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# The Case for a Defect Genome Initiative

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**The Materials Genome Initiative (MGI) has streamlined the materials discovery effort by leveraging generic traits of materials, with focus largely on perfect solids. Defects such as impurities and perturbations, however, drive many attractive functional properties of materials. The rich tapestry of charge, spin, and bonding states hosted by defects are not accessible to elements and perfect crystals, and defects can thus be viewed as another class of "elements" that lie beyond the periodic table. Accordingly, a Defect Genome Initiative (DGI) to accelerate functional defect discovery for energy, quantum information, and other applications is proposed. First, major advances made under the MGI are highlighted, followed by a delineation of pathways for accelerating the discovery and design of functional defects under the DGI. Near-term goals for the DGI are suggested. The construction of open defect platforms and design of data-driven functional defects, along with approaches for fabrication and characterization of defects, are discussed. The associated challenges and opportunities are considered and recent advances towards controlled introduction of functional defects at the atomic scale are reviewed. It is hoped this perspective will spur a community-wide interest in undertaking a DGI effort in recognition of the importance of defects in enabling unique functionalities in materials.**

## 1. Introduction

Defined as imperfections in crystalline structures, defects are ubiquitous and drive many important properties and functionalities of materials. Defects in solid-state materials are broadly classified into point defects and extended defects. A point defect is located at a specific location or point in a crystal and involves a small number of atomic sites, while an extended defect covers a large region of the lattice (e.g., line defects involve anomalously arranged rows of atoms). The most elementary point defect is a vacancy or a missing atom. Another intrinsic point defect is a self-interstitial where an atom is transferred from a lattice site to the interstitial region. A vacancy and interstitial can form simultaneously, for example, under bombardment with energetic ions. A dopant—an impurity atom added to a crystal—can occupy a substitutional or an interstitial site. Combinations of point defects can also occur such as di-vacancies and defect complexes involving various combinations of vacancies and dopants that can stabilize under effects of Coulomb interactions and strain fields.

Unwanted defects in bulk, thin-films, low-dimensional, and/or nanoscale solids can be deleterious to material properties and device performance. For instance, the efficiency of energy conversion devices, such as light-emitting diodes and solar cells, is often adversely affected by defect-induced energy losses. Controlled introduction of specific defects, however, can induce novel electronic, quantum, energetic, and other attractive functionalities. The past two decades have witnessed rapid development of theoretical and experimental tools to advance the understanding of defects in materials. Progress in thin-film growth, microscopy, spectroscopy, and theoretical and computational modeling is leading to unprecedented, atomic-level glimpses into the nature of point as well as extended defects and enabling the deliberate incorporation of novel defect-enabled functionalities in engineering devices. To discuss our case for a Defects Genome Initiative (DGI), herein, we will focus on point defects, although the concepts introduced here are applicable more generally.

Traditional experimental and theoretical tools and protocols for defect studies are largely based on Edisonian trial-and-error approaches. For instance, a robust understanding of doping and defect-induced signatures in optical spectra of common semiconductors has taken decades to mature because many different types of defects can co-exist in a solid, depending on thermodynamic, chemical, kinetic and other environmental factors. Theoretical modeling of defect-related properties from first principles is also challenging due to limitations of available exchange-correlation functionals, and large cells needed to incorporate electrostatic and strain effects in realistic simulations.

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The Materials Genome Initiative (MGI) has ushered in a new paradigm beyond the traditional "trial and error" approaches for materials discovery[1] based on datasets generated through experimental, theoretical, and computational studies by deploying high-throughput data-driven techniques. Growing efforts to coordinate access and sharing of data and algorithms present a unique opportunity to accelerate scientific discovery in materials sciences. Data-driven materials research has been greatly accelerated by the recent proliferation of novel machine learning (ML) algorithms and rapid advances in high-performance computing and multimodal experimental workflows. The complementarity of experiments, theory, and simulation with data science has generated a new research paradigm, enabling the realization of accurate predictions and comprehensive understanding of the physical and chemical behaviors of a vast number of materials.

Herein, we argue that the viewpoint of thinking of defects as functional elements has the potential to change the way defects are studied by integrating community efforts to build up open platforms that host algorithms, methods, and databases of defects and their complex properties. We begin with a discussion of the significant advances that have been made over the last decade under the MGI across the continuum of research, development and deployment. Moving from perfect crystals to defected materials, we call for both theoretical and experimental research efforts toward a DGI to harness the power of data science and accelerate the discovery and design of functional defects in solid-state materials. We provide a brief review of the critical role of defects in functional materials, and identify goals for the DGI in terms of the objectives and actions appropriate to be undertaken by the community, including defect platform construction and data-driven functional defect design. We also discuss some of the challenges and opportunities for the DGI.

## 2. MGI and Data-Driven Materials Discovery and Design

MGI[2,3] was launched in 2011 to accelerate the discovery, design, development, and deployment of new materials at a fraction of the cost by harnessing the power of data and computational tools in concert with experiments. The MGI seeks to provide access to data-driven tools for knowledge exchange in materials research communities. As a result, tremendous advances have been made in building the foundation of the data-driven infrastructure of the MGI, including materials databases, computational and experimental tools, and prediction and analysis models driven by artificial intelligence (AI).

As a central objective of the MGI, the use of data-driven techniques offers an excellent opportunity for the accelerated discovery and rational design of functional materials. In the last decade, high-throughput computations with density-functional theory (DFT) led to the creation of representative materials databases, including the Materials Project,[4] Automatic FLOW for Materials Discovery (AFLOW or Aflowlib),[5] Open Quantum Materials Database (OQMD),[6] Joint Automated Repository for Various Integrated Simulations (JARVIS),[7] and Novel Materials Discovery (NOMAD).[8] Based on data-mining in these databases and high-throughput computational screening, new functional materials across a number of applications have been theoretically predicted,[9] and many of these materials have been realized later experimentally.[9e,10] In the fast-growing field of 2D materials, several material databases have been constructed as well,[11] and many functional 2D materials have been proposed using data-driven approaches.[12] **Figure 1** displays a timeline of the development of these platforms and repositories.

Figure 1 makes it clear that the information in materials databases over the last decade has been evolving in time from fundamental structural and thermodynamic properties of perfect crystalline materials to encompass complex electronic/magnetic/optical responses and electronic structures. Symmetry information of crystals and electronic states has been incorporated in several databases, including the topological materials database based on topological quantum chemistry[13] and its expansion to magnetic topological material systems.[14] Another trend is that the complexity of the materials encoded in the databases is growing, from perfect materials systems to heterogeneous systems such as 2D heterojunctions, surfaces, and interfaces. Development of such enhanced public materials repositories is in line with the parallel advances taking place in the development and deployment of new materials.

## 3. Why is a Defect Genome Initiative Desirable?

Macroscopic functionalities of large-scale materials systems are seldom predictable from only the electronic, atomistic, or structural features of perfect crystals. Downstream tasks in materials design and deployment of realistic systems often involve other aspects such as synthesis conditions, dynamics, excitations, and perturbations. Here, the field of semiconductors offers an obvious case study. Defects present the major source of uncertainty in the design of semiconductors with desired functionalities under realistic working conditions. However, by leveraging data-driven approaches, we are now in a position to start categorizing defects genomically. A DGI effort will help us better understand and control electronic and optoelectronic functionalities, such as carrier transport, which are key for the intentional and unintentional incorporation of defects or dopants in semiconductors.

In the past three decades, theoretical, experimental, and computational studies of defect properties in semiconductors and other crystalline solids have created a mature research field that is critical to engineering materials design. Despite rapid developments in data-driven functional materials design under the MGI, only a limited effort has been devoted to predicting and understanding defect properties in a data-driven paradigm. This could be attributed to a number of factors, including that the properties associated even with point defects involve significant complexities, such as the coexistence of many defect types, existence of multiple charge states, and the need to distinguish between deep and shallow defects. Moreover, many different computational approaches are often needed to account for interactions involving defect spins, excitons, and phonons. In fact, in fields such as quantum information science and single-atom catalysis, the defect *is* the functional device and hence it becomes the focus of research. Owing to the inherent complexity of the problem, a comprehensive understanding of point defects in a single material can take years of study.

To harness the power of data science and accelerate the discovery and design of functional defects in a diverse set of solid-state materials, we call for both theoretical and experimental research

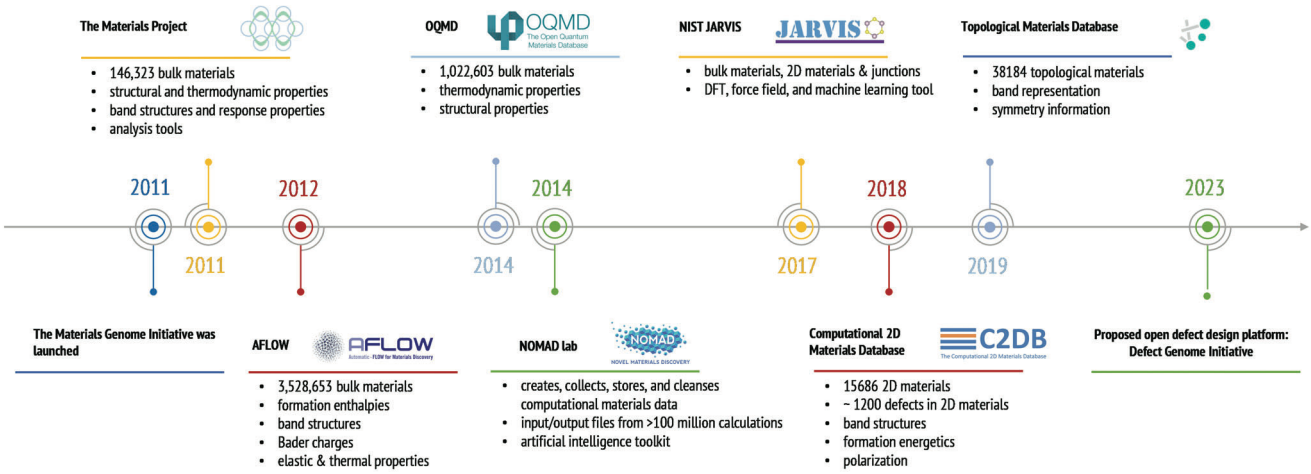


**Figure 1.** A timeline showing the development of materials data platforms and repositories in the past decade after the launch of the Materials Genome Initiative in 2011. The present proposal (in 2023) is shown symbolically at the right end of this timeline.

efforts in the material research community toward developing a DGI that is focused on constructing an open data platform for defect design. Our proposal goes well beyond earlier suggestions that incorporate defects into existing MGI frameworks,[1b] or the genomic efforts that have been applied to investigate selected sets of halide perovskites.[15] Data-driven design frameworks for studying defects are, however, emerging. For instance, the creation of defected crystal structures has been incorporated in python tools such as Python Material Genomics (Pymatgen).[16] Python Charged Defect Toolkit (PyCDT)[17] code has been especially developed to handle many charged defects. These frameworks allow reproducible modeling of charged point-defect properties, which are a foundation for automated, high-throughput defect computations envisioned under the umbrella of a DGI rich.

