MARVEL

NATIONAL CENTRE OF COMPETENCE IN RESEARCH

Materials' Revolution: Computational Design and Discovery of Novel Materials

Progress Report

Year 6

February 2019 - January 2020

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NCCR: 6th Progress Report - Cover Sheet

Title of the NCCR	Materials' Revolution: Computational Design and Discovery of Novel Materials (MARVEL)
NCCR Director Name, first name Institution	 Nicola Marzari EPFL
Date of submission	10 February 2020

Contents

1 Executive summary

MARVEL is a National Centre of Competence in Research (NCCR) on Computational Design and Discovery of Novel Materials; it was approved in December 2013, started its operations in May 2014, and currently is reaching the middle of phase II (May 2018-April 2022).

MARVEL Mission: *Accelerated design and discovery of novel materials in order to achieve improved properties and performance or witness the emergence of original physical properties, via a materials' informatics platform of high-throughput quantum mechanical simulations, powered by advanced electronic-structure capabilities, for predictive accuracy; innovative sampling methods to explore configurational and compositional space; and the application of big-data concepts to computational materials science.*

MARVEL Metrics: The Centre has 4 metrics to score its mission during phase II; in order of importance these are:

- *Design and discovery of novel materials displaying novel physics or improved properties or performance*
- *Conceptual, methodological, and algorithmic developments: Their dissemination and usage*
- *Open Science: codes, data, tools, and workflows/turnkey solutions*
- *Engagement in the four management areas — especially tech transfer and equal opportunity*

In phase I MARVEL worked to unite a community of outstanding computational scientists towards the goals of materials design and discovery, aligning this community to the needs and the challenges coming from the experimental world, and developing the computational tools required for realistic, accurate and predictive simulations, to explore the complex landscape of materials' space, to leverage the new ideas coming from the world of data mining and machine learning, and to adapt those to the domain of computational materials science.

In phase II MARVEL has dedicated itself — the capabilities and the teams developed during phase I — to the core goal of design-and-discovery of novel materials, with 6 ambitious projects that address outstanding scientific problems that have either direct or promising technological implications: organic crystals, complex metal alloys, nanomaterials and nanodevices, metal-organic frameworks, correlated oxides, and topological materials — with relevance to the high-tech, high-value industrial landscape of Switzerland, and to the broader fields of energy and information-and-communication technologies. The key long-term structural effort is embodied in the Open Science platform, powered by the AiiDA materials' informatics infrastructure and the Materials Cloud for the deployment and dissemination of user services, open-source tools, curated and raw data, and educational material, providing in the process archival solutions and FAIR-compliant data-management plans. Very close and strategic collaborations with the Swiss National Supercomputing Centre, the Paul Scherrer Institute, and Empa embed these efforts in the scientific infrastructure of the country.

As director, I am particularly impressed by the outstanding scientific results that are being delivered by the Centre, by the major initiatives that have been initiated both at the Swiss and European level, leveraged and made possible by the Open Science platform, by the new industrial engagements, and by the sustained and visible efforts in improving diversity and opportunities in our research environment.

2 Reaction to the recommendations of the review panel

There was no site visit in year 5. The review panel received the year 5 progress report and the "Equal opportunities, analysis and plans" document. They sent a very positive feedback on both documents. A SNSF delegation came for a short visit about equal opportunities, with as well a very positive outcome. We present hereafter a short reaction to the feedback of the review panel.

The panel members who gave their feedback on the short progress report were pleased with the progress of the NCCR. The reorganization of the NCCR towards the new phase II structure seems to be progressing well. The projects are advancing well, as measured by output in terms of publications, databases, software releases etc. The various teams continue to perform excellently at the international level. New collaborations have arisen as a result of MARVEL.
One panel member resumed: "I would like to congratulate the MARVEL team for the constructive spirit in which they have taken the feedback received in the previous round of review; it is clear that they have made determined efforts to respond positively to the suggestions received, and to incorporate many of these suggestions into the way that MARVEL has evolved within the past year."

We are glad that the review panel is pleased by the progress of MARVEL, as well as its reorganization for phase II, the quality of the research and all outcomes.

The panel members had also received the new action plan on Equal Opportunities. Their replies comment the plan positively and appreciate that the MARVEL leadership took the raised concerns seriously. They are confident that the various initiatives will have a positive impact. One reviewer is pleased to see that also EPFL as the home institution is helping with a number of specific actions.

We are grateful for the positive feedback of the review panel and the SNSF about this document. While continuing our various successful activities for equal opportunities, always working in close collaboration with EPFL as home institution, we have started to implement the new initiatives presented in this document. All our actions in year 6 are presented in Chapter 7, Equal opportunities.

One panel member has a suggestion in view of the next site visit: "It would be nice to know, during next year's site visit, what the response is to these Gender Training sessions, both in terms of the number of people who attended them, and what feedback they gave about these sessions. It may also be nice during the coming year, to have some mechanism of collecting feedback from all the participants in MARVEL, about how they feel about the gender climate (and perhaps about the working climate in MARVEL in general?). After all, the review panel's impressions were collected during a site visit of two days and may not reflect the ground reality, it would be good to have some mechanism to know what the faculty/postdocs/students themselves think."

As described in more detail in Chapter 7, 75 MARVEL members/associated members participated in the six Gender Training sessions organized in year 6, with very positive feedback. A short survey to assess the gender and working climate in the MARVEL laboratories is currently under preparation. It will be sent out in February and collected feedback will be presented at the April site visit.

3 Management

3.1 Structure and organisation of the NCCR

3.1.1 Structure of the NCCR

In phase II, MARVEL is organized around 6 Design & Discovery projects (major collaborative projects covering key topics in energy, ICT, manufacturing, chemistry and pharma, and ranging from molecular crystals to complex metal alloys to topological materials), 2 Incubator projects (more focused efforts, dedicated presently to solid-state ionic conductors and machine learning). In addition, 2 core structural efforts are represented by the Open Science platform (which provides open access to simulation services and tools, dissemination and preservation of curated and raw data, and educational material) and the HPC and Future Architectures platform, which supports the deployment of the next hardware resources for MARVEL, and the co-design of software and hardware (Fig. 1). EPFL is the home institution and participating scientists are affiliated with 11 Swiss academic and industrial institutions.

In phase II, along with the director, Nicola Marzari, Berend Smit (EPFL) is deputy director, and Clémence Corminboeuf (EPFL) and William Curtin (EPFL) complete the Executive Committee in charge of day-to-day activities. Strategic decisions (reallocation of funding, establishment and termination of projects) are taken by an enlarged committee, the Strategic Committee, which was slightly reorganized in year 6, now including, in addition to the Executive Committee, Thomas Schulthess (CSCS and ETHZ), Frithjof Nolting (representative of PSI) and Daniele Passerone (representative of Empa). Finally the Scientific Committee is composed of the Strategic Committee and all project leaders; it has the formal role of discussing the science and an advisory role on strategy. A Scientific Advisory Board (SAB) of nine members, chaired by Giulia Galli (Univ. Chicago), and an Industrial Advisory Board (IAB), chaired by Erich Wimmer (Mat. Design)

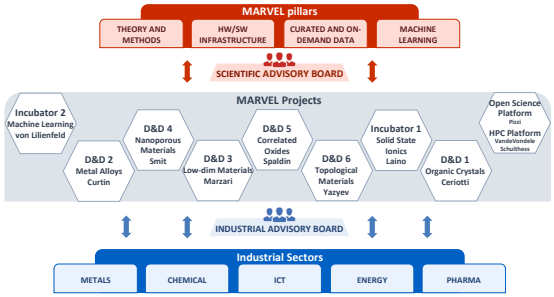

Figure 1: *Structure of NCCR MARVEL in phase II.*

and with F. Diologent (Richemont), T. Eckl (Robert Bosch), A. Ryoji (Toyota), N. Cudré-Mauroux (Solvay) and A. Grandeury (Novartis), provide feedback on all activities and convene once a year (SAB, at the Review and Retreat) and one every two years (IAB). The management team is unchanged this year.

3.1.2 Organisation of the NCCR

Claire Villevieille left PSI in February 2019 to become a senior research scientist at the Université Grenoble Alpes. The MARVEL effort in her group continued throughout the year, with the postdoc supported by her funding managed by Daniele Pergolesi, PI in the same Inc1 project. Piero Macchi left UniBE in August 2019. Arrangements were made to allow him use his year 6 funding and conclude his contribution to the project. Kyriakos Stylianou left EPFL in August 2019. His research effort and budget are taken over by Christopher Ireland, also part of Berend Smit's group. Following the passing of Alexey Soluyanov in October 2019, his MARVEL budget is maintained. Prof. Titus Neupert, recently arrived also at UZH, and an expert in topological insulators, joins as new MARVEL PI and continues the collaboration within the D&D6 project.

3.2 Management activities and status of collaboration/integration

3.2.1 Activities and measures

Events organisation

In year 6, MARVEL management organized meetings, lectures and events, described in chapters 5 – 8. We mention, for example:

- MARVEL stand at International Women's Day "50 years of EPFL Women", EPFL, March 8.
- Julie Birenbaum, the light painting artist, ETHZ, March 21–28.
- MARVEL stand at the EPFL Industry Day, March 20, and at the ETHZ Industry Day, September 4.
- Ig Nobel Award Tour Show, EPFL, March 25.
- AiiDA plugin migration workshop, EPFL, March 25–29.
- 2 CECAM/MARVEL Classics in molecular and materials modeling lectures, EPFL, "Molecular dynamics under (holonomic) constraints", April 30, and "Car-Parrinello molecular dynamics", July 25.
- Mary Ann Mansigh conversation "Computer modeling for industrial applications", EPFL, May 8.
- "Mini-site-visit" about Equal Opportunities with SNSF Research Council members, EPFL, May 15.
- AiiDA tutorial on "writing reproducible workflows for computational materials science, using AiiDA 1.0", EPFL, May 21–24.
- 2 MARVEL workshops "Overcome implicit bias in recruiting and supervising a diverse workforce" with Marianne Schmid Mast, for PIs and senior scientists, EPFL and UZH, May.
- 4 MARVEL workshops "Beyond Bias: Diversity and Team Work" with Marianne Schmid Mast, for PhD students and postdocs, EPFL and UZH, June and October.
- The summer camp for high school students *Des atomes aux ordinateurs, à la découverte de la programmation scientifique*, EPFL, July 1–5.
- CPMD Meeting 2019 — Pushing the Boundaries of Molecular Dynamics, EPFL, July 22–24.
- MARVEL Review and Retreat, EPFL, September 5–6.
- MARVEL stand at EPFL Open House, September 14–15.
- CCMX-MARVEL Materials Science Day, Bern, October 8.
- MARVEL stand at "Digital in our daily lives, today and tomorrow", *Lycée cantonal de Porrentruy*, October 30.
- 4^{th} annual AiiDA coding week, Fiesch, December 9–13.
- INSPIRE Potentials — MARVEL Master's Fellowships for female Master students, with 2 calls in April and October.
- 2 Industry Sector Days at EPFL, on metals, February 15, and pharma and fine chemistry, October 10.
- 2 MARVEL distinguished lectures at EPFL, Emily Carter (Princeton), June 17, and Giulia Galli (Chicago), November 13.
- 9 junior seminars at EPFL, continuing on a monthly basis.
- 3 "CECAM/MARVEL soirées", movie nights featuring movies with scientific themes, EPFL, March – May.
- 16 seminars at EPFL.

Other activities

MARVEL members organized 21 conferences, tutorials or workshops (not counting the AiiDA tutorials), of which MARVEL sponsored 4.

Measures

The management team has prepared a "welcome letter" with information and requirements regarding finances, acknowledging MARVEL in publications, visual identity, equal opportunities and tech transfer themes. It has been sent to all MARVEL members in January 2019 and is being sent progressively to the new members.
A set of regulations finalizing the set of rules that the NCCR is operating with, updating the current one, has been prepared and signed.

Research data management activities

MARVEL publications in 2019 generated about 70 MARVEL related entries on Materials Cloud *Archive* as well as some entries on other open repositories. The MARVEL dataset index is available on the website on nccr-marvel.ch/publications/dataset-index. The use of Materials Cloud *Archive* as open repository is well accepted by MARVEL members. Moreover, for entries already on Materials Cloud *Archive* by mid November, we could generate the bibliography file with the related publications, asking the PIs to complete the file with the missing publications (i.e. publications with no dataset deposited on Materials Cloud or publications for which the datasets have been deposited between November and January).
On the side of Materials Cloud *Archive* moderation, the biggest challenge is identifying what changes can improve the entry while still respecting the needs and wishes of the authors. This requires to strike a delicate balance between observance of the rules and openness to each individual case. A careful review also

takes time, and since the rate of submissions is steadily increasing (about which we are very happy), the total time commitment has become non-trivial.

On the MARVEL data manager side, one challenge is to track, for each MARVEL publication with no indication about datasets, whether datasets exist, are already deposited somewhere and, if not, ask the authors to do so. This is mostly the case for publications submitted before 2019, before the writing of the MARVEL research data management strategy. The authors did not plan the publication of the data at the time of the writing of the paper. We hope that next year such cases will drastically decrease. Another issue is to convince authors that even small datasets in an article are still data and should be made openly available. We will work on this in the next months with informative campaigns.

Also related to publications is the openness of the publications themselves. The policy of the SNSF is known by the researchers and most of them use at least the green road, when allowed by the journals, although sometime constraint by an embargo of more than 6 months. On the physics side of the community, the preprint server arXiv.org has already been well implemented for a long time. Now most of the institutional repositories (InfoScience at EPFL, DORA at PSI and Empa, Research Collection at ETHZ, edoc at UniBas, etc.) manage by themselves the embargo, giving already at the time of publication an open access link that will allow free access to the publication at the end of the embargo, releasing the responsibility of the authors to come back later.

3.2.2 Status of collaboration/ integration

All projects in phase II are intensely collaborative, with a project leader integrating the different computational efforts of the co-PIs, and in most cases including funded or matching experimental efforts. It's only because of MARVEL that these projects were nucleated and grown, and the ongoing close interest and support taking place at CSCS, PSI and Empa reassure us of the long-term effects and collaborations that these projects are establishing. Having a common hardware and software infrastructure deployed at CSCS, a data infrastructure in the Materials Cloud, and an operating system with AiiDA make MARVEL a think thank and a working laboratory to develop the vision, synergies and facilities/infrastructure for computational science.

On the other hand, the new Equal Opportunities effort required a reallocation of funding in order to free the 887'000 CHF dedicated to this. In addition, the budget of phase II was front-loaded in the original proposal for the projects during their first 2 years, in order to ramp up the most promising efforts; this implied a 15% cut in their allocation in the second half of phase II. For these reasons, and with the goal of protecting as much as possible all the ongoing and very successful projects, we took the decision to keep all 6 design-and-discovery projects and cut them by just an additional 3%; the platforms have been reduced by 10%, and the two incubators by $\sim$ 50% (in particular, Incubator 1 stops at the end of year 7, while Incubator 2 will continue to year 8, but at a reduced allocation for the last two years). Happily, the MARVEL match to PSI cash funding remains unaltered for the experimental groups involved.

4 Research

4.1 Results since the last report

Design & Discovery 1 — Understanding Complex Molecular Crystals: Structure and Properties

Project leader: Michele Ceriotti (EPFL, 1MC)

Computational partners: Clémence Corminboeuf (EPFL, 2MC), **Stefan Goedecker** (UniBas, 1.5MC), **Michele Parrinello** (USI and ETHZ, 1.5MC), Anatole von Lilienfeld (UniBas)
Experimental partners: Esther Amstad (EPFL), Lyndon Emsley (EPFL)

Molecular crystals pose significant modeling challenges due to the presence of competing polymorphic forms, which has important industrial repercussions, and the interplay of intra- and inter-molecular forces, molecular flexibility and crystal packing [1]. This project aims at extending the reach of modeling approaches for molecular materials, using machine-learning techniques and advanced statistical sampling to address molecular flexibility, finite-temperature thermodynamics and nucleation kinetics. We focus here on summarizing advances achieved in the past year; we refer to the proposal and to past years' reports for a discussion of the project scope, milestones, and previous achievements.

1 Progress of the different efforts

The progress of the project in the first one and a half year has been outstanding, and essentially all of the milestones planned for the first two years have been reached. In the following paragraphs we briefly summarize the main achievements of each subproject.

a) *Molecular and crystal energetics by machine learning (Corminboeuf, Ceriotti)* Building a transferable, accurate model of the energetics of molecular materials has been the driving force for a major software development effort, jointly with the D&D2 and the Inc2 projects, taking the form of LibRascal, an efficient library to compute representations of atomic structures, and machine-learning (ML) models. We are currently performing a systematic comparison of the performance of different ML schemes – including Gaussian process regression and neural networks – when used to evaluate energy and forces of oligopeptides. We plan to use these models to sample the conformational landscape of these flexible

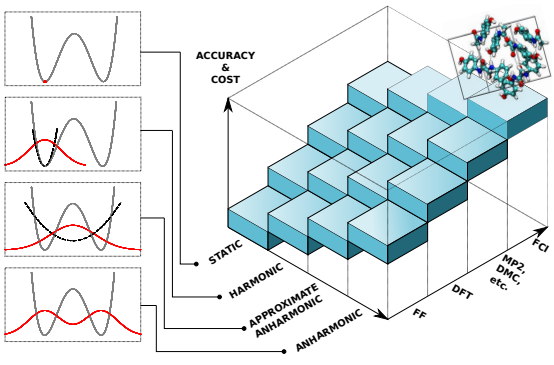

Figure 1: *Multiple levels of sampling approximations combine with the accuracy of the potential energy surface to determine the predictive power of molecular crystals thermodynamics. We implemented and tested harmonic free energies, perturbative corrections on top of them, and self-consistent phonons, against fully-converged thermodynamic integration.*

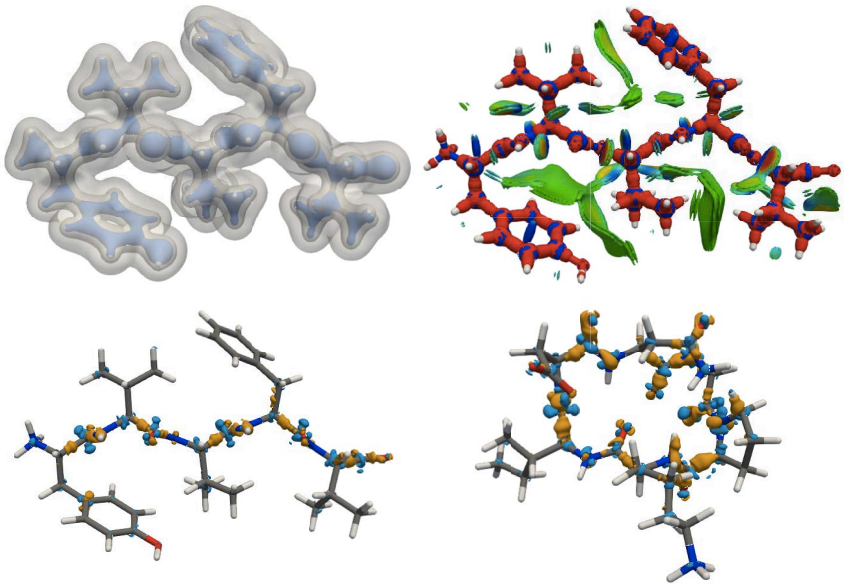

Figure 2: *Machine-learned prediction of the charge density of the peptide enkephalin. The charge is sufficiently accurate to capture the fine details of the charge density that are associated with covalent and non-covalent interactions, as identified by the DORI fingerprint [2]. The error (shown in the bottom panels for enkephalin and a cyclopeptide) concentrates in the region of the peptide bond, as the training set contains no examples of such chemical feature.*

molecules, and as a baseline to predict the interactions between molecules in the condensed phase. The need to model electrostatic and dispersion interactions has triggered a parallel effort to tackle the difficulty in incorporating long-range physics [3], which will be implemented in LibRascal.

b) *Anharmonic and quantum free energies (Ceriotti)* Temperature and zero-point energy induce fluctuations that affect the thermodynamic stability and the properties of materials, and are particularly important in molecular crystals, where few meV per molecule differentiate between stable and unstable polymorphs. We have introduced the concept of a "Jacob's checkerboard" combining multiple tiers of electronic structure and thermodynamic sampling approximations (Fig. 1). We have implemented in i-PI [4] several methods to approximate anharmonic free energies, and benchmarked them on different classes of materials. We found that even though approximate methods perform well in typical inorganic materials, they are insufficient to treat flexible modes, librations and group rotations in molecular compounds [5]. More systematic tests of different classes of molecular crystals are underway.

c) *Predicting complex properties by machine learning (Ceriotti, Corminboeuf)* Armed with a unique framework to construct machine-learning models of properties that have a tensorial nature [28], we have been able to develop models for several observables that are of high relevance to molecular compounds.
We introduced a framework to predict the electron charge density in molecules, based on an atom-centered decomposition

$$\rho(\mathbf{r}) = \sum_{i,nlm} c_{nlm}(i) R_n(\mathbf{r} - \mathbf{r_i}) Y_l^m(\mathbf{r} - \mathbf{r_i}) \quad (4.1)$$

and a symmetry-adapted machine-learning algorithm [6]. Progress on this front have been very fast, and we could move from the proof-of-principle level to a model that can, after training on dimers of small organic molecules, predict the charge density of polypeptides [7] (Fig. 2). A symmetry-adapted model has also been developed, in a separate project, to predict the polarizability of molecules [8, 9], which is going to be used to estimate the Raman spec-

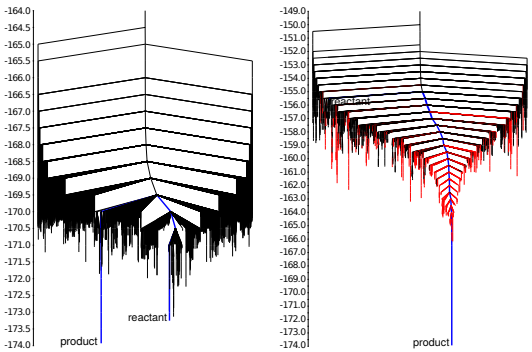

Figure 3: *Disconnectivity graphs of the LJ_{38} cluster with two different bias strengths. The red minima indicate fcc like structures and the blue path shows the transition from the second lowest icosahedral to the global fcc minimum. Without a bias (left) it is extremely difficult to find a reaction pathway that crosses the high barrier between the icosahedral minimum and the global fcc minimum, which is the global minimum since the barriers that have to be overcome are quite high. The bias renders these barriers smaller and makes it easy to hop from the initial structure into the final one (right).*

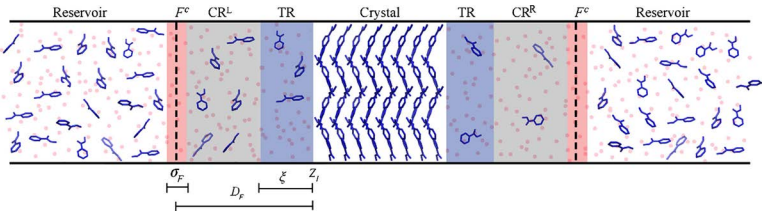

Figure 4: *A schematic of the simulation box that is used to run CμMD simulations. The crystal is kept in the middle of an orthorhombic simulation box. The solution concentration in the CRs (blue slab) is maintained at a target concentration.*

tra of different polymorphs of molecular compounds [29].
Finally, a collaboration with the experimental group of Lyndon Emsley (EPFL, chemistry) has led to the development of ShiftML [10], a model that predicts, using GIPAW-DFT references, the chemical shieldings of nuclei in molecular materials, and that can be used in tandem with solid-state NMR measurements, to determine the crystal structure of a compound based on small amounds of powder samples. ShiftML has been used to create an online tool, hosted on Materials Cloud [11], that predicts in seconds the chemical shieldings for molecular materials containing C, H, N, O, S atoms. Thanks to the availability of an inexpensive uncertainty estimation on the predictions [12], we could also introduce a Bayesian scheme to determine the reliability of NMR structural determination based on the comparison between experiment and ShiftML [13] – a first example of the convergence of machine learning, modeling and experiments that constitutes one of the grand challenges in this project.

d) *Molecular conformers and crystal structure search (Goedecker)* Training a machine-learning scheme requires a large and very diverse dataset that includes all relevant structural motifs. The minima hopping structure prediction method is an ideal tool to generate such a dataset. By its built-in feedback it escapes from regions that were already sampled and explores instead new regions. This method has been already applied to generate an initial dataset of amino acids and dipeptides. Another important application of conformational search involves the study of polymorphic transitions, that is complicated by the fact that it is not a priori known which atom or molecule in the initial structure will be mapped on the atoms in the final structure. To avoid the combinatorial explosion of possible mappings, we have introduced a permutationally invariant bias, that pulls the system toward the chosen final structure. In this way, we can bias the potential energy surface such that a minima hopping run will rapidly find a path that connects the initial structure with the final structure [14]. This bias can selectively target a single configuration and allows us to follow the transformation on the full 3N dimensional potential energy surface. We expect that this method in combination with machine-learned potentials will allow to describe complex polymorphic transformations. A demonstration of this approach is shown in Fig. 3.

e) *Interface energies and nucleation kinetics (Parrinello)* Kinetic effects occurring during the crystallization process are known to play an important role in determining the experimentally-observed polymorphs. Molecular dynamics (MD) simulations can shed light upon the atomistic details of the crystallization process. To achieve this goal, one has to deal with two main limitations of standard MD – finite-size effects and the time-scale problem.
In a standard simulation of crystallization from solution, finite-size effects arise due to the continuous extraction of solutes from the solution to the growing crystal phase resulting in solution depletion. We recently developed a method, constant chemical potential molecular dynamics (CμMD), to tackle this finite-size problem [15].
The CμMD method and its *cannibalistic* variant have been applied to study solvent-dependent morphology selection of an anti-tuberculosis drug, isoniazid [16], and supersaturation-dependent crystal shape prediction of naphthalene in ethanol solution [17]. Furthermore, a spherical variant of the CμMD has been developed to study homogeneous nucleation from supersaturated solution [18]. The basic CμMD method and its variant provide a solid platform to carry out crystallization simulations at constant chemical potential conditions mimicking realistic experiments (Fig. 4).
In order to address the time-scale problem, collective variables (CVs) based enhanced sampling has proven to be a great tool. New strategies have been developed to better deal with the problem of suboptimal CVs [19, 20], and in the study of complicated free energy landscapes [21]. Furthermore, a new method has been developed to focus the sampling on the transition state [22], and the multithermal-multibaric sampling scheme introduced last

year has been extended to investigate also phase transitions [23]. These methodological improvements will help in studying difficult phenomena such as crystal nucleation [24] and growth of molecular systems.

2 Contribution to overall goals and initial proposal

The different subprojects have addressed many of the fundamental challenges that were identified to advance the state-of-the-art in modeling complex molecular materials – computing accurate energetics [25], describing finite-temperature thermodynamics, predicting experimental observables, sampling conformational space and modeling kinetics and nucleation effects. Furthermore, many of these advances integrate into the broader objectives of MARVEL; ML software is being developed in close connection with D&D2, making sure that the open-science and dissemination metrics are met; data from scientific publications has been shared on Materials Cloud, and online tools demonstrating ML models for chemical shifts and molecular polarizabilities have been contributed to the portal [26, 11].

3 Collaborative and interdisciplinary components

Several of the core developments of this project, discussed above, have a strong collaborative component. This is especially true on the machine-learning side, where the need of complementary expertise on the generation of accurate reference data, the development of a model, and its efficient implementation, make cross-disciplinary efforts particularly important. Here we want to highlight an effort that combines the expertise in the Corminboeuf, Ceriotti, and Goedecker labs, that targets modeling of the interactions within and between peptides, in line with the original objective triggered by the development of peptides-based clinical therapeutics. The two main challenges associated with accurately describing peptides with QM-based ML potentials are the conformational space (i.e. large flexibility with 100s to 1000s atoms) and the chemical diversity inherent to the 20 natural amino acids with different protonation states and forms. Training such a robust and transferable ML model relied upon the generation of a large and diverse set of energies and forces (i.e. 48k conformations for 17 charge-neutral gas-phase amino acid dipeptides) that were generated by the Goedecker group using the minima hopping structure prediction method. The Ceriotti and Corminboeuf groups exploited two different ML strategies coupled with accelerated sampling techniques (i) a kernel ridge regression coupled with an Hamiltonian-reservoir replica-exchange Monte Carlo sampling [27] and (ii) an artificial Behler-Parrinello type neural network with a metadynamics simulation. These two models can predict the free energy surfaces of charge-neutral dipeptides within 0.05 kcal/mol/atom of accuracy. Current efforts are placed into the transferability and scalability of the models to larger and charged oligopeptides as well as to the generalization to the crystal phase.

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

- [1] G. Gryn'ova, K.-H. Lin, and C. Corminboeuf, *Read between the Molecules: Computational Insights into Organic Semiconductors*, Journal of the American Chemical Society **140**, 16370 (2018).
- [2] B. Meyer, S. Barthel, A. Mace, L. Vannay, B. Guillot, B. Smit, and C. Corminboeuf, *DORI Reveals the Influence of Noncovalent Interactions on Covalent Bonding Patterns in Molecular Crystals Under Pressure*, The Journal of Physical Chemistry Letters **10**, 1482 (2019).

[3] A. Grisafi and M. Ceriotti, *Incorporating long-range physics in atomic-scale machine learning*, The Journal of Chemical Physics **151**, 204105 (2019).

