

Atomic-Scale Modeling for Materials and Chemistry

Nam Q. Le, Janna Domenico, K. Michael Salerno, and Christopher D. Stiles

ABSTRACT

Atomic and molecular modeling techniques have developed over the past 75 years into a vibrant field of computational science, used to understand and predict materials properties and phenomena in academic, industrial, and government labs. Researchers today have the benefit of decades of Moore's law growth in computer processors, decades of algorithm and software development, experiments capable of atomic-scale characterization for validation, and a deeper understanding of the strengths, limitations, and complementary features of different computational methods. It is not surprising then that important problems in many fields—battery chemistry, drug design, mechanics of materials, biocompatibility, and catalyst design—are routinely studied using atomic-scale simulation and modeling. In this article, we first outline a brief history and background of the density functional theory and molecular dynamics methods. Next, we discuss several case studies that exemplify how scientists and engineers at the Johns Hopkins University Applied Physics Laboratory (APL) use these computational methods to attain APL's broader goals and mission. Finally, we discuss future directions for atomic-scale modeling and calculations, such as integration with modeling methods at other scales and with artificial intelligence-enabled frameworks, to meet the next generation of sponsor challenges.

INTRODUCTION

If, in some cataclysm, all of scientific knowledge were to be destroyed, and only one sentence passed on to the next generations of creatures, what statement would contain the most information in the fewest words? I believe it is the *atomic hypothesis* (or the *atomic fact*, or whatever you wish to call it) that *all things are made of atoms* . . .

—Richard Feynman,
The Feynman Lectures on Physics¹

We strive to build all physical human technology using materials with just the right stiffness, clarity to light, or push or pull to electrons. We rely on chemicals with just the right stick or slip, taste and smell, and interference with toxicity and disease. All these properties of materials and chemicals arise, albeit in complex ways, from two attributes: the elements that compose them and their spatial arrangement, atom by atom.

Humans have not always needed to know that all things are made of atoms, but our capabilities have expanded immeasurably now that we do. In many cases, the behavior of huge numbers of nearby atoms does not depend at all on the details of a few. In those cases, one can step back from the trees and use models at larger scales based on average properties of the forest, as described in this issue by Darragh et al. However, in many other cases, differences at the atomic scale have measurable, human-scale effects. Researchers and engineers have rapidly developed ways to exploit these differences to design polymers, drugs, doped semiconductor devices, and nanostructured materials whose incredible properties can vary drastically based on the identity and placement of individual atoms in a motif.

As humans continue to invent novel materials, our capabilities to physically manipulate and engineer materials on these scales improve as well, so that the shoreline of new possibilities is expanding rather than shrinking. Exhaustive searches of all possible materials and chemicals will be impossible even if the most optimistic projections of experimental technologies are realized.² Models and simulations are therefore critical to predict the properties of materials based on new compositions and atomic structure, sight unseen, in tandem with physical synthesis and characterization methods.

This article provides a brief survey of important methods for atomic-scale modeling and illustrates how they can impact missions relevant to APL and its sponsors. Methods such as density functional theory (DFT) and classical molecular dynamics (MD) have decades-long histories of development and fruitful application across materials science, chemistry, and biology. This includes not just fundamental scientific work but applied research as well: just a few examples are designing macromolecules for pharmaceuticals,³ silicate glass for consumer devices,⁴ and polymers for defense applications.⁵ DFT and MD have become critical links in a chain of physical modeling methods that capture different physical phenomena across length and timescales. As improvements continue in theory, software, and computational hardware, the range of practicality and applicability of these methods also expands. We describe a few example challenges of interest to APL that require atomic-scale modeling, and we conclude with an outlook on the future of atomistic modeling.

BACKGROUND

DFT and MD are the two most widely used atomistic modeling techniques today. They are complementary tools, capturing different physics to answer different questions. They were developed largely in parallel, with only loose coupling between them.

After the formal theory of quantum mechanics was laid out in the mid-1920s, scientists realized its nearly universal predictive power, but they also immediately realized the importance of developing approximate models that could be solved efficiently. Paul Dirac wrote in 1929 that “the underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much too complicated to be solvable.”⁶ In principle, all thermal, mechanical, electronic, and chemical properties can be predicted by solving the Schrödinger equation, but the exponential scaling of its complexity makes a solution impossible for systems with more than a few dozen electrons.