## 4. Defects in Functional Materials

Defects in conventional semiconductors such as silicon and gallium arsenide deserve special attention. Here, very small concentrations of defects and impurities can induce significant changes in physical properties, such as unintentional conductivity, optical absorption/emission, carrier trapping and scattering, and Fermi level pinning.[18] Native defects (vacancies and self-interstitials) control basic processes involving self- and dopant diffusion.[18b] Defects can introduce electronic levels into the bandgaps of host materials, while dopants can create acceptor and donor levels that are associated with free charge carriers for device applications. For these reasons, defect control and effective doping in traditional semiconductors and wide-bandgap systems, such as group-III nitrides,[19] has remained a vibrant field of research for decades.

Defects are often viewed as being deleterious for device performance. For instance, in silicon-based solar cells, energy levels created in the silicon bandgap can reduce photovoltaic efficiency via nonradiative recombination of charge carriers. Defects in wide bandgap systems such as group-III nitrides tend to form deep levels that trap charge carriers,[20] and cause unwanted luminescence[21] and nonradiative carrier recombination,[22] all of which act to degrade the performance of nitride-based electronic and light-emitting devices. The efficiency of light-emitting diodes and solar cells can be effectively enhanced by controlling defect populations.

An important aspect of defects concerns mechanical properties of functional materials, such as alloys and ceramics, where vacancies, dislocations, and grain boundaries are critical microstructural components[23] whose presence can degrade elastic strength and breaking point. For this reason, thermal treatment or deformation processing is often applied after casting to control the microstructure and remove defects. In high-entropy alloys, a balance of strength and ductility can be obtained via thermal treatment and/or thermo-mechanical processing, and in some cases, mechanical properties can even be enhanced through the control of point defects. For instance, vacancies can weaken hardness but improve brittleness of $Ta_5Si_3$,[24] and in B1 $VN_x$ ceramics, while the control of anion vacancy concentration allows simultaneous enhancement of hardness and toughness.[25]

Many studies explore effects of defects and how defects can be used to manipulate in-plane and out-of-plane mechanical properties of 2D materials, which present very large values of the Young's moduli.[26,27,b,28,29] Formation of undetectable defects during the synthesis process and understanding their role in controlling mechanical properties, however, remains a challenge in the field.[30] Although theoretically predicted Young's moduli for graphene and other 2D materials are in accord with experimental findings,[31] this is not the case when it comes to predicting their strengths. Topological and other defects in 2D materials offer new opportunities for manipulating mechanical properties of 2D heterostructures such as twisted bilayer graphene.[26,27c,32,33,34,35]

## 5. Defects as Sources of Functionality in Materials

Electrical, optical, and magnetic phenomena related to defects have been observed experimentally and studied theoretically in various solid-state materials, including not only traditional semiconductors but also emergent functional materials, including

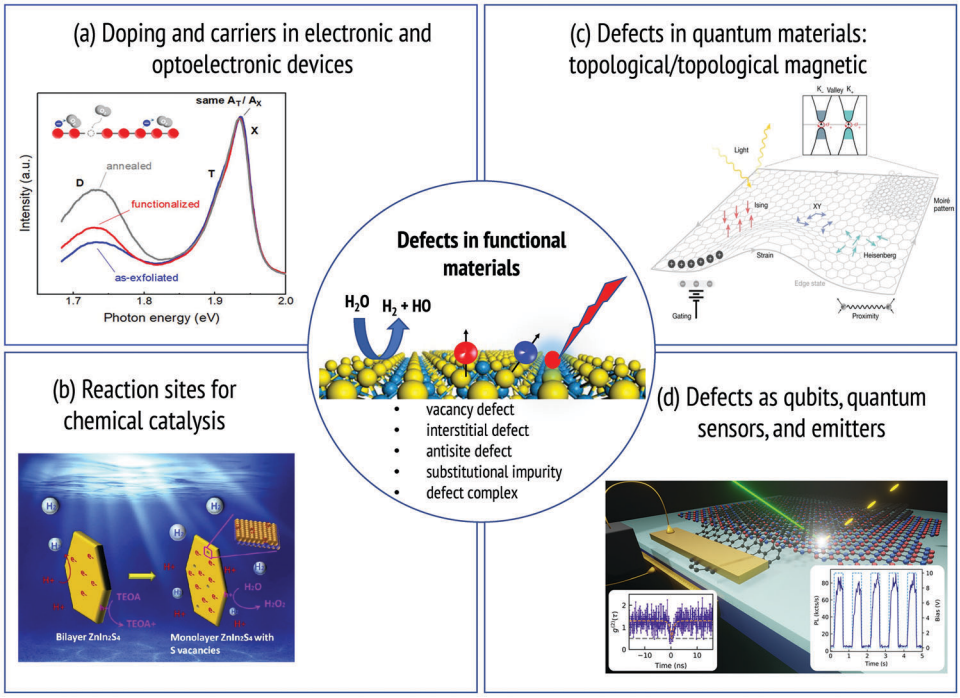


**Figure 2.** Examples of functional defects in different classes of materials: a) Doping and carriers in electronic and optoelectronic devices, b) reaction sites for chemical catalysis, c) defects in quantum materials: topological systems, superconductors, strongly correlated systems; d) electrical control of defect-based quantum emitters in a van der Waals heterostructure. a) Reproduced with permission.[40] Copyright 2020, American Chemical Society. b) Reproduced with permission.[41] Copyright 2019, Elsevier. c) Reproduced with permission.[42] Copyright 2018, Springer Nature. d) Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[43] Copyright 2022, The Authors, published by Springer Nature.

multifunctional complex oxides,[36] organic semiconductors,[37] superconductors,[38] and topological materials.[39] Although often considered detrimental for many functional applications, point defects that are either carefully designed/incorporated or naturally formed can endow materials with novel functionalities, including magnetism, superconductivity, chemical catalysis, and quantum computing and sensing, as showcased in **Figure 2**. For instance, electron spins at optically active defect centers can serve as promising qubits for quantum computing and quantum sensing even at ambient temperature.

Many excellent reviews of defects in solids and their applications are available. We refer to the review by Freysoldt et al. for first principles calculations of point defects in solids,[44] by Van de Walle for applications to group-III nitrides for solid-state lighting and power devices,[19] by Dreyer et al. on defect calculations for quantum technologies,[45] and Bassett et al.[46] for the design of quantum defects in wide-bandgap materials. We now turn to briefly review recent defect-related progress in several functional classes of materials that justify the immediate need for data-driven functional defect design.

### 5.1. 2D Material Hosts as a First Step in Building the DGI

2D layered materials as hosts for functional defects are a natural first step for building the relevant theoretical and experimental tools in connection with the DGI. 2D materials are promising candidates for the next generation nanoelectronics devices, a field that has matured significantly over the past two decades. The atomically thin planar geometry of 2D hosts presents a platform for controlled creation, manipulation, and scalability of theory-predicted functional defects with greater ease compared to the bulk hosts. Hence, we believe that investigation of point defects in 2D materials will form a natural stepping-stone toward DGI in more complex thin films and bulk materials, and/or the investigation of extended defects. It will also teach us how to step toward a DGI in hosts of lower dimensions, such as nanowires, nanotubes, nano/quantum dots, and macromolecules.

Defects can be formed in 2D materials during the growth process or created during postprocessing steps.[47] For instance, in hexagonal boron nitride (h-BN) and transition-metal dichalcogenides (TMDs), defects can be generated by a number of existing approaches,[48] and characterized and manipulated using atomic-level scanning probe techniques.[49] Although formation of point defects in materials is expected to be energetically favorable in thermodynamic equilibrium, the type and number of defects generated depend sensitively on the synthesis and growth processes and involve kinetics considerations.[50–53] In plane mechanical strain at the interface can break atomic bonds to create point, topological, or other defects, which can also induce phase transitions.[54–56] Theoretical approaches for understanding the interplay between thermodynamics, kinetics, and defects in materials include DFT based high-throughput calculations and machine learning (ML) models.[57]

From a computational point of view, due to the inclusion of a vacuum region in 2D materials simulations, both the band edge

energies of the host and the energy positions of the deep defect levels with respect to the vacuum level can be ascertained. This information is helpful for multiple tasks in a data-driven material design framework, including material discovery and ML. Also, 2D heterojunction structures create infinite possibilities for functional defect design. We envision studies of defects in 2D materials to provide a fundamental first step toward the development of the DGI.

### 5.2. Functional Defects in 2D Materials

One of the common defect types in TMDs is a topological defect resulting from vacancies at the chalcogen atomic sites, which can tune the optical, electrical, and magnetic properties of host materials. For example, pristine $WSe_2$ is a semiconductor and exhibits intriguing optical properties due to its high light-matter coupling constant,[58] making it a promising candidate for optoelectronics applications.[47] Silverman and co-workers demonstrated that the valley lifetime of excitons increases due to chalcogen atom (Se atom) vacancies in monolayer $WSe_2$.[59] Several other groups have demonstrated the effect of chalcogen vacancy on the excitonic lifetime and the optical transition process in 2D-TMDs.[40,60] Therefore, the chalcogen atomic defects in 2D-TMDs provide a good playground for future optoelectronic and quantum devices.

Fast, high-density, and energy-efficient microelectronic devices are essential components for quantum information processing and energy-efficient computing beyond Moore's era. The current challenge is to design logic and memory devices that are small enough to fit inside a transistor but large enough to allow individual, separate conduction channels to minimize energy loss during electron transport. One promising avenue is to build a transistor using TMDs. This has motivated extensive research on the electrical properties of various defects in TMDs. Wee and co-workers recently demonstrated that including oxygen atoms in place of Se atoms of $PdSe_2$ enhanced the electrical conductivity and surface absorption capacity of the electron acceptors molecules.[61] Similarly, Shawkat et al. explained that the high Se deficiency or vacancy concentration would transform $PtSe_2$ from semiconductor (2H phase) to a metal (1T).[62] This semiconductor to metal transition depends on the thickness of the materials.[62] $MoS_2$ and $WS_2$ are two prominent 2D-TMDs whose thickness dependent properties have been investigated widely. Several studies have experimentally demonstrated that the carrier mobility of monolayer $MoS_2$ and $WS_2$ depends on the concentrations and types of defects.[63] Hence, introduction of appropriate defects is rapidly gaining interest for modulating electronic functionalities of 2D materials.

Ultrathin ferroelectric materials are essential components of energy-efficient, high-density electronic devices. Defect-induced ferroelectricity in nonpolar 2D-TMDs has been proposed recently as a means to go beyond symmetry-dependent ferroelectricity in the 2D-TMDs.[64,65] Surface iridium (I) vacancy has been suggested to induce switchable out-of-plane electric polarizations in $CrI_3$.[66] First-principles calculations also demonstrate that point defects in $SrTiO_3$ could introduce ferroelectricity.[67] Robust ferroelectricity has been reported in the nonpolar semiconductor $\alpha Ga_2Se_3$ due to the native Ga vacancy at the asymmetrical sites.[65] These results indicate that defect-induced ferroelectricity is an attractive route for DGI, especially in nonpolar 2D-TMDs.

