

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 2025.

1. Spin-dependent photovoltaic effects in monolayer semiconductor and layered magnetic material hetero-interface with P - and T - symmetry broken system

Recently, the introduction of shift current concepts from topological physics drastically extend the possibilities for photovoltaic devices because of their unconventional spontaneous photocurrent properties. The shift current as one of the bulk photovoltaic effects are the most promising photovoltaic responses to be able to overcome the Shockley-Queisser limit in the conventional p-n junction photovoltaic devices^{5–8}. These anomalous photovoltaic responses including extremely large open-circuit voltage, not limited by band-gap energy, and robust photocurrent to defects and impurities of materials, have been observed in the polar bulk materials. The shift current could be explained as topological photocurrent depending on Berry connection, which is a vector in real space indicating the information of center-of gravity position of electrons. In the crystal with broken spatial inversion (P -) symmetry, the imbalance electron “shift” due to optical interband transitions before and after photoexcitation is occurred, which then generates the spontaneous photocurrent under zero-bias conditions as shift current. In addition to the shift current, the nonlinear topological photoresponses also induce the injection current, which is caused by difference of group velocity and imbalance photoexcitation with circular polarized light in k-space^{16–18}. Since the shift current and injection current are induced by the linear- and circular-photogalvanic effects (LPGE and CPGE) of linearly and circularly polarized light, respectively, their effects are generally observed separately. However, in time-reversal (T -) symmetry broken crystals such as some layered magnetic materials, each spin band is

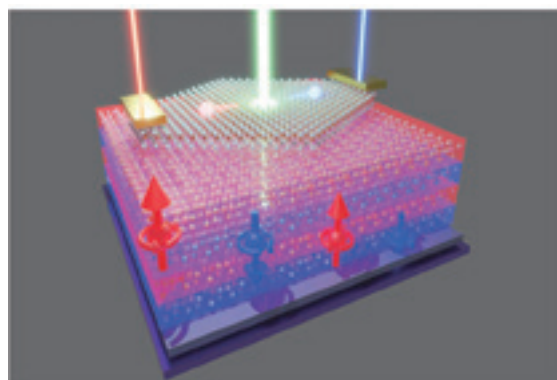


Figure 1 Schematics of bulk photovoltaic effect in nonlinear photocurrent responses of MoS₂/CrPS₄ vdW heterostructure with the system with broken P - and T -symmetry.

lifted by spin polarization and shows an imbalanced band structure in momentum space. The electron is photoexcited unevenly in terms of momentum, and the group velocity of the electron is not perfectly cancelled, which would induce a linear injection current in systems with broken T -symmetry. The shift current is also induced by the CPGE after the magnetic transition. These magnetic PGEs have odd parity under magnetization changes; thus, they are not observed in nonmagnetic materials where T -symmetry is preserved.

The emerging two-dimensional (2D) materials, including semiconducting transition metal dichalcogenides have opened the new research fields in the fundamental science and potential applications in various electronic and optical applications. The artificial van der Waals (vdW) heterostructures fabricated by stacking 2D materials lead to induce the emerging periodicity of crystal structures such as moiré superlattice and provide us the new pathways to control the P -symmetry at their heterointerfaces. Recently, it has been reported the nonlinear photocurrent arising from shift current in the artificially P -symmetry breaking vdW heterointerface by stacking three-fold symmetry MoS₂ and two-fold symmetry black phosphorous. Moreover, the artificial symmetry breaking would extend the exploration for high efficiency shift current photovoltaic devices, however, no promising candidates have been shown. Accordingly, it is crucial to experimentally investigate the shift current and injection current depending on Berry connection and Berry curvature in

the novel interface of vdW heterostructure with the systems with broken P - and T -symmetry. The understanding of nonlinear photocurrent responses in the vdW heterostructure with the system with broken P - and T -symmetry would provide the ground breaking importance for fundamental physics and applications for emerging photovoltaics.

We studied nonlinear photocurrent responses of $\text{MoS}_2/\text{CrPS}_4$ vdW heterostructure with the system with broken P - and T -symmetry. The finite spontaneous photocurrent arising from shift current along the parallel axis to mirror plane at the interface of $\text{MoS}_2/\text{CrPS}_4$ has been observed. Moreover, we demonstrated that the spontaneous photocurrent drastically changes below Neel temperature of A-type AFM CrPS_4 in $\text{MoS}_2/\text{CrPS}_4$ heterostructure. The spontaneous photocurrent also changes depending on external magnetic field below the Neel temperature, accompanied by field induced magnetic phase transition. The critical suppression (enhancement) of spontaneous photocurrent below Neel temperature is caused by competition of the shift current and sign of magnetic injection current in $\text{MoS}_2/\text{CrPS}_4$ vdW heterointerface. Our results of magnetic switchable photovoltaic responses demonstrated here provide the new aspects on the nonlinear photovoltaic effects in P - and T -symmetry breaking vdW heterointerface, and new strategy for next-generation solar cells.

