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Big-Data-Driven Materials Science and its FAIR Data Infrastructure

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Abstract

This chapter addresses the challenges and chances of big-data driven materials science, and it describes concepts and implementation of the forth paradigm of materials research. The importance of an all-embracing sharing (open data), of an efficient data infrastructure, and of the consideration of all computer codes used in the community is emphasized: Data must be FINDABLE, ACCESSABLE INTEROPERABLE and RE-PURPOSABLE. Recent developments of methods from artificial intelligence are reviewed and demonstrated, and the chapter is concluded by a forward looking perspective addressing important not yet solved challenges.

1. Introduction

Materials science is entering an era where the growth of data from experiments and computations is expanding beyond a level that can be handled by established methods. The so-called "4 V challenge" – concerning *Volume* (the amount of data), *Variety* (the heterogeneity of form and meaning of data), *Velocity* (the rate at which data may change or new data arrive), and *Veracity* (the uncertainty of data quality) is clearly becoming eminent. Most importantly, however, is that big data of materials science offer novel, extraordinary, and expansive opportunities for achieving scientific knowledge and insight. These opportunities require new research concepts and lines of thought. While this chapter focusses on computational materials science, we emphasize that the described concepts and methods are valid for experimental data as well.

In most cases, the results of computational materials science studies are published as focused research studies, reporting only those few data that are directly relevant for the respective publication. Thus, even from very extensive computations (thousands or millions of CPU core hours) just very few results are shared with the community. Most data, in particular when they were not deemed of immediate relevance, are kept private or even thrown away. In November 2014, the principal investigators of the NOMAD Center of Excellence (NOMAD) launched an initiative at the Psi-k community (Psi-k network), proposing “a change in scientific culture” of computational materials science and engineering, i.e. the extensive sharing of data of this field – of *all* data: If and only if the full input and output files of computations would be shared, calculations don’t need to be repeated again and again, and the field will have access to big data which can be used in a totally new research manner, i.e. by artificial-intelligence methods. As will be elaborated in sections 2 and 5 below, one can find structure and patters in big data, gaining new insight that cannot be obtained by studying small data sets, and this way even allegedly useless data get value. A popular example from real life on the value of big-data analytics is the tracking of the movement of mobile phones which provides instantaneous information on traffic flow and jam. Another example is the local information of google searches for *flu symptoms and medicine* which reflect the spreading of a flue wave. Initially, many but not every colleague in computational material science liked the initiative towards all-embracing open access, but four years later it is clear that the overwhelming part of the community supports an extensive data sharing (sections 2 and 3).[1]

The NOMAD Center of Excellence (NOMAD) has assumed a pioneering role in data sharing and analytics, also considering all aspects (Draxl and Scheffler 2018) of what is now called the FAIR handling of data (Wilkinson et al. 2016):[2] Data are *FINDABLE* for anyone interested; they are stored in a way that make it easily *ACCESSABLE*; their representation follows accepted standards (Ghiringhelli et al. 2016), and all specifications are open – hence data are *INTEROPERABLE*.[3] All of this enables the data to be used for research questions that could be different from their original purpose; hence data are *RE-PURPOSABLE*.[4]

The chapter is structured as follows. In section 2 we briefly summarize the history of the four research paradigms of materials science, with particular focus on the fourth one, “big-data-driven materials science”. section 3 then stresses the importance of an extensive data sharing for the advancements of science and

[1] As important side remark, we note that the all-embracing sharing of research raw data helps the prevention of scientific misconduct.

[2] The concept of the NOMAD Repository and Archive (NOMAD) was developed in 2014, independently and parallel to the “FAIR Guiding Principles” (Wilkinson et al. 2016). Interestingly, the essence is practically identical.

[3] The understanding of “interoperable” is somewhat controversial. It may be defined as: 1) Metadata and data use a formal, accessible, shared, and widely accepted language for data representation; 2) The vocabulary of metadata and data follow FAIR principles; 3) Metadata and data include qualified references to other (meta)data and to the authors who created the results.

[4] The NOMAD CoE uses the term re-purposable, while in the FAIR concept it was termed re-usable. Both obviously mean the same in this context.

engineering and the (so far) empty rhetoric of scientific societies and funding agencies (Nature editorial 2017). In section 4, the NOMAD concept is highlighted and it is discussed how it is orthogonal to and how it plays synergistically together with other important databases, particularly AFLOW (AFLOW), Materials Project (Materials Project), and OQMD (OQMD)[5]. Section 5 addresses artificial-intelligence concepts for materials-science data with some specific examples. Finally, in section 6, we give an extensive outlook on the developments and open questions of big-data driven materials science.

2. Development of the four paradigms of material sciences

Figure 1 sketches the historical evolution of methods and concepts of materials science. Experimental research dates back to the Stone Age, and the basic techniques of metallurgy were developed in the Copper and Bronze Ages which started in the late 6th millennium BCE. The control of fire prompted a major experimental breakthrough. Towards the end of the 16th century, scientists began to describe physical relationships through equations. Well-known names from the early days are Tycho Brahe (1546-1601), Tomas Harriot (ca.1560-1621), Galileo Galilei (1564-1642), Johannes Kepler (1571-1630), Isaac Newton (1643-1727), and Gottfried Wilhelm Leibniz (1646-1716). The latter two also developed the concept of the mathematical differential and derivatives. Thus, analytical equations became the central instrument of theoretical physics. Second from the left in Fig. 1, this new way of thinking – paradigm – is symbolized by the Schrödinger equation. Needless to say, the first paradigm, the empirical and experimental sciences, did not become obsolete, but theoretical physics represents an important complementary methodology.

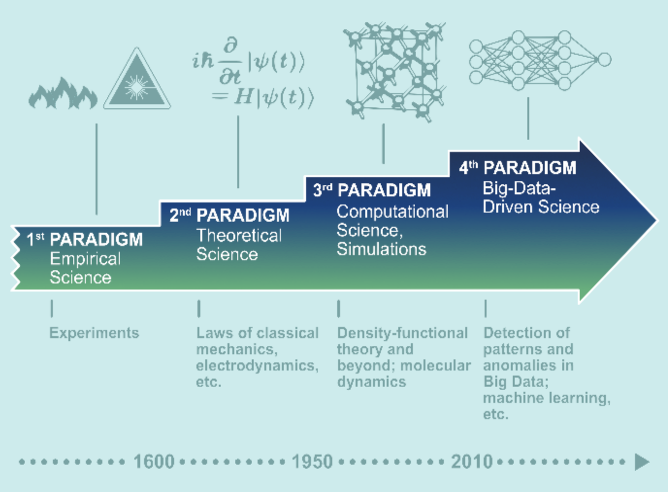

$i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle = H|\psi(t)\rangle$
1st PARADIGM Empirical Science
2nd PARADIGM Theoretical Science
3rd PARADIGM Computational Science, Simulations
4th PARADIGM Big-Data-Driven Science
Experiments
Laws of classical mechanics, electrodynamics, etc.
Density-functional theory and beyond; molecular dynamics
Detection of patterns and anomalies in Big Data; machine learning, etc.
1600
1950
2010

Figure 1. Development of the paradigms of materials science and engineering.

[5] Other important, databases with largely experimental data are the PAULING FILE and the Crystallography Open Database (COD), both presented in the handbook (Blokhin and Vilhars 2018, Gražulis et al. 2018)

Since the 1950s electronic-structure theory was advanced for materials by J. C. Slater (e.g. Slater 1937, 1957, 1963, 1965, 1967, Slater and Johnson 1972), the Monte-Carlo method was introduced by (Metropolis et al. 1953), and (Alder and Wainwright 1958, 1962, 1970) and (Rahman 1964) introduced molecular dynamics. (Hohenberg and Kohn 1964) and (Kohn and Sham 1965) laid the foundation of density-functional theory (DFT)[6] in the mid nineteen-sixties. All these developments enabled the numerical calculation and analysis of thermodynamics and statistical mechanics on the one hand, and the quantum-mechanical properties of solids and liquids on the other hand. They define the beginning of computational materials science, what is nowadays considered the 3rd paradigm. Herewith "computer experiments" were introduced, i.e. simulations, whose results are often treated and analyzed analogous to those of experimental studies. Initially developed independently, the fields of electronic-structure theory and statistical mechanics and thermodynamics are now growing together (Reuter et al. 2005 and references therein). Likewise, in parallel to DFT, many-body techniques, based on Green functions were introduced (Hedin 1965), which are now synergistically interleaved with DFT to form the forefront of electronic-structure theory, including excitations.

