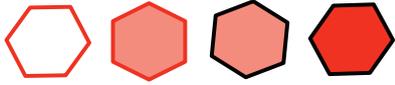


MARVEL



NATIONAL CENTRE OF COMPETENCE IN RESEARCH

**Materials' Revolution:
Computational Design and
Discovery of Novel Materials**

Progress Report
Year 4
February 2017 - January 2018

NCCR: 4th Progress Report - Cover Sheet

Title of the NCCR	Materials' Revolution: Computational Design and Discovery of Novel Materials (MARVEL)
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1 Reaction to the recommendations of the review panel

1 Response to general impressions

The general impression of the review panel was very positive. Few representative excerpts mention that “The panel continues to be impressed by the quality of the scientific output”, “a very large number of outstanding researchers”, “some exceptional, extremely original collaborations that set the agenda internationally”, “great examples of success among the younger PIs”, while mentioning also the 16 ERC awards by the team (that have now become 18). The panel also commented favorably on the actions taken in response to previous recommendations, on the organization of the site visit, on the collaborations taking place, and in general of the overall awareness and spirit present in all participants of the project. Also measures taken during phase I or proposed for phase II, such as the “Agility Plus” grants or the increased financial flexibility planned, were positively commented upon. The two recommendations coming from this general impressions paragraph were those pertaining to a clear graphical and chronological presentation of collaborative links, and to focus on a subset of target materials for the collaborative grand challenge. For the first recommendation, detailed in the final part of this response, we will indeed provide this overall presentation of the collaborations to the review panel before the site visit; for the second, detailed in the Full Proposal, we also note that we have removed two of the proposed Design and Discovery projects, to continue the focusing effort that has been consistently suggested.

2 Response to scientific progress

The project leaders respond, for the project of their competence, to the recommendations made by the review panel.

VP1, Spaldin and Yazyev

Following the recommendations of the review panel junior PI Yazyev joined the committee in charge of the communication strategy of the

NCCR. His group also accepted two INSPIRE Potentials fellowship students in order to address the issue of gender balance in the group.

VP2, Röhrlisberger and Pasquarello

The grand challenges are getting better defined as the project progresses and the collaboration between the members strengthen. In particular, the reorganization and restructuring in a limited number of research directions has allowed us to define more clearly the themes that will be pursued in phase II, which will naturally lead to a narrowing of the focus. As far as the comment concerning the competition is concerned, we are convinced that (i) the complementary expertise of the various PIs within MARVEL and (ii) the infrastructure including computer codes, machine learning protocols, and screening engines will ultimately provide an important competitive advantage. As far as the comment on the machine learning approaches, their integration within the research projects has undergone a steady increase since the start of MARVEL. This integration is bound to increase further as the project moves beyond its initial phase, which mainly involved the building of understanding and of simulation protocols.

HP3, Hutter

The review panel acknowledged the importance of HP3 as an enabler for other parts of the NCCR. It was also recommended to continue to use the dissemination paths for new developments available through the large user bases of the DFT codes. The main concern of the panel was on the lack of clear collaborations between the groups focusing on methods for strong correlation and the groups working on post-DFT methods. Although several attempts have been made to overcome this gap with targeted projects, no successful collaboration could be established as of today. Different cultures (languages) have still to meet, and it seems that a clearly defined common interest problem is still missing.

HP4, Goedecker

The investigators of HP4 appreciate that the review panel stresses the importance and expertise in the development of methods of the HP4 team. We also believe that our methodological leadership will allow us to tackle some of the most challenging problems in materials science in phase II of MARVEL. A high degree of collaboration has been reached in HP4 and will further be developed.

HP5, Curioni

It was pointed out that HP5 members need to strengthen collaborative efforts within the HP5 framework as well as within the general MARVEL community. This recommendation was adopted meticulously, resulting in several key collaborations for the development of novel machine learning schemes for the prediction of physical properties (mostly within HP5) as well as for the application of the Knowledge driven approach in other vertical projects (collaborations with VP2 and HP4). In addition, the reviewers suggested the design and implementation of illustrative demos to validate the approaches. HP5 members performed a number of such demonstrations, and in addition made available their methods/codes/services to the MARVEL community.

PP6, Schulthess and Marzari

The panel was highly positive about these activities, noting the wider adoption of AiiDA, and progress on the Materials Cloud — commenting that the *Learn* section for the latter might be the most “difficult” one to accomplish. In response to this, we developed and deployed on the Materials Cloud a “Quantum Mobile”, i.e. a virtual machine targeted first and foremost at teaching. The virtual machine can be run on any computer architecture (Windows, Mac, Linux) very straightforwardly through the use of the Virtual-Box software, and provides a Linux environment with an Ubuntu distribution, and major open-source codes compiled and ready to run, in stand alone mode or through AiiDA. The codes include CP2K (mixed Gaussians and plane waves), FLEUR (all-electron FLAPW), Quantum-ESPRESSO (plane-wave pseudopotentials), SIESTA (atomic orbitals and pseudopotentials), and Yambo (many-body perturbation theory and Bethe-Salpeter), in addition to AiiDA and different tools (torque, openmpi, xmgrace, gnuplot, xcrysden, jmol). This will be tested in classes taught by Marzari at EPFL in

the spring 2018, and a similar effort will be deployed by Smit, and used at EPFL in the same semester, for classical molecular dynamics and Monte Carlo simulations. We believe that this could represent a model to offer a high-quality, optimal environment on which to build homeworks, tutorials, and to explore novel computational tools (different codes, and AiiDA management).

PP7, Nolting, Kenzelmann and Gröning

In phase II the experimental groups will be integrated from the beginning into the Design and Discovery projects, which will enable to reinforce the feedback loops between experiment and theory/modeling, and also lead to more pronounced interactions to challenge and confront theory as well as confirm it.

3 Response to structure-related criteria

Knowledge and technology transfer

We thank the panel for the positive feedback and the recommendation regarding the need for additional resources for TT actions. In 2017, we continued our efforts to identify and contact companies with Dr. Van Landuyt, hired at 0.2 FTE for industrial liaison and TT activities. We have created an Industrial Advisory Board (IAB) that will convene at our Review and Retreat in Sep 2018 (part of our broader conference on materials’ design and discovery COMDI2018 (sites.google.com/view/comdi2018)) and a first workshop with 4 representatives from industry has been organized during the annual Review and Retreat to have a better insight on the industrial needs. The other members of the IAB will be enrolled at the beginning of the phase II. Moreover, Carey Sargent has been hired, as a journalist at 0.2 FTE, to increase significantly our visibility and our communication efforts towards industry. As part of this effort, she is preparing “Feature Stories”, accessible to non-specialists in modeling, which are disseminated via the MARVEL Newsletter to Industry and the website. A Twitter account has been launched and Carey is our community manager.

Education and training

We very much appreciate the positive feedback on educational and training activities. We are keeping up the momentum encouraging bottom-up collaborations via the junior retreat



and the junior seminars, and with the presentation of new junior projects during the 2017 Review and Retreat. Efforts towards the younger generations have been accentuated with the organizational aspects for the first 2-week summer camp starting from June 25, 2018. The next step is to materialize the MARVEL academy, that involves extended discussions with EPFL leadership, and the initiative on *Frontiers for Young Minds* (kids.frontiersin.org). The communication strategy for phase II includes measures to attract new talents to MARVEL.

Equal opportunities

We thank the panel for the positive feedback regarding the activities, which are planned to continue in phase II. Following the panel recommendations, the allowances for recipients of the INSPIRE Potentials Master's Fellowships have been raised in July from 1'600 CHF to 2'000 CHF per month (maximum allowed), plus coverage of medical insurance for extra-European students, who are not covered by their own country insurance, and any tuition fees. The panel also expressed the wish to see PIs involved in MARVEL make efforts to recruit more female PhD students and post-docs and the wish to evolve a feedback mechanism to ensure that the working climate is favorable to and aware of gender balance. The MARVEL strategy for equal opportunities in phase II will address these issues, in particular through a pilot seminar on "Diversity in Recruitment & Supervision" for MARVEL PIs led by Prof. Franciska Krings (UniL), which is currently in preparation, and a planned survey among all MARVEL members. Furthermore, additional resources allocated to MARVEL in phase II would be used to a pilot scheme where a dedicated person supports the proactive recruitment of female PhD students and post-docs in MARVEL labs.

Communications

We thank the panel for the very helpful comments. The Twitter account that was launched in Nov 2017 is now a crucial instrument to relay important information from and to the entities and people involved in materials modeling worldwide. The use of social media will be a key measure for the second phase on MARVEL, as indicated in the communication strategy. Thanks to the monthly internal newsletters, issued since last October, we have further energized the MARVEL community and network, added to the yet established opportunities to interact physically. The wider commu-

nity has also been addressed with the regular issue of "Highlight Papers" and "Feature Stories" on the website (relayed as well through other means), 5 distinguished lectures, and the launch of the scientific newsletter in Jan 2018. Many lectures, seminars and talks were filmed: they are now accessible online to an extensive audience. The COMDI2018 conference next September in Lausanne will help making our effort even more visible and will be an excellent opportunity to mix MARVEL members with worldwide researchers in materials modeling.

4 Response to pre-proposal for the next phase

The review panel "applauded the focus on grand challenges, the engagement of new PIs, as well as the competitive and more dynamic budget allocation in phase II", "fully supporting the pre-proposal for the next phase".

The panel suggested that the Bonus project on complex metal alloys (Curtin) should not be contingent to bonus funding, and indeed this is now integrated fully in the planned activities for phase II — albeit the experimental component would grow contingent to bonus funding. The panel suggested that greater resources should be given to the two Incubator projects (solid-state conductors, Laino, and machine learning, von Lilienfeld). The project on machine learning has been increased by the involvement of the new PI, Prof. Volker Roth (UniBas); the machine learning activities of Ceriotti have been preserved, but are taking place in the framework of the complex metal alloys project, to ensure a direct focus of the methodology towards applications. The Laino project has added one additional experimental counterpart (Dr. Claire Vielleville, PSI). The panel commented favorably on the mechanism set up for the identification of the Design and Discovery projects of phase II, involving the MARVEL directorate and the Scientific Advisory Board, that led to streamline the initial proposals down from 8 (including the Bonus) to 6. We believe that this represented the optimal number in the present landscape, balancing the need for focus and major collaborative effort with (as recommended by the Scientific Advisory Board) the need for diversity, and capacity to adapt. Clear metrics are provided in the full proposal and will be presented during the site visit; broadly speaking, phase I aimed at TRL (technology readiness levels) 1 – 3 (from basic principles to proof of concept), while phase II aims at TRL 3 – 6 (from experimental proof of concept to technology demon-

strated in relevant environment). More precisely, this NCCR has two objectives — a structural long-term one, that is the creation of a community and an infrastructure for the computational design and discovery of materials, and a scientific and technological one, aimed at the design and discovery of materials. For the first objective, we aim directly at TRL 6 for phase II (technology demonstrated in relevant environment, where the environment is that of a research laboratory, experimental facility, or industrial lab), and TRL 8 for phase III (system completed and qualified). For the second objective, different projects will aim at TRL 3 – 6 in phase II, and specify their metrics, specific milestones, the materials targeted, and how they are different from similar activities elsewhere. Most projects will maintain, as requested, a strong methodological component — but we feel that embedding these efforts within the Design and Discovery project will help focus and streamline these efforts, that are in any case leveraged by the fairly substantial non-MARVEL group funding of all the PIs involved, and especially by the requirements of matching the MARVEL funds 1-to-1 for all PIs involved. Guidance from industry or industrially focused academics will take place through the formation and guidance of the Industrial Advisory Board, and will be helped by the addition of Bill Curtin in the Executive Committee of MARVEL, and the elevation of Berend Smit to the position of Deputy Director.

5 Response to structural aspects and support by the home institution

The panel members “were impressed by the ongoing support for MARVEL from EPFL, CSCS and PSI, and noted that the continuation of this support to phase II was a strong endorsement of the project”. In particular, the panel underscored positively the importance and the commitment to install a new tenure-track position in phase II, in the field of computational materials science.

6 Response to final recommendations

We provide here an itemized reply for all the points presented in the final recommendations.

- A full graphical and chronological presentation of the collaborations (and resulting publications) will be provided to the panel before the site visit.
- The list of each PI’s contribution to the Centre’s activities will be updated and provided to the panel before the site visit.

- The INSPIRE Potentials Fellowships have been enhanced and expanded, and the PIs are becoming ever more aware of the importance of recruiting more female students and postdocs. Our goal is to double the number of female PhD students in MARVEL during phase II. The director has been pushing for a pilot on gender training at EPFL, to ensure that the working climate is favorable to and aware of gender balance.
- Indeed, the Software Infrastructure will remain a significant part of phase II. Methodological development has been embedded into the Design and Discovery projects, but preserving the need for development efforts. Also, the machine learning activities have been expanded.
- Each Design and Discovery project will present its metrics, and should be scored around these and the metrics of MARVEL
- The Bonus project has been integrated into the full proposal, and it is not contingent on bonus funding.
- An Industrial Advisory Board has been created, and will first meet at the Review and Retreat + COMDI2018 in September. Berend Smit has been elevated to Deputy Director, and Bill Curtin has entered the executive committee — both have extensive experience in industrial settings and in industry-focused projects.
- The expertise of Design and Discovery project 1 has been transitioned to hard materials systems, including direct inclusion of Ceriotti in the machine learning activities of the complex alloys project.
- Greater resources to the two Incubators projects have been provided.
- A senior academic in machine learning has been recruited (Prof. Volker Roth, UniBas).
- The junior retreat will perfect the mechanism of igniting research projects formulated by young researchers. Bonus funding will also be devoted to supporting MARVEL researchers that have moved to Ambizione grants and Eccellenza professorships.
- An additional 0.2 FTE has been allocated to technology transfer and communication for technology transfer. A social media strategy has become part of the communication effort, and MARVEL has now an active Twitter account.

2 Management

2.1 Structure and organisation of the NCCR

2.1.1 Structure of the NCCR

NCCR MARVEL is organized around 2 vertical projects of materials design and discovery, focusing on novel materials physics (VP1) and novel materials applications (VP2), supported by 3 horizontal projects on advanced quantum simulations (HP3), advanced sampling methods (HP4), and materials informatics (HP5), that are interfaced to 2 platform projects on the informatics infrastructure (hardware and software) (PP6) and on the experimental pipeline (PP7). EPFL is the home institution and participating scientists are affiliated with 12 Swiss academic and industrial institutions forming a strong network in the field of computational design and discovery of novel materials. MARVEL is organized around three bodies responsible of its operation, the Executive Committee, the Scientific Committee and the Scientific Advisory Board. Members (unchanged) can be found on the MARVEL [website](#).

2.1.2 Organisation of the NCCR

Two new members, William Curtin (EPFL) and Martin Jaggi (EPFL), entered MARVEL in May 2017 through the collaborative “Agility Plus” effort. Following the reallocation of the year 4 budget to projects, as presented in the Chapter 10 Finances, new projects started, one for an existing MARVEL member, Berend Smit (EPFL), to directly contribute to the Materials Cloud, and one for a new member, Mathieu Luisier (ETHZ), thanks to a collaboration with Empa. With the remaining money allocated to PP7, two small 6-month projects were launched, led by existing MARVEL members, Rüegg (PSI) and Curioni (IBM). Matthias Troyer is still on a 2-year leave in the Quantum Architectures and Computation Group of Microsoft Research, part of a major effort of Microsoft to develop quantum computing;

the activity on topological insulators continues strongly at ETHZ; for the rest of phase I Oleg Yazyev is replacing him as co-project leader in VP1, together with Nicola Spaldin. Antoine Georges directs, from Sep 2017, the new Center for Computational Quantum Physics in New York City, funded by the Simons Foundation. These two are examples of how strong the MARVEL community is, and will lead to additional important international synergies. Updates to the management team saw Monika Salas-Tesar serve as ad-interim administrative assistant from Jan to Apr 2017, with Sonia Collaud starting in March 2017 at 60%. Carey Sargent joined the management team in Nov 2017 at 20% as support to Pascale Van Landuyt for communication and events in the framework of industrial liaison and tech transfer activities. The 4 management areas have Marzari, Passerone, Curtin and Van Landuyt in KTT, Smit, Ceriotti and Jongen in education and training, Corminboeuf, Hutter, Favre-Quattropiani and Fügler in equal opportunities, and Pasquarello, Yazyev, von Lilienfeld, Favre-Quattropiani, and Jongen in communication.

2.2 Management activities and status of collaboration/integration

2.2.1 Activities and measures

Events organisation

In year 4, MARVEL organized meetings, lectures and events that are described in Chapters 5-8; we mention, e.g., here:

- MARVEL booth at the spring meeting of the Swiss Association of Computational Chemistry, Empa, Dübendorf, Feb 9, 2017.

- MARVEL booth at the *Salon des Technologies et de l'Innovation de Lausanne (STIL)*, Mar 2, 2017.
- Advanced AiiDA tutorial for MAX and MARVEL members at EPFL, Mar 6 – 7, 2017.
- MARVEL booth at the Industry Day organized by EPFL, Mar 16, 2017.
- Ig Nobel Award Tour Show on the EPFL campus, Mar 21, 2017.
- Site visit, with the review panel at EPFL, Apr 25 – 26, 2017.
- MARVEL/MaX/Psi-K tutorial on high-throughput computations: general methods and applications using AiiDA at EPFL, May 29 – 31, 2017.
- Third MARVEL junior retreat in Magliaso, Jul 3 – 7, 2017, with 48 participants.
- MARVEL booth at the Industry Day organized by ETHZ, Aug 29, 2017.
- Fourth MARVEL Review and Retreat, gathering annually all MARVEL members, Sep 7 – 8, 2017 at EPFL, with 156 participants, including 5 Scientific Advisory Board members.
- CCMX-MARVEL Materials Day 2017 at Empa, Dübendorf, Oct 5, 2017
- Second AiiDA coding week in Leukerbad, Oct 16 – 20, 2017.
- 2017 ACCES Visualization Contest launched mid-Sep until Dec 31, 2017, including the seminar by Prof. W. Craig Carter (MIT and invited professor at EPFL) on Nov 10, 2017.
- Screening of the movie "Hidden Figures" jointly with CECAM and EPFL Equal Opportunities Office, Nov 13, 2017.
- Exclusive female lunch with Mary Ann Mansigh Karlsen together with the EPFL Equal Opportunities Office and gathered 84 women, Nov 14, 2017.
- Event "Almost famous, a woman behind the codes" featuring Mary Ann Mansigh Karlsen, jointly with CECAM, Nov 15, 2017.
- "Agility Plus" workshop at EPFL, Dec 4, 2017
- INSPIRE Potentials — MARVEL Master's Fellowships for female Master students, with 2 calls in Apr and Oct 2017.
- 5 MARVEL distinguished lectures at EPFL: Prof. Markus Reiher (ETHZ), Mar 8, 2017; Prof. Annabella Selloni (Princeton), May 16, 2017; Prof. Steven G. Louie

(Univ. California, Berkeley), Jul 21, 2017; Prof. Yang Shao-Horn (MIT), Sept 11, 2017; and Prof. Chris G. Van De Walle (Univ. California, Santa Barbara), Sept 21, 2017. A lecture by Prof. Kieron Burke (Univ. California, Irvine) is already planned for Feb 20, 2018.

- 9 MARVEL junior seminars at EPFL, continuing on a regular monthly basis.
- 3 meetings with the computational group leaders and the PP7 leaders at EPFL, Mar 21 and 28, and Jul 10, 2017.
- 13 seminars at EPFL

Other activities

MARVEL members organized 15 conferences, tutorials or workshops (not counting the AiiDA tutorials), of which MARVEL sponsored 11. The Psi-k Network and the Quantum-ESPRESSO Foundation were supported through group membership fees.

Measures

The management team is finalizing a *welcome letter* for all new MARVEL members with information and requirements regarding finances, acknowledging MARVEL in publications, visual identity, gender issues, and tech transfer.

2.2.2 Status of collaboration/integration

The management team has now been reinforced with the arrival of Carey Sargent. The arrival of Curtin in the NCCR will reinforce industrially-minded activities. As it was apparent in year 3, the meetings, retreats, and reviews have built a sense of community and true collaborations with different groups that are exclusively the result of MARVEL — these will be highlighted especially in the poster session of the April site visit, but we are seeing this very clearly at many levels — from group leaders to students and postdocs, and also between different institutions, with, e.g., PSI, Empa and CSCS having developed many new links with groups at EPFL, ETHZ, and all the other Universities involved. The Sep 2018 [COMDI2018](#) will act as a focal point and summary of all the MARVEL activities of phase I. It is also worthwhile mentioning that Marzari has been elected chairman of the [Psi-k](#) network, and that the 2020 Psi-k conference will be held at EPFL, following the very successful event in 2015 in San Sebastian (Spain), with more than 1'200 participants.

3 Research

3.1 Results since the last reporting phase

Vertical Project **1**

VP1 — Novel Materials Physics

Project leaders: Nicola Spaldin (ETHZ), Oleg Yazyev (EPFL)

Participating members: Matthias Troyer (ETHZ), Nicola Spaldin (ETHZ), Antoine Georges (UniGE), Oleg Yazyev (EPFL), Ulrich Aschauer (UniBE)

1 Main goals and achievements

The correlated oxides component of VP1 has focused on three areas. Firstly, our earlier successes in the rare earth nickelates have been reinforced by the DFT+U studies of the trends in structural parameters across the nickelate series, RPA/GW calculations of the effective screened interaction, and the elaboration of a Landau theory of the coupled bond density-wave/spin density-wave orders in these materials. Second, we addressed $\text{CaVO}_3/\text{LaAlO}_3$ and $\text{CaVO}_3/\text{SrTiO}_3$ heterostructures, identifying the conditions for and the nature of a metal-insulator transition in thin-film CaVO_3 , as well as elucidating the electrostatic boundary conditions in the heterostructures. Finally, we have launched a new research direction, which is a collaboration between Aschauer, Ederer and Spaldin, on the interplay between defect chemistry, strain and functionality in complex oxides.

As far as the topological materials part of VP1 is concerned, during the last year of phase I, our efforts were mostly focusing on completing the database containing the results of high-throughput search of topological materials and integrating it into Materials Cloud. In addition, we implemented a number of new functionalities in Z2PACK and WannierTools packages, as well as several AiiDA workflows, and investi-

gated in detail recently found candidate topological materials.

2 Progress of the different efforts

2.1 The rare earth nickelates (Antoine Georges — UniGE, Nicola Spaldin — ETHZ)

The series of rare earth perovskite nickelates, $R\text{NiO}_3$, exhibit a complex phase diagram where all members except LaNiO_3 undergo a metal-insulator transition (MIT) accompanied by a structural distortion. This “breathing mode” distortion leads to alternating large and small oxygen octahedra surrounding the Ni^{3+} cations on adjacent sites. At low temperatures, a magnetically ordered antiferromagnetic (AF) phase with wave-vector $\mathbf{k} = (\frac{1}{4} \frac{1}{4} \frac{1}{4})$ (in pseudo-cubic units) occurs. For NdNiO_3 and PrNiO_3 , the MIT coincides with the AF transition, whereas for all other members of the series the AF transition occurs at lower temperatures than the MIT.

The goal of this project has been to understand the interplay between structural distortion, MIT, and magnetic order in rare earth nickelates, and to quantitatively describe trends across the series. Our work has solved the long-standing problem in the field of how to describe the electronic structure of these materials by focusing only on the low-energy e_g

states and taking electronic correlations into account [1], has revealed the key interplay between Mott and Peierls-like physics and has been verified experimentally in joint collaborations [2].

Since two-particle spectroscopies, such as neutron scattering or inelastic X-ray spectroscopy, provide crucial insight into the interplay of spin and orbital degrees of freedom in transition metal oxides, we have developed software infrastructure enabling the computation of such spectra within DFT+DMFT (and beyond). This approach has previously only been applied as proof of concept, e.g. studying the incommensurate magnetic response of Sr_2RuO_4 [18]. We intend to apply this formalism to study the $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ magnetic response in LaNiO_3 [19, 20, 21] and the bond disproportionation transition [22, 23].

As part of the MARVEL effort for code development, we have contributed to the development and testing of two-particle Green's function functionality in the open-source software stack using the TRIQS library [24]. This development effort is an international collaboration with O. Parcollet at IPHT-CEA, Paris. The main developments are listed hereafter.

(i) A versatile set of two-particle Green's function samplers in the continuous-time quantum Monte Carlo impurity solver TRIQS/CTHYB has been implemented and tested.

(ii) A draft implementation of a Bethe-Salpeter equation [25] solver is tested now on a simple model. The solver is intended to be turned into an official TRIQS application once it has passed community review.

In addition, the methods development that we made in the general framework for combined density functional theory + dynamical mean-field theory (DFT+DMFT) calculations are all available within the TRIQS/DFTTools library [3].

2.2 DFT+DMFT study of strain and interface effects in d^1 and d^2 t_{2g} perovskites (Nicola Spaldin — ETHZ)

Our DFT+DMFT calculations have revealed that free-standing unstrained CaVO_3 slabs become insulating only in the ultrathin limit of less than four unit cells. The metal-insulator transition in this case can be understood to originate from a very strong surface-induced splitting between the t_{2g} orbitals, leading to strong orbital polarization and favoring the insulating state. Our calculations for heterostructures indicate that no charge transfer is observed at the $\text{CaVO}_3\text{-SrTiO}_3$ surface,

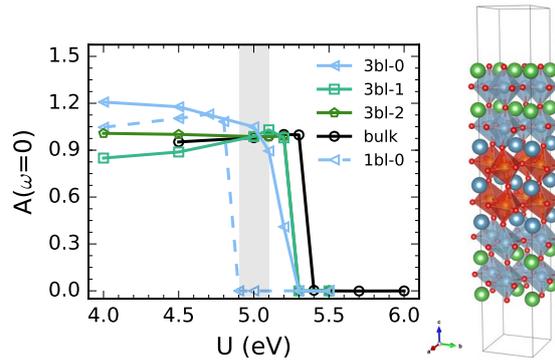


Figure 1: Right: unit cell of a symmetric stoichiometric slab containing 1 bilayer (1bl) of CaVO_3 sandwiched between 2 layers of LaAlO_3 on both sides and separated by vacuum. Left: spectral weight around the Fermi level for bulk CaVO_3 , for the 1bl slab, and for the three different layers of a 3 bilayer (3bl) slab of CaVO_3 between LaAlO_3 as function of Hubbard U . Different layers are numbered according to their distance from the interface layer. It can be seen that the critical U for the Mott transition is reduced in the 1bl slab compared to bulk CaVO_3 , whereas the 3bl slab is already nearly bulk-like. The shading indicates a realistic U value of 5 eV where the 1bl slab is insulating while bulk and 3bl slab are metallic.

whereas rather strong effects can occur at the polar $\text{CaVO}_3\text{-LaAlO}_3$ interface, depending on whether stoichiometric or non-stoichiometric slabs are used in the calculations (Fig. 1).

2.3 Stoichiometry as a materials design parameter (Nicola Spaldin — ETHZ, Ulrich Aschauer — UniBE)

Oxygen vacancies are a common defect in perovskite oxides, where they play a key role in a variety of emergent phenomena, such as superconductivity, the two-dimensional electron gas, metal-insulator transitions (MIT), and magnetoresistance. Moreover, many of the promising functionalities that are being studied in the field of complex oxide thin films and heterostructures are in many cases triggered by the presence of vacancies [26]. Many of these systems fall under the category of strongly correlated: they are Mott insulators, or live close to a Mott MIT. The presence of oxygen vacancies both creates states which are highly localized and releases carriers to the system, suggesting a need for an appropriate beyond-DFT treatment for the vacancy states as well as for the host material.

In this project, we have begun to explore the well-established density functional theory with dynamical mean-field theory (DFT+DMFT) framework to investigate the ef-

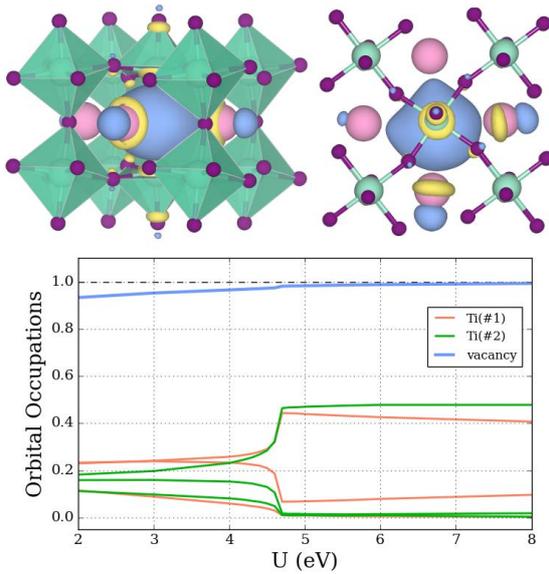


Figure 2: Top: LaTiO_3 with an oxygen vacancy at the center, together with the Wannier function of the vacancy state (blue and yellow lobes) from a planar (left) and an axial (right) perspective. Bottom: DMFT occupations for the $\text{Ti-}t_{2g}$ (green and orange lines) and the vacancy (blue line) orbitals as a function of the Hubbard U . Despite the MIT displayed by the $\text{Ti-}t_{2g}$ around 4.6 eV, the vacancy orbital stays close to full occupation.

fect of oxygen vacancies on several titanate perovskites, such as LaTiO_3 , a prototypical Mott insulator close to the MIT [27]. This will be a focus direction in the second phase of MARVEL. To date, we have focussed on correlation effects within the electronic states created by the oxygen vacancies, and the interplay between the release of carriers and the Mott insulating state. We use standard DFT codes, such as VASP, to relax and calculate structures and WANNIER90, to express the correlated subspace, i.e. the $\text{Ti-}t_{2g}$ bands, as maximally localized Wannier functions. Then we solve the DMFT problem with the tools and libraries provided by TRIQS/DFTTools [24][3]. Our preliminary results (Fig. 2) suggest that for a vacancy concentration of $\sim 8\%$, the electrons released by a single vacancy form a band around 1 eV below the t_{2g} manifold. This band stays doubly occupied for the whole range of Hubbard U explored on both the Ti-site and the vacancy-site, and has almost no effect on the MIT that is still observed when the U is increased beyond ~ 4.6 eV.

In parallel to this methodological development, we have started implementing AiiDA workflows for the DFPT linear-response determination of U in cells with defects, in order to streamline the calculations at the DFT+ U level. Our approach is to determine inequiv-

alent Hubbard sites with respect to the defect and to compute U for each of them using a double loop. The inner loop computes U in a U -in U -out fashion until convergence of U , starting from a pure GGA ($U = 0$ eV) calculation. The outer loop performs this series of U calculations before each structure relaxation, which is required to obtain U values that depend on the altered geometry around the defect.

2.4 High-throughput search of novel topological materials (Oleg Yazyev — EPFL)

The primary goal of the project is the discovery of novel materials realizing topological electronic phases, such as topological insulators, Dirac and Weyl semimetals. This objective is achieved by means of first-principles high-throughput screening of databases of known compounds. Promising candidate materials are subject to in-depth theoretical investigation and attempted for experimental realization.

In the final year of phase I, our research has largely focused on completing the database of high-throughput calculations for the search of novel topological materials. At present, the database contains the results of calculations on 16'705 materials produced by our high-throughput computation workflows. The data include basic information such as the crystal structure, chemical composition and point group symmetry, a band structure plotted over high-symmetry k -point path, Z2PACK analysis [4], and indicators of various topological phases. Promising candidates are classified and collected into a smaller database. Currently, the database is installed locally in the Yazyev group, and made available to collaborators. The web-based frontend with advanced search capabilities (Fig. 3) allows exploring the data in different ways.

Thanks to the “Agility Plus” grant and close collaborations with the group of Marzari, our high-throughput screening workflow has been implemented within AiiDA and the data is in process to be made available via Materials Cloud. Initially, we have planed sharing a curated dataset of known topological materials collected by the community on the website www.topologicalmaterials.org created by one of the group members. This curated dataset will help developing the capabilities of Materials Cloud and obtaining feedback from its users worldwide. The entire set of data will be made publicly available shortly after.

The high-throughput screening of topological materials resulted in a large number of can-

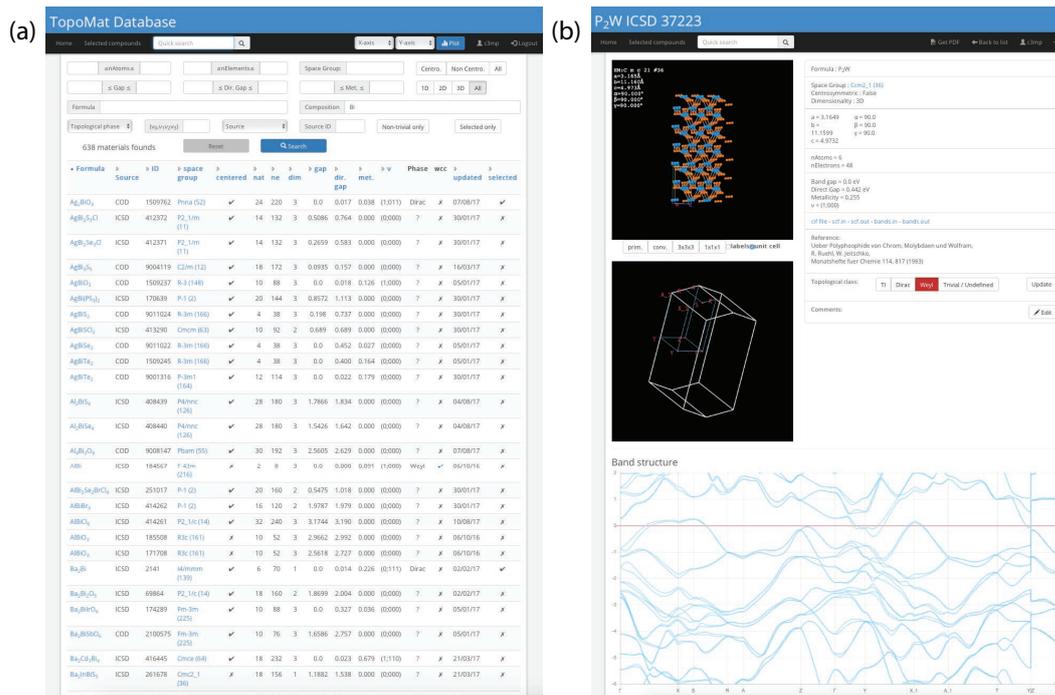


Figure 3: (a) Web interface of the search engine of the topological materials database. In this case, the filter for showing only Bi-containing materials is applied. (b) Example of a detailed record, in this case showing the newly discovered type-II Weyl semimetal WP₂.

didates that were subject to a detailed theoretical investigation and attempted for experimental realization. In this respect, our work evolved along the following two lines. Firstly, we continued investigations of the confirmed materials, in close collaboration with experimentalists. For the quasi-1D topological insulator β -Bi₄I₄ discovered by us [5], the transport properties and emergent superconducting phase under pressure were addressed [6]. Other structurally related materials belonging to the family of bismuth halides are being investigated. The robust Weyl semimetals MoP₂ and WP₂ discovered by us [7] have recently been synthesized in the Max Planck Institute for Chemical Physics of Solids directed Claudia Felser and showed a number of exceptional properties, such as the largest ever measured magnetoresistances [8], hydrodynamic fluid transport regime [28] and negative resistances [29]. We are extensively involved in collaborations addressing the properties of these “rising star” materials, e.g. the ARPES measurements performed by the group of Ming Shi at PSI. Secondly, a very large number of candidate materials are in the early stage of investigation. Among them is the predicted broad family of topological materials with composition AM_4Pn_3 (A = alkali metal; M = Zn, Cd; Pn = P, As), for which we initiated another collaboration with the institute of Claudia Felser.

