MM 39: Computational Materials Modelling V

Time: Thursday 16:15–19:00   Location: IFW A

MM 39.1   Thu 16:15   IFW A

First-principles phase stability calculations of $Al_3(Ti_{1-x}Nb_x)$ pseudobinary alloys — •Yuhong Liu[1,2], Peter Puschnig[1], Ernst Gamsjäger[2], and Claudia Ambrosch-draxl[1] — [1]Atomistic Modelling and Design of Materials, University of Leoben, Austria — [2]Institute of Mechanics, University of Leoben, Austria

In the TiAlNb-system(mole fraction of Al amounts to 0.75), the intermetallic ε-phase can exist in the $L1_2$, $D0_{22}$, or $D0_{23}$ structure. Experimental results indicate that the addition of Nb shows a site preference for the Ti sublattice. Therefore, we calculate the thermodynamic properties of $Al_3(Ti_{1-x}Nb_x)$ by assuming a pseudo-binary alloy system. We utilize the ATAT package to perform the sublattice cluster-expansion approach. Total energies of various ordered supercell structures are obtained within density-functional theory using the projector augmented wave (PAW) method as implemented in the VASP code. Effective cluster interactions derived from the structure-inversion method are then used in an Ising-type Hamiltonian, which is solved by Monte-Carlo simulations. Our results show that the addition of Nb stabilizes the cubic $L1_2$ structure relative to the $D0_{22}$ and $D0_{23}$ ones. We find the effect of short-range-order on the calculated formation energies to be small compared to random mixing on the Ti-Nb sublattice. At high temperature, thermodynamic quantities such as the enthalpy and free energy are extracted from the Monte-Carlo simulations and compared to results from the CALPHAD method.

MM 39.2   Thu 16:30   IFW A

Hydrogen solution enthalpies derived from first principles: Chemical trends along the series of transition metals — •Ugur Aydin[1], Sixten Boeck[2], Tilmann Hickel[1], and Jörg Neugebauer[1] — [1]Max-Planck Institut für Eisenforschung — [2]Gemmantics IT-Consulting

Since the mid-19th century it has been known that some transition metals (TM) can absorb significant amounts of hydrogen, whereas others cannot. Since the presence of this interstitial atom can lead to serious materials failures, the energetics and dynamics of hydrogen in TMs are of critical importance in state-of-the-art materials design. Most of the previous studies, considering the hydrogen solubility for a larger number of TMs, rely on experimental observations and/or semi-empirical models. However, the ability of these models to provide a deeper understanding and insight into the decisive underlying mechanisms involved in hydrogen-solubility is limited. In our work, we therefore employed ab initio calculations to systematically investigate the mechanical and chemical mechanisms governing hydrogen solution in a complete set of TM under comparable conditions. The solution enthalpy for H in the high symmetry interstitial sites of TMs has been calculated consistently assuming various crystallographic / magnetic structures. For the data management a sophisticated automatic database has been developed. The analysis of this complete set of data allowed us to detect an universal dependence of the H solution enthalpy on the crystal lattice constant. Further a material dependent interplay of chemical and strain contributions has been found.

MM 39.3   Thu 16:45   IFW A

Interplay between magnetism and defects in Fe, Cr and their alloys from first principles — •Chu-Chun Fu[1], Romain Soulairol[1], and Cyrille Barreteau[2] — [1]CEA,DEN, Service de Recherche de Metallurgie Physique, 91191 Gif sur Yvette, France — [2]CEA-Saclay, DSM/IRAMIS/SPCSI, 91191 Gif sur Yvette, France

Iron-based alloys, which show complex structural-magnetic phase diagrams, play a major role in metallurgical and nuclear technology. In this work, we investigate, by means of Density Functionnal Theory (DFT), the influence of magnetism on structural and energetical properties of such alloys.

Fist, we give a detailed description of various non-conventional magnetic phases such as spin spiral and spin density waves in bulk Fe and Cr, in particular, we predict the polarization of spin density waves in the ground state of Cr in good agreement with experimental evidence [1].

Then, we report on the properties of defects (vacancies, substitutional impurities and interfaces) in Cr and FeCr alloys with various magnetic configurations. Our results show a significant interplay between magnetism and defects, e.g. stabilization of non-collinear magnetic configurations near both FeCr (110) interface and small Cr clusters in Fe. The dissolution energy of *3d* elements in bcc Fe is also detailed, pointing out the influence of both d-band filling and magnetism, the latter being crucial in the case of Cr in Fe.

