MM 8: Materials for the Storage and Conversion of Energy II

Time: Monday 15:45–18:30 Location: SCH/A216

MM 8.1 Mon 15:45 SCH/A216

Self-Consistent Hubbard Corrections for Accurate Modelling of Li- and Na-Ion Cathodes — •Valentina Sanella[1,2], Stefan Schären[1,3], Cristiano Malica[4], Livia Giordano[5], Nicola Marzari[1,3,4], Claude Ederer[2], and Iurii Timrov[1] — [1]PSI, Switzerland — [2]ETHZ, Switzerland — [3]EPFL, Switzerland — [4]U Bremen, Germany — [5]UniMiB, Italy

Accurate first-principles modelling of layered transition-metal oxides is essential for understanding and optimizing cathode materials for Li- and Na-ion batteries.

In this work, we investigate two widely studied systems: $LiCoO_2$ and P2-Na_xMnO_2, using density-functional theory (DFT) with self-consistent Hubbard corrections (DFT+U+V). We apply onsite U parameters to both transition-metal $3d$ orbitals and oxygen $2p$ states, together with intersite V derived from density-functional perturbation theory (DFPT), enabling a consistent treatment of electron localization and metal-oxygen hybridization.

For $LiCoO_2$, we show that including a U correction on oxygen significantly improves predictions of the intercalation voltages, resolving known inaccuracies associated with standard DFT approaches. In Na_xMnO_2, the extended Hubbard formalism provides a more realistic description across varying sodium concentrations, leading to reliable predictions of structural stability and electrochemical trends.

Our results demonstrate that self-consistent U and V parameters provide significant improvements over empirical or fixed values, enabling reliable predictions of structural and electronic properties.

MM 8.2 Mon 16:00 SCH/A216

Voltage Profile Predictions for Na-Ion Cathodes: A Hybrid MLIP / PBEsol+U+V approach — •Stefan Schären[1,2], Valentina Sanella[1,3], Manuel Dillenz[4], Nicola Marzari[2], and Iurii Timrov[1] — [1]PSI, Villigen, Switzerland — [2]EPFL, Lausanne, Switzerland — [3]ETHZ, Zürich, Switzerland — [4]DTU, Copenhagen, Denmark

Sodium-ion batteries are emerging as a sustainable alternative to lithium-ion batteries, but are limited by the lack of a cathode that offers high voltage, energy density and long-term stability. Here, we present a computational approach that accelerates the prediction of voltage profiles during cathode discharge, using a combination of self-consistent Hubbard-corrected DFT (PBEsol+U+V), and foundational machine-learning interatomic potentials (MLIPs). Using $Na_xV_2(PO_4)_3$ (NVP) as a test case, we show that MLIP-based prescreening reduces the number of required DFT calculations to a fraction of those needed for a traditional cluster expansion, while retaining comparable accuracy. Furthermore, the self-consistent Hubbard U and V corrections yield electronic occupations that closely match the chemical intuition for oxidation states, enabling a direct analysis of vanadium redox activity and associated volume changes during desodiation. Altogether, the approach delivers a useful and predictive toolkit for identifying high-performance Na-ion cathodes in the future.

MM 8.3 Mon 16:15 SCH/A216

Electronic structure of perovskite oxides calculated with dynamical mean field theory — •Daniel Mutter[1], Jannis Ehrlich[1], Frank Lechermann[2], Daniel F. Urban[1], and Christian Elsässer[1] — [1]Fraunhofer IWM, Freiburg — [2]Institut für Theoretische Physik III, Ruhr-Universität Bochum

Perovskite oxide materials containing first-row transition-metal (TM) elements are regarded as promising alternatives to platinum-based electrodes for high-temperature solid-oxide fuel and electrolyser cells. This is due to the strong electronic correlation effects of the localized 3d electrons of the TM, which directly influence the mechanism and activity of the oxygen reduction reaction. We analyze the electronic structure of perovskite oxides by means of a combined approach of density functional theory and the dynamical mean field theory (DFT+DMFT) to account for the correlation of the electrons in the TM-3d orbitals. We exemplarily demonstrate the influences of static versus dynamic correlation. In addition, we consider correction methods to treat the correlation in the O-2p orbitals, namely the self-interaction correction (SIC) or the DFT-1/2 method. A systematic study of different A- and B-site occupations of the ABO_3 perovskite phases will be presented.