One can also mention K₂Sn₂Se₄ as a minimum model Weyl semimetal, and others. Detailed investigations of candidate topological materials make ideal subjects for the MARVEL INSPIRE Potentials fellowships Master’s projects.

2.5 Novel aspects of topology and explanation of transport observations in topological devices (Matthias Troyer — ETHZ)

Within this year we concentrated on developing public numerical software for identifying topological materials, and on analysis of experimental data in the field that attracted a lot of attention, but had no theoretical explanation. In particular, we developed and published Z2PACK and WannierTools packages [9, 10] that allow even a complete novice in the field of topological materials identify candidate compounds and make predictions for ARPES and STM experiments (Fig. 4). Among novel functionalities of the WannierTools package, we would like to stress calculations of magnetotransport properties using the Boltzmann transport theory and the calculation of Landau levels in 2D and bulk materials.

We also developed theoretical basis for explaining strange magnetoresistive behavior of a family of centrosymmetric paramagnetic compounds [4]. We provided a robust theoretical and numerical explanation of the data ob-



Z2Pack



Figure 4: The Z2PACK and WannierTools [9, 10] codes allow non-experts to study topological features in materials.

served in most experiments on Majorana wires. In particular, it was observed that contrary to previous understanding, the g -factors of semiconducting wires are much larger than the bulk values [11]. We proved and numerically illustrated that this occurs due to orbital contribution, similar to the one occurring in the textbook example of Hydrogen atom [11]. We also made theoretical and numerical studies of InAs/GaSb quantum wells to provide help to the group of Profs. Ihn and Ensslin at ETHZ in establishing a robust device exhibiting the quantum spin Hall effect [12, 13]. In collaboration with the STM group of Prof. Wu at Rutgers University (USA), we made the quasiparticle interference study of the surface states of type-II Weyl semimetal WTe₂ [14]. We finally predicted novel topological phenomena in HfC [15], PdTe₂ [16], and TiB₂ (ZrB₂) compounds [17].

3 Contribution to overall goals and initial proposal

The topological materials component identified novel topological electronic phases (type-II Weyl semimetals, nodal-chain and triple point topological metals), candidate materials realizing topological insulator phases (β -Bi₄I₄), as well as new type-II Weyl semimetals MoP₂ and WP₂, with a major impact in the community. The goal of high-throughput classification is also largely attained by now. The methodological developments resulted in two public software packages, Z2PACK and WannierTools, that in turn powered some of the above efforts. The correlated oxides component of VP1 has contributed directly to the overall goal of the NCCR of designing new correlated materials systems with specific targeted functionalities by providing fundamental understanding of the interplay between strain, defect chemistry and functionality in complex oxides and their heterostructures. It has contributed to the methodological goals in our develop-

ment of methodologies for calculation of two-particle spectroscopies, our extension of the DFT+DMFT method to the description of vacancy point defects, and our development of AiiDA workflows for defect calculations.

4 Collaborative and interdisciplinary components

In the past year the transition-metal oxides work has involved collaborations between the groups of Georges, Ederer and Spaldin (DFT+DMFT), and Aschauer, Ederer and Spaldin (defects) within VP1, as well as with multiple experimental groups (Medarde, van der Marel, Schmitt, Rüegg, Rossell) within the PP7 project. The core activity of the topological materials project was carried out by the groups of Troyer and Yazyev (VP1) with the extensive involvement of the experimental group of Ming Shi at PSI (PP7) and a number of external partners.

MARVEL-related publications

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Vertical Project 2

VP2 — Novel Materials Applications

Project leaders: Ursula Röthlisberger (EPFL), Alfredo Pasquarello (EPFL)

Participating members: Ursula Röthlisberger (EPFL), Alfredo Pasquarello (EPFL), Jürg Hutter (UZH), Nicola Marzari (EPFL), Alessandro Curioni (IBM), Clémence Corminboeuf (EPFL), Daniele Passerone (Empa), William Curtin (EPFL)

Summary and highlights The restructuring of the VP2 project introduced in year 3 proved successful and is maintained in year 4. The current focus of VP2 is on four grand-challenge problems, which consist in identifying optimal materials for solar cells, for water-splitting catalyst, for solid-state ionic conductors, and for identifying and screening 2D materials. VP2 has been successful in bringing the computational PIs to collaborate in a synergetic way and to join forces with experimental teams with the ultimate goal of attaining common objectives. The various subprojects are now entering a more consolidated stage in which a material search can be envisaged, entailing a growing attention to machine learning techniques and to the AiiDA platform. As most of the research activities started at the beginning of the funding period are now producing their most significant results, an important scientific output in terms of publications is recorded for year 4. The current shape of VP2 is also ideal for envisaging the continuation of the research activity in phase II.

Project overview

In year 3, VP2 has undergone significant reorganization and restructuring with the goal of focusing the effort more effectively. This has led to the identification of four grand challenge themes, which can in short be described as: (1) materials for solar cells; (2) catalysts for water splitting; (3) solid-state ionic conductors; (4) low-dimensional materials. While subprojects 1 to 3 focus on the search of materials with a specific functionality, subproject 4 offers a catalogue of 2D materials that could potentially target different functionalities, as evidenced by the transversal collaborations that this subproject has generated with other research activities both in VP1 and VP2.

This reorganization has proved effective as it has been instrumental, on the one hand, to bring various PIs to collaborate organically in the attempt of attaining common objectives as demonstrated by the increasing number of research activities involving more than one PI, and, on the other hand, to focus the research activities on well defined goals in view of the restructuring of the MARVEL project in phase II. It has therefore not been deemed necessary to further evolve the organization of the VP2 project. The only modification in year 4 consists of the integration within VP2 of subproject 5 led by William Curtin, which represents the first extension of MARVEL efforts into structural metals and offers an interesting perspective for phase II.

VP2 truly lies at the heart of MARVEL and re-

lies heavily on the global infrastructure that has been set up. Indeed, as described in detail in the following, the VP2 subprojects often rely on methods developed in HP3, on sampling techniques proposed in HP4, on machine learning protocols defined in HP5, and/or on the AiiDA platform supported in PP6. As scientific data are being produced, we can already anticipate that VP2 is going to make an extensive use of the Materials Cloud facility, as an important resource both for data preservation and for making such data available to the internal and external community. Furthermore, it is worth to mention that all subprojects have developed ties with experimental teams. These contacts not only challenge the computational PIs to account for actual measurements, but also provide access to valuable expertise in guiding the search efforts.

The global scientific output of VP2 is outstanding, and year 4 is no exception. As phase I is coming to an end, almost all subprojects are now publishing the most significant results achieved over this funding period. Year 4 has therefore produced a particularly rich output with 50 publications, a large fraction of which in high-impact journals. A second major objective that has been pursued during year 4 is the proper formatting of the projects in view of phase II. This has led to the definition of more compact and focused research activities involving jointly several computational PIs and experimental researchers. This formatting has led to the proposal of several grand-challenge

projects issued from VP2 in view of the competitive selection of the projects for phase II of MARVEL.

1 Novel materials for solar cells

Ursula Röthlisberger — EPFL, Alfredo Pasquarello — EPFL, Jürg Hutter — UZH, Michele Ceriotti (HP5) — EPFL, Stefan Goedecker (HP4) — UniBas, Michele Parrinello (HP4) — USI, Anatole von Lilienfeld (HP5) — UniBas, Expt.: Michael Grätzel — EPFL, Jürg Osterwalder — UZH

1.1 Goals and achievements

The overall goal of this project is finding new routes for the improvement of existing solar cell materials [1] and the design of new ones using the tools of computational materials science. During the last year, we have mainly investigated electronic and optical properties of halide perovskites, that are very promising materials for solar cells, and their interface with electron transporting materials such as TiO₂. In addition, some promising candidates for new dye-sensitized solar cells were also explored.

a) *Dye-sensitized solar cells* Co-Pyrphyrin (CoPy) has been recently synthesized and successfully used as homogeneous water reduction catalyst. The **Hutter** group has investigated the adsorption of CoPy on the rutile(110) surface [2]. Apart from CoPy/oxide interactions, CoPy interactions with a metal surface are also investigated by adsorbing CoPy on a reconstructed Au(111) surface [3]. The **Hutter** group further investigated [4] the effect of the herringbone reconstruction on Au(111) on the adsorption strength and on the modification of the molecular properties. Insertion of Co atom in Pyrphyrin stabilizes adsorption. Formation of a monolayer and the geometrical configuration of the molecular assembly are mainly driven by molecule/molecule interactions. The **Hutter** group investigates the assembly and metalation process by means of *ab initio* molecular dynamics and shows the presence of intermediate states that hinder the process. Moreover, re-arrangements within the monolayer are observed upon metalation, which are in agreement with experimental evidence. In a joint experimental and computational effort with the **Osterwalder** group [5], the **Hutter** group has determined the local adsorption geometry at low coverage, the long-range molecular ordering at higher coverage and the electronic structure for both the bare ligand and the cobalt-metalated Py molecule on TiO₂. The

energy level alignment of CoPy/TiO₂ supports electron injection into TiO₂ upon photoexcitation of the CoPy complex and thus renders it a potential sensitizer dye. Importantly, Co incorporation is found to stabilize the Py molecule against photo-induced degradation, while the bare ligand is decomposed rapidly under continuous UV irradiation.

b) *Halide perovskites* In a collaboration between the **Pasquarello** and the **Röthlisberger** groups, different levels of theory were tested to calculate electronic and optical properties of halide perovskites (general formula ABX₃) including density functional theory with different exchange-correlation functionals and many-body perturbation theory. Using these methodologies, the electronic properties of different inorganic perovskites were calculated taking also spin-orbit coupling and temperature effects into account [6].

Furthermore, in an effort to understand the origin of the slow recombination of photo-generated carriers in CH₃NH₃PbI₃, the evolutions of extra carriers were simulated by the **Pasquarello** group through hybrid-functional molecular dynamics [7]. The carriers were found to localize as polaronic states and to hop on a sub-picosecond time scale. The localization is mainly induced by thermal vibrations in the inorganic sublattice rather than by the orientation of the organic cations. Electrons and holes localize in spatially distinct regions slowing down the radiative bimolecular recombination. The separate polaronic localization of electrons and holes emerges as the key feature for achieving exceptional photovoltaic properties.

The extensive collaboration between the **Röthlisberger** and **Grätzel** groups has continued and achieved several significant results. It was possible to rationalize the temperature dependence of the photoluminescence spectra of MAPbI₃ [8], to explain *J* – *V* hysteresis in MAPbI₃ [9], to suggest new routes for the band gap tuning in halide perovskites [10], and to characterize the dependence of the effective mass on chemical composition [11]. To improve the phase stability of perovskite materials, it was proposed to mix FAPbI₃ with CsPbI₃ [12] and dope FAPbI₃ by MA, Cs, and Rb cations (Fig. 1) [13]. In addition, the band gap evolution in FAPb_xSn_{1-x}I₃ and FAPb_xSn_{1-x}Br₃ mixtures was explained and the band structure and absorption spectra of CsPbBr_{3-x}Cl_x systems were determined.

c) *Phase discovery and phase diagrams of halide perovskites* The **Röthlisberger** group works

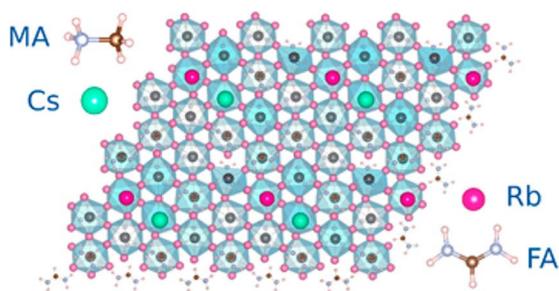


Figure 1: Perovskite phase of FAPbI_3 doped by MA, Cs and Rb cations.

in collaboration with the groups of **Goedecker** (HP4) and **Ceriotti** (HP5) to characterize different crystalline forms of halide perovskites via minima hopping simulations and sketch-map analysis (this project is detailed in the reporting for HP4).

d) *Crystal growth and nucleation* In collaboration with the **Parrinello** group (HP4), the **Röthlisberger** group is applying force field based molecular dynamics simulations together with variational enhanced sampling techniques to study crystal growth and nucleation processes of lead halide perovskites (this project is detailed in the reporting for HP4).

e) *Development and applications of an evolutionary algorithm toolbox for chemical compound and materials design* Together with the group of **von Lilienfeld** (HP5), the **Röthlisberger** group is developing combinations of genetic algorithm optimisations and machine learning models (this project is detailed in the reporting for HP4). The **Pasquarello** group is also involved in this project and is expected to generate high-level (beyond DFT) reference data for band gaps of metal halide perovskites, as validation of the training data used for the machine learning.

1.2 Current and future progress

Our current research is based on establishing the factors that govern stability and performance of perovskite solar cells. Currently, we are investigating electronic properties of $\text{FAPb}_x\text{Sn}_{1-x}\text{I}_3$ and $\text{FAPb}_x\text{Sn}_{1-x}\text{Br}_3$ mixtures and these studies are close to submission. In the future, we plan to investigate new halide perovskite structures obtained by modulating A, B, and X sites and to calculate their electronic and excited state properties. Additionally, we plan to look for possible alternatives for lead-free perovskite materials.

In a second research line, we are investigating possible interfaces between MAPbI_3 and TiO_2 .

We created the interface structures for different possible mutual arrangements of these two materials and calculations of the electronic properties of the interfaces are ongoing. In the future, we plan to broaden our study to different halide perovskites and oxides. Additionally, we plan to investigate how the treatment of the interface in terms of passivation and atom substitution affects solar cell performance.

2 Superior electrocatalysts for the oxygen evolution reaction

Clémence Corminboeuf — EPFL, *Jürg Hutter* — UZH, *Nicola Marzari* — EPFL, *Alfredo Pasquarello* — EPFL, *Expt.: Xile Hu* — EPFL, *Greta Patzke* — UZH, *Grigory Smolentsev* — PSI, *Raffaella Buonsanti* — EPFL

The two principal research objectives have been reinforced. They concern (i) exploiting tools and design principles to screen superior molecular and solid-state electrocatalysts for the oxygen evolution reaction (2.1) and (ii) achieving an accurate electronic structure description of the band alignment at semiconductor-water interfaces (2.2). In addition, close interactions with the SCCER on Heat and Electricity Storage, as postulated in Article 10 of the MARVEL contract, have taken place with the group of Prof. T. J. Schmidt in the context of catalysis and water splitting, and are detailed in the relevant section of PP7.

2.1 Tools and design principles

The efficiency of current state-of-the-art solid-state catalysts for the oxygen evolution reaction (OER) is limited by a minimum overpotential of 0.4 eV, which has been identified from the linear free energy scaling relations (LFESR). This subproject aims at establishing efficient strategies that can be used to identify superior electrocatalysts for the OER. Our efforts have focused on two approaches: a) an alternative *bifunctional pathway*, which opens the door to lower overpotentials and b) *molecular catalysts*, which can more easily be modified and tuned than solid-state catalysts to maximize the thermodynamic drive of each reaction step.

a) *The bifunctional pathway* Recently, the **Corminboeuf** and **Hu** groups exploited an alternative bifunctional mechanism involving two materials: the standard catalyst as well as a catalytically active hydrogen acceptor support material [14]. The novel bifunctional mechanism was cast into a revised 3D volcano plot

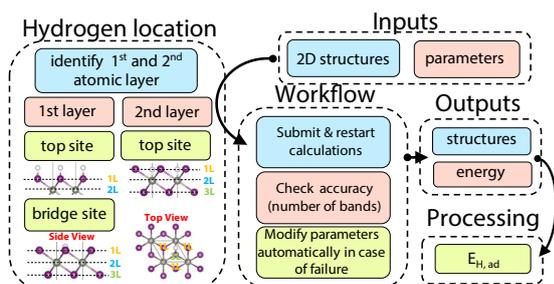


Figure 2: The overall AiiDA workflow for the automated high-throughput screening of ΔG_H

that featured two descriptors used to characterize each material component (i.e. ΔG_H and $\Delta G_O - \Delta G_{OH}$). In year 4, the **Corminboeuf** and **Marzari** groups exploited the same ΔG_H (the free energy of adsorption atomic hydrogen) descriptor for screening materials that might serve as hydrogen acceptor candidates. Preliminary focus was placed on examining a database of exfoliated 2D layered structure constructed by the **Marzari** group [15]. The screening procedure involved locating the most stable hydrogen binding sites and identifying the most suitable binding energies (1.2 eV) using an automated AiiDA workflow (Fig. 2). To date, 267 non-magnetic and 56 magnetic structures out of the 1844 2D exfoliable materials in the database have been screened. Among them, only 5% are characterized as having attractive binding energy and only 2% (i.e. 6 2D structures) are potentially stable hydrogen acceptor candidates. While the screening is still ongoing, the most promising candidates are planned to be experimentally characterized by the **Hu** group.

b) *Molecular catalysts* As an alternative to the bifunctional pathway, the **Hutter** and **Corminboeuf** groups are exploring another strategy to generate superior OER electrocatalysts. This effort also involves the experimental teams of **Patzke** at UZH and of **Smolentsev** at PSI. Because they can be fine tuned, molecular catalysts are ideally suited to challenge the robustness of the universal linear scaling relationships. Considering the distinct nature of homogeneous catalysts compared to solid-state systems, the applicability of the LFESR generated for solids may not be valid for molecules. Within MARVEL, the design and the synthesis of efficient molecular water oxidation catalysts [16, 17, 18] and the extensive examination of the influence of ligands and their environment on the catalytic activity [19, 20, 21, 22] have been achieved and serve as motivation for our current research focus. Inspired by

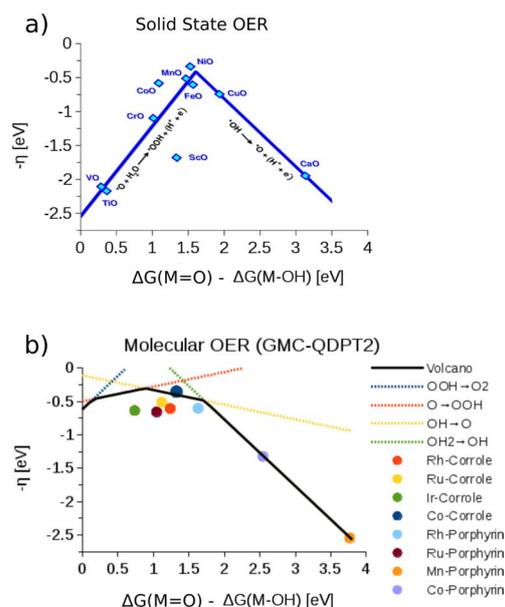


Figure 3: Comparison between (a) the volcano plot for standard solid-state catalysts and (b) the proposed molecular volcano plot based on the GMC-QDPT2 LFESR. For the scaling between *OH and *OOH , a slope of 1 was assumed.

these examples, an extended set of transition metal catalysts coordinated by corrole and porphyrin ligands was constructed prior to validating the robustness of scaling relations using various density functionals and a multireference GMC-PT2 method. Accordingly, we identified a contrast between the solid state and molecular volcano plots (Fig. 3) that results in a different shaped volcano plot displaying a broad plateau at the top with a slightly lower overpotential of 0.3 eV. Owing to the broadness of the plateau, a larger number of catalysts should show good performance, although further improvement of the reaction thermodynamics through alternative reaction mechanisms will not be possible [23].

From a methodological perspective, the LFESR for molecular catalysts strongly depend on the underlying methods used to create them. The problem is principally rooted in the complex electronic structure of the metal-doped porphyrins and corroles as well as of other Co-based water oxidation catalysts. More generally, the performance of each density functional depends on its ability to simultaneously minimize the delocalization error and mimic the effects of static electron correlation. Comparisons with benchmark GMC-QDPT2 results generally show a poor performance for density functionals, with the exception of low exact-exchange hybrids. Ongoing work from the **Hutter** and **Corminboeuf** groups focusing on



more accurate electronic structure frameworks (e.g. full configuration quantum Monte Carlo with **A. Alavi**, MPI Stuttgart, DMRG with **M. Reiher**, ETHZ) are currently underway.

In a related context, the concept of molecular volcanoes introduced by the **Corminboeuf** group in years 2 and 3 [24, 25] has been further expanded and generalized through new developments [26, 27, 28]. Excitingly, these efforts are currently being exploited in a joint “Agility Plus” project between the **von Lilienfeld** (HP5) and **Corminboeuf** groups. This project aims at developing machine learning models that can rapidly screen thousands of prospective species and identify the most suitable catalyst for a given chemical reaction.

2.2 Accurate electronic structure description

The **Pasquarello** group achieved one of the main objectives of MARVEL phase I, which consisted in assessing the accuracy by which the band alignments at semiconductor-water interfaces can be determined [29]. Molecular dynamics simulations of atomistic interface models and electronic structure calculations at the hybrid-functional and GW level were combined. The study comprised GaAs, GaP, GaN, CdS, ZnO, SnO₂, rutile TiO₂, and anatase TiO₂. The most accurate results within a fully *ab initio* scheme were obtained with one-shot GW calculations based on hybrid-functional starting points leading to a mean average error of ~ 0.25 eV. The molecular or dissociated nature of the adsorbed water molecules is shown to be critical.

As a first step towards the modeling of catalytic water-splitting processes, the **Pasquarello** group developed a theoretical scheme for studying the pH-dependent coverage of the semiconductor surface in aqueous environment through advanced electronic structure calculations, molecular dynamics simulations, and the thermodynamic integration method [30]. Application to the BiVO₄(010)-water interface yields excellent agreement with experiment for the pH at the point of zero charge and enables the construction of a diagram for the adsorbed species at the interface as a function of the pH (Fig. 4). Furthermore, to model electrochemical half-reactions at electrodes with varying potential, the **Pasquarello** group has implemented a molecular dynamics setup to control the Fermi level as an external variable. This technique has been successfully benchmarked to determine redox potentials of aqueous species [31] and to control the generation of defects in

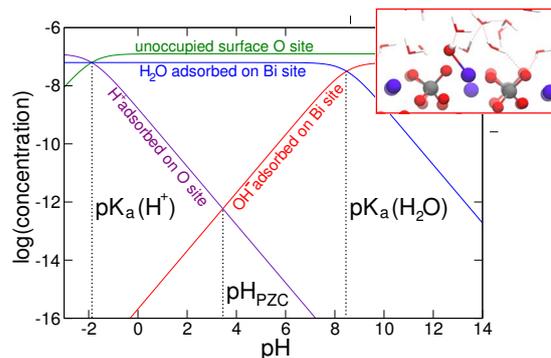


Figure 4: *Ab initio* diagram for the coverage of the BiVO₄(010) surface vs pH in aqueous environment.

semiconductors [32, 33].

In close interaction with the **Buonsanti** group (PP7), the **Pasquarello** group focused on BiVO₄ as a benchmark photoanode and studied its electronic structure through quasiparticle self-consistent GW (QSGW) calculations [34]. Spin-orbit coupling, electron-hole interaction, nuclear quantum motions, and thermal vibrations were accounted for. In this way, the optical band gap and the absorption spectrum were found in good agreement with experimental data. The next step involves paramagnetic Cu₂V₂O₇, which will enable direct comparisons with the experimental work currently pursued in the **Buonsanti** group.

3 A search for solid-state ionic conductors

Alessandro Curioni — IBM, *Nicola Marzari* — EPFL, *Expt.:* *Daniele Pergolesi* — PSI, *Thomas Lippert* — PSI

3.1 Scientific goals

Safety challenges in rechargeable Li-ion batteries inhibit the employment of mobile applications [42]. The electrolyte is a critical component to improve the safety of a battery [43, 44], and replacing the organic counterpart with solid-state electrolytes (SSE) offers an attractive option [45, 46]. However, no electrochemically stable structures with a sufficient ionic conductivity have been found [47]. This project aims at deepening our understanding of the underlying mechanisms of superionic conduction to develop a robust framework for the screening of SSEs candidates.

3.2 Recent results

The work during the reporting period first led to the finalization of the *pinball model* (**Marzari** group), which will serve as a first screening

step for the platform of phase II. Second, it enabled progress in the development of *polarizable force fields* (IBM group), which will be used as a second screening criterion. Third, both the IBM and the Marzari group started assembling the first part of the phase II *screening platform* for ionic conductors, leading to the first screening process based on the pinball model. Finally, the Marzari group also finalized two studies on *first-principles simulations*, which have improved our understanding of the role of ion-ion correlations and strain in ionic transport.

The pinball model has as underlying assumption that the lithium ions move in an energy landscape defined by a frozen charge density, with the host lattice constrained to equilibrium. Due to the introduction of four empirical parameters to account for the electrostatic screening of the different contributions, the Marzari group achieved excellent agreement between simulations in the model and self-consistent calculations. During the reporting period, a more efficient algorithm was developed to calculate the four parameters. In this scheme, one relies on the generation of configurations through the use of random displacements and on a force-matching technique to regress the model forces to self-consistently calculated values. While the pinball model at this level of approximation is accurate enough for screening applications, the hope is to gain in accuracy by removing the constraint on the host lattice. Implementations of harmonic vibrations of the host lattice and updates of the charge density from linear-response theory were carried out. A first working scheme of this Hamiltonian framework is currently being tested extensively, with the goal of producing stable dynamics for realistic systems in the near future.

Regarding polarizable force fields, the IBM group can now parametrize them efficiently from first-principles and empirical data. The charge splitting and force constant of the polarizable Dick-Overhauser model are determined using the polarizability of an isolated atom surrounded by point charges. The remaining parameters of the Buckingham potentials are fitted to reproduce first-principles forces of selected structures. The fitting procedure is necessary since the data found in the literature are generally not transferable. Understanding the dependency of the resulting force field on the various parameters is therefore of paramount importance for its use as a screening tool. The IBM group showed that the polarizability has a large impact on the measured properties. Through the use of fitted potentials,

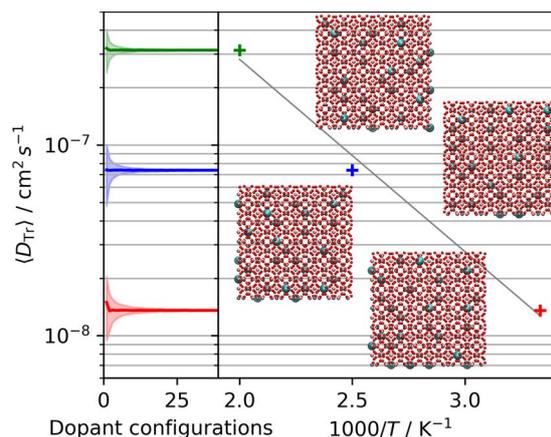


Figure 5: Left: diffusion coefficient and error estimate as a function of the number of dopant configurations used for the averaging of the diffusion coefficient at 300 K (red), 400 K (blue) and 500 K (green). Right: Arrhenius plot of the converged diffusion coefficients with four examples of the dopant configurations used.

the effect of doping on the diffusive behavior of lithium ions in tungsten doped materials of the garnet class was investigated. Besides stabilizing the more conductive cubic phase, the dopant atoms were found to act as barriers. The use of explicit doping lowers the conductivity with respect to an implicit model, due to local changes in polarization (Fig. 5). An important dependence of the diffusion coefficient on the spatial arrangement of the doping agents is also observed and, consequently, the IBM group introduced an averaging procedure over several doping arrangements. In order to finalize the conclusions, investigations of finite size effects are in progress. As for the comparison with experiment, the simulated conductivity decreases linearly with carrier concentration, whereas the experimental one shows an increase in conductivity at low concentration. The differences between simulations and measurements could be explained by thermodynamical and interfacial effects, the experiments being performed on granular samples. It is also not yet possible to rule out that the model is not sufficiently accurate to reproduce all diffusive processes in the crystals.

The Marzari group interfaced the initial steps of the high-throughput screening envisioned for phase II with the AiiDA materials' informatics platform [35]. The first step relies on tools for automatically importing structures from external repositories and selecting suitable candidates in terms of geometry, composition, and bond types. The next step comprises a variable-cell relaxation and calculation of band-gaps from first principles. Recent ef-

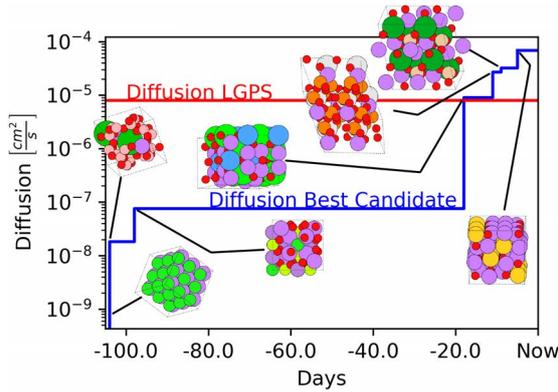
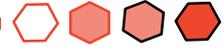


Figure 6: The screening progress is shown in blue as an increase of the best diffusion coefficient (at 1000 K) in the pinball model over time for the set of candidates (best respective candidate in insets). The red curve shows our estimate for $\text{Li}_{10}\text{GeP}_2\text{S}_{24}$, one of the best ionic conductors currently known.

forts were targeted on the efficient fitting of the pinball parameters and the automatized calculation of diffusion coefficients from molecular dynamics in the pinball model with a frozen host lattice using a custom workflow to converge transport coefficients to a pre-defined tolerance. The screening started with all Li-containing structures from the COD and ICSD repositories (1362 suitable unique candidates), resulting in 973 electronic insulators that have undergone a successful variable-cell relaxation. These structures are currently processed in the third stage of the screening, the dynamics in the pinball model. The candidate pool for the next stages of the screening is continuously updated (Fig. 6).

First-principles analyses were performed in order to analyze selected candidates or materials of broad interest to the field. The **Marzari** group finalized studies on the ion-ion correlation effects in $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ [36] and on its oxide analogue $\text{Li}_{10}\text{GeP}_2\text{O}_{12}$. Within MARVEL, in collaboration with the experimental teams of **Pergolesi** and **Lippert**, strain and proton trapping effects in BaZrO_3 were investigated and finalized through a publication [37]. Currently, Raman spectra and Born effective charges are being computed for members of the thio-lisicon family. An analysis of the potential energy surfaces for lithium ions using persistent-homology is in progress, in collaboration with the **Smit** group (HP4).

3.3 Future work

The **Marzari** group will focus first and foremost on the first-principles screening and experimental realization of the many new can-

didates being suggested by the pinball model. In addition, it will continue the development of the pinball model in its next level of approximation (releasing the fixed host lattice condition) using the newly developed linear response formulation. In the **IBM** group, the work on doped garnets is being finalized and the fitting procedure will be generalized and tested for different cases. A concerted effort will be made to implement the next stages, which will be based on polarizable force fields or Car-Parrinello dynamics, and will also include new candidate structures from the Pauling files database. The **Marzari** group is in the meanwhile performing first-principles calculations to elucidate the effect of polymorphism (orthorhombic/tetragonal structure) on the conductivity and stability of the $\text{Li}_{10}\text{GeP}_2(\text{O}/\text{S})_{12}$ ionic conductor.

4 AiiDA-based discovery of low-dimensional materials and nanostructures

Nicola Marzari — EPFL, Daniele Passerone — Empa, Mathieu Luisier — ETHZ, Expt.: Empa, Kumar V. Agrawal — EPFL

Low-dimensional materials provide novel opportunities to venture into largely unexplored regions of materials space. In this respect, the availability of a broad portfolio of two-dimensional (2D) candidate materials resulting from a screening procedure [15] is an outstanding asset. From the extensive database of exfoliable materials, classified into groups of easily (1'036) and potentially (789) exfoliable compounds, the **Marzari** group proceeded further by first identifying the most common prototypes, which will allow future expansion of the list of novel 2D materials by chemical substitutions and alternative site decorations. For 258 easily exfoliable compounds with up to 6 atoms/cell, the **Marzari** group then comprehensively studied at the DFT-PBE level their stability, as well as their vibrational, electronic, magnetic, and topological properties, revealing a wealth of magnetic systems (56 magnetically ordered, including 37 ferromagnets, 19 antiferromagnets, 14 half-metals, and 6 half-semiconductors — Fig. 7) and highlighting a relative scarcity of insulators with Z2 topological order. Around 1'000 of all easily or potentially exfoliable compounds were also screened for quantum spin Hall insulators, with and without additional strain applied, allowing the discovery of a handful of novel topological insulator candidates for which further characteri-

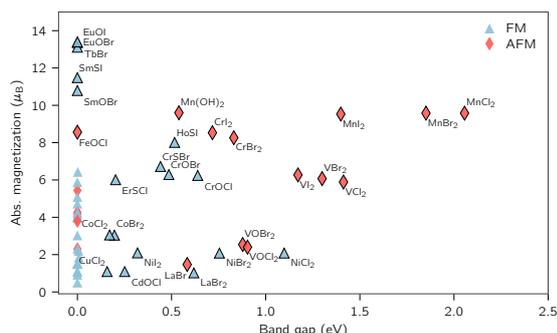


Figure 7: Absolute magnetization (in μ_B /unit cell) vs PBE band gap (in eV) for a set of magnetic 2D materials. Structures with ferromagnetic ground state are marked with triangles, while diamonds correspond to antiferromagnetic ordering.

zation is ongoing. Properties such as electronic band structure and density of states, projected density of states and vacuum levels have been studied at the PBE level for nearly 1'500 monolayers. Effective mass tensors, at each band extrema relevant for transport, have been computed for all the insulators and semiconductors within the aforementioned set. Accurate band gaps using the GLLB-sc functional have been obtained for nearly 900 materials. These properties allowed us to identify 80 possible candidates for photocatalytic water splitting and 20 materials suitable for thermionic devices that are currently being studied in detail. Furthermore, the **Marzari** group identified a handful of new candidates for FET channels, which are currently being screened in collaboration with the **Luisier** group. A geometric screening over all ICSD and COD layered compounds allowed the **Marzari** group to identify 110 layered nanoporous materials and their binding energy is currently being computed. Forty materials have already been identified as easily or potentially exfoliable, and are being studied for gas and ion separation in collaboration with the experimental group of **Agrawal**.

