

MM 6: Transport in Materials: Ion, Charge and Heat Transport

Time: Monday 10:15–13:00

Location: SCH A 118

MM 6.1 Mon 10:15 SCH A 118

Large-scale Atomistic and Quantum Mechanical Study of the Na⁺ Transport Mechanism in Sodium-ion Battery Electrolytes — ●AMAL KANTA GIRI¹ and HARALD OBERHOFER^{1,2} — ¹University of Bayreuth — ²TU Munich

A steady increase of the greener and safer energy sources have led to a rise in the need for energy storage technologies. In the last three decades, the lithium ion batteries (LIBs) dominated the global market from small to large scale storage of the energy. Yet, the shortage of the lithium and its localized geographical distribution raises concerns. New charge carriers for batteries beyond the lithium such as Na, is viable alternative for safe and large-scale energy storage, mainly due to its large abundance and high electrochemical potential.

Here, to improve our understanding of the Na⁺ transport mechanism, aggregation, and electrolyte performance in sodium ion batteries, we perform a theoretical investigation using a combination of all-atoms molecular dynamics (MD) simulations based on the OPLS forcefield and density functional theory calculations (DFT). In this regard, we simulate NaPF₆ salt in various organic electrolytes including ethylene carbonate (EC), propylene carbonate (PC), dimethoxyethane (DME), and dimethyl carbonate (DMC), and their binary mixtures at 320 K. Specifically, we focus on the conductivity, diffusivity and solvation of Na⁺ in the liquid electrolyte mediums. Furthermore, the solvation structure and the binding energy of ions in the electrolytes are thoroughly analyzed. We find the diffusivity of Na⁺ ions in the order DMC>DME>EC>PC, which follow the viscosity of the electrolytes.

MM 6.2 Mon 10:30 SCH A 118

Fast ion conduction in glassy and crystalline phases of Na₃PS₄: Insight from a machine-learning potential molecular dynamics study — ●YU-TAO LI, TABEA HUSS, CARSTEN STAACKE, KARSTEN REUTER, and CHRISTOPH SCHEURER — Fritz-Haber-Institute of the Max-Planck-Society

Its earth-abundance and chemical similarity makes sodium a promising substitute for lithium in future, sustainable solid-state batteries. In that respect the sodium thiophosphate (NaPS) material class has proven to provide electrolytes with high ionic conductivities [1]. Similar to the lithium thiophosphate (LiPS) material class, the NaPS class is characterized by a high degree of structural variety. It limits the transferability of *ab initio* studies, which are restricted to small model systems or short time scales for more complex models. Machine learning force-fields (ML-FF) provide a computationally cheaper yet sufficiently accurate alternative.

We have previously reported on a universal ML-FF for the whole LiPS material class [2]. In this work, we now develop a related ML-FF for glassy and crystalline phases of Na₃PS₄. Using the obtained ML-FF, we probe sodium ion conductivity in the glassy and crystalline phases. We systematically compare ion conductivity in LiPS vs. NaPS, and disentangle cation and anion dynamics in both material classes. Our aim is to understand the influence of stoichiometry and thiophosphate microchemistry on phase stability and ion conductivity.

[1] A. Hayashi *et al.*, J. Power Sources 258, 420 (2014).[2] C.G. Staacke *et al.*, Nanomaterials 12, 2950 (2022).

MM 6.3 Mon 10:45 SCH A 118

Accelerating structure prediction of solid-solid interfaces in solid electrolytes using Machine Learning Potentials — ●TABEA HUSS, CARSTEN STAACKE, KARSTEN REUTER, and CHRISTOPH SCHEURER — Fritz-Haber-Institut der MPG

The lithium thiophosphate (LPS) material class provides promising candidates for solid-state electrolytes (SSE) in lithium ion batteries due to high lithium ion conductivities, non-critical elements, and low material cost. LPS materials are characterized by structural disorder and occur in a multitude of glassy and crystalline phases, depending on their stoichiometry. The most performant glass-ceramic SSEs from the LPS class are characterized by omnipresent two-dimensional interfaces between crystalline and glassy domains, which can dominate the materials performance and cycle stability. To address this complexity we present a protocol for the construction of polycrystalline solid-solid interfaces in the LPS system. Within our protocol, expensive *ab-initio* random structure search (AIRSS) calculations are replaced by a Machine Learning surrogate accelerated approach. We present a pathway

towards a full assessment of partially amorphous interfaces in the LPS material class.

