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Exploiting Ultrafast Carrier Transfer in Van der Waals Heterostructures for THz Optoelectronics

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14. ABSTRACT Here we summarize the milestones achieved for the three years project (AFOSR FA9550) in the chronicle order, with the total publication listed at the end of the report. We have published a total of 29 journal publications, which include high-impact journals such as Nature Physics, PRX, 5 Nature Communications, ACS Nano, and 6 Nano Lett. The notable highlights achieved include: • Correlated insulating states in WSe₂/WS₂ moiré superlattices. (Nature Physics 2020) • Revealing Rydberg exciton up to 11s in monolayer WSe₂ (Nano Lett. 2020) • Landau quantization of exciton-phonon interaction in monolayer WSe₂ (Nature Comm. 2020) • Electrically tunable exciton annihilation to exciton funneling transition in vdW heterostructure (Nature Comm 2020) • Landau quantization of exciton resonance in monolayer WSe₂ (PRX 2020) • Giant Zeeman splitting of interlayer exciton singlet and triplet in TMD heterobilayer (Nano Lett. 2019) • Chiral phonon replica of the dark exciton in monolayer WSe₂ (Nature Comm. 2019) • Direct imaging of dark exciton and dark trion in monolayer WSe₂ (Nano Lett. 2019) • Chiral phonon replica of the intervalley exciton in monolayer WSe₂ (ACS Nano 2019) • Emerging exciton phonon interlayer in monolayer TMD alloy (Nano Lett. 2018) • Revealing intrinsic biexciton and charged biexciton in monolayer WSe₂ (					
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Final Report

Here we summarize the milestones achieved for the three years project (AFOSR FA9550) in the chronicle order, with the total publication listed at the end of the report. We have published a total of **29** journal publications, which include high-impact journals such as **Nature Physics**, **PRX**, **5 Nature Communications**, **ACS Nano**, and **6 Nano Lett.**

Notable highlights achieved in this project

- 1) Correlated insulating states in WSe₂/WS₂ moiré superlattices. (Nature Physics 2020)
- 2) Revealing Rydberg exciton up to 11s in monolayer WSe₂ (Nano Lett. 2020)
- 3) Landau quantization of exciton-phonon interaction in monolayer WSe₂ (Nature Comm. 2020)
- 4) Electrically tunable exciton annihilation to exciton funneling transition in vdW heterostructure (Nature Comm 2020)
- 5) Landau quantization of exciton resonance in monolayer WSe₂ (PRX 2020)
- 6) Giant Zeeman splitting of interlayer exciton singlet and triplet in TMD heterobilayer (Nano Lett. 2019)
- 7) Chiral phonon replica of the dark exciton in monolayer WSe₂ (Nature Comm. 2019)
- 8) Direct imaging of dark exciton and dark trion in monolayer WSe₂ (Nano Lett. 2019)
- 9) Chiral phonon replica of the intervalley exciton in monolayer WSe₂ (ACS Nano 2019)
- 10) Emerging exciton phonon interlayer in monolayer TMD alloy (Nano Lett. 2018)
- 11) Revealing intrinsic biexciton and charged biexciton in monolayer WSe₂ (Nature Comm. 2018)
- 12) Unique light emission from 2D InSe flake (Nano Lett. 2018)

Invited talks

- 1) Quantum Nanoscience/Spin Canada – I of Quantum Day in Canada, 2021 (correlated states in WS₂/WSe₂ moiré heterojunction);
- 2) 4th International Workshop on Rydberg Excitons in Semiconductors (Rydex2020) (Rydberg excitons in WSe₂);
- 3) Physics Department seminar at Iowa State University (Excitonics in 2D, 2021);
- 4) Physics Department seminar at Austin Univ. (Excitonics in 2D, 2020).
- 5) Physics Department Seminar, RPI, “Light-matter interactions in Flatland: Excitonic Physics in 2D”. Sep 2020. Virtual seminar.
- 6) Lawrence Symposium on Epitaxy, Phoenix, AZ, “2D Excitons: Current Status and Feature”, Feb 2020.
- 7) Ultrafast Bandgap Semiconductor workshop, DC Washington, “Excitonic complexes and dark exciton in WSe₂”, April 2019.

