Research Update: Towards designed functionalities in oxide-based electronic materials

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One of the grand challenges facing materials-by-design approaches for complex oxide deployment in electronic devices is how to balance transformative first-principles based predictions with experimental feasibility. Here, we briefly review the functionality-driven approach (inverse design) for materials discovery, encapsulated in three modalities for materials discovery (m^3D) that integrate experimental feedback. We compare it to both traditional theoretical and high-throughput database-directed approaches aimed at advancing oxide-based materials into technologies. © *2015 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License.* [http://dx.doi.org/10.1063/1.4928289]

Ternary and multinary complex oxide materials could offer a superior platform for electronics owing to the allure of phenomena that silicon-based electronics simply cannot deliver:[1] transparent electronics leading to invisible circuitry,[2] optoelectronics based on absorbers having direct bandgaps, high-temperature electronics that can operate in oxidizing hostile environments, high-power electronics, and hyper-sensitive sensors that significantly outperform today's sensors. However, to achieve this potential, one would have to overcome numerous critical obstacles, including realization of bipolar dopeability and defeating polaron formation to the benefit of high-mobility carriers *at room temperature*. Finding oxides with an optimal feature set has been difficult: on one hand, empirical trial-and-error approaches have their limitations. On the other hand, theoretical first principles methods are reasonably accurate when applied one at a time to specific compounds. Yet, the selection problem is immense—there are many thousands of candidate oxides from which to choose.[3] This clearly calls for sophisticated materials selection strategies. The problem is challenging in that the physical properties "living" in each compound depends sensitively on (local and crystal) structure and composition. This fact is to some extent magnified in oxides and is at origin of their widespread and continued interest to the community.

To innovate in the field of complex oxides for electronics requires a change to the standard discovery operating procedure—new cooperative approaches for materials-by-design (MBD) and property prediction in complex ceramics that lead to efficient experimental realization. By this, we mean that such approaches should attempt to balance prediction with experimental feasibility, while at the same time, challenging synthetic chemists and materials scientists to advance new growth techniques. One solution includes the reformulation of the MBD challenge into a manageable workflow that relies on drawing a distinction between *properties* of materials (electronic, optical, thermodynamic, mechanical, magnetic, etc.), which describe the response to a generalized force via a generalized displacement,[4] and the very description of *what defines a material* that we dub as "'ACS." It consists of the **A**tomic identities involved (elemental species), **C**omposition (either

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integer stoichiometry in compounds or continuous variable, "subscript engineering," in alloys and ceramics), and **S**tructure (i.e., where the atoms are as pertaining to crystallographic structure, or broadly, from nanoscale to microscale and mesoscopic morphology).

Property prediction has made tremendous strides owing to (i) advanced material-dependent electronic structure methods that (unlike material agnostic model Hamiltonian theories) accept ACS as input: primarily, density functional theory (DFT) and its ACS-recognizing extensions that incorporate many-electron effects (Configuration Interaction, Dynamical Mean Field Theory, GW, and Quantum Monte Carlo methods);[5–9] and (ii) new protocols to compute complex functionalities, combining a few target properties such as dopeability,[10,11] coexistence of transparency and conductivity,[12–14] and topological insulation.[15,16] However, protocols to identify specific, including unknown, ACS for oxides that have the required technology-enabling properties and functionalities have only recently been offered. This in part relies on establishing structure-property relationships and stability conditions from structural-chemistry data and calculable material properties,[17–23] which are often established from disparate yet physically motivated models and domain knowledge. Such protocols address the question of how to go beyond calculations of *individual* materials ("given an ACS, predict its properties," i.e., the "direct approach") and simple exploitation of available materials data to intelligent exploration of those ACS that have a target property ("inverse approach"), including unknown compounds? Doing so requires approaches that both are conscientious of experimental feasibility and embrace functionality-first predictive methods.[24]

Figure 1 illustrates the recently proposed modern modalities for functionality-directed materials discovery, which we refer to as m^3D. Each poses the question of "given the property, find the material/structure (ACS)," subjected to multiple constraints. Different modalities address different circumstances of material selection and synthesis. Note that other useful approaches exist; owing to our focus on oxide materials for electronics, we elect to describe those methods we believe could spur successful experimental validation of materials predictions. Below, we describe the essential tasks associated with each modality (m), which provide solutions to this question as additional unknown features, i.e., phase stability, properties, and ACS, are examined.