Developing environmentally friendly catalysts is of great importance, a field that faces challenges due to stability, poor charge transfer kinetics, appropriate bandgap values, and cost.[68] Defect engineering is one effective pathway for tuning the properties, overcoming these challenges, and increasing the photocatalysts' efficiency. Several strategies, such as creating oxygen and chalcogenide vacancies have been proposed.[69] $H_2$ evolution can be enhanced using sulfur vacancy in monolayer $ZnIn_2S_4$.[41] The hydrogen evolution performance of 2H-$MoS_2$ nanosheets is believed to be due to the sulfur vacancy (point defects and sulfur stripes).[70] Defects can enable single-molecule photocatalytic dynamics on the surface of InSe.[71] Hence, a DGI-based approach to advance defect-induced catalysis is another attractive avenue.

Future generations of nanodevices will benefit tremendously from magnetic 2D materials. Beyond intrinsically magnetic 2D materials,[72] defect engineering provides an additional pathway for introducing magnetism. Transition metal vacancies have been suggested to induce magnetism in $PtSe_2$[73] and $ReSe_2$[74] chalcogenides, and Se-deficient line defects can potentially lead to ferromagnetism in $VSe_2$.[75] Fluorination can induce phase transition in nonmagnetic 2H-$WS_2$ to magnetic 1T phase of $WS_2$,[76] where defect site and interlayer spacing have been suggested to play important roles. These studies justify a DGI-based approach for exploring defect-induced magnetism in 2D materials.

The ongoing second quantum revolution calls for exploiting unique quantum features of materials in transformative new technologies for quantum computation and quantum information sciences.[77] Here point defects in wide-bandgap materials can host coherent quantum states that can be addressed optically and electronically. Quantum superpositions of localized defect states can be prepared and maintained for a long enough period for quantum computing and communication applications. These defects in wide-bandgap materials have been called "quantum defects," and are considered building blocks for various quantum applications, including quantum computing, quantum sensing, and quantum communication.[78] The most well-studied quantum defect is the NV (nitrogen-vacancy) center in diamond. The electronic spin state of the NV center can be initialized, controlled, and read out even at room temperature.[79] Each NV center also constitutes a quantum register of electron and nuclear spins and it can be used to perform quantum operations, including teleportation[80] and error correction.[81] Spurred by the potential to identify other spin qubits with improved properties and new functionalities, exploration of quantum defects has grown rapidly,[82] including the Si–V center in diamond,[83] defects in SiC,[84] and rare-earth ions in oxides.[85] Other defects have been suggested as promising qubit candidates in SiC[86] and AlN.[87]

Quantum defects in ultrathin 2D systems, which could be controlled by electrostatic gating, offer potential advantages over NV centers, including optical indistinguishability[88] and high brightness.[89] In recent work, antisite defects in monolayer TMDs have been identified as viable spin qubits.[90] Beyond spin qubits, the desirable optical transitions created by deep

in-gap states or defect-bound excitons in 2D materials can be utilized as quantum emitters or even single-photon emitters (SPEs) for quantum communication applications. Defects in h-BN show promise as polarized and ultrabright SPEs at room temperature.[91] Carbon-vacancy complex $C_B$–$V_N$ in h-BN has been proposed as a quantum emitter as well as a spin qubit,[92] and its optical signature has been measured.[93] NV centers in diamond[78c] and other defects in wide-bandgap materials such as ZnO[94] and GaN[95] have been recognized as SPEs. On the other hand, the difficulty of integrating traditional wide-bandgap materials into quantum devices by lithography techniques motivates the exploration of other material systems, including 2D materials. SPEs in 2D materials, including $WSe_2$[96] and h-BN[89,97] have been observed. The emission source in h-BN has been identified to be carbon related.[98] As an exciting and emerging research field, progress beyond initial experimental demonstration of quantum defects in 2D materials such as h-BN and TMDs calls for accelerated discovery and rational design of novel quantum defects under the MGI for quantum computing and information processing and opens up a huge opportunity for a DGI-based approach for their discovery.

### 5.3. Modification of Material Properties by Defects

Chemical substitution has proven to be one of the most efficient strategies for tuning material properties.[99] Thermodynamically stable materials such as 2D-TMDs are ideal playgrounds for examining the effect of chemical substitution in crystals,[100,101] and the DGI needed to guide the community holistically on the impact of defect engineering. For example, substituting carbon atoms with boron and nitrogen atoms opens a bandgap in graphene[102] but also adversely impacts its conductivity,[103] mechanical properties, and chemistry.[99a,b] Another important aspect will be to provide guidance on different pathways to chemical substitution.[104] For example, chemical substitutions in $MX_2$ are more effective than charge transfer and doping.[99c] Recent experimental reports demonstrate that chemical substitution of Bi by Sb is an effective avenue for realizing the quantum anomalous Hall effect (QAHE) in bulk $MnBi_2Te_4$ (MBT).[105] and tuning the Fermi level toward the bulk bandgap.[106] Chowdhury et al. have shown that chemical substitutions could tune MBT's topological states and electronic properties.[107]

Intrinsic point defects (vacancies, interstitials, and antisites) form during the growth of 2D materials and can significantly change their properties. A common intrinsic defect in TMDs is the chalcogen vacancy.[108] The formation of vacancies in TMDs depends on the chemical potentials,[108c] and their properties can be significantly altered via vacancy defects. For instance, Wang et al. demonstrated the constructive effects of chalcogen vacancies in $MoS_2$ on catalytic properties.[109]

ML algorithms can address effects of chemical substitutions to predict the phase stability of solid-state materials[110] and the evolution of properties such as magnetic behavior[100a] and proton conductivity.[15] An exploration of the defect genome in 2D materials, including the complex effects induced by intrinsic point defects as well as substitutional defects, can be expected to significantly enhance our ability to control novel materials functionalities.

## 6. Defect-Data Platform Construction and Data-Driven Defect Design

### 6.1. Defects by Design—Defect Data Generation, Database Construction, and Data Sharing

Design of functional defects in a data-driven paradigm faces immediate theoretical, computational, and experimental challenges, so that the execution of the DGI will require combined efforts of the entire materials research community. We turn now to discuss the challenges and opportunities associated with the development of the DGI. Targeted design of functional defects is a difficult task since many design criteria must be satisfied simultaneously. Lack of data standards and centralized repositories in defect research hinders reproducibility and discovery. For instance, the current design of defect-spin-based qubits largely relies on trial and error, although a set of criteria and strategies for the discovery and design of quantum defects have been proposed years ago.[46,111] Going beyond identifying individual defect candidates in a small set of materials faces several questions: How to construct a uniform and open defect design platform and connect it seamlessly to existing data-driven material design infrastructure? What defect properties can be or should be included in both computational and experimental input? How to identify functional defects in the vast solid-state compound space?

Another important task within the DGI is the incorporation of common design rules for handling specific defect properties in a data-driven design framework. As an illustrative example, we consider the defect formation energy which, in a relatively simple picture, involves contributions from both strain-dependent terms and electron–electron interactions.[112] Along this line, Frey et al. discuss random-forest-based ML models that encode information about local relaxation and electronic interactions to classify deep centers and predict formation energies.[112a] Empirical models have also been developed to predict the formation energetics of point defects (especially vacancies) that take into account both electronic and thermodynamic features.[113,114] One of the first electronic features invoked involved partial charges on metals atoms,[113] which are strongly correlated with electrostatic attraction of the atoms to $O^{2-}$ and hence to the formation energies of vacancies. Later, Deml et al. proposed a more comprehensive model for vacancy formation energies of 45 metal oxides[114a] by introducing two new electronic features (O 2p band center and bandgap) and two thermodynamic features (electronegativity and formation enthalpy of the host).[114a] A recent study of defects in $La_{1-x}Sr_xBO_3$ perovskites (B = Cr, Mn, Fe, Co, and Ni) shows that the formation enthalpy of oxides and the bandgap are strongly correlated with oxygen vacancy formation energies.[115]

Notably, design rules have been proposed to correlate the presence of defects to address the tendency in certain materials to form stable off-stoichiometric compositions. A recent example is the valence-balanced rule for the discovery of half-Heusler alloys with defects by Anand et al.[116]: the ground states of both stoichiometric and off-stoichiometric alloy structures are found to have a common net valence of 0 within Zintl chemistry, and the valence-balanced rule (with the 18-electron rule as a special case) predicts the formation of off-stoichiometric phases with a large number of vacancies.[116]

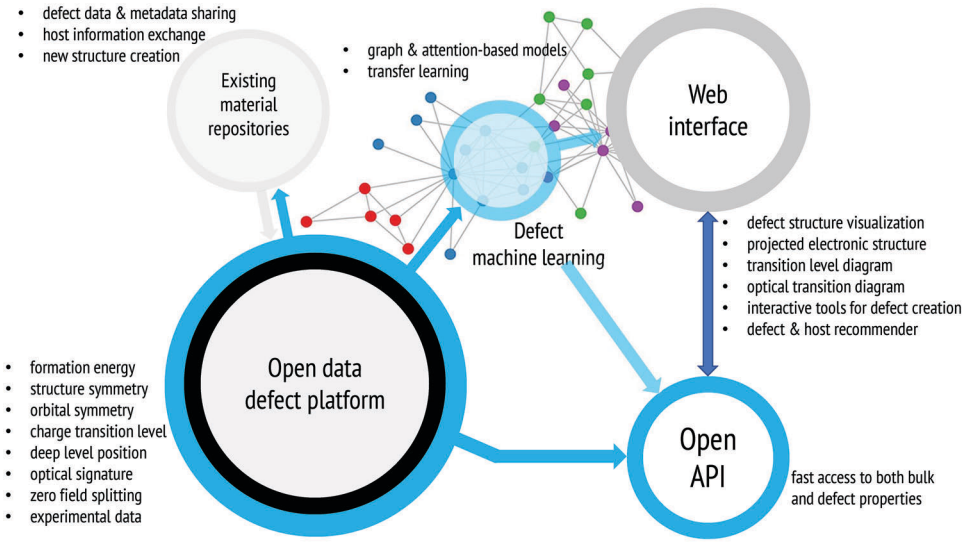


**Figure 3.** A proposed open-data defect design platform under the Defect Genome Initiative (DGI), including a defect database with comprehensive defect properties and connected to existing databases, defect machine learning (ML) models, web interface, and an open API interface.

Understanding and incorporating defect design rules in ML will be important for realizing feature engineering and for enhancing the model training process of shallow learning models. Empirical rules could also be incorporated in more complex learning frameworks involving multifidelity models that combine rules-based simple models with deep learning models. For the development of generative learning models, design rules such as the valence-balanced rule, could serve as a constraint to reduce the exploration space of candidate structures.

