Probing bandgap renormalization, excitonic effects, and interlayer coupling in 2D transition metal dichalcogenide semiconductors

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Abstract

Atomically-thin transition metal dichalcogenide (TMD) semiconductors have generated great interest recently due to their remarkable physical properties. In particular, reduced screening in 2D has been predicted to result in dramatically enhanced Coulomb interactions that should cause giant bandgap renormalization and excitonic effects in single-layer TMD semiconductors. Here we present direct experimental observation of extraordinarily high exciton binding energy and band structure renormalization in a single-layer of semiconducting TMD[1]. We have determined the binding energy of correlated electron-hole excitations in monolayer $MoSe_2$ grown via molecular beam epitaxy[2] on bilayer graphene (BLG) by using a combination of high-resolution scanning tunneling spectroscopy and photoluminescence spectroscopy. We have measured both the quasiparticle electronic bandgap and the optical transition energy of monolayer $MoSe_2$/BLG, thus enabling us to obtain an exciton binding energy of 0.55 eV for this system, a value that is orders of magnitude larger than what is seen in conventional 3D semiconductors. In a more highly screened environment (on top of graphite), we find that single layers of $MoSe_2$ show a strong 51% reduction in the exciton binding energy and an 11% reduction in the quasiparticle electronic gap, without significantly changing the optical gap. We have corroborated these experimental findings through ab-initio GW and Bethe-Salpeter equation calculations, which show that the large exciton binding energy arises from enhanced Coulomb interactions that lead to blue-shifting of the quasiparticle bandgap. We have also studied the role of interlayer coupling and layer-dependent carrier screening on the electronic structure[3] of few layer $MoSe_2$. We find that the electronic quasiparticle bandgap decreases by nearly 1 eV when going from one layer to three. Our findings paint a clear picture of the evolution of the electronic wave function hybridization in the valleys of both the valence and conduction bands as the number of layers is changed. These results are of fundamental importance for the design and evaluation of room-temperature electronic and optoelectronic nanodevices involving single layers of semiconducting TMDs as well as more complex layered heterostructures.

References

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