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)	Apply V_g — Si, J — ON Remove V_g — Si, $J = 0$ — OFF	Slow switching (limited by voltage switching)	Stable over long periods of time
Valleytronics in 2D TMDCs [1-3]	Change light polarization — TMDC — Excite K exciton / Excite K' exciton	Fast switching (limited by light oscillation)	Disappears within 10^{-12} seconds ("valley relaxation")

Why does this happen?

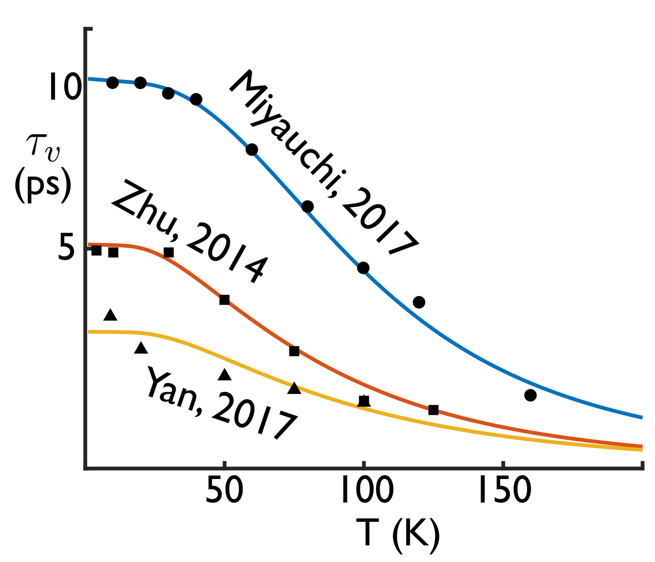
- Previous work [4-5]: long-range electron-hole exchange interactions induce fast valley relaxation.
- Screening effects from carrier dopants critically affect this process [5].
- Model exciton valley scattering as Maille-Silva-Sham depolarization of valley pseudospin [4-5]:

$$\tau_v \propto k_{TF}^2 \Big/ \Gamma_h \quad (1)$$

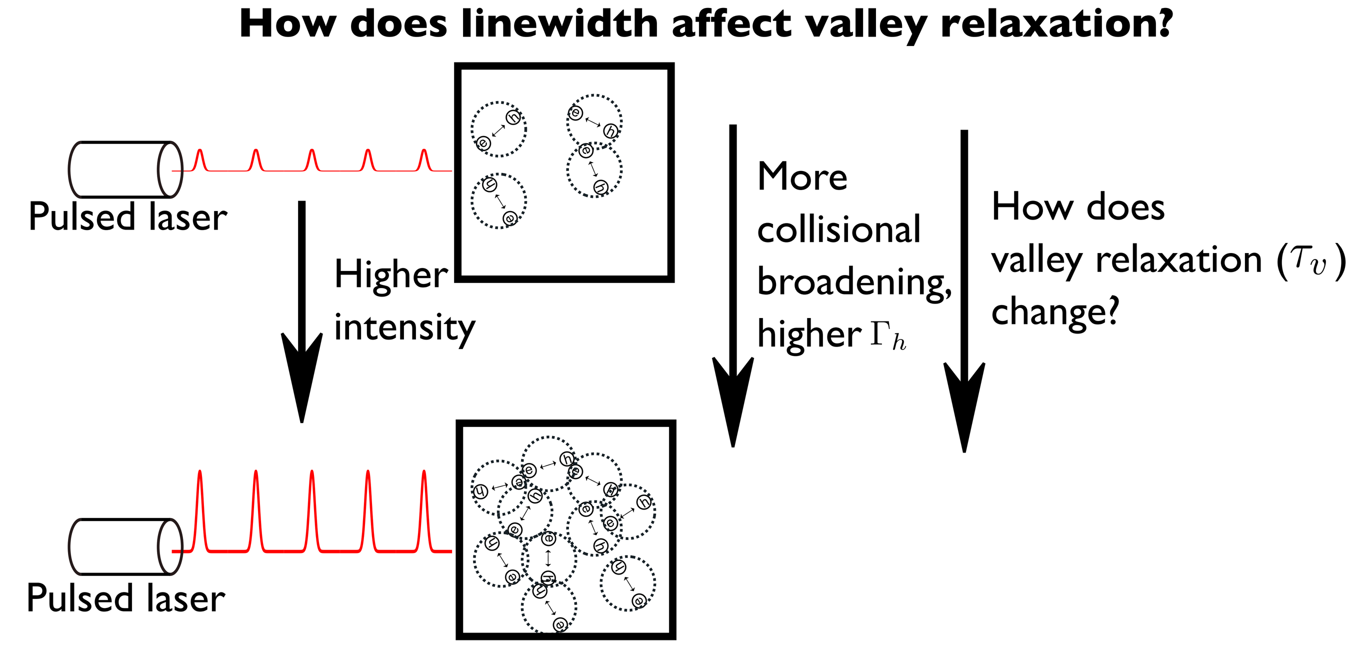
- τ_v is valley relaxation time.
- k_{TF} is 2D Thomas-Fermi wave vector (screening effect).
- Γ_h is homogeneous line width.

