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The FAIRmat Project for Condensed-Matter Physics and the Chemical Physics of Solids

Claudia Draxl[a], Matthias Scheffler, and the whole FAIRmat team[b]

Prosperity and lifestyle of our society largely rely on improved or even novel materials that make new products possible for the energy, environment, health, mobility, IT sectors, and more. Materials Science has been producing for decades a huge amount of possibly extremely valuable data, which, however, are seldom re-used or even accessed outside the single project they were produced for. Besides, data that are not used for a publication are simply forgotten and wasted. In order to turn this gold mine of data into value, FAIR (Findable, Accessible, Interoperable, and Re-usable) data infrastructures are necessary. Even more, data need to be ready to be harvested, explored, and analyzed by means of artificial intelligence. In this sense, our forward-looking re-interpretation of the FAIR acronym is that research data should be "Findable and AI-Ready".

The *FAIRmat* consortium of the German Research-Data Infrastructure (NFDI) has been established in October 2021 with the mission of implementing a FAIR research-data infrastructure that interweaves data and tools from and for materials synthesis, experiment, theory, and computation [1, 2].

FAIRmat builds upon and expands the achievements of the first phase of the NOMAD Center of Excellence, which had established a FAIR data infrastructure for *ab initio* computational materials-science. The core of the infrastructure is the development and deployment of a hierarchical, modular, and extensible metadata schema, which maps the information contained in atomistic-simulation codes, stored in the *NOMAD Repository*, into a standardized representation, stored in the *NOMAD Archive*. The content of the *Archive* is accessed via a flexible API and can be browsed in the *NOMAD Encyclopedia* or analyzed with AI tools in the *NOMAD AI toolkit*. The NOMAD infrastructure, which is web based and operable without registration, has also a local counterpart, the *NOMAD Oasis*, which allows users to operate with exactly the same tools and interfaces on local, private data, also behind strong fire walls.

In this three-panel poster, we present the current results together with established achievements in the NOMAD Infrastructure and *FAIRmat* consortium. In particular, the challenge of representing complex workflows from synthesis, experiments, and computation is addressed and a roadmap is provided.

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The NOMAD (Novel Materials Discovery) Center of Excellence (CoE): Tackling Exascale Computing and Extreme-Scale Data

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Exascale computing will have a profound impact on everyday life in the coming decades. At 10^{18} operations per second, exascale supercomputers will be able to quickly analyze massive volumes of data and more realistically simulate complex processes. The goal of the *NOMAD Center of Excellence CoE* https://www.nomad-coe.eu/ is to bring computational materials science to the next level of supercomputing.

The *NOMAD CoE* assesses and exploits the characteristics of extreme-scale data and exascale computing for computational materials science, to enable investigations of systems of higher complexity (space and time), consideration of metastable states and temperature, and all this at significantly higher accuracy and precision than what is possible today. Systematic studies and predictions of novel materials to solve urgent energy, environmental, and societal challenges require such significant methodological advancements targeting the upcoming exascale computers. Key NOMAD examples are catalytic water splitting for hydrogen production and the transformation of waste heat into useful electricity.

The infrastructure developed with the *NOMAD CoE* builds on three pillars: "Codes" extends and develops *ab initio* computational materials science for entire code families to be able to exploit exascale for attacking new problem categories that are not feasible on today's top supercomputers. "Workflows" develops tools to manage high-throughput computations that take full advantage of exascale resources. "Big-Data Analytics" advances the existing big-data tools and brings them towards near-real-time performance. Here, we present the first achievements of the *NOMAD CoE*: upgrade of the ELPA eigensolver to the exascale, development of an interface to CC4S for coupled-cluster calculations in solids, creation of a libraries of "recipes" for massive *ab initio* calculations, applied to creation of a large data-set of vibrational calculations, and deployment of a massive parallel framework for symbolic regression calculations.