Today, big data and artificial intelligence revolutionize many areas of our lives, and materials science is no exception (Agrawal and Choudhary 2016). Jim Gray had probably first discussed this 4th paradigm (Gray 2007) arguing explicitly that big data reveal correlations and dependencies that cannot be seen when studying small data sets. A further important difference to the second paradigm is that we accept that many materials properties, i.e. patterns and correlations in big data, cannot be described in terms of a closed mathematical formulation, as they are governed by several, intermingled theoretical concepts and multilevel, intricate processes.

Figure 2 provides a schematic view of our vision: The chemical and structural space of different materials is practically infinite. However, when asking e.g. for

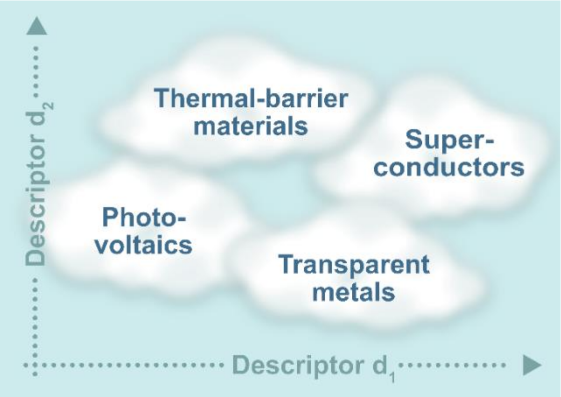


Figure 2. Big data contain correlations and structure that are invisible in small data sets. Finding descriptors that determine a specific property or function of a material is a crucial challenge. Once this being in place, we will be able to machine learn the data and eventually draw maps of materials.

[6] See the Chapter by (Li and Burke 2018)

good thermal-barrier coatings, there are just a few. Thus, in terms of functional materials the space is sparsely populated. Finding these regions or patterns that correspond to materials with superior functional performance requires the identification of descriptors, noted as d_1 and d_2 in Fig. 2. Obviously, in general the dimensionality will likely be higher than just two. At this point, Fig. 2 is just a sketch and for most properties the appropriate descriptors are unknown. Compressed sensing, machine learning, and other methods of artificial intelligence are able to identify these descriptors and patterns, and we will address these methods in section 5 below.

We close this section by noting the radically decreasing time scales of new developments and paradigms: 6th millennium BCE, 1600, 1950, 2010. Thus, a next research paradigm may be just ahead of us.

3. Extensive data sharing – why and how?

Data is a crucial raw material of this century. Our community is producing materials data by CPU-intensive calculations since many years. The results are stored on PCs, workstations, or local computers. Most of these data are not used and often even thrown away, though the information content is significant. The field is now slowly changing its scientific culture towards *Open Science* and *Open Data*, and there are many reasons for doing so. Open access of data implies that data can be used by anyone, not just by the experts who develop or run advanced codes. If data were openly available, many more people would work with the data, e.g. computer scientists, applied mathematicians, analytic condensed matter scientists, and more. We will be surprised what people will do with data when they are made available, probably using tools that the present computational materials science community does not even know. The NOMAD Kaggle competition with 883 teams participating in a study of transparent conducting metal oxides is a nice example (Kaggle/NOMAD2018; Sutton et al. 2018). We note that our considerations are not project specific but general, even though several examples below will be taken from the NOMAD Center of Excellence which reflect the synergetic relationship of NOMAD with other major key data bases, in particular AFLOW, Materials Project, OQMD (see references: AFLOW, Materials Project, OQMD, NOMAD)[7].

Without a proper data infrastructure that enables efficient collection and sharing of data, the envisioned success of big-data-driven materials science will, however, be hampered. For the field of computational materials science, the NOMAD Center of Excellence (NOMAD) has changed the scientific culture toward comprehensive data sharing. On a more formal basis, and in parallel to NOMAD, the "proper" way of collecting data was suggested as the *FAIR Guiding Principles* (Wilkinson et al.

[7] The MARVEL consortium recently designed and implemented "Materials Cloud", a web portal that is coupled to the AiiDA workflow manager (see Pizzi 2018 for details). Contributions to their database is upon application.

2016). We emphasize that NOMAD had employed the same principles, and thus NOMAD data are an example of FAIR data.

So what is behind FAIR? What does it mean for computational materials science?

The *F* stands for ***findable***. Making research data open and keeping them for at least ten years is now requested by many research organizations. Seen from a practical point of view, it is also useful to avoid doubling of work and thus save human and computational resources and energy. Since individual researchers create their data on various platforms – from workstations to compute clusters to high-performance computing (HPC) centers –, it is often impossible to find a student's or postdoc's data, some time after she/he has left the research group. Besides matters of organization, issues may be related to automatic erasure of data in HPC centers, missing permissions on local machines, data protection, and alike. Clearly, making data findable requires a proper data infrastructure, including documentation, search engines, and hardware. This is one of the reasons why the NOMAD Repository was established (NOMAD).

The *A* stands for ***accessible***. Accessibility in materials science has different facets. First, we should not forget about the proper hardware that allows for swift access to data. Second, we need to provide application programming interfaces (APIs). Perhaps not formally required in the FAIR sense, but extremely useful for getting a first insight into materials data, is to provide the data not only in a machine-readable but also in a human-accessible form. Seeing results of calculations clearly supports understanding – as realized in the NOMAD Encyclopedia (NOMAD) and Advanced Visualization Tools (NOMAD). To make data fully accessible requires an important additional step which is the formal description of the data, its metainfo (NOMAD Metainfo). This connects to the ***I*** in FAIR.

The *I* stands for ***interoperable***. Here we need to consider in a first place the extreme heterogeneity of computational materials data. The wider community is using about 40 major codes (considering electronic-structure, molecular-dynamics, and quantum-chemistry packages for materials) that differ in various aspects of methodology and implementation. Consequently, the necessity arises to make their results comparable, which is a major challenge not only in the sense that they need to be brought to a common format and common units; we also recall that one quantity may be named differently in different (sub-)communities or one and the same expression may have a different meaning in one or the other area. Thus, "dictionaries" are needed to translate between them.

Still, we need to ask the question whether tools to make data human-accessible, as mentioned above, or artificial-intelligence approaches can operate upon all available data in a meaningful way. Apart from formats and units, there are more severe restrictions that may hamper such important tasks. These concern the computational parameters – and consequently the numerical precision – that are used in different calculations. We recall here that all NOMAD Repository data can be considered valid in the sense that, besides code and codes version, input and output files[8] are known. Thus all data are valid and reproducible as such. Nevertheless, data have been created for different purposes which may require different levels of convergence in terms of basis-set size and alike.[9] More than that, we may even ask whether different codes aiming at the solution of one and the same problem with "safe" settings give the same results. For the latter, we point to the community effort led by Stefaan Cottenier (Lejaeghere et al. 2016), where the equations of state for 71 elemental solids were calculated with many different *ab initio* electronic-structure packages. Over a time span of a few years, it turned out that upon improvements of codes, basis sets, and in particular pseudopotentials, all codes lead to basically the same answer. In fact, the first thirteen entries in the list[10], differ by an average error of less than 0.5 meV per atom in total energies. Such investigations are extremely helpful and have set the stage towards a culture of benchmarking, which had been established in quantum chemistry for molecules already decades ago. This study by (Lejaeghere et al. 2016) is, however, only the beginning. Clearly, other properties like energy barriers, band gaps, spectra, etc., and systems like surfaces, interfaces and inorganic/organic hybrid materials, etc. will be much less forgiving than total energies of simple bulk solids and will make discrepancies more obvious. Therefore, more investigations along these lines are on the way.

