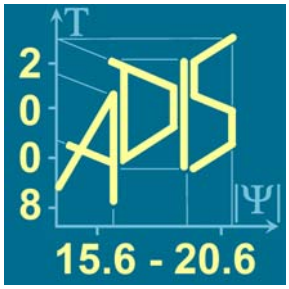
International Workshop on

Ab initio Description of Iron and Steel Magnetism and Phase Diagrams

- Final Report -



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1. Summary

This workshop has already been the second meeting in a series of workshops on “Ab initio description of Iron and Steel”. Ab initio based materials design is a quickly developing and very promising field of research. Nevertheless, its application in particular to steels has just started a few years ago and the workshop showed that there are still a lot of unresolved challenges. The workshop, therefore, served as a forum where internationally leading experts from different scientific communities (basic research, applied science, experiments) come together, presented their methods, exchanged the latest results, and identified future trends and developments. The success of this concept became particularly apparent in lively and fruitful discussions after each of the presented talks. 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 particitants the opportunity to discuss their latest results.

The scientific topic of the meeting, the ab initio description of magnetism in metals, is a topic of great relevance for iron and steels. Several speakers at the workshop have stressed the influence of the specific ordering of magnetic moments (ferro-/antiferro-/paramagnetic) on the phase stabilities and transitions in steels. In some of the contributions also the dependence of the thermodynamic properties of iron and steel on magnetic entropy contributions has been addressed. In the sections devoted to fundamental aspects of magnetism, sophisticated methodological investigations, including model Hamiltonians, spin dynamics, spin-fluctuations etc., have been present. It turned out that a lot of progress has recently been achieved in describing the temperature dependence of magnetic quantities, in developing magnetic interaction potentials and in treating disordered magnetic systems. Therefore, several speakers have already applied some of these concepts successfully to practical questions of materials design, ranging from bulk properties up to grain boundaries and surfaces. For iron-based materials the influence of carbon, but also of other alloying elements such as Mn, Cr, Co, Ni or Si, has been of particular interest and has been intensively discussed.

2. Description of the scientific content

The main contributions to the international workshop ADIS2008 were the tutorial-like lectures of the invited speakers. The large number (17) 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 (60min+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:

Lars Nordström started the course with giving a very informative overview over various aspects of magnetism in Fe-based compounds. He particularly focused on the description of non-constrained magnetization densities within accurate full-potential calculations. Secondly, he discussed the magnetic anisotropy of different materials. Thirdly, he covered several aspects on the magnetic behaviour at elevated temperature, through the concept of spin dynamics. Results for various simulations were presented on atomic scale spin dynamics, where the spin degree of freedom has been mapped from DFT calculations on to effective spin models.

Wolfgang Nolting gave an overview on how the temperature dependence of magnetic properties can be simulated by means of many-body theory. He pointed out that ferromagnets can roughly be divided into two different classes, the band ferro-magnets and the local-moment ferromagnets, and discussed appropriate models for both classes. He presented many-body approaches to these models which had been connected with band structure calculations to study prototypes of the mentioned classes, Fe and Ni as band ferromagnets and Gd, GdN and EuO as local-moment metals. He also commented on the double-counting problem.

Malcolm Stocks reviewed in his presentation some of his studies of the magnetic structure of some Fe-based alloys and nanostructures. The calculations were based on implementation of relativistic density functional theory within state of the art order-N multiple scattering Green's function methods. Results were presented for some interesting nanostructures consisting of Fe-chains and impurities in Cu as well as FeNi, FeMn, and FeCu alloys.

Jürgen Kübler has spoken on magnetism of metals at finite temperatures as described within the local-spin-density functional approximation (LSDA). The essential ingredients to his thermodynamic theory were orientational fluctuations of the local magnetization for which an adiabatic principle was postulated. Moriya's unified theory was used to derive the spherical approximation (SA) for the description of magnetic phase transitions. He presented a large range of applications of the SA including ferromagnets, iron, inter-metallic compounds, Heusler compounds etc.

Jeff Lynn discussed the topics of the workshop from an experimental point of view. He has mainly spoken on his longstanding interest in the properties of isotropic ferromagnets, beginning with the spin dynamics of the band magnets Fe and Ni. He

discussed the interaction of the magnons with the so-called Stoner continuum, and the nature of the collective excitations in the paramagnetic phase. In the second part of his talk he has spoken about experimental results for amorphous magnets and Invar systems (Fe-Ni, Fe-Pt, amorphous Fe-B). The highlight of the talk was the part on iron-based high-T_C superconductors.