An open data platform for defect design will rely on the development of three components: defect data creation and sharing, ML of defect properties, and functional defect design, as outlined schematically in **Figure 3**. There are many existing challenges related to high-throughput defect computations. It is well known that the energetic and charge stability of defects should be evaluated by computing formation energies in different charge states.[117] Relative formation energies of charge states as a function of the Fermi level determine the ground-state charge state and the thermodynamic transition levels, from which we can evaluate the conditions when these defects host desirable functionalities. Therefore, possible charge states of a given defect in each host material need to be carefully determined to avoid the superficial effects of adding electrons or holes in bulk materials instead of at the defects. The choice and proper use of finite-size correction approaches, however, have yet to be fully automatized. The continued development of automatized tools to estimate possible charge states and evaluate corrections is critical to high-throughput defect data generation.[90]

A challenge for the defect-by-design problem is the lack of publicly available data for defected materials, which can be addressed by utilizing existing and developing new tools for high-throughput defect computations. Several existing python-based open packages can be adopted for defect-data creation, analysis, and storage. For instance, pymatgen includes modules for structure generation, symmetry analysis, and electronic structure analysis. PyCDT contains specialized modules for creating and analyzing defect properties (formation energy and transition levels). Recently, an extension to pymatgen (called pymatgen-analysis-defects) has been developed for performing defect analysis. An automatized computational pipeline can be created utilizing the Atomate and Atomate2 packages[118] to customize workflows, manage high-throughput defect computations, as well as store computed defect data in centralized materials databases.

A comprehensive point-defect database under the DGI should include all five types of point defects: cation/anion vacancies, cation/anion antisites, substitutional impurities, interstitial defects, and vacancy-impurity complexes. To guarantee the reproducibility of defect data, computational parameters for the employed DFT calculations, as well as the choice of supercell size and charge-state correction should be clearly stated and stored together with other original data and metadata for all defect systems. Automatized defect analysis tools should be developed to identify localized defect states in the electronic band structures based on wavefunction localization. These defect states should be labeled with atomic-orbital and local-symmetry information in terms of irreducible representations. To amplify the impact of the defect-design platform, all computed defect data should be connected to existing public materials repositories such as the Materials Project, Aflowlib, and NOMAD.

Thanks to recent advances in computational methodologies, a powerful toolkit is available for predicting complex phenomena related to defects, including formation energetics, ground, and excited states, optical properties, and nonequilibrium relaxations involving triplet and singlet states.[119] Leveraging the dramatic growth in computational power, defect modeling studies span from standard DFT with local or semilocal functionals (such as local density approximation (LDA) and generalized gradient approximation (GGA)) to meta-generalized gradient approximation (meta-GGA) (such as strongly constrained and appropriately normed (SCAN) and the second revision of the SCAN meta-GGA functional (r2SCAN)), hybrid functional,[120] many-body perturbation theory,[121] and even quantum Monte

Carlo.[122] For instance, defects for quantum technologies have been investigated using constrained DFT (CDFT) calculations and many-body perturbation theory within the GW approximation, and the Bethe–Salpeter equation (BSE) has been applied to study the excited states of defect spin qubits in diamond,[123] SiC,[124] and h-BN.[92b,125] Triplet–singlet intersystem crossing process of $NV^-$ in diamond has been studied within a rigorous theoretical framework.[126] Spin coherence phenomena have been explored theoretically for bulk compounds[127] and 2D materials.[128] Progress has been made in addressing the artificial electrostatic interaction in both bulk[129] and 2D systems,[92b,130] which paves the way for high-throughput defect computations.

Creation of materials platforms for defect studies has been an ongoing effort.[119b] A Quantum Point Defects (QPOD) 2D materials database[131] was recently released and linked to the C2DB 2D material database.[132] QPOD includes about 500 vacancy and antisite defects in different charge states, hosted by 82 different 2D semiconductors and insulators. High-throughput DFT calculations using the semilocal PBE functional were performed by using a defect workflow built around Atomic Simulation Recipes (ASR), which include calculations of formation energies, charge transition levels, equilibrium defect and carrier concentrations, point group symmetry labels of in-gap defect states, transition dipole moments, hyperfine couplings, and zero-field splittings. This work is an important step toward constructing an open data platform with centralized defect databases, which will greatly accelerate the discovery and design of functional defects. Recently, the Jarvis-FF materials database[133] released the formation energy data for ≈1000 vacancy defects, which is another high-throughput defect data generation/dissemination effort.

As we climb the Jacob's ladder for increasingly accurate electronic structures of materials,[134] the computational cost of defect calculations will increase dramatically, and eventually high-throughput computations of defects in solids will likely become impractical. This challenge is common to heterogeneous materials systems involving surfaces, interfaces, and heterostructures. Within the DFT framework, the defects-in-solids problem in heterogeneous materials requires a density functional that can accurately capture both the bulk and surface electronic structures as well as the localized defect states. The hybrid functional has been applied to many defect systems but it is known to be relatively expensive for high-throughput computations. A reasonable approach will be to evaluate the relaxed geometries and electronic structures of point-defect systems using high-throughput computations based on a meta-GGA functional such as SCAN (or r2SCAN).[135] To push the DGI forward, it will be important to develop efficient computational models to accurately describe electronic structures and responses of localized defect states in crystalline environments. The continued development of meta-GGA functionals that satisfy physical constraints[135b] is a promising direction. Another strategy is to design effective embedding methods to treat defect states and hosts on different theory levels separately and then effectively merge the simulation results. Quantum embedding method[136] is one of the promising approaches to accurately describe the correlation effects associated with localized defect states in crystals.

Effective data-driven functional defect design relies on the incorporation of domain knowledge. Defect design and discovery is inherently complex because many native defects and potential impurities must be investigated to construct a comprehensive picture of defects in even a single material. Notably, defect formation energetics as a screening criteria does not need to be applied strictly, as desirable defects could be created via nonequilibrium processes.[48] Beyond the basic structural and energetic properties, complex properties of defects should also be included in the databases to allow the development of effective defect design strategies for various specific applications. For instance, it will be very useful to take advantage of local site-symmetry and orbital-symmetry information that are highly relavant to defect properties. Local symmetry of a defect defines its orbital and spin structures through the interplay of molecular orbitals and group theory considerations. It is therefore crucial to develop symmetry-based design principles, which will dramatically reduce the defect-host design space. The storage and sharing of the symmetry information in the defect databases should be carefully designed with both an API and a user-friendly web interface.

Eventually, the vast amount of open defect data created under the DGI combined with the associated defect analysis tools will provide a data-driven paradigm to accelerate the identification of promising defect-host candidates for functional applications. We emphasize that computations on defected systems are time consuming because they involve large supercells. In a recent study, Mannodi-Kanakkithodi et al.[137] provide an estimate for the cost of high-throughput defect computations in traditional semiconductors: approximately 32 million core hours would be required for structural optimization and prediction of 12474 defect properties at the GGA-PBE level.[137] Development of effective ML models is, therefore, needed for addressing key properties such as formation energies and defect concentrations, and classifying the deep versus shallow nature of various defects. The defect datasets created under the MGI will provide essential training samples for developing ML models especially designed to predict the complex properties of point defects as discussed below.

### 6.2. Construction of Defect Genome by Machine Learning

ML has upended many facets of everyday life, from speech recognition and generation to advertising and entertainment recommendations.[138] Physics, chemistry, and materials sciences are no exception. ML provides a novel opportunity to reduce computational costs and speed up the pace of materials discovery by utilizing data-driven paradigms.[139] Combined with data-driven technologies, ML has become a powerful tool in materials research.[140] Supervised learning has proven effective in materials property predictions, including phase stabilities of molecules and crystals,[139a,141] crystal structures,[140c,142] electronic features such as bandgaps[140e,143] and densities of states,[144] effective potentials,[139b] effective Hamiltonians,[145] and energy functionals.[146]

The renaissance of AI is largely driven by breakthroughs in feature learning boosted by deep learning techniques.[147] The existing deep learning techniques, such as convolutional neural networks (CNN),[148] however, are limited in handling the great diversity of materials that cannot be described by rigid grids. Solid-state materials require many tiers of fundamental information involving atoms and electrons and their complicated interactions

in the presence of local and global symmetries. The recent development of graph-based deep learning, especially the graph neural network (GNN),[149] offers a novel tool for creating an innovative representation of crystal structures.[150] Atom-based GNN has been applied to predict formation energetics and features of electronic structures of materials, and shows considerable improvement over shallow learning models.[151] A motif-based graph neural network has been developed to include higher-tier materials information within the GNN framework.[152] Other graph structures, such as line graphs[153] and GNNs with three-body interactions,[154] have been applied to property predictions and creating interatomic potentials. Recently, a GNN leveraging the strength of Transformer architectures and incorporating equivariant features based on irreducible representations has been proposed.[155]

Defects add another tier of materials information in the design of ML frameworks, so that the development of novel ML frameworks that are tailored for defects in crystals is needed to accelerate functional defect design. Unlike the solid-state materials and molecules, however, we do not currently have public databases with large amounts of labeled data for defected systems. This data limitation makes it challenging to apply deep learning models to the defect problem and, as a result, the existing ML work is limited to shallow learning models.[112a,156] This data challenge can only be addressed by community-wide efforts toward defect-data generation, collection, and sharing.

A critical task for the DGI is the creation of comprehensive and carefully labelled datasets for training complex ML models that are specifically designed for defects. In a recent work,[157] a dataset was created for defects in 2D materials containing 5933 defect configurations for $MoS_2$ and $WSe_2$, with the latter relaxed using the PBE functional, indicating that datasets with samples on the order of ten thousand are currently achievable. To effectively catalog point defects in a large variety of materials, attention- and/or contrastive-learning based classification models may be designed within a graph-based learning framework. It is expected that ML models with DFT-level accuracy will be developed for specific types of defects. Models that consider local symmetries, such as the equivariant neural networks, would help improve the accuracy of property predictions using training datasets of affordable size. GPU-based computations will substantially accelerate the creation of high-quality defect data based on higher-level functionals such as the hybrid and meta-GGA functionals.

Another fundamental question regarding ML for defect systems is: How much of the information learned from pristine systems can be transferred to address defected systems? Understanding model-transferability will be important for handling of learning of defect properties more effectively. Novel ML models are needed to extend the predictability of macroscopic properties of materials with point defects based on both global and local electronic/atomic level information. Accordingly, efforts should be made to develop learning tasks at various levels to establish ML models based on transfer learning to utilize the rich information available on host materials. Pretraining on GNNs in a self-supervised manner has been proposed recently, and self-supervised learning (SSL) has been widely used in image representation learning,[158] natural language processing,[159] and applied in molecular ML.[160] SSL is a way to augment data through a pretraining process by generating easy-to-get labels from unlabeled data and learning informative representation, which will improve the performance of downstream tasks. Novel self-supervised learning architectures can be designed to connect to the DGI to circumvent the data challenge by generating a large amount of labeled "easy-to-get data", which can then be used in the pretraining process to improve the performance of the deep learning models for defects.

