TAming COmplexity in Materials Modeling (TACO)

4th Annual Ph.D. Workshop

Date: February 02–05, 2025
Place: Sporthotel Royer, Europaplatz 583, 8970 Schladming

Sunday, February 02

08:00 – 12:00	Bus transfer from Vienna (Wiedner Hauptsr./Resselg.) to Schladming
12:00 – 16:00	Outdoor discussions
16:00 – 16:25	Check-in Sporthotel Royer

Chair: Alexander Imre (P02)

16:45 – 16:50	**Ulrike Diebold, TU Wien (P01):** *Welcome address*
16:50 – 17:30	**Binqing Wei, University of Delaware (invited)** *Boost the Power Conversion Efficiency of Single-Junction Silicon Solar Cells through Temperature-Regulating*
17:30 – 17:50	Break
17:50 – 18:30	**Karsten Reuter, Fritz Haber Institute (invited)** *Machine Learning Accelerated Materials Discovery for Energy Conversion and Storage*
18:30 – 19:00	**David Kugler, TU Wien (P04)** *Stabilization of the Polar Spinel $MgAl_2O_4$(001) Surface by Al-rich Reconstruction*
19:00 – 20:00	Dinner
20:00 – 20:15	**Poster Speed Talks** (chair: Stefan Uttenthaler, P01)
20:15 – open-end	**Poster session**

Monday, February 03

08:00 – 09:00	Breakfast
09:00 – 15:00	Outdoor discussions

Chair: Marco Corrias (P07)

15:00 – 15:40	**Andreas Hauser, TU Graz (invited)** *Metallic Systems of Increasing Size: Computational Approaches on a Mesoscopic Scale*
15:40 – 16:10	**Marie Kienzer, TU Wien (P02)** *Progress on ViPErLEED*
16:10 – 16:40	**Parinya Tangpakonsab, TU Wien (P08)** *Oxide-Based Catalysts for Selective CO Oxidation*
16:40 – 17:00	Break

Chair: Faith Lewis (P04)

17:00 – 17:40	**Martin Sterrer, University of Graz (invited)** *Controlling Charge Transfer through Ultrathin Dielectric Films*
17:40 – 18:10	**Deborah Steiner / Silvio Bellomi, TU Wien (P10)** *Studies on Structure and Reactivity of Metal-Oxide Nanoparticles*
18:10 – 18:40	**Moritz Zelenka, University of Vienna (P11)** *Skiing Down the Co_3O_4 Mountain: Gaseous and Liquid 2-Propanol*
19:00 – 20:00	Dinner
20:00 – 21:00	**Executive Board meeting (PI, Co-PIs & student representatives)**
20:00 – open-end	**Poster session**

Tuesday, February 04

08:00 – 09:00	Breakfast
09:00 – 15:00	Outdoor discussions

TACO
TAMING COMPLEXITY TOGETHER

Chair: Viktor Birschitzky (P07)

15:00 – 15:40 **Ralf Drautz, Ruhr University Bochum (invited)**
Efficient Machine Learning Potentials

15:40 – 16:10 **Luca Leoni, University of Vienna (P07)**
Small Polaron Dynamics with Machine-Learned Force Fields

16:10 – 16:40 **Salvatore Romano, University of Vienna (P12)**
Structure and Dynamic of Water at the Magnetite Interface from Neural Network Potential-Based MD Simulations

16:40 – 17:00 Break

Chair: Sylwia Gutowska (P03)

17:00 – 17:40 **Andrea Grisafi, Sorbonne Université Paris (invited)**
Machine Learning of Electronic Charge Densities in the Simulation of Electrochemical Interfaces

17:40 – 18:10 **Florian Buchner, TU Wien (P09)**
msmJAX: Fast and Differentiable Electrostatics on the GPU

18:10 – 18:40 **Payal Wadhwa, University of Vienna (P03)**
Machine Learning-Driven Surface Reconstructions of the Cu_2O (111) Plane

