Detection of thermodynamic “valley noise” in monolayer semiconductors: access to intrinsic valley relaxation timescales

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The new class of atomically-thin transition-metal dichalcogenide (TMD) semiconductors such as monolayer MoS_2 and WSe_2 has focused broad attention on the concept of “valleytronics”, founded on the idea of encoding information in an electron’s valley degree of freedom. The key parameter for any valleytronic technology is the intrinsic timescale of an electron’s inter-valley relaxation, and recent optical pump-probe studies have shown long (exceeding microseconds at low temperatures) valley relaxation of resident carriers in monolayer TMDs [1,2]. However, a significant drawback of all such experiments is that they are by design perturbative: they require optical pumping to intentionally drive the electrons out of equilibrium. Such pumping inevitably introduces so-called “dark” exciton states, whose presence can mask carriers' intrinsic valley relaxation.

Here we present a completely alternative approach, based on the idea of passively “listening” to the random spontaneous scattering of carriers between K and K' valleys (Fig. 1(b)) in strict thermal equilibrium. We demonstrate that the stochastic *valley noise* is measurable by optical means (Fig. 1(a)) and encodes the true intrinsic timescales of valley relaxation, free from any pumping or excitation [3].

Using this new fluctuation-based methodology we measure very long valley relaxation dynamics of both electrons and holes in a single electrostatically-gated monolayer of WSe_2. Valley noise frequency spectra (Fig. 1(c)) reveal long intrinsic valley relaxation with a single sub-microsecond time scale. Moreover, they validate both the relaxation times and the spectral dependence of conventional pump-probe measurements, thereby resolving concerns about the role of dark excitons and trions in studies of long-lived valley relaxation.

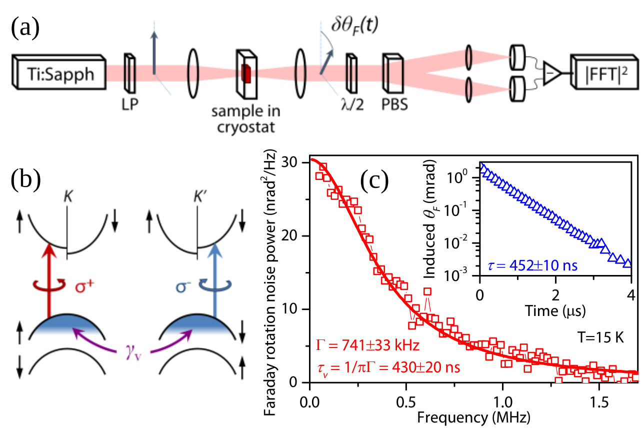

Fig. 1. (a) Experiment: stochastic valley fluctuations impart Faraday rotation $\delta\theta(t)$ on the linearly polarized probe laser detected with balanced photodiodes. (b) Band structure and $\sigma^{\pm}$ optical transitions of hole-doped monolayer WSe_2. (c) The valley noise spectrum of resident holes in monolayer WSe_2. Lorentzian lineshape indicates an exponentially-decaying valley correlation with relaxation time of 430 ns. Inset: valley relaxation measured separately using pump-probe technique.

[1] J. Kim *et al.*, Science Advances **3**, e1700518 (2017).
[2] P. Dey *et al.*, Phys. Rev. Lett. **119**, 137401 (2017).
[3] M. Goryca *et al.*, arXiv:1808.01319 (2018).

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Supplementary Pages

Figure 2 shows the temperature dependence of the valley relaxation time τ_v determined by noise experiments, along with a comparison to τ_v measured by conventional time-resolved Faraday rotation (TRFR) methods. Fig. 2A presents normalized valley noise spectra obtained at T=15, 20, 25, and 29 K. All are well described by Lorentzian line shapes, with full-width $\Gamma(= 1/\pi\tau_v)$ increasing significantly with temperature, indicating a reduction of τ_v. Separately, conventional TRFR measurements of valley dynamics (Fig. 2B) also show faster decays at higher temperature. Figure 2C compares τ_v as measured by the two methods, showing very close agreement and establishing that the long-lived TRFR signals arise from intrinsic valley polarization of resident carriers and are not dark exciton effects.

