Poster List 2023 of The NOMAD Laboratory

Recent work of *The NOMAD Laboratory* is displayed on 12 posters

In the Communication Room of the MP (Molecular Physics) Department

Poster Location Index *Poster Title and Authors*

NOMAD #1 **All-Electron Large-Scale Density Functional Simulations with FHI-aims**

Sebastian Kokott, Florian Merz, Christian Carbogno, Andreas Marek, Peter Karpov, Mariana Rossi, Markus Rampp, Volker Blum, and Matthias Scheffler

NOMAD #2 **Towards Robust Periodic *GW* Calculations with Numeric Atom-Centered Orbitals in FHI-aims**

Min-Ye Zhang, Florian Merz, Uthpala Herath, Markus Rampp, Volker Blum, Xinguo Ren, Matthias Scheffler

NOMAD #3 **Periodic Coupled-Cluster Theory for the Ground and Excited States with Atom-Centered Basis Functions**

Evgeny Moerman, Felix Hummel, Andreas Irmler, Alejandro Gallo, Andreas Grüneis, and Matthias Scheffler

NOMAD #4 **Artificial-Intelligence-Driven Discovery of Materials with Exceptional Performance**

Lucas Foppa, Mario Boley, Felix Luong, Simon Teshuva, Daniel Schmidt, and Matthias Scheffler

NOMAD #5 **Artificial-Intelligence-Guided Workflows for Exploring Materials Space**

Thomas A. R. Purcell, Yi Yao, Sebastian Eibl, Markus Rampp, Luca M. Ghiringhelli, Christian Carbogno, and Matthias Scheffler

NOMAD #6 **The Max-Planck-Cardiff Centre on the Fundamentals of Heterogeneous Catalysis (FUNCAT): Advancing Critical Chemical Processes for a Sustainable Future**

Ray Miyazakia, Herzain I. Rivera-Arrieta, Kendra Belthle, Jonathan Mauß, Igor Kowalec, Andrew Logsdail, David Willock, Richard Catlow, Michael Bowker, Graham Hutchings, Harun Tüysüz, Ferdi Schüth, Lucas Foppa, and Matthias Scheffler

NOMAD #7 **AI-Driven Workflows for the Discovery of Water Splitting Catalysts Based on DFT with Non-local Exchange-Correlation**
Akhil Sugathan Nair, Osama Jalil, Lucas Foppa, Evgeny Moerman, Andreas Gruneis, Stefano Americo, Kristian Sommer Thygsen, and Matthias Scheffler

NOMAD #8 **Accelerating Training of Machine-Learned Interatomic Potentials for Strongly Anharmonic Materials through Active Learning**
Kisung Kang, Christian Carbogno, and Matthias Scheffler

NOMAD #9 **Anisotropic Thermal Conductivity Studied with the *ab initio* Green-Kubo Approach: the Example of Ga_2O_3**
Shuo Zhao, Thomas A. R. Purcell, Kisung Kang, Christian Carbogno, and Matthias Scheffler

NOMAD #10 **Non-perturbative Electronic Transport for Strongly Anharmonic Materials**
Jingkai Quan, Christian Carbogno, and Matthias Scheffler

NOMAD #11 **Towards an *ab initio* Kinetic Monte Carlo Model for The Growth of β-Ga_2O_3 (100)**
Qaem Hassanzada, Konstantin Lion, Claudia Draxl, and Matthias Scheffler

NOMAD #12 **Artificial Intelligence Discovery of Novel Memristor Materials**
Wahib Aggoune and Matthias Scheffler

All-Electron Large-Scale Density Functional Simulations with FHI-aims

Sebastian Kokott, Florian Merz[a], Christian Carbogno, Andreas Marek[b], Peter Karpov[c], Mariana Rossi[d], Markus Rampp[b], Volker Blum[e], and Matthias Scheffler