19:00 – 20:00 Dinner

20:00 – 20:30 **General Assembly**

20:30 – open-end **Poster Session**

Wednesday, February 05

07:00 – 07:45	Breakfast, Check-out
8:00 – ca. 12:00	Bus transfer to Vienna

Abstracts

Boost the Power Conversion Efficiency of Single-Junction Silicon Solar Cells through Temperature-Regulating

Bingqing Wei

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Abstract

Solar cells are pivotal in addressing some of the most pressing challenges facing our planet today. As a clean and renewable energy source, solar cells convert sunlight into electricity, offering a sustainable and environmentally friendly alternative to conventional energy sources, significantly reducing our dependence on fossil fuels, mitigating climate change, and curbing environmental degradation. The widespread adoption of solar technology promises to achieve energy independence for nations, reduce greenhouse gas emissions, and promote a more sustainable and resilient energy infrastructure. Additionally, solar power can potentially empower communities in remote or off-grid areas, providing access to electricity where traditional power sources may be impractical or unavailable. The importance of solar cells lies not only in their capacity to generate clean energy but also in their role as catalysts for a more sustainable and equitable future.

Enhancing the power conversion efficiency (PCE) of solar cells is a constant and essential endeavor to advance the utilization of renewable electricity and build sustainable technology and society. However, the primary factor hindering PCE is the energy loss in the form of heat dissipation, imposing a PCE limitation of < 33% for single-junction Si solar cells, the most popular solar conversion technology thus far. Improving the PCE further to the next level is a fundamental and practical challenge. In this talk, I will discuss the possibilities of boosting the PCE of Si solar cells to over the S-Q limitation from temperature regulation, light penetration, and carriers' density and mobility perspectives.

Beyond Crystallinity and Throughput: Machine Learning Accelerated Materials Discovery for Energy Conversion and Storage

Karsten Reuter
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More performant and durable materials are urgently needed to further drive the transition to a sustainable energy system. Unfortunately, accelerated materials discovery is in this field presently still more claim than practical reality. Computational screening approaches hinge on efficient descriptors that only reflect nominal material properties of the crystalline bulk, simple bulk-truncated surfaces or idealized lattice-matching interfaces. They can thus not account for the substantial, complex and continuous structural, compositional and morphological transitions at the working surfaces or interfaces of catalysts, electrolyzers or batteries. Accelerated experimental discovery, in turn, still suffers from severe throughput limitations, as easily automatable human steps are rarely limiting the overall workflows. In my talk I will illustrate how modern machine learning (ML) approaches help to overcome these challenges. ML surrogate models, in particular in conjunction with agile, active learning-based training, boost the capabilities of predictive-quality multiscale modeling. Starting to tackle the true complexity of working interfaces, the derived mechanistic understanding promises to establish improved descriptors for more reliable computational screening campaigns. For experimental discovery, ML-guided experiment planning allows to optimally use available throughput capacities. In particular, most data-efficient adaptive design of experiment (DoE) requires only a limited number of experiments to provide a global (trend) understanding of the materials search space, while simultaneously allowing to batch-wise modify and refine both this search space or even the original research objective on the basis of the hitherto accumulated information.

Metallic Systems of Increasing Size: Computational Approaches on a Mesoscopic Scale

Andreas W. Hauser

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The description of metallic nanoparticles in various size regimes necessitates different computational treatments. Smallest particles are fully accessible via electronic structure theory, typically employing atomic-centered basis functions, while bulk systems are often treated via plane wave descriptions, taking advantage of translational symmetry. However, for metallic many-body systems in between these two extrema, where particle number and composition, surface effects and structural deviations play an important role, often the application of alternative strategies is required.

