Toward self-organizing low-dimensional organic–inorganic hybrid perovskites: Machine learning-driven co-navigation of chemical and compositional spaces

Jonghee Yang, Sergei V. Kalinin, Ekin D. Cubuk, Maxim Ziatdinov, and Mahshid Ahmadi*

Low-dimensional hybrid perovskites combine the richness of physical functionalities of inorganic materials and complexity and stimulus responsiveness of organic molecules in a single bulk dynamic material. The unique aspect of these materials is the thermodynamic (meta) stability, allowing for self-organized formation of complex large-period structures. Combined with the ease of fabrication, these materials not only have extensively demonstrated state-of-the-art high-performance optoelectronics, but also offer the pathway toward versatile applications, including sensors, electronic, and neuromorphic devices as well as their cost-effective mass production. However, discovery and optimization of this material require joint optimization of the composition of the inorganic components and selection of the molecular moieties, to harness the phase formation and self-assembly processes on the material level, and extend it to micro- and macroscale functional devices. Here, we discuss the potential of machine learning-driven automated experiments to accelerate the discovery of these materials, optimize the processing pathways, and transition from the lab-level to the product-level manufacturing.

Introduction

Within a decade since discovery,[1] the rapid development of hybrid perovskites (HPs) achieved an outstanding optoelectronic performance (power conversion efficiency approaching 26% and light-emitting efficiency >20% for photovoltaics and light-emitting diodes, respectively), surpassing silicon-based devices.[2–4] This incredibly fast rate of progress of HPs for semiconductor applications can be attributed to the combination of excellent optoelectronic functionalities and scalability of solution-processing. The broad range of functional responses in these materials suggests potential for other high-performance and mass-producible device applications, such as x-ray and chemical sensors, neuromorphic, and ferroelectric devices.[5–8] However, practical realization of HP as a universal functional material platform that constrains its realization to date is the chemical lability and low phase stability of the inorganic lattice.

Two-dimensional (2D) HPs offer substantially improved robustness compared to three-dimensional (3D) HP systems, increasing their potential for device applications. In these materials, the HP lattices are spatially confined in 2D space by organic cations binding onto the surface. This structure not only protects the functional inorganic lattice from external chemical and physical stresses, but also results in strong quantum and dielectric confinement effects. The latter, in turn, leads to intriguing physical characteristics that are not observed in 3D counterparts, including tunable bandgap, strong exciton binding energy, anisotropy in optoelectronic properties, strong Rashba effect, and so on.[9,10] These functionalities, in turn, can readily be tuned by changing the composition outlay, including

Jonghee Yang, Department of Materials Science and Engineering, Institute for Advanced Materials and Manufacturing, The University of Tennessee, Knoxville, USA; jyang70@utk.edu
Sergei V. Kalinin, Department of Materials Science and Engineering, Institute for Advanced Materials and Manufacturing, The University of Tennessee, Knoxville, USA; sergei2@utk.edu
Ekin D. Cubuk, Brain Team, Google Research, Mountain View, USA; cubuk@google.com
Maxim Ziatdinov, The Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, USA; Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, USA; ziatdinovma@ornl.gov
Mahshid Ahmadi, Department of Materials Science and Engineering, Institute for Advanced Materials and Manufacturing, The University of Tennessee, Knoxville, USA; mahmadi3@utk.edu
*Corresponding author
doi:10.1557/s43577-023-00490-y

both the composition of inorganic component and the nature of organic moieties separating them.[11] Moreover, as the 2D HP shares the inorganic lattice structure with the 3D counterpart, it is convenient to mix them with 3D HPs to form intergrowth phases. These so-called as quasi-2D HPs demonstrated excellent functionalities and performances for photovoltaics and light-emitting technologies.[12] As a result, the promising potential and functionalities of 2D and quasi-2D HPs make these materials an attractive material platform for versatile applications.

Structurally, low-dimensional HPs are composed of several key components: (1) large organic cation (L), called a spacer, that spatially separates and maintains the layer-structured HP sheets within quantum-confined dimensionalities. Metal cations (B), halide anions (X), and small monovalent cations construct the HP sheets based on metal halide octahedra $[BX_6]^{4-}$ building blocks. Depending on the charge of the spacer cations, 2D HPs have generally a chemical formula of $L_2A_{n-1}B_nX_{3n+1}$ or $LA_{n-1}B_nX_{3n+1}$.[10,13,14] Here, L is a monovalent or divalent molecular cation—two monovalent cation heads in a molecule, respectively, forming a discrete inorganic 2D monolayer (Ruddlesden–Popper structure) or interconnected 2D bilayer sheets (Dion–Jacobson structure). The spacer cation has a positive-charged functional head (typically, ammonium functional group) in its molecular structure, binding onto the surface of 2D sheets by filling the A cation site. The *n* indicates the layer thickness of the HP sheets (i.e., *n* times unit cell length of $[BX_6]^{4-}$ octahedron comprising the sheet) primarily determining the electronic structure—for example, bandgap and exciton binding energy—which can be tuned by precursor composition.[9]

