Interlayer exciton formation, relaxation, and transport in TMDs van der Waals heterostructures

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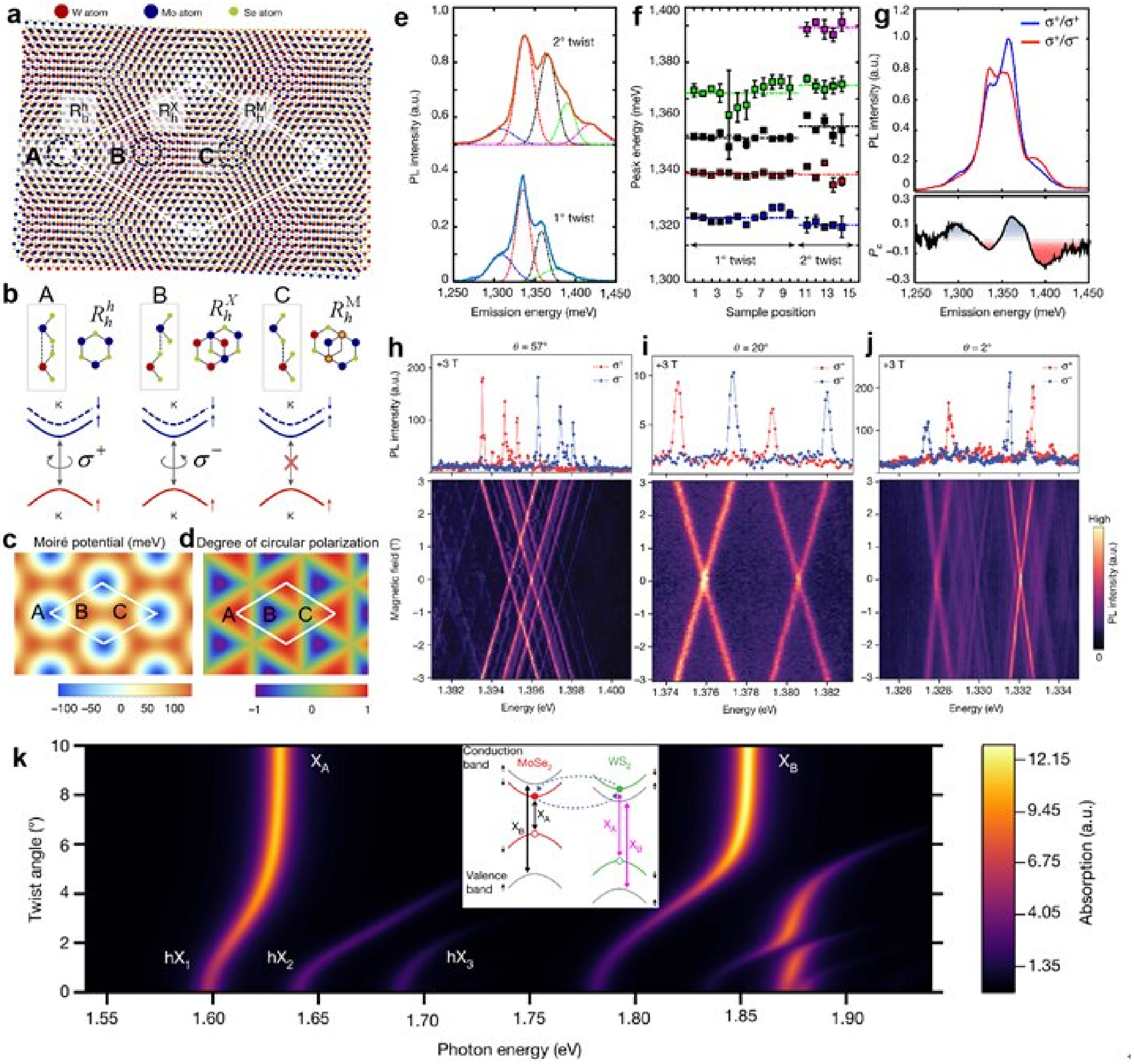
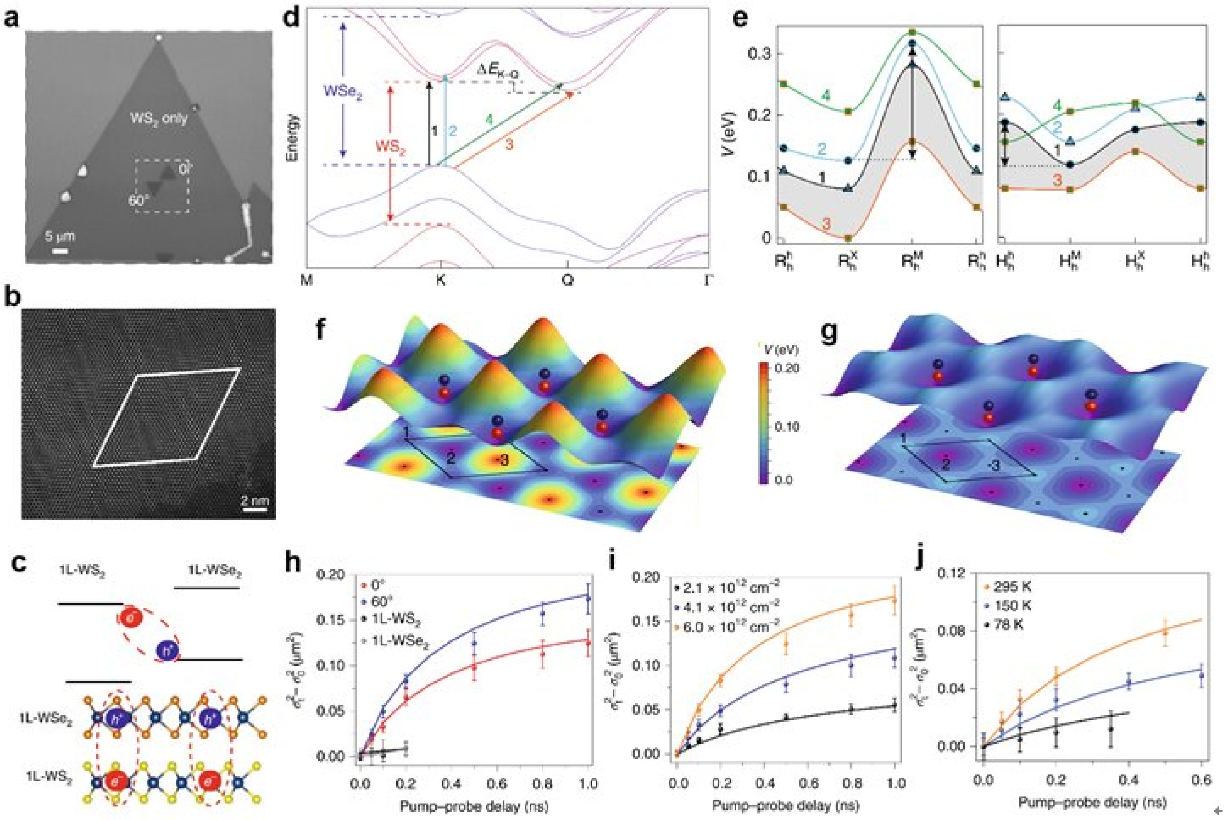


