

HL 6: Focus Session: Frontiers of Electronic-Structure Theory III (joint session O/HL)

Electronic-structure calculations, based on density-functional theory (DFT) and methodology beyond, are getting increasingly involved as they face the following challenges: First, investigations of modern materials typically require large unit cells, owing to complex crystal structures, mixed compositions, internal interfaces, etc. Second, at the same time, they often require advanced methods, including hybrid functionals of DFT, Green-function techniques from many-body perturbation theory (MBPT), high-level wavefunction-based methods like coupled-cluster (CC) theory, or quantum Monte-Carlo simulations. All these methods should ideally be implemented in scientific software that is running efficiently on modern supercomputers. With both methodology and computer architectures exhibiting increasing complexity, collaborative development and shared tools, including ready-to-use libraries and codes, are becoming indispensable. This interdisciplinary symposium covers recent progress in the broad area of electron-structure methods and highly-sophisticated tools that enable the entire community to explore most exciting materials from different perspectives to either predict peculiar features or get insight into measured counterparts.

Organizers: Claudia Draxl (HU Berlin), Dorothea Golze (TU Dresden), Xavier Gonze (U Louvain), and Andris Gulans (U Latvia)

Time: Monday 10:30–13:00

Location: TRE Ma

HL 6.1 Mon 10:30 TRE Ma

Testing the hell out of DFT codes with virtual oxides — EMANUELE BOSONI¹, ●STEFAN COTTENIER², and GIOVANNI PIZZI³ — ¹ICMAB-CSIC, Spain — ²Ghent University, Belgium — ³EPFL, Switzerland

If you use DFT to predict a property of a crystal, how confident can you be that the prediction is computed in a bug-free way? And if your DFT-code uses pseudopotentials, can you trust that the pseudopotential does not modify your predictions? Answering such questions has been the goal of a study a few years ago, in which 71 unary crystals were examined in exactly the same way by 40 different DFT methods and codes [DOI 10.1126/science.aad3000]. In a next step, a consortium of 41 scientists (*) has done a similar exercise for a much larger pool of crystals: all elements of the periodic table up to Z=96, each in 10 different crystal structures, 6 of them being (virtual) oxides that sample a range of chemical bond types and 4 being unary crystals that sample low to high coordination environments. In this presentation, we will discuss the reasons to choose these crystals, the different quality criteria by which results can be compared, we will demonstrate how this exercise leads to more precise and more trustworthy pseudopotential libraries, and we will show how this data set is shared with the community in order to foster better-tested codes and pseudopotentials for all.

(*) Unfortunately the size of this abstract does not allow to mention them all.

HL 6.2 Mon 10:45 TRE Ma

High-throughput absorption spectra obtained by beyond-DFT workflows — ●FABIAN PESCHEL, ALEXANDER BUCCHERI, and CLAUDIA DRAXL — Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany

Fully converging *ab initio* calculations can be a challenging task, in particular when it comes to excited states, which require multiple ground-state calculations for different physical quantities. In this work, we aim at computing highly precise absorption spectra by employing the Bethe-Salpeter equation of many-body perturbation theory, as implemented in the all-electron full-potential package **exciting** [1,2]. To obtain benchmark data for a wide range of material classes, we have developed workflows where Python tools automatically create input files, start calculations, and evaluate results. For each material, all relevant input parameters, such as the number of k-points for the Brillouin-zone sampling, basis-set basis cutoff and the number of unoccupied states, are varied until the targeted convergence criteria are reached. With the help of a workflow manager, the calculations can be executed in a high-throughput fashion on a high-performance computing cluster. We demonstrate our approach with core-level spectra of elemental and binary solids, and provide an in-depth analysis of the obtained data. This work is carried out in the framework of the NOMAD Center of Excellence [3] and the CRC FONDA [4].

[1] A. Gulans et al., *J. Phys. Condens. Matter* **26**, 363202 (2014). [2] C. Vorwerk, B. Aurich, C. Cocchi, and C. Draxl, *Electron. Struct.* **1**, 037001 (2019). [3] <https://nomad-coe.eu> [4] <https://fonda.hu-berlin.de>

Topical Talk

HL 6.3 Mon 11:00 TRE Ma

Large-scale machine-learning assisted discovery and characterization of materials — ●MIGUEL ALEXANDRE LOPES MARQUES — Institut für Physik Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany

In this talk we discuss our recent attempts to discover, characterize, and understand inorganic compounds using *ab initio* approaches accelerated by machine learning. We start by motivating why the search for new materials is nowadays one of the most pressing technological problems. Then we summarize our recent work in using crystal-graph attention neural networks for the prediction of materials properties. To train these networks, we curated a dataset of over 2 million density-functional calculations with consistent calculation parameters. Combining the data and the newly developed networks we have already scanned more than two thousand prototypes spanning a space of more than one billion materials and identified tens of thousands of theoretically stable compounds. We then discuss how simple, interpretable machine learning approaches can be used to understand complex material properties, such as the transition temperature of superconductors. Finally, we speculate which role machine learning will have in the future of materials science.

15 min. break

HL 6.4 Mon 11:45 TRE Ma

Predicting the electronic structure at any length scale with machine learning — ●ATTILA CANGI — Helmholtz-Zentrum Dresden-Rossendorf, Görlitz, Germany

The properties of electrons in matter are of fundamental importance. They give rise to virtually all molecular and material properties and determine the physics at play in objects ranging from semiconductor devices to the interior of giant gas planets. Calculations rely primarily on density functional theory (DFT), which has become the principal method for predicting the electronic structure of matter. While DFT calculations have proven to be very useful, their computational scaling limits them to small systems. We have developed a scalable machine learning framework for predicting the electronic structure on any length scale [1,2,3]. It shows up to three orders of magnitude speedup on systems where DFT is tractable and, more importantly, enables predictions on scales where DFT calculations are infeasible. Our work demonstrates how machine learning circumvents a long-standing computational bottleneck and advances science to frontiers intractable with any current solutions.