Typical inorganic 2D materials are grown by vapor synthesis in gas phase, resulting in highly ordered and organized structures via epitaxial growth.[15–17] In contrast, 2D HPs are mainly synthesized via a solution-based process where the nucleation process is difficult to control. As a result, rather than a phase-pure system, multiple phases emerge, subsequently rendering somewhat disordered and imperfectly organized 2D assemblies on large scales.[10,14,18] Notwithstanding their fascinating physical properties, the aforementioned structural variability, and the presence of multiple layered systems with similar compositions, hamper practical applications and developments of this rising 2D materials system.

Depending on their molecular structures and the consequent chemical properties, spacer cations can induce multitude effects on the physical and structural properties of 2D HPs. Increasing the molecular length of these spacers separates the interval between neighboring 2D sheets farther, suppressing the electronic coupling over the 2D HP stacks.[19] Steric hindrance of the spacer molecule can restrict the possible binding site onto the 2D HP surface, selectively allowing them to bind at specific sites such as lattice edge or kink points. The shape and structure of the spacer cation (aliphatic/aromatic, specific chirality, etc.) not only determine the stacking features of the resulting 2D HPs, but also influence the in-plane atomic alignments, including lattice strain in 2D sheets, directly changing the physical properties.[10,20] Also, add-ons with other functional groups at the spacer body impose additional interaction between the spacer-HP lattice,[20,21] which not only further influence to the stacking behaviors between the 2D sheets, but also uncover the hidden properties of the HP system. Although it is not feasible to bind the larger spacer cations onto the 2D sheet surfaces, they can stabilize the edge sites and thereby contribute to dimension-regulated block formations. Hence, a subtle change in physicochemical properties in spacer cation not only regulates the shapes, structures, and geometries of the 2D HPs, but also induces substantial changes in their optoelectronic properties and the resulting functionalities.

The judicious design, selection, and/or combination of appropriate spacer cation, as well as elemental composition of the HPs can realize a self-organized 2D HP platform showing intriguing functionality, which can expand the versatility toward various applications. As an example, it is envisaged that systematic deployment of molecular interaction/reaction along the spacer cation in HP synthesis realize rigid molecular assemblies in a reaction system, serving as a template for the 2D HP growths (**Figure 1**). This subsequently allows synthesizing and optimizing unique "self-organized" 2D HP superlattice systems, opening a new avenue to maximize its functionality.[22] However, there are massive amounts of organic spacer with different functional groups, sizes, shapes, electronic structures, etc. In addition, various synthetic parameters—concentration, composition ratio, temperature, solvent, growth time, etc.—should be controlled to find an optimal condition for synthesis of each material system. Thus, the degree of freedom that we have to control with an appropriate combination of the spacer candidates and the synthetic parameters in this regard are infinite, whereas the evaluation of the resulting product with appropriate characterizations becomes extremely complex and thereby beyond human power.

Automated high-throughput synthesis is an ideal approach to explore this giant pool of molecular and synthetic spaces. The robot-based automated synthesis platform, coupled with a high-throughput, fast absorption, and emission spectroscopy, enables precise and quick assessments of the synthetic parameters while excluding human-oriented errors. As a result, dynamic changes of 2D phases by stoichiometric/compositional tuning, temporal stabilities, and the function of solvent on the crystallization kinetics—dependent on the physicochemical properties of solvents (i.e., dielectric constant, Lewis acidity/basicity, etc.)—can be precisely and readily assessed. Also, by simply changing the spacer molecules in precursors, this strategy allows comprehensive explorations of the optoelectronic properties and functionalities, effectively revealing the key effect and/or mechanism of the molecules on the formation, packing, and electronic structures of 2D HP