In the first part of the talk, possible approaches for various levels of system size are briefly discussed on selected systems, covering a range from less than 10 up to 10^4 metal atoms. While advanced multi-reference methods are needed to capture the quantum mechanics involved e.g. in electronic excitation processes, [1] a meaningful prediction of energy-to-structure relationships may not always necessitate a full quantum treatment, which extends the applicability in terms of particle number by an order of magnitude at least. Finally, metallic systems in the range of 10^4 particles may be approached from an entirely different perspective: in this size regime, atomic resolution will be abandoned for the sake of computational efficiency and replaced by a voxel-based ansatz for the description of surface diffusion and cluster shape stability, e.g. as a function of temperature. [2]

In the second part, the focus will shift from metal clusters to metallic complexes. Recently, we proposed a new strategy for the manipulation of nuclear spins within the context of quantum computing and information storage. [3] The vibrational excitation of 'pseudorotational' motions in suitable, highly symmetric molecules is identified as a possible way to generate localized magnetic fields. Within the solid-state community, this effect has been referred to as 'dynamical multiferroicity'. [4] With the help of locally induced magnetic fields, even spin states of identical atoms within the same molecule might become individually addressable.

(1) D. Buceta, S. Huseyinova, M. Cuerva, H. Lozano, L. J. Giovanetti, J. M. Ramallo-Lopez, P. Lopez-Caballero, A. Zanchet, A. O. Mitrushchenkov, A. W. Hauser, G. Barone, C. Huck-Iriart, C. Escudero, J. C. Hernandez-Garrido, J. J. Calvino, M. Lopez- Haro, M. P. de Lara-Castells, F. G. Requejo, and M. A. Lopez-Quintela. Stability and Reversible Oxidation of Sub-Nanometric Cu_5 Metal Clusters: Integrated Experimental Study and Theoretical Modeling. *Chemistry – A European Journal*, 49(29):e202301517, **2023**

(2) A. W. Hauser, M. Schnedlitz, and W. E. Ernst. A coarse-grained Monte Carlo ap- proach to diffusion processes in metallic nanoparticles. *The European Physical Journal D*, 71(6):150, **2017**

(3) Wilhelmer, R.; Diez, M.; Krondorfer, J. K.; Hauser, A. W. Molecular Pseudorotation in Phthalocyanines as a Tool for Magnetic Field Control at the Nanoscale. *J. Am. Chem. Soc.*, *146* (21), 14620–14632. **2024**

(4) D. M. Juraschek, M. Fechner, A. V. Balatsky and Nicola A. Spaldin. Dynamical multiferroicity. *Phys. Rev. Materials* 1, 014401, **2017**

Controlling the Charge Transfer through Ultrathin Dielectric Films

Martin Sterrer

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Charge transfer processes through ultrathin, supported oxide films have received increasing attention in recent years because of the possibility of controlling the charge state of adsorbates or the direction of catalytic reactions. The main driving force for the occurrence of charge transfer in these systems is the reduction of the substrate work function induced by the deposition of the oxide film in combination with an adsorbate of relatively high electron affinity. While previous studies have focused on the charging of metal atoms (e.g. Au) or small molecules (e.g. O_2, NO_2), we have recently extended these investigations to charge transfer processes into organic molecules. In this contribution, we present results on the adsorption and charging of model organic semiconductors (e.g. pentacene (5A), 2H-tetraphenylporphyrin (2H-TPP) and others) on ultrathin MgO(001) films supported on Ag(001). By combining scanning tunneling microscopy and photoemission spectroscopy and tomography, we identify and quantify charge transfer into the organic monolayer film. In addition, we show that by tuning the work function and/or the MgO thickness, it is possible to control the ratio of charged and neutral species, the number of electrons transferred and, concomitantly, the conformation of the adsorbates. In the case of 2H-TPP, charge transfer also appears to strongly influence the self-metalation of 2H-TPP to Mg-TPP. Thus, our investigations lay the basis for the ultimate control of charge transfer and the related chemistry on ultrathin oxide film systems.

Efficient Machine Learning Potentials

Ralf Drautz, Ruhr University Bochum

Machine learning interatomic potentials (MLIPs) are revolutionizing atomistic simulation as they enable computations at quantum mechanical accuracy for millions of atoms and chemically complex systems. Over the past two decades many different MLIPs were developed that represent non-experts with an impenetrable maze of methods and acronyms. In my talk, I will show that all MLIPs can be understood from a single model, despite the seemingly overwhelming complexity of the field. To this end, I will briefly introduce the graph atomic cluster expansion (GRACE). The complete basis of GRACE enables the representation of many other methods and I will outline some important links. I will then discuss how efficiency is a guiding principle for the development of MLIPs, as naive approaches tend to scale poorly and quickly hit exponential walls. Finally, I will illustrate applications of GRACE and highlight its scores on the matbench discovery leaderboard.