In collaboration with the **Corminboeuf** group, the portfolio of 2D compounds is also searched for catalysts for the hydrogen and oxygen evolution reactions using the volcano plot approach (see Sec. 2.1). At the same time, elastic properties, phonon dispersions and Raman activity of multilayer compounds are investigated in collaboration with the experimental group of **A. Ferrari** at the University of Cambridge. On similar grounds, a collaboration has been initiated with an industrial company, to try to understand better the relation between elastic properties and exfoliability. Finally, all 3D compounds extracted from the ICSD and COD are also screened for Kitaev-type mag-

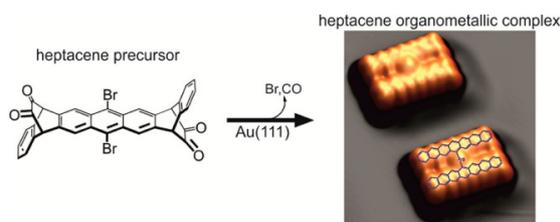


Figure 8: On-surface synthesis scheme of Au-directed organometallic complexes and corresponding STM image of the heptacene product (from [38]).

netic interactions, in collaboration with the **Yazyev** group (VP1).

Concerning 1D carbon based nanomaterials, the **Empa** group has attained new objectives in the synthesis and characterization. A route for the on-surface fabrication of acenes, a family of polycyclic aromatic hydrocarbons (PAH) formed by linearly fused benzene rings, has been defined and applied to the fabrication of heptacene (Fig. 8) [38] and nonacene [39] on $\text{Au}(111)$ by means of α -diketone-type precursor molecules that can be sublimed remaining intact and converted to heptacene or nonacene by thermal activation on the metal surface.

Comparison of simulated and experimental STM images (by means of the techniques developed in the previous years) together with total energy calculations allowed the **Empa** group to discriminate among different side products in the fabrication process. For the characterization of the electronic properties of acenes on $\text{Au}(111)$, the collaboration with the **Hutter** group was essential. The cubic scaling approach implemented in CP2K to compute quasiparticle energies through self-consistent GW was employed to compute the fundamental gap of acenes of different length (Fig. 9) in the hypothesis of closed shell system and open

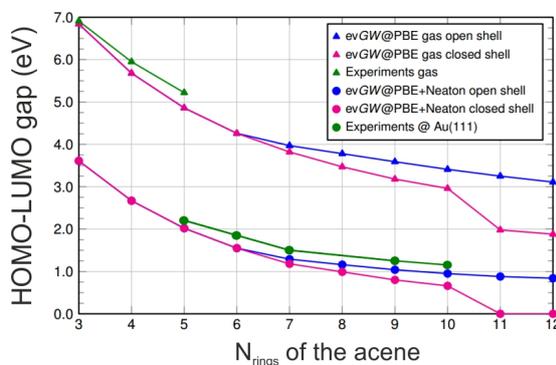


Figure 9: Singlet open-shell GW gaps show better agreement to the experimental values than closed-shell GW gaps. This finding may indicate that higher acenes possess significant open-shell character on $\text{Au}(111)$. Image credit: Ignacio Urgel, Empa.



shell system. Total energy calculations confirm the increased reactivity of nonacene compared to heptacene. Moreover, the GW results were found to match the STS measurements only in the hypothesis of open shell systems for acenes longer than octacene. The results provide and support the first experimental evidence of the radical character of PAH when their length exceeds eight units.

More recently, the **Empa** group reported a novel synthetic pathway that uses methyl groups to form 5-membered rings upon cyclohydrogenation in a controlled fashion. Starting from a monomer design including bromine functionalization and methyl groups, polymers containing indenofluorenes as base unit were created [40]. In this respect, the technique developed for the simulation of non-contact AFM images turned out to be crucial for the characterization of the geometric features of the fabricated materials. This technique has already been integrated within AiiDA workflows by the **Smit** group (HP4). Also in this context, the collaboration with the **Hutter** group was instrumental to apply the GW level of theory to the long anti-aromatic chains.

All the results related to this project (in particular the novel 2D structures predicted) will be made available online on the Materials Cloud platform (www.materialscloud.org).

5 Predicting strength of precious metal high-entropy alloys

William Curtin — EPFL

High-entropy alloys (HEA) are a new class of metals consisting of five or more elements at near-equal compositions. HEA exhibit high strength and ductility and new materials are emerging rapidly. Schroers *et al.* [48] reported an fcc PtPdRhIrCuNi alloy with impressive properties (yield strength 526 MPa, ultimate strength 1'800 MPa, elongation of 30%) rivaling the strongest and toughest steels. The goal of this project is to use a new theory [41] to predict the initial yield strength of the PtPdRhIrCuNi alloy based solely on material properties computed via first-principles density functional theory (DFT).

We compute three key properties needed in the theory: misfit volumes of each elemental addition in the alloy, elastic constants, and stable stacking fault energy of the alloy. Being random alloys, large computational cells, multiple random realizations, and special quasi-random structures (SQS) are needed. Mis-

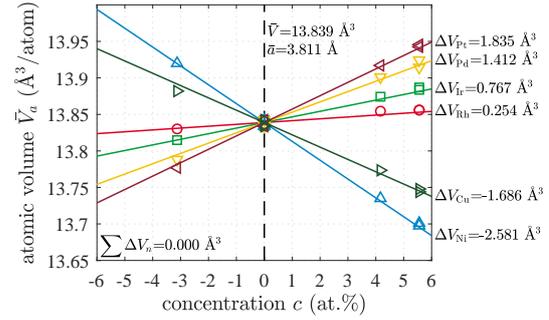


Figure 10: Changes in fcc atomic volumes for solute concentration deviating from the average concentration for the PtPdRhIrCuNi high-entropy alloy, from which individual solute misfit volume is obtained.

fit volumes are computed from derivatives of the lattice constant at compositions around the central composition (Fig. 10). Elastic constants are computed using standard methods, but recognizing that the finite-size random alloy samples break cubic symmetry so that averaging is necessary. Isotropic elastic constants are computed as $\mu = \sqrt{\frac{(C_{11}-C_{12})C_{44}}{2}}$ and the Poisson ratio is obtained from the computed bulk modulus B and μ . The stacking fault energy is estimated using the ANNNI model based on the difference in fcc and hcp cohesive energies. Using all DFT-computed properties, the estimated fcc partial dislocation separation is found to satisfy the requirements for applying the analytic model.

Using all DFT-computed properties in the analytic model for yield strength, the predicted initial yield strength at the experimental conditions of $T = 300$ K and strain rate of 10^{-4} /s is 510 MPa. The prediction is in excellent agreement with the experimental value (526 MPa), with no adjustable/fitted parameters, and well within the uncertainty of both the DFT inputs and the approximations embedded in the model itself. This success represents the first fully predictive result for yield stress of any HEA, and establishes the basic methodology for application to other HEA.

This project extends the MARVEL efforts into structural metals. The predictions demonstrate that the computational methodology can be used for designing new alloy compositions of noble metals that possess even higher strengths. The methodology points to a systematic set of material properties needed to make predictions, with the possibility to establish workflows in AiiDA for automated computation across a wide range of alloys and exportation of such data to the Materials Cloud. These directions will be pursued in the continuation of this project.

MARVEL-related 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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Horizontal Project **3**

HP3 — Advanced Quantum Simulations

Project leader: Jürg Hutter (UZH)

Participating members: Jürg Hutter (UZH), Philipp Werner (UniFR), Matthias Troyer (ETHZ), Stefan Goedecker (UniBas), Nicola Marzari (EPFL)

1 Main goals and achievements

The development of new improved methods for the description of the electronic structure of materials beyond density functional theory (DFT) using local functionals is the primary goal. One main approach is based on DMFT and the development of a parameter-free *ab initio* method using GW+DMFT. Such a development has been successfully achieved and applications to realistic multi-band materials conducted. The non-equilibrium extension of DMFT has emerged as a powerful and versatile tool for the study of non-equilibrium phenomena in correlated lattice models. We are considering time-periodic steady states, which can be achieved when a system is coupled to a thermal bath and continuously driven by time-periodic excitations. By applying this method to correlated electron and electron-phonon systems, we hope to identify new strategies for the non-equilibrium control of materials.

Another line of research in this project is to establish Quantum Monte Carlo (QMC) as a reliable tool to predict properties of materials. We aim to introduce novel approaches to simulate and characterize many-body systems both at and out of equilibrium. Finding an efficient approach based on artificial neural networks is currently our central research theme to progress along this line.

Extending DFT using Koopmans-compliant spectral functionals has been a successful strategy for more reliable predictions of quasiparticle states. Recent progress in extending this approach to condensed matter systems has been achieved and documented with the accurate calculation of band gaps of semiconductors and insulators.

Methods that include an explicit treatment of electron correlation can yield reliable results for a wide range of weaker correlated systems. They have the potential to provide both high accuracy and general applicability, while still providing an efficiency that allows applications to large systems and a sampling of many

configurations. Further progress has been achieved in the development of MP2/RPA and GW methods. Another project develops implicit solvent models to facilitate the simulation of complex electrochemical solid/liquid interfaces. These efforts are made available for high-performance computers through the interface to the AiiDA software and their implementations in the freely available computer codes Quantum-ESPRESSO, BIGDFT, and CP2K.

2 Progress of the different efforts

2.1 GW+DMFT (Philipp Werner — UniFR)

We develop a parameter-free *ab initio* simulation method for correlated materials based on the GW+DMFT scheme [33], which allows a self-consistent treatment of screening and correlation effects in the solid. While self-consistent GW+DMFT calculations have previously been implemented for one-band Hubbard models [34, 35][1], we extend this approach here to realistic multi-band materials.

Our *ab initio* GW+DMFT scheme treats the different orbitals of the material with an appropriate level of accuracy. Starting from a LDA-DFT calculation for the full range of bands in the solid, a one-shot GW calculation (G_0W_0) is performed. We then define an intermediate subspace I , for which the goal is to construct an accurate low-energy model, and a possibly smaller correlated orbital subspace C , whose effective local interactions are treated by means of an impurity construction within an extended DMFT approach similar to [1]. Apart from the definition of the subspaces, there are *no free parameters* in this *ab initio* simulation approach.

The first application of this GW+DMFT formalism to SrVO₃ [3], in which we chose the t_{2g} subspace for both I and C , resulted in a new interpretation of the electronic structure of this material. SrVO₃ is not a strongly correlated metal with Hubbard bands, as was claimed by numerous previous studies [36, 37], but a weakly correlated metal with pronounced plasmonic

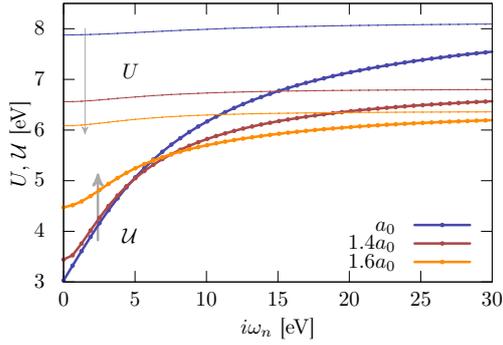
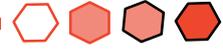


Figure 1: Comparisons between the bare (cRPA) local interaction U and the self-consistently determined GW+DMFT interaction \mathcal{U} for Na with different lattice constants (from [2]).

satellites. We have recently extended this study to SrMoO₃, where we also found evidence for plasmonic satellites, which is in agreement with previous interpretations of photoemission data [38]. The formalism has furthermore been tested with stretched sodium as a model system (Fig. 1).

A detailed description of our implementation of *ab initio* GW+DMFT and a discussion of the performance of this new simulation approach has been presented in [2].

2.2 Floquet DMFT (Philipp Werner — UniFR)

Floquet DMFT is developed as a method to study non-equilibrium phenomena in correlated lattice models [39]. By applying this method to correlated electron and electron-phonon systems, we hope to identify new strategies for the non-equilibrium control of materials.

We have applied Floquet DMFT to electron-phonon coupled systems in order to study the effects of phonon driving on superconductors [4]. Experimentally, it has been reported that the resonant excitation of a certain phonon mode in K₃C₆₀ induces a characteristic behavior in the optical conductivity, which is reminiscent of superconductivity [40]. Our simulations showed that even though the effective interaction can be tuned by the phonon driving, heating effects dominate so that the superconductivity is generically suppressed.

In a second project, we have implemented the Floquet DMFT using a strong coupling expansion method as an impurity solver. We used this method to study the high-harmonic generation (HHG) in Mott insulators. This work revealed the general HHG profile (Fig. 2) and its relation to the dynamics of doublons and holons.

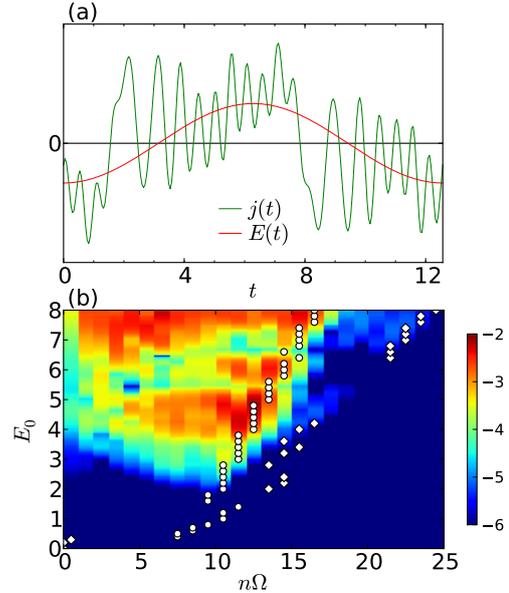


Figure 2: Results of the Floquet DMFT for the AC driven Mott insulator (Hubbard model with $U = 8$). (a) The current $j(t)$ and electric field $E(t)$ during one period for field strength $E_0 = 4.0$. (b) HHG spectrum in the plane of field strength E_0 and harmonic energy $n\Omega$.

2.3 Topological systems (Philipp Werner — UniFR)

The study of topological properties of band structures is one of the most active research fields and has resulted in new strategies of classifying materials. In topological systems, the effect of strong electron-electron interactions leads to highly nontrivial new effects that challenge usual classification via the Chern number [41]. Parallel to these developments, the question of how topological properties manifest themselves in non-equilibrium setups — where a classification via the Chern number fails — arose recently. Our goal is to identify experimentally accessible quantities that reflect the topological nature of weakly and strongly correlated materials in both equilibrium and non-equilibrium scenarios.

We have adapted and extended the recently suggested classification via the circular dichroism [42] to model systems for HgTe quantum wells [5]. By tuning and optimizing the parameters of the left circularly polarized (LCP) and right circularly polarized (RCP) light pulses, we showed how the Berry phase can be mapped out by the circular dichroism in optical absorption (Fig. 3). This concept goes beyond the usual band-structure classification and hence is robust against electron-electron and electron-phonon interactions. We have demonstrated this by including electron-

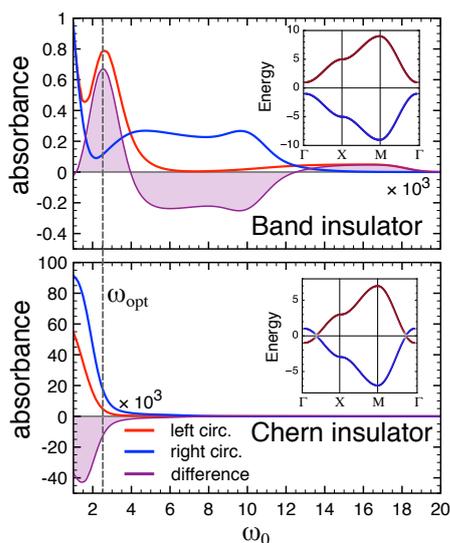


Figure 3: Photoabsorption probability of the massive Dirac model in the band insulator (upper panel) and Chern insulator state (lower panel) as a function of the photon energy, resolved with respect to LCP and RCP light. The insets depict the band structure. The vertical dashed line indicates the optimal photon energy ω_{opt} which allows to clearly distinguish the different states (from [5]).

phonon coupling using a quantum kinetic approach [5].

Furthermore, extending the setup to *transient* absorption allows to trace the non-equilibrium time evolution after the transition across topologically distinct phases. In particular, we demonstrated how the transient dynamics of the topology after a quench can be traced via pump-probe spectroscopy. Moreover, we discussed the nontrivial steady states after electron-phonon relaxation and their manifestation in angle-resolved photoemission spectroscopy (ARPES).

2.4 Quantum Monte Carlo methods for molecular dynamics and neural network quantum states (Matthias Troyer — ETHZ)

We work to establish Quantum Monte Carlo (QMC) as reliable tool to predict properties of materials with strong electronic correlations. Finding an efficient technique to combine the QMC electronic simulations with a molecular dynamics (MD) based ionic sampling is also needed. Another goal is to introduce approaches to simulate strongly-correlated many-body systems both at and out of equilibrium. Finding an efficient approach based on artificial neural networks is a central research theme to progress along this line. In the last year we developed a novel MD framework, based on a covariant formulation

of the Langevin equations, to accelerate the sampling of finite temperature properties [6]. The method can be combined with DFT and QMC, and it has been applied to dense hydrogen [6] and hydrogen-helium mixtures at planetary conditions [7]. This achievement allows us to perform large scale and well equilibrated simulations using QMC forces. As test case we consider hydrogen under high pressure.

Another methodological advance concerns the calculation of quantum chemical rates from standard equilibrium path-integral molecular dynamics simulations (PIMD) [8]. We discovered an important property of PIMD, which links the autocorrelation time of the simulations with the real quantum tunneling rate (Fig. 4).

A main research line developed concerns the use of artificial neural networks (ANN) to study many-body quantum systems. We express the many-body state in terms of a complex-valued ANN, whose output is the value of the wavefunction's amplitude on a given many-body state in the Hilbert space. ANN can then be used as a highly flexible variational ansatz to solve both the static and the time-dependent SE [9]. Describing the many-body state as an ANN constitutes a controlled expansion of the wavefunction, since upon increasing the capacity of the network one can systematically approximate the full many-body state. In [9] we have introduced a restricted Boltzmann machine (RBM) network architecture, and obtained results both for 1D and 2D lattice spins systems. Neural-network quantum states can also be used to perform quantum state tomography (QST) of large many-body systems. The problem of reconstructing the wavefunction from experimental measurements shares very important analogies with the statistical inference problem solved by machine learning (ML). QST can be then seen as a ML problem where ANN are used to “reproduce” the observed measure-

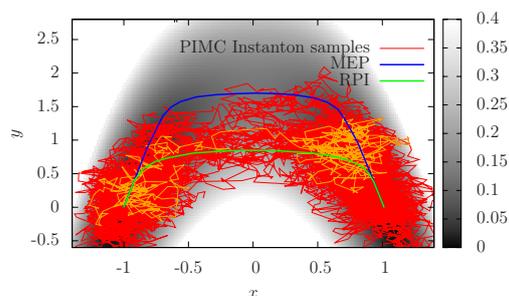


Figure 4: Ensemble of transition states generated by QMC simulations on a proton transfer reaction model [8].



ments, and generate arbitrary new measurements. This paradigm is called *unsupervised* learning in the ML community, and its application to QST has been introduced in [10], where new results have been obtained for QST. In particular, we did QST of very large quantum systems, previously inaccessible to brute-force QST methods. The representational power of ANN for many-body states has been also further investigated in [43], where the first use of deep networks in an unsupervised setting has been achieved.

2.5 Koopmans-compliant spectral functionals for extended systems (Nicola Marzari — EPFL)

Koopmans-compliant functionals [44][11] have been shown to provide accurate spectral properties for molecular systems [12, 13]; this accuracy is driven by the generalized linearization condition imposed on each charged excitation, i.e. on changing the occupation of any orbital in the system, while accounting for screening and relaxation from all other electrons [14]. We worked on the practical implementation of this formalism to the case of extended systems. We are looking for reliable, and computationally efficient approaches to predict spectral properties in nanostructures, interfaces, or solids.

A third condition, namely, *localization* of Koopmans' orbitals, becomes determinant for extended systems: the condition of Koopmans' compliance not only works better when states are localized [14], but it relies in an essential way on localization when considering larger and larger systems, where the variational Koopmans' orbitals converge rapidly to their thermodynamic limit [15].

To prove the effectiveness of the Koopmans' formalism, we applied it to predict the band gap E_g for a set of 30 compounds [15], including small gap semiconductors and large gap insulators (Fig. 5). The results show that in PBE the value of the E_g is underestimated with a mean absolute error (MAE) and mean absolute percent error (MAPE) of about 2.5 eV and 50% with respect to experiments, respectively. Notably, MAE in KI and KIPZ are down to 0.27 and 0.22 eV; this latter is comparable with qp-scGW [45][16] and more accurate than G_0W_0 [PBE] [16][46].

These results reiterate the role of Koopmans-compliant functionals as simple and accurate quasiparticle approximations to the exact spectral functional [17][47]. The broad range of applicability and the reduce computational cost with respect to diagrammatic technique, makes

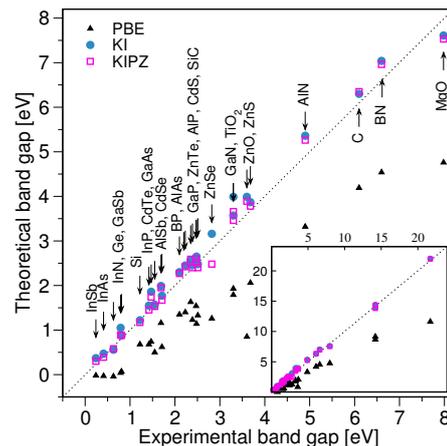


Figure 5: Band gaps of 30 semiconductors and insulators, calculated using PBE, KI, or KIPZ functionals, compared with available experimental data, shown in an energy range between -0.5 and 8.2 eV. A wider energy range (up to 22 eV) is shown in the inset.

those functionals very attractive to study electronic levels in complex materials and devices.

2.6 Implicit solvation models (Stefan Goedecker — UniBas, Nicola Marzari — EPFL)

Following the lines of the self-consistent continuum solvation (SCCS) model [48], and exploiting the impressive performances of a recently proposed preconditioned conjugate gradient algorithm for the generalized Poisson equation [18], we developed, parametrized and tested a new implicit solvation approach [19]. Here the interface between the quantum-mechanical solute and the surrounding environment is described by a fully continuous permittivity built up with atomic-centered “soft” spheres. It is able to describe accurately both neutral and charged systems. We show that, with given, fixed atomic radii, two parameters are sufficient to mimic various aqueous and non-aqueous solvents. Tuning of the model on experimental data of contact angle measurements allowed to improve its accuracy on surface properties of materials that represent one of the innovative targets of the developed methodologies.

To explore the feasibility of coupling implicit solvation models with first-principle structure prediction methods, we coupled the soft-sphere model with the minima hopping method [49]. We systematically explored reconstructions on the (100) surface of calcium fluoride (CaF_2) and other fluorites (MF_2), $M = \text{Sr, Cd, Ba}$, in vacuum and water environments [20]. We performed a global structural search both by explicitly including wa-

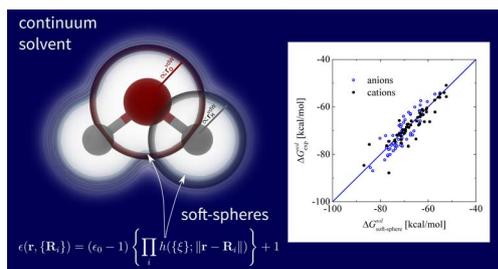


Figure 6: The soft-sphere cavity.

ter molecules and by employing the implicit soft-sphere model. The implicit approach correctly reproduces both our findings with the explicit-water model and the experimentally reported contact angles for the partial-hydrophobic (111) and hydrophilic (100) surfaces.

To further improve the accuracy of the models, we implemented a non-local definition of the cavity, able to correct artifacts coming from inaccessible regions of space smaller than a pre-determined solvent radius (Fig. 6). Typical examples of these situations include empty pockets in complex molecular systems and hollow sites or channels in crystals. The new solvent-aware formulation allows to maintain a continuously differentiable expression of the interface with a moderate computational overhead [21].

2.7 Explicitly correlated methods (Jürg Hutter — UZH)

Methods that include an explicit treatment of electron correlation can yield reliable results for a wide range of systems. They have the potential to provide, both, high accuracy and general applicability. We are looking for algorithms to calculate the energy at these levels of theory while still providing an efficiency that allows applications to large systems and a sampling of many configurations.

Analytic gradients and stress tensor are available for MP2 [22, 23]. We investigated properties of liquid water using these methods and compared them with a series of density functionals [24, 22]. Further investigation of the properties of solvated electrons and redox properties of water [25] are performed.

We developed [26] a method to calculate the RPA correlation energy with $\mathcal{O}(N^3)$ operations. The $\mathcal{O}(N^3)$ -RPA method relies on the RI with a local metric, imaginary time and imaginary frequency grids and sparse matrix multiplications [27]. New treatments of the local [28] as well as fully periodic two center integrals resulted in additional efficiency im-

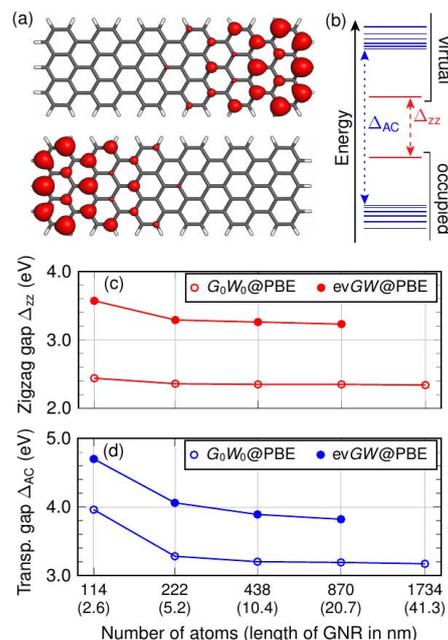


Figure 7: (a) Molecular geometry of the (7,12) GNR (6-anthene) together with an isosurface of the zigzag edge states obtained from DFT. (b) Sketch of the spectrum of the (7,12) GNR with the HOMO-LUMO gap Δ_{zz} between zigzag (zz) states and the transport gap Δ_{AC} (HOMO-1-LUMO+1) between bulk states. (c) Zigzag gap Δ_{cc} anthenes with a horizontal length from 2.6 nm to 41.3 nm.

provements. The same techniques have been transferred to our implementation of the G_0W_0 method [29]. The method has been successfully applied to study the electronic structure of graphene nanoribbons [30] (Fig. 7).

2.8 Verification of electronic structure methods (Stefan Goedecker — UniBas, Nicola Marzari — EPFL)

a) *Basis sets in all-electron codes* While any user of a density functional code is aware of the fact that the calculation is based on an approximated exchange correlation functional, users are generally not so much aware of other intrinsic approximations such as basis sets, integration grids, pseudopotentials, PAWs, that can significantly compromise the accuracy. To assess the influence of these approximations on the accuracy of density functional calculations, highly accurate reference data are required. Multi-wavelet basis sets form an adaptive and systematic basis set that allows to perform all-electron calculations with arbitrary high accuracy. Using the MRChem [50] density functional code, we were able to calculate total energies and atomization energies of a dataset of 211 molecules with μ Hartree precision [31]. In addition we also provided highly accurate

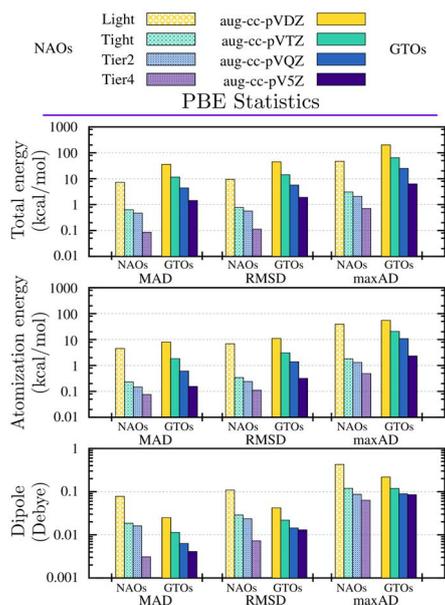


Figure 8: Deviation of the PBE total energy, atomization energy and dipole moment of molecules for two basis sets GTOs and NAOs of different sizes with respect to the MRChem reference values.

dipole moments for these molecules. These reference values were used further to estimate the error arising due to basis sets used in different all-electron codes. The basis sets considered were GTOs as implemented in NWChem [51] and numeric atomic orbitals (NAOs) as implemented in FHIaims [52]. From these comparison studies it was found that GTOs on average can achieve chemical accuracy (1 kcal/mol) using the largest basis set as shown in Fig. 8. However, medium size basis sets give significant errors comparable to the ones arising from the xc potential. NAOs, on the other hand, can achieve chemical accuracy with basis sets used for production calculation.

We started also to assess the accuracy of pseudopotentials. First results show that the errors in atomization energies are comparable or larger than errors arising from xc functionals. It became also obvious that good values in the Delta test of elemental solids is not sufficient to obtain accurate atomization energies.

b) *The standard solid-state pseudopotential (SSSP) library, version 1.0* We updated and finalized the definition of a standard protocol, namely the SSSP protocol, to assess the precision and efficiency of pseudopotential libraries and implemented it as an automated workflow using the AiiDA infrastructure [32]. In particular, in addition to the Δ -factor and the convergence of phonon frequencies, cohesive energy and pressure of elemental solids,

we now added also the convergence of band structures.

We systematically applied this protocol to several publicly available pseudopotential libraries using the Quantum-ESPRESSO package and identified two new sets of optimal pseudopotentials, called SSSP efficiency and SSSP accuracy (version 1.0), for 85 elements of the periodic table. As of today, the SSSP accuracy is the most accurate open-source pseudopotential library with the smallest average Δ -factor. All the data produced is stored in an AiiDA database and is freely available on the Materials Cloud (www.materialscloud.org) where it is also possible to track the full provenance of the results obtained in order to ensure their reproducibility.

3 Contribution to overall goals and initial proposal

The main goal of this project is the development of novel many-body simulations techniques for improving accuracy and efficiency of electronic structure calculations, which then are used in the vertical projects VP1 and VP2 as well as in projects HP4 and HP5 for the investigation of new materials. The development of continuous-time QMC and GW+DMFT methods within this project reached this goal for strongly correlated materials. The development of Koopmans-compliant functionals, implicit solvent methods, and wavefunction-based correlation methods for large complex systems has opened new possibilities for the simulation of materials related to electrochemistry and heterogeneous catalysis as studied in VP2. Improvements of the DFT codes (Quantum-ESPRESSO, BIGDFT, CP2K) in performance on HPC systems are directly benefiting high-throughput sampling and verification. Cross-verification and validation of methods have been performed for a limited set of methods and approaches, although at a yet smaller scale than originally anticipated.

4 Collaborative and interdisciplinary components

The groups within HP3 are working mainly in bilateral collaborations. Such collaborations exist within HP3 as well as with PIs from other projects. The Troyer and Werner groups collaborated on continuous-time QMC and on downfolding methods. They have several international collaborations, e.g. with S. Sorella (SISSA, Italy), and Ferdi Aryasetiawan (Lund, Sweden). The Hutter and VandeVondele groups jointly worked on MP2

and RPA implementations in CP2K, as well as on high-performance algorithms for petascale computing. MARVEL collaborations on the application of the RPA and GW methods are active with the Passerone group, and the Marzari and Corminboeuf groups. The Marzari and Goedecker groups have a joint development on implicit solvent methods, that resulted in a common library for the Quantum-ESPRESSO and BIGDFT codes. This independent library is suitable for integration in other codes, including packages developed outside MARVEL. Testing frameworks are developed by the Hutter and Goedecker groups, coordinated with the group of Marzari.

MARVEL-related publications

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

- [1] L. Huang, T. Ayral, S. Biermann, and P. Werner, *Extended dynamical mean-field study of the Hubbard model with long-range interactions*, *Physical Review B* **90**, 195114 (2014).
- [2] F. Nilsson, L. Boehnke, P. Werner, and F. Aryasetiawan, *Multitier self-consistent GW + EDMFT*, *Physical Review Materials* **1**, 043803 (2017).
- [3] L. Boehnke, F. Nilsson, F. Aryasetiawan, and P. Werner, *When strong correlations become weak: Consistent merging of GW and DMFT*, *Physical Review B* **94**, 201106 (2016).
- [4] Y. Murakami, N. Tsuji, M. Eckstein, and P. Werner, *Nonequilibrium steady states and transient dynamics of conventional superconductors under phonon driving*, *Physical Review B* **96**, 045125 (2017).
- [5] M. Schüler and P. Werner, *Tracing the nonequilibrium topological state of Chern insulators*, *Physical Review B* **96**, 155122 (2017).
- [6] G. Mazzola and S. Sorella, *Accelerating ab initio Molecular Dynamics and Probing the Weak Dispersive Forces in Dense Liquid Hydrogen*, *Physical Review Letters* **118**, 015703 (2017).
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- [9] G. Carleo and M. Troyer, *Solving the quantum many-body problem with artificial neural networks*, *Science* **355**, 602 (2017).
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Horizontal Project **4**

HP4 — Advanced Sampling Methods

Project leader: Stefan Goedecker (UniBas)

Participating members: Stefan Goedecker (UniBas), Michele Parrinello (USI and ETHZ), Ursula Röthlisberger (EPFL), Berend Smit (EPFL)

1 Main goals and achievements

Perovskites are a highly promising material for photovoltaic applications. Within a few years the efficiency of perovskites based solar cells has reached 22 percent and is thus comparable to the efficiency of solar cells based on materials that are more expensive to manufacture. In particular, lead halide perovskites of the form ABX_3 (A = mono or divalent cation, B = divalent cation, X = halide anion) have recently become an intensely investigated area of research as light-harvesting materials due to low production costs and favorable band gap and charge carrier transport properties [12, 13]. Several groups within HP4 and MARVEL (Röthlisberger, Goedecker, Parrinello, von Lilienfeld, Ceriotti, Pasquarello) jointly focused their research activities on perovskites. State-of-the-art methods developed by these groups, such as methods to predict the structure, methods to simulate growth and nucleation as well as machine learning methods, were applied in a concerted way to advance our understanding of this important class of materials. The challenges posed by perovskites also spurred further methodological developments such as variationally enhanced sampling methods or the development of improved force fields for perovskites materials. The numerous collaborations that have been established within HP4 in this context would not have come true outside an NCCR and will be the basis for further progress in the next phase of the NCCR. The close collaboration of the Röthlisberger group with the experimental group of Grätzel establishes in addition a valuable link to experiment. The nucleation and growth simulation methods developed by the Parrinello group address a fundamental problem in materials sciences and will find also many other applications outside the perovskites project. In addition to perovskites other materials that are of interest for energy related applications were studied such as materials for hydrogen storage, superconducting

materials and materials for high-voltage power electronics. Another topic that is of interest to several groups in HP4 is to find descriptors or fingerprints. Many projects in MARVEL have in common to investigate in how far materials can be described by their shapes, which can be the topological-type of a bond network, the geometrical shape of atom positions, pore shapes of nanoporous materials, or the form of energy or density landscapes. The methods used range from traditional geometric descriptions like volumes over graph-theoretical approaches to methods from algebraic topology. Such methods allowed the Smit group to identify in large databases porous materials with optimal pore structures.

