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Artificial Intelligence for Advanced Functional Materials: Progress and Emerging Frontiers

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ABSTRACT

Artificial intelligence (AI) is transforming the way materials are designed, understood, and manufactured. This Perspective examines how recent advances in data-driven modeling, high-performance simulation, and autonomous experimentation are converging to accelerate the discovery of functional materials for next-generation technologies—from energy storage and biomedicine to

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nanoelectronics and quantum devices. We outline ongoing strategies to embed AI across the materials design workflow—from synthesis and characterization to large-scale simulations enabled by machine learning techniques and approaching ab initio accuracy—and discuss key challenges that remain on the path toward intelligent (bio)materials discovery.

1 | Introduction

(Cristiano Malica, Stephan Roche, Kostya S Novoselov)

Artificial intelligence (AI) is progressively reshaping the scientific and technological landscape, opening unprecedented avenues for accelerating materials discovery and design. What once relied primarily on human intuition and incremental experimentation is now being complemented by data-driven approaches that co-integrate the analytical power of AI with the creativity and interpretive depth of human mastery. Far from replacing scientists, AI augments human intelligence, transforming how questions are posed, data interpreted, and strategies developed in both academic and industrial research. This emerging synergy is fostering a new generation of collaborative frameworks that bridge fundamental science with industrial design toward accelerating practical innovation, responding to an increasing demand for AI-enabled solutions in materials and chemical industries.

Within this rapidly evolving context, the integration of AI and machine learning (ML) methods across the materials design workflow—from synthesis and characterization to predictive modeling—has become a defining trend. Autonomous laboratories and robotics-assisted experimentation are beginning to accelerate the traditional cycle of hypothesis, testing (trial and errors), and refinement, while data-driven methodologies are enabling the prediction of non-intuitive synthesis pathways, stability landscapes, and target properties for both large ensemble of complex molecules and advanced innovative materials. Simultaneously, the rise of universal ML potentials trained on quantum-mechanical data is unlocking the possibility of simulating disordered, amorphous and other complex/heterogeneous systems with near ab initio accuracy, extending the predictive power of computational materials science to unprecedented scales.

The present Perspective builds upon the foundations established in the first AI4AM Perspective [1] (C Malica, K S Novoselov et al., 2025 *J. Phys. Mater.* **8**, 021001), and brings together complementary views from leading experts worldwide. Rather than aiming to provide a fully exhaustive or uniquely unified roadmap of the rapidly evolving applications of AI in materials science, the manuscript is conceived as an ensemble of thematic snapshots reflecting representative directions, methodologies, and challenges currently explored across different areas of the field. The Perspective therefore collects a broad range of viewpoints, from autonomous laboratories for materials synthesis to data-enabled molecular design, machine-learned interatomic potentials (MLIPs), ML-optimized exchange–correlation functionals, and AI-driven strategies for quantum technologies. Together, these contributions highlight emerging trends, methodological opportunities, and conceptual challenges associated with the integration of AI into materials research, while also emphasizing the need for innovative solutions and stronger partnerships between academia, industry, and policymakers.

This rapidly evolving and multidisciplinary field benefits from continued efforts to organize and exchange knowledge across experimental, computational, and theoretical domains. In this spirit, international initiatives such as AI4AM (www.ai4am.net) provide open and collaborative platforms for connecting communities, sharing tools, and fostering dialog across different research directions in materials science.

This scope also requires a clearer statement of the limits of the present article. The contributions originate from a conference-based community and therefore should not be read as a geographically or thematically exhaustive survey of AI for materials science. The emphasis on European infrastructures, MLIPs, electronic-structure surrogates, and data-workflow platforms reflects the composition of the present collection. At the same time, several rapidly expanding directions are insufficiently represented in the body of the article and deserve explicit recognition, including US-based work on autonomous scientific reasoning, LLM-driven hypothesis generation beyond synthesis prediction, graph-native discovery platforms, hierarchical and bioinspired materials design, and multi-agent systems that combine retrieval, simulation, critique, and planning in closed loops [2–6].

In this broader landscape, the transition from prediction to discovery increasingly depends on agentic workflows rather than isolated models. Recent systems such as multi-agent graph-reasoning frameworks, protein- and alloy-design agents, and distributed scientific-agent ecosystems illustrate a shift from single-step property prediction toward autonomous hypothesis generation, tool orchestration, experimental or computational planning, and iterative validation [2–7]. These developments do not replace the contributions collected here, but they sharpen the interpretation of the Perspective: the vignettes should be viewed as selected entry points into an evolving field whose next phase will likely combine generative models, physics-based simulators, knowledge graphs, autonomous laboratories, and human oversight into auditable discovery loops.

The same caution applies to the concept of trust. Uncertainty quantification is indispensable, but it is not by itself sufficient to establish reliable AI-driven discovery. Trustworthy workflows also require physical grounding, adversarial falsification of proposed hypotheses, compositional world-model construction, explicit provenance, negative or failed experiments, and reproducible digital-to-physical validation. Emerging decentralized-science and artifact-exchange frameworks further suggest that provenance-aware coordination among independent agents may become an important complement to centralized databases and self-driving laboratories [7, 8].

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2 | Internal Sections

2.1 | Machine Learning for Inorganic Synthesizability and Synthesis Planning

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With advances in machine learning, a wide range of promising hypothetical materials can now be generated; however, their experimental realization remains one of the major challenges. Since it is impractical to attempt synthesis for the enormous number of hypothetical candidates, studies for estimating synthesizability in advance are needed. In solid-state materials, thermodynamic stability has been widely used as an approximate indicator, and it often provides a useful first-pass filter for large chemical spaces. Recently, as another alternative approach, data-driven approaches have increasingly been adopted to capture complex underlying patterns for determining inorganic synthesizability. Among them, one representative work is structure-based synthesizability prediction using partially supervised learning, where positive and unlabeled learning is adapted to accumulated materials databases [9]. Using this model, hypothetical structures can be classified as synthesizable or unsynthesizable based on structural patterns, enabling high-throughput workflows to prioritize candidates for experimental validation. More recently, large language models (LLMs) have emerged as practical tools for chemistry using domain-specific fine-tuning. Using this strategy, fine-tuned LLMs have been evaluated for predicting synthesizability of inorganic compounds, showing comparable performance relative to bespoke machine learning models [10]. Beyond prediction, interpretability is important for understanding the factors that govern the synthesizability of solid-state materials. To explore the explainability of inorganic material synthesizability, one study showed an LLM fine-tuned on inorganic synthesizability knowledge can be utilized to extract reasons for why a material is predicted to be synthesizable or unsynthesizable [11]. They fine-tuned an LLM on textual expressions of inorganic structures and, after predicting synthesizability, extracted the rationale behind each decision, generating human-readable explanations. These LLM-based workflow for explainable synthesizability prediction identified diverse factors that govern synthesizability.

Collectively, these data-driven studies have demonstrated that their predictions align well with experimental outcomes, supporting their potential as practical tools. However, cross-study analyses showed that the fraction of hypothetical materials predicted to be synthesizable remains quite low. In other words, a large fraction of virtual candidates is predicted to be unsynthesizable and filtered out, motivating the need for approaches that can recover such discarded candidates. To address this issue, recent work has introduced LLM-based methods that redesign structures with unsynthesizable structures into synthesizable alternatives [12]. Using this approach, it successfully redesigned 11,753 low-synthesizability structures into 3395 high-synthesizability structures. Moreover, among the top 100 redesigned structures, 34 had been experimentally reported despite being absent from the dataset used, indicating that the fine-tuned LLM's redesigned candidates were not hallucinations but experimentally realistic.

After assessing the synthesizability of inorganic materials, the next step is to determine how to synthesize those synthesizable materials. For the synthesis planning, there have also been efforts to develop models that predict specific synthesis recipes, such as precursors and heating temperatures [13]. They used a dataset of text-mined inorganic synthesis recipes from the literature to construct precursor-templates, and developed a model that can score candidates within this predefined template space to produce ranked recommendations. Through this approach, the model produced more chemically valid and experimentally realistic precursor predictions, and demonstrated its ability to provide recipe suggestions that experimentalists can prioritize for initial trials.

Collectively, using accumulated materials and recipe datasets, these data-driven approaches have been shown to work well and have been studied as effective methodologies; however, several limitations still remain. They are highly sensitive to the underlying data, which can contain anthropogenic biases, and the available datasets are often dominated by positive (reported) cases with few or no reliable negative examples. Therefore, it is important to reduce dataset bias and to expand reliable negative examples through high-throughput experiments and emerging self-driving labs (SDLs). If these issues are addressed, data-driven machine learning studies for inorganic synthesizability and synthesis planning are expected to become more robust.

2.2 | Computational Materials Science With Machine Learning: From Data to Insights

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The rational design of functional materials—light absorbers, transparent electrodes, thermoelectrics, and beyond—remains one of the grand challenges of materials science. The combinatorial space of possible compounds is far too vast to explore by trial and error or even by systematic first-principles calculations alone, and yet innovative technologies are still waiting for the right materials. Data-driven approaches powered by machine learning are increasingly central to addressing this challenge, provided that the underlying data are reliable, openly accessible, chemically meaningful, and that the representations used to encode materials preserve physical intuition.

A prerequisite for effective machine learning in materials science is the availability of large, high-quality datasets. The ALEXANDRIA materials database provides density functional theory (DFT) calculations for approximately seven million periodic compounds spanning one-, two-, and three-dimensional crystal structures, computed with PBE, PBEsol, and SCAN

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exchange–correlation functionals [14–16]. The database is openly distributed, accessible via an Optimade interface, and integrated into the NOMAD repository. With more than 200,000 thermodynamically stable compounds identified on the convex hull, compared to around 50,000 experimentally known unique stable phases in ICSD, ALEXANDRIA substantially expands the known materials landscape. Yet scale alone is not sufficient. Analysis of property distributions across this dataset reveals that all target quantities follow non-normal distributions, a statistical feature that directly governs machine learning training behavior and prediction errors, and that must be explicitly accounted for in model design and evaluation [16]. This insight points to a broader principle: understanding the data is as important as accumulating it.

Representing materials in a way that is both computationally efficient and physically interpretable is an equally pressing challenge. We developed a non-orthogonal, composition-based fingerprint rooted in chemical similarity through a generalization of the Pettifor scale, that encodes materials as vectors whose geometry reflects chemical relationships between elements [17]. Unlike one-hot encodings, which treat all elements as equally dissimilar, or black-box neural embeddings, our representation preserves chemical intuition: elements of the same group cluster naturally together, and chemically analogous compounds appear geometrically close. When combined with structural information and visualized via dimensionality reduction, the resulting maps correctly group known material families on ALEXANDRIA's convex hull, and reveal non-obvious but electronically meaningful proximities, such as Ge and SiC, but also a less obvious $NiSi_3P_4$, appearing near diamond silicon. This interpretability, alongside competitive prediction accuracy, makes the embedding a practical tool for hypothesis generation and for navigating vast chemical spaces in a human-understandable way.

Looking beyond static property prediction, the recent proliferation of universal machine learning interatomic potentials trained on millions of DFT calculations raises the question of whether these models are ready for more demanding physical quantities. A systematic benchmark of state-of-the-art potentials for phonon properties, including maximum phonon frequency, vibrational entropy, heat capacity, and free energy, yields an encouraging answer [18]. The best-performing models achieve impressing DFT-like accuracies for several thermodynamic quantities, suggesting that universal machine learning force fields have reached sufficient maturity for phonon calculations. However, performance varies substantially across models, and some widely used potentials still exhibit large systematic errors, underscoring that benchmark transparency and community standards are not optional but essential to progress.

Taken together, these results sketch both the promise and the honest boundaries of the current state-of-the-art. The accuracy of machine learning models is fundamentally limited by the quality and diversity of their training data; biases in chemical coverage, crystal symmetry distributions, and the accuracy ceiling of DFT itself all propagate into predictions. Addressing these limitations requires sustained investment in open, well-curated, and physically diverse datasets, alongside representation strategies that make learned models interpretable, transferable, and trustworthy for the field to move from collecting data to genuine materials design insights.

2.3 | Modeling Disordered Materials With Experiment-Constrained Atomistic Machine Learning

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With the introduction of highly flexible message-passing machine-learning interatomic potentials (MLIPs) that can learn potential energies close to the accuracy of the reference electronic-structure method (usually DFT), and scale favorably with respect to the number of species, there is a growing consensus among the community that the *short-range* atomistic ML modeling problem is essentially "solved". In addition, foundation models have lowered the barrier to training accurate models and improved the training data efficiency through the fine-tuning approach. Although the short-range atomistic modeling problem is compounded in practice by the existence of long-range interactions, which are currently being actively researched within the field, models that capture short-range physics are indeed sufficient for tackling many problems pertaining to the elucidation of the structure of materials, disordered materials in particular. Equipped with these powerful new tools to accurately model the potential energy surface (PES), one formidable problem remains: configurational sampling.

While most sampling techniques within atomistic modeling concentrate on approximating the partition function, and thus focus on finding thermodynamically stable (equilibrium) structures, many disordered materials are metastable and/or experimentally synthesized via non-equilibrium processes. These traditional sampling methods, like regular molecular dynamics (MD) and Monte Carlo (MC) approaches, notoriously fail at generating atomistic structural models of disordered materials with quantitative agreement with experiment, even when using state-of-the-art MLIPs, requiring expensive and/or specialized simulations matching the experimental conditions, which are only possible in selected cases [19]. Thus, experimental agreement is the next frontier in atomistic modeling of disordered materials.

The typical, traditional approach to enforce experimental agreement of simulation with experimental structure characterization data has been reverse MC (RMC) targeting available diffraction patterns, for example, X-ray (XRD) or neutron diffraction (ND). However, the mapping between structure and observable is injective, not bijective, that is, a structure has a single observable associated with it but a single observable may be compatible with multiple candidate structures. The consequence of this is that unconstrained RMC often leads to unrealistic structures which are high in potential energy. Hybrid RMC (HRMC) was introduced to address this issue by incorporating a physical constraint to the optimization, where only structures that improve experimental agreement and are low in potential energy are accepted, according to the Metropolis criterion [20]. While the idea of HRMC is very promising, quantitative accuracy at affordable computational cost has been lacking until the emergence of MLIPs a few years ago, which has hindered applicability of the method for real-life materials modeling problems.

