared to exfoliated flakes. Compared with the solution or chemical assisted transfer of CVD grown TMDCs, the dry transfer should keep the intrinsic properties of as grown TMDCs because the solution processes induce physical or chemical property modulations of TMDCs. To expand the versatility of this growth-assisted approach, it is highly desirable to demonstrate the dry transfer and investigate its effects on physical properties.

Here, we report the dry transfer and optical properties of CVD-grown TMDCs using a simple acrylic resin stamp [1]. We introduced the melting and solidification process of an acrylic resin stamp in contact with the sample to improve the adhesion of the stamp. This improvement allows us to efficiently pick up the single crystals of various TMDC monolayers with desired grain size and density from the SiO<sub>2</sub> surface, and to perform high-throughput and continuous dry transfer. Furthermore, we also fabricated the hBN-encapsulated TMDC monolayers and various twisted bilayers including MoSe<sub>2</sub>/MoS<sub>2</sub>, MoSe<sub>2</sub>/WSe<sub>2</sub>, and MoSe<sub>2</sub>/WS<sub>2</sub>. The interlayer interaction and quality of dry-transferred, CVD-grown TMDCs were characterized by using photoluminescence (PL), cathodoluminescence (CL) spectroscopy, and cross-sectional electron microscopy. The prominent PL peaks of interlayer excitons can be observed for the MoSe<sub>2</sub>/MoS<sub>2</sub> with small twist angles at room temperature (Fig.1). We also found that the optical spectra were locally modulated due to the nanosized bubbles, which are formed by the presence of interface carbon impurities (Fig.2). The present findings provide a widely applicable potential of the present method and enable an efficient search of the emergent optical and electrical properties of TMDC-based vdW heterostructures.



**Figure 1.** (a) Optical and (b) PL images of hBN encapsulated MoS<sub>2</sub>/MoSe<sub>2</sub> heterobilayers. Scale bars are 10  $\mu$ m. (c) Room-temperature PL spectra of twisted area with various twist angle. (d) PL peak positions of intralayer exciton from MoS<sub>2</sub> and MoSe<sub>2</sub>, and interlayer exciton from MoSe<sub>2</sub>/MoS<sub>2</sub> heterobilayer with different twist angles.



**Figure 2.** (a) CL analysis of hBN-encapsulated monolayer MoSe<sub>2</sub>. (b) STEM image of hBN-encapsulated monolayer MoSe<sub>2</sub>. (c) CL map at 1.57 eV of the same area as (a). Scale bars in (a) and (b) are 1  $\mu$ m. (d) CL spectra recorded at the positions in (a).

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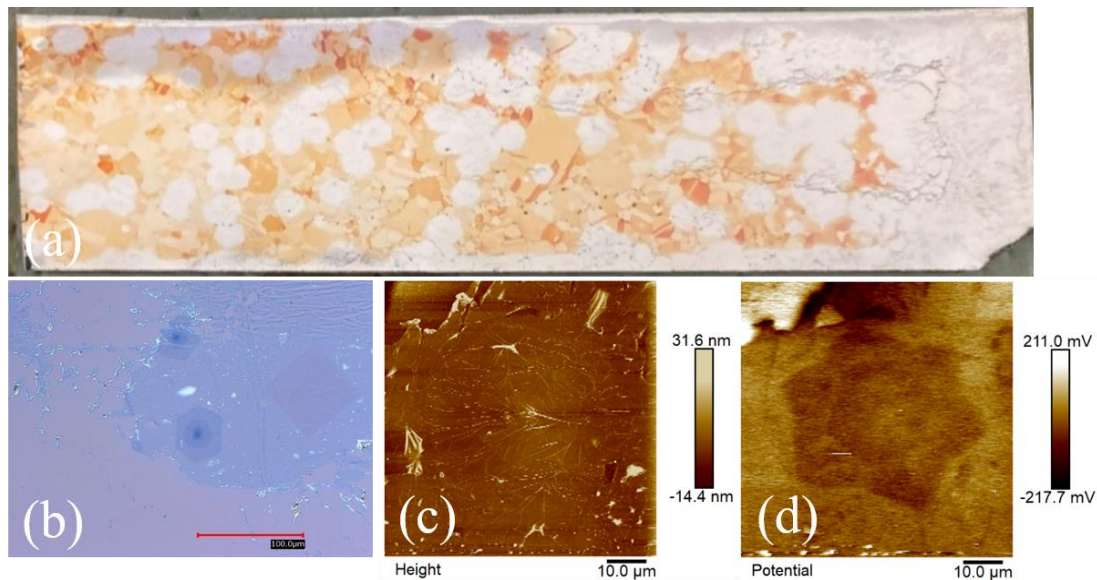
## Structural and Electrical Properties of Millimeter Scale CVD Graphene

Toyota Technological Institute, Surface Science Lab., Chinnasamy Sengottaiyan, Kazunori Hirose,

Yuta Kurachi, Masanori Hara, Masamichi Yoshimura\*

\*Email: yoshi@toyota-ti.ac.jp

Despite the large single-crystal graphene is highly desired and important for the applications of graphene in electronics, it is still a challenge to precisely control the nucleation site of graphene to develop wafer-scale high-quality graphene. Herein, we synthesized millimeter-scale high-quality graphene on Cu-foil by chemical vapor deposition (CVD). To synthesize graphene, a copper foil was annealed in Ar and H<sub>2</sub> atmosphere to remove impurities and increase grain size. Then, methane (CH<sub>4</sub>) was introduced at 1035 °C for 200 min to grow high-quality graphenes on the copper surface (**Figure a**). Graphene was transferred from Cu-foil to SiO<sub>2</sub>/Si substrate by the bubble transfer method (**Figure b**). The synthesized graphenes before and after transfer are studied for structural and electrical properties by AFM (**Figure c**) and Kelvin probe force microscope (**Figure d**). The Raman spectra specify the quality and defects structure in the graphene lattice, while energy dispersive spectroscopy shows oxidation on the graphene/Cu surface. Investigate a deeper understanding of the structural and electrical properties of hexagon graphene structure to meet future developments in the research relevant to the scalable growth of high-quality graphene.



**Figure:** (a) Photograph of a synthesized hexagonal graphene/Cu-foil. The transferred graphene on SiO<sub>2</sub>/Si substrate for (b) Optical, (c) AFM, and (d) Surface potential images.

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