MM 8: Development of Computational Methods: Diverse Topics and Machine Learning

Time: Monday 15:45–17:45 Location: SCH A 251

MM 8.1 Mon 15:45 SCH A 251

Adaptively Compressed Exchange in LAPW — •Davis Zavickis, Kristians Kacars, Janis Cimurs, and Andris Gulans — Electronic Structure Group, University of Latvia, Jelgavas st. 3, LV-1004, Riga, Latvia

We address precision and reproducibility issues in DFT calculations with hybrid functionals. Linearized augmented plane waves (LAPW) method currently serves as the de facto reference tool within the electronic structure community. In the current implementation of the Fock exchange in LAPW, the total and band energies depend on the number of orbitals used. We overcome these issues by implementing the adaptively compressed exchange (ACE) method [Lin Lin, J. Chem. Comput., 2016, 12, 5] in `exciting` code that introduces a low rank approximation and apply it to light atoms, molecules and solids [D. Zavickis et al., Phys. Rev. B., 2022, 106, 165101]. In case of atoms and molecules, we show that ACE leads to highly precise total energies which are within a few microhartrees off the results obtained by multiresolution analysis method. In solids we calculate band structures that are compared with other all-electron hybrid implementations. Lastly, we apply optimizations and fine tuning to ACE, analyze its complexity and computational performance by comparing it to the previous Fock-exchange implementation in the `exciting` code.

MM 8.2 Mon 16:00 SCH A 251

How much laser power can two-photon 3D printed microoptics withstand? — •Sebastian Klein, Pavel Ruchka, Tobias Steinle, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany

In recent years, the manufacturing of small 3D printed microoptics has seen a rise in importance. 3D printed lenses are used for applications where space is of big concern. Tailored free form optics are ideally suited to improve the performance for specific use cases. New applications also include printing these optics onto optical fibers, for example for endoscopes in the biomedical field or for focusing a laser beam inside a compact fiber laser. In the latter case, the sustainability of high intensities of the pump laser is important. This is directly dependent on the maximum power the material of the lens can withstand.

In this work, we obtain a first insight into laser damage thresholds of the 2PP photoresist IP-S in the near-IR spectral range. We determine damage thresholds with femtosecond laser pulses for wavelengths in the range of 1550-2000 nm. We conduct these experiments on cube samples printed from IP-S and evaluate the damage of the exposed samples visually under a microscope, employing also differential interference contrast. The observed damage is mostly of thermal nature. Knowledge of damage thresholds of 3D printed micro-optics further pushes the applications to these limits, enabling high-power applications at safe operating intensities.

MM 8.3 Mon 16:15 SCH A 251

Molecular Dynamics Simulation of Selective Laser Melting — Fabio Oelschläger[1], Azad Gorgis[1], Dominc Klein[1], Sarah Müller[2], and •Johannes Roth[1] — [1]FMQ, Universität Stuttgart, Germany — [2]GSaME, Universität Stuttgart, Germany

Traditionally, manufacturing has been subtractive which means removing material from a workpiece. Additive manufacturing on the other hand is defined by successive addition of material and fusion with the help of heat for example. Here we report atomistic simulations of selective laser melting (SLM) used to produce additive manufactured objects. After a short introduction into the subject we present challenges to SLM. Next, modifications to basic molecular dynamics simulation are described which are required to simulate the annealing process. Although the sample sizes studied are already impressively large, scaling of system parameters are required to relate simulation and experiment. First results from the study of single and rows of spheres will be reported and further developments and improvements will be addressed.

MM 8.4 Mon 16:30 SCH A 251

Exploring Enhanced Sampling Concepts based on Boltzmann Generators — •David Greten, Karsten Reuter, and Johannes T. Margraf — FHI Theory Department, Berlin, DE

Computational surface science and catalysis research is still mainly conducted with static density functional theory (DFT) calculations. This approach is computationally convenient, but misses important aspects of surface chemistry, such as anharmonic free energy contributions. In principle, DFT-based molecular dynamics (MD) simulations (ideally combined with enhanced sampling algorithms) would allow a much more accurate description of these processes. Unfortunately, these are far too expensive to be routinely applied to complex surface/adsorbate systems. This is due to the fact that configurations in MD are generated sequentially. As a consequence, MD configurations are not statistically independent so that a very large number of samples is required to obtain converged ensemble properties. To overcome this limitation, Noé and co-workers recently proposed a generative machine learning model called the Boltzmann Generator, which was used to generate independent configurations of biomolecules. In this contribution, we explore how we can expand ML based sampling concepts utilizing Boltzmann Generators. In particular, training protocols and validation metrics will be discussed.

15 min. break

MM 8.5 Mon 17:00 SCH A 251

When does the Tamura model of phonon-isotope scattering break down? — •Nakib Protik and Claudia Draxl — Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany

A standard approach to the phonon-isotope scattering problem is the Tamura model [1]. This non-self-consistent 1st Born approximation of the scattering T-matrix expansion is exact for the low energy phonons, and higher order perturbative corrections for the higher energy, dispersive acoustic phonons have been argued to be small [1]. To our knowledge, the validity of this approach for the optic phonons has not yet been demonstrated. In this talk, we compare the Tamura model to the ab initio computed non-perturbative phonon-isotope scattering T-matrix for a set of well-studied materials. We show under what conditions the Tamura model breaks down.

[1] Tamura, S. I. (1983). Isotope scattering of dispersive phonons in Ge. Physical Review B, 27(2), 858.

MM 8.6 Mon 17:15 SCH A 251

Physics-inspired Machine Learning for Predicting Ionization Energies of Electronically Localized Systems — •Ke Chen[1,2,3], Christian Kunkel[1], Bingqing Cheng[3], Karsten Reuter[1], and Johannes T. Margraf[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]Technische Universität München, Garching, Germany — [3]Institute of Science and Technology, Klosterneuburg, Austria

Machine learning (ML) has been successfully applied to predict many chemical properties, most prominently energies and forces in molecules and materials. The strong interest in predicting energies in particular has lead to a 'local energy'-based paradigm for modern chemical ML models, which ensures size extensivity and linear scaling of computational cost. However, some electronic properties (such as excitation energies or ionization potentials) are not size-extensive and may even be spatially localized. Using extensive models in these cases can lead to large errors. In this work, we explore different strategies for predicting intensive and localized properties, using ionization energies in organic molecules as a test case. In particular, we compare size-intensive aggregation functions and effective, machine-learned Hamiltonians. The physical interpretability and cost/benefit ratios of the approaches will be discussed.

MM 8.7 Mon 17:30 SCH A 251

Kernel Charge Equilibration: Machine Learned Interatomic Potentials With Full Long-Range Electrostatics — •Martin Vondrak, Johannes T. Margraf, and Karsten Reuter — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

Machine learning (ML) techniques have recently been shown to bridge the gap between accurate first-principles methods and computationally cheap empirical potentials. This is achieved by learning a mapping between a systems structure its physical properties. State-of-the-art models typically represent chemical structures in terms of local atomic environments to this end. This inevitably leads to the neglect of long-range interactions (most prominently electrostatics)

and non-local phenomena (e.g. charge transfer), resulting in significant errors in the description of polar molecules and materials (particularly in non-isotropic environments). To overcome these issues, we recently proposed a ML framework for predicting charge distributions in molecules termed Kernel Charge Equilibration (kQEq). Here, atomic charges are derived from a physical model using environment-dependent atomic electronegativities. In this contributions, strategies for creating kQEq interatomic potentials are discussed, including the combination of short-ranged Gaussian Approximation Potentials with kQEq.