Detection of Thermodynamic "Valley Noise" in Monolayer Semiconductors: Access to Intrinsic Valley Relaxation Timescales

Goryca, M., Dey, P. and Crooker, S.A. (NHMFL-Los Alamos); Wilson, N.P. and Xu, X. (U. Washington)

Introduction

Novel 2D materials such as graphene and monolayer transition-metal dichalcogenide (TMD) semiconductors has rejuvenated long-standing interests in harnessing *valley degrees of freedom*. Indeed, encoding information in an electron's momentum state forms the conceptual basis of the burgeoning field of "valleytronics". In particular, the family of monolayer TMDs such as MoS_2 and WSe_2 have focused attention on valley physics because they provide a facile means of addressing specific valleys in momentum space using light: Owing to strong spin-orbit coupling and lack of crystalline symmetry, monolayer TMDs possess *valley-specific* optical selection rules, wherein right- and left-circularly polarized light couples selectively to transitions in the distinct K and K' valleys of their hexagonal Brillouin zone. Valley-polarized electrons, holes, and excitons can therefore be readily injected and detected optically, in marked contrast to most conventional semiconductors. The intrinsic time scales of valley scattering and relaxation are of obvious critical importance. Recent optical pump- probe studies have shown that while valley-polarized electron-hole pairs (excitons) scatter very quickly on picosecond timescales, the valley relaxation of *resident* carriers in electron- or hole-doped TMD monolayers can be orders of magnitude longer. However, all such pump-probe measurements are necessarily perturbative in nature. This is because optical pumping injects nonequilibrium electrons and holes which scatter, dissipate energy, and interact, thereby perturbing the resident carriers' valley polarization away from thermal equilibrium via processes not yet well understood. Moreover, optical pumping of both majority *and* minority carriers can create optically-inactive "dark" excitons and trions whose presence, if sufficiently long-lived, could in principle mask detection of carrier valley relaxation, as recently suggested. An alternative means of accessing the truly *intrinsic* valley relaxation of resident carriers in monolayer TMDs, free from perturbative or dark exciton effects, is therefore highly desired.

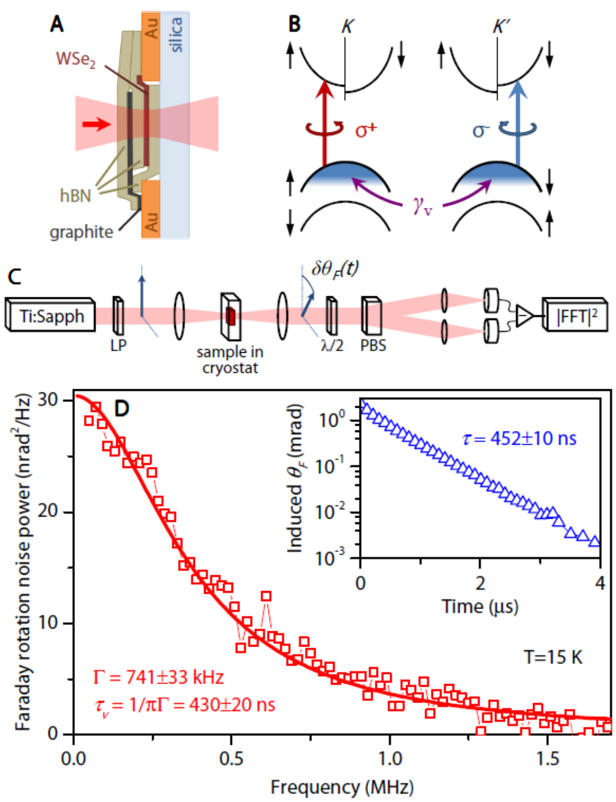

Figure: Sample, experimental setup, and valley noise spectrum of resident holes in monolayer WSe_2. (**A**) A single WSe_2 monolayer is sandwiched between hBN layers and electrically gated. (**B**) Band structure and optical transitions of hole-doped WSe_2. Even in thermal equilibrium, resident holes spontaneously scatter between K and K' valleys, giving a randomly fluctuating valley polarization noise. (**C**) To detect valley noise, a CW probe laser is linearly polarized and focused through the sample. Thermodynamic valley fluctuations impart Faraday rotation fluctuations $\delta\theta_F(t)$ on the probe laser, which are detected using balanced photodiodes. (**D**) The valley noise power spectrum of resident holes in monolayer WSe_2. Its Lorentzian lineshape (solid line) with full-width Γ indicates an exponentially-decaying valley correlation with relaxation timescale $\tau_v = 1/\Gamma$ = 430 ns. Inset: valley relaxation measured separately in a perturbative pump-probe experiment.

Results and Discussion

We introduce and demonstrate an entirely passive, noise-based approach for exploring intrinsic valley dynamics in atomically-thin transition-metal dichalcogenide (TMD) semiconductors. Exploiting the valley-specific optical selection rules in monolayer TMDs, we use optical Faraday rotation to detect, under conditions of strict thermal equilibrium, the stochastic thermodynamic fluctuations of the valley polarization in a Fermi sea of resident carriers. Frequency spectra of this spontaneous "valley noise" reveal narrow Lorentzian lineshapes and therefore long exponentially-decaying intrinsic valley relaxation. Moreover, the valley noise signals are shown to validate both the relaxation times and the spectral dependence of conventional (perturbative) pump-probe measurements, thereby resolving concerns about the role of dark excitons and trions in studies of long-lived valley relaxation. These results provide a viable route toward quantitative measurements of intrinsic valley dynamics, free from any external perturbation, pumping, or excitation [1].

References

[1] Goryca, M. *et al.*, *Science Advances, accepted and in press* (2018).