- [4] V. Kapil, M. Rossi, O. Marsalek, R. Petraglia, Y. Litman, T. Spura, B. Cheng, A. Cuzzocrea, R. H. Meißner, D. M. Wilkins, B. A. Helfrecht, P. Juda, S. P. Bienvenue, W. Fang, J. Kessler, I. Poltavsky, S. Vandenbrande, J. Wieme, C. Corminboeuf, T. D. Kühne, D. E. Manolopoulos, T. E. Markland, J. O. Richardson, A. Tkatchenko, G. A. Tribello, V. V. Speybroeck, and M. Ceriotti, *i-PI 2.0: A universal force engine for advanced molecular simulations*, Computer Physics Communications **236**, 214 (2019).
- [5] V. Kapil, E. Engel, M. Rossi, and M. Ceriotti, *Assessment of Approximate Methods for Anharmonic Free Energies*, Journal of Chemical Theory and Computation **15**, 5845 (2019).
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Design & Discovery 2

From Atoms to Additive Manufacturing: Computational Design of Complex Metal Alloys

Project leader: W. A. Curtin (EPFL, 2MC)

Computational partners: Michele Ceriotti (EPFL, 1MC), Anatole von Lilienfeld (UniBas)
Planned experimental partners: Helena Van Swygenhoven (PSI), Christian Leinenbach (Empa)

1 Progress

1.1 LibRascal

A major effort in D&D2 has been the development of the *library for representation of atomic scale learning* (LibRascal), an effort involving Junge and Stricker of LAMMM and Goscinksi, Musil, Veit, Fraux, and Cersonsky of COSMO. LibRascal aims to be an efficient, scalable and versatile library for using, comparing, and developing atomic representations for use as a predictor for atomistic/molecular properties including, but not limited to, energies and forces. These representations are the input to any supervised or unsupervised learning algorithm and are often the computational bottleneck. LibRascal is a stand-alone code written in C++ with flexibility to interface to other established codes like LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator) and PLUMED-2.0 (Plugin for Molecular Dynamics) and Python bindings to the core functionality provide ease of use. Computationally-expensive operations are implemented in the C++14 standard with a special focus on efficient data structures and memory-safe design of iteration and data access.
The core of LibRascal is the StructureManager abstraction. It represents an atomic structure (anything from a single molecule in a training set to a polycrystalline metal MD simulation) and can be

- *iterated over*, e.g., iterate over atoms, pairs, triplets, ...
- *filtered*, e.g., iterate only over iron-nickel pairs, or only over pairs with a distance smaller than r_{cut}, or
- *extended*, e.g., StructureManager contains only atoms and pairs, but we need triplets.

The StructureManager can then be used to associate so-called Properties — contiguous data structures — to any cluster size (1 for atoms, 2 for pairs, etc), accessible with the StructureManager's iterate as direct access key. This is for efficiency. For instance, to evaluate a set of symmetry functions $G(\{r_{ij}\})$ that represent the contribution of Ni neighbors to an Fe atom, one filter operation of the StructureManager yields only Fe-Ni pairs within the specified cutoff. The resulting adapted StructureManager can be iterated over again just once to obtain all pair distances in a contiguous Property. The G functions can then be computed by evaluating contiguous input arrays into contiguous output arrays without evaluating any neighborhood-related operations. More generally, StructureManager in combination with Properties forms the data structure of LibRascal suitable for current and future implementations of atomic descriptors of any kind. To our knowledge, no other library provides this kind of flexible framework to implement and compare multiple descriptors both efficiently and safely.
Because LibRascal has interfaces with LAMMPS and PLUMED (with more interfaces to come), users can concentrate effort on implementing new novel descriptors when developing machine-learned potentials without worrying about framework integration. Further, the Python bindings allow users to take advantage of modern tested machine-learning frameworks (scikit-learn, PyTorch, TensorFlow) for training and evaluation.
LibRascal is currently in α development phase with a β release expected in mid-2020.

1.2 Machine-learning potentials

A second major focus for D&D2 has been the development of machine-learned interatomic potentials for complex alloys. The main focus to date has been on two technologically-relevant lightweight alloys, Al-Mg-Si and Al-Cu, as the intermediate steps toward the Al-Mg-Si-Cu and other advanced Al alloys. We currently use the established Behler-Parrinello atomistic descriptors (symmetry functions)

and a neural-network machine-learning (ML) framework using the open-source n2p2 code. These choices enable the demonstration that machine-learned potentials can capture a very broad range of metallurgically-relevant properties while maintaining a computational cost suitable for large-scale simulations of alloy processing and performance. Significant parallel efforts are being made in D&D2 (COSMO) toward the development of better descriptors and machine learning using kernel methods that have a number of advantages but remain computationally costly for multicomponent alloys [1, 2].

In Al-Mg-Si, we have worked to improve the prediction of the shear modulus C_{44} of pure Al, which is typically rather larger than DFT-computed values that are, in turn, somewhat larger than experiments. Dislocation energies, and strengthening mechanisms, scale with the elastic constants and hence predictive simulations require accurate elastic constants. However, as second derivatives of energy versus distortion, the C_{ij} are generally difficult to capture accurately in both ML potentials and traditional potentials. Understanding the optimization/training methods of the n2p2 code, we have enhanced our DFT training set to guide better representations of pure shear deformations. Results show improved C_{44} can be achieved although still not to the level we would like. Given trade-offs in predicting a wide scope of properties, and an initial focus on precipitate evolution that does not directly depend strongly on C_{44}, we are developing a final Al-Mg-Si Neural Net Potential (NNP) now.

A useful potential for metallurgical studies of a complex alloy must accurately predict quite a range of complex atomic environments. We have thus expanded our evaluation of Al-Mg-Si to include the prediction of the generalized stacking fault energies (GSFE) for the β'' precipitates that are crucial in the Al-6000 series Al-Mg-Si alloys and the anti-site defects in these precipitates. Standard studies of the GSFE reveal that initial GSFE configurations with atomic positions that are unusually close lead to a structural collapse and deep unphysical minimum energy structure; problems can also arise in direct DFT simulations. Our previous NNPs did not include high-compression equation-of-state (EOS) data in the training sets, and so we have now included this information. Also, the computation of the GSFE can be modified to avoid the collapse problem (which would never arise in any realistic study). Overall, the GSFE predictions for the three typical β'' compositions are in good agreement with full DFT over the entire SF surface(s) expected to be relevant for precipitate shearing inside the Al matrix. Several unrelaxed anti-site defects among all possible configurations were predicted to have negative energies, in contrast to DFT, and so this data is being introduced into the training set. We anticipate a final Al-Mg-Si potential that will accurately predict all important aspects of Al, the β'' precipitates, the most-stable β precipitate, and the precursor clusters that develop in the early stages of precipitate evolution.

We have also pursued development of an NNP for binary Al-Cu (Fig. 1). We developed an extensive dataset for training including many possible (metastable) precipitate structures, solid-solution configurations, and other configurations based on our extensive experience with the Al-Mg-Si system. The resulting NNP accurately captures energies for many precipitates, various precipitate GSFEs, and various precipitate anti-site defects, none of which were in the training set. The NNP also predicts the Cu misfit volume in Al with good accuracy and various other properties. A careful study of the most-widely-accepted and used Al-Cu EAM-type potential (the ADP potential by Mishin *et al.* [7]) shows it to be very inaccurate in a number of respects (Cu misfit volume is 3× DFT and experiment; crucial θ'

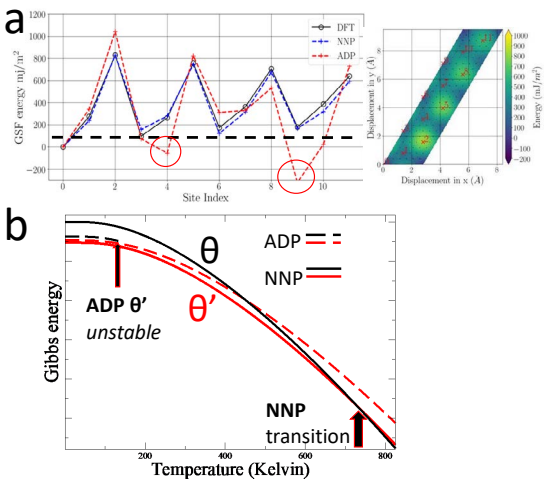

Figure 1: *Neural Net Potential (NNP) for Al-Cu versus DFT and/or state-of-the-art ADP potential. (a) Stable and unstable stacking fault energies of θ'' (full NNP stacking fault surface on the right). The NNP agrees well with DFT and ADP predicts sheared precipitates to be more stable. (b) Anharmonic Gibbs free energy versus T. The NNP predicts the $\theta - \theta'$ transition at 730 K (vs DFT harmonic estimate $>$ 473 K) and the ADP predicts the low-temperature θ' phase to be unstable at only $\sim$ 150 K.*

precipitate has a negative GSFE region indicating lack of stability; thermodynamic properties of the competing θ and θ' precipitates are very poor — losing stability at moderate temperatures). In contrast, the important crossover in thermodynamic stability between θ and θ' precipitates — identified first via DFT in classic work of Wolverton *et al.* [8] — is captured by our new NNP using fully anharmonic analysis. This Al-Cu potential will next be used to simulate the evolution of so-called Cu GP zones using kinetic Monte Carlo, with subsequent molecular statics/dynamics simulations of the strengthening and then simulations of strengthening due to θ' precipitates.

Mg alloys have a high technological importance, but the creation of potentials that capture all of the different deformation features in hcp Mg has proven very difficult. So, we have pursued application of the machine-learning machinery in D&D2 to develop a comprehensive neural-network potential for Mg. Again using the Behler-Parrinello framework, we have created a family of NNPs using a DFT database computed with high accuracy. We include many key features for plasticity, such as generalized stacking fault curves for the basal, prismatic, and pyramidal I and II slip systems. In application of an early version that included standard decohesion curves, we found that fracture is poorly predicted, with some unphysical behaviors. We thus added a series of atomic configurations containing edges and corners of various types and have now achieved good agreement between fracture simulations and fracture theories for the next-generation Mg potential. The potential captures a range of dislocation core structures in agreement with DFT. The one exception is the pyramidal II screw dislocation, for which the NNP predicts a structure that appears to be a combination of Pyr. I and II screw partial dislocations. The structure and stability of the Pyr. II screw dislocation relative to the Pyr. I screw is crucial for understanding how high ductility is achieved in Mg alloys. However, current DFT studies remain uncertain about the relative stability, and so training a potential based on DFT may lead to an incorrect result. In all other respects, our Mg NNPs are far superior to the best MEAM (Modified Embedded Atom Method) potential (created by our group in a separate program). It is also suitable for extension to Mg alloys whereas our best-effort MEAM potential for Mg-Y is not fully satisfactory for study of crucial deformation modes in Mg-Y. Extension of the new Mg NNP to dilute Mg-Y alloys will be done in the near future, taking advantage of our existing database on dilute Y in a range of Mg configurations.

1.3 Kinetic Monte Carlo of early-stage precipitation

We have further developed the i-PI code (COSMO) for kinetic Monte Carlo (kMC) simulations, using NNPs in the n2p2 code with LAMMPS as the engine for energy computation. A key feature is a caching algorithm to store previously-visited configurations and thus accelerate the kMC. Concepts for algorithms to retain only local environments around the instantaneous vacancy position are developed but not implemented.
We have performed preliminary simulations of early-stage Si-Mg cluster development in an Al – 2 at% Mg – 1 at% Si alloy approaching the commercial Al – 1 at% Mg – 0.5 at% Si alloy. The goal of this initial study is to determine whether these early-stage clusters trap vacancies, aiming to verify the concept of "vacancy prisons" that has been put forth based on indirect experimental analyses. To examine vacancy trapping, we have been developing SOAP-type descriptors and dimensional reduction methods to identify the characteristics of dynamically-evolving local clusters that trap a vacancy and determine the trapping times. This trapping retards precipitate evolution, and also controls eventual alloy properties. To date, we have identified that small Si-Mg do trap vacancies, with trapping times that are $10^3 - 10^5$ times longer than the fundamental diffusive step of the vacancy in the Al lattice. We have deferred the generation of a large-scale dataset pending the finalization of the Al-Mg-Si NNP (see above), and expect to start large-scale simulations at the beginning of 2020.

1.4 Precipitate-strengthening in Al-Mg-Si

In parallel with atomistic studies, we have pursued a higher-level continuum discrete-dislocation modeling strategy for computing the strength of an alloy due to precipitation in the regime of Orowan looping. At peak strengthening, standard theory says that the strength associated with Orowan looping is equal to that for precipitate shearing. This enables us to avoid, at present, the need for an atomistic representation of the shearable precipitates. Our goal has been to quantitatively predict the strengthening of the peak-aged Al-6xxx microstructure with β'' precipitates, and to identify the role of precipitate-induced coherency misfit-stresses on the strengthening.

We have developed an approach for (i) creating a series of increasingly-realistic β'' precipitate morphologies, (ii) computing the entire stress field due to the precipitate eigenstrains (relative to the Al matrix) as computed in DFT (and captured by our NNPs), and then (iii) simulating the motion of screw and edge dislocations through the alloy microstructure. The full 3D stress field is computed using a new spectral method developed by Junge in the LAMMM lab. The relevant resolved shear stress (RSS) field acting on the (111)/110 dislocations is then obtained by projection. The RSS field is then input to the open-source parallel discrete-dislocation code ParaDis. The precipitates, for which elastic mismatch with the matrix can be neglected, are represented by prismatic dislocation loops, which enables algorithms within ParaDis to automatically prevent the gliding dislocations from shearing the precipitates but without creating any spurious effects on the RSS field. The dislocations in ParaDis are also represented using our new calibration method for the ParaDis dislocation core energy to that of an atomistic potential, replacing the previous ad-hoc representations in ParaDis [3].

Results to date are mainly on the minimal-periodic morphology at the peak-aging conditions. We show that the coherency stresses have a comparatively small effect on the RSS fields and hence on strengthening. The use of a realistic atomistic core energy is seen to affect the strength, demonstrating its importance for the first time. Preliminary results on more-realistic microstructures that match experiments show, as expected, a reduction in strength. We are approaching experimental strengths, and believe we are making the most concerted effort to date in realistic modeling of strengthening in a precipitate microstructure.

1.5 High entropy alloys

We have continued to work on first-principles-based predictions following upon earlier Agility Plus funding. High entropy alloys (HEAs) are separate from the precipitation-strengthened alloys of main interest in D&D2 but represent a major opportunity for additive manufacturing (AM) because HEA properties are robust against the severe processing conditions typical of AM. The HEA effort thus remains within the ultimate goals of D&D2.

We have compute misfit volumes of alloying elements using our DFT method in several medium entropy and binary alloys (NiCoV, NiCoCr, Ni_2Cr, Ni_2V, Ni_2Pd) relevant to recently-discovered alloys. Applying our analytic theory for yield strength, we have predicted the very high strength of NiCoV, relative to all alloys in the well-known Cantor family of Co-Cr-Fe-Mn-Ni, within 10% of new experiments [9, 10]. That paper proposed a more complex but non-derived correlation of strength with electronegativity differences among the alloying elements, and did not make actual predictions. In contrast, we made parameter-free predictions, demonstrating that the strengthening is due to the (computable) large atomic volume of V in these fcc alloys. Vanadium is interesting — it is large in fcc alloys and small in bcc alloys. We have shown that when V is added to either fcc or bcc HEAs, higher strengths are achieved, and have applied our theory to explain these trends across several classes of fcc and bcc alloys [4].

We have also made an in-depth study of NiCoCr due to the high interest in this strong and tough alloy and the possible role of chemical short-range order (SRO). We drove careful experiments by Tsuji *et al.* (Kyoto) so as to obtain the misfit volumes of Ni, Co, and Cr in NiCoCr. We then studied the alloy using standard PBE PAW spin-polarized DFT. Magnetic interactions between Co and Cr lead to predicted misfit volumes for Cr in NiCoCr that are only about 1/2 the experimental value. DFT is thus questionable for subtle structural aspects of NiCoCr. This makes DFT likely unsuitable as a basis for interpretation of EXAFS studies aimed at detecting SRO. This also has implications for work by others on developing interatomic potentials for NiCoCr using DFT. With accurate experimental misfit volumes and elastic constants, we have applied our theory for the random alloy and accurately predict the yield strength of NiCoCr studied to date. We conclude that SRO, whether it exists or not, has no measurable effect on NiCoCr alloys that have been widely fabricated and tested to date [5].

Finally, a recent *Nature* paper showed that Pd additions to the known CoCrFeNi alloy to give CoCrFeNiPd yield (i) very high strength, 3× that of CoCrFeNi, and (ii) non-random fluctuations in the distribution of alloying elements, especially Pd [11]. They speculated that the high strength was due to these fluctuations. We determined the misfit volume of Pd in Ni_2Pd via DFT and from experiments, computed several relevant elastic constants to verify the softening due to Pd addition, and then predicted the strength of *random* CoCrFeNiPd. We obtained good agreement — approximately 20% above experiments at $T = 300$ K and matching experiments at $T = 77$ K. Studying the quasi-

binary alloy $(CoCrFeNi)_{1-x}Pd_x$, we showed that the strength is weakly varying over a wide range of Pd compositions. The theory then also predicts length scales relevant to the dislocation motion that are rather larger than the scale of the non-random fluctuations. Thus, we can firmly state that the high strength in CoCrFeNiPd is due to the large misfit of Pd in CoCrFeNi, in spite of elastic softening, and can rationalize why the alloy strength is insensitive to the observed fluctuations [6].

Overall, our work on HEAs is demonstrating the power of the theory for HEA strength. With first-principles or experimental inputs, the theory is thus extremely useful for the design of complex alloys and for setting limits on performance across a family of alloys. For instance, in the Co-Cr-Fe-Mn-Ni-V family, we predict, and it was experimentally measured, that the $Ni_{0.64}V_{0.37}$ alloy is the strongest alloy in this family. Thus, the complexity alone does not provide special or exceptional strengthening. Instead, the complexity provides a means for easy fabrication of high strength alloys attractive for additive manufacturing.

2 Collaborative and interdisciplinary components

This project is an intimate collaboration between LAMMM (Curtin), providing the main expertise in metallurgy and mechanical properties, and COSMO (Ceriotti), providing the main expertise in advanced machine-learning methods and thermodynamics. The only sub-efforts solely in the Curtin lab are the efforts on dislocation-level modeling of Al-6xxx alloys and on HEAs and those in the Ceriotti lab are efforts on new descriptors for machine learning.

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

- [1] F. Musil, M. J. Willatt, M. A. Langovoy, and M. Ceriotti, *Fast and Accurate Uncertainty Estimation in Chemical Machine Learning*, Journal of Chemical Theory and Computation **15**, 906 (2019).
- [2] M. J. Willatt, F. Musil, and M. Ceriotti, *Feature Optimization for Atomistic Machine Learning Yields a Data-Driven Construction of the Periodic Table of the Elements*, Physical Chemistry Chemical Physics **20**, 29661 (2018).
- [3] Y. Hu, B. Szajewski, D. Rodney, and W. Curtin, *Atomistic dislocation core energies and calibration of non-singular discrete dislocation dynamics*, Modelling and Simulation in Materials Science and Engineering **28**, 015005 (2020).

[4] B. Yin, F. Maresca, and W. A. Curtin, *Vanadium is the optimal element for strengthening in both fcc and bcc High Entropy Alloys, submitted* (2019).

[5] B. Yin, S. Yoshida, N. Tsuji, and W. A. Curtin, *Yield strength and misfit volumes in NiCoCr and implications for short-range-order, submitted* (2019).

[6] B. Yin and W. A. Curtin, *Origin of high strength in CoCrFeNi-Pd High Entropy alloys, submitted* (2019).

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Design & Discovery 3

MARLON: MARVEL Design, Discovery and Engineering of Low-Dimensional Materials and Nanostructures

Project leader: Nicola Marzari (EPFL, 1.5MC)

Computational partners: Mathieu Luisier (ETHZ, 1MC), **Daniele Passerone** (Empa, 1MC), **Alfredo Pasquarello** (EPFL, 0.5MC), Clémence Corminboeuf (EPFL)
Experimental partners: Kumar Agrawal (EPFL), Oliver Gröning (Empa), **Roman Fasel** (Empa, 0.5MC)

1 Progress of the different efforts

D&D3 is dedicated to the design or discovery of novel low-dimensional materials and nanostructures, and in the engineering and integration of these into novel architectures, devices, and processes.

1.1 Novel two-dimensional materials and their properties

This subproject was ignited by the high-throughput study in *Nature Nanotechnology* [1], where we identified more than 1'800 inorganic materials that could be exfoliated into novel monolayers. The project has evolved in many different directions, detailed below, including substantial new collaborations with Swiss experimental groups. We also note in passing that this early effort led to clarify the size-thickness relationships in experimental liquid exfoliation [2] and inclusion, authorship in the general review of the EU Graphene Flagship on production and processing of graphene and related materials [22]. The screening also continues, and thanks to the recent inclusion of the Pauling File and new updates to the COD and ICSD databases (part of the Open Science platform effort), we have doubled the number of novel exfoliable materials.

The first search effort has been dedicated to identifying novel topological insulators; this has recently completed [3], showing that $\sim$ 1% of the materials identified are topological. Among these, the most exciting discovery is that of jacutingaite [4], a mineral first identified in Brazil in 2008, and predicted to be the first ever Kane-Mele quantum spin Hall insulator, and to remain topological even at room temperature. This MARVEL discovery has generated much interest in the community, and several collaborations have started — the first completed one is with the group of Felix Baumberger at UniGE, with the growth (E. Giannini) and measurement (A. Tamai) of high-quality 3D crystals showing dual topology [5] in close agreement with the theoretical predictions, where we generalized the Kane-Mele model to 3-dimensional materials having long-range interlayer hoppings [6].

A second search effort is now dedicated to the identification of high-mobility semiconductors, exploiting the automated AiiDA framework in place [7]. This has led to understanding the role of intervalley scattering as a dominant mechanism for resistivity, and the suggestion that even very small amounts of uniaxial strain can lead to the breaking of the degeneracy between multiple valleys, inhibiting phonon-induced scattering, and increasing conductivity, e.g. of 600% in arsenene, in response to a 2% strain [8]. Currently we are screening our entire portfolio for candidates able to beat the performance of transition-metal dichalcogenides. Several side efforts are also notable — from understanding electron-phonon interactions and their effect on Raman spectra in multivalley materials [23], to the characterization (in collaboration with the experimental group of C. Stampfer in Aachen) of the highest mobility ever recorded in a heterostructure device [9]. The increasing portfolio of electron-phonon interactions will pave the way for a year 7 screening of BCS supercondcutivity in both the portfolio of 2D and 3D parent materials.

A third effort, led by Mathieu Luisier at ETHZ, focuses on materials that could be used as active layers of future field-effect transistors (FETs) beyond the currently manufactured silicon FinFETs. Starting from the database [1], 100 most promising monolayers have now been examined in detail: their bandstructure has been mapped into maximally localized Wannier functions from which a device Hamiltonian matrix is created. Using these data, the current-voltage characteristics of single-gate FETs with a 2D channel material have been simulated with the OMEN quantum transport solver. For all components, both n- and p-type

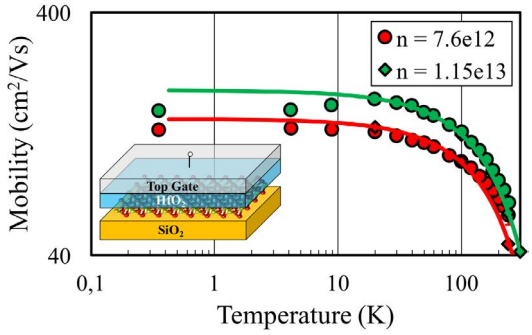

Figure 1: *Comparison between the measured (dots) and calculated (solid lines) é lectron mobility vs temperature" of single-layer MoS_2 at two different carrier concentrations. The MoS_2 monolayer is embedded between a SiO_2 and HfO_2 dielectric environment.*

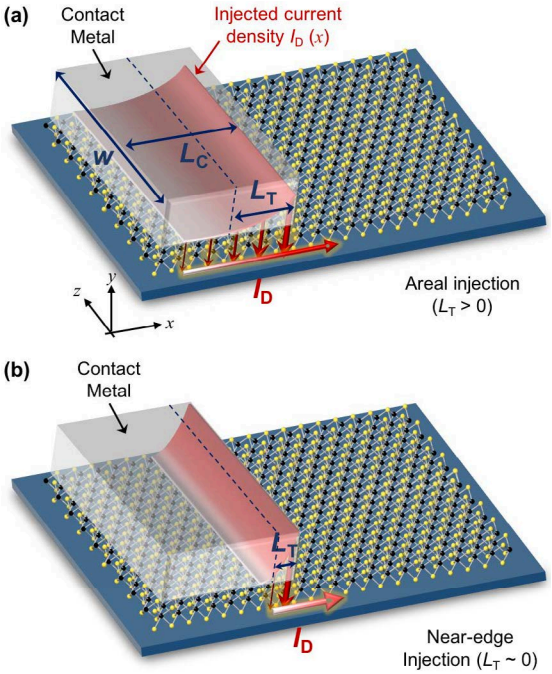

Figure 2: *(a) Area- and (b) edge-dependent electron injection into a MoS_2 monolayer from a titanium contact. L_T refers to the transfer length, L_c to the contact length, and w to its width.*

configurations have been investigated at gate lengths ranging from 15 down to 5 nm. A careful analysis of the results obtained reveals at least 12 compounds with electron and hole currents potentially much higher than future Si FinFETs. Among these, some are already known, e.g. GeS, GeSe, Sb, or black phosphorus (P_4), but others have not been identified before; for example Ag_2N_6, O_6Sb_4, or TiNCl. The most promising materials have been further simulated with a double-gate architecture, which increases their ON-state current and improves scalability down to ultra-short gate lengths. To go one step further we complemented the effort on carrier mobilities limited by electron-phonon interactions, adding the influence of charged impurities and surface optical phonon scattering. When all these effects are taken into account, excellent agreement with experimental data can be achieved, as illustrated in Fig. 1 for single-layer MoS_2 [10] (measurements taken from [24]).

Since two dimensional devices, especially those relying on single-layer materials, are often limited by their contact resistance, metal-semiconductor structures were created at the atomic level and the electrical current flowing through them evaluated to better understand the underlying physics. It was found that electrons tend to be transferred at the edge of the "contact to the 2D material" interface if the latter is clean (edge process), while the presence of an interfacial layer, for instance a native oxide, extends the transfer region (area process) [11]. Both transfer mechanisms are illustrated in Fig. 2. To be able to perform device simulations beyond the ballistic limit of transport while reducing the computational burden, the numerical algorithms of the OMEN tool were enhanced, in particular the communication scheme in case of self-heating. For large structures composed of 10'000 atoms or more, a speed up factor of 140 could be demonstrated and a sustained performance of $>$ 85 Pflop/s was reached. These results were awarded the ACM Gordon Bell Prize 2019 [12].

A fourth search effort has also started, dedicated to other properties than electronics — these include the systematic study of these novel 2D materials for photocatalytic water splitting and their application as membranes for gas separation (the latter involves an experimental collaboration with Kumar Agrawal at EPFL Sion, with a joint PhD student [13]). As part of the effort in identifying novel materials for photocatalysis, the effect of the solvent on the oxygen evolution reaction (OER) was studied by the group of Alfredo Pasquarello. First, it was elucidated that the main effect consists in modifying the free energy steps due to the electrostatics associated with the charging of the reaction intermediates at the interface [14]. Then, following the proposal by the group of Clémence Corminboeuf that a bifunctional mechanism [15] in the oxygen evolution reaction can lead to a reduction of the reaction overpotential below the limit imposed by the linear scaling relationships, a focus was put on identifying materials where a second active site acts as a hydrogen acceptor, creating a thermodynamically more favorable reaction interme-

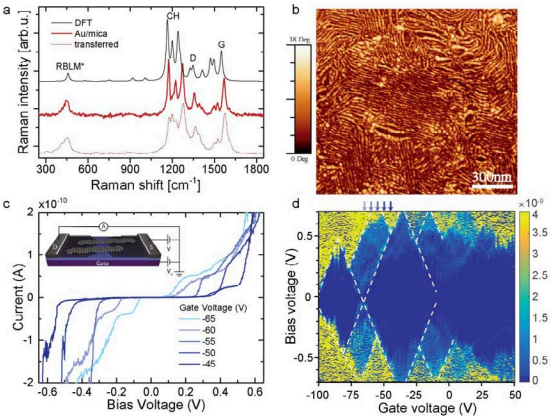

Figure 3: Ex situ *characterization of pGNR films and pGNR devices. (a) Raman spectra of pGNR (top) simulated by DFT, (middle) acquired directly on the Au/mica growth substrate, and (bottom) after substrate transfer to device substrates. (b) AFM topography image of pGNRs transferred onto an Al_2O_3 substrate. (c) Current-voltage traces at various gate voltages for a typical device, recorded at 9 K. The gate voltages at which these curves were recorded are marked with arrows in panel (d). The inset shows the schematic of the device and measurement layout. (d) dI/dV-map of the device.*

diate. The performance of this mechanism was investigated by considering a selection of materials and their combinations, finding that several pairings of catalysts can lead to a reduction of the overpotential with respect to the regular OER mechanism, with some combinations resulting in overpotentials as low as 0.27 eV.
Last, on the fundamental theoretical side, we have shown how the edges of polar 2D materials can host intrinsic excitons [16] and how the hydrodynamics regime of heat propagation, that we first suggested in 2015 [17] and that was first experimentally seen in 2019 [25], can be described with a novel mesoscopic theory that coarse grains the Boltzmann transport equation into coupled viscous heat equations [18] that encompass both the Fourier and the hydrodynamic regimes.