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In the context of such inverse structure optimization methods and given the improved capabilities afforded by atomistic machine learning techniques, we proposed an implementation of HRMC which we referred to as the "modified-Hamiltonian" approach [21]. We used the method to infer the structure of oxygen-rich amorphous carbon using HRMC-based sampling with experimental X-ray photoelectron spectroscopy (XPS) as the target, thus going beyond the previous limitation of HRMC, which inferred structures solely from diffraction-derived data. Furthermore, the formulation of the modified Hamiltonian in terms of analytically differentiable functions of the atomic positions allowed us to generalize the notion of forces to include gradients of "experimental agreement", which we call "molecular augmented dynamics" (MAD) [22, 23]. MAD benefits from the linear-scaling (in the number of atoms) behavior of MD, which is superior to the quadratic scaling of MC methods, to unlock system sizes previously out of reach, even of our earlier, but still rather recent, work [21]. In addition, MAD can use *multiple* experimental constraints as long as the computational version of the experimental observable can be given in terms of differentiable atomic descriptors. Generalized barostatting, enabled by this addition of "experimental forces", allows for simultaneous optimization of the mass density and atomic positions to find configurations compatible with experiment.

As atomistic ML technology matures, and simulations of materials become more accurate and computationally efficient, simulations that were either impractical or *unthinkable* just a few years ago have now become possible and *imaginable*. Within the realm of atomistic ML simulations of real materials, we expect that the range of the possible will continue to expand in the years to come.

2.4 | Machine Learning You Can Trust

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Atomistic machine learning (ML) models have become instrumental to materials modeling and discovery, enabling large-scale simulations with an accuracy approaching that of first-principles methods at a fraction of the computational cost. In their practical deployment, however, it is crucial to recognize that the ML models are fundamentally interpolative, and therefore one must determine when and where their predictions can be trusted. Despite this, the majority of atomistic ML-assisted scientific results are still reported without any consideration of uncertainties in the model predictions. This lack of uncertainty awareness risks hindering overall scientific progress, particularly as the atomistic ML models become increasingly embedded into complex simulation workflows. This is in part due to the lack of easy-to-implement and computationally efficient approaches that make uncertainty quantification (UQ) readily accessible to the general atomistic ML practitioner.

To this end, we have devised two complementary methods for uncertainty quantification (UQ) in atomistic ML models: direct propagation of shallow ensembles (DPOSE) [24] and last-layer prediction rigidities (LLPR) [25]. DPOSE constructs and trains an ensemble of models at the last linear layer of the neural network (NN) model, drastically reducing the computational cost relative to conventional ensemble-based UQ schemes. LLPR applies the "prediction rigidity" formalism [26, 27] to the last-layer features of an already-trained NN model, requiring a computational cost equivalent to only a single additional training epoch during preparation. With no additional overhead at inference time, the LLPR approach allows for cheap and efficient UQ. The two approaches can be naturally combined into a best-of-both-worlds UQ strategy, in which the weights of the last-layer ensembles are sampled via the LLPR formalism (Equation (25) of [25]) and subsequently fine-tuned to produce an expressive UQ module. This hybrid framework is capable of capturing non-Gaussian error distributions, and allows uncertainties to be propagated to any downstream observable of interest.

Beyond UQ on global targets, prediction rigidities further provide quantitative assessments of the robustness of "raw" model predictions, namely those made at the local or component-wise level (the latter relevant, for example, in explicitly body-ordered or range-separated ML models) before aggregation into a global prediction. The local prediction rigidity (LPR) reveals degeneracies in training formulations that can lead to models with reasonably accurate global predictions yet arbitrary local predictions, which severely hinders their transferability outside the training domain [26]. Similarly, the component-wise prediction rigidity (CPR) detects unwanted information spillover between different prediction components, which undermines physical interpretability and prevents the model from fully exploiting the training data [27].

The proposed UQ workflows are readily generalizable across a broad range of model architectures and prediction targets, spanning ML interatomic potentials (MLIPs), models that directly learn first-principles observables (e.g., electronic density of states, charge density, etc.), as well as universal direct molecular dynamics propagators. A user-friendly implementation of these methods is provided in our open software suite metatrain [28], where the UQ module can be seamlessly combined with any of the available NN models and target quantities. By coupling methodological advances with accessible, community-driven software, we aim to lower the barrier to achieving uncertainty awareness in ML-assisted atomistic simulations. Ultimately, we envision UQ becoming a standard and indispensable component of atomistic ML, on equal footing with accuracy and efficiency, thereby ensuring that the integration of ML methods can be transparent, reliable, and scientifically meaningful.

2.5 | Machine Learning Exchange–Correlation Functionals: From Accuracy to Interpretability and Control

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Density functional theory (DFT) underpins modern computational materials science by enabling predictive simulations

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of electronic structure at manageable computational cost. Despite its success, the accuracy of DFT is fundamentally limited by the approximations used for the exchange–correlation (XC) functional, which encodes the many-body effects of electron–electron interactions. Traditional functional development relies on physically motivated ansätze constrained by known limits, but this strategy has struggled to deliver systematic improvements across diverse bonding environments and material classes.

Recent advances in machine learning (ML) offer a qualitatively different route: instead of proposing new analytic forms, ML-based XC functionals learn the functional dependence of the XC energy directly from data. In this framework, neural networks are trained on reference electronic-structure calculations to map the electronic density (and, in some cases, its derivatives or nonlocal descriptors) to XC energies and potentials. Early demonstrations showed that ML-XC functionals can surpass standard generalized gradient approximations and even hybrid functionals in selected systems, achieving chemical accuracy while retaining the favorable scaling of semilocal DFT [29]. More recent approaches leverage differentiable programming to train functionals end-to-end through the self-consistent Kohn–Sham cycle, and to use the Kohn–Sham equations as a regularizer that injects prior physical structure into the learning problem [30–32].

However, embedding ML into the XC functional raises challenges that go beyond those encountered in other areas of ML for materials, such as interatomic potentials. Unlike force fields, XC functionals enter self-consistently into the Kohn–Sham equations: errors in the functional affect not only total energies but also the electron density itself. This feedback loop makes ML-XC development uniquely sensitive to issues of stability, transferability, and physical consistency.

A central insight emerging from recent work is the importance of distinguishing functional-driven and density-driven errors in ML-based DFT. As demonstrated by Kim, Sim, and Burke [32] and highlighted by Medvedev et al. [33], high accuracy on fixed reference densities does not guarantee accurate self-consistent results. In some cases, ML-XC functionals that appear excellent in non-self-consistent tests perform poorly once the density is allowed to relax. This realization shifts the focus from fitting energies alone toward understanding how ML-XC approximations modify the Kohn–Sham fixed point and the resulting electronic structure.

Another key challenge concerns physical constraints and interpretability. While neural networks are highly expressive, unconstrained models can violate known exact conditions of DFT, such as sum rules, scaling relations, or correct asymptotic behavior. Current efforts aim to incorporate these constraints either explicitly—by enforcing them during training—or implicitly, through architecture design and carefully curated training data. Achieving the right balance between flexibility and physical structure remains an open problem, especially for systems far from those represented in the training set.

At the same time, ML-XC functionals open new opportunities that extend beyond incremental accuracy gains. By providing access to differentiable, data-driven representations of the XC functional, ML enables systematic analysis of how errors propagate through the self-consistent cycle and how different physical regimes challenge existing approximations. This perspective reframes functional development as a problem of learning and controlling approximations, rather than merely proposing better formulas.

Looking forward, several directions appear particularly promising. First, coupling ML-XC development with high-quality, diverse training datasets—including strongly correlated systems—will be crucial for generalization. Second, integrating ML-XC functionals into established electronic-structure codes in a transparent and reproducible way is essential for community adoption. Finally, combining ML-based functionals with diagnostic tools that explicitly monitor density-driven errors may provide a pathway toward adaptive or system-aware DFT approximations.

In this sense, ML-XC functionals represent not only a technical advance but also a conceptual shift in electronic-structure theory. They force us to confront long-standing questions about what limits DFT accuracy, how errors arise and propagate, and how data-driven methods can complement—rather than replace—physical insight.

2.6 | Accelerated Materials Property Prediction With Quantum-Chemical Bonding Analysis and Machine Learning Interatomic Potentials

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The search for new battery and thermoelectric materials frequently employs ab initio property simulations as a key tool for predicting structural and functional characteristics. In recent years, machine learning models have increasingly complemented—and in some cases replaced—traditional ab initio approaches. Among these, machine learning interatomic potentials (MLIPs) stand out for their versatility, as they enable the prediction of energies, forces, and stresses directly from materials structures. Despite the emergence of foundation models such as MACE-MP-0 [34], significant challenges remain.

For instance, current foundation models often require fine-tuning—or even retraining from scratch—to achieve sufficient accuracy for specialized tasks, such as modeling amorphous phases. Another major limitation of current MLIPs is their lack of interpretability in terms of chemical bonding concepts, such as bond orders derived from quantum-chemical analysis. This makes it challenging to anticipate failures or relate predictions to established chemical knowledge. Representations based on bonding descriptors—such as Crystal Orbital Hamilton Populations (COHPs) or Crystal Orbital Bond Indices (COBIs)—may offer a more chemically intuitive approach. For instance, during MLIP training, these quantities could be incorporated as edge attributes that encode bond strength or bonding character. Such chemically informed edge attributes could supplement purely distance-based descriptors, enabling the model to learn

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interactions within a representation space that also reflects the underlying bonding characteristics. This approach may prove particularly advantageous in cases where similar geometries correspond to different bonding situations. Prior work using natural bond orbital (NBO) analysis data on transition-metal complexes indicates this [35].

Developing automated simulation workflows can help streamline both bonding analysis and data generation for MLIPs. These workflows, implemented in a workflow language and executed via a manager, ensure scalability and reproducibility. While multiple workflow languages exist, we rely on the Materials Project infrastructure and the atomate2 library [36], which uses jobflow for workflow definition and jobflow-remote for execution. Using the atomate2 framework, we implemented two automated workflows to address these needs.

The first workflow we developed enables high-throughput bonding analysis by computing COHPs and COBIs. A pilot version of this workflow was implemented in its predecessor, atomate, and was used to generate a quantum-chemical bonding database covering several thousand materials [37]. This data can be transformed into descriptors (features) ready for use in predictive machine learning models. Our recent work demonstrates that incorporating such descriptors into model training pipelines not only improves predictive accuracy but can also help uncover intuitive relationships between bonding and material properties [38]. While these initial efforts focused on their use as descriptors in tabular form, we anticipate that they can also play an essential role in graph-based representations.

The second workflow provides a framework for automated training and fine-tuning of MLIPs [39]. In its first applications, the framework was successfully applied to systems including titanium–oxygen, SiO_2, crystalline and liquid water, and phase-change memory materials [39]. The approach employs an iterative random-structure search: new structures are generated, down-selected, labeled, and used to fit a potential. After one generation, the structures are optimized using the previous generation's potential, and the optimization trajectories are also used within the training of the next potential. All structures are ultimately labeled with ab initio calculations.

With the implementation of these two automated workflows, we are well-positioned to assess whether the accuracy of MLIP can be further improved through the training of chemistry-aware MLIPs, which may yield increased reliability and interpretability. Nevertheless, a thorough evaluation remains crucial to ascertain the degree to which it provides consistent improvements.

2.7 | Bridging the Digital-Physical Divide: From Virtual Screening to Autonomous Discovery

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The discovery of functional organic materials has historically been a problem of scale. For the past decade, the dominant strategy has been High-Throughput Virtual Screening (HTVS) [40]. By funneling large libraries of commercially available compounds through quantum-mechanical calculations, we have successfully identified rare "needles in a haystack" [41]. These are molecules exhibiting exotic photophysical properties simply by virtue of their existing electronic structure; such as singlet fission, thermally-activated delayed fluorescence, non-fullerene acceptors and inverted singlet-triplet gaps.

A prime example of this approach is our recent work on "anti-Kasha" fluorophores [42]. These are rare molecules that violate standard photophysical rules by emitting from higher excited states, which is a property highly sought after for photonic applications, such as sensing. By screening a massive library of existing compounds and rigorously filtering for specific energy gap criteria, we successfully identified and experimentally validated two new anti-Kasha emitters. This work highlights the critical importance of bridging the digital-physical barrier because it is not enough to predict theoretical candidates.

However, while effective, this chemical-database screening approach is inherently limited by what has already been synthesized and cataloged. To transition from finding materials to designing them, we are now witnessing a shift toward AI that builds its own search space.

The first major leap in this new regime is the application of LLMs to molecular science. We demonstrated that a fine-tuned GPT-3 model could accurately predict complex electronic properties, such as the HOMO to LUMO gap and dipole moment, by treating chemical strings (SMILES) as a language [43]. Unlike standard regression, this model learns "chemical intuition" and maintains predictive accuracy even when input data is noisy or incomplete. This opens the door to inverse design where a model generates novel molecular structures that meet specific optoelectronic criteria rather than screening existing ones.

The second leap is in predictive scale. Reliable prediction of excited-state properties usually requires expensive time-dependent density functional theory (TD-DFT) calculations. To circumvent this, we have developed a "Delta-Machine Learning" (Δ-ML) protocol [44]. By training models to predict the difference between a cheap, low-level calculation (like ZINDO) and a high-level result (TD-DFT), we can approximate high-level accuracy at a fraction of the cost. This allows us to map the excited-state landscape of millions of molecules, which is orders of magnitude more than traditional HTVS, and expands the accessible chemical space significantly.

These technologies are now converging across academia and industry, that is, in platforms like DiaDEM. Moving beyond simple automation, researchers are employing agentic AI to construct and manage complex computational workflows autonomously, also helping bring predictive power into the hands of the non-expert. In combination, AI can orchestrate the screening of millions of candidates using Δ-ML protocols and integrate directly with molecule-generating models to explore novel chemical spaces with little intervention and a direct link to autonomous labs, which are rapidly gaining traction. By coupling these autonomous digital agents with experimental feedback loops, we are shifting the paradigm from brute-force screening to intelligent, generative discovery.