MM 8.4 Mon 16:30 SCH/A216

Unraveling the connection between Jahn-Teller dynamics and charge transport in $LiMn_2O_4$ — •Manuel Dillenz, José Maria Castillo Robles, Mikkel Agerbo Essendrop, Ivano Eligio Castelli, and Juan Maria García Lastra — Department of Energy Storage and Conversion, Technical University of Denmark, Kgs. Lyngby, DK-2800, Denmark

Future energy storage solutions demand batteries that are energy-dense, fast-charging, and based on abundant materials. However, ultrafast dynamics in battery electrodes remain insufficiently characterized despite their crucial role in energy storage performance. Moreover, charge transport in Mn- or Ni-based cathodes is strongly influenced by local structural distortions originating from Jahn-Teller active centers. $LiMn_2O_4$ (LMO) exemplifies this behavior, featuring a ferrodistortive tetragonal ground state that undergoes an order-disorder transition at 290K, where pseudorotations continuously reorient the Jahn-Teller axis. These processes occur on timescales relevant for to polaron and ion transport in the electrode. By combining ab initio molecular dynamics (AIMD) simulations with nudged elastic band (NEB) calculations, we reveal that the interplay of JT and polaron dynamics governs the kinetics of charge transfer in LMO. Our work provides essential mechanistic understanding for enhancing charge transport in Jahn-Teller active electrode materials.

MM 8.5 Mon 16:45 SCH/A216

Defect Landscape of $Ba_2In_2O_5$: The Role of Oxygen Interstitials — •Rachele Sciotto and Karsten Albe — achgebiet Materialmodellierung, Institut für Materialwissenschaft, Technische Universität Darmstadt, Otto-Berndt-Str. 3, D-64287 Darmstadt, Germany

$Ba_2In_2O_5$ (BIO) is a promising n-type mixed ionic and electronic conductor (MIEC) for oxygen permeation membranes and solid oxide fuel cells (SOFCs). At ambient conditions, BIO adopts an orthorhombic brownmillerite structure with alternating InO_6 octahedral and InO_4 tetrahedral layers that can host interstitial oxygen atoms at sites of structural vacancies. Existing defect thermodynamics models consider only this type of oxygen interstitial; however, oxygen interstitials forming O-O dumbbells, found in oxides like ZnO and In_2O_3, are also possible. In this work, we investigate the potential existence of O-O dumbbells in BIO using density functional theory (DFT). By calculating formation enthalpies as a function of Fermi level and concentrations as a function of oxygen chemical potential, we identify two distinct classes of interstitial configurations. Neutral O-O dumbbells are stabilized over a wide Fermi-level range, while negatively charged interstitials at the structural oxygen vacancy site become favorable under n-type conditions. Although the neutral dumbbells do not contribute to charge compensation, they form in high concentrations. They could therefore serve as intermediate positions for oxygen diffusion pathways, thereby influencing the material's ionic and electronic conductivity.

15 min. break

MM 8.6 Mon 17:15 SCH/A216

Interplay of Dynamic Defects and Ultrafast Carrier Dynamics in Lead-Free Double Oxide Perovskites Toward Stable, High- Performance Photovoltaics — •Manasa Gattavadi Basavarajappa, Arijeet Sarangi, and Sudip Chakraborty — Harish-Chandra Research Institute, Chhatnag Road, Jhunsi, Prayagraj - 211019, India

Oxide-based double perovskites are gaining increasing attention as stable and environmentally benign alternatives to halide perovskites in photovoltaic and optoelectronic devices. Their structural versatility and chemical flexibility open pathways for fine-tuning both electronic and lattice properties, yet their carrier dynamics and defect behavior remain less understood. In this work, we investigate these aspects using a combination of first principles calculations and advanced simulation techniques. Non-adiabatic molecular dynamics is employed to capture ultrafast carrier relaxation and to quantify the role of electron*phonon coupling in determining charge transport and recombination lifetimes. The results highlight efficient charge separation and suppressed nonradiative recombination channels, which are essential for high photovoltaic efficiency. To establish the dynamical stability of the materials, we carry out a phonon-based symmetry analysis that confirms the absence of imaginary modes and reveals the interplay between structural distortions and vibrational spectra. In parallel, we explore the formation and dynamic evolution of intrinsic point defects through defect-mediated molecular dynamics simulations.

MM 8.7 Mon 17:30 SCH/A216

Morphological study of $\mathrm{LiNi_{0.5}Mn_{1.5}O_4}$ synthesized with alkali molybdates by density functional theory — •Liang-Yin Kuo[1], Tavinder Singh[1], Rio Akbar Yuwono[2], Fu-Ming Wang[2], and Harald Oberhofer[1] — [1]Chair for Theoretical Physics VII and Bavarian Center for Battery Technologies, University of Bayreuth — [2]Graduate Institute of Applied Science and Technology, National Taiwan University of Science and Technology

High-voltage spinel $\mathrm{LiNi_{0.5}Mn_{1.5}O_4}$ (LNMO) has emerged as a promising cathode active material for next-generation lithium-ion batteries due to its high operating voltage ($\sim$ 4.7 V vs. $\mathrm{Li/Li^+}$), manganese abundance, and potential for high energy density. Single-crystal (SC) LNMO has attracted particular interest because it offers enhanced structural integrity, reduced surface area for electrolyte interaction, and improved cycling stability. Among the synthesis methods, molten-salt synthesis has shown promise for producing SC LNMO. However, the role of the precursor salts remains unclear. In this study, the effects of alkali molybdates, specifically $\mathrm{Li_2MoO_4}$ and $\mathrm{Na_2MoO_4}$, on the SC LNMO formation mechanism are investigated by density functional theory (DFT). Surface energies of various facets are calculated to analyze the resulting Wulff shapes. Our results indicate that LNMO synthesized with $\mathrm{Li_2MoO_4}$ tends to form a truncated octahedron shape, whereas $\mathrm{Na_2MoO_4}$ leads to an octahedron morphology with aggregation into polycrystalline structures. Furthermore, the influence of surface energy on nucleation is discussed, providing insight into morphology control in SC LNMO synthesis.