1.2 Novel two-dimensional nanostructures

Here we explore how novel two-dimensional nanostructures can be predicted by simulations, and how these can actually be assembled in a real device. The general paradigm is based at this stage on graphene and graphene nanoribbons (GNRs), thanks to the exquisite synthesis capabilities at Empa.
Earlier transport simulations pinpointed poor electronic coupling at the graphene-GNR interface as the major intrinsic factor limiting the current modulation and hence the device performance in terms of transistor application. The low injection efficiency of charge carriers from the leads results in a resonant transmission characteristic which translates to a weak modulation of the current with gate voltage. So, a major effort has been put in the optimization of the contact between graphene leads and nanoribbons, with a focus on 9-AGNR (armchair GNR) and pyrene. Contacting pyrene graphene nanoribbons (pGNRs) to graphene leads was our first attempt towards "all carbon devices". The first step towards device integration is to demonstrate the robustness of pGNRs under ambient conditions. We use Raman spectroscopy to verify the structural integrity of the ribbons after exposing them to air comparing to DFT Raman calculations which allow us to attribute the Raman peaks to specific phonon modes of pGNR (Fig. 3a). For the integration of pGNRs into FET devices we use graphene as electrode material because of its atomic flatness. This allows a "GNRs-last" fabrication process that avoids additional processing steps, which might introduce defects in the transferred pGNRs films. We probed the electrical properties of the device by measuring the current as a function of both the bias and gate voltage. At room temperature, we observe a linear dependence of the current on the applied bias voltage, and a negligible dependence of the applied gate voltage. We attribute this quasi-metallic behavior to the small bandgap of the pGNR. Figs. 3c and d display transport measurements at low temperature (9 K). In the $I-V$ curves we observe Coulomb blockade of charge transport at low bias voltages. The onset of current at higher bias is tunable via the applied gate voltage. This is attributed to transport channels of the pGNRs entering the bias window. We observe currents of up to 10 nA at 0.7 V bias, which we assume to be limited by coupling of the graphene electrodes to the pGNR channel. Fig. 3d shows the corresponding stability diagram (differential conductance dI/dV as a function of bias and gate voltages) in which diamond-like features are visible. These Coulomb diamonds are characteristic of weakly coupled quantum dots and indicate that transport occurs through multiple pGNRs but is dominated by few ribbons. The observed blockade regions range from ~ 0.2 to ~ 0.75 V bias. This agrees well with the range of transport gaps expected for pGNRs if one takes into account GW-corrections for electron-electron interaction, the insulating substrate and the range of observed ribbon lengths.

a) *Key achievements: GNR-lead orientation, adatom intercalation* A selection of structures

was created at the atomic level and the electrical transmission has been examined: the geometries were relaxed, and the optimized DFT Hamiltonian and overlap matrices were passed to OMEN to compute the transmission characteristics (fundamental here are the interactions between Empa and Mathieu Luisier at ETHZ). The analysis of the results revealed a potential configuration with high injection efficiency, where the GNR-lead relative orientation must match that of the AB stacking in graphite. As suggested by Marzari [26] the inclusion of metal adatoms as electronic bridges can enhance the coupling between the electronic states of the GNR and the leads: the d-orbitals of the adatoms lie close in energy to the π-manifolds of the two regions and hybridize, forming a continuum of states for an efficient charge transfer.

b) *Connection to experiments* To apply the theoretical findings, the following strategies are currently considered in real experimental setups. A single crystal graphene sheet will be grown on SiO_2 substrate for the contacts. Contrary to the CVD strategy employed at the present, which delivers a graphene layer made of non-aligned patches, the method allows to know the (single) graphene orientation prior to the GNRs transfer. As for the intercalation of the adatoms, experiments are on the way to allow the setup of the contact/GNR device in presence of a gold atom atmosphere. Deployment of routine computational tasks to scientists from the experimental unit was possible thanks to recent ameliorations to the "On surface chemistry" app of AiiDA lab. The increased yield observed in simulations boosted the research activity towards novel nanostructures and allowed us to obtain a number of successful results opening the route towards nanoribbons incorporating porphynes [19] as well as exploring nanostructures with open shell character [20, 21]. These include the bow-tie-shaped nanographene $C_{38}H_{18}$ (Clar's goblet) envisioned in 1972 by Erich Clar where topological frustration in the π-electron network renders it impossible to assign a classical Kekulé structure without leaving unpaired electrons, driving the system into a magnetically non-trivial ground state appealing for room-temperature carbon-based spintronics.

c) *Theoretical developments* The evidence of Coulomb blockade in the $I-V$ curves poses a major challenge for simulations. To go one step further, dynamics of out-of-equilibrium excitations such as screening of added electrons or holes, e.g. from the leads to the GNR and vice versa, are currently under study. In the specific, a self-consistent transport calculator based on non-equilibrium Green's function (NEGF) and dynamical mean-field theory (DMFT) is under development and has already proved its validity for small model systems. Starting from the DFT calculation, a model Hamiltonian is constructed by subdiagonalizing the DFT Hamiltonian in the full basis set and extracting the atomic orbitals at low energy and involved in the electron transmission. In this reduced active space DMFT self-consistency is obtained and the converged correlated self-energy used in NEGF calculations to compute transmission quantities. For large structures, composed of thousands of atoms, the efficiency of the numerical algorithms must be enhanced.

2 Contribution to overall goals and initial proposal

D&D3 is very closely aligned to the overall goals of the project: to discover novel materials, to ramp up simulations to be able to investigate entire devices, and to do this in close collaboration with experimentalists, in addition to developing novel methods. EPFL, ETHZ and Empa are doing this by joining forces to shed light on the performance of next-generation transistors based either on novel 2D materials or graphene nanoribbons. "$I-V$" characteristics, scalability, mobility, and contact resistance have been explored at the *ab initio* level by combining several tools (Quantum-ESPRESSO, CP2K, Wannier90, and OMEN — this latter receiving this year the ACM Gordon Bell Award, and now integrated to work with both Quantum-ESPRESSO/Wannier90 and CP2K).

3 Collaborative and interdisciplinary components

Besides the collaborations between EPFL, ETHZ, and Empa internal to the project, several key efforts within Switzerland, and outside, have started. For EPFL, synthesis of jacutingaite has started at UniGE (Dr. Enrico Giannini), with high-quality 3D samples that have shown emergent dual topology in the ARPES experiments of Dr. Anna Tamai and Prof. Felix Baumberger [5], followed by STM (Prof. Christoph Renner) and transport experiments (Prof. Alberto Morpurgo), with exfoliation of monolayers in the group of Prof. Francesco Stellacci (EPFL). ETHZ is working closely with the groups of Profs. Lukas Novotny (ETHZ) and Aaron Franklin (Duke

Univ.) on MoS_2 contact issues, of Prof. Eric Pop (Stanford) on mobility calculations, of Prof. Adrian Ionescu (EPFL) on the modeling of 2D band-to-band tunneling FETs, and of Prof. Torsten Höfler (ETHZ) on the improvement of the OMEN software. Empa has started a dedicated collaboration with the Technical University of Vienna (Dr. Angelo Valli) for the development of DMFT/NEGF self-consistent transport calculators.

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

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Design & Discovery 4 Nanoporous Materials Genome: Optical, Catalytic and Electronic Properties

Project leader: Berend Smit (EPFL, 2MC)

Computational partners: Jürg Hutter (UZH, 1.5MC), **Alfredo Pasquarello** (EPFL, 1.5MC), **Ivano Tavernelli** (IBM, 1MC)
Experimental partners: Marco Ranocchiari (PSI, 0.33MC + 0.33MC from PSI), **Piero Macchi** (UniBE, 0.33MC), **Kyriakos Stylianou** (EPFL, 0.33MC)

1 Progress of the different efforts

D&D4 aims to develop and apply computational method to better understand the properties of metal organic frameworks (MOFs). The development of novel technics have focussed on optical and electronic properties of MOFs, while the application focus on separations (e.g. capture CO_2 from flue gasses). The experimental work has focussed on the exploration of catalysis and other applications of MOFs.

The group of *Hutter* has focussed on the development methodologies for the large-scale calculation of absorption spectra for nanoporous materials, with the focus on absorption spectra. We therefore implemented a recently proposed semi-empirical ansatz dubbed Simplified Tamm Dancoff approximation in the CP2K program package. This implementation allows for the calculation of periodic systems and it is also possible to combine it with tight-binding ground-state references, enabling to treat large system sizes. Logarithmic timings comparing again the conventional and the simplified approach show a gain in computation time of about two orders of magnitude, reducing the time to calculate a spectrum of 100 excited states from minutes to seconds (Fig. 1).

The second project in the group of *Hutter* focussed on benchmarking tight-binding approach denoted GFN-xTB. This method was examined for its suitability for high-throughput screening of periodic systems. GFN-xTB was already proven to be a robust and efficient tight-binding ansatz for molecular systems and, with this study, we extended the application range to metal organic frameworks, examining a test set of 10 diverse MOFs and 800 DFT optimized structures taken from the CoRE MOF database. Logarithmic timings reveal that an overall gain of about 2 orders of magnitude can be expected regarding computation time. While GFN-xTB geometry data (volumes, RMSDs) differ by up to 10% in comparison to GGA-optimized structures, thereon-based pore analytics (pore limiting diameter (PLD), largest cavity diameter (LCD) and accessible volume fraction) yields similar results for both GFN-xTB and GGA functionals, suggesting to use the semi-empirical ansatz for pre-screening of pore properties.

The group of *Pasquarello* developed a time-dependent approach in which a dielectric-dependent hybrid functional (TD-CAM) determined in a self-consistent manner without any empirical parameters is used for the calculation of the optical absorption. The proposed method correctly captures the electron-hole interaction in the full range. Thus, TD-CAM allows us to calculate the spectra at a fraction of the computational cost of the Bethe-Salpeter equation-GW scheme. As shown in Fig. 2, the spectra calculated with TD-CAM are in good agreement with both experiment and the more demanding BSE-*GW* approach. The main benefit of our approach is the fact that the most demanding part of the calculation (*GW*) can be circumvented, allowing us to address in perspective metal-organic frameworks of much larger size.

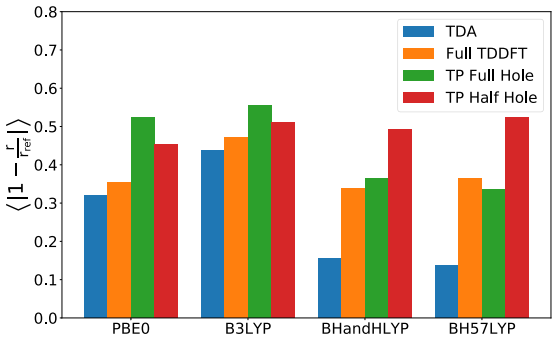

Figure 1: *A mean absolute deviation of the first oscillator strength ratios with respect to a CCSD reference. TDDFT methods in general and the Tamm-Dancoff approximation (TDA) in particular show systematic improvement with respect to transition potential (TD) methods, in particular for functionals with high level of exact exchange.*

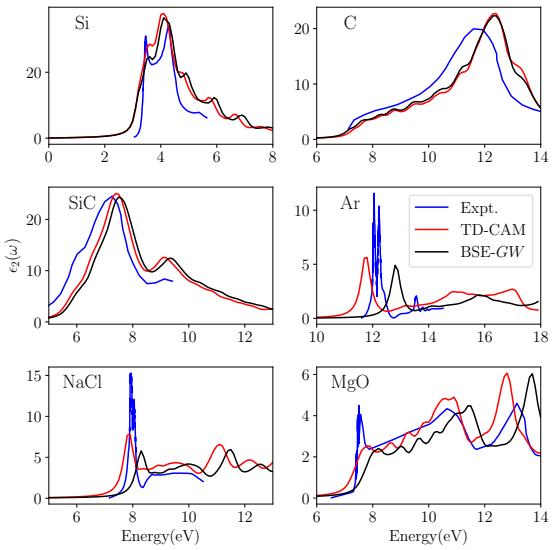

Figure 2: *Absorption spectra calculated for Si, C, SiC, Ar, NaCl, and MgO with BSE-GW and the time-dependent hybrid functional approach.*

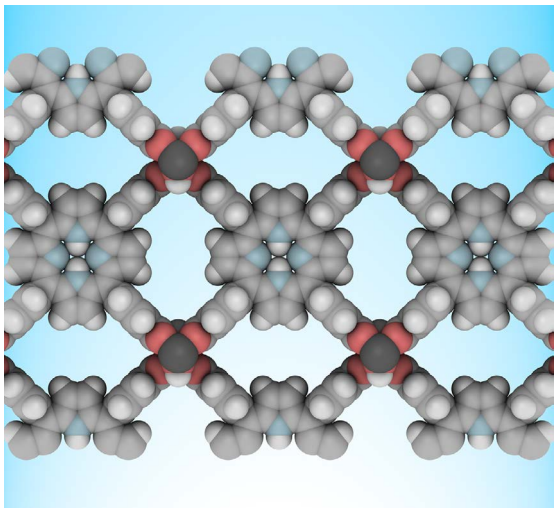

Figure 3: *A molecular representation of the carbon capture material Al-PMOF (iRASPA software was used for visualization).*

A second project in the group of *Pasquarello* involves an investigation of the role of NiO and Ni_2P as co-catalysts by calculating the band edge alignment at NiO/H_2O and Ni_2P/H_2O interfaces. We demonstrated that both NiO and Ni_2P show a favorable energy alignment for the hydrogen evolution reaction, while only NiO is suitably aligned for the oxygen evolution reaction. Our results are in agreement with photoluminescence experiments of NiO and Ni_2P used in combination with the metal organic framework MIL-125-NH_2 .This project is done in collaboration with the experimental group of *Stylianou*.

The work in the group of *Tavernelli* involves setting up an automatized screening protocol to address the potential of metal-organic frameworks (MOFs) to be used as photocatalysts. The requirements that a MOFs must fulfill to be a good photocatalyst, are, (1) absorption in the visible, (2) appropriate band alignment, (3) large charge separation, and (4) high mobility. The aim of this project is to establish descriptors that translate these required properties into computable values. That is (1) optical gap, (2) ionization potential/electron affinity, (3) spatial electron-hole overlap, and (4) effective mass. These descriptors have been computed at high level of theory and we evaluated cost-effective ways to estimate these values. A linear correlation was established of +0.85 eV between the band gap at PBE level and the optical gap at PBE0 level. This protocol was applied to a test-set of MOFs and we evaluated the performance of the approximations to discriminate between promising candidates already found in the literature.

The work in the group of *Smit* focussed on screening materials for carbon capture for wet flue gasses. A few promising materials were identified (Fig. 3) and subsequently synthesized in the group of *Stylianou*. These materials could indeed capture CO_2 from wet flue gasses and outperformed commercial materials [1]. A related project in the group of *Smit* focussed on creating an AiiDA workflow to computed the parasitic energy for a carbon capture process using covalent organic frameworks (COFs) [2]. In addition, the group of *Smit* developed an efficient method to compute diffusion coefficients from energy grids [3].

The group of *Ranocchiari* focussed on the application of microporous materials in catalysis. The micropores in metal-organic frameworks (MOFs) push homogeneous catalytic reactions into kinetic regimes inaccessible under standard conditions. Such property allows branched selectivity up to 90% in the Co-catalysed hydroformylation of olefins without directing groups, not achievable with existing catalysts. This finding has a big potential in the production of aldehydes for the fine chemical industry. Monte Carlo and density functional theory simulations, combined with kinetic models show that the micropores of MOFs with UMCM-1 and MOF-74 topologies increase the olefins density beyond neat conditions while partially preventing the adsorption of syngas leading to high branched selectivity. This effect, which we call adsorption-driven kinetic modulation (AKM), has important consequences in catalysis. The aim is to find catalytic applications that would strongly benefit from

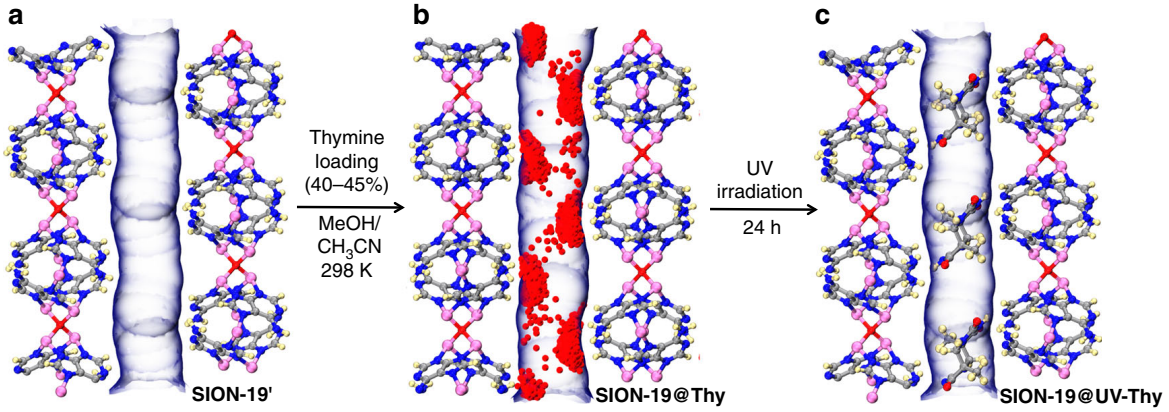

Figure 4: *Schematic representation of thymine loading, location, and dimerization within SION-19'. (a) 1-dimensional base-pore surface for SION-19' prepared using HOLE2. (b) Thy snapshots (red dots) from an MD trajectory of 44% thymine loading in the base pore of SION-19. Here, Ade acts as a structure directing agent that "lock" Thy molecules into positions close enough to another Thy molecule for photodimerization to occur. (c) DFT optimized positions of Thy<>Thy dimers arising from a 44% loading of Thy. Atom color code: pink, Zn; red, O; blue, N; gray, C; light yellow, H (from [4]).*

this effect leading to catalysis that cannot be done in any other way. This work is done in collaboration with the group of *Smit*.

The group of *Macchi* investigated the effects of the adsorption of guest molecules on the dielectric properties of the MOFs HKUST-1, and $\{Zn_3(BTC)_2\}_n$, and in particular the role of chemi- and physisorbed guest molecules and that of specific intra-framework and framework-guest linkages [5]. This study included electron charge density analysis, impedance spectroscopy, density functional theory simulations, and atomic partitioning of the polarizabilities. These analyses at different degrees of pores filling enabled one to observe structural and electronic changes induced by guest molecules, especially when chemisorbed. The electrostatic potential inside the pores allows one to describe the absorption mechanism and to estimate the polarization of guests induced by the framework. The dielectric constant shows very diverse frequency dependence and magnitude of real and imaginary components as a consequence of (i) capture of guest molecules in the pores during synthesis, (ii) MOF activation, and (iii) water absorption from the atmosphere after activation.

The group of *Stylianou* applied rational design to synthesis of a novel bio-MOF featuring unobstructed Watson-Crick faces of adenine (Ade) pointing towards the MOF cavities [4]. The thymine (Thy) molecules are packed within the channels in a way that fulfill both the Woodward-Hoffmann and Schmidt rules, and upon UV irradiation, Thy molecules dimerize into Thy<>Thy. This study highlights the ability to "lock" molecules in specific positions that can be subsequently dimerized upon light irradiation, extending the use of MOFs as nanoreactors for the synthesis of molecules that are otherwise challenging to isolate (Fig. 4). Other studies performed in the group of *Stylianou* involve the development of a MOF-based sensor to accurately measure Fluor in drinking water [6], the capturing of chemical intuition from failed and partially successful experiments [7], the synthesis of metal-doped MOFs, which were computationally predicted to have improved optical properties [8].

2 Contribution to overall goals and initial proposal

2.1 Objectives

The aim of this design and discovery project is to extend the existing Nanoporous Materials Genome effort to new applications. The common aspect of these new applications is that they require a fundamental understanding of the electronic structure of these materials and how the electronic structure changes upon external stimuli. The overall project involves the following three components:

1. generation of novel photoactive and conductive materials and sensors,
2. prediction of the materials' properties,
3. screening databases of materials to discover materials with optimal performance.

Significant progress has been made in the development of computational methods to study

the electronic and optical properties of MOFs. We expect that in the next period these methods will be used for the first screening study. On the experimental side, the highlights are that some of the computational predicted materials have been successfully synthesized. In particular, the material for carbon capture [1] and the metal doped MOF [8] are both materials that have been computationally designed before the actual synthesis.
Of concern is that both the groups of *Macchi* and *Stylianou* have left Switzerland, although Stylianou had been replaced by Chris Ireland.

3 Collaborative and interdisciplinary components

D&D4 relies on collaboration between the different groups. Several publications have appeared that involve collaborations between the experimental and computational groups (e.g. [1, 8, 4, 9], other are submitted or are in an advanced stage of being written). Also articles that involve collaborations between the computational groups are submitted.
Work that involves different D&D groups includes [10], which discribes an extension of energy surfaces to orbital surfaces.

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

[1] P. G. Boyd, A. Chidambaram, E. García-Díez, C. P. Ireland, T. D. Daff, R. Bounds, A. Gładysiak, P. Schouwink, S. M. Moosavi, M. M. Maroto-Valer, J. A. Reimer, J. A. R. Navarro, T. K. Woo, S. Garcia, K. C. Stylianou, and B. Smit, *Data-driven design of metal-organic frameworks for wet flue gas CO_2 capture*, Nature **576**, 253 (2019).

[2] D. Ongari, A. V. Yakutovich, L. Talirz, and B. Smit, *Building a Consistent and Reproducible Database for Adsorption Evaluation in Covalent-Organic Frameworks*, ACS Central Science **5**, 1663 (2019).

[3] A. Mace, S. Barthel, and B. Smit, *Automated Multiscale Approach To Predict Self-Diffusion from a Potential Energy Field*, Journal of Chemical Theory and Computation **15**, 2127 (2019).

[4] S. L. Anderson, P. G. Boyd, A. Gładysiak, T. N. Nguyen, R. G. Palgrave, D. Kubicki, L. Emsley, D. Bradshaw, M. J. Rosseinsky, B. Smit, and K. C. Stylianou, *Nucleobase pairing and photodimerization in a biologically derived metal-organic framework nanoreactor*, Nature Communications **10**, 1612 (2019).

[5] R. Scatena, Y. T. Guntern, and P. Macchi, *Electron Density and Dielectric Properties of Highly Porous MOFs: Binding and Mobility of Guest Molecules in $Cu_3(BTC)_2$ and $Zn_3(BTC)_2$*, Journal of the American Chemical Society **141**, 9382 (2019).

[6] F. M. Ebrahim, T. N. Nguyen, S. Shyshkanov, A. Gładysiak, P. Favre, A. Zacharia, G. Itskos, P. J. Dyson, and K. C. Stylianou, *Selective, Fast-Response, and Regenerable Metal–Organic Framework for Sampling Excess Fluoride Levels in Drinking Water*, Journal of the American Chemical Society (2019), doi:10.1021/jacs.8b11907.

[7] S. M. Moosavi, A. Chidambaram, L. Talirz, M. Haranczyk, K. C. Stylianou, and B. Smit, *Capturing chemical intuition in synthesis of metal-organic frameworks*, Nature Communications **10**, 539 (2019).

[8] M. A. Syzgantseva, C. P. Ireland, F. M. Ebrahim, B. Smit, and O. A. Syzgantseva, *Metal Substitution as the Method of Modifying Electronic Structure of Metal-Organic Frameworks*, Journal of the American Chemical Society **141**, 6271 (2019).

[9] A. Gładysiak, S. M. Moosavi, L. Sarkisov, B. Smit, and K. C. Stylianou, *Guest-dependent negative thermal expansion in a lanthanide-based metal-organic framework*, CrystEngComm **21**, 5292 (2019).

[10] B. Meyer, S. Barthel, A. Mace, L. Vannay, B. Guillot, B. Smit, and C. Corminboeuf, *DORI Reveals the Influence of Noncovalent Interactions on Covalent Bonding Patterns in Molecular Crystals Under Pressure*, The Journal of Physical Chemistry Letters **10**, 1482 (2019).

Design & Discovery 5 Correlated Transition Metal Oxides and Heterostructures

Project leader: Nicola Spaldin (ETHZ, 1MC)

Computational partners: Ulrich Aschauer (UniBE, 1MC), **Claude Ederer** (ETHZ, 1MC), **Philipp Werner** (UniFR, 1MC)
Experimental partners: Marisa Medarde (PSI, 0.5MC + 0.5MC from PSI), Urs Staub (PSI)

1 Progress of the different efforts

1.1 Nickelates (theory)

Our two papers demonstrating how the metal-insulator transition in the rare earth nickelates stabilizes the structural breathing mode distortion have now been published in *npj Quantum Materials* [1] and in *Physical Review B* [2]. Alexander Hampel has graduated and has taken on a prestigious postdoc position with the Simons Foundation (the Flatiron Institute) in New York.
Further calculations for nickelates have been performed including the effect of magnetic order. The results are not yet fully conclusive, nevertheless they demonstrate that DFT+DMFT calculations using only a minimal e_g-based correlated subspace do indeed allow to reproduce the complex magnetic order with wave-vector $\boldsymbol{q} = (1/4, 1/4, 1/4)$ observed in the nickelates. The calculations also show that the emergence of magnetic order enhances the charge disproportionation and thus pushes the system towards the charge-disproportionated insulating state. However, the calculations also seem to indicate certain limitations of the minimal e_g-only description. Specifically, it appears to be difficult to stabilize the magnetic order in the fully charge-disproportionated case, when the occupation of the short-bond Ni site approaches zero. This could indicate the importance of the "uncorrelated" bands with predominant oxygen p character for mediating the magnetic coupling.
We have further used $LuNiO_3$ as a prototypical example for a charge-ordered (or charge-disproportionated) system to study the effect and importance of full charge self-consistency in DFT+DMFT calculations. Thereby, we have compared the *site-polarized* case of $LuNiO_3$ with the *orbitally polarized* case of strained $CaVO_3$. This allowed us to clearly distinguish the effect resulting from the DMFT charge-density correction, which tends to produce a more homogeneous charge distribution, from effects related to the double-counting correction, which tend to strongly enhance a potential site polarization. A manuscript summarizing our findings has been published on *arXiv* [3] and is currently under review.
We have also started to extend our studies to the case of $CaFeO_3$. The Fe^{4+} cation in this material is isoelectronic (d^4) to the Mn^{3+} cation in the prototypical Jahn-Teller-distorted insulator $LaMnO_3$, but $CaFeO_3$ exhibits the same breathing mode distortion as the rare earth nickelates [13]. This material is thus a very interesting system to study a possible competition between Jahn-Teller and breathing mode distortions and the corresponding interplay with the electronic properties of the material.

1.2 Nickelates (experiment)

After the successful growth of the first $RNiO_3$ single crystals during MARVEL phase I, we have pursued our re-investigation of the nickelate phase diagram. Our efforts have been focused in two directions: on one side, obtaining new, high-quality structural and physical property data to be contrasted with the predictions of our theory partners; on the other, exploring regions of the $RNiO_3$ phase diagram where novel physics and exotic phenomena are likely to occur.
In the first case, we started with a detailed investigation of the gap opening and the simultaneous emergence of charge order in $PrNiO_3$ [4]. We combined electrical resistivity, magnetization, and heat capacity measurements with high-resolution neutron and synchrotron X-ray powder diffraction (Fig. 1). In order to ease comparisons with our DFT and DMFT calculations, we analyzed the diffraction data in terms of symmetry-adapted distortion modes. This type of analysis allowed us to identify the contribution of the different modes to the global distortion, their evolution across the metal-to-insulator transition (MIT), and their coupling schemas in the metallic and insulating phases. Moreover, it unraveled the existence of a nearly perfect linear correlation between

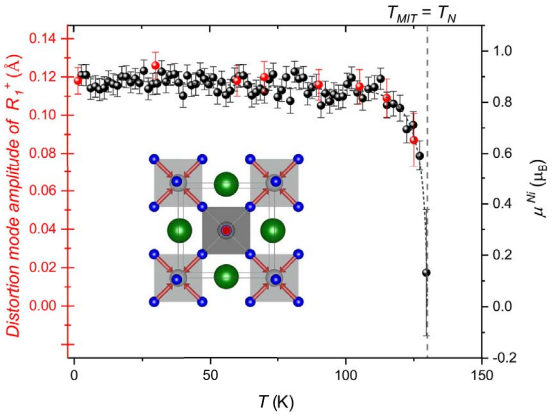

Figure 1: *Temperature dependence of the breathing mode amplitude (left axis) and the Ni staggered magnetization (right axis), as obtained from neutron powder diffraction measurements for $PrNiO_3$. The vertical dashed line marks $T_{MIT} = T_N$ ($\sim$ 130 K). The small-dashed line is a guideline for the eye. The insert illustrates the atomic displacements in the breathing mode (from [4]).*

the amplitude of the breathing mode associated to the charge order and the Ni staggered magnetization below T_{MIT}. Our data also uncovered a previously unnoticed anomaly at $T^* \sim 60$ K ($\sim 0.4 \times T_{MIT}$), clearly visible in the electrical resistivity, lattice parameters and some mode amplitudes. Since phase coexistence is only observed in a small temperature region ($\sim \pm 10$ K) around T_{MIT}, these observations suggest the existence of a hidden symmetry in the insulating phase. A possible origin could be the theoretically predicted existence of polar distortions induced by the non-centrosymmetric magnetic order [5]. We plan to check this hypothesis on $YNiO_3$, where the magnetic and electronic transitions are well separated and the existence of lattice anomalies at the Néel temperature T_N, if any, should be easier to detect.

In $RNiO_3$ nickelates, the region of the phase diagram where T_{MIT} approaches zero limit has been very little investigated, both experimentally and theoretically. However, this region may host exotic physics due to the possible existence of charge, magnetic, and even ferroelectric quantum fluctuations. An open question, recently addressed by the theory partners, is how the lattice and electron-lattice energy scales evolve along the nickelate phase diagram. Particularly open is how the electron-lattice term, usually much smaller than the electronic energies, develops in the vicinity of $T_{MIT} = 0$, where the electronic energies become also very small. To address these questions, we started an experimental investigation of this region, that we accessed through the preparation of $Pr_{1-x}La_xNiO_3$ solid solutions ($0 < x < 1$). In order to evaluate the role of the lattice in the MIT, we exchanged the oxygen isotope (mostly ^{16}O in natural oxygen) by heavier ^{18}O. This resulted in a substantial T_{MIT} increase (+8.5 K for $PrNiO_3$), which grows in an approximately exponential way with x on approaching the $T_{MIT} \to 0$ limit. Synchrotron X-ray diffraction experiments aimed to investigate the impact of the O isotope effect in the crystal structure and the expansion coefficient will be carried out at the Swiss Light Source in February 2020. Transport measurements are in progress in order to evaluate the impact of the O isotope substitution in the electronic mobility.

1.3 Complex oxide heterostrutures

We have continued our studies of metal-insulator transitions in oxide heterostructures, focusing in particular on potential charge transfer across interfaces between two different complex oxides with different transition metal cations and different filling of the corresponding d bands. Such charge transfer can lead to the emergence of a quasi-two-dimensional metallic layer, which is very promising for potential applications such as, e.g., Mott transistors. In particular, we have identified the emergence of a metallic layer at the interface between the two Mott insulators $LaTiO_3$ and $LaVO_3$, and demonstrated the high tunability of this metallic layer, e.g., by applying strain (Fig. 2). A paper on these results has been published in *Physical Review Materials* [5].

Continuing our efforts along these lines, we are now investigating $LaVO_3/SrVO_3$ heterostructures. Here, the transition metal (V) is chemically identical on both sides of the interface, but changes its oxidation state from V^{3+} (d^2) in $LaVO_3$ to V^{4+} (d^1) in $SrVO_3$. Furthermore, the stoichiometry of the system does not support an integer d level occupation in the interfacial layer (for symmetric interfaces), leading to a robust metallic character and an intermediate valence of $V^{3.5+}$ in the interfacial layer. We are currently exploring the possibility for a spontaneous charge ordering in this interfacial layer, as suggested by previous DFT+U calculations [14].

