MM 39: Methods in Computational Materials Modelling V: Kinetics and Beyond DFT

Time: Wednesday 15:45–17:45 Location: H 0106

MM 39.1 Wed 15:45 H 0106

Exploring nucleation mechanisms in nickel: Novel insight from transition path sampling simulations. — •Grisell Diaz Leines, Ralf Drautz, and Jutta Rogal — Atomistic Modelling and Simulation, ICAMS, Ruhr-Universität Bochum, Universitätstr. 150, 44780 Bochum, Germany

Molecular dynamics simulations can provide valuable atomistic insight into solidification, but the modelling of the initial nucleation during solidification remains challenging due to the extended timescales of the process. Nowadays, advanced computational methods like transition path sampling (TPS) have enabled the investigation of nucleation on the atomistic level. In this work, we employ TPS to investigate the nucleation during solidification in nickel. We initially focus on homogeneous nucleation in elemental nickel as a function of undercooling. As a second step towards more complex materials, we extend our study by including small Ni-clusters as seeds during heterogeneous nucleation. The transition state ensemble obtained from our TPS simulations provides atomistic insight into the structure and size of critical nuclei for different nucleation mechanisms (homogeneous and heterogeneous nucleation at defects), as well as nucleation barriers/rates. Such results provide valuable information to validate and improve existing thermodynamic models describing nucleation. Furthermore, the information obtained about nucleation rates and the distribution of nucleation centres can also directly be connected to phase field models.

MM 39.2 Wed 16:00 H 0106

Annealing of coper nanoparticles on substrate by surface diffusion — •Raphael Schiedung[1], Ingo Steinbach[1], and Ulrich Köhler[2] — [1]ICAMS, Bochum, Deutschland — [2]Ruhr Universität Bochum, Institute for Experimental Physics, Surface Physics, Bochum

Copper nanoparticles on zinc-oxide substrates are discussed as materials for heterogeneous catalysis. An electric field gradient at the contact line between particles, substrate and gas is a possible mechanism for the chemical activity of otherwise only chemically interactive materials. In processing of the nanoparticles an annealing step is employed after deposition on the substrate at moderate temperatures. Experimental studies revealed, that the nanoparticles are sightly sucked into the substrate during annealing that a small crater remains after removing individual particles. In order to reveal the mechanism and the dynamics of this process a phase-field study has been performed. Surface diffusion is assumed to be the underlying physical process for the structural changes of the nanoparticles on the surface. The multiphase-field theory as implemented in the open-source software OpenPhase has been applied. It is based on the minimization of the free surface energies in the three phase system copper, zinc-oxide and gas. Results of the simulation are presented and compared to the experimental observations. This is a first step in theoretically investigating the multi-physics problem of deformation, piezoelectric activity of the substrate and electrostatic charging of the copper particles. Future work will concentrate the elastic and electric properties of the system in order to reveal the catalytic activity of the compound structure.

MM 39.3 Wed 16:15 H 0106

Simulation of fractal abnormal grain growth in nanocrystalline materials — •Mingyan Wang[1], Jules Dake[1], Rainer Birringer[2], and Carl Krill[1] — [1]Ulm University, Germany — [2]University of the Saarland, Germany

Despite an implicit rarity in its name, abnormal grain growth (AGG) appears to be a common mode of coarsening in nanocrystalline materials regardless of the specimen's composition or synthesis route. During AGG, a subpopulation of grains manifests rapid growth, leading to grain volumes that not only are much larger than those of their neighbors, but also are sometimes highly irregular in shape. The nature of this irregularity can be described by the geometric concept of fractals. This fractal morphology suggests that, in certain cases, AGG might proceed by some kind of percolation process along the "grid" defined by the initial ensemble of grains. We have investigated this possibility by extending a phase field algorithm for simulating grain growth to include selection rules for percolation. For properly chosen parameter values, the abnormal grains generated by simulation can be strikingly similar in shape to their experimental counterparts. Quantifying the comparison between simulation and experiment by fractal dimensionality, we hope to gain insight into at least one of the underlying physical mechanisms behind AGG in nanocrystalline materials.

