Optical Nano-science Research Section

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1. Introduction

We are engaged in fundamental and applied research of nano-materials from a viewpoint of optics and material science. Our research aims to explore new physical and chemical phenomena leading to the applications of novel nano-materials including carbon nanotubes, layered transition metal dichalcogenides, perovskites for the efficient utilization of light energy and the development of future optoelectronic devices with ultra-low energy consumption. The followings are main the research achievements in the year of 2023.

1. Quantum Phenomena of Moiré Excitons in a WSe_2/$MoSe_2$ Heterobilayer

A quantum two-level system has garnered considerable attention in recent years due to its numerous potential applications in the fields of physics, such as quantum simulation, quantum computing, and quantum information processing. The development of these systems facilitates the construction and utilization of quantum bits (qubits), which serve as fundamental units for quantum computing and quantum information. Resonant light–matter interactions, such as Rabi oscillation, Ramsey interference, and Hahn echoes enable the manipulation of quantum two-level systems by generating superposition states. However, the superposition states of qubits suffer from the interaction and fluctuation from the environment, resulting in an accelerated decoherence process. This decoherence process imposes a temporal limitation on the precise manipulation of quantum systems, hindering their potential applications.

Recent progress in artificial van der Waals (vdW) structures, achieved by stacking atomically thin two-dimensional (2D) materials, has opened up opportunities for designing novel quantum platform. vdW heterobilayer assembled from monolayers of semiconducting transition metal dichalcogenides exhibited various intriguing physical phenomena, including strongly correlated insulator phases, superconductivity, and novel ferromagnetism. Moiré superlattices with varying atomic registries in vdW heterobilayers can be constructed using monolayer semiconducting transition metal dichalcogenides with a small lattice mismatch or twist angle. The resulting moiré superlattice leads to the formation of periodic, ordered potential traps, confining and spatially organizing optically generated bound electron–hole pairs (excitons) into

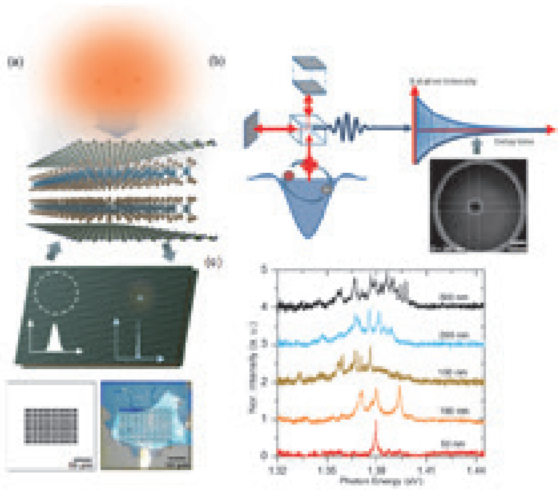

Figure 1 (a) (b) Schematic of the concept in this study. Nanoscale fabrication using reactive-ion etching enables to obtain emissions from a single moiré exciton and the observation of its quantum coherence with Michelson interferometer in $MoSe_2$/WSe_2 heterobilayer. Designed pattern for RIE (left) and the optical image of $MoSe_2$/WSe_2 heterobilayer with an array of nanofabricated structures (right) are presented in (a). A SEM image of typical pillar is presented in (b). The inner dotted circle shows a pillar with a diameter of 240 nm corresponding to the optical excitation and observation area of moiré potential. Optical spectra from the pillars are Fourier-transformed to temporal interferograms using the Michelson interferometer. (c) PL spectra of $MoSe_2$/WSe_2 heterobilayers with various pillar sizes at 4 K. (d) Integrated PL intensity and calibrated intensity of heterobilayer as a function of pillar size.