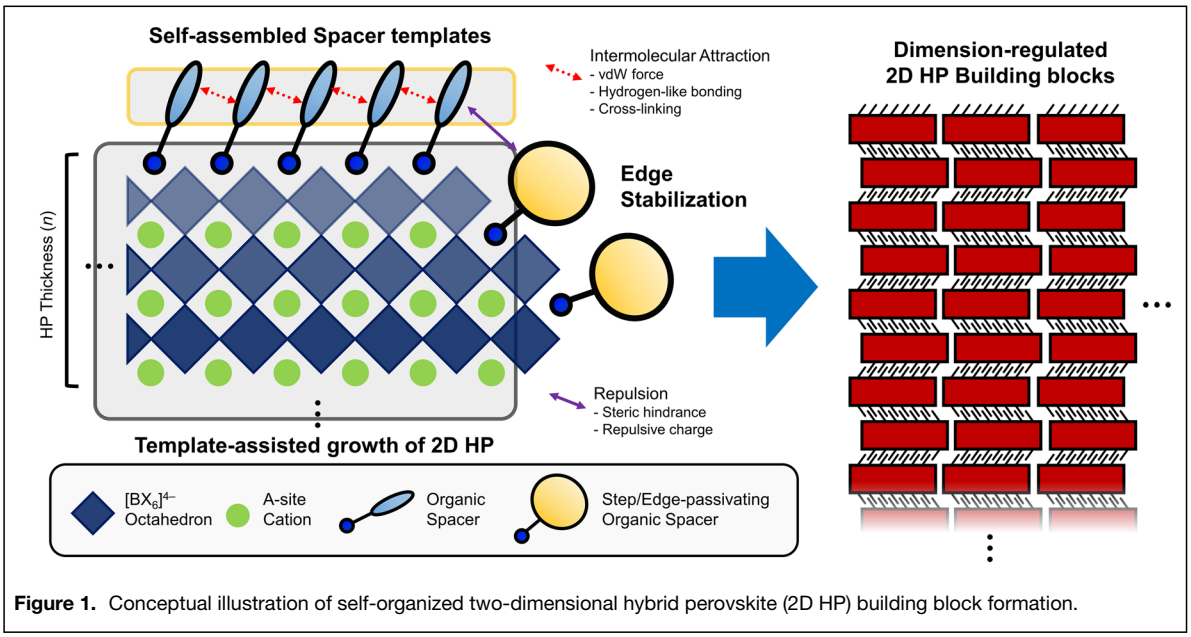

Figure 1. Conceptual illustration of self-organized two-dimensional hybrid perovskite (2D HP) building block formation.

phases and consequently reaching to the solution that we pursue.[23–25]

The critical component of materials discovery and optimization are advanced characterization techniques that can be employed in a high-throughput manner, allowing us more comprehensive insight into the quasi-2D HP system beyond optical characterizations. For example, high-throughput grazing incidence wide-angle x-ray scattering (GIWAXS) characterization utilizing a synchrotron facility opens a new avenue to explore the structural evolutions in quasi-2D HPs, including lattice orientations, interlayer spacings, phase (im)purities, and distributions;[26] these are complementary with the optical spectroscopies. Thus, the combination of these powerful characterizations provides a complete understanding of a vast compositional space of HPs, suggesting a practical way to realize the desired functionalities and/or new materials discovery. Here, incorporation of machine learning (ML) techniques is a powerful strategy to accelerate the process by guiding key signatures appearing in the massive amounts of data collected in the high-throughput approach.[23–25] As a result, the workflow coupled with high-throughput synthesis/evaluation and ML (**Figure 2**) enables effective and accurate co-navigation of the compositional, structural, chemical space, thereby realizing demonstration of novel low-dimensional functional materials.

In this article, we describe how ML can navigate the rapid exploration of low-dimensional HP materials. We describe principles and progress of ML in both theoretical calculations and experiments on the materials, and propose a conceptual strategy to realize a novel versatile self-organized 2D HP system in the aid of ML, which can be directly implemented to various device applications.

ML in theoretical calculations

Novel material discovery requires the ability to predict the relative stability of different compositions in a chemical family and the relative formation energy of different structural phases of a composition. An attractive promise of computational methods such as density functional theory (DFT) has been to allow such predictions using computer simulations.[27] This can be a challenging task, as the difference between different structural phases can be on the order of several mega-electronvolts per atom.[28] Recent advances in modeling the exchange-correlation functional, such as SCAN,[29] have shown promising results for being able to rank the energies of different phases accurately.[30]

Although much needed progress has been made on improving first-principles calculations, a complementary effort has been ongoing for aiding or bypassing the DFT simulations using ML tools. The basic idea is to collect a set of inputs and outputs from DFT simulations to train a neural network or some other ML model. Broadly, ML models that can predict the formation energy of a material can be classified into two categories: compositional models,[31,32] which predict the ground-state formation energy of a composition only based on the elements and their fractions that make up the material, and structural models,[33,34] which predict the energy of a crystal based on its composition as well as its structure (positions and lattice vectors). State-of-the-art ML models can predict the formation energy of materials from Materials Project (MP) with 28 meV/atom[33,34] and 60 meV/atom[32,35] mean-absolute error using structural and compositional models, respectively. However, it has been challenging to utilize these low-error models to discover novel stable materials. Bartel et al. have found that compositional models can have difficulty predicting the relative stability of competing compositions in a