Awards/honors

- 1) NSF Career Award, 2020
- 2) RPI Engineering School Research Excellence Award, 2019
- 3) ACS PRF New Investigator Award, 2018

Significant collaborations

- 1) Prof. Chuanwei Zhang, Univ of Texas at Dallas, supported by ARO, AFOSR. Collaborated on Rydberg exciton states and many-body interactions in WSe₂ (Nano Lett 2020, PRX 2020).
- 2) Prof. Arka Majumdar, the University of Washington at Seattle, supported by AFOSR, ONR, and DARPA. Collaborated on strong coupling between WSe₂ and photonic crystal cavity (Nano Lett. 2020).
- 3) Prof. Ali Adibi, Georgia Institute of Technology, supported by AFOSR. Collaborated on monolayer TMD alloy and lateral heterostructure (ACS Nano. 2020, ACS Nano 2018)
- 4) Prof. Li Yang, Washington University at St Louis, supported by AFOSR. Collaborated on phonon replica of excitons in WSe₂ (Nature Comm. 2019).
- 5) Prof. Di Xiao, the University of Washington at Seattle, supported by AFOSR and DOE. Collaborated on correlated states in WSe₂/WS₂ moiré superlattice (Nature Physics 2021, Nature Comm 2021).

Student(s)/postdoc(s) supported and their current status(es).

- 1) Postdoc: Zhipeng Li. Current position: Associate Professor at Shanghai Jiaotong University.
- 2) Graduate student: Zhen Lian (one semester). Current position: Graduate student at RPI. Expected to graduate with Ph.D. in 2022.
- 3) Graduate student: Shengnan Miao (one semester and one summer). Current position: Graduate student at RPI. Expected to graduate with M.E. in 2021.

I also included the milestone details in the following.

(1) Year 3 (June 15-May 14, 2021)

Despite the challenges presented by the pandemic, we managed to achieve a few milestones and finish the final project. We have managed to publish 6 journal publications.

We have been able to construct a moiré superlattice using the angle-aligned WS₂/WSe₂ heterobilayer. In this unique moiré system, the electron correlation is particularly strong, and we have identified additional correlated states corresponding to a fractional charge per moiré unit cell (fractional fillings), in addition to the Mott insulating states and Wigner crystal states (Nature Physics 2021). Further, we have found that the interlayer exciton PL is strongly modulated by the presence of the correlated states (Nature Comm. 2021). These exciting findings inspire us to utilize the electron correlation to engineering the quantum emitters in the WS₂/WSe₂ moiré superlattice.

We have also been able to extensively study the photocurrent response from the topological insulator (Nanotechnology, 2021). Our gate dependence and magnetic field dependence study suggested a unique photodoping effect from bulk. Interestingly, this effect does not compromise the quantum Hall effect (QHE) but only shifts the QHE onset in a nonvolatile fashion, enabling a new type of quantum memory device.

We have also developed a photocurrent spectroscopy method and reveal the highest Rydberg exciton states in 2D, up to 11s, in the monolayer WSe₂ (Nano Lett. 2020). This is an unprecedented technology improvement that enables the Rydberg exciton study in 2D, which is critical for quantum computation and simulation.

In collaboration with others, we have also characterized and developed new quantum dots in the infrared regime (Nanoscale 2020). We have also utilized Raman spectroscopy to characterize Burn severity (Burn 2020).

We have also probed the ultrafast photocurrent response from the WS₂/WSe₂ moiré heterojunction and found that the photocurrent dynamics are sensitively dependent on the dissociation of the interlayer exciton. We also developed a new schematic of controlling correlated states in the WS₂/WSe₂ moiré heterojunction. Due to the covid, these works are still under review/preparation.