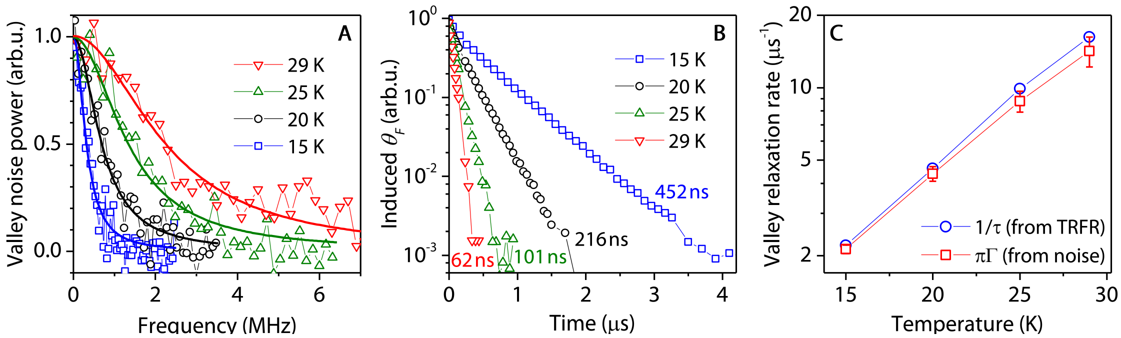

Fig. 2. (A) Valley noise spectra of hole-doped WSe_2 monolayer at different temperatures (solid lines are Lorentzian fits). (B) Hole valley relaxation measured separately by TRFR. (C) Temperature dependence of valley relaxation rate extracted from the valley noise ($1/\tau_v = \pi\Gamma$; red) and relaxation rate measured by TRFR (blue), showing very close agreement.

In Fig. 3 we investigate the dependence of the hole valley noise on the probe photon energy E, and show that the noise is largest when the probe laser is tuned near to (but not on) the X^+ charged exciton transition. Fig. 3A shows the transmission spectrum of the lightly hole-doped WSe_2 monolayer. The X^+ absorption appears as a very narrow resonance below the neutral exciton resonance (X^0). Fig. 3B shows the result of a conventional CW pump-probe Faraday rotation (FR) experiment, in which a weak circularly-polarized CW pump laser generates a steady-state nonequilibrium valley polarization in the hole Fermi sea, while the induced FR spectrum $\theta_F(E)$ is concurrently measured by a tunable CW probe laser. Antisymmetric resonance line in $\theta_F(E)$ spectrum is centered on the X^+ absorption resonance, in agreement with the expectation that charged exciton transitions (and not neutral excitons) depend explicitly on the presence of resident carriers. Therefore the polarization of the hole Fermi sea due to thermodynamic valley fluctuations should generate a similar spectral response centered on the X^+ transition, and the spectrum of the valley noise *power* should resemble the *square* of $\theta_F(E)$. Indeed we find this to be the case. The blue points in Fig. 3C show the total valley noise power versus the probe laser's energy. For comparison, red points show the square of the induced $\theta_F(E)$ shown in Fig. 3B, revealing a nearly identical spectral dependence. Thus, valley fluctuations can be detected by FR using light tuned in energy well below the lowest absorption resonance in the WSe_2 monolayer, which assures the non-perturbative nature of this noise-based approach.

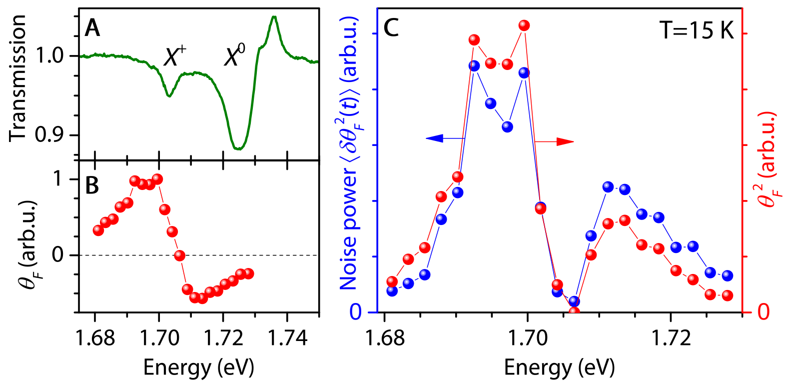

Fig. 3. (A) Optical transmission spectrum of the lightly hole-doped WSe_2 monolayer, showing neutral (X^0) and positively-charged exciton (X^+) resonances. (B) Energy-dependent FR spectrum $\theta_F(E)$ induced by an intentional (optically-pumped) valley polarization of the resident holes. $\theta_F(E)$ is antisymmetric and centered on X^+, as expected. (C) Spectral dependence of the total valley noise power. The red points show the square of $\theta_F(E)$ from panel (B), showing close agreement.