DFT is widely used because it addresses this challenge of capturing enough quantum mechanical effects to predict properties accurately, while being computationally efficient enough to solve quickly. Hohenberg, Kohn, and Sham developed the fundamental theories in 1964 and 1965, showing that a system’s ground-state properties can be determined from its total electron density instead of the more complex many-body wavefunction.⁷ In other words, DFT is a method of obtaining an approximate solution to the Schrödinger equation for a many-body system.

The wide adoption of DFT began in earnest decades later in the 1980s and 1990s enabled by joint progress in theory, software, and computational hardware. With time, physicists developed more accurate functionals, and these theoretical developments were steadily incorporated into commercial and open-source software,^{8,9} making them available to chemists and materials

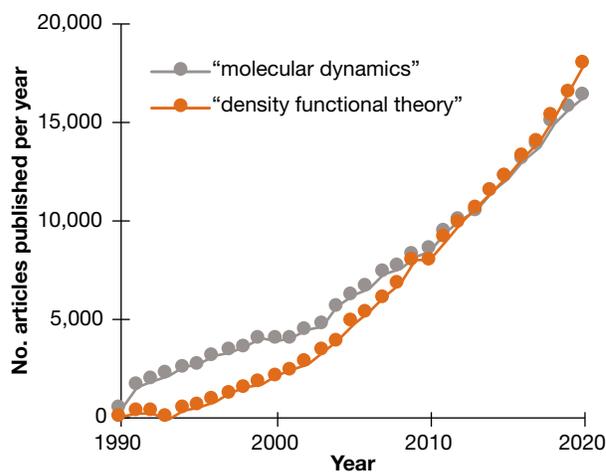


Figure 1. Number of publications using atomistic modeling since 1990. The number of journal articles published per year containing the phrases *molecular dynamics* and *density functional theory* in the title or abstract continues to rise, as indexed by the Clarivate Web of Science. Data from Web of Science, provided by Clarivate. Web of Science and Clarivate are trademarks of their respective owners and used herein with permission.

scientists as well. Accordingly, the volume of research based on DFT grew rapidly, as shown in Figure 1. Today, researchers use DFT to model inorganic and organic materials across academia, industry, and government institutions.

In parallel, several efforts in the mid-20th century began to use computers to model materials at the atomic and molecular level using a classical description to capture phenomena in which quantum mechanical effects can be neglected. In 1964, the first MD simulation was performed by Aneesur Rahman at Argonne National Laboratory. It simulated just 864 atoms of liquid argon for only 10^3 time steps ($\approx 10^{-12}$ s), but allowed for measuring the atomic radial distribution function, velocity autocorrelation, and diffusion.¹⁰ Molecular interaction models developed throughout the 1940s and 1950s (used for finding minimum-energy, optimum molecular structures) were subsequently used in MD simulations to model increasingly complex systems.

Early researchers recognized the shortcomings of MD's classical description and limited length and time-scales and worked to overcome them. In 2013, the Nobel Prize in Chemistry recognized two different flavors of "multiscale models," developed in the 1970s, that (1) presented a systematic way to simulate some parts of a molecule quantum-mechanically while treating other parts classically and (2) combined distinct amino acids within a protein into pseudoatom units, enabling much larger and faster simulations of proteins and peptides.¹¹ As shown in Figure 1, the utility of MD paired with increasing accessibility to software and hardware has led to rapid growth in usage in parallel with DFT. Today, all manner of materials from metals to organic molecules and biomolecules are regularly simulated in systems with more than 10^7 atoms, and for times greater than 1 μ s. Modern simulations run on thousands of computer processors and specially designed hardware for weeks or months and can generate terabytes of data.

Despite the obvious increase in computing resources since the early days of molecular simulation, researchers face many similar challenges today. Treating molecular systems both classically and quantum-mechanically can be critical when modeling many systems. In particular, APL efforts to understand and design quantum-biological systems rely on designing and understanding these dual-fidelity models. Many physical phenomena arise from coupling of physics across many scales, requiring a multiscale modeling approach.

METHODS

DFT and classical MD are the two best-established modeling methods to account for electronic and atomic effects on the properties of materials and chemical compounds. In the following sections, we briefly describe how they work and what phenomena they can model.