MM 8.8 Mon 17:45 SCH/A216

Thermodynamic Stability of $\mathrm{MgSc_2Se_4}$ Surfaces — •Sebastian Utz[1] and Axel Gross[1,2] — [1]University Ulm, Ulm, Germany — [2]Helmholtz Institute Ulm, Ulm, Germany

$\mathrm{MgSc_2Se_4}$ represents a promising candidate for post-lithium battery technologies as a solid-state magnesium ion conductor. With magnesium being significantly more abundant and cheaper than lithium, magnesium-based materials could offer sustainable alternatives for next-generation energy storage systems. While the bulk properties and ion diffusion of $\mathrm{MgSc_2Se_4}$ have already been studied extensively, its surface structure and properties remain largely unexplored. However, understanding its surface behaviour is crucial as surfaces and interfaces can behave substantially differently from the bulk. In real applications, like batteries, the properties of surfaces and their interfaces may be limiting factors for stability and mass transfer. This work addresses this gap in the literature by investigating the low-index surfaces of $\mathrm{MgSc_2Se_4}$ using grand-canonical density functional theory. Our computational approach enables the characterisation of thermodynamic surface stability and structure under varying chemical potentials. The results provide fundamental insights into surface terminations and their stability in different chemical environments. Combined with experimental validation, these findings could enable detailed atomistic studies of mass transfer at and through $\mathrm{MgSc_2Se_4}$ surfaces, supporting the rational assesment of $\mathrm{MgSc_2Se_4}$ as a battery material.

MM 8.9 Mon 18:00 SCH/A216

LDH Under Stress: Assessing Degradation Pathways of Ni-Fe-V Catalysts Under Technical Operating Conditions — •Juan Manuel Lombardi, Charles Pare, Karsten Reuter, and Christoph Scheurer — Fritz-Haber-Institut der MPG, Berlin

The transition to a sustainable energy landscape relies on electrocatalysts that are not only active and affordable but also structurally robust under operating conditions. Ni-based layered double hydroxides (LDHs) doped with Fe and V are among the most promising OER candidates for anion-exchange membrane water electrolyzers (AEMWE), owing to their tunable lattice chemistry and rich redox behavior. However, the mechanistic influence of these dopants and how they reshape catalytic performance and structural stability remains insufficiently understood due to the immense configurational complexity.

In this contribution, we present an integrated strategy tailored to address this complexity directly. Our evolutionary exploration framework EZGA provides a systematic route through the high-dimensional composition-structure landscape, employing chemically informed operators and explicit diversity control to generate physically meaningful structural candidates. Machine-learning interatomic potentials (MLIPs) enable high-throughput sampling of thermally accessible configurations with near first-principles fidelity, revealing how dopants modulate stability and reactivity. Together, these elements deliver a predictive workflow that reveals how dopants reshape the accessible configurational landscape and provides a mechanistic picture of their influence on stability and reactivity.

MM 8.10 Mon 18:15 SCH/A216

Quantum-mechanical study of miscibility of Sn in $\mathrm{LaNi_5}$ — •Martin Friák[1], Petr Čípek[2,1], Anna Huňařová[2,1], Kateřina Dočkalová[2,1], Jana Pavlů[2,1], and Ondřej Zobač[1] — [1]Institute of Physics of Materials, v. v. i., Czech Academy of Sciences, Žižkova 22, Brno, 61600, Czech Republic — [2]Institute of Chemistry, Masaryk University in Brno, Kotlářská 2, Brno, 61137, Czech Republic

The $\mathrm{LaNi_5}$ compound is a prototypical hydrogen-storage material intended for future energy applications. Some of its properties have not been sufficiently studied so far, and some critically important data are missing. In particular, there are conflicting literature reports related to the substitution of Ni atoms with Sn atoms in $\mathrm{LaNi_5}$. We have employed quantum-mechanical calculations implementing the density functional theory (DFT) within the generalised gradient approximation (GGA) to determine the ground-state structural, electronic, thermodynamic, and vibrational properties of several substituted $\mathrm{La(Ni,Sn)_5}$ phases. Our computational cell containing 48 atoms allowed for simulating different Sn/Ni ratios as well as different configurations of substituting Sn atoms. Our results indicate that the free energy of formation is concave and there is a miscibility gap and a decomposition into two phases, one with a low and one with a high Sn concentration. Our theoretical study was complemented by experiments which found a similar miscibility gap, too.