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Momentum Conservation Driven Ultrafast Charge Transfer Dynamics of Interlayer Excitons in vdW Heterostructure

Heterostructures comprising vdW stacked transition metal dichalcogenide (TMDC) monolayers are a fascinating class of two dimensional (2D) materials with a host of unique properties [1-3]. Presence of interlayer exciton is an intriguing feature of these heterostructures. In an interlayer exciton, the electron and the hole remain spatially separated in the two layers due to the ultrafast charge transfer, which compete with other relaxation channels [4]. Inevitably, the efficiency of devices with 2D heterostructures is critically dependent on the charge transfer dynamics. Investigating MoS₂/WSe₂ heterostructures with monochromated low-loss electron energy loss spectroscopy (EELS) combined with aberration-corrected scanning transmission electron microscopy (STEM), we report for the first time that momentum conservation is a critical factor in the charge transfer dynamics of TMDC heterostructures. From the low-loss electron energy loss (EEL) spectra of the heterostructures with various rotation angles, we demonstrate that- in the aligned case, the charge transfer rate can be about three times faster than the anti-aligned case. Our results provide a deeper insight into the role of the fundamental principle of momentum conservation in the 2D TMDC heterostructure charge transfer dynamics, which could have substantial implications in the rational design of efficient devices based on interlayer excitons. This work also demonstrates the strength of combined STEM-EELS with aberration-correctors and monochromators for investigating optical properties at high spatial resolution discriminating local nanoscale inhomogeneities far beyond the resolution limits of conventional optical techniques.

References

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Figures

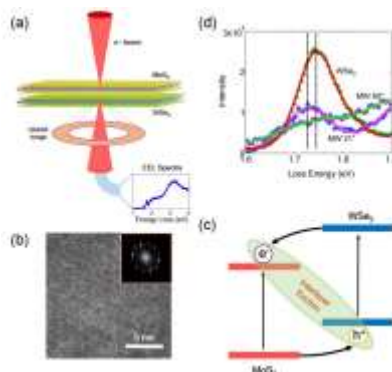


Figure 1: (a) Schematic of the STEM-EELS experimental set-up. (b) HAADF STEM image of the Moiré superlattice due to 29° relative rotation angle between the MoS₂ and WSe₂ monolayers (inset show the FFT pattern). (c) Schematic representation of interlayer exciton formation after charge transfer to the other layer. (d) Comparison of representative low-loss EEL spectra for monolayer WSe₂, aligned MoS₂/WSe₂ heterostructure (60°), and mis-aligned MoS₂/WSe₂ heterostructure (21°).