I have also given a few invited talks based on these results, including Quantum Nanoscience/Spin Canada – I of Quantum Day in Canada, 2021 (correlated states in WS₂/WSe₂ moiré heterojunction); 4th International Workshop on Rydberg Excitons in Semiconductors (Rydex2020) (Rydberg excitons in WSe₂); Physics Department seminar at Iowa State University (Excitonics in 2D, 2021), Physics Department seminar at Austin Univ. (Excitonics in 2D, 2020).

(2) Year 2 (June 15, 2019-May 14, 2020)

In the span June 15, 2019-May 14, 2020, we continued our success of high-quality vdW heterostructure devices and further developed precise rotation angle control to fabricate moiré superlattice based on twisted WSe₂/WS₂ hetero-bilayer. We have conducted optical spectroscopy and scanning photocurrent spectroscopy of the vdW heterostructure. We have also developed photocurrent spectroscopy techniques. We have also built the time-resolved photocurrent setup with time-resolved information.

We have published **15** journal publications from June 1, 2019. There are a couple of papers currently under review, and I did not include them in the publication list but will mention them in the work progress description. Most of the papers are published in journals with high-impact factors.

Among the **15** journal publications, there are a few worth particular attention.

We have, for the first time, identified the intrinsic biexciton in WSe₂ (Nature Comm. 2018), the dark exciton in WSe₂, and chiral phonons interacting with dark exciton (Nature Comm. 2019). We have shown emerging electron-phonon coupling in BN encapsulated monolayer alloy: WSSe (Nano Lett. 2018). We have also explored the optical properties of InSe, a new 2D semiconductor with superior electrical and optical properties (Nano Lett. 2018). We have employed time-resolved PL spectroscopy to investigate the dynamics in the hybrid halide perovskite (Scientific Reports, 2018). We have fabricated InSe and perovskite devices for photocurrent measurement and photodetector applications (Advanced Optical Materials, 2019). Based on these results, we have published eight high-impact journal papers, with six from our group and two from collaboration.

These publications included three Nature Communications, two Nano Letters, and one ACS Nano. One of the Nature Communications papers (2018), reporting the important dark exciton effect on the formation of exotic biexciton state in WSe₂, has drawn worldwide attention is highlighted in a news article by Nature Nanotechnology and other sources.

We have given two conference talks with the results from the AFSOR sponsored research (one APS and one MRS) and one invited talk at the Ultrafast Widebandgap Semiconductor workshop at DC.

(3) Year 1 (June 15, 2018-May 14, 2019)

In the span of one year, we have developed the fabrication recipe for extremely high-quality vdW heterostructure devices. We have conducted optical spectroscopy and scanning photocurrent spectroscopy of the vdW heterostructure. We have also built the time-resolved photocurrent setup with time-resolved information.

We have, for the first time, identified the intrinsic biexciton in WSe₂ (Nature Comm. 2018), the dark exciton in WSe₂, and chiral phonons interacting with dark exciton (Nature Comm. 2019). We have shown emerging electron-phonon coupling in BN encapsulated monolayer alloy: WS₂ (Nano Lett. 2018). We have also explored the optical properties of InSe, a new 2D semiconductor with superior electrical and optical properties (Nano Lett. 2018). We have employed time-resolved PL spectroscopy to investigate the dynamics in the hybrid halide perovskite (Scientific Reports, 2018). We have fabricated InSe and perovskite devices for photocurrent measurement and photodetector applications (Advanced Optical Materials, 2019). Based on these results, we have published eight high-impact journal papers, with six from our group and two from collaboration. These publications included three Nature Communications, two Nano Letters, and one ACS Nano. One of the Nature Communications papers (2018), reporting the important dark exciton effect on the formation of exotic biexciton state in WSe₂, has drawn worldwide attention is highlighted in a news article by Nature Nanotechnology and other sources.

We have given two conference talks with the results from the AFSOR sponsored research (one APS and one MRS) and one invited talk at the Ultrafast Widebandgap Semiconductor workshop at DC.

Publications:

#=equal contributions, *= corresponding authors.

The student and postdoc I supervised are underscored.