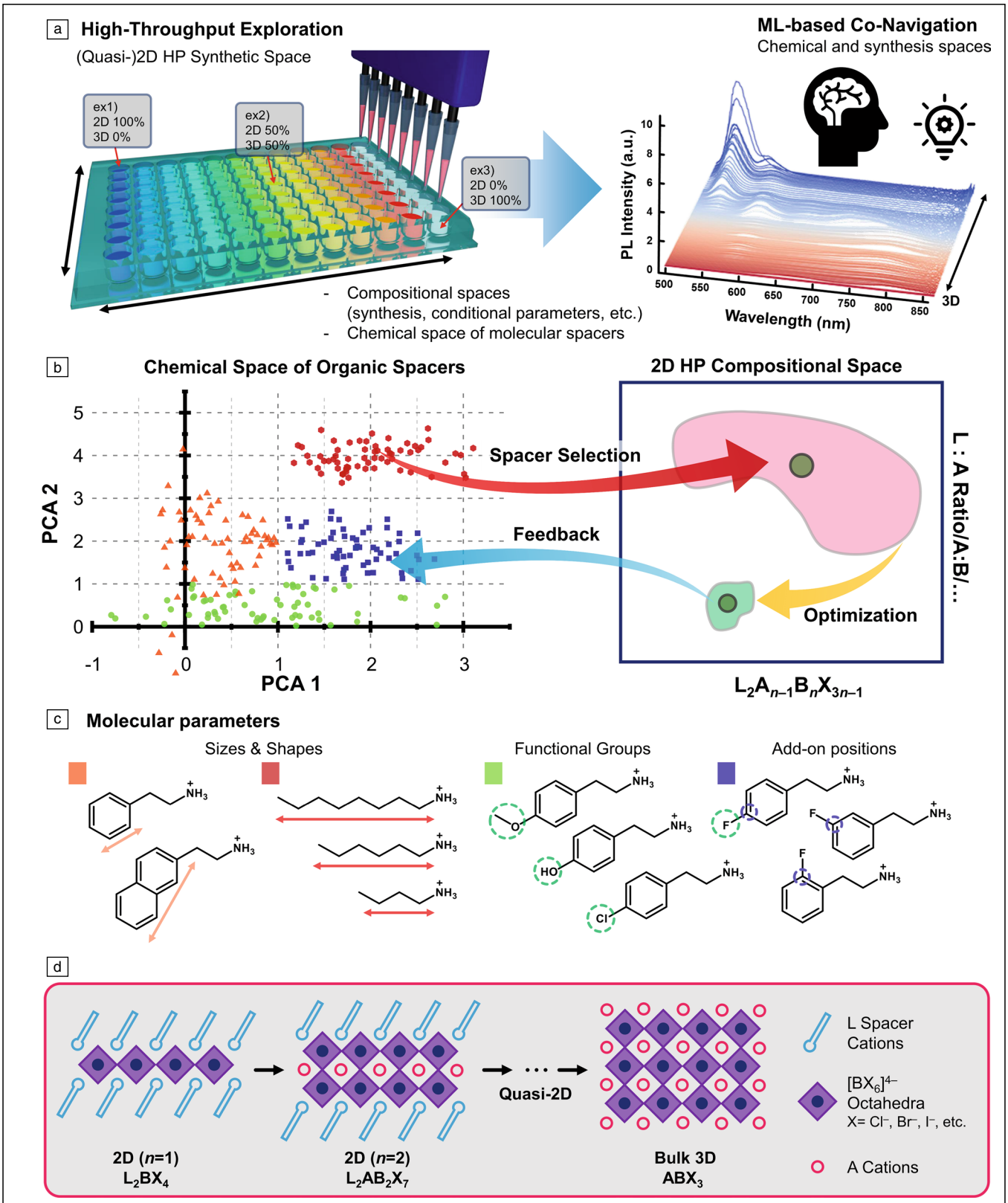

Figure 2. (a) A representative workflow for (quasi-) two-dimensional hybrid perovskite (2D HP) explorations, including high-throughput automated synthesis (here, a pipetting robot platform is presented), rapid material characterizations, and machine learning (ML) techniques. (b) Illustration of co-navigation in chemical and compositional spaces for 2D HP system. (c) Molecular parameters categorized by physicochemical properties of the spacer candidates. (d) Schematics illustrating basic structure of (quasi-) 2D and three-dimensional (3D) HP lattices.

chemical family.[35] They also reported that although structural models can have sufficient predictive power for distinguishing the relative stability of different compositions, they require knowledge of the ground-state structure beforehand, which can defeat the purpose of discovery.

