First-principles modelling of dopants at interfaces in TCO materials

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Abstract

Nanometer-sized thin films, multi-layers, or nano-crystals of metal oxides in integrated functional devices are increasingly attracting interest in science and technology as potential alternatives to various silicon-based components, for instance dielectric thin-film capacitors for data storage, ferroelectric multi-layer stacks for mechanical actuators, or transparent and conducting oxides for optoelectronics or photovoltaics. The functional metal oxides in such devices contain structural defects and are in contact with electrodes. Upon size miniaturisation to nanometers, the functional properties of the devices are increasingly influenced by atomistic and electronic structures at defects and interfaces in the oxides and towards the electrodes. At such small dimensions, experimental atomic-scale characterisations are usually difficult. Alternatively, first-principles density-functional-theory (DFT) studies can provide detailed and predictive insight into relationships of interfacial structures and electronic properties.

This will be illustrated for ZnO [1] and TiO_2 with focus on their use as transparent conducting oxide (TCO) materials [2]. The impact of grain boundaries in a doped polycrystal on thermodynamic and electronic properties of atomic defects is investigated by DFT pseudopotential calculations for supercells containing both interfaces and dopants. For oxygen vacancies, cation dopants Al and Ga substituting Zn, or anion dopants N and P substituting O, energies of defect formation and interface segregation are determined in the local density approximation (LDA) of DFT. Defect levels in the electronic band structure of ZnO are analysed in terms of densities of states, which are calculated by means of the LDA with a self-interaction-correction (SIC). This SIC-LDA approach yields a theoretical band structure of bulk ZnO with sufficiently accurate band widths and band gaps, as compared to experiment, and it is applicable to large supercells needed for interfaces with no more computational effort than LDA. The important outcome of this study is a detailed microscopic information on how much positions and shapes of electronic defect levels can be altered at grain boundaries with respect to a doped single crystal.

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