MM 52: Methods in Computational Materials Modelling (methodological aspecst, numerics)

Atomistic sampling techniques

Time: Thursday 10:15–11:30 Location: TC 006

MM 52.1 Thu 10:15 TC 006

Finding reaction coordinates for crystal nucleation in Ni — •Grisell Diaz Leines, Ralf Drautz, and Jutta Rogal — Ruhr-Universität Bochum, Universitätsstrasse 150, 44801 Bochum, Germany

Despite the technological importance of crystal nucleation, fundamental insight into the early stages of solidification is often hampered by the long timescales and complexity of the process. In this work we employ transition path sampling simulations together with a maximum likelihood analysis of the committor function to identify the reaction coordinates that best describe the nucleation mechanism in nickel. The analysis of the reweighted path ensemble on a set of candidate structural order parameters shows that the nucleation pathway in Ni is governed by the initial formation of mesocrystal regions and a subsequent emergence of fcc-hcp crystallites embedded within the core of these prestructured clusters. Our findings indicate that the prestructured liquid cloud is an order parameter that enhances the reaction coordinate and therefore has an essential role in the structural description of the nucleus and its interfacial free energy. Moreover, we show that the prestructured regions predetermine the coordination of the fcc-hcp polymorphs selected, acting as precursors of the crystallization. These results shed light not just on the atomistic mechanism of nucleation during solidification in Ni but also on the process of polymorph selection.

MM 52.2 Thu 10:30 TC 006

A Preconditioning scheme for Minimum Energy Path finding methods. — •Stela Makri[1], James Kermode[1], and Christoph Ortner[2] — [1]Warwick Centre for Predictive Modelling, School of Engineering, University of Warwick, Coventry, UK — [2]Mathematics Institute, University of Warwick, Coventry, UK

In transition state theory, the study of thermally activated transitions between energy minima is achieved by finding transition paths connecting the minima. These paths provide information on the energy barrier and reaction rates of the system without going through long and expensive simulations. To find them, current techniques use steepest descent-like minimisation to relax a discretised initial guess. However, steepest descent typically gives slow convergence rates in the presence of ill-conditioned potentials. In this talk I will be discussing how to reduce the condition number of the potential of an arbitrary system and improve the convergence speed and robustness of transition path finding methods, using a preconditioning scheme.

Our key assumption is that the cost of constructing a preconditioner is much smaller than the cost of computing the potential; for density functional theory the cost of single point evaluations is much more expensive than the computation of a preconditioner and thus the proposed approach improves computing times significantly. We have developed a local preconditioning scheme, where the preconditioner acts as a coordinate transformation on the discrete images along the path and a global preconditioning scheme is currently in development, in which the entire path is preconditioned as one entity.

MM 52.3 Thu 10:45 TC 006

One shot calculation of multicomponent phase diagrams with combined umbrella and nested sampling — •Robert Baldock[1], Christopher Sutton[2], Luca Ghiringhelli[2], and Nicola Marzari[1] — [1]Theory and Simulation of Materials (THEOS), and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), EPFL, Switzerland — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

The automated calculation of complete phase diagrams, directly from a first-principles or empirical potential energy function, is one of the outstanding challenges in computational materials science. Here we show how nested sampling, a Bayesian Markov chain Monte Carlo algorithm, can be transformed into a powerful tool for exactly this task. In particular, the introduction of umbrella sampling within nested sampling enables the efficient, one-shot calculation of composition-temperature phase diagrams, including for materials that exhibit a miscibility gap whereby the material separates into domains of different composition. Since our nested sampling algorithm does not require previous information about the location of phase transitions, or the atomic structures of phases formed by the material, it can be used as a black-box tool for phase diagram calculation. I will showcase the efficacy of the approach by presenting the binary phase diagrams of a Lennard-Jones alloy (continuous atomistic state space) and gallium indium phosphide as described using a lattice model (discrete atomistic state space).

MM 52.4 Thu 11:00 TC 006

Accurate free energies from *ab initio*: Applications of the TU-TILD technique — •Xi Zhang[1], Dominique Korbmacher[1], Lifang Zhu[1], Albert Glensk[1], Andrew Duff[3], Fritz Körmann[1,2], Blazej Grabowski[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 — [3]Scientific Computing Department, STFC Daresbury Laboratory, Hartree Centre, Warrington, UK

Calculations of free energies have been long pursued to predict and understand many important phenomena in materials, e.g., thermodynamic properties, defect properties, or phase transitions. Computing free energies with sufficient accuracy fully from *ab initio* is a demanding challenge requiring special methodological techniques. Based on density functional theory, the recently developed TU-TILD (*two-stage upsampled thermodynamic integration using Langevin dynamics*) [Phys. Rev. B 91, 214311 (2015)] technique provides a very efficient framework retaining the desired accuracy in the free energies, capturing in particular explicit anharmonicity. We show the efficiency and accuracy of the TU-TILD technique by focussing on stacking fault Gibbs energies. Successful extensions to point defect Gibbs energies, dynamically unstable phases, phonon lifetimes and liquid free energies within the TOR-TILD technique will be briefly highlighted.

MM 52.5 Thu 11:15 TC 006

Approach for ab initio simulations of materials under extreme conditions — •Jacob Wilkins and Matthew Probert — University of York, York, United Kingdom

To date, the ab initio exploration of materials in extreme conditions has been primarily focussed on equilibrium studies of heat and pressure to a stable crystal structure which fails to capture the methods used to produce these conditions in experiments. In order to attempt to more accurately reproduce the mechanisms present we have adopted a partially self-parametrising fire-and-forget approach for application to moderately sized systems to induce a series of independent shock-waves into a sample medium for testing shock response of the material.

Using the approach of the Hugoniostat, optimised for application to ab initio time-scales we apply a number of isolated shocks at stactic volumes and use a predictor-corrector algorithm to approach the target pressure in even steps. This method allows us to extract the material response to the compression in order to predict the ideal coupling, allowing the method to minimise convergence times even for extreme compressions.