Ultrafast Exciton Dynamics in 2D Semiconductors Probed by Time-Resolved THz-STM

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Excitons in atomically thin two-dimensional (2D) transition metal dichalcogenide (TMDC) semiconductors exhibit exceptionally large binding energies owing to the strong Coulomb interaction and reduced dielectric screening. As a result, excitons remain stable even at room temperature and dominate the optical response of TMDCs. Because exciton dynamics are strongly influenced by atomic- and nanoscale features such as defects, impurities, wrinkles, and moiré superlattices, it is essential to elucidate how these local structures affect exciton behavior at the microscopic level.

To address this issue, we have employed time-resolved terahertz scanning tunneling microscopy (THz-STM) to probe local exciton dynamics in monolayer TMDCs with nanometer spatial resolution. Free-space coupling of THz pulses into the STM junction generates sub-picosecond transient voltages across the tunneling gap, thereby driving ultrafast tunneling currents. In this study, THz-driven tunneling signals were used to capture exciton dynamics in a MoS_2/WSe_2 heterobilayer prepared by mechanical exfoliation on an insulating SiO_2 substrate. An optical pump pulse train (517 nm, 309 fs) was employed to generate excitons, and their temporal evolution was probed by measuring the delay-time–dependent THz-induced tunneling current (Fig 1(a)).

Fig 1(b) shows a representative time-resolved THz-STM spectrum obtained on the MoS_2/WSe_2 heterobilayer. The observed exponential decay at positive delay times corresponds to the relaxation of interlayer excitons. This decay arises from exciton dissociation induced by the strong THz near-field at the STM tip, followed by carrier tunneling (electrons or holes). The local exciton lifetime can thus be determined from the decay time constant. The presentation will also discuss real-space imaging of exciton dynamics with nanometer resolution.

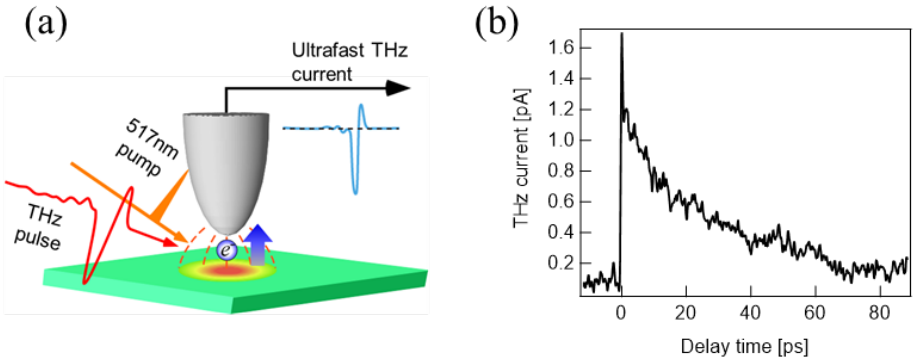

Fig. 1. (a) experimental setup. (b) time-resolved THz-STM spectroscopy on MoS_2/WSe_2 hetero-bilayer