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2.8 | From Simulation to Screening: How Federated Databases, Machine Learning, and Workflow Automation Are Transforming Materials Discovery

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The landscape of materials informatics has undergone rapid expansion, and its toolkit is growing at an unprecedented pace. This evolution builds on three foundational pillars: simulations, databases, and machine learning (ML) algorithms. Researchers now have access to a diverse array of simulation software that enables physical modeling of materials across scales and formalisms. Pushed by the progress in computing power, this capability has facilitated the creation of extensive and curated databases that are accessible via dedicated platforms or APIs. Inspired by the broader success of ML, materials science has seen the development of specialized algorithms tailored to its unique challenges in a community-driven approach. The next frontier lies in integrating these tools synergistically to fully exploit the exciting opportunities of this data-driven transformative shift.

Workflow automation and standardization have emerged as key players in this endeavor. Workflows bridge the gap between the three pillars, automating processes and simplifying access to collective expertise while abstracting technical intricacies. A notable example is the Python package *atomate2*, which offers a comprehensive library of materials workflows, ranging from density functional theory (DFT) calculations and structure optimization to molecular dynamics simulations with pre-trained machine learning interatomic potentials [36]. This user-friendly tool streamlines high-throughput computations by managing input generation, output storage, and database formatting, all while accommodating multiple software in a single workflow. Concurrently, the adoption of standards is becoming indispensable for seamless data and tool interoperability. Although establishing these frameworks demands significant community effort, their value is undeniable: standards ensure efficient and secure navigation across diverse data sources. The *OPTIMADE* consortium exemplifies this progress, as it has standardized the representation and querying of over 30 million crystal structures and their properties across more than 20 providers, many of which are hypothetical materials generated through ML-based approaches [45].

The efficacy of these tools has been demonstrated in the search for new optical and nonlinear optical (NLO) materials which are critical components in laser technology, biomedical imaging, and quantum information processing [46, 47]. However, the vastness of chemical and structural space, coupled with the need to balance competing material properties, has historically presented a challenge. By training ML models to predict properties such as the refractive index (linear optical regime) or the second-harmonic generation (SHG) coefficient (NLO regime), researchers can efficiently screen a vast pool of candidates to identify materials that meet multi-objective optimization criteria, that is, maximizing the optical performance in a given spectral range. The initial candidate pool is sourced via the *OPTIMADE* API and can be periodically updated with the same query as the federated databases grow. Promising candidates are then evaluated using *atomate2* DFT workflows for rigorous validation.

A persistent challenge is the scarcity of large, curated, and openly accessible datasets for specific target properties, such as SHG. To address this, researchers can initiate the process using a limited dataset [48] and employ active learning to iteratively refine the ML model while acquiring informative or optimal data with minimal computational resources. This approach (training, predicting, selecting, annotating, and repeating) proves more efficient in data acquisition than one-shot methods, even when the initial model is accurate. Further enhancements are achievable through multi-fidelity learning, where abundant low-accuracy data is leveraged to improve predictions for high-accuracy targets with limited training data. Even simple techniques, such as correction learning, which aims at predicting the difference between high- and low-fidelity data, have shown great results.

The convergence of federated databases, ML, and high-throughput computation is reshaping materials discovery. By leveraging standardized open data, iterative learning, and advanced modeling, researchers can navigate the intricate landscape of materials space with unprecedented speed and precision. The future of materials science lies not only in uncovering new compounds but also in doing so more efficiently and collaboratively than ever before.

2.9 | Uncertainty-Aware Machine Learning for Quantitative Electron Energy-Loss Spectroscopy

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The rapid development of machine learning (ML) and artificial intelligence (AI) methods in recent years has opened new avenues for extracting quantitative physical information from spectroscopic data in advanced materials. In the specific case of electron energy-loss spectroscopy (EELS), which provides nanoscale access to electronic structure, dielectric response, and collective excitations, these advances have been particularly impactful. They enable a fuller exploitation of EELS measurements, whose information content is often constrained by noise, background modelling, and the need for robust uncertainty quantification. Recent work has demonstrated that physics-informed and uncertainty-aware ML frameworks can transform EELS from a largely qualitative probe into a quantitative, statistically controlled tool for materials discovery.

Applications of ML to EELS have largely focused on denoising, clustering, and automated feature recognition, typically treating the background subtraction and uncertainty estimation as preprocessing steps. A conceptual shift was introduced in [49, 50] by reframing spectroscopic analysis itself as an inference

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problem. In the low energy loss regime, MC replica techniques combined with neural networks enable bias-free removal of the zero-loss peak background, allowing the extraction of dielectric functions and band gaps with faithful uncertainty propagation. This approach reveals subtle spatial variations in low-dimensional materials that were previously inaccessible due to the dominant background in the low-loss region. Importantly, these studies demonstrate that ML models can learn physically meaningful spectral representations while preserving a rigorous statistical interpretation of both signal and background contributions. Crucially, the methods of [49, 50] are validated through rigorous statistical closure tests using synthetic data generated from known underlying physical laws.

Subsequent work extended these ideas to increasingly complex spectroscopic regimes and to other material platforms. By integrating physics-based constraints directly into the learning process, ML models were shown to interpolate and extrapolate spectral behavior in a controlled manner. This strategy is essential when dealing with regions of the spectrum where training data are sparse or absent, a situation that arises naturally in EELS due to overlapping excitations and limited energy windows. Applications of this approach to excitonic features [51] and to band gap modulations in low-dimensional van der Waals systems highlighted how uncertainty-aware ML can distinguish genuine physical signals from artifacts induced by noise or fitting bias, enabling new insights such as the heterostrain-driven bandgap increase in twisted WS_2 [52]. Likewise, this approach can be seamlessly extended to the energy-gain region [53], where many interesting signals of the underlying material arise.

A major remaining challenge lies in the core-loss regime of EELS, where quantitative analysis critically depends on accurate background subtraction. Conventional parametric background models provide single best-fit estimates and offer limited insight into uncertainty, especially when extrapolating into the ionization-edge window. Ongoing work, building on low-loss case, aims to address this limitation by introducing an uncertainty-aware ML framework that reformulates core-loss background subtraction as a statistically controlled inference problem, analogous to the. The method combines Monte Carlo replica generation, neural network ensembles trained on the pre-edge region, and physics-informed extrapolation constraints that enforce physically consistent behavior at the edge onset. This framework yields pixel- and energy-resolved probability distributions for the inferred background, enabling direct propagation of uncertainty to observables such as white-line intensities. Application of this approach to twisted CrSBr demonstrates its power in resolving spatially modulated core-loss signals associated with moiré superlattices, which reveal stacking-correlated variations in the white-line intensities. This result confirms previous findings that uncertainty-aware ML does not merely improve numerical precision but fundamentally changes the type of subtle electronic responses that can be identified in advanced functional materials.

Despite this progress, important challenges remain in uncertainty-aware ML for EELS. Computational cost, interpretability of learned representations, and the integration of prior physical knowledge across multiple length and energy scales are active areas of development. More broadly, the community must continue to move beyond binary estimates toward probabilistic descriptions of spectroscopic observables, particularly as datasets grow in size and complexity.

Looking ahead, a natural next step is the integration of Neural Simulation-Based Inference (NSBI) techniques to extract underlying physical parameters of moiré van der Waals materials directly from spectroscopic data. By combining uncertainty-aware EELS analyses with NSBI frameworks, one can envision end-to-end pipelines that connect raw spectra to physically meaningful parameters, such as interlayer coupling strengths, local dielectric screening, or symmetry-breaking fields, while preserving rigorous uncertainty estimates. Such approaches would be particularly powerful for moiré systems, where small structural variations often lead to major changes in their optical and electronic responses. Embedding NSBI within ML-driven spectroscopic workflows offers therefore a promising route toward quantitatively linking structure, spectroscopy, and emergent functionality in complex quantum materials.

2.10 | Toward Data Engineering and Model Selection in Predictive Regression of Advanced Materials Properties

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Intensive use of machine learning (ML) in the inference of materials properties requires consistency of datasets to guarantee a robust feature engineering and a reliable model tuning. Existing datasets on properties of 2D materials and others emerging materials, which have been released based on the theoretical simulations through high-throughput DFT computations, have been served as consistent input data for ML workflow [54]. However, the amount of data remains limited compared to massive labelled data for large models like computer vision or large language models. In this context, the construction of descriptors and the feature transformation have been developed not only to generate additional data for data-driven workflows but also to better represent the characteristics of materials. The construction should be designed so that used ML algorithms are able to load the data and extract their common patterns with respect to the target properties.

In the supervised regression of materials properties, constructed descriptors regardless the choice of models in the framework of feature engineering have been developed in several works. A straightforward way is to generate additional descriptors with respect to atomic structure or elemental atoms of compounds [55, 56]. The main results have shown efficient training and prediction of materials properties with the help of universal fragment descriptors and a rapid scanning of potential ferromagnetic 2D materials candidates based on crystal graph multilayers for spintronic applications. Practically, the generated descriptors enabling to capture the relevant features of materials may be transformed to matrix or graphical entries before feeding the ML pipeline [57].

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Physical and chemical relationships between descriptors and target properties are not consistently easy to access owing to technical constraints in design and construction of descriptors. Furthermore, the generalized aspect of descriptors with respect to studied datasets and ML algorithms has been fairly scrutinized. Therefore, gathering criteria in terms of homogeneity of datasets, ML algorithms, appropriate descriptors with respect to target properties remains challenging in the regressive ML of materials properties. In this perspective, a holistic analysis of ML performance for advanced materials should be performed by considering mutual interactions between the primary building blocks of a ML workflow, that is, datasets, descriptor engineering and model selection. There is still a limited number of studies that unveil the nested relationships of these building blocks. Such a study could provide a comprehensive picture on the trusted ML performance on the data-driven prediction of materials properties. A recent work has attempted to provide some insights into this by investigating the ML performance within the descriptor engineering on different dataset characteristics of defect formation energy in 2D materials [58]. This study enables a fair comparison of diverse ML algorithms when applying the descriptor engineering on two datasets with varying numbers of entries and distributions of the target property. As a whole, the descriptors help to enhance the ML training for both neural networks and statistical algorithms across the considered datasets. The results have emphasized that back propagation-based neural networks interact with descriptors in different manner as compared to statistical algorithms. The neural networks exhibit a poorer training for the considered datasets, but they have shown a higher stability in performance regarding input data scaling. By applying the same descriptor design to different datasets, the study has also indicated the efficiency and the generalized aspect of the developed descriptors despite their low dimensions compared to the aforementioned descriptors.

Further investigations of other types of materials datasets would support the future development of ML for materials science. This opens rooms for universal design of descriptors and generalized models with the aim of accelerating the scanning and property analysis of advanced functional materials. Another aspect in supervised ML is data labeling which may be extended to experimental data. Since ML works for materials science have mainly relied on datasets generated by theoretical simulations, data-driven ML on property-labelled experimental datasets would be of great interest for several purposes, for instance benchmarking, discovery of new and relevant patterns related to practical scenarios.

2.11 | Composable Operator Learning for Large-Scale Electronic Structure

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First-principles electronic-structure methods underpin much of modern materials science, yet a fundamental mismatch persists between the length and time scales accessible to quantum-mechanical simulations and those relevant for many technologically important phenomena. Despite its comparatively favorable scaling, density functional theory (DFT) remains computationally prohibitive for simulations reaching mesoscopic or macroscopic regimes, or for dynamical processes extending beyond nanoseconds. Consequently, phenomena such as dielectric response in amorphous networks, charge transport in disordered materials, phase transitions, and structure–property relations governed by medium-range correlations remain largely inaccessible to direct first-principles modeling.

Machine-learned interatomic potentials (MLIPs) have partially bridged this gap by enabling near-DFT-accurate molecular dynamics at unprecedented scales. Descriptor-based kernel methods, systematically improvable many-body expansions, atom-centered neural networks, and equivariant message-passing architectures now allow simulations involving millions of atoms while faithfully reproducing forces and energies [59]. However, MLIPs alone are insufficient when the target observables are intrinsically electronic. Quantities such as electronic spectra, dielectric functions, and transport coefficients cannot be reliably inferred from structure alone without explicit quantum operators.

This limitation motivates an operator-centric paradigm in which machine learning is used to infer quantum-mechanical operators, most notably the Kohn–Sham Hamiltonian and related response operators, directly from atomic structure. Once learned, these operators become reusable and composable primitives, enabling efficient evaluation of diverse electronic observables through standard linear-algebra or polynomial-expansion solvers on large, realistic systems. While deep neural networks trained directly on Hamiltonians or derived observables can achieve high accuracy, they often require large datasets, offer limited interpretability, and show reduced transferability. An emerging consensus is that embedding physical structure into ML models, most notably symmetry and equivariance, substantially improves both accuracy and data efficiency. In particular, SO(3)-equivariant tensorial representations aligned with quantum mechanics have demonstrated clear advantages across chemical and structural diversity [60, 61].

These insights motivate a shift from purely numerical surrogates toward physics-informed learning architectures that balance expressivity with transparency, modularity, and scalability. In this context, learning frameworks that prioritize interpretability and systematic improvability are especially appealing. Hamiltonian matrix elements admit a formal expansion into one-, two-, and higher-center contributions. Building on tight-binding theory and bond-order concepts, higher-order terms can be expanded as products of lower-order interactions, enabling controlled increases in fidelity while retaining analytical tractability [62]. Angular dependencies are treated analytically, ensuring exact rotational equivariance. Crucially, such analytical construction can be made linear in its parameters, allowing efficient least-squares training and resulting in high data efficiency, stability, and interpretability.

Preliminary applications to disordered materials, such as amorphous boron nitride, illustrate the promise of this approach. Training on a limited set of amorphous configurations yields

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Hamiltonians with near-DFT accuracy that generalize seamlessly to crystalline phases excluded from training, demonstrating the extraction of transferable electronic motifs from sparse data.

Beyond Hamiltonian prediction, this framework points toward a broader vision: a composable quantum operator inference architecture. In this paradigm, specialized modules infer individual quantum operators, such as Hamiltonians, velocity operators, or response kernels, from atomic structure and can be composed into higher-level models for emergent properties. For example, combining learned Hamiltonian and velocity operators enables direct prediction of optical and transport response functions, bridging microscopic quantum mechanics and macroscopic functionality. More broadly, scalable, physics-informed operator inference offers a principled pathway to extend quantum-mechanical modeling into regimes traditionally accessible only to empirical or coarse-grained methods, enabling predictive simulations of complex functional materials across previously inaccessible length and time scales.

