Long-lived Spin/Valley Dynamics of Resident Electrons and Holes in Monolayer Semiconductors

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Interest in atomically-thin transition metal dichalcogenide (TMD) semiconductors such as MoS_2 and WSe_2 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 & 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 MoS_2 is found to accelerate rapidly in small transverse magnetic fields B_y. 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[3] also allow observation of very long spin/valley relaxation of resident holes, a consequence of spin-valley locking.

These dynamic signals are observed in a variety of TMD samples and are studied as a function of applied field, temperature, and carrier density, providing direct insight into the physics underpinning the spin and valley dynamics of electrons and holes in the monolayer semiconductors.

References

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

Figures

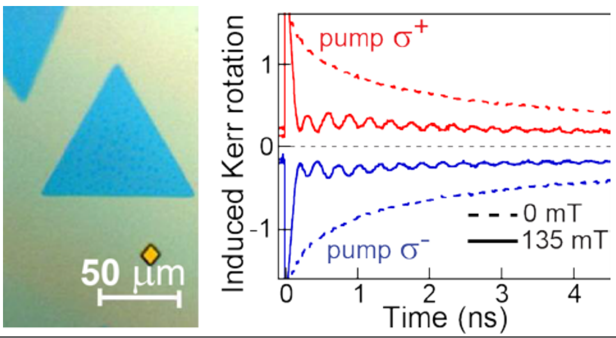

Figure 1: And example of time-resolved Kerr rotation of spin and valley polarization in *n*-type monolayer MoS_2. Long-lived polarization decay is observed, as well as an oscillatory spin coherence signal in applied magnetic fields.