

Poster Abstracts

Ultrahigh Out-of-Plane Piezoelectricity in 2D monolayers

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The simultaneous occurrence of gigantic piezoelectricity and Rashba effect in two-dimensional materials are unusually scarce. Inversion symmetry occurring in MX_3 ($\text{M}=\text{Ti, Zr, Hf}$; $\text{X}=\text{S, Se}$) monolayers is broken upon constructing their Janus monolayer structures MX_2Y ($\text{X}\neq\text{Y}=\text{S, Se}$), thereby inducing a large out-of-plane piezoelectric constant, d_{33} (~ 68 pm/V) in them. d_{33} can be further enhanced to a super high value of ~ 1000 pm/V upon applying vertical compressive strain in the van der Waals bilayers constituted by interfacing these Janus monolayers. Therefore, d_{33} in these Janus transition metal tri-chalcogenide bilayers reach more than four-fold times that of bulk ceramic PZT material (~ 268 pm/V). The absence of horizontal mirror symmetry and the presence of strong spin-orbit coupling (SOC) cause Rashba spin splitting in electronic bands in these Janus 2D monolayers, which shows up as an ultrahigh Rashba parameter, $\alpha_R \sim 1.1$ eVÅ. It can be raised to 1.41 eVÅ via compressive strain. Most of the 2D materials reported to date mainly show in-plane electric polarization, which severely limit their prospects in piezotronic devices. In this present work, the piezoelectricity shown by the Janus monolayers of Group IV transition metal tri-chalcogenides and their bilayers is significantly higher than the ones generally utilized in the form of three-dimensional bulk piezoelectric solids, e.g., α -quartz ($d_{11} = 2.3$ pm/V), wurtzite-GaN ($d_{33} = 3.1$ pm/V), wurtzite-AlN ($d_{33} = 5.6$ pm/V). It is exceedingly higher than that in Janus multilayer/bulk structures of Mo and W based transition metal dichalcogenides, e.g., MoSTe ($d_{33}\sim 10$ pm/V). The 2D Janus transition metal trichalcogenide monolayers and their bilayers reported herewith straddle giant Rashba spin splitting and ultrahigh piezoelectricity, thereby making them immensely promising candidates in the next generation electronics, piezotronics, spintronics, flexible electronics and piezoelectric devices. We have also performed a high-throughput computational screening for two-dimensional (2D) piezoelectric materials. From the existed 2D materials database, we have screened 252 materials which show piezoelectricity out of 1000 stable 2D materials.

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Machine Learning-Enabled Prediction of Electronic Properties of Radical-Containing Polymers at Coarse-Grained Resolutions

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Non-conjugated radical-containing polymers are a class of charge-carrying polymers that rely on pendant stable radical sites to transport charges successfully. They constitute promising materials for applications in all-organic energy storage or memory devices. The properties of these and other soft electronic materials depend on the coupling of electronic and conformational degrees of freedom over a wide range of spatiotemporal scales. Predictive modeling of such properties requires multiscale approaches that efficiently connect quantum-chemical calculations to mesoscale coarse-grained (CG) methodologies.

We present an efficient computational scheme that leverages supervised machine learning (ML) to predict electronic-structure information pertaining to charge transport at CG resolutions. ML models are trained on data coming from quantum-chemical calculations on all-atom conformations sampled from condensed-phase simulations. Predictions of electronic couplings, spin densities, and energy levels are studied as a function of CG mapping and resolution. Trained ML models subsequently enable electronic property predictions directly from CG polymer morphologies. We evaluate both systematic and building-block coarse-graining techniques. We validate the approach by comparing to the standard methodology that requires backmapping to atomistic resolution and subsequent quantum-chemical calculations. As such, the proposed ML-assisted scheme drastically accelerates multiscale computational workflows that connect electronic properties to mesoscale morphological features, thereby enabling high-throughput modeling aimed at the understanding and rational design of radical-containing polymers and other soft electronic materials.

GreenX

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The *GW* approximation has become a popular method for predicting quasiparticle excitations in both solids and molecules alike, due to its high accuracy with moderate computational effort [1, 2]. Direct implementation of the *GW* method scales as $O(N^4)$ with system size, where both the dielectric function and the correlation self-energy are formulated in terms of a product basis of Kohn-Sham wave functions [3]. We propose an efficient LAPW implementation of *GW* in the all-electron code, *exciting* [4], based on the space-time method [5], which will reduce the scaling of *exciting*'s G_0W_0 method to $O(N^3)$.