FHI-aims (Fritz Haber Institute *ab initio* molecular simulations) is a versatile electronic-structure code designed for computational research in molecular and materials science. This software is employed by a global community of developers, researchers at the Fritz Haber Institute, academic institutions, and industrial sectors. FHI-aims utilizes numeric atom-centered basis sets, providing computational precision that rivals the best benchmark codes in density functional and many-body theory. Notably, it achieves this precision while maintaining computational efficiency comparable to plane-wave pseudopotential methods. The code demonstrates remarkable applicability, routinely handling systems comprising thousands of atoms with semi local and hybrid density functionals. Additionally, it exhibits excellent scalability on modern high-performance computing platforms. FHI-aims boasts advanced electronic-structure capabilities for both molecules and solids and seamlessly integrates into complex simulation environments. This integration includes the ability to serve as a parallel library accessible through Python or via internet sockets or through its graphical user interface GIMS, making it a powerful tool for a wide range of scientific investigations. Here, we focus on the two main bottlenecks of large-scale hybrid density functional simulations: The computation of the Hartree-Fock exchange and the solution of the generalized eigenvalue problem. The localized resolution-of-identity approach enables O(N) hybrid density functional simulations and, thus, the computation of accurate electronic properties of large-scale atomistic models in the range of ten thousand of atoms in FHI-aims. In this range, parallelization and memory requirements of the exact exchange part, and the evaluation of the Hartree potential remain challenging. The solution of the generalized eigenvalue problem with direct eigensolvers like ELPA naturally becomes a computational bottleneck due to O(N^3) scaling. In this work, we present recent algorithmic advancements for the exact exchange part and the evaluation of the Hartree potential, as well as optimizations of the ELPA library. We systematically perform benchmarks tests on CPU and GPU-accelerated architectures covering inorganic solids, large molecules, and organic crystals with up to 30,000 atoms.

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Towards Robust Periodic *GW* Calculations with Numeric Atom-Centered Orbitals in FHI-aims

Min-Ye Zhang, Florian Merz[a], Uthpala Herath[b], Markus Rampp[c], Volker Blum[d], Xinguo Ren[e], Matthias Scheffler

The electronic band structure plays a fundamental role for the electrical, electronic and optical properties of materials. In terms of first-principles simulations, an accurate description of the electronic structure of weakly correlated systems is provided by the many-body perturbation *GW* approach. For a plane-wave basis and pseudo-potentials, the approach has been implemented and used for decades. [1] However, an all-electron full-potential treatment is more accurate. Such method was implemented for clusters in FHI-aims [2] using compact numeric atom-centered orbitals (NAOs), [3] based on the resolution-of-identity (RI) technique, and was benchmarked in the *GW*100 molecular set. [4] Recently, the one-shot variant of *GW* approach (G_0W_0) in FHI-aims was extended to periodic systems. [5] Here the localized RI approximation (LRI) [6] is applied, but a thorough benchmarking of accuracy and efficiency is still needed.

In this poster, we present our recent efforts to benchmark and improve the *GW* functionality for periodic systems in FHI-aims. After sketching the current algorithmic implementation, we benchmark the efficiency with strong scaling of time and memory for unit and super cells. In terms of accuracy, we investigate the error due to the localized approximation of RI by carefully converging the band gap of 40 gapped systems with respect to the auxiliary basis functions (ABFs) on top of the NAO presets in FHI-aims. A robust choice of additional functions to build ABFs is proposed based on the convergence test. Furthermore, code improvements to facilitate an out-of-the-box and user-friendly experience are introduced briefly, including an implementation of dry run with memory estimate. It is particularly useful for systems containing large number of atoms (~10^2) or with dense Brillouin zone sampling.

