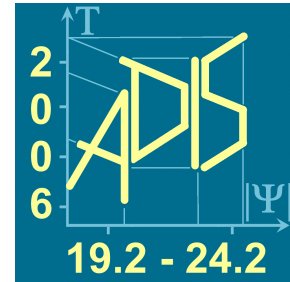


International Workshop on

Ab initio Description of Iron and Steel Status and Future Challenges

- Final Report -



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Convener Name : Tilmann Hickel

1. Summary

The aim of this workshop has been to provide a forum where internationally leading experts from different scientific communities (basic research, applied science, industry) may gather, present tutorial-like lectures on their methods, exchange the latest results, and identify future trends and developments. After each talk, there were lively and fruitful discussions about the suggested methods, final results, and future directions. The lively atmosphere and the tutorial character of the talks encouraged the participants to contribute to this exchange of ideas. Several contributed talks as well as a poster session gave all participants the opportunity to discuss their latest results.

The scientific topic of the meeting, the ab initio based description of materials properties and processes, is a field of rapidly growing interest. The talks discussed various combinations of accurate quantum-mechanical description at the atomic scale with meso- and/or macroscale approaches such as thermodynamics, kinetics and phase field simulations and showed that this approach provides a qualitatively new level of predictive power. Examples, which have been successfully tackled by this approach ranged from fundamental aspects, e.g. science in the earth core, to industrial research activities in designing novel materials for engine blocks. The talks made also clear that despite recent tremendous success, these methods are still in their infancy and many open problems remain. Main challenges which have been discussed are the quest for an improved description on the atomistic level to obtain accurate ground states and potential energy surfaces, construction of efficient and accurate tools to map the first-principles results onto Ising-like Hamiltonians, incorporation of vibrational and anharmonic entropy contributions or accurate description of magnetism and its temperature-dependent effects. The participants of the workshop felt that to address these challenges a joint effort of the scientific groups working in the fields of ab-initio calculations and thermodynamic / kinetic modeling of materials is needed and that the ADIS2006 workshop served as an important step in this direction.

2. Description of the scientific content

The main contributions to the international workshop ADIS2006 were the tutorial-like lectures of the invited speakers. The large number (15) of these talks allowed a detailed overview of a broad scope of state-of-the-art simulation techniques for the description of iron, steel and related materials. The longer than usual talks (75min+15min discussion) allowed the speakers to give a profound insight into the fundamentals, the applicability and the outcome of specific ab-initio methods, and were the basis for very lively discussions. In the following a brief summary of the lectures:

Dario Alfè started the course with his concepts for a calculation of Gibbs free energies of iron from first principles. He performs density functional theory in conjunction with the technique of *thermodynamic integration*. Different approaches to melting, based on free energies and the coexistence method, were discussed. The theory was applied to the melting conditions of iron under Earth's core conditions.

Frank Lechermann considered in his talk the ternary system Ni-Fe-Al as a test case for how far one can come with the calculation of finite-temperature phase diagrams of multi-component systems by a combination of the ab initio electron theory with statistical mechanics. For the calculation of the whole Gibbs triangle a mixed basis pseudopotential code was combined with the *cluster-expansion method*. Lechermann discussed the *cluster-variation method* of statistical mechanics in order to calculate the ab initio phase diagrams at finite temperatures.

Mojmír Šob focused in a first part on magnetic behavior changes of solids when a high-strain deformation is applied. He displayed total energies in contour plots as functions of distortions and volume, calculated by *spin-polarized full-potential LAPW* method. In a second part he briefly outlined the *CALPHAD method* for the construction of phase diagrams in alloys containing complex intermetallic phases. Šob showed ab initio total energy differences (lattice stabilities) for sigma phase in Fe-Cr, Fe-Cr-Ni and Fe-Cr-Mo systems.