The task of going from the composition of a material to its ground-state structure is known as structure prediction. ML models have also been utilized for structure prediction. A common pathway is to train a force field using ML. The first general-purpose ML force fields have been proposed by Behler and Parrinello.[36] Follow-up work has reported that Behler–Parrinello neural networks can learn to represent different structural phases of a composition[37] and model the thermodynamic stability and dynamics of different compositions.[38] More recent work[39] has evaluated the promise of ML force fields for random structure search.[40] Another application of ML to structure prediction has been via more advanced deep learning methods, such as learned optimizers[41] and generative models.[42,43] As a proof of concept, generative models have been used to sample already known HP structures, as evaluated by their reconstruction error.[42,43]

In addition to generating and predicting the stability of a new HP, ML can also be used to predict electronic properties such as the bandgap. Although the formation energy prediction error has been improving with developments in deep learning, electronic property prediction has proven to be difficult. For example, MEGNet reported a mean-absolute error of 0.33 eV for predicting the DFT-based bandgaps of nonmetallic materials in MP.[34] Given the further problems with DFT's inaccuracy for predicting bandgaps,[44] it will be unlikely that an ML model trained on DFT-based labels of bandgaps will be able to make useful predictions in the near future. However, a promising direction could be to train the ML model on experimental labels of bandgap measurements, done on a predefined family of HPs.

Designing 2D layered perovskites presents unique challenges for DFT calculations. The ability to choose the composition of the inorganic components and selection of the organic molecules allow for unprecedented flexibility. However, navigating through this combinatorial explosion only using simulations is prohibitive, given the computational expense of DFT. For this reason, we believe that ML can play a transformative role for this design problem. Graph neural networks have already been shown to model organic molecules effectively.[45] Graph neural networks have also shown promise for modeling more complex systems involving both inorganic surface slabs and organic molecules in the context of catalysis.[46] Further developments in graph neural networks as well as meta-learning hold the promise to make 2D layered perovskite design more tractable.

From end members to solid solutions

The unique aspect of the quasi-2D HPs is the potential for formation of larger numbers of low-dimensional phases with various compositions as solid solutions between the pure 2D and 3D HP end members. This gives rise for complex multicomponent phase diagrams containing single- and multicomponent regions and morphotropic phase boundaries between crystallographically incompatible phases, which subsequently renders drastic functional variabilities. Although the properties of the end members can often be ascertained using DFT calculations, the variety of possible solid solutions renders theoretical investigations intractable. Similarly, the properties of experimentally synthesized materials differ from theoretical predictions, both due to the systematic errors of theoretical models and also the presence of the exogenous (to theory) factors such as point and extended defect populations, nonstoichiometry, etc. Many of these factors are sensitively affected by the synthesis conditions including the precursor solutions, solvent and antisolvent, and annealing conditions.

These considerations necessitate the use of high-throughput experimental methods based on automated platforms for HP explorations, allowing precise control of compositional, dimensional, and synthetic parameters and thereby drawing a complete chemical landscape in the HP system.[47–49] However, a simple increase of throughput is insufficient for materials optimization and especially discovery. For example, assuming that the materials behavior can change with 1% composition variation, exploring a four-dimensional (4D) phase diagram using grid search requires 10^8 samples, equally intractable for manual or automated synthesis. This, in turn, requires developing ML methods capable of navigating the multidimensional composition and synthesis (e.g., solvent composition or annealing trajectories/gas regime) spaces.

The by now well-established approach for such optimization problems is Bayesian optimization (BO) based on Gaussian process (GP) regression. Generally, GP refers to an approach for reconstruction of function of interest and its uncertainty from a relatively small number of measurements. BO is an active learning approach in the sense that a surrogate GP model interacts with the data generation process, and it aims to reconstruct the physical property (or properties) of interest in the minimal number of steps (exploration), or discover the regions of the parameter space where this property is maximized (exploitation). The limitation of the classical GP methods is their data-driven nature, where the kernel function learnt from experimental observations represents the correlations across the parameter space. As such, simple GP methods have limited applicability in high-dimensional spaces such as annealing trajectories.