Machine Learning of Electronic Charge Densities in the Simulation of Electrochemical Interfaces

Andrea Grisafi, Sorbonne Université Paris

Machine-learning methods are playing an increasingly prominent role in the atomistic simulation of materials. By inferring the relevant electronic properties from the sole knowledge of the spatial atomic coordinates, these data-driven techniques make it possible to attain the accuracy of quantum mechanics while sidestepping the computational burden of electronic-structure methods. When it comes to electrochemical systems, however, the effective implementation of these approaches presents unique challenges. Notably, the presence of a heterogeneous interface between an electrode material and an electrolyte solution under an applied electric bias requires predicting complex long-range interactions that drive the dynamics of the system at the nanoscale. In this context, I will introduce a machine-learning method suitable to treat the three-dimensional electron density of a system [1] and show how it can be used to predict the nonlocal electronic polarization of metallic electrodes that are embedded in concentrated ionic solutions [2]. I will then continue discussing the integration of this method within the MetalWalls molecular dynamics program in order to accurately simulate the electrostatic properties of model ionic capacitors [3]. By taking a gold/electrolyte interface as an example, I will show how these advancements allow us to overcome the shortcomings of classical approaches in the description of the electrical double layer, by matching the accuracy of density-functional QM/MM methods while reaching time scales of several nanoseconds.

1. Andrea Grisafi, Alberto Fabrizio, Benjamin Meyer, David M. Wilkins, Clemence Corminboeuf, Michele Ceriotti, *Transferable machine learning model of the electron density*, ACS Central Science 5, 57-64 (2019) [https://doi.org/10.1021/acscentsci.8b00551]

2. Andrea Grisafi, Augustin Bussy, Mathieu Salanne and Rodolphe Vuilleumier, *Predicting the charge density response in metal electrodes*, Physical Review Materials 7, 125403 (2023) [https://doi.org/10.1103/PhysRevMaterials.7.125403]

3. Andrea Grisafi and Mathieu Salanne, *Accelerating QM/MM simulations of electrochemical interfaces through machine learning of electronic charge densities*, The Journal of Chemical Physics 161, 024109 (2024) [https://doi.org/10.1063/5.0218379]

Posters

Marco Corrias (P07)	*Computer-Assisted Pattern Recognition of Quasicrystalline Surfaces*
Tobias Dickbreder (P11)	*Investigating the Water Organization at the Calcite (10.4)-Water Interface at High pH*
Eva Doloszeski (P09)	*Exploring (2×4) Ti-Rich SrTiO3(110) Reconstructions Using Machine-Learned Force Fields Combined with Evolutionary Search*
Florian Dörr (P02)	*ViPErLEED: Finally in Use*
Karin Föttinger (P10)	*Surface Chemistry, Structure, and Reactivity of Multi-Component Spinel Nanoparticles*
Susanne Gross (P08)	*La-Modified Cobalt Oxide Thin Films: An XPS and LEIS Study*
Alexander Imre (P02)	*Parameter-Tree Tensor-LEED*
Darin Joseph (P07)	*Coupling between Small Polarons and Ferroelectricity in* $BaTiO_3$
Harsharan Kaur (P11)	*Understanding the Water Structure at Iron-Oxide/Water Interfaces*
Faith Lewis (P04)	*Imaging Rhodium Gem-Dicarbonyls on* TiO_2*(110)*
Christoph Rameshan (P10)	*Modified Pechini Synthesis of Perovskite-Type Oxide Catalysts*
Thomas Ruh (P01)	*Simulating Membrane Capture of* CO_2 *with Citizen Scientists*
Stefan Uttenthaler (P01)	*Dust Formation in Mira Variable Stars*
Ralf Wanzenböck (P09)	*Exploring Inhomogeneous Surfaces: Reconstructions via Active-Learning*