Density Functional Theory

DFT is the most widely used method today for incorporating electronic effects in atomistic models of materials and molecules. The goal of any electronic structure method is to compute the energies of electrons in a chemical system, and by doing so, any related property can be determined. The crux of DFT is that only the electron *density*—a function of three coordinates—is needed to predict properties of the system. The density is just a scalar field that depends on position, regardless of the number of electrons in the system. This is an enormous simplification from the many-body wavefunction of quantum mechanics, which is a complex field that depends on the coordinates of all N electrons in the system: $\Psi(\vec{r}_1, \vec{r}_2, \dots, \vec{r}_N)$. The relationship between the density perspective and the many-body perspective is illustrated schematically in Figure 2. These two approaches have been proven equivalent if the exact dependence of energy on $\rho(\vec{r})$ is known. This dependence is called the exact DFT *functional*. Unfortunately, the form of the exact functional is not known, and the utility of DFT hinges on the development of *approximate* functionals that balance accuracy and computational efficiency. Physicists have proposed many functionals, and each has its own benefits and drawbacks; for more on the selection of functionals, the authors recommend Ref. 12. Given a system of N electrons, whereas the computational complexity of the many-body problem scales exponentially as 2^N , the cost of DFT scales as N^3 . In practice, this makes DFT calculations currently tractable for systems with up to 10^3 to 10^4 valence electrons, as opposed to tens of electrons for solving the full many-body problem.

Given a set of atomic elements and positions in space, a DFT software package works to compute the

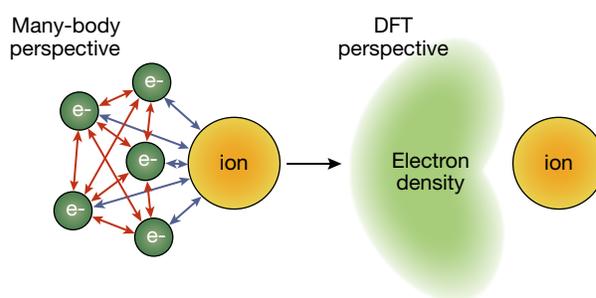


Figure 2. Relationship between electron density perspective and many-body perspective. More direct approaches to modeling quantum mechanical systems of N electrons solve for the many-body wavefunction, a function of $3N$ coordinates. DFT, however, relies only on the electron density, which is a function of three spatial coordinates and results in a simpler mathematical problem. (Reprinted by permission from Springer, *MRS Bulletin*, M. T. Lusk and A. E. Mattsson, "High-performance computing for materials design to advance energy science," vol. 36, no. 3, pp. 169–174, © 2011, <https://doi.org/10.1557/mrs.2011.30.13>)

corresponding ground-state electron density in a system. If the system is periodic, such as in a crystalline solid material, symmetries of the lattice are included here. A DFT practitioner will then prescribe calculation details that provide the best compromise among desired accuracy, time, and computing resources. These additional details include the DFT functional, basis functions, and pseudopotentials, which incorporate the chemical environments of the atoms, among others. The energy of the system is computed iteratively until self-consistency is achieved, and a converged electron density and energy are the main results.

The output of a DFT calculation is the ground-state electronic density of a system, from which any related property can be computed. Examples of these properties include the optimized equilibrium geometry, free energies (from which one can compute relative stability of related structures), and energetics of defects.

The most basic applications, however, are to calculate electronic and optical properties (e.g., energies of molecular orbitals in nonperiodic systems or band structure in periodic systems). The total energy as a function of atomic geometry is also a useful result and allows a researcher to determine binding preferences (e.g., an adsorbent and surface for a chemical sensor or a substrate and an enzyme in a biochemical reaction). This is computed by taking a simple difference between the energy of the bound system and the energies of the constituents.¹⁴ The converged density of electrons in a system can also provide insight into its charge distribution, which in turn elucidates the partial charges in a system. In short, DFT is widely used for property prediction and screening of materials or molecules for a variety of specific applications.