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Periodic Coupled-Cluster Theory for the Ground and Excited States with Atom-Centered Basis Functions

Evgeny Moerman, Felix Hummel[a], Andreas Irmler[a], Alejandro Gallo[a], Andreas Grüneis[a], and Matthias Scheffler

In recent years, the periodic formulation of coupled-cluster (CC) theory has proven itself repeatedly as a reliable and highly accurate benchmark electronic structure method in materials science [1-3]. The all-electron code FHI-aims [4], which employs numeric atom-centered orbitals, has recently been interfaced to the CC theory for solids (CC4S) code [5] via the CC-aims library [6], making CC theory for both the ground state and for excited states in the equation-of-motion coupled-cluster (EOM-CC) theory accessible to FHI-aims. In particular, the EOM-CC framework allows for the systematic inclusion of electronic correlation in the calculation of band gaps and band structures [7] and has been shown to predict quasi-particle energies more accurately and reliably than the GW approximation in the molecular case [8].

To fully capitalize on the utility of the CC and EOM-CC method, however, two sources of error have to be addressed: Firstly, periodic coupled-cluster theory – like most correlated wave function methods – exhibit excessively slow convergence to the thermodynamic limit. Even though a very powerful finite-size correction technique based on the transition structure factor has been formulated for plane waves [9], an equally effective treatment for atom-centered basis sets does not exist yet.

Secondly, wave function methods suffer from equally slow convergence to the complete basis set (CBS) limit. While this can be effectively addressed by employing basis sets that can be extrapolated to the CBS, convergence with respect to the auxiliary basis of the localized resolution-of-identity (RI-LVL) scheme employed in FHI-aims needs to be ensured.

As an all-electron code, FHI-aims treats core- and valence electrons on equal footing without resorting to approximations as they are encountered in pseudopotentials or the projector augmented wave method, which potentially can contribute to an erroneous description of the electronic structure. We present the current state of the coupled-cluster theory framework available in FHI-aims and possible avenues to address the auxiliary basis- and finite-size error.

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Artificial-Intelligence-Driven Discovery of Materials with Exceptional Performance

Lucas Foppa, Mario Boley[a], Felix Luong[a], Simon Teshuva[a], Daniel Schmidt[a], and Matthias Scheffler

Artificial intelligence (AI) has the potential to revolutionize the design of materials by uncovering correlations and complex patterns in data. However, current AI methods attempt to describe the entire, immense materials space with a single interpolation-based model, while different mechanisms govern the materials behaviors across the materials space. In this poster, we highlight recent advances on the subgroup-discovery (SGD) approach that enable the efficient identification of rules that can focus on mechanisms leading to exceptional performance. In particular, we discuss the notion of SG exceptionality and analyze the tradeoff between exceptionality and generality based on a Pareto front of SGD solutions. Additionally, we analyze challenges in AI-driven sequential materials-property optimization frameworks, namely the need for sound uncertainty estimates and for modelling the relationship between features and property in a spatially adaptive way. We propose measures of model performance that can be computed on benchmark datasets and that correlate better with the expected reward of the discovery process than the traditional measures based on average predictive performance.

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Artificial-Intelligence-Guided Workflows for Exploring Materials Space

Thomas A. R. Purcell, Yi Yao, Sebastian Eibl, Markus Rampp, Luca M. Ghiringhelli, Christian Carbogno, and Matthias Scheffler

Accurate artificial-intelligence (AI) models are key to accelerate the discovery of new functional materials for myriad applications. However, advancements in this field are often hindered by the scarcity of available data and the significant effort required to acquire new data. For such applications, reliable surrogate models that help guide materials space exploration using easily accessible materials properties are urgently needed. Here, we present a general, AI-guided workflow framework for materials discovery, significant advancements to the portability of the symbolic regression methods, and apply them to find new thermal insulators. The framework provides both quantitative predictions and qualitative rules for steering data creation using a combination of symbolic regression and sensitivity analysis. We first use the sure-independence screening and sparsifying operator (SISSO) [1, 2] to build an analytical model that describe thermal conductivity of a material and then extract out the most important input properties using a variance-based sensitivity analysis [3]. The SISSO results highlight the improvements to the algorithms using the performance-portability framework Kokkos to offload the high-cost parts of the code onto accelerators such as graphics processing units (GPUs), getting a factor of fifteen speed up to the code. Using the information gained from the analysis we screen over a set of 732 materials and find the region of space most likely to contain strong thermal insulators, and confirm four of these predictions by calculating their thermal conductivity using the *ab initio* Green-Kubo technique [4, 5]. These results suggest the general applicability of this approach to other fields including catalysis. Finally, we demonstrate how we can use active learning to further accelerate the discovery of new materials.