MM 39.4 Wed 16:30 H 0106

On the Propagation of two en passent cracks upon mutual interaction: A pase field study — •Markus Thäter, Michael Fleck, Martin Lautenschläger, and Heike Emmerich — Materials and Process Simulation, University of Bayreuth, Germany

A phase field model for the simulation of crack propagation in brittle materials is applied to the problem of two mutually interacting "en passent" cracks. Thereby, crack growth is described as a first order phase transformation process, where the solid parent phase transforms into an infinitely weak *broken* phase, driven by elastic energy dissipation. We discuss the problem of "en passent" cracks in a two dimensional plain strain geometry, subjected to a constant uniaxial pulling velocity of mode I type. Our model reproduces a number of basic features that are also observed in corresponding experimental setups [1]: Initially, when the two cracks propagate independently, they approach each other along straight paths. Then, during the early stage of the mutual interaction and for certain geometrical circumstances the principle of local symmetry may even force the cracks to turn slightly away from each other. When the line connecting the two crack tips alines with the pulling direction, the two cracks curve towards each other upon mutual tip-tip interaction until each crack tip reaches the other*s crack tail, finally releasing a lenticular fragment. Here, we investigate the crack propagation dynamics as well as the chosen crack paths as a function of all relevant physical dependences.

15 min. break

MM 39.5 Wed 17:00 H 0106

Random phase approximation up to the melting point: The impact of anharmonicity and many-body effects on the thermodynamics of Au — Blazej Grabowski, •Stefan Wippermann, Albert Glensk, Tilmann Hickel, and Jörg Neugebauer — Max-Planck-Institut für Eisenforschung GmbH, D-40237 Düsseldorf, Germany

Triggered by the Materials Genome Initiative the development of *ab initio* based materials databases has gained immense momentum. A crucial requirement is that the databases reliably provide quantitatively accurate data. However, the application of generalized-gradient corrected functionals within standard density-functional theory results in a dramatic failure for e. g. the heat capacity of Au, leading to divergent thermodynamic properties well below the melting point.

Here we present an extension of the upsampled thermodynamic integration using Langevin dynamics (UP-TILD) technique to compute accurate finite temperature properties including many-body effects. We employ the random phase approximation (RPA) within the adiabatic connection fluctuation dissipation (ACFD) framework, showing that the inclusion of many-body effects leads to a stabilization and to an excellent agreement with experiment. The study demonstrates the capabilities of the RPA – previously confined to T=0K – at finite temperatures.

MM 39.6 Wed 17:15 H 0106

Correlation energy in the Adiabatic Connection Fluctuation-Dissipation Theory beyond RPA: Systematic development and simple approximations — •Nicola Colonna[1], Maria Hellgren[2], and Stefano de Gironcoli[3,4] — [1]Theory and Simulation of Materials (THEOS), École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland — [2]Physics and Materials Science, University of Luxembourg, L-1511 Luxembourg, Luxembourg — [3]International School for Advanced Studies (SISSA), I-34136 Trieste, Italy — [4]CNR-IOM Democritos, I-34136 Trieste, Italy

We present a systematic approach to compute accurate correlation energies in the adiabatic connection fluctuation dissipation (ACFD) framework beyond RPA. To first order our scheme leads to the RPA plus exact-exchange kernel (RPAx), for which a novel and efficient implementation is proposed. It is based on an eigenvalue decomposition of the time dependent response function of the Many Body system in the limit of vanishing coupling constant, evaluated by density functional perturbation theory. The accuracy of the RPAx approximation

has been tested calculating the correlation energy of the homogeneous electron gas and studying the dissociation energy curves of selected diatomic molecules. A sensible improvement of the total energy description is disturbed by a pathological behavior of the response function. Staying within an exact first-order approximation to the response function, we use an alternative resummation of the higher-order terms. This slight redefinition of RPAx fixes the instability in total energy calculations without compromising the overall accuracy of the approach.

MM 39.7 Wed 17:30 H 0106

Comparison between exact and semilocal exchange potentials: An all-electron study for solids — •Fabien Tran[1], Markus Betzinger[2], Peter Blaha[1], and Stefan Blügel[2] — [1]Institute of Materials Chemistry, Vienna University of Technology, Getreidemarkt 9/165-TC, A-1060 Vienna, Austria — [2]Peter-Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, D-52425 Jülich, Germany

The exact-exchange (EXX) potential, which is obtained by solving the optimized-effective potential (OEP) equation, is compared to various approximate semilocal exchange potentials in selected solids (C, Si, BN, MgO, Cu_2O, and NiO). This is done in the framework of the linearized augmented plane-wave method, which allows for a very accurate all-electron solution of electronic structure problems in solids. In order to assess the ability of the semilocal potentials to approximate the EXX-OEP, we considered the EXX total energy, electronic structure, electric-field gradient, and magnetic moment. An attempt to parameterize a semilocal exchange potential is also reported.