Gerbrand Ceder also pointed out that within the last fifteen years the *cluster expansion technique* has become a practical solution to predict the most stable structures and phase diagrams on a chosen parent lattice. By *data mining* more than 15,000 first principles energy calculations more than 85 alloys, he furthermore showed that the energies of different crystal structures are strongly correlated between different chemical systems, and demonstrated how this correlation can be used to boost phase stability investigations of new systems.

Mark Asta discussed applications of *atomic-scale molecular-dynamics (MD)* computer simulations to the calculation of interfacial thermodynamic and kinetic properties of both molecularly rough and faceted crystal-melt interfaces. A large range of aspects was mentioned, including the capillary fluctuation method, dendrite growth and structure selection in crystal nucleation from the melt, thermodynamic properties of

steps on faceted crystal-melt interfaces and their mobilities. Animated examples for Fe, Mg, Si as well as other materials were shown.

Stefan Müller considered *ab initio* concepts for alloy properties, which possess a delicate temperature-dependence like short-range order or precipitate evolution. For this purpose he used the combination of DFT calculations with methods from statistical physics, namely *Cluster Expansions (CE)* and *(Kinetic-) Monte-Carlo (MC) simulations*. The mixed-space presentation for the treatment of long-range interactions and a new surface cluster-expansion allowing for an efficient selection and reduction of relevant interactions was explained.

Andrei V. Ruban presented his calculations of effective interactions in bcc Fe-Cr and fcc Fe-Cr-Ni alloys for different alloy compositions. He used the *screened generalized perturbation method (SGPM)* in conjunction with the *KKR-ASA-CPA method*. It was shown that magnetism plays crucial role in the ordering behavior of these alloys. Interactions were consequently used in the MC simulations of the atomic ordering effects and the results were compared with experimental data.

Peter Entel used first-principles calculations in order to obtain microscopic information of structural instabilities in transition metal compounds and alloys. These calculations allow to characterize martensitic trends associated with *anharmonicities and phonon softening, magnetic ordering, electronic and chemical features*. At finite temperatures, simulation of reactions of solids, thin films and nanoparticles were presented with the help of *optimized interatomic potentials*.

Pavel Korzhavyi presented various examples of theoretically derived composition-structure-property relations for several industrially relevant alloy phases, which include Fe-Ni-Cr austenite phase, oxi-carbo-nitrides of titanium, and iron-based sigma phases. He showed that *ab initio calculations* have reached the level at which they can provide quantitatively accurate description of the energetics of these phases as a function of their structure, composition, atomic and magnetic order and can be used for the *CALPHAD approach*.

Helmut Eschrig pointed out that the phase equilibrium under external fields (as stress or electromagnetic one) is preferably described by the *Free Enthalpy*, whereas the *Free Energy* is more easily obtained from total energy calculations. The interrelations between both were analyzed beyond linear response, and examples were discussed. Besides general elasticity, this included epitaxial paths as well as a fixed magnetic moment approach for invar and magnetostriction.

François Willaime studied from first principles defects and defect clusters in iron. He presented the methodological issues of calculations based on the *SIESTA code*, which changed in particular the energetic landscape of interstitial-type defects, as compared to what was known before from empirical potentials. The results were used as input data for the *simulation of the kinetic evolution* of radiation induced defects. This multi-scale approach allows reaching macroscopic time and length

scales. The validity of empirical potentials was improved by including ab initio results in the database of fitted properties.

Matt Probert introduced a *novel genetic algorithm* for atomistic structure determination. It has been implemented in the ab initio DFT code CASTEP, where it can be used for the prediction of crystal structures from first principles, and in a simple pair potential code for algorithm development and testing. Results from both codes were presented, showing how the algorithm performs for a number of difficult problems, and how to best choose the various controlling parameters for optimal performance.

Peter E. Blöchl's talk was devoted to the growth of atomically perfect, epitaxial oxide films on semiconductors as done using molecular beam epitaxy. In order to guide experiment he performed ab initio calculations using *ab initio molecular dynamics* and the *projector augmented wave method*. He investigated and discussed the atomic processes leading to perfect interfaces of semiconductors with high-K oxides.

