# Internal Disorder-Induced Transition from 2D to 3D Dielectric Screening in Single-Layer $WS_2$

**C. M. Smyth[1‡], A. Boehm[1§], K. Burns[2], C. Spataru[3], A. Kim[1], D. Bethke[1], T.-M. Lu[4], T. Ohta[1]**

[1] *Sandia National Laboratories, Albuquerque, NM, 87123, USA*
[2] *Materials Science & Engineering, University of Virginia, Charlottesville, VA, 22903, USA*
[4] *Sandia National Laboratories, Livermore, CA, 94550*
[3] *Center for Integrated Nanotechnologies, Albuquerque, NM 87123, USA*

The quasiparticle band gap ($E_{g,QP}$) and dielectric permittivity represent fundamental properties of bulk semiconductors as a result of the three-dimensional (3D) character of the dielectric screening. In two-dimensional (2D) materials, the lattice is confined well below the typical dielectric screening length, and the non-local characteristic of the dielectric function emerges, resulting in unusually large exciton radius and binding energy in the weak screening environment. Consequently, the $E_{g,QP}$ and exciton binding energy become highly responsive to perturbations in the dielectric screening with extrinsic origins. Ubiquitous monovacancies in 2D semiconductors host a polarizable dipole, which can impact the screening strength and $E_{g,QP}$ through the internal defect density ($n_v$). Similarly, substrate-induced screening can impact the $E_{g,QP}$ and exciton behavior, which often obscures the combined effects of multiple screening mechanisms.

We present an experimental investigation of the isolated and combined impacts of structural disorder and external dielectric screening on the $E_{g,QP}$ and exciton behavior in single-layer $WS_2$. Introducing dilute structural disorders, primarily sulfur monovacancies, to weakly screened $WS_2$ results in a significant 190 meV renormalization of the $E_{g,QP}$, while the optical bandgap remains effectively unchanged at around 2.0 eV, as confirmed by photoemission spectroscopy (PES) and electron energy loss spectroscopy (EELS). When the $n_v$ reaches a threshold of $5 \times 10^{12}$ cm$^{-2}$, the interactions between isolated excitons and defects in poorly screened $WS_2$ transition to correlated interactions. The exciton-defect interaction changes because the screening radius and interdefect distance converge at ~4 nm. The emergence of correlated exciton-defect interactions is attributed to the dielectric function transitioning from a 2D to 3D character. In contrast, the $E_{g,QP}$ and exciton binding energy of $WS_2$ remain unaffected by vacancies when the screening environment is dominated by a strongly screening Au substrate. Therefore, the sensitivity of the electronic band structure and exciton stability in 2D materials to the screening environment hinges on their polarizability.

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 was performed, in part, at CINT, a User Facility operated for the U.S. DOE SC.

‡ Author for correspondence: cmsmyt@sandia.gov
§ Current Affiliation: Los Alamos National Laboratory, Los Alamos, NM 87545, USA