

MM 51: Computational Materials Modelling VII - Oxides

Time: Thursday 11:45–13:00

Location: TC 006

MM 51.1 Thu 11:45 TC 006

Structural and electronic properties of oxides within a transferable tight binding model — ●ALESSANDRO PARMA, GEORG H. K. MADSEN, and RALF DRAUTZ — ICAMS Ruhr-Universität Bochum, Stiepel Strasse 129, 44801 Bochum, Germany

Using the Tight Binding (TB) approximation charge transfer and magnetism as well as covalent bonding can be modelled. Thereby a TB approximation is obtained by a systematic coarse graining of the density functional theory energy functional. We have implemented an orthogonal pd TB model for transition metal oxides. The optimized minimal basis is obtained by downfolding a multiple- ζ LCAO basis. The bond integrals within the two center approximation are then calculated from a down-folded DFT Hamiltonian. They are found to be continuous and transferable, and allow for a parametrization of the bond energy in terms of exponential functional forms. We use a Stoner model for the inclusion of magnetism and charge transfer is approximated by monopole interactions.

We present the common oxide structures in terms of simple building blocks and analyze them within a close packing approach. Their stability is calculated across the transition metal series and is rationalized in terms of covalent bonding, atomic volume and electronegativity using simple TB models. The Hematite and Wüstite structures are presented in special detail.

MM 51.2 Thu 12:00 TC 006

Molecular Dynamics of Metal Oxide Systems with Polarizable Force Fields — ●PHILIPP BECK¹, PETER BROMMER^{1,2}, STEPHEN HOCKER³, JOHANNES ROTH¹, HANS-RAINER TREBIN¹, and SIEGFRIED SCHMAUDER³ — ¹Institut für Theoretische und Angewandte Physik (ITAP), Universität Stuttgart — ²Département de physique, Université de Montréal, QC, Canada — ³Institut für Materialprüfung, Werkstoffkunde und Festigkeitslehre (IMWF), Universität Stuttgart

Both force field generation and simulation of oxide systems are computationally much more demanding than those of metals or covalent materials due to long-range electrostatic interactions. We used the Wolf [1] direct, pairwise summation method with spherical truncation for Coulomb interactions and extended it to dipolar interactions. The polarizability of oxygen atoms is modeled with the Tangney-Scandolo [2] interaction force field approach. Due to the Wolf summation, the computational effort in simulation scales linearly in the number of particles, despite the presence of electrostatic interactions. Thus, this model allows to perform large-scale molecular dynamics simulations of metal oxides with realistic potentials. In the present contribution, we show the application to several metal oxide systems where simulations of microstructural, thermodynamic and vibrational properties are performed.

[1] D. Wolf *et al.*, J. Chem. Phys. **110**, 8254 (1999).

[2] P. Tangney and S. Scandolo, J. Chem. Phys. **117** 8898 (2002).

MM 51.3 Thu 12:15 TC 006

Thermodynamic stability of Cd₂SnO₄ and Zn₂SnO₄ and their point defects: A comparative study using first-principles methods — ●ARNO FEY, PÉTER ÁGOSTON, and KARSTEN ALBE — Institut für Materialwissenschaft, Technische Universität Darmstadt, Petersenstr. 32, 64287 Darmstadt

Ternary oxides represent a possible alternative to current transparent

conducting oxides (TCOs). Although high electric conductivities were found for Cd₂SnO₄, the presence of toxic Cadmium in widespread devices is a massive drawback. A possible alternative is Zn₂SnO₄ which is non-toxic and chemically similar, but until now its conductivity is more than one order of magnitude lower. In this study we investigate both, Cd₂SnO₄ and Zn₂SnO₄, by quantum mechanical calculations within density functional theory (DFT) in order to elucidate the reasons for the different conductivities. We study the thermodynamic stability of various arrangements of the cations in the inverse spinel structure, and the intrinsic point defects as a source for the conductivity. Our results show that Cd₂SnO₄ is thermodynamically stable for certain cation distributions, while no stable phase for Zn₂SnO₄ can be identified. The Sn_{Cd}- and Sn_{Zn}-antisite are the source of the *n*-type conductivity in these materials.

MM 51.4 Thu 12:30 TC 006

Interplay between defect dipoles and ferroelectric polarization in a lead-free piezoelectric material — ●SABINE KÖRBEL and CHRISTIAN ELSÄSSER — Fraunhofer-Institut für Werkstoffmechanik IWM, Wöhlerstraße 11, 79108 Freiburg, Germany

KNbO₃ is a ferroelectric perovskite-type compound and a special case of the solid-solution system (K,Na)NbO₃ (KNN), which is regarded as a prospective lead-free substitute for today's best material Pb(Zr,Ti)O₃ (PZT) in piezoelectric components. The piezoelectric properties of PZT and KNN are commonly optimized by doping. Depending on the application, ferroelectric hardness or large strain is desirable. Defect dipoles consisting of aliovalent dopants and associated oxygen vacancies can impede domain wall motion and lead to ferroelectric hardening, and in aged ferroelectrics defect dipoles can cause extraordinarily large strains [1]. Combining density-functional theory and classical empirical interatomic potentials, like in [2], we studied defect dipoles consisting of Cu substitutionals and oxygen vacancies in KNbO₃. In this way we obtained the energy needed for rotating the ferroelectric polarization around a defect dipole. This energy determines to which extent defect dipoles can pin domain walls, hence contribute to ferroelectric hardening and aging.

[1] X. Ren, Nature Materials **3**, 91 - 94 (2004).

[2] S. Körbel and C. Elsässer, Phys. Rev. B **84**, 014109.

MM 51.5 Thu 12:45 TC 006

Attracting shallow donors: Hydrogen passivation in (Al,Ga,In)-doped ZnO — ●MOZHGAN AMINI, MASAHIKO MATSUBARA, ROLANDO SANIZ, DIRK LAMOEN, and BART PARTOENS — CMT & EMAT, Departement Fysica, Universiteit Antwerpen, Groenenborgerlaan 171, B-2020 Antwerpen, Belgium

We show by ab initio calculations that hydrogen passivates the dopant in Al, Ga or In doped ZnO, although hydrogen and (Al, Ga or In) are both shallow donors in ZnO. This puts a limit on the *n*-type conductivity of the transparent conducting oxide ZnO, in agreement with recent experimental results. Ab initio studies have confirmed the expected attraction between donor and acceptors in ZnO, and between deep and shallow donors in ZnO. Shallow donors, however, are expected to repel each other, as they both donate electrons to the system and become positively charged. But in this work we predict that they can also attract, and, even more important, can form a deep donor level. This is a new passivation mechanism that may also be relevant for other materials.