We first provide an overview of the three modalities, before describing each separately. Modality 1 explores artificial materials, e.g., those realized through (generally metastable) layer-by-layer growth, whereas $m2$ and $m3$ concern mostly stable compounds grown in bulk form. Also, $m2$ focuses on finding the targeted functionality among groups of *previously made* compounds (so, the structure and composition are generally given and need not be optimized), whereas $m3$ deals with
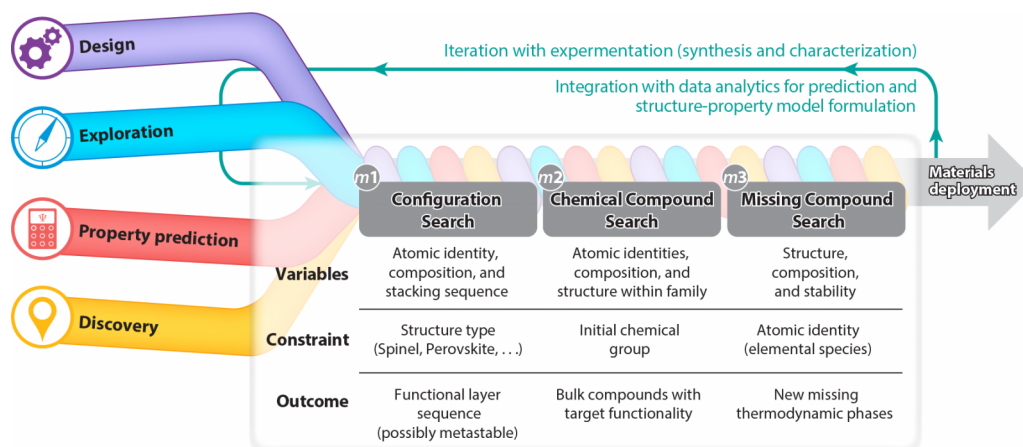

FIG. 1. The objective of rational discovery and laboratory realization of functional oxides requires a number of tasks heuristically described by the braided cords on the left. Such requirements can be implemented through three search modalities ($m1$-$m3$). Each modality pertains to a different set of growth conditions from which functional oxides may be discovered. Synchronously, data analytic methods can be applied to refine materials selection and aid knowledge discovery through the creation of suitable descriptors for predicting functionality. Uncovering the optimal oxides may take several development cycles indicated by the green loop before a new missing and functional compound is ultimately deployed.

discovery of *unreported* compounds ("missing materials") with desirable properties (so, stability and stable crystal structures need to be computed). The distinction between the various modalities stems in part from the different theoretical approaches required: $m1$ focuses on properties of an artificially grown, kinetically frozen layer sequence which need not be globally stable as long as it is locally stable, while $m2$ (sometimes) and $m3$ (always) require thermodynamic structure stability calculations. Also, $m1$ involves exploration of the almost infinite number of possible assemblies of building blocks (as pulsed-laser deposition (PLD) and molecular beam epitaxy (MBE) layer sequences) and thus cannot be addressed, even in principle by database approaches that attempt to enumerate and compute all structures. Instead, $m1$ would require *search methods* that do not require "visiting" all possible structures (such as genetic algorithms and other evolutionary approaches).

All modalities focus on functionality-driven discovery and thus require that a computable expression or *functionality metric* (FM), being a quantitative model of the target functionality, exists. Such functionality metrics are created in practice using a combination of detailed study of the underlying physics of the problem, and more recently facilitated by statistical learning tools.[25–28] Note that whereas a FM is an *a priori* quantity developed from a theoretical or empirical model from domain knowledge, the related but distinct term of "descriptors" is typically distilled *ex post facto* from mining data. Calculations such as those based on DFT then provide a theoretical value for the functionality metric—usually some composite parameters beyond total energies and standard single-particle band structures—that reflects the desired functionality. The FM may then be used "on-the-fly" to refine the material's atomic configuration having the target property (as in $m1$) or chemical makeup (as in $m2$ and $m3$). It is important to emphasize that $m2$ and $m3$ consider phase stability in parallel with the functionality, because an oxide could decompose into simpler composition-equivalent phases and make the intriguing predicting properties experimentally inaccessible. Such compounds, nonetheless, can contribute to understanding and inform functionality metric and model generation needed to both predict behavior that has not yet to be observed and connect oxide properties to device functionality.

