Polar Discontinuity Governs Surface Segregation and Interface Termination: a case study of LaInO3/BaSnO3

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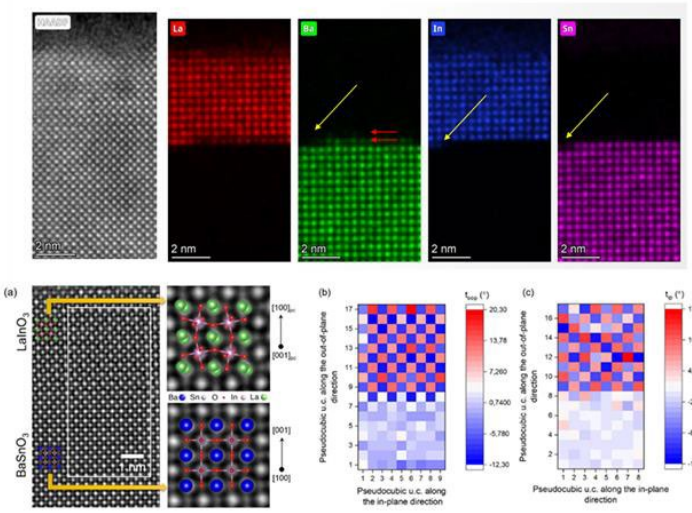
Background and aims.
Interfacial polar discontinuities are a unique way to manipulate charge states at interfaces and to create novel two-dimensional electron states. The polar-nonpolar interface between ABO3 perovskite oxides offers new degrees of freedom to tune interface states by the accessibility of mixed-valence states. The commonly accepted model for the formation of a two-dimensional electron gas (2DEG) is based on the concept of charge transfer between the layers terminating the polar-nonpolar interface. To realize a 2DEG or a two-dimensional hole gas (2DHG), control of the interface is a prerequisite. When growing the heterostructures, it is commonly assumed that the surface termination of the non-polar layer controls the interface. In this work, we provide experimental evidence that polar discontinuity compensation can drive surface segregation and thus control the interface

Methods.
We study the surface termination of BaSnO3 and the interface formation between the cubic wide bandgap semiconductor BaSnO3, and orthorhombic LaInO3, by negative cs imaging and direct spectral imaging of the LaInO3-BaSnO3 interface using Scanning Transmission Electron Microscopy Energy Dispersive X-Ray Spectroscopy (STEM-EDS) with the Thermo Fisher Spectra Ultra electron microscope equipped with Ultra X EDS detector. In parallel we perform Integrated Differential Phase Contrast STEM (iDPC-STEM) technique to study the oxygen octahedral tilt at the interface, providing further insights into the compensation of polar charges at that interface. Density functional theory (DFT) calculations are used to rationalize our experimental results. The samples were grown by plasma assisted molecular beam epitaxy on DyScO3 substrates at 835°C using a mixture of Sn and SnO2 as a SnO source.

Results.
While TEM experiments of BaSnO3 bulk crystals and DFT agree that BaO is the most stable surface termination of BaSnO3 over wide range of chemical potentials, we find by EDX spectral imaging the interface between BaSnO3 and LaInO3 is terminated by SnO2 (Fig. 1). This is consistent with the presence of a 2DEG but also with our DFT calculations which show this interface to be the most energetically favorable. STEM EDX shows the presence of BaO on the surface of thin LaInO3 films indicating Ba surface segregation. Based on our DFT calculations we find that the driving force for Ba segregation is the compensation of the polar discontinuity at the interface. This compensation is an effect the gradual reduction of the octahedral tilt from the orthorhombic LaInO3 to the cubic BaSnO3 and the polar and non-polar distortions at the interface as evidenced by the evaluation of iDPC images (Fig. 2). In the case of the n-type SnO2 interface, this leads to an expansion of the out-of-plane lattice spacing at the interface which most efficiently compensates for the discontinuity. At the BaO terminated p-type interface it leads to non-polar distortions in the BaSnO3, while polar distortions remain mainly in the LaInO3 which compensate the polar discontinuity less efficiently. This shows that in Perovskites in addition to surface energy and strain, the response of the system to compensate for the polar discontinuity must be considered as an additional driving force for segregation which may control the interface termination.

Graphic:



Keywords:

EDX, EELS, iDPC, Perovskites, 2DEG