2 Progress of the different efforts

2.1 Perovskites (Ursula Röthlisberger — EPFL, Stefan Goedecker — UniBas, Michele Parrinello — USI and ETHZ, Anatole von Lilienfeld — UniBas)

a) *Phases of organic-inorganic lead halide perovskites* Due to the liquid processing of organic-inorganic lead halide perovskites, there is only limited control of the phase that is formed. Since the crystalline structure crucially affects the optical, electrical and transport properties, a complete knowledge of the stability ranges of different crystalline phases is crucial for the design of suitable photovoltaic materials but an exhaustive exploration of the space of possible phases is a methodological and computationally highly demanding task.

In a collaboration between the Goedecker and the Röthlisberger group, the minima hopping method (MH) [14] was applied to identify crystalline structures of halide perovskites. Minima hopping is an efficient method to find the ground state as well as other low-energy configurations of condensed matter systems. The application of MH, to identify the phases of a crystalline system requires an accurate



model of the atomic forces. Density functional theory (DFT) has proven to work quite well for lead halide perovskites, however it is methodological and computationally rather expensive which makes it unsuitable for an extensive screening of the phase space. For this reason MH simulations were performed for methyl-ammonium lead iodide (MAPbI₃) using a force field. MH was coupled with a low-cost inter-atomic model potential [15] derived from first-principles simulations that was refitted to a new functional form suitable for the TINKER molecular simulation package. This model is able to describe the main features of MAPbI₃ but it fails to describe the transition from the low-temperature orthorhombic to the room temperature tetragonal phase. In these force field based MH runs, ~ 6'500 regular as well as defect structures were found. To classify and further analyze this large pool of structures, they had to be systematized with respect to their geometric similarity. To this end, in collaboration with the group of Ceriotti, a sketch-map [16] was constructed. Each point on the map represents a configuration and the close proximity of a pair of points indicates their structural similarity. All the experimentally known crystal structures of MAPbI₃ were identified and, in addition, a new δ like structure was predicted that could exist at very low temperatures showing that MH sampling in combination with sketch-map analysis is a powerful way to predict the different crystalline phases of this kind of materials.

b) *Improved force fields for MAPbI₃* The R thlisberger group is currently developing more accurate force fields for various halide perovskites systems. First tests with a zeroth order polarizable force field showed that the tetragonal phase of MAPbI₃ can indeed be stabilized when introducing polarization. However, the lowest energy structure was still not predicted correctly. To develop an automated way of generating optimal fixed point charge as well as polarizable force fields for various systems, a force-matching approach has been developed and implemented based on the method originally introduced by Ercolessi [17]. As first test case system, this approach has been applied for the parametrization of a fixed-point charge model of CsPbI₃ for which no force field is currently available. To employ the force matching method, the MINPACK library of FORTRAN for least squares minimization of the residual of a set of nonlinear equations has been coupled with the TINKER package. This allowed us to build a general protocol for generating force fields for organic-inorganic

halide perovskites. The newly developed fixed point charge model can reproduce the experimentally known δ and cubic perovskites structures over a wide range of temperatures (up to 650 K), predicting as most stable structure at 0 K the δ phase with an energy difference $\Delta E = E_{cubic} - E_{\delta} = 1.53$ kcal/mol consistent with DFT calculations $\Delta E = 4.09$ kcal/mol.

c) *Nucleation and crystal growth of lead halide perovskites* Since the emergence of perovskites solar cells, it has become clear that the efficiency of these devices is directly related to the quality of their crystalline morphology. Therefore it is important to understand the nucleation and surface growth of the prototypical system methyl-ammonium lead iodide (MAPbI₃) by performing standard and enhanced sampling molecular dynamics (MD) simulations. However, nucleation is a typical example of a rare event occurring on a time scale that is much longer than what atomistic simulations [1] can typically afford. To overcome the time scale problem in MD simulations of nucleation, the R thlisberger group employed enhanced sampling methods [18] in a collaboration with the Parrinello group. We have found that solvation of lead (Pb²⁺) and its coordination with iodide (I⁻) dominate the pre-nucleation clusters in the precursor solution. In a supersaturated solution, Pb²⁺ reacts with I⁻ and forms PbI₄²⁻, which is the thermodynamically most stable complex. Furthermore, we have simulated the seeded crystal growth of the cubic phase of MAPbI₃ in γ -butyrolactone (GBL). We observed that crystals grew with the surface adsorption of different layers consisting of PbI₂ and MAI. An important finding from this study is that the PbI₂ layer grows with a “rough-growth” mechanism while the formation of the MAI layer is controlled by entropic and electrostatic contributions. In addition, we have qualitatively analyzed the effect of dielectric strength of the solvent on the evolution of defects in the crystal micro-structure. This result can be very helpful to understand the effect of different solvents on crystal morphology and thus help rationalize the choice of solvents to produce superior perovskites thin films. As a next step, we have performed meta-dynamics simulations of nucleation of MAPbI₃ in a solution of dimethylsulfoxide (DMSO) as displayed in Fig. 1. We found that at first, lead and iodide ions form small clusters that consist of two main distinguishable geometries: (i) edge sharing and (ii) face sharing octahedra between lead and iodide. However, these aggregates are short-lived and we classify them as metastable

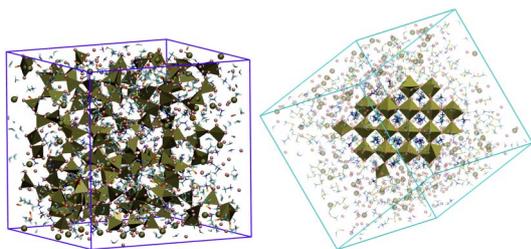


Figure 1: A typical simulation of nucleation of MAPbI_3 in dimethylsulfoxide (DMSO).

states in our simulations. The destruction of these metastable structures is accompanied by charge balance provided by MA^+ ions and the formation of a crystalline perovskites structure as shown in Fig. 1, right.

In the future, it is planned to investigate the effects of additives on the basic nucleation mechanism to form higher order lead-iodide complexes in different solvents. To this end, we will carry out *ab initio* MD simulations of precursor solutions that can serve as a basis for the generation of DFT consistent force fields based on force-matching. Additionally, we will extend the methods of constant chemical potential [2] to extract the free energy profiles of nucleation and crystal growth. Based on these studies, we will propose novel strategies for tuning the parameters for nucleation and crystal growth, thereby gaining better control over the resulting perovskites morphology, which is crucial for obtaining high performance of perovskites solar cells.

d) *Machine learning properties of perovskites*
The R othlisberger group has created a flexible, modular, and application-driven package of evolutionary algorithms (EA) called EVOLVE. This package allows to efficiently search chemical space, facilitating accelerated material and chemical design. This EA based optimization is being combined with machine learning models developed in the von Lilienfeld group to enable efficient fitness evaluations of electronic properties. As initial training set, a database of Kohn-Sham gaps is generated, which are compared with GW results from the Pasquarello group.

EVOLVE was applied to the optimization of machine learning models of molecular properties using “global” representations, i.e. descriptions of molecules without decomposition into atomic contributions. It was found that using genetic algorithms (GA) to optimize training set composition yields a significant improvement of the out-of-sample mean absolute errors (MAE). Consequently, much fewer training examples are required to reach a cer-

tain level of accuracy. There are systematic trends in training set composition upon going from randomly generated to GA-optimized training sets, including flattening of distance distributions and significantly increased sampling of the non-linear property tails. Given this, the a priori selection of training data was investigated and found to be nontrivial. Using GA provides a means to generate training sets with better-than-random performance within databases for which no GA has been performed. This work has been published [3]. Now the possibility to extend this technique to machine learning (ML) models based upon local representations [4] is investigated with the goal to see whether one can improve generalizability towards larger out-of-sample molecules.

The set of possible elements which can feasibly create perovskites materials is large, thus the search for the composition with optimal properties is a daunting task. Therefore a computational protocol for perovskites design was developed, that couples EVOLVE, the Quantum-ESPRESSO *ab initio* code and AiiDA [5]. This protocol can effectively find solutions close to the global minimum for a fixed composition. Structures produced by this technique are capable of being uploaded directly to the Materials Cloud. Furthermore, in collaboration with the von Lilienfeld group, a machine learning model of band gaps for a database of perovskites materials was developed which can be included in EVOLVE as an additional fitness metric. Fig. 2 details parity plots of DFT(PBE)- and ML-predicted band-gaps for a benchmark

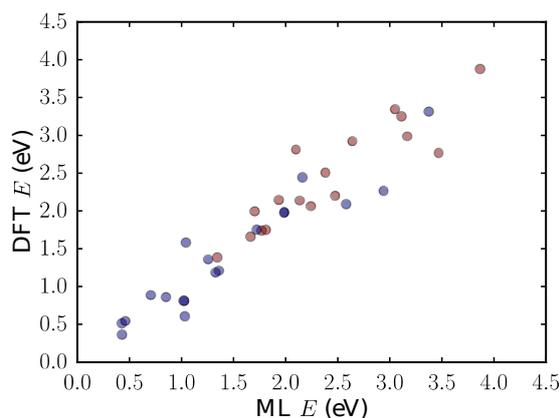


Figure 2: Parity plots of DFT(PBE)- and ML-predicted band-gaps for a benchmark database of ABX_3 perovskites; $B = [\text{Pb} (\text{blue}), \text{Sn} (\text{red})]$. 40 compositions used for training, predicting on 40 test samples. ML model: MARAD (molecular, alchemical, radial and angular distribution) descriptor, Gaussian kernel and KRR regressor.



of ABX_3 perovskites where $B = [\text{Pb} \text{ (blue), Sn} \text{ (red)}]$, $X = [\text{I}, \text{C}, \text{Br}]$ and $A = [\text{Cs}, \text{FA}, \text{MA}]$. A suitable fitness metric to describe thermostability with respect to unfavorable phase transitions is now developed to provide experimentally relevant GA-based predictions of potential perovskites compositions with suitable band-gaps for light-harvesting.

In a joint collaboration of the groups of Pasquarello, von Lilienfeld and R othlisberger, QSGW calculations were performed for the band gaps of cubic inorganic perovskites belonging to the $\text{Cs}BX_3$ class ($B = \text{Pb}, \text{Sn}$ and $X = \text{Cl}, \text{Br}, \text{I}$). Thermal vibrations were accounted for through *ab initio* molecular dynamics. Additionally, the spin-orbit coupling effects were evaluated at levels of theory of increasing accuracy and it was shown that semi-local density functionals significantly underestimate these corrections. In this way, reliable estimates for the band gaps of halide perovskites could be obtained [6]. The proposed scheme can now be used to study novel materials from the family of halide perovskites and to generate a reference dataset for machine learning models.

e) *Variationally enhanced sampling for phase transitions in perovskites* A powerful way to deal with a complex system is to build a coarse-grained (CG) model capable of catching its main physical features, while being computationally affordable. Inevitably, such CG models introduce a set of phenomenological parameters, which are often not easily deducible from the underlying atomistic system. The Parrinello group has proposed a systematic approach to go from a fine scale to a coarser one, taking advantage of the variational principle at the heart of VES [7]. This approach allows us to obtain the CG parameters from atomistic simulations, by minimizing the relative entropy of the CG model, while at the same time enhancing the sampling. This method was applied to the PbTiO_3 perovskites. A realistic Landau free energy was constructed and the Landau parameters were obtained by the VES method. By using these Landau parameters, and with the Landau free energy as Hamiltonian, the second-order phase transition temperature T_c and order parameters of PbTiO_3 were well reproduced with the coarse-grained model.

2.2 Other energy materials (Stefan Goedecker — UniBas)

a) *Silicon carbide for power electronics* Silicon carbide (SiC) is among the most promising materials for high-power switching electronics at

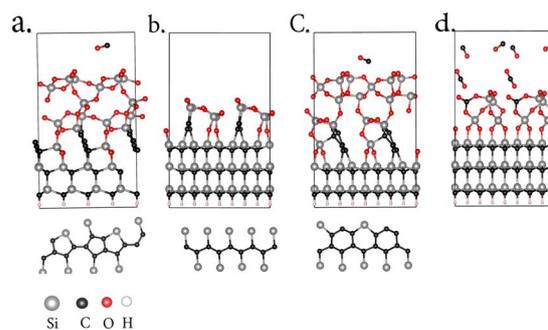


Figure 3: Different carbon cluster after oxidation at the SiC/SiO₂ interface; a) carbon five ring chain; b) v-shape carbon molecule; c) carbon six ring; d) without any carbon cluster.

elevated temperatures in field effect transistors. Usually a gate oxide (SiO₂) is grown by thermal oxidation technique. However, the mobility in the near interface region decreases during the thermal oxidation of SiC. This behavior has been attributed to new defect states, arising near the interface during the complex oxidation procedure but the nature of these defects was hitherto unknown.

To better understand the character of these oxidation defects, 4H-SiC(0001) surfaces at different oxygen concentrations were simulated and explored thoroughly by the Goedecker group using the minima hopping algorithm coupled with the density functional tight-binding method. Highly stable 5-ring (Fig. 3a) or 6-ring (Fig. 3c) were found that can either exist isolated or form ribbon like structures. In addition zigzag carbon ribbons (Fig. 3b) were also found. All these carbon ribbons gain a significant additional stabilization by anchoring to surface Si atoms. They also lead all to states in the band gap which lower the mobility.

b) *Transparent conductive oxides* The combination of optical transparency and high electrical conductivity enables transparent conductive oxide (TCO) materials to be used for a wide range of applications — from simple smart window coatings to OLEDs and futuristic see-through displays. Doped tin-dioxide (SnO₂) is an important semiconductor that is already used for these applications. However, in order to uncover the entire potential of this material in more advanced applications of optoelectronics, further improvements in electrical properties are necessary. In an extensive search for useful substitutional dopants of SnO₂, the entire periodic chart was scanned for stable charges and hole-electron dopants by the Goedecker group. These findings are in excellent agreement with current known

dopants and predict other possible substitutional dopants that have been not experimentally examined to date [8].

2.3 Descriptors and fingerprints (Michele Parrinello — USI and ETHZ, Berend Smit — EPFL)

a) *Fingerprints to distinguish liquid and solid environments* In investigations of crystallization events, collective variables are normally constructed using knowledge of the geometry of the final crystal structure and, therefore, they tend to be system specific. This limitation hinders the applicability of enhanced sampling methods for high-throughput calculations, for instance to all systems in a large database.

The Parrinello group has tackled this limitation by developing a set of general collective variables applicable to all systems [9]. Recalling that in crystallization, as in all first order phase transitions, there is an interplay between enthalpy and entropy, two collective variables are defined, one enthalpic and the other entropic. These two variables can be used to study the crystallization of Na, Al, and Si. More remarkable, they can be employed to study the superionic transition in AgI [10].

In many scenarios, it is also useful to assign a measure of crystallinity to each atom. Fingerprints [11] were devised for the identification of local crystal structures by projecting the enthalpy and entropy collective variables onto each atom. The entropy fingerprint is useful

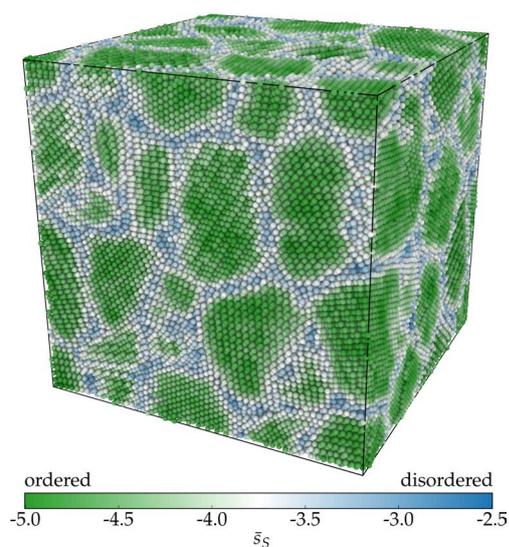


Figure 4: Nanocrystalline Al with mean grain size 5 nm at 300 K. Atoms are colored using our fingerprint. The color scale is such that green and blue atoms have ordered and disordered environments, respectively.

to distinguish between liquid-like and solid-like atomic environments (Fig. 4) and, in combination with the enthalpy fingerprint, it can be used to identify the crystal structure an atom is embedded into. In the future, these ideas will be applied to other systems, such as ionic and molecular crystals.

These tools are developed within the PLUMED 2 enhanced sampling plugin, that can be used with many molecular simulation packages.

b) *Nanoporous materials* The Smit group finished a project relating methane storage in nanoporous materials, in particular zeolites, to their pore-shapes as described by an adaption of persistent homology (PH) that was developed for that purpose. First steps were taken towards the integration of the nanoporous genome with AiiDA and the Materials Cloud. These include the adaptation of the AiiDA core to be able to efficiently deal with structures with large numbers of atoms, as required for the study of nanoporous materials, and the creation of a plugin template for jump-starting the development of AiiDA plugins. These improvements not only form the basis for all screening studies on nanoporous materials, but also for making the tools to compute PH descriptors available.

c) *Prediction of 2D zeolites* Based on the assumption that 2D layers can be extracted from zeolites if there are many more bonds in two directions than in the third, a maximum flow – minimum cut algorithm was used by the Smit group to find potential crystal surfaces. Zeolites with high potential for 3D → 2D synthesis via Ge-O hydrolysis were predicted.

d) *Distinguishing metal-organic frameworks (MOFs)* The question whether two MOFs are the same or not is very basic and fundamental. The existing algorithms to compare crystalline structures are based on symmetry comparison and cannot be used for comparing MOFs, which can exist in different spacial arrangements. The Smit group developed an algorithm that decides whether two MOFs are the same. It is based on distinguishing structures by comparing a set of mainly graph-theoretical descriptors that are obtained from the bond network (Fig. 5).

e) *Accurate pore volume* The Smit group provided an algorithm to compute the volume of a microporous materials (complement of the Connolly volume) that corresponds to the volume measured experimentally as obtained by the nitrogen uptake.

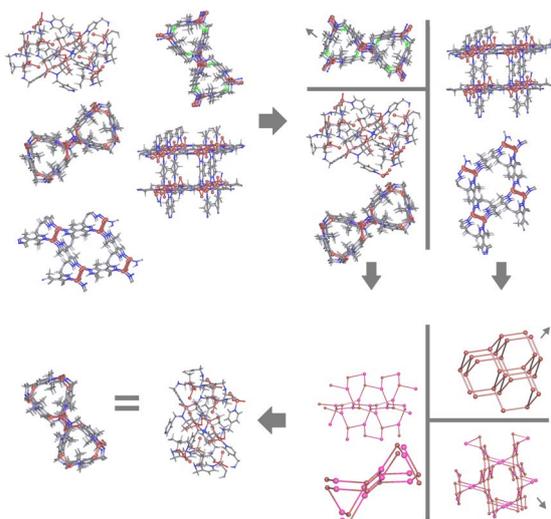


Figure 5: Distinguishing metal-organic frameworks.

f) *Evaluating different methods to generate the density overlap region indicator (DORI)* In collaboration with Corminboeuf’s group, the Smit group compared the shapes of DORI’s obtained from differently computed electron densities with PH. The aim is to rank the different approaches to compute electron densities by accuracy to find the cheapest method that gives a sufficiently good DORI for the description of bonds.

g) *Ionic conductivity of superionic conductors* In collaboration with Marzari’s group, the Smit group tested descriptors for ionic conductivity from the shape of potentials. An algorithm was developed to screen hypothetical conductors that detects breakthrough values for the formation of tunnels together with their direction, as well as the basins and transition states.

3 Contribution to overall goals and initial proposal

The goal of the initial proposal was to generate an interleaved research structure of horizontal methodological oriented projects with vertical application oriented projects. This interleaving turned out to be highly successful in HP4. A unifying application oriented topic was found, namely perovskites, that in addition is also an energy related material, which according to the initial proposal should be one the main thrusts of MARVEL. In addition other energy related materials were also studied. The work in HP4 really lead to the theoretical discovery of new materials such as new silicon allotropes with improved properties for photovoltaic applications, new porous materials for gas storage,

new perovskites formed out of different elements or new two-dimensional titanium dioxide sheets. The challenges posed by these materials lead to the development of new and improved methods that will allow to tackle other challenges in the area of energy related materials in the future.

4 Collaborative and interdisciplinary components

As outlined previously, the work in HP4 was highly collaborative. It had also a clear interdisciplinary character. Concepts and methods originating in physics, mathematics and computer science were used to solve problems in materials sciences.

MARVEL-related publications

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

- [1] M. Salvalaglio, C. Perego, F. Giberti, M. Mazzotti, and M. Parrinello, *Molecular-dynamics simulations of urea nucleation from aqueous solution*, Proceedings of the National Academy of Science of the USA **112**, E6 (2015).
- [2] C. Perego, M. Salvalaglio, and M. Parrinello, *Molecular dynamics simulations of solutions at constant chemical potential*, The Journal of Chemical Physics **142**, 144113 (2015).
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- [4] B. Huang and O. A. von Lilienfeld, *The “DNA” of chemistry: Scalable quantum machine learning with “amons”*, arXiv:1707.04146 (2017).
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- [17] F. Ercolessi and J. B. Adams, *Interatomic Potentials from First-Principles Calculations: The Force-Matching Method*, *Europhysics Letters* **26**, 583 (1994).
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Horizontal Project 5

HP5 — Materials Informatics

Project leader: Alessandro Curioni (IBM)

Participating members: Alessandro Curioni (IBM), Michele Ceriotti (EPFL), Volkan Cevher (EPFL), Anatole von Lilienfeld (UniBas)

1 Main goals and achievements

Work in HP5 focused on three main axes.

- Machine learning (ML) based models with increased accuracy for the prediction of crucial physical properties. These are physically inspired models. It was shown that, under the right training set conditions, ML based methodologies can serve as effective surrogates for classic density functional theory (DFT) based methods.
- New core ML methods that exhibit superior numerical behavior and are amenable to particular preconditioning and acceleration. More than 15% statistically significant improvement in prediction accuracy was demonstrated.
- Knowledge extraction and representation for accelerated material discovery. The support of deep search contextual and quantitative queries over a corpus of scientific literature. A demo was developed that showed how the approach can dra-

matically accelerate the pace of discovery in solid-state electrolytes.

In addition, the HP5 members strived to strengthen cross-group collaboration and in addition to practically demonstrate the validity of the proposed approaches in end-to-end demonstrations. This was repeatedly demonstrated at MARVEL meetings and reviews as well as with a number of key collaborative publications.

2 Progress of the different efforts

2.1 Machine learning, perturbation theory, and compositional space (Anatole von Lilienfeld — UniBas)

New rigorous structure-property models were developed using (i) machine learning (ML) and (ii) perturbation theory.

a) *Machine learning* A first key finding is that ML based models can outperform hybrid density functional theory in predicting various electronic structure properties including heats of atomization, HOMO/LUMO eigenvalues, polarizabilities, heat-capacities and others both in terms of performance and accuracy, when adequate training data exists [12, 13] (Fig. 1). Secondly, the new “amon” machine learning (AML) has been devised, which has been shown to afford systematically improving learning curves even for very large query molecules [1].

b) *Perturbation theory* It was shown that first order based estimates of atomic adsorption energies on metal surfaces, a crucial indicator of catalytic activity [2], can reach sufficient predictive power for the computational design of heterogeneous catalysts.

2.2 Machine learning, from molecules to materials (Michele Ceriotti — EPFL)

Previous years have focused on the extension of the smooth overlap of atomic posi-

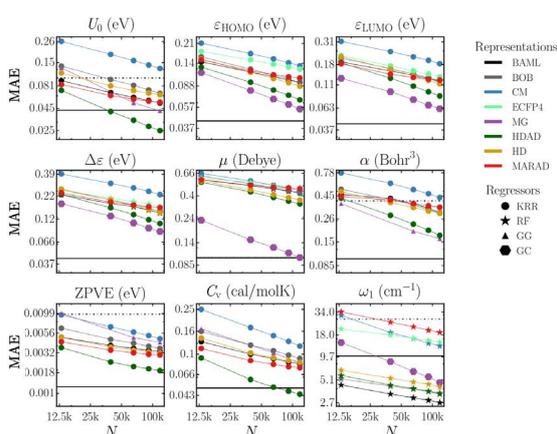


Figure 1: Learning curves (mean absolute error (MAE) as a function of training set size N) for 9 properties of QM9 molecules, displaying the best regressor for each representation and property. Horizontal solid lines correspond to target accuracies, vertical dotted lines correspond to approximated B3LYP accuracies (unless off-chart).

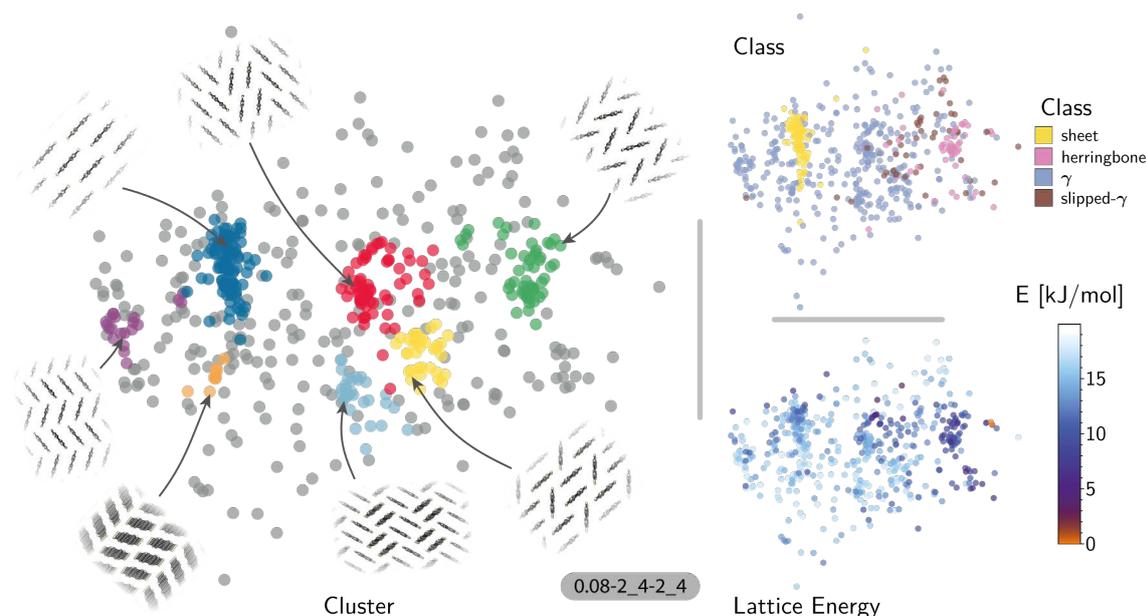


Figure 2: Sketch-map [14] representations of the crystal structure landscape of pentacene, as seen through the lenses of a REMatch-SOAP [3]. Each point corresponds to a configuration, color-coded according to their relative lattice energy (bottom right), heuristic packing pattern classification (top right) and automatic clustering (left, gray structures do not belong to a cluster). The structure of a representative of each cluster is also shown.

tions (SOAP) local similarity kernel [15] to treat multiple chemical species and to describe non-extensive properties through a weighted combination of local environments [3] inspired by optimal transport theory [16]. This period demonstrated that the combination of this kernel and Gaussian process regression provides a very general machine learning framework for the prediction of atomistic properties. In particular, it can be used to build a potential for Si that reproduces the balance of electronic surface states and elastic strain that governs the complex reconstruction of the (100) and (111) surfaces; it predicts the atomization energies of small molecules with an accuracy (< 1 kJ/mol error with the best combination of parameters) on par or better than the current state-of-the-art; it classifies molecules according to their binding behavior on a database of protein targets [4].

With better understanding and control of the ML framework, we could apply it to more complex systems. Following an early analysis of a database of gas-phase oligopeptides [5], we could tackle the challenging problem of predicting and interpreting the structure-energy-property landscape of substituted pentacene molecular crystals, which are relevant as materials for organic electronics [6] (Fig. 2). The maturity and the impact reached by SOAP-REMatch descriptors and sketch-map representations is also apparent in their use by collaborators, both inside

(Clémence Corminboeuf: sketch-maps to interpret the density overlap region indicator (DORI) in floppy molecules [7], Volkan Cevher: alternative norm regression on the SOAP kernel, Ursula Röthlisberger: mapping the landscape of lead-iodide perovskites) and outside MARVEL (C. Baldauf: SOAP kernels for quantum chemistry, G. Day: predictions and maps for crystal-structure predictions, J. Keith: rationalizing ion solvation energies based on the structure of solvation shells [8], C. Pickard: mapping the phase diagram of water ices).

2.3 Nonlinear prediction for materials discovery (Volkan Cevher — EPFL)

Work focused on achieving better accuracy beyond the root mean squared error (RMSE). It was shown that one can obtain improved mean absolute error (MAE) and max absolute error (MaxAE) for ML based DFT surrogate models. In particular, progress included:

- A new formulation for the molecule energy prediction via statistical learning theory with the intuition that different loss functions affect different statistical accuracy metrics.
- The design of a flexible machine learning framework for problem of predicting quantum properties. The new framework was applied with two new models: Ridge-

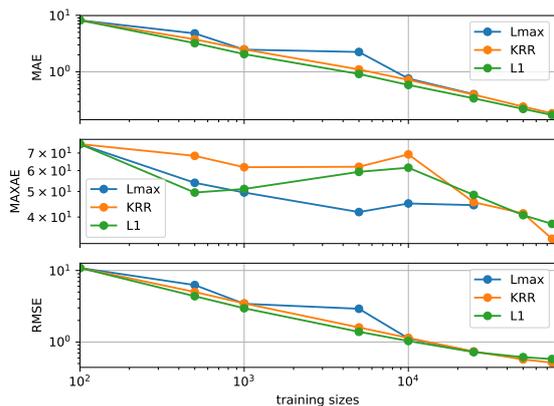


Figure 3: Learning curve (unit: kcal/mol), which is computed on the same training set (GDB9) with partitions suggested by Ceriotti’s group.

	KRR	L1	Lmax
MAE	0.4	0.34	0.39
MaxAE	45.53	48.54	44.4
RMSE	0.74	0.82	0.73

Table 3.1: Accuracy (unit: kcal/mol)

regularized ℓ_1 -loss minimization

$$\|\mathbf{K}\mathbf{x} - \mathbf{y}\|_1 + \lambda \mathbf{x}^\top \mathbf{K}\mathbf{x}, \quad (\text{L1})$$

and the same with ℓ_∞ -loss minimization:

$$\|\mathbf{K}\mathbf{x} - \mathbf{y}\|_\infty + \lambda \mathbf{x}^\top \mathbf{K}\mathbf{x}, \quad (\text{Lmax})$$

It was shown by experiments that kernel regressions with two proposed models achieve a better MAE and MaxAE than traditional kernel ridge regression (KRR).

- Standard optimization approaches cannot achieve the desired numerical accuracy. As a result, a novel preconditioning for these criteria was developed that can obtain accurate solutions, which are necessary for prediction.
- While the ML models proposed use kernels as inputs, different types of kernels, such as Gaussian and Laplacian kernels, were used to simulate the numerical results. It was demonstrated that the SOAP-REMatch kernel [3] achieves the state-of-the-art accuracy. See Table 3.1 and Fig. 3 for experimental results that show an overall 15% improvement in prediction accuracy.

2.4 Advanced technical document understanding for the discovery of novel materials (Alessandro Curioni — IBM)

Work focused on the integration and further development of four main components of the

cognitive strategy for the discovery of novel materials. These four components are:

- Document-ingestion: the ingestion of highly complex technical documents, which consist out of text, tables, images, formulas, etc. A major breakthrough has been achieved here by building default models. These default models significantly improve accuracy and reduce drastically the need for manual, laborious annotation.
- Data-integration: the integration of curated databases (e.g. ICSD or COD, technical domain ontologies, etc) into the ingested documents.
- Entity- & relationship-detection: the detection of entities of interests (such as inorganic materials and their properties).
- Knowledge engineering through knowledge graphs: the creation and advanced querying capability of knowledge graphs [9, 10].

We have focused a part of the effort into the development of the API’s for each cognitive component and their corresponding back-end implementation. We have collaborated with Empa to engage with an AiiDA implementation that drives the search result of the ingested literature to a targeted simulation (barrier computations, Fig. 4). In particular, 1’000 papers in the area of solid-state electrolytes (SSE) have been ingested. Then search for materials with properties within a certain envelop (using the knowledge graph) dramatically narrowed down the possibilities. The resulting materials were then pipelined for simulations using AiiDA. In a matter of one week we identified three new promising materials for SSE.

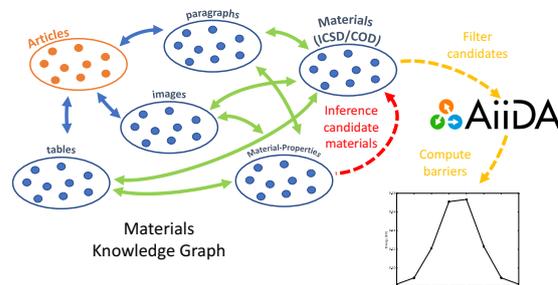


Figure 4: A sketch of the cognitive discovery pipeline demo for solid-state electrolytes, using the data ingestion pipeline and knowledge graph technology to scan potential candidates and AiiDA as a computational back-end to compute the barriers of the proposed materials. The height of the barriers are a key indicator for the suitability of the material as a SSE.

3 Contribution to overall goals and initial proposal

The research plan in HP5 followed very closely the initial proposal, with the needed adaptations of course, which however were not of altering nature. The mission of HP5 has been to develop horizontal machine learning and knowledge/data based approaches for the acceleration of materials discovery. In addition, the target was to demonstrate this by the application of the methodologies in a number of applications in collaboration with the vertical projects. Indeed, a number of such cross-project collaborations resulted in publications as well as in “live” demos that were not only just presented to our reviewers but also made available to the wider MARVEL community.