Classical MD

In many material systems, electronic degrees of freedom can be ignored at larger length scales, and classical MD can be used to model atoms and molecules. The method is a discretization and integration of Newton's second law, $\vec{F} = m\vec{a}$, for every atom in a material system. Intramolecular forces are calculated from simple potential energy functions that contain terms representing molecular bonds (characterized by equilibrium distances and vibrational frequencies), three-atom angles, and four-atom torsions. Intermolecular van der Waals forces and electrostatic interactions are also included. All these interactions together make up a molecular "force field" that parameterizes interactions for different atomic species in different chemical environments.¹¹

Taking atoms rather than electrons as the fundamental unit enables orders of magnitude larger simulations (partially because of better scaling), but simulations are currently limited to $\approx 10^7$ degrees of freedom (atoms). As the name molecular dynamics (MD) implies, capturing

atomic and molecular dynamics is an important feature; however, resolving atomic bond vibrations requires an integration time on the order of a femtosecond (10^{-15} s). Even a relatively long simulation of 10^8 time steps only captures ≈ 100 ns, so processes that evolve over much longer timescales require special techniques to simulate. Many advances in algorithms and computational methods have sought to overcome these length and timescale limitations. Advances addressing the former involve sophisticated methods to couple different length resolutions; for example, continuum flow or solid media surrounding a computational region with atomic detail.¹⁵ Methods to address the latter go by the name "enhanced sampling" because they "push" simulations to sample statistically unlikely states. For example, a protein unfolding transition, which might naturally occur on a millisecond timescale, can be "pushed" to sample the transition state in a physical way on a realizable simulation timescale $\approx 10^7$ times faster.¹⁶

One of the inherent strengths of MD is the natural, central position of temperature in simulations. The motion of atoms and molecules is explicitly simulated and the kinetic energy of this motion defines the temperature. Intuition suggests this is an important feature! Decrease the motion or temperature of a system, and the simulated materials change their properties: a solid's mechanical response becomes more brittle, or the water surrounding a protein freezes. Increase the temperature, and a polymer softens or a protein becomes denatured. This natural realization of temperature in simulation is important for understanding many material properties and for connecting explicitly with experimental results and trends.

Explicitly simulating atomic and molecular motion also allows researchers to model nonequilibrium and dynamic phenomena, typical conditions found in materials processing. Example applications include modeling the intricate folding and binding of biological macromolecules,¹⁷ crack propagation during fracture of inorganic materials,¹⁸ and heat transfer in nanoscale devices.¹⁹ Phenomena like nonelectronic heat and mass diffusion, vibrational spectra, and rheological response in flow are all routinely modeled using MD. In all these cases, the relevant material properties are impractical to calculate analytically, but emerge from the complex motion and interactions of constituent atoms. Dynamic phenomena such as flow rheology, adatom or interstitial/vacancy diffusion, mixing, phase (or glass) transitions, and many others can be modeled to better understand the effects of processing or operating conditions at an atomic scale.

CASE STUDIES

These modeling methods have a range of applications: at APL, many internally funded and sponsor-funded initiatives are employing atomistic modeling to achieve

their goals. Calculations for these projects have made use of existing mature software packages: Quantum Espresso⁸ and NWChem⁹ were used for periodic and nonperiodic DFT calculations, and NAMD²⁰ and LAMMPS²¹ were used for organic and inorganic MD simulations, respectively.

Designing Reactive Oxides to Destroy Toxic Environmental Compounds

APL is currently applying atomic-scale models in collaboration with researchers at the US Naval Research Laboratory (NRL) and the US Army Chemical Biological Center (CCDC-CBC) to develop novel materials for destroying toxic chemical compounds in the environment. Responding to a need from the Defense Threat Reduction Agency (DTRA), APL aims to improve on NRL-developed aerogel material that harnesses light to decompose toxic environmental compounds²² by extending their activity for hours after the light is removed. Current technologies based on photocatalytic oxide materials can effectively break down toxic compounds under UV illumination, as shown in Figure 3a. We are therefore researching ways to keep this solar-powered chemistry working “around the clock.”

The team’s proposed solution is to develop composite, high-surface-area aerogel materials based on certain oxides, including engineered defects that can bind and extend the lifetime of photo-generated reactive oxygen species (ROS) such as superoxide ($\text{O}_2^{\bullet-}$).²³ As illustrated in Figure 3b, this will result in a material that can generate ROS under illumination (left) and store them to oxidize compounds hours later (right). However, significant development is still required to improve performance and translate this concept into mission-ready materials. The overall challenge requires optimizing optical, electronic, chemical, and thermodynamic properties in a large design space of material composition and structure.

