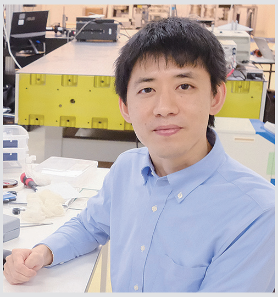
Nanostructure Fabrication, Optical Property Control, and Photonic Functionalization of Atomic-Layer Materials

Department of Materials Molecular Science
Division of Electronic Properties



SHINOKITA, Keisuke
Associate Professor
[shinokita@ims.ac.jp]

Education
2008 B.S. Kyoto University
2010 M.S. Kyoto University
2013 Ph.D. Kyoto University

Professional Employment
2010 JSPS Research Fellow (DC1)
2013 Postdoctoral Researcher, University of Groningen
2015 Visiting Researcher, Max Born Institute
2017 Specially Appointed Assistant Professor, Kyoto University
2020 Assistant Professor, Kyoto University
2025 Associate Professor, Institute for Molecular Science
Associate Professor, The Graduate University for Advanced Studies

Awards
2011 Young Researcher Award, International Symposium on Terahertz Nanoscience (TeraNano 2011), Workshop of International Terahertz Research Network (GDR-I)
2013 Poster Award, International Workshop on Optical Terahertz Science and Technology 2013 (OTST2013)
2020 FY2019 Research Encouragement Award, Institute of Advanced Energy, Kyoto University
2022 18th Osawa Encouragement Award, The Fullerenes, Nanotubes and Graphene Research Society

Member
Graduate Student
OH, Shuto
Secretary
NODA, Ayaka

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Modern electronic devices are approaching their miniaturization limits, necessitating innovative solutions through the integration of optical and quantum effects. Our research group focuses on atomic layer materials, particularly transition metal dichalcogenides (TMDs), which exhibit remarkable optical properties due to their single-atom thickness.

These materials possess direct bandgaps with strong light–matter interactions and unique electronic properties stemming from their extreme two-dimensionality. By stacking different atomic layers through van der Waals forces, we can create artificial heterostructures. In these structures, excitons—bound pairs of electrons and holes generated by photoexcitation—become spatially confined within periodic moiré potentials.

Our team has successfully observed exciton localization phenomena in $WSe_2/MoSe_2$ heterostructures induced by moiré potentials. Through detailed investigations of interlayer exciton formation and their optical responses, including circular polarization characteristics and quantum coherence measurements, we continue to elucidate the quantum states of moiré excitons. Currently, we are advancing this research by developing nanofabrication techniques for atomic layer materials and precisely controlling light–matter interactions to explore novel optical phenomena and functionalities.

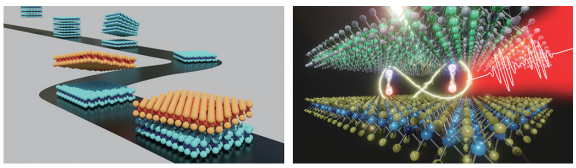

Figure 1. Left: Schematics of nanofabrication process for atomic-layer materials. Right: Visualization of quantum optical phenomena emerging from the engineered atomic-layer nanostructures.

Selected Publications

- H. Wang, H. Kim, D. Dong, K. Shinokita, K. Watanabe, T. Taniguchi and K. Matsuda, “Quantum Coherence and Interference of a Single Moiré Exciton in Nano-Fabricated Twisted Monolayer Semiconductor Heterobilayers,” *Nat. Commun.* **15**, 4905 (2024).
- K. Shinokita, K. Watanabe, T. Taniguchi and K. Matsuda, “Valley Relaxation of the Moiré Excitons in a $WSe_2/MoSe_2$ Heterobilayer,” *ACS Nano* **16**, 16862 (2022).
- K. Shinokita, Y. Miyauchi, K. Watanabe, T. Taniguchi and K. Matsuda, “Resonant Coupling of a Moiré Exciton to a Phonon in a $WSe_2/MoSe_2$ Heterobilayer,” *Nano Lett.* **21**, 5938 (2021).
- K. Shinokita, X. Wang, Y. Miyauchi, K. Watanabe, T. Taniguchi and K. Matsuda, “Continuous Control and Enhancement of Excitonic Valley Polarization in Monolayer WSe_2 by Electrostatic Doping,” *Adv. Funct. Mater.* **29**, 1900260 (2019).

1. Tailoring Exciton Dimensionality and Unveiling Prolonged Valley Polarization

Interlayer excitons (IXs) in twisted TMD heterostructures are a promising platform for novel optoelectronic devices. However, the moiré potential arising from the lattice mismatch often traps these IXs, forming zero-dimensional (0D) quantum emitters known as moiré excitons. While interesting for quantum optics, these localized states are less suitable for applications like photodiodes or solar cells, which benefit from mobile two-dimensional (2D) excitons.