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The Max-Planck-Cardiff Centre on the Fundamentals of Heterogeneous Catalysis (FUNCAT): Advancing Critical Chemical Processes for a Sustainable Future

Ray Miyazaki[a], Herzain I. Rivera-Arrieta, Kendra Belthle[b], Jonathan Mauß[b], Igor Kowalec[c], Andrew Logsdail[c], David Willock[c], Richard Catlow[c], Michael Bowker[c], Graham Hutchings[c], Harun Tüysüz[b], Ferdi Schüth[b], Lucas Foppa, and Matthias Scheffler

Heterogeneous catalysis involves a complex and intricate interplay of several underlying processes taking place at different space and time scales. This makes the atomistic modeling and design of improved catalytic materials extremely challenging. By bringing together the expertise of different Max Plank Institutes (MPI), namely the MPI for Chemical Energy Conversion, the MPI für Kohlenforschung, and the Fritz Haber Institute, with the one from the Cardiff Catalysis Institute (CCI) in the UK, the FUNCAT collaboration aims to advance the rational design of heterogeneous catalysts and tackle grand challenges on the field [1]. Within this partnership, we exploit the fusion of theoretical and experimental data through Artificial Intelligence (AI) to reveal nontrivial correlations between key descriptive parameters, characterizing the materials and relevant underlying processes. These key parameters were called “materials genes" [2] in analogy to biological systems. Although such parameters might not explicitly uncover the mechanism behind the material’s performance, they describe the materials behavior in a similar manner as certain human genes relate to the color of our eyes or possible health issues. Therefore, the identification of the “materials genes”, which are obtained by experiment or calculation, can accelerate the design of catalysts for critical chemical processes. Specifically, we will present our results for the CO_2 reduction [3] and the selective hydrogenation of acetylene.

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AI-Driven Workflows for the Discovery of Water Splitting Catalysts Based on DFT with Non-local Exchange-Correlation

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The water splitting reaction is crucial for the sustainable production of hydrogen (H2) from renewable energy sources [1]. However, an efficient water splitting process requires materials with appropriate surface properties to catalyze the sluggish oxygen evolution reaction (OER). Many of the known oxide-based catalysts lack stability under harsh aqueous conditions and are not earth-abundant [2]. The computational discovery of oxides is hindered by the challenging description of their electronic structure and vast compositional and structural space [3]. Here, we conduct an accelerated discovery of oxide catalysts for water splitting by employing the recent NOMAD CoE developments in Eigenvalue Solvers for Petaflop Application (ELPA) libraries, hybrid functionals [4] and artificial intelligence (AI) workflows. We perform non-local DFT calculations for computing the stability of oxides using the recent Heyd–Scuseria–Ernzerhof (HSE) exchange–correlation functional implementation in FHI-aims package [5]. We develop a hierarchical symbolic regression sequential learning (HSL) method which guarantees the effective utilization of data available at different fidelities. The application of sure-independence screening operator (SISSO) approach leverages the limited amount of high-fidelity data [6]. We develop a unified computational workflow based on Atomic Simulation Recipes (ASR) which integrates AI and DFT calculations for the automated discovery of stable oxides [7]. The combination of the HSL method with an efficient HSE implementation will facilitate the discovery of water splitting catalysts.