Year 3 (June 15, 2020-May 14, 2021)

- 1) Shengnan Miao, Tianmeng Wang, Xiong Huang, Dongxue Chen, Zhen Lian, Chong Wang, Mark Blei, Takashi Taniguchi, Kenji Watanabe, Sefaattin Tongay, Zenghui Wang, Di Xiao*, Yong-Tao Cui*, Su-Fei Shi*. “Strong interaction between interlayer excitons and correlated

electrons in WSe₂/WS₂ moiré superlattice". **Nature Communications** 12, Article number: 3608 (2021).

- 2) Xiong Huang[#], Tianmeng Wang[#], Shengnan Miao[#], Chong Wang[#], Zhipeng Li, Zhen Lian, Takashi Taniguchi, Kenji Watanabe, Satoshi Okamoto, Di Xiao^{*}, **Su-Fei Shi^{*}**, Yong-Tao Cui^{*}. "Correlated Insulating States at Fractional Fillings of the WS₂/WSe₂ Moiré Lattice." **Nature Physics** 17, 715 (2021).
- 3) Faji Xie, Zhen Lian[^], Shuai Zhang, Tianmeng Wang[^], Shengnan Miao[^], Zhiyong Song, Zhe Ying, Xing-Chen Pan, Mingsheng Long, Minghao Zhang, Fucong Fei, Wei-Da Hu, Geliang Yu, Fengqi Song, Tingting Kang and **Sufei Shi^{*}**. "Reversible Engineering of Topological Insulator Surface State Conductivity through Optical Excitation." **Nanotechnology** 32 17LT01, (2021).
- 4) Ingrid J. Paredes, Clara Beck, Scott Lee Shuzhen Chen, Mersal Khwaja, Michael R. Scimeca, Shuang L, Sooyeon Hwang, Zhen Lian[^], Kevin McPeak, **Sufei Shi**, Ayaskanta Sahu. "Photoluminescent core/shell α -Zn₃P₂/ZnS quantum dots from benign zinc carboxylates". **Nanoscale**, 12 20952 (2020).
- 5) Tianmeng Wang[#], Zhipeng Li[#], Yunmei Li[#], Zhengguang Lu[#], Shengnan Miao, Zhen Lian, Yuze Meng, Mark Blei, Takashi Taniguchi, Kenji Watanabe, Sefaattin Tongay, Dmitry Smirnov, Chuanwei Zhang, **Su-Fei Shi^{*}** "Giant Valley-polarized Rydberg Excitons in Monolayer WSe₂ Revealed by Magneto-photocurrent Spectroscopy." **Nano Letters**, 10 7635 (2020).
- 6) Hanglin Ye, Rahula, Uwe Krugera, Tianmeng Wang, **Sufei Shi**, Jack Norfleetc and Suvrana De. "Raman Spectroscopy Classifies Burn Severity with High Accuracy." **Burn**, 2020 (accepted).
<https://doi.org/10.1016/j.burns.2020.08.006>

Year 2 (June 15, 2019-May 14, 2020)

- 7) Zhen Lian, Zeyu Jiang, Tianmeng Wang, Mark Blei, Ying Qin, Morris Washington, Toh-Ming Lu, Sefaattin Tongay, Shengbai Zhang, **Su-Fei Shi^{*}**. "Anisotropic Band Structure of TiS₃ Nanoribbon Revealed by Polarized Photocurrent Spectroscopy." **Applied Physics Letters**, 117, 073101 (2020).
- 8) Kyle Vogt, **Su-Fei Shi**, Feng Wang, Matt Graham. "Ultrafast Photocurrent and Absorption Microscopy of Few-Layer TMD Devices Isolate Rate-Limiting Dynamics Driving Fast and Efficient Photoresponse." **The Journal of Physical Chemistry C** 124,15195 (2020).
- 9) Yueyang Chen[#], Shengnan Miao[#], Tianmeng Wang[#], Ding Zhong, Abhi Saxena, colin chow, James Whitehead, Dario Gerace, Xiaodong Xu, **Su-Fei Shi**, and Arka Majumdar. "Metasurface Integrated Monolayer Exciton Polariton." **Nano Letters** 7, 5292 (2020).
- 10) Zhipeng Li[#], Tianmeng Wang[#], Shengnan Miao[#], Yunmei Li[#], Zhenguang Lu, Chenhao Jin, Zhen Lian, Yuze Meng, Mark Blei, Takashi Taniguchi, Kenji Watanabe, Sefaattin Tongay, Wang Yao, Dmitry Smirnov, Chuanwei Zhang, **Su-Fei Shi^{*}**. "Phonon-exciton Interactions in

