BNL-105467-2014-CP

Excitons in quasi-two-dimensional van der Waals materials

Alexey Chernikov

Alexey Chernikov[1], Timothy C. Berkelbach[2], Heather M. Hill[1], Albert Rigosi[1],Yilei Li[1], Ozgur, B. Aslan[1], David R. Reichman[2], Mark S. Hybertsen[3], Tony F. Heinz[1]

[1] Department of Physics, Columbia University, 538 West 120th Street, New York, NY 10027, USA
[2] Department of Chemistry, Columbia University, 3000 Broadway, New York, NY 10027, USA
[3] Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY 11973-USA

Presented at the Nonlinear Optics and Excitation Kinetics in Semiconductors Conference
Bremen, Germany

September 22-25, 2014

Center for Functional Nanomaterials

Brookhaven National Laboratory

U.S. Department of Energy
Office of Science

Notice: This manuscript has been authored by employees of Brookhaven Science Associates, LLC under Contract No. DE-AC02-98CH10886 with the U.S. Department of Energy. The publisher by accepting the manuscript for publication acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

This preprint is intended for publication in a journal or proceedings. Since changes may be made before publication, it may not be cited or reproduced without the author's permission.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

BNL-105467-2014-CP

Excitons in quasi-two-dimensional van der Waals materials

Alexey Chernikov[1], Timothy C. Berkelbach[2], Heather M. Hill[1], Albert Rigosi[1], Yilei Li[1], Özgur B. Aslan[1], David R. Reichman[2], Mark S. Hybertsen[3], Tony F. Heinz[1]

[1] *Department of Physics, Columbia University, 538 West 120th Street, New York, NY 10027, USA*
[2] *Department of Chemistry, Columbia University, 3000 Broadway, New York, NY 10027, USA*
[3] *Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY 11973-5000, USA*

Atomically thin van der Waals materials such as graphene and transition metal dichalcogenides (TMDs) exhibit remarkable physical properties resulting from their reduced dimensionality [1]. The TMDs are direct-gap semiconductors in the monolayer limit [2] and are thus an especially promising platform both for fundamental studies of two-dimensional systems and potential applications in ultra-thin optoelectronic devices. The quasi-two-dimensional (2D) character of these materials suggests a strong enhancement of the Coulomb interaction and the resulting formation of tightly bound electron-hole pairs, i.e., excitons. The understanding of the excitonic properties is critical both for the elucidation of the underlying many-body physics in such materials and for their use in electronic and photonic applications.

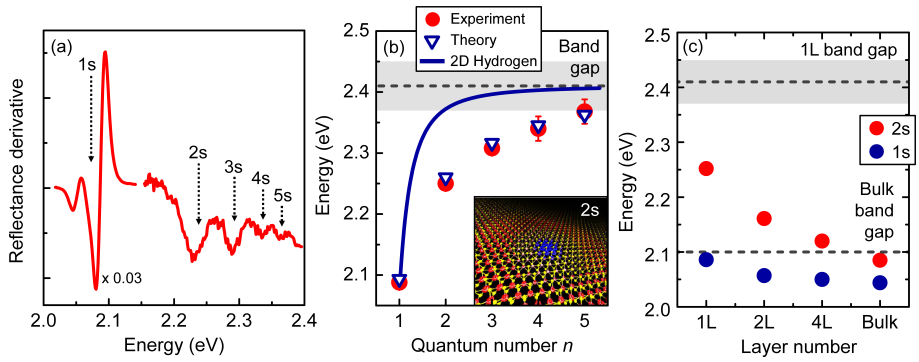

Figure 1: (a) The derivative of the reflectance spectrum of the WS_2 monolayer at T = 5 K with the exciton states are labeled by their respective quantum numbers. (b) Experimentally and theoretically obtained transition energies for the exciton states. The best fit of the data to the 2D hydrogen model for Wannier excitons is shown for comparison. An exemplary representation of the exciton wavefunction of the first excited state in real space is presented in the inset. (c) Energies of the ground and first excited states for various layer thicknesses. Band gaps of the bulk and the monolayer are represented by the dashed lines.

Here, we experimentally and theoretically investigate the properties of excitons in mono- and few-layer TMDs, identifying and characterizing a sequence of excited exciton states, i.e., Rydberg series. Analyzing the optical reflection spectra of these materials (Fig. 1(a)), both empirically and using a physically motivated model, yields an exciton binding energy as large as 0.32($\pm$0.04) eV. Remarkably, we also find significant deviations from the conventional hydrogenic model typically employed for the description of Wannier excitons (Fig. 1(b)). We explain both the large binding energy and the unusual spectrum of excited states within a theoretical approach that takes into account the non-uniform dielectric screening in 2D systems. A pronounced dependence of the exciton binding energy on material thickness is further observed by carefully monitoring the evolution of the excitonic states with increasing layer number (Fig. 1(c)). The findings bolster our understanding of the fundamental physics of 2D materials and lay the foundation for future studies of many-body phenomena in TMDs and TMD-based devices.

References

[1] K. S. Novoselov, D. Jiang, F. Schedin, T. J. Booth, V. V. Khotkevich, S. V. Morozov, and A. K. Geim, Proc. Natl. Acad. Sci. U.S.A. **102**, 10451 (2005)

[2] K.F. Mak, C. Lee, J. Hone, J. Shan, and T.F. Heinz, Phys. Rev. Lett. **105**, 136805 (2010)

Research carried out in part at the Center for Functional Nanomaterials, Brookhaven National Laboratory, which is supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886