

2D Transition-Metal Dichalcogenides: Doping, Alloying and Electronic Structure Engineering Using Electron Beam

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By combining first-principles simulations with high-resolution transmission electron microscopy experiments, we study the evolution of atomically thin layers of transition metal dichalcogenides (TMDs) under electron irradiation. We show that vacancies produced by the electron beam agglomerate and form line structures, which can be used for engineering materials properties. We also study the radiation hardness of 2D TMD materials [1]. We further show that TMDs can be doped by filling the vacancies with impurity atoms. We also study the stability and electronic properties of single layers of mixed TMDs, such as $\text{MoS}_{2x}\text{Se}_{2(1-x)}$, which can be referred to as 2D random alloys [2]. We demonstrate that 2D mixed ternary $\text{MoS}_2/\text{MoSe}_2/\text{MoTe}_2$ compounds are thermodynamically stable at room temperature, so that such materials can be manufactured by CVD or exfoliation techniques. By applying the effective band theory approach we further study the electronic structure of the mixed ternary 2D TMD compounds and show that the direct gap in these material can continuously be tuned. Using GW first-principles calculations for few-layer MoS_2 , we further study [3] the effects of quantum confinement on the electronic structure of this layered material. By solving the Bethe-Salpeter equation, we evaluate the exciton energy in these systems. Our results are in excellent agreement with the available experimental data. Exciton binding energy is found to dramatically increase from 0.1 eV in the bulk to 1.1 eV in the monolayer. The fundamental band gap increases as well, so that the optical transition energies remain nearly constant.

[1] H.-P. Komsa, et al., *Phys. Rev. Lett.* 109 (2012) 035503.

[2] H.-P. Komsa and A. V. Krasheninnikov, *J. Phys. Chem. Lett.* 3 (2012) 3652.

[3] H.-P. Komsa and A. V. Krasheninnikov, *Phys. Rev. B (Rapid Comm.)* 86 (2012) 241201.