Atomic-scale modeling is a powerful complement to experimental synthesis and characterization in navigating that space: DFT provides a “virtual laboratory” for modeling oxide surface structures and calculating coupled electronic and chemical properties with control not attainable experimentally. This will help answer critical questions: What types of defects maximize the stabilization of ROS? How can we synthetically incorporate those desired defects and optimize their density? We are therefore using DFT to calculate and compare the intrinsic stability of different defect types (surface and subsurface oxygen vacancies) on selected material surfaces, along with the relative binding strength of neutral oxygen molecules to form $\text{O}_2^{\bullet-}$ (Figure 3c), and finally coadsorption with organic toxic compounds, questions that have only been partially addressed in previous literature.^{24,25} This modeling work is proceeding hand in hand with experimental work at NRL testing strategies to incorporate those defects in synthesized samples and at CCDC-CBC to assess their effectiveness in destroying toxic compounds.

Bridging Scales in Quantum Biology

Can migratory birds use radical pairs and spin chemistry to sense the direction of an external magnetic field?²⁶ How could molecular simulations and quantum calculations predict magnetic field sensitivity and help us to engineer more sensitive molecular sensors?

A multiscale model of MD and electron dynamics is being used to understand how an external magnetic field influences electron-scale interactions and how information about the electron interactions propagates to larger scales. The work builds from a long history of linking classical and quantum mechanical calculations to model complex molecular systems where both electron-scale and molecule-scale dynamics are important.

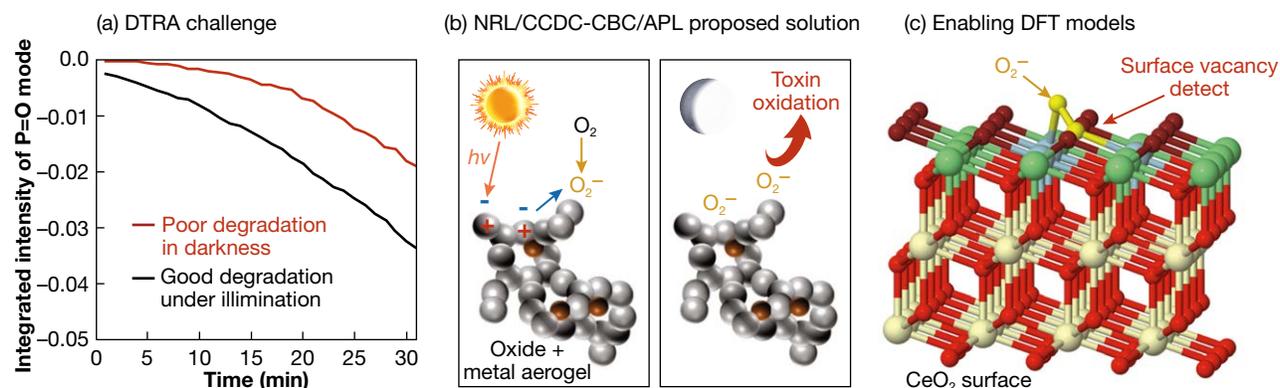


Figure 3. Overview of a collaborative effort among APL, NRL, and CCDC-CBC to address a current performance gap. (a) Existing photo-driven materials for destruction of toxic compounds quickly lose efficacy in darkness. (b) Materials with tailored composition and defects could stabilize reactive chemical species for much longer, enabling continued efficacy in darkness. (c) APL is performing DFT calculations to understand and optimize the effect of chemical and structural features on lifetimes of reactive species.