This work is part of a larger effort towards developing an excited-state library, GreenX, under the NOMAD CoE. Like other libraries in the CoE, this will be a mix of code-agnostic implementations, which can be shared between code families with different basis-set types, and code-specific features. Abstraction of the most computationally-intensive routines required for excited-state calculations into an efficient library, and the pooling of developer resources through the CoE, underpins the broader ambition of migrating the code families towards excited-states simulations on exascale hardware.

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- [3] Jiang, Hong, et al. „FHI-gap: A *GW* code based on the all-electron augmented plane wave method.“ *Computer Physics Communications* 184.2 (2013): 348-366.
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Denoising Autoencoder Trained on Simulation-Derived Structures Reveals Tetranucleosome Motifs in STEM Images of Chromatin

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Advances in scanning electron microscopy have led to an increase in the quantity and quality of imaging data. While existing algorithms can determine descriptive physical parameters, resolvability remains a challenge for many of these methods. Therefore, there is a need for data-driven approaches that reduce noise and allow for accurate structural predictions from experimental images. ChromSTEM, a combination of DNA-specific staining in scanning transmission electron microscopy, as allowed for the 3D study of genome organization. By leveraging convolutional neural networks and molecular dynamics simulations, we have developed a denoising autoencoder (DAE) capable of providing nucleosome-level resolution. Our DAE is trained on synthetic images generated from simulations of the chromatin fiber using the τ CPN model of chromatin. We find that our DAE is capable of removing noise commonly found in high angle annular dark field (HAADF) STEM experiments and is able to learn structural features driven by the physics of chromatin folding. We find that our DAE outperforms other well-known denoising algorithms without degradation of structural features. In addition, we are able to identify several tetranucleosome motifs and observe the absence of a 30-nm fiber, which has been suggested to serve as the higher-order structure of the chromatin fiber.

Thermodynamics of Ionic Bonding with Crown Ethers in Aqueous Solution

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The usage of Lithium for powering consumer electronics has increased its extraction from the land. However, the extraction process is complicated and polluting, and therefore, it has encouraged interest in Lithium recycling methods. In this work, Molecular simulations of aqueous solutions of three different crown ethers (12-Crown-4, 15-Crown-5 and 18-Crown-6) have been performed using Molecular Dynamics to explore their binding processes to Li^+ . Free energy calculations with umbrella sampling were used to compute the potential of mean force PMF of each system. The simulations were carried out on GROMACS using the GAFF force field and the explicit-solvent model TIP3P for water. In addition, the binding processes of the crown ethers to Na^+ were also examined for comparison.

Infrared-active acoustic-optical phonon modes in two dimensional organic/inorganic interfaces

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The growing interest in hybrid inorganic/organic interfaces formed by conjugated molecules adsorbed on transition-metal dichalcogenide monolayers has stimulated a number recent studies on the electronic properties of these compounds^{1,2,3}. However, little is known about the phonon characteristics of these hybrid materials, which are crucial for a full understanding of their photophysical properties. In this work, we use first-principles calculations based on density functional (perturbation) theory, to study the hybrid system formed by the pyrene molecule adsorbed on either MoS₂ or MoSe₂ monolayers. We find that, in these hybrid materials, new phonon modes are activated, compared to those of the individual constituents and play a crucial role to determine some of the main prominent optical peaks in the infrared spectra. Furthermore, we analyze the coupling modes that appear due to the hybridisation.

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- [2] D. Sercombe, S. Schwarz, O.D. Pozo-Zamudio, F. Liu, B.J. Robinson, E.A. Chekhovich, I.I. Tartakovskii, O. Kolosov, and A.I. Tartakovskii, Optical investigation of the natural electron doping in thin MoS₂ films deposited on dielectric substrates, *Scientific Reports* (2013).
- [3] Jannis Krumland and Caterina Cocchi, *IOP 2021 Electron. Struct.* 3 044003

Long Range Interactions in Atomistic Machine Learning

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Machine learning (ML) based methods to study materials at the atomistic scale have experienced a surge in interest in the last couple of years. One central assumption in many models for atomistic simulations is locality: the behavior of an atom primarily depends on its neighborhood. This is the assumption that leads to more efficient algorithms, making the methods applicable to large systems that could only be studied using classical physics, as opposed to quantum mechanical approaches like density functional theory, effectively bridging the two worlds.

Despite of the successes, effects including Debye-Hückel screening or nonlocal charge transfers cannot be captured in this way. The long distance equivariant (LODE) representation, first introduced in 2019, allows one to create ML models that go beyond this approximation. The approach as well as its implementation are strongly inspired by the Ewald based techniques in conventional molecular dynamics (MD).