MM 6.4 Mon 11:00 SCH A 118

Understanding Oxide Ion Transport In Yttria Stabilized Zirconia: Fresh Insights from Molecular Dynamics Simulations — ●SUDESHNA MADHUAL, KRISHNANJAN PRAMANIK, and PADMA KUMAR PADMANABHAN — Indian Institute of Technology Guwahati, Guwahati, Assam, India, 781039

A comprehensive molecular dynamics investigation of yttria stabilized zirconia, Y_xZr_{1-x}O_{2-x/2}, is carried out for a wide range of compositions, x = 4 to 40 mol%, and over temperatures spanning 800 - 2200 K. The lattice parameter of the fluorite cell shows a monotonic increase with concentration, while the self-diffusivity of oxide ion as well as the resulting ionic conductivity, shows an optimum value around x = 10 mol%. These gross structural and transport properties of the system from the present study is in good agreement with previous experimental and theoretical investigations. It is noted that the oxygen migration occurs along straight channels parallel to the crystallographic axes, connecting the tetrahedral holes of the fluorite lattice occupied by them. A microscopic investigation of distinct oxygen environments, variably coordinated to Y³⁺ and Zr⁴⁺ cations, and of the channels connecting them is carried out. Analysis of these local channels for their energetics and their contribution to overall oxygen transport, resolved in terms of the cationic edges connecting them, provides fresh insights on the oxygen migration mechanism in the system.

MM 6.5 Mon 11:15 SCH A 118

Modelling accelerated ion transport in porous metal organic frameworks — ●THOMAS BERGLER^{1,2} and HARALD OBERHOFER^{1,2} — ¹University of Bayreuth — ²Bavarian Center for Battery Technologies

To date, metal organic frameworks (MOFs) have found a number of successful applications, for example in gas storage or as a filter for gas mixtures. So far, these mostly incorporated them as passive materials, but recent research points the way towards a more active role, possibly through the external manipulation of the materials's internal properties. One recent example for such a property is the susceptibility of the lattice parameters of multiple specific MOFs towards electric fields, which have been shown to distort on application of the fields. The aim of this project is to further investigate this behaviour from a theoretical point of view and gain insight into it through the use of molecular dynamics (MD) and meta-dynamics (MTD) simulations. These simulations are based on density functional theory (DFT) with the Perdew-Burke-Ernzerhof (PBE) functional but also on extended tight-binding (xTB) models and MOF-specialized force-fields. After confirmation that xTB-DFT and force-field MDs yield similar results to PBE-DFT MDs, MTDs have been performed with xTB-DFT to gain information on the energy-barriers for linker rotations inside some MOFs. Afterwards, MDs of multiple ps-length and an applied external E-field were performed with these force-fields to find the minimum strength of the E-field required to have an influence on the linker's rotation statistics.

15 min. break

MM 6.6 Mon 11:45 SCH A 118

Opposite in-plane anisotropy in thermal diffusivity and resistivity — ●FEI SUN¹, SIMLI MISHRA¹, PHILIPPA MCGUINNESS¹, ZUZANNA FILIPIAK¹, IGOR MARKOVIC¹, DMITRY SOKOLOV¹, SEAN HARTNOLL², ANDREW MACKENZIE^{1,3}, and VERONIKA SUNKO⁴ — ¹MPI, CPFS, Dresden, Germany — ²Univ. of Cambridge, Cambridge, UK — ³Univ. of St Andrews, St Andrews, UK — ⁴UC Berkeley, California, USA

We report the temperature dependence of the anisotropic thermal and electrical transport in the orthorhombic bilayer ruthenate Ca₃Ru₂O₇. Measurements are performed using two bespoke experimental techniques: optical measurement of thermal diffusivity, and resistivity measurements on devices micro-structured using Focused Ion Beam (FIB) sculpting. Intriguingly, we find that the electrical conductivity is higher along the direction where thermal conductivity is lower at all temperatures. The mechanism underlying this unusual behavior will be discussed in this talk.

MM 6.7 Mon 12:00 SCH A 118

Non-local microwave electrodynamics in ultra-pure PdCoO₂ — ●GRAHAM BAKER¹, TIMOTHY W BRANCH¹, JAKE BOBOWSKI¹, JAMES DAY¹, DAVIDE VALENTINIS², MOHAMED OUDAH¹, PHILIPPA MCGUINNESS³, SEUNGHYUN KHIM³, PIOTR SURÓWKA⁴, YOSHITERU MAENO⁵, RODERICH MOESSNER⁶, JÖRG SCHMALIAN², ANDREW MACKENZIE³, and DOUG BONN¹ — ¹University of British Columbia — ²Karlsruhe Institute of Technology — ³Max Planck Institute for Chemical Physics of Solids — ⁴Wrocław University of Science and Technology — ⁵Kyoto University — ⁶Max Planck Institute for the Physics of Complex Systems

