

NATIONAL HIGH MAGNETIC FIELD LABORATORY 2017 ANNUAL RESEARCH REPORT

Bright-Dark Exciton Splitting in Monolayer MoSe2

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Introduction

Atomically thin group-VIB transition metal dichalcogenides (TMDs) have recently attracted vast interest as a new class of gapped semiconductors. When TMDs are thinned down from bulk to monolayers, a striking change in their electronic structure is the crossover from indirect to a direct band gap at the degenerate but inequivalent K and –K valleys at the corners of the hexagonal Brillouin zone. Since the conduction band (CB) edges at K/-K valleys are spin-split, the lowest energy excitonic state could be optically bright or dark depending on the nature of relevant spin-allowed or spin-forbidden transitions. Application of an external in-plane magnetic field mixes the components of the spin-split CB, thus brightening the originally dark excitons. Magnetic field brightening of dark excitons has been observed in WSe₂ monolayers [1], although the magnetic field induced splitting of bright and dark exciton in TMDs remains unexplored.

Experimental

The experiments were performed on a MoSe₂ monolayer encapsulated by h-BN. The photoluminescence and reflectance spectra were measured in Voigt geometry using a direct-optics micro-spectroscopy setup coupled either to the 14.5T (EMR facility) or 17.5T (DC field facility) superconducting magnet.

Results and Discussion

The PL spectrum of MoSe2/hBN features two peaks corresponding to the emission from bright neutral and negatively charged excitons. The in-plane magnetic field brightens the spin-forbidden neutral dark exciton making it clearly distinguishable at B>10T. The energy separation between bright and dark excitons varies quadratically with the magnetic field strength, consistent with theoretical expectations, in which the CB spin is linearly perturbed by the in-plane magnetic field. However, this simple model does not explain two important experimental observations: (i) the asymmetry of bright and dark exciton branches, (ii) the deviation from the B² behavior of exciton branches that appears in reflectance spectra at high fields above 14-15T.

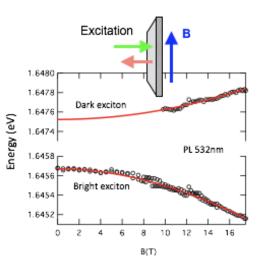


Fig.1. Splitting of dark and bright excitons of monolayer MoSe₂ as a function of in-plane magnetic field. Symbols represent the energies of PL peaks measured with non-resonant excitation.

Conclusions

PL and reflectance spectroscopy measurements on h-BN encapsulated MoSe₂ monolayers performed with inplane magnetic fields up to 17.5T reveal the effect of bright-dark exciton splitting. Further measurements at higher fields are needed to investigate the high-field anomalies of the splitting.

Acknowledgements

Z.J., S.M. and D.S. acknowledge the support from the U.S. Department of Energy (grant number DE-FG02-07ER46451) and the NHMFL User Collaboration Grants Program (grant UCGP No. 5087). A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490 and the State of Florida.

References

[1] Zhang, X.-X.; Cao, T.; Lu, Z.; Lin, Y.-C.; Zhang, F.; Wang, Y.; Li, Z.; Hone, J.C.; Robinson, J.A.; Smirnov, D.; Louie, S.G. and Heinz, T.F., *Magnetic brightening and control of dark excitons in monolayer WSe*₂, **Nature Nanotechnology**, **12**, 883 (2017)