Superradiant Coupling Effects in Transition-Metal Dichalcogenides

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Introduction

Cooperative effects allow for fascinating characteristics in light–matter interacting systems. Here, we study naturally occurring superradiant coupling in a class of quasi–two-dimensional, layered semiconductor systems. We perform optical absorption experiments of the lowest exciton for transition-metal dichalcogenides with different numbers of atomic layers. We examine two representative materials, $MoSe_2$ and WSe_2, using incoherent broadband white light. The measured transmission at the "A" exciton resonance does not saturate for optically thick samples consisting of hundreds of atomic layers, and the transmission varies nonmonotonously with the layer number. High magnetic fields are used to obtain the diamagnetic shift of the exciton and therefore its radius. A self-consistent microscopic calculation reproduces the experimental observations, clearly identifying superradiant coupling effects as the origin of this unexpected behavior.

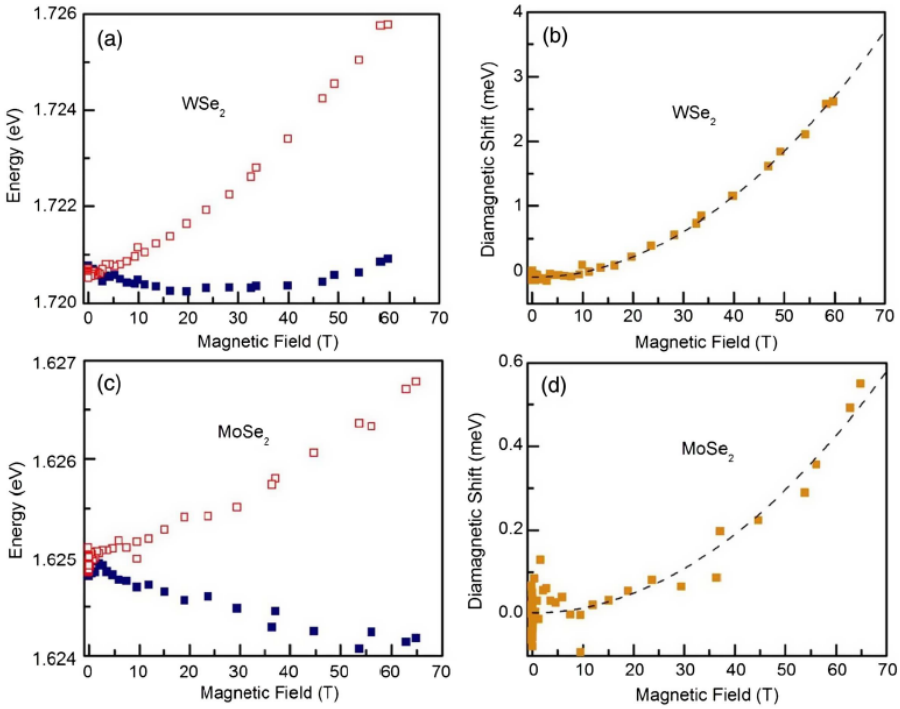

Figure: (a) Energy position of the "A" exciton in WSe_2 as a function of magnetic field strength up to +60 T (blue squares) and up to −60 Tesla (red squares). (b) Obtained diamagnetic shift for the "A" exciton in WSe_2 (yellow squares). The dashed line is a quadratic fitting. (c) Energy position of the A exciton in MoSe2 as a function of magnetic field strength up to +60 T (blue squares) and up to −60 T (red squares). (d) Obtained diamagnetic shift for the "A" exciton in MoSe2 (yellow squares). The dashed line is a quadratic fitting.

Results and Discussion

We analyze the optical transmission/absorption properties of exfoliated TMDC samples for different numbers of layers, ranging all the way from monolayers to bulk-like configurations with more than a hundred layers. We carefully characterize the respective sample thicknesses using AFM; for thinner samples, photoluminescence and Raman spectroscopy were additionally used. The absorption was quantitatively measured for the A exciton resonance by carefully measuring transmission and reflection simultaneously, and by taking into account all the possible losses. The resulting optical density as a function of sample thickness deviates significantly from the Beer–Lambert law. We model the observed effects employing the semiconductor Maxwell–Bloch equations for a classical optical field interacting with equidistantly spaced two-dimensional layers, where the "A" excitons are assumed to be well localized within the individual layers. To validate the model assumptions, we carefully measure the excitonic Bohr radii in the two representative bulk TMDs using the diamagnetic energy shift of the excitonic resonance at magnetic fields up to 65 T. The exciton wave functions obtained from these measurements are well localized within the layers and are also in agreement with recent angle-resolved photoemission studies performed in bulk WSe_2, which reveal a two-dimensional character of the bands at the K -point. The theoretical calculations reveal that the experimentally observed variation of the resonant optical absorption can be uniquely attributed to the superradiant coupling between the excitons in the different TMDC layers.

References

[1] Stevens, C. E. *et al.*, Optica **5**, 749 (2018).