Bulk and surface properties of Sr_2TaO_3N by density functional theory

M. Bouri[1], U. Aschauer[1]*

[1]University of Bern

Ruddlesden-Popper structured oxides are promising candidate materials for water-splitting catalysts [1], superconductors [2] and photovoltaic materials [3]. Furthermore, the substitution of oxygen with nitrogen in perovskite oxides lead to novel physical and chemical properties due to their difference in electronegativity [4-5]. However, the electronic features of oxynitrides with layered perovskite structure has not been studied rigorously.
In the present work, we perform first-principles density functional theory (DFT) calculations to investigate the electronic properties of the bulk Ruddlesden-Popper oxynitride Sr_2TaO_3N as well as its (001) surface. We show that the energetically favoured Sr_2TaO_3N bulk structure contains octahedral rotations combined with a *cis*-type ordering of the nitrogen anions on the equatorial sites of the octahedra. Besides the anion-*p* valence band and the Ta-*d* conduction band, the electronic density of states (DOS) of the bulk shows an unoccupied Ta-*d* state below the conduction band. This state also exists at the TaON terminated (001) surface but is slightly higher in energy than in the bulk, which means that excess electrons are preferentially accommodated in the bulk and subsurface layers. At the same time, the N-derived valence-band edge is shifted to higher energies at the surface, which implies that holes are preferentially located in the surface layer. These results show that photo-excited charge carriers remain spatially separated in the surface region, which should drastically reduce their recombination and lead to improved photocatalytic properties.

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