m1—Searching the Configuration Space for Structures with Target Functionality. Many *artificial* oxides can be realized in the laboratory, via layer-by-layer growth, whereby cations can be digitally arranged in numerous sequences by interleaving various oxide building blocks such as those obtained in $m2$ or $m3$.[29,30] In oxide electronics, two-dimensional thin films are obtained through layer-by-layer growth methods. These approaches include PLD, MBE, or sputtering. The selection of suitable substrate surface orientations and layering sequences available to these approaches produces the pertinent configurational problem: Which sequence of oxide-A and oxide-B (or oxide-C, etc.) layer arrangements and layer orientations gives the property of interest? The ensuing multi-layered configuration is not necessarily the global ground state of the system (hence, *global* stability calculations need not be applied), but represents a *locally* stable structure. This modality addresses the question of how to examine the very large (in fact, astronomic) number of spatial configurations, each having, in principle, different properties and finding the configuration(s) that have a target functionality such as maximal electric polarization or optical response. This modality thus involves a search of atomic identities (A) and composition (C) under the constraint that the structure (S) is limited to locally relaxed layer-by-layer configurations of a specified lattice type, mimicking that of the building blocks and the substrate (say, perovskite units). Under such constraints, configurational search optimization techniques (such as genetic algorithms) are used to search for the atomic configurations that satisfy the targeted function by on-the-fly calculation of properties[24,31–33] or via a surrogate model such as cluster expansion.[34,35] Good optimization methods find the optimal configurations by sampling only a tiny fraction of the configurational space—typically, a couple of thousands when the size of the space is 10^{14}–10^{20}.

Examples of $m1$ from non-oxides include finding the sequence of indirect gap Si and Ge building blocks that produce a superlattice with direct and optically strong absorption, a classic[36] and, until recently, unsolved problem in semiconductor physics.[31] A functional oxide example was recently demonstrated for the case of ferroelectric heterostructures comprised of $PbTiO_3$ and $SrTiO_3$ building blocks, where the polarization does not obey a simple Vegard's law behavior as a function of mixing the end members.[37] Approximately 3×10^6 heterostructures were searched for those that yield the maximal ferroelectric polarization by calculating the polarization of only

~50 configurations to construct a robust cluster expansion.[35] The outcome was that new configurations consistent with recent design guidelines for obtaining additional contributions to ferroelectric polarizations in perovskites through improper mechanisms[38] immediately appear. Thus, $m1$ makes it possible to both identify optimal chemical sequences to be recommended for experimentation and to discover novel physical mechanisms in cases where such knowledge may not exist for property enhancement. Feedback from experiment to theory includes observations on the degree on interfacial intermixing—a fact that can be incorporated in the next step theory as a constraint on the search procedure (such as "find configurations with the target properties *and* interfacial sharpness not exceeding ε").

m2—Searching Chemical Compound Space for Target Functionality. Electronic based properties of most oxides such as photovoltaic absorption, transparent conductivity, photo-electrochemical water-splitting ability, or thermoelectricity are intimately tied to metal-oxide coordination preferences and polyhedral connectivity, which change with cation-anion ratios and structure in a material. Modality 2 explores bulk materials where structures and compositions are known, e.g., from previous studies, but the properties for a set of atomic identities are unknown. The concept here is to search through ACS-space (albeit, within a specific compositional group such as A_2BO_4, ABO_3, and ABO_2) to design a material with the set attributes that give the optimal value to the functionality metric. The steps then involve (a) identifying a calculable FM that is simpler to compute than the final functionality sought; (b) calculating the FM in the ACS space while (c) navigating in the space of compounds to specifically identify the "best of class" compounds deemed to have the highest probability for laboratory realization through some evaluation of material stability. The rationale for the above functionality-driven strategy is that it is much faster to screen the functionality assuming a given crystal structure than to investigate the existence of a functionality and the ground state crystal structures simultaneously.