It is conventionally accepted that discovery and optimization of materials functionalities will be enhanced if the underpinning physical mechanisms are known, allowing for straightforward answers to the interventional and counterfactual questions. However, this is generally *not* the case, and deciphering the quantitative mechanisms is a herculean effort. The pathway out from this conundrum is a physics-based structured GP (*s*GP) and hypothesis learning as shown

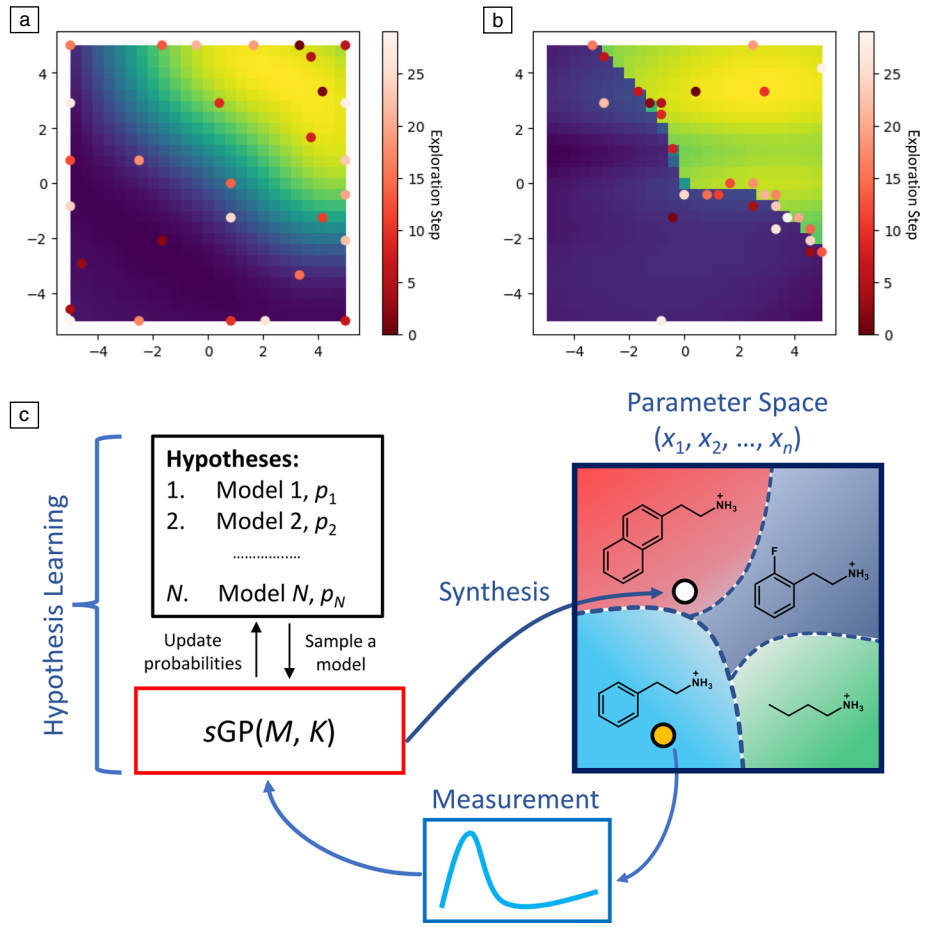

Figure 3. Comparison between vanilla Gaussian process (GP) (a) and structured GP (b) for active learning of magnetization behavior in the 2D Ising model.[50] Unlike vanilla GP, the structured GP with only a crude model of system behavior is able to localize a phase boundary within a small number of "measurements" as well as provide a reasonable reconstruction of the phase diagram. (c) Schematic of hypothesis learning, where at each step, we sample a probabilistic model *M* of possible system behavior according to a predefined policy (e.g., epsilon-greedy) and wrap it into a structured GP with kernel *K*. The GP posterior predictive distribution is used to derive the next measurement point and model probabilities are updated based on the difference in total values of predictive uncertainty over the unmeasured parameter space between the current and the previous step.

in **Figure 3**.[50] In the structured GP, prior mean function of the GP is no longer constant, but represents the possible physical behavior of the system, for example, the concentration dependence of bandgap, extrema of stability in the vicinity of the morphotropic boundaries, etc. The model is defined in the probabilistic sense, with the prior knowledge about the physics of the system summarized in associated prior distributions on the parameters. Hence, during the active learning, the physical model is learned jointly with the GP kernel toward the desired outcome. This approach can be naturally extended to the scenarios where mutually exclusive hypotheses that combine the physical mechanistic frameworks and weakly known nature of corresponding parameters and mechanisms are available, the approach referred to as hypothesis learning.[51]

