

Rydberg Excitons & Dielectric Environment Effects in Monolayer Semiconductors: Insight from High Magnetic Fields

Andreas Stier¹, Nathan Wilson², Junichiro Kono³, Xiaodong Xu², & Scott Crooker¹

¹ National High Magnetic Field Lab, Los Alamos National Lab, Los Alamos, NM 87545

² Department of Physics, University of Washington, Seattle, WA 98195

³ Department of Physics, Rice University, Houston, TX

Excitons in atomically-thin semiconductors necessarily lie close to a surface, and therefore their properties are expected to be strongly influenced by the surrounding dielectric environment. However, systematic studies exploring this role are challenging, in part because the most readily accessible exciton parameter—the exciton’s optical transition energy—is largely *unaffected* by the surrounding medium. In this work we show that the significant role of the dielectric environment on 2D materials can be directly revealed through its systematic influence on the *size* of the exciton, which can be measured via the diamagnetic shift of the exciton transition in high magnetic fields [1].

Using exfoliated WSe₂ monolayers affixed to single-mode optical fibers, we tune the surrounding dielectric environment by encapsulating the flakes with different materials [2] and perform polarized low-temperature magneto-absorption studies to 65 T. The systematic increase of the exciton’s size with dielectric screening, and concurrent reduction in binding energy (also inferred from these measurements), is quantitatively compared with leading theoretical models based on the Keldysh potential for 2D materials. These results demonstrate how exciton properties and the free-particle bandgap can be tuned in 2D van der Waals heterostructures, via the surrounding dielectric environment. We also present recent 65T measurements of high-quality hBN/WSe₂/hBN structures that permit an unambiguous identification and quantification of excited *1s*, *2s*, *3s*, and *4s* Rydberg states of neutral excitons [3], which allows a direct measurement of exciton mass in 2D materials.

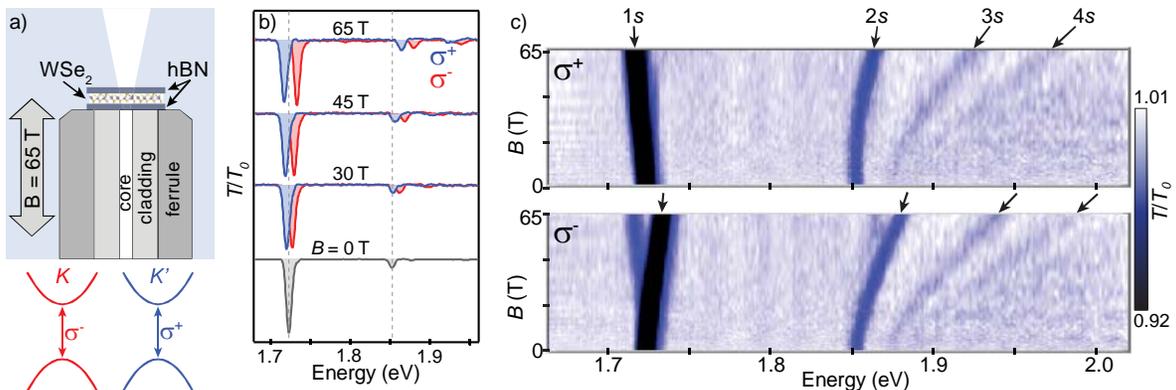


Figure 1: Polarized optical spectroscopy of hBN-encapsulated monolayer WSe₂ to 65T, showing the distinct energy shifts of the *1s*, *2s*, *3s*, and *4s* Rydberg excitons, from which mass can be measured.

[1] A. V. Stier *et al.*, *Nano Letters* **16**, 7054 (2016).

[2] A. V. Stier *et al.*, *Nature Communications* **7**:10643 (2016).

[3] A. V. Stier *et al.*, *submitted* ; arXiv:1709.00123

⁺ Author for correspondence: crooker@lanl.gov