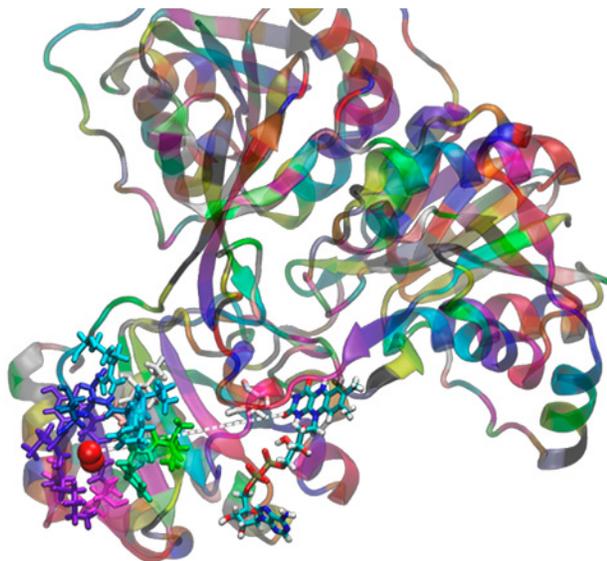


Figure 4. Common protein model with embedded cofactor molecule in “wet” biological environment (solvent not shown). The backbone of the electron-transfer flavoprotein molecule (translucent multicolored) surrounds the FAD molecule (bottom center, thick bonds) and oxygen molecule (red). The oxygen molecule is caged by the side-chain atoms (thick bonds) of the amino acid residues at a potential electron-transfer reaction site. Each amino acid residue is uniquely colored.

The physical system modeled comprises a common protein (e.g., certain cryptochrome proteins), an embedded cofactor molecule (flavin adenine dinucleotide [FAD]), and the surrounding “wet” chemical environment, as illustrated in Figure 4. Electron transfer can occur among these three subsystems when an incident photon creates a photoexcitation in the FAD. The electron transfer creates a pair of radicals, leading to the phenomenon of “spin chemistry,” where the quantum spin state of the radical pair affects the resulting reaction products. The shift in the reaction products from one species to another depends on orientation within the earth’s magnetic field and is believed to give birds the measurement needed to guide flight. Researchers hope this novel sensing mechanism could prove to be tunable in the lab and exploitable in future devices.

Currently, APL is developing protein samples to test their magnetosensitivity. At the same time, MD simulations have pinpointed several molecule positions and orientations where radical pair formation may occur. These candidate molecular configurations are passed to DFT calculations of electron transfer that results in magnetically sensitive radical pairs. Current work in the multiscale modeling of quantum biomolecular phenomena is supported both through internal research and development funds at APL and through funding from the National Science Foundation, which also provides high-performance computing resources.

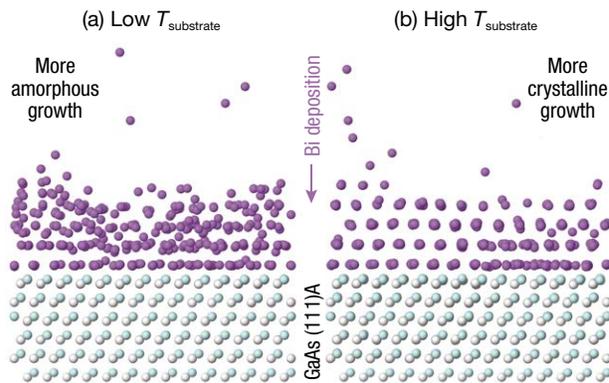


Figure 5. Snapshots from MD simulations of thin film growth to complement experimental MBE. Simulations at low temperature (a) exhibit more amorphous growth while high temperatures (b) exhibit more crystalline growth. Such simulations can help assess hypotheses explaining experimental observations and guide further process development.

Understanding Fabrication Quality of Inorganic Thin Films

MD simulations are also used frequently outside of biological systems to model materials in different thermodynamic conditions or nonequilibrium processes. One such process is molecular beam epitaxy (MBE), which is used to grow extremely high-quality thin films one atomic layer at a time. The high quality of these thin films is critical for application areas like thermoelectrics²⁷ and inorganic phase change materials.²⁸ Successful MBE growth requires optimizing multiple process parameters. One important parameter is the temperature of the substrate onto which the film is deposited. In Figure 5, simulation snapshots show the growth of bismuth (Bi) on substrates of gallium arsenide (GaAs). These results show a transition from the growth of an amorphous Bi film at low substrate temperature (Figure 5a) to a crystalline Bi film at higher temperatures (Figure 5b), an effect that is seen experimentally in other material systems.²⁹ The results highlight the utility of using MD to model and understand nonequilibrium, dynamic phenomena relevant to experiments or materials processing.