WSe₂ under a Quantizing Magnetic Field." **Nature Communications**, 11, Article number: 3104 (2020).

- 11) Yuze Meng[#], Tianmeng Wang[#], Chenhao Jin[#], Zhipeng Li, Shengnan Miao, Zhen Lian, Takashi Taniguchi, Kenji Watanabe, Fengqi Song^{2*}, **Su-Fei Shi**^{*}. "Electrical Switching between Exciton Dissociation to Exciton Funneling in MoSe₂/WS₂ Heterostructure." **Nature Communications** 11, Article number: 2640 (2020).
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- 13) Tianmeng Wang[#], Zhipeng Li[#], Zhengguang Lu[#], Yunmei Li[#], Shengnan Miao, Zhen Lian, Yuze Meng, Mark Blei, Takashi Taniguchi, Kenji Watanabe, Sefaattin Tongay, Wang Yao, Dmitry Smirnov, Chuanwei Zhang^{*}, and **Su-Fei Shi**^{*}. "Observation of Quantized Exciton Energies in Monolayer WSe₂ under a Strong Magnetic Field." **Phys. Rev. X** 10, 021024 (2020).
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- 15) Zhipeng Li, Tianmeng Wang, Shennan Miao, Zhen Lian, **Su-Fei Shi**^{*}. "Fine structures of valley-polarized excitonic states in monolayer transitional metal dichalcogenides." **Nanophotonics** 9, 1811 (2020).
- 16) Dali Shao, Weiguang Zhu, Guoqing Xin, Xueqing Liu, Tianmeng Wang, **Sufei Shi**, Jie Lian, Shayla Sawyer. "High performance UV-visible dual-band photodetector based on inorganic Cs₂SnI₆ perovskite/ZnO heterojunction structure." **Journal of Materials Chemistry C** 8 1819 (2020).
- 17) Tianmeng Wang[#], Shengnan Miao[#], Zhipeng Li[#], Yuze Meng[#], Zhengguang Lu, Zhen Lian, Mark Blei, Takashi Taniguchi, Kenji Watanabe, Sefaattin Tongay^{*}, Dmitry Smirnov, **Su-Fei Shi**^{*}. "Giant Valley-Zeeman Splitting from Spin-Singlet and Spin-Triplet Interlayer Excitons in WSe₂/MoSe₂ Heterostructure." **Nano Letters** 20, 694 (2019).
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- 19) Zhipeng Li[#], Tianmeng Wang[#], Chenhao Jin[#], Zhengguang Lu, Zhen Lian, Yuze Meng, Mark Blei, Mengnan Gao, Takashi Taniguchi, Kenji Watanabe, Tianhui Ren, Ting Cao, Sefaattin Tongay, Dmitry Smirnov, Lifa Zhang, **Su-Fei Shi**^{*}. "Momentum-Dark Intervalley Exciton in Monolayer Tungsten Diselenide Brightened via Chiral Phonon." **ACS Nano** 13, 14107-14113 (2019).

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Year 1 (June 15, 2018-May 14, 2019)

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- 27) Yanwen Chen[#], Tianmeng Wang[#], Zhipeng Li, Huanbin Li, Tao Ye, Christian Wetzel, Hanying Li, **Su-Fei Shi***. "Communicating Two States in Perovskite Revealed by Time-Resolved Photoluminescence Spectroscopy." **Scientific Report** 8, 16482 (2018).
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