Nano Optical Science Research Section

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1. Introduction

We are working on basic and applied research of nano-materials from a viewpoint of optics and material science. Our research aims at exploring new physical and chemical phenomena leading to applications of novel nano-materials including carbon nanotubes, layered transition metal dichalcogenides, perovskite for efficient utilization of light energy and development of future optoelectronic devices with ultra-low energy consumption. Followings are main research achievements in the year of 2021.

1. Moiré exciton-phonon coupling in a $WSe_2/MoSe_2$ heterobilayer

The interference of two similar patterns is a universal concept in physics that plays a pivotal role in modern science and technology such as in gravitational wave detection, optical frequency combs, superconducting quantum interference devices (SQUIDs), and cold atoms in optical lattices. The moiré patterns of van der Waals heterostructures arising from interference of angular- or lattice-mismatched atomically thin materials with honeycomb structures, such as graphene and semiconducting transition metal dichalcogenides (TMDs), have attracted increasing attention because of the potential for engineering a range of emergent quantum phenomena. Examples include superconductivity, ferromagnetism near ¾ filling, and correlated insulator phases in twisted bilayer graphene. In a two-dimensional (2D) semiconducting TMD heterostructure, the stacking of two different monolayer TMDs usually results in staggered type II band alignment, which causes separation of electrons and holes in different layers, or interlayer excitons (Coulomb-bound electron-hole pairs). The nature of the interlayer excitons is modulated by the moiré pattern because of the spatially varying atomic registry. The moiré pattern works as a periodic trap potential to confine the interlayer exciton in zero dimensions (0D) (moiré exciton, Fig. 1) and spatially organize the moiré-trapped excitons, which results in an array of quantum-dot-like 0D systems composed of a moiré exciton ensemble. In addition, the moiré period and interaction between the moiré excitons can be tailored by the stacking angle. Therefore, moiré exciton ensembles in periodic moiré potentials have potential for dense coherent quantum emitters and quantum simulation of many-body physics, which could result in a number of applications in quantum optics, including quantum dot lasers, entangled photon lasers, and Dicke superradiance.

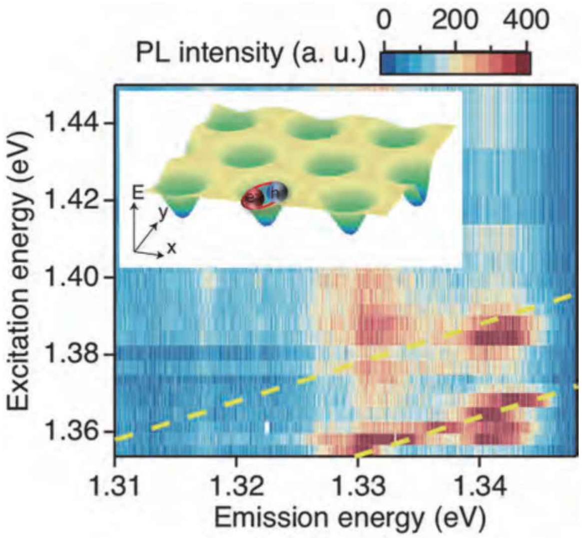

Fig. 1 2D PLE map of $WSe_2/MoSe_2$ heterobilayer, measured at low temperature. Dotted lines correspond to phonon resonances. Inset shows schematic of moiré exciton.

The electronic, optical, and transport properties of solids are frequently dominated by electron-phonon or exciton-phonon interactions. Extensive works on exciton and phonon interactions in 2D materials and their van der Waals heterostructures have been performed. For instance, emergent interlayer exciton-phonon coupling was observed in a WSe_2/*h*-BN heterostructure system, which provides important information for the generation and control of intriguing physical properties of 2D materials. The exciton-phonon interaction of the heterostructure can also be modified by the periodic moiré potential, which would pave a new way for control of diverse fascinating physical behaviours of 0D-like moiré excitons towards coherent quantum emitters and quantum simulation of many-body physics. To date, the signatures of moiré excitons have been optically studied by absorption and photoluminescence (PL) measurements, where the moiré exciton was confirmed by the appearance of sharp peaks in low-temperature PL spectra under low excitation power conditions, reflecting the trapping of excitons in the moiré potential. However, the interaction between the moiré exciton and phonon have yet to be studied experimentally. To explore novel quantum phenomena in moiré superlattices, it is important to understand the moiré exciton-phonon interaction, which play a dominant role in the intriguing properties of moiré exciton ensembles and quantum applications.