Simultaneously, we have studied different ways to model interfaces between two materials exhibiting a so-called *polar discontinuity*, using multilayers of $LaAlO_3$ and $CaVO_3$ as a model system. Different ways of building slab supercells, either in the form of non-

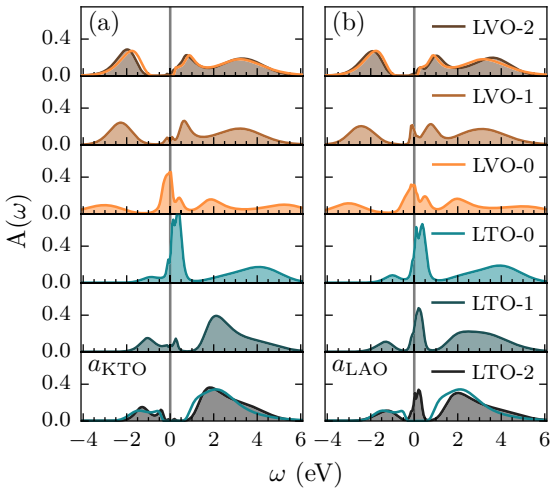

Figure 2: *Layer-resolved spectral function (corresponding to the transition metal d orbitals) for a $LaVO_3/LaTiO_3$ multilayer under (a) tensile and (b) compressive strain. In the tensile case the metallic character is constrained to the two interface layers (LVO-0 and LTO-0), while the innermost layers (LVO-2 and LTO-2) are very similar to the corresponding insulating bulk spectral functions (shown as orange and blue lines). Under compressive strain, the metallicity reaches further into the $LaTiO_3$.*

stoichiometric but symmetric, stoichiometric but asymmetric (polar), or both stoichiometric and symmetric slabs (using a vacuum-separated tri-layer), impose different electrostatic boundary conditions on the system, and according to our calculations can lead to drastically different properties of the corresponding interface. Considering the fact that our previous calculations have also shown a high sensitivity of the metal-insulator transition in $CaVO_3$ both to strain and layer thickness [6, 7], this suggests an extremely high tunability of the $LaAlO_3/CaVO_3$ heterostructure, provided that the specific boundary condition can be accurately controlled experimentally. A manuscript on these insights is currently under preparation.

1.4 Mott solar cells

In this collaboration between the Werner and Ederer groups we implemented a semi-realistic simulation of light harvesting in Mott insulating solar cells, which are few-layer heterostructures of $LaVO_3$ (or $YTiO_3$) on top of $SrTiO_3$. Our paper discussing the role of Hund's interaction on the impact ionization processes in a semi-realistic $SrTiO_3/LaVO_3/SrTiO_3$ heterostructure was published this year in *Physical Review B* [8]. This work represents the first combination of non-equilibrium dynamical mean field theory with *ab initio* band structure input.

1.5 DFT+U+V treatments of defects in complex oxides

The first application of the self-consistent site-dependent DFT+U formalism was finalized in year 6, published [9] and presented at various international and national conferences. After this successful application to $SrMnO_3$, we continued testing the method on $SrTiO_3$ but found that the underestimation of the band gap causes severe artifacts for delocalized defect states. In a partially covalent transition-metal oxide, the band gap can be corrected for using the DFT+U+V methodology that complements the intra-site interactions on transition metals in DFT+U with inter-site interactions between the transition metal and the oxygen ligands. We found this self-consistent approach to work remarkably well, resulting in band gaps and oxygen vacancy formation energies in very good agreement with experiment and hybrid functional calculations, however at a fraction of the computational cost of the latter (Fig. 3). This work is still pending publication. It is also encouraging that the approach delivers qualitatively the same electronic structure in terms of trapped and free electrons as those obtained with a more involved DFT+DMFT

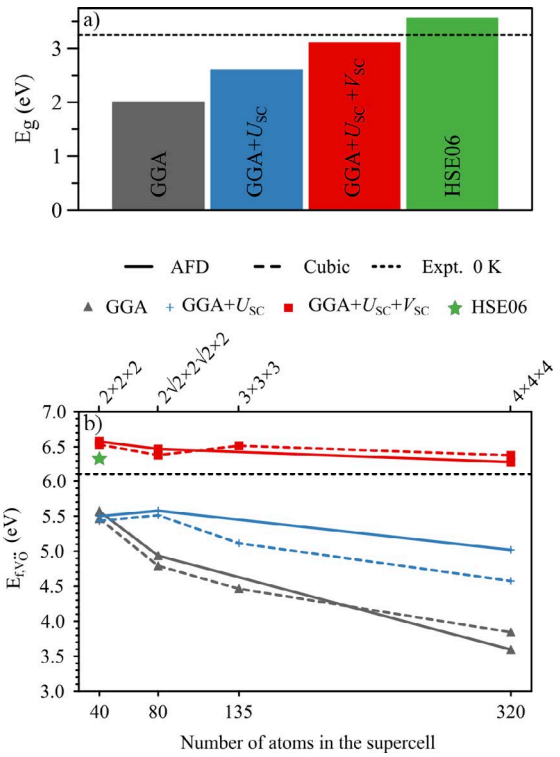

Figure 3: *(a) Band gaps and (b) formation energies of oxygen vacancies computed at the GGA, GGA+U, GGA+U+V and HSE06 levels of theory.*

treatment.
Given that the self-consistent site dependent DFT+U was shown to work well for $SrMnO_3$, we also applied it to the related material $LaMnO_3$, finding an interesting interplay of the Jahn-Teller distortion with the vacancy formation and for polar defect pairs in $SrMnO_3$, where we show that defects can support ferroelectricity at lower strains than in the stoichiometric material. These manuscripts are currently in preparation and are expected to be published during year 7.

1.6 DFT+DMFT description of defects in complex oxides

We continued to study the effect of defects on the functional properties of complex transition metal oxides, and to assess the predictive capabilities of the DFT+DMFT approach with respect to the corresponding properties. Specifically, we completed our study of oxygen vacancies in the prototypical Mott insulator $LaTiO_3$, demonstrating that the electrons released by introducing a neutral vacancy into the system populate an emerging vacancy band that is energetically below the partially filled Ti d states. Consequently, the presence of (neutral) oxygen vacancies does not dope the Mott insulator, resulting in a surprisingly robust insulating character. A paper summarizing these findings has been published in *Physical Review B* [10].
For the case of $SrTiO_3$, where the Ti d states are completely empty in the stoichiometric case, a different picture emerges. Here, the empty d bands are energetically only slightly above the vacancy-related band, and thus electrons can be transferred into the d bands. Thereby, we can distinguish two different scenarios. Depending on the electron-electron interaction parameters used in the DFT+DMFT calculations, either both electrons released by the (neutral) vacancy fill the lower-lying vacancy band, or one of these electrons is transferred into the Ti d bands leading to partially filled bands and metallic behavior. The latter scenario is consistent with a model proposed by Lin and Demkov [15], where the first and second electron would correspond to "shallow" and "deep" donors, respectively. Our calculations further show that the preference for one or the other scenario is also strongly influenced by the Ti-Ti distance across the vacancy.
To obtain realistic estimates for the strength of the screened Coulomb interaction in the Ti d bands and also for the vacancy-related band, we have performed calculations using the constrained random phase approximation (cRPA). These calculations clearly show that the electron-electron interaction in the vacancy band should not be neglected and is approximately half of that corresponding to the Ti d states.

1.7 GW+DMFT

The main research effort of postdoctoral researcher Francesco Petocchi (Werner group) during the reporting period was devoted to the testing and further development of the GW+DMFT formalism for realistic materials. In particular, we performed a systematic comparison of 3-band and 5-band models for several transition metal perovskites ($SrXO_3$, $X = V, Mo, Mn$) [11]. In the DMFT community, these materials are usually treated as 3-band (t_{2g}) models. One goal of this study was to test if our parameter-free GW+DMFT scheme yields consistent results for the low-energy electronic structure in a t_{2g} and $t_{2g} + e_g$ description. On the methodological side, we extended the GW+DMFT code to two-sublattice magnetic order, which allowed us to study $SrMnO_3$ in the antiferromagnetic state. It was shown that antiferromagnetic order, or strong short-ranged antiferromagnetic correlations are needed to correctly reproduce the electronic structure of this material.
Additional GW+DMFT based materials investigations are under way (cuprates, nickelates, iron), some of which involve a real-space extension of GW+DMFT, i.e. an implementation which allows to treat strong correlation effects for several atoms in a unit cell.

1.8 Other projects

We have recently also published a paper still resulting from an experimental validation project of MARVEL phase I, together with the experimental group of Marta Rossell at Empa, on strain relaxation and cation distribution in ferroelectric Aurivillius phase thin films [12].

2 Collaborative and interdisciplinary components

A collaboration with Cesare Franchini (University of Vienna) on cRPA calculations of screened Coulomb interaction parameters has resulted in a joint publication [1].
Our work in the collaboration with the TRIQS project (triqs.github.io) has served as an important test case for the implementation of full charge self-consistency with the VASP/PLO interface.

Continued collaboration with former MARVEL members Oleg Peil and Antoine Georges has resulted in a joint publication [2].

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

- [1] A. Hampel, P. Liu, C. Franchini, and C. Ederer, *Energetics of the coupled electronic-structural transition in the rare-earth nickelates*, npj Quantum Materials **4**, 5 (2019).
- [2] O. E. Peil, A. Hampel, C. Ederer, and A. Georges, *Mechanism and control parameters of the coupled structural and metal-insulator transition in nickelates*, Physical Review B **99**, 245127 (2019).
- [3] A. Hampel, S. Beck, and C. Ederer, *Charge self-consistency and double-counting in DFT+DMFT calculations for complex transition metal oxides*, arXiv:1907.10339 (2019).
- [4] D. J. Gawryluk, Y. M. Klein, T. Shang, D. Scheptyakov, L. Keller, N. Casati, P. Lacorre, M. T. Fernández-Díaz, J. Rodríguez-Carvajal, and M. Medarde, *Distortion mode anomalies in bulk $PrNiO_3$: Illustrating the potential of symmetry-adapted distortion mode analysis for the study of phase transitions*, Physical Review B **100**, 205137 (2019).
- [5] S. Beck and C. Ederer, *Charge transfer in $LaVO_3/LaTiO_3$ multilayers: Strain-controlled dimensionality of interface metallicity between two Mott insulators*, Physical Review Materials **3**, 095001 (2019).
- [6] S. Beck, G. Sclauzero, U. Chopra, and C. Ederer, *Metal-insulator transition in $CaVO_3$ thin films: Interplay between epitaxial strain, dimensional confinement, and surface effects*, Physical Review B **97**, 075107 (2018).
- [7] D. E. McNally, X. Lu, J. Pelliciari, S. Beck, M. Dantz, M. Naamneh, T. Shang, M. Medarde, C. W. Schneider, V. N. Strocov, E. V. Pomjakushina, C. Ederer, M. Radovic, and T. Schmitt, *Electronic localization in $CaVO_3$ films via bandwidth control*, npj Quantum Materials **4**, 6 (2019).
- [8] F. Petocchi, S. Beck, C. Ederer, and P. Werner, *Hund excitations and the efficiency of Mott solar cells*, Physical Review B **100**, 075147 (2019).
- [9] C. Ricca, I. Timrov, M. Cococcioni, N. Marzari, and U. Aschauer, *Self-consistent site-dependent DFT+U study of stoichiometric and defective $SrMnO_3$*, Physical Review B **99**, 094102 (2019).
- [10] J. Souto-Casares, N. A. Spaldin, and C. Ederer, *DFT+DMFT study of oxygen vacancies in a Mott insulator*, Physical Review B **100**, 085146 (2019).
- [11] F. Petocchi, F. Nilsson, F. Aryasetiawan, and P. Werner, *Screening from e_g states and antiferromagnetic correlations in $d^{(1,2,3)}$ perovskites: A GW+EDMFT investigation*, arXiv:1911.09955 (2019).
- [12] M. Campanini, M. Trassin, C. Ederer, R. Erni, and M. D. Rossell, *Buried In-Plane Ferroelectric Domains in Fe-Doped Single-Crystalline Aurivillius Thin Films*, ACS Applied Electronic Materials **1**, 1019 (2019).

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[13] P. M. Woodward, D. E. Cox, E. Moshopoulou, A. W. Sleight, and S. Morimoto, *Structural studies of charge disproportionation and magnetic order in $CaFeO_3$*, Physical Review B **62**, 844 (2000).

[14] S. Y. Park, A. Kumar, and K. M. Rabe, *Charge-Order-Induced Ferroelectricity in $LaVO_3/SrVO_3$ Superlattices*, Physical Review Letters **118** (2017).

[15] C. Lin and A. A. Demkov, *Electron Correlation in Oxygen Vacancy in $SrTiO_3$*, Physical Review Letters **111**, 217601 (2013).

Design & Discovery 6 Search for Novel Topological Materials

Project leader: Oleg Yazyev (EPFL, 2MC)

Computational partners: Alexey Soluyanov (UZH, 1MC),
Experimental partner: Ming Shi (PSI, 0.5MC + 0.5MC from PSI), Hugo Dil (EPFL), Christian Rüegg (PSI and UniGE), Arnaud Magrez (EPFL)

1 Progress of the different efforts

The project is currently making steady progress towards the objectives defined in phase II proposal. The core activity is the discovery of novel topological phases and materials hosting them, including experimental realization. The new directions of research of our project include materials design, correlated topological phases and spin systems. Search for new topological phases is at the heart of the activity of project D&D6. The following achievements in this direction need to be reported.

A joint project with Tomáš Bzdušek at Stanford University resulted in the development of a new type of topological classification of metals. This classification shows that non-Abelian topological charges can be used to characterize nodal-line band degeneracies in metals with space-time inversion (PT) symmetry and weak spin-orbit coupling [1]. The non-Abelian charges put strict constraints not only on the possible nodal-line configurations, but also on their transformations (Fig. 1). Our analysis goes beyond the standard approach to band topology and implies the existence of one-dimensional topological phases not present in existing classifications. Current efforts in this direction aim at applying the introduced formalism to realize reciprocal braiding of Weyl nodes with ZrTe being the model material [2].

We have also achieved progress in understanding the Hopf insulator phase, a three-dimensional topological insulator phase that cannot be described in terms of the 10-fold classification. In particular, we investigated the possibility for the Hopf insulator to possess localized Wannier representation and the existence of its topological obstructions. We propose that Wannier functions are exponentially localized and preserve the symmetries of the system. Furthermore, we have shown that the surface states of the Hopf insulator phase can be gapped out by surface potential without violating the symmetry or closing the bulk gap. The surface states were shown to have a non-trivial first Chern number that equals to the bulk Hopf invariant [3]. Our study also gives hints regarding possible materials realizations of this elusive topological phase.

Further progress in extending the classification of topological band degeneracies to include composite Weyl fermions beyond the minimal chiral charge $\chi = \pm 1$ has been achieved. We developed a simple algorithm to identify and classify singular and critical points on Fermi surfaces. This approach allows to explore the classification of the band degeneracies according to the topology of the energy dispersion. Two new type-II band degeneracies with $\chi = \pm 2$ giving rise to exotic Lifshitz transition have been found. We have also started studying the physical consequences of these new types of band crossings, in particular their density of states and chiral Landau levels, as well as searching for the materials hosting these new composite Weyl fermion quasiparticles.

Recent discovery of the superconducting and correlated insulator phases in twisted bilayer graphene (TBG) at the so-called "magic" angle has created attractive opportunities for project D&D6 given the expertise of its participants. Indeed, the appearance of flat bands around the Fermi level in moiré heterostructures of weakly correlated two-dimensional materials opens a totally new way to study strongly correlated physics. At the same time, the twist degree of freedom creates a new paradigm for designing novel physical systems in a fully controlled manner. Finally, it becomes clear that topological arguments are at the heart of TBG and related twisted systems. This intersection of materials design, correlation and topology allowed Yazyev and Soluyanov to quickly enter the emerging field of twistronics and reinforce collaboration between their groups.

The group of Soluyanov primarily focused on the correlated aspects of TBG. A 12-band tight-binding model developed by Arkadiy Davydov provides an accurate description of the flat-band manifold and the neighboring remote

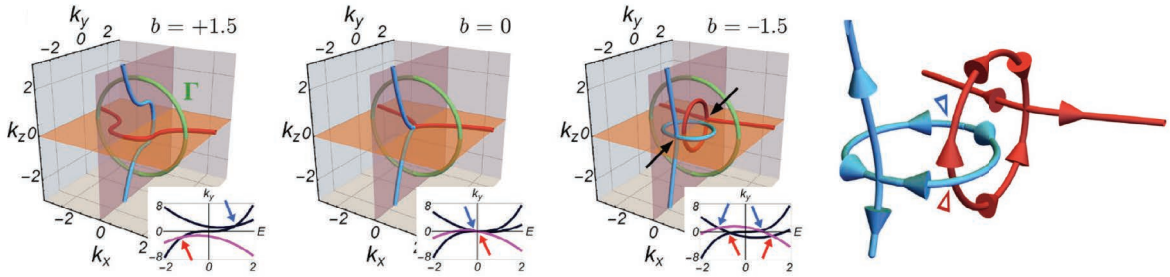

Figure 1: *Left: transformation of the nodal lines upon the change parameters in the three-band Hamiltonian governed by the quaternion topological charges. Right: details of the nodal-line linking (from [1]).*

bands in TBG in the non-interacting regime, thus providing a firm starting point for studying correlated physics in this system. First results on the fractional quantum Hall phase in this system using the DMRG techniques have been obtained. The group of Yazyev has been focusing on lattice relaxation effects, intrinsic crystal field and polarization effects as well as Hofstadter butterfly physics in TBG and related systems. In particular, the intrinsic polarization effect revealed in twisted *double* bilayer graphene (TDBG) allows accurate tight-binding description of this more complex moiré superlattice [4]. This progress has also created a new collaboration with the group of Guangyu Zhang (IOP CAS, China) performing experimental studies of TDBG. A work on probing the topological phase of the flat-band manifold in twisted multilayer graphene systems by means of magnetotransport measurements is approaching completion.

As far as spin systems are concerned, the search for novel quantum spin liquid (QSL) materials has been focusing on novel candidates realizing the Kitaev model. We identified two interesting candidate materials $Na_3Co_2SbO_6$ and $Na_3Co_2TeO_6$ that may realize the Kitaev model scenario in an unusual d^7 electronic configuration. Our configuration interaction calculations with embedding revealed that both the antiferromagnetic Heisenberg exchange interaction and the Kitaev interaction are significant, which implies these materials are likely to be magnetically ordered. Nevertheless, experimental realization of this two compounds and attempts to find structurally similar materials in this family has already begun (crystal growth facility led by Arnaud Magrez).

We then further extended our search to $4f$-electron systems. The recently synthesized $YbCl_3$ was identified as a candidate for hosting the Kitaev model with antiferromagnetic interactions, a model that was argued to undergo successive phase transitions from the low-field QSL connected to the topological QSL to another type of topological QSL. Our multi-reference wavefunction calculations mapped onto the generalized spin Hamiltonian yield a dominant Kitaev exchange interaction parameter that exceeds the ferromagnetic Heisenberg counterpart by a factor of 3. The Kitaev-to-Heisenberg ratio is similar to that in isostructural $RuCl_3$, but the magnitude of exchange couplings is an order of magnitude smaller, which highlights the importance of further exploration of magnetic phase diagram of $YbCl_3$ in magnetic field, as this might suppress the long-range magnetic order and lead to a QSL phase.

Using the multi-reference wavefunction calculations we have also addressed magnetic exchange interactions in the single-layer ferromagnet CrI_3 [5]. Our calculations identify ferromagnetic Heisenberg exchange parameter of $J = -1.44$ meV as the dominant term, while the other anisotropic couplings are one order of magnitude weaker. The reference results allow to assess the performance of common density-functional approximations, but also suggest that finding conditions (e.g. strain) at which J vanishes may emphasize the Kitaev model physics in this honeycomb lattice material as well.

The core experimental efforts of D&D6 led by Ming Shi (PSI) resulted in the following important results. Firstly, the ARPES investigation of WP_2, the robust Weyl semimetal discovered by us previously [7], has been concluded. Our ARPES measurements confirm the predicted Weyl semimetal phase and unveil the Weyl points that originate from the splitting of 4-fold degenerate band-crossing points with Chern numbers $C = \pm 2$ induced by the crystal symmetries. The manuscript reporting these results has been published in *Physical Review Letters* with the Editor's Suggestion distinction [8]. Second, we observed the Weyl semimetal state induced by spin fluctuation in the paramagnetic phase of $EuCd_2As_2$ [6].

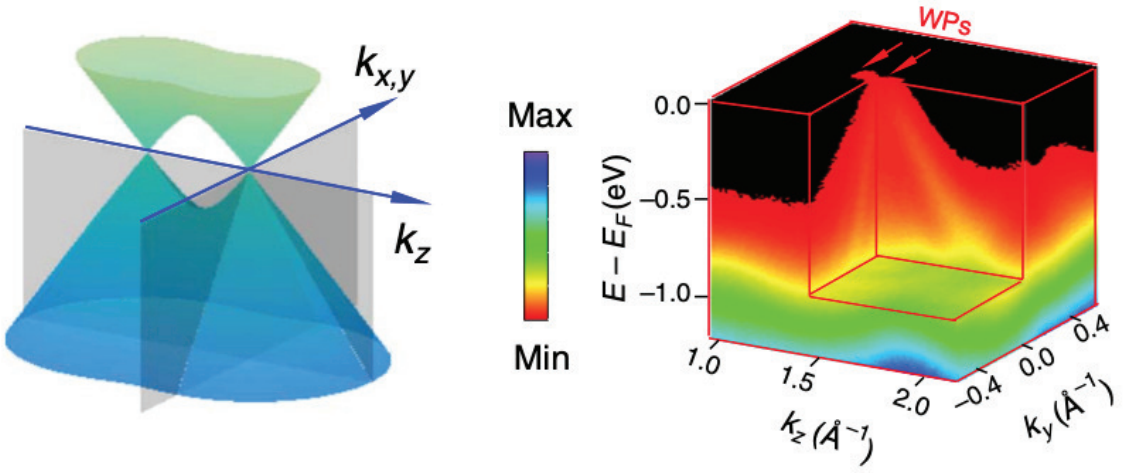

Figure 2: *Left: Schematic of the 3D Weyl cone band structure in the $k_y - k_z$ plane. Right: ARPES spectrum of $EuCd_2As_2$ showing two point-like Fermi surfaces along the $\Gamma - A$ line and the cone-shaped dispersions in the $k_y - k_z$ plane assigned to Weyl points (from [6]).*

We reveal that the degeneracy of Bloch bands is already lifted in the paramagnetic phase of $EuCd_2As_2$, and attribute this effect to the itinerant electrons experiencing quasi-static and quasi-long-range ferromagnetic fluctuations. The spin-nondegenerate band structure harbors a pair of ideal Weyl nodes near the Fermi level (Fig. 2). Hence, we show that the long-range magnetic order and the spontaneous breaking of time-reversal symmetry are not essential requirements for Weyl semimetal states in centrosymmetric systems.

Experimental work outside project D&D6 involved several groups, including the newly created collaborations. The continuing project with the group of Gabriel Aeppli (PSI) addressed the electronic structure of Fe_3Sn_2, a Kagome lattice ferromagnet in which the easy axis changes from out-of-plane to in-plane orientation with decreasing the temperature. A direct consequence of this effect is that the positions of topological band degeneracies in momentum space drastically change with temperature. In order to discuss the possible effect on the transport properties one needs an accurate description of the electronic structure. The ARPES measurements performed on this material reveal surprising complexity, that is now being rationalized with the help of DFT+U calculations [9].

Collaboration with the group of Marco Grioni (EPFL) addressed several topological materials by means of photoemission spectroscopy. One project focuses on trigonal Te, a material that, due to its screw symmetry, presents a new type of spin texture which is radial along the momentum direction. Another project addresses ZrSiTe, a material that hosts the nodal-line semimetal phase protected by nonsymmorphic symmetries. Even though the crystal structure of ZrSiTe preserves inversion symmetry, each ZrTe layer breaks inversion symmetry locally. The experimental data shows that the local inversion symmetry breaking in these layers results in the hidden spin polarization. Our calculations help interpreting the data, and also explain a new type of surface states, the so-called floating surface states, originating from the local breaking of the nonsymmorphic symmetry on the surface.

A related work addressed the magnetotransport properties of the isostructural nodal-line semimetal ZrSiS in collaboration with the group of Mario Novak at University of Zagreb. This material shows an unusual in-plane butterfly-shaped angular magnetoresistance (AMR) that was claimed to be related to a sharp topological phase transition. Our calculations performed using the previously developed methodology combining first-principles calculations with the Boltzmann semiclassical transport theory [10] reveal the mechanism underlying this peculiar shape of angular magnetoresistance [11]. We conclude that the dominant contribution to the cusp-like AMR is related to the open-orbit scenario in the hole pocket and, in general, AMR is strongly influenced by charge-carrier compensation effect and the off-diagonal conductivity tensor elements, which give rise to the peculiar butterfly-shaped AMR with no relation to any topological phase transition whatsoever.

2 Contribution to overall goals and initial proposal

Overall, project D&D6 is making steady progresses towards the main objectives of the NCCR related to the discovery of novel topological materials and method development activities. Some of the specific plans defined in the phase II full proposal, for instance related to the search for novel spin systems, proceed with no significant deviations. A research direction on graphene-based moiré superlattices has been initiated as this field provides interesting opportunities both as an emerging paradigm for materials design and a fertile ground for investigating correlated and topological phases. On the other hand, the initially planned research on device-scale simulations did not see much progress due to the definitive departure of Matthias Troyer.
Alexey Soluyanov has tragically passed away on October 26, 2019. The members of his group are now accommodated in the group of Titus Neupert (UZH), who will be involved in D&D6. This will require adapting some of the research plans, which is currently being discussed.

3 Collaborative and interdisciplinary components

The reported year was particularly fruitful in terms of establishing new collaborations with both theory and experimental groups outside MARVEL, at different institutions in Europe, USA and China. The project also profited from two horizontal cooperations with other D&D projects of MARVEL. A visiting Master student (Shiyu Deng) from the group of Nicola Spaldin has carried out a project in the group of Oleg Yazyev on the crystal structure search of novel bismuth halide topological materials (publication in preparation). Another spontaneous project with the group of Alfredo Pasquarello allowed acquiring new expertise [12].
As far as the interdisciplinary component is concerned, the project relies on numerous collaborations with experimental groups to complement its core theory and computational research. The own capabilities of experimental participants of D&D6 cover the majority of work related to materials synthesis and ARPES measurements, while other types of experimental activities are done by the collaborators outside MARVEL. The project also makes progress in introducing in its research novel methodologies such as machine learning [13].

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

- [1] Q. Wu, A. A. Soluyanov, and T. Bzdušek, *Non-Abelian band topology in noninteracting metals*, Science **365**, 1273 (2019).
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- [10] S. Zhang, Q. Wu, Y. Liu, and O. V. Yazyev, *Magnetoresistance from Fermi Surface Topology*, Physical Review B **99**, 035142 (2019).
- [11] M. Novak, S. N. Zhang, F. Orbanić, N. Biliškov, G. Eguchi, S. Paschen, A. Kimura, X. X. Wang, T. Osada, K. Uchida, M. Sato, Q. S. Wu, O. V. Yazyev, and I. Kokanović, *Highly anisotropic interlayer magnetoresistance in ZrSiS nodal-line Dirac semimetal*, Physical Review B **100**, 085137 (2019).
- [12] M. Pizzochero, F. Ambrosio, and A. Pasquarello, *Picture of the wet electron: a localized transient state in liquid water*, Chemical Science **10**, 7442 (2019).
- [13] R. Fournier, L. Wang, O. V. Yazyev, and Q. Wu, *An Artificial Neural Network Approach to the Analytic Continuation Problem*, arXiv:1810.00913, *to be published in* Physical Review Letters (2020).

Incubator 1

Design and Discovery of Novel Solid-State Ionic Conductors

Project leader: Teodoro Laino (IBM, 1MC)

Computational partners: Nicola Marzari (EPFL, 1MC)
Experimental partners: Daniele Pergolesi (PSI, 1MC + 1MC from PSI)

The development of all-solid-state rechargeable batteries is a fundamental step for next-generation energy storage solutions. The ultimate goal of this project is to find a solid-state material suitable for replacing the liquid electrolytes used in batteries today. This would bring enormous advantages in terms of packaging, capacity, and safety. Solid-state electrolyte materials may also be deposited as a thin film, thereby allowing the fabrication of miniaturized batteries for on-chip power sources and portable devices. A good solid-state electrolyte requires a high ionic and negligible electronic contribution to the total electrical conductivity. It should contain earth-abundant non-toxic elements, and offer suitable mechanical, thermal and electrochemical stability. To maximize the capacity of the battery the electrolyte should be stable in contact with Li metal. Meeting all of the requirements listed above is a daunting task.
Incubator 1 is a synergic project based on continuous feedback between theory and experiment to accelerate the discovery of new solid-state ionic conductors. A high-throughput computational screening of materials databases and the theoretical simulations of fundamental materials properties, such as ionic diffusivity and chemo-mechanical stability, are applied to select suitable candidates. Selected materials are then characterized experimentally, addressing the problem of their actual synthesis and measuring their ionic conductivity. Conventional ceramic processing routes are used to fabricate powder and sintered pellets. Materials characterizations include structure, morphology, composition and electrical properties. The experimental results are then fed back to the simulation for further refinement.

1 Status of the project

Experimentally, in 2019, the team focused on the synthesis and characterization of two materials, Li_7TaO_6 [1] and $Li_{10}GeP_2O_{12}$ [2], chosen among the candidates suggested by the computational screening realized during phase I of the project [3]. From a computational perspective, we developed new methodologies to evaluate the electrochemical stability windows (ESW) of solid-state electrolytes [4] and to screen partially delithiated structures in order to suggest materials whose diffusion properties can be improved by the creation of vacances through doping [5]. We also tested a methodology for computing mechanical and elastic properties at finite temperature. These developments were made available for immediate consumption by the MARVEL community through AiiDA workflows. Finally, novel training schemes were developed to construct models based on artificial neural network used to simulate solid-state electrolyte materials [6].

