Excitons, electrons, and holes in monolayer semiconductors: Insights from spectroscopy in (really) high magnetic fields

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Historically, magnetic fields have played an essential role in revealing the properties of semiconductors, and the many-body physics that can emerge when they are doped with mobile carriers. However, for atomically-thin 'transition metal dichalcogenide' (TMD) semiconductors such as MoS₂ and WSe₂, the relevant field scale is substantial (of order 100 tesla!) due to heavy carrier masses, huge exciton binding energies, and typically large Fermi energies. Fortunately, modern pulsed magnets can achieve this scale. This talk will discuss a few recent optical studies that probe the physics of -- and many-body correlations between -- excitons, electrons, and holes in TMD monolayers. These experiments used dual-gated TMD monolayers, assembled via van der Waals stacking directly atop single-mode optical fibers to enable polarized absorption spectroscopy at low temperatures in 60-100T fields.

*In <u>charge-neutral</u> monolayers, spectroscopy up to ~90T reveals the diamagnetic shifts of the neutral exciton's *Is* ground state *and* its excited *2s*, *3s*,... *ns* Rydberg states, revealing exciton masses, radii, binding energies, dielectric properties, and free-particle bandgaps – essential ingredients for the rational design of optoelectronic van der Waals structures. [1]



*In <u>hole-doped</u> monolayer WSe₂, high-field spectroscopy of both neutral and charged exciton transitions revealed the (often-hypothesized) spontaneous valley polarization of mobile holes, due to exchange interactions, occurring at ~40T. [2]

*In <u>electron-doped</u> WSe₂ monolayers, the ordering of the conduction bands in the *K* and *K'* valleys allows studies of not only neutral excitons (*X0*) and charged excitons (*X*- trions) at low carrier density, but also many-body states that can emerge at higher doping. We investigate the so-called *X*-' resonance that emerges at high electron density, known since 2013 but never understood. The data suggest that *X*-' is, in fact, a six-particle "hexciton" state that arises when the photoexcited electron-hole pair couples simultaneously to <u>two</u> Fermi seas having quantum-mechanically distinguishable spin/valley quantum numbers. This state also appears in WS₂ and may appear in MoS₂, and appears in MoS₂ at the B-exciton resonance [3,4]

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[1] Revealing exciton masses and dielectric properties of monolayer semiconductors with high magnetic fields, *Nat. Commun.* **10**, 4172 (2019).

[2] Spontaneous Valley Polarization of Interacting Carriers in a Monolayer Semiconductor, PRL 125, 147602 (2020).

[3] Many-body exciton and intervalley correlations in heavily electron-doped WSe₂, *Nano Letters* 22, 426 (2022).

[4] Six-body and Eight-body Exciton States in Monolayer WSe₂, PRL 129, 076801 (2022).

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