a Moiré pattern in a R-type MoSe2/WSe2 heterobilayer. The three highlighted regions (A, B, and C sites) correspond to the local atomic configurations with three-fold rotational symmetry. b The side- and top-view of the three R-type local atomic registries (A, B, and C sites) and the corresponding optical selection rules for the interlayer exciton in these atomic registries. c Moiré potential of the interlayer exciton transition with a local minimum at A site. d Optical selection rules for K-valley interlayer excitons. e PL spectra of multiple moiré interlayer excitons in MoSe2/WSe2 heterobilayers with twist angles of 1° (bottom) and 2° (top). Each spectrum is fitted with four (1°) or five (2°) Gaussian functions. f The centre energy of each moiré interlayer exciton resonance at different spatial positions across each sample. g Circularly polarized PL spectrum of the 1° sample under σ+ excitation (top). The degree of circular polarization versus the emission wavelength is shown in the bottom, demonstrating the multiple moiré interlayer excitons with alternating co- and cross-circularly polarized emission. h-j Magnetic-field-dependent PL from moiré-trapped interlayer excitons in MoSe2/WSe2 heterobilayers with twist angles of 57° (h), 20° (i) and 2° (j). Top: circularly polarization-resolved PL spectra with narrow linewidth (100 μeV) at 3 T. Bottom: total PL intensity as a function of magnetic field, displaying a linear Zeeman shift of the σ+ and σ? polarized components. k Absorption spectrum of the MoSe2/WS2 heterobilayer as a function of twist angle. The MoSe2 A- and B-exciton resonances (XA and XB) are indicated for large twist angles where hybridization effects become negligible. The three resonances labelled hX1,2,3 appearing at θ ? 0° correspond to the hybridized excitons in the vicinity of XA. Credit: Ying Jiang, Shula Chen, Weihao Zheng, Biyuan Zheng and Anlian Pan

Interlayer excitons in transition metal dichalcogenides (TMDs) van der Waals (vdW) heterostructures exhibit fascinating physics and hold great promise for developing excitonic devices. Scientists in China present a systematical and comprehensive overview of the interlayer exciton formation, relaxation, transport, and potential applications of TMDs vdW heterostructures, in order to provide valuable guidance for new researchers in this field as well as to present the most important issues present in the field for future deep studies.