periodic arrays of quantum two-level systems. The trapped excitons in moiré potentials are expected to exhibit long quantum coherence due to their limited degree of freedom as 0D quantum systems; furthermore, coupling interactions can be formed between spatially separated moiré potentials, leading to quantum interference of emitted photons. These results establish moiré exciton quantum systems as not only a promising platform for achieving extended coherence but also an effective tool for exploring interactions within or between quantum systems. However, experimentally, important information on the quantum coherence and interference of moiré excitons in vdW heterobilayers remains unexplored. This is due to the overlapping of multiple emission peaks from moiré excitons in their inhomogeneously broadened spectra, hindering these intrinsic insights within the diffraction limit of light. The experimental approaches of strain-induced exciton traps introduced by metallic nanopillars in heterobilayers have been previously reported, to observe spectra with discrete emission lines; however, the intrinsic properties of the moiré exciton system might be inadvertently concealed due to the additional strain-induced effects.

Figures 1a and 1b illustrate schematics of the concept using a nanostructure fabrication process, and experimental setup employed in this study. Nanoscale fabrication using reactive-ion etching (RIE) enables emissions from a single moiré exciton and the observation of its quantum coherence in the $MoSe_2/WSe_2$ heterobilayer. During usual optical measurements, obtaining clear spectrum from moiré excitonic states is difficult due to the inhomogeneity of the moiré potential, leading to ensemble-averaged and broadened emissions comprising multiple peaks. This issue arises because the focused laser light with a spot size of approximately 1.5 μm is determined by the diffraction limit of light, which excites a large number of moiré potentials due to the much smaller spatial period of moiré potentials. To address this, in the microfabrication process, we applied a nanoscale fabrication technique to reduce the optical excitation and detection area of the $MoSe_2/WSe_2$ heterobilayer with a nano-pillar structure. The nanofabricated heterobilayer, with a pillar size smaller than the wavelength of light, will result in a reduced number of spectral peaks beyond the diffraction limit of light. Thus, this approach allows for the anticipated observation of emission from a single moiré potential, enabling the revelation of the quantum coherence of moiré excitons.

Figure 1c displays the photoluminescence (PL) spectra of the nanostructure-fabricated $MoSe_2/WSe_2$ heterobilayer, measured at 4 K with varying pillar sizes. In the heterobilayer with a pillar size of 500 nm, the PL spectrum reveals an inhomogeneously broadened ensemble average of multiple peaks from large number of moiré excitons. This finding agrees with previously reported results. The broadened spectrum, characterized by a Gaussian distribution, arises from the inhomogeneity of moiré potentials in the heterobilayer. As the pillar size decreases, the number of peaks in the spectra significantly decreases. Consequently, the PL spectrum of the heterobilayer with a 50-nm pillar size reveals a singular emission peak from a moiré exciton, attributed to the reduced number of moiré potentials within the optical excitation and detection area determined by the pillar size.

To obtain the information on the quantum coherence, the first-order correlation function of emission signals from a moiré exciton is measured using a Michelson interferometer. Figure 2a presents the counter map of interferometry of the PL spectra as a function of delay time at 4 K for an excitation power density of 14 W/cm^2. The amplitude of the oscillation fringe between the maximum and minimum intensities gradually decreases with increasing delay time, indicating

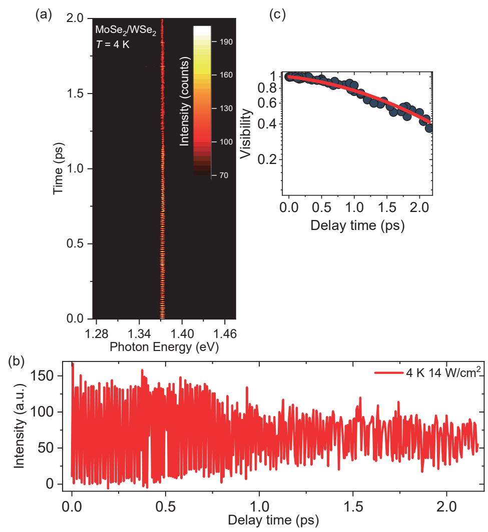

Fig. 2 (a) Counter plot of the first-order correlation function of PL signals as a function of delay time, measured using the Michelson interferometer. (b) Interferogram of the moiré exciton peak in the time domain at 4 K in the excitation power condition of 14 W/cm^2. (c) Decay profile of visibility in the interferogram, with solid curve representing the fitted result of the product of an exponential and a Gaussian func-

the process of decoherence, as presented by the temporal interferogram in Fig. 2b. Figure 2c shows the visibility as a function of delay time from Fig. 2b.

