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Ultrafast Charge Transfer and Recombination Dynamics at the Monolayer-Multilayer WSe₂ Homojunction Revealed by Time-Resolved Photoemission Electron Microscopy

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Abstract

Transition metal dichalcogenides (TMDs) have rapidly emerged as one of the most attractive families of two-dimensional materials due to their numerous outstanding electronic and optoelectronic properties. TMDs have layer-dependent band structures, exhibiting a dramatic transition from a direct bandgap in the monolayer (1L) to an indirect bandgap in the multilayer (NL). TMD homojunctions form when a 1L TMD interfaces with an NL TMD of the same chemical composition. Such 1L/NL TMD homojunctions are predicted to have similar properties as heterojunctions, which comprises two different TMDs. In this work, we employ time-resolved photoemission electron microscopy (TR-PEEM) to study the ultrafast carrier dynamics of a WSe₂ 1L/NL Type-I homojunction with high temporal (50 fs) and spatial (70 nm) resolution. Analysis of the TR-PEEM signal obtained from the individual components of the homojunction reveals timescales for carrier cooling and exciton-exciton annihilation. Beyond the carrier dynamics confined to the individual components of the homojunction, the TR-PEEM image also revealed the accumulation of holes at the interface of the homojunction; the width of the depletion zone is found to be $0.60 \pm 0.17 \mu\text{m}$. Finally, TR-PEEM reveals electron-hole recombination across the homojunction with a time constant of 108 ± 9 ps. These results shed light on the ultrafast carrier dynamics of TMD homojunctions.

Keywords: Transition metal dichalcogenides, Time-resolved photoemission electron microscopy, Type-I homojunction, Ultrafast carrier dynamics.