[1] R. Soulairol et al., J. Phys. Condens. Matter, 22, 295502 (2010)

MM 39.4   Thu 17:00   IFW A

Superheating Gallium Clusters — •Nicola Gaston[1] and Krista G. Steenbergen[2] — [1]MacDiarmid Institute, IRL, Wellington, New Zealand — [2]Victoria University Wellington, New Zealand

The experimental discovery of superheating in gallium clusters [1] contradicted the clear and well-demonstrated paradigm that the melting temperature of a particle should decrease with its size [2]. However the extremely sensitive dependence of melting temperature on size also goes to the heart of cluster science, and the interplay between the effects of electronic and geometric structure [3]. In the case of gallium, the extreme polymorphism displayed by the bulk metal introduces additional complications. We use our understanding of the dimeric bulk structure of gallium to elucidate the patterns of bonding in the clusters, which also display dimeric structural motifs for small sizes. In particular, the deviation of gallium from the extremely general linear relationship of melting temperature and cohesive energy for elemental solids demonstrates that the low melting temperature of gallium corresponds to the melting of a molecular solid. We present systematic calculations of the melting-like transition in small gallium clusters, and a comparison with aluminium clusters of similar sizes. Finally, we discuss the importance of features of the electronic structure for the melting-like transition.

[1] G. A. Breaux, R. C. Benirschke, T. Sugai, B. S. Kinnear, and M. F. Jarrold, Phys. Rev. Lett. 91, 215508 (2003) [2]P. Pawlow, Z. Phys. Chem. 65, 1 (1909) [3]M. Schmidt, R. Kusche, B. von Issendorff, and H. Haberland, Nature 393, 238 (1998)i

MM 39.5   Thu 17:15   IFW A

Lithium oxide nanoparticles in lithium-air batteries: an ab-initio study — •Nicola Seriani — The Abdus Salam International Centre for Theoretical Physics

Lithium-air batteries may become an attractive solution for energy storage thanks to their high energy density. A key process in this kind of battery is the reversible lithium oxidation at the cathode. To get an atomistic insight into this process, we have performed density functional theory calculations. We show how the resulting oxide phase might be either Li_2O or Li_2O_2, depending on temperature and particle size. At room temperature, Li_2O_2 is the stable phase for particles smaller than 2.5 nm, whereas at larger diameters Li_2O becomes stable. Consequences for battery performance are discussed.

MM 39.6   Thu 17:30   IFW A

Multiscale modeling of nanowire-based Schottky-barrier field-effect transistors for sensor applications — •Daijiro Nozaki[1], Jens Kunstmann[1], Felix Zoergiebel[1], and Gianaurelio Cuniberti[1,2] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, Dresden University of Technology, 01062, Dresden, Germany — [2]Division of IT Convergence Engineering and National Center for Nanomaterials Technology, POSTECH, Pohang, Republic of Korea

We developed a theoretical framework for the calculation of charge transport through nanowire-based Schottky-barrier field-effect transistors that is conceptually simple but still captures the relevant physical mechanism of the transport process. Our multiscale model combines two approaches on different lengthscales: (1) the finite element method to calculate the electrostatic potential across the Schottky-contact and (2) the Landauer approach combined with the method of non-equilibrium Green's functions to calculate the electron transmission through the device. Our model correctly reproduces typical I-V characteristics of FETs and we obtained current saturations and high on/off ratios that are in good agreement with the experiment.

MM 39.7   Thu 17:45   IFW A

A molecular dynamics approach to simulate Raman and IR spectra in silicon nanowires — Felix Zörgiebel, •Jens Kunstmann, Daijiro Nozaki, and Gianaurelio Cuniberti — Insitute

for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany

We developed a method to filter phonons from molecular dynamics (MD) simulations based on a density functional tight binding approach. We are able to extract phonon frequencies, polarizations, and q-vectors. Our results for silicon bulk and silicon nanowires show good agreement with experimental data and exceed the accuracy and applicability of the Hesse matrix approach. In particular we are able to determine phonon occupations and temperature effects. Furthermore we filtered Raman, Infrared, and silent modes from MD trajectories via symmetry analysis. This enables us to predict Raman and Infrared spectra of silicon nanowires and explain their origin in terms of phonon polarizations.

