

Dynamics of Non-radiative Spin-valley Polarized Carriers in Gate-tunable Suspended WSe₂ Monolayers

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The long lifetime of optically accessible spin-valley polarization in transition-metal dichalcogenides (TMDs) monolayers makes them attractive for a plethora of applications such as optoelectronics, valleytronics, and spintronics. To adopt the benefits of such a degree of freedom for high-mobility two-dimensional carriers in TMDs, we need to suppress the inhomogeneous potential landscape caused by local strain pockets or contaminations originating from the substrate supporting the monolayer. One of the solutions is to suspend the monolayer over a well etched in the substrate, which can improve carrier mobility [1] and even enable the tuning of strain and doping simultaneously [2]. So far, previous studies have investigated suspended TMD monolayers mainly by photoluminescence (PL) spectroscopy [2]. However, much of information derived by this method has been limited to the phenomena involving radiative excitonic complexes that recombine in short time scales. Here, we report the magneto-optical Kerr rotation (KR) spectroscopy of a gate-tunable suspended WSe₂ monolayer, conducted at 7 K. The KR signal gives an additional information about non-radiative carriers and reveals the contributions of spin and valley polarization of long-lived complexes such as dark excitons or resident carriers.

A gold-assisted exfoliation was used to prepare large-area WSe₂ monolayers that were directly transferred on a field-effect transistor device etched with holes or trenches with a depth of 1 μm , as shown in Fig. 1a, b. The vertical gap between the top electrode and the back gate allowed the suspension of the monolayer, which was not in direct contact with any material. The back-gate voltage allowed a simultaneous control of the strain by electrostatic pressure and doping of the monolayer. The optical reflectance in Fig. 1c, measured for different wavelengths at the center of the trench in Fig. 1b, shows symmetrical oscillations with respect to $V_g = 0$ due to the light interference, which confirmed that the monolayer was deflected by the electrostatic force toward the substrate.

The dynamics of the spin-valley polarization were measured by a two-color time-resolved KR spectroscopy, which was developed in our previous experiments [3]. Circularly-polarized pump pulses (1.734 eV) from a Ti:Sapphire laser initialized the spin-valley polarization. Linearly polarized probe pulses (1.65-1.77 eV) from another synchronized laser were focused on the pump beam spot. The KR angle of the reflected probe pulses was measured to investigate the transient dynamics of the spin and valley polarization after the excitation.

Figure 1d shows that the application of $V_g = \pm 30$ V resulted in the survival of the KR signal even after ~ 12 ns (corresponding to ~ 0.15 ns), indicating the accumulation of the polarization due to a lifetime longer than several tens of ns. The asymmetry of the KR signals with respect to V_g inferred the possibility of V_g -dependent formation of different trion complexes [4]. We also confirmed the effects of strain, wavelength, and excitation power on KR. These results indicate that we can use multiple tuning knobs to control the charge state of the exciton complexes and the spin valley polarization lifetime in a single sample.

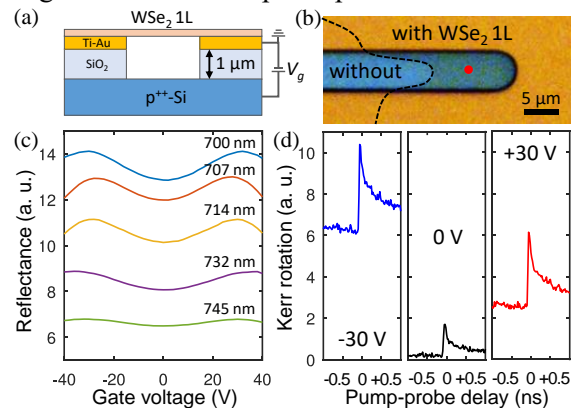


Fig. 1: (a) Schematic of the device. (b) Optical microscope image of a WSe₂ monolayer partially suspended on a trench. (c) Reflectance spectrum at the center of the device (red point in (b)) as a function of the gate voltage. (d) Time-resolved Kerr rotation measured at gate voltages of -30, 0, and 30 V. The probe beam energy was tuned around 1.70 eV (730 nm) near the trion resonance.

References

- [1] T. Jin *et al.*, J. Appl. Phys. **114**, 164509 (2013).
- [2] P. Hernández López *et al.*, Nat. Commun. **13**, 7691 (2022).
- [3] H. Sanada *et al.*, Phys. Rev. Lett. **106**, 216602 (2011).
- [4] J. Li, *et al.*, Phys. Rev. Materials **5**, 044001 (2021).