2.12 | AI-Enabled Multiscale Optimization for Functional Materials

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Multiscale modelling has long been recognized as a central challenge in computational materials science. The difficulty lies not only in the computational cost of linking atomistic, electronic, and device-level descriptions, but also in the breadth of expertise required at each scale. Atomic structure prediction, electronic structure theory, and transport or response calculations are traditionally treated as distinct problems, often addressed by different communities using separate tools. Integrating these layers into a unified and automated workflow remains an open challenge, particularly when the objective is not only simulation but optimization.

This challenge becomes more acute in bottom-up materials optimization. Optimization workflows typically rely on repeated evaluations of an objective function in order to explore a high-dimensional design space. When the target quantity involves advanced functional properties, such as carrier mobility, dielectric response, or spin–orbit torque efficiency, each evaluation may require a full quantum-mechanical calculation. Performing this process iteratively and at scale is generally impractical with conventional first-principles approaches.

Heuristic optimization strategies, such as genetic algorithms, offer a pragmatic alternative [63]. By avoiding explicit derivatives, they are well suited to complex and mixed parameter spaces and have been successfully applied to materials discovery problems. Their main limitation, however, lies in representation. Any genetic algorithm requires a compact and expressive encoding of the system under optimization, commonly referred to as the genotype, from which the physical structure, or phenotype, can be reconstructed efficiently. Moreover, the objective function must be computable in a fully automated manner, as even minimal human intervention breaks the optimization loop.

Recent advances in machine-learning interatomic potentials address part of this challenge [59]. Modern equivariant models can reproduce first-principles potential energy surfaces with high accuracy at a fraction of the computational cost, enabling efficient structural relaxation and large-scale sampling. These approaches have matured to the point where they can be considered a reference tool for atomic-scale exploration, rather than a mere approximation.

The construction of the electronic Hamiltonian, however, remains a critical bottleneck. While plane-wave methods offer systematic accuracy, they scale poorly and are ill suited to large, disordered, or low-symmetry systems. In contrast, localized atomic-orbital frameworks, such as those implemented in SIESTA or OpenMX, naturally lead to sparse Hamiltonians whose structure is closely tied to atomic positions. This locality is essential for scalability and for subsequent transport or response calculations.

In this context, recent progress in symmetry-aware machine learning offers a promising route forward that exploit first principles based on linear combination of atomic orbitals. Equivariant graph neural networks have demonstrated the ability to learn not only scalar quantities but structured objects that obey physical symmetries. Extending these ideas to the direct prediction of electronic Hamiltonians provides a natural analog to machine-learning interatomic potentials, but at the electronic level [64]. By preserving rotational and translational equivariance and maintaining locality, such models can generate sparse Hamiltonians suitable for large-scale simulations.

This capability is particularly relevant for linear-scaling quantum transport methodologies, which rely on sparse Hamiltonians to compute electronic and spintronic response functions in systems containing millions of degrees of freedom [65]. Once an accurate Hamiltonian is available, quantities such as conductivity, spin Hall response, or spin–orbit torque can be evaluated efficiently, without explicit diagonalisation. The combination of automated Hamiltonian construction and linear-scaling solvers therefore enables end-to-end workflows that were previously inaccessible.

We have demonstrated that this integration can be realized in practice by combining equivariant graph neural networks with automated quantum transport calculations. In particular, spin–orbit torque efficiencies can be computed without manual intervention and directly used as objective functions within a genetic algorithm framework. This approach enables genuine bottom-up optimization of functional properties, rather than post hoc screening of predefined structures, and allows the exploration of material configurations that would otherwise be computationally prohibitive.

More broadly, these developments point toward a new paradigm for AI-driven materials design [1]. Rather than replacing physics-based models, machine learning acts as a connective tissue between scales, enforcing symmetry, reducing cost, and enabling automation. The key opportunity lies not in isolated models, but in the coherent integration of structure generation, electronic modelling, and functional evaluation. Achieving this integration will be essential for transforming multiscale optimization from a conceptual ambition into a practical tool for application-driven materials discovery.

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2.13 | Machine Learning for Powder X-Ray Diffraction

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Powder X-ray diffraction (PXRD) is a powerful, non-destructive technique widely used in materials science to identify crystalline phases, determine crystal structures, and assess the purity and composition of powder samples. Its ability to provide rapid, high-resolution data makes it indispensable for the characterization of new materials, the study of phase transitions, and the optimization of synthesis processes. Over the past few decades, the volume of PXRD data has surged, driven by advancements in laboratory diffractometer technology and the growing use of *in situ* and *in operando* experimental setups—particularly at synchrotron beamlines. This explosion of PXRD data—combined with the accessibility of online crystal structure databases—has created the ground for the application of ML approaches by chemists, crystallographers, and data scientists. The ability to rapidly simulate powder PXRD patterns from Crystallographic Information Files (CIFs) sourced from databases like the ICSD, CSD, COD, or Materials Project provides rich and diverse datasets. This capability is particularly valuable for training and applying ML models to analyze, predict, and interpret diffraction data efficiently.

In recent years, PXRD has been increasingly coupled with supervised and unsupervised machine learning. Among the most common problems addressed using ML is the determination of crystal systems and/or crystallographic space groups [66]. More recently, the determination of structure types has also been considered [67]. Deep learning models based on Convolutional Neural Networks approach are most often used to solve such classification problems and have shown relatively high accuracies. To go beyond such classification, more recent works have focused on generative AI models to solve the crystal structures. Such interesting approaches have demonstrated its potential with the successful structure determination of few unknown inorganic compounds. For multiphasic samples, ML models have also been used with the objective of automatizing the data analysis. With this purpose, CNN models have been constructed to carry out phase identification and quantification in an autonomous manner [68]. In addition, unsupervised ML methods such as clustering trough Principal Component Analysis (PCA) and non-negative matrix factorization (NMF) have been used to quickly identify the single-phase domains in series of PXRD patterns collected in synchrotron [69].

Owing to the availability of extensive crystallographic databases—such as the Inorganic Crystal Structure Database (ICSD), with over 300,000 entries, and the Cambridge Structural Database (CSD), containing more than 1.2 million structures—machine learning (ML) models could be trained on vast datasets of simulated XRD patterns derived from known crystal structures. However, critical aspects of data curation are frequently overlooked. For instance, these databases often contain duplicate or highly similar entries (e.g., the same crystal structure determined at similar temperatures or isostructural analogs), which can lead to data leakage and artificially inflate model accuracy. Another significant challenge arises when ML models trained on simulated patterns are tested on experimental PXRD data, often resulting in diminished performance. To address this, various data pre-processing techniques have been proposed, such as adding experimental noise to simulated patterns or denoising experimental data. Yet, these approaches struggle to accurately reproduce complex experimental parameters—such as preferential orientation or specific crystallite shapes—during data augmentation, limiting their effectiveness.

ML holds significant promise for the analysis of powder PXRD data, particularly in high-throughput characterization of multiphasic samples and in assisting in the structure determination of unknown materials. While CNNs have been the most widely used approach for these tasks, emerging generative machine learning models based on diffusion processes are gaining increasing attention, as they have demonstrated the ability to solve crystal structures from noisy diffraction patterns acquired in real world experiments [70]. Furthermore, innovative data preprocessing techniques such as dimensionality reduction and data augmentation are essential to address the persistent challenge of training models on simulated PXRD datasets while ensuring robust performance on experimental PXRD data.

2.14 | Wannier Functions as a Bridge for Artificial Intelligence in Advanced Functional Materials

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Recent advances in computational materials science—particularly those driven by machine learning—have led to an unprecedented expansion of predicted crystal-structure libraries. Translating this structural abundance into functional understanding, however, remains a central challenge. The relationship between crystal structure and material properties can be highly nontrivial—especially for electronic, topological, optical and magnetic properties—and is often governed by subtle features of the electronic structure, which demand high physical and computational accuracy. An accurate and compact mapping from crystal structure to electronic structure is thus often required to bridge large-scale databases and the corresponding physical properties. Such a representation must be low-dimensional to enable efficient storage and manipulation, yet sufficiently accurate for studying a broad range of materials properties.

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Maximally localized Wannier functions (MLWFs) [71] provide an ideal framework to meet these requirements. By restoring the chemical intuition of localized orbitals in crystalline solids—often resembling atomic s/p/d/f orbitals or bonding and antibonding states—MLWFs yield a minimal and physically transparent basis that represent an exact mapping of the initial Bloch states. In the MLWF representation, quantum-mechanical operators such as the Hamiltonian and position operators are expressed using a minimum number of orbitals with rapidly decaying real-space matrix elements. This compactness enables efficient and essentially lossless Fourier interpolation of many electronic-structure quantities [72]. More broadly, MLWFs serve as an intermediate representation that decouples the complexity of property calculations from the underlying electronic-structure methods, thereby enabling ultra-fine Brillouin-zone integrations for novel materials properties that were previously computationally prohibitive, including topological properties involving Berry curvature and electronic transport considering electron–phonon interactions.

Despite these advantages, the practical construction of MLWFs has traditionally relied on substantial user input, chemical intuition, and manual tuning of parameters, limiting their applicability in high-throughput settings. To over- come these challenges, we have developed two automated Wannierization algorithms in recent years. Projectability- disentangled Wannier functions (PDWFs) provide an accurate description of the low-energy electronic structure for both metals and insulators [73]. On top of this, manifold-remixed Wannier functions (MRWFs) further decompose PDWFs into two independent subspaces spanning the fully occupied valence manifold and the unoccupied conduction manifold [74].

Using these automated approaches, we showed that it is possible to systematically investigate the electronic structures of large sets (22085 entries) of unique, stoichiometric, and experimentally known inorganic crystals from the Materials Cloud 3D structure database (MC3D) [75], encoding these in approximately 2 million MLWFs. Such strategy enables a "compute-once, analyze-many" paradigm: the electronic structure is evaluated a single time, after which diverse materials properties can be explored efficiently in a post-processing fashion. As a result, the computational cost of high-throughput materials discovery is dramatically reduced, and one can apply this approach to search for compound that have, for example, optimal thermoelectric figure of merit, large Bardeen–Cooper–Schrieffer superconducting temperature, large nonlinear Hall responses, or extreme effective masses, to name only a few [76].

Beyond targeted studies, a MLWF-based framework is inherently scalable, enabling exploration of both larger experimental/theoretical structural databases and a broader spectrum of physical properties. In addition, these MLWF databases can serve as a rich training set for machine learning models, facilitating rapid predictions of electronic structures and derived properties across vast chemical spaces. We anticipate that the widespread adoption of Wannier-based methodologies, in close synergy with experimental validation, will play a pivotal role in accelerating the discovery of next-generation functional materials.

2.15 | A Closed-Loop Framework for AI-Driven Growth and Transformer-Based Spectral Analysis of 2D Materials

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The scalable synthesis of two-dimensional (2D) materials remains one of the central bottlenecks for their technological deployment. Despite remarkable advances in conventional chemical vapor deposition, many 2D systems still lack reproducible, automated routes that can guarantee uniform crystallinity, controlled layer number, and minimal defect density. Artificial intelligence (AI) has recently emerged as a powerful approach to transform materials synthesis, enabling autonomous experimentation loops capable of navigating high-dimensional parameter spaces and learning optimal growth protocols directly from experimental feedback. In parallel, AI-enhanced characterization methods are beginning to overcome the limitations of traditional spectral analysis [77], especially in systems where the signatures of different structural phases are strongly overlapping.

A representative example of this emerging paradigm is the adaptive Monte Carlo (aMC)-driven synthesis framework we recently demonstrated for epitaxial graphene on silicon carbide (SiC) [78]. In this approach, an artificial neural network (ANN) learns a time-dependent temperature protocol for SiC graphitization without any prior knowledge of effective growth recipes. Each iteration of the autonomous loop generates a new thermal profile, executes the synthesis, evaluates the resulting material via Raman spectroscopy, and updates the ANN based on a score function sensitive to graphene's 2D-peak intensity and FWHM. Unlike optimization strategies restricted to static parameters, this method directly learns dynamic synthesis protocols, allowing the system to discover physically non-intuitive trajectories that improve material quality within only a few tens of experimental cycles. The result is a fully closed-loop growth paradigm in which the synthesis "brain" adapts, potentially in real time, to hardware-level feedback.

While our initial work targeted monolayer graphene growth, the same methodology naturally extends to the synthesis of epitaxial buffer-layer graphene, a material whose importance has been revitalized by recent evidence that a well-ordered buffer layer constitutes a genuine wide-gap 2D semiconductor with room-temperature mobilities exceeding 5000 $cm^2\ V^{-1}\ s^{-1}$ [79]. The challenge, however, lies in its spectral detectability: the Raman response of the buffer layer is extremely weak and is almost entirely masked by the dominant SiC substrate features [80]. As a result, conventional background-subtraction approaches become unreliable, as they require acquisition of a reference SiC spectrum under *identical* experimental conditions: identical focus, identical laser intensity, identical exposure time and confocal alignment. In practice, even small variations in spot size, focus conditions or local surface morphology introduce intensity and baseline mismatches that compromise direct subtraction. Furthermore, the Raman response of SiC itself is not uniform, exhibiting variations across a single wafer as well as differences between wafers arising from local structural,

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morphological, and processing-related heterogeneities. Consequently, obtaining a truly representative reference spectrum becomes challenging, making conventional subtraction procedures cumbersome and often unsuitable for quantitative fast feedback.

To overcome this limitation, we introduce Transformer-based spectral decomposition, which acts as an AI-enhanced set of "eyes" for the aMC optimization loop. Transformer encoders, equipped with self-attention layers [81], learn to recognize the SiC contribution directly within mixed spectra and subtract it with high fidelity, without requiring a separately measured reference spectrum. By training on curated datasets of pristine SiC spectra and mixed buffer-layer/SiC spectra, the model learns a latent representation of the substrate background that is far more flexible and robust than any analytic multi-peak fitting. This approach enables the reliable extraction of the weak buffer-layer signature, establishing a viable feedback metric for the same closed-loop optimization strategy previously demonstrated for monolayer graphene.