While the above comparison (Lejaeghere et al. 2016) could only be made with parameter sets that represent full convergence, calculations performed in daily life, are often far from this optimal case, and are, in fact, often sufficient. This situation obviously leads to the question how to compare and operate on calculations that have been performed with different settings, e.g. in terms of basis sets, meshes for Brillouin zone integrations, and alike. Below, it is shown that this is, in fact, possible (Carbogno et al. 2018).

Let us demonstrate that fully converged results of complex materials can be predicted by learning from errors of calculations of simple materials (Carbogno et al. 2018). Four different codes – **exciting** and FHI-aims, two all-electron full-potential codes, and GPAW and VASP, two projector augmented planewave (PAW) codes – have been employed for this study. For the 71 elemental solids adopted from (Lejaeghere 2016),

[8] Without the full input file and the main output file(s) (NOMAD) does not accept uploads.
[9] For example, a band structure may needs less stringent parameters than atomic forces. We also note that what we regarded "converged" a few years ago, may not be considered precise enough today. This should not devalue older calculations (see also the discussion of Figs. 4 and 5).
[10] At https://molmod.ugent.be/deltacodesdft, one has to choose a reference where obviously an all-electron code is the natural choice. In fact, the precision of the all-electron packages WIEN2k, **exciting**, and FHI-aims are practically identical, and these codes are leading the whole list.

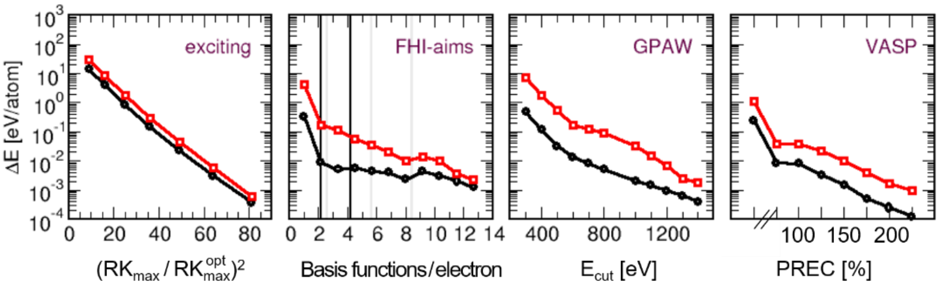


Figure 3. Convergence of total energies of 71 elemental solids with increasing basis-set quality for four different electronic-structure codes. Red symbols indicate materials with maximal error, black symbols display averages over all 71 materials. All values are taken with respect to the respective fully converged results. RK_{max} is the LAPW-specific parameter defining the basis set size, E_{cut} the GPAW cutoff energy, PREC is the VASP-specific parameter for basis set quality. The black lines for FHI-aims indicate light (left) and tight (right) settings, the gray lines Tier1, Tier2, and Tier3 (from left to right).

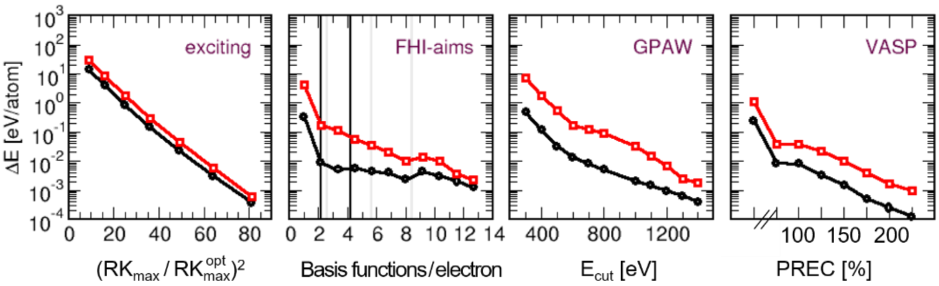
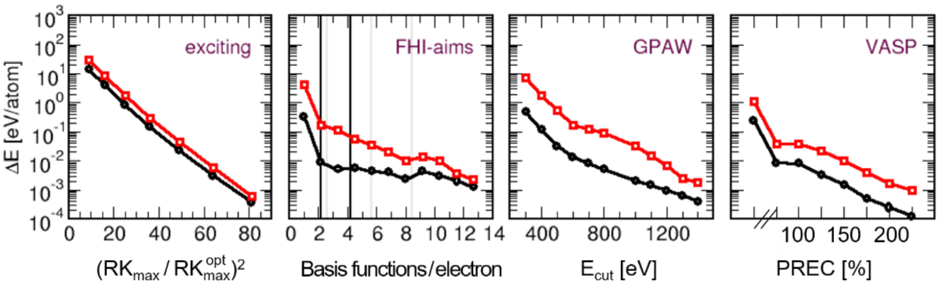


Figure **3** shows how the total energies for fixed geometries change as a function of basis set quality. Incomplete basis sets are, indeed, the most severe source of discrepancies of different calculations in publications and/or the various data repositories. The red symbols in Figure **3** mark the materials exhibiting the largest error and the black symbol refer to the average taken over all materials. The error, ΔE, for each material is defined with respect to the fully converged value. In all cases, the error decreases systematically from the order of 1eV for small basis sets by about four orders of magnitude below meV precision.[11] Based on these errors we now estimate for multi-component systems the errors arising from the basis-set incompleteness, utilizing a simple analytic model, i.e., linearly combining the respective errors of the constituents (elemental solids) for the same settings:



$$\overline{\Delta E_{tot}} = \frac{1}{N} \sum_N N_i \Delta E_{tot}^i \ .$$

[11] The fact that not all codes show a monotonous behavior for small basis set sizes has to do with specifics of the basis set. Here we refer the reader to (Carbogno 2018).

Here N_i is the number of atoms of species i present in the alloy and ΔE_{tot}^{i} is the error of this species in the respective elemental solid. This model is applied to 63 binary solids that were chosen such to optimally cover the chemical space (one for each element with atomic number up to 71, without noble gases). The results are confronted to those of corresponding DFT calculations for these binaries, depicted in Figure 4. The authors of this work find a clear correlation between predicted and actual errors for all four codes (with the details depending on peculiarities of the specific basis set). In particular, it is confirmed that the better converged the calculations are for the simple systems – what is, in fact, easy to reach – the smaller the errors are in the predictions. For more detailed analysis, also including relative energies and ternary alloys and an in-depth discussion about the role of the employed method/basis set, we refer the reader to (Carbogno et al. 2018). In essence, the conclusion can be drawn that even not having fully converged values for complex materials in hand, one can predict the energetics for the fully converged case. This study sets the stage for the NOMAD data fulfilling the stringent condition for the ***I***.

The *R* stands for ***re-usable***. In fact, we prefer the term re-purposable that gives a better impression about what is meant in the materials-science context. It means that we can use data that has been created for some specific question, in a different connection. Indeed, one and the same material can be considered for various applications. So why should a researcher working on one aspect not allow another researcher to use the same data for focusing on another aspect? Let us illustrate this with the example of TiO_2 which is an important support material for heterogeneous catalysis. The detailed results are not only useful for researchers working in this area, but also in a different context: for example for photovoltaics where TiO_2 is a component of dye-sensitized solar cells, or cosmetic products where TiO_2 is used, e.g., in sunscreens.