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FHI-aims: The *ab initio* Materials Simulation Package

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FHI-aims (Fritz Haber Institute *ab initio* materials simulations) is an all-electron, electronic-structure code used for computational molecular and materials research by a global community of developers and users in academia, and in industry [1]. Numeric atom-centered functions are used as basis sets, offering numerical precision on par with the best available benchmark codes in density-functional [2, 3] and many-body [4] theory, at a computational cost that is comparable with plane-wave pseudopotential methods. The code is routinely applicable to thousands of atoms using semi local and hybrid functionals and shows excellent scalability on current high-performance platforms. Further strengths include advanced electronic-structure developments for molecules and solids and seamless integrability into complex, externally managed simulation environments, including as a parallel library through python or using internet sockets. Here, we describe recent developments in FHI-aims and in open-source high-performance libraries on which the code relies.

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The TEC1p Project: Thermal and Electrical Conductivity from First Principles

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Heat and charge transport are ubiquitous phenomena in solid-state physics of paramount importance for both scientific and industrial applications. In particular, tailoring these transport coefficients and finding materials with optimal properties is a key step to enable thermoelectric waste-heat recovery devices as well as new-generation batteries and thus a transition to a sustainable energy economy.

To address these challenges, we have developed novel, non-perturbative methodologies that allow to assess transport coefficients from first principles [1, 2]. In contrast to existing formalisms, anharmonic effects are accounted for to all orders, hence allowing predictions with unprecedented accuracy [3]. In this poster, we review the fundamental physical concepts and their application to material science. Moreover, we discuss how such calculations can be accelerated so to enable a rapid, high-throughput screening through material space [4].

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Learning Interpretable Models with Artificial Intelligence

Thomas A. R. Purcell, Lucas Foppa, Luca M. Ghiringhelli[a], Christian Carbogno, and Matthias Scheffler

Artificial intelligence (AI) frameworks that are capable of creating reliable and interpretable models are paramount for discovering new functional materials. Here, we present the sure independence screening and sparsifying operator (SISSO) [1] and the subgroup discovery (SGD) [2] approaches. Both methods identify analytical equations (SISSO) or Boolean (SGD) expressions from a set of user-given physical parameters, which are able to model a chosen property. In particular we will highlight a new implementation of SISSO, SISSO++ [3], which provides not only a high-performance library for SISSO, but also a user-friendly python interface that facilitate interfacing SISSO into existing frameworks. Finally, we showcase the ability of both of these approaches by applying them to better understand thermal conductivity and catalysis.

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The BiGmax Project on Big-Data-Driven Materials Science

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BiGmax is a Max Planck Network of 10 Max Planck Institutes that addresses the various challenges of big-data-driven materials. It is coordinated by Peter Benner (Max Planck Institute for Dynamics of Complex Technical Systems, Magdeburg) and Matthias Scheffler [1]. *The NOMAD Laboratory* contributes with projects in heterogeneous catalysis (see poster #10) and together with Claudia Draxl on the *FAIRmat* project (see Poster #1).

This poster here describes our project on "crystal-structure identification via Bayesian deep learning". Due to their ability to recognize complex patterns, neural networks can drive a paradigm shift in the analysis of materials science data. Here, we introduce *ARISE*, a crystal-structure identification method based on Bayesian deep learning [2]. As a major step forward, *ARISE* is robust to structural noise and can treat more than 100 crystal structures, a number that can be extended on demand. While being trained on ideal structures only, *ARISE* correctly characterizes strongly perturbed single- and polycrystalline systems, from both synthetic and experimental resources. The probabilistic nature of the Bayesian-deep-learning model allows obtaining principled uncertainty estimates, which are found to be correlated with crystalline order of metallic nanoparticles in electron tomography experiments. Applying unsupervised learning to the internal neural-network representations reveals grain boundaries and (unapparent) structural regions sharing easily interpretable geometrical properties. This work enables the hitherto hindered analysis of noisy atomic structural data from computations or experiments.

