MM 59: Computational Materials Modelling - Phase Stability III

Time: Thursday 15:45–17:00 Location: H24

MM 59.1 Thu 15:45 H24

Phase diagram of Cr_xSb_y - A theoretical study on the structural and magnetic properties — •Gerhard Kuhn[1], Svitlana Polesya[1], Sergiy Mankovsky[1], Hubert Ebert[1], Matthias Regus[2], and Wolfgang Bensch[2] — [1]Ludwig-Maximilians-Universität München — [2]Christian-Albrechts-Universtität zu Kiel

We present results of first-principles investigations on the electronic structure, magnetic properties and phase stability of stoichiometric Cr_xSb_y compounds. The work is based on electronic structure calculations performed within the frame of DFT. The magnetic properties at finite temperature are studied using the Monte Carlo method based on the Heisenberg model.

The focus will be on $CrSb_2$ with marcasite structure exhibiting interesting electronic and magnetic properties. GGA as well as GGA+U calculations have been performed to show the role of local correlation effects, for electronic and magnetic properties. Comparing different types of magnetic order - i.e. ferromagnetic (FM) and antiferromagnetic (AFM) order along different directions - we have found in accordance with experiment the AFM order to be the stable one. Furthermore total energy calculations show a phase transition from marcasite to $CuAl_2$-structure for hight pressures according to experimental investigations.

Different types of structures are considered for the Cr_3Sb phase that is observed in several experiments. Total energy calculations for the most probable structures were performed to compare their stabilities and properties.

MM 59.2 Thu 16:00 H24

Rationalizing and screening high throughput DFT calculations using systematic tight binding models of transition metal - group 14 compounds. — •Alessandro Parma, Eunan J. McEniry, Ingo Opahle, Georg K. H. Madsen, and Ralf Drautz — ICAMS, Ruhr-Universität Bochum Universität Strasse 90a, 44789 Bochum, Germany

High throughput Density Functional Theory calculations are a powerful tool in materials design, since they allow for the discovery of new stable structures with relevant electronic or mechanical properties.

We rationalize the phase stabilities of transition metal-group 14 compounds based on systematically derived orthogonal Tight Binding (OTB) models. 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 this down-folded DFT Hamiltonian. They are found to be continuous and transferable and show trends across the transition metal period, allowing us to implement a general *pd* OTB model.

Our focus will be on the prediction of structural stabilities in terms of number of electrons in the *d* shell and principal quantum number of the *p*-states. An efficient screening strategy is developed based on the structural energy difference theorem. A reliable OTB model that reproduces DFT trends is crucial in the framework of high throughput structure search, since it allows for much more efficient energy calculations.

MM 59.3 Thu 16:15 H24

Phase diagram and thermodynamic properties of aluminum nitride from *ab initio* calculations — •Steve Schmerler and Jens Kortus — TU Bergakademie Freiberg, Institut für Theoretische Physik, Leipziger Str. 23, 09599 Freiberg, Germany

The AlN system is of interest in the context of phase diagram studies of the Si–Al–O–N system where AlN is one important binary compound. Recent experiments have generated interest in the calculation of a *P-T* phase diagram for the AlN system in order to supplement the avaliable *ab initio* based data.

We used density functional perturbation theory in order to obtain the phonon density of states and the quasi-harmonic approximation to calculate thermodynamic properties of the low pressure wurtzite and high pressure rocksalt phase.

We present results on the *P-T* behavior of thermodynamic properties (heat capacity, bulk modulus, thermal expansion) and compare to experimental data. Especially, we show how the anisotropic thermal expansion of the wurtzite phase is calculated. A phase diagram constructed from calculated Gibbs enthalpy data for the wurtzite - rocksalt phase transition is presented. We also discuss the limits of the method and report results regarding the thermal stability of the high pressure phase.

We would like to thank the DFG for financial support within the DFG Priority Program 1236: *Strukturen und Eigenschaften von Kristallen bei extrem hohen Drücken und Temperaturen*

MM 59.4 Thu 16:30 H24

Structure optimization via "local heat pulse"-quench cycles — •Arnulf Möbius[1] and Christian Schön[2] — [1]Institute for Theoretical Solid State Physics, IFW, Dresden — [2]Max Planck Institute for Solid State Research, Stuttgart

Structure prediction for crystals with a large number of atoms in the primitive cell, as well as for clusters of many atoms, are often impeded by the corresponding energy landscape exhibiting a huge number of local minima. For such tasks, we present an optimization procedure which is based on "local heat pulse"-quench cycles (LHPQC). It was originally developed for combinatorial optimization tasks [1].

This approach is applied to a lattice structure prediction problem. In that, we use the general utility lattice program (GULP) by J.D. Gale and co-workers [2] as local search code. As a test case, the energy landscape of the $Mg_{10}Al_4Ge_2Si_8O_{36}$ lattice is investigated, where the interactions are modelled by Coulomb, Buckingham, and three-body potentials [3], and where the cell parameters are free to vary.

The results of our computer experiments testify that the LHPQC procedure is robust and far more efficient than the previous approaches to the same test problem in Ref. 3. Finally, we show how our procedure can be easily parallelized: Its efficiency is considerably improved by treating an ensemble of local minima instead of a single one.

[1] A. Möbius, A. Neklioudov, et al., Phys. Rev. Lett. 79 (1997) 4297.
[2] J.D. Gale and A.L Rohl, Mol. Simul. 29 (2003) 291.
[3] A.R. Oganov, J.C. Schön, et al., in "Modern methods of Crystal Structure Prediction", ed. A.R. Organov, (Wiley, 2011), p. 223.

MM 59.5 Thu 16:45 H24

Improving the magneto-caloric effect in layered alloys: an ab-initio investigation — •Biswanath Dutta[1], Tilmann Hickel[1], Jörg Neugebauer[1], and Andreas Hütten[2] — [1]Max-Planck-Institut für Eisenforschung GmbH, D-40237, Düsseldorf, Germany — [2]Department of Physics, Thin Films and Physics of Nanostructures, Bielefeld University, PB 100131, 33501 Bielefeld, Germany

The study of magnetocaloric effect (MCE) based magnetic refrigeration is primarily focused on increasing the magnetic entropy difference ΔSmag(H,T) between the two phases across the coupled magnetic and structural transition. Here, we demonstrate a novel strategy consisting of alternating layers of different magnetocaloric materials. For this study, we choose Heusler alloys Ni-Mn-Sn and Ni-Mn-Ga for which the details of MCE in the bulk phase are well investigated. The presence of different chemical species across the interface in the layered structure, however, opens up the possibility of significant changes in the magnetic and structural properties which we investigate with the help of density functional theory. Our calculations reveal that the atomic magnetic moments get modified near the interface leading to the conclusion that interfaces could influence the transition temperatures and the entropy change and thus alter the MCE. In order to further clarify this influence, we carefully investigated strain contributions, interlayer diffusion and possible segregation effects near the interface.