The visibility as a function of delay time, corresponds to the Fourier transform of the emission spectrum, with the convolution result of extrinsic inhomogeneous and intrinsic inhomogeneous linewidth, in form of Lorentz and Gaussian functions, respectively. Consequently, the delay time-dependent visibility in Fig. 2c can be modeled by the product of exponential and Gaussian functions. The fitted result by the model function of Lorentz and Gaussian functions well reproduce the experimental results of visibility as a function of delay time. With consideration of the energy relaxation lifetime, the population relaxation process hardly contributes to the dephasing process. As a result, pure dephasing time of this position is evaluated to be 7.1 ps, corresponding to a homogeneous linewidth of 184 μeV. The long coherence of moiré exciton revealed in this study offer potential applications of moiré quantum systems in quantum technologies.

Collaboration Works

松田一成，University of Bordeaux（フランス），単一ナノ物質における先端分光

Financial Support

1. Grant-in-Aid for Scientific Research

松田一成, 基盤研究(S), 原子層人工ヘテロ構造におけるバレースピン量子光学の開拓と応用

松田一成, 基盤研究(S), 低次元状態の埋め込みに基づく新物質科学（分担金）

松田一成, 学術変革領域研究(A)（計画研究）, ２.５次元構造の分析技術開発

松田一成, 学術変革領域研究(A), ２.５次元物質科学の総括（分担金）

松田一成, 挑戦的研究（萌芽）, バレースピン制御とデバイス応用：バレートロニクスに向けた課題と挑戦

篠北啓介, 基盤研究(B), モアレ超構造における協力的量子光学現象の開拓

朝田秀一, 特別研究員奨励費, 革新的エネルギー変換に向けた原子層人工ヘテロ構造の自発的光起電力に関する研究

2. Others

松田一成, 東京都公立大学, 配向制御ナノチューブを基盤にした日米共同クリーンエネルギー技術開発

篠北啓介, 科学技術振興機構 創発的研究支援, 半導体モアレ超構造を用いた量子電磁力学の創生

Publications

H. Kim, K. Aino, K. Shinokita, W. Zhang, K. Watanabe, T. Taniguchi, K. Matsuda, Dynamics of Moire Exciton in a Twisted $MoSe_2/WSe_2$ Heterobilayer, Advanced Optical Materials, 11, 2300146, 2023

Y-R. Chang, R. Nanae, S. Kitamura, T. Nishimura, H. Wang, Y. Xiang, K. Shinokita, K. Matsuda, T. Taniguchi, K. Watanabe, K. Nagashio, Shift Current Photovoltaics based on A Noncentrosymmetric Phase in in-plane Ferroelectric SnS, Advanced Materials, 35, 2301172, 2023

H. Kim, D. Dong, Y. Okamura, K. Shinokita, K. Watanabe, T. Taniguchi, and K. Matsuda, Dynamics of Moiré Trion and Its Valley Polarization in a Microfabricated $WSe_2/MoSe_2$ Heterobilayer, ACS Nano, 17, 14, 13715-13723, 2023

H. Naito, Y. Makino, W. Zhang, T. Ogawa, T. Endo, T. Sannomiya, M. Kaneda, K. Hashimoto, L.E. Lim, Y. Nakanishi, K. Watanabe, T. Taniguchie, K. Matsuda, Y. Miyata, High-throughput dry transfer and excitonic properties of twisted bilayers based on CVD-grown transition metal dichalcogenides, Nanoscale Advances, 5, 18, 5115-5121, 2023

