Correlated oxygen states and Schottky barrier height in transition metal oxides from first principles

S. Apte,[1] I. Kim,[1] H. Jin,[1] A. Demkov[1+]
[1]Department of Physics, The University of Texas at Austin, Austin, Texas 78712, USA.

Schottky barrier height (SBH) [1] that measures the potential barrier for charge transfer across a metal-semiconductor interface, is one of the central quantities for semiconductor and electro-optic devices. SBH directly affects contact resistance, rectification, leakage current, and turn-on voltage. As such, accurately predicting Schottky barrier heights from first principles is of crucial scientific and technological importance.

In the ideal Schottky-Mott picture [2], the SBH is simply defined as the difference between the work function of the metal and the electron affinity of the semiconductor. However, in real materials the presence of interface dipoles, chemical bonding, metal-induced gap states and defect-driven Fermi-level pinning, makes the determination of the SBH extremely complicated. In particular semi-local density functional theory (DFT) suffers from a self-interaction error which results in underestimated band gaps and misaligned band edges [3]. In transition-metal oxides, a common remedy is to add a Hubbard U term on the transition metal d-orbitals that form the conduction band bottom [4]. While this strategy improves bulk gaps and d-state localization, it often remains insufficient for interfaces. For several materials the calculated barrier heights significantly deviate from experimental values unless much more expensive methods like GW are employed.

Many transition metal oxides have tops of their valance bands comprise oxygen 2p states with a relatively flat dispersion. These bands have a large effective mass and are particularly susceptible to self-interaction error, leading to a distortion in the band alignment at the interface. This observation motivates treating the oxygen 2p electrons as correlated degrees of freedom. In this work we show that introducing a Hubbard U on the oxygen 2p states systematically lowers the valence manifold, corrects the offsets, and yields quantitatively accurate barrier heights. We demonstrate this idea by choosing ten candidate oxides and constructing an interface with platinum. Platinum is a high work-function, chemically stable contact that is widely used experimentally. Importantly platinum does not readily scavenge oxygen under typical growth conditions. Beyond improving agreement, our approach is computationally efficient relative to hybrid functionals or many-body perturbation theory, making it practical for material screening and device-scale modeling.

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[+] Author for correspondence: demkov@physics.utexas.edu

Supplementary Pages

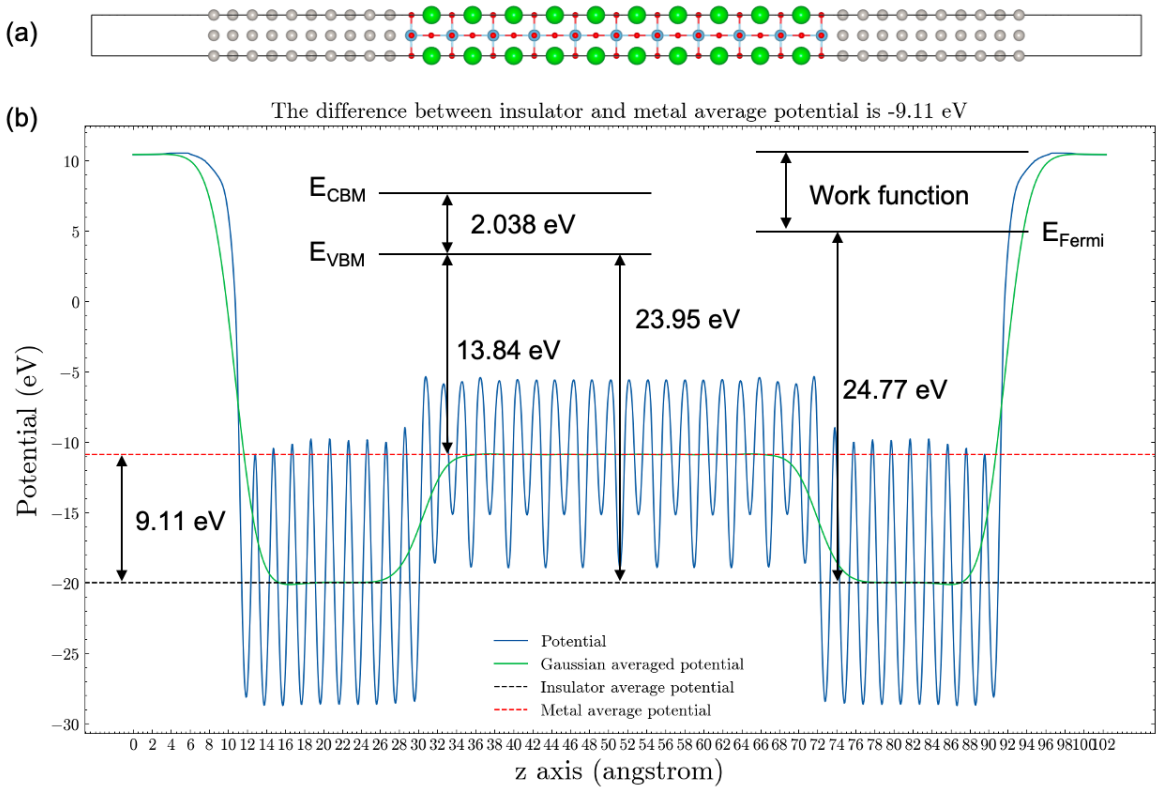

Figure 1. Schottky barrier height and electrostatic potential of a $SrTiO_3$-Pt interface. a) atomic structure of a Pt-$SrTiO_3$-Pt heterostructure. b) Planar averaged electrostatic potential along the out-of-plane direction. A Hubbard U of 8 eV is applied on the oxygen $2p$ states. Grey and red dashed lines show the average potential in the oxide and metal layers respectively. The relative positions of the valance and conduction band of the oxide and the metal Fermi level are marked. This yields a p-type Schottkey barrier height of 1.83 eV, which is in close agreement with the experimental value of $1.7 - 1.9\ eV$.