Thermodynamic "valley noise" in monolayer semiconductors

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From the point of view of encoding information in an electron's valley degree of freedom, a key parameter of atomically thin transition-metal dichalcogenide (TMD) semiconductors is the intrinsic timescale of an electron's inter-valley relaxation. Such relaxation has been shown to be long (microsecond timescale) by recent optical pump-probe studies of resident carriers in monolayer TMDs [1,2]. However, a significant drawback of all such pump-probe experiments is that they are by design perturbative: the optical pumping that drives the carrier polarization away from equilibrium also inevitably introduces "dark" excitons, whose presence may mask the intrinsic valley relaxation of resident carriers.

Here we present a completely alternative approach for measuring valley dynamics, based on the idea of passively "listening" to the random spontaneous scattering of carriers between K and K' valleys that occurs even in strict thermal equilibrium. We demonstrate that the stochastic valley noise is measurable by optical means and, in accord with the fluctuation-dissipation theorem, encodes the true intrinsic timescales of valley relaxation, free from any pumping, excitation, or other perturbative effects [3]. Using this new fluctuation-based methodology we measure very long valley relaxation dynamics of both electrons and holes in a single electrostatically-gated WSe2 monolayers. Noise spectra reveal long intrinsic valley relaxation with a single sub-microsecond time scale. Moreover, they validate both the relaxation times and the wavelength dependence observed in conventional pump-probe measurements, thereby resolving concerns about the role of dark excitons and trions in studies of long-lived valley relaxation.

[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., Science Advances **5**, eaau4899 (2019).