There has been significant recent interest in unconventional electronic transport regimes in which a local, Ohmic relationship between electric current and field breaks down. To date, the vast majority of experimental work has been done in the DC limit. Here we introduce a novel approach via AC measurements with a bespoke microwave spectrometer, using the skin effect rather than sample dimensions to induce non-local transport. Through measurements on Sr₂RuO₄ and Sn, we verify the predictions for the magnitude and frequency dependence of the surface resistance for the classical and anomalous skin effects. Data from PdCoO₂, in contrast, deviate from any previous prediction of frequency-dependent surface resistance. Analysis of such data required the construction of a more complete electrodynamic theory of metals than previously existed, and allows us to conclude that the PdCoO₂ data are due to the combination of a highly anisotropic Fermi surface and a contribution from momentum-conserving scattering.

MM 6.8 Mon 12:15 SCH A 118

Spin Hall effect in tungsten and tantalum via first-principles calculations — ●REENA GUPTA, STEFANO SANVITO, and ANDREA DROGHETTI — School of Physics and CRANN, Trinity College, 2, Dublin, Ireland

We study theoretically the spin Hall effect (SHE), a phenomenon where a longitudinal charge current in a material sample gets converted into a transverse spin current and leads to spin accumulation on the sample boundaries. The common theoretical methods to calculate the intrinsic SHE conversion efficiency (i.e., the "spin Hall angle") are semi-classical or rely on the relaxation time approximation. To go beyond these limitations, we combine the non-equilibrium Green's function approach with Density Functional Theory. We study the SHE in bulk as well as slab geometries for different phases of tungsten and tantalum. We find values for the spin Hall angles, which are comparable to the most recent experimental results. Furthermore, in the slab geometries, we find that the spin Hall effect is accompanied by current-induced spin polarization (CISP). We then discuss the relative magnitude of the SHE and of CISP suggesting how the two effects can be distinguished in experiments.

MM 6.9 Mon 12:30 SCH A 118

Transport properties of Dirac materials: the role of plasmons — ●KITINAN PONGSANGANGAN — TU Dresden, Dresden, Germany

We study the thermoelectric transport of Coulomb interacting Dirac electrons with Keldysh quantum field theory. We study it from a weak-coupling and a strong-coupling perspective. We demonstrate that long-range Coulomb interactions play two independent roles: (i) they provide the inelastic and momentum-conserving scattering mechanism that leads to fast local equilibration; (ii) they facilitate the emergence of collective excitations, for instance plasmons, that contribute to transport properties on equal footing with electrons. Our approach is based on an effective field theory of the collective field coupled to electrons. Within a conserving approximation for the coupled system we derive a set of coupled quantum-kinetic equations. This builds the foundation of the derivation of the Boltzmann equations for the interacting system of electrons and plasmons. From this, we explicitly derive all the conservation laws and identify the extra contributions of energy density and pressure from the plasmons. We demonstrate that plasmons show up in thermo-electric transport properties as well as in quantities that enter the energy-momentum tensor, such as the viscosity.

MM 6.10 Mon 12:45 SCH A 118

Dragging effect of the Berry curvature in ferromagnetic Weyl semimetals NiMnSb and PtMnSb — SUKRITI SINGH¹, ●ANA GARCÍA-PAGE¹, JONATHAN NOKY¹, SUBHAJIT ROYCHOWDHURY¹, MAIA G. VERGNIORY^{1,2}, HORST BORRMANN¹, HANS-HENNING KLAUSS³, CLAUDIA FELSER¹, and CHANDRA SHEKHAR¹ — ¹Max-Planck-Institute for Chemical Physics of Solids, Dresden 01187, Germany — ²Donostia International Physics Center, Donostia-San Sebastián 20018, Spain — ³Institute for Solid State and Materials Physics, Technische Universität Dresden, Dresden 01069, Germany

The Anomalous Hall effect is a transport phenomenon in ferromagnets, which exhibit currents even in the absence of a magnetic field. Their inner magnetization breaks Time Reversal Symmetry, allowing the Berry Curvature (BC) to be finite. As a result, topological features close to the Fermi energy have a deep impact in the transport properties, leading to huge Anomalous Hall Conductivities (AHC). This has been a well-established paradigm for the last years for the linear AHC.

However, recent experimental results in the Weyl semimetals NiMnSb and PtMnSb might be pointing to something beyond it. By combining ab-initio calculations with a toy-model, we show that the steep slope-bands both compounds exhibit are capable of dragging the BC originated in Weyl nodes far away from the Fermi level, leading to a huge AHC in these compounds. We propose that this dragging effect of the BC can be generalized to the other materials exhibiting this band structure behaviour, which enriches the paradigm described above. Further research in this direction is currently being done.