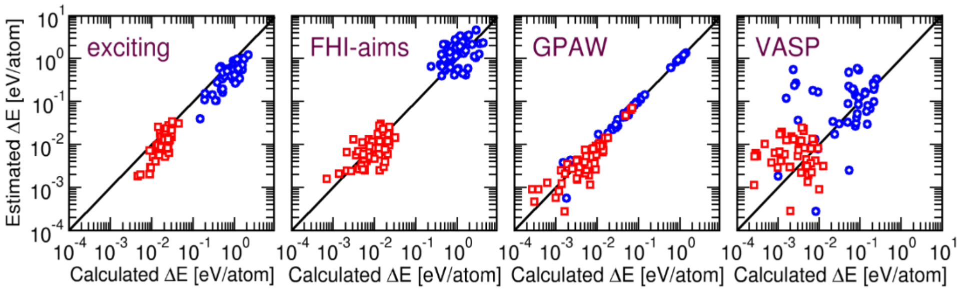


Figure 4: Predicted versus actual errors in total energy for 63 binary alloys, obtained from the errors in elemental solids for four different electronic-structure codes.

It is generally agreed that research results obtained in academia should be published. In view of what was discussed above, it should be a duty to publish all the results, i.e. making also the complete data underlying a publication available. This has been said by many people, funding agencies, politicians, and research journals. Indeed, a few research journals have starting doing so. However, research agencies do not support the necessary data infrastructure, so far. Let us cite from a recent Nature Editorial “Empty rhetoric over data sharing slows science” (Nature editorial 2017): “Everyone agrees that there are good reasons for having open data. It speeds research, allowing others to build promptly on results. It improves replicability. It enables scientists to test whether claims in a paper truly reflect the whole data set. It helps them to find incorrect data. And it improves the attribution of credit to the data’s originators. But who will pay? And who will host?” – and further “Governments, funders and scientific communities must move beyond lip-service and commit to data-sharing practices and platforms.” For computational materials science though, NOMAD had already changed the culture and implemented the platform (Draxl et al. 2017).

4. The NOMAD concept

As briefly outlined in the previous section, NOMAD (NOMAD) has initiated extensive data sharing in computational materials science. The NOMAD Laboratory processes, cleanses, and hosts computational materials science data, computed by all important materials-science codes available today and makes this data accessible by providing several related data services. The big picture is to advance materials science by enabling researchers in basic science and engineering to understand and utilize materials data to identify new materials and, in turn, pave the way to novel products and technologies. NOMAD also collaborates with many researchers and all other big databases, specifically AFLOW (AFLOW), Materials Project (Materials Project), and OQMD (OQMD), Computational Materials Repository (CMR), and others. In the following, NOMAD’s corner stones are described in some more detail.

The **NOMAD Repository** forms the basis of all NOMAD tools by processing and hosting the raw materials data. Made publically available in early 2014, it contains by now the input and output files from millions of high-quality calculations and became the world’s largest collection of computational materials science data. It comprises calculations that have been produced with any of the leading electronic-structure codes and increasingly also with codes from quantum chemistry. Presently, NOMAD supports about 40 codes, and less-frequently used or newly established codes will be added on demand. We point to the orthogonality compared to these latter databases, and emphasize that the NOMAD Repository is not restricted to selected computer codes or closed research teams but serves the entire community with its ecosystem of very different computer codes.

By hosting raw data, the NOMAD Repository not only helps researchers to organize their results and make them available to others keeping scientific data for free for at least 10 years[12]. The NOMAD Repository was the first repository in materials science recommended by Scientific Data as stable and safe long-time storage.

[12] NOMAD guarantees to keep data for at least ten years from the last upload. In practice this means that overall, data are kept much longer than ten years.

Upload of data is possible without any barrier. Results are requested in their raw format as produced by the underlying code. For downloading data, not even registration is needed. NOMAD sticks to an open data policy, where data are published according to the Creative Commons Attribution 3.0 License. Nevertheless, uploaders can keep their data secret for a certain period that can be used for publishing the results and / or restricting the access to a selected group of colleagues (or referees). After a maximum period of three years though, all data become open access.

Figure 3 reflects how actively the entire community is participating in the NOMAD initiative. It depicts a snapshot of the NOMAD Archive by March 15, 2018, showing the number of uploads (total-energy calculations) of data from various community codes.

The **NOMAD Archive** hosts the normalized data, i.e. the open-access data of the NOMAD Repository converted into a common, code-independent format. In other words, numerous parsers have been developed that read out and translate all the information contained in in- and output files. This ensures the ***I*** in FAIR, namely that data from different sources can be compared and, hence, collectively operated upon by various NOMAD (and other) tools.

As mentioned already above, a clear metadata definition is a prerequisite for this normalization step to make sure that the results/values obtained atomistic or *ab initio* calculations are correctly interpreted by the parsers. Such activity should be community driven and the result of a key workshop on the goal was published in (Ghringhelli et al. 2016, 2017). The development of an open, flexible, and hierarchical metadata classification system (NOMAD Metainfo), as achieved by NOMAD, was indeed challenging. Obviously, as codes are continuously updated and extended, and new codes may be developed, this is an ongoing process, to which everybody is welcome to contribute.

The **NOMAD Visualization Tools** allow for remote visualization of the multi-dimensional NOMAD data through a dedicated infrastructure developed within the NOMAD CoE. A centralized service is provided that enables users to interactively perform comprehensive data visualization tasks on their computers without the need for specialized hardware or software installations. Interactive data exploration by virtual-reality (VR) tools is a special and most successful focus. Also for this branch of the toolbox, users have access to data and tools using standard devices (laptops, smartphones), independent of their location. Such VR enhances training and dissemination and even were a great success, e.g. when presented to the general public during the Long Night of Sciences, taking place in Berlin, in June 2017 (see Figure 6). As an example, we note that 360-degree movies can be even watched with simple Google cardboard glasses as demonstrated, e.g. for CO_2 adsorption on CaO and excitons in LiF. The latter, being six-dimensional objects, cannot easily be visualized in a standard way . Taking the position of an electron

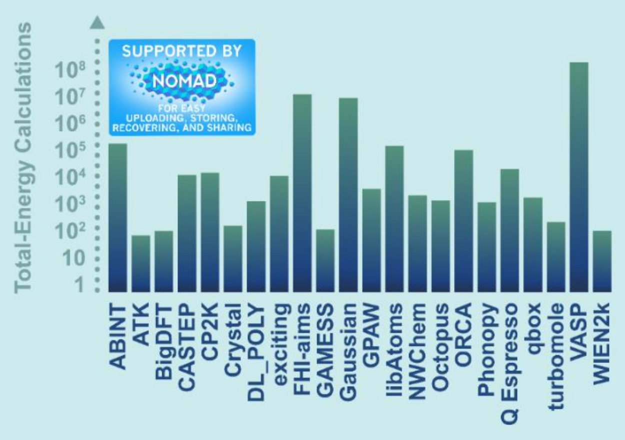
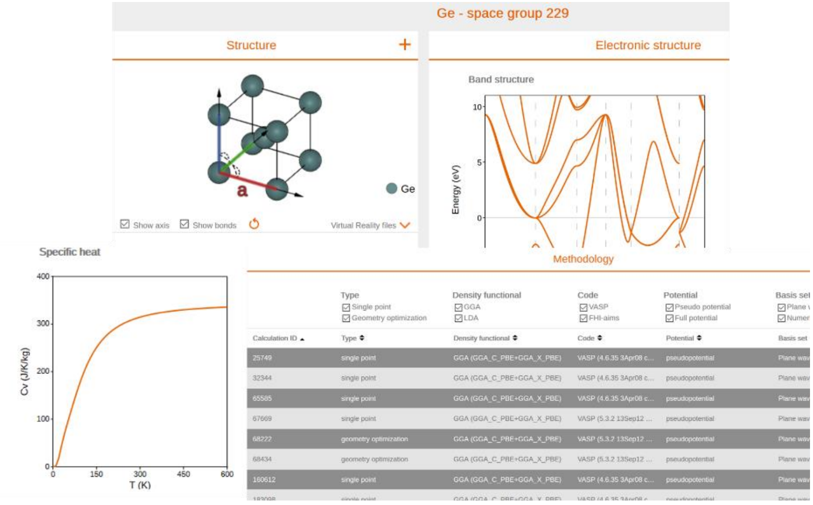


Figure 5. The NOMAD Laboratory supports all important codes in computational materials science. The figure shows the number of uploaded open-access total-energy calculations at the NOMAD Repository (NOMAD) as of March 15, 2018. The abscissa shows the various codes with more than 80 uploads. The total number of open-access total-energy calculations at the NOMAD Repository is more than 50 million, corresponding to billions of CPU-core hours. The stamp “supported by NOMAD” can be found at the homepage of many *ab initio* software packages of computational materials science.

