# Atomic and Electronic Structure Prediction for Heterostructural Interfaces with Ultra-Wide Gap Materials

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First-principles calculations can be an invaluable tool for modeling of interface properties. However, such calculations require detailed knowledge of the atomic structure, which is often not known experimentally. This is especially true for heterostructural interfaces between materials with different bulk crystal structures. Thus, interface atomic structures are often constructed by making plausible assumptions but without confirmation that the structure actually represents a free energy minimum. Atomic force relaxation alone is generally insufficient, because it only yields a local total energy minimum. A more rigorous approach requires the sampling of the configuration space. In general, we distinguish commensurate and incommensurate interfaces, where the former displays a 1:1 matching of the in-plane primitive cell lattice vectors whereas the latter require construction of a coincidence site lattice for the matching of two 2D supercells. As an example of incommensurate interfaces, we recently adopted a bulk crystal structure prediction method to the slab geometry, so to predict heterostructural $SnO_2$/CdTe interfaces with atomically thin $CdCl_2$ interlayers facilitating the transition from the rutile to zincblende structure.[1]

As an example of a commensurate heterostructural interface, we here consider the case of wurtzite (wz) AlGaN alloys on rocksalt (rs) TaC (111) substrates, where the lattice matching of the hexagonal interface primitive cell occurs at high Al compositions for ultra-widegap applications. Using an algorithm for systematic enumeration of all possible different stacking sequences promoting the transition from the rocksalt to the wurtzite structure, we perform high-throughput density functional theory (HT-DFT) calculations to obtain the most favorable structures for each combination of substrate termination, film nucleation, and film polarity. Subsequent electronic structure calculations reveal the band alignment, Schottky barrier, and electronic interface density of states. We will further discuss the outlook for future work on commensurate and incommensurate heterostructural interfaces.



Figure 1. Lowest energy structures for rs-TaC/wz-AlN interfaces obtained from HT-DFT sampling of stacking sequences. There are 8 combinations of Ta/C termination, Al/N nucleation, and Al/N polarity.

[1] A. Sharan, M. Nardone, D. Krasikov, N. Singh, S. Lany, Appl. Phys. Rev. **9**, 041411 (2022).

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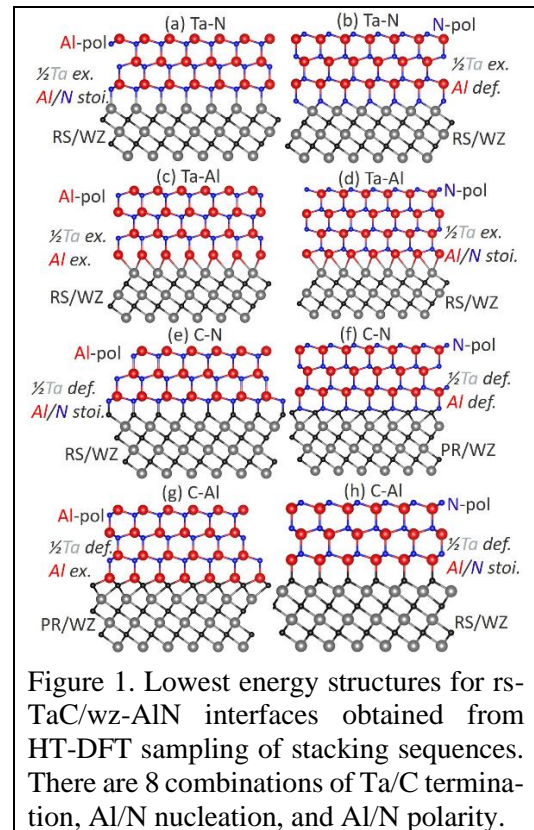
# Supplementary Pages