Britta Nestler presented a recently developed *non-isothermal phase-field model* for alloys with multiple components and phases. The model has been derived from an entropy functional in a thermodynamically consistent way and allows to model arbitrary phase diagrams. She showed solidification microstructures on different time and length scales leading to the formation of dendritic and eutectic grain structures and the subsequent process of grain growth.

Christopher Wolverton described the role of atomic-scale first-principles computations in the Integrated Computational Materials Science methodology applied to Al castings at Ford. He focused on coupling *first-principles methods* with other computational approaches such as *phase-field microstructural evolution models*, computational thermodynamics or *CALPHAD methods*, *cluster expansion methods* and *kinetic Monte Carlo*. Applications were shown to prediction of precipitation, microstructure evolution, and ultimately yield strength during heat treatment.

Since all participants are currently doing research in the field defined by the scope of the workshop, there were extensive discussions after each lecture. Experienced researchers had ample time to debate different approaches. Junior researchers had plenty of opportunities to improve their understanding with specific questions.

In addition to the tutorial-like lectures several participants presented results of their research in 30minute talks and a poster session. These parts of the workshop were also characterized by vivid discussions and exchanges of ideas. In particular junior researchers had a chance to get a feedback of their research.

3. Impact of the event

A main achievement of the workshop ADIS2006 was to provide a platform for bringing together distinguished experts in ab-initio calculations and thermodynamic/kinetic modeling of metals and to provide an opportunity for exchange of knowledge and for fruitful scientific discussions. In the following a few highlights of the conference:

- There was consensus that the computational methods of density functional theory (DFT) have now reached the level at which they can provide a quantitatively accurate description of the energetics of metals and metallic alloy phases as a function of their atomic and magnetic order, composition and (defect) structure.
- For the simulation of binary and ternary alloys two classes of highly successful approaches have been discussed: The screened generalized perturbation method (SGPM) in conjunction with the KKR-ASA-CPA and the cluster expansion technique.
- The participants agreed that the current challenge of ab initio calculations is an extension towards the description of finite temperatures. A variety of approaches, including statistical mechanics, kinetic Monte Carlo and molecular dynamics, have been suggested and discussed at the workshop. Some participants reported that these approaches are already in a stage that they are relevant for industrial materials design. Nevertheless, a joint effort is still necessary to improve and validate these methods.
- Several participants have voted for a collection of electronic data obtained with ab initio calculations for various compounds and alloys. Such data bases would be a valuable complement to existing empirical data used for CALPHAD.
- Many participants that worked in different communities did not know each other before they came to Ringberg. Contacts have been established and people agreed to stay in contact after the workshop.

We, the organizers of the workshop, received a very positive feedback from the participants. There was a strong wish to repeat this meeting regularly (every two years) in order to exchange the latest results, and identify new trends and developments. We like to thank the Psi-k network in the framework of the ESF for financial support of the workshop ADIS2006.



4. Final programme

Monday, February 20 (Ab initio Thermodynamics)

Chair: Jörg Neugebauer	8:00 - 8:45	Breakfast
	8:45 - 9:00	Opening
	9:00 - 10:30	Dario Alfe (University College London, UK): <i>Thermodynamics of iron at high pressure and temperature from first principles calculations</i>
Chair: Jörg Neugebauer	10:45 - 12:15	Frank Lechermann (École Polytechnique, France): <i>Ab initio thermodynamics for alloys: The ternary system Ni-Fe-Al as test case</i>
	12:30	Lunch
	14:30 - 16:00	Mojmir Sob (Masaryk University in Brno, Czech Republic): <i>Ab initio studies of iron magnetism at high shape and volume deformations and predictions of structure of superaustenitic steels</i>
Chair: Martin Stratmann	16:30 - 18:00	Gerbrand Ceder (MIT, USA): <i>New approaches to the ab initio prediction of phases diagrams and crystal structure in metals and alloys</i>
	18:30 - 20:00	Dinner
	20:00 - 21:30	Mark Asta (University of California, USA): <i>Atomic-scale simulations of crystal-melt interface properties for multiscale modeling of solidification microstructure</i>