4 Collaborative and interdisciplinary components

The document ingestion pipeline of the Curioni group was applied to IOP 16'000 papers in collaboration with VP1 (Spaldin). Training on a mere 100 pages led to the development of document models with an overall accuracy of 99.67%. Focus was given for the discovery of solid-state electrolytes. The entire COD database of materials was scanned. A complete demo was developed using the AiiDA framework at Empa (Fig. 4). The von Lilienfeld and the Röthlisberger groups have collaborated in the exploratory study on the improvement of the predictive power of machine learning models of molecular properties due to genetic optimization of the training set composition [11]. Besides working on development and application of ML, the Ceriotti group also made the automatic mapping of structural landscapes easily accessible, deploying an infrastructure for interactive representation that is available as a demo (interactive.sketchmap.org) and ready to be integrated in Materials Cloud. In collaboration with the von Lilienfeld group, they are generating a systematic database of elemental solid structures (QMAT-1), to be used as a benchmark and test system for future ML developments. The Ceriotti and Cevher groups collaborated on training sets.

MARVEL-related publications

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

- [1] B. Huang and O. A. von Lilienfeld, *The “DNA” of chemistry: Scalable quantum machine learning with “amons”*, arXiv:1707.04146 (2017).

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- [12] T. Hastie, R. Tibshirani, and J. Friedman, *The Elements of Statistical Learning: Data Mining, Inference, and Prediction, Second Edition* (Springer, New York, 2009).
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Platform Project 6

PP6 — Informatics

Project leader: Thomas Schulthess (CSCS and ETHZ)

Participating members: Thomas Schulthess (CSCS and ETHZ), Nicola Marzari (EPFL), Daniele Passerone (Empa), and Stefan Goedecker (UniBas)

1 Main goals and achievements

This project coordinates the development of the infrastructure underpinning the Open Science Platform of phase II; namely, the development and deployment of AiiDA and of the Materials Cloud, the integration of these services at CSCS, and the delivery of an optimal HPC infrastructure that is able to take advantage of novel hardware developments (e.g. GPUs) and that is verified and validated. Year 4 has seen core updates to AiiDA (new workflow engine, new plugin system, full REST API), together with proof-of-concept efforts for services at CSCS (web servers, databases, object storage, and authentication and authorization). Materials Cloud has been deployed publicly, with all 5 sections operative (*Learn, Work, Discover, Explore, and Archive*), providing FAIR-compliant data-management plans, and with core developments in the provision of quantum simulation services through the novel Appmode in Jupyter. The SIRIUS domain-specific library is being developed and tested to provide verified results on the new Piz Daint XC50 hybrid CPU-GPU architecture. Version 1.0 of the SSSP (standard solid-state pseudopotentials) has been released. CSCS continues to provision HPC resources to all the MARVEL community, while moving to an Infrastructure-as-a-Service model (where web, data, and storage services are provided), with mirror and federation to other Tier-0 European supercomputing centres, starting from CINECA.

2 Progress of the different efforts

2.1 The AiiDA materials' informatics platform

The AiiDA open-source platform implements the four ADES pillars of Automation, Data, Environment and Sharing as described in [1]. AiiDA is continuously being developed and has matured into an ecosystem with multiple backend options for increased performance and flexibility, a powerful graph querying tool for easy result analysis, a redesigned plugin

system to simplify external user contributions, new and more powerful workflow engine for easy-to-write workfunctions and workchains, and a continuous integration system to ensure the stability of the platform. Since the last report, sustained progress has been made. Three more major versions have been released: v0.8.0 on Apr 24, 2017, v0.9.0 on June 15, 2017, and v0.10.0 on Nov 20, 2017. These three releases have brought major improvements to the workflow system, a fully compliant REST API to interface with the Materials Cloud and a brand new plugin system. The user friendliness of the platform has also been enhanced, by improvements to the command line interface, the documentation and simplification of the installation procedure. Independently of the AiiDA releases, we also provide, via our website, the AiiDA virtual machines of our tutorials and the corresponding exercises.

The plugin system The new plugin system, released with AiiDA v0.9.0, was designed to make it as easy as possible for developers to extend the functionality of AiiDA and allow it to be fully interoperable with any other computational tool or code. This is not just limited to the necessary code input generators and output parsers, but also includes workflows. A schematic overview of the AiiDA plugin system is shown in Fig. 1. Plugin developers can develop and maintain their plugin independently and separated from the core AiiDA repository. This enables full flexibility on how to host and distribute their code. Since the plugin system leverages the

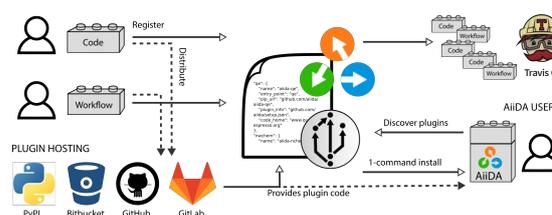


Figure 1: The new AiiDA plugin system.

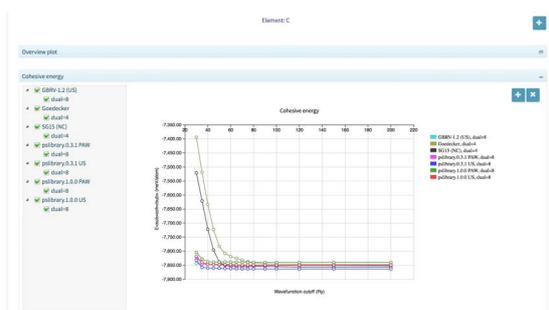


Figure 3: Browsable data in the Discover section: Cohesive energies in the SSSP.

DOI service provided by the ETHZ library).

The architecture of the Materials Cloud is shown in Fig. 4. New backends have been implemented for *Learn*, to allow to organize videos from different sources (youtube, slideshot) in sections and complement these with additional files; and for *Archive*, to provide HTML, JSON, and XML endpoints, and to provide DOIs.

The Jupyter portal has been implemented from scratch, using JupyterHub with a Docker-Spawner that launches docker instances per each user, with home directories mounted locally. Moreover, an “app mode” has been developed to provide a seamless user interface to Jupyter notebooks. Each tool (e.g. seekpath) is now distributed and deployed as a docker container. The whole infrastructure has been migrated to the very new OpenStack service at CSCS. Ansible scripts for each server have been written to completely automate the deployment. Currently, 7 servers are deployed to deliver various microservices, and most of the servers are duplicated for development. Data in the *Archive* and *Learn* section are stored in the object store Swift service provided by OpenStack. The infrastructure is already online and is undergoing a beta phase for a few months, where feedback is collected from

MARVEL members to improve its usability.

As an additional deliverable, the quantum mobile (github.com/marvel-nccr/quantum-mobile) virtual machine has been released, containing AiiDA and a number of popular simulation software packages (including Quantum-ESPRESSO, CP2K, Yambo, FLEUR, SIESTA), automated AiiDA workflows for properties calculations, a job scheduler, and a number of useful tools (torque, openmpi, xmgrace, gnuplot, xcrysden, jmol).

Beside these activities, regular meetings (every 2 – 3 weeks) with CSCS take place, in addition to a 3-day visit of the whole EPFL team to CSCS to coordinate the development and deployment of the novel services at CSCS and to discuss about technical aspects, security and legal (contractual) details. Moreover, regular meetings are being held with the EPFL TTO to prepare all legal documents (terms of use, licenses for codes and data, etc.) needed for the dissemination platform. Finally, discussions are ongoing between EPFL and CSCS to draft the contract between MARVEL and CSCS for a long-term storage service that would make it seamless for researchers to comply with data management plans when they publish data on the Materials Cloud.

2.3 AiiDA and Jupyter for one-dimensional materials

Concerning low dimensional carbon-based nanomaterials, simulation strategies have been consolidated taking into account the new needs from the experimental side. The recent success of Empa’s nanotech@surfaces laboratory in demonstrating a graphene-nanoribbon field-effect transistor (FET) with characteristics comparable to standard FETs [23] required standardization of procedures for the calculation of electronic and optical properties beyond DFT. The collaboration with Ivano Tavernelli [4] from the IBM group allowed to apply TDDFT methods to recognize the chemical termination of armchair nanoribbons from optical fingerprints, while collaborations with Jan Wilhelm from the Hutter group and Leopold Talirz from the Smit group allowed to establish and test an approach to compute the electronic properties of low-dimensional materials at the GW level within an eigenvalue self-consistent approach that scales quadratically up to thousands of atoms [5]. The novel approach, that will be integrated in the AiiDA based work-package for 1D nanomaterials developed by Ole Schütt and Aliaksandr Yakutovich, allows to employ atomistic models that match the size

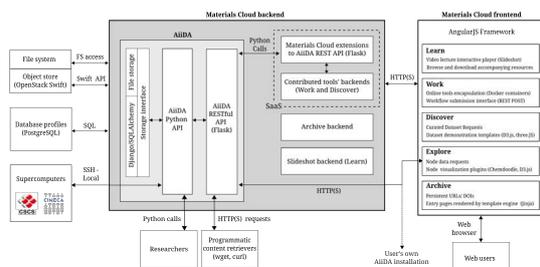


Figure 4: The architecture of the Materials Cloud, leveraging AiiDA and federated supercomputing services providing HPC, database solutions, and long-term storage.

and the structural details of the nanomaterials (such as heterojunctions) that are fabricated in the laboratory. Concerning development of tools for the community, in addition to the Two-Probe Particle Model developed by Aleksandr Yakutovich in collaboration with Marcella Iannuzzi from the Hutter group, to simulate non contact AFM images, the Appmode results obtained by Ole Schütt have to be mentioned. In fact, the workflow that was developed to fulfill the need to simulate nanoribbons in a standardized and high-throughput manner, was redesigned and embedded in the Materials Cloud project with the aim of creating an ecosystem of turn-key solutions, ie. apps. The workflow takes as input a structure and runs a sequence of Quantum-ESPRESSO calculations. As output it provides a thoroughly optimized geometry, the band structure, relevant Kohn-Sham orbitals, the spin density, and the projected density of states. In order to make the workflow and its results accessible to experimentalists, a custom web application was created using Jupyter. To enhance the user experience, a novel Jupyter extension, called Appmode, was developed. This combination allows for the creation of web applications in pure Python and to leverage the ecosystem of existing libraries including ipywidgets, matplotlib, bqplot, nglview, and ase. Since Python knowledge is widespread in the community, many scientists will be able to contribute new apps. In order to create an ecosystem of apps, they have to be easily shareable among scientists. The major hurdle in this respect is the complex software stack upon which apps typically rely. As a solution the software stack was standardized using Docker containers, which provide every user with the identical software environment. In order to minimize the entrance barrier for new users, the containers are hosted in the novel OpenStack cloud at CSCS using JupyterHub. Fig. 5 gives an architectural overview.

We believe that the combination of AiiDA, Jupyter, Appmode, and Docker provides a very suitable architecture for building turn-key solutions. While the nanoribbon workflow can be considered a proof of concept, more apps are already under development. Since quantum-chemical simulations usually employ very specific approximations, the Materials Cloud will have to accommodate numerous turn-key solutions to serve the community at large. It is therefore important to focus on scalability by enabling as many scientist as possible to contribute to the Materials Cloud.

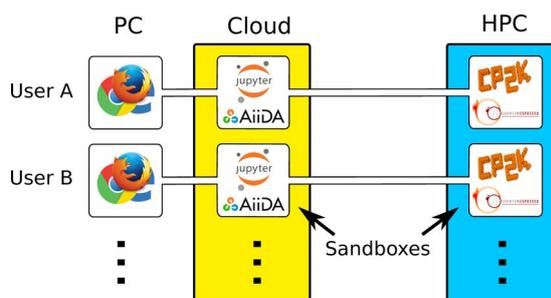


Figure 5: Materials Cloud services: End-to-end sandboxing provides each user with identical software environments; this ensures both reproducibility and security. The GUI runs as JavaScript in the user’s browser and the application logic runs together with the AiiDA middleware in Docker containers. These containers are hosted at the CSCS OpenStack cloud; eventually, also the HPC calculations will be sandboxed via Shifter containers.

2.4 ETHZ/CSCS software development services

As a partner of the MARVEL Informatics platform, ETHZ/CSCS is involved in two software projects for the Materials Cloud database.

a) *SIRIUS-enabled Quantum-ESPRESSO on GPUs*
Quantum-ESPRESSO (QE) is one of the flagship material science codes widely used by the MARVEL community and outside of it. Unfortunately, it is also one of the codes that still lacks the official support for GPU devices, which makes it inefficient on the Piz Daint hybrid supercomputer. To overcome this problem CSCS is working on a domain specific library for electronic structure calculations, SIRIUS [6]. The goal of SIRIUS is to separate and optimize the low-level quantum engines from the electronic structure codes [7]. The library supports several most popular diagonalisation-based methods, such as plane-waves (PW) pseudopotential and projected augmented waves (PAW), as well as full-potential linearized augmented plane waves (LAPW). The following features have been implemented in SIRIUS and interfaced with the QE code:

- non-magnetic, magnetic collinear and non-collinear ground state (“scf” type of calculations),
- atomic forces and stress tensor (“relax” and “vc-relax” type of calculations),
- spin-orbit interaction,
- Gamma-point treatment.

At the end of Oct 2017, CSCS has made a first release of the SIRIUS-enabled QE code for beta testing and to receive feedback from the users (Figs. 6 and 7). Based on this feedback we plan

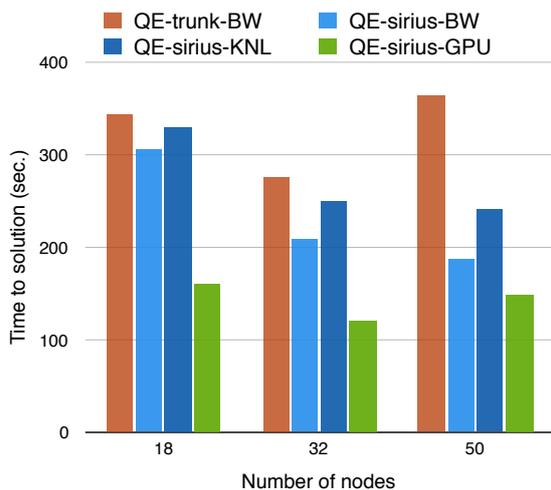


Figure 6: Performance benchmark of QE and SIRIUS-enabled QE for a 288-atom unit cell of Pt cluster embedded in the water. The runs are performed on dual socket 18-core Intel Broadwell @2.1 GHz nodes (BW), on hybrid nodes with 12-core Intel Haswell @2.5 GHz + NVIDIA Tesla P100 card (GPU) and on nodes with 64-core Intel Xeon Phi processor @1.3 GHz (KNL).

to continue the development and optimization of QE/SIRIUS in the following way:

- LDA+U+V ground state calculation, correction to stress tensor and forces,
- tweaking of the iterative solver,
- tweaking of the GPU code for small unit cells and large number of k -points,
- tweaking for the KNL system.

b) *AiiDA plugins for Exciting code* During the past year, AiiDA has introduced a new plugin engine to decouple the external code plugins

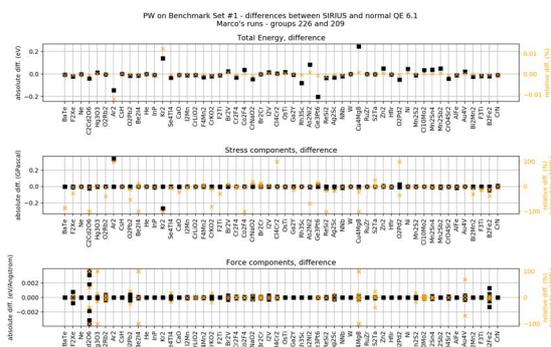


Figure 7: Differences between normal and SIRIUS-enabled QE results for total energies, stress tensor components and atomic forces on a reference high-throughput dataset of 56 materials (insulators, metals, magnetic). The main source of discrepancy comes from the different numerical integration schemes of the radial functions.

from the main AiiDA source and a new workflow systems that allows to keep a full provenance track of the complex calculations. This had an impact on the development of AiiDA Exciting plugins:

- the Exciting code plugin was adapted to the new AiiDA engine and moved to a separate repository (github.com/electronic-structure/aiida-exciting);
- equation of states (EoS) plugin was adapted to a new workflow system;
- stress tensor plugin was implemented for the old workflow system and now is being adapted to a new workflow system (github.com/cocteau/twins/stress-tensor-for-AiiDA).

2.5 Verification and validation

We have released version 1.0 of our curation effort for standard solid-state pseudopotentials (SSSP, Fig. 3), that is now fully available on the Materials Cloud with clickable data and AiiDA graphs for all properties (equation of state, phonons, bands, formation energies, stress tensors). An ongoing collaboration with the group of S. Cottenier (U. Ghent) is dedicated to extending the testing protocols to binaries and multinaries — notably, the group has adopted AiiDA for their VASP workflows. Ongoing collaborations include also the Goedecker group, for verification against wavelet calculations, and the group of C. Wolverton, for verification of formation energies against VASP, and validation against experimental results.

2.6 CSCS service provisioning

CSCS is responsible for the hardware infrastructure of MARVEL as well as provisioning core services, such as computing, storage, identity management and security. Furthermore, CSCS is engaged in collaboration with other HPC centres in order to build a federated infrastructure that should benefit MARVEL as well. At the end of 2016, the necessary computing infrastructure (initially installed at CSCS at the end of 2014) was upgraded from Haswell to Broadwell processors. Installed hardware now include 180 dual-socket compute nodes (2x Intel Broadwell 18-Core, E5-2695 v4 @ 2.1 GHz), with 64 GBytes of memory. The initially provided scratch filesystem of 2.7 PetaBytes, accessible from all compute nodes, was complemented with an additional 6.2 PBytes Sonexion3000 file system at that time as well. The available resources are distributed among the

different MARVEL projects in a shape of a compute quota of node-hours per quarter and a storage quota in TBytes. Production workload began on Apr 1, 2015, and resources have been utilized steadily. The permanent storage assigned to MARVEL projects is of 200 TBytes. Utilization of all these resources is reported quarterly.

In order to further develop the Materials Cloud platform, starting from Q4'2016 (and for a period of around 6 months), 4 proof-of-concept (PoC) services have been deployed at CSCS between the AiiDA team and CSCS: web servers, databases, object storage and authentication and authorization infrastructure (AAI). These PoC Services were jointly followed in parallel to the regular collaboration meetings, yielded very good results and expanded the mutual understanding between CSCS and MARVEL at all levels. As a result of the mentioned series of PoC Services, the needs of other communities (such as the Human Brain Project) and the collaboration with JÜLICH and CINECA, a new OpenStack infrastructure service was installed at CSCS and offered to MARVEL to continue deploying the services needed for AiiDA and Materials Cloud, in addition to the existing resources on Piz Daint. This OpenStack infrastructure was presented in a face-to-face meeting in Jul 2017 and the needed services were migrated by the AiiDA team during the following months with the help of CSCS. The work on the new OpenStack infrastructure still continues and work has started in order to provide better resilience, long-term storage and OpenStack-to-HPC bridging, among other things, in preparation for the public release of the Materials Cloud.

3 Contribution to overall goals and initial proposal

The Informatics project is a key long-term structural effort for MARVEL, and we believe it has outperformed its initial plans of the original full proposal, that listed explicitly the 5 goals of 1) developing an infrastructure able to manage high-throughput simulations, 2) establish data standards for database-reading/database-writing, 3) automatically transform a structure into a calculation, 4) encode in workflows all the steps that are needed to obtain a complex property out of a microscopic calculation, and 5) verify and validate the computational results. In addition, we have developed a very close infrastructural link with CSCS, that has allowed to development and deployment of the Materials Cloud,

and we have pushed the efforts of modernizing codes to be able to run on complex, ever-evolving hardware infrastructures, through the SIRIUS domain-specific library. This project has leveraged and supported several other efforts, most notably with our participation as workpackage leaders in the H2020 MaX Centre of Excellence for e-infrastructure (WP3 Ecosystem, 2015 – 18, under renewal), in the H2020 MarketPlace project (WP2, Data and Services, 2018 – 22), and in the Coordination and Support Action for the creation of the European Materials Modelling Council and its standards (2015 – 19).

4 Collaborative and interdisciplinary components

This project is very closely collaborative — the AiiDA team at EPFL has regular meetings within the team (10+ people, leveraging other projects) and with CSCS, that in response of the needs of MARVEL has started working on its deployment of Infrastructure-as-a-Service and has set up a matching-team there, with considerable human resources. Close interactions take place with the MARVEL groups involved in AiiDA and/or Materials Cloud efforts, including, e.g., Leopold Talirz, Aliaksandr Yakutovich and Elsa Passaro (Smit), Felix Musil and Marco di Gennaro (Ceriotti and von Lilienfeld), QuanSheng Wu (Troyer and Yazyev), Tiziano Müller (Hutter), Chiara Ricca (Aschauer), and the Empa effort from the nanotech@surfaces laboratory (Passerone) pioneering services to the experimental counterparts. These collaborations have also made it easy, e.g., for Ole Schütt to move from the VandeVondele group to the Passerone group, where he developed the Appmode in Jupyter, or for Aliaksandr Yakutovich to move from the Passerone group to the Smit group, where he contributes to the Materials Cloud effort of Smit. Outside Switzerland we should mention the AiiDA efforts in FLEUR, Yambo, and SIESTA, through MaX efforts, the ones by the Cottenier group as part of our verification activities, and by SINTEF as part of their industrial licence to VASP. CSCS and CINECA are working on mirroring parts of the Materials Cloud between each other, as a pilot on federated services across supercomputing centres. Last, we are working with a Swiss company (MPDS, owners of the Pauling file) on a model for cross-referencing the computational results of MARVEL against the experimental results in their databases and their online platform.



MARVEL-related 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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Platform Project **7**

PP7 — Experiments

Project leader: Frithjof Nolting (PSI)

Participating members: Frithjof Nolting (PSI), Michel Kenzelmann (PSI), Pierangelo Gröning (Empa), Raffaella Buonsanti (EPFL), Claudia Cancellieri (Empa), Thomas Lippert (PSI), Marisa Medarde (PSI), Daniele Pergolesi (PSI), Wendy Queen (EPFL), Marco Ranocchiari (PSI), Marta Rossell (Empa), Christian Rüegg (PSI), Thomas Schmidt (PSI), Thorsten Schmidt (PSI), Ming Shi (PSI), Grigory Smolentsev (PSI), Urs Staub (PSI), Dirk van der Marel (UniGE)

1 Main goals and achievements

The general aim of PP7 for the fourth year was to bring the individual projects towards a successful end and to prepare for a closer integration between experiment and theory/modeling in the Design and Discovery projects for phase II. Five projects from the first call received an extension of a few months, for which they had to submit a short proposal. Almost all 15 projects from the first and second call will be finished by April 2018. One new 6-month project on modeling spin-waves (section 2.6) was started, which is a first project towards the Open Science Platform — Computational Spectroscopies and Microscopies.

2 Progress of the different efforts

VP1

2.1 $R\text{NiO}_3$ perovskites: exploring the boundary between localized and itinerant behavior

Marisa Medarde — PSI, Antoine Georges — UniGE, Nicola Spaldin — ETHZ, from Aug 2015, Dariusz Gawryluk

The activity during the last year was focussed on the investigation of the lattice and magnetic anomalies of $R\text{NiO}_3$ perovskites (R = rare

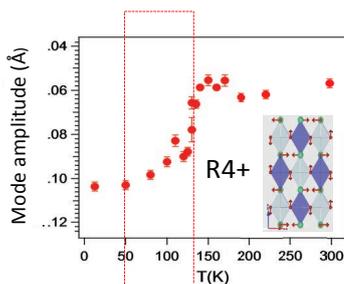


Figure 1: Temperature dependence of the $R4^+$ distortion mode amplitude in PrNiO_3 across T_{MIT} .

earth and Y) using synchrotron and neutron diffraction. Our results, supported by the inelastic neutron scattering measurements performed together with S. Petit (LLB Saclay), and by the calculations of our theory partners Claude Ederer [1], Antoine Georges [2, 3], and P. Ghosez (Uni Liège) [20], clearly favor the existence of two distinct Ni sites with $S \sim 0$ and $S \sim 1.6\mu_B$ and a collinear magnetic order. This result has important implications for the magnetic interactions and should indicate up to which extent their magnetism can be exclusively described by localized magnetic moments. We also investigated the lattice anomalies across the metal-insulator transition (MIT) and the associated breathing distortion for the full nickelate family. Our results are again in excellent agreement with the calculations of our theory partners [1, 2, 3][20], which used them to explore the capabilities of DFT+U for describing materials intrinsically close to the boundary between localized and itinerant behavior. For PrNiO_3 , where the MIT coincides with the Néel temperature, we analyzed the lattice distortions across $T_{MIT} = T_N$ in terms of symmetry-adapted distortion modes, and found that *all* mode amplitudes display anomalous behavior between T_{MIT} and $T = 0.6 T_{MIT}$ (~ 80 K), Fig. 1. These findings suggest that the symmetry of the insulating phase could be actually lower, in line with the theoretically predicted appearance of ferroelectric distortions below T_N [21].

2.2 Testing ultrafast processes in condensed matter

Urs Staub — PSI, Philipp Werner — UniFR, Nicola Spaldin — ETHZ, from Sept 2015, Michael Porer

Simulations of Hubbard-type models within the framework of dynamical mean-field the-

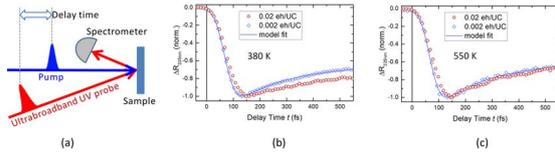
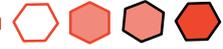


Figure 2: (a) UV-pump/broadband-UV probe experiment. (b,c) Transient reflectivity of a 17 nm thin film of NiO on MgO substrate probed close to the band gap (335 nm) after photoexcitation with 266 nm below (b) and above (c) the Néel temperature in two regimes of excitation intensity (electron-hole (eh) pairs per unit cell (uc)).

ory have predicted a series of novel transient states in antiferromagnetic (AF) Mott insulating systems. Using ultrafast pump-probe techniques, we aim for first experimental tests of key predictions and to provide feedback for an improved and material specific modeling. We tested predictions on the relation between an AF spin background and the relaxation dynamics of photoexcited carriers in Mott-Hubbard insulators by UV-pump/UV-probe studies (Fig. 2a) on AF NiO (collaboration M. Chergui, EPFL). We monitored the carrier cooling by recording the transient optical reflectivity ΔR around the band gap after above band gap photoexcitation. Fig. 2b and c show ΔR for excitation in two doping regimes with a base temperature below and above the Néel temperature, respectively. The timescale of carrier accumulation at the band edges (i.e the cooling) is reflected by the rise time of ΔR . We observe a rise dynamics independent on the initial magnetic state and a slight slow-down at increased excitation intensity. Similar timescales in both magnetic phases might indicate that short-range spin correlations, which are also present in the paramagnetic phase, provide the dominant scattering channel/heat reservoir independent of long-range order. In addition we studied the magnetic dynamics in TbMnO₃, where we found a slow demagnetization after photoexcitation [4] that could be related to trapped magnetic states reported by theory.

2.3 Mott localization in CaVO₃

Thorsten Schmitt — PSI, Nicola Spaldin — ETHZ, from Jan 2016, Daniel McNally

Bulk CaVO₃ and SrVO₃ are archetypal examples of weakly correlated metals, that were recently reported to undergo a metal-insulator transition (MIT) when synthesized in ultra-thin film form. By means of X-ray absorption spectroscopy and resonant inelastic X-ray

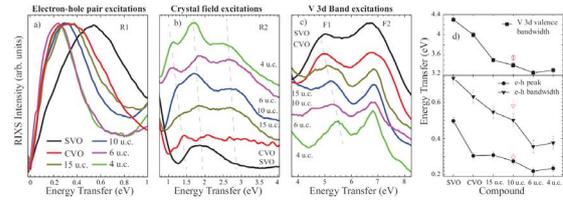


Figure 3: (a,b,c) RIXS of the different valence-band excitations as a function of energy transfer for the films indicated for an incident energy of 520 eV. SrVO₃ and CaVO₃ are 50 uc films and also shown are 15 uc, 10 uc, 6 uc and 4 uc CaVO₃ films. (d) Top: V 3d bandwidth of the two features F1 and F2 for different films indicated, as extracted from the data in (c). Bottom: bandwidth and peak position of the electron-hole pair excitations for different films indicated. All data were recorded at a temperature of 20 K and normalized to the total integrated intensity and offset by an additive constant for clarity except data in (a) that were multiplied by small factors for clarity.

scattering (RIXS) at the vanadium $L_{2,3}$ edges, we investigated the evolution of the valence band excitations across the MIT in thin films of CaVO₃ grown by pulsed laser deposition on SrTiO₃ (001) substrates for different thicknesses. We find a continuous reduction in V-O hybridization and the bandwidth of the charge excitations with reduced film thickness, as well as a modified crystal field (Fig. 3). The 10 uc film undergoes a temperature-driven MIT with analogous evolution of the RIXS response. This work shows that the MIT in ultra-thin CaVO₃ films is of more Mott than Anderson type, which is of importance for the future rational design of correlated oxide heterostructures. The experimental results were compared to DFT+DMFT calculations of CaVO₃ under tensile strain and thickness confinement performed within VP1. This comparison highlights that the combined effects of strain and dimensionality need to be considered in order to rationalize the development of the XAS and RIXS spectra as a function of film thickness.

2.4 Raman spectroscopy of transition metal oxides

Dirk van der Marel — UniGE, Nicola Spaldin — ETHZ, from May 2015, Adrien Stucky

We investigated the Raman active modes of a new series of unique single crystals of rare earth nickelates and BaBiO₃ films prepared by the PSI team (Marisa Medarde and Nicholas Plumb). The small size of these nickelate crystals poses a challenge to the experimental method, but the results are promising and

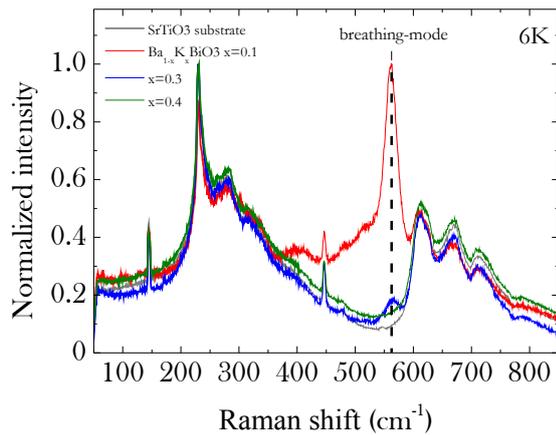


Figure 4: Raman spectroscopy of $Ba_{1-x}K_xBiO_3$ with 3 doping level (red, blue and green) and $SrTiO_3$ substrate (gray) at 6 K.

several additional measurements are planned. The Raman spectra of $x = 0.1$ K-doped $BaBiO_3$ (Fig. 4) clearly shows the breathing mode peak while in the $x = 0.3$ and 0.4 samples, the same feature gets progressively weaker. Further analysis (in progress) aims at understanding the origin of the other strong features at low wavenumber and intermediate temperatures.

2.5 Single-band Hubbard model in new fluorides

Christian Rüegg — PSI, Nicola Spaldin — ETHZ, Thomas Schulthess — ETHZ, Matthias Troyer — ETHZ, from May 2016, Fan Xiao

This PP7 project aims to experimentally verify the predictions from an *ab initio* search concerning the properties of materials potentially realizing a single-band Hubbard model [5]. During the last twelve months we have achieved many of the goals as set in the original proposal. We have synthesized the first single crystals of the candidate material $NaCuF_3$ and the achieved crystal sizes and quality are suitable for all planned experiments. We successfully performed an inelastic neutron scattering (INS) experiment on the spectrometer MERLIN (ISIS, UK) on these single crystals and revealed the excitation spectrum. The compound shows a two-spinon continuum, characteristic of a one-dimensional Heisenberg spin chain along the $(-1,1,0)$ direction in an insulating material (Fig. 5). A new band structure calculation was then provided by our theoretical MARVEL collaborators (Spaldin’s group) based on our findings and the now confirmed crystal structure of the material. We carried out first single-crystal neutron diffraction experiments on the instrument ZEBRA (SINQ, PSI), and determined the 3D long-range ordering temperature and or-

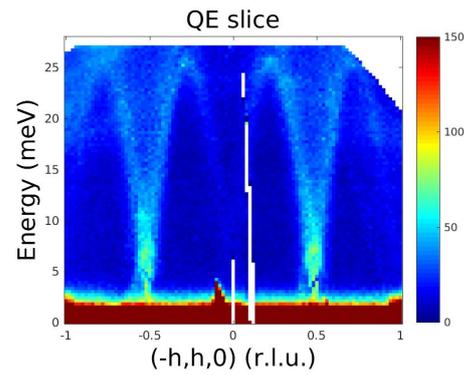


Figure 5: Magnetic excitations in $NaCuF_3$. Two-spinon continuum measure by neutron spectroscopy on single crystals.

dered spin structure in $NaCuF_3$. The results will help to establish the complete Hamiltonian of the material including interchain interactions.

2.6 Modeling spin-waves with SpinW

Christian Rüegg — PSI, Thomas Schulthess — ETHZ and CSCS, from Sept 2017, Sinom Ward

This PP7 project is primarily based around the SpinW code for numerically simulating magnetic structures and excitations of a given spin Hamiltonian using classical Monte Carlo simulation and linear spin-wave theory. The aims are to modularize, improve and develop this code so that it can be used to simulate big datasets which result from neutron spectroscopy experiments and to implement advanced statistical analysis. Spectroscopy data in combination with such an analysis will enable, e.g., to benchmark *ab initio* calculations of exchange interactions.

We have analyzed the existing SpinW code and have started re-factoring it into two sub-modules, the first dealing with structural and exchange manipulations and the second to compute the associated spin-wave dispersions. The first module will be able to interface with a number of numerical solvers. The SpinW code was originally single threaded. We have analyzed the most computationally expensive parts of the code and made them both multi-threaded and multi-processor capable. The result is an order of magnitude increase in computational speed, which enables efficient simulations of complex spectra. Together with IDIDS and CSCS the code will be further optimized and new statistical tools will be implemented.

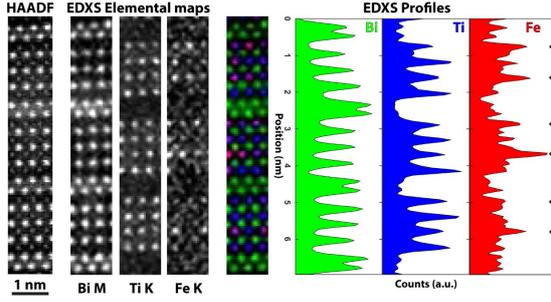


Figure 6: Atomic-resolution elemental maps of the $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ thin film grown on NdGaO_3 . The preferential sites for Fe cations are marked by the black arrows.