Here, we study the moiré exciton and phonon interaction in a twisted $WSe_2/MoSe_2$ heterobilayer based on near-resonant photoluminescence excitation (PLE) spectroscopy, taking advantage of extraction of coupling of specific moiré exciton to phonon. The experimentally observed PL spectrum strongly depending on the excitation energy shows highly selective excitation of the ground state of the moiré exciton at phonon resonances. On the other hand, the negligibly small off-resonant PLE signal in the interlayer region suggests δ-function discrete energy levels, which reflects density of states of a 0D-like system for the interlayer moiré exciton. In addition, the excitation power dependence of the PL spectra reveals the moiré exciton dynamics between different potential minima with discrete energy levels via the resonant phonon scattering process.

Our results shed light on new aspects of moiré exciton and phonon coupling and lay the groundwork to explore quantum phenomena in moiré superlattices for quantum emitters with extremely low threshold lasing and so on.

2. Experimental Evidence of Magonon-Moiré Trion Complex in Monolayer Semiconductor and Antiferromagnet vdW Heterostructures

Since the discovery of 2D layered ferromagnetic and antiferromagnetic materials, the intriguing magnetic properties of 2D materials have attracted considerable interest in the research field of material science. Among 2D layered magnetic materials, transition metal phosphorous dichalcogenides ($TMPX_3$; TM = Mn, Ni, Fe, Co; X = S, Se) have been extensively studied as a model system of layered magnetic materials. For instance, free excitons coupled to magnons were reported in both bulk antiferromagnetic $MnPS_3$ and heterostructures composed of semiconducting $MoSe_2$ and antiferromagnetic $MnPSe_3$. Moreover, novel excitons with an ultra-narrow PL peak were found in bulk $NiPS_3$; these excitons arise from the many-body states of Zhang–Rice singlets and reach a coherent state assisted by the antiferromagnetic order. However, the interaction between moiré excitonic states (excitons and trions) and magnetic elementary excitations in magnetic materials has yet to be experimentally studied. To explore novel quantum phenomena in moiré superlattices, it is important to prove the existence of moiré excitonic states (excitons and trions) coupled with magnetic elementary excitations, which would give rise to the intriguing properties of moiré excitonic systems with magnetic functionalities.

Here, we report the emergence of intralayer trions

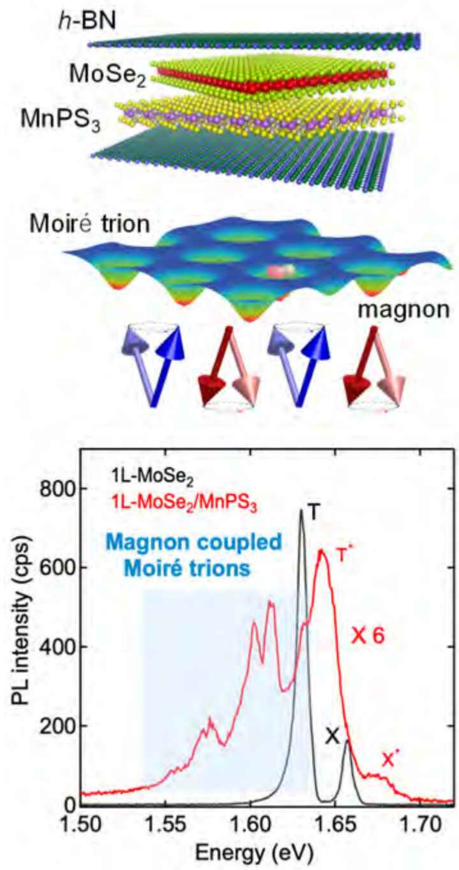

Fig. 2 Schematic of $MoSe_2/MnPS_3$ vdW heterostructure. Schematic of magnon-moiré charged exciton (trion) complex. Low temperature photoluminescence spectra of reference monolayer $MoSe_2$ and $MoSe_2/MnPS_3$ vdW heterostructure. Exciton and trion related peaks are denoted as X and T, respectively.

localized in the moiré potentials formed by twisted monolayer $MoSe_2$ and antiferromagnetic $MnPS_3$ vdW heterostructure. We carefully investigated the low-temperature PL spectra of the vdW heterostructure and found additional fine spectral structures on the low-energy side of the coupled magnon–trion peaks below the Néel temperature (78 K) of $MnPS_3$ (Figure 2). The fine spectral structures with long lifetime and coherence time are assigned to localized intralayer trion–magnon complexes in the moiré potentials (moiré trion–magnon complexes), which makes the moiré excitons different from those frequently observed in bilayer semiconducting TMD heterostructures, implying the appearance of magnetic moiré excitonic states.

We envisage that novel vdW heterostructures characterized by a combination of monolayer semiconductor TMDs and antiferromagnetic $TMPX_3$ will reveal the existence of magnetic moiré excitonic states. The precise energies and light emission intensities of these magnetic moiré-excitonic states can be tuned and controlled by applying external magnetic fields. Therefore, these states also provide a good platform toward for future application of quantum emitters with magnetic functionalities.

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

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，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），高次データー科学による原子層物質のバレースピン制御

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

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