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The Leibniz Science Campus on Growth and Fundamentals of Oxides for Electronic Applications (GraFOx)

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This project is part of the Berlin-centered Leibniz Science Campus "Growth and Fundamentals of Oxides (*GraFOx*) for electronic applications" which combines the expertise of its 8 partner institutions in the field of oxide research [1]. 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 Ga_2O_3, exhibiting a band gap of about 4.9 eV, is a very promising candidate for a number of 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 stability of all low-index β-Ga_2O_3 surfaces from first principles. Using *ab initio* atomistic thermodynamics, we show that (-201) faceting is energetically favored on (100) substrates, supporting results obtained during homoepitaxial growth on off-oriented substrates [2]. While all surface energies are slightly reduced by explicitly including harmonic vibrational contributions, the relative stability between the surfaces is not affected. We construct full phase diagrams for all surfaces to identify the stable phases in realistic temperature and pressure conditions. In addition, we illustrate our current work to simulate surfaces in a reactive oxygen atmosphere to find novel metastable structures and assess the effect of anharmonic vibrations on surface stability.

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Materials Genes of Heterogeneous Catalysis from Clean Experiments and Artificial Intelligence

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The performance in heterogeneous catalysis is an example of a complex materials function, governed by an intricate interplay of several processes (e.g., the different surface chemical reactions, and the dynamic restructuring of the catalyst material at reaction conditions). Modeling the full catalytic progression via first-principles statistical mechanics is impractical, if not impossible. Instead, we show how a tailored artificial-intelligence approach can be applied, even to a small number of materials, to model catalysis and determine the key descriptive parameters ("materials genes") reflecting the processes that trigger, facilitate, or hinder catalyst performance. We start from a consistent experimental set of "clean data" [1], containing nine vanadium-based oxidation catalysts. These materials were synthesized, fully characterized, and tested according to standardized protocols. By applying the symbolic-regression based SISSO approach, we identify correlations between the few most relevant materials properties and their reactivity. This approach highlights the underlying physicochemical processes, and accelerates catalyst design.

A poster discussing the experimental aspects of this work is shown in the AC Department.

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Learning Design Rules for Selective Oxidation Catalysts from High-Throughput Experimentation and Artificial Intelligence

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The design of heterogeneous catalysts is challenged by the complexity of materials and processes that govern reactivity and by the fact that the number of good catalysts is very small compared to the number of possible materials. Here, we show how the subgroup-discovery (SGD) artificial-intelligence approach can be applied to an experimental plus theoretical data set to identify constraints on key physicochemical parameters, the so-called SG rules, which exclusively describe materials and reaction conditions with outstanding catalytic performance [1]. By using high-throughput experimentation, 120 SiO_2-supported catalysts containing ruthenium, tungsten and phosphorus were synthesized and tested in the catalytic oxidation of propylene. As candidate descriptive parameters, the temperature and ten parameters related to the composition and chemical nature of the catalyst materials, derived from calculated free-atom properties, were offered. The temperature, the phosphorus content, and the composition-weighted electronegativity are identified as key parameters describing high yields towards the value-added oxygenate products acrolein and acrylic acid. The SG rules not only reflect the underlying processes particularly associated to high performance but also guide the design of more complex catalysts containing up to five elements in their composition.

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The FUNCAT (Fundamentals of Heterogeneous Catalysis) and Center-to-Center Collaboration: CO_2 Conversion into Useful Chemicals and Fuels

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The *FUNCAT* center and the *C2C* collaboration establish a link between the Max Planck Society (Max Planck Institute for Chemical Energy Conversion, Fritz Haber Institute and Max-Planck-Institut für Kohlenforschung) and the Cardiff Catalysis Institute in the UK, which enables the sharing resources and expertise in order to tackle major challenges in heterogeneous catalysis. In particular, CO_2 hydrogenation is one of the focuses of this project [1, 2]. Because the formation of valuable products from CO_2 relies on a complex interplay of several processes, such as the multiple surface reactions and the dynamic restructuring of the material under reaction conditions, a rational approach to design efficient materials has not been established so far. Herein, we combine leading-edge techniques of the involved centers to develop artificial intelligence (AI) approaches able to blend consistent "clean" data from experiments and first-principles calculations [3]. The goal is to unveil the intricate correlations between key physicochemical descriptive parameters ("materials genes") and the catalytic performance in the CO_2-conversion processes [4]. Crucially, the inclusion of theoretical descriptive parameters in the AI analysis will help identify and understand the microscopic processes that govern catalysis, while circumventing the atomistic description of the full catalytic progression.

