MM 5: Computational Materials Modelling: Materials at finite temperatures

Time: Monday 10:15–11:30 Location: IFW B

MM 5.1 Mon 10:15 IFW B

Accurate electronic free energies of the transition metals at high temperatures — •Xi Zhang[1], Blazej Grabowski[1], Fritz Körmann[1,2], Christoph Freysoldt[1], and Jörg Neugebauer[1] — [1]Max-Planck-Institut für Eisenforschung GmbH, D-40237, Düsseldorf, Germany — [2]Department of Materials Science and Engineering, Delft University of Technology, Mekelweg 2, 2628 CD Delft, Netherlands

Free energies of bulk materials are nowadays routinely computed by density functional theory. In particular for metals, electronic excitations can significantly contribute to the free energy. Neglecting non-adiabatic contributions, electronic free energies can be obtained at relatively low computational cost, e.g., from the electronic density of states (DOS) at $T = 0$ K or by utilizing the Sommerfeld approximation. However, the actual error introduced by such approximations and the electron-phonon coupling effect at elevated temperatures is rarely known, even though it could affect, e.g., theoretical phase stability predictions. In order to clarify these issues we have computed the electronic free energies for all 3*d*, 4*d*, and 5*d* transition elements in the bcc, fcc, and hcp structures. For selected elements we have performed *ab initio* molecular dynamic simulations to investigate the electron-phonon coupling effect. We provide an analysis of the observed chemical trends in terms of the electronic DOS and the canonical *d* band model. The error in the approximate methods as well as the specific validity ranges for their application are clarified. The electronic heat capacities of all investigated elements as well as the error analysis within the Sommerfeld model are also presented.

MM 5.2 Mon 10:30 IFW B

Sampling of the magnetic structure in bcc and fcc iron using bond-order potentials — •Martin Staadt[1], Jutta Rogal[1], Tilmann Hickel[2], Thomas Hammerschmidt[1], Jörg Neugebauer[2], and Ralf Drautz[1] — [1]Interdisciplinary Centre for Advanced Materials Simulation, Ruhr-Universität Bochum, Germany — [2]Max-Planck-Institut für Eisenforschung, Düsseldorf, Germany

The phase transition between bcc and fcc iron is well known for its importance in manufacturing and processing of steels. We use a Metropolis Monte Carlo sampling algorithm with a non-collinear magnetic bond-order potential to sample the magnetic structure of bcc and fcc iron at various temperatures. Our approach includes simultaneous sampling of atomic and magnetic degrees of freedom and therefore implicitly accounts for magnon-phonon coupling. We show that we achieve a sensible description of the magnetic phase transformation and discuss the structural first order bcc-fcc phase transition. We will also present inital work on the prediction of temperature dependent local magnetic properties at grain boundaries and bcc-fcc interfaces.

MM 5.3 Mon 10:45 IFW B

Hot carriers Relaxation Dynamics in Hybrid Perovskites — •Mohamed El-Amine Madjet — Qatar Environment and Energy Research Institute(QEERI), Hamad bin Khalifa University(HBKU), Doha, Qatar

Hybrid organic-inorganic perovskites, such as methylammonium lead iodide CH3NH3PbI3, have attracted a lot of attention as promising new absorber materials for solar energy. Recently, perovskite-based solar cells reported a conversion efficiency of more than 22%. However, many fundamental processes related to the photophysics of these materials remain not fully understood. The excitation of electrons from the valence band to conduction band results in the formation of hot electrons and holes. Right after this photo-excitation an ultrafast thermalization of the hot carriers takes place driven by electron-phonon interaction.

A mixed quantum-classical approach based on trajectory surface hopping is used in order to investigate the process of non-radiative carrier relaxation in perovskite. Results will be presented for different perovskite materials depending on their structures and compositions and will discuss ways how the thermalization time could be enhanced.

MM 5.4 Mon 11:00 IFW B

Dynamical Material Parameters in Molecular Dynamics Simulations of Laser Ablation — •Johannes Roth[1], Eugen Eisfeld[1], Stefan Scharring[2], Marco Patrizio[3], Hans-Albert Eckel[2], and Mikhail Povarnitsyn[4] — [1]FMQ, Universit\"at Stuttgart — [2]DLR Stuttgart — [3]TU Darmstadt — [4]JIHT, RAS Moskau

Laser ablation is studied with classical molecular dynamics simulations. The interaction of the laser with the free electrons in the metallic material is modeled with a two-temperature model with separate temperatures for the electrons and the atoms. The properties of the electrons, heat capacity and conductivity and the electron-phonon-coupling as well as the reflectivity of the laser are model as constants for femto-second pulses since they do not change during the short time of interaction. This assumption is no longer valid for pico- and nano-second pulses. We have implemented wide-range models for the electronic parameters similar to the work of Povarnitsyn [1] which describe the transition from the metal to the plasma state. The reflectivity is calculated dynamically with the Fresnel equations and the absorption with the help of the Helmholtz equations instead of the simple Lambert-Beer law. The improved results are compared to simulations carried out by hydrodynamic simulations.

[1] Povarnitsyn et al., Appl. Surf. Sci \textbf{258} (2012) 9480.

MM 5.5 Mon 11:15 IFW B

Investigation of low temperature features in the heat capacity of metals — •Ankit Gupta[1], Bengü Tas Kavakbasi[2], Biswanath Dutta[1], Blazej Grabowski[1], Tilmann Hickel[1], Sergiy Divinski[2], Gerhard Wilde[2], and Jörg Neugebauer[1] — [1]Max-Planck-Institut für Eisenforschung, Düsseldorf, Germany — [2]Inst. of Materials Physics, Univ. of Münster, Münster, Germany

Heat capacities are often used to assess the predictive capability of computational thermodynamics, since they are experimentally directly accessible and as second derivatives very sensitive to energy changes. Finite-temperature first-principles calculations have accurately described the heat capacity of various material systems up to the melting point. In contrast, their assessment for the competition and coupling between the electronic and vibrational excitation mechanisms in the low-temperature regime is still in its infancy.

In this work, we explore this competition and coupling in the heat capacity C_p below 20 K for the example of Al and Al_3Sc. In order to enhance the sensitivity with respect to low temperature features, we analyze C_p/T^n for $n = 0, \ldots, 3$. We show that, density functional theory successfully resolves all the features observed in the recent experimental calorimetric data, including the low-temperature limit and localization effects in the heat capacity. The relevance of peaks in the phonon density-of-states beyond the Debye level and non-adiabatic electron-phonon coupling phenomena are evaluated. All mechanisms are found to be described accurately at the level of semi-local exchange-correlation functionals.