Determining Thermal Equilibrium Defect Densities for Transition Metal Dichalcogenides from First Principles and Experimental Thermodynamics

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Two-dimensional transition metal dichalcogenides (TMDs) have gained interest due to their novel optical and electronic phenomena and potential for optoelectronic applications. Chemical vapor deposition (CVD) and chemical vapor transport (CVT) are commonly used to synthesize TMDs due to their fast growths and high yields; however, they produce highly defective TMDs, negatively impacting the novel properties. Alternatively, the self-flux growth method has been shown to yield TMDs with defect densities several orders of magnitude lower than those from CVD and CVT. Yet, it is unknown how close the self-flux TMDs quality is to the thermal equilibrium defect density. In this work, we perform density functional theory calculations for intrinsic defects and substitutional oxygen impurities in WSe_2. We use ab initio modeling in conjunction with experimental thermodynamic data to calculate equilibrium formation energies and defect densities as a function of growth temperature. Theoretical chalcogen point defect densities at the self-flux growth conditions were shown to be an order of magnitude less than experimental values. The defect density difference between experiment and theory suggests that equilibrium was not reached, and kinetic or impurity factors alter the self-flux TMDs defect densities.