Metric #1: Design and discovery of novel materials displaying novel physics or improved properties or performance

Li_7TaO_6 (LTaO)

In 2018, the *PSI group* performed ionic conductivity and X-ray diffraction (XRD) spectra measurements of both doped and undoped Li_7TaO_6 (LTaO) materials, suggested by a computational screening performed at EPFL. In 2019, the new processing method of spark plasma sintering (SPS) was explored to significantly improve the relative density of the LTaO pellets and check its influence on Li-ion conductivity. The electronic conductivity of the LTaO pellets was also tested to ensure their insulating nature, which is a key requirement for solid-state electrolytes (SSEs). The corresponding conductivity value was 2×10^{-9} S cm^{-1}, which is on par with known SSE materials such as LLZO and LPS. With the addition of a new postdoctoral researcher to the PSI group in early 2019, the surface reactivity and chemical/electrochemical stability of LTaO were explored for the first time. X-ray photoelectron spectroscopy (XPS) experiments revealed lithium carbonate (Li_2CO_3) surface contamination on the LTaO pellets irrespective of polishing in an inert atmosphere (e.g. Ar-filled glove-

box). The exposure to ambient air is unavoidable due to the synthesis steps and pellet sintering procedures and is in good agreement with the DFT reaction energetics between Li_7TaO_6 and CO_2. In addition, XPS measurements elucidated the chemical reactivity of LTaO against metallic lithium, which showed no oxidation state changes for tantalum in the presence of lithium even after exposure to high temperature (80°C). From this result, metastability of LTaO against metallic lithium can be inferred. The electrochemical stability of LTaO was evaluated experimentally using a cyclic voltammetry (CV) protocol against a metallic lithium counter electrode over a wide voltage range (−0.5 V to 6 V vs Li/Li+), showing onset oxidation and reduction potentials for LTaO at 2.7 V and 0.7 V vs Li/Li+, respectively. These experimental results agree fairly well with the ESW evaluated computationally by the IBM group, ∼2.5 V and ∼0.3 V vs Li/Li+, as reported in Fig. 1. Due to the presence of Li_2CO_3 on the LTaO pellet surface, there is an extra oxidation peak around 3.8 − 3.9 V vs Li/Li+ that decreases in intensity with subsequent CV scans. Overall, LTaO was identified as a relatively stable material from the latest experimental results in 2019. The *EPFL group* performed long first-principles molecular dynamics (FPMD) simulations of LTaO. The analysis of the trajectories shows this material to be a fast ionic conductor with an activation barrier of diffusion below 0.3 eV, which is lower than that of the well-know LiPON [7] or cubic LLZO [8]. The *IBM group* used polarizable force fields (PFF) to refine the FPMD simulations performed at EPFL. Thanks to the know-how acquired simulating *W*-doped garnet LLZO [5], PFF simulations were able to provide longer trajectories for larger system sizes, albeit more approximate. Volume and temperature effects were found not to have a big impact on diffusion. The diffusion coefficient was found to be anisotropic in nature, with a factor of 2 between in-plane and out-of-plane diffusion. Finally, two types of grain boundaries were simulated, considering coincidence site lattice theory or pressing together grains created computationally. In both cases, the order of magnitude of the diffusion coefficient was found to be stable, confirming the experimental results from PSI. Computational investigations (IBM) also helped to interpret the ESW of LTaO, associating the shoulders appearing in the CV curves with decomposition reactions. In particular, the phase stability method, discussed in the next section, predicts the formation of Ta and Li_2O below ∼0.3 V,

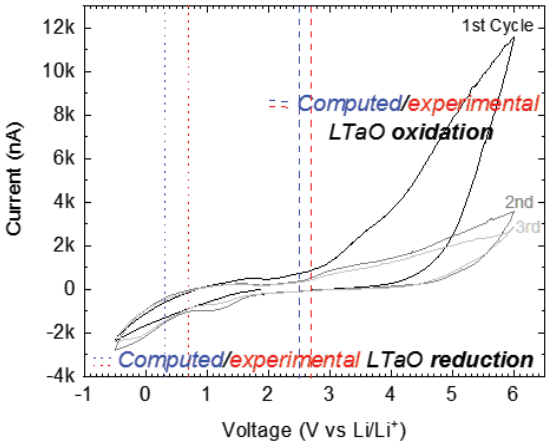

Figure 1: *CV curves of a Au-coated LTaO pellet with Li as the reference and counter electrode measured by the PSI group. The voltages computed by the IBM group for the redox reactions, limiting the electrochemical stability of LTaO, are reported as vertical blue lines and compared with the experimental ones.*

whereas Li_3TaO_4, O_2 and Li should form above ∼2.5 V.

$Li_{10}GeP_2O_{12}$ (LGPO)

Starting from the results from the last year we tested the effect of the morphology on the transport properties of LGPO. Thin films of LGPO were deposited by the *PSI group* via pulsed laser deposition at 500 and 25°C. XRD showed the formation of polycrystalline thin films. A difference in the peak intensities in the XRD of the samples deposited at 500°C and 25°C can be observed clearly. This indicates a difference in the microstructure, i.e. smaller grains or an amorphous phase in the latter. Conductivity of the samples was then tested through impedance spectroscopy; stoichiometry was also analysed by RBS (Rutherford backscattering) and ERDA (Elastic recoil detection analysis). It is noteworthy that LGPO can be experimentally deposited as thin film without sensibly altering the stoichiometry, which is not trivial for the deposition of lithium compounds [9, 10]. There is no significant effect of the microstructure on the conductivity for both films and pellet. Instead there is an effect on the activation energy (E_a) (0.51 eV for the sample deposited at 500°C and 0.58 eV for the sample deposited at 25°C). We calculated the charge distribution and diffusivity in LGPO (EPFL): the analysis revealed isotropic lithium diffusion in LGPO without a preferential direction. The E_a calculated by FPMD from the diffusivity (0.37 eV) is lower but comparable to the one obtained experimentally on thin films de-

posited at 500°C. The diffusivity and E_a simulated by FPMD can be considered to be representative of a defect free single crystal. Therefore, the deviation between the experimental and calculated electrical properties highlights the effect of defects on the lithium ion transport: increment in grain boundary density or amorphous phases in LGPO hinder the ionic transport. Nevertheless, these differences in the electrical properties are still small enough to allow the use of LGPO as thin film electrolytes, also when deposited at low temperature. The *EPFL group* performed extended FPMD simulations on LGPO solid-state electrolyte in different phases. A moderate conductivity ($\sigma_{RT} \simeq 10^{-5}$ S cm^{-1}, $E_a = 0.37$ eV) of LGPO in its orthorhombic phase *Pnma* [11] (here o-LGPO) was calculated through fixed-cell (NVT) FPMD simulations and compared with experimental results at PSI on the synthesized pellet and thin film (Fig. 2). Additionally, in MARVEL phase I a highly conductive ($\sigma_{RT} \simeq 10^{-3}$ S cm^{-1}, $E_a = 0.21$ eV) tetragonal phase (here t-LGPO), analogue to LGPS ($P4_2/nmc$ [12]) was predicted by the same group through NVT FPMD. This phase has so far not been described in literature, although evidence of oxosulphide structures of LGPS [13] has been found. With the goal of assessing its stability toward o-LGPO, variable-cell (NPT) FPMD simulations were conducted on the two structures at 600 K. Both phases were found to be stable, but t-LGPO with a significantly higher enthalpy (1 meV/Li-atom). From the time-dependent strain tensor, we calculated the elastic moduli for o-LGPO. This material is found to be harder than the corresponding sulphides, but with comparable ductility, which makes it appealing as SSE on the battery manufacturing side. In order to investigate the occurrence of an entropy-driven phase transition from o-LGPO to t-LGPO, we conducted NPT simulations at 1200 K. At this temperature the o-LGPO structure was found to undergo several phase transitions in a time range of 300 ps. Two new phases were found, one of which is pseudo-tetragonal, possessing a similar structure as t-LGPO up to length scales equal to 2/3 of the cell length. Although being highly conductive at 1200 K, cooling to 600 K showed a significantly higher activation barrier of these phases than the one of t-LGPO. NPT FPMD additional simulations are ongoing on larger commensurate supercells that would allow an o- to t-LGPO phase transition. The resulting theoretical XRD spectra will be compared with experiments of annealing and quenching of o-LGPO conducted at PSI.

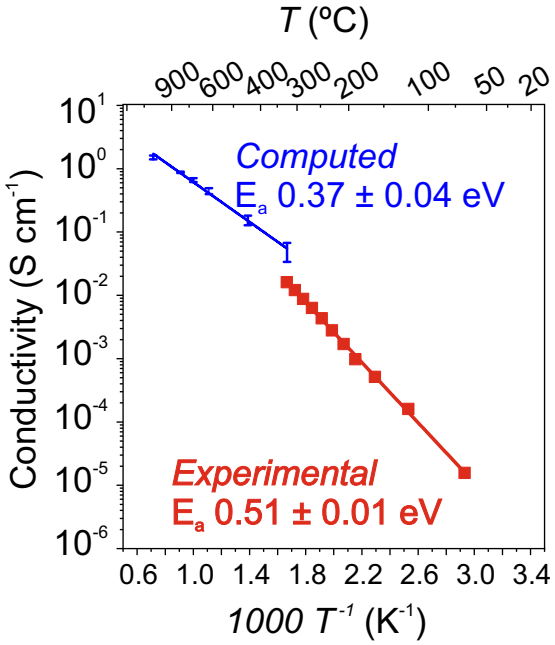

Figure 2: *Comparison between the conductivity dependence on the temperature measured on a thin film of LGPO deposited at 500°C and simulated by FPMD.*

Metric #2: Conceptual, methodological and algorithmic developments: Their dissemination and usage

Electrochemical stability (IBM)

The electrochemical stability window (ESW) of a SSE determines its stability in contact with candidate anodes/cathodes and is therefore directly related to the energy density of the entire battery. The team performed a systematic study of state-of-the-art computational methodologies to evaluate ESWs of SSEs, considering seven well known fast ionic conductors: LGPS, LIPON, LLZO, LLTO, LATP, LISICON and NASICON [4]. The *grand potential phase diagram method* considers all the possible decomposition products and is found to provide ESWs systematically narrower than the ones obtained by analyzing Lithium insertion/extraction reactions, which are considered in the *method of stable stoichiometry*. This result was found to be independent of the level of theory (PBE/HSE). For the stable stoichiometry method, the IBM group developed a methodology based on a Monte Carlo sampling of the electrostatic energy contributions to find low energy states for Lithium. Validation experiments show that, depending on the material and the setup, the measured ESW is related to one of the two instability processes covered either by the *grand potential phase diagram method* or the *method of stable stoichiometry*.

Therefore, the team suggested the use of the two methodologies in a complementary strategy.

Materials screening (EPFL)

After a large high-throughput screening for novel solid-state Li-ion conductors of all Li-containing insulators without partial occupancies and at full stoichiometry in the ICSD and COD structural repositories, as reported in previous reports, we assessed methods to screen structures that can be doped into becoming good ionic conductors. We performed a large screening of the Li-containing structures in the ICSD and COD during the reporting period, removing 20% of the lithium ions in the system to find materials that might be doped into high ionic conductivity through the introduction of Li vacancies. We investigated 772 structures with the pinball model [14] and are currently conducting FPMD simulations on the 26 most interesting compounds, out of which 12 were already suggested to the PSI group for synthesis. Another promising class of materials to screen for application as solid-state electrolytes are structures with partial occupancies. We are currently investigating how to create derivative supercells from structures that are reported with partial occupancies. Using cubic LLZO as a case study, the EPFL group is testing the electrostatic energy as a proxy to find low-energy configurations. This procedure originates from the synergy with the IBM group, which exploited the same method to estimate ESWs [4]. Our next step is to prepare a screening study of structures reported with partial occupancies at full and reduced Li-ion stoichiometry.

Neural networks for force-fields (IBM)

Recently, force-fields based on deep neural network (DNN) architectures have brought the promise of maintaining the level of accuracy of *ab initio* molecular dynamics simulations at a reduced computational cost. For the first time the IBM group tested the capability of the DeepMD model, a novel DNN-based force field, to predict diffusion in fast ionic conductors [6]. A novel training scheme was developed, based on a training set iteratively augmented until stationarity is achieved on the diffusion coefficient. The workflow developed was found necessary to avoid overfitting and the possibility of building poor generalizable models. As a bonus, the models can be trained starting with very short FPMD simulations. The training workflow was tested for three well-known SSEs: LGPS, LLZO and LLTO. The results compare very well with previous FPMD simulations from the EPFL group and show that the DeepMD model can indeed be used as a tool to discover novel solid-state electrolytes.

Statistical computation of elastic moduli (EPFL)

We computed the SSE mechanical properties in the elastic regime and derived the elastic moduli B, G, E and Poisson's ratio of the benchmark electrolytes LGPS and LGPO. This was done by calculating the matrix of the elastic compliance from the strain fluctuations, which we can access through our constant stress molecular dynamics simulations (NPT) [15]. This automatically includes the temperature effects beyond the harmonic approximation that fails for ionic conductors solid materials.

Metric #3: Open Science: codes, data, tools, and workflows/turnkey solutions

Suite for Analysis of Molecular Simulations — SAMOS (EPFL)

Our tools to analyze molecular dynamics simulations have been made public within a GitHub repository (github.com/lekah/samos). These tools allow an efficient and automatized analysis of trajectories with a user-friendly Python interface.

ZRL-AiiDA toolbox (IBM)

The workflows based on AiiDA plugins and used to evaluate the ESW under different approximations [4] have been made public under the following GitHub repository: github.com/zrl-aiida-toolbox/zrl-aiida-toolbox. The algorithms for fitting PFF force-fields have been uploaded to the same repository.

2 Report on milestones

M13: Milestone achieved. Candidate compounds have been proposed to the experimental partners and the feedback loop with the computational members of the project is active. Further PFF simulations to refine FPDM results have been performed. A second screening using the pinball model is active and ready to propose new candidate materials.
M19: Milestone achieved. AiiDA workflows have been developed and exploited to evaluate diffusion properties and electrochemical stability windows. Data and codes have been made public through the Materials Cloud platform and GitHub.

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

- [1] L. Kahle, A. Marcolongo, and N. Marzari, *High-throughput computational screening for solid-state Li-ion conductors*, Energy & Environmental Science (2020), doi:10.1039/c9ee02457c.
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- [13] K. Suzuki, M. Sakuma, S. Hori, T. Nakazawa, M. Nagao, M. Yonemura, M. Hirayama, and R. Kanno, *Synthesis, structure, and electrochemical properties of crystalline Li–P–S–O solid electrolytes: Novel lithium-conducting oxysulfides of $Li_{10}GeP_2S_{12}$ family*, Solid State Ionics **288**, 229 (2016).
- [14] L. Kahle, A. Marcolongo, and N. Marzari, *Modeling lithium-ion solid-state electrolytes with a pinball model*, Physical Review Materials **2**, 065405 (2018).
- [15] M. Parrinello and A. Rahman, *Strain fluctuations and elastic constants*, The Journal of Chemical Physics **76**, 2662 (1982).

Incubator 2

Active Machine Learning for Computational Materials Design

Project leader: Anatole von Lilienfeld (UniBas, 1.5MC)

Computational partners: Volker Roth (UniBas, 1MC), Michele Ceriotti (EPFL), Martin Jaggi (EPFL)

1 Progress of the different efforts

1.1 Progress of the von Lilienfeld group

Activities during the first part of MARVEL phase II in the von Lilienfeld group included several research projects.

a) *Revision of FCHL representation* In order to enhance the applicability and usability of our quantum-machine-learning models, we have mainly focused on a revision of the FCHL representation, published in 2018 [1] (FCHL18). While the use of FCHL18 within kernel ridge regression based machine-learning results in highly competitive predictive accuracy, its computational efficiency decays rapidly for larger training set sizes. We have alleviated the computational burden by using a discretized version of a Fourier series expansion fitted to the interatomic three-body terms in FCHL18. Fig. 1 illustrates this effect for two angles in the water molecule. Training timings for the resulting new FCHL model [2] (FCHL19) on training sets consisting of 1'000 different snapshots drawn from MD data for various organic molecules are also shown in Fig. 1. Comparison between FCHL19 and FCHL18 models indicates potential CPU time savings of one order of magnitude.
Due to its superior computational efficiency, more extensive hyper-parameter optimization runs of FCHL19-based machine-learning models are possible. This can result, for some datasets, in vastly superior predictive power. In the case of predicting the binding energy in clusters with sum formula, $(H_2O)_{40}$ for example, learning curves indicate that the advantageous hyper-parameters of FCHL19 enable predictions twice as accurate after training on ∼1'500 geometries (Fig. 2). Thanks to the Operator approach [3], energies and forces can now be efficiently modeled (crucial for molecular dynamics or geometry relaxation of larger systems), and are always among the top 3 methods in terms of predictive power for systems that have been studied in the literature (Fig. 2).

b) *Other efforts* Independently, we have also studied the application of machine-learning methods to the (i) modeling of X-ray powder diffraction, (ii) melting and boiling points of molecules, and (iii) energetics of conformational isomers. Unfortunately, the team member working on the first project decided to stop her PhD studies. Manuscripts reporting on projects (ii) and (iii) will be submitted soon.

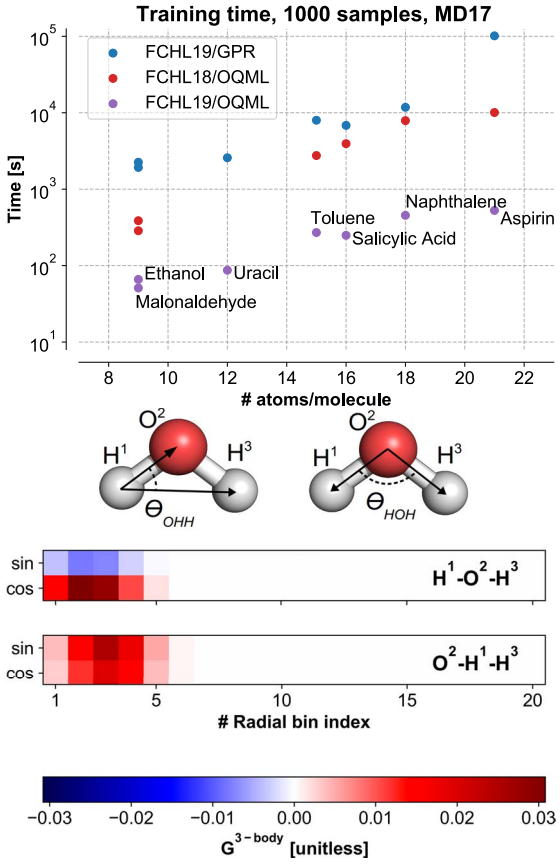


Figure 1: *Methodological illustrations for FCHL19 [2]. Top: training timings for kernels of rank 1'000 with three different methods for ethanol, malonaldehyde, uracil, toluene, salicylic acid, naphthalene, and aspirin. Timings are calculated as averages over 5 kernels using different random splits on a 24-core node equipped with two Intel Xeon E5-2680v3 @ 2.50 GHz CPUs. Bottom: the three-body basis functions are plotted for the two unique three-body terms in the water molecule, corresponding to the O^2-H^1-H^3 and H^1-O^2-H^3 angles displayed at the top. The atoms are numbered for clarity.*

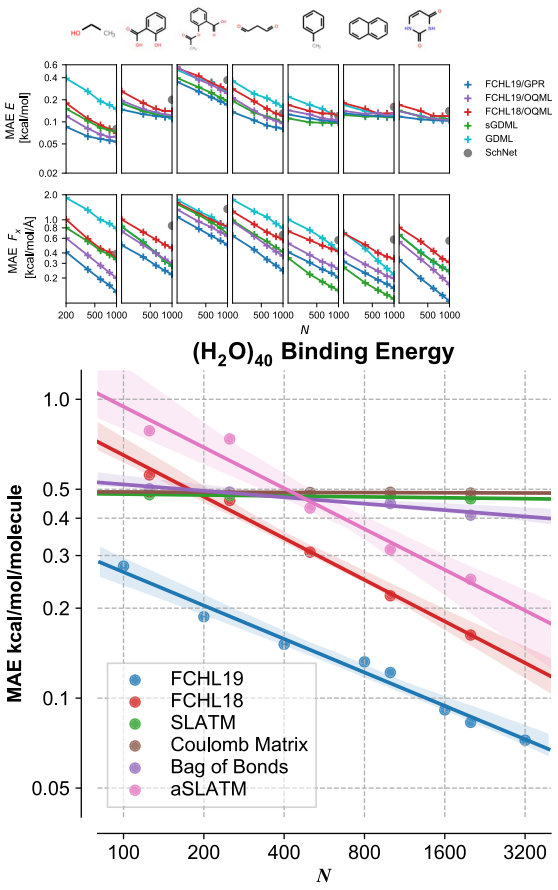


Figure 2: *Performance illustrations for FCHL19 [2] Top: learning curves for various machine-learning models predicting forces and energy for snapshots of 7 molecules from the MD17 dataset. The first and second rows refer respectively to energy (E) and force component (F) mean absolute errors (MAE) for out-of-sample predictions. Bottom: learning curves for the Water40 dataset: the mean absolute error (MAE) binding energy per molecule is plotted for 6 different representations versus the training set size. Linear fits are displayed for clarity, and shaded areas denote the 95% confidence intervals for the fits as obtained via boot-strapping.*

1.2 Progress of the Roth group

During the first year of MARVEL phase II, we worked on deep latent variable models to uncover structures in the chemical space.
In the first part, we developed a method with side information to represent molecules as convex combinations of extremal molecules [4, 5] with respect to certain properties.
In the second part, we designed an invariant subspace method to investigate the chemical space with respect to a set of desired properties. To do so, we created two disentangled latent spaces where one space encodes the property information, and the second space represents accounts for the residual variation of molecules having this property. For this purpose, we introduced a mutual information regularizer in combination with adversarial training. This model allows us to actively explore and propose novel molecules having predefined properties. On the more applied side, we trained our invariant subspace model to investigate if DFT simulations produce a systematic error for certain molecules. Therefore, we focused on a subset of QM9 with the same stoichiometry. We were able to infer a subspace that represents the structural variation of molecules conditioned on the predefined DFT error. Correlating this subspace with structural fingerprints of molecules, we then showed that high DFT error occur frequently for molecules with certain ring structures.

2 Contribution to overall goals and initial proposal

As specified in the phase II full proposal, the work of this incubator serves the further development and implementation of machine-learning models for the wider sake of the overall MARVEL effort. In order to facilitate usage by other MARVEL teams, we have open sourced our developments at github.com/qmlcode/qml. Manuals and documentation have also been made available at qmlcode.org.

3 Collaborative and interdisciplinary components

In order to support the organic crystal research in D&D1, we have assessed the performance of kernel ridge regression models based on the FCHL representation [1] for the modeling of non-covalent interactions. More specifically, corrections to the density functional SCAN for the prediction of van der Waals interactions in common benchmarks have been studied. This work has been submitted for publication [6].

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

[1] F. A. Faber, A. S. Christensen, B. Huang, and O. A. von Lilienfeld, *Alchemical and structural distribution based representation for universal quantum machine learning*, The Journal of Chemical Physics **148**, 241717 (2018).

[2] A. S. Christensen, L. A. Bratholm, F. A. Faber, and O. A. von Lilienfeld, *FCHL revisited: faster and more accurate quantum machine learning*, arXiv:1909.01946 (2019).

[3] A. S. Christensen, F. A. Faber, and O. A. von Lilienfeld, *Operators in quantum machine learning: Response properties in chemical space*, The Journal of Chemical Physics **150**, 064105 (2019).

[4] S. M. Keller, M. Samarin, M. Wieser, and V. Roth, *Deep Archetypal Analysis*, in *Pattern Recognition. DAGM GCPR 2019. Lecture Notes in Computer Science*, G. A. Fink, S. Frintrop, and X. Jiang, eds. (Springer, Cham, 2019), vol. 11824, p. 171.

[5] S. M. Keller, F. Arend Torres, M. Samarin, M. Wieser, and V. Roth, *Exploring Data Through Archetypal Representatives*, in *NeurIPS 2019 Learning Meaningful Representations of Life Workshop, to be published* (2020).

[6] P. D. Mezei and O. A. von Lilienfeld, *Non-covalent quantum machine learning corrections to density functionals*, arXiv:1903.09010 (2019).

Open Science — Open Science Platform

Project leader: Giovanni Pizzi (EPFL)

Computational partners: Nicola Marzari (EPFL), **Michel Kenzelmann** (PSI)

1 Progress of the different efforts

1.1 AiiDA

All efforts on AiiDA focused on improving stability, robustness and efficiency of the code. The result is version 1.0, released on October 31, 2019, including all developments of more than 1.5 years of work. The major changes since the latest stable 0.12.x releases are summarized below.

Speed and efficiency A new event-based workflow engine has been implemented, powered by RabbitMQ, providing almost instantaneous reaction to events. AiiDA can now support tens of thousands of processes per hour (Fig. 1). Moreover, significant efforts have been put on increasing query speed and reducing storage size of the AiiDA database, by switching to JSONB fields for storing node attributes and extras.

Improving stability AiiDA now deals seamlessly with remote calculation problems and network issues. AiiDA attempts again failed tasks with an exponential back-off policy and, if the problem persists, it "pauses" the process until user's intervention.

Standardization and homogeneity The code interface (`verdi` command line, Python API for calculations and workflows, etc.) has been greatly simplified and homogenized, providing a consistent interface to users across all codes and commands, together with powerful auto-documentation features.

Extended provenance model In AiiDA 1.0, a clear separation between data provenance (calculations, data) and logical provenance (workflows) has been introduced, giving more power to users in understanding and inspecting the provenance graphs.

Backward compatibility While the internal format for data storage has changed for efficiency reasons, data is automatically migrated without user intervention (even from very old AiiDA versions) via well-tested migrations that we implemented. Moreover, in AiiDA 1.0 we had to introduce some changes in the Python API to simplify its usage. To ensure a smooth transition, we provide extensive migration documentation and we have organized a 1-week event (March 2019) to support plugin developers in the migration.

Python 2 & 3 Support for Python 3 has been added (both in the core and in the plugins): AiiDA 1.0 is compatible with Python 2.7, 3.5, 3.6, 3.7. Since Python has discontinued support for the 2.x series from January 2020, future versions of AiiDA (from 1.1) will be compatible with Python 3 only.

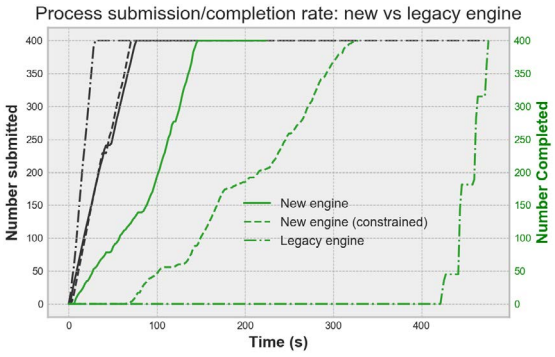


Figure 1: *Number of submitted (left axis) and completed (right axis) processes over time for the old and new engine. Submission of the old engine was slightly faster but, despite this, the completion rate of the new engine is clearly higher.*

1.2 Materials Cloud

a) *New entries in* Learn *and* Work *sections* A significant number of new entries has been added to Materials Cloud over the last year, in close collaboration with other MARVEL D&D projects. The *Learn* section has been populated with 21 new lecture videos (19 AiiDA tutorials with materials, 2 MARVEL distinguished lectures) and it has been reorganized in sections and subsections to simplify navigation. The *Work* section has been populated with additional research tools, including a Quantum ESPRESSO input generator, the AlphaML machine-learning (ML) tool to predict molecular polarizabilities, and the oxiMACHINE ML tool to predict the oxidation state for metal sites of a crystal structure. Moreover, the ShiftML tool has been upgraded to estimate

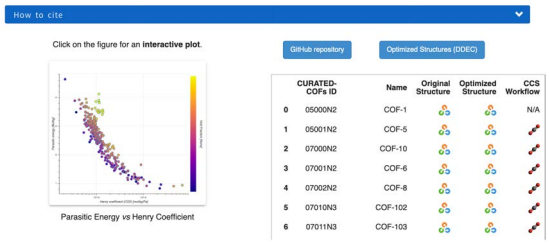


Figure 2: *An example of* Discover *section associated with the* Archive *entry "Building a consistent and reproducible database for adsorption evaluation in Covalent-Organic Frameworks". The interactive plot corresponds to a figure in the paper and connects every material with the history of the simulations to generate it.*

the uncertainty in predicting chemical shifts.

b) Discover, Explore *and* Archive *sections* An important component of Materials Cloud is its long-term archiving capability to store inputs and results of computational research, as required by SNSF for all publications. In the past year, over 70 new entries were contributed to the *Archive* section. Various new data entries were generated using AiiDA, and four of them are connected to corresponding *Explore* sections, allowing to interactively browse the data provenance. In particular, one entry associated with [1] contains not only the AiiDA database, but a fully reproducible virtual machine (based on the Quantum Mobile) that allows to reproduce all results of the paper, even with different numerical parameters. Moreover, eight new *Discover* sections originating from various MARVEL projects have been published, hosting curated datasets (see an example in Fig. 2).

c) *Technological advances* In addition to the visible changes, the Materials Cloud portal has evolved technologically. All machines have been migrated to the new NFS backend for storing OpenStack volumes, recently provided by CSCS, resulting in performance and stability improvements and automated data backups. A significant effort has been put to migrate the *Archive* section to the Invenio v3 framework (developed by CERN, the backend of Zenodo) that will provide users with a wide list of new features including user login with private workspace, search by keywords and free text, integrated submission and moderation workflows, record history tracking.

1.3 AiiDA lab and Quantum Mobile

The AiiDA lab service and all apps have been migrated to AiiDA 1.0. Moreover, AiiDA lab has seen a number of major improvements to make it ready to be adopted within a wider scientific community, along the following four main directions.

Scalability We reimplemented the deployment of the platform using Kubernetes, a *de facto* industry standard to orchestrate container-based systems allowing for automatic deployment, scaling, and management.

Stability We enabled automated testing of most of the service core features. To allow different apps with different requirements to coexist, we are in the process of defining and implementing apps version compatibility policies.

Ease of use We extended the AiiDA lab App Store to install and update apps (and switch between versions) with just one click via a tight and transparent integration with `git`. Additionally, we created a robust app that allows to run CP2K or Quantum ESPRESSO calculations (relaxation, equation of state, band structure) with just a few clicks (Fig. 3).