[1] J. A. Ellis, L. Fiedler, G. A. Popoola, N. A. Modine, J. A. Stephens, A. P. Thompson, A. Cangi, S. Rajamanickam, *Phys. Rev. B* **104**, 035120 (2021). [2] L. Fiedler, N. Hoffmann, P. Mohammed, G. A. Popoola, T. Yovell, V. Oles, J. A. Ellis, S. Rajamanickam, A. Cangi, *Mach. Learn.: Sci. Technol.* **3** 045008 (2022). [3] L. Fiedler, N. A. Modine, S. Schmerler, D. J. Vogel, G. A. Popoola, A. P. Thompson, S. Rajamanickam, A. Cangi, arXiv:2210.11343 (2022).

HL 6.5 Mon 12:00 TRE Ma

Demonstrating temperature transferability of neural network models replacing modern density functional theory — •LENZ FIEDLER and ATTILA CANGI — Helmholtz-Zentrum Dresden-Rossendorf / CASUS

Due to its balance between accuracy and computational cost, Density Functional Theory (DFT) is one of the most important computational methods within materials science and chemistry. However, current research efforts such as the modeling of matter under extreme conditions demand the application of DFT to larger length scales as well as higher temperatures. Such investigations are currently prohibited due to the computational scaling of DFT.

We have recently introduced a machine-learning workflow that replaces modern DFT calculations [1,2,3]. This workflow uses neural networks to predict the electronic structure locally. We show that by employing such an approach, models can be trained to predict the electronic structure of matter across temperature ranges. This paves the way for large-scale simulations of thermodynamically sampled observables relevant to modeling technologically important phenomena such as radiation damage in fusion reactor walls.

- [1] J. A. Ellis *et al.*, Phys. Rev. B **104**, 035120
- [2] L. Fiedler *et al.*, Mach. Learn.: Sci. Technol., **3** 045008
- [3] L. Fiedler *et al.*, arXiv:2210.11343

HL 6.6 Mon 12:15 TRE Ma

Pure non-local machine-learned density functional theory for electron correlation — •JOHANNES T. MARGRAF — Fritz-Haber-Institut der MPG, Berlin, Germany

Density-functional theory (DFT) is a rigorous and (in principle) exact framework for the description of the ground state properties of atoms, molecules and solids based on their electron density. While computationally efficient density-functional approximations (DFAs) have become essential tools in computational chemistry, their (semi-)local treatment of electron correlation has a number of well-known pathologies, e.g. related to electron self-interaction. Here, we present a type of machine-learning (ML) based DFA (termed Kernel Density Functional Approximation, KDFA) that is pure, non-local and transferable, and can be efficiently trained with fully quantitative reference methods. The functionals retain the mean-field computational cost of common DFAs and are shown to be applicable to non-covalent, ionic and covalent interactions, as well as across different system sizes.

HL 6.7 Mon 12:30 TRE Ma

Predicting the response of the electron density to electric field using machine learning — •ALAN LEWIS and MARIANA ROSSI

— MPI for Structure and Dynamics of Materials, Hamburg, Germany

The response of the electron density of a molecule or material to a homogeneous electric field defines its dielectric constant, along with its Raman and sum-frequency spectrum. We present a local and transferable machine learning approach capable of predicting the density response of molecules and periodic system on the same footing. This uses a very similar framework to that of the SALTED method recently introduced by these authors,[1,2] requiring only a small modification to the λ -SOAP descriptors used to represent the atomic environments. This allows us to predict the density response of liquid water to a field applied in each Cartesian direction from a single machine learning model. The tensorial dielectric constant can then be derived from this predicted density response, dramatically reducing the computational cost of calculating these properties relative to the standard approach of using density functional perturbation theory. We discuss the transferability of the model to different phases, and demonstrate the extrapolative power of this approach.

- [1] Lewis, Grisafi, Ceriotti, Rossi, JCTC **17**, 11, 7203 (2021)
- [2] Grisafi, Lewis, Rossi, Ceriotti, accepted JCTC (2022)

HL 6.8 Mon 12:45 TRE Ma

Analysis of Batching Methods in Graph Neural Network Models for Materials Science — •DANIEL SPECKHARD, TIM BECHTEL, JONATHAN GODWIN, and CLAUDIA DRAXL — Humboldt-Universität zu Berlin, Physics Department and IRIS Adlershof, Berlin, Germany

Graph neural network (GNN) based models have shown promising results for materials science [1]. These models often contain millions of parameters, and like other big-data based models, require only a portion of the entire training dataset to be fed as a mini-batch to update model parameters. The effect of batching on the computational requirements of training and model performance has been thoroughly explored for neural networks [2] but not yet for GNNs. We explore two different types of mini-batching methods for graph based models, static batching and dynamic batching. We use the Jraph library built on JAX to perform our experiments where we compare the two batching processes for two data-sets, the QM9 dataset of small molecules and the AFLOW materials database [3]. We show that dynamic batching offers significant improvements in terms of computational requirements for training. We also present results on the effect of the batch size and batching method on model performance.

- [1] T. Xie *et al.*, Physical Review Letters, **120**, 14 (2018).
- [2] M. Li *et al.*, Proceedings of the 20th ACM SIGKDD (2014).
- [3] S. Curtarolo *et al.*, Comp. Mat. Science, **58**, 227-235 (2012).