It has been recognized that electronic structures are much more complicated targets than formation energetics for deep learning models.[151,152] To tackle the deep-learning problem for defected systems, especially for quantum defects, two questions must be addressed. (1) How can local interactions between the point defects and their surrounding atoms be modeled efficiently? And (2), how can we learn material properties at the electronic structure level? Notably, there have been recent developments of novel ML architectures, including self-supervised learning[158] and attention-based learning including graph attention network (GAN)[161] and graph transformer network.[162] Attention-based learning models[161] provide a unique opportunity for ML models that are especially designed for point defects to pay special attention to local interactions involving defects and their neighboring atoms. The development of state-of-the-art ML models with graph attention (e.g., the graph transformer network[162]) and orbital information is promising to realize the learning and prediction of complex electronic features of point defects in solid-state materials, such as the deep levels and their splittings.

## 7. Promising Techniques for Fabrication and Characterization

Introduction of impurities and defects into crystals has remained a key enabler in various advanced technologies. Techniques such as ion implantation, milling, and chemical etching have been utilized routinely for defect/impurity modification in materials of macroscopic scales. These techniques can introduce controlled levels of dopants and/or defects over large areas. For example, controlled doping of impurities in semiconductors has led to the development of transistors, solar cells, and lasers, which have widely impacted modern life. Defect-induced mid-gap states have provided new paradigms for catalytic and optoelectronics applications, and for designing optically addressable spin qubits. As we move from nanoscale to atom-scale, experimental techniques in parallel will need increased control and precision.[163] In the context of the DGI, experimental techniques will need to insert specific species of impurities and defects at specific locations with atomic precision in specific hosts, to obtain the required functionalities. In this connection, we discuss below several promising techniques that could be developed further to achieve ultra-precise controlled introduction of functional defects at the atomic scale. The combined use of theory, simulation, and AI models will help expand the reach of the techniques beyond their current capabilities.

### 7.1. Controllable Manipulation of Atoms

One of the earliest and most promising approaches for manipulating individual defects was introduced in the field of scanning

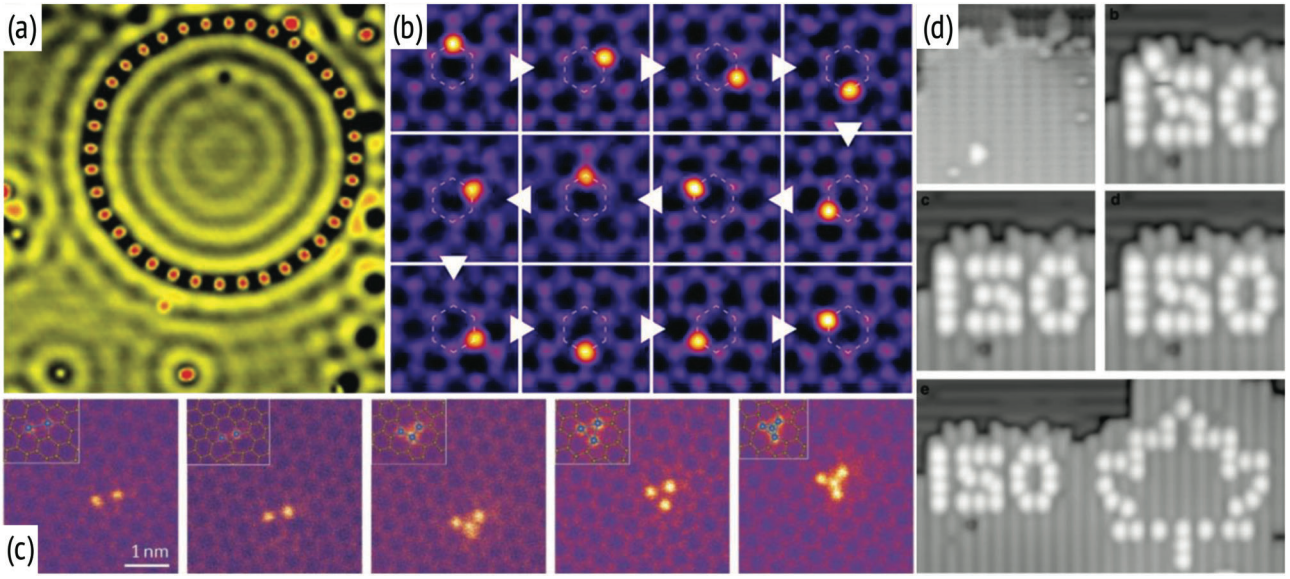


**Figure 4.** Promising experimental approaches for engineering, manipulating, and assembling atoms: a) scanning tunneling microscopy (STM), b) scanning transmission electron microscopy (STEM), c) electron-beam irradiation, d) hydrogen depassivation lithography. a) Reproduced with permission.[164] Copyright 2014, IOP Publishing. b) Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[165] Copyright 2018, The Authors, published by American Chemical Society. c) Reproduced with permission.[166] Copyright 2019, Springer Nature. d) Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[167] Copyright 2018, The Authors, published by Springer Nature.

tunneling microscopy (STM). Crommie et al.[168] used an STM to arrange a group of 48 iron ad-atoms on a Cu surface in order to confine electrons to artificial geometries and demonstrated the celebrated "quantum corrals" (**Figure 4**a). Such an "atom-by-atom" assembly has advanced much since then,[164,169] whereby an STM tip can "touch" and "take" atoms, and perform lateral and vertical manipulations. Atoms can be extracted by crashing tips into bulk materials (e.g., Ag or Au) or breaking molecules on substrates.[170] Depending on the atom–substrate and atom–tip interaction strengths, and the nature of the final destination, the STM can procure, move, and assemble atoms into predetermined structures. In the context of the DGI, the STM approach certainly appears to provide an encouraging method for atom/impurity manipulation. However, there are limits to what an STM can do. First, STMs rely on tunneling current for their operation and are thus mostly limited to conductive surfaces. Second, generation and manipulation of point defects (e.g., vacancies) using an STM is still a challenge, although encouraging new results have been achieved: for example, selenium-vacancy defects ($V_{Se}$) could be manipulated using an STM near the surface of $PdSe_2$.[171] These demonstrations, however, have been limited to specific substrates and types of atoms so far, and much further work is needed to understand how STM can become a versatile tool for engineering theory-predicted defects on functional substrates.

### 7.2. Structural Modifications and Evolution of Defects

While the STM can manipulate atoms across a surface, it is limited when it comes to the creation or manipulation of defects. Here, controlled ion implantation and electron irradiation remain more attractive methods for on-demand defect generation in materials. Control of irradiation energy has been shown to generate specific types of defects in materials, such as the Stone–Wales defect in graphene.[172a] Ion implantation also has the advantage that it can exploit a broad range of elements, and in principle, it can be applied to any material. It will be interesting to eventually encompass within the scope of the DGI the ability to predict the type of defects that are likely to be generated via irradiation with various energetic ions and electrons under different conditions. Such a database will guide experimentalists and reduce time to discovery for tailored defect engineering. Through a combination of theory, modeling, and experimental characterizations, data-driven approaches could be developed to achieve real-time control of defect engineering protocols.

In contrast to ion-implantation, which can access many different ionic species, e-beam irradiation only involves the impinging electrons, and electron energy is the main free parameter involved in defect generation. Recent advances in aberration corrected TEMs have enabled electron beams to focus on very few atoms to engineer and manipulate single defects and atoms with unprecedented control. Defects such as vacancies can be created, and ad-atoms can be moved with reproducible control under appropriate e-beam fluxes. In their 2016 comment, "Fire up the atom Forge," Kalinin et al. envision how TEM could be used to "build quantum materials from scratch," and lay down a pathway for the realization of precision defect engineering in quantum materials[172b] through a combination of e-beam control, real-time monitoring, and AI-assisted real-time adjustment of beam control. In combination with the DGI for engineering theory-predicted new defects, such an approach would provide valuable experimental validation for building a much-needed defect-manufacturing database. Figure 4b,c showcases the

atomically precise control and assembly of defects using e-beam irradiation.

### 7.3. Fabrication of Defect-Enabled Devices and Applications

Applications such as catalysis will perhaps draw immediate benefits from the DGI effort, since often the position of defects in their macroscopic hosts need not be atomically precise. For example, bulk cryo-mechanical treatments followed by controlled chemical, thermal, and electrochemical processes lead to large numbers of point defects in a host such as h-BN. These defects can be activated by charge/discharge with a metal Li counter-electrode or other individual metal atoms for improved electrochemical technologies.[173] More controlled defect generation will benefit nanoscale quantum point-defect-engineered applications, such as single photon sources for quantum information systems. Here, DGI-guided discovery will help identify other hosts suitable for such applications.

Developing methods for fabricating sophisticated quantum devices, for example, scalable arrays of defect qubits on a specific host, such as NV defect arrays in diamond, or antisite defect arrays in 2D materials, continue to remain a challenge. DGI-guided choice of hosts could help narrow down the search for specific approaches for engineering such precision quantum devices. On the experimental side, in addition to STM-manipulation and implantation/irradiation methods, more scalable techniques will need to be developed. One promising avenue is hydrogen depassivation lithography,[167,174] an STM-guided lithography/etching technology that can position arrays of P atoms on Si down to a few-nanometer accuracy. This technology could potentially improve its accuracy to single atoms scale (Figure 4d) and expand its reach to handle other atoms and substrates. A concerted effort combining theory, experiment, and AI/ML will enable defect-based devices and applications to become a practical reality more rapidly.

### 7.4. Experimental Characterizations of Defects

As new defects with novel functionalities are designed and fabricated, a range of experimental techniques will be required to characterize them. Structural information can be obtained from atomically precise techniques such as scanning transmission electron microscopy (S/TEM) and STM. High-resolution electron tomography[175] can now provide 3D information on crystal defects with unprecedented accuracy,[176] as a powerful tool for correlating 3D atomic defects and their electronic properties obtained by first-principles calculations.[176] STM can additionally provide the electronic density-of-states information for mapping electronic properties of defects. Surface probes such as scanning NV magnetometry provide a new approach for characterizing magnetic properties at the nanoscale. This technique, however, will likely need improvements in sensitivity and spatial resolution for characterizing single defect structures. For characterizing larger, bulk-defected materials, traditional approaches such as deep-level transient spectroscopy, X-ray photoelectron spectroscopy (XPS), and nonlinear spectroscopic techniques[177] are appropriate. For energy applications, electrochemical characterizations, such as cyclic voltammetry and impedance spectroscopy will allow access to redox properties of defect-loaded hosts. Recent development of techniques based on inelastic X-ray scattering as a spectroscopic tool for imaging redox orbitals and dopants at the atomic level offer novel opportunities for rational design of battery and other materials.[178] Nanoscale and bulk structure–function correlations so obtained will provide a robust basis for achieving iterative improvements in the prediction–synthesis–characterization loop of the DGI.