Mehmet Acet was the second experimentalist in the list of invited speakers. He has spoken about the stabilization of bcc (ferritic) and fcc (austenitic) structures in steels by short range ferromagnetic correlations. He reported their X-ray emission spectroscopy data obtained under high-pressure (30 GPa) and high-temperature (1400 K) conditions, which demonstrate that a magnetic instability in the form of a high-spin to low-spin transition is present in fcc-Fe in the temperature range of its stability (1184 - 1665 K). This observation was mentioned in the context of the anti-Invar effect. Furthermore, the differences in the Gibbs free energies between the various crystallographic phases of Fe and its alloys were discussed.

Graeme Ackland presented in his talk his recent developments for dealing with the FeCrC system. Using the second-moment tight binding (Finnis-Sinclair) picture he explained why to a first approximation magnetism is automatically covered in the formalism, and why this fortuitous feature does not apply to FeCr. He further discussed how Fe-C interactions can be incorporated into the same embedded-atom framework. In the last part he has spoken about the anomalous solubility of Cr in Fe.

Mike Finnis presented DFT and self-consistent tight-binding (TB) approaches to describe interstitials and magnetism in Fe-rich Fe-Cr alloys. The TB method starts by expanding the exchange and correlation energy functional to second order in the deviation of the electron density from the initial guess. Finnis showed in a very instructive way how this can be extended straightforwardly to the case of spin-polarised electrons, giving a self-consistent tight-binding description of magnetism.

Ralf Drautz' presentation was based on his recently derived analytic interatomic bond-order potential (BOP) that depends explicitly on the valence of the transition metal element. He showed that this potential predicts the structural trend from hcp to bcc to hcp to fcc observed across the non-magnetic 4*d* and 5*d* transition metal series. In the second part he showed how this BOP may be extended to include magnetic contributions to the binding energy. The resulting magnetic potential describes the experimental trend from antiferromagnetism to ferromagnetism across the 3*d* transition metal series.

Sergei Dudarev has chosen the effect of magnetism on interatomic interactions in iron, and on the structure and thermally activated mobility of radiation defects in iron and iron-based alloys as the topic of his talk. He reviewed ab-initio studies of these radiation defects and new parameterisations of conventional as well as "magnetic" semi-empirical many-body interatomic potentials. Furthermore, he described recent applications of these methods to the interpretation of experimental data on the high-temperature mechanical properties of iron and steels.

Reinhard Singer devoted his talk to the atomistic modeling of the adiabatic magnetic energy landscape. He first showed calculations of the adiabatic magnetic energy for selected reference configurations by means of constrained SDFT. Therein, atomic Lagrange constraint fields were applied. It has been argued that the spin-cluster expansion (SCE) technique is an appropriate functional model to determine the energy landscape. Singer showed how the construction of an SCE model of bulk fcc-Fe as obtained by the full-potential ASW code is performed practically. Afterwards, the application of the obtained model to a systematic search for magnetic groundstate candidate configurations was presented.

Christian Elsässer presented first-principles calculations and atomistic simulations of properties of grain and phase boundaries in transition metals. He addressed the computational atomic-scale simulation of bcc transition metals on the basis of DFT, TB and BOPs. The capabilities and limitations were illustrated for selected case studies, incl. bulk structures and energetics of non- and ferromagnetic crystals; adiabatic potentials and vibrational spectra of interstitial hydrogen isotopes; structure, bonding, and stability of grain boundaries; effects of segregated impurities at grain boundaries, metal/metal and metal/carbide phase boundaries; and interactions of lattice dislocations with grain boundaries.

Stefan Blügel investigated the interrelation of magnetism and stability of ultrathin magnetic films. He came to the conclusion that under certain circumstances there should exist magnetically stabilized two-dimensional surface alloys. Blügel presented *ab initio* results for the formation energy and the interdiffusion energy of Cu(001)c(2×2)3*d* surface alloy films, which indicate the existence of a thermodynamically stable surface alloy. He pointed out that several of these surface alloys have recently been found experimentally. He discussed this in the context of large magneto-volume effects.

Göran Grimvall addressed different thermophysical properties of iron and steels and the role of ab initio calculations for their description. He showed that the average phonon frequency, obtaind from DFT, and the related vibrational entropy show a remarkably smooth variation in Fe as the temperature is increased, in spite of structural changes from bcc to fcc, and in spite of the associated changes in the magnetic properties. He also investigated the electrical and thermal conductivities of bcc and fcc Fe and various steels. He explained its variation as a function of temperature and composition on the basis of saturation effects in the electron mean free path.

