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 2022.

1. Valley Relaxation of the Moiré Excitons in a WSe_2/$MoSe_2$ Heterobilayer

Due to their unusual physical properties and ability to function as extreme two-dimensional (2D) systems, atomically thin materials such as graphene and semiconducting transition-metal dichalcogenides (TMDs) and their van der Waals (vdW) heterostructures have gained increasing attention. The physical properties of atomically thin materials originating from a particular atomic arrangement are significantly altered by emerging moiré superlattices made up of lattice- or angular-mismatched vdW heterostructures. The moiré superlattice is gaining attention for its ability to engineer optically excited, tightly bound electron-hole pairs (excitons) and can result in exciton trapping through in-plane periodic moiré potentials in twisted semiconducting TMDs heterobilayers. The quantum confined zero-dimensional (0D) as a two-level system created by the moiré exciton trapped in the moiré potentials is encouraging for the development of quantum optics and quantum information processes.

The confinement of moiré potential of exciton alters the optical characteristics of the system. Under low-excitation power density conditions, the moiré exciton results in the appearance of sharp peaks in the low-temperature photoluminescence (PL) spectrum. Additionally, the moiré pattern (atomic registry) has a significant impact on the optical selection rule of the exciton. The intrinsic valley degrees of freedom control the optical properties of monolayer TMDs. The K and -K valleys at the edge of the Brillouin zone are inequivalent but energetically degenerate due to the strong spin-orbit interaction and lack of inversion symmetry. The optical selection rule and corresponding circular polarized light emission are both valley-dependent due to the inherent coupling of the valley and spin degrees of freedom.

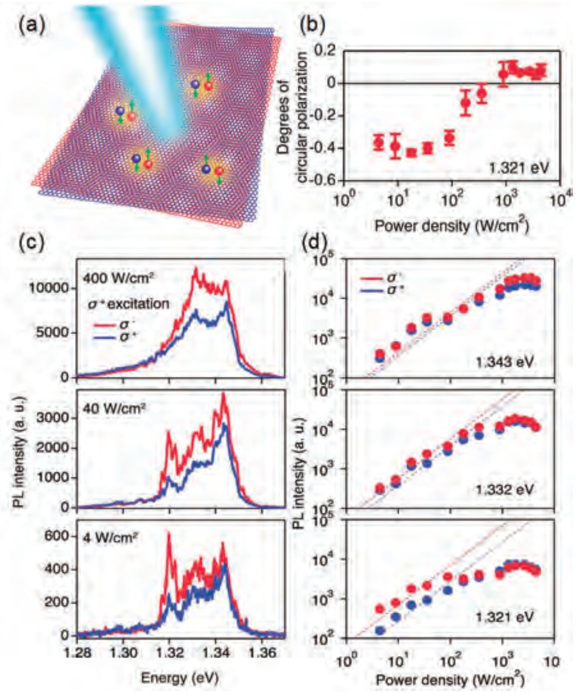

Fig. 1 (a) Schematic of moiré exciton in WSe_2/$MoSe_2$ heterobilayer. (b) Degree of circularly polarization as a function of excitation power density. (c) Low-temperature circularly polarized PL spectrum under σ+ circularly polarized excitation (1.676 eV) with different excitation power densities. The σ+ and σ– circularly polarized components are shown as the blue and red lines, respectively. (d) Excitation power density dependence of the PL intensity monitored at 1.321, 1.332, and 1.343 eV in the polarization-resolved PL spectra.

Contrary to monolayer TMDs, the optical selection rule of moiré superlattice also influences the circularly polarized light emission due to the atomic registry's C_3 symmetry and the valley degrees of freedom. The fascinating optical and electrical properties requires an understanding of the intra- and intervalley relaxation processes. The valley relaxation process in monolayer TMDs has thus been successfully explained and controlled through extensive experimental and theoretical studies. Electro-static carrier doping techniques were used to continuously control the valley relaxation process by altering the electron-hole exchange interaction. It has been discovered that momentum-dependent long-range electron-hole exchange interactions dominate the valley relaxation of neutral excitons in monolayer TMDs. On the other hand, it is still unclear how the valley relaxation process in the moiré superlattice works, where the reduced dimensionality might also have an impact.