Figure 6. Virtual-reality setup as demonstrated during the Long Night of Sciences, Berlin 2017.

or a hole, VR allows for inspecting the space of its counterpart. With this example, we demonstrate how seeing indeed helps understanding.

The NOMAD CoE has created its data infrastructure not only for collecting and sharing data but also to let us see what all this data contain. The **NOMAD Encyclopedia** is a web-based public infrastructure that gives a materials-oriented view on the Archive data that helps us to search for the properties of a large variety of materials. It serves the purpose of in-depth characterization and understanding of materials by providing knowledge of their various properties. This includes structural features, mechanical and thermal behavior, electronic and magnetic properties, the response to light, and more. Whatever property of a given material has been computed, it should be easily accessible through a user-friendly graphical user interface (GUI). Having all this information in one place, gives us an impression about the wealth of available materials data and allows for comparing even very different systems in terms of certain features. The NOMAD Encyclopedia allows us see, compare, explore, and comprehend computations obtained with various tools and different methodology (for a collage of snapshots,



Type
☑ Single point
☑ Geometry optimization

Density functional
☑ GGA
☑ LDA

Code
☑ VASP
☑ FHI-aims

Potential
☑ Pseudo potential
☑ Full potential

Basis set
☑ Plane
☑ Numer

Calculation ID ▲	Type ⬍	Density functional ⬍	Code ⬍	Potential ⬍	Basis set
25749	single point	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (4.6.35 3Apr08 c...	pseudopotential	Plane wav
32344	single point	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (4.6.35 3Apr08 c...	pseudopotential	Plane wav
65585	single point	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (4.6.35 3Apr08 c...	pseudopotential	Plane wav
67669	single point	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (5.3.2 13Sep12 ...	pseudopotential	Plane wav
68222	geometry optimization	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (5.3.2 13Sep12 ...	pseudopotential	Plane wav
68434	geometry optimization	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (5.3.2 13Sep12 ...	pseudopotential	Plane wav
160612	single point	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (4.6.35 3Apr08 c...	pseudopotential	Plane wav
183098	single point	GGA (GGA_C_PBE+GGA_X_PBE)	VASP (4.6.35 3Apr08 c...	pseudopotential	Plane wav

Figure 7. Collage from Encyclopedia snapshots.

see Figure 7). We can directly assess the spread of results and, as for instance, measure the impact of a functional on a given feature and materials class.

So far, we process structural and electronic properties and thermal behaviors, and alike for bulk materials and low-dimensional systems. The NOMAD Encyclopedia is constantly extended in terms of new data, other system types, and properties. And soon, the Encyclopedia will handle molecules, surfaces and adsorbate systems, the response to external excitations, elastic properties, Fermi surfaces, molecular-dynamics, and more.

Furthermore, the Encyclopedia provides a material classification system, information on various levels, e.g. about computed quantities or the methodology behind the calculations, as well as links to external sources, like to useful Wikipedia pages. We also point to the error-reporting tool, implemented for the case of problematic data. Should there be a dataset or a graph that does not appear to be correct, making use of a simple pull-down menu, the user can let us know about it.

Data-driven science will be discussed in detail in the next section. Therefore, we only briefly mention the **NOMAD Analytics Toolkit** for the sake of completeness. It provides a collection of examples and tools to demonstrate how materials data can be turned into knowledge and understanding. They concern topics like crystal-structure prediction, property prediction, error estimates, classification of materials, and more. To explore these tools, there is no need to install any software, no need for registration, and no need for computational capacity. Like the visualization tools, also the analytics toolkit allows for remote usage.

Let us conclude this section by pointing at informative YouTube movies that describe the NOMAD project as a whole https://youtu.be/yawM2ThVlGw, the Repository https://youtu.be/UcnHGokl2Nc, and the Analytics Toolkit https://youtu.be/UcnHGokl2Nc.

5. Artificial intelligence concepts for materials science data

In this chapter, we are using the term artificial intelligence (AI) as umbrella term of computational methods that "learn from experience". We like to emphasize upfront, that a proper analysis of data needs to consider at least some aspects of the causality that drives the correlations of interest, i.e. one needs to include domain knowledge in the learning process in order to achieve a predictive description, interpretability and possibly even deeper understanding of the cause behind the structure or patterns in the data. The lower the employed domain knowledge is the more data are needed in the learning process, and it may happen that data are fitted but predictions and even interpolations are not reliable. AI is a wide and interdisciplinary field, and machine learning (learning from data) and compressed sensing (originating from signal compression; aiming at modeling in terms of low dimensional descriptors) are important subdomains.

As noted above (see the discussion of Fig. 2), big data may reveal correlations (structure and patters) if and only if the data are arranged in a proper way, e.g. represented by appropriate descriptors. These correlations can be found by AI but the identification of such correlations does not necessarily go along with deeper insight or understanding. To some extent we like to argue, that the wish for insight is often overrated. This is well documented by the Periodic Table of the Elements, that may arguably be considered as one of the most impacting achievements for chemistry, condensed matter physics, engineering, and biophysics:. When Mendeleev published his table in 1871, based on knowledge of 63 atoms (their weights and chemical behavior), there was no understanding of the deeper cause behind the structure of the table (Scerri 2008; Pyykkö 2012). Still, the table predicted the existence of at that time unknown atoms, and even their properties where described. However, the underlying causality, i.e. that the different rows reflect the different number of nodes of the radial wave functions of the outer valence electrons, and that the different columns refer to the number of valence electrons, was unknown when the table was created. It only was understood about 50 years later, when the shell structure of electrons in atoms was described by quantum mechanics.

Thus, identifying correlations, structures, and patterns in big data is an important step by its own. When the relationship between a property of interest, P, and a set of useful descriptive parameters (the descriptors d_1, d_2, ... – sometimes called representation) is known, graphs as in Figure 2 or approximate equations can be obtained for the relationship $P(d_1, d_2, ...)$. For the example of the Periodic Table, the descriptors are the row and column numbers. Obviously, as the number of possible materials is practically infinite, building a map as in Fig. 2 is a highly demanding task, of much higher complexity than building the Periodic Table of the Elements.

How to find the descriptors for materials properties? The direct descriptor for a quantum-mechanical problem is given by the position of all atoms, the nuclear numbers and the total number of electrons: $\{R_I, Z_I\}$, N^e. This fully describes the many-body Hamiltonian but it is too complex to learn. Thus, the amount of data needed for a training (fitting) a function based on $\{R_I, Z_I\}$, N^e is unrealistically high. We distinguish 3 concepts for choosing proper descriptors: a) the descriptor may be selected out of a huge, systematically created pool of candidates; b) the descriptor may be built in the machine-learning step in an abstract manner; and c) one may just use a (known) descriptor assuming that with many data the actuals choice may be not so important. Concept a) will be discussed below when we describe compressed sensing and subgroup discovery. Concept b) is realized in neural-network approaches which, in the learning step, minimizes an objective function that quantifies the difference between the predicted and the correct (known) data. Through this minimization, the weights (i.e. parameters) of the neural network are optimized (Hinton 2006, Hinton et al. 2006), and in this way, the network learns the descriptors. Doren and coworkers (Blank et al. 1995, Lorenz et al. 2004, 2006) have shown early examples of neural network potential energy surfaces. Hellström and Behler describe recent advances in their chapter (Hellström and Behler 2018).

Concept c) is probably the most widely used approach. It is nicely described in the chapter by Ceriotti, Willatt, and Csányi (Ceriotti et al. 2018).