Peter Entel described the magnetism in Fe-Mn, Co-Mn and Fe-Pt nanoclusters, compounds and alloys. He had investigated ordered $Fe_{(100-x)}Mn_x$ alloys in view of their structural and magnetic stability by using the FLAPW method. He calculated mixing energies and determined the martensitic transition temperature of different compositions. He also pointed out that Fe-Mn alloys exhibit a double magneto-volume instability, i.e. they show Invar at low temperatures and anti-Invar behavior at elevated temperatures. Co-Mn and Fe-Pt were mainly investigated because of their unusual cluster properties in comparison to bulk.

Andrei Ruban demonstrated in his talk the influence of the local and global magnetic state on the effective chemical interactions on examples of several different Fe alloys. He pointed out that there are several techniques for calculating effective cluster interactions from first principles, but that the complex magnetic state makes problems for their application. Ruban presented a simple model for the strain induced interactions in random alloys.

Igor Abrikosov gave the last of the invited talks. He showed that magnetic and chemical interaction in itinerant alloy systems are deeply interconnected, and strongly affect each other. Starting with relatively simple examples, he showed that there exists a very strong dependence of thermodynamic properties, like elastic constants, structural distortions, and mixing enthalpies, on the underlying magnetic state in Fe alloys with Cr, Mn and Ni. He then illustrated the very strong effect of the underlying magnetic state on the interactions and phase stability in Fe-Si alloys.

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 used the many 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.

The participants of the workshop ADIS2008.

3. Impact of the event

A main achievement of the workshop ADIS2008 was to provide a platform for bringing together distinguished experts in the ab-initio simulation of magnetism and thermodynamic 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 a level at which they can provide an accurate description of the magnetism of simple metals as a function of temperature and magnetic order.
- Nevertheless, it turned that most of the concepts presented at the workshop are optimized for certain elements and compounds. The form of the DFT functional has an influence on the results. Furthermore, there is a large diversity of DFT codes with respect to the treatment of magnetism. A general theory/code covering all relevant aspects, such as constraint magnetic moments, quantum mechanical effects at low temperatures, spin-fluctations, and paramagnetic contributions is still missing.
- It turned out that the biggest challenge for further research is the simulation of the combined effects of magnetism and other thermodynamic excitations (lattice vibrations, electronic excitations and configurational entropy). Some of the presentations have revealed that the development is currently going in this direction.
- There has been a substantial progress within the last years in constructing interatomic potentials, which have magnetic properties incorporated. However, since these activities are limited to a few elements and have their limitations in predicting the properties of compounds, a lot of work still needs to be done in this field.
- Many participants who 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.

Similarly to the workshop ADIS2006 we again received a very positive feedback from the participants. We have, therefore, decided to establish this ADIS workshops now as a series (every two years) and provide in this way the opportunity to regularly exchange the latest results, and identify new trends and developments. We like to thank again the Psi-k network in the framework of the ESF for financial support of the workshop ADIS2008.

4. Final programme

Monday, June 16

	8:00	Breakfast
	8:45	Opening
Fundamentals of magnetism *Chair:* **Tilmann Hickel**	9:15 - 10:15	**Lars Nordström** (Uppsala University, Sweden): *Various aspects of magnetism in Fe-based compounds; non-collinear magnetism, magnetic anisotropy and spin dynamics*
	10:35	Coffee Break
	11:00 - 12:00	**Wolfgang Nolting** (Humboldt-Universität zu Berlin, Germany): *Ferromagnetism in correlated particle systems*
	12:15	Lunch
	15:00 - 16:00	**G. Malcolm Stocks** (Oak Ridge National Laboratory, USA): *Spin dynamics in iron based alloys and nanostructures*
	16:15	Coffee Break
Finite temperature magn. *Chair:* **Peter Entel**	16:40 - 17:40	**Jürgen Kübler** (Technische Universität Darmstadt, Germany): *Magnetism of metals at finite temperatures: density functional studies and beyond*
	17:55 - 18:25	**Fritz Körmann** (MPIE Düsseldorf, Germany): *The free energy of bcc iron: Integrated ab initio derivation of vibrational, electronic, and magnetic contributions*
	18:30	Dinner
	20:00	Poster session