For example, a quantum-mechanical generalization of the Shockley-Queisser criteria, called the "Spectroscopic-Limited Maximum Efficiency" (SLME) functionality metric, was created in step (a). It was tested on several hundred ternary materials using $m2$ to identify candidate photovoltaic absorbers,[18] followed by exploration of a select chemical group of semiconductors in search of maximal SLME. Note that $m2$ is distinct from making *all* possible chemical substitutions and optimizing over that set as in high-throughput searches. In $m2$, there is no need to compute many properties of a broad range of compounds, albeit that information could be useful to data-driven discovery approaches. In $m2$, one thus uses the strategy of narrowing down a large number of an *a priori* set of compounds by using the FM as a preliminary initial filter. This filter does not address all aspects of the functionality (such as material performance, durability, and economic factors), but focuses on a number of "first-filter," *critical attributes* needed for a particular functionality (such as FM = dopeability for making junctions or FM = strong SLME absorption for solar cells, depending on the need). Following such initial screening, one is left with a handful of candidate materials. The second step involves application of more accurate high-level methods (such as beyond-DFT methods). Now the calculation of a broader range of material functionality that transcends the simpler principles involved in the FM may occur; efforts to include such features as durability, reliability, and economics are already appearing.[17] It is important to emphasize that this difficult detailed screening occurring in the second step is required only for a much smaller group of "best candidates," rather than for all materials, thus providing a significant discovery acceleration factor. In the last step, the "best of class" materials are handed over to experimental validation, which very often leads to a refined FM, to be used in the next iteration. This protocol defines an iterative loop, whereby theory and experiment work hand in hand.

Examples of $m2$ applications include both successful confirmation of known absorbers and identification of previously unappreciated new candidate compounds that have been proposed and experimentally realized.[39] Another example is the use of FM = dopeability to search the space of ABX "half Heusler" compounds looking for transparent conductors. This resulted in the theoretical identification and eventual laboratory synthesis of the new, previously never before made, high hole mobility (2700 cm^2/V s at room temperature) TaIrGe transparent conductor.[14]

m3—Exploring Missing Compounds for Target Functionality. This modality explores "*missing compounds,*" that is, members of known chemical groups, that were never made. For example,

in A_2BX_4 spinels or ABX half-Heusler like compounds, one can readily identify hundreds of cases—specific selection of A, B, and X elements—that are undocumented. Here, $m3$ searches theoretically the thermodynamic stability and stable structure thus establishing which of the missing compounds are inherently unstable (thus, understandably missing) and which is predicted to be stable (thus, just overlooked). In $m3$, the crystal structure is initially unknown and phase stabilities of numerous structures with variable composition require exploration under the constraint of atomic identities, such as spinel and non-spinel (variable 2:1:4 stoichiometry and competing phases) structures.[40] Here, a thermodynamic-based search for missing but stable (and low-energy metastable) compounds is essential to reduce the large chemical phase space into a digestible set of materials for the synthetic chemist or thin film grower. (We include in the identified list also low energy metastable predicted structures because in some cases, oxides with low formation energies or those deemed to be metastable can indeed be realized in the laboratory, but may require non-equilibrium, kinetic-limited growth approaches.) The key tasks include (a) screening of candidate structure types,[41,42] (b) unconstrained composition and structure prediction,[43–45] and (c) evaluation of competing phase stability with respect to decomposition to simpler phases under the constraint that the desired atomic identities are fixed; iteration of $m3$ for different element species enables exploration of broad chemical space. An outcome of $m3$ is accelerated discovery of materials in the laboratory. Note that in the ideal case where a handful of compounds are recommended for synthesis and growth following identification of phase stability, the required processing conditions may be unknown. Realizing any new materials in the laboratory, therefore, can become a challenge to perseverance.

As an example of $m3$, Gautier *et al.* recently reported the first-principles evaluated phase stability of more than 400 ternary ABX compounds which fulfill the 18-electron rule and predicted 54 of the previously unidentified compounds to be stable; shortly afterwards, 15 new materials were obtained, including new topological insulators.[42] Implicitly, the properties are also unknown in $m3$, and a parallel property search can occur, or alternatively, once a stable new material is identified, strategies in $m1$ or $m2$ may be applied. Feedback from experiment to theory to be used in the next iteration includes focusing attention on previously overlooked daughter structure types. A recent example of functionality search includes the discovery of a previously missing half Heusler compound TaIrGe that was predicted to be stable[14] and a wide gap compound (despite the strongly held intuition that high-Z compounds are generally narrow gap or metals) while also being conductive. This new type of transparent conductor was recently synthesized and confirmed experimentally to have high hole mobility.[14]