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Ab initio Description of Surface Restructuring and Phase Boundary under Realistic Conditions

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A reliable description of surfaces structures phase equilibria in a reactive environment is a prerequisite for understanding mechanism of, e.g., heterogeneous catalysis. However, studying phase equilibria at *ab initio* level, is a formidable challenge, especially for highly anharmonic systems with sluggish barriers. Pioneering techniques require the prior knowledge of relevant phases and do not yield to unexpected phases and phase transitions. Furthermore, only free energy differences among the considered phases have been used to assess the phase stability regions, whereas the modelling of the singularities of a response function (e.g., heat capacity) is needed to locate phase boundaries and distinguish phase transitions from smooth transitions.

In this work, we introduce a fully *ab initio* approach to determine temperature-pressure (T, p) surface phase diagram and apply it to evaluate phase equilibria of surfaces in a reactive environment [1]. For this purpose, our replica-exchange grand-canonical (REGC) method [2] is extended by evaluating the heat capacity, $C_V(T, p)$, as function of T and p, thus locating phase boundaries including triple and critical points where $C_V(T, p)$ shows ridges. Furthermore, we show how the crucial limitation of the GC approach, formally defined only for a constant-volume ensemble, is circumvented by sampling different simulations cells and connecting all the phases in both cells via a reference phase. The approach is demonstrated by addressing open questions for the Si(100) surface in a hydrogen gas phase. By defining microscopic descriptors, 25 distinct thermodynamically stable surface phases are identified, most of which including few order-disorder phase transitions, have not been observed experimentally, so far. The results also show that Si-Si-bonds forming/breaking is the driving force behind the phase transition between the experimentally confirmed 3×1 and 2×1 adsorption patterns.

The REGC approach yields an *ab initio* description of surface restructuring as well as phase equilibria at technologically relevant (T, p) conditions. This constitutes an important advancement in the field of surface science and relevant for important applications, such as heterogeneous catalysis.

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Characterizing Point Defects in 2D Transition Metal Dichalcogenides for Novel Electronic Properties

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Defects can strongly influence the electronic, optical and mechanical properties of 2D materials. However, their stability and distribution under different conditions of temperature, pressure and strain are not well characterized from an atomistic perspective.

We have investigated the structural and electronic properties, as well as the thermodynamic stability of point defects (vacancies and adatoms) in semiconductor monolayer transition metal dichalcogenides (TMDC) MX_2 with M = Mo/W

and X = S/Se, through density-functional theory (DFT) simulations with hybrid exchange correlation functional, as implemented in the all-electron package FHI-aims. We show quantitatively that in rich X conditions an X adatom is most favorable while, in a poor X environment, an X mono-vacancy is most favorable. Interestingly, an interplay between adatom and divacancies takes place as temperature increases.

Through the development of a novel method to compute tip-enhanced Raman spectroscopy images that combines time dependent density functional theory and density functional perturbation theory to obtain realistic local fields, we computed the local vibrational fingerprints of these defects. We expect this data to be fundamental for the characterization of defect structures in these systems in the near future, and to aid electronic structure engineering that also exploits nuclear vibrational modes and electron-phonon coupling in these systems.

Finally, in order to gauge the importance of vibrational entropy on the engineering of gap states in the 2D monolayers, we compare the formation energies of point defects and adsorbed organic molecules at various thermodynamic conditions. We perform *ab initio*-based random structure searches for flexible molecules on surfaces, in order to efficiently navigate the conformational space. With a handle on the stability of these systems, we study the origin and temperature-dependence of singly occupied gap states induced by molecular anchoring at defects on semiconductor TMDCs.

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