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Probing ultrafast valley dynamics in 2D semiconductors via time-resolved Kerr rotation JIANI HUANG, THANG HOANG, Department of Physics and Center for Metamaterials and Integrated Plasmonics, Duke University, TIAN MING, JING KONG, Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, MAIKEN MIKKELSEN, Department of Physics and Department of Electrical and Computer Engineering, Duke University — Monolayer transition metal dichalcogenides (TMDCs) offer a tantalizing platform for controlling spin and valley degrees of freedom, enabling future optoelectronic devices with enhanced and novel functionalities. Here, we experimentally probe the valley dynamics in monolayer MoS₂ and WSe₂ using time-resolved Kerr rotation (TRKR) from $T = 10$ K to 300 K. This pump-probe technique offers sub-picosecond temporal resolution, providing insight into ultrafast valley dynamics inaccessible by polarized and time-resolved photoluminescence spectroscopy. Bi-exponential decay dynamics were observed for both materials at low temperatures. Strong long-range exchange interactions between the K valleys led to a rapid exciton valley depolarization time (< 10 ps), while the valley polarization of the trion and defect states decays within several tens of ps. Moreover, spatial distributions of the TRKR amplitude across monolayer flakes indicated weaker valley polarizations near the edges of MoS₂, which is likely associated with the Mo- or S-zigzag terminations at the boundaries. These temporal and spatial TRKR measurements reveal insight into the complex dynamics of valley excitonic states in monolayer TMDCs.

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