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Electron dynamics and valley relaxation in 2D semiconductors

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Single layer transition metal dichalcogenides are 2D semiconducting systems with unique electronic band structure. Two-valley energy bands along with strong spin-orbital coupling lead to valley dependent carrier spin polarization, which is the basis for recently proposed valleytronic applications. Since the durations of valley population provide the time window in which valley specific processes take place, it is an essential parameter for developing valleytronic devices. These systems also exhibit unusually strong many body effects, such as strong exciton and trion binding, due to reduced dielectric screening of Coulomb interactions. But there is not much known about the impact of strong many particle correlations on spin and valley polarization dynamics. Here we report direct measurements of ultrafast valley specific relaxation dynamics in single layer MoS₂ and WS₂. We found that excitonic many body interactions significantly contribute to the relaxation process. Biexciton formation reveals hole valley spin relaxation time. Our results also suggest initial fast intervalley electron scattering and electron spin relaxation leads to loss of electron valley polarization, which then facilitates hole valley relaxation via excitonic spin exchange interaction.