## 8. Outlook

Substantial advances have been made over the past decade under the umbrella of the MGI toward creating, sharing, and using centralized materials data for the discovery and design of new materials. Given the key role defects play in driving many functional properties of materials, the time is ripe for undertaking a DGI effort to harness the full power of data science and accelerate the discovery and design of defected solid-state materials. The construction of an open defect data platform through high-throughput computations and web interface construction will make it possible to achieve a comprehensive theoretical and experimental understanding of how defects form in crystals and how the associated defect structures and properties are correlated. Novel machine-learning models that are specifically designed for point defect systems, which would be a natural starting point, will enable rapid and accurate prediction of electronic structures and properties of defects. Parallel development of experimental techniques, including atomic-scale characterization of defect systems and controllable creation of defects, will close the gap between theory and experiment to advance defect studies. The proposed DGI effort will provide a comprehensive umbrella for the rational design and accelerated realization of functional defects for quantum information, energy and other applications.

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## Conflict of Interest

The authors declare no conflict of interest.

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[1] a) A. White, *MRS Bull.* **2012**, *37*, 715; b) J. J. De Pablo, N. E. Jackson, M. A. Webb, L.-Q. Chen, J. E. Moore, D. Morgan, R. Jacobs,

T. Pollock, D. G. Schlom, E. S. Toberer, J. Analytis, I. Dabo, D. M. Delongchamp, G. A. Fiete, G. M. Grason, G. Hautier, Y. Mo, K. Rajan, E. J. Reed, E. Rodriguez, V. Stevanovic, J. Suntivich, K. Thornton, J.-C. Zhao, *npj Comput. Mater.* **2019**, *5*, 41.

[2] *Materials Genome Initiative for Global Competitiveness*, National Science and Technology Council, Washington, D.C., USA **2011**.

[3] Materials Genome Initiative, https://www.mgi.gov/about, **2021**.

[4] A. Jain, S. P. Ong, G. Hautier, W. Chen, W. D. Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder, K. A. Persson, *APL Mater.* **2013**, *1*, 011002.

[5] a) S. Curtarolo, W. Setyawan, G. L. W. Hart, M. Jahnatek, R. V. Chepulskii, R. H. Taylor, S. Wang, J. Xue, K. Yang, O. Levy, M. J. Mehl, H. T. Stokes, D. O. Demchenko, D. Morgan, *Comput. Mater. Sci.* **2012**, *58*, 218; b) S. Curtarolo, W. Setyawan, S. Wang, J. Xue, K. Yang, R. H. Taylor, L. J. Nelson, G. L. W. Hart, S. Sanvito, M. Buongiorno-Nardelli, N. Mingo, O. Levy, *Comput. Mater. Sci.* **2012**, *58*, 227.

[6] J. E. Saal, S. Kirklin, M. Aykol, B. Meredig, C. Wolverton, *JOM* **2013**, *65*, 1501.

[7] K. Choudhary, K. F. Garrity, A. C. E. Reid, B. Decost, A. J. Biacchi, A. R. Hight Walker, Z. Trautt, J. Hattrick-Simpers, A. G Kusne, A. Centrone, A. Davydov, J. Jiang, R. Pachter, G. Cheon, E. Reed, A. Agrawal, X. Qian, V. Sharma, H. Zhuang, S. V. Kalinin, B. G. Sumpter, G. Pilania, P. Acar, S. Mandal, K. Haule, D. Vanderbilt, K. Rabe, F. Tavazza, *npj Comput. Mater.* **2020**, *6*, 173.

[8] C. Draxl, M. Scheffler, *J. Phys Mater.* **2019**, *2*, 036001.

[9] a) S. Curtarolo, G. L. W. Hart, M. B. Nardelli, N. Mingo, S. Sanvito, O. Levy, *Nat. Mater.* **2013**, *12*, 191; b) G. Hautier, A. Miglio, G. Ceder, G.-M. Rignanese, X. Gonze, *Nat. Commun.* **2013**, *4*, 2292; c) Y. Wu, P. Lazic, G. Hautier, K. Persson, G. Ceder, *Energy Environ. Sci.* **2013**, *6*, 157; d) I. E. Castelli, T. Olsen, S. Datta, D. D. Landis, S. Dahl, K. S. Thygesen, K. W. Jacobsen, *Energy Environ. Sci.* **2012**, *5*, 5814; e) A. Jain, Y. Shin, K. A. Persson, *Nat. Rev. Mater.* **2016**, *1*, 15004; f) M. G. Vergniory, L. Elcoro, C. Felser, N. Regnault, B. A Bernevig, Z. Wang, *Nature* **2019**, *566*, 480; g) M. G. Vergniory, B. J. Wieder, L. Elcoro, S. S. P. Parkin, C. Felser, B. A Bernevig, N. Regnault, *Science* **2022**, *376*, eabg9094.

[10] a) L. Zhou, Q. Yan, A. Shinde, D. Guevarra, P. F. Newhouse, N. Becerra-Stasiewicz, S. M. Chatman, J. A. Haber, J. B. Neaton, J. M. Gregoire, *Adv. Energy Mater.* **2015**, *5*, 1500968; b) Q. Yan, J. Yu, S. K. Suram, L. Zhou, A. Shinde, P. F. Newhouse, W. Chen, G. Li, K. A. Persson, J. M. Gregoire, J. B. Neaton, *Proc. Natl. Acad. Sci. USA* **2017**, *114*, 3040.

[11] a) G. Cheon, K.-A. N. Duerloo, A. D. Sendek, C. Porter, Y. Chen, E. J. Reed, *Nano Lett.* **2017**, *17*, 1915; b) M. Ashton, J. Paul, S. B. Sinnott, R. G. Hennig, *Phys. Rev. Lett.* **2017**, *118*, 106101; c) A. K. Singh, K. Mathew, H. L. Zhuang, R. G. Hennig, *J. Phys. Chem. Lett.* **2015**, *6*, 1087; d) F. A. Rasmussen, K. S. Thygesen, *J. Phys. Chem. C* **2015**, *119*, 13169; e) N. Mounet, M. Gibertini, P. Schwaller, D. Campi, A. Merkys, A. Marrazzo, T. Sohier, I. E. Castelli, A. Cepellotti, G. Pizzi, N. Marzari, *Nat. Nanotechnol.* **2018**, *13*, 246.

[12] a) Y.-F. Zhang, J. Pan, H. Banjade, J. Yu, H. Lin, A. Bansil, S. Du, Q. Yan, *Nano Res.* **2021**, *14*, 584; b) Di Wang, F. Tang, J. Ji, W. Zhang, A. Vishwanath, H. C. Po, X. Wan, *Phys. Rev. B* **2019**, *100*, 195108; c) J. Pan, Y.-F. Zhang, J. Zhang, H. Banjade, J. Yu, L. Yu, S. Du, A. Ruzsinszky, Z. Hu, Q. Yan, *npj Comput. Mater.* **2020**, *6*, 154; d) A. Agarwal, S. Goverapet Srinivasan, B. Rai, *Front. Mater.* **2021**, *8*, 292.

[13] B. Bradlyn, L. Elcoro, J. Cano, M. G. Vergniory, Z. Wang, C. Felser, M. I. Aroyo, B. A Bernevig, *Nature* **2017**, *547*, 298.

[14] L. Elcoro, B. J. Wieder, Z. Song, Y. Xu, B. Bradlyn, B. A Bernevig, *Nat. Commun.* **2021**, *12*, 5965.

[15] J. Balachandran, L. Lin, J. S. Anchell, C. A. Bridges, P. Ganesh, *J. Phys. Chem. C* **2017**, *121*, 26637.

[16] S. P. Ong, W. D. Richards, A. Jain, G. Hautier, M. Kocher, S. Cholia, D. Gunter, V. L. Chevrier, K. A. Persson, G. Ceder, *Comput. Mater. Sci.* **2013**, *68*, 314.

[17] D. Broberg, B. Medasani, N. E. R. Zimmermann, G. Yu, A. Canning, M. Haranczyk, M. Asta, G. Hautier, *Comput. Phys. Commun.* **2018**, *226*, 165.

[18] a) J. S Park, S. Kim, Z. Xie, A. Walsh, *Nat. Rev. Mater.* **2018**, *3*, 194; b) H. J. Queisser, E. E. Haller, *Science* **1998**, *281*, 945.

[19] C. G. Van De Walle, J. Neugebauer, *J. Appl. Phys.* **2004**, *95*, 3851.

[20] A. Alkauskas, M. D. McCluskey, C. G. Van de Walle, *J. Appl. Phys.* **2016**, *119*, 181101.

[21] a) Q. Yan, A. Janotti, M. Scheffler, C. G. Van De Walle, *Appl. Phys. Lett.* **2012**, *100*, 142110; b) J. L. Lyons, A. Janotti, C. G. Van De Walle, *Phys. Rev. Lett.* **2012**, *108*, 156403.

[22] A. Alkauskas, M. D. McCluskey, C. G. Van De Walle, *J. Appl. Phys.* **2016**, *119*, 181101.

[23] D. B. Miracle, O. N. Senkov, *Acta Mater.* **2017**, *122*, 448.

[24] J. Chen, X. Zhang, C. Ying, He Ma, J. Li, F. Wang, H. Guo, *Ceram. Int.* **2020**, *46*, 10992.

[25] A. B. Mei, H. Kindlund, E. Broitman, L. Hultman, I. Petrov, J. E. Greene, D. G. Sangiovanni, *Acta Mater.* **2020**, *192*, 78.

[26] a) F. Liu, P. Ming, Ju Li, *Phys. Rev. B* **2007**, *76*, 064120; b) C. Lee, X. Wei, J. W. Kysar, J. Hone, *Science* **2008**, *321*, 385.

[27] a) D. Akinwande, C. J. Brennan, J. S Bunch, P. Egberts, J. R. Felts, H. Gao, R. Huang, J.-S. Kim, T. Li, Y. Li, K. M. Liechti, N. Lu, H. S. Park, E. J. Reed, P. Wang, B. I. Yakobson, T. Zhang, Y.-W. Zhang, Y. Zhou, Y. Zhu, *Extreme Mech. Lett.* **2017**, *13*, 42; b) Z. Xiong, L. Zhong, H. Wang, X. Li, *Materials* **2021**, *14*, 1192; c) L. Fan, J. Xu, Y. Hong, *RSC Adv.* **2022**, *12*, 6772.

[28] a) Li Song, L. Ci, H. Lu, P. B. Sorokin, C. Jin, J. Ni, A. G. Kvashnin, D. G. Kvashnin, J. Lou, B. I. Yakobson, P. M. Ajayan, *Nano Lett.* **2010**, *10*, 3209; b) S. Bertolazzi, J. Brivio, A. Kis, *ACS Nano* **2011**, *5*, 9703; c) A. Castellanos-Gomez, M. Poot, G. A. Steele, H. S. J. Van Der Zant, N. Agraït, G. Rubio-Bollinger, *Adv. Mater.* **2012**, *24*, 772; d) R. C. Cooper, C. Lee, C. A. Marianetti, X. Wei, J. Hone, J. W. Kysar, *Phys. Rev. B* **2013**, *87*, 035423; e) K. Liu, Q. Yan, M. Chen, W. Fan, Y. Sun, J. Suh, D. Fu, S. Lee, J. Zhou, S. Tongay, J. Ji, J. B. Neaton, J. Wu, *Nano Lett.* **2014**, *14*, 5097.