A systematic understanding of the suitability of various machine-learning (ML) models and thorough benchmarking studies are still lacking in materials science. It was only recently addressed in terms of a public data-analytics competition that was hosted on the internet platform Kaggle using a dataset of 3,000 $(Al_xGa_yIn_z)_2O_3$ compounds ($x+y+z=1$), spanning 6 different crystal structures. The aim of this challenge was to identify the best machine-learning model for the prediction of two key physical properties that are relevant for optoelectronic applications: the electronic band gap energy and the crystal formation energy. Impressively, 883 teams contributed, using many different methods (Kaggle/NOMAD2018). The interested reader is referred to a recent publication (Sutton et al. 2018).

Some warnings may be appropriate. In general, an observed correlations will have a causal reason – provided that it is supported by a large data set (Pearl 2009). Thus, a correlation that is described by the function $P(d_1, d_2, ...)$ may reflect that $(d_1, d_2, ...) = \boldsymbol{d}$ are the actuators: $\boldsymbol{d} \rightarrow P$. However, it could well be that the reverse is true: $P \rightarrow \boldsymbol{d}$. Thirdly, it is possible that there is an "external master", *M*, who controls both $\boldsymbol{d}$ and *P*, with no direct relationship between $\boldsymbol{d}$ and *P*. And fourthly, the data may be selected with a significant bias of the researcher or research community. We fear that this may be happening much more frequently than realized. But then the observed correlation may just reflect this bias. All this needs to be kept in mind when tools of artificial intelligence are applied to big data and when we ask for interpretability.

Let us add another warning about big data of materials science. The number of possible materials is practically infinite, and we like to identify new materials that have better performance or functionality than the materials that are used today. Clearly, the amount of available data in materials science is getting big though from the few (about 300,000) materials that have been synthesized up to now, we often just know the atomic structure and hardly their electronic, elastic or other properties. Getting more and more data, however, does not imply that all the data is relevant for all properties of interest. Materials science is ruled by a significant number of different properties and mechanisms, and experience seems to show that at the end, the number of materials that are good for a certain group of functions is very small. Thus, the amount of available data may be big but the number of *relevant* data is small. For example, if we ask for a highly transparent materials with excellent heat conductivity and scratch resistance there is probably nothing better than and nothing even close to diamond. Thus, in materials science and engineering, we are looking for "needles in a hay stack". This is different to typical AI challenges in social science and medicine where a 80% chance of being right is often considered a great success. In materials science we need values as high as or even higher than 99.9%. Alternatively, we need methods that identify the relevant subgroups in the large amount of data right away.

Let us sketch this challenge for kernel methods of machine learning approaches. The property of interest is written as a sum over a large, appropriate subset of all known data *j*

$$P(\boldsymbol{d}) = \sum_{j=1}^{N} c_j K(\boldsymbol{d}, \boldsymbol{d}_j) \quad . \tag{1}$$

There are many options for the kernel *K*. A most popular and very successful choice is the Gaussian kernel

$$K(\boldsymbol{d}, \boldsymbol{d}_j) = \exp(-(\boldsymbol{d} - \boldsymbol{d}_j)^2 / 2\sigma_j^2) \quad . \tag{2}$$

Fitting a set of say *N*=100,000 known data is achieved by determining 100,000 coefficients by minimizing

$$Min\,\{\sum_{i=1}^{N} (\hat{P}_i - P(\boldsymbol{d}_i))^2 - \lambda \| m \| \} \ . \tag{3}$$

Here $\hat{P}_i$ are the actually known data that should be fitted, and we also introduced a regularization which prevents overfitting and creates a result that does not go exactly trough the known data points but is smooth. This regularization is noted as "norm of the applied model, *m*". Figure 8 gives an example of such fitting/machine-learning approach: Obviously, fitting *N* data points with a function that contains *N* free parameters must work, but the regularization creates some uncertainty (a smooths curve), and the interpretability of the many determined coefficients is typically lacking. Getting back to the "needles in the hay stack" analogue, we note that if >99% of the data reflect the hay, and just <1% data points reflect the needles, the latter will not play a noticeable role in the fitting process when minimizing the

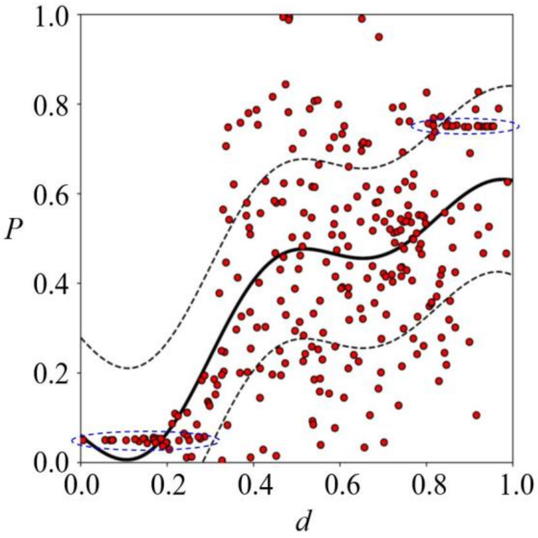


Figure 8. Sketch for a kernel method (Eq. (1)) fit of 1,000 data points (full line) and the confidence interval which contains 69% of all the data using Gaussian process regression. Also noted are two subgroups (regions of blue dashed lines) that are statistically exceptional but ignored in the global kernel-method description (Boley 2017).

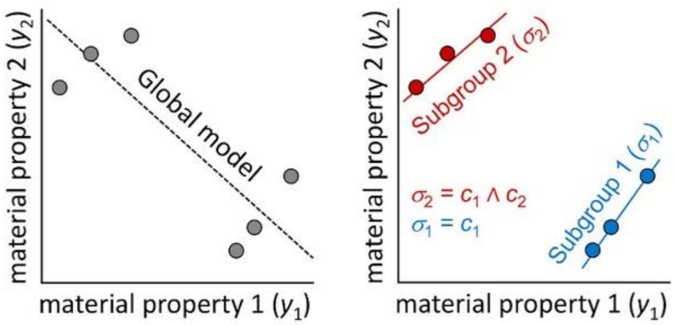


Figure 9. Left, data points and a global regression machine-learning model predicting a negative relationship between material properties y_1 and y_2 . Right, subgroup discovery identifies statistically exceptional regions marked as red and blue, and machine learning of these two regions exhibits positive slope. Subgroup discovery aims at describing such subpopulations by Boolean selector functions (σ_1 and σ_2) defined as conjunctions (logical AND, denoted as $\wedge$) of basic selectors (the c_i).

regularized least square error (Eq. 3). In other words, the regularized least square error minimization puts the overwhelming weight on the description of the hay. Two subgroups (statistically exceptional regions) are also noted in Figure 9. These are data parallel to the *d* axis but not described well by the employed kernel approach. This example reveals that the general statement “more data provide a better description” is wrong in ML for materials science. It essentially means: Add more irrelevant information (more hay) to the information pool (the hay stack). Obviously, this will not help to find the needles. Alternatively, could we turn this around? Can we attempt to fit the hay and then consider, the few materials that are distinguished by a high fitting error as an interesting subgroup that contains the needles? The difficulty here is that materials are very heterogeneous, and this heterogeneity is not just restricted to the direct hay-needle comparison. Also the “hay” is heterogeneous. Obviously, the goal should be to optimize e.g. the maximum error, but this requires new methods.

Nevertheless, whenever we attempt a global description, machine learning is a great tool. The chapter by (Huang et al. 2018) gives an excellent description, and the (Kaggle/NOMAD2018, Sutton et al. 2018) competition is a good example.

Two interpretability-driven approaches have recently been adopted by materials science. These are *subgroup discovery* on the one hand and *compressed sensing* on the other. Let us introduce them briefly.