Here, we used circularly polarized PL and photoluminescence excitation (PLE) spectroscopy to investigate the intra- and intervalley relaxation of the moiré excitons in a twisted $WSe_2/MoSe_2$ hetero-bilayer. Contrary to 2D monolayer TMDs, the experimentally observed circularly polarized emission is strongly dependent on the excitation power density. The excitation power density dependence of circularly polarized emission leads to the inter-valley relaxation of the moiré exciton, which are originated from Pauli blocking due to the low density of states typical of 0D systems. The intravalley relaxation of the moiré exciton states from the triplet to singlet state via Γ_5 phonon emission is also revealed by the resonant PLE measurement. Circularly polarized quantum light emitter may be used in quantum optics and information processing as a result of our discoveries regarding the valley relaxation of moiré excitons.

2. Dynamics of Moiré Exciton in a Twisted $MoSe_2/WSe_2$ Heterobilayers

Studies in artificial vdW heterostructures made of two atomically thin 2D materials have received a lot of attention. A number of distinct physical phenomena, such as superconductivity, ferromagnetism near 3/4 filling, correlated insulator phases, and Hofstadter butterflies in graphene moiré superlattices, have new pathways thanks to the recent discovery of moiré superlattices at the interface of vdW hetero-structures with small lattice mismatches or twist angles between two monolayers. When optically generated electrons and holes in a spatially separated layer interact via Coulomb interactions, an interlayer exciton is created. This exciton is then contained within the moiré trap potential and spatially organized as a moiré exciton ensemble. The dense array of coherent quantum emitters is one application of the moiré trapped exciton that has great potential for many-body physics. However, little is known about the discrete excitonic states and dynamics of the confined exciton within the moiré potential.

The excitons can be divided into optically active bright excitons and inactive dark excitons in semiconductors. The dipole-allowed bright exciton exhibits radiative recombination during the emission process and directly couples to light via strong light-matter interaction. Optical absorption and PL spectroscopy makeit possible to observe and track the dynamic behaviors of bright excitons. However, due to the weak light-matter interaction and reduced oscillator strength, optical access to the dark exciton is challenging. Dark excitons can be used in research on spin storage and long-lived qubits. The dark exciton state in monolayer 2D materials exhibits nonequilibrium dynamics as well as optical responses, and studies have shown that

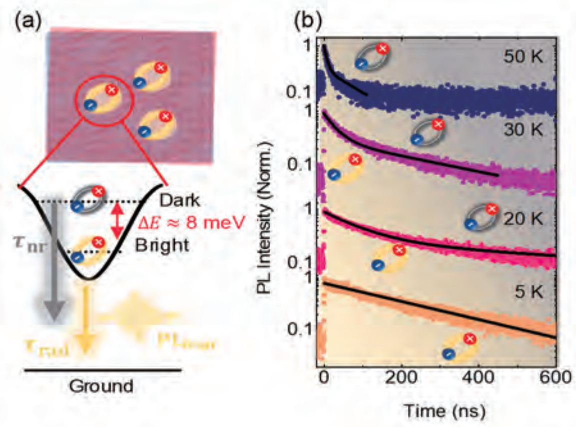

Fig. 2 (a) Schematic of moiré excito states confined in the potential. The splitting of bright and dark moire exciton states is about 8 meV. (b) PL decay profiles at various temperatures. The contribution of bright and dark moiré exciton states is changed from low to high temperature.

it is energetically inferior to the bright exciton state.

The energy structures and characteristics of the bright and dark moiré exciton states in the moiré potential, however, may raise important and crucial questions. Therefore, it is essential to conduct experiments to demonstrate the existence of dark exciton within moiré potential. The dynamics of bright and dark moiré excitonic states in twisted semiconducting vdW heterobilayers requires further research in order to fully understand their revolutionary significance for fundamental moiré physics and potential quantum optics applications in the novel platform of moiré exctonic systems.

Here, we used PL spectroscopy and rate-equation analysis to investigate the moiré exciton states and their dynamics in twisted $MoSe_2/WSe_2$ heterobilayers. The experimental results demonstrate the existence of a dark moiré exciton state above the lowest emissive bright singlet exciton state, and the dynamics of bright excitons are determined by radical recombination and transition to the dark exciton state with the aid of phonons. In-depth information is provided regarding the dynamics and nature of moiré exciton states.

Our findings lay the groundwork for further investigation into quantum phenomena in moiré physics for use in quantum optics while illuminating novel aspects of quantum states of moiré excitons and fascinating moiré exciton dynamics.

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，学術変革領域研究(A)，２.５次元構造の分析技術開発

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

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）

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

田中絢也，特別研究員奨励費，室温動作超広帯域光検出器の実現に向けた量子物質赤外応答の解明

2. Others
篠北啓介，創発的研究支援事業，半導体モアレ超構造を用いた量子電磁力学の創生

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