[29] Z. Liao, L. Medrano Sandonas, T. Zhang, M. Gall, A. Dianat, R. Gutierrez, U. Mühle, J. Gluch, R. Jordan, G. Cuniberti, *Sci. Rep.* **2017**, *7*, 211.

[30] Y. Yang, X. Li, M. Wen, E. Hacopian, W. Chen, Y. Gong, J. Zhang, Bo Li, Wu Zhou, P. M. Ajayan, Q. Chen, T. Zhu, J. Lou, *Adv. Mater.* **2017**, *29*, 1604201.

[31] a) T. Li, *Phys. Rev. B* **2012**, *85*, 235407; b) J. Wu, B. Wang, Y. Wei, R. Yang, M. Dresselhaus, *Mater. Res. Lett.* **2013**, *1*, 200; c) Q. Peng, X. Wen, S. De, *RSC Adv.* **2013**, *3*, 13772; d) B. Wang, J. Wu, X. Gu, H. Yin, Y. Wei, R. Yang, M. Dresselhaus, *Appl. Phys. Lett.* **2014**, *104*, 081902; e) Q. Wei, X. Peng, *Appl. Phys. Lett.* **2014**, *104*, 251915; f) E. Gao, B. Xie, Z. Xu, *J. Appl. Phys.* **2016**, *119*, 014301; g) H. Wang, Q. Li, Y. Gao, F. Miao, X.-F. Zhou, X. G. Wan, *New J. Phys.* **2016**, *18*, 073016; h) Z. Zhang, Y. Yang, E. S. Penev, B. I. Yakobson, *Adv. Funct. Mater.* **2017**, *27*, 1605059.

[32] Y. Hou, S. Zhang, Q. Li, L. Liu, X. Wu, Z. Zhang, *Opt. Lasers Eng.* **2022**, *152*, 106946.

[33] X. Li, H. Gao, *Natl. Sci. Rev.* **2015**, *2*, 133.

[34] K. E. Eshkalak, S. Sadeghzadeh, M. Jalaly, *Comput. Mater. Sci.* **2018**, *149*, 170.

[35] a) I. K. Petrushenko, K. B. Petrushenko, *Comput. Mater. Sci.* **2017**, *128*, 243; b) S. Thomas, M. Asle Zaeem, *Phys. Chem. Chem. Phys.* **2020**, *22*, 22066; c) H. Tang, D. Hu, Z. Cui, H. Ye, G. Zhang, *J. Phys. Chem. C* **2021**, *125*, 2680; d) A. Bagri, S.-P. Kim, R. S. Ruoff, V. B.

Shenoy, *Nano Lett.* **2011**, *11*, 3917; e) Y. Fu, T. Ragab, C. Basaran, *Comput. Mater. Sci.* **2016**, *124*, 142; f) S.-P. Wang, J.-G. Guo, Li-J Zhou, *Phys. E* **2013**, *48*, 29; g) X. Liu, G. Zhang, Y.-W. Zhang, *Nano Lett.* **2016**, *16*, 4954; h) Z. Song, V. I. Artyukhov, J. Wu, B. I. Yakobson, Z. Xu, *ACS Nano* **2015**, *9*, 401.

[36] P. Gao, C. T. Nelson, J. R. Jokisaari, S.-H. Baek, C. W. Bark, Yi Zhang, E. Wang, D. G. Schlom, C.-B. Eom, X. Pan, *Nat. Commun.* **2011**, *2*, 591.

[37] C. Wang, H. Dong, L. Jiang, W. Hu, *Chem. Soc. Rev.* **2018**, *47*, 422.

[38] G. Xiao, M. Z. Cieplak, A. Gavrin, F. H. Streitz, A. Bakhshai, C. L. Chien, *Phys. Rev. Lett.* **1988**, *60*, 1446.

[39] A.-M. Netsou, D. A. Muzychenko, H. Dausy, T. Chen, F. Song, K. Schouteden, M. J. Van Bael, C. Van Haesendonck, *ACS Nano* **2020**, *14*, 13172.

[40] K. Greben, S. Arora, M. G. Harats, K. I. Bolotin, *Nano Lett.* **2020**, *20*, 2544.

[41] C. Du, Q. Zhang, Z. Lin, Bo Yan, C. Xia, G. Yang, *Appl. Catal., B* **2019**, *248*, 193.

[42] K. S. Burch, D. Mandrus, Je-G Park, *Nature* **2018**, *563*, 47.

[43] S. J. U. White, T. Yang, N. Dontschuk, C. Li, Z.-Q. Xu, M. Kianinia, A. Stacey, M. Toth, I. Aharonovich, *Light: Sci. Appl.* **2022**, *11*, 186.

[44] C. Freysoldt, B. Grabowski, T. Hickel, J. Neugebauer, G. Kresse, A. Janotti, C. G. Van De Walle, *Rev. Mod. Phys.* **2014**, *86*, 253.

[45] C. E. Dreyer, A. Alkauskas, J. L. Lyons, A. Janotti, C. G. Van De Walle, *Annu. Rev. Mater. Res.* **2018**, *48*, 1.

[46] L. C. Bassett, A. Alkauskas, A. L. Exarhos, K.-M. C. Fu, *Nanophotonics* **2019**, *8*, 1867.

[47] Q. Liang, Q. Zhang, X. Zhao, M. Liu, A. T. S. Wee, *ACS Nano* **2021**, *15*, 2165.

[48] L. Zhong, R. C. Bruno, K. Ethan, L. Ruitao, R. Rahul, T. Humberto, A. P. Marcos, T. Mauricio, *2D Mater.* **2016**, *3*, 022002.

[49] D. Wong, J. Velasco, L. Ju, J. Lee, S. Kahn, H.-Z. Tsai, C. Germany, T. Taniguchi, K. Watanabe, A. Zettl, F. Wang, M. F. Crommie, *Nat. Nanotechnol.* **2015**, *10*, 949.

[50] N. Khossossi, D. Singh, A. Ainane, R. Ahuja, *Chem. Asian J.* **2020**, *15*, 3390.

[51] G. S. B. McKee, K. S. Vecchio, *J. Phys. Chem. B* **2006**, *110*, 1179.

[52] Y. R Lim, J. K. Han, S. K. Kim, Y. B. Lee, Y. Yoon, S. J. Kim, B. K. Min, Y. Kim, C. Jeon, S. Won, J.-H. Kim, W. Song, S. Myung, S. S. Lee, Ki-S An, J. Lim, *Adv. Mater.* **2018**, *30*, 1705270.

[53] M. Yi, Z. Shen, *J. Mater. Chem. A* **2015**, *3*, 11700.

[54] S. Caneva, R. S. Weatherup, B. C. Bayer, R. Blume, A. Cabrero-Vilatela, P. Braeuninger-Weimer, M.-B. Martin, R. Wang, C. Baehtz, R. Schloegl, J. C. Meyer, S. Hofmann, *Nano Lett.* **2016**, *16*, 1250.

[55] a) P. Chen, W. Xu, Y. Gao, J. H. Warner, M. R. Castell, *ACS Appl. Nano Mater.* **2018**, *1*, 6976; b) X. Zhang, Fu Zhang, Y. Wang, D. S. Schulman, T. Zhang, A. Bansal, N. Alem, S. Das, V. H. Crespi, M. Terrones, J. M. Redwing, *ACS Nano* **2019**, *13*, 3341; c) T. S. Millard, A. Genco, E. M. Alexeev, S. Randerson, S. Ahn, A. Jang, H. S. Shin, A. Tartakovskii, *npj 2D Mater. Appl.* **2020**, *4*, 1.

[56] W. Li, X. Qian, Ju Li, *Nat. Rev. Mater.* **2021**, *6*, 829.

[57] K. Momeni, Y. Ji, Y. Wang, S. Paul, S. Neshani, D. E. Yilmaz, Y. K. Shin, D. Zhang, J-Wu Jiang, H. S. Park, S. Sinnott, A. Van Duin, V. Crespi, L.-Q. Chen, *npj Comput. Mater.* **2020**, *6*, 22.

[58] F. H. L. Koppens, T. Mueller, Ph. Avouris, A. C. Ferrari, M. S. Vitiello, M. Polini, *Nat. Nanotechnol.* **2014**, *9*, 780.

[59] G. Moody, K. Tran, X. Lu, T. Autry, J. M. Fraser, R. P. Mirin, Li Yang, X. Li, K. L. Silverman, *Phys. Rev. Lett.* **2018**, *121*, 057403.

[60] a) Z. Lin, B. R. Carvalho, E. Kahn, R. Lv, R. Rao, H. Terrones, M. A. Pimenta, M. Terrones, *2D Mater.* **2016**, *3*, 022002; b) V. Carozo, Y. Wang, K. Fujisawa, B. R. Carvalho, A. McCreary, S. Feng, Z. Lin, C. Zhou, N. Perea-López, A. L. Elías, B. Kabius, V. H. Crespi, M. Terrones, *Sci. Adv.* **2017**, *3*, e1602813; c) C. Chakraborty, N. Vamivakas, D. Englund, *Nanophotonics* **2019**, *8*, 2017; d) E. Mitterreiter, B. Schuler, A. Micevic, D. Hernangómez-Pérez, K. Barthelmi, K. A. Cochrane, J. Kiemle, F. Sigger, J. Klein, E. Wong, E. S. Barnard, K. Watanabe, T. Taniguchi, M. Lorke, F. Jahnke, J. J. Finley, A. M. Schwartzberg, D. Y. Qiu, S. Refaely-Abramson, A. W. Holleitner, A. Weber-Bargioni, C. Kastl, *Nat. Commun.* **2021**, *12*, 3822; e) Y. J. Zheng, Y. Chen, Y. L. Huang, P. K. Gogoi, M.-Y. Li, L.-J. Li, P. E. Trevisanutto, Q. Wang, S. J. Pennycook, A. T. S. Wee, S. Y. Quek, *ACS Nano* **2019**, *13*, 6050; f) P. K. Gogoi, Z. Hu, Q. Wang, A. Carvalho, D. Schmidt, X. Yin, Y.-H. Chang, L.-J. Li, C. H. Sow, A.?H. C Neto, M. B. H. Breese, A. Rusydi, A. T. S. Wee, *Phys. Rev. Lett.* **2017**, *119*, 077402.

[61] Q. Liang, Q. Zhang, J. Gou, T. Song, Arramel, H. Chen, M. Yang, S. X. Lim, Q. Wang, R. Zhu, N. Yakovlev, S. C. Tan, W. Zhang, K. S. Novoselov, A. T. S. Wee, *ACS Nano* **2020**, *14*, 5668.

[62] M. S. Shawkat, J. Gil, S. S. Han, T.-J. Ko, M. Wang, D. Dev, J. Kwon, G.-H. Lee, K. H. Oh, H.-S. Chung, *ACS Appl. Mater. Interfaces* **2020**, *12*, 14341.