2.7 Atomic-scale investigation of magnetic ion ordering in Aurivillius $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ thin films

Marta Rossell — Empa, Claude Ederer and Nicola Spaldin — ETHZ, from June 2016, Marco Campanini

Aurivillius phases ($\text{Bi}_2\text{A}_{m-1}\text{B}_m\text{O}_{3m+3}$) are a family of ferroelectric compounds displaying a layered-perovskite structure (m : number of layers), particularly interesting for their high Curie temperatures and low fatigue [22]. The possibility of incorporating magnetic ions into the perovskite structure paves the way for the design of new multiferroic materials.

Ab initio calculations [23, 24] predicted that the epitaxial strain could play a pivotal role in controlling the Fe cations distribution in $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ thin films. The goal of our project is to understand the microscopic relationship between the epitaxial strain and the magnetic ion distribution, and to correlate the structure to ferroelectric and magnetic properties. Such analysis has been carried out by probe-corrected scanning transmission electron microscopy (STEM), combining different imaging (HAADF, ABE, DPC) and analytical (EDXS) techniques.

$\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ thin films were grown on different substrates (NdGaO_3 and DyScO_3) with different lattice mismatches. Our results demonstrate that despite the mismatch due to the different lattice parameters, in the thin films, the strain is released due to the formation of anti-phase boundaries. The Fe distribution was elucidated by atomically-resolved EDXS (Fig. 6). In the unstrained Aurivillius phase, the preferential configuration is mixed (Fe cations occupy an inner and an outer site of the perovskite blocks) as predicted by DFT.

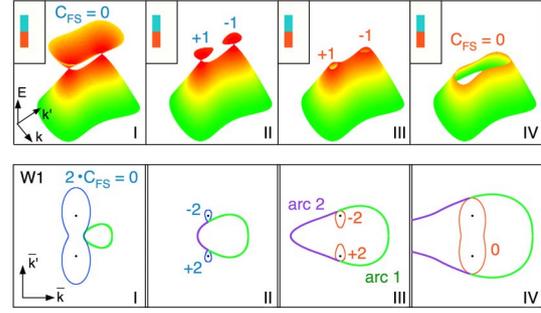


Figure 7: Illustrations of the Fermi surface (FS) topology and FS Chern number evolutions with chemical potentials in Weyl semimetals. Low panels: corresponding evolution of the Fermi arc surface states in TaP and NbP.

2.8 ARPES studies of novel topological semimetals

Ming Shi — PSI, Oleg Yazyev — EPFL, Matthias Troyer — ETHZ, from Sept 2016, Mengyao Yao

Continuing the efforts in realizing novel topological materials and understanding the evolution of electronic structure in the transition from a topological trivial phase to a topological nontrivial phase, we have made several important progresses in the studies of topological semimetals by joint experimental and theoretical investigations. These include:

(i) We revealed that there is a distinct evolution of Weyl fermions and Fermi arcs in Weyl semimetals (Fig. 7) [6]. While Weyl fermion quasiparticles exist only when the chemical potential is located between two saddle points of the Weyl cone features, the Fermi arc states extend in a larger energy scale and are robust across the bulk Lifshitz transitions. These findings not only provide insight into the relationship between the exotic physical phenomena and the intrinsic bulk band topology in Weyl semimetals, but also resolve the apparent puzzle of the different magneto-transport properties observed in TaAs, TaP, and NbP, where the Fermi arc states are similar.

(ii) Since MoP_2 and WP_2 were predicted to be a new type (type II) of Weyl semimetals [7], it attracted great attention to verify this prediction experimentally. Our stage ARPES results on recently available WP_2 samples provide insights about the electronic structure of this transition metal diphosphides [8], which buttress the prediction and could account for the transport properties of this material.

VP2

2.9 Theory and experiment synergy for artificial photosynthesis

Thomas Lippert — PSI, Nicola Marzari — EPFL, from Jun 2015, Wenping Si

In its final stage, the research project focused on three main aspects of fundamental design and application of oxynitride semiconductors: (i) the role of the crystallographic orientation of the surface, (ii) the effects of the N substitution into the O site, and (iii) the effect of different co-catalysts.

It was for the first time proved that different crystallographic surface orientations have different electrochemical properties (Fig. 8). In particular epitaxial LaTiO_2N films (001)-oriented show almost 5 times higher adsorbed photon to current conversion efficiency compare to polycrystalline samples [9]. The role of the nitrogen substitution was investigated by comparing the results of X-ray absorption and emission spectroscopy of a La-Ti oxide and a La-Ti oxynitride samples with the same crystal structure. The N incorporation reduced the total band gap by affecting both band edges [10]. Finally the role of different co-catalysts and passivation layers was investigated to improve performance and stability during operation using La-Ti oxynitride powder samples [11].

2.10 Dynamic surface self-reconstruction is the key of highly active perovskite electrocatalysts for water splitting

Thomas Schmidt — PSI, Nicola Marzari — EPFL, from Aug 2015, Daniel Abbott

The main goal of the present project is to provide a fundamental understanding of the water splitting reaction on perovskite electrocatalysts, which could represent a breakthrough in the development of cost-effective water electrolyzers. The novelty of this research project lies in the use of operando X-ray absorption spectroscopy (XAS) which can provide dynamics of the electrocatalyst electronic and local structure. In a recent publication in *Nature Materials* [12] we have revealed, for the first time, by operando XAS that the electronic and local structure of perovskite oxide catalysts changes during the water splitting reaction. Particularly, we could demonstrate that the key for highly active catalysts is a self-assembled, (oxy)hydroxide top layer. This is completely different from the message of several water electrolysis-related publications, which con-

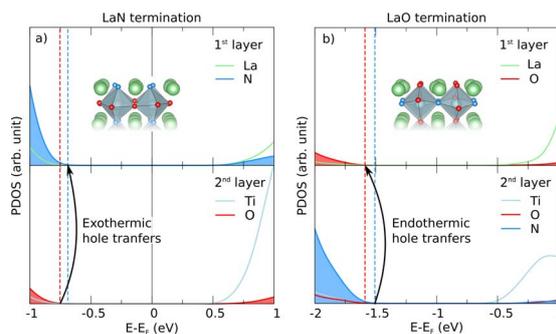


Figure 8: Layer-resolved electronic densities of states for the first and second layer of a) the LaN terminated (001) surface and b) the LaO terminated (from [9]).

sider the surface of oxide catalysts as an ideal, atomically flat surface. This new concept completely revolutionizes the currently most accepted view of design principles for highly active perovskite catalysts. It also points towards the paramount necessity of investigating other perovskite properties under operando conditions in order to develop accurate design principles for highly active perovskite catalysts. An active collaboration with the group of Nicola Marzari at EPFL has been established and it has already resulted in several joint publications [13, 14].

2.11 *In situ* structural and electrochemical characterization of phase transformations at oxide-liquid interface

Claudia Cancellieri — Empa, Alfredo Pasquarello — EPFL, Daniele Passerone — Empa, from Jan 2016, Fabio Evangelisti

In the first part of our MARVEL project, the microstructure and electronic properties of amorphous Al-oxide films, crucial for applications in the fields of, e.g., corrosion-protection, catalysis and adhesion, were investigated and compared with their crystalline polymorphs [15]. Through successful collaboration between experimentalists and modelers, fundamental understanding of the interrelationships between the growth conditions, the oxide density and the dielectric properties of amorphous oxides has been rationalized as a function of the growth anodizing potential. Our approach consisted in combining different experimental techniques: spectroscopic ellipsometry, X-ray photoelectron spectroscopy (XPS), SEM, TEM, electrochemical impedance spectroscopy (EIS) and scanning Kelvin probe force microscopy (SKPFM) to derive a methodology to assess electrical properties in amorphous ox-

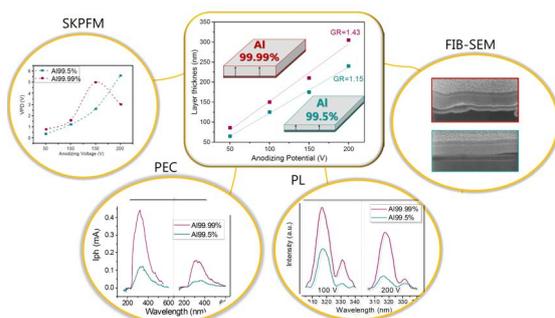


Figure 9: Schematic summary of the second year of MARVEL activity. Al anodic barrier oxides on Al substrates with 2 different purities.

ides which could be modeled by theory. As a next step, we wanted to identify the type of structural defects, which govern the density and electric properties of the amorphous oxide phase. Applied research of amorphous and highly defective oxides is an extremely relevant industrial topic which can indeed attract interest of industrial partners already involved in Empa activities. We investigated oxide grown on Al substrate with different purities to rationalize the role of impurities and defects in determining the oxide properties. In Fig. 9 a summary of the experimental findings of anodic Al oxide grown on 2 different purities substrates are represented. The presence of defects was assessed by measuring defective properties by photoluminescence (PL) and photo-electrochemistry (PEC). Surprisingly the oxide grown on the less pure substrate contains less defects and has less roughness than the one grown on the purer Al, indicating that the presence of foreign atoms stabilizes the oxide and its properties [16].

2.12 X-ray absorption study of $\text{Co}_4^{\text{II}}\text{O}_4$ cubane water oxidation catalyst

Grigory Smolentsev — PSI, Jürg Hutter — UZH, from Apr 2016, Nicolò Azzaroli

$\text{Co}_4^{\text{II}}\text{O}_4$ -dpc cubane catalyst represents the first molecular water oxidation catalyst with the characteristic $\text{H}_2\text{O}-\text{Co}_2(\text{OR})_2-\text{OH}_2$ edge-site motif which is expected in the most efficient heterogeneous water oxidation catalysts based on Co-oxides. We have investigated this catalyst experimentally under operando conditions and using freeze-quench approach. Theory partners explored the same catalyst using the density functional theory based molecular dynamics (DFT-MD) simulations of the cubane in a box of water molecules. Re-

sults are reported in our joint publication [17]. To determine the average oxidation state of catalyst under operando conditions and under light illumination we have measured Co K edge X-ray absorption near edge structure (XANES) spectra with the time-resolution ~ 3 minutes. These data demonstrate that the Co(II) centers are readily oxidized to Co(III) or higher oxidation states with slow recovery. Structural information on $\text{Co}_4^{\text{II}}\text{O}_4$ -dpc after illumination was obtained based on the extended X-Ray absorption fine structure (EXAFS) analysis. The chemical oxidation process was monitored with different equivalents of $[\text{Ru}(\text{bpy})_3]^{3+}$ using freeze quench XANES spectroscopy in search of intermediate oxidation states. Results demonstrate that complete conversion of the Co(II) centers to Co(III) is not mandatory for O_2 evolution.

2.13 Colloidal nanocrystals as model systems to uncover structure/properties relations in CO_2 electroreduction

Raffaella Buonsanti — EPFL, Nicola Marzari — EPFL, from May 2016, Jianfeng Huang

a) *Novel degradation mechanism of Cu nanocubes based electrocatalysts during electrochemical CO_2 reduction* Understanding the degradation pathways of electrocatalysts of electrochemical CO_2 reduction is essential for developing corresponding mitigation strategies and eventually improving catalyst design for successful implementation of the promising technology. In this project, we have revealed a unique degradation mechanism of copper nanocatalysts. In contrast with the most common coalescence and dissolution/precipitation mechanisms, we found a potential-driven nanoclustering to be the predominant degradation pathway (Fig. 10A, B). In collaboration with the team of Nicola Marzari, we confirmed the role of the negative potential applied to reduce CO_2 as the driving force for the clustering (Fig. 10C). This study opens new venues towards the understanding of the degradation mechanisms of new electrocatalysts for CO_2 reduction and, more generally, other electroreduction reactions.

b) *Geometry-dependent electrocatalytic CO_2 reduction of CuAg bimetallic nanocrystals* Bimetals hold great promise in electrochemical CO_2 reduction, because they are capable of breaking “scaling relationship” of intermediates formed in the reaction pathway from CO_2 to hydrocar-

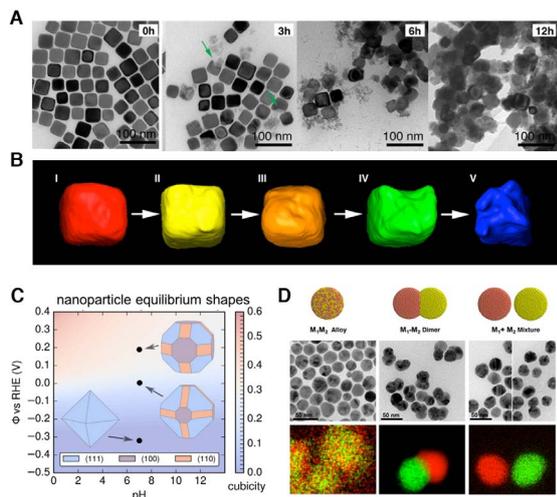


Figure 10: (A,B) Morphological evolution of Cu nanocubes during electrolysis. (C) pH and potential dependence of the Wulff-construction of Cu nanoparticles. (D) Schematics, TEM images and elemental mapping of three types of CuAg nanocrystals: alloy, dimer and mixture.

bons. Nevertheless, the question as to what kind of specific configuration of the two metals should adopt to maximize their potential remains to be answered. In this project, we have successfully developed three types of intriguing CuAg bimetallic nanocrystals, i.e. alloy, dimer and mixture (Fig. 10D). Preliminary results have shown that dimer nanocrystals exhibit a high selectivity up to 35% towards ethylene. Future efforts will be continued to the measurements of the electrocatalytic activities of the other two bimetallic nanocrystals. In addition, a theoretical corroboration of geometric effects on the electrochemical CO_2 reduction will be conducted in collaboration with the theory team led by Nicola Marzari.

2.14 The search for new ionic conductors: High-throughput screening and experimental synthesis and characterization

Daniele Pergolesi — PSI, Nicola Marzari — EPFL, from May 2016, Elisa Gilardi

Compared to the previous year the project extended to the computational study and experimental validation of different ionic conductors. We reported for the first time the differences in the ionic conductivity (via proton and oxygen vacancies) along different crystallographic planes of $\text{Ba}_2\text{In}_2\text{O}_5$ [18]. Also we reported the first measurement of enhanced proton conductivity under controlled biaxial tensile strain in thin films of Y-BaZrO_3 . Results have been con-

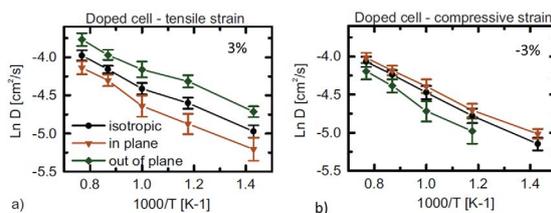


Figure 11: FPMD simulation of the diffusion coefficients Y-BaZrO_3 . The diffusion coefficient at different temperatures is shown for (a) tensile strain and (b) compressive strain for the doped system.

firmed by first-principles molecular dynamic (FPMD) simulations of proton diffusivity. The presence of the dopant and the consequent proton-trapping effect make tensile strain favorable for proton conduction (Fig. 11) [19]. We studied in detail the structure and conductivity of interfaces of Sm-CeO_2 and Y-ZrO_2 . The conductive properties of these interfaces may have remarkable impact on the performance of solid oxide fuel cells.

Concerning lithium ions conductors, the project aims to identify new potential solid-state conductors for use as electrolyte in lithium ions batteries. A first benchmark study identified the tetragonal structure of $\text{Li}_{10}\text{GeP}_2\text{O}_{12}$ (LGPO) as a new promising material. The experimental validation up to now focused on the synthesis of the more stable orthorhombic structure of LGPO following different synthetic routes (solid-state synthesis, ball milling, sol-gel synthesis).

2.15 Metal-organic frameworks invert molecular reactivity: Lewis acidic phosphonium zwitterions catalyse the Aldol-Tishchenko reaction

Marco Ranocchiari — PSI and Wendy Queen — EPFL, Berend Smit — EPFL, from Jul 2016, Gerald Bauer

Metal-organic frameworks (MOFs) have a striking influence when used as additives for the reaction of *n*-alkyl aldehydes in the presence of methylvinylketone and triphenylphosphine. In the absence of a MOF, the expected Morita-Baylis-Hillman product — α β -hydroxyenone — is observed. In the presence of MOFs with UMCM-1 and MOF-5 topologies, the reaction is selective to Aldol-Tishchenko products — the 1 and 3 *n*-alkylesters of 2-alkyl-1,3-diols — that is unprecedented in organocatalysis (Fig. 12). The (3-oxo-2-butenyl)triphenylphosphonium zwitterion, a commonly known nucleophile,

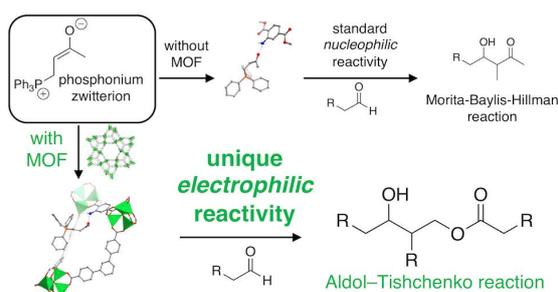


Figure 12: Schematic summary showing the influence of MOF additives to drive a phosphonium zwitterion intermediate towards unprecedented reactivity.

is identified as the catalytic active species (Fig. 12). This zwitterion favors nucleophilic character in solution, whereas once confined within the framework, it becomes an electrophile yielding Aldol-Tishchenko selectivity. The role of the catalytically active phosphonium zwitterion was studied in solution as well as in the UCMC-1 environment using a mix of density functional theory and force field calculations. Computational investigations reveal a structural change in the phosphonium moiety induced by the steric confinement of the framework that makes it accessible and a Lewis acid. This work shows a novel way of doing catalysis in which MOFs can be used as additive to trap reaction intermediates yielding unprecedented reactivity, inaccessible under standard reaction conditions.

2.16 Small molecule conversion in metal-organic frameworks

Wendy Queen — EPFL and Marco Ranocchiari — PSI, Berend Smit — EPFL, from April 2017, Olga Trukhina

Metal-organic frameworks (MOFs) represent an ideal platform for developing efficient and selective heterogeneous catalysts for biomass valorization that overcomes the main technological limitations of currently employed enzymatic systems. Within this teamed project, we aim to refine the catalytic performance of MOFs by varying pore size/shape and surface functionality of these solids. At the current stage, we have developed a series of stable MOF-based catalysts with highly reactive Lewis acid sites such as Zr(IV) and Hf(IV), which are capable to promote the selective conversion of C5 and C6 sugars. As an example, a porphyrin-based Zr-MOF prepared in Queen's lab showed selective conversion of glucose to mannose in aqueous media, reach-

ing 20% yields with over 70% selectivity, the highest value observed to date. Additionally, we have succeeded in doping the Zr-based robust UiO-67 framework and MIL101(Cr)-SO₃H with Sn active sites, showing a nearly 100% selective conversion of glucose to 5-HMF in water at room temperature. Currently, we are working on the optimization of the yield of this reaction which has reached 12%. Given Queen's group expertise in MOFs chemistry and *in situ* crystallographic characterization, and Ranocchiari's group knowledge in heterogeneous catalysis, we will carry out structure-property correlations using *in situ* diffraction and spectroscopy to help understand how minor changes in the framework structure direct the materials catalytic activity. Our combined and unique expertise will help us unveil mechanisms associated with these chemical transformations and hence certainly promote the advancement of MOFs in the bio-refining processes. Worthy to mention, our collaborative efforts and interdisciplinary focus — in terms of characterization techniques and participants' expertise — fully support the global goal of MARVEL network for the progress of materials science and continuous MOFs design for future commercialization.

3 Contribution to overall goals and initial proposal

The PP7 projects are scientifically contributing to the design and discovery of novel materials with novel physics or improved properties or performance mainly via collaborations with partners from VP1 and VP2. Another important aspect is the building of a joint community of experimental and theory/modeling experts for materials design. We also note the effort of MARVEL to collaborate with the SCCER on Heat and Electricity Storage, with collaborations focusing on electrocatalytic water splitting and CO₂ reduction.

4 Collaborative and interdisciplinary components

The concept of PP7 is based on the collaboration between an experimental and a theory/modeling group for a single project. In 2017, one new project was started adding a new collaboration between PSI and the CSCS/ETHZ. The partners of the different projects have regular joint meetings and, at a one day PP7 workshop at PSI, all groups of PP7 and also several PhD students and post-docs from MARVEL, which are not involved in PP7, met and discussed the research projects.

MARVEL-related 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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- [10] M. Pichler, J. Szlachetko, I. E. Castelli, N. Marzari, M. Döbeli, A. Wokaun, D. Pergolesi, and T. Lippert, *Determination of Conduction and Valence Band Electronic Structure of LaTiO_xN_y Thin Film*, ChemSusChem **10**, 2099 (2017).
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3.2 New projects

Two projects were already announced in year 3 through the collaborative “Agility Plus” effort and have been added in May 2017, to include two new MARVEL group leaders, William Curtin (EPFL) and tenure-track assistant professor Martin Jaggi (EPFL).

With the remaining money allocated to PP7, two small projects for 6 months were launched led by existing MARVEL members, Christian Rüegg (PSI) and Alessandro Curioni (IBM).

Following the reallocation of the year 4 budget to projects, as presented in the chapter 10. Finances, two further projects started, one for an existing MARVEL member, Berend Smit (EPFL), to disseminate data on the Materials Cloud, and one for a new member, Mathieu Luisier (ETHZ), thanks to the collaboration with Daniele Passerone (Empa), on the properties of low-dimensional devices.

4 Knowledge and technology transfer

Knowledge and technology transfer in MARVEL occurs at different levels: knowledge transfer takes place in the dissemination of open-source simulation codes, training in the use of those codes and of the newly developed materials informatics framework, in the sharing of all results from materials simulations, and verification and validation of the calculations. The technology transfer activities focused this year on approaching companies in five industrial sectors of interest, while developing list of target companies in Switzerland and abroad. This led to numerous meetings with companies and visits on campus, as well as an enhanced visibility thanks to the launch of the Newsletter to Industry and the MARVEL Twitter account. Several negotiations with companies are ongoing. By building an industrial community around MARVEL, and with the help of the Industrial Advisory Board being formed, we aim to nucleate new collaborations and projects with the industrial sector.

Software

AiiDA

AiiDA is an open source computational platform which is presented in detail in the scientific report of PP6. In the past year, 4 major versions of the code have been released, v0.8.0, v0.9.0, v0.10.0 and v0.11.0, on Apr 2017, June 2017, Nov 2017, and Jan 2018, respectively. In addition to the regular team and CSCS meetings, now monthly meetings are held with CINECA in Italy, to allow the native integration of AiiDA inside their supercomputer environment. AiiDA has been presented at various conferences: the Molecular Simulations Software Institute in Berkeley, CA, USA (Feb 2017), the TERATEC conference in Paris, France (June 2017), the International Supercomputing Conference in Frankfurt, Germany (June 2017), the International Workshop on Materials Genome Infrastructure in Beijing, China (June 2017), the plenary public lecture at PASC 2017 in Lugano (June 2017), the Materials Research and Data Science Conference, in Rockville, MD, USA (Sept 2017), and the workshop on interoperability by the European Materials Modelling Council in Cambridge, UK (Nov 2017), to name a few. To support the community, three tutorials have been organized in 2017. The PSI-K, MAX and MARVEL tutorial (nccr-marvel.ch/events/aiida-tutorial-may-2017) with 50 participants in Lausanne (May 29 – 31, 2017), an advanced AiiDA tutorial for MAX and MARVEL members (nccr-marvel.ch/events/marvel-max-meeting-march-2017) with 50 participants in Lausanne

(Mar 6 – 7, 2017) and an AiiDA tutorial for users within ICTP with 75 participants in Trieste, Italy (Jan 24 – 25, 2017). Another tutorial is planned for late spring 2018 at CINECA, Italy. Finally, in close collaboration with the AiiDA plugin developers of the [Quantum-ESPRESSO](#), [FLEUR](#), [SIESTA](#) and [Yambo](#) codes, the second AiiDA coding week was organized in Oct 2017 in Leukerbad, Switzerland. The participants propose a filmed summary on YouTube (youtu.be/GI4mIU7lmbw). A series of short demonstrations of AiiDA features and capabilities is also available on YouTube (youtube.com/playlist?list=PL19kfn4sO-XDUeXu5jwUq9k7TId8mM0).

Materials Cloud

The first public beta release of Materials Cloud took place in Dec 2017. All components in *Learn*, *Work*, *Discover*, *Explore* and *Archive* (besides the new Jupyter section) are now publicly available under www.materialscloud.org, and open for submissions from our scientific partners, i.e. MARVEL, EPFL, MaX, EMMC and NFFA. Accounts for the Jupyter section are available upon request, and the service is currently used and tested by early adopters from EPFL, Empa and FZ Jülich. Materials Cloud has been presented in the form of posters, talks and live demos at the TeraTec Forum (June 2017), the MARVEL Review and Retreat (Sept 2017), the MaX Meeting (Nov 2017, Flo-

rence), the EUDAT conference (Jan 2018, Porto) and the MaX conference (Jan 2018, Trieste). The [Quantum Mobile](#) virtual machine, which makes it possible to run several quantum mechanics codes together with AiiDA on Linux, MacOS and Windows hosts out of the box, has already been downloaded from 19 different countries since its release in Nov 2017 and is becoming a valuable educational resource for the MARVEL community and beyond — it will be used in classes taught by Smit and by Marzari in the spring of 2018.

New releases, open source codes

In Feb 2017 a public release of the Z2PACK software for computing topological invariants from first-principles, tight-binding and k.p models was made available. In Oct 2017, CP2K v5.0 was released. Before, in Jul 2017, the “CP2K User Tutorial on Advanced ab-initio MD methods” was organized by Jürg Hutter at UZH and co-sponsored by MARVEL. 21 researchers and students (~50% MARVEL members, others outside Switzerland) in the field of molecular simulations were provided a survey of the most relevant computational MD tools implemented within the CP2K program package. Mar 2017 saw a public and free release of the WannierTools software for analysis of topological materials (STM and ARPES predictions) based on tight-binding compound description. In Dec 2017, Quantum Mobile (v17.11.0), a virtual machine based on Ubuntu Linux that comes with a collection of quantum simulation codes (Quantum-ESPRESSO, Yambo, FLEUR, SIESTA, CP2K) was released. All codes are set up and ready to be used through the AiiDA Python framework for automated workflows and provenance tracking. Quantum Mobile may be useful for exercises in physics, chemistry and materials science courses, for running quantum simulations without any setup, for experimenting with new codes, but also for managing production simulations on external supercomputers through AiiDA. The Ceriotti group developed and distributed [GloSim](#) for atomistic modeling, and [SketchMap](#) for data mining and visualization. The Schulthess group, in collaboration with the Marzari group, continued the development of the SIRIUS-enabled Quantum-ESPRESSO code and the associated development of a domain-specific library for GPU acceleration of electronic structure codes.

Conferences and collaborations

European and worldwide synergies: MaX CoE, NFFA, EMMC CSA, MarketPlace, and MICCoM

We are actively engaged in many H2020 European projects, that provide strategic synergies with MARVEL’s objectives and goals — these include 1) the [MaX](#) Centre of Excellence on “Materials’ design at the eXascale” (2015 – 18, under renewal), where we lead the WP3 workpackage on the materials’ modeling ecosystem; MaX actively engages the FLEUR, SIESTA and Yambo developers in AiiDA plugins and workflows, as well as CINECA and Jülich in federating services with CSCS; 2) the [NFFA](#) “Nanoscience Foundries & Fine Analysis” (2015 – 19) has an “installation” of theory and simulation, where we provide modeling services to the experimental community (currently, electronic-structure of cathode materials for Li-ion batteries, redox couples in room-temperature ionic liquids); in addition, it has an activity on standards for storing and disseminating experimental data, to which we participate; 3) the Coordination and Support Action for the European Materials Modelling Council (2016 – 19, [EMMC](#)), whose goal is to bring the field of materials modeling closer to the demands of manufacturers (both small and large enterprises) in Europe; 4) the MarketPlace project (2018 – 22), whose goal is to design, create and maintain a sustainable web-based platform providing a central access point for the entire materials community to databases, modeling services, integrated open simulation platforms, and translation and knowledge services. One of the key activities related to these last two items was a joint session at the first EMMC international workshop in Vienna, Apr 5 – 7, 2017, on “The need for Marketplace services: Requirements from stakeholders (model developers, translators, software owners and end users)” chaired by Nicola Marzari and Georg J. Schmitz (Access Technology e.V, DE), and the EMMC Workshop on Interoperability in Materials Modelling (IntOP2017), Cambridge, UK, Nov 7 – 8, 2017. Last, MARVEL participated in a MICCoM international meeting in Paris, May 31 – June 2 ([paris-meeting-2017.uchicago.edu](#)), convened to bring together all the senior principal investigators of computational materials science centers and of major computational materials projects worldwide. The goal was to discuss the respective strategic views in these efforts, in order to seek possible collaborations or by developing complementary capabilities,



and to brainstorm about strategic position papers on the topic.

Conferences organized by MARVEL members

MARVEL members organized or co-organized more than 15 conferences, tutorials or workshops, some of them being also sponsored by MARVEL. All are listed in the NIRA database and the list can be found on the website (nccr-marvel.ch/ctw). The MARVEL School on “Variationally Enhanced Sampling”, organized by members of the Parrinello group took place Feb 14 – 17, 2017, at USI and attracted 48 young researchers (17 from Swiss institutions, 31 from 8 different countries). This school allowed to disseminate concepts and methods developed within MARVEL and enhanced the project visibility outside the Swiss scientific community. The conference “New Trends in Topological Insulators 2017 (NTTI2017) in Monte Verità on Jul 16 – 21, 2017, was co-organized by Oleg Yazyev with MARVEL financial support. It gathered 84 participants (10% from MARVEL) in a friendly atmosphere. More than a half (13) of the 24 invited speakers were junior researchers. As one particularly important topic is the valuation of theoretical proposals of novel materials with regard to their experimental realization, a public webpage for collecting information about all known and predicted materials was implemented, with the capability of experimental materials researchers to freely comment on the predictions (www.topologicalmaterials.org). The list of collected materials will be used for the first curated dataset on topological materials made publicly available via Materials Cloud. Two symposia were sponsored and organized by MARVEL members at the Joint Annual Meeting of SPS and ÖPG, Geneva, Aug 21–28, 2017. Claude Ederer, Antoine Georges and Marisa Medarde proposed the session “Correlated-Electron Physics in Transition-Metal Oxides” which gathered 27 contributions (18 talks and 9 posters), while Marco Gibertini and Oleg Yazyev organized the session “Emergent Phenomena in Novel Low-Dimensional Materials” with 13 talks. Mikhail Langovoy (in the group of Martin Jaggi), Michele Ceriotti and Anatole von Lilienfeld organized the “Machine Learning meets Advanced Manufacturing and Materials Science” workshop that took place on Jan 28, 2018, in the framework of the “Applied Machine Learning Days” at EPFL.

Technology transfer

In 2015, we have prepared a portfolio of MARVEL activities that could be of interest for companies. We have defined 5 industrial sectors of interest.

- Energy harvesting, conversion and storage.
- Information-and-communication technologies.
- Organic crystals/pharmaceuticals compounds.
- Catalysis and chemical synthesis.
- Watch making industry.

In 2016, we have approached companies from these 5 sectors, and started to create a list of “companies of interest” in Switzerland and also abroad. The final aim is to create an industrial community around MARVEL and to increase the number of collaborations. In 2017, we maintained our efforts to reach companies with potential interest in MARVEL research thanks to targeted contacts by email or LinkedIn, events, industrial newsletters and social media (Twitter). We have hired a journalist at 20% to support TT communication. In parallel, we have implemented 2 new collaborations, beside 3 consulting agreements and 2 new NDA signatures.

Creation of an industrial community

Our efforts are going mainly in the direction of large companies which have the potential to use and transfer materials modeling at quantum level. We have approached large Swiss companies and, in parallel, large foreign companies, some of them investing already in the modeling approach, beside the experimental approach in materials research. In one year, we have increased our industrial community from 24 to 56. This represents 80 people that have been in contact with MARVEL at different levels during the last 3 years in the 5 industrial sectors of interest. We have mainly large companies, except in the sector of modeling, in which the 4 companies are SMEs. The following sectors and countries have been engaged:

	watch-making/materials	energy	fine chemistry/pharmaceuticals	ICT/electronics	modeling	total
2017	22	14	10	6	4	56
2016	9	3	8	2	2	24
2015	3	2	4	2	2	13

Countries:

	CH	EU	USA	JP	Total
2017	31	12	4	9	56
2016	10	7	3	4	24
2015	5	5	1	2	13

Increase the awareness/promotion

In 2017, we have participated in several events with a MARVEL booth (roll-up, flyers and MARVEL representatives).

- Computational chemistry for industry, organized by the Swiss Association of Computational Chemistry, Feb 9, 2017, Empa Akademie, Dübendorf.
- *Salon des Technologies et de l'Innovation de Lausanne* (STIL), Mar 2, 2017, Lausanne.
- EPFL Industry Day, Mar 16, 2017, Lausanne.
- ETHZ Industry Day, Aug 29, 2017, Zurich.

Moreover, we have co-organized with CCMX on Oct 5 at Empa a "Materials Day", a full day dedicated to research in materials science. The morning session included a general presentation of MARVEL and 3 MARVEL projects combining modeling and experimental approaches. In the afternoon, the 5 current CCMX challenges have been presented. About 100 participants were attending, among them 23 participants from Swiss industry. Out of these promotion activities, we were able to gather new contacts, which have been used for the "MARVEL Newsletter to Industry". First issue has been sent in Nov 2017 to about 80 industrial contacts. Three issues per year are planned. In Nov, we have also launched a Twitter account (@nccr_marvel) with the aim to relay information from and to scientific, industrial and associative entities involved in materials modeling.

Company visits

In 2017, 5 companies came for a first visit to one or several MARVEL laboratories. Among them, 1 Japanese, 1 German, 1 French, 1 Swiss and 1 industrial research institute from Taiwan. The main topics of interest were machine learning, compounds for lithium ion batteries and metallurgy.

Ongoing discussions/negotiations

- 1 large European chemical company has signed a NDA in Jan 2017. We have

organized a 1.5-day workshop between 4 MARVEL PIs and 20 people on their side. Out of this, 2 propositions of collaboration emerged and are still under negotiation.

- 1 French company has signed a NDA to prepare further discussion.
- 3 contracts of consulting for companies have been started by PIs from EPFL, ETHZ and UniBas.