Similarly, BO methods can be extended toward the continuous processes (e.g., optimization of annealing trajectories). Here, a number of approaches based on the encoding of possible trajectories in the latent space of the generative model or deep kernel learning are possible. Finally, recently BO networks were proposed for more complex spaces including chemical organics with various functional groups, and associated chemical reactions.[52]

Engineering the molecules toward a self-organized 2D HP system

Organic molecules allow multiple venues for engendering not only intermolecular electrostatic interactions such as π–π stacking or hydrogen bonding, but also the dynamic responses to external stimuli, including light-induced isomerization, cross-linking, pH, and ionic strength-induced conformational changes. However, the charge-transport behaviors in such organic molecular semiconductor systems, based on the long-range ordering of discrete molecular arrangement, are generally limited. In comparison, inorganic materials exhibit a rich gamut of the electronic, optical, and magnetic phenomena highly owing to the proximity in atomic arrangements over a continuous lattice, stimulating decades of effort to harness them in multifunctional low-cost devices for nonvolatile electronics, environmental monitoring, and medical sensing. However, accessing and tuning these functionalities has remained limited, with the field effect being the primary control knob.

Over the last decade, the field of optoelectronics was revolutionized with the advent of HPs that combine excellent photo-physical and electronic properties with low-cost solution-based fabrication.[53] The key aspect of the 2D HPs is that not only tuning the HP building block tunes the functionality, but also the dynamic behavior of the component in the organic layer can impose similar effects on continuous layers. An additional degree of control is the layer spacing that affects quantum confinement. Thus, engineering the spacer cation is the indispensable step to control the dimensionality and structure of the low-dimensional HP lattice, thereby realizing the

phase-pure self-organized system with on-demand lattice orientations. As an example, by judiciously deploying the molecular interactions appropriately, a dimensionally confined free space with a periodic arrangement of organic spacer cations could be established, in which the inorganic 2D or quasi-2D HP lattice can grow along the spacer template. This subsequently expands and maximizes the coherence in functionalities along the entire system, making it fully utilized in device applications.

Of course, it is challenging to find such ideal spacer candidates which simultaneously serve appropriate spatial periodicity in binding head position compatible with the HP lattice unit cell and the rigid molecular arrangement that are not decomposed readily during synthetic progress. The high-throughput robotic platform can play a crucial role swiftly confirming each molecular spacer whether it has feasibility on these aspects or not among the massive gamut of molecular candidates. Mapping the synthetic/chemical/compositional spaces on specific organic molecules, and a series of workflows established in this regard could be an invaluable reference to go forward with another molecular system. Systematic theoretical calculation based on model structures of atomic/molecular arrangements further suggest clues on the key design principles. This not only provides us with the general parameters such as optimized formation energy, chemical potential, etc., but also gives us detailed insights into molecular/atomic geometries including preferential orientation of spacer cation, signatures for hydrogen bonding, spacer-induced lattice strain, and spacer–spacer interaction associated with layer stacking behaviors over the low-dimensional HP ensembles.

From materials to devices

In over a decade since becoming mainstream, 3D HPs have successfully demonstrated their indispensable functionality ideal to rich gamut of optoelectronic device applications, and now they are also showing potential compatibilities with flexible electronics, neuromorphic devices, and sensors.[4,5,7,8,10,54] Notwithstanding their superiority, the notorious phase stability of 3D HPs has constrained the realization, which is the most crucial drawbacks in this system. For 2D and quasi-2D cases, the surface covering organic molecules protects the HP lattice from external stress, thereby bestowing notably improved stability on these functional systems. This subsequently opens an avenue to utilize multifunctional 2D and quasi-2D HPs as a universal building block toward solution-processible device applications, including electronics, sensors, and so on. The self-organized low-dimensional HP superlattices, where the major challenges are associated with these functional systems, can establish long-range coherence in fundamental properties of the materials—such as charge-transport behaviors or electronic structures—over the entire system,[22] thereby maximizing the optoelectronic functionalities.[55] The key principles on the mechanism and optimal conditions for synthesis, which are already proposed in the high-throughput material exploration stage, make the discovered materials directly employed to versatile device application, thereby manifesting its functionality by performance evaluations. Such a workflow allows us intuitive realization of intriguing characteristics and functionalities observed from the materials level directly to device applications.