Previous work: Screening effect — Current work: Linewidth effect

Using screening to predict valley lifetime [5]

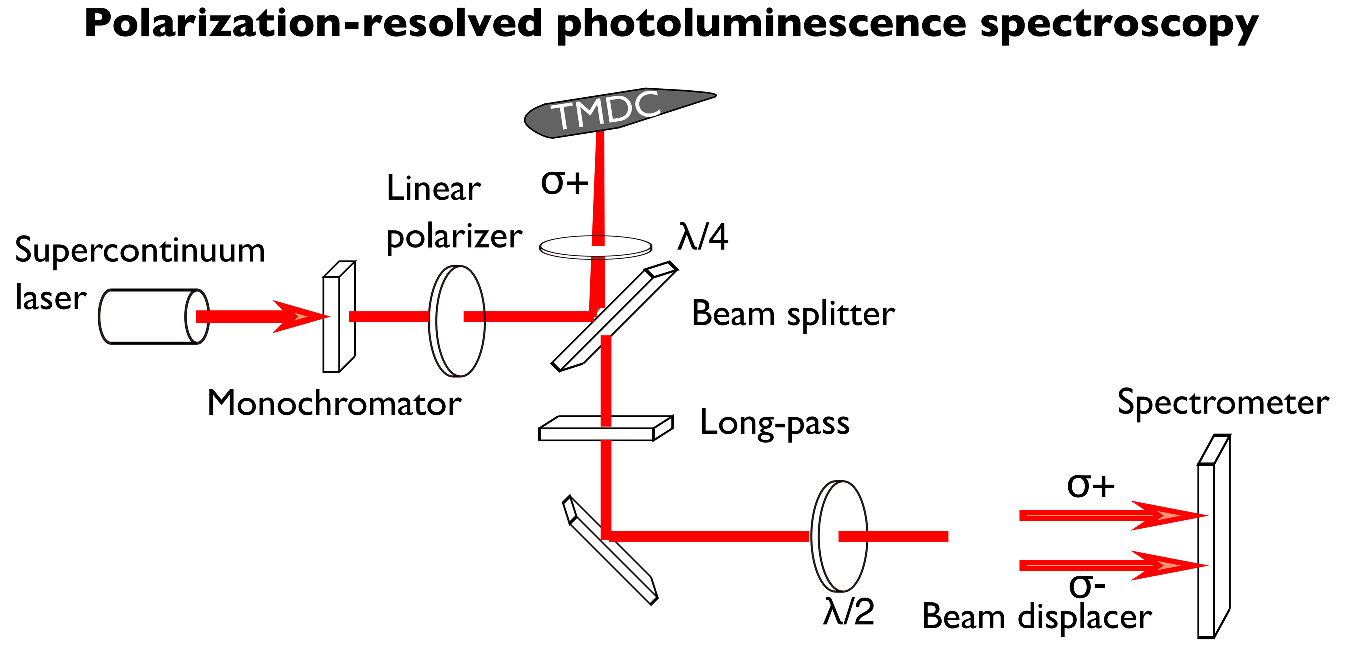


How does linewidth affect valley relaxation?