In this combined framework, AI controls both synthesis and metrology: the aMC engine suggests new growth trajectories, while the Transformer provides physically interpretable, noise-robust spectral descriptors to evaluate each outcome. This dual-AI workflow significantly accelerates convergence in regimes where the spectral contrast is intrinsically low or where multiple phases coexist, as in buffer-to-graphene transitions or the formation of SiC-induced superstructures. The approach is consistent with the broader vision outlined in recent AI-materials perspectives, where autonomous synthesis, foundation models, and high-fidelity characterization are expected to merge into self-driving laboratories for advanced functional materials.

Beyond epitaxial graphene, the methodology is generalizable to heterostructures, doped buffer layers, and intercalated systems, where spectral complexity increases and traditional analytics struggle. The combination of dynamic protocol learning and Transformer-grade spectral intelligence points toward a future in which synthesis optimization and materials characterization co-evolve autonomously, reducing human intervention to high-level supervision and interpretation.

2.16 | Foundation Models for Microscopy Images

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Foundation models have revolutionized various fields of artificial intelligence by providing pre-trained models with very strong generalization capabilities. For microscopy image analysis, these models have significant potential to improve performance and efficiency. However, due to the unique characteristics of microscopy data, it is difficult to effectively train and deploy these types of models.

Off-the-shelf vision foundation models are trained on vast amounts of natural images. While they perform well on in-distribution data, their performance often degrades when applied to specialized microscopy datasets. Models like SAM [82] have shown impressive results in various segmentation tasks and even in some microscopy applications, yet the final performance often leaves much to be desired, as boundaries between objects can be smeared or interesting objects with low contrast are missed. Training vision foundation models from scratch for microscopy image analysis also presents challenges. Firstly, the availability of large-scale annotated microscopy datasets is limited compared to natural image datasets like ImageNet [83]. Getting high-quality annotations for microscopy images requires expert knowledge, making collection and annotation a costly and time-consuming process. Secondly, the objects of interest in microscopy images are often very small compared to the image size, or are very sparse, making it difficult for models to learn robust representations without very large amounts of data, extensive data augmentation, or specialized training techniques.

The solutions to these challenges lie in engineering the data and models specifically for microscopy applications. Microscopy images, in contrast to natural images, show much less variability in terms of the shapes, textures, and structures present. This means the final data distribution is much more constrained and can often be simulated using known physical principles. By using physical simulation, large amounts of realistic training data can be generated that closely resemble real-world microscopy images. This allows the models to be pre-trained on large synthetic datasets to learn robust initial representations. By further downstream fine-tuning on smaller real datasets, these initial representations can be calibrated to the specific microscopy application, whether it is fluorescence microscopy, electron microscopy, or others. This fine-tuning can even be done with as little as five to ten annotated images per class [84], if the initial priors are well aligned.

Another important aspect is the introduction of physics-based inductive biases into the model architecture itself. While general-purpose end-to-end architectures have shown great success in various domains, incorporating domain-specific knowledge can further enhance performance and even interpretability. One example would be splitting the task of detecting the objects of interest into a segmentation task followed by a classification task. Because the initial segmentation task is class-agnostic, the model is able to focus on learning the shapes and structures present in microscopy images. The classification of objects can then be performed by using engineered physics-based features [85] that are relevant to the specific microscopy application, outperforming end-to-end models [84]. This also allows for modular reuse of the segmentation model across different microscopy tasks, as only the classification module needs to be retrained for new applications.

To train a foundation model tailored for microscopy image analysis, it is important to address the unique challenges of this domain. By pre-training on simulated data, splitting tasks, and incorporating physics-based inductive biases, we can effectively use expert knowledge about our specific microscopy application to improve performance. The use of these specialized foundation models has the potential to significantly advance automated microscopy image analysis in the future.

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2.17 | Application of Artificial Intelligence to Electrochemical Sensing: How Deep Learning Can Open to New Perspective in the Analysis of Complex Samples by the Help of Tailored Materials

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Chemical sensing has emerged as a promising field to leverage the recent opportunities offered by artificial intelligence (AI) [86]. In particular, Machine Learning (ML) can transform the interpretation of complex signals deriving from the analysis of complex biological samples, facilitating rapid on-site diagnosis and detection of defined biomarkers. Indeed, ML models can process massive datasets to: (1) classify samples based on the concentration of a target analyte; (2) enhance accuracy by mitigating matrix effects and environmental variables; (3) compensate for sensor fouling drift; (4) increase signal-to-noise ratio to lower detection limits; (5) enable swift pattern recognition; and (6) tailor diagnostic models to individual patient metrics, from physical activity to vital signs. Thanks to these advantages, the application of supervised ML models is transforming modern healthcare, enabling the development of sophisticated wearable devices for fast, easy diagnosis of important diseases. Such devices are often made of amperometric sensors which are compact, cheap and easy to use [87, 88]. The signal, in this case, derives from oxidation or reduction processes directly involving the analyte or secondary species included in the system.

Typically, the analysis of multiple biomarkers is realized in parallel using independent sensors, each one dedicated to the detection of a specific analyte. Attention is paid in this case to the use of well selective sensitive materials, which can recognize the target, avoiding the interference due to the complex components of the biological matrix.

We recently proposed an alternative approach, consisting in the use of an array of poorly selective materials to detect and quantify electrolytes, such as Na^+ and K^+, in complex biological samples like sweat and saliva [89]. We first trained an artificial neural network (ANN) using signals produced by various sensors of the array in test solutions of known chemical composition. Then, we used the trained ANN to determine the unknown concentration of the target ions in test solutions, featuring different ionic strengths and also containing other ions as interferents.

The sensing mechanisms exploited the unique properties of redox active iron and nickel hexacyanoferrates (HCF); these materials feature a crystalline cubic structure where small ions, such as Na^+ and K^+, can penetrate with a different extent, resulting in redox responses distinctive of the chemical composition of the solution. Additionally, the sensors were externally coated by a layer of monoatomic 2-dimensional sheets of reduced graphene oxide (RGO), which forced the ions to undergo a tortuous path before reaching the active area of the sensor [90]. Thanks to these complementary mechanisms, ions experienced differential intercalation and diffusion processes within both RGO and HCF according to their ionic radius and chemical interaction with the coating, yielding distinct voltammetric signals.

At difference with conventional amperometric sensors, which estimate analyte concentration measuring a single current value at a specific potential, in our approach all the electrochemical data points acquired during the voltage scan are used for the analysis. This could only be achieved by training an ANN with a massive dataset consisting of current values recorded from the sensors using 327 variables as columns and over 4 million voltammetric traces as rows. To account for possible effects of sensor aging and fouling, also the age of the electrodes was used as input for the ANN.

In conclusion, while ML is typically employed to interpret data from specific selective sensors or to classify samples using poorly selective sensor elements of electronic noses/tongues, our approach opens the possibility of using ANNs to quantify target species with non-specific sensors. Of note, the strategy enables to use electrochemistry to quantify non-electroactive ions (such as Na^+ and K^+) using amperometric sensors, providing an alternative to the use of more conventional ion-selective electrodes (ISE), which possess a fixed and limited sensitivity.

By following a similar route, we are now trying to expand the range of materials included in the multi-sensor array to increase the information deriving from sensing elements and to extend the type of ions which can be quantified by our ANN (e.g. NH_4^+ and Ca^{2+}).

2.18 | Toward a Materials Commons for Europe

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In recent years, the combination of richer data sets, more and cheaper computing power and tools incorporating artificial intelligence, has accelerated scientific and technological advances. These developments make it possible to substantially shorten the time needed to find new materials, bring them to practical applications and scale-up their production. European researchers hence face a significant paradigm change in the way new materials are discovered, characterized, developed, tested and produced.

Advanced materials are critical building blocks for technology and innovation across different sectors such as electronics, construction, renewable energy and mobility. While their development and deployment enable new technological capabilities that fuel economic growth, they are also key to addressing societal challenges, such as sustainable growth, climate change and strategic autonomy. Accelerating research and innovation of advanced materials through digitalization is therefore a key priority for the European Commission. At national level this is also

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recognized; several European countries have created major national initiatives to bring together and strengthen the digitalization in this field, ranging from building data sets and developing tools, to integrating artificial intelligence into the research and innovation process.

In 2025, the European Union has launched a topic under a Horizon Europe call for proposals – *Materials Commons for Europe* – aiming to build a pan-European digital infrastructure that will accelerate the design, development, characterization and testing of advanced materials. The initiative aims to bring together public research organizations, industry partners and national digital platforms under a common framework that can serve both academia and the industry.

Why a Materials Commons for Europe?

Whereas advanced materials are a cornerstone of Europe's industrial leadership, digital aspects of research in this field are fragmented. Researchers and innovators face isolated data repositories with ad-hoc interfaces, incompatible data formats and conflicting laboratory workflows. The Materials Commons for Europe envisions a pioneer federated infrastructure that will give researchers seamless access to FAIR, heterogeneous data sources and interoperable computational tools, thereby shortening the time from laboratory discovery to market uptake. By integrating AI and lab automation, the platform will enable new techniques for automatic discovery, predictive modelling and automated decision-making, fostering a new era of data-centric materials science.

The project is expected to deliver the following outcomes:

1. A federated digital infrastructure, supporting industrial use cases and providing a feedback loop to academic research;
2. FAIR data access for industry and academia, linking heterogeneous datasets and computational services;
3. Tailored solutions for experimental workflows, ensuring high-quality, well-documented primary data;
4. A framework for self-driving labs, integrating state-of-the-art AI and predictive models;
5. Long-term sustainability mechanisms allowing the infrastructure to evolve beyond the initial project phase.

This approach aligns with the Materials 2030 manifesto [91] ("*High performance computing, big data and AI revolutionize the digital modelling, simulation and screening of materials properties, materials development and production processes, unlocking the merge of computational and experimental material science.... This will lead to new safe and sustainable products / systems by design, which utilise smart and advanced materials with targeted functionalities and physical properties*") and the European Commission's Communication on Advanced Materials for Industrial Leadership [92], aiming at strengthening the European research and innovation ecosystem on advanced materials. The Materials Commons for Europe should also be seen in the context of the European AI in Science strategy [93], which presents a European approach to accelerate the adoption of AI by European scientists in all disciplines, including building leading European AI scientific models and supporting their innovative potential to boost the impact, quality and productivity of science.

To address these challenges, the Materials Commons for Europe will put forward common vocabularies, semantic descriptions and interfaces. While keeping the human in the loop, it will also rely on federated machine learning that allow for training models on open as well as private datasets. The ultimate expectation is a resilient, evolving ecosystem that continuously translates scientific breakthroughs into safe, sustainable products and systems, delivering economic growth and societal benefits across Europe, in the context of other actions in this area taken under the strategy on advanced materials.

2.19 | Digital Maturity and Regulatory Readiness of European Advanced Materials Communities

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Companies in the export-oriented European industry are integrated into European and worldwide, complex supply networks. The shortage of raw materials in Europe makes it necessary to ensure industrial value creation through constant innovations in materials, manufacturing and production, based on raw and recycled materials and intermediate products in an internationally drastically changing context. On the European policy side [92, 94, 95], strategic goals have been proclaimed, based on the European Green Deal and the Clean Industrial Deal, that have already been taken up legislatively. Above all, the Data Act, the AI Act, the TEN-E Regulation, the Critical Raw Materials Act, and the Net Zero Industry Act should be mentioned here as key initiatives supporting the accelerated roll-out of clean energy and manufacturing; elements of which can already be found for example in the Ecodesign for Sustainable Product Regulation (ESPR [96]), in particular the plans to introduce Digital Product Passports (DPPs) as measures to support the transition to a circular European economy. These legislations/regulations effect even international, cross-EU border value networks, since European importers will be responsible to issue data for European DPPs once materials and/or (intermediate) products cross European borders. Therefore, the European Twin Green&Digital Transition process is not an option, but rather the future basis of economic activities in the European Union.

Industrial transition and innovation processes in general call for accelerating advanced materials design, development and upscaling as a basis for manufacturing. In this context, the development of cost-effective, cloud-based, and interoperable digital solutions, such as DPP platforms, digital twins, predictive lifetime modelling tools (both AI- and physics-based), and AI-driven systems for automated regulatory updates, will be essential to speed-up the uptake of advanced materials by product innovators/manufacturers.

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The EU project DigiPass Coordination and Support Action (DigiPass-CSA) [97] takes the position that accelerating the design, development, and production of regulatory compliant advanced, safe, and sustainable chemicals and materials calls for a collaborative approach involving many stakeholders within value networks to support durability, R-strategies and recyclability of products. A central objective of DigiPass-CSA is to couple regulatory aspects with collaborative materials and products innovation processes on federated dataspaces, thus allowing for a seamless transition from research to innovation to production:

- the ESPR is the central regulatory instrument which strives to improve for instance on products' circularity, carbon footprint, and toxicity.
- the Safe and Sustainable by Design (SSbD) framework on the other hand represents a structured (voluntary) framework to drive innovation processes, and the Key Performance Indicators of SSbD closely matches the ESPR requirements for DPPs.

DigiPass-CSA builds a digital demonstration platform [98], which combines production and (materials) innovation by advising a seamless digitalized transition from SSbD driven innovation processes (SSbD-KPI data) to industrial production (ESPR-DPP data). Moreover, the platform design follows a user-centric approach. One DigiPass ideas is the introduction of a Digital Materials Passport (DMP), that enters a DPP in parallel to the flow of materials in production. For fully functional scalable systems, digital infrastructures are necessary, including their streamlined governance. The infrastructure can support the collection, storage, access, and analysis of datasets for all types of Advanced Materials – across entire value networks. Such infrastructure is beyond of DigiPass-CSA scope.

Another overarching objective of the DigiPass-CSA is to bolster the digital maturity of European stakeholders from manufacturing and materials development communities, with a particular emphasis on small and medium enterprises (SMEs). The project develops recommendations and clear routes toward digitalized circular business models. The key result of DigiPass is to create a sustainable digital platform which includes support for Digital Materials & Product Passport (DMPP) and for collaborative innovation-by-design processes in a circular economy served by Advanced Materials. The status of the ongoing platform development can be traced online [98].