We will present an overview of recent developments on how to incorporate long range interactions in atomistic ML models. Starting from extensions of the original LODE framework, we will demonstrate preliminary results showing the use of these descriptors, as well as the challenges that arise when combining this approach with traditional MD.

Ab-initio Prediction of Adsorption Isotherms of Water with a Metal- Organic Framework

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The interaction of water with the metal organic framework $\text{Mg}_2(2,5\text{-dioxido-1,4-benzenedicarb-oxylate})$ (MOF-74-Mg), will be studied using chemically accurate quantum chemical methods. Adsorption structures with one, two and five H_2O molecules per Mg^{2+} site will be investigated. The calculated isotherms will be compared with experimental data.

Subsequently, the formation of defects in the crystalline structure by hydrolysis will be studied to understand the wide variations of experimental isotherms. The calculations will use a hybrid QM:QM approach, which provides coupled cluster corrections to density functional theory and has been shown to yield chemical accuracy for molecule-surface interactions.

Empowering Carbon & Energy Catalysis with HPC, Data, AI and the Cloud

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Catalytic processes and materials play a key role in enabling power-2-X (P2X) transformations. C2CAT specializes in the design of customized catalysts for crucial P2X transformations such as the electrocatalytic nitrogen reduction reaction and conversion of CO₂, using proprietary technology which boosts the catalytic activity. Our approach is rooted in fundamental research on design and discovery of new materials using state-of-the-art computational modelling and simulations techniques. We implement and commercialize these multi-scale discoveries in real-world applications through synthesis of novel materials and their scale-up via various unique technologies.

This poster will discuss some of the key P2X applications that leverage from multiscale modelling of catalysis combining quantum chemistry with microkinetic modelling, empowered via artificial intelligence (AI), and high-performance computing (HPC) powered over the cloud.

High-throughput ab-initio calculations as a tool for screening tabulated experimental data

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In this contribution we present calculated band gaps for a set of ca. 250 semiconductors for which experimental band gaps are available in standard tabulations [1]. Calculations were performed with the LAPW code WIEN2k [2], using the PBE and modified Becke-Johnson (mBJ) functional. Correlations between the two sets of calculations and experiment are presented and discussed, as well as correlations between the two sets of calculations themselves. It is shown to which extent such an approach can be used to identify “suspicious” entries in tabulated experimental data.

[1] CRC Handbook of Chemistry and Physics, 91st Edition, CRC Press, William M. Haynes, National Institute of Standards and Technology, Boulder, Colorado, USA

[2] P. Blaha et al., WIEN2K, An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties (Karlheinz Schwarz, Techn. Universitaet Wien, Austria), 1999. ISBN 3-9501031-1-2

Uncertainty Modelling for Property Prediction of Double Perovskites

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Statistical predictive models for double perovskite properties are of high interest, because the perovskite structure allows relatively accurate property prediction and at the same time provides enough flexibility to yield a huge number of different materials of which some are likely relevant for important applications. While promising performance results have been published for this class of materials, they typically refer only to the predictive performance as, e.g., measured by the root mean squared error. However, active learning strategies for effective materials screening rely critically not only on accurate predictions but also on adequate uncertainty estimates as provided by probabilistic models.

Here, we study the predictive performance of two popular machine learning models, Gaussian processes and random forests, together with the quality of their uncertainty estimates. This study is based on a dataset of over 800 single (ABO₃) and double (A₂BB'O₆) cubic perovskite oxides with computed bulk modulus, cohesive energy, and bandgap. As a first observation, we show that random forests outperform Gaussian processes in terms of predictive performance for all three properties.

Moreover, we show that Gaussian processes, while providing sound Bayesian uncertainty estimates, can have inferior performance when their assumption of isometric smoothness of the target property is not met. In this case, as exemplified by the double perovskite bandgaps, random forests provide a better alternative, despite their rather ad-hoc uncertainty estimates. Improving these estimates thus appears to be a promising direction for future research, potentially leading to substantially improved procedures for computationally efficient sequential candidate screening.