MM 39.8   Thu 18:00   IFW A

TDDFT study of the momentum-dependent loss function of palladium — ●Stephan Sagmeister[1], Kathrin Glantschnig[1], Gerald Kothleitner[2], Cecile Hebert[3], and Claudia Ambrosch-Draxl[1] — [1]Chair of Atomistic Modelling and Design of Materials, University of Leoben , Austria — [2]Institut für Elektronenmikroskopie und Feinstrukturforschung, TU Graz , Austria — [3]Ecole Polytechnique Federale de Lausanne (EPFL) (Institute of Condensed Matter Physics) , Switzerland

The momentum-dependent loss function of several bulk metals, including palladium, is investigated by means of time-dependent density functional theory (TDDFT) as well as experimental data based on EELS (electron energy loss spectroscopy) measurements. On the theoretical side, density-functional theory within the full-potential LAPW (linearized augmented planewave) method is employed to obtain the Kohn-Sham band structure, using the exciting code. The framework of TDDFT is utilized within the linear response regime for the calculation of the macroscopic dielectric function and hence the loss function, including crystal local-field effects. The loss function is studied as a function of momentum transfer q for several directions in the Brillouin zone. While TDLDA, or even the independent-particle approximation, can well reproduce the low energy range, for higher energies local-field effects become crucial. For the case of Pd this is reflected in the spectra within the range of 40 to 60 eV, where the local-field effects can reduce the spectral weight by up to 50%. The calculations are in excellent agreement with experimental data.

MM 39.9   Thu 18:15   IFW A

Lead-free piezo-electrics: A combined investigation via synchrotron measurements and first-principles calculations — ●Sascha B. Maisel, Henry E. Mgbemere, Rodrigo P. Fernandes, Gerold A. Schneider, and Stefan Müller — Hamburg University of Technology, Institute of Advanced Ceramics, Denickestrasse 15, 21073 Hamburg, Germany

We present structural and chemical investigations of lead-free piezomaterials based on perovskite niobates like $NaNbO_3$, $KNbO_3$, $LiNbO_3$ and their multinary compounds from ab-initio calculations. Since some intrinsic properties of these materials (like ferroelectricity) are very sensitive to cell geometry and the precision of relaxations, the choice of the correct super cells and exchange-correlation functionals is crucial. To make the proper choices, we compare the data from density functional theory with experimental data from both single crystals and

sintered ceramics, with very good agreement. The cluster expansion formalism is applied to these DFT results in order to obtain concentration resolved information on short range ordering of the materials. Once the proper short range ordering is known, we are able to calculate piezoelectric response tensors for realistic super cells by means of density functional perturbation theory. The eventual aim of these investigations is to give a quantitative description of the electric and piezoelectric behavior as a function of compound concentration.

MM 39.10   Thu 18:30   IFW A

Superabundant vacancies in fcc metals: A combination of ab-initio, thermodynamic and kinetics approaches — ●Roman Nazarov, Tilmann Hickel, and Jörg Neugebauer — Max-Planck-Institut für Eisenforschung, Düsseldorf, Germany

A dramatic increase of the vacancy concentration in a H-rich atmosphere, the so called superabundant vacancy formation, has been experimentally observed in several metals and alloys. In order to study this phenomenon we systematically applied density functional theory to a large set of fcc metals. We found that a large amount of H can be trapped by a monovacancy with, e.g., up to 15 H atoms in an Al vacancy, up to 12 H atoms in the case of Pd and more than 17 H atoms for Pb.

Based on the defect formation energies from DFT calculations, we have constructed a thermodynamic model that determines the equilibrium concentration of point defects as a function of temperature and H chemical potential. By applying this approach we revealed that the vacancy concentration can indeed strongly increase if H is added.

To understand the phenomenon of accelerated self-diffusion in a H-rich atmosphere we coupled the information on the number of vacancies from the thermodynamic treatment with self-diffusion barriers obtained from DFT calculations. Using this approach we found that the self-diffusion coefficient is reduced not only due to the increased vacancy concentration, but also as a result of a H-induced lubricant effect.

MM 39.11   Thu 18:45   IFW A

Mechanism of Hydrogen Binding to Metal-Doped Carbon Nanostructures — ●Mina Yoon[1,2] and Matthias Scheffler[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft — [2]Oak Ridge National Laboratory, USA

Using quntum mechanical first-principles calculations based on numerical atom-centered orbitals as all-electron basis functions we study the mechanism of hydrogen binding to metal-doped nanoscale carbons. These materials have been widely studied as potential building blocks for hydrogen storage. We systematically investigate and compare the performance of different approaches in describing the interaction between hydrogen and metals doped on nanocarbons and the importance of the vibrational contribution in the formation enthalpy. The employed approaches include various DFT xc functionals, a semi-empirical approach, and Møller-Plesset perturbation theory. By comparing the chemical potential with that of free hydrogen gas at a finite temperature (T) and pressure (p), we identify the (T,p) conditions for hydrogen absorption/desorption. The vibrational contribution to the chemical potential of hydrogen is prominent for dihydrogen adsorption to metals, where its significance dramatically changes depending on the binding characteristics. This feature is illustrated by the example of metal-doped fullerenes and graphenes.