New signed collaborations

- 1 Swiss start-up has obtained a Commission of Technology and Innovation (CTI) project for 2 postdocs during 18 months in the groups of Anatole von Lilienfeld (UniBas) and Nicola Marzari (EPFL) to develop a tool for "Machine Learning Driven Quantum Chemistry at Break-Neck Speeds: Now-Casting Chemical Reactions for Online Point-of-Sales". The project will start in spring 2018.
- 1 large US company has given 100 kCHF as a gift to Anatole von Lilienfeld to support research in machine learning.

Ongoing collaborations

- 1 large company signed for an AiiDA licence with 2 years of technical support (2016 – 2017).
- 1 Swiss large company signed a 3 year research contract (2015 – 2017) for a PhD.

Industrial Advisory Board

In Sept 2017, we have invited 3 industrial guests to participate in the Review and Retreat and to share with us, during a short meeting, about their main interests regarding research activities within MARVEL. One of them, Erich Wimmer, CEO of Materials Design, a French company active in materials modeling, has accepted to become the chairperson of the Industrial Advisory Board. The latter will be formed in 2018, as soon as the Design and Discovery Projects of phase 2 are confirmed. Our aim is to have one industrial representative in each sector of activity.

Recommendations from the site visit

Greater resources were already allocated to TT thanks to Carey Sargent (hired at 0.2 FTEs), to write "Feature Stories" for the MARVEL Newsletter to Industry and the website. She is managing the Twitter account.

5 Education and training

In year 4, the particular emphasis on junior scientist interactions and stimulation for new collaborations has continued (junior retreat, junior seminars). Also, the preparation of the camp for high-school students has advanced well. More generally, MARVEL has carried on focusing education and training activities on its PhD and postdoctoral researchers, and on the wider community, through training schools and workshops of broad, fundamental interest dedicated to researchers worldwide.

PhD students and postdoctoral researchers

In the MARVEL community

Junior retreat and junior seminars

The third edition of the MARVEL junior retreat took place on Jul 3 – 7, 2017, at Hotel Paladina in Magliaso, Ticino, and was attended by 48 participants (including 10 women), most of whom were MARVEL PhD students and postdocs, as well as 4 INSPIRE Potentials students, from almost all MARVEL involved institutions. Organized by a committee of four postdocs from EPFL and UZH — Senja, Vamshi, Fernando and Vladimir — as a means of vigorously addressing challenges revealed over the course of the first three years of the NCCR, its main goal was to stimulate collaborations between young researchers by proposing realistic research projects to be pursued independently. Oral presentations focused on outlining the topics of interest and the available expertise/tools within each MARVEL research group. The program also included in-

vited talks: the introductory lecture by the MARVEL PI Michele Parrinello, a talk on intellectual property by Mauro Lattuada, from EPFL Technology Transfer Office, and a lecture on industrial applications of quantum simulations by Ansgar Schäfer from BASF. These additional insights together with social activities facilitated the creation a friendly working atmosphere and boosted collaborative brainstorming (Fig. 1). On Jul 7, 9 projects, conceived by researchers from at least 2 and up to 3 institutions, were presented to the whole group. Two proposals were officially presented at the MARVEL Review and Retreat 2 months later. The full story can be found on nccr-marvel.ch/junior-retreat-2017.

The MARVEL junior seminars continue taking place monthly at EPFL to stimulate collaborations between research groups and to maintain a vibrant community. The initiative is described in the Communication chapter.

Education platform

Five more MARVEL distinguished lectures were made available (slideshot.epfl.ch/events/14). The 4 highlight talks presented at the MARVEL/MaX/Psi-k tutorial on “High-throughput computations: general methods and applications using AiiDA” are visible on slideshot.epfl.ch/events/21. All these lectures and tutorial are now directly accessible from the *Learn* platform of Materials Cloud on www.materialscloud.org/learn. Antoine Georges also contributed to educational material (www.college-de-france.fr/site/antoine-georges/_audiovideos.htm) with his open access lectures at the Collège de France.



Figure 1: Third MARVEL junior retreat on Jul 3 – 7, 2017, in Magliaso: group picture made from above; © EPFL, Sandip De.

Success stories

Sandra Luber, MARVEL senior researcher at UZH, was awarded in Feb 2017 an SNSF professorship. Dariusz Gawryluck, MARVEL postdoc in the experimental project PP7, has been hired as tenure track staff member at PSI. Two MARVEL postdocs, Marco Gibertini (EPFL) and Vladimir Rybkin (UZH) made a first step towards scientific independence, being awarded an Ambizione grant by the SNSF.

Outside MARVEL

Courses

In June 2017, Michele Ceriotti taught the EPFL PhD class “Advanced Statistical Methods for Atomistic Simulations”. More than 25 students, including many MARVEL students, participated. Several more students even chose that class without the goal of getting ECTS credits. The demand for this kind of course stresses the importance of offering graduate classes in the subjects that are at the core of MARVEL. The class was also followed by Wycliffe Owmansu, a student from Kenya involved in an ICTP graduate student program, as a pilot for future joint education activities.

At the level of Master students

Courses

The existing offer of computational classes at the Master and PhD level in the participating institutions is available from the MARVEL website, regularly updated (nccr-marvel.ch/references/education-and-training/Courses-Master-level). Furthermore, some students also join MARVEL groups for their semester works or Master’s theses.

For the younger generation

MARVEL high-school summer camp

The MARVEL education team materialized its plans for the high-school students summer camp *Des atomes aux ordinateurs*, à la découverte de la programmation scientifique that will take place on June 25 – Jul 6, 2018 (gymnases.epfl.ch/camp-materiaux-2018). In addition to 6 junior researchers from the Ceriotti and Marzari groups, the EPFL high-performance computing team SCITAS is enthusiastically involved in the organization, and is providing both facilities and instructors. The

scientific content is overseen by Jean-Luc Desbiolles (SCITAS) and Michele Ceriotti (MARVEL). The promotion of the camp benefits from the precious help of the EPFL Study Programs Promotion Service. The camp is open to teenagers aged 16 to 19 years, corresponding to the last two years of high-school, with no high-level in maths or physics prerequisite and no prior programming knowledges required. Half of the places are reserved for girls. Participants may propose a scientific project that could be realized during the second week of the camp. MARVEL will fund the organization costs, and transport or accommodation when needed. This first edition will enable the development of similar camps in other MARVEL-involved institutions.

Other activities

Thibault Sohier and Rico Häuselmann, junior researchers from the Marzari group, contributed to the success of EPFL’s visit to the Collèges Sainte-Croix and Gambach in Fribourg on Oct 31, 2017 under the supervision of the MARVEL management team, with 3D movies on materials simulation and hands-on experiments to explain the concept of phonons (nccr-marvel.ch/fribourg-high-schools). The Röthlisberger group hosted 2 high-school students for a maturity project on MARVEL related topics. Matthieu Mottet (IBM), as member of the organization committee of the Swiss Chemistry Olympiads promoting the transfer of chemistry to high-school students, mentored the Swiss student delegation to the 49th International Chemistry Olympiads in Bangkok.



Figure 2: Flyer to advertise the high-school students summer camp *Des atomes aux ordinateurs*, à la découverte de la programmation scientifique.

6 Equal opportunities

In year 4, MARVEL has continued established measures and activities in line with its strategy, as well as the exchange and cooperation with other NCCRs, the EPFL Equal Opportunities Office and Science Outreach Department.

Recommendation of the review panel

The implementation of the equal opportunities strategy took into account recommendations of the review panel. These included the wish to see PIs involved in MARVEL make efforts to recruit more female PhD students and postdocs, to expand and increase allowances for recipients of the INSPIRE Potentials Master’s Fellowships, and to evolve a feedback mechanism to ensure that the working climate is favorable to and aware of gender balance.

Numbers

Table 6.1 provides an overview of the number and share of women directly involved in MARVEL, as reported in NIRA. The number and the share of women among postdocs increase every year since the beginning of MARVEL, finally reaching 14 female postdocs and 19% in year 4. Time will tell us if this trend will continue. At the level of PhD students, the number of women remains stable, but, as the number of men increases, the share of women is decreasing, reaching 18% in year 4. However this share can vary rapidly because of the still small total number of PhD students in MARVEL (49

	year 1		year 2		year 3		year 4	
	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%
Senior res.	1 13%	7 87%	0 0%	8 100%	2 9%	21 91%	3 10%	28 90%
Postdocs	5 11%	39 89%	8 11%	65 89%	13 16%	69 84%	14 19%	60 81%
PhD	6 26%	17 74%	9 20%	35 80%	9 19%	38 81%	9 18%	40 82%

Table 6.1: Number and share of women (W) and men (M) involved in MARVEL in years 1, 2, 3 and 4 (From NIRA).

in year 4). We hope the INSPIRE Potentials Master’s Fellowships will help to hire more female PhD students in the next years. To check the representativeness of the share of women measured in MARVEL, we initiated a comparison of these figures regarding PhD students and postdocs with those obtained when considering the research groups as a whole. This comparison revealed a variation below 4% in Dec 2017. We will pursue these comparisons to monitor their evolution.

Raising gender awareness

Talk on stereotypes and implicit bias

MARVEL invited Marianne Schmid Mast, professor in organizational behavior at the Univ. of Lausanne for a talk at its Review and Retreat on Sept 7, 2017. To ensure maximal participation and sufficient time for discussion, the talk entitled “Gender & Genius” was organized ahead of the social dinner. It was followed by a generous time of exchanges and questions, and generated fruitful and animated discussions around the tables during the dinner.

Screening of the movie “Hidden Figures”

In the context of the MARVEL/CECAM lecture by Mary Ann Mansigh Karlsen, a public screening of the movie “Hidden Figures” by Theodore Melfi, featuring the story of female African-American mathematicians at NASA in the early years of the US space program, was organized for a public of ~ 80 people.

Gender Training

MARVEL has continued efforts asking for the introduction of formal gender trainings for all new employees at EPFL to prevent discrimination and harassment. MARVEL and

the EPFL Equal Opportunities Office contacted Prof. Franciska Krings at Univ. of Lausanne to lead a pilot seminar on “Diversity in Recruitment & Supervision” dedicated, in the first instance, to MARVEL PIs. With the objective to foster an attractive and productive working culture and climate that are favorable to and aware of diversity and gender balance, this half-day seminar should be organized in Lausanne and Zurich, hopefully before summer.

Advance women scientists

INSPIRE Potentials fellowships

The INSPIRE Potentials — MARVEL Master’s Fellowships (nccr-marvel.ch/inspire) have the goal of empowering excellent female students to conduct a 6-month Master’s thesis research in a computational laboratory belonging to MARVEL and are a common initiative with the NCCR QSIT on “Quantum Science and Technology”. They were introduced in 2016, with 2 calls per year. The 4 students granted in 2016 did their Master’s research work (Fig. 1). These are Norma Rivano (from Cagliari, IT, who was in the Marzari group), Alice Cuzocrea (Trieste, IT, in the Ceriotti group), Cecilia Chiaracane (Trieste, IT, in the Yazyev group), and Cecilia Vona (Trieste, IT, in the Röthlisberger group). They were followed by 5 granted after the Apr 2017 call. These are Andriani Keliri (ETHZ, CH, Spaldin), Özge Kadioğlu (Koç, TR, Smit), Nataliya Lopanitsyna (MIPT, Moscow, RU, Ceriotti), Yuting Qian (UCAS, Beijing, CN, Yazyev), and Simran Kumari (IISER, Kolkata, IN, Marzari) — last 3 still ongoing in Feb 2018. 5 fellowships were granted after the Oct 2017 call and will begin their Master’s research work between Feb and Aug 2018. These are Henglu Xu (EPFL, CH, Smit), Rong Zhang (NTU, Singapore, Marzari), Chiara Cignarella (La Sapienza, Rome, IT, Hutter), Martina Danese (Trieste, IT, Passerone), and Melania D’Aniello (La Sapienza, Rome, IT, Goedecker). In total, up to now, 14 students were granted for a Master’s project in 9 different MARVEL groups in 5 institutions. 4 INSPIRE Potentials students participated to the MARVEL junior retreat and 6 (all the ones involved at that time) attended the Review and Retreat, one even presenting a poster. It is still too early to know where these students will go after their Master, but there are already a few signs that some students may continue at the PhD level in the group in which they performed their Master’s project. We requested some anonymous feedback from the students who finished their project. 4 already answered



Figure 1: Informal lunch with all the INSPIRE Potentials students present at EPFL in Aug 2017.

and the feedbacks are very good. About career prospects, the INSPIRE Potentials opportunity, even if not always easy and considered as a challenging experience, contributed to empower them. It also encouraged them to pursue a career in research, even if 2 of them are planning to do a PhD in different field than computational design and discovery of novel materials. A survey was also sent to the supervising PIs.

The overall number of applications is increasing with time, but with 11 applications in Oct 2017, we still have to increase publicity, e.g., contacting more proactively professors outside MARVEL in Switzerland and abroad (granted-students learned about the fellowship through their professors), using the social networks, publishing testimonies of granted-students on the INSPIRE Potentials webpage. Following the recommendation of the review panel, and after discussion with the EPFL Student Office, the amount of the fellowship was raised in July from 1’600 CHF to 2’000 CHF per month, plus coverage of medical insurance for extra-Europeans students, who are not covered by their own country insurance.

Lunch events

In the context of the MARVEL/CECAM lecture by Mary Ann Mansigh Karlsen, an exclusive female lunch with her was organized together with the EPFL Equal Opportunities Office and gathered 84 women. Besides, MARVEL has advertised other lunch events and industry site visits organized by the EPFL Equal Opportunities Office to its female PhD students and researchers. Counting the one with Mary Ann Mansigh Karlsen, 11 events have been organized in 2017, providing privileged opportunities to personally interact with role models from academia, industry, start-ups, science policy and entrepreneurship. They were attended



by more than 50 participants on average (between 20 and 100). Some MARVEL female PIs are also engaged in sharing about their career path in events organized for students and post-docs in various institutions.

Training, mentoring and coaching

MARVEL encourages its female PhD students and postdocs to participate in training, mentoring and coaching programs offered at the various institutions. The up-to-date offer is on the website and MARVEL female researchers are regularly informed of their existence and of the new programs.

Recognition for female researchers' excellence

MARVEL Distinguished Lectures MARVEL has strived to increase the number of women invited for distinguished lectures. In year 4, 2 of the 5 distinguished lectures were delivered by women. So far 4 out of 13 distinguished lectures have been presented by women.

Women participation in conferences The MARVEL School on "Variationally Enhanced Sampling" was organized by members of the Parrinello group and took place in Feb 2017 at USI. Among the 48 participants, nearly one third were women, 6 of them affiliated with MARVEL and 8 with other organizations. Thanks to the financial support of MARVEL, Oleg Yazyev co-organized the conference "New Trends in Topological Insulators 2017" (NTTI2017) in Monte Verità in Jul 2017. Three invited speakers were women, while overall 12 female participants, of a total of 84, were present at the conference. This ratio appears to be representative in the field.

Prize and distinctions In year 4, several distinguished prizes have been awarded to female PIs. Nicola Spaldin has been elected as fellow of the Royal Society. She also received the 2017 MRS Mid-Career Researcher Award. Clémence Corminboeuf receives the 2018 Early-Career Award in Theoretical Chemistry of the ACS Physical Chemistry Division. In Feb 2017, Sandra Lubser, MARVEL senior researcher at UZH, was awarded an SNSF professorship.

Support work-life balance

Information on work-life balance policies and support for parents at the different institution has been made available on the MARVEL website. MARVEL has advertised new actions in this field, such as the SNSF Flexibility Grants, to the community.

Cooperation with other NCCRs

MARVEL equal opportunities representatives participate regularly to the meetings gathering all NCCRs' equal opportunities representatives to enable a closer cooperation in these activities and to share good practices. The next meeting will be organized at EPFL by the NCCRs Robotics and MARVEL in late spring 2018.

Actions for young girls

EPFL Science Outreach Department and Equal Opportunities Office work together with MARVEL to enhance the interest of young girls for STEM fields. With the support of MARVEL, specific activities in MARVEL related domains are developed. Through the organization of these activities, MARVEL acts to change behavior, decrease stereotypes and obstacles related to gender, support the development of knowledge and interest of young girls for MARVEL related fields, and increase their confidence in their capacities for STEM fields. These activities receive strong interest and are always fully booked. The running activities are presented below. A 2-weeks summer camp in scientific programming for high-school students will be organized in Jul 2018. Registration will open soon and 50% of the places are reserved for young girls. More details can be found in the Education chapter.

Polythèmes workshop on materials

This workshop, developed especially for MARVEL and entitled *Diamant, alu, caoutchouc, ils sont fous ces matériaux !*, has as goal to give the opportunity to 7 to 10 years old girls to discover the world of material sciences over 3 Wednesdays afternoon. The 6th and 7th editions took place on Nov 29, Dec 6 and 13, 2017 (Fig. 2 left), and Jan 10, 17 and 24, 2018. The workshops were run by a female scientist who also acted as a role model.

Summer camp Matériaux super géniaux

The 3rd edition of the summer camp *Matériaux super géniaux*, developed especially for MARVEL, took place from Aug 14 to 18, 2017 (Fig. 2 center-left). Throughout the week, participants could run various hands-on experiments around materials and their properties (e.g. conductivity, density, ferromagnetism). The 11-13 years old girls also had the opportunity to discover the research running in different labs of MARVEL. They prepared a presentation to explain to their families what they



Figure 2: Workshops and camp for young girls at EPFL. From left to right: workshop Diamant, alu, caoutchouc, ils sont fous ces matériaux !; summer camp Matériaux super géniaux; summer camp in chemistry; mathematics workshop Maths en jeu.

had learned during the lab visits for the closing ceremony of the camp.

Chemistry summer camp

This 5-days camp was organized in a real chemistry lab from Aug 14 to 18, 2017, and gave the opportunity to 10 girls and 10 boys aged 11 to 13 years to discover different fields of chemistry and chemical engineering. Research labs also opened their doors to the participants who could discuss with researchers and learn about the interest and the usefulness

of their research for the society (Fig. 2 center-right).

Mathematics workshops Maths en jeu

The *Maths en jeu* workshops are organized thanks to the joint funding of MARVEL and the EPFL Science Outreach Department. Workshops at 4 different levels were successfully organized from Mar to June and from Sept to Dec 2017 (Fig. 2 right). Some workshops were girls-only and some others for girls and boys (with 50% girls). The ages were 7 to 13.

7 Communication

During year 4, MARVEL continued implementing its communication strategy. The website was regularly updated, including “Highlight Papers” and “Feature Stories” on recent work by MARVEL researchers. The MARVEL community and network was energized thanks to the release of the internal newsletter and the organization of various meetings, including the junior seminars and the Review and Retreat. MARVEL took opportunities to team up with external events to open up its 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

1 Website and newsletter

The website nccr-marvel.ch serves two main communication purposes: internal and external communications on MARVEL. It is regularly updated with pertinent information on projects, people, events and news for ensuring smooth internal communication. A toolkit for editing a newsletter from website entries was implemented, enabling the release of the first internal newsletter in Oct 2017. The internal newsletter fosters transversal communication between the MARVEL research groups and features various items such as events, seminars and courses organized by MARVEL members/management in Switzerland and abroad, stories about MARVEL research and its scientists, etc. The “MARVEL Newsletter to Industry” (3 issues/year) was first released in Nov 2017 while the first issue of the monthly scientific newsletter (external audience) was released in Jan 2018.

2 MARVEL distinguished lectures

MARVEL continued inviting worldwide recognized experts in computational materials science to present a “MARVEL distinguished lecture”. This year, 5 lectures were organized at EPFL, gathering a public of 50 to 80 people per event. A specific effort was made to invite 2 world renown women.

- On Mar 8, 2017, Prof. Markus Reiher (ETHZ) gave the lecture “Interactive and automated exploration of reaction mechanisms”.
- On May 16, 2017, Prof. Annabella Selloni (Princeton) presented on “Photocatalysis

on TiO₂: insights from simulations”.

- On Jul 21, 2017, Prof. Steven G. Louie (Univ. California, Berkeley) talked about “The fascinating quantum world of two-dimensional materials: Interaction and topological effects”.
- On Sept 11, 2017, Prof. Yang Shao-Horn (MIT) presented on “The Future of electrochemistry”.
- On Sept 21, 2017, Prof. Chris G. Van De Walle (Univ. California, Santa Barbara) gave the lecture “Using the right criteria for design and discovery”.

All lectures were recorded and made available through the MARVEL website on slideshow.epfl.ch/events/14.

Internal communication

1 Review and Retreat

The fourth Review and Retreat took place at EPFL on Sept 7 and 8, 2017 (Fig 1). This edition gathered 156 participants, including 116 from MARVEL (group leaders, senior researchers, postdocs, PhD students and INSPIRE Potentials Master’s students), representing the various research groups. The Scientific Advisory Board was invited to attend the whole meeting. The program on Thursday included 8 highlight talks covering MARVEL achievements, a poster parade with live flash poster presentations and 2 presentations on potential collaborative projects, all presented by junior scientists. The poster session took place during the standing lunch and the afternoon break. This allowed ample time around the 80 posters



Figure 1: MARVEL Review and Retreat at EPFL on Sept 7 and 8, 2017. From left to right: group picture; highlight talks; poster session; interesting discussions; social evening.

for discussing possible synergies and collaborations. A social evening was organized on-site at Gina Ristorante with a talk by Marianne Schmid Mast entitled “Gender & Genius” followed by the dinner. Friday was attended by computational group leaders, experimental groups representatives and the Scientific Advisory Board. The day was devoted to the presentations of phase II projects by the relevant PIs and discussions. The Scientific Advisory Board presented its report with comments and recommendations.

To be noted that the April site visit in the presence of the review panel was another great opportunity for the community to meet and increase synergies between research groups.

2 Other meetings

Computational group leaders met several times at EPFL to discuss the results and the evolution of the NCCR. In Mar 2017, 2 meetings were organized, one with a small group specifically on machine learning, and the second with all the other computational group leaders. Another meeting took place on Jul 10, 2017, including use of videoconference for those who could not attend physically. An “Agility Plus” workshop with more than 30 participants, on Dec 4, 2017, consisted of 7 presentations and discussions on new collab-



Figure 2: Scientific and informal discussion around a pizza slice at the junior seminars. In the background the photography exhibition by Julie Birenbaum is still visible.

orative and synergistic projects that started as of Jan 2017. The members of the PP7 experimental projects met on Apr 4, 2017, at PSI.

3 MARVEL junior seminars

The MARVEL junior seminars continue taking place monthly (except Jul, Aug, Sept), with 45 – 60 participants, to initiate additional collaborations between research groups, and to maintain a vibrant community. The organizing committee consists of 9 delegates among PhD students and postdocs representing EPFL research groups and acting as chairpersons. Each seminar includes two presentations on a scientific question explored in depth and followed by a discussion facilitated by the chairperson of the day, whose mission is to ensure active lively interactions between the audience and the speakers. The program is communicated to the MARVEL community and to the associated EPFL research groups, and advertised on the website (nccr-marvel.ch/references/education-and-training/marvel-junior-seminars), in the internal and scientific newsletters, and on EPFL online agenda. Pizza is offered before the seminar, and coffee and pies after, to allow for scientific and informal discussion (Fig. 2).

External communication

1 Events

1.1 Ig Nobel Award Tour Show

MARVEL sponsored and co-organized with EPFL the second visit of the Ig Nobel Award Tour Show on Mar 21, 2017 (Fig. 3). On



Figure 3: Ig Nobel Award Tour Show 2017 on Mar 21, 2017, at EPFL; © EPFL, Alain Herzog.



Figure 4: Public discussion with Mary Ann Mansigh Karlsen on Nov 15, 2017, at EPFL; © EPFL, Alain Herzog.

this occasion, a public of about 700 people could once again learn about “achievements that first make people laugh, and then make them think” as, e.g., that yawning is not contagious for tortoises or that organizations would become more efficient if they promoted people at random. The show will come again on Mar 27, 2018, with 3 new Ig Nobel laureates.

1.2 Almost famous, a woman behind the codes

Many of the breakthroughs of the early days of simulation would not have been possible without skilled programmers who translated new scientific ideas into efficient codes that would run without errors on the supercomputers of the 1950s and 1960s. CECAM and MARVEL had the great pleasure to organize a public discussion with a truly outstanding and female representative of the first generation of coders, Mary Ann Mansigh Karlsen, on Nov 15, 2017 as a conversation with Daan Frenkel (Univ. of Cambridge, UK) (Fig. 4, www.cecama.org/upload/videos/Part1.CECAM). It was preceded by a presentation from Prof. Michel Mareschal (Univ. Libre Bruxelles) about the “Early years of computational statistical mechanics: It’s only numerics” (www.cecama.org/upload/videos/Part2.CECAM). In collaboration with the EPFL Equal Opportunities Office, the week was completed with the screening of the movie “Hidden Figures” by Theodore Melfi featuring the story of female African-American mathematicians at NASA in the early years of the US space program, and with an exclusive lunch with Mary Ann Mansigh Karlsen for EPFL female researchers.

1.3 Open days and public conferences

Ulrich Aschauer and his team were at the *Nacht der Forschung* at UniBE on Sept 16, 2017 (Fig. 5, right). They presented to the general public interactive molecular dynamics with posters and discussions providing information about computational research. On Nov 25, 2017, Ulrich



Figure 5: Left: Festival science+fiction 2017, panel discussion on artificial intelligence with Anatole von Lilienfeld; © Festival science+fiction 2017, Raphael Hünerfauth und Katharina Good. Right: Nacht der Forschung on Sept 16, 2017, at UniBE; © Universität Bern, Ramon Lehmann.

Aschauer gave a public lecture for *(Bio)chemie am Samstag* at UniBE for a ~ 100 people audience, with a high proportion of high-school students and teachers as well as interested general public. A section of the lecture detailed the general importance of this kind of research in Switzerland and in particular provided information about the MARVEL project. On May 7, 2017, Anatole von Lilienfeld participated in a panel discussion entitled *Wann werden wir überflügelt* on artificial intelligence within the *Festival science+fiction: machina sapiens* in Basel (Fig. 5, left). Watch the discussion on YouTube (youtu.be/6j3t-iqQxfm).

On Feb 27, 2017, Nicola Marzari gave a public lecture entitled “Computational Materials Science Enters a New Age” at the ICTP Graduation Ceremony for the Masters in high performance computing in Trieste. He was also the speaker for the PASC17 public conference “The Times They Are a-Changin’ — Computational Discovery in the 21st Century” on June 27, 2017, in Lugano. Nicola Spaldin was invited to give two Lise-Meitner-Lectures (event series from DPG and ÖPG dedicated to the public at large), one on Mar 8, 2017, in Erlangen entitled “From Materials to Cosmology: Studying the early universe under the microscope” and the second on “New Materials for a New Age” on Nov 13, 2017, in Vienna.

2 Artistic work

2.1 ACCES visualization contest 2017

The second ACCES Visualization Contest, with management and financial supports from MARVEL, was launched mid-Sept with a deadline for the submission of entries on Dec 31, 2017. ACCES is an EPFL platform for Application-Centered Computational Engineering Science. The contest is part a broader effort at EPFL to embed computational thinking into the education of scientists and engineers. It aims at fostering a culture of insight-

ful and visually-engaging representations of scientific/engineering computational concepts and solutions, and solicits contributions from all students and postdocs of EPFL. 29 entries were received in 3 categories (static, dynamic or interactive). The winners are not yet known as the award ceremony is scheduled for Feb 26, 2018, together with the exhibition of all contributions in the ME D hall at EPFL.

2.2 MARVEL/ACCES seminar by W. Craig Carter

In the framework of the ACCES Visualization Contest 2017, MARVEL and ACCESS organized on Nov 10, 2017, a seminar by Prof. W. Craig Carter (MIT and invited professor at EPFL) entitled “What is — and what isn’t — computational thinking? And, why and how should universities include it in curricula”. More than 150 participants followed the lecture either directly or from afar thanks to video streaming. The seminar was recorded and is available through the MARVEL website.

2.3 Photography exhibition by Julie Birenbaum

The photography exhibition by Julie Birenbaum, inaugurated on Sep 8, 2016, was displayed in the ME D hall at EPFL for more than one year until Nov 2, 2017 (Fig. 2). It is planned to present it at ETHZ in 2018.

3 Social media

In Nov 2017, a twitter account was launched (@nccr_marvel) enabling us to relay information from and to scientific, industrial and

associative entities involved in materials modeling.

4 Other communication-related actions

MARVEL participates in the monthly meeting of all EPFL communication officers to guarantee a good cooperation with the host institution partners.

5 MARVEL in web news and in the press

A press release for the MARVEL supported work published by Schmidt *et al.* in *Nature Materials* [1] appeared on the PSI website on Jul 17, 2017. It received a warm echo on several web-based scientific news services (e.g. Phys.org, *idw – Informationsdienst Wissenschaft*, or innovations report) as well as in the local press (Neue Zürcher Zeitung). Alexey Soluyanov in Troyer’s group published a viewpoint on topological materials for *Physics* [2].

Some news in relationship with MARVEL were published on the websites of EPFL and other MARVEL-involved institutions, in particular for the promotion of hot research topics. A few articles mentioning MARVEL were published in the internal *EPFL Magazine* newspaper, including 2 interviews, of Marc Abraham, creator of the Ig Nobel Awards, in the Apr issue, and of Mary Ann Mansigh Karlsen, a woman behind the codes, in the Dec issue. Such news are usually relayed on the MARVEL website and since fall on Twitter and in the newsletters. They are also listed in the NIRA database.

8 Structural aspects

Most of the contractual structural measures have already been implemented during years 1 and 2, with the set-up of the visualization room “CoViz1” in MXC 320 and the long-term data storage co-located at CSCS. In addition, extra measures, not in the initial commitment, included moving the headquarters of MARVEL to the new ME D building and creating the distant education classroom “CoViz2”.

The promised tenure-track positions in computational materials science and condensed matter physics were allocated to Michele Ceriotti in Materials and to Oleg Yazyev in Physics. The third tenure-track assistant professor in the School of Computer and Communication Sciences was allocated to Martin Jaggi, who works in the field of machine learning and optimization, and who has been associated to MARVEL from May 2017 through an “Agility Plus” project. All the home institution’s commitments were thus accomplished or improved upon already before the end of year 3

9

Key achievements during phase I

The NCCR MARVEL has four major goals: world-class research in materials, and especially in the computational design and discovery of novel materials; the creation of a sustainable world-class infrastructure for the computational design and discovery of novel materials; the creation of a community in this field in Switzerland, with world-class visibility; the implementation of long-term structural measures that support the three goals above, and the four strategies of KTT, education and training, equal opportunities, and communication. We believe that we have had key achievements in all these; we first highlight the achievements around the two goals of world-class research in computational design and discovery (VP1, VP2, HP3 HP4, HP5 and PP7) and in infrastructure (PP6), followed by those focusing on the two goals of creating a community, of long-term structural measures, and of strategic developments.

VP1 — Novel Materials Physics

As far as the topological materials part of VP1 is concerned, during the entire period of phase I the following key results were obtained thanks to the collaboration established between the groups of Troyer and Yazyev. Novel topological electronic phases, namely the type-II Weyl semimetals [3], nodal-chain [4] and triple-point [5] topological metals were discovered making a very strong impact in the topological community. The methodological developments resulted in two public software package, Z2PACK [6] and WannierTools [7], that in turn allowed the realization of high-throughput computational workflows for identifying topological materials among known compounds. The results of first-principles calculations performed within this high-throughput screening are collected in the database that, by now, contains over 16'000 materials. A number of candidate materials identified by us were attempted for experimental verification. This resulted in several discoveries of new topological materials, e.g., the β -Bi₄I₄ quasi-1D topological insulator [8] and the MoP₂ and WP₂ type-II Weyl semimetals [9] that are becoming rising-star materials due to extraordinary physical properties. Collaboration with the experimental group of Ming Shi at PSI realized within the PP7 project became an important part of our topological materials discovery research already resulting in several joint publications on Weyl semimetals [10, 11]. For the correlated materials part of VP1, the main phase I achievements have been the understanding of the low-energy physics

of the nickelates as well as d^1 and $d^2 t_{2g}$ perovskites, and the design of a new class of room-temperature multiferroics with an entirely new mechanism for stabilizing spiral magnetic order. In addition, our rational design approach has been illustrated with the prediction of new materials with unusual yet appealing Hamiltonians — the single-band Hubbard model and the strong coupling between structural Higgs and Goldstone modes — which are currently being sought experimentally.

VP2 — Novel Materials Applications

A prominent and highly-cited result of the subproject on *halide perovskites* in VP2 is the discovery of the atomistic mechanism of how mixing of monovalent cations can be used for a preferential phase stabilisation of perovskites [12]. We also showed that the radiative bimolecular recombination in halide perovskites is slowed down by the formation of polarons [13]. In the VP2 subproject on *water splitting*, we proposed cubane structures, including open cubane core structures [14], as novel and highly efficient catalysts for the water oxidation reaction [15, 16, 17, 14]. We also developed a unified perspective on catalysis through exploiting the paradigms of both homogeneous and heterogeneous catalysis. Our concept of the “molecular volcano plots” [18, 19, 20, 21, 22] combined with machine learning models is instrumental to identify superior catalysts for homogeneous applications. In turn, we achieved superior electrocatalysts for the oxygen evolution reaction borrowing strategies from homogeneous catalysis [23, 24]. In view of identifying cat-

alysts with band edges that straddle the water redox levels, we determined the accuracy of advanced electronic-structure calculations in predicting band alignments at semiconductor-water interfaces [25]. The VP2 subproject on *solid ion conductors* has reached the stage of screening up to a thousand Li-containing compounds at the rate of 100 materials per month, leading to novel candidates with diffusion coefficients larger than the current state-of-the-art. In addition, we developed a computational framework to understand the effect of doping on structural stability and transport phenomena in garnet solid-state electrolytes, and analyzed how correlation effects related to ionic conductivity [26] and how strain can decrease activation energies [27]. The subproject on *low-dimensional materials* has extensively characterized 110'000 experimentally known materials, identifying 1'825 that can be exfoliated into novel 2D monolayers [28]. The large portfolio of materials resulting from this study is currently being screened for various properties, and has already led to the discovery of the first room-temperature Kane-Mele quantum spin Hall insulator [29] and a variety of novel 2D magnetic systems. Furthermore, the quest for novel carbon based 1D nanomaterials culminated in a joint experimental and theoretical demonstration of edge states in atomically precise nanostructures [30]. In parallel, an automated computational platform has been developed for this experimental community.