Integrating the described modalities with feedback from experimental synthesis. Often referred to as "closing-the-loop," the process is schematically illustrated in Figure 1. An interesting question arises as to the necessity and number of iterations required and to what extent the number of cycles may be predicted. Ideally, a single cycle should give convergence to the optimal oxide. This is possible if our materials knowledge relating structure and chemistry to properties and functionalities is formally uniquely quantifiable and approximations in electronic structure methods are fully controlled. Advances in laboratory discovery of missing materials by *in situ* studies and combinatorial synthesis methods also make it possible to navigate broad areas of phase space efficiently.[46] However, our knowledge is not complete, and this is reflected in the limited size of the data from which most of our models are based. Iterative progress in the discovery loops is therefore far more common before immediate convergence of the best oxide occurs. One may then want to assess and estimate the extent to which improvement can be achieved by an additional iteration through the workflow cycle. This return on investment can be informed through application of adaptive learning strategies[47] and should be considered in any materials discovery program.

Finally, we contrast the functionality-driven m^3D platform to the data-driven discovery paradigms.[48] The latter focuses on high-throughput computations of many properties for many materials being cataloged in databases[22,49–51] or combinatorial synthesis for MBD.[52,53] The high-throughput and combinatorial synthesis approaches have in part been energized by national materials discovery initiatives around the globe and have encouraged data organization and curation, which is invaluable to achieving data-mined materials discoveries.[54] These efforts generate enormous amounts of

data quickly. Materials selection is made *ex post facto* and based on sorting/ranking a pre-defined figure-of-merit and occur within a limited ACS space.[55] Thus, high-throughput approaches make it possible to identify previously known but unidentified functional materials from a database of known compounds. New understanding may then be gleaned from the down-selected compound and become useful for practitioners of first-principles materials-by-design methods that fall outside the mentioned modalities to predict new functional materials.[56] The functionality-directed approach m^3D is distinct from the high-throughput approach in that if it focuses directly on the subset of materials that has the required target property rather than attempting all combinations. Computationally speaking, the m^3D approach then focuses on optimization theory and search methods, whereas the high-throughput focuses on fast direct computations and databases.

Given availability of such data, more researchers are applying big data science analytics to address the materials chemistry and structure-property challenges.[57] We contend, however, that it remains to be determined whether the real challenges in oxide-based electronics are of the sort suitable for only data-driven scientific discovery approaches. For instance, the subject of m1—different configurations of artificially grown structures—cannot be captured even in principle by any database because of the limitless configuration space. Second, the data sets, features, and prior knowledge in condensed matter systems are significantly smaller than those available to biology and health, communication networks, and finance, where big data science dominates. One therefore needs to consider nuances in the informatics methods for materials science data[58–60] and whether the "correct" questions are asked about the data.[61] Black-box application of machine-learning algorithms to small data sets without careful data cleaning and feature selection could also lead to further complexity and minimal understanding of the underlying structure. Note that in some areas of materials science, "big" data sets exist, for example, from high-resolution imaging techniques or synchrotron based scattering and spectroscopy measurements.[62,63]

Perhaps the oxide electronics challenges are rather better described as "small" and "deep" functionality-targeted data problems, where the necessary prior knowledge required to make progress still remains to be uncovered from hypothesis- and exploration-driven scientific discovery methodologies and personal communications between theorists and experimentalists. The inability of informatics-based data analyses alone to create *robust*, i.e., optimal model design and error estimation, mechanistic knowledge from *small* data sets, typical of what we can expect in oxides, has been borne out in biology and genetics;[64–66] similar pitfalls should be avoidable in materials science. The field should establish to what extent statistical inference and learning methods can benefit materials discovery by including authentic structure-property relationships guided by physical models. This is an important open question that will be addressed in the next few years as the number and the large size of materials structure, property, and calculated quantity databases available for scientific query continue to grow.[67]

In summary, we described a functionality-first view for oxide electronics materials discovery through three computational materials science modalities that can synchronously dovetail with experimentation. Once theory has narrowed the range of oxide materials that are predicted to have the target properties, *interactive* rather than sequential cycles of synthesis, characterization, and theory validation should ensue. If these approaches are applied and fostered while searching, designing, or predicting the properties of oxides, we are hopeful that the promised impact of oxides for future electronics is not far off. Many more predictions are likely to be realized and found in new electronic devices.

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