TMDs vdW heterostructures generally possess a type-II band alignment which facilitates the formation of interlayer excitons between constituent monolayers. Manipulation of the interlayer excitons in TMDs vdW heterostructures hold great promise for developing excitonic integrated circuits that serve as the counterpart of electronic integrated circuits, which allows photons and excitons to transform between each other and thus bridges optical communication and signal processing at the integrated circuit. Consequently, numerous researches have been carried out in order to get a deep insight into the physical properties of interlayer excitons, including the revealing of their ultrafast formation, long population recombination lifetimes, and intriguing spin-valley dynamics. These outstanding properties ensure the interlayer excitons with good transport characteristics and may pave the way for their potential applications in efficient excitonic devices. At present, a systematical and all-round overview of these fascinating physics as well as the exciting applications of interlayer excitons in TMDs vdW heterostructures is still lacking and highly desirable for the scientific community.

In a new review paper published in *Light Science & Applications*, a team of scientists, led by Professor Anlian Pan from Key Laboratory for Micro-Nano Physics and Technology of Hunan Province, School of Physics and Electronics, and College of Materials Science and Engineering, Hunan University, China, and co-workers have given a comprehensive description and discussion of the interlayer exciton formation, relaxation, transport, and the potential applications in excitonic optoelectronic devices, based on TMDs vdW heterostructures. An outlook for future opportunities for interlayer excitons in TMDs-based heterostructures was also presented in this review.



a Optical image of two CVD-grown WS2/WSe2 heterobilayers with twist angles of 0 and 60° on the same WS2 underlayer. b High-resolution annular dark-field scanning transmission electron microscopy image of the 60° heterobilayer. The white diamond outline shows the moiré superlattice with a periodicity of ~7.6?nm. c Schematic illustration of the WS2/WSe2 heterobilayer with a type-II band alignment to facilitate the interlayer exciton formation. d Schematic representation of a typical electronic band structure of a WS2/WSe2 heterobilayer in a (strained) primitive unit cell. The four lowest-energy transitions are indicated by arrows (K-K valley transitions are denoted by vertical arrows 1 and 2, and K-Q valley transitions are denoted by vertical arrows 3 and 4). The K-K transitions in individual WS2 and WSe2 monolayers are marked by vertical arrows WS2 and WSe2, respectively. e Approximate moiré potentials for the twist angles of 0° (left) and 60° (right) plotted along the main diagonal of the moiré supercells (black lines in f). f, g Illustrations of the 2D K-K moiré potentials in both 3D graphs and 2D projections to trap interlayer excitons (red and black spheres) in the local minima for 0° (f) and 60° (g) heterobilayers. h

Time-dependent mean squared distances (σt2-σ02) travelled by interlayer excitons in 0° and 60° heterobilayers as well as by intralayer excitons in WS2 and WSe2 monolayers (1L-WS2, 1L-WSe2). i Exciton density-dependent interlayer exciton transport at room temperature for the 60° heterobilayer. j Temperature-dependent interlayer exciton transport for the 60° heterobilayer. Credit: Ying Jiang, Shula Chen, Weihao Zheng, Biyuan Zheng and Anlian Pan

Specifically, the content of this review includes four sections. The first section discussed the band alignment, ultrafast charge transfer, and the interlayer exciton formation as well as its fundamental properties in TMDs vdW heterostructures. Moiré interlayer excitons, as a newly emerged research hotspot, were also detailed in this section.

The second section discussed the interlayer exciton relaxation processes including the population recombination dynamics, the intervalley scattering process, and the valley-polarized dynamics in TMDs vdW heterostructures. The recombination lifetimes of interlayer excitons in various TMDs vdW heterostructural systems were summarized, and the role of moiré superlattice on interlayer exciton lifetimes was also discussed in this section.

The third section reviewed the transport behaviors of interlayer excitons in TMDs vdW heterostructures, including the interlayer exciton diffusion without external electric field, the (valley-polarized) interlayer exciton transport with external electric field, and the manipulation of the interlayer exciton transport under various potential landscapes such as potential wells or barriers. Moreover, the influences of the moiré potential and the atomic reconstructions on the interlayer exciton transport were also detailed in this section. These related works offer a novel way to control the exciton transport behavior in potential excitonic devices.

After a detailed description of the interlayer exciton formation, relaxation and transport properties in TMDs vdW heterostructures, the final section of this review gave a brief introduction of the potential applications of interlayer excitons in various excitonic devices such as excitonic switches, lasers, and photodetectors. Quantum light based on moiré-trapped interlayer excitons was also discussed here. Nevertheless, the research on excitonic devices based on interlayer excitons in TMDs vdW heterostructures is still in early stages. Improving the performance of the already developed excitonic devices for practical applications and exploring more functional excitonic devices like waveguides and modulators are expected in further works. Moreover, the integration of individual excitonic devices such as light sources, switches, modulators, and detectors on a single chip is very likely and highly desirable in future to realize the on-chip integrated optoelectronics based on two-dimensional vdW heterostructures.

More information: Ying Jiang et al, Interlayer exciton formation, relaxation, and transport in TMD van der Waals heterostructures, *Light: Science & Applications* (2021). DOI: 10.1038/s41377-021-00500-1

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