Outlook

Despite tremendous advances and developments in the HP system, still only a handful of compositions have been majorly utilized. Automated high-throughput exploration in 3D HP opens a new avenue to comprehensively investigate this functional material platform by systematically changing composition and its ratio with small increments. Additionally, incorporation of compatible ML methods allows us to efficiently analyze massive data sets and derive proper solutions, a sequence of which is essentially challenging by relying on human power. As a result, a series of these workflows are now accelerating discovery of 3D HPs, including optimal conditions with desired functionality. Such a joint approach becomes much more powerful and indispensable for exploration of low-dimensional 2D HP systems where the vast number of determinants is associated with materials synthesis—particularly the chemical space associated with the physicochemical properties of spacer cations. High-throughput theoretical predictions and automated synthesis can realize synergistic co-navigation of both chemical and compositional spaces. Effectively, this can give us a genuine solution for realization of the phase-pure self-organized 2D HP system, which is one of the conundrums in this functional and versatile material platform. This, in turn, opens an avenue to be employed in a wide range of high-performance next-generation optoelectronic applications including light and chemical sensors, neuromorphic devices, memristors, or other electronics that can be employed as integrated circuits.

Acknowledgments

J.Y. and M.A. acknowledge support from the National Science Foundation (NSF), Award Number No. 2043205 and Alfred P. Sloan Foundation, Award No. FG-2022-18275. All authors acknowledge support from the Center for Nanophase Materials Sciences (CNMS) user facility, which is a US Department of Energy Office of Science User Facility, Project No. CNMS2021-B-00922.

Data availability

The data sets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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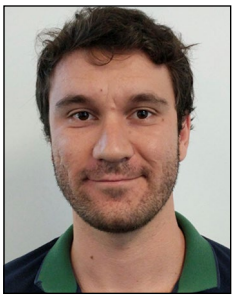
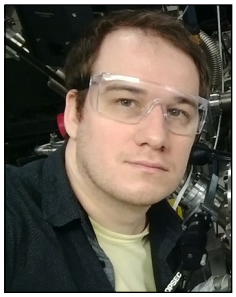
Jonghee Yang is a postdoctoral research associate in the Institute for Advanced Materials and Manufacturing, Department of Materials Science and Engineering, at The University of Tennessee, Knoxville. He received his PhD degree in chemistry from Hanyang University, South Korea. His research focuses on the high-throughput synthesis of perovskite nanomaterials and fundamental explorations on the functional organic–inorganic semiconductors, including perovskite materials using spatially resolved imaging techniques coupled with machine learning methods. Yang can be reached by email at jyang70@utk.edu.

Sergei V. Kalinin is a professor at The University of Tennessee, Knoxville, following 20 years at Oak Ridge National Laboratory. His research focuses on the applications of machine learning in scanning transmission electron microscopy and scanning probes for physics discovery and atomic fabrication, as well as mesoscopic studies of electrochemical and ferroelectric phenomena. He is a Fellow of the Materials Research Society, American Physical Society, Microscopy Society of America, Institute of Physics, IEEE, Foresight Institute, and the American Vacuum Society; a recipient of the Blavatnik Award for Physical Sciences (2018), Burton Medal of the Microscopy Society of America (2010), four R&D100 Awards, and a number of other distinctions. Kalinin can be reached by email at sergei2@utk.edu.



Ekin D. Cubuk is a staff research scientist at Google Brain, where he works on deep learning and its applications to solid-state physics and materials science. He received his PhD degree from Harvard University, where he studied the physics of disordered solids and battery materials. Recently, he has been studying scaling and robustness properties of large neural networks, and their applications to materials discovery for applications, including clean energy. Cubuk can be reached by email at cubuk@google.com.



Maxim Ziatdinov is a research scientist and team lead at Oak Ridge National Laboratory. His research is directed primarily toward the synergy of the experiment, theory, and machine learning for accelerated scientific discovery. He is particularly interested in developing physics-based machine learning approaches to enable the "smart labs" of the future. He has pioneered the development of artificial intelligence-guided autonomous experimentation in microscopy ("self-driving microscopes") and developed the hypothesis learning framework for co-navigation of the hypothesis and experimental spaces in automated experiments. Ziatdinov can be reached by email at ziatdinovma@ornl.gov.

Mahshid Ahmadi is an assistant professor of materials science and engineering at The University of Tennessee, Knoxville. She graduated from Nanyang Technological University, Singapore, in 2013. Her research focuses on machine learning-driven automated synthesis and characterization for materials discovery and design with focus on hybrid perovskites. Other interests include understanding chemistry of hybrid perovskites using different techniques with spatial resolution such as scanning probe microscopy, time-of-flight secondary ion mass spectrometry, and scanning electron microscopy cathodoluminescence. Ahmadi can be reached by email at mahmadi3@utk.edu.