Ectronic and structural reconstructions in $LaAlO_3/SrTiO_3$ heterointerfaces imposed by oxygen vacancies and hydrogen adatoms

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Keywords: Heterostructures, defects, electronic structure, DFT+*U*.

Abstract

Recently, it was established that a two-dimensional electron system can arise at the interface between the two non-magnetic oxide insulators $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) [1]. Since then, different properties of this layer structure, such as metal-insulator transition (MIT) [2], superconductivity [3], ferromagnetism [4], were under investigation. However, despite huge amounts of both theoretical and experimental work a thorough understanding of most properties has not yet been achieved. The electronic reconstruction is caused by a charge transfer from the surface of LAO to the STO in order to avoid a polarization catastrophe which arises due to the sequence of polar LAO layers. Since surface states have a strong influence on the electrical conductivity, the generation of oxygen vacancies [5] and adsorption of hydrogen are key processes which can control the electronic reconstruction [6]. Previous findings related to surface chemistry in LAO/STO layer structures revealed reversible changes of the electrical conductivity, regulated by LAO surface protonation [6]. The aim of the present study is to investigate the electronic properties and structural distortions of surfaces and interfaces based on LAO and STO by means of density functional theory (DFT) in stochiometric structures and in the presence of defects.

In the present work we investigate the electronic properties of $LaAlO_3/SrTiO_3$ layer structures using varying values of an on-site Coulomb repulsion *U* term on Ti 3*d* and La 4*f* states within the GGA+*U* approach. We find that the position of the metal-insulator transition in the parameter space spanned by the *U* values on both Ti and La is sensitive to both values: While U_{Ti} increases the band gap of the layer structure, U_{La} shifts the Ti 3*d* states to lower energies and thereby leads to a reduction of the

band gap. Within the considered parameter space the number of $LaAlO_3$ overlayers required to induce metallic conductivity varies from five to six.

Based on these findings we have selected a set of physically reasonable *U* values and investigated structural deformations of the LAO and STO (001) slabs induced by hydrogen adatoms, oxygen vacancies, and a combination of these defects located both at the surface and inside the slabs. We found that defects lead to a strong buckling inside the slabs for all considered surfaces and interfaces. In addition, conducting surface states in the LAO slab may be fully occupied or emptied by surface defects, and at a certain concentration the surface may lose its conductivity. In contrast, defects at an insulating STO surface will provide electrons or holes and as a result the Fermi-level is shifted into the valence or conduction band.

The situation becomes even more complicated in the case of the layered LAO/STO structures. Surface defects provide electrons to the system as before, but they are not confined to the surface. For instance, the bare layer structure with three LAO overlayers, which is an insulator, may become metallic in the presence of defects.

We also calculated the formation energies for defects located inside different layers of LAO and STO slabs, and also inside the LAO/STO slab. Interestingly, the defect formation energies show a peculiar behaviour depending on the position of the defect within the slab. In addition, it strongly depends on the defect concentration as well as on the number of LAO overlayers in the case of LAO/STO layer structures.

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