Coherent Exciton Dynamics in Atomically Thin Semiconductors
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The near band-edge optical response of an emerging class of semiconductors, known as the transitional metal dichalcogenides (TMDs), is dominated by tightly-bound excitons and charged excitons (i.e. trions). A fundamental property of these quasiparticles (excitons and trions) is quantum decoherence time, which reflects irreversible quantum dissipation arising from system (excitons and trions) and bath (vacuum and other quasiparticles) interactions and determines the timescale during which excitons can be coherently manipulated. Dephasing time is also equivalent to the intrinsic homogeneous linewidth of exciton resonance. In addition, excitons in TMDs are localized at the corners of the Brillouin zone and provide a convenient way to optical manipulate the valley degree of freedom, which may act as a useful information carrier analogous to electronic charge or spin. Direct measurement of valley coherence time is challenging because it corresponds to a non-radiative coherence between two degenerate states. Using ultrafast multi-dimensional optical spectroscopy, we investigate the intrinsic homogeneous linewidth of excitons, exciton valley coherence as well as coupling between excitons and trions. Our studies reveal coherent electronic dynamics on the order of ~100 fs in monolayer TMDs. We gratefully acknowledge financial support from NSF, AFOSR, and the Welch Foundation.