HP3 — Advanced Quantum Simulations

The Werner group develop a parameter-free *ab initio* simulation method for correlated materials based on the GW+DMFT scheme [101], which allows a self-consistent treatment of screening and correlation effects in realistic multi-band materials. The new method was applied to SrVO₃ [31], resulting in a new interpretation of the electronic structure of this material. The Troyer and Werner groups have developed efficient continuous-time quantum Monte Carlo methods (LCT-QMC) [32, 33] and new algorithms to substantially reduce the sign problem [34]. Enabling simulations of electrochemical processes at the many-body correlation energy level using efficient algorithms for second-order Møller-Plesset perturbation theory (MP2) [35], the random phase approximation (RPA) [36, 37], and G₀W₀ approximation [38] have been the focus of the VandeVondele and Hutter groups. Applications to liquid water [39, 40] and aqueous redox systems [41, 42] by *ab initio* molecular dynamics and Monte

Carlo sampling were performed, and the VandeVondele and Ceriotti groups collaborated on new advanced sampling algorithms combining DFT and MP2 electronic structure methods with nuclear quantum effects [43]. In collaboration, the Marzari and Goedecker groups developed the ENVIRON library. This library implements state-of-the-art versions of the self-consistent continuum solvation (SCCS) model [102] including major new developments in algorithms [44] and parametrizations [45]. This library is suitable for integration in other codes, including packages developed outside MARVEL. Last, Koopmans-compliant functionals [103][46] have been shown to provide accurate spectral properties for molecular systems [47, 48]. The Marzari group finished recently an implementation of this formalism for extended systems. This opens the door for reliable, and computationally efficient approaches to predict spectral properties in nanostructures, interfaces, solids [49, 50], and liquids — for the latter an ongoing collaboration between Pasquarello, VandeVondele and Marzari is dedicated to rationalizing the connection between the microscopic structure of water and its electronic properties.

HP4 — Advanced Sampling Methods

The Goedecker group has discovered numerous energy related materials such as silicon allotropes for photovoltaic applications [51], superconductivity [52, 53], or novel two-dimensional TiO₂ sheets for water splitting [54]. They also succeeded to perform the first calculation of all-electron DFT atomization energies for a test set of small molecules with micro-Hartree accuracy [55]. This allows for the first time to assess unambiguously the accuracy of commonly used approximations in density functional calculations. The Goedecker group also developed a neural network interatomic potential that correctly describes long-range charge transfer [56] as well as high-resolution fingerprints for quantifying similarities between crystalline structures [57]. Structure prediction of complex surfaces and interfaces [58] were also performed. In a collaboration between the Goedecker and Røthlisberger groups, minima hopping simulations were performed that could identify all known crystalline phases of methyl-ammonium lead iodide (MAPbI₃) and predict a novel low-temperature phase. In a collaboration between the Parrinello and Røthlisberger groups, metadynamics simulations of the nucleation process in a highly multicomponent system were per-



formed that allowed to characterize the crystalline growth and nucleation mechanism of MAPbI_3 and to identify the prime role of the monovalent cation for crystallization of lead halide perovskites materials. The Parrinello group has developed a novel advanced sampling method, variationally enhanced sampling (VES), which has been proved to be robust and extremely successful in many ways, such as exploring high-dimensional free energy landscapes, obtaining kinetic information from atomistic simulations and computing free energy differences. They also developed several novel methods for simulating crystal nucleation and growth from the melt and solution, e.g., constant chemical potential molecular dynamics, nucleation rate calculations using metadynamics and VES, and the test particle insertion approach that uses metadynamics as well. The Parrinello group also proposed a set of general collective variables, entropy and enthalpy, which can be used for a wide variety of applications, including crystallization, crystal identification and prediction. The Smit group developed a methodology to identify similarity of pore shapes. This is important as for applications of nanoporous materials having the optimal pore shape is one of the criteria that is used for the design of materials. The state-of-the-art in this field was determining similarity by visual inspection. This approach allows us to screen large databases to find the optimal material [59]. They also published a review about the nanoporous materials genome for *Nature Materials Review* [60].

HP5 — Materials Informatics

The focus and aim of HP5 is to develop machine learning (ML) and knowledge/data driven methodologies for the acceleration of material discovery. The nature of this work is by definition interdisciplinary and thus the horizontal nature of HP5. During phase I, the project demonstrated that ML based methodologies, when supplied with adequate but not enormous high-quality training sets, can indeed be generalized to previously unseen physical models and deliver an accuracy that is often competing with established computational techniques, but at a fraction of the cost [61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71]. In parallel, HP5 demonstrated that massive knowledge extraction and subsequent relevant representation is indeed possible in spite of the extremely complicated nature of the native scientific literature [72]. Moreover, it was shown that, thanks to modern computational frame-

works for the driving of simulations, such as AiiDA, we can for the first time close the loop and go from literature to derived simulation in a seamless manner. Important awards in key conferences (e.g. ACM/IEEE IPDPS [73]) of other fields demonstrate the appeal of the work in HP5. Indeed, the last few years, the most important ML venues, such as NIPS, routinely host data driven approaches for the discovery of new materials. This serves as strong evidence of the validity of the HP5 mission, members of which routinely now frequent these events, as well as of the overall NCCR mission.

PP6 — Informatics

The PP6 project aimed in its original goals to provide a platform for high-throughput simulations, to offer verification and validation, and to integrate with the high-performance computing platform at CSCS. It has largely exceeded its objectives, leveraging also additional funding and collaborations through the H2020 Centre of Excellence MaX, on “Materials Design at the eXascale”, of which Marzari and Schulthess are PIs. A first achievement has been the full development and deployment of AiiDA as an operating system for computational materials science [74], with stable version 1.0 available in April 2018. AiiDA covers the four pillars of Automation, Data, Environment, and Sharing, and it greatly benefits from a new workflow engine, backend-agnostic structure that supports multiple databases (currently, Django and SQLAlchemy), an ever-growing set of plugins for major codes (Quantum-ESPRESSO, VASP, CP2K, FLEUR, SIESTA, Yambo, WANNIER90, to mention a few) and stable and robust workflows for the high-throughput calculation of materials properties, and direct pipelines to and from major repositories, including ICSD, COD, OQMD, and the Materials Project. The verification and validation effort [55] has led to a first major contribution to the effort led by S. Cottener [75] that is currently continuing according to protocols developed for binary and multinary. The SSSP pseudopotential library that we developed is the most accurate available (molmod.ugent.be/deltacodesdft, comparable to VASP PAW GW-ready when compared to Wien2K, and slightly superior when compared to FHI-aims or Exciting). The combination of these two achievements has made high-throughput calculations achievable with open-source codes, and has led to, e.g., the recent work on 2D exfoliation as a first high-profile demonstrator. A third achieve-

ment, not part of the original proposal, has been the deployment of the first version of the Materials Cloud (www.materialscloud.org), as a portal built to enable the seamless sharing and dissemination of resources [76] in computational materials science, encompassing educational material, interactive tools, simulation services, and curated and raw data. These underpin published results and empower data-based discovery, compliant with data management plans and the FAIR principles (findable, accessible, interoperable, reusable). Thanks to the full provenance model of AiiDA, we believe this represents one of the clearest and most open implementations. A major effort has also gone into rethinking the current computational models for high-performance computing [77, 78], with the development of domain-specific libraries, like SIRIUS [79], that provide optimal implementation on complex heterogeneous architectures (GPUs, multi-core, accelerators) of the core building blocks of electronic-structure calculations, such as the application of a Hamiltonian to a wavefunction in a pseudo-potential or all-electron formulation. Last but not least, close integration with CSCS has led to an offering to MARVEL researchers of world-class infrastructure to which NCCR MARVEL directly contributed: Piz Daint is both the 3rd most powerful supercomputer in the world, and the 10th greenest, and the only one to be in both top-10 lists (www.top500.org/green500/list/2017/11). These same collaborations have led to an architecture for the Open Science Platform of phase II that morphs supercomputer centres in providers of Infrastructure-as-a-Service, where HPC cycles, database solutions, and long-term storage are integrated (and, thanks to the H2020 CoE MaX, are federated across supercomputing centres) — see www.materialscloud.org/infrastructure for the description of the integration of the centres with the AiiDA “operating system” and the Materials Cloud dissemination.

PP7 — Experiments

To demonstrate the power and potential of computational driven materials design, an intense interaction with sample growth and characterization experts is required. PP7 is the corresponding linking point and provides funding for experimental verification projects, mainly financing 2 years postdoc positions for collaborative research projects between a computational MARVEL PI and an experimental group. For this, a proposal system was in-

stalled with two calls for regular projects and one for extension of projects. These calls were accompanied by workshops bringing experimental and computational groups together. In total 17 projects have been funded, resulting in 17+ new collaborations, partly including more than two partners. These projects are scientifically contributing to the design and discovery of novel materials with novel physics or improved properties or performance, mainly via collaborations with partners from VP1 and VP2. The aim is not only to verify (or challenge) predictions but to have an integrated feedback loop in both directions between theory and experiment, in order to improve and optimize the computational models and the experimental conditions. Scientific highlights range from shedding light on novel topological materials [80, 11, 10], time-dependent phenomena [81], rare-earth nickelates [82, 83], multi-ferroics [84], Hubbard single-band model systems [85], water splitting and catalysis [86, 87, 88, 1, 89, 90, 91, 92, 93, 94, 95, 96], solid-state ionic conductors [97, 27, 98], and anodic barriers [99, 100]. The platform, based on proposal calls and workshops, was very successful and, besides intriguing results, lead to new and closer collaborations between the computational and experimental groups. This will form the basis of several Design and Discovery projects, where experimental groups will be integrated from the beginning into selected teams tackling grand challenges.

Community and structural measures

First and foremost, the NCCR MARVEL wants to create and sustain a community in the field, greatly benefiting from the outstanding environment in Switzerland, in the field of computational condensed matter physics, chemistry, and materials science, the close proximity with world-class facilities at PSI, the presence on campus at EPFL of CECAM, and close links with Psi-k, of which Marzari is current chairman. EPFL has provided exceptional support, through three PATT positions in phase I (Cerioti, Yazyev, and Jaggi), and a new one in computational materials science for phase II; these investments have been matched by the delivery of dedicated space in the new ME D building, the creation of the CoViz1 and CoViz2 visualization and distant education classrooms (aces.epfl.ch/coviz1 and [/coviz2](http://aces.epfl.ch/coviz2)), and the cash commitments that make long-term storage on the Materials Cloud possible and sustainable. Most importantly, many of the activities detailed below are contributing to creating



a community of young researchers in the field that leverages each other's strengths (e.g. PhDs from one group moving as postdocs in another, transferring knowledge), knows each other's work, and is proud to be part of this effort. The redirection of ~ 1 MCHF to ignite the "Agility Plus" projects has also been instrumental in supporting junior or new members.

Knowledge and technology transfer

The KTT mission statement for phase I focuses on the development and dissemination of open-source materials simulation codes and to the training in the use of those codes; to the development and application of the materials informatics framework; to the verification and validation of calculations; to the sharing of all results from materials simulations. These goals have been accomplished through more than 50 activities (nccr-marvel.ch/ctw), the development and deployment of AiiDA, Materials Cloud, and the SSSP verification and validation effort. Continuous development of open-source software has taken place — from Quantum-ESPRESSO to CP2K to BIGDFT for density-functional theory and beyond; TRIQS and ALPS for correlated-electron calculations, PLUMED and i-PI for sampling, WANNIER90, Z2PACK, and WannierTools for electronic and topological properties, and ENVIRON for embedding electronic-structure codes into electrochemical simulations. We have engaged key stakeholders: Tier-0 supercomputing centres through the H2020 CoE MaX; the experimental community through the H2020 NFFA; independent software vendors thanks to the agreement between the QE Foundation and Schrödinger Inc. and the one with MPDS to share and cross-reference computational and experimental data; policy-makers through the H2020 CSA for the European Materials Modelling Council. A new 5-year H2020 Market-Place project, of which EPFL is partner, aims to deliver simulations services to the community through a Materials Cloud model.

For tech transfer, we have had close to 100 informal contacts, leading to 12 company visits, the signature of 5 collaborations, and ongoing discussions (including a strategic alliance with a large European chemical company). Among the signatures, notable are a major project funded by a Swiss company in the watch industry, the establishment of a CTI project with a local start-up that merges machine learning at UniBas and AiiDA at EPFL, and engagement in machine learning with a major US company.

Education and training

Three core activities have greatly contributed to forming a MARVEL community: the junior retreat, held over 3 days every summer, and self-organized by students and postdocs - with research presentations, tutorials of key interest (grant writing, funding opportunities), and nucleating original research projects; the junior seminars, held every month during term-time, as a focal point for all the community; and the MARVEL summer camp on scientific computing for high-school students, that will be held for 2 weeks in Jun-Jul 2018. Many of the 50+ activities nccr-marvel.ch/ctw have often an educational component.

Equal opportunities

The NCCR aims to engage the entire pipeline of the educational process, so key activities are those targeted at girls 7 – 19 years old, with the workshops *Diamant, alu, caoutchouc, ils sont fous ces matériaux !* and on maths, the summer camp *Matériaux super géniaux*, and the mixed-gender (50/50) summer camps on chemistry and on scientific computing. A key initiative for us has been the INSPIRE Potentials Master's Fellowships, that was started in the third year of the project, and that aims at doubling the number of female PhD students in MARVEL. Last, a personal highlight has been the invitation of Mary Ann Mansigh Karlsen, a pioneer "computer", developing the first molecular dynamics codes, co-organized with CECAM, and preceded by the screening of the movie "Hidden Figures".

Communication

The internal communication efforts have centered around our two annual events (the SNSF site visit, and the Review and Retreat) that have become a major appointment and exchange for all the community. The external communication efforts have first focused on developing the technology to record events, so that the MARVEL distinguished lectures and tutorials could all be made available to the website; these have now been complemented by the launch of internal and external (scientific and to Industry) regular newsletters. For the general public, MARVEL organises dedicated events (IgNobel, visualization contest, Birenbaum exhibition) or participates in broader efforts (Scientastic, open doors events) to communicate widely about the long-term impact of its research.

MARVEL-related publications

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

- [1] E. Fabbri, M. Nachtegaal, T. Binninger, X. Cheng, B.-J. Kim, J. Durst, F. Bozza, T. Graule, R. Schäublin, L. Wiles, M. Pertoso, N. Danilovic, K. E. Ayers, and T. J. Schmidt, *Dynamic surface self-reconstruction is the key of highly active perovskite nano-electrocatalysts for water splitting*, *Nature Materials* **16**, 925 (2017).
- [2] A. A. Soluyanov, *Viewpoint: Type-II Dirac Fermions Spotted*, *Physics* **10**, 74 (2017).
- [3] A. A. Soluyanov, D. Gresch, Z. Wang, Q. Wu, M. Troyer, X. Dai, and B. A. Bernevig, *Type-II Weyl semimetals*, *Nature* **527**, 495 (2015).
- [4] T. Bzdušek, Q. Wu, A. Rüegg, M. Sigrist, and A. A. Soluyanov, *Nodal-chain metals*, *Nature* **538**, 75 (2016).
- [5] Z. Zhu, G. W. Winkler, Q. Wu, J. Li, and A. A. Soluyanov, *Triple Point Topological Metals*, *Physical Review X* **6**, 031003 (2016).
- [6] D. Gresch, G. Autès, O. V. Yazyev, M. Troyer, D. Vanderbilt, B. A. Bernevig, and A. A. Soluyanov, *Z2Pack: Numerical implementation of hybrid Wannier centers for identifying topological materials*, *Physical Review B* **95**, 075146 (2017), <http://z2pack.ethz.ch>.
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- [8] G. Autès, A. Isaeva, L. Moreschini, J. C. Johannsen, A. Pisoni, R. Mori, W. Zhang, T. G. Filatova, A. N. Kuznetsov, L. Forró, W. Van den Broek, Y. Kim, K. S. Kim, A. Lanzara, J. D. Denlinger, E. Rotenberg, A. Bostwick, M. Grioni, and O. V. Yazyev, *A novel quasi-one-dimensional topological insulator in bismuth iodide β -Bi₄I₄*, *Nature Materials* **15**, 154 (2016).
- [9] G. Autès, D. Gresch, M. Troyer, A. A. Soluyanov, and O. V. Yazyev, *Robust Type-II Weyl Semimetal Phase in Transition Metal Diphosphides XP₂ (X = Mo, W)*, *Physical Review Letters* **117**, 066402 (2016).
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Annex 2 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 1st, 2017 to January 31st, 2018.

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

Group of Ulrich Aschauer

- M. PICHLER, W. SI, F. HAYDOUS, H. TÉLLEZ, J. DRUCE, E. FABBRI, M. E. KAZZI, M. DÖBELI, S. NINOVA, U. ASCHAUER, A. WOKAUN, D. PERGOLESI, AND T. LIPPERT
LaTiO_xN_y Thin Film Model Systems for Photocatalytic Water Splitting: Physicochemical Evolution of the Solid-Liquid Interface and the Role of the Crystallographic Orientation
Advanced Functional Materials **27**, 1605690 (2017).

Group(s): Aschauer, Lippert / Project(s): PP7

Group of Claudia Cancellieri

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 Journal of Electron Spectroscopy and Related Phenomena **219**, 72 (2017).
 Group(s): Yazyev / Project(s): VP1
- N. XU, G. AUTÈS, C. E. MATT, B. Q. LV, M. Y. YAO, F. BISTI, V. N. STROCOV, D. GAWRYLUK, E. POMJAKUSHINA, K. CONDER, N. C. PLUMB, M. RADOVIC, T. QIAN, O. V. YAZYEV, J. MESOT, H. DING, AND M. SHI
Distinct Evolutions of Weyl Fermion Quasiparticles and Fermi Arcs with Bulk Band Topology in Weyl Semimetals
 Physical Review Letters **118**, 106406 (2017).
 Group(s): Shi, Yazyev / Project(s): PP7, VP1
- L. YANG, M. JEONG, P. BABKEVICH, V. M. KATUKURI, B. NÁFRÁDI, N. E. SHAIK, A. MARGREZ, H. BERGER, J. SCHEFER, E. RESSOUCHE, M. KRIENER, I. ŽIVKOVIĆ, O. V. YAZYEV, L. FORRÓ, AND H. M. RØNNOW
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 Physical Review B **96**, 024445 (2017).
 Group(s): Yazyev / Project(s): VP1

2. Scientific articles in journals without peer review

Group of Antoine Georges

W. WU, M. S. SCHEURER, S. CHATTERJEE, S. SACHDEV, A. GEORGES, AND M. FERRERO
Pseudogap and Fermi surface topology in the two-dimensional Hubbard model

arXiv:1707.06602 (2017).

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

- M. KIM, J. MRAVLJE, M. FERRERO, O. PARCOLLET, AND A. GEORGES
Spin-Orbit Coupling and Electronic Correlations in Sr_2RuO_4

arXiv:1707.02462 (2017).

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

Group of Nicola Marzari

- D. DRAGONI, T. D. DAFF, G. CSÁNYI, AND N. MARZARI

Achieving DFT accuracy with a machine-learning interatomic potential: thermomechanics and defects in bcc ferromagnetic iron

arXiv:1706.10229 (2017).

Group(s): Marzari / Project(s): HP5

- A. MARRAZZO, M. GIBERTINI, D. CAMPI, N. MOUNET, AND N. MARZARI

Prediction of a room-temperature and switchable Kane-Mele quantum spin Hall insulator

arXiv:1712.03873 (2017).

Group(s): Marzari / Project(s): VP2

- N. COLONNA, N. L. NGUYEN, A. FERRETTI, AND N. MARZARI

Screening in orbital-density-dependent functionals

arXiv:1711.04485 (2017).

Group(s): Marzari / Project(s): HP3

- N. L. NGUYEN, N. COLONNA, A. FERRETTI, AND N. MARZARI

Koopmans-compliant spectral functionals for extended systems

arXiv:1708.08518 (2017).

Group(s): Marzari / Project(s): HP3

Group of Matthias Troyer

- X. FENG, C. YUE, Z. SONG, Q. WU, AND B. WEN

Topological Dirac Nodal-net Fermions in AlB_2 -type TiB_2 and ZrB_2

arXiv:1705.00511 (2017).

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

- G. TORLAI, G. MAZZOLA, J. CARRASQUILLA, M. TROYER, R. MELKO, AND G. CARLEO

Neural-network quantum state tomography for many-body systems

arXiv:1703.05334 (2017).

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

- A. A. SOLUYANOV

Viewpoint: Type-II Dirac Fermions Spotted

Physics **10**, 74 (2017).

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

Group of Anatole von Lilienfeld

- B. HUANG AND O. A. VON LILIENFELD
The "DNA" of chemistry: Scalable quantum machine learning with "amons"

arXiv:1707.04146 (2017).

Group(s): von Lilienfeld / Project(s): HP5

- R. RAMAKRISHNAN AND O. A. VON LILIENFELD

Machine Learning, Quantum Chemistry, and Chemical Space

in *Reviews in Computational Chemistry*, A. L. PARRILL AND K. B. LIPKOWITZ, eds. (John Wiley & Sons, Inc., Hoboken, NJ, USA, 2017), vol. 30, pp. 225–256, doi:10.1002/9781119356059.ch5.

Group(s): von Lilienfeld / Project(s): HP5



3. Publications involving several groups

- M. PICHLER, W. SI, F. HAYDOUS, H. TÉLLEZ, J. DRUCE, E. FABBRI, M. E. KAZZI, M. DÖBELI, S. NINOVA, U. ASCHAUER, A. WOKAUN, D. PERGOLES, AND T. LIPPERT
LaTiO_xN_y Thin Film Model Systems for Photocatalytic Water Splitting: Physicochemical Evolution of the Solid-Liquid Interface and the Role of the Crystallographic Orientation
Advanced Functional Materials **27**, 1605690 (2017).
Group(s): Aschauer, Lippert / Project(s): PP7
- D. SUTTER, C. G. FATUZZO, S. MOSER, M. KIM, R. FITTIPALDI, A. VECCHIONE, V. GRANATA, Y. SASSA, F. COSSALTER, G. GATTI, M. GRIONI, H. M. RØNNOW, N. C. PLUMB, C. E. MATT, M. SHI, M. HOESCH, T. K. KIM, T.-R. CHANG, H.-T. JENG, C. JOZWIAK, A. BOSTWICK, E. ROTENBERG, A. GEORGES, T. NEUPERT, AND J. CHANG
Hallmarks of Hund's coupling in the Mott insulator Ca₂RuO₄
Nature Communications **8**, 15176 EP (2017).
Group(s): Georges, Shi / Project(s): VP1
- J. RUPPEN, J. TEYSSIER, I. ARDIZZONE, O. E. PEIL, S. CATALANO, M. GIBERT, J.-M. TRISCONE, A. GEORGES, AND D. VAN DER MAREL
Impact of antiferromagnetism on the optical properties of rare-earth nickelates
Physical Review B **96**, 045120 (2017).
Group(s): Georges, van der Marel / Project(s): VP1, PP7
- G. FISICARO, L. GENOVESE, O. ANDREUSSI, S. MANDAL, N. N. NAIR, N. MARZARI, AND S. GOEDECKER
Soft-Sphere Continuum Solvation in Electronic-Structure Calculations
Journal of Chemical Theory and Computation **13**, 3829 (2017).
Group(s): Goedecker, Marzari / Project(s): HP3
- J. WILHELM, D. GOLZE, L. TALIRZ, J. HUTTER, AND C. A. PIGNEDOLI
Towards GW Calculations on Thousands of Atoms
The Journal of Physical Chemistry Letters **9**, 306 (2018).
Group(s): Hutter, Passerone / Project(s): HP3
- F. SONG, R. MORÉ, M. SCHILLING, G. SMOLENTSEV, N. AZZAROLI, T. FOX, S. LUBER, AND G. R. PATZKE
Co₄O₄ and Co_xNi_{4-x}O₄ Cubane Water Oxidation Catalysts as Surface Cut-Outs of Cobalt Oxides
Journal of the American Chemical Society **139**, 14198 (2017).
Group(s): Hutter, Smolentsev / Project(s): VP2, PP7
- M. PICHLER, J. SZLACHETKO, I. E. CASTELLI, N. MARZARI, M. DÖBELI, A. WOKAUN, D. PERGOLES, AND T. LIPPERT
Determination of Conduction and Valence Band Electronic Structure of LaTiO_xN_y Thin Film
ChemSusChem **10**, 2099 (2017).
Group(s): Lippert, Marzari / Project(s): PP7
- A. FLURI, A. MARCOLONGO, V. RODDatis, A. WOKAUN, D. PERGOLES, N. MARZARI, AND T. LIPPERT
Enhanced Proton Conductivity in Y-Doped BaZrO₃ via Strain Engineering
Advanced Science **4**, 1700467 (2017).
Group(s): Lippert, Marzari, Pergolesi / Project(s): VP2, PP7
- A. FLURI, E. GILARDI, M. KARLSSON, V. RODDatis, M. BETTINELLI, I. E. CASTELLI, T. LIPPERT, AND D. PERGOLES
Anisotropic Proton and Oxygen Ion Conductivity in Epitaxial Ba₂In₂O₅ Thin Films
The Journal of Physical Chemistry C **121**, 21797 (2017).
Group(s): Lippert, Marzari, Pergolesi / Project(s): PP7, VP2
- E. GILARDI, E. FABBRI, L. BI, J. L. M. RUPP, T. LIPPERT, D. PERGOLES, AND E. TRAVERSA
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The Journal of Physical Chemistry C **121**, 9739 (2017).
Group(s): Lippert, Pergolesi / Project(s): PP7
- B.-J. KIM, D. F. ABBOTT, X. CHENG, E. FABBRI, M. NACHTEGAAL, F. BOZZA, I. E. CASTELLI, D. LEBEDEV, R. SCHÄUBLIN, C. COPÉRET, T. GRAULE, N. MARZARI, AND T. J. SCHMIDT
Unraveling Thermodynamics, Stability, and Oxygen Evolution Activity of Strontium Ruthenium Perovskite Oxide
ACS Catalysis **7**, 3245 (2017).
Group(s): Marzari, Schmidt / Project(s): VP2, PP7
- D. LEBEDEV, M. POVIA, K. WALTAR, P. M. ABDALA, I. E. CASTELLI, E. FABBRI, M. V. BLANCO, A. FEDOROV, C. COPÉRET, N. MARZARI, AND T. J. SCHMIDT
Highly Active and Stable Iridium Pyrochlores for Oxygen Evolution Reaction

Chemistry of Materials **29**, 5182 (2017).

Group(s): Marzari,Schmidt / Project(s): VP2, PP7

- J. WIKTOR, U. RÖTHLISBERGER, AND A. PASQUARELLO

Predictive determination of band gaps of inorganic halide perovskites

The Journal of Physical Chemistry Letters **8**, 5507 (2017).

Group(s): Pasquarello, Röthlisberger / Project(s): VP2

- F. FRANCO DE CARVALHO, C. A. PIGNEDOLI, AND I. TAVERNELLI

TDDFT-Based Spin-Orbit Couplings of 0D, 1D, and 2D Carbon Nanostructures: Static and Dynamical Effects

The Journal of Physical Chemistry C **121**, 10140 (2017).

Group(s): Passerone, Curioni / Project(s): VP2

- G. BAUER, D. ONGARI, X. XU, D. TIANA, B. SMIT, AND M. RANOCCHIARI

Metal-Organic Frameworks Invert Molecular Reactivity: Lewis Acidic Phosphonium Zwitterions Catalyse The Aldol-Tishchenko Reaction

Journal of the American Chemical Society **139**, 18166 (2017).

Group(s): Ranocchiaro, Smit / Project(s): HP4, PP7

- N. J. BROWNING, R. RAMAKRISHNAN, O. A. VON LILIENFELD, AND U. RÖTHLISBERGER

Genetic optimization of training sets for improved machine learning models of molecular properties

The Journal of Physical Chemistry Letters **8**, 1351 (2017).

Group(s): Röthlisberger,von Lilienfeld / Project(s): VP2, HP5

- N. XU, G. AUTÈS, C. E. MATT, B. Q. LV, M. Y. YAO, F. BISTI, V. N. STROCOV, D. GAWRYLUK, E. POMJAKUSHINA, K. CONDER, N. C. PLUMB, M. RADOVIC, T. QIAN, O. V. YAZYEV, J. MESOT, H. DING, AND M. SHI

Distinct Evolutions of Weyl Fermion Quasiparticles and Fermi Arcs with Bulk Band Topology in Weyl Semimetals

Physical Review Letters **118**, 106406 (2017).

Group(s): Shi, Yazyev / Project(s): PP7, VP1

- E. M. BOTHSCHAFTER, E. ABREU, L. RETTIG, T. KUBACKA, S. PARCHENKO, M. PORER, C. DORNES, Y. W. WINDSOR, M. RAMAKRISHNAN, A. ALBERCA, S. MANZ, J. SAARI, S. M. KOOHPAYEH, M. FIEBIG, T. FORREST, P. WERNER, S. S. DHESEI, S. L. JOHNSON, AND U. STAUB

Dynamic pathway of the photoinduced phase transition of TbMnO₃

Physical Review B **96**, 184414 (2017).

Group(s): Staub, Werner / Project(s): PP7

- D. GRESCH, G. AUTÈS, O. V. YAZYEV, M. TROYER, D. VANDERBILT, B. A. BERNEVIG, AND A. A. SOLUYANOV

Z2Pack: Numerical implementation of hybrid Wannier centers for identifying topological materials

Physical Review B **95**, 075146 (2017), <http://z2pack.ethz.ch>.

Group(s): Troyer, Yazyev / Project(s): VP1

Annex 3 Status of structural measures implementation

Planned measures according to annex 3 of the NCCR contract for phase I	Current status of implementation and comments
Infrastructure	
<i>Planned</i>	
Visualization facility (48 sqm, MXC 320, 100'000 CHF)	Implemented CoViz1, aces.epfl.ch/coviz1 .
Database Cluster (500'000 CHF)	Implemented at CSCS, see note 1 after the tables.
<i>Additional measures</i>	
New Mechanical Engineering Building (ME D)	Relocation of all MARVEL and Marzari offices in new space in the ME D building.
Distant-education classroom (38 seats, 120'000, ME D2 1124)	Implemented as an additional commitment made by EPFL and co-located on the same space as the MARVEL offices CoViz2, aces.epfl.ch/coviz2 .
In-kind support for the MARVEL hardware at CSCS	ETHZ has been providing, through an agreement with CSCS, 205'000 CHF/year of in-kind support for the MARVEL hardware, from May 2015 now to Apr 2020.
Faculty	
<i>Planned new professorships</i>	
Assistant professor (TT) in Computational Material Science, planned for 2014	Michele Ceriotti, in Materials, Computational Science and Modeling, from 2014
Assistant professor (TT) in Data Mining, planned for 2014	Martin Jaggi, in Computer Science, Machine Learning and Optimization, from 2017
Assistant professor (TT) in Computational Physics, planned for 2015	Oleg Yazyev, in Physics, Theory of Dirac Fermion Materials, from 2014
<i>Additional measures</i>	
Full professorship in Chemistry	Berend Smit in Chemistry, Computational Molecular Simulation, from 2014

Planned measures according to annex 3 of the NCCR contract for phase I**Current status of implementation and comments**

Education

Planned

Establishment of a common Doctoral School in Computational Materials Science between ETHZ and EPFL with common summer/winter schools

Organization of graduate courses via tele-teaching between ETHZ and EPFL

As the NCCR is fully delocalized over most Swiss Institutions, the decision was made to work at something that could benefit all institutions (see *New proposed measures* below).

As we thought it was more important to broaden the effort to all Swiss sites (not to mention that the departure of Troyer and VandeVondele from the ETHZ faculty made it more important to focus on an all-site effort), we are actively promoting tele-teaching. We have put a considerable effort in developing the open-source technologies to make deployment more straightforward — all the material prepared by MARVEL is now deployed in the *Learn* section of the Materials Cloud (www.materialscloud.org/learn), including an up-to-date 2-week, 2 credits graduate class in computational materials science: www.materialscloud.org/learn/sections/lfYatW/hpc-and-high-throughput-materials-modeling-ictp-trieste-2017.

New proposed measures

H2020 MSCA COFUND proposal (“The MARVEL Academy”)

Smit and Marzari prepared a H2020 MSCA COFUND proposal (“The MARVEL Academy”), aiming to support 36 PhD students based on all MARVEL sites. As part of this effort, the MARVEL members would offer 10 1-week doctoral schools common to all students in the program. The first submission (Sept 2017) was rated very highly (87/100, Jan 2018), but below the threshold for funding. A second submission is planned for the summer of 2018.

Support of summer schools for doctoral and postdoctoral researchers

Education of MARVEL junior researchers on advanced topics was also supported by selective funding of in-depth summer and winter schools on relevant topics, often co-funded by CECAM or Psi-k (see nccr-marvel.ch/ctw for a list). The next one, in June 2018, can be found at www.cecam.org/workshop-1528.html.



Specific conditions and requirements according to Article 10 of the NCCR contract for phase I	Current status of implementation and comments
Special attention to KTT	Very close attention has been given to KTT, as amply detailed in the strategies, deliverables, and achievements presented in the yearly reports; this has become even more central in the proposal of phase II, with the inclusion of faculty in the executive committee (Smit, Curtin) that have had extensive experience in industry, the creation of an industrial advisory board, and dedicating an additional 0.2 FTE to communication to industry. All funding received and efforts initiated are detailed in the reports.
Cooperation with SCCERs	Close collaboration with the SCCER in Heat and Electricity Storage has led to 3 joint publications with the groups of Schmidt and Coperet.

Note 1: Database cluster

200'000 CHF are redirected to upgrade the MARVEL hardware at CSCS, and extend it from the end of phase I (Apr 2018) for two extra years (until Apr 2020). ETHZ matches this with 205'000 CHF/year of in-kind support (electricity, cooling, maintenance, sysadmin). Original hardware was 180 compute nodes on Cray XC30 Piz Daint, 24 cores Intel Xeon E5-2780v3; current is 180 compute nodes of 36 cores Intel Xeon E5-2695v4 (details in MARVEL Y3 Progress Report, p. 60). This upgrade was part of a general refurbishment of Piz Daint that made it the third most powerful supercomputer worldwide (first in Europe) in June 2017 and Nov 2017 (www.top500.org/lists/2017/06 and www.top500.org/lists/2017/11), after two Chinese systems. Piz Daint is also the only top-10 supercomputer that figures in the top-10 greenest architectures (www.top500.org/green500/lists/2017/06 and www.top500.org/green500/lists/2017/11). The remaining funds are supporting the Materials Cloud hardware infrastructure, now implemented and deployed (www.materialscloud.org) and in particular data storage of all the data present that is compatible with the long-term data management plans required by funding agencies.

Cover picture

Sketch-map representations of the crystal structure landscape of pentacene, as seen through the lenses of a REMatch-SOAP (from Michele Ceriotti, EPFL, Horizontal Project 5).

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