MM 19: Development of Computational Methods: Simulation Methods – Theory

Time: Tuesday 14:15–15:30 Location: SCH A 251

MM 19.1 Tue 14:15 SCH A 251

Multi-methodological study of temperature trends in Mössbauer effect in beta-Sn — •Martin Friák[1], Nikolas Masničák[1], Oldřich Schneeweiss[1], Pavla Roupcová[1], Alena Michalcová[2], Šárka Msallamová[2], and Mojmír Šob[3,1] — [1]Institute of Physics of Materials, Czech Academy of Science, Brno, CZ — [2]Department of Metals and Corrosion Engineering, University of Chemistry and Technology in Prague, Prague, CZ — [3]Department of Chemistry, Faculty of Science, Masaryk University, Brno, CZ

We have performed a multi-methodological theoretical study of impact of thermal vibrations on the Mössbauer effect in the tetragonal beta-phase of tin. We have seamlessly combined (i) atomic-scale numerical data in the form of mean square displacements of Sn atoms determined by quantum-mechanical calculations, (ii) continuum-level thermodynamic modeling based on the quasi-harmonic approximation and (iii) theoretical analysis of Mössbauer effect resulting in the prediction of temperature dependence of Mössbauer factor. The computed results were compared with our Mössbauer and X-ray experimental data. We show that classical theoretical approaches based on simplistic Debye model of thermal vibrations of solids can be nowadays replaced by exact ab initio calculations of individual thermal vibrations. For details see M. Friák at al., Computational Materials Science 215 (2022) 111780, doi:10.1016/j.commatsci.2022.111780.

MM 19.2 Tue 14:30 SCH A 251

Increasing the Diversity of One-Sided Transition State Searches with Biasing Potentials — •Nils Gönnheimer, King Chun Lai, Karsten Reuter, and Johannes T. Margraf — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Chemical processes on catalytic surfaces are commonly described in terms of complex networks of chemical reactions. One-sided transition state (TS) searches are a powerful tool for exploring these networks, when not all reactants, products and intermediates are known. To this end, the dimer method is commonly used, since it avoids the computationally demanding calculation of second derivatives. However, a prominent disadvantage of the dimer method is the convergence to the same few TSs even with differently initialised simulations, which raises the possibility of overlooking reaction pathways. Here, we propose a method to overcome this limitation. Specifically, we use a biasing potential to modify the potential energy surface (PES) based on information gathered from previous calculations. This biasing potential is adaptively generated in a series of dimer searches to drive the searching algorithm away from already known TSs. Intuitively, such potentials can be built within the Cartesian space by placing an energy bias at the atomic positions of a found TS. Alternatively, a similarity kernel between the structure and the known TS based on a local environment descriptor can be employed. This ensures that the biasing potential is invariant to permutations between identical atoms, as well as rotations and translations. The performance of both approaches is discussed for adatom diffusion and island formation on the Pd(100) surface.

MM 19.3 Tue 14:45 SCH A 251

How to Speed up First-Principles Based Geometry Optimization with Small Numerical Basis Sets — •Elisabeth Keller, Johannes T. Margraf, and Karsten Reuter — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

First-principles based geometry optimizations are often the most expensive part of high-throughput virtual screening studies for functional materials. This is particularly true for large systems, i.e. when studying complex surface reconstructions or nanoparticles. Here, the computational cost is strongly influenced by the size of the basis set. Large, converged basis sets result in precise equilibrium geometries, yet demand high computational cost and thus limit the simulation scale. Semiemprical methods using minimal basis sets offer a much lower computational cost, but may yield unacceptably large and uncontrolled errors. Furthermore, the availability of adequate parameterizations is rather sparse across the periodic table.

In this presentation, we will discuss the potential of using near-minimal basis sets for accelerating and enabling large-scale geometry optimizations at the DFT level. For this purpose, we studied how the size of the numeric atom-centered orbital (NAO) basis set in FHI-aims impacts the accuracy of bulk geometries. We recover equilibrium geometries at a nearly converged level with a highly compact basis by employing a simple short-ranged pair-potential correction. We show the scheme's ability to treat different systems across the periodic table ranging from small molecules and clusters to large-scale bulk and surface structures.

MM 19.4 Tue 15:00 SCH A 251

Automated Wannierizations — •Junfeng Qiao[1,2], Giovanni Pizzi[1,2,3], and Nicola Marzari[1,2,3] — [1]THEOS, EPFL — [2]NCCR MARVEL — [3]LMS, Paul Scherrer Institut

Maximally localized Wannier functions (MLWFs) are widely used in computational condensed matter physics. The standard approach to construct MLWFs often requires initial guesses which are based on chemical intuition and some measure of trial and error. Here, we first introduce an algorithm based on "projectability disentanglement" that provides reliably and automatically atom-centered Wannier functions describing both occupied and empty states. Then, we show how to mix these again into target subspaces; e.g., to describe valence and conduction bands separately. We test these algorithms on 200 representative materials (77 insulators), showing that the final MLWFs are very well localized and can accurately interpolate band structures at the meV scale. Such approaches enable automated Wannierizations for both metals and insulators, promoting further applications of MLWFs in physical applications and high-throughput calculations.

MM 19.5 Tue 15:15 SCH A 251

Higher-order finite difference method for accurate calculation of Wannier centers and position matrix elements — •Minsu Ghim[1,2,3], Jae-Mo Lihm[1,2,3], and Cheol-Hwan Park[1,2,3] — [1]Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea — [2]Center for Correlated Electron Systems, Institute for Basic Science, Seoul 08826, Korea — [3]Center for Theoretical Physics, Seoul National University, Seoul 08826, Korea

An accurate calculation of Wannier function centers and position matrix elements is crucial for calculating many physical quantities such as the electric polarization and optical responses from first-principles Wannier interpolation. Since the Fourier transform of the position operator is the gradient in momentum space, a finite difference technique is used to calculate the position matrix elements for Wannier functions. However, so far, only the first-order finite difference has been considered therein. We present a higher-order finite difference method and demonstrate the advantage of our method over the conventional first-order finite difference method.