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Gate-Controlled Spin-Valley Locking of Resident Carriers in WSe₂ Monolayers

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Introduction

Besides their obvious promise for 2D optoelectronics, monolayer transition-metal dichalcogenide (TMD) semiconductors such as MoS₂ and WSe₂ have also revitalized interests in exploiting both the spin *and valley pseudospin* of electrons and holes for potential applications in (quantum) information processing. This notion of ``valleytronics" arises due to their crystalline asymmetry and strong spin-orbit coupling, which leads to spin-dependent band structure, spin-valley locking, and valley-specific optical selection rules. These rules mandate that the K or K' valleys in momentum space can be selectively populated and probed using polarized light, in contrast with most conventional III-V, II-VI, and group-IV semiconductors. Therefore, information may be readily encoded not only by whether an electron (or hole) has spin "up" or "down", but *also* by whether it resides in the K or K' valley -- or, indeed, in some quantum-mechanical superposition thereof.

The intrinsic timescales of carrier spin and valley dynamics in monolayer TMDs are therefore of considerable importance. However, most studies to date have focused on photogenerated neutral and charged *excitons*, whose dynamics are inherently limited by their short (3-30~ps) recombination lifetimes. An essential but altogether different question, however, concerns the intrinsic spin/valley lifetimes of the *resident* electrons and holes that exist in *n*-type and *p*-type TMD monolayers. In future valleytronic devices, it is likely the properties of these resident carriers that will determine performance -- analogous to how the scattering timecales and mobility of resident carriers (not excitons) determines the performance of modern-day transistors and interconnects.

Results

Using time-resolved Kerr rotation, we measure the spin/valley dynamics of resident electrons and holes in single charge-tunable monolayers of WSe₂ an archetypal TMD semiconductor. In the n-type regime, we observe long (~130 ns) polarization relaxation of electrons that is sensitive to in-plane magnetic fields B_{ν} , indicating spin relaxation. In marked contrast, extraordinarily long (~2 µs) polarization relaxation of holes is revealed in the p-type regime, that is unaffected by B_{y} , directly confirming long-standing expectations of strong spin-valley locking of holes in the valence band of monolayer TMDs. Supported by continuous-wave Kerr spectroscopy and Hanle measurements. these studies provide a unified picture of carrier polarization dynamics in monolayer TMDs, which can guide design principles for future valleytronic devices.



References

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References

[1] P. Dey et al., Phys. Rev. Lett. 119, 137401 (2017)