

Revealing exciton masses and dielectric properties of monolayer semiconductors with high magnetic fields

M. Goryca,¹⁺ J. Li,¹ A.V. Stier,¹ T. Taniguchi,² K. Watanabe,² E. Courtade,³ S. Shree,³ C. Robert,³ B. Urbaszek,³ X. Marie,³ and S. A. Crooker¹

¹ National High Magnetic Field Laboratory, Los Alamos, NM 87545, USA

² National Institute for Materials Science, Tsukuba, Ibaraki 305-0044, Japan

³ Universite de Toulouse, INSA-CNRS-UPS, LPCNO, 31077 Toulouse, France

In semiconductor physics, many of the essential material parameters relevant for optoelectronics can be experimentally revealed via optical spectroscopy in sufficiently large magnetic fields. For the new class of monolayer transition-metal dichalcogenide (TMD) semiconductors, this field scale can be substantial – many tens of teslas or more – due to the relatively heavy carrier masses and the very large electron-hole (exciton) binding energies. For that reason many fundamental parameters of TMDs were – up to now – still assumed from density functional theory calculations [1-3] and have not been experimentally measured.

Here we report circularly-polarized absorption spectroscopy of the monolayer semiconductors MoS₂, MoSe₂, MoTe₂, WS₂, and WSe₂ in very high magnetic fields up to 91 T. By encapsulating exfoliated monolayers in hexagonal boron nitride (hBN), we achieve very high optical quality structures that allow to follow the diamagnetic shifts and valley Zeeman splittings of not only the 1s ground state of the neutral exciton but also its excited 2s, 3s, ..., ns Rydberg states (see Fig. 1). The energies and diamagnetic shifts provide a direct determination of the effective (reduced) exciton masses and the dielectric properties of these monolayer semiconductors [4-5]. Unexpectedly, the measured exciton masses are significantly heavier than predicted for Mo-based monolayers. Moreover, we also measure other important material properties, including exciton binding energies, exciton radii, and free-particle bandgaps. These results provide essential and quantitative parameters for the rational design of optoelectronic van der Waals heterostructures incorporating 2D semiconductor monolayers.

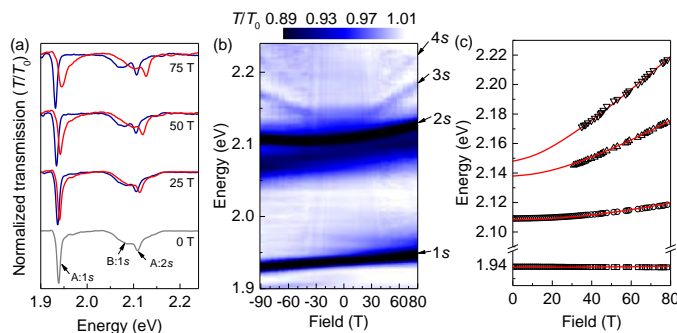


Figure 1. (a) Transmission spectra through monolayer MoS₂ encapsulated in hBN, in both circular polarizations. (b) Intensity map showing all spectra from -91 T to +80 T. The excited Rydberg states of the neutral exciton are visible. (c) The energies of each *ns* exciton state. Solid curves show model calculations.

- [1] A. Kormányos *et al.*, 2D Materials **2**, 049501 (2015).
- [2] T. C. Berkelbach *et al.*, Phys. Rev. B **88**, 045318 (2013).
- [3] D. Wickramaratne *et al.*, J. Chem. Phys. **140**, 124710 (2014).
- [4] A. V. Stier *et al.*, Phys. Rev. Lett. **120**, 057405 (2018).
- [5] M. Goryca *et al.*, Nat. Commun. **10**, 4172 (2019).

⁺ Author for correspondence: mgoryca@lanl.gov