Tuning Phonon and Exciton Dynamics through alloying in 2D Transition Metal Dichalcogenides

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Quasiparticles significantly influence thermal transport and electronic behavior in 2D materials, where dipole interactions and lattice dynamics can be tuned through alloying. However, controlling momentum transitions of phonons, exciton coupling strength, and their locality based on intrinsic lattice stresses remains a challenge due to limitations in bulk characterization techniques, which average signals over many unit cells. To address this, we studied how alloying in $W_xMo_{1-x}S_2$ modulates local dipole moments and phonon/exciton populations. By varying stoichiometry across five compositions, we observed how atomic-scale disorder affects vibrational properties. X-ray photoelectron spectroscopy (XPS) analysis was used to confirm chemical uniformity. Raman and Infrared (IR) spectroscopy measured bulk vibrational modes, while photoluminescence (PL) revealed band transitions and valence band splitting. Monochromated electron energy loss spectroscopy (EELS) in an aberration corrected scanning transmission electron microscope (STEM) enabled atomic-scale mapping of phonon and exciton populations beyond the diffraction limit (Figure 1 and 2). This combined approach revealed how alloying impacts spin-orbit coupling and exciton-phonon interactions in low-dimensional semiconductors.

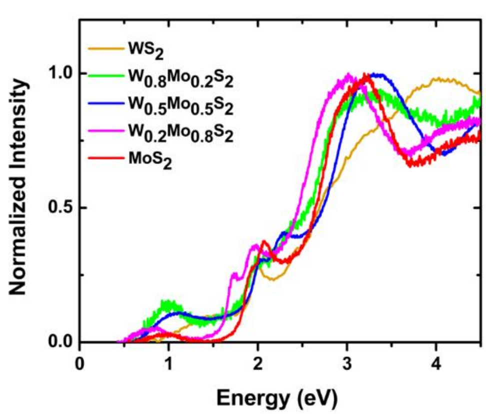

Figure 1. Optical measurements of WS_2, MoS_2, and three alloys of $W_xMo_{1-x}S_2$.

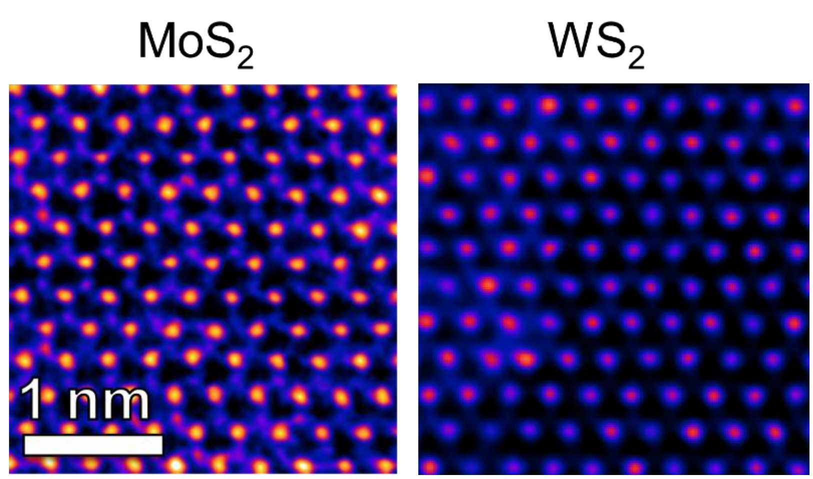

Figure 2. STEM images of pure MoS_2 and pure WS_2.