Ease of development AiiDA lab should not only provide a user-friendly interface to run simulations in the browser, but also an easy-to-use infrastructure for apps development. For this purpose, we provide a versatile set of Jupyter

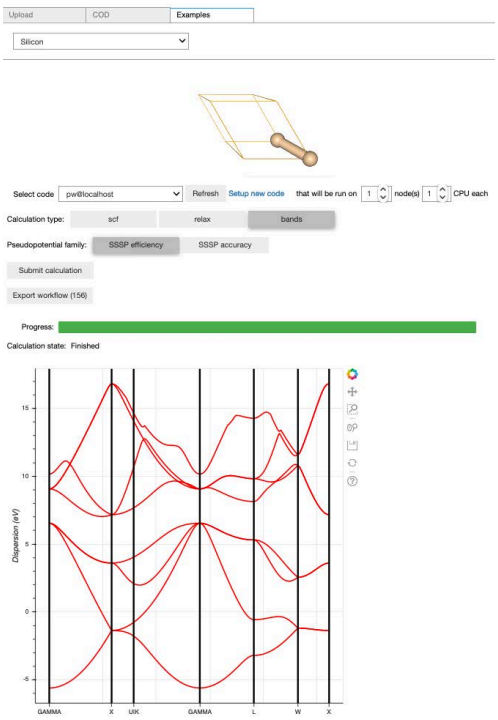


Figure 3: *A snapshot of band structure computed with* Quantum ESPRESSO *through the AiiDA lab app. The calculation requires almost no setup, since it is supervised by an turnkey workflow provided by the aiida-quantumespresso plugin developed within the MARVEL OSP.*

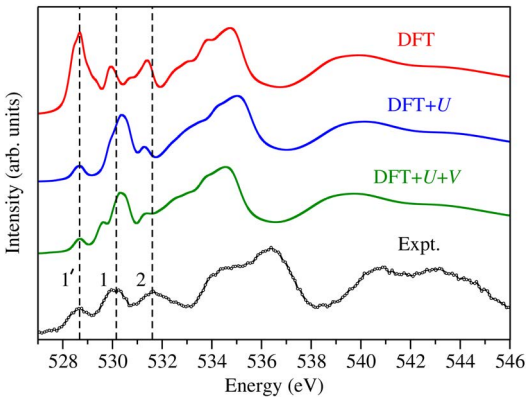


Figure 4: *Oxygen K edge XAS spectra of $LaFeO_3$ as computed using Fermi's golden rule on top of DFT, DFT+U, and DFT+U+V, and as measured in the experiment.*

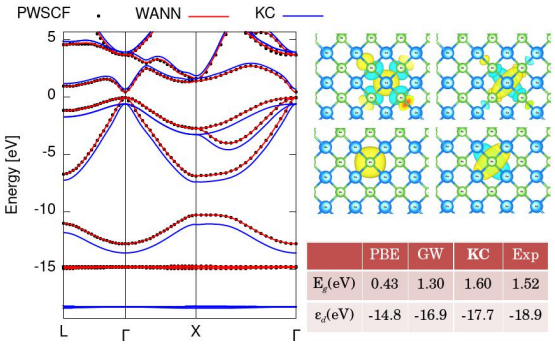


Figure 5: *Band structure of GaAs. The band Gap (E_g) and the position of the d bands (ε_d) relative to the top of the valence band are in good agreement with experimental results.*

widgets[1] that allows to create fully operational apps with only a few lines of Python code.
In addition to this, the Quantum Mobile virtual machine is continuously upgraded, with a new release every two months, and is serving as the basis for many courses at universities and a number of Schools and tutorials.

1.4 Computational spectroscopies and microscopies and curated data

a) *XAS at the K edge* Most of the work focused on the methodological development, implementation, and application of the XAS method for modeling *K* edge spectra of transition metal oxides. We interfaced the XSpectra code of Quantum ESPRESSO with the Hubbard-corrected DFT scheme by adding on-site *U* and inter-site *V* Hubbard corrections. The Hubbard parameters are computed from first principles using DFPT, using our public implementation in Quantum ESPRESSO [2]. E.g., O *K* edge spectrum of $LaFeO_3$ is in very good agreement with the experimental spectrum, when both on-site *U* and inter-site *V* corrections are taken into account [3] (Fig. 4).

b) *Koopmans-compliant spectral functionals for photoemission spectroscopies* During the last year a significant effort has been devoted to a re-structuring of the Koopmans-compliant (KC) code. This was primarily to have an implementation compatible with the *k*-point sampling of the Brillouin zone and therefore capable to exploit the Bloch symmetry in periodic crystals. A preliminary version of the code which uses Wannier functions as a reasonable guess for the true functional minimum is now available in a private repository. The band structure of a prototypical semiconductors, i.e. GaAs, is shown in Fig. 5, together with the *s*-like and *p*-like Wannier functions for the valence and conduction manifolds and a table with a comparison with GW and experimental results.

c) *XAS and RIXS at the $L_{2,3}$ edge* We are currently exploring possible strategies to simulate X-ray absorption and resonant X-ray scattering at the $L_{2,3}$ edge of transition metal ions. This is done fully *ab initio* at the GW+BSE level with the OCEAN interface or by modeling the crystal field on the emitting atom at the DFT level and importing it into an atomic multiplet calculator.

2 Contribution to overall goals and initial proposal

2.1 AiiDA, Materials Cloud and AiiDA lab

AiiDA serves as the "operating system" providing the infrastructure to run high-throughput simulations and turnkey simulation workflows, orchestrating multiple simulation softwares. Over the last year the high-throughput capabilities were greatly improved and extensively tested. With the release of v1.0, the code has a stable and robust interface. Moreover, support of about 40 simulation packages (and over 80 simulation codes) is now available[2] thanks to the combined support of MARVEL, a number of partner projects, and individual contributors.
Materials Cloud continues to serve as the dissemination platform to share educational ma-

[1] AiiDA widgets for AiiDA lab applications, github.com/aiidalab/aiidalab-widgets-base

[2] See the full list of plugins in the AiiDA plugin registry: aiidateam.github.io/aiida-registry.

terial, scientific tools and research results. It is being continuously enriched by contributions of data and tools by many different MARVEL D&D projects.
AiiDA lab, as an integral part of Materials Cloud, provides access to run AiiDA turnkey workflows and to analyze the results via simple web interfaces. AiiDA lab also greatly simplifies the effort needed to build new applications and to share them with the community.

2.2 Computational spectroscopies and microscopies and curated data

a) *Vibrational spectroscopies.* The long-term goal is to provide turnkey solutions that cover vibrational spectroscopies, such as phonons with Hubbard corrections. To this aim, we publicly released the DFPT+U code via Quantum ESPRESSO for modeling phonons with the Hubbard U correction, and demonstrated its strength by applying it to CoO and $LiCoO_2$ [4]. This implementation can be used directly with AiiDA.

b) *Photoemission spectroscopies.* KC functionals emerge as a reliable and efficient alternative to Green's function techniques for the simulation of spectral properties [5, 6]. Having a modular KC code fully compatible with the Quantum ESPRESSO package and Wannier90 code would make it easily accessible to the scientific community and particularly suitable for an AiiDA workflow implementation.

3 Collaborative and interdisciplinary components

3.1 AiiDA, Materials Cloud and AiiDA lab

AiiDA has evolved as a truly collaborational project over the past few years. We highlight fruitful and continuous collaborations with Dominik Gresch (Microsoft), Espen Flage-Larsen (SINTEF), Tiziano Müller (UZH, MARVEL), Chris Sewell (Imperial College London) who contributed to the code and participated in many code design discussions.
The Materials Cloud *Archive* now is part of a broader community of data repositories. Besides being a recommended repository for Materials Science by the Nature's journal *Scientific Data*[3], since June 2019 it supports the Open Archive Protocol for Metadata Harvesting (OAI-PMH). It has also joined the B2FIND international network and it is now indexed by Google Dataset Search. Moreover, tight collaboration with CSCS (where the web services are hosted) allows for continuous improvements and co-design of the deployment and service technologies.
AiiDA lab is evolving as a tight collaboration with Empa teams (Passerone, Fasel) who contribute in the design of the service, provide user feedback and contribute with apps. Moreover, we have setup additional AiiDA lab instances on the Kubernetes clusters provided by CSCS, and the CESNET center (Czech Republic), also integrating with external authentication services (CSCS Keycloak, EGI check-in).

[3] Materials Cloud recommended by *Scientific Data*: www.nature.com/sdata/policies/repositories#materials

3.2 Computational spectroscopies and microscopies and curated data

Close collaboration between the EPFL team (groups of Marzari and Pizzi) and Empa team (group of Passerone) was crucial to develop and apply a new approach for modeling XAS at the O K edge of transition metal oxides including Hubbard corrections.
We are currently providing support to the EPFL Laboratory of Ultrafast Spectroscopy of Prof. Chergui for the simulation of ARPES spectra of lead halide perovskites [7].

MARVEL publications

List of publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR.

- [1] V. Vitale, G. Pizzi, A. Marrazzo, J. Yates, N. Marzari, and A. A. Mostofi, *Automated high-throughput Wannierisation*, arXiv:1909.00433 (2019).
- [2] I. Timrov, N. Marzari, and M. Cococcioni, *Hubbard parameters from density-functional perturbation theory*, Physical Review B **98**, 085127 (2018).
- [3] I. Timrov, P. Agrawal, X. Zhang, S. E. R. Liu, A. Braun, M. Cococcioni, M. Calandra, N. Marzari, and D. Passerone, *X-ray absorption near-edge structure of pure and homovalent No-substituted $LaFeO_3$: First-principles simulations and experiments*, (in preparation) (2020).
- [4] A. Floris, I. Timrov, B. Himmetoglu, N. Marzari, S. de Gironcoli, and M. Cococcioni, *Hubbard-corrected density functional perturbation theory with ultrasoft pseudopotentials*, arXiv:1910.06195, *to be published in* Physical Review B (2020).
- [5] N. Colonna, N. L. Nguyen, A. Ferretti, and N. Marzari, *Koopmans-Compliant Functionals and Potentials and Their Application to the GW100 Test Set*, Journal of Chemical Theory and Computation **15**, 1905 (2019).
- [6] J. D. Elliott, N. Colonna, M. Marsili, N. Marzari, and P. Umari, *Koopmans Meets Bethe-Salpeter: Excitonic Optical Spectra without GW*, Journal of Chemical Theory and Computation **15**, 3710 (2019).
- [7] M. Puppin, S. Polishchuk, N. Colonna, A. Crepaldi, D. N. Dirin, O. Nazarenko, R. De Gennaro, G. Gatti, S. Roth, T. Barillot, L. Poletto, R. P. Xian, L. Rettig, M. Wolf, R. Ernstorfer, M. V. Kovalenko, N. Marzari, M. Grioni, and M. Chergui, *Evidence of large polarons in photoemission band mapping of the perovskite semiconductor $CsPbBr_3$*, arXiv:1909.00248 (2019).

HPC

HPC and Future Architectures

Project leaders: Joost VandeVondele (CSCS and ETHZ), **Thomas Schulthess** (CSCS and ETHZ)

1 Progress of the different efforts

HPC and Future Architectures platform is involved in two major activities: (i) hardware provisioning and support for MARVEL, located at CSCS and (ii) software development of electronic-structure library SIRIUS for the GPU acceleration of Quantum ESPRESSO code.

1.1 Hardware resources

CSCS is responsible for the hardware infrastructure of MARVEL as well as provisioning core services, such as computing, storage and identity management for its several research groups. The MARVEL project owns 180 dual-socket compute nodes (2x Intel Broadwell 18-core, E5-2695 v4 @ 2.10 GHz), with 64 GBytes of memory (DDR4 @ 2133 MHz) which are clustered to the multi-core partition of Piz Daint and during 2019 were fully utilized. MARVEL compute nodes have full access to the scratch filesystem of 2.7 PetaBytes. Besides that, 150 TB of permanent storage is available exclusively for MARVEL, which is key for the *Archive* section of Materials Cloud.
The Materials Cloud infrastructure (web frontend, AiiDA backend and storage) is hosted on the OpenStack instance at CSCS. Around 250 cores, 850 GB RAM and 50 TB of storage are assigned for this service. CSCS invest significantly with its own resources, providing expertise and support to MARVEL, as MARVEL is considered a strategic partner in building a converged cloud-HPC infrastructure, where user communities build and grow their domain-specific platforms.

1.2 SIRIUS library

SIRIUS is a domain-specific library with GPU backend designed to accelerate plane-wave electronic structure codes such as Quantum ESPRESSO and Exciting, or to be used as a Quantum Engine in programs using localized basis sets. The development of SIRIUS is supported by CSCS as part of its strategic effort to separate the concerns between domain and computational scientists in the today's complex software development processes. In the context of MARVEL project, SIRIUS is used to accelerate the major workflows of materials simulations — ground states, lattice relaxations and atomic forces — performed by the Quantum ESPRESSO code (QE, Marzari), and has integrated a second important MARVEL code, namely CP2K (Hutter). CSCS officially supports the patched version of QE with SIRIUS bindings, which is now available to all users (see the link).
In year 6, the following developments have been accomplished:

- atomic forces, stress tensor and total energies for magnetic collinear and non-collinear ground states;
- spin-orbit coupling;
- Python bindings to major C++ classes of SIRIUS, allowing binding to and integration in i-PI (Ceriotti);
- low level optimizations and performance tuning, reduction of GPU memory consumption (using the concept of memory pool) and code refactoring;
- initial ROCm port for AMD GPU cards;
- prototype code for robust wavefunction optimization.

An important feature that was added to SIRIUS is a set of Python bindings (Fig. 1 for example). This allows to create interactive Python-based programs for demonstrational, educational and prototyping purposes. Using the Python bindings, several flavors of robust wavefunction optimizers were developed as a prototype.

1.3 Robust wavefunction optimization

The self-consistent iterative algorithms commonly used in DFT calculations, based, e.g., on charge-density or potential mixing, are not guaranteed to converge. The success rate of the iterative approach, observed by the Marzari group for a single-point calculation, is of the order of 90% and, for a large-scale geometry optimization of magnetic systems, is of the order of 80%. Direct minimization algorithms offer

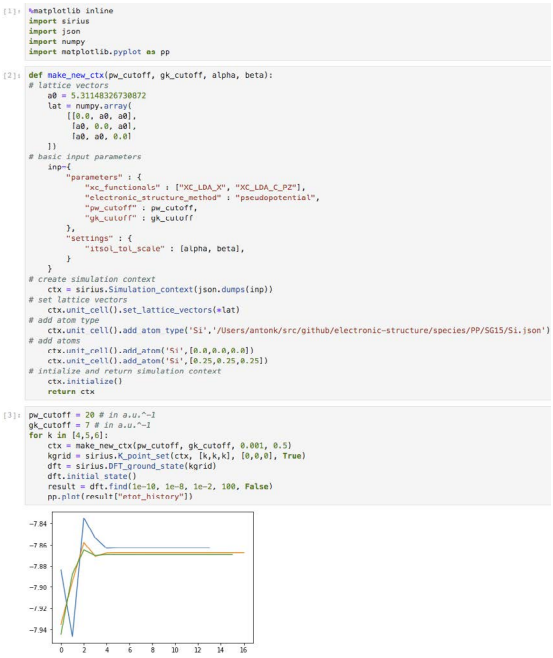


Figure 1: *Jupyter notebook running ground state of silicon with SIRIUS. In the present example three k-meshes are tested for total energy convergence.*

a robust fallback method, since if properly implemented provide an algorithm that is bound to converge (ensemble DFT, Marzari, Vanderbilt and Payne [1]). We have thus implemented such nonlinear conjugate gradient methods for metallic and non-metallic systems using the SIRIUS Python API. For insulators, we have implemented the orbital-transformation method (VandeVondele [2]); for metals, ensemble DFT, as referenced above, where one has to deal with the minimization over wavefunctions, occupation numbers, and unitary rotations (the last two combined in a non-diagonal Fermi-Dirac occupation matrix). We have implemented these methods and benchmarked them rigorously (Fig. 2). The methods differ with respect to how the orthogonality constraint of the wavefunctions are taken into account, for example by projection, or by formulations on the Stiefel manifold (Baarman [3]), and how the search over the occupation numbers is performed under the constraint that a single orbital has an occupation number between 0 and 1 and the total number of electrons in constant. The original Marzari-Vanderbilt-Payne method splits the optimization in an outer loop (wavefunction optimization) and an inner loop optimizing the occupation matrix while wavefunctions are kept fixed. A variant of this employs a pseudo-Hamiltonian to parametrize the occupation numbers. This approach allows to form a single gradient with respect to the wavefunctions and the occupation numbers, with the drawback that it performs progressively worse at small smearings, but with the advantage that it can exhibit faster convergence

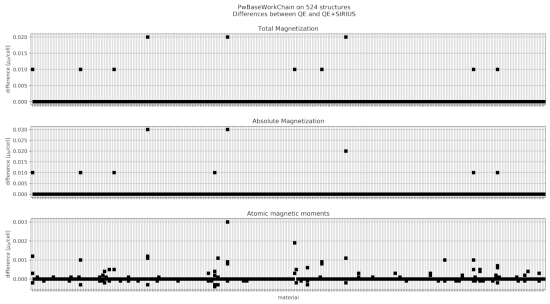
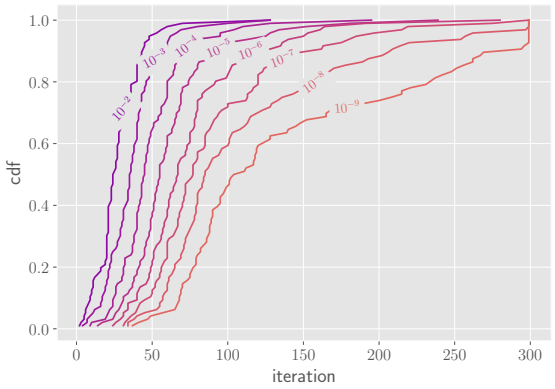


Figure 2: *Comparison of cell magnetization and atomic moments between native and SIRIUS-enabled QE calculations performed for 524 magnetic structures. Small difference in results originates from the slightly different numerical implementation schemes.*

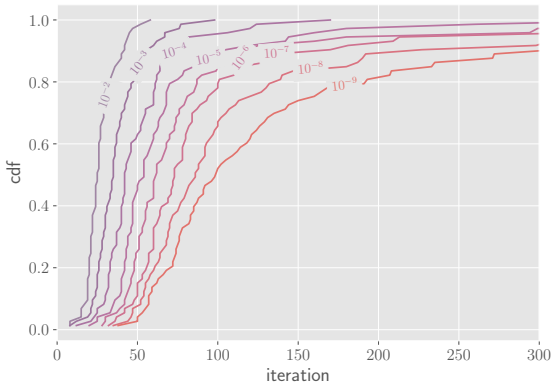
Our test set consists of 100 magnetic structures which failed to converge with the standard SCF protocols. Using ensemble DFT together with kinetic energy preconditioning, we are able to converge 100% of these structure to 10^{-7} residuals, and 90% of these structures to 10^{-9} residuals, within less than 300 iterations leading to an expected success rate of 100% and 98% respectively (Fig. 3). In order to manage the benchmark calculations we have setup a SIRIUS plugin for AiiDA. Currently we are working on a separate library written in C++ for robust optimization that will work with SIRIUS, Quantum ESPRESSO and CP2K.

1.4 Python API

The Python API is built on pybind11 to expose a large fraction of the functionality of the SIRIUS C++ classes. It allows both coarse and fine-grained access to SIRIUS components. As an example, for coarse-grained access, input parameters can be defined in a Python dictionary and then be passed on to the DFT ground-state object of SIRIUS to run the SCF cycle. Afterwards, the results can be queried from Python: for example, the atomic forces or the total energy. A practical use case for the coarse-grained interface is the integration of SIRIUS into multiple high-level engines, such as i-PI, a Python code for path-integral molecular dynamics. The Python interface allowed us to propose a future replacement for the i-PI socket interface based on an MPI pool, working with MPI sub-communicators using the mpi4py package. The socket interface, which relies on port forwarding and polling of re-



(a) *Gaussian-spline smearing* $T = 0.14$



(b) *Fermi-Dirac smearing* $T = 300K$

Figure 3: *Number of calculations converged to a given residual for a given number of CG iterations. We can converge* 100% *of the structures to* 10^{-7} *residual.* 90% *of all calculations converge within at most* 300 *iterations to* 10^{-9} *accuracy. The results were obtained for the PBE GGA functional and SG15 norm-conserving Vanderbilt pseudo-potentials.*

sults, requires that the DFT client codes implement their specific protocol and does not scale well on HPC systems. On the other hand, the MPI pool approach does not suffer from such problems, but requires Python bindings of the client code. SIRIUS provides both Python bindings, and, thanks to its modular design, a SIRIUS instance can be created based on a mpi4py MPI (sub)-communicator created in the Python domain. Similarly, such Python interface would facilitate for example the implementation of frozen-density embeddings or as demonstrated in a student semester project the exploration of wavefunction extrapolation for *ab initio* molecular dynamics. The fine-grained Python API exposes the internal data structures of SIRIUS, such as the plane-wave coefficients of the wavefunctions, using the buffer protocol to interface with NumPy, and allows composition of operators. On the Python side the application code can then be written in a NumPy compatible way, which

$$\frac{\partial L}{\partial \eta_{nn'}} = -\frac{\delta_{nn'}}{kT}(\langle\psi_n|\hat{H}|\psi_n\rangle - \epsilon_n)f_n(1-f_n) + \delta_{nn'}\frac{\partial \mu}{\partial \eta_{nn'}}\frac{\partial A}{\partial \mu} + \frac{f_{n'}-f_n}{\epsilon_{n'}-\epsilon_n}\langle\psi_n|\hat{H}|\psi_{n'}\rangle(1-\delta_{nn'})$$

```
def grad_eta(Hij, ek, fn, T, kw):
    g_eta_1 = -1/kT * diag(diag(Hij) - kw*ek) * fn * (1-fn)
    dFdmu = np.sum(np.real(
    1/kT * einsum('i,i',
                  (diag(Hij) - kw*ek).asarray().flatten(),
                  fn * (1-fn))))
    sumfn = np.sum(kw*fn*(1-fn))
    g_eta_2 = diag(kw * fn * (1-fn) / sumfn * dFdmu)
    II = diag(ca.ones_like(fn))
    Eij = ek-ek.T + II
    Fij = fn-fn.T

    g_eta_3 = Fij / Eij * Hij * (1-II)
    return (g_eta_1 + g_eta_2 + g_eta_3)
```

Figure 4: *Expression for the gradient of the Lagrangian with respect to the pseudo-Hamiltonian as formula (top) and in Python (bottom). The dependency on the k-points and the spin-channels is omitted for the sake of simplicity; the SIRIUS Python interface handles it implicitly.*

greatly facilitates rapid prototyping of new algorithms. An example how directly complicated mathematical expression can be implemented in our Python API is depicted in Fig. 4. Computations implemented in the NumPy-like API will be carried out in NumPy (e.g. on CPU), while operators (Hamiltonian, overlap matrix) will be executed in C++ SIRIUS, and can thus be accelerated on GPU. This provides the optimal balance between performance and flexibility for rapid prototyping.
The implementation of nonlinear conjugate gradient algorithms is simplified by our array class wrapping wavefunctions, occupation numbers, etc. which are in general depending on spin-channels and k-point index into a single object.

- Plane-wave coefficients of the wavefunctions $|\psi_i\rangle$ and the occupation numbers f_n are treated as vector spaces and wrapped in the `Array` class.
- Arithmetic operators `+,-,/,*,@` are applied separately (threaded) for each k-point and spin-channel of the underlying object.
- Reductions happen implicitly, for example `np.sum(fn * (1-fn))` will trigger a reduction over all spin-channels, k-points and call MPI if needed.

- Overloading for NumPy functions `diag`, `transpose`, `einsum`, etc. and matrix decompositions (QR, SVD, etc.)

- User defined functions can be decorated using the `threaded` decorator, in order to apply operations over *k*-points/spin-channels.

2 Contribution to overall goals and initial proposal

The HPC platform is contributing significantly to the overall goals of the NCCR in various ways. First, the hardware and services provided by CSCS are essential to run the Materials Cloud platform, in particular the OpenStack service as well as the long term storage. In this way the HPC platform is key for the open science objectives. Second, the compute services provided form a foundation for much of the computational work in MARVEL, and contribute to the discovery of new materials aspect of the NCCR. Third, the software developments presented contribute to the NCCR in multiple ways. This includes: (i) increased efficiency in the usage of resources, enabling 2× performance per CHF and/or per Watt, (ii) future-proofing of the basic computational tools by providing re-architected, re-factored, GPU-accelerated software, (iii) addition of new features, such as robustness, that will greatly help the high-throughput workflows of MARVEL users, and (iv) establishing for and disseminating this computational approach to a wider audience.

3 Collaborative and interdisciplinary components

The collaboration between CSCS and several groups of MARVEL is going strong. First, with the Materials Cloud team, building the platform, with regular technical meetings. Second, for the software effort, we have been very actively collaborating with multiple MARVEL PIs. This includes the group of Marzari — who has formulated key requirements for SIRIUS, and did early testing and verification, and provided feedback and testcases, the group of Hutter — who has provided guidance and help for the integration of SIRIUS in CP2K, the group of Ceriotti — who has provided guidance and help for the integration of SIRIUS in i-PI, and the group of Goedecker — who has used SIRIUS-enabled Exciting for benchmarking.
Furthermore, MARVEL has lead to additional collaborative projects related to SIRIUS in the context of various other projects, including the Platform for Advanced Scientific Computing (PASC), and the H2020 MaX Centre of Excellence.

Other references

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[2] J. VandeVondele and J. Hutter, *An Efficient Orbital Transformation Method for Electronic Structure Calculations*, The Journal of Physical Chemistry **118**, 4365 (2003).

[3] K. Baarman, V. Havu, and T. Eirola, *Direct Minimization for Ensemble Electronic Structure Calculations*, Journal of Scientific Computing **66**, 1218 (2016).

4.2 New projects

In year 6, two PIs were added to the OSP platform led by Giovanni Pizzi to implement the part of the project on "computational spectroscopies and microscopies and curated data". Michel Kenzelmann (PSI) and Nicola Marzari (EPFL) thus join Giovanni Pizzi as PIs in this project and work in close collaboration. More details on their research is given in the report on OSP.
As already mentioned in the Management chapter, some other changes also occurred in year 6.

- Claire Villevieille left PSI in February 2019 to become a senior research scientist at the Université Grenoble Alpes. The MARVEL effort in her group continued throughout the year, with the postdoc supported by her funding managed by Daniele Pergolesi, PI in the same Inc1 project.
- Piero Macchi left UniBE in August 2019. Arrangements were made to allow him using his year 6 funding and conclude his contribution to the project.
- Kyriakos Stylianou left EPFL in August 2019. His research effort and budget are taken over by Christopher Ireland, also part of Berend Smit's group.
- Following the passing of Alexey Soluyanov in October 2019, his MARVEL budget is maintained. Prof. Titus Neupert, recently arrived also at UZH, and an expert in topological insulators, joins as new MARVEL PI and continues the collaboration within the D&D6 project.

As discussed in the "Equal opportunities, analysis and plans" document, a call for new Agility Plus funding, targeted at junior women PIs, has now been completed and is under evaluation, with four new PIs starting in year 7.

5 Knowledge and technology transfer

MARVEL's knowledge and technology transfer activities are broad and varied. We transfer knowledge through our dissemination of open-source simulation codes, training in their use as well as, critically, through the Materials Cloud platform. Offering a range of tools, services and data, this materials informatics framework is built to enable the seamless sharing and dissemination of resources in computational materials science and is central to MARVEL's work. Our technology transfer activities this year have focused on the organization of Industry Sector Days meant to allow our researchers to gain insights into the broad needs of industry in five sectors of particular interest. We have signed additional research agreements and are pursuing discussions on new collaborations with at least ten companies. We continue to build awareness of our activities through participation in and organization of industry days, our industrial newsletter and a growing presence on social media.

Software

AiiDA

After thorough beta-testing, last year saw the release of AiiDA 1.0 on October 31, 2019. This milestone is the result of almost two years of development on top of version 0.12.x, and comes with many major improvements described in the OSP section.

Beside the code development, one major activity is its dissemination through the scientific community and beyond. Six AiiDA tutorials were organized. Three were focused on AiiDA aiming at "writing reproducible workflows for computational materials science, using AiiDA 1.0" at EPFL (May 2019) and in Mandi, India (October 2019, Fig. 1, left), and an "AiiDA tutorial" at ISSP University of Tokyo, Japan (December 2019). Other were included as a part of larger events in September 2019: in Xiamen, China, as a component of a workshop on computational chemistry and machine learning, in Ljubljana, Slovenia, as part as the "Summer School on Advanced Materials and Molecular Modelling with Quantum ESPRESSO (QE-2019)", and at SINTEF, Oslo, Norway, as part of a "VASP & AiiDA workshop". Giovanni Pizzi, head of the Open Science platform, provided a training on the OSP, delivering a 1-week course for the PhD school of the University of Udine (Italy), giving an introduction to computational simulations of materials, in particular high-throughput computing. This was the opportunity to provide an introduction to both AiiDA and Materials Cloud. Advanced events for AiiDA developers and contributors also took place in year 6. A workshop at EPFL (March 2019) helped AiiDA 25 developers with the migration of their plugins to support both Python 2 and Python 3 as well as with changes introduced in AiiDA 1.0. The fourth annual AiiDA coding week in Fiesch attracted 15 participants (December 2019, Fig. 1, right). In February 2020, an AiiDA Hackathon is organized at CINECA Bologna, Italy, to develop code plugins and robust scientific workflows. The full list of events (past and future) is available on www.aiida.net/events/.

The AiiDA community is continuously informed through Twitter and Facebook posts.

Materials Cloud

Materials Cloud is central to MARVEL. It is built to enable the seamless sharing and dissemination of resources in computational materials science, offering educational, research, and archiving tools; simulation software and services; and curated and raw data. All its sections (*Learn*, *Work*, *Discover*, *Explore*, *Archive*)

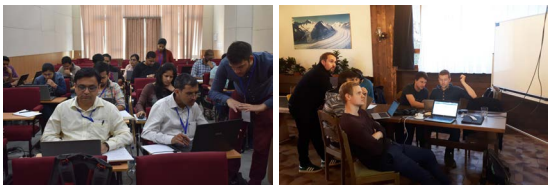

Figure 1: *Left: AiiDA tutorial at ITT Mandi, India, October 9–11, 2019. Right: Fourth annual AiiDA coding week, Fiesch, Switzerland, December 9–12, 2019.*

were continuously populated during year 6, as also mentioned in the OSP section. As already stated in the Management chapter, the *Archive* is used as open data repository for the MARVEL research data management strategy, with about 70 MARVEL related entries in year 6.
Materials Cloud furthermore benefits from a swissuniversities P-5 grant to develop the web platform to ultimately allow users to autonomously contribute hundreds of different data entries in the different sections without having to interact with one of the platform maintainers.

