O 58: Poster Wednesday: New Methods and Developments, Frontiers of Electronic Structure Theory

Time: Wednesday 18:00–20:00 Location: P4

O 58.1 Wed 18:00 P4

Fully Atomistic Modelling of Tip-enhanced Raman Spectra from First Principles — •Yair Litman, Franco Bonafe, Alaa Akkoush, Heiko Appel, and Mariana Rossi — MPI for the Structure and Dynamics of Matter, Hamburg, Germany.

Tip-enhanced Raman scattering (TERS) has emerged as a powerful tool to study surfaces with subnanometer spatial resolution [1]. In particular, single-molecule TERS studies have shown the capability to visualize high-resolution images of individual molecular normal modes in real space [2]. Theoretical simulations that can provide an unambiguous interpretation and atomic description of obtained TERS images often rely on crude approximations of the local electric field [3]. In this work, we present a novel method to compute TERS images by combining Time Dependent Density Functional Theory (TDDFT) and Density Functional Perturbation Theory (DFPT) to calculate Raman cross sections with realistic local fields. The new approach allows for a fully *ab initio* atomistic description of the tip-molecule-surface system, and naturally incorporates chemical effects arising from the molecule-surface interaction. We show results for benzene and pyridine and discuss the importance of a realistic description of the local field, and self consistent evaluation of the electronic density response. Finally, we evaluate the use of 2D TERS imaging as an identification tool for defects in 2D materials. [1] M. Richard-Lacroix, *et al.*, Chem. Rev. **56**, 3922 (2017) [2] L. Joonhee, *et al.*, Nature **568**, 78 (2019) [3] P. Liu, X. Chen, H, Ye, and L. Jensen, ACS Nano **13**, 9342 (2019)

O 58.2 Wed 18:00 P4

Deep learning based signal processing for touch-sensitive surfaces — •Jakob Elsner, Viktor Fairuschin, and Thorsten Uphues — Institute for Sensor and Actuator Technology, Coburg, Germany

Touch-sensitive surfaces enable intuitive and efficient operation of electronic devices and eliminate the need for external peripherals and mechanical components, making touch technology increasingly important in the modern society. However, conventional touch technologies, i.e. capacitive, resistive or optical, are usually limited to non-metallic materials that hardly meet the stringent requirements for robustness and hygiene in a medical environment. Stainless steel is one of the most commonly used materials in medical fields due to its high strength, chemical resistance and excellent hygienic properties. In this work, we present a novel approach based on Lamb wave technology and deep learning analytics, and apply this new principle to design a stainless steel touch-sensitive surface. Compared to Rayleigh wave-based touch technology, our approach requires no additional reflective structures and involves only a single piezoelectric transducer used to monitor the entire surface, while position-sensitive information is extracted from raw Lamb wave signals using a trained deep neural network.

O 58.3 Wed 18:00 P4

Enhanced Sampling of Surface Reactions Using Boltzmann Generators — •David Hering, Johannes T. Margraf, and Karsten Reuter — 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 Boltzmann Generators can also be used to sample the free energy surface of surface/adsorbate systems relevant for heterogeneous catalysis. In particular, training protocols and validation metrics of generated ensembles will be discussed.

O 58.4 Wed 18:00 P4

theoretical and experimental investigation of Fe and Ni-TCNQ on graphene — •azin shahsavar, zdeněk jakub, anna kurowská, jakub planer, ondrej herich, lenka černá, lukáš kormoš, pavel procházka, and jan čechal — CEITEC Brno University of Technology Purkyňova 656/123 612 00 Brno, Czech Republic.

Due to the outstanding properties of the 2D metal-organic frameworks (MOF), intensive computational and experimental studies have been done. However, the lack of fundamental studies of MOFs on the graphene backbone is observed. This work studies Fe and Ni as metal and tetracyanoquinodimethane (TCNQ) with a high electron affinity as an organic linker functionalized on graphene. Here we present DFT calculations results to unveil the electronic and magnetic properties of iron and nickel-TCNQ physisorbed on graphene. Adsorption and Fermi energies, structural, and magnetic properties will be reported. Our experimental observations prove Fe- and NiTCNQ@Gr/Ir(111) are thermally highly stable up to 500 and 250 °C, respectively, making them promising materials for single-atom catalysts or high-density storage media [1]. [1] Z. Jakub et al., Nanoscale., 1-9 (2022). DOI: 10.1039/d2nr02017c

