Examining Radiation Effects on the Electronic Structure and Defect Density of 1L WS_2 through *in-situ* Photoemission Spectroscopy

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Two-dimensional (2D) transition metal dichalcogenides (TMDs) exhibit a unique combination of high radiation tolerance and promising computing performance in an atomically thin profile. However, the relationship between ion irradiation, defect density, and electronic structure in TMDs has not been identified, and must be understood to characterize radiation resilience. Heavy ion fluences of $<10^{12}$ cm^{-2} degrade TMD device performance, but are currently untraceable by conventional TMD characterization techniques. Hence, the relationship between radiation-induced defect density and carrier concentration in monolayer (1L) transition metal sulfides (TMS) remains unresolved. Some reports indicate ion irradiation induces hole doping in TMS, whereas theory predicts electron doping by sulfur vacancies, which are the most abundant defect formed in TMS under ion irradiation.

In this talk, we irradiate suspended 1L WS_2 films with 5 MeV Fe^{2+} ions at fluences of 10^{11}-10^{13} cm^{-2} to controllably introduce defects, and examine the valence band density of states (DOS) using photoemission spectroscopy. Photoemission spectra (PES) show the highest occupied valence band DOS at the Γ-point in the Brillouin zone shift towards the vacuum level and Γ-point peak width increases up to an ion fluence of 10^{13} cm^{-2}. These spectral modifications substantiate a reduced band gap and excited photo-hole lifetime by enhanced screening with increased defect density. By calculating the defect density as a function of ion fluence using the Shockley-Read-Hall recombination model, we demonstrate that the PES exhibits an unprecedented sensitivity to defect concentrations on the order of 10^{12} cm^{-2} generated at an ion fluence of 10^{11} cm^{-2}. We further verify ion beam-induced defect densities using scanning transmission electron microscopy. Additional ion irradiations up to 10^{13} ions cm^{-2} cause electron doping as predicted by theory, but we detect hole doping after ion irradiation beyond the 10^{13} cm^{-2} fluence. This study highlights the high sensitivity of the Γ-point valence band PES to radiation-induced changes in defect density, band gap, and excited photo-hole state behavior in 1L WS_2 down to ion fluences of 10^{11} cm^{-2}.

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