AiiDA lab and Quantum Mobile

Implementation developments of the AiiDA lab platform have made it possible to couple to external authentication services. We have therefore now published a demo instance of AiiDA lab where any researcher with an academic affiliation in Europe can login, thanks to the EGI Checkin service. In addition, we have started investigating which companies or other universities might be interested in having a custom on-site deployment of the platform on their premises, and we are scheduling a custom support and deployment program for next year.
Quantum Mobile, periodically upgraded, is being increasingly adopted as the platform for university courses, schools and tutorials. Last year, it was adopted in 7 different courses (EPFL, ETHZ, Amsterdam, Ghent, Addis Ababa, ICTP Trieste) in addition to all the AiiDA tutorials. Teachers and lecturers are enthusiastic about this tool, saying, for example, that "Quantum Mobile is absolutely the right tool to have" and that "it really gives a gigantic help for the organization of schools focusing on simulation codes" (testimonials on www.materialscloud.org/work/quantum-mobile).

New releases, open source codes

The non-equilibrium systems simulation package NESSi (nessi.tuxfamily.org) has been officially released. This library was developed with partial support from MARVEL in the group of Philipp Werner and allows efficient and accurate calculations based on non-equilibrium Green's functions.
ENVIRON (www.quantum-environ.org) is a module to handle environment effects in first-principles condensed-matter simulations, in particular for applications in surface science and materials design. It is developed in the groups of Nicola Marzari and Stefan Goedecker and version 1.1 was released on April 22, 2019.

Conferences and collaborations

Other Swiss and EU synergies

The long-term planning and strategy of MARVEL have made it an ideal partner for shorter-term European projects that share the same vision — so the SNSF investment has allowed to leverage complementary funding from a significant number of projects. Most notably these are the H2020 MaX Centre of Excellence for Materials Design at the Exascale (2015–2018, 2018–2021), H2020 MarketPlace — Materials Modelling Marketplace for Increased Industrial Innovation (the-marketplace-project.eu) —(2018–2022), aimed at developing a single point of access for materials modeling activities in Europe, and H2020 INTERSECT — Interoperable Material-to-Device simulation box for disruptive electronics (intersect-project.eu) —(2019–2021). This last one, started in year 6, will drive the uptake of materials modeling software in industry, bridging the gap between academic innovation and industrial novel production, with a goal of accelerating by one order of magnitude the process of materials' selection and device design and deployment; all of these projects leverage the AiiDA and Materials Cloud IT infrastructure of MARVEL. In addition, strategic efforts have taken place in the Coordination and Support Action that led to the creation of the European Materials Modelling Council, and in the provision of simulation services of the H2020 NFFA — Nanoscience Fine Foundries and Analysis (nffa.eu) —(2015–2020).
Clémence Corminboeuf is involved in two Marie Skłodowska-Curie actions, LIDOS (Light-Induced Spin Switch using Dynamic Organic Species) and D3AiSF (Screening Database to Discover Donor-Acceptor copolymers for intramolecular Singlet Fission), and Jürg Hutter in one, MOFdynamics (Investigating metal-organic frameworks using excited-state dynamics and theoretical spectroscopy).
In Switzerland, in addition to the swissuniversities P5 grant for Materials Cloud, we have to mention the launch in 2019, in collaboration with the Centre Européen de Calcul Atomique et Moléculaire (CECAM), of OSSCAR (Open Software Services for Classrooms and Research) thanks to the EPFL Open Science fund to build an open, collaborative online hub to host simulation and data-analysis tools with

an environment that offers software tools as easy-to-use services requiring little or no setup time.

Conferences organized by MARVEL members

MARVEL members organized or co-organized more than 25 conferences, tutorials or workshops, and some were also sponsored by MARVEL. All are listed in the NIRA database and on the website (nccr-marvel.ch/ctw). A selection is given here.

- The EMMC International Workshop 2019 (Vienna, Austria, February 25–27, 2019); this "invitation-only" workshop was attended by more than 170 European and international experts to discuss and contribute in setting a common direction among stakeholders in all areas of materials modeling.
- The topical symposium "Defects, Electronic and Magnetic Properties in Advanced 2D Materials Beyond Graphene" co-organized by Oleg Yazyev at the Materials Research Society spring meeting (Phoenix, AZ, USA, April 22–26, 2019).
- The PASC19 Conference (ETH Zurich, June 12–14, 2019); two mini-symposiums were organized by MARVEL members, "Artificial Intelligence and Knowledge Representation in Chemical Sciences" (Laino's group) and "Scientific Applications Written in Fortran to the Exascale Era: How Software Engineering Can Help to Fill the Gap" (Hutter's group).
- The CPMD Meeting 2019 — Pushing the Boundaries of Molecular Dynamics — co-organized by Michele Ceriotti, Nicola Marzari and Alfredo Pasquarello with CECAM (EPFL, July 22–24, 2019).

Technology transfer

Industry Sector Days

We are now able to capitalize on the industrial community we have built up around MARVEL since 2016 and our main objective is now organizing Industry Sector Days in the five following domains: 1. metallurgy, 2. organic crystals/pharmaceuticals compounds (pharma & fine chemistry), 3. energy harvesting, conversion and storage (materials for energy), 4. information-and-communication technologies (materials for electronics), and 5. catalysis and chemical synthesis (chemistry & catalysis), that align with the MARVEL D&D projects.

The purpose of the Sector Days is for the researchers to gain insights into the broad needs of industry. One main activity of MARVEL is the development of platforms for high-throughput and turnkey computations of material properties and for databases of material properties with full provenance. The collective input from Sector Day participants spanning a range of industries from the five sectors covered by MARVEL research will help guide our choices for applications of these platforms. We are looking for insights at the pre-competitive level, and hope that the industrial participants across different application domains might have similar interests. In each Sector Day, one or two specific D&D projects are presented.

To reach the above goals, we have invited both already known and newly targeted industrial representatives, senior experts and innovation managers from large companies. We identified new companies and their representatives thanks to the networks of the Industrial Advisory Board, the MARVEL PIs and the EPFL Vice-Presidency for Innovation.

In February 2019, we held the first Sector Day on metals involving six companies from different levels of the value chain and from different European countries (1 leader in aluminum, 1 car manufacturer, 2 leaders in aerospace, 1 watch/jewelry manufacturer, 1 automotive supplier). William Curtin and Michele Ceriotti presented modeling activities within the framework of D&D2, while Roland Logé (EPFL) and Helena van Swygenhoven (PSI/EPFL), not affiliated with MARVEL, shared experimental capabilities and accomplishments in metallurgy. After an extended closed industrial panel, the PIs received collective feedback on their research as well as input regarding the most important and valuable outstanding problems in alloy metallurgy, across different application domains. The main areas of industrial interest/concern were the most complicated and highest-scale problems of fatigue and fracture and hydrogen embrittlement. These are outside the domain of MARVEL research, but within the scope of other research by the participating EPFL PIs. William Curtin will continue previous discussions on alloy design and performance with two of the companies and provide information about Hydrogen embrittlement to a third. D&D2 will continue to focus on alloy design.

In October 2019, we held another Sector Day tailored towards pharmaceutical and chemical companies, sectors for which structure, stability and crystallization kinetics play strategic

roles. Representatives from eight large companies (5 pharma, 2 fine chemistry, 1 food) attended. Presentations by MARVEL PIs Ceriotti and Corminboeuf on topics ranging from crystal structure prediction to machine learning of molecular and crystal properties provided an understanding of the work being done within D&D1. Esther Amstad (EPFL) then presented microfluidic solutions to formation and control of capsules and Lyndon Emsley (EPFL) presented the potential of the NMR crystallography platform, which includes new machine-learning work in D&D1. Following the closed industrial session, the panel gave feedback on the most interesting directions for them. Several companies expressed interest in continuing discussions on a modeling platform for a consortium of companies using shared data, polymorphism, ligand-binding estimations, and chemical reactivity. Followup actions are underway to form thematic working sub-groups to develop avenues for collaboration along these lines, and to find mechanisms by which companies can share data with EPFL securely.

The program is set for a third Sector Day on materials for energy on February 7, 2020. Ten companies have confirmed their participation. Five PIs and researchers from EPFL, PSI and IBM Research will provide an overview of MARVEL research on Lithium compounds for batteries, materials for electrolysers and the use of deep neural networks in modeling.

These three Sector Days are an opportunity to invite companies not yet in contact with MARVEL, namely 1 out of 6 in metals, 7 out of 8 in pharma and 4 out of 9 in energy. Two other Sector Days, one on materials for electronics and one on chemistry & catalysis, will be organized in 2020.

Industrial collaborations

Beside the two ongoing collaborations with a large European chemical company, two additional research agreements have been signed with a leading Asian company in electronics. In the field of chemistry, another collaboration has been established with a Swiss company. Moreover, discussions for new collaborations on machine learning, chemical compounds, materials for electronics and metallurgy are going on with at least ten companies and at least two non-disclosure agreements (NDA) have been signed.

Awareness/promotion

Our efforts are going mainly in the direction of large companies, which have the potential to use and transfer materials modeling at quantum level. In 2019, we participated in two events with a MARVEL stand including a demo of AiiDA and Materials Cloud: the first was the EPFL-STI Industry Day in Lausanne (March 20, 2019), the second was the ETHZ Industry Day in Zurich (September 4, 2019). Moreover, we co-organized the third edition of the "CCMX-MARVEL Materials Science Day" on October 8 in Bern, hosting 70 participants including 24 from industry. A full day was dedicated to research in materials science with a focus on materials for energy. The morning session included an overview of MARVEL activities, followed by two MARVEL projects: "High-throughput computational screening for solid-state Lithium-ion conductors" given by Leonid Kahle (EPFL, Inc1) and "Multiferroics for energy-efficient magnetic memories" by Marisa Medarde (PSI, D&D5), combining modeling and experimental approaches. Guest speakers from Empa and Leclanché SA enriched the program with battery challenges at the experimental and industrial production levels, respectively. In the afternoon, the five current CCMX challenges were presented.

The industrial newsletter was issued three times and the number of subscribers rose to 228 in 2019 from 80 at the end of 2017 and 153 at the end of 2018. In parallel, we maintained our effort to promote MARVEL on social media (Twitter, LinkedIn).

Industrial Advisory Board

The Industrial Advisory Board provided excellent advice during a first meeting held in September 2018 during the MARVEL annual Review and Retreat. A second meeting will be held on February 6, 2020 where, in particular, progress and outcomes from the three executed Sector Days will be discussed. To fulfill another aspect of our strategic plan, we will hold a panel session for PhD students and postdocs in MARVEL and at EPFL on the topic of "Scientific research in industry: What you want to know". It will be be held after the IAB meeting and involve these board members. The goal here is to inform junior MARVEL members about the research environment and dynamic challenges of industrial research, especially as related to computational sciences, since many of them will ultimately work in industry.

6 Education and training

In year 6, we continued to encourage junior scientists to take the lead in interacting and stimulating new collaborations through a student-organized summer school and junior seminars, chaired by PhD students or postdocs. We engaged in numerous collaborations in various educational activities, in particular with CECAM for a new series, the CECAM/MARVEL Classics in molecular and materials modeling. We supported pre-university students with a summer camp as well as an information stand at a regional high school. In short, MARVEL continues to emphasize education and training activities for a broad range of interested parties through schools and workshops on topics of broad, fundamental interest.

PhD students and postdocs

In the MARVEL community

Junior seminars

MARVEL junior seminars continue taking place monthly at EPFL. They aim to intensify interactions between MARVEL junior scientists from to different research groups. Members of the EPFL community likely to be interested in MARVEL research are also invited and attend regularly. More details are given in the Communication chapter.

Summer school on Advanced Electronic Structure Methods in Condensed Matter Physics

This year, a group of MARVEL PhD students — Stefano Falletta, Patrick Gono, Michele Pizzochero (all EPFL) and Sophie Beck (ETHZ) (Fig. 1) — organized a summer school entitled "Advanced Electronic Structure Methods in Condensed Matter Physics". The organizers managed to obtain ETH Board funding for the school (for summer schools organized exclusively by doctoral students), and wanted to organize their effort somewhat independently of MARVEL. Because of the significant overlap with MARVEL themes, the NCCR opted to fully promote and support the effort of these students by providing significant sponsorship and by covering the registration costs for MARVEL members rather than organize a competing Junior Retreat.

Figure 1: *Organizing committee of the "Advanced Electronic Structure Methods in Condensed Matter Physics" summer school held at EPFL.*

The school took place on July 8–10, 2019 at EPFL and was a big success, attracting 146 participants, including 53 members of current MARVEL groups, and a few more members of phase I MARVEL groups. The 3-day school provided an overview of advanced methodologies in computational condensed matter physics. The covered topics included density functional theory, many-body perturbation theory, molecular dynamics simulations, and machine-learning approaches. The summer school consisted of lectures given by 14 internationally renowned speakers, many of whom are domain experts from leading international institutes. A doctoral course was associated with the school, offering ECTS credits for students from the ETH domain. Apart from a social dinner, a poster presentation and an apéro facilitated the open exchange of ideas within the community of computational condensed matter physics.

Education platform

The 2019 MARVEL distinguished lectures were made available on the *Learn* platform of Materials Cloud at www.materialscloud.org/learn.

Figure 2: *Left and center: MARVEL students at the 2019 Many Electron Collaboration Summer School in New York. Right: Eastern Africa School on Electronic Structure Methods and Applications in Addis Ababa.*

The AiiDA plugin migration workshop of March 2019 and the tutorial on reproducible workflows in computational materials science of May 2019 are also available on the same platform, which has been reorganized in sections and subsections to simplify navigation. This platform, central to MARVEL, is a space for students and experts to gather knowledge and consolidate expertise in the domain of materials simulation.

CECAM/MARVEL Classics in molecular and materials modeling

MARVEL and CECAM launched a new series of lectures at EPFL on "Classics in molecular and materials modeling". In this series, lecturers explain their pioneering contributions in the field of molecular and materials simulations at a level appropriate for second year master and graduate students. The lectures are followed by an interview with the presenters: they are asked to recall the period, problems, people and circumstances that accompanied the creation of milestone methods and algorithms that are now routinely used. The events are recorded. The two first lectures of the series were:

- Jean-Paul Ryckaert and Giovanni Ciccotti, April 30, 2019, on "Molecular dynamics under (holonomic) constraints"
- Michele Parrinello and Roberto Car, July 25, 2019, on "Car-Parrinello molecular dynamics".

They attracted about 40–50 participants. Both lectures are available on the *Learn* platform of Materials Cloud.

Outside MARVEL

Many electron collaboration

Five MARVEL PhD students travelled in June to the 2019 Many Electron Collaboration Summer School held at the Simons Foundation in New York (Fig. 2, left and center). The local expenses were covered by the Simons Foundation and MARVEL supported the travel. The experience there was "absolutely amazing!" The students spent a week attending lectures on topics ranging from many body physics to topological quantum chemistry to machine-learning methods. They said that the school was "a wonderful opportunity to take a step back and admire the much wider spectrum of the research currently being carried out all over the world".

Other schools

MARVEL PIs regularly organize and/or lecture to various schools in the domains of MARVEL, e.g. the yearly MolSim school — Understanding Molecular Simulation — with Berend Smit, or other CECAM schools. MARVEL students are encouraged to participate to these schools. Conferences, tutorials, workshops, and schools organized by MARVEL members are listed on the website (nccr-marvel.ch/ctw).

Initiatives in sub-Saharan Africa

MARVEL supports the African School series on Electronic Structure Methods and Applications (ASESMA). The mentoring and training is led in collaboration with the International Center for Theoretical Physics (ICTP) in Trieste. ASESMA is a bi-annual 2-week school providing an introduction to the theory of electronic structure, with an emphasis on the computational methods for practical calculations, that brings together students from countries in Africa. The next edition will take place at Kigali, Rwanda, June 1–12, 2020.
MARVEL initiated the Atomistic Simulations, Electronic Structure, Computational Materials Science and Applications: the African Network (ASESMANET), with support now from Psi-k, CECAM and MARVEL (12'000 Euros/year) matched by another 12'000 Euros/year by

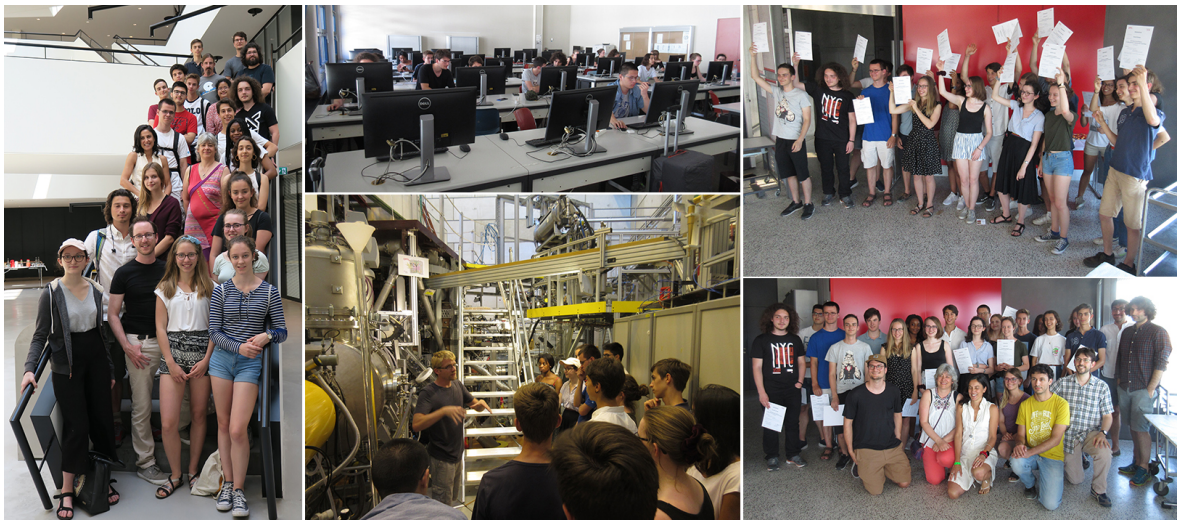

Figure 3: *MARVEL summer camp* Des atomes aux ordinateurs, à la découverte de la programmation scientifique *in 2019.*

ICTP. This network funds "African researchers to visit other researchers or research groups in Africa for focused research collaboration for two months or more at a time and also support exchange of scientists between Africa and Europe for research collaborations and conference participation".
On July 1–5, 2019, the "Eastern Africa School on Electronic Structure Methods and Applications: electrochemical energy storage and conversion materials" took place in Addis Ababa (Ethiopia), with three MARVEL members, Iurii Timrov, Riccardo de Gennaro and Tommaso Chiarotti, participating as lecturers with presentations and hands-on trainings (Fig. 2, right). The workshop brought together about 40 senior and junior researchers in the field of advanced computational methods and emerging electrochemical energy materials to share ideas, experiences, and disseminate knowledge and skills.

At the level of Master students

Courses

The MARVEL website lists the Master- and PhD-level computational courses currently offered at participating institutions and is regularly updated (nccr-marvel.ch/outreach/education-and-training/Courses-Master-level).
Oleg Yazyev has made an important contribution to teaching computational subjects at EPFL. With his colleague Henrik Rønnow, he obtained support from the Digital Resources for Instruction and Learning (DRIL) fund at EPFL. The project aims to develop a set of computational thinking tools for bachelor-level solid state physics courses. Oleg Yazyev and Michele Ceriotti are members of the Computational Thinking Working Group at EPFL.

For the younger generation

MARVEL high-school summer camp

This year's MARVEL summer camp *Des atomes aux ordinateurs, à la découverte de la programmation scientifique* organized in collaboration with the EPFL Education Outreach Department attracted 21 high school students — 10 women and 11 men — for a full week of lectures, exercises and lab visits built around the theme of scientific programming (Fig. 3). With motivations ranging from a general desire to learn more about EPFL overall to a "passion for the chemical properties of different materials," students came from French-, German- and, Italian-speaking Switzerland — as well as France, the U.S. and even Hong Kong — to deepen their knowledge of the programming language Python. Led by Michele Ceriotti, and EPFL HPC application experts Vincent Keller and Nicolas Richart, a team of six MARVEL volunteer postdocs and PhD students led the students through a basic introduction to Python to applied exercises in cellular automaton, molecular dynamics and machine learning. The week was enhanced by visits to the clusters of SCITAS — the home of high-performance computing at EPFL — Clémence Corminboeuf's Laboratory of Computational Molecular Design, and the Swiss Plasma Center. Student evaluations indicated that the week was a success: participants over-

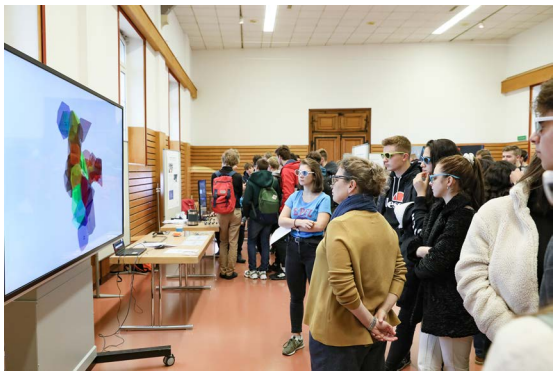

Figure 4: *Thematic day at the* Lycée cantonal de Porrentruy *on October 30, 2019.*

whelmingly agreed that they enjoyed attending the camp, that it motivated them to learn more about the topic and that they would recommend it to their friends. The decision of reducing the length of the camp to one week was positively received. This makes an activity that takes place during the school holidays more attractive. A new edition is planned on June 29 – July 3, 2020.

Thematic day in Porrentruy

EPFL Education Outreach Department organizes thematic days in high schools that allow students to discover various research domains within their own four walls, with conferences and feature demonstrations. MARVEL participated in the most recent event, "Digital in our daily lives, today and tomorrow", at the *Lycée cantonal de Porrentruy* on October 30, 2019, focused on the theme of digital education. With their stand *Concevoir les matériaux du futur grâce aux ordinateurs*, the MARVEL team (Lidia Favre-Quattropani, Antimo Marrazzo and Emilie Vuille-Dit-Bille, an EPFL Master student) introduced the teenagers to concepts such as sound, heat, vibrations and, ultimately, phonons, and discussed research in computational materials discovery. The stand acted as a showcase for the entire NCCR, with 3D videos providing an overview of our research projects (Fig. 4).

7 Equal opportunities

In year 6, we are pursuing our various successful activities promoting equal opportunities, in close collaboration with EPFL as a home institution as well as implementing novel initiatives. The latter were presented in our "Equal opportunities, analysis and plans" document. which was positively received by the review panel and the SNSF. Our rich portfolio includes INSPIRE Potentials MARVEL Master's fellowships, activities for girls and young women at the high school level, newly implemented gender trainings, upcoming Agility Plus grants and a survey of MARVEL members.

Numbers

At the national level, the share of women in MARVEL disciplines (physics, chemistry, materials science and computer science) is very low. Table 7.1 gives the number and share of women and men involved in MARVEL in years 1 to 6 (from NIRA). These figures are close to those found in Swiss academic institutions in these disciplines. The positive point is the increases achieved for postdocs and PhD students. The latter category reached 25% in years 5 and 6, with 16 female PhD students in year 6. This increase is a direct impact of the INSPIRE Potentials MARVEL Master's fellowships (see below). In sharp contrast, we note a decrease of female group leaders, which essentially arises from the reorganization of the phase II projects, with larger D&D projects and the termination of the PP7 experimental platform of phase I, resulting in an overall decrease of PIs and particularly female PIs. This situation will be mitigated in years 7 and 8 thanks to the Agility Plus call (see below). As always, the low share of women prompted us to act all over the path from young girls to high school students, with University students and then throughout the academic path.

Raising gender awareness

Gender training

The gender trainings, proposed by the Director as a followup from the site visit of year 2, were implemented in year 6. MARVEL offered 3-hours training sessions on gender and ethical/respectful behavior taught by Prof. Marianne Schmid Mast, chair of Organizational Behaviour at the Faculty of Business and Economics (HEC) at UniL to PIs and people with leadership positions ("workshops for PIs") as well as to PhD students and postdocs ("workshops for students"). Two "workshops for PIs", one at EPFL and one at UZH, were organized in May 2019, with respectively 8 (3 women) and 9 (2 women) participants. Four "workshops for students" were organized, two in June and two in October 2019, one at EPFL and one at UZH each time, with 58 participants in total, with about 30% women and 60% PhD students. Three quarters of the MARVEL groups were represented overall.

The goals of these workshops consist in raising awareness on how implicit biases and stereotypes can affect how we see and judge others and to develop skills on how to overcome bias. The "workshops for PIs" also provide concrete advices on how to avoid bias in recruitment and how to create an inclusive environment. An aperitif was organized after the workshops to promote open discussions. The interaction with and between participants was central to the sessions. The feedback from the participants was very positive. In the anonymous questionnaire, the workshops were described as "very clear and well presented", "never boring", "very instructive with a relaxed and enjoyable atmosphere", with "many real life examples", "a lot of interaction" and "room for discussion from the participants", "in a respectful manner". Regarding criticisms, "it would have been interesting to focus a bit more on the ways to avoid to be driven by biases", "on how to combat implicit bias", "on what we can do, at our level, to be more inclusive and create a good work environment". The requests for practical, usable solutions should be addressed for the next edi-

	year 1		year 2		year 3		year 4		year 5		year 6	
	W	M	W	M	W	M	W	M	W	M	W	M
Group leaders	4 17%	20 83%	6 18%	27 82%	9 22%	32 78%	8 19%	34 81%	4 13%	27 87%	3 10%	27 90%
Senior res.	1 13%	7 87%	0 0%	8 100%	2 9%	21 91%	3 10%	28 90%	4 19%	17 81%	3 15%	17 85%
Postdocs	5 11%	39 89%	8 11%	65 89%	13 16%	69 84%	15 20%	61 80%	13 21%	48 79%	9 18%	40 82%
PhD	6 26%	17 74%	9 20%	35 80%	9 19%	38 81%	10 19%	43 81%	14 25%	41 75%	16 26%	46 74%

Table 7.1: *Number and share of women (W) and men (M) involved in MARVEL in years 1, to 6 (From NIRA).*

tions that are planned in the second half of year 7. One difficulty was to convince people to participate. While the four workshops for students were ultimately all full, multiple reminders were needed for people to register and students needed to be encouraged by their PIs to apply.

Gender and working climate

A short survey to assess the gender and working climates in the MARVEL laboratories is currently under preparation. It will be sent out in February and collected feedback will be presented at the April site visit.

Advance women scientists

Leadership of projects & allocation of funding

It was announced in our "Equal opportunities, analysis and plans" document that the reallocation of MARVEL funding for equal opportunities will enable the funding of four Agility Plus projects supporting a PhD student or half a postdoc between May 2020 and April 2022 for a total amount of 95'000 to 120'000 CHF per grant. The call was opened at the end of November 2019, with a deadline on January 15, 2020. We identified in the Swiss ecosystem six junior woman scientists (typically non-tenured assistant professors) in computational or experimental fields related to MARVEL activities. Clémence Corminboeuf, as head of the selection committee, proactively contacted them and encouraged them to submit a project that needed to be integrated into one of the existing MARVEL projects (D&Ds, Incs or platforms). The call was also announced in the December edition of the MARVEL internal newsletter. Nine applications were received, all from women, including four of the proactively contacted ones. The selection committee is composed of Clémence Corminboeuf, William Curtin, Joost VandeVondele and Nicola Marzari. The selection process is in progress and a decision is planned for mid-February, to be able to launch the new projects in May.

INSPIRE Potentials fellowships

The INSPIRE Potentials MARVEL Master's fellowships are continuing as a flagship action for MARVEL. They offer 6-month fellowships to female students to conduct their Master's research thesis in a MARVEL computational group. With eight fellowships every year, this initiative seeks to increase the number of women researchers in the field of simulation science. The objective is to double the number of female PhD students in MARVEL during phase II. Since 2016, and after 8 rounds, 55 applications and 26 fellowships have been awarded. Of these, 6 are already continuing at the PhD level in the same groups, 2 will start a PhD in a MARVEL group in February 2020, and an additional 2 received offers to stay for a PhD (Fig. 1, left). Of those who left, most (at least 7) are continuing at the PhD level abroad. The two calls of 2019 awarded 5 fellowships for a total of 15 applications. The three in the spring went to Jigyasa Nigam (Indian Institute of Space Science and Technology (IIST), Trivandrum, IN) and Linnea Mørch Folkmann (Univ. Copenhagen, DK) in the Ceriotti group at EPFL and Joanna Stoycheva (Sofia Univ. "St. Kliment Ohridski", BG) in the Spaldin group at ETHZ. The two in autumn went to Dune André (Ecole Normale Sup., Paris, FR) in the Corminboeuf group at EPFL and to Yuting Chen (ETHZ) in the Passerone group at Empa.

The feedback from the students is excellent. We could read for example "Dear MARVEL Equal Opportunities team, I would like to thank you all for the provided help starting from the very beginning. Indeed I enjoyed carrying out my masters research and I thing it is an amazing opportunity for master students. Also I went to EPFL to the summer school on computational chemistry which also had a big im-

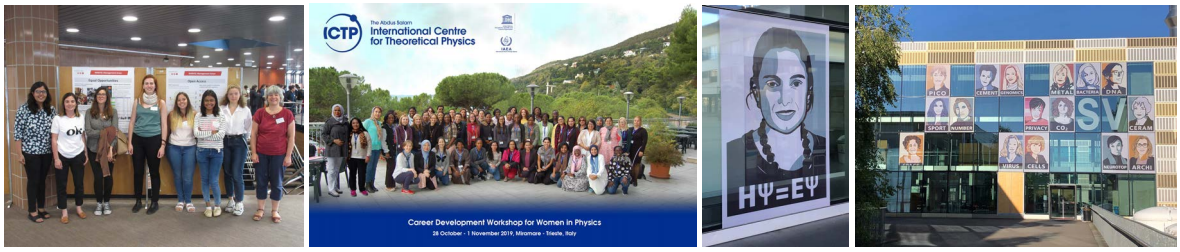

Figure 1: *Left: present and previous MARVEL INSPIRE Potentials fellows at the MARVEL Review and Retreat, September 2019, EPFL. From left to right. Jigyasa Nigam, Prof. Clémence Corminboeuf, Norma Rivano, Martina Danese, Julieta Trapé, Sinjini Bhattacharjee, Nataliya Lopanitsyna, and Dr Lidia Favre-Quattropani. Center: group photo at the Career Development Workshop for Women in Physics, ICTP, Trieste (© ICTP). Right (2 pictures): "Female Leadership in Science" Exhibition for EPFL's 50th anniversary, with Clémence Corminboeuf on the left picture.*

pact on my decision staying in academia." We also ask the PIs for a short midterm assessment to make sure everything is going well. One big challenge however is to obtain more applications. We publicize the action on different platforms, websites, Twitter, LinkedIn, specific mailing lists (as psi-k.net/jobs), and through the PIs and the Swiss institutions' equality offices. Yet, it is difficult to attract a larger number of (good) candidates per round. A likely reason is the "narrow range" of computational material science. We just launched a new series of interviews of participating students to populate the MARVEL website and newsletters with new testimonies. Additional effort is being made to ensure that we retain them within MARVEL as much as possible (see the figures above).