Designing Novel Materials

There is great potential in coupling machine learning (ML) methods with atomistic modeling. At APL, we attempt to address this through an ongoing internally funded initiative known as Material Invention Through Hypothesis-unbiased, Real-time, Interdisciplinary Learning (MITHRIL). The overall goal of MITHRIL is to link ML, experimental synthesis and characterization, and atomistic-scale modeling to accelerate materials discovery, a process that currently takes many years. The approach aims to (1) use ML to better guide searches of

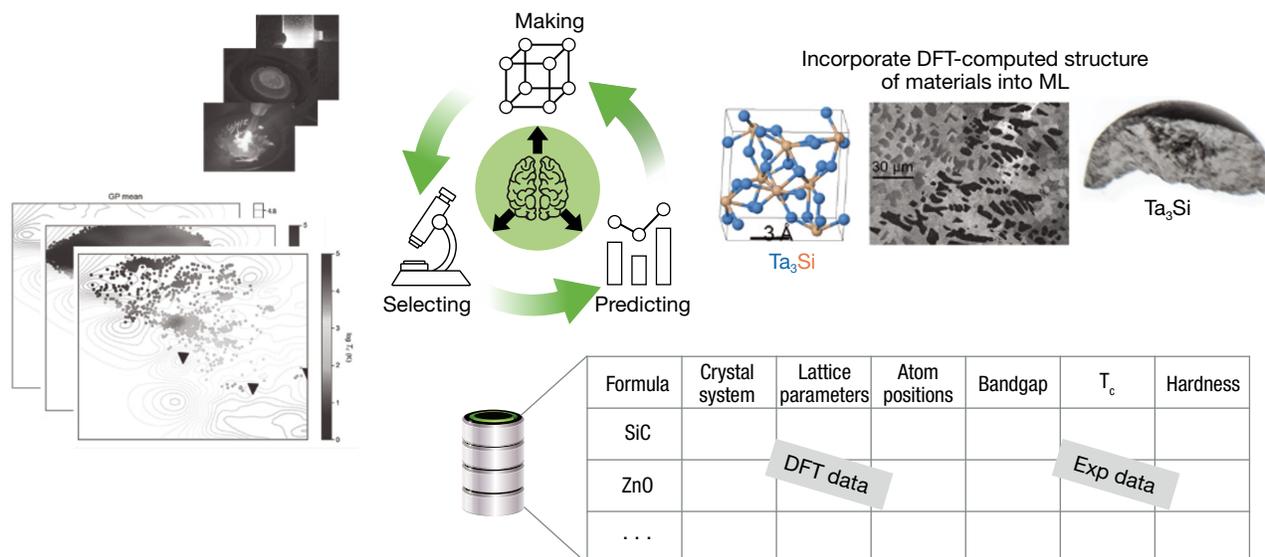


Figure 6. Role of DFT within the scope of MITHRIL. Databases of DFT-generated structure and property data are bootstrapped with relevant experimental (Exp) data and used to feed ML models to make property predictions.

materials space and suggest material that satisfies requirements for a specific application, (2) experimentally synthesize and test those suggestions, and (3) incorporate new data to enable further design iterations. Example roles of atomistic modeling are to predict stable crystal structures and to predict the physical properties of materials that have not yet been synthesized. MITHRIL leverages existing databases of computational materials data in the form of DFT results, as shown schematically in Figure 6, to generate models for property prediction. Because of the sizes of the databases (~800,000 materials), ML methods are obvious candidates to help efficiently explore the vast and complex spaces of possible new materials.

The DFT-generated data provides the ML models with structure and property relationships governed by the laws of physics and chemistry and at lower cost than running full-fledged physical experiments. The current properties of interest in MITHRIL are related to superconductivity (e.g., critical temperature [T_c]), but the approach is transferable to any field. Leveraging resources developed via ML and experiments, MITHRIL seeks to enable closed-loop materials discovery through the optimal exploitation of well-mapped materials spaces and efficient exploration of poorly understood regions. This is where potential novel and impactful discovery occurs.

FUTURE DIRECTIONS

What does the future hold for atomic-scale modeling methods? Hardware and software will certainly continue to improve, meaning that researchers will be able to apply DFT and MD to capture larger length and timescales in more complex systems. In addition,

new modeling approaches will emerge as a result of integrating DFT and MD with other methods—both physics-based approaches such as finite element analysis (FEA) and computational fluid dynamics (CFD), and data-driven approaches such as ML models.