To address this, we demonstrated a strategy to tune the dimensionality of IXs in a $MoSe_2/WSe_2$ heterostructure. By inserting an atomically thin hexagonal boron nitride (h-BN) layer as a spacer, we effectively modulated the moiré potential landscape, thereby transforming the trapped 0D moiré excitons into 2D IXs. The transition was unambiguously confirmed through systematic optical spectroscopy, which revealed a significant blue-shift in photoluminescence (PL) energy and a change from nonlinear saturation to linear power dependence of the PL intensity, a hallmark of the transition from a localized to a delocalized system. A remarkable feature of these engineered 2D IXs is their prolonged valley relaxation lifetime, which reaches up to 100 nanoseconds at low temperatures—orders of magnitude longer than the picosecond lifetimes typically observed in monolayer TMDs. This longevity is attributed to the suppression of the electron–hole exchange interaction, a dominant valley depolarization mechanism, which is weakened by the spatial separation of the electron and hole across the h-BN spacer. Our findings provide an effective strategy to tailor exciton dimensionality and harness the long valley lifetime of 2D IXs for future opto-valleytronic applications.

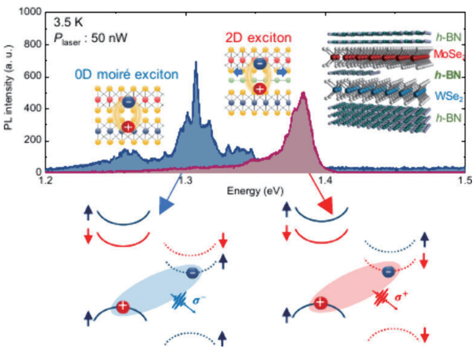

Figure 2. Dimensionality control and valley polarization of interlayer excitons in a $MoSe_2/WSe_2$ heterostructure. The insertion of an h-BN spacer layer (center schematic) transforms 0D moiré excitons with sharp emission peaks (blue spectrum) into 2D excitons with broad emission (red spectrum). These excitons exhibit a unique optical response to circularly polarized light (σ^+/σ^-, bottom schematic), enabling long-lived valley polarization.

2. Observation of Magnetically Switchable Nonlinear Photocurrents

The bulk photovoltaic effect (BPVE), which generates spontaneous photocurrents in non-centrosymmetric materials, offers a pathway to overcome the Shockley-Queisser efficiency limit of conventional solar cells. We investigated these nonlinear photoresponses in a vdW heterostructure that breaks both spatial inversion (P) and time-reversal (T) symmetry, composed of monolayer MoS_2 and the layered antiferromagnet $CrPS_4$. At the hetero-interface, the broken P-symmetry gives rise to a spontaneous photocurrent under illumination, which we identified as a shift current.

More strikingly, we discovered that this photocurrent is highly sensitive to the magnetic state of the $CrPS_4$ layer. The spontaneous photocurrent changed drastically below the Néel temperature of $CrPS_4$ (~40 K), where it transitions into an antiferromagnetic (AFM) phase. This phenomenon is attributed to the emergence of a "magnetic injection current," a distinct nonlinear photocurrent that arises in systems with broken T-symmetry and is superimposed on the existing shift current. We further demonstrated that the net photocurrent can be actively switched with an external magnetic field, which triggers distinct magnetic phase transitions in $CrPS_4$ (*e.g.*, from canted-AFM to ferromagnetic) and alters the contribution of the magnetic injection current. This work demonstrates a magnetic-field-controllable photovoltaic effect and opens a new avenue for "magneto-photovoltaics," a new class of devices that merge magnetism with solar energy conversion.

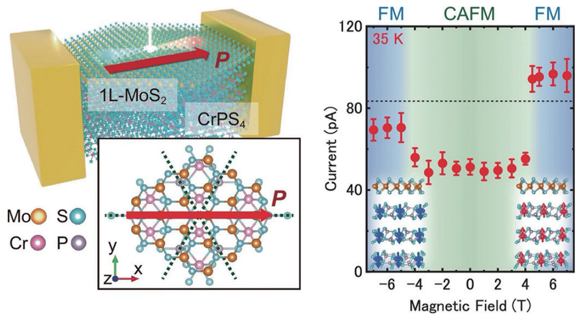

Figure 3. Magnetic field-switching of nonlinear photocurrent at a $MoS_2/CrPS_4$ hetero-interface with broken P- and T-symmetry. In the heterostructure (left schematic), a spontaneous photocurrent is observed. As the magnetic phase of the $CrPS_4$ layer is changed by an external magnetic field (right plot, from canted-AFM to ferromagnetic phase at 35 K), the photocurrent increases in a step-like manner, demonstrating active control of the optical response.

References

1) H. Kim, H. Wang, Y. Wang, K. Shinokita, K. Watanabe, T. Taniguchi, S. Konabe and K. Matsuda, *ACS Nano* **19**, 322 (2025).

2) S. Asada, K. Shinokita, K. Watanabe, T. Taniguchi and K. Matsuda, *Nat. Commun.* **16**, 4827 (2025).