For incommensurate interfaces, the identification of a lattice-matched coincidence site lattice (CSL) is often not sufficient to construct realistic interface atomic structures. In a recent work (Ref. [1]), we used first principles structure prediction methods to sample the most likely atomic arrangements at the $SnO_2$/CdTe (rutile/zincblende) interface (Fig. S1). The atomic structure prediction were followed by band-gap corrected electronic structure calculations of the band alignment and interface density of states (Fig. S2).
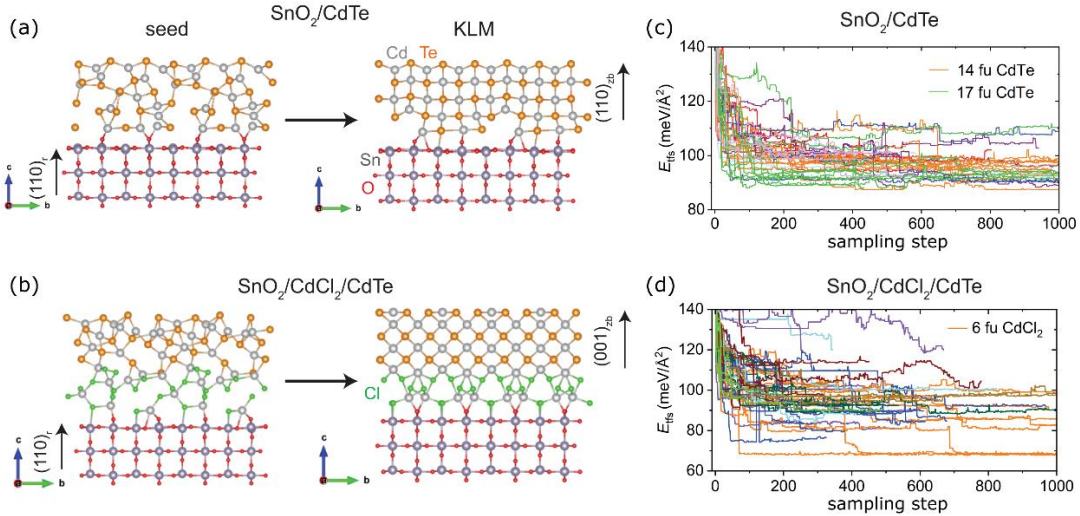


Figure S1: Atomic structure prediction for $SnO_2$/CdTe interfaces without (a,c) and with (b,d) a $CdCl_2$ interlayer. For the direct $SnO_2$/CdTe interface, a configuration with a fractional first CdTe layer is more favorable than the interface obtained by joining the two free surfaces. For the $SnO_2$/$CdCl_2$/CdTe interface, a new 2D phase is identified which reduces the interface energy and facilitates the transition for the rutile to zincblende structure without formation of defects.
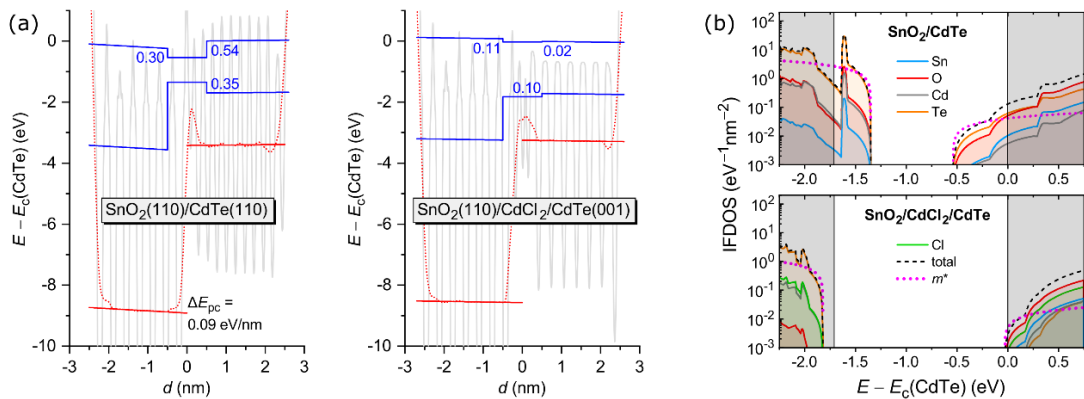


Figure S2: Band-gap corrected electronic structure prediction for the $SnO_2$/CdTe and $SnO_2$/$CdCl_2$/CdTe interfaces. (a) Band diagram with a 1 nm wide interface region. (b) Calculated electronic density of states at the interface in relation to the CdTe band gap.