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Accelerating Training of Machine-Learned Interatomic Potentials for Strongly Anharmonic Materials through Active Learning

Kisung Kang, Christian Carbogno, and Matthias Scheffler

Machine-learned interatomic potentials (MLIP) promise to conduct highly efficient molecular dynamics (MD) simulations with a level of accuracy comparable to *ab initio* methods. Recently, an exact and computationally efficient definition of the heat flux that incorporates semi-local interactions in MLIPs was proposed, paving the way to fully anharmonic MLIP-based thermal transport calculations [1]. However, this also requires that all strongly anharmonic effects are correctly captured by the MLIP, including rare events such as intrinsic defect formations and early signs of phase transitions [2]. To ensure that such infrequent events, often overseen by standard MLIP training approaches, are correctly incorporated, we devise an active learning approach. This combines MD simulations based on the MLIP (MLIP-MD) with a measure of uncertainty, enabling the identification of qualitative deviations from the training area of MLIP models, for which the model is iteratively improved Our investigations with $KCaF_3$ and CuI demonstrate noticeable improvements in training speed through the uncertainty evaluation of potential energy, atomic forces, and degree of anharmonicity [3]. The MLIP-MD method adeptly captures essential dynamic features, including anharmonicity measures along molecular dynamics trajectories, anharmonic vibrations, and the occurrence of rare anharmonic events. Furthermore, MLIP-MD enables MD simulations with larger supercells and longer simulation time, a capability that has been beyond the reach of *ab initio* MD. This capability opens an efficient avenue for investigating carrier transport in strongly anharmonic materials using the *ab initio* Kubo-Greenwood approach.

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Anisotropic Thermal Conductivity Studied with the *ab initio* Green-Kubo Approach: the Example of Ga_2O_3

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In practical applications, the anisotropy of thermal transport plays a fundamental role, since the thermoelectric figure-of-merit as well as the efficiency of heat dissipation crucially depends on the crystal axis along which heat is to be conducted [1]. Usually, such anisotropic effects are discussed in terms of a phonon picture. However, very little is known about how anharmonicity affects the anisotropy of thermal transport, despite the fact that such effects are known to break the phonon picture and fundamentally alter heat transport in strongly anharmonic materials [2]. To clarify this open question, we employ the *ab initio* Green-Kubo (aiGK) method, which accounts for all anharmonic effects through aiMD [3,4], and extend its applicability to anisotropic heat transport. We discuss the intricacies of anisotropic transport using Ga_2O_3 as an example, a promising candidate material for field-effect transistors. Our results reveal that the anisotropic thermal conductivity of Ga_2O_3 originates from the anisotropic coordination of Ga-O. In its α phase that only features octahedral coordination, an isotropic thermal transport is observed, whereas the β-, and κ-polymorphs exhibit distinct degrees on anisotropy due to the different ratios of octahedral and tetrahedral Ga-O coordination. While anisotropy remains largely unaffected by temperature effects, it is highly sensitive on the actual film thickness viz. on the maximum mean free path accessible in the sample. In this light, we discuss the optimal growth direction and film thickness to tailor and boost heat transport in nano- and microscale Ga_2O_3 devices.

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Non-perturbative Electronic Transport for Strongly Anharmonic Materials

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First-principle approaches for phonon-limited electronic transport are typically based on many-body perturbation theory [1] and thus rely on the validity of a quasi-particle picture for phonons and electrons. However, both these pictures can become questionable in strongly anharmonic systems [2,3]. We overcome this hurdle by combining *ab initio* molecular dynamics (aiMD) calculations with the Kubo-Greenwood (KG) formalism [4]. This non-perturbative, stochastic method allows us to account for all orders of anharmonic and vibronic couplings in the calculation of carrier mobilities. We discuss the implementation of this technique in the *ab initio* material simulation package FHI-aims [5] with particular focus on those numerical acceleration strategies that enable to obtain converged bulk mobilities, i.e., using Fourier interpolation for the Brillouin zone integration, efficient k-grid parallelization, and strategies to extrapolate to the direct current limit [6]. By this means, we are able to correctly reproduce and explain the temperature-dependent electron mobility of strongly anharmonic SrTiO3 at high temperatures.