O 58.5 Wed 18:00 P4

Assessment of Structural Descriptors for the Construction of High-Dimensional Neural Network Potentials — •Moritz R. Schäfer[1], Jonas Finkler[2], Stefan Goedecker[2], and Jörg Behler[1] — [1]Georg-August-Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Tammannstraße 6, 37077 Göttingen, Germany — [2]Basel University, Department of Physics and Astronomy, Klingelbergstrasse 82, 4056 Basel, Switzerland

High-dimensional neural network potentials (HDNNPs) can be used to efficiently compute close-to ab initio quality energies and forces for performing large-scale molecular dynamics simulations of complex systems. In this method, the total energy is constructed as a sum of environment-dependent atomic energy contributions. Also electrostatic interactions based on environment-dependent charges can be included. Hence, a set of reliable structural descriptors for the atomic local environments is crucial to develop accurate potentials. Often, atom-centered symmetry functions (ACSFs) are used for this purpose in HDNNPs. In this work, we benchmark the accuracy and transferability of HDNNPs with respect to alternative descriptors like the recently proposed overlap matrix descriptor.

O 58.6 Wed 18:00 P4

Fortnet, a software package for training Behler-Parrinello neural networks — •Tammo van der Heide[1], Jolla Kullgren[2], Peter Broqvist[2], Vladimir Bačić[3], Thomas Frauenheim[4,5,1], and Bálint Aradi[1] — [1]BCCMS, University of Bremen, Bremen, Germany — [2]Dept. of Chemistry - Ångström Laboratory, Uppsala University, Uppsala, Sweden — [3]Dept. of Physics and Earth Sciences, Jacobs University Bremen, Bremen, Germany — [4]Beijing CSRC, 100193 Beijing, P. R. China — [5]Shenzhen JL CSAR Institute, Shenzhen 518110, P. R. China

A new, open source, parallel, stand-alone software package (Fortnet) has been developed, which implements Behler-Parrinello neural networks. It covers the entire workflow from feature generation to the evaluation of generated potentials, coupled with higher-level analysis such as the analytic calculation of atomic forces. The functionality is demonstrated by driving the training for the fitted correction functions of the density functional tight binding (DFTB) method, which are commonly used to compensate the inaccuracies resulting from the DFTB approximations to the Kohn-Sham Hamiltonian. Their usual two-body form limits the transferability of parameterizations between very different structural environments. After investigating various approaches, we have found the combination of DFTB with a near-sighted artificial neural network, acting on-top of baseline correction functions, the most promising one. It allows to introduce many-body corrections on top of two-body parametrizations, while excellent transferability to deviating chemical environments could be demonstrated.

O 58.7 Wed 18:00 P4

Machine learning enhanced DFTB method for periodic sys-

tems — •Wenbo Sun, Guozheng Fan, Tammo van der Heide, Adam McSloy, Thomas Frauenheim, and Bálint Aradi — Bremen Center for Computational Materials Science, University of Bremen, Am Fallturm 1, Bremen 28359, Germany.

The Density Functional based Tight Binding (DFTB) is an approximative density functional based quantum chemical simulation method with low computational costs. In order to increase its accuracy, we have introduced a machine learning algorithm to optimize several parameters of the DFTB method, concentrating on solids with defects. The backpropagation algorithm was used to reduce the error between DFTB and DFT results w.r.t. the training dataset and to obtain adjusted DFTB Hamiltonian and overlap matrix elements. Afterwards, the generalization capability of the trained model was tested for geometries not being part of the training set. In the current work, we have focused on defective periodic silicon and silicon carbide systems as target materials and the density of states (DOS) as target property to demonstrate the feasibility of our approach. The trained model was able to reduce the differences between the DFTB and the DFT DOS significantly, while other derived properties (e.g. charge distribution, partial DOS) remained physically sound. Also, the transferability of the obtained model could be verified. Our method allows to carry out relatively fast simulations with high accuracy and only moderate training efforts, and represents a good compromise for cases, where long range effects make direct machine learning predictions difficult.

O 58.8 Wed 18:00 P4

Electronic properties of Density Functional Tight Binding by Machine Learning — •Guozheng Fan[1], Adam McSloy[1], Balint Aradi[1], Chi-Yung Yam[2], and Thomas Frauenheim[1,2] — [1]Bremen Center for Computational Materials Science (BCCMS), University of Bremen, Bremen, Germany — [2]Beijing Computational Science Research Center (CSRC), Beijing, China

We have introduced a machine learning workflow, which could optimize electronic properties in density functional tight binding method. With this workflow, we can train and predict electronic properties in a cheap, accurate and transferable way. The implementation features of batch calculations greatly improve the calculation efficiency, especially for high throughput calculations. This workflow could optimize electronic properties by train basis functions or train a spline model to generate two center integrals for off-diagonal and onsite for diagonal Hamiltonian and overlap. The results show that compared with previous Slater-Koster parameters, the dipole moments, charges, the ratios of the on-site populations and the atomic numbers in charge population analysis method can be improved by both tuning basis function parameters or optimizing integrals in spline model directly. The training on basis functions could prevent the two center integrals go randomly and keep Hamiltonian and overlap in reasonable range. Besides, the multiple electronic properties could be improved simultaneously.