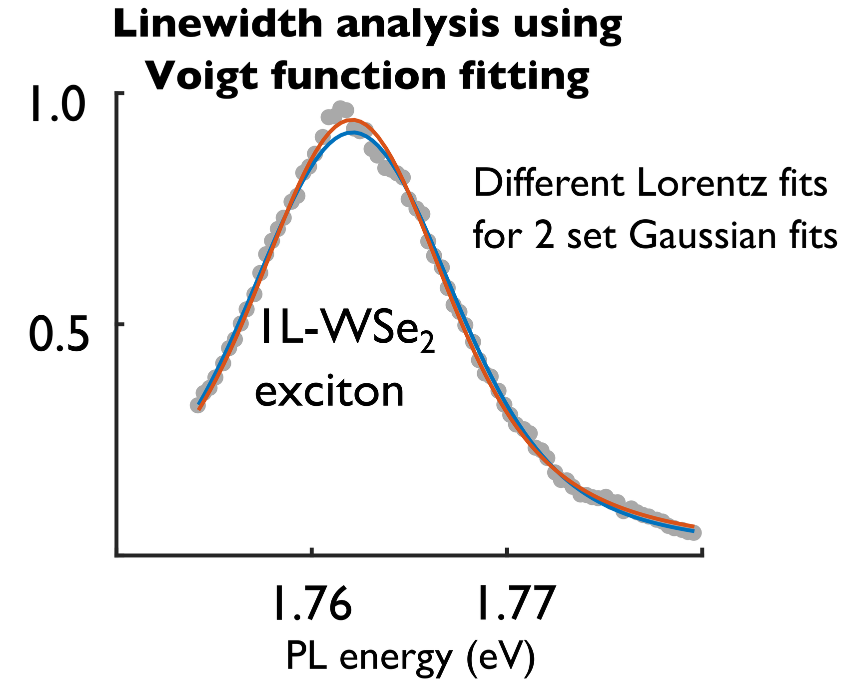


2. METHODS

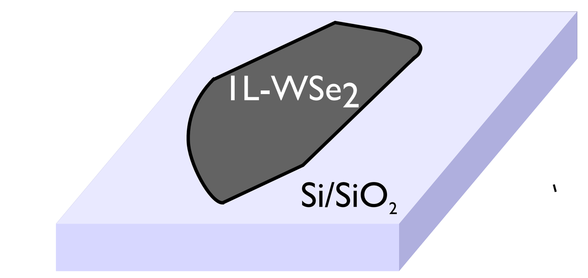
Polarization-resolved photoluminescence spectroscopy