2.20 | Getting Better Energy Materials Faster With ML and Distributed Platforms - *Interoperable Data Infrastructures and Federated Labs*

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Harvesting the full potential of AI-orchestration, machine-learned models and automated data generation to accelerate the discovery and industrial adoption of advanced energy materials hinges on the close integration of three pillars: (i) interoperable data infrastructures with shared semantics and governance; (ii) federated self-driving laboratories (SDLs) that can operate across laboratories; and (iii) dynamic and interoperable modular workflows that integrates simulations, ML and experimental workflow nodes at materials and device level.

Federated SDLs for advanced materials as a European stronghold. SDLs will be most valuable when they form a distributed capability with well-defined interfaces and data standards, providing (real-time) access to bespoke models, instruments, or facilities. Proof-of-concept demonstrations have shown the path. In the large-scale EU project BIG-MAP, a distributed battery Materials Acceleration Platform (MAP) based on the FINALES framework orchestrated asynchronous experiments and simulations across multiple labs (tenants) in multiple countries, while leaving control with partners, an essential ingredient for cross-lab validation and IP-aware collaboration [99]. In parallel, a community perspective argues for SDLs that embed human-in-the-loop decision-making and accessibility, so the benefits scale beyond a single site and enable rapid cross-lab validation. Large-scale, interoperable workflows for battery simulation and multi-modal characterization [100], and multitiered data-arching [101] also performed in BIG-MAP further illustrate how coordinated data generation, archiving, and correlative analysis can be standardized across facilities to accelerate advanced materials discovery. The long-term opportunity is to develop an interoperable pan-European network of SDL nodes, spanning simulations, synthesis, characterization, and testing of materials for energy, mobility, engineering/construction, electronics, and exposed through standard APIs and governed by data/ontology agreements.

Workflows and ML models that adapt, interoperate and provide uncertainty quantification. Method development is the core that enables proof-of-concept studies to evolve into generalizable and operationalizable tools. The workflow orchestrator PerQueue illustrates the kind of dynamism that will be needed: active-learning loops that can switch between DFT, MLIPs, molecular dynamics, and experimental segmentation, with the ability to re-plan at runtime and to hand off between virtual and physical tasks [102]. Within electrochemical energy materials, asynchronous MAPs demonstrate co-optimization of materials and device-level KPIs by integrating ontology-mapped data (e.g., electrolyte formulation, cycling, early-life prediction) with ML models and molecular simulations [99]. Looking forward, the community should converge on exchange formats that permit a workflow or workflow node to connect to different engines and institutions while carrying its metadata, uncertainty estimates, and validation records.

Open foundation and fine-tuned models with scientific validation. Open-source, general-purpose (atomistic) models reduce the barrier to simulating complex chemistries and morphologies; combined with fine-tuning and in-loop validation, they have reached a level where they can capture complex interactions, such as long-range electrostatics, and may ultimately drive targeted "synthesis by design" experiments. The MACE-family foundation model exemplifies this trend and has been applied across liquids, crystals, defects, and surfaces [59] Access to curated large-scale computational datasets that go beyond

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ground-state energies and describe, e.g, transition states will be required to unlock their full potential.

In a multi-sourced, multi-fidelity, and multi-modal workflow environment, adopting a tomographic perspective suggests that requiring faithful prediction of diverse observables can constrain learned latent spaces and improve reliability and interpretability, offering a principled route to representation learning in AI4materials [103, Ortega-Ochoa et al. 2025].

Case studies as stepping-stones. Proofs of concept illustrate what a mature, federated ecosystem could deliver at scale. In electrocatalysis, the autonomous platform "FastCat" explored >500 multielement Ni-based LDH compositions for alkaline OER, identifying high-performance PGM-free alloys while operating at ~75 compositions per day [104]. In structure-targeted nanoparticle synthesis for CO_2 reduction, an autonomous loop combined a fine-tuned potential with real-time total-scattering/PDF at a synchrotron to match simulated target patterns, successfully producing decahedral (~5 nm) and fcc (~10 nm) Au nanoparticles and documenting the synthesis protocol [105]. These are not endpoints, but blueprints for what a networked European capability can make routine.

Governance, incentives and policy alignment. The European Commission and Technology Council for Advanced Materials is in the process of preparing the Advanced Materials Act. Here, a federated materials infrastructure should align with these processes, offering: (i) tiered access and compute-to-data to protect IP while enabling AI; (ii) mechanisms for accreditation and onboarding of new nodes; and (iii) measurable targets (time-to-validated-device, cross-lab reproducibility, fraction of automated workflows) that link infrastructure to industrial impact. The newly funded MaterialsCommons4.eu project (Horizon Europe, #101293545) will contribute here by operationalizing a pan-European knowledge graph, a semantic foundation and workflow exchange that can host SDL endpoints and compute services. The materials commons platform should facilitate access to unique research infrastructure and capability, and accelerate and incentivize industry adoption of advanced materials, by providing uncertainty quantification and de-risking to the decision makers.

2.21 | Novelty-Generating Materials as a Substrate for Open-Ended Computation

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The fundamental limitation of contemporary artificial intelligence lies not in its computational power but in its conceptual foundations. Current AI systems operate within fixed datasets, static learning paradigms, and i.i.d. (independent and identically distributed) assumptions that constrain them to predefined tasks. In contrast, biological intelligence exhibits continuous learning, evolving complexity, and open-ended novelty—characteristics that emerge from hierarchical structures and dynamic feedback loops operating at the edge of chaos. This section proposes a radical departure from conventional computing: leveraging physical materials whose intrinsic dynamics naturally implement open-ended computational processes.

2.21.1 | The Open-Ended Evolution Framework

Bio-intelligence operates within an evolutionary framework characterized by three essential properties. First, hierarchy continuously creates new organizational levels, enhancing adaptive complexity. Second, adaptivity emerges through novelty generation via variation and selection mechanisms [106]. Third, feedback loops arise through emergent higher-order patterns that recursively influence lower-level dynamics. This interplay across three levels—materials (L1), physical constraints (L2), and optimization principles (L3)—produces "edge-of-chaos" dynamics that enable continual novelty and adaptive emergence.

Open-ended computation, by definition, never exhausts its space of possibilities. It exhibits non-i.i.d., history-dependent evolution and unbounded complexity growth without fixed sophistication limits: properties that traditional machine learning pipelines, designed for static optimization, fundamentally cannot accommodate, necessitating entirely new computational substrates.

The insufficiency is not algorithmic but physical. Software open-endedness—novelty search [107], quality-diversity [108, 109], POET [110]—generates variety within a fixed state space defined by the programmer; the rule set never expands itself. Clocked von Neumann logic enforces discrete state updates with memory separated from compute—precisely the conditions that preclude genuine non-equilibrium dynamics, continuous history-dependence, and the open thermodynamic flux from which biological novelty arises. New rules can only be added from outside the system. Open-endedness therefore requires substrates whose physics, not whose code, supplies the unbounded degrees of freedom.

2.21.2 | State-of-the-Art: Physical Substrates for Novelty Generation

The most promising candidate materials share critical dynamical properties observed in biological neural systems. Magnetic skyrmions—topologically protected spin textures in magnetic thin films—exemplify these principles [111]. They exhibit topological stability with dynamical flexibility: maintaining structural identity under perturbations while enabling interaction, merging, splitting, and evolution. Their nonlinear, nonlocal spin dynamics support complex collective behavior across spatial scales, analogous to long-range neural interactions. Crucially, skyrmions display intrinsic memory through hysteresis and exhibit history-dependent, context-aware behavior essential for novelty generation.

Beyond skyrmions, several material classes show potential: spintronic systems combining topological stability with dynamic evolution; resistive switching and memristive devices offering rich nonlinear dynamics for synaptic and neuronal functions; metal-insulator transition (MIT) devices like VO_2 with intrinsic threshold and excitability properties; and electroactive metal-organic frameworks with tunable band gaps enabling adaptive reconfiguration. These materials share a common thread: their dynamical complexity arises from fundamental physical mechanisms rather than engineered circuitry.

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Experimental signatures of novelty-generating capacity include dynamical properties (transient metastable states), emergent behaviors (spontaneous spatiotemporal pattern formation), and statistical hallmarks (power-law distributed event sizes characteristic of avalanche dynamics) [112]. Harnessing these dynamics requires a read-out and steering scheme rather than a program. In the physical reservoir-computing paradigm [113], the material's intrinsic evolution serves as a high-dimensional nonlinear map; only a lightweight output layer is trained, while inputs are injected as physical perturbations (fields, voltages, currents) and outputs read from local observables (resistance, magnetization, emitted patterns). Steering is exercised through L2 constraints—boundary geometry, drive amplitude, coupling topology—that bias emergence without prescribing it. A minimal demonstrator would inject a temporal signal into a skyrmion or VO_2 film and recover task-relevant structure from its avalanche statistics, testing whether $\Delta CP > 0$ coincides with computational utility.

2.21.3 | *Critical Challenges*

Three interconnected challenge domains must be addressed. Theoretically, open-ended computation requires new mathematical foundations: recursive procedural formalisms for intrinsic novelty, scalable methods for non-equilibrium systems, and robust multiscale materials modeling. From a materials-science standpoint, the challenge is to engineer the requisite dynamical complexity while imposing constraints that guide emergence without collapsing the possibility space. Most significantly, causal-emergence frameworks offer principled measures for characterizing novelty generation itself.

Causal Emergence 2.0 quantifies how macro-level descriptions carry irreducible causal power ($\Delta CP > 0$) through noise reduction and degeneracy reduction channels [114]. Scale boundaries manifest as structural pathologies: commutativity failures where interventions at different scales produce inconsistent predictions [115], DAG instabilities indicating theory breakdowns, or interaction range discontinuities signaling organizational transitions. Causal representation learning extends beyond discovering relationships to discovering the variables themselves [116]—when abstraction maps fail to commute, the breakdown localizes where new physical concepts must emerge. This reframes the measurement challenge: rather than syntactic complexity (Kolmogorov, sophistication, assembly index), emergence becomes detectable from causal dynamics. Material substrates can be evaluated by whether their intrinsic physics naturally supports hierarchical causal structures with $\Delta CP > 0$ at multiple scales—a testable, physically grounded criterion bridging observables with computational capability.

2.21.4 | *Conclusion*

Novelty-generating materials offer physical substrates where emergent behavior arises from intrinsic dynamics rather than programmed algorithms. This paradigm shift reframes computation as discovery rather than execution, yielding results unreachable by current AI.

3 | Conclusions

This Perspective has outlined a collection of snapshots reflecting current developments in the application of artificial intelligence to materials science. The individual contributions collectively illustrate a diverse and rapidly evolving landscape of research directions spanning data-driven discovery, atomistic modeling and experimental automation. A central conclusion of the revision is therefore one of scope and positioning.

The article is best understood as a community snapshot rather than as a single, fully integrated roadmap. Its value lies in juxtaposing active directions presented within the AI4AM community, while making explicit that important areas remain less developed in the present collection. This clarification is essential because the field is moving faster than any single conference-based Perspective can comprehensively capture.

Among the clearest emerging frontiers is agentic and generative discovery. Multi-agent scientific systems now begin to combine large language models, structured knowledge graphs, retrieval, physics-based simulation, automated critique, and planning to generate hypotheses, design workflows, and refine candidate materials or biomolecular systems [2–6]. Related decentralized frameworks show how independent agents can exchange artifacts, preserve computational lineage, and coordinate discovery without a single centralized planner [7]. These developments broaden the meaning of AI for materials beyond prediction and optimization toward scientific reasoning, falsifiable hypothesis generation, and auditable autonomous workflows.

A first key theme concerns the increasing use of machine learning not only for property prediction, but also for generation, inverse design, and synthesis planning within expanding chemical spaces. In this context, large language models and generative approaches are enabling new strategies for assessing synthesizability, redesigning candidate structures, and proposing synthesis pathways.

At the same time, advances in atomistic machine learning and electronic-structure modeling are pushing the boundaries of accuracy and scale. Universal interatomic potentials, operator learning frameworks, and machine-learned exchange–correlation functionals collectively point toward a framework in which quantum-mechanical accuracy can be extended to previously inaccessible regimes. However, these developments also highlight fundamental challenges in these approaches, particularly in terms of transferability, interpretability, and robustness.

A recurring theme is the central role of *data*. The availability of large-scale databases has enabled unprecedented progress, yet issues of bias, data quality, and data distribution remain limiting factors. Addressing these challenges requires not only larger datasets, but better representations, standardized infrastructures, and interoperable platforms that connect simulation, experiment, and machine learning in a seamless manner.

Equally critical is the question of *trust*. The integration of uncertainty quantification into machine learning workflows is emerging as an essential requirement for scientific reliability. Without

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rigorous assessment of prediction confidence, the risk of propagating errors across increasingly complex pipelines becomes significant. In this sense, uncertainty-aware methodologies are poised to become as fundamental as accuracy and efficiency in future developments.

Trust must consequently be framed more broadly than confidence intervals or model errors alone. For AI-driven materials discovery to become scientifically robust, proposed predictions and hypotheses should be exposed to adversarial falsification, checked against physical constraints, connected to compositional world models, and validated through traceable digital-to-physical feedback. In this sense, uncertainty-aware methods, provenance-rich data infrastructures, closed-loop experiments, and agent-based critique should be regarded as complementary safeguards rather than separate topics [7, 8].

Another major frontier lies in bridging the digital and physical domains. From experiment-constrained simulations to autonomous laboratories and AI-driven spectroscopy, the boundary between computation and experiment is progressively dissolving. Closed-loop frameworks, in which models iteratively guide experiments and learn from feedback, represent a transformative step toward self-driving materials discovery.

Taken together, these developments signal a paradigm shift: from sequential, human-guided workflows to interconnected, adaptive, and partially autonomous ecosystems. Realizing this vision will require sustained interdisciplinary collaboration, combining advances in physics, chemistry, computer science, and engineering, as well as strong coordination between academia, industry, and policy frameworks.

In this evolving landscape, the central challenge lies in the convergence of tools into interoperable and adaptive ecosystems. The next phase of materials discovery will rely on tightly integrated frameworks combining data infrastructures, AI models, and experimental platforms, capable of operating across scales while ensuring reliability, interpretability, and scientific robustness.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data sharing is not applicable to this article as no datasets were generated or analyzed during the current study.

References

1. C. Malica, K. S. Novoselov, A. S. Barnard, et al., "Artificial Intelligence for Advanced Functional Materials: Exploring Current and Future Directions," *Journal of Physics: Materials* 8, no. 2 (2025): 021001.

