Measuring the Exciton's Mass in Monolayer WS_2 via 65T Magneto-absorption

Goryca, M., Li, J., Stier, A. and Crooker, S.A. (NHMFL-Los Alamos); Courtade, E., Shree, S., Robert, C., Urbaszek, B. and Marie, X. (INSA-Toulouse)

Introduction

In comparison with the other members of the monolayer TMD semiconductor family, WS_2 exhibits exceptionally strong exciton absorption. Signatures of excited Rydberg excitons in monolayer WS_2 were first revealed by optical reflection spectroscopy at zero magnetic field by Chernikov. Subsequently, the small diamagnetic shift and valley Zeeman splitting of the ground state (1*s*) exciton were measured via polarized magneto-reflection to 65 T by Stier. More recently, charged excitons and also the 2*s* state of the neutral exciton in monolayer WS_2 were studied to ~30 T using photoluminescence. Crucially, however, in all these field-dependent studies the sample quality was *not* sufficient to observe highly-excited Rydberg states, and therefore the exciton's mass – a fundamental material parameter – could not be experimentally determined. Rather, the exciton's reduced mass m_r (≈ 0.15−0.16 m_0) was assumed from density-functional theories which, together with the measured diamagnetic shifts, allowed estimates of the exciton's size and binding energy.

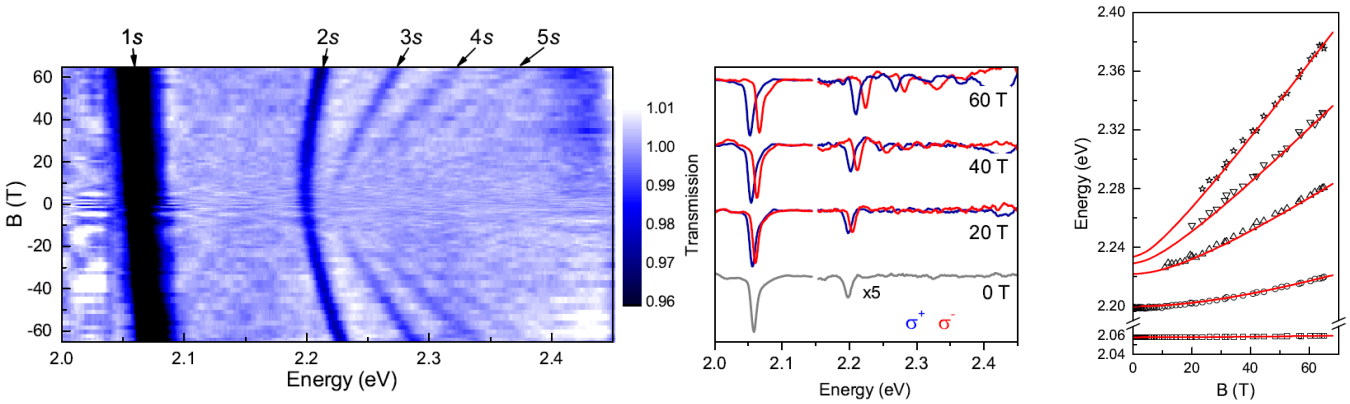

Figure: Normalized transmission spectra of monolayer WS2 versus magnetic field to +-65T. Blue/red curves correspond to right- and left-circularly polarized spectra. Excellent sample quality allows observation of the 2*s*, 3*s*, 4*s*, and 5*s* excited Rydberg states of the neutral "A" exciton. (d) Average energies of the 1*s*-5*s* excitons reveals distinct diamagnetic shifts. Solid lines show calculated energies using the Rytova-Keldysh model, using m_r = 0:175 m_0.

Results and Discussion

Here, using high-quality WS_2 monolayers encapsulated in hBN, we observe the field-induced shifts of the 1*s*, 2*s*, 3*s*, 4*s*, and 5*s* states of the neutral "A" exciton to 65 T. Most importantly, these data provide the first experimental measurement of m_r in monolayer WS_2. Moreover these results also allow a detailed quantitative comparison with the Rytova-Keldysh model that describes the non-hydrogenic electrostatic potential $V(r)$ between an electron and hole in a 2D material, from which various other material parameters can also be determined (dielectric properties, exciton size, etc.).

To prepare monolayer samples for high-field magnetotransmission studies, exfoliated TMD monolayers were sandwiched between slabs of exfoliated hexagonal boron nitride (hBN) using a dry-stamp technique. The thicknesses of the hBN slabs were selected to maximize the absorption of light by the exciton resonances in the TMD monolayer. Each van der Waals structure was assembled directly over the core of a single-mode optical fiber. Crucially, this ensures a rigid drift- and vibration-free alignment of the optical path through the TMD monolayer during the experiment. The fiber/sample assembly was then mounted in 4 K exchange gas in the tail of a liquid helium cryostat located in the bore of a 65 T pulsed magnet. Broadband transmission spectroscopy was performed by coupling unpolarized white light from a Xe lamp into the single-mode fiber. After passing through the sample and a thin-film circular polarizer, the transmitted light was retro-reflected back through a separate multimode collection fiber, and was detected using a spectrometer and high-speed CCD camera. Spectra were continuously acquired every 1 ms throughout the magnet pulse. Access to σ+- circularly polarized transitions (corresponding to transitions in the *K* and *K′* valley of the TMD monolayer) was achieved by reversing the direction of the magnetic field *B*.

References

[1] Goryca, M. *et al.*, submitted (2018).