Researchers have devised many strategies for combining physics-based modeling methods at different scales, but much remains to be done to increase their computational efficiency and transferability among different material systems. Two examples within atomistic methods are *ab initio* MD, in which forces calculated from DFT (rather than classical force fields) are used to drive MD simulations³⁰ and spatially decomposed multifidelity methods such as QM/MM (“quantum mechanics/molecular mechanics”) and ONIOM.³¹ At larger scales, the quasicontinuum method³² and heterogeneous multiscale method³³ are examples of approaches for bridging atomic-scale and continuum methods such as FEA and CFD. These methods remain highly application specific at present, but we envision exciting progress toward more robust multiscale models in the near future, as technologies rely on increasingly advanced materials with tailored structure and properties at multiple scales.

Clever application of data-driven ML methods could provide one path to better integration of atomistic methods with other modeling approaches and with experiments. One view of scientific progress counts data-driven discovery as the “fourth paradigm” of science,³⁴ arriving now after the empirical, theoretical, and computational paradigms. Applying ML techniques is a natural next step because of the vast space of possible materials or molecules, which is impractical to search completely through experiments alone. ML techniques are being used to rapidly map a molecule or material’s structure to

its properties and require a sufficient amount of generated data as training data. But an ML model is only as good as its data, and for this field to advance, researchers need findable, accessible, interoperable, and reusable data. Further, ML algorithms applied so far have not deeply incorporated chemical theory, which would help accelerate our understanding. We expect developments in chemistry-informed ML to decrease the time to solution, facilitated by a push toward open chemistry that is generating more openly available chemical and materials data. It is important to note that ML is not going to replace computation, but rather augment and expedite it while enhancing understanding.

SUMMARY

The field of atomistic modeling is a product of the corresponding development of the fields of chemical and physical theory and computing, and since its nascence in the mid-20th century, it has become a key component of materials, chemical, and biological sciences. These tools are being increasingly integrated into projects at APL to validate and drive experimental measurements or designs and to predict properties of molecules and materials. As these tools become more widespread at APL, there is great opportunity to make key contributions across the fields of biology, chemistry, and materials science, and to incorporate these tools into the process of discovering new defining innovations.

ACKNOWLEDGMENTS: The authors acknowledge support from the Defense Threat Reduction Agency (DTRA), the National Science Foundation (NSF) under Grant No. 2051510, and internal research and development funding through APL's Research and Exploratory Development Department Mission Area.

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Nam Q. Le, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Nam Q. Le is a computational materials scientist in APL’s Research and Exploratory Development Department. He has a BS in mechanical engineering and a PhD in mechanical and aerospace engineering from the University of Virginia. He was a postdoctoral fellow in the chemistry division at the US Naval Research Laboratory before joining APL in 2018. He has experience in applying both quantum and classical atomic-scale modeling methods to address research problems across materials science and chemistry. His email address is nam.q.le@jhuapl.edu.



Janna Domenico, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Janna Domenico is a computational chemist in APL’s Research and Exploratory Development Department. She has a BS in chemistry and a BA in Spanish language and literature from William Paterson University of New Jersey and a PhD in physical chemistry from Drexel University. Janna tackles problems at the intersection of chemistry, materials science, and data science, with a focus on coupling machine learning with materials science and quantum chemical calculations. Her areas of expertise include computational materials science, materials modeling and simulation, and nanomaterials for energy. Her email address is janna.domenico@jhuapl.edu.



K. Michael Salerno, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

K. Michael Salerno is a computational physicist in APL’s Research and Exploratory Development Department. He has an AB in political science from Princeton University and a PhD in physics and astronomy from the Johns Hopkins University. Michael has worked with research teams across academia, government, and industrial labs modeling materials with atomistic and coarse-grained simulations. He has experience and expertise in developing new computational models to simulate polymeric, “soft,” and disordered materials. His previous research has focused on reproducing fundamental material properties across multiple modeling scales and on rationalizing and understanding experimental results with molecular simulation data. His email address is michael.salerno@jhuapl.edu.



Christopher D. Stiles, Research and Exploratory Development Department, Johns Hopkins University Applied Physics Laboratory, Laurel, MD

Christopher D. Stiles is a computational physicist and supervisor of the Multiscale Mathematical Modeling Section in APL’s Research and Exploratory Development Department. He holds a BS in mathematics and physics and a PhD in nanoscale science and engineering, both from the State University of New York at Albany. He specializes in computational approaches of materials science to model real-world systems—developing, using, and advising on the use of cutting-edge computational methods. His email address is christopher.stiles@jhuapl.edu.