Linewidth analysis using Voigt function fitting



3. RESULTS: Monolayer WSe2

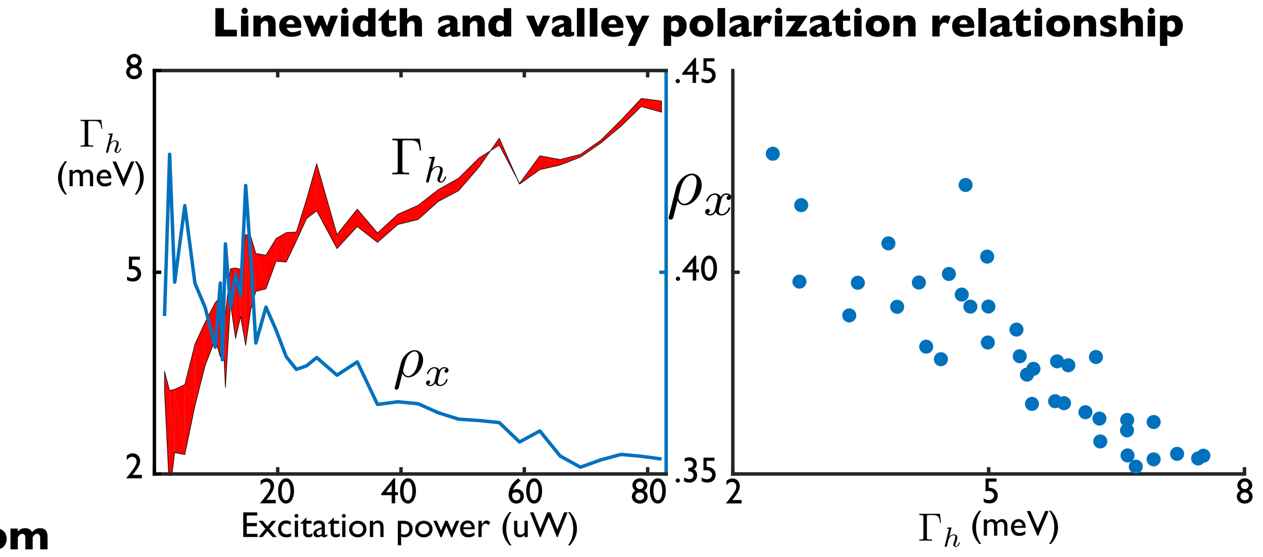


- Measure steady-state valley polarization, related to valley lifetime by:
- ρ_x is valley polarization; ρ_0, A, and J are constants; $\langle\tau_x\rangle$ is exciton lifetime.
- Expect inverse relationship between valley polarization and linewidth.

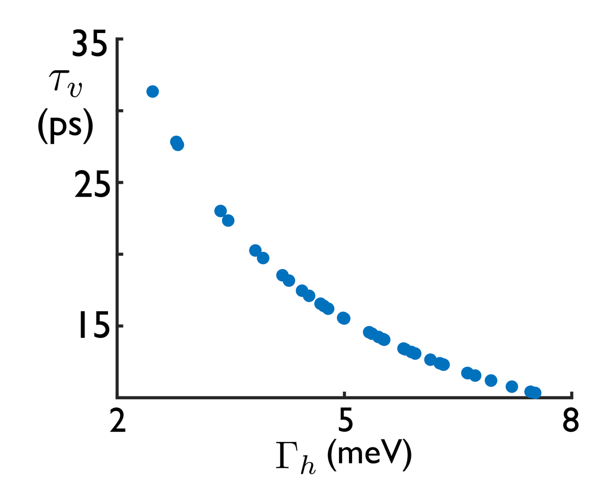
$$\rho_x = \frac{\rho_0}{1+\frac{\langle\tau_x\rangle}{\tau_v}} = \frac{\rho_0}{1+\left(\frac{\langle\tau_x\rangle A J^2}{\hbar k_{TF}^2}\right)\cdot\Gamma_h} \quad (2)$$

- With increasing excitation power, linewidth broadens and valley polarization decreases.
- Plotting linewidth vs. valley polarization reveals an inverse relationship, as predicted by (2).

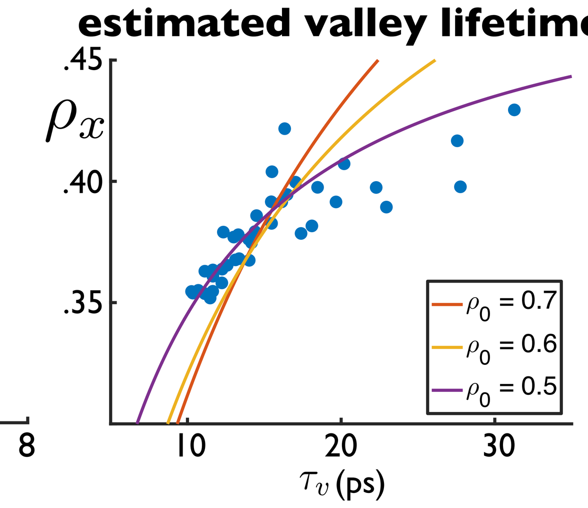
Linewidth and valley polarization relationship



