MM 5: Materials for Storage and Conversion of Energy I

Time: Monday 10:15–11:30 Location: C 264

MM 5.1 Mon 10:15 C 264

"Watt's Up" with ^{7}Li: Computing NMR Parameters in Battery Materials Using an All-Electron DFT QM/MM approach — •Federico Civaia[1], Simone S. Köcher[2], Karsten Reuter[1], and Christoph Scheurer[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]Institute of Energy and Climate Research (IEK-9), Forschungszentrum Jülich GmbH, Jülich, Germany

Solid-state electrolytes are crucial in lithium-ion battery research, because of the pressing need for safe and durable high-energy storage solutions. Understanding Li-ion dynamics in these materials is essential for developing improved battery materials. Owing to its non-destructive nature and sensitivity to atomic environments, solid-state nuclear magnetic resonance (SS-NMR) spectroscopy has become an invaluable tool for probing diverse Li-ion environments and investigating Li-ion mobility.

In order to facilitate interpretation of experimental Li SS-NMR spectra, we are setting up a new computational reference scale for a wide range of diamagnetic Li compounds. The efficient calculation of NMR parameters such as the electric field gradient tensor is enabled through the use of the linear-scaling *ab initio* all-electron density-functional theory code FermiONs++ [1] and a solid-state embedding method. The latter hybrid QM/MM (quantum mechanics/molecular mechanics) approach is validated against traditional periodic calculations of SS-NMR parameters for ideal reference systems.

[1] J. Kussman *et al.*, J. Chem. Phys. **138**, 134114 (2013); J. Chem. Theory Comput. **11**, 918 (2015).

MM 5.2 Mon 10:30 C 264

Correlated Motion of Li-Ions and Polarons in Disordered LTO — •Tavinder Singh[1,2] and Harald Oberhofer[1,2] — [1]University of Bayreuth — [2]Bavarian Center for Battery Technology

Lithium Ion Batteries are of great importance to our daily lives. Yet, especially with a view on the latter, low reliability and energy density of batteries hamper their adoption. This drives the search for new materials exhibiting favourable properties like high transport efficiencies, wider ranges of electrochemical stability, and, at least for the emerging class of all-solid-state batteries-zero strain on ion insertion for anode materials. In this respect, Lithium Titanium Oxide is a potential candidate exhibiting such properties. In the past, we have shown that in this material the electronic conductivity is due to the formation of polarons, which in turn might influence the movement of ions. Yet, a full study of the possibly correlated motion of both types of carriers is still lacking due to the combinatorial explosion of possible polaron and ion sites making it hard to compute necessary energies and barriers from first principles. Thus, we used a compressed sensing model based on Density Functional Theory energetics to increase the efficiency of our sampling and obtain a full description of conductivity and correlated motion in $Li_4Ti_5O_{12}$.

MM 5.3 Mon 10:45 C 264

High-throughput computational screening of fast Li-ion conductors for solid-state electrolytes — •Tushar Thakur[1], Loris Ercole[1], and Nicola Marzari[1,2] — [1]École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland — [2]Paul Scherrer Institut, Villigen, Switzerland

We present a high-throughput computational screening to find fast lithium-ion conductors to identify promising candidate materials for application in solid-state electrolytes. Starting with ~30,000 experimental structures sourced from the COD, ICSD and MPDS repositories, we perform highly automated calculations using AiiDA at the level of density functional theory (DFT) to identify electronic insulators. On these ~1000 structures, we use molecular dynamics (MD) to estimate lithium-ion diffusivities using the pinball model [1], which describes the potential energy landscape of diffusing lithium at near DFT accuracy while being 200-500 times faster. Then, we study the 50 most promising unknown fast conductors with full first-principles MD simulations. We present this entire screening protocol, including the workflows where the accuracy of the pinball model is improved self-consistently, necessary to automatically running the required calculations and analysing their results.

[1] L. Kahle, A. Marcolongo and N. Marzari, Modeling lithium-ion solid-state electrolytes with a pinball model. Phys. Rev. Mater. 2, 65405 (2018)

MM 5.4 Mon 11:00 C 264

Mechanical testing of LiCoO2 battery electrode and effect of de-/lithiation — •Robert Löser, Yug Joshi, and Guido Schmitz — Institut für Materialwissenschaft, Universität Stuttgart, Deutschland

The mechanical characteristics of lithium-ion cathode materials play a crucial role in determining key battery performance such as durability, cycle life, and safety. This study focuses on LiCoO2 (LCO), a widely used layered-structured cathode material for lithium-ion batteries, and investigates its mechanical properties during delithiation using sputtered thin films and nanoindentation techniques. The research reveals a significant decrease in Young's modulus from 275.9GPa to 243.4GPa upon lithium-ion deintercalation, attributed to alterations in bonding interactions. After subjecting the sample to extensive cycling, the decline in Young's modulus after the delithiation is reduced to 4.3% with a Young's modulus for the fully lithiated state of 255.5GPa. Additionally, this investigation identifies a linear increase in the proportion of elastic deformation with decreasing lithium content. Furthermore, the experiment in comparison to finite element method (FEM) simulations suggest the propagation of a Li0.5CoO2 phase from the top of the layer during delithiation. This work sheds light on the dynamic changes in mechanical properties of LCO, providing valuable insights for the design and optimization of lithium-ion batteries.

MM 5.5 Mon 11:15 C 264

Modelling LLZO Grain Boundaries with Amorphous Domains by Adaptively Trained Machine-Learning Interatomic Potentials — •Yuandong Wang, Yute Chan, Kyeonghyeon Nam, Karsten Reuter, and Christoph Scheurer — Fritz-Haber-Institut der MPG, Berlin, Germany

Garnet $Li_7La_3Zr_2O_{12}$ (LLZO) is a highly promising solid electrolyte for lithium batteries. However, its practical application faces challenges, primarily arising from Li dendrite formation and the impact of grain boundaries (GBs) on Li transport and stability. The amorphous LLZO exhibits desirable properties like blocking Li dendrite growth, high Li mobility and high electronic impedance. Control over amorphous domains between crystalline grains promises a method for tuning electrolyte performance. An atomistic understanding of the interplay between composition, structural characteristics and the properties of LLZO glass-ceramics is required for rational electrolyte and GBs design.

Machine Learning Interatomic Potentials (MLIPs) offer an accurate and efficient way to explore complex structures by enabling large-scale molecular dynamics (MD) simulations. In our study, we first establish a baseline MLIP using a dataset containing c-LLZO with varied Li configurations. Then an iterative training protocol employing simulated annealing to gradually produce amorphized GB motifs is utilized. The converged MLIP enable the construction of realistic models for amorphous LLZO and GBs in LLZO electrolytes, paving the way for their detailed exploration.