Periodic Coupled-cluster Theory and Applications: Interface to FHI-aims and VASP

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Coupled-cluster (CC) theory achieves a high level of accuracy for a wide range of properties in atoms and molecules at affordable computational cost and is generally considered the "gold-standard" in quantum chemistry. However, only few codes exist for CC calculations of periodic systems. The Cc4s code[1] allows for periodic CCSD(T)-level calculations and it has been interfaced to VASP[2] and, recently, to FHI-aims[3, 4]. Together these interfaces, which cover atom-centered basis and plane-wave codes, can be generalized to interface most other ab initio software packages to Cc4s. This enables a wide range of applications ranging from molecular systems in the gas phase to solids and surfaces. The numerical heavy lifting in Cc4s is outsourced to a tensor contraction framework, which currently is the Cyclops Tensor Framework[5]. However, a generalized interface has been implemented, so that in principle any parallel tensor contraction library can be used by Cc4s. Using this strategy, it has been possible to compute systems with over 300 electrons and about 2000 virtual states. With the exploitation of the block-sparsity of tensors in periodic Coupled-cluster theory, which is currently being implemented in Cc4s, it is expected that conventional Coupled-cluster calculations with up to a 1000 electrons will become accessible.

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[2] Kresse et al. (1996). "Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set". *Computational materials science* 6 (1), 15–50. [https://doi.org/10.1016/0927-0256\(96\)00008-0](https://doi.org/10.1016/0927-0256(96)00008-0).

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AI with Experimental and Theoretical Data toward the Understanding CO₂ Hydrogenation Catalysis: The Role of the Support Materials

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The genesis of organic molecules from CO₂ at a hydrothermal vent, which is a fissure on the seafloor, is one of the theories for the origin of life [1]. We focus on CO₂ hydrogenation catalyzed by cobalt nanoparticles supported on SiO₂, which mimic the environment of a hydrothermal vent. In particular, we investigate the role of support materials by using several amorphous SiO₂ supports incorporating different elements (e.g., Ti, Zr). In this study, the experimental selectivity toward organic molecules (e.g., methanol, formic acid) is modeled by the sure-independence screening and sparsifying operator (SISSO) AI approach [2]. Our approach identifies the key descriptive parameters correlated to the selectivity, which lead to a better understanding of the origin of life and to design of novel CO₂ hydrogenation catalysts.

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[2] R. Ouyang et al., Phys. Rev. Mater., 2, 083802 (2018)

Hydrogen adsorption on Pd surfaces and its effect on CO₂ activation

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An accurate description of the surface of Pd-based catalysts under reaction conditions is a critical step toward a deeper understanding of catalyst reactivity. Herein, by modeling the phase diagram of the (111) and (100) surfaces of face-centered cubic Pd via ab initio atomistic thermodynamics, we predict the stable hydrogen coverages for a wide range of temperatures and H₂ pressures. The hydrogen coverage at the experimental conditions used for CO₂ hydrogenation plays a major role in the reactivity, as it hinders the chemisorption of activated CO₂. The calculated data will be the basis for subsequent subgroup-discovery analysis on CO₂ activation.

Surface reconstructions and diffusion properties of beta-Ga₂O₃ surfaces from first principles

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This project is part of the Berlin-centered Leibniz ScienceCampus “Growth and Fundamentals of Oxides (GraFOx) for electronic applications” which combines the expertise of its eight partner institutions in the field of oxide research. The primary focus is on synthesizing oxides to the highest material quality and thoroughly studying them in terms of their surface structure, microstructure, and optical as well as optoelectronic properties.

The transparent conducting oxide Ga₂O₃, exhibiting a band gap of about 4.9eV, is a very promising candidate for several applications, such as semiconducting lasers and transparent electrodes for UV optoelectronic devices and solar cells. As such, the bulk properties of its thermodynamically stable β phase have been extensively studied in the last two decades. The surface properties, however, playing a vital role in epitaxial growth, electrical contacts, and gas sensors are still not well understood.

In this project, we study the reconstructions of β -Ga₂O₃(001) in realistic conditions from first principles. With the replica-exchange grand-canonical (REGC) approach [1], we screen for possible surface reconstructions in a reactive oxygen atmosphere in an unbiased way which includes vibrational contributions with full anharmonicity. The obtained metastable structures are used to construct a surface phase diagram using conventional ab initio atomistic thermodynamics (aiAT). By combining the two approaches, we can identify two novel 1x2 reconstructions that exhibit a region of stability in the phase diagram. Our structural model is consistent with STEM images of homoepitaxially-grown (001) films. We also illustrate our current work on the diffusion of gallium and oxygen atoms on the (001) surface. We outline how we will combine the obtained energy barriers, machine learning, and kinetic Monte Carlo methods to model the growth of group-III sesquioxides alloys by molecular beam epitaxy.

[1] Y. Zhou, M. Scheffler, L. M. Ghiringhelli, Phys. Rev. B 100, 174106 (2019).