Estimated low-temp. valley lifetime

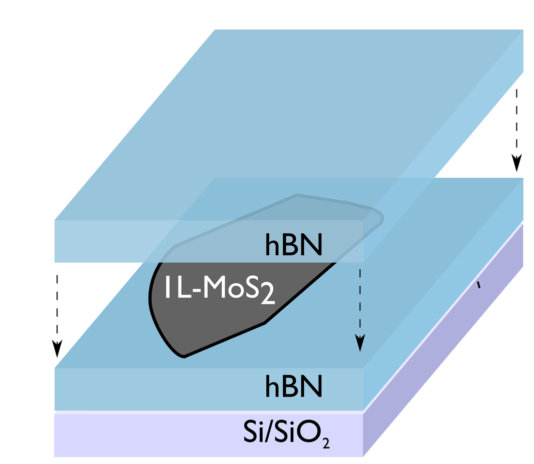


Reasonable fits for valley polarization from estimated valley lifetime



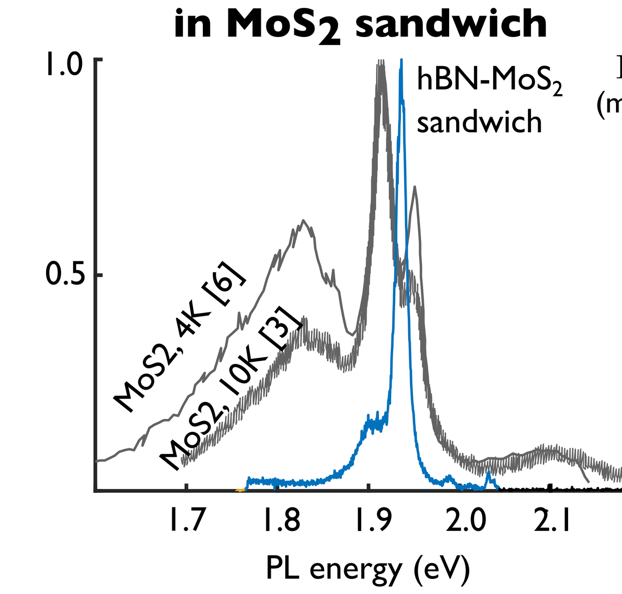
- Low-temperature linewidth and valley lifetime have a simple relationship [5]: $\tau_v(T\approx 0) \approx 118\hbar/\Gamma_h(T\approx 0)$
- Plugging into (2) above, we do a simple fit for $\langle\tau_x\rangle$.
- We obtain reasonable fits ($\langle\tau_x\rangle$ from 4-12 ps).
- ρ_0 is a known constant (approx 0.7) from previous experiments.

4. RESULTS: hBN-MoS2 sandwich

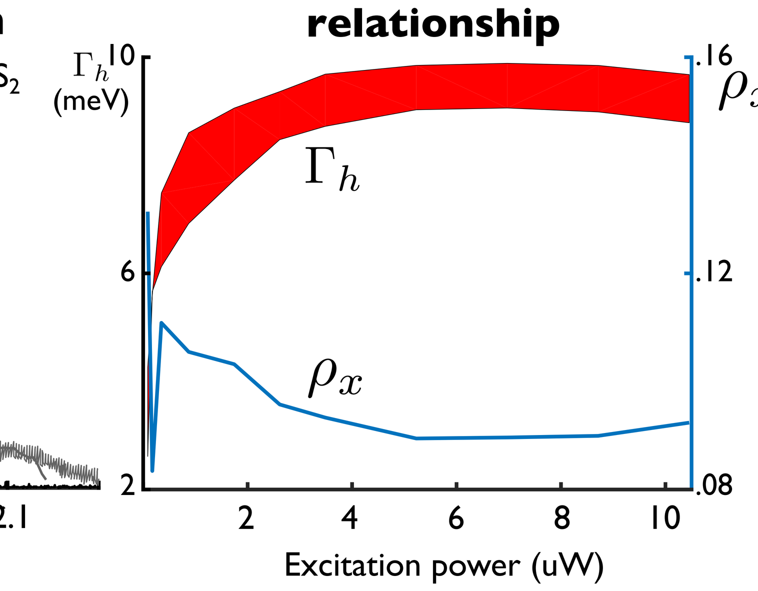


- hBN encapsulation common for graphene, but only recently used for TMDCs. [6]
- Observed very narrow PL spectrum with this technique.

PL spectral narrowing in MoS2 sandwich



Linewidth and valley polarization relationship



- Excitation-power response of linewidth matches response of valley polarization.
- Results consistent with WSe$_2$.

5. CONCLUSION

- Exciton linewidth inversely related to valley polarization in 2DTMDCs.
- 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

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