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Long-lived Spin/Valley Dynamics of Resident Electrons and Holes in Monolayer Transition Metal Dichalcogenides

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Interest in atomically-thin transition metal dichalcogenide (TMD) semiconductors such as MoS2 and WSe2 has exploded in the last few years, driven by the new physics of coupled spin/valley degrees of freedom and their potential for new spintronic and 'valleytronic'devices. Although robust spin and valley degrees of freedom have been inferred from polarized photoluminescence (PL) studies of excitons, PL timescales are necessarily constrained by short-lived (1–30 ps) recombination timescales of excitons. Direct probes of spin and valley dynamics of the resident electrons and holes in n-type or p-type doped TMD monolayers, which may persist long after recombination ceases, are still at a relatively early stage.

In this work, we directly measure the coupled spin-valley dynamics of resident electrons and resident holes in n-type and p-type monolayer TMD semiconductors using time-resolved Kerr rotation. Very long relaxation timescales in the nanosecond to microsecond range are observed at low temperatures – orders of magnitude longer than typical exciton lifetimes. In contrast with III-V or II-VI semiconductors, electron spin relaxation in monolayer MoS2 is found to accelerate rapidly in small transverse magnetic fields. This indicates a novel mechanism of electron spin dephasing in monolayer TMDs that is driven by rapidly-fluctuating internal spin-orbit fields that, in turn, are due to fast electron scattering between the K and K'conduction bands [1]. Additionally, a small but surprisingly long-lived oscillatory signal is also observed, indicating the spin coherence of a small population of localized states [2]. More recent studies of gated TMD monolayers also allow observation of very long spin/valley relaxation of resident holes, a consequence of spin-valley locking [3]. These studies provide direct insight into the physics underpinning the spin and valley dynamics of electrons and holes in the monolayer semiconductors.

[1] L. Yang *et al.*, *Nature Physics* **11**, 830 (2015).
[2] L. Yang *et al.*, *Nano Letters* **15**, 8250 (2015).
[3] P. Dey *et al.*, submitted.

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