Subgroup discovery

As noted above, a global model addressing the quantitative description of the entire dataset may be difficult to interpret. In many materials-science applications, local models describing subgroups would be advantageous. For illustration (see Figure 9), a globally optimal regression model could predict a negative relationship between the material properties. However, a subgroup discovery analysis may reveal that there are two statistically exceptional data groups (indicated by blue and red color in the figure). Thus the relationship in the data set does not have a negative slope (the global model) but a positive slope (the two subgroups). As a physical example, the transition metals of the Periodic Table are a subgroup, and the actinides, lanthanides, and halogens are other subgroups. Thus, identification of subgroups is useful to gain an understanding of similarities and differences between systems.

The concepts of subgroup discovery (SGD) was introduced in the early 1990s, when the advent of large databases motivated the development of explorative and descriptive analytics tools as an interpretable complement to global modeling (Duivesteijn et al. 2016, Klösgen 1996, Atzmueller 2015, Herrera et al. 2011, Siebes 1995, Wrobel 1997, Friedman and Fisher 1999). Simply speaking, the identification of subgroups is built on 3 components: *i)* The use of a description language for identifying subpopulations within a given pool of data. These are called selectors and typically Boolean expressions, e.g. "the ionization potential of atom A minus the ionization potential of atom B should be smaller than *X*" where *X* is a number that may be chosen iteratively. *ii)* The definition of utility functions that formalize the interestingness (quality) of subpopulations. This may include requests as "the band gap of the material should be in between 1.1 and 1.5 eV AND the cohesive energy should be larger than 3 eV"; and *iii)* The design of a Monte Carlo search algorithm to find selectors that describe the subpopulations of interest (Goldsmith et al. 2017). Figure 10 illustrates the idea for a recent study of heterogeneous catalysis: Finding potential catalysts that can transform the greenhouse gas CO_2 into useful chemicals or fuels (Mazheika et al. 2018). This study concentrated on metal oxides and realized that a global description (fitting all the data at once) did to provide an accurate description. However, searching for subgroups by considering many potentials selectors reveals that the adsorption energies of CO_2 , contain a subgroup that is statistically exceptional (marked in red in Figure 10). The best selector is determined as follows: The center of the O 2*p* density of states should be at an energy above 7.2 eV (relative to the vacuum level) AND the work function of the surface should be smaller than 5.3 eV. These are properties of the clean oxide surface, and the subgroup identified in this way contains surface sites that are more ionic than those of the remaining (black) systems.

The "interestingness (quality) of the subpopulation" was defined in this study in terms of a quality function that simultaneously maximizes the size of the subgroup and the deviation of the median of the target property distribution within the subgroup from the median for the whole data set, and minimizes the width of the target property distribution within the subgroup. For the example in Figure 10, the target property is the CO_2 adsorption energy. Details can be found the (Mazheika et al 2018).

The identified red subgroup is characterized by the fact that the surface oxygen sites of these materials are more strongly ionic than the oxygen sites for the remaining systems (Mazheika et al. 2018). Fitting the identified subgroup (the red systems) and the remainder (the black systems) independently, provides a good description of the CO_2 adsorption energy.

Compressed Sensing and the SISSO Approach

As noted in the discussion of Fig. 2, finding a descriptor (2-dimensional in Fig, 2), i.e. the set of parameters that capture the underlying mechanism of a given materials property or function, is the key, intelligent step toward identification of structure or patterns in (big) data. This central role of the descriptor was only recently addressed explicitly and systematically in the works of Ghiringhelli and coworkers (Ghiringhelli et al. 2015, 2016; Ouyang et al. 2018). These authors recast the descriptor-identification challenge into a systematic formulation using compressed sensing (CS).

The CS approach had been originally introduced for efficiently reproducing a high-quality signal from a very small set of observations (Candès and Wakin 2008, Nelsen et al. 2013, Candès et al. 2006, Donoho 2006)]. Mathematically, this can be sketched as follows. Given a set of data P_1, P_2, … P_N, where j=1-N labels different materials (or different conditions), CS finds the sparse solution $\boldsymbol{c}$ of an underdetermined system of linear equations

$$P(\boldsymbol{d}_i) = \sum_{k=1}^{M} \hat{c}_k \, d_{ki} \quad = \hat{\boldsymbol{c}} \cdot \boldsymbol{D} \quad . \tag{4}$$

$\{d_{ki}\} = \boldsymbol{D}$ is called the sensing matrix with the number of rows k=1-M significantly bigger than the number of columns, i=1-N. Thus, the sensing matrix is built from N vectors (the columns), each of length M. Material i is characterized by vector i, i.e. by k=1-M descriptive parameters, d_{ki}. Equation (4) corresponds to Eq. (1) when the linear kernel is used. If most elements of the vector $\hat{\boldsymbol{c}}$ were be zero, specifically when the number of nonzero elements of $\hat{\boldsymbol{c}}$ is smaller than N, the dimensionality of the problem is reduced (Candès et al. 2006; Donoho 2006; Candès and Wakin 2008). In order to achieve this reduction, the coefficients c_k are determined by solving Eq. (3) with the norm ||m|| taken as the l_0 norm of $\hat{\boldsymbol{c}}$. The norm zero is defined

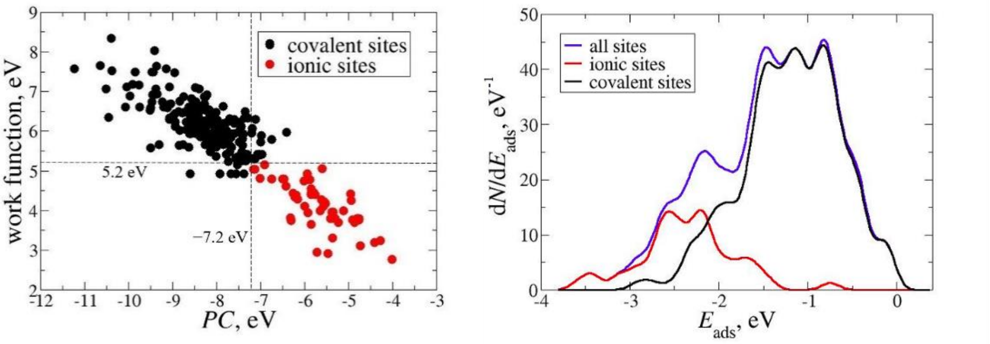


Figure 10. Subgroup of oxide materials in the CO_2 adsorption energy data that may be characterized as "largely ionic" surfaces. Left: The selectors determined by SGD are the work function (W) and the O 2p-band center (PC). Right: Density of samples with respect to adsorption energy for the whole data set as well as for identified "ionic subgroup". The rest subgroup of "high adsorption energies" (black dots and black line) is defined by (PC > −7.207 eV) AND (W < 5.182 eV).

as the number of non-zero elements. Thus, the regularization $\lambda\|\boldsymbol{c}\|_0$ can be also called a constraint that favors solutions for $\hat{\boldsymbol{c}}$ where most elements of $\hat{\boldsymbol{c}}$ are zero. However, using the norm zero poses a mathematical combinatorial problem, and it has been shown that this is (asymptotically) NP hard. As a consequence it has been suggested to approximate the norm zero by the norm l_1, and a popular approach then is LASSO (least absolute shrinkage and selection operator) (Tibshirani 1996). For materials science this has been developed by Ghiringhelli and coworkers (Ghiringhelli et al. 2015, 2017).

Thus, the ideas is to offer many descriptor candidates and then let the optimization approach (Eq. (3)) find out which of these candidates are really good. Since Eq. (4) is linear, it is necessary that the offered descriptor candidates contain the potential nonlinearities. As a consequence, different descriptors, i.e. different columns of the sensing matrix, may become correlated. As a result, when e.g. more than about M = 5,000 candidate descriptors are used, the LASSO approach is getting unstable.