O 58.9 Wed 18:00 P4

Unsupervised regression-based measures for applications on atomistic features — •Alexander Goscinski[1], Guillaume Fraux[1], Giulio Imbalzano[1], Félix Musil[1,2], Sergey Pozdnyakow[1], and Michele Ceriotti[1] — [1]Laboratory of Computational Science and Modeling, Institute of Materials, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland — [2]National Center for Computational Design and Discovery of Novel Materials (MARVEL), Lausanne, Switzerland

The quality of the features as input for a machine learning model is a crucial factor for the prediction quality and the computational efficiency. Commonly, to assess the quality of features, they are compared by benchmarking the regression performance on several properties. Complementary to such a quality assessment, this work presents certain measures for direct feature-to-feature comparisons without the need of a target property. These measures are used to quantify the capacity of features representing geometrical space in atomistic applications and derive an understanding of the information encoded in features.

O 58.10 Wed 18:00 P4

Accelerating plane-wave-based *ab initio* molecular dynamics by optimization of Fast-Fourier transforms for modern HPC architectures — •Christian Ritterhoff, Tobias Klöffel, Sagarmoy Mandal, and Bernd Meyer — Interdisciplinary Center for Molecular Materials and Computer Chemistry Center, FAU Erlangen-Nürnberg, Germany

The most important advantage of plane-wave basis sets is that wave functions can be transformed efficiently from reciprocal to real space and back by using the Fast-Fourier transform (FFT) algorithm. This allows to evaluate the kinetic and potential energy in reciprocal and real space, respectively, where both operators are diagonal. This reduces the computational cost for applying the Hamilton operator from N^2 to $N \log N$. However, the scalability of current FFT libraries is rather limited on today's HPC systems, which offer large numbers of compute nodes, each of them with many cores. Here we present our optimization of the FFTX library of the Quantum Espresso software package. Data distribution and communication patterns have been revised to make optimal use of combined MPI and OpenMP parallelization. Scalability is further increased by combining FFTs into batches and by introducing overlapping computation and communication. We implemented the revised FFTX library in our optimized version of the CPMD code [1], and we demonstrate the achieved acceleration by a series of benchmark simulations.

[1] T. Klöffel, G. Mathias, B. Meyer, Comput. Phys. Commun. **260** (2021) 107745

O 58.11 Wed 18:00 P4

Surface tension measurement of pure water in vacuum — •Paul T. P. Ryan, Jiri Pavelec, Jan Balajka, Michael Schmid, and Ulrike Diebold — Institute of Applied Physics, TU Wien, Austria

Very little is known about the surface tension of pure liquids in contact with their pure gaseous phases, i.e. without the presence of other gases or liquid phase contaminants. This is surprising given that contaminants are known to greatly affect surface tensions values[1]. Recently we have developed a method to dose liquid water onto pristine surfaces in UHV using a small cryostat [2,3]. We combine this approach with the pedant drop method [4] to measure the surface tension of ultra-clean liquids in contact with their pure gaseous phases. The upgraded version of the small cryostat, replaces the syringe typically used in the pendant-drop method. The ultra-clean liquid is condensed onto a small cryostat placed in a vacuum chamber. A pendant drop is formed and carefully photograph allowing the surface tension of the liquid to be directly determined. The design of the apparatus will be discussed and preliminary measurements of ultra-clean water will be presented. [1] Yuki Uematsu, et. al., Current Opinion in Electrochemistry, Volume 13, (2019) [2] Jan Balajka, et. al. , Review of Scientific Instruments 89, (2018) [3] Jan Balajka, et. al., Science, 361, (2018) [4] Berry, J. D. et. al., J. Coll. Interface Sci. 454, 226*237, (2015).

O 58.12 Wed 18:00 P4

Home-Built UHV Suitcase — •Luca Lezuo[1], Lucie Dockalová[2], Gareth Parkinson[1], Ulrike Diebold[1], and Jiri Pavelec[1] — [1]Institute of Applied Physics, TU Wien, Wiedner Hauptstrasse 8-10/134, Vienna, Austria — [2]Institute of Physical Engineering, Brno University of Technology

Due to their extreme sensitivity to adsorbing molecules, most surface experiments have to be carried out in ultra-high vacuum (UHV). Ideally, multiple different techniques are used to explain and understand the phenomena happening on an atomic scale. To this end, it is often necessary to transfer a sample from one chamber to another.