2. A. Ghafarollahi and M. J. Buehler, "SciAgents: Automating Scientific Discovery Through Bioinspired Multi-Agent Intelligent Graph Reasoning," *Advanced Materials* 37 (2025): 2413523.

3. A. Ghafarollahi and M. J. Buehler, "Sparks: Multi-Agent Artificial Intelligence Model Discovers Protein Design Principles," preprint, arXiv:2504.19017, 2025, https://doi.org/10.48550/arXiv.2504.19017.

4. A. Ghafarollahi and M. J. Buehler, "ProtAgents: Protein Discovery via Large Language Model Multi-Agent Collaborations Combining Physics and Machine Learning," *Digital Discovery* 3 (2024): 1389–1409, https://doi.org/10.1039/D4DD00013G.

5. A. Ghafarollahi and M. J. Buehler, "Rapid and Automated Alloy Design With Graph Neural Network-Powered Large Language Model-Driven Multi-Agent AI," *MRS Bulletin* 50 (2025): 1309–1324, https://doi.org/10.1557/s43577-025-00953-4.

6. A. Ghafarollahi and M. J. Buehler, "Autonomous Inorganic Materials Discovery via Multi-Agent Physics-Aware Scientific Reasoning," preprint, arXiv:2508.02956, 2025, https://doi.org/10.48550/arXiv.2508.02956.

26404567, 0, Downloaded from https://advanced.onlinelibrary.wiley.com/doi/10.1002/aisy.70496 by Universite Catholique De Louvain, Wiley Online Library on [28/08/2026]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

7. F. Y. Wang, L. Marom, S. Pal, et al., "Autonomous Agents Coordinating Distributed Discovery Through Emergent Artifact Exchange," preprint, arXiv:2603.14312, 2026, https://doi.org/10.48550/arXiv.2603.14312.

8. M. J. Buehler, "Why We Must Break the World," preprint, ChemRxiv, 2026, https://doi.org/10.26434/chemrxiv.15001674/v2.

9. J. Jang, G. H. Gu, J. Noh, J. Kim, and Y. Jung, "Structure-Based Synthesizability Prediction of Crystals Using Partially Supervised Learning," *Journal of the American Chemical Society* 142, no. 44 (2020): 18836–18843.

10. S. Kim, Y. Jung, and J. Schrier, "Large Language Models for Inorganic Synthesis Predictions," *Journal of the American Chemical Society* 146, no. 29 (2024): 19654–19659.

11. S. Kim, J. Schrier, and Y. Jung, "Explainable Synthesizability Prediction of Inorganic Crystal Polymorphs Using Large Language Models," *Angewandte Chemie International Edition* 64, no. 19 (2025): e202423950.

12. J. Choi, S. Kim, and Y. Jung, "Synthesis-Aware Materials Redesign via Large Language Models," *Journal of the American Chemical Society* 147, no. 43 (2025): 39113–39122.

13. S. Kim, J. Noh, G. H. Gu, S. Chen, and Y. Jung, "Predicting Synthesis Recipes of Inorganic Crystal Materials Using Elementwise Template Formulation," *Chemical Science* 15, no. 3 (2024): 1039–1045.

14. T. Cavignac, J. Schmidt, P.-P. De Breuk, et al., "AI-Driven Expansion and Application of the Alexandria Database," *Journal of Physics: Materials* 9 (2026): 025014, https://doi.org/10.1088/2515-7639/ae6620.

15. J. Schmidt, N. Hoffmann, H. Wang, et al., "Machine-Learning-Assisted Determination of the Global Zero-Temperature Phase Diagram of Materials," *Advanced Materials* 35 (2023): 2210788, https://doi.org/10.1002/adma.202210788.

16. J. Schmidt, T. F. T. Cerqueira, A. H. Romero, et al., "Improving Machine-Learning Models in Materials Science Through Large Datasets," *Materials Today Physics* 48 (2024): 101560, https://doi.org/10.1016/j.mtphys.2024.101560.

17. T. F. T. Cerqueira, H.-C. Wang, S. Botti, and M. A. L. Marques, "A Non-Orthogonal Representation for Materials Based on Chemical Similarity," *npj Computational Materials* 12 (2026): 48, https://doi.org/10.1038/s41524-025-01916-8.

18. A. Loew, D. Sun, H.-C. Wang, S. Botti, and M. A. L. Marques, "Universal Machine Learning Interatomic Potentials Are Ready for Phonons," *npj Computational Materials* 11 (2025): 178, https://doi.org/10.1038/s41524-025-01650-1.

19. M. A. Caro, G. Csányi, T. Laurila, and V. L. Deringer, "Machine Learning Driven Simulated Deposition of Carbon Films: From Low-Density to Diamondlike Amorphous Carbon," *Physical Review B* 102 (2020): 174201.

20. G. Opletal, T. C. Petersen, I. K. Snook, and S. P. Russo, "HRMC_2.0: Hybrid Reverse Monte Carlo Method With Silicon, Carbon and Germanium Potentials," *Computer Physics Communications* 184 (2013): 1946.

21. T. Zarrouk, R. Ibragimova, A. P. Bartók, and M. A. Caro, "Experiment-Driven Atomistic Materials Modeling: A Case Study Combining X-Ray Photoelectron Spectroscopy and Machine Learning Potentials to Infer the Structure of Oxygen-Rich Amorphous Carbon," *Journal of the American Chemical Society* 146 (2024): 14645.

22. T. Zarrouk and M. A. Caro, "Molecular Augmented Dynamics: Generating Experimentally Consistent Atomistic Structures by Design," preprint, arXiv:2508.17132 (2025).

23. T. Zarrouk and M. A. Caro, "Linear-Scaling Calculation of Experimental Observables for Molecular Augmented Dynamics Simulations," preprint, arXiv:2509.22388 (2025).

24. M. Kellner and M. Ceriotti, "Uncertainty Quantification by Direct Propagation of Shallow Ensembles," *Machine Learning: Science and Technology* 5, no. 3 (2024): 035006.

25. F. Bigi, S. Chong, M. Ceriotti, and F. Grasselli, "A Prediction Rigidity Formalism for Low-Cost Uncertainties in Trained Neural Networks," *Machine Learning: Science and Technology* 5, no. 4 (2024): 045018.

26. S. Chong, F. Grasselli, C. B. Mahmoud, J. D. Morrow, V. L. Deringer, and M. Ceriotti, "Robustness of Local Predictions in Atomistic Machine Learning Models," *Journal of Chemical Theory and Computation* 19, no. 22 (2023): 8020–8031.

27. S. Chong, F. Bigi, F. Grasselli, P. Loche, M. Kellner, and M. Ceriotti, "Prediction Rigidities for Data-Driven Chemistry," *Faraday Discussions* 256 (2025): 322–344.

28. F. Bigi, J. W. Abbott, P. Loche, et al., "Metatensor and Metatomic: Foundational Libraries for Interoperable Atomistic Machine Learning," *Journal of Chemical Physics* 164 (2026): 064113, https://doi.org/10.1063/5.0304911.

29. S. Dick and M. Fernández-Serra, "Machine Learning Accurate Exchange and Correlation Functionals of the Electronic Density," *Nature Communications* 11 (2020): 3509.

30. S. Dick and M. Fernández-Serra, "Highly Accurate and Constrained Density Functional Obtained With Differentiable Programming," *Physical Review B* 104 (2021): L161109.

31. L. Li, S. Hoyer, R. Pederson, et al., "Kohn–Sham Equations as Regularizer: Building Prior Knowledge Into Machine-Learned Physics," *Physical Review Letters* 126 (2021): 036401.

32. M.-C. Kim, E. Sim, and K. Burke, "Understanding and Reducing Errors in Density Functional Calculations," *Physical Review Letters* 111 (2013): 073003.

33. M. G. Medvedev, I. S. Bushmarinov, J. Sun, J. P. Perdew, and K. A. Lyssenko, "Density Functional Theory Is Straying From the Path Toward the Exact Functional," *Science* 355 (2017): 49–52.

34. I. Batiata, P. Benner, Y. Chiang, et al., "A Foundation Model for Atomistic Materials Chemistry," *Journal of Chemical Physics* 163 (2025): 184110.

35. H. Kneiding, R. Lukin, L. Lang, et al., "Deep Learning Metal Complex Properties With Natural Quantum Graphs," *Digital Discovery* 2 (2023): 618–633.

36. A. M. Ganose, H. Sahasrabuddhe, M. Asta, et al., "Atomate2: Modular Workflows for Materials Science," *Digital Discovery* 4 (2025): 1944–1973, https://doi.org/10.1039/d5dd00019j.

37. A. A. Naik, C. Ertural, N. Dhamrait, P. Benner, and J. George, "A Quantum-Chemical Bonding Database for Solid-State Materials," *Scientific Data* 10 (2023): 610.

38. A. A. Naik, N. Dhamrait, K. Ueltzen, et al., "A critical assessment of bonding descriptors for predicting materials properties," preprint, arXiv, 2026, https://doi.org/10.48550/arXiv.2602.12109.

39. Y. Liu, J. D. Morrow, C. Ertural, et al., "An Automated Framework for Exploring and Learning Potential-Energy Surfaces," *Nature Communications* 16 (2025): 7666.

40. Ö. H. Omar, M. del Cueto, T. Nematiaram, and A. Troisi, "High-Throughput Virtual Screening for Organic Electronics: A Comparative Study of Alternative Strategies," *Journal of Materials Chemistry C* 9 (2021): 13557–13583.

41. Ö. H. Omar, T. Nematiaram, A. Troisi, and D. Padula, "Organic Materials Repurposing, a Data Set for Theoretical Predictions of New Applications for Existing Compounds," *Scientific Data* 9 (2022): 54.

42. Ö. H. Omar, D. Padula, K. Morrison, et al., "Breaking the Digital-Physical Barrier in Organic Electronics Materials Discovery: Application to Finding Anti-Kasha Molecules," preprint, ChemRxiv, 2025, https://doi.org/10.26434/chemrxiv-2025-5z172.

43. Z. Xie, X. Evangelopoulos, Ö. H. Omar, A. Troisi, A. I. Cooper, and L. Chen, "Fine-Tuning GPT-3 for Machine Learning Electronic and

26404567, 0, Downloaded from https://advanced.onlinelibrary.wiley.com/doi/10.1002/aisy.70496 by Universite Catholique De Louvain, Wiley Online Library on [28/08/2026]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

Functional Properties of Organic Molecules," *Chemical Science* 15 (2024): 500–510.

44. A. Coxson, Ö. H. Omar., M. del Cueto, and A. Troisi, "Predicting S1 TDDFT Energies from ZINDO Calculations using Message-Passing Delta-ML with Electronically-Informed Descriptors," *ChemRxiv*, 2025, https://doi.org/10.26434/chemrxiv-2025-gln8z-v2.

45. M. L. Evans, J. Bergsma, A. Merkys, et al., "Developments and Applications of the OPTIMADE API for Materials Discovery, Design, and Data Exchange," *Digital Discovery* 3, no. 8 (2024): 1509–1533, https://doi.org/10.1039/D4DD00039K.

46. V. Trinquet, M. L. Evans, C. J. Hargreaves, P.-P. De Breuck, and G.-M. Rignanese, "Optical Materials Discovery and Design With Federated Databases and Machine Learning," *Faraday Discussions* 256 (2025): 459–482, https://doi.org/10.1039/D4FD00092G.

47. V. Trinquet, M. L. Evans, and G.-M. Rignanese, "Accelerating the Discovery of High-Performance Nonlinear Optical Materials Using Active Learning and High-Throughput Screening," *Journal of Materials Chemistry C* 13, no. 35 (2025): 18197–18212, https://doi.org/10.1039/D5TC01335F.

48. V. Trinquet, F. Naccarato, G. Brunin, et al., "Second-Harmonic Generation Tensors From High-Throughput Density-Functional Perturbation Theory," *Scientific Data* 11, no. 757 (2024): 1–10, https://doi.org/10.1038/s41597-024-03590-9.

49. L. I. Roest, S. E. van Heijst, L. Maduro, J. Rojo, and S. Conesa-Boj, "Charting the Low-Loss Region in Electron Energy-Loss Spectroscopy With Machine Learning," *Ultramicroscopy* 222 (2021): 113202, https://www.sciencedirect.com/science/article/pii/S0304399121000012.

50. A. Brokkelkamp, J. ter Hoeve, I. Postmes, et al., "Spatially-Resolved Band Gap and Dielectric Function in Two-Dimensional Materials From Electron Energy Loss Spectroscopy," *Journal of Physical Chemistry A* 126 (2022): 1255–1262, https://pubs.acs.org/doi/full/10.1021/acs.jpca.1c09566.

51. H. La, A. Brokkelkamp, S. van der Lippe, J. ter Hoeve, J. Rojo, and S. Conesa-Boj, "Edge-Induced Excitations in Bi_2Te_3 From Spatially-Resolved Electron Energy-gain Spectroscopy," *Ultramicroscopy* 254 (2023): 113841, https://www.sciencedirect.com/science/article/pii/S0304399123001584.

52. S. E. van Heijst, M. Bolhuis, A. Brokkelkamp, J. J. M. Sangers, S. Conesa-Boj, "Heterostrain-Driven Bandgap Increase in Twisted WS_2: A Nanoscale Study," *Advanced Functional Materials* 34, no. 8 (2024): 2307893, https://advanced.onlinelibrary.wiley.com/doi/full/10.1002/adfm.202307893.

53. S. van der Lippe, A. Brokkelkamp, J. Rojo, and S. Conesa-Boj, "Localized Exciton Anatomy and BandGap Energy Modulation in 1D MoS_2 Nanostructures," *Advanced Functional Materials* 33, no. 52 (2023): 2307610, https://advanced.onlinelibrary.wiley.com/doi/full/10.1002/adfm.202307610.

54. S. Haastrup, M. Strange, M. Pandey, et al., "The Computational 2D Materials Database: High-Throughput Modeling and Discovery of Atomically Thin Crystals," *2D Materials* 5 (2018): 042002.

55. O. Isayev, C. Oses, C. Toher, E. Gossett, S. Curtarolo, and A. Tropsha, "Universal Fragment Descriptors for Predicting Properties of Inorganic Crystals," *Nature Communications* 8 (2017): 15679.