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Towards an *ab initio* Kinetic Monte Carlo Model for The Growth of β-Ga_2O_3 (100)

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Ga_2O_3 is attracting much attention as a promising candidate for various types of applications including light-emitting diodes, solar cells, field-effect transistors, photodetectors, and gas sensors [1-4]. Significant advances have been made in controlling its growth quality, but the theoretical understanding is shallow. Experimentally, it has been observed that different Ga-to-O ratios lead to different growth modes in molecular beam epitaxy (MBE) [5], but the underlying mechanisms at the atomic level are unknown. It is experimentally confirmed that volatile suboxide (Ga_2O) desorption limits growth rates under Ga-rich conditions [6], but the desorption mechanisms of the suboxide are not understood. Furthermore, changes in substrate and growth conditions lead to different crystal phases [1]. However, a comprehensive understanding of the stabilization of these different phases remains elusive. For these reasons, it is important to understand the growth processes at the atomic level. In this project, we study the growth of Ga_2O_3 at its most stable surface, β-Ga_2O_3 (100). Using density functional theory (DFT) calculations with the exchange-correlation functional PBEsol, we investigated all stable adsorption sites for single Ga and O atoms as well as multi-atom clusters (up to 5 adatoms) at the surface for the two stoichiometric terminations, (100)-A and (100)-B. We find that the stable sites for single adatoms are drastically different from the stable sites for two-atom and multi-atom clusters. We also calculated the energy barriers for all possible diffusion paths. Our findings reveal that Ga adatoms freely diffuse along the crystallographic b direction, with energy barriers of less than 0.14 eV for the B termination and 0.17 eV for the A termination. While, O adatoms face notably higher energy barriers when diffusing along the b direction, reaching up to 0.56 eV for the B termination and 0.28 eV for the A termination. When considering diffusion along the crystallographic c direction, both Ga and O adatoms encounter even higher energy barriers. For Ga adatoms, these barriers reach up to 0.37 eV (0.46 eV) for the B (A) termination, while for O adatoms, they can be as high as 1.41 eV (1.9 eV) for the B (A) termination. Additionally, we investigated the diffusion of Ga-O pairs and calculated associated energy barriers. These are the base for a comprehensive *ab initio* kinetic Monte Carlo (kMC) model of Ga_2O_3 growth.

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Artificial Intelligence Discovery of Novel Memristor Materials

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Memristor [1] is a non-volatile resistive switch (RS) where the resistivity of the material can be programmed upon applying a voltage [2]. Its ability to store and process digital information with fast operation speed and low energy consumption makes it a key for neuromorphic and other in-memory computers [3]. Such devices can potentially overcome a current bottleneck and enable a hardware breakthrough. However, in some cases, scaling down the dimensions of memristive devices has been limited by high leakage currents, thus hampering further progress [4]. Recently, the RS property was observed in some 2D materials, paving the way to develop novel memristors with extremely small size (atomristor) [5, 6]. Our goal is to benefit from the broad platform provided by the portfolio of 2D materials and discover the best candidates, by a systematic linkage of first-principles approaches and recent, successful artificial intelligence (AI) methods. In this poster, we present the workflow of our project which aim to search for the most promising 2D materials for atomristor applications. *Ab initio* approaches will be used to provide important materials characteristics and features. Then, the SISSO method [7], which is a combination of symbolic regression and compressed sensing, will be used to learn a predictive model of the RS as it is less data-intensive and (better) interpretable than traditional AI methods. Validation with respect to available experimental results (e.g. [6]) will be carefully considered as well. Overall, this project will provide guidance to industrial R & D to develop an efficient atomristor technology.

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