Y. Zhang, K. Kamiya, T. Yamamoto, M. Sakano, X. Yang, S. Masubuchi, S. Okazaki, K. Shinokita, T. Chen, K. Aso, Y. Yamada-Takamura, Y. Oshima, K. Watanabe, T. Taniguchi, K. Matsuda, T. Sasagawa, K. Ishizaka, T. Machida, Symmetry Engineering in Twisted Bilayer WTe_2, Nano Letters, 23, 20, 9280-9286, 2023

M. Shimasaki, T. Endo, K. Watanabe, T. Taniguchi, K. Matsuda, T. Nishihara, Y. Miyata, Y. Miyauchi, Exciton energy transfer under low temperature in a lateral heteromonolayer of WSe_2–$MoSe_2$, Japanese Journal of Applied Physics, 62, 11, 112002, 2023

H. Wu, T. Nishihara, A. Takakuraa, K. Matsuda, T. Tanaka, H. Kataura, Y. Miyauchi, Birefringent optical responses of single-chirality carbon nanotube membranes, Carbon, 218, 118720, 2023

W. Zhang, Z. Liu, H. Nakajo, S. Aoki, H. Wang, Y. Wang,Y. Gao, M. Maruyama, T. Kawakami, Y. Makino, M. Kaneda,T. Chen, K. Aso, T. Ogawa, T. Endo, Y. Nakanishi,K. Watanabe, T. Taniguchi, Y. Oshima, Y. Yamada-Takamura, M. Koshino, S. Okada, K. Matsuda, T. Kato, Y. Miyata, Chemically Tailored Semiconductor Moiré Superlattices of Janus Heterobilayers, Small Structures, 2300514, 2024

篠北啓介, 松田一成, ねじれて積層した 2 次元半導体のモアレ構造における新規な励起子光物性, 応用物理, 92, 9, 524-529, 2023

越野幹人, 松田一成, 岩佐義宏, 2 次元ファンデルワールス物質の概観, 固体物理, 58, 11. 1-14, 2023

篠北啓介, 松田一成, 2 次元半導体とそのモアレ構造の光科学, 固体物理, 58.11. 117-132, 2023

松田一成, 二次元半導体ヘテロ構造におけるモアレの物理と光科学, 遷移金属ダイカルコゲナイドの基礎と最新動向, 第 5 章, 2023

Presentations

K. Matsuda, Optical science in two-dimensional material and its hetero-structures, The 12th A3 Symposium on Emerging Materials, Hybrid (Korea), 2023.2.14-16

松田一成, 研究会「１次元、２次元物質科学の展望と課題」, ナノサイエンスに立脚した光科学：未来へ向けて, 東北大学青葉山キャンパス, 2023.3.10

K. Matsuda, Optical science in nanocarbon and artificial atomically thin 2D materials, Japan-US Network for Clean Energy Technologies Involving Oriented Nanotubesin PIRE, 横浜情報文化センター, 2023.5.22-23

松田一成, ナノサイエンスで拓く二次元半導体の光科学とその展望, 大阪公立大学工学研究科電子物理セミナー, 大阪公立大学, 2023.7.13

K. Matsuda, Recent progress on optical physics in two-dimensional materials and its hetero-structures, The 13th A3 Symposium on Emerging Materials, Nanjing University, 2023.7.20-22

K. Matsuda, Optical science and applications of quantum materials for carbon negative energy science, The 14th International Symposium of Advanced Energy Science, 京都大学宇治キャンパス, 2023.9.1

H. Kim, D. Dong, Y. Okamura, K. Shinokita, K. Watanabe, T. Taniguchi, and K. Matsuda, Dynamic of moiré trion in mircrofabricated $WSe_2/MoSe_2$ heterobilayer, The 65th Fullerenes-Nanotube-Graphene General Symposium, Nishijin Plaza, 2023. 9.5

S. Shomura, K. Shinokita, K. Watanabe, T. Taniguchi, and K. Matsuda, Valley polarized hybridized excitons in $MoSe_2/WS_2$ heterobilayers, The 65th Fullerenes-Nanotube-Graphene General Symposium, Nishijin Plaza, 2023. 9.5