56. S. Lu, Q. Zhou, Y. Guo, Y. Zhang, Y. Wu, and J. Wang, "Coupling a Crystal Graph Multilayer Descriptor to Active Learning for Rapid Discovery of 2d Ferromagnetic Semiconductors/Half-Metals/Metals," *Advanced Materials* 32, no. 29 (2020): 2002658.

57. C. B. Owens, N. Mathew, T. W. Olaveson, et al., "Feature Engineering Descriptors, Transforms, and Machine Learning for Grain Boundaries and Variable-Sized Atom Clusters," *npj Computational Materials* 11 (2025): 21.

58. A. El Alouani, M. Al Khalfioui, A. Michon, S. Vézian, P. Boucaud, and M. T. Dau, "Defect Formation Energy of Impurities in 2D Materials: How Does Data Engineering Shape Machine Learning Model Selection?," *Materials Today Physics* 61 (2026): 102006.

59. I. Batatia, D. P. Kovacs, G. Simm, et al., "MACE: Higher Order Equivariant Message Passing Neural Networks for Fast and Accurate Force Fields," *Advances in Neural Information Processing Systems* 35 (2022): 11423–11436.

60. X. Gong, H. Li, N. Zou, R. Xu, W. Duan, and Y. Xu, "General Framework for E(3)-Equivariant Neural Network Representation of Density Functional Theory Hamiltonian," *Nature Communications* 14 (2023): 2848.

61. C. Qian, V. Vitartas, J. Kermode, and R. J. Maurer, "Equivariant Electronic Hamiltonian Prediction With Many-Body Message Passing," preprint, arXiv:2508.15108, 2025.

62. K. F. Garrity and K. Choudhary, "Fast and Accurate Prediction of Material Properties With Three-Body Tight-Binding Model for the Periodic Table," *Physical Review Materials* 7 (2023): 044603.

63. P. C. Jennings, S. Lysgaard, J. S. Hummelshøj, T. Vegge, and T. Bligaard, "Genetic Algorithms for Computational Materials Discovery Accelerated by Machine Learning," *npj Computational Materials* 5 (2019): 46.

64. Y. Zhong, H. Yu, M. Su, X. Gong, and H. Xiang, "Transferable Equivariant Graph Neural Networks for the Hamiltonians of Molecules and Solids," *npj Computational Materials* 9 (2023): 182.

65. Z. Fan, J. H. Garcia, A. W. Cummings, et al., "Linear Scaling Quantum Transport Methodologies," *Physics Reports* 903 (2021): 1–69.

66. W. B. Park, J. Chung, J. Jung, et al., "Classification of Crystal Structure Using a Convolutional Neural Network," *IUCrJ* 4 (2017): 486–494.

67. F. Massuyeau, T. Broux, F. Coulet, A. Demessence, A. Mesbah, and R. Gautier, "Perovskite or Not Perovskite? a Deep-Learning Approach to Automatically Identify New Hybrid Perovskites From X-Ray Diffraction Patterns," *Advanced Materials* 34 (2022): 2203879.

68. J.-W. Lee, W. B. Park, J. H. Lee, S. P. Singh, and K.-S. Sohn, "A Deep-Learning Technique for Phase Identification in Multiphase Inorganic Compounds Using Synthetic XRD Powder Patterns," *Nature Communications* 11 (2020): 86.

69. V. Stanev, V. V. Vesselinov, A. G. Kusne, G. Antoszewski, I. Takeuchi, and B. S. Alexandrov, "Unsupervised Phase Mapping of X-Ray Diffraction Data by Nonnegative Matrix Factorization Integrated With Custom Clustering," *npj Computational Materials* 4 (2018): 43.

70. G. Guo, T. L. Saidi, M. W. Terban, M. Valsecchi, S. J. L. Billinge, and H. Lipson, "Ab Initio Structure Solutions From Nanocrystalline Powder Diffraction Data via Diffusion Models," *Nature Materials* 24 (2025): 1726–1734.

71. N. Marzari and D. Vanderbilt, "Maximally Localized Generalized Wannier Functions for Composite Energy Bands," *Physical Review B* 56 (1997): 12847.

72. A. Marrazzo, S. Beck, E. R. Margine, et al., "Wannier-Function Software Ecosystem for Materials Simulations," *Reviews of Modern Physics* 96 (2024): 045008.

73. J. Qiao, G. Pizzi, and N. Marzari, "Projectability Disentanglement for Accurate and Automated Electronic-Structure Hamiltonians," *npj Computational Materials* 9 (2023): 208.

74. J. Qiao, G. Pizzi, and N. Marzari, "Automated Mixing of Maximally Localized Wannier Functions Into Target Manifolds," *npj Computational Materials* 9 (2023): 206.

75. S. P. Huber, M. Minotakis, M. Bercx, et al., "MC3D: The Materials Cloud Computational Database of Experimentally Known Stoichiometric Inorganics," *Digital Discovery* 5 (2026): 1114–1131.

76. M. Bercx, S. Poncé, Y. Zhang, et al., "Charting the Landscape of Bardeen-Cooper-Schrieffer Superconductors in Experimentally Known Compounds," *PRX Energy* 4 (2025): 033012.

77. H. Jia, C. Wang, C. Wang, and P. Clancy, "Machine Learning Approach to Enable Spectral Imaging Analysis for Particularly Complex Nanomaterial Systems," *ACS Nano* 17, no. 1 (January 2023): 453–460, https://doi.org/10.1021/acsnano.2c08884.

26404567, 0, Downloaded from https://advanced.onlinelibrary.wiley.com/doi/10.1002/aisy.70496 by Universite Catholique De Louvain, Wiley Online Library on [28/08/2026]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License

78. L. Sabattini, A. Coriolano, C. Casert, et al., "Towards AI-Driven Autonomous Growth of 2D Materials Based on a Graphene Case Study," *Communications Physics* 8, no. 1 (April 2025): 180, https://doi.org/10.1038/s42005-025-02086-1.

79. J. Zhao, P. Ji, Y. Li, et al., "Ultrahigh-Mobility Semiconducting Epitaxial Graphene on Silicon Carbide," *Nature* 625, no. 7993 (January 2024): 60–65, https://doi.org/10.1038/s41586-023-06811-0.

80. G. Radtke and M. Lazzeri, "Vibrational Properties of Epitaxial Graphene Buffer Layer on Silicon Carbide," *Physical Review B* 111, no. 22 (June 2025): L220303, https://doi.org/10.1103/xk69-4n46.

81. A. Vaswani, N. Shazeer, N. Parmar et al., "Attention is All You Need," preprint, arViv, accessed November 24, 2025, https://arxiv.org/abs/1706.03762v7.

82. A. Kirillov, E. Mintun, N. Ravi et al., "Segment Anything," in *Proceedings of the IEEE/CVF International Conference on Computer Vision* (ICCV, 2023): 3992–4003.

83. J. Deng, "Imagenet: A Large-Scale Hierarchical Image Database," in *IEEE Conference on Computer Vision and Pattern Recognition* (2009): 248–255.

84. J.-L. Uslu, A. Nekrasov, A. Hermans, et al., "MaskTerial: A Foundation Model for Automated 2D Material Flake Detection," *Digital Discovery* 4 (2025): 3744–3752.

85. J. L. Uslu, T. Ouaj, D. Tebbe, et al., "An Open-Source Robust Machine Learning Platform for Real-Time Detection and Classification of 2D Material Flakes," *Machine Learning: Science and Technology* 5 (2024): 015027.

86. F. Cui, Y. Yue, Y. Zhang, Z. Zhang, and H. S. Zhou, "Advancing Biosensors With Machine Learning," *ACS Sensors* 5 (2020): 3346–3364, https://doi.org/10.1021/acssensors.0c01424.

87. F. Poletti, B. Zanfrognini, L. Favaretto, et al., "Continuous Capillary-Flow Sensing of Glucose and Lactate in Sweat With an Electrochemical Sensor Based on Functionalized Graphene Oxide," *Sensors and Actuators B: Chemical* 344 (2021): 130253, https://doi.org/10.1016/j.snb.2021.130253.

88. F. Poletti, A. Scidà, B. Zanfrognini, et al., "Graphene-Paper-Based Electrodes on Plastic and Textile Supports as New Platforms for Amperometric Biosensing," *Advanced Functional Materials* 32 (2022): 2107941, https://doi.org/10.1002/adfm.202107941.

89. L. Malavolta, I. Bracaglia, G. Cazzador, et al., "Multivariate Sensing of Ions Using Machine Learning and Composite 2D-3D Graphene Oxide-Hexacyanoferrate Electrodes," *Sensors and Actuators B: Chemical* 427 (2025): 137194, https://doi.org/10.1016/j.snb.2024.137194.

90. L. Lancellotti, A. Bianchi, A. Kovtun, et al., "Selective Ion Transport in Large-Area Graphene Oxide Membrane Filters Driven by the Ionic Radius and Electrostatic Interactions," *Nanoscale* 16 (2024): 7123, https://doi.org/10.1039/d3nr05874c.

91. Materials 2030 Manifesto, https://www.ami2030.eu/wp-content/uploads/2022/06/advanced-materials-2030-manifesto-Published-on-7-Feb-2022.pdf.

92. COM2024 98 final (https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex:52024DC0098).

93. https://research-and-innovation.ec.europa.eu/strategy/strategy-research-and-innovation/our-digital-future/european-ai-science-strategy_en.

94. https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=COM:2019:640:FIN.

95. https://commission.europa.eu/document/download/10017eb1-4722-4333-add2-e0ed18105a34_en.

96. https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32024R1781&qid=1719580391746.

97. https://ms.hereon.de/digipass/; https://cordis.europa.eu/project/id/101138510.

98. https://digital-passport.io (registration required).

99. M. Vogler, S. K. Steensen, F. F. Ramírez, et al., "Autonomous Battery Optimization by Deploying Distributed Experiments and Simulations," *Advanced Energy Materials* 14 (2024): 2403263, https://doi.org/10.1002/aenm.202403263.

100. F. Cadiou, C. Herrera, D. Atkins, et al., "A Large Scale Multi-Modal Workflow for Battery Characterization: From Concept to Implementation," preprint, arXiv, 2026, https://doi.org/10.48550/arXiv.2602.09771.

101. V. Granata, F. Liot, X. Wang et al., Implementing a Scalable, Redeployable and Multitiered Repository for FAIR and Secure Scientific Data Sharing: The BIG-MAP Archive, arXiv, 2025, https://doi.org/10.48550/arXiv.2512.15815.

102. B. H. Sjølin, W. S. Hansen, A. A. Morin-Martinez, et al., "PerQueue: Managing Complex and Dynamic Workflows," *Digital Discovery* 3 (2024): 1832–1841, https://doi.org/10.1039/D4DD00134F.

103. R. Ortega-Ochoa, A. Aspuru-Guzik, T. Vegge, and T. Buonassisi, "A Tomographic Interpretation of Structure-Property Relations for Materials Discovery," preprint, arXiv, 2025, https://doi.org/10.48550/arXiv.2501.18163.

104. N. Fisker-Bødker, E. R. Moretti, J. H. Chang, and T. Vegge, "FastCat: Autonomous Discovery of Multielement LDH Alloy Catalysts for Alkaline OER," *Advanced Intelligent Discovery* (2025): e202500138, https://doi.org/10.1002/aidi.202500138.

105. A. S. Anker, J. H. Jensen, M. González-Duque, et al., "Autonomous Synthesis of Nanoparticles With Target Scattering Patterns," *ACS Nano* 20 (2026): 6767–6782, https://doi.org/10.1021/acsnano.5c15488.

106. C. P. Kempes and D. C. Krakauer, "The Multiple Paths to Multiple Life," *Journal of Molecular Evolution* 89 (2021): 415–426, https://doi.org/10.1007/s00239-021-10016-2.

107. J. Lehman and K. O. Stanley, "Abandoning Objectives: Evolution Through the Search for Novelty Alone," *Evolutionary Computation* 19 (2011): 189–223.

108. J. K. Pugh, L. B. Soros, and K. O. Stanley, "Quality Diversity: A New Frontier for Evolutionary Computation," *Frontiers in Robotics and AI* 3 (2016): 40.

109. J.-B. Mouret and J. Clune, "Illuminating Search Spaces by Mapping Elites," preprint, arXiv:1504.04909, 2015.

110. R. Wang, J. Lehman, J. Clune, and K. O. Stanley, "Paired Open-Ended Trailblazer (POET): Endlessly Generating Increasingly Complex and Diverse Learning Environments and Their Solutions," preprint, arXiv:1901.01753, 2019.

111. K. M. Song, J.-S. Jeong, B. Pan, et al., "Skyrmion-Based Artificial Synapses for Neuromorphic Computing," *Nature Electronics* 3, no. 3 (2020): 148–155, https://doi.org/10.1038/s41928-020-0385-0.

112. J. M. Beggs and D. Plenz, "Neuronal Avalanches in Neocortical Circuits," *Journal of Neuroscience* 23, no. 35 (2003): 11167–11177, https://doi.org/10.1523/JNEUROSCI.23-35-11167.2003.

113. G. Tanaka, T. Yamane, J. B. Héroux, et al., "Recent Advances in Physical Reservoir Computing: A Review," *Neural Networks* 115 (2019): 100–123.

114. E. P. Hoel, "Causal Emergence 2.0: Quantifying Emergent Complexity," preprint, arXiv: 2503.13395v3, 2025, https://arxiv.org/abs/2503.13395.

115. S. Beckers and J. Y. Halpern, "Abstracting Causal Models," *Proceedings of the AAAI Conference on Artificial Intelligence* 33, no. 01 (2019): 2678–2685, https://arxiv.org/abs/1812.03789.

116. B. Schölkopf, F. Locatello, S. Bauer, et al., "Toward Causal Representation Learning," *Proceedings of the IEEE* 109, no. 5 (2021): 612–634, https://doi.org/10.1109/JPROC.2021.3058954.

26404567, 0, Downloaded from https://advanced.onlinelibrary.wiley.com/doi/10.1002/aisy.70496 by Universite Catholique De Louvain, Wiley Online Library on [28/08/2026]. See the Terms and Conditions (https://onlinelibrary.wiley.com/terms-and-conditions) on Wiley Online Library for rules of use; OA articles are governed by the applicable Creative Commons License