Tuesday, June 17

	8:00	Breakfast
Experimental magnetism *Chair:* **Ingo Steinbach**	8:45 - 9:45	**Jeffrey Lynn** (NIST Center for Neutron Research, USA): *Highly correlated electron systems - half a century of progress in isotropic ferromagnets*
	10:00 - 10:30	**Mehmet Acet** (Universität Duisburg-Essen, Germany): *Short range ferromagnetic correlations as the stabilizing agent of bcc (ferritic) and fcc (austenitic) structures in steels*
	10:35	Coffee break
Tight binding and empirical potentials *Chair:* **Jörg Neugebauer**	11:00 - 12:00	**Graeme Ackland** (University of Edinburgh, UK): *Lattice models and interatomic potentials: How much physics do we need for an atomistic description of steel?*
	12:15	Lunch
	14:15 - 15:15	**Mike Finnis** (Imperial College London, UK): *Interstitials and magnetism in Fe-rich Fe-Cr alloys; DFT and self-consistent tight-binding approaches*
	15:30	Coffee break
	16:00 - 17:00	**Ralf Drautz** (ICAMS Bochum, Germany): *Development of analytic bond-order potentials including magnetism*
	17:15 - 18:15	**Sergei Dudarev** (Imperial College London, UK): *The effect of magnetism on interatomic interactions in iron*
	18:30	Dinner
Cluster expansion *Chair:* **Ralf Drautz**	20:00 - 20:30	**Reinhard Singer** (MPI-MF Stuttgart, Germany): *Atomistic modeling of the adiabatic magnetic energy landscape: constrained SDFT and spin-cluster expansion*
	21:10 - 21:40	**Stefan Müller** (Universität Erlangen-Nürnberg, Germany): *Segregation induced surface magnetism by defects in intermetallic compounds*

Wednesday, June 18

	8:00	Breakfast
Boundaries and surfaces *Chair:* **Stefan Müller**	8:45 - 9:45	**Christian Elsässer** (Fraunhofer-Institut für Werkstoff-mechanik, Freiburg, Germany): *First-principles calculation and atomistic simulation of properties of grain and phase boundaries in transition metals*
	10:00 - 10:30	**Adam Kiejna** (University of Wrocław, Poland): *Cohesion and segregation at grain boundaries in iron and chromium*
	10:35	Coffee break
	11:00 - 12:00	**Stefan Blügel** (Forschungszentrum Jülich, Germany): *Magnetically stabilized surface alloys*
	12:15	Lunch
	13:00	Excursion: Wallberg
	18:30	Dinner
Effect of C in iron *Chair:* **Martin Friák**	20:00 - 20:30	**Chu-Chun Fu** (CEA Saclay, France): *Effects of carbon on defect properties in α-Fe from first principles*
	20:35 - 21:05	**Marcel Sluiter** (TU Delft, Netherlands): *Diffusion of interstitial and substitutional solutes in bcc Fe from electronic density functional calculations*
	21:10 - 21:40	**Lars Ismer** (MPIE, Düsseldorf, Germany): *Effect of interstitial carbon on the magnetic structure of fcc iron - Towards an ab-initio simulation of austenitic steels*

Thursday, June 19

	8:00	Breakfast
Ab initio thermodynamics *Chair:* **Igor Abrikosov**	8:45 - 9:45	**Göran Grimvall** (KTH Stockholm, Sweden): *Thermophysical properties of iron and steels - noteworthy features and the role of ab initio calculations*
	10:00 - 10:30	**Blazej Grabowski** (MPIE Düsseldorf, Germany): *Ab initio up to the melting point: Influence of vacancies and explicit anharmonicity on thermodynamic properties*
	10:35	Coffee break
	11:00 - 12:15	**Peter Entel** (University of Duisburg-Essen, Germany): *Magnetism in Fe-Mn, Co-Mn and Fe-Pt nanoclusters, compounds and alloys*
	12:15	Lunch
Physics of iron-based alloys *Chair:* **Jürgen Kübler**	15:00 - 16:15	**Andrei V. Ruban** (Royal Institute of Technology, Sweden): *Effective chemical, magnetic exchange and strain-induced interactionsin bcc and fcc Fe substitutional and interstitial alloys*
	16:15	Coffee break
	16:40 - 17:40	**Igor A. Abrikosov** (Linköping University, Sweden): *Magneto-structural coupling in itinerant systems*
	17:55 - 18:25	**Martin Friák** (MPIE, Düsseldorf, Germany): *Ab initio study of the α-iron stability limits*
	18:30	Conference dinner