Tuesday, February 21 (Applications)

Chair: Martin Friak	8:00 - 9:00	Breakfast
	9:00 - 10:30	Stefan Müller (University Erlangen-Nürnberg, Germany): <i>Density functional theory meets statistical physics: From the local atomic structure to the mesoscopic properties of metal alloys</i>
	10:45 - 12:15	Andrei V. Ruban (Royal Institute of Technology, Sweden): <i>Magnetism, effective interactions and atomic ordering effects in steels</i>

Tuesday, February 21 (continuation)

	12:30	Lunch
Chair: Tilmann Hickel	14:30 - 16:00	Peter Entel (University Duisburg-Essen, Germany): <i>First-principles investigations of structural transformations in magnetic solids, thin films and nanoparticles</i>
	16:30 - 18:00	Pavel Korzhavyi (Royal Institute of Technology, Sweden): <i>Theoretical design of hard materials</i>
	18:30 - 20:00	Dinner
	20:00 - 20:30	Blazej Grabowski (MPIE Düsseldorf, Germany): <i>Ab initio calculation of thermo dynamic properties of metals: xc-related error bars and chemical trends</i>
	20:30 - 21:00	Hans L. Skriver (Technical University of Denmark): <i>Stability of nitrided stainless steel</i>
	21:00 - 21:30	Martin Friak (MPIE Düsseldorf, Germany): <i>Anomalous equilibrium volume change of magnetic Fe-Al crystals</i>

Wednesday, February 22 (Fundamentals)

	8:00 - 9:00	Breakfast
Chair: Mark Lusk	9:00 - 10:30	Helmut Eschrig (IFW Dresden, Germany): <i>General theory of phases of single crystals under stress</i>
	10:45 - 12:15	François Willaime (CEA/Saclay, France): <i>Defects in iron from first principles: structure, stability and mobility</i>
	12:30 - 13:15	Lunch
	13:15 - 18:30	Excursion: Technical Museum Munich
	18:30 - 21:00	Dinner in Munich

Thursday, February 23 (Methods and Surfaces)

	8:00 - 9:00	Breakfast
Chair: Gerd Ceder	9:00 - 10:30	Matt Probert (University of York, UK): <i>A novel genetic algorithm for atomistic structure determination</i>
	10:45 - 12:15	Peter Blöchl (Clausthal Technical University, Germany): <i>Towards computational materials design of oxide films</i>
	12:30	Lunch
	14:00 - 17:00	Excursion
Chair: Alexander Bluemenau	17:00 - 17:30	Adam Kiejna (University of Wroclaw, Poland): <i>First principles study of oxygen adsorption on iron surfaces</i>
	17:30 - 18:00	Duc Nguyen-Manh (UKAEA Abingdon, UK): <i>Nano-clustering and impurity-defect interaction in Fe-Cr alloys: an ab initio study</i>
	18:00 - 18:30	Liverios Lymperakis (MPIE Düsseldorf, Germany): <i>Ab initio based multiscale calculations of extended defects in condensed matter</i>
	18:30 - 19:00	Sergey Ostanin (University of Warwick, UK): <i>Ab initio study of the soft-magnetic properties of permalloys</i>
	19:00 - 21:00	Conference Dinner
	21:00 -	Poster Session

Friday, February 24 (Towards Meso-/Macroscale)

	8:00 - 9:00	Breakfast
Chair: Mark Asta	9:00 - 10:30	Britta Nestler (Karlsruhe University, Germany): <i>A thermodynamically consistent phase-field model for alloys with multiple components and phases</i>
	10:45 - 12:15	Christopher Wolverton (Ford Research Laboratory, USA): <i>What can we learn about precipitation in Al from first-principles?</i>
	12:30	Lunch