The mentioned problems have been recently solved by Ouyang and coworkers (Ouyang et al. 2018). These authors solve the l_0 challenge in an iterative approach called SISSO (sure independence screening and sparsifying operator). Interestingly, the mentioned correlations are not causing problems, and the number of candidate descriptors can be increased in SISSO to many billions and even trillions. From the previously mentioned "basic descriptor" $\{R_I, Z_I\}$, N^e only Z_I derived quantities were used explicitly, e.g. the ionization potentials of the atoms, the electron affinities, and information about the extension of the atomic wave functions. Then, a combination of algebraic/functional operations is recursively performed for extending the space. The operators set is defined as $+, -, \times, /, \exp, \log, |-|, \sqrt{}, ^{-1}, ^{2}, ^{3}$. Details are described in (Ouyang et al. 2018). Clearly, when different structures are considered or different charge states $\{R_I\}$, N^e related features are needed as well.

Let us demonstrate the mentioned concept of building "maps of materials" by a recent example that employed compressed sensing for the descriptor identification.

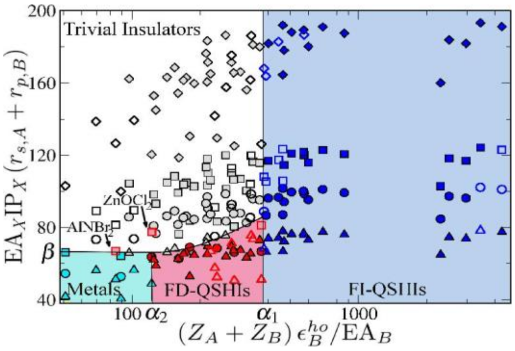


Figure 11. A map of materials classifying two-dimensional functionalized honeycomb structures as functionalization-independent Quantum Spin Hall Insulators (QSHIs), functionalization-dependent (FD) QSHIs, metals, and trivial insulators. Representation of DFT results for the training (filled) and test (unfilled symbols) set in the domain defined by the two-dimensional descriptors. Compounds functionalized with F, Cl, Br, I are represented by diamonds, squares, circles and triangles, respectively. Just two systems are classified incorrectly: $AlNBr_2$ and $ZnOCl_2$. For more details see (Acosta et al 2018). Note that the *x*-axis scale is logarithmic.

Specifically, we here summarize a recent work by Acosta and coworkers (Acosta et al. 2018) who used the approach to build a materials map for two-dimensional honeycomb structures in order to analyze and identify two-dimensional topological insulators (also called quantum spin Hall insulators, QSHIs). The characteristic symmetry-protected band inversion of QSHIs is characterized by the so-called topological invariant, Z_2. The authors calculated 220 functionalized honeycomb-lattices that are isoelectronic to functionalized graphene. Specifically, these are built of group IV, or III-V, or IV-VI, elements, and all atoms are bonded to (functionalized by) a group VII element. Besides confirming the QSHI character of well-known materials, the study revealed several other yet unreported QSHIs. In a next step, the authors used the SISSO approach. They offered 10 million candidate descriptors, and then SISSO identified the best low-dimensional descriptor. Figure 11 shows the map, created by the found two-dimensional descriptor. The scientifically important point is which of the primary (atomic) features were selected and which were not selected (see the formulas at the x- and y axis of the figure). While these atomic properties are actuating the materials property, the specific formulas have only limited scientific meaning as they largely reflect the goal of a linear equation (Eq. 4). This study yields fundamental insights in the mechanisms driving topological transitions, especially in the atypical case of topological insulators with low spin-orbit coupling. Furthermore, the map predicts several new QSHIs that were not part of the calculated materials. (Acosta et al. 2018)

6. **Outlook**

Computational materials science took off with impressive early work by Moruzzi, Janak, and Williams (Moruzzi et al. 1978) on various properties of metals and by Cohen and coworkers (Yin 1982) on the cohesion and phase transition of silicon and germanium[13]. A number of computer codes for solving the Kohn-Sham equations have been developed since them, initially involving approximations like pseudopotentials (removing the core electrons, creating smooth potentials) or introducing touching or even overlapping atom-centered spheres in which the potential was sphericalized. During the 1980’s significant advancements in the original pseudopotential approach have been made (see the work of Vanderbilt and coworkers: Garrity 2014 and references therein), and all-electron codes now treat the “full potentials” (e.g. Blaha et al. 1990, Gulans 2014, Blum 2009). The work by (Lejaeghere et al. 2016) provides a nice overview of the precision of modern electronic-structure codes for elemental bulk solids. Clearly, this kind of work is important and needs to be extended to more complicated structures and compositions, defects, surfaces, and interfaces. Work in this direction is underway, as are studies for advanced electronic-structure methods, like e.g. the *GW* approach (van Setten et al. 2015). Furthermore the field urgently needs benchmarks for the various numerical approximations and for exchange-correlations potentials in order to address also accuracy, not only numerical precision. The MSE (materials science

[13] See the chapter by M. Cohen in this handbook, in particular Fig. 4 (Cohen 2018).

and engineering) project is a first step (Zhang et al. 2018).

Computational materials science is still dominated by the third paradigm (cf. Fig. 1), but advancements in AI methods has been significant in recent years, and the fourth paradigm is playing an increasing role. Still, at present there is more hype than realism in what AI can do. Much of this relates to the domain-specific standards and requirements of materials science and engineering. Machine-learning techniques can already now help a lot when general trends are of interest and when one needs to fit and predict "the behavior of a big crowd" (see e.g. the methods used in the Kaggle competition for predicting properties of transparent conductors (Kaggle/NOMAD2018). Usually, the exciting questions in materials science and engineering are, however, different: We are typically not looking for a crowd behavior but we are searching for materials with extraordinary performance on certain functions or properties, often even a combination of several properties. There are typically just ten or a few hundred materials in the enormous number of possible options. Thus, the number of *relevant* data is rather small, and the few materials that we are searching for are statistically exceptional. How can we distinguish which data / materials are relevant and which are not? Learning about less than 0.01% relevant materials from thousands or millions of irrelevant data is obviously problematic, and standard methods, that optimize the regularized root-mean-square-error, even emphasize the importance of the irrelevant data, while surpassing the special cases. Here advancements of subgroup discovery are urgently needed.

The field is just developing the methods for the 4th paradigm. The learning curve connecting paradigms 1, 2, 3 and 4 is apparently getting steep. Thus the next paradigm may be close, even though the 4th has not been developed well. What could be the next paradigm? Considering that "the future is already here – it's just not very evenly distributed" (Gibson 1993), it may hintingly visible already today. We guess that it may be virtual reality with direct and instantaneous connection to new calculations or a merger of theory (predictions and analysis) and experiment.

What is urgently missing at present? Let us list some topics:

- Close coupling of materials property prediction with stability analysis and prediction of routes towards synthesis;
- High-throughput studies of metastable materials and the lifetime of these metastable states;
- Materials under real conditions (T, p, and reactive environment): stability and properties. This very much concerns multiscale modeling with robust, error-controlled links with knowledge of uncertainty between the various simulation methodologies – often stated in the past but still not fully realized;
- Error estimates of calculations in terms of numerical approximations (basis sets, pseudopotentials, etc.) for specific properties (structure, elastic and electronic properties, etc.);

- Computations beyond standard DFT: coupled cluster methods and calculations for solids (possibly also getting prepared for quantum computers) and quantum Monte Carlo approaches;
- Complete description of scientific results accounting for the heterogenity of data: still metadata are missing. While significant progress has been made toward transformation of computational data from the many computer codes and the development of corresponding metadata (Ghringhelli et al. 206, 2017, NOMAD Metainfo) the advantage will only fully become apparent when the same will have been achieved also for experimental data. The latter challenge is even bigger than the first. The sample material used in the experimental study corresponds to the input file of a calculation; the experimental condition (*T*, *p*, environment) and the experimental equipment to the computer code. The not fully solved challenge is the definition of the sample materials.

Clearly, the discovery of improved and novel – not just new – materials or unknown properties of known materials to meet specific scientific or industrial requirements is one of the most exciting and economically important applications of high-performance computing (HPC). The prosperity and lifestyle, including important developments in the health sector, of our society depend on materials to that enable such developments. The field is very active to meet these challenges and indeed experiences significant advances in the needed methodologies. There are exciting times ahead of us.

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