Exciton Linewidth Effects on Valley Relaxation in 2D TMDCs

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Monolayer transition metal dichalcogenides (TMDCs) have shown exceptional promise as valleytronic materials. Their strongly coupled spin-valley physics allow selective valley population using circularly polarized light. Understanding the physical mechanisms behind valley relaxation (loss of binary valley information) in these materials is an important, ongoing research topic. We have accumulated experimental evidence for a comprehensive theory [1] of two-dimensional screened, electron-hole exchange-interaction-mediated valley relaxation processes in TMDCs [2]. Our results can also explain temperature-dependent and excitation-density-dependent valley relaxation phenomena in a variety of previous studies. According to our theory, valley relaxation times should show strong dependence on exciton homogeneous linewidth. Through changing excitation density with a pulsed laser, we recently showed an inverse relationship between steady-state valley polarization and exciton homogeneous linewidth at low temperature, consistent with our theory. We also showcase recent efforts to enhance valley physics by encapsulating TMDCs in thin layers of hexagonal boron nitride (hBN). This is a common practice for enhancing the optical and electronic properties of graphene, but has been only recently been utilized for TMDCs. We demonstrate the effects of hBN encapsulation on improving low-temperature excitonic spectra of monolayer MoS$_2$, and report on how this affects valley relaxation physics within the context of our current theoretical understanding. Our results help gain insight into the fundamental valley physics of monolayer TMDCs, a class of exciting valleytronic materials.

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1. BACKGROUND

Conventional electronics (e.g. MOSFET) vs. Valleytronics in 2D TMDCs

- Previous work [4-5]: long-range electron-hole exchange interactions induce fast valley relaxation.
- Screening effects from carrier dopants critically affect this process [5].
- Model exciton valley scattering as Maille-Silva-Sham depolarization of valley pseudospin [4-5]:

$$\tau_v \propto \frac{k_T^2}{\Gamma_h}$$

1. Why does this happen?

2. METHODS

- Polarization-resolved photoluminescence spectroscopy

3. RESULTS: Monolayer WSe$_2$

- Measure steady-state valley polarization, related to valley lifetime by:

$$\rho_v = \frac{\rho_0}{1 + \frac{\lambda}{k_T^2 \tau_v}}$$

4. RESULTS: hBN-MoS$_2$ sandwich

- hBN encapsulation common for graphene, but only recently used for TMDCs. [6]
- Observed very narrow PL spectrum with this technique.

5. CONCLUSION

- Exciton linewidth inversely related to valley polarization in 2D TMDCs.
- Agrees with broader theory of TMDC valley relaxation by carrier-screened, long-range electron-hole exchange interactions.
- Results suggest that narrowing exciton linewidth can help extend valley lifetimes and increase valley polarization.

6. REFERENCES