[63] a) H. Qiu, T. Xu, Z. Wang, W. Ren, H. Nan, Z. Ni, Q. Chen, S. Yuan, F. Miao, F. Song, G. Long, Y. Shi, L. Sun, J. Wang, X. Wang, *Nat. Commun.* **2013**, *4*, 2642; b) Z. Wu, Z. Luo, Y. Shen, W. Zhao, W. Wang, H. Nan, X. Guo, L. Sun, X. Wang, Y. You, Z. Ni, *Nano Res.* **2016**, *9*, 3622; c) B. Radisavljevic, A. Radenovic, J. Brivio, V. Giacometti, A. Kis, *Nat. Nanotechnol.* **2011**, *6*, 147; d) Xu Cui, G.-H. Lee, Y. D. Kim, G. Arefe, P. Y. Huang, C-Ho Lee, D. A. Chenet, X. Zhang, L. Wang, F. Ye, F. Pizzocchero, B. S. Jessen, K. Watanabe, T. Taniguchi, D. A. Muller, T. Low, P. Kim, J. Hone, *Nat. Nanotechnol.* **2015**, *10*, 534.

[64] a) F. Liu, L. You, K. L. Seyler, X. Li, P. Yu, J. Lin, X. Wang, J. Zhou, H. Wang, H. He, S. T. Pantelides, W. Zhou, P. Sharma, X. Xu, P. M. Ajayan, J. Wang, Z. Liu, *Nat. Commun.* **2016**, *7*, 12357; b) Yu Zhou, Di Wu, Y. Zhu, Y. Cho, Q. He, X. Yang, K. Herrera, Z. Chu, Yu Han, M. C. Downer, H. Peng, K. Lai, *Nano Lett.* **2017**, *17*, 5508; c) Z. Fei, W. Zhao, T. A. Palomaki, B. Sun, M. K. Miller, Z. Zhao, J. Yan, X. Xu, D. H. Cobden, *Nature* **2018**, *560*, 336; d) S. Yuan, X. Luo, H. L. Chan, C. Xiao, Y. Dai, M. Xie, J. Hao, *Nat. Commun.* **2019**, *10*, 1775; e) Q. Peng, D. Li, Pu Huang, Y. Ren, Z. Li, L. Pi, P. Chen, M. Wu, X. Zhang, X. Zhou, T. Zhai, *ACS Nano* **2021**, *15*, 16525.

[65] W. Xue, Q. Jiang, F. Wang, Ri He, R. Pang, H. Yang, P. Wang, R. Yang, Z. Zhong, T. Zhai, X. Xu, *Small* **2022**, *18*, 2105599.

[66] Y. Zhao, L. Lin, Q. Zhou, Y. Li, S. Yuan, Q. Chen, S. Dong, J. Wang, *Nano Lett.* **2018**, *18*, 2943.

[67] a) T. Xu, T. Shimada, M. Mori, G. Fujimoto, J. Wang, T. Kitamura, *Phys. Rev. Mater.* **2020**, *4*, 124405; b) R. He, J. L. Lin, Q. Liu, Z. Liao, L. Shui, Z. J. Wang, Z. Zhong, R.-W. Li, *ACS Appl. Mater. Interfaces* **2020**, *12*, 45602.

[68] L. Ran, J. Hou, S. Cao, Z. Li, Y. Zhang, Y. Wu, Bo Zhang, P. Zhai, L. Sun, *Sol. RRL* **2020**, *4*, 1900487.

[69] Z. Zafar, S. Yi, J. Li, C. Li, Y. Zhu, A. Zada, W. Yao, Z. Liu, X. Yue, *Energy Environ Mater.* **2022**, *5*, 68.

[70] L. Li, Z. Qin, L. Ries, S. Hong, T. Michel, J. Yang, C. Salameh, M. Bechelany, P. Miele, D. Kaplan, M. Chhowalla, D. Voiry, *ACS Nano* **2019**, *13*, 6824.

[71] T.-X. Huang, B. Dong, S. L. Filbrun, A. A. Okmi, X. Cheng, M. Yang, N. Mansour, S. Lei, N. Fang, *Sci. Adv.* **2021**, *7*, eabj4452.

[72] a) Z. Sun, H. Lv, Z. Zhuo, A. Jalil, W. Zhang, X. Wu, J. Yang, *J. Mater. Chem. C* **2018**, *6*, 1248; b) X. Lin, W. Yang, K. L. Wang, W. Zhao, *Nat. Electron.* **2019**, *2*, 274; c) R. Zhu, W. Zhang, W. Shen, P. K. J. Wong, Q. Wang, Q. Liang, Z. Tian, Ya Zhai, C.-W. Qiu, A. T. S. Wee, *Nano Lett.* **2020**, *20*, 5030; d) Y. Zhang, S. Talapatra, S. Kar, R. Vajtai, S. K. Nayak, P. M. Ajayan, *Phys. Rev. Lett.* **2007**, *99*, 107201.

[73] A. Avsar, A. Ciarrocchi, M. Pizzochero, D. Unuchek, O. V. Yazyev, A. Kis, *Nat. Nanotechnol.* **2019**, *14*, 674.

[74] S. Horzum, D. Çakir, J. Suh, S. Tongay, Y.-S. Huang, C.-H. Ho, J. Wu, H. Sahin, F. M. Peeters, *Phys. Rev. B* **2014**, *89*, 155433.

[75] R. Chua, J. Yang, X. He, X. Yu, W. Yu, F. Bussolotti, P. K. J. Wong, K. P. Loh, M. B. H. Breese, K. E. J. Goh, Y. L. Huang, A. T. S. Wee, *Adv. Mater.* **2020**, *32*, 2000693.
[76] S. Radhakrishnan, D. Das, L. Deng, P. M. Sudeep, G. Colas, C. A. Los Reyes, S. Yazdi, C. W. Chu, A. A. Martí, C. S. Tiwary, T. Filleter, A. K. Singh, P. M. Ajayan, *Adv. Mater.* **2018**, *30*, 1803366.
[77] R. Bharath, *Choice: Curr. Rev. Acad. Libr.* **2011**, *49*, 345.
[78] a) D. D. Awschalom, L. C. Bassett, A. S. Dzurak, E. L. Hu, J. R. Petta, *Science* **2013**, *339*, 1174; b) W. B. Gao, A. Imamoglu, H. Bernien, R. Hanson, *Nat. Photonics* **2015**, *9*, 363; c) I. Aharonovich, D. Englund, M. Toth, *Nat. Photonics* **2016**, *10*, 631; d) M. Atatüre, D. Englund, N. Vamivakas, S.-Y. Lee, J. Wrachtrup, *Nat. Rev. Mater.* **2018**, *3*, 38.
[79] M. W. Doherty, N. B. Manson, P. Delaney, F. Jelezko, J. Wrachtrup, L. C. L. Hollenberg, *Phys. Rep.* **2013**, *528*, 1.
[80] W. Pfaff, B. J. Hensen, H. Bernien, S. B. Van Dam, M. S. Blok, T. H. Taminiau, M. J. Tiggelman, R. N. Schouten, M. Markham, D. J. Twitchen, R. Hanson, *Science* **2014**, *345*, 532.
[81] G. Waldherr, Y. Wang, S. Zaiser, M. Jamali, T. Schulte-Herbrüggen, H. Abe, T. Ohshima, J. Isoya, J. F. Du, P. Neumann, J. Wrachtrup, *Nature* **2014**, *506*, 204.
[82] J. R. Weber, W. F. Koehl, J. B. Varley, A. Janotti, B. B. Buckley, C. G. Van De Walle, D. D. Awschalom, *Proc. Natl. Acad. Sci. USA* **2010**, *107*, 8513.
[83] a) C. Hepp, T. Müller, V. Waselowski, J. N. Becker, B. Pingault, H. Sternschulte, D. Steinmüller-Nethl, A. Gali, J. R. Maze, M. Atatüre, C. Becher, *Phys. Rev. Lett.* **2014**, *112*, 036405; b) B. C. Rose, D. Huang, Zi-H Zhang, P. Stevenson, A. M. Tyryshkin, S. Sangtawesin, S. Srinivasan, L. Loudin, M. L. Markham, A. M. Edmonds, D. J. Twitchen, S. A. Lyon, N. P. De Leon, *Science* **2018**, *361*, 60; c) L. J. Rogers, K. D. Jahnke, M. H. Metsch, A. Sipahigil, J. M. Binder, T. Teraji, H. Sumiya, J. Isoya, M. D. Lukin, P. Hemmer, F. Jelezko, *Phys. Rev. Lett.* **2014**, *113*, 263602.
[84] a) W. F. Koehl, B. B. Buckley, F. J Heremans, G. Calusine, D. D. Awschalom, *Nature* **2011**, *479*, 84; b) M. Widmann, S.-Y. Lee, T. Rendler, N. T. Son, H. Fedder, S. Paik, Li-P Yang, N. Zhao, S. Yang, I. Booker, A. Denisenko, M. Jamali, S. A Momenzadeh, I. Gerhardt, T. Ohshima, A. Gali, E. Janzén, J. Wrachtrup, *Nat. Mater.* **2015**, *14*, 164; c) D. J. Christle, A. L. Falk, P. Andrich, P. V. Klimov, J. U. Hassan, N. T. Son, E. Janzén, T. Ohshima, D. D. Awschalom, *Nat. Mater.* **2015**, *14*, 160.
[85] P. Siyushev, K. Xia, R. Reuter, M. Jamali, N. Zhao, N. Yang, C. Duan, N. Kukharchyk, A. D. Wieck, R. Kolesov, J. Wrachtrup, *Nat. Commun.* **2014**, *5*, 3895.
[86] a) J. R. Weber, W. F. Koehl, J. B. Varley, A. Janotti, B. B. Buckley, C. G. Van de Walle, D. D. Awschalom, *J. Appl. Phys.* **2011**, *109*, 102417; b) A. Gali, *Phys. Status Solidi B* **2011**, *248*, 1337.
[87] H. Seo, M. Govoni, G. Galli, *Sci. Rep.* **2016**, *6*, 20803.
[88] L. J. Rogers, K. D. Jahnke, T. Teraji, L. Marseglia, C. Müller, B. Naydenov, H. Schauffert, C. Kranz, J. Isoya, L. P. McGuinness, F. Jelezko, *Nat. Commun.* **2014**, *5*, 4739.
[89] T. T. Tran, K. Bray, M. J. Ford, M. Toth, I. Aharonovich, *Nat. Nanotechnol.* **2016**, *11*, 37.
[90] J.-Y. Tsai, J. Pan, H. Lin, A. Bansil, Q. Yan, *Nat. Commun.* **2022**, *13*, 492.
[91] I. Aharonovich, M. Toth, *Science* **2017**, *358*, 170.
[92] a) S. A. Tawfik, S. Ali, M. Fronzi, M. Kianinia, T. T. Tran, C. Stampfl, I. Aharonovich, M. Toth, M. J. Ford