村瀬大騎, 篠北啓介, 若藤祐斉, 小野寺桃子, 町田友樹, 谷口尚, 渡邊賢司, 松田一成, エッチングフリー金剥離法による二次元半導体作製手法の確立, 第 84 回応用物理学会秋季学術講演会, 熊本城ホール, 2023.9.19

平林弘暉, 篠北啓介, 渡邊賢司, 谷口尚, 松田一成, 面内分極を持つ MoS_2/SnS ヘテロ界面による自発的光起電力, 第 84 回応用物理学会秋季学術講演会, 熊本城ホール, 2023.9.19

K. Shinokita, Moiré excitonic states in a twisted $WSe_2/MoSe_2$ heterobilayers, JSAP-Optica Joint Symposia 2023, 熊本城ホール, 2023.9.20

篠北啓介, ツイスト二次元ヘテロ構造におけるモアレ励起子の光科学, 第 84 回応用物理学会秋季学術講演会, 熊本城ホール, 2023.9.21

松田一成, ナノサイエンスによる二次元半導体の光科学とその進展, 光圧マニュピレーション研究会, オンライン, 2023.10.20

Y. Yubei, K. Shinokita, K. Wanatabe, T. Taniguchi, and K. Matsuda, Magnetic Brightening of Defect localized Exciton Emission in Monolayer WSe_2, 13th A3 Symposium on Emerging Materials: Nanomaterials for Electronics, Energy, and Environment, Korea University, 2023.10.30-11.1

S. Asada, K. Shinokita, K. Wanatabe, T. Taniguchi, and K. Matsuda, Nonlinear Spontaneous Photocurrent in $MoS_2/CrPS_4$ van der Waals Heterointerface, 13th A3 Symposium on Emerging Materials: Nanomaterials for Electronics, Energy, and Environment, Korea University, 2023.10.30-11.1

K. Matsuda, Quantum optics based on moiré excitonic systems in artificial van der Waals semiconducting heterostructures, 13th A3 Symposium on Emerging Materials: Nanomaterials for Electronics, Energy, and Environment, Korea University, 2023.10.30-11.1

W. Idehara, F. Yang, K. Shinokita, K. Matsuda, Efficient robotic fabrication of large area monolayer semiconductors, The 66th Fullerenes-Nanotube-Graphene General Symposium, Nagoya, 2024.3.6

Y. Wang, K. Shinokita, and K. Matsuda, Strain-influenced optical properties of moiré excitonic states in $MoSe_2/WSe_2$ van der Waals heterostructures, The 66th Fullerenes-Nanotube-Graphene General Symposium, Nagoya, 2024.3.7

Y. Yubei, K. Shinokita, K. Wanatabe, T. Taniguchi, and K. Matsuda, Dynamic of defect-localized excitons in monolayer WSe2 under in-plane magnetic field, The 66th Fullerenes-Nanotube-Graphene General Symposium, Nagoya, 2024.3.8

呉柊斗, 篠北啓介, 杉本泰, 藤井稔, 松田一成, Mie 共鳴を示すシリコンナノ粒子と結合した単層 WSe_2 の光学特性, 第 71 回応用物理学会春季学術講演会, 東京都市大学, 2024.3. 22

出原渉, 篠北啓介, 松田一成, Y. Fan, ロボット技術支援による高効率・大面積な単層半導体作製システムの構築, 第 71 回応用物理学会春季学術講演会, 東京都市大学, 2024.3. 22

朝田秀一, 篠北啓介, 松田一成, MoS_2/$CrPS_4$ 人工ヘテロ構造デバイスにおける磁気光起電力特性, 第 71 回応用物理学会春季学術講演会, 東京都市大学, 2024.3. 23

松田一成, 二次元半導体人工ヘテロ構造におけるモアレ光科学とその応用, 第 71 回応用物理学会春季学術講演会, 東京都市大学, 2024.3.24