Sulfur vacancy-induced dielectric screening crossover in single-layer WS_2

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Dielectric property in two-dimensional (2D) semiconductors is set by the external environment, charge carriers, and disorders, making defects a tuning parameter to modulate their electronic response. Here, we reveal quasiparticle electronic band gap ($E_{g,QP}$) renormalization in single-layer (1L) WS_2 by independently tuning sulfur vacancy density and substrate screening. Photoelectron spectroscopy and electron energy loss spectroscopy show that a defect density (n_V) of $1 \times 10^{13}\mathrm{cm}^{-2}$ reduces $E_{g,QP}$ from 2.4 eV to 2.2 eV in weakly-screened, suspended WS_2 with the diminished exciton binding energy (E_b) by 0.2 eV. In Au-supported WS_2 with strong external screening, $E_{g,QP}$ remains ~2.0 eV showing no $E_{g,QP}$ renormalization upon increasing n_V. Enhancing disorder additionally drives the reduced photoelectron core-hole relaxation in suspended WS_2 when the exciton screening length (estimated from E_b) exceeds the mean interdefect spacing of ~4 nm at $n_V \approx 6 \times 10^{12}\mathrm{cm}^{-2}$. This threshold n_V signifies the crossover from weak to strong polarizability in 2D dielectric screening.

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