2. Enhanced single-photon emission from defect-localized exciton in monolayer two-dimensional semiconductor

Two-dimensional transition-metal dichalcogenides (TMDs), specifically MX_2 compounds ($M = \text{Mo}, \text{W}$; $X = \text{S}, \text{Se}, \text{Te}$), have emerged as promising platforms for nanoscience and novel optical devices due to unique electronic and optical properties, including layer-thickness-dependent bandgaps. Monolayer semiconducting TMDs exhibit a direct bandgap at the K and K' valleys in momentum space. The coupling of valley and spin degrees of freedom, or valley-spin locking, results in valley-dependent optical selection under circularly polarized light. Under this light, optically generated electron-hole pairs form valley excitons through strong Coulomb interactions and dielectric screening effects at the K (K') valleys. Spin-dependent exciton transitions are classified as either optically allowed singlet bright or optically forbidden singlet dark. Experimental and theoretical studies have explored dark exciton control through defect engineering and external magnetic fields, both in-plane and out-of-plane. Additionally, precise control over magnetic field strength and orientation enables manipulation of exciton spins, enhancing radiative recombination and brightening dark excitons.

Single-photon emitters are vital for quantum technologies, enabling advancements in quantum key distribution, quantum computing, and quantum sensing.

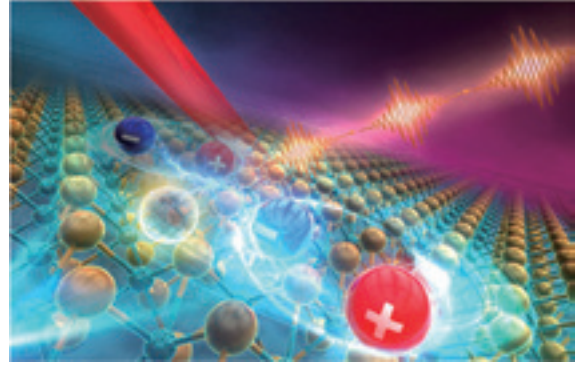


Figure 2 Schematic of dynamics and single photon emission of defect-localized excitons in monolayer WSe_2 under a magnetic field.

Developing stable, efficient, and controllable single-photon sources is essential. Monolayer tungsten diselenide (WSe_2) with a single defect shows promise as a single-photon source. Single-photon emission has been achieved from point defects, such as vacancies and antisites, introduced into monolayer WSe_2 through annealing, plasma treatment, or ion irradiation. Defect-localized excitons feature two spin states, resulting from defect-induced mismatch and disorder, which disrupt spin-momentum locking in monolayer WSe_2 . This spatial localization leads to the hybridization of bright and dark states, extending lifetimes. However, the dynamics of defect-localized excitons with two spin states remain unclear, and understanding the mixing and transition processes of these excitons under an external magnetic field is crucial for controlling single-photon emissions.

We investigated the nature and dynamics of defect-localized excitons in monolayer WSe_2 under a magnetic field. A sharp PL peak, attributed to defect-localized emission, exhibited photon antibunching behavior in second-order correlation function measurements, indicating it acts as a single-photon emitter. This sharp PL peak, comprising doublet peaks with fine-structure splitting of less than 1 meV, originates from defect-localized exciton emissions of two hybridized intra- and inter-valley bright and dark excitons trapped and hybridized at the defect site. Additionally, magnetic brightening of the $|\text{IDE}\rangle$ exciton state occurs even in relatively low magnetic fields below 1 T, which is much weaker than similar effects in other spin-related free dark excitons in monolayer semiconductors and carbon nanotubes. Furthermore, the dynamics of localized excitons in the $|\text{IDE}+\rangle$ and $|\text{IDE}\rangle$ states under magnetic fields demonstrate field-induced mixing, driving the magnetic brightening phenomena of defect-localized exciton emission. Our findings on the microscopic physical properties and dynamics of defect-localized exciton states illuminate magnetic brightening and suggest a new strategy for single-photon emitters controllable by an external magnetic field for quantum optics applications.

Collaboration Works

松田一成, University of Bordeaux (フランス), ナノ物質における光科学の開拓

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

松田一成, 基盤研究(S), ロボット技術支援によるモアレ量子プラットフォームの創生と応用

松田一成, 科学技術振興機構 戦略的創造研究推進事業チーム型研究 (CREST), 二次元半導体・ヘテロ構造の量子光プラットフォームの構築と応用

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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)

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