Quasiparticle Band Gap Renormalization in Semiconducting Single-Layer WS_2 Induced by Dielectric Screening

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Pristine transition metal dichalcogenides (TMDs) exhibit a unique combination of radiation tolerance and energy-efficient carrier transport in an atomically-thin profile. However, synthesis and device fabrication processes introduce defects and interface inhomogeneity into TMDs, which perturb the electronic structure and excitation dynamics by altering the dielectric screening environment. The dielectric screening strength, and the electronic structure perturbation, are proportional to the dielectric constant (ε) of the external environment (ε_{ext}) and the TMD (ε_{WS2}). Yet, the renormalizing effect of dielectric screening variations on the TMD electronic structure is unclear.

Here, we elucidate the impact of varying the external and internal dielectric screening strength on the electronic structure of single-layer WS_2, using ε_{ext} and ε_{WS2} as proxies for screening strength. We systematically vary the external screening strength through sample design including suspended WS_2 ($\varepsilon_{ext} \approx 2$) and Au-supported WS_2 ($\varepsilon_{ext} \approx 62$), and the internal screening strength by varying the WS_2 defect density through ion irradiation (5 MeV Fe^{2+}, 10^{11}-10^{13} cm^{-2}). We estimate the quasiparticle band gap and exciton binding energy as a function of ε_{ext} and ε_{WS2} using photoemission and electron energy loss spectroscopy (PES, EELS).

PES shows the valence band density of states exhibit an increasing shift towards the vacuum level with increasing dielectric screening strength. From the offset between the charge neutrality point and valence band edge, we estimate the band gap of suspended WS_2 is reduced by up to 160 meV up to an ion fluence of 10^{13} cm^{-2}, coinciding with an increased ε_{WS2}. More substantial band gap reductions are estimated in Au-supported WS_2. The estimated reduction in quasiparticle band gap primarily manifests from reduced exciton binding energy according to EELS. To enable broader application of our findings, we estimate the WS_2 defect density using Shockley-Read-Hall recombination. This study highlights the profound impact of the dielectric screening environment on TMD electronic structure.

This work was supported by a LDRD program at SNL. A.R.K. acknowledges support from the U.S. DOE SC, Division of MSE. SNL is a multimission laboratory managed and operated by NTESS, LLC, a subsidiary of Honeywell International, Inc., for the NNSA under contract DE-NA-0003525. The views expressed in the correspondence do not necessarily represent the views of the U.S. DOE or the U.S. Government. This work leveraged CINT, a U.S. DOE SC User Facility.