Career development

- MARVEL supported (5'000 CHF) the 2019 edition of the Career Development Workshop for Women in Physics, held at the ICTP, Trieste, on October 28–31, and co-organized by Nicola Spaldin (MARVEL PI) and Shobhana Narasimhan (member of the MARVEL review panel). This nearly biennial workshop "brings together women physicists from all over the world" to share thoughts and experiences and develop "the various non-academic skills that are needed to succeed in a career in science" (Fig. 1, center).
- We were approached by the NCCR Digital Fabrication to co-organize a new series of negotiation workshops for women in science with Nancy Houfek. A first series of workshops was organized by MARVEL in October 2015 with participants from nine other NCCRs. Preliminary contacts were made with Nancy Houfek. Dates in October 2020 have been set aside at ETHZ and EPFL with, in both places, separate workshops for professors and for PhD students and postdocs. Remaining money from the 2015 edition (about 3'500 CHF) will be used. Other NCCRs were informally approached and have shown interest to join.
- MARVEL female researchers were regularly informed about dedicated activities (lunch events, conferences or workshops) organized at EPFL or in other participating institutions. Female PhD students and postdocs are encouraged to participate in training, mentoring and coaching programs offered at the various institutions.
- On March 8, 2019, the EPFL Alumni and Equal Opportunities offices organized for the second time their event for the International Women's Day "50 years of EPFL Women", which also launched the EPFL 50th anniversary celebrations, with testimonies from five generations of women graduates. Once more, MARVEL had a stand with information about its activities regarding equal opportunities. The event will be repeated on March 5, 2020, and MARVEL will be present.
- MARVEL decided to contribute 5'000 CHF to the EPFL-WISH Foundation fellowships for Masters projects abroad.

Recognition for female researchers' excellence and increase of their visibility

We take care and promote the visibility of MARVEL women scientists.

- In preparation of 2019 international women's day, we have been speaking

Figure 2: *Action for girls in 2019. From left to right:* Coding club des filles *in Bienne;* Polythèmes *workshop on materials;* Maths en jeu *(© EPFL SPS for the three pictures); lab visit during the the summer camp* Matériaux super géniaux.

with the women of MARVEL about their triumphs as well as the challenges they face in pursuing their careers and put their testimonies on a dedicated webpage.

- Based on an idea from Nicola Marzari, MARVEL's director, and sponsored by MARVEL, the EPFL Materials science and engineering department commissioned an artwork by Alban Kakulya, unveiled in November 2018. The project was extended by the EPFL Equal Opportunities Office to "Female Leadership in Science" portraying 50 women professors at EPFL at the occasion of EPFL's 50^{th} anniversary, associated with short documentaries from the French-speaking Swiss television company, the *Court du jour*, highlighting women in science. Clémence Corminboeuf is part of both the exhibition (Fig. 1, right) and the movies.
- Clémence Corminboeuf was also selected as one of the "100 Women and Thousands More" (100femmes.ch), an interregional communication campaign aiming to show young people, especially girls, the wide range of exciting paths and exceptional careers taken by women living in French- and Italian-speaking Switzerland as well as neighboring France.
- In 2019, Nicola Spaldin was awarded the Swiss Science Prize Marcel Benoist, also known as the "Swiss Nobel Prize". She was also elected as foreign member of the National Academy of Engineering in the United States. Clémence Corminboeuf received the Teaching Excellence Award of the EPFL chemistry and chemical engineering section.
- We continue to invite renowned female scientists for the MARVEL distinguished lectures, with, in 2019, all given by women: Profs. Emily A. Carter (Princeton Univ., now Executive Vice Chancellor and Provost at UCLA) in June and Giulia Galli (Univ. Chicago) in November, also as an EPFL campus lecture part of the EPFL's 50^{th} anniversary, giving increased visibility.
- We put a lot of attention on women representation in educational or public events organized by MARVEL or in which MARVEL participates, e.g., asking that at least one of the three Ig Nobel recipients presenting each year at the Ig Nobel Award Tour Show is a woman, having women among the supervising students for the summer camp for high school students (2 out of 6 in 2019 and in 2020) and at the MARVEL stand at EPFL open days (5 out of 24).

Actions for girls, young women and future scientists

MARVEL continues to support the activities for girls organized by EPFL Science Outreach Department. In 2019, we had

- two editions of the *Polythèmes* workshop on materials *Diamant, alu, caoutchouc, ils sont fous ces matériaux !* for girls only in March-April and October-November 2019; next edition April-May 2020 (Fig. 2, center left);
- the fifth edition of the summer camp *Matériaux super géniaux* for girls only in August 2019; some girls are also welcomed in MARVEL groups for a lab visit (Fig. 2, right); next edition in August 2020;
- the chemistry summer camp for girls and boys (with 50% girls) in August 2019; next edition in August 2020.

To this, we can add the continued support of

- the mathematic workshops *Maths en jeu* (Fig. 2, center right);

- the *Coding club des filles*, offering coding workshops for girls 11 to 15 years old organized throughout French-speaking Switzerland with great success (Fig. 2, left) ; a virtual platform has been built to allow the participants to share their experiences and projects and to be put in contact with coaches and mentors.

Moreover, we have decided to set aside for girls half of the spots of the MARVEL-organized summer camp for high-school students *Des atomes aux ordinateurs, à la découverte de la programmation scientifique*; target met in 2019, with 10 women and 11 men (more details in the Education and Training chapter). This condition is renewed in 2020. Indeed, high-school is a good time to help young women consider university studies in a scientific domain and the summer camp is an opportunity show them a little bit what it looks like.

Other actions

- Two SNSF Flexibility grants to help cover the external childcare costs assigned to MARVEL parents in 2018 were renewed in 2019.
- Some women in MARVEL — and supportive men — participated to the women strike on June 14, 2019.
- Together with EPFL representatives, MARVEL participated in the SNSF-organized NCCR Exchange Platform on Equal Opportunities on November 8, 2019.

8 Communication

During year 6, MARVEL carried on implementing its communication strategy. We developed communication on MARVEL research by regularly updating the website with news, scientific highlights and feature stories, by expanding the use of EurekAlert with the publication of press releases on topics deemed to be of interest to the wider scientific community, and by associating with existing events such as the World Conference of Science Journalists or the "My Thesis in 180 Seconds" contest. We continued to build the MARVEL community with the internal newsletter as well as the organization of various meetings, including junior seminars and the Review and Retreat. MARVEL also organized events for a wider public (IgNobel Award Tour Show, Mary Ann Mansigh conversation series) and participated in external events to open up communication to scientists in the domain of material simulation at the national and international level, industrial partners, media and the general public.

Internal and external communication

Website and newsletter

We continued adding to the website, contributing more than 30 news items on various activities, events, awards and other news of interest to the broader community. We also wrote 21 (as of January 31, 2020) feature stories and science highlights focused on the research of the groups.
We published ten internal and scientific newsletters during the year. External readership of the scientific newsletter grew to 132 subscribers, up from 63 the year before.

MARVEL distinguished lectures

MARVEL invites internationally prominent experts in the field of computational materials science to present MARVEL distinguished lectures. This year we invited:

- Prof. Emily A. Carter (Princeton University) presenting "Quantum Simulations of Sustainable Energy Materials" on June 17;
- Prof. Giulia Galli (University of Chicago) presenting "Marveling at materials through in-silico lenses" on November 13. This MARVEL distinguished lecture was included as an EPFL campus lecture part of EPFL's 50^{th} anniversary celebrations, enhancing the visibility of MARVEL (Fig. 1, left).

As always, to favor work-life balance, a specific effort has been made to organize these events at 16:15, a time suitable for parents, for whom a late end to a lecture might be problematic.

Internal communication

Review and Retreat

The sixth Review and Retreat took place at EPFL on September 5–6, 2019. A total of 150 researchers attended the event. The eight core projects (D&Ds and Incs) were presented on the first day, each being assigned a slot of 45 minutes, with 35 min. for the presentation itself (delivered by the sole project leader) and 10 min. for Q&A (where other partners could participate). The PIs were informed that the Scientific Advisory Board (SAB) would evaluate in detail the status, achievements and potential of their project, and match of the project against the four NCCR metrics. In addition to the presentations themselves, the posters were an integral part of the evaluation: a poster session for SAB review was scheduled during lunch on the first day. In order to ensure a manageable review, a total of 62 posters (7 max. per project) were presented by the community. This event, in addition to showcasing MARVEL research, is a precious opportunity to build the MARVEL community.

MARVEL junior seminars

The MARVEL junior seminars continue taking place monthly at EPFL (except July, Au-

Figure 1: *From left to right: MARVEL distinguished lecture and EPFL campus lecture by Giulia Galli, November 2019; MARVEL Transport Day at Empa, May 2019; MARVEL Lunch@Lab for the World Conference of Science Journalists, July 2019.*

gust, September). They aim to intensify interactions between the MARVEL junior scientists belonging to different research groups (i.e. PhD students and postdocs either funded directly by the NCCR or by a matching contribution). The EPFL community interested in MARVEL research topics is welcome to attend, and does regularly. On average, 40 to 45 people attend. The organizing committee consists of seven delegates among PhD and postdoctoral students, who each represent MARVEL EPFL research groups and act as chairs, with the mission to hold a lively Q&A session. Each seminar consists of two 25-minute presentations, followed by time for discussion. Pizza is served before the seminar, and the participants are invited after the seminar for coffee and dessert to continue the discussion with the speakers.

Other meetings

D&D3 project organized on May 13, 2019 a MARVEL Transport Day at Empa to discuss the results so far as well as future perspectives in the project. External groups involved in experimental and theoretical collaborations participated (Fig. 1, center). Other MARVEL projects also met regularly during the year, either in person or remotely.

External communication

Communicating MARVEL research

EurekAlert

We expanded use of EurekAlert, first started in January 2019, with the publication of ten press releases on topics deemed to be of interest to the wider scientific community. All of them were viewed more than 1'000 times, according to EurekAlert statistics, and the majority were viewed more than 2'000 times. This is in line with our strategy of increased outreach to the wider scientific and journalist community.

Social media

Our social media work has focused on Twitter account @nccr_marvel, which now has a following of 929 subscribers as of January 31, 2020, up from 550 at the end of the last reporting period. We posted a total of 156 tweets during the year, earning nearly 200'000 impressions, and an average engagement rate, which reflects the number of times that readers interact with a tweet, of 2.5%. According to 2018 industry benchmarks by Rival IQ, the median Twitter engagement rate across every industry is 0.046%. We have also launched an account on LinkedIn, primarily with the intention of building and reinforcing ties within the MARVEL community. This community is continuing to grow.

MARVEL in web news and in the press

In addition to being frequently featured on MARVEL website and the websites of their home institutions, the research of MARVEL teams has been covered in numerous press articles. For example, the EPFL news story "New material design tops carbon-capture from wet flue gases" on a result of Berend Smit's group (December 11) received a warm echo in the local and national press. An interview with Nicola Marzari, "Toccare il futuro" by Valentina Ravizza, featured in *Style Magazine*, monthly supplement of the *Corriere della Sera* (March 27).

World Conference of Science Journalists

MARVEL participated in the Lunch@Lab program organized as part of the 11^{th} edition of the World Conference of Science Journalists (WCSJ) that took place at the SwissTech Convention Center of EPFL on July 1–5, 2019. The event attracted journalists from all over the world for five days of presentations, workshops, discussions, visits and field trips. Lunch@Lab gave participants a chance to better understand the work going on in the labs

Figure 2: *From left to right: Marc Abraham presenting the Ig Nobel Award Tour Show at EPFL; opening of Julie Birenbaum's light painting exhibition in the presence of the artist, March 2019; CECAM/MARVEL Mary Ann Mansigh conversation series with Massimo Noro, May 2019.*

they visited. We organized a program giving both an overview of MARVEL as well as information on the specific research of four MARVEL groups, all based around the Materials Cloud platform. The four 5-minute presentations were filmed and these videos will serve as the basis for continued scientific outreach. Though few journalists attended, there was engaged conversation with those present (Fig. 1, right).

My Thesis in 180 Seconds

Three NCCR MARVEL members — Bardiya Valizadeh and Daniele Ongari, both PhD students in Smit's group as well as Leonid Kahle in Marzari's group — were invited to participate in the EPFL final of "My Thesis in 180 Seconds" on March 7, 2019. This international contest asks competitors to present their research, in plain language, to a non-specialist audience and a jury made up of researchers, journalists and business people in just three minutes.

Events

Ig Nobel Award Tour Show

On March 25, EPFL welcomed for the fourth time the Ig Nobel Award Tour Show with the support of the NCCR MARVEL. Mark Abraham presented the prize and the actuality of research that makes people laugh and then think (Fig. 2, left). This year's program included Claire Rind (Institute of Neuroscience, Newcastle Univ., UK), 2005 Peace Prize winner — "Monitoring a brain cell of a locust while that locust watches selected highlights from the movie Star Wars" and Marc-Antoine Fardin (Lab. de Physique, ENS Lyon, FR), the 2017 Physics Prize winner speaking on "Can a cat be both a solid and a liquid?". The third Ig Nobel laureate, Mark Dingemanse (Max Planck Inst. for Psycholinguistics, Nijmegen, NL), the 2015 Literature Prize winner for his work "The word 'huh?' seems to exist in every human language" could not come because of flight cancellation due to bad weather. Pierre Barthelemy, science journalist, came to the rescue, doing an impromptu talk, in French, about Ig Nobel Prize-winning research he has written about in *Le Monde*. The next edition will take place on March 30, 2020.

Movies

With CECAM (Centre Européen de Calcul Atomique et Moléculaire), MARVEL hosted a series of three movie nights, the "Soirées CECAM/MARVEL", showing movies with scientific themes (A Beautiful Mind, Gravity and Good Will Hunting). We aimed to expand awareness of NCCR MARVEL and our work and to offer more after-hours activities on campus. While those who attended appreciated the introduction to MARVEL and CECAM as well as the films selected, participation was not high enough to warrant continuing the initiative.

CECAM/MARVEL Mary Ann Mansigh conversation series

MARVEL, together with CECAM, organized the event "Computer modeling for industrial applications", with Massimo Noro, Business Development Director, Science and Technology at STFC Daresbury, on May 8, 2019, as first event of the Mary Ann Mansigh conversation series.
The goal was to provide the broad EPFL community with an opportunity to meet leading figures in domains in close proximity with the development and application of molecular and materials simulations. The guest engaged in a conversation to describe his or her expertise, approaches to research, and career paths in this area from a perspective not usually encountered in "standard" scientific presentations.
In this conversation, Massimo Noro, formerly at Unilever and current Business Development Director at Daresbury labs, discussed with William Curtin and with the audience the rele-

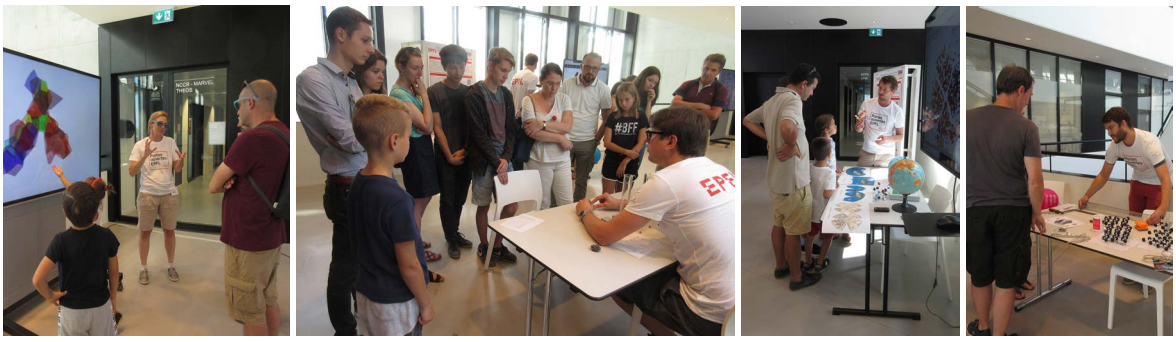

Figure 3: *MARVEL stand at the EPFL Open days, September 2019, with 3D movies and fun experiments.*

vance of simulation for industry and his role as the leader of an important computing facility that interacts directly with industry. This conversation offered insight into how to promote and facilitate industrial use of simulation and modeling, and allowed the audience to meet a "living example" of a carrier path for simulators outside academia (Fig. 2, right). The event was live-streamed and recorded.

EPFL Open House

As part of the celebrations for its 50^{th} anniversary, EPFL opened its doors to nearly 40'000 science and engineering enthusiasts on September 14–15, 2019. On this occasion, MARVEL presented a stand with 3D movies and fun experiments to show how novel materials are created by computational design (Fig. 3). New creations from the groups of Ceriotti and Corminboeuf completed the already fascinating collection of 3D movies. Curtin's group interested the public with intriguing properties of metals and glass and how their different atomic structure and bonds lead to very different observable behavior. Ceriotti's group showed how complex molecular conformations can be simplified through sketch-map representations, which can help to understand the properties and interactions of structurally-complex materials. To better convey the message they encouraged the visitors to play with molecular toy models and explore different sketch-maps on a computer. Marzari's group tolda story about sound, heat and, ultimately, phonons, with direct presentation of scientific results on the Materials Cloud platform.

Julie Birenbaum, the light painting artist

The photo exhibition by Julie Birenbaum, the light painting artist, was displayed at ETH Zurich Hönggerberg on March 21–28, 2019, thanks to the support of Spaldin's Materials Theory group and MARVEL. At the frontier between physics and metaphysics, Julie Birenbaum explores the mysterious beauty of the light that materializes in her hands, seemingly sparking with life. Through the photography, she presents her vision of the relation between art and science (Fig. 2, center).

9 Structural aspects

The call for the promised tenure-track position in computational materials science is currently being discussed in the faculty.

Annex 2 Structural measures implementation status

Planned measures according to annex 3 of the NCCR contract for phase II	Current status of implementation and comments
Infrastructure	
Additional measures	
250'000 CHF in cash for data storage and services for Materials Cloud	Ongoing
Faculty	
Planned new professorship	
New Assistant professor (PATT) in Computational Material Science (Institute of Materials), planned for 2020	The call is not yet open
Continuation of professorships of phase I	
- Michele Ceriotti	Ongoing
- Oleg Yazyev	Ongoing
- Martin Jaggi	Ongoing

Specific conditions and requirements according to Article 10 of the NCCR contract for phase II	Current status of implementation and comments
Equal opportunities, analysis and plans	The implementation of the new initiatives has started. The details are given in chapter 7, Equal opportunities

Annex 3 Publication list

All publications have been entered in NIRA, and are listed below, sorted by group leaders. We list publications either resulting directly from the NCCR (marked with a red hexagon) or with minor contributions from the NCCR. The following lists cover the period from February 1, 2019 to January 31, 2020.

1. Scientific articles in journals with peer review
2. Scientific articles in journals without peer review
3. Publications from lists 1 and 2 involving several groups

1. Scientific articles in journals with peer review

Phase II PIs

Group of Ulrich Aschauer

- C. Ricca, I. Timrov, M. Cococcioni, N. Marzari, and U. Aschauer
  Self-consistent site-dependent DFT+U study of stoichiometric and defective $SrMnO_3$
  Physical Review B **99**, 094102 (2019).
  Group(s): Aschauer, Marzari / Project(s): DD5, DD3

Group of Michele Ceriotti

- E. A. Engel, A. Anelli, A. Hofstetter, F. Paruzzo, L. Emsley, and M. Ceriotti
  A Bayesian approach to NMR crystal structure determination
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Group of Joost VandeVondele

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Group of Claire Villevieille

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Group of Anatole von Lilienfeld

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Group(s): Curtin, Marzari, Pizzi, Spaldin, Troyer, Yazyev / Project(s): OSP, PP6

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Group(s): Soluyanov, Yazyev / Project(s): DD6

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Phase I PIs, not active in phase II

Group of Raffaella Buonsanti

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Group(s): Buonsanti, Corminboeuf / Project(s): VP2, PP7

Group of Volkan Cevher

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International Journal of Quantum Chemistry **119**, e25872 (2019).

Group(s): Cevher, Ceriotti / Project(s): HP5

Group of Antoine Georges

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Group(s): Georges / Project(s): VP1

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Group(s): Shi, Georges / Project(s): DD6

Group of Thomas Lippert

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Group of Wendy Queen

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An In-Situ Neutron Diffraction and DFT Study of Hydrogen Adsorption in a Sodalite-Type Metal-Organic Framework, Cu-BTTri
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Group of Marta Rossell

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Group of Thomas J. Schmidt

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Group of Thorsten Schmitt

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Group(s): Hutter, Schmitt / Project(s): PP7

H. Elnaggar, R.-P. Wang, S. Lafuerza, E. Paris, Y. Tseng, D. McNally, A. Komarek, M. Haverkort, M. Sikora, T. Schmitt, and F. M. F. de Groot
Magnetic Contrast at Spin-Flip Excitations: An Advanced X-Ray Spectroscopy Tool to Study Magnetic-Ordering
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Group(s): Schmitt / Project(s): PP7

F. A. Garcia, O. Ivashko, D. E. McNally, L. Das, M. M. Piva, C. Adriano, P. G. Pagliuso, J. Chang, T. Schmitt, and C. Monney
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O. Ivashko, M. Horio, W. Wan, N. B. Christensen, D. E. McNally, E. Paris, Y. Tseng, N. E. Shaik, H. M. Ronnow, H. I. Wei, C. Adamo, C. Lichtensteiger, M. Gibert, M. R. Beasley, K. M. Shen, J. M. Tomczak, T. Schmitt, and J. Chang
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Group of Matthias Troyer

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Wannier90 as a community code: new features and applications

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Group(s): Curtin, Marzari, Pizzi, Spaldin, Troyer, Yazyev / Project(s): OSP, PP6

MARVEL visitors

François Gygi (UC Davis)

M. D. LaCount and F. Gygi

Ensemble first-principles molecular dynamics simulations of water using the SCAN meta-GGA density functional

The Journal of Chemical Physics **151**, 164101 (2019).

Group(s): NCCR / Project(s): NCCR

2. Scientific articles in journals without peer review

Group of Ulrich Aschauer

C. Ricca, I. Timrov, M. Cococcioni, N. Marzari, and U. Aschauer
Self-consistent DFT+U+V study of oxygen vacancies in $SrTiO_3$
arXiv:2001.06540 (2020).
Group(s): Aschauer, Marzari / Project(s): DD5, DD3

Group of Claude Ederer

A. Hampel, S. Beck, and C. Ederer
Charge self-consistency and double-counting in DFT+DMFT calculations for complex transition metal oxides
arXiv:1907.10339 (2019).
Group(s): Ederer / Project(s): DD5

Group of Jürg Hutter

T. Duignan, G. K. Schenter, J. Fulton, T. Huthwelker, M. Balasubramanian, M. Galib, M. D. Baer, J. Wilhelm, J. Hutter, M. D. Ben, X. S. Zhao, and C. J. Mundy
Quantifying the hydration structure of sodium and potassium ions: taking additional steps on Jacob's Ladder
ChemRxiv. Preprint (2019), doi:10.26434/chemrxiv.7466426.v2.
Group(s): Hutter / Project(s): HP3

Group of Nicola Marzari

C. Ricca, I. Timrov, M. Cococcioni, N. Marzari, and U. Aschauer
Self-consistent DFT+U+V study of oxygen vacancies in $SrTiO_3$
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L. Banszerus, T. Sohier, A. Epping, F. Winkler, F. Libisch, F. Haupt, K. Watanabe, T. Taniguchi, K. Müller-Caspary, N. Marzari, F. Mauri, B. Beschoten, and C. Stampfer
Extraordinary high room-temperature carrier mobility in graphene-WSe_2 heterostructures
arXiv:1909.09523 (2019).
Group(s): Marzari / Project(s): DD3

I. Cucchi, A. Marrazzo, E. Cappelli, S. Riccò, F. Y. Bruno, S. Lisi, M. Hoesch, T. K. Kim, C. Cacho, C. Besnard, E. Giannini, N. Marzari, M. Gibertini, F. Baumberger, and A. Tamai
Bulk and surface electronic structure of the dual-topology semimetal Pt_2HgSe_3
arXiv:1909.05051 (2019).
Group(s): Marzari / Project(s): DD3

P. D'Amico, M. Gibertini, D. Prezzi, D. Varsano, A. Ferretti, N. Marzari, and E. Molinari
Intrinsic edge excitons in two-dimensional MoS_2
arXiv:1909.01613 (2019).
Group(s): Marzari / Project(s): DD3

A. Marrazzo, N. Marzari, and M. Gibertini
Emergent dual topology in the three-dimensional Kane-Mele Pt_2HgSe_3
arXiv:1909.05050 (2019).
Group(s): Marzari / Project(s): DD3

A. Floris, I. Timrov, B. Himmetoglu, N. Marzari, S. de Gironcoli, and M. Cococcioni
Hubbard-corrected density functional perturbation theory with ultrasoft pseudopotentials
arXiv:1910.06195, *to be published in* Physical Review B (2020).
Group(s): Marzari / Project(s): OSP

F. Nattino and N. Marzari
Operando XANES from first-principles and its application to iridium oxide
arXiv:1912.09769 (2019).
Group(s): Marzari / Project(s): HP3, OSP

M. Puppin, S. Polishchuk, N. Colonna, A. Crepaldi, D. N. Dirin, O. Nazarenko, R. De Gennaro, G. Gatti, S. Roth, T. Barillot, L. Poletto, R. P. Xian, L. Rettig, M. Wolf, R. Ernstorfer, M. V. Kovalenko, N. Marzari, M. Grioni, and M. Chergui
Evidence of large polarons in photoemission band mapping of the perovskite semiconductor $CsPbBr_3$
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Group(s): Marzari / Project(s): OSP

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A High-Throughput Computational Study Driven by the AiiDA Materials Informatics Framework and the PAULING FILE *as Reference Database*
in *Materials Informatics: Methods, Tools and Applications*, O. Isayev, A. Tropsha, and S. Curtarolo, eds. (John Wiley & Sons, Ltd,

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Group(s): Marzari, Pizzi / Project(s): OSP

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  arXiv:1909.00433 (2019).
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Group of Giovanni Pizzi

M. Uhrin, G. Pizzi, N. Mounet, N. Marzari, and P. Villars
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Group(s): Marzari, Pizzi / Project(s): OSP

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  Automated high-throughput Wannierisation
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Group of Michele Parrinello

- M. Invernizzi and M. Parrinello
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  arXiv:1909.07250 (2019).
  Group(s): Parrinello / Project(s): DD1

Group of Volker Roth

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Exploring Data Through Archetypal Representatives
in *NeurIPS 2019 Learning Meaningful Representations of Life Workshop, to be published* (2020).
Group(s): Roth / Project(s): Inc2

Group of Alexey Soluyanov

A. Alexandradinata, A. Nelson, and A. A. Soluyanov
Teleportation of Berry curvature on the surface of a Hopf insulator
arXiv:1910.10717 (2019).
Group(s): Soluyanov / Project(s): DD6

Group of Anatole von Lilienfeld

- A. S. Christensen, L. A. Bratholm, F. A. Faber, and O. A. von Lilienfeld
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  arXiv:1909.01946 (2019).
  Group(s): von Lilienfeld / Project(s): Inc2

F. A. Faber and O. A. von Lilienfeld
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in *Materials Informatics: Methods, Tools and Applications*, O. Isayev, A. Tropsha, and S. Curtarolo, eds. (John Wiley & Sons, Ltd, 2019), pp. 171–179.
Group(s): von Lilienfeld / Project(s): Inc2

- F. A. Faber, A. S. Christensen, and O. A. von Lilienfeld
  Quantum Machine Learning with Response Operators in Chemical Compound Space
  in *Machine Learning for Quantum Simulations of Molecules and Materials*, K. T. Schütt, S. Chmiela, A. von Lilienfeld, A. Tkatchenko, K. Tsuda, and K.-R. Müller, eds. (*To be published*, 2020).
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arXiv:1908.06714 (2019).
Group(s): von Lilienfeld / Project(s): Inc2

P. D. Mezei and O. A. von Lilienfeld
Non-covalent quantum machine learning corrections to density functionals
arXiv:1903.09010 (2019).
Group(s): von Lilienfeld / Project(s): Inc2

Group of Oleg Yazyev

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arXiv:1911.12150 (2019).
Group(s): Yazyev / Project(s): DD6

3. Publications involving several groups

S. L. Anderson, P. G. Boyd, A. Gładysiak, T. N. Nguyen, R. G. Palgrave, D. Kubicki, L. Emsley, D. Bradshaw, M. J. Rosseinsky, B. Smit, and K. C. Stylianou
Nucleobase pairing and photodimerization in a biologically derived metal-organic framework nanoreactor
Nature Communications **10**, 1612 (2019).
Group(s): Smit, Stylianou / Project(s): DD4

O. Andreussi, N. G. Hörmann, F. Nattino, G. Fisicaro, S. Goedecker, and N. Marzari
Solvent-Aware Interfaces in Continuum Solvation
Journal of Chemical Theory and Computation **15**, 1996 (2019).
Group(s): Goedecker, Marzari / Project(s): DD1, DD3

M. Asgari, R. Semino, P. Schouwink, I. Kochetygov, O. Trukhina, J. D. Tarver, S. Bulut, S. Yang, C. M. Brown, M. Ceriotti, and W. L. Queen
An In-Situ Neutron Diffraction and DFT Study of Hydrogen Adsorption in a Sodalite-Type Metal-Organic Framework, Cu-BTTri
European Journal of Inorganic Chemistry **2019**, 1147 (2019).
Group(s): Ceriotti, Queen / Project(s): DD1, PP7

D. Beyer, S. Wang, C. A. Pignedoli, J. Melidonie, B. Yuan, C. Li, J. Wilhelm, P. Ruffieux, R. Berger, K. Müllen, R. Fasel, and X. Feng
Graphene Nanoribbons Derived from Zigzag Edge-Encased Poly(para-2,9-dibenzo[bc,kl]coronenylene) Polymer Chains
Journal of the American Chemical Society **141**, 2843 (2019).
Group(s): Fasel, Hutter, Passerone / Project(s): DD3

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Data-driven design of metal-organic frameworks for wet flue gas CO_2 capture
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Cover picture

A molecular representation of the carbon capture material Al-PMOF (iRASPA software was used for visualization) (from Berend Smit, EPFL, D&D4).

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