Materials Science & Engineering Doctoral Defense

Computational Design of Interfacial Properties for Materials Discovery

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Abstract

Interfacial interactions between materials in complex heterostructures can dominate the material's response in many modern-day energy-related devices and processes. Considerable research has been dedicated towards addressing the profound effects of interfaces. Here, first-principles-based quantum mechanical simulations are discussed to characterize the interfacial materials properties of two systems. First, density-functional theory (DFT) calculations were performed for ceramic oxide grain boundaries in undoped and doped CeO2. Second, the development, theoretical framework, and utilization of high-throughput, workflow-based, DFT calculations are presented to model the synthesis of two-dimensional (2D) heterostructured materials. Utilizing this workflow, predictive machine learning models were created to elucidate key interface-property relationships in 2D heterostructured materials.

The DFT simulations reveal that the Σ3(111)/[101] grain boundary was energetically more stable than the Σ3(121)/[101]grain boundary due to the larger atomic coherency in the Σ3(111)/[101] grain boundary plane. The alkaline-earth metal-doped grain boundary energies demonstrate a parabolic dependence on the size of the solutes, interfacial strain, and packing density of the grain boundary. The grain boundary energies were stabilized upon Ca, Sr, and Ba doping whereas Be and Mg render them energetically unstable. The electronic density of states reveals that no defect states were present in/above the band gap. The thermodynamic trapping of oxygen vacancies in the near grain boundary region was not significantly impacted by the presence of Ca-solute ions. However, the migration energy barriers within the grain boundary core were dramatically reduced with high local Ca-solute concentrations, around 0.3 eV-0.5 eV.

Chapter 5 and Chapter 6 discusses the development of the open-source, high-throughput computational "synthesis" based workflow package Hetero2d and the application of Hetero2d using 52 Janus 2D materials and 19 metallic, cubic phase, elemental substrates. The 438 Janus 2D-substrate pairs were analyzed by identifying substrate surfaces that stabilize metastable Janus 2D materials, characterizing their effects on the post-adsorbed 2D materials, and identifying the bonding between the 2D material and substrate. Machine learning models were applied to predict the binding energy, z-separation, and charge transfer of the Janus 2D-substrate pairs providing insight into the critical properties which factor into these properties.

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Zoom Link: https://asu.zoom.us/j/81957031669