As a showcase, we discuss the analysis of perovskite oxides, produced by pulsed laser deposition (PLD) as thin films, in another UHV chamber that allows atomically resolved STM/AFM imaging with a Q+-sensor at liquid N_2/He - temperatures. The home built UHV suitcase consists of three stages divided by gate valves. It has a scroll pump for rough vacuum, a turbo pump to reach high vacuum, a NEG pump and a cryopump to ensure a clean transfer and an ION/NEG combination to provide UHV conditions long term in the storage stage, where the sample is transported.

O 58.13 Wed 18:00 P4

Wettability investigation of microscale water droplets on silicon substrate using atomic force microscopy — •Mohammadali Hormozi[1], Marvin Hoffer[1,2], Pauline Brumm[2], and Regine von Klitzing[1] — [1]Soft Matter at Interfaces, Department of Physics, Technical University of Darmstadt, 64289 Darmstadt, Germany — [2]Institute of Printing Science and Technology (IDD), Technische Universität Darmstadt, Magdalenenstraße 2, 64289 Darmstadt, Germany

The wettability of a particular substrate by a liquid drop is of interest in many scientific fields. This phenomenon is often described by the contact angle between the considered liquid and substrate. This pa-

rameter is an important boundary condition, especially for the wetting of mixtures and solutions. This study shows a methodology for evaluating the contact angle of different droplets with a base diameter down to 0.5 micrometers. 3D topography of water droplets -generated through condensation and Inkjet printing on a silicon substrate- has been determined using Atomic Force Microscopy. Also, the topography of printed mixture droplets including water-glycerol and water-glycerol-isopropanol has been measured. Different curves have been fitted to a 2D cross-section of each droplet, which provides information about their contact angle. The contact angle of the mixtures deviates from the macroscale contact angle at the vicinity of the three-phase contact line; however, this phenomenon cannot be seen in pure water droplets. While no effect of droplet diameter could be detected for diameters, ranging from 0.5 to 30 micrometers, the macroscopic contact angle of droplets is several degrees higher.

O 58.14 Wed 18:00 P4

High-frequency shot-noise STM to study correlated electron systems — •Maialen Ortego Larrazabal[1], Jiasen Niu[2], Koen M Bastiaans[3], Jianfeng Ge[2], Tjerk Benschop[2], Milan P Allan[2], and Ingmar Swart[1] — [1]Debye Institute for Nanomaterials Science, Utrecht University, PO Box 80000, 3508 TA Utrecht, The Netherlands — [2]Leiden Institute of Physics, Leiden University, Niels Bohrweg 2, 2333 CA Leiden, The Netherlands — [3]Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, Netherlands

The fluctuations in time of a measured signal provide information that is not present in the time averaged value. For example, the discreteness of the electric charge leads to fluctuations in the tunneling current in an STM, known as shot-noise. Shot-noise measurements convey information about the correlations among the electrons in condensed matter systems, such as the effective charge of the carrier or their distribution in the tunneling process. However, other contributions to the measured noise, such as 1/f and thermal noise, make it difficult to isolate the shot-noise component. For this reason, we use custom-built electronics that allows us to read out the noise signal of the STM at high frequencies and cryogenic temperatures and that does not interfere with conventional STM measurements.

O 58.15 Wed 18:00 P4

Optimization of a Simple Electrospray Deposition Device — •Ken Kolar[1], Miriam Meyer[2], Henrik Siboni[1], Christophe Nacci[1], Grant Simpson[1], and Leonhard Grill[1] — [1]Institute for Chemistry, Department of Physical Chemistry, University of Graz, Austria — [2]Institute for Ion Physics and Applied Physics, University of Innsbruck, Austria

A commercially available design of an electrospray deposition apparatus was optimized for cleaner and more controllable deposition in a high vacuum. The apparatus consists of a series of 5 differentially pumped chambers separated by skimmer cones or apertures. An angle-adjustable bellow was introduced between the second and third pumping chamber for better control over the alignment and, consequently, the ion beam flux reaching the final stage. Monitoring the flux/alignment was done by current measurements around the apertures and behind the last one with a conductive probe. Also, heating of the transfer capillary and enclosing the emitter-transfer capillary interface in a small transparent chamber (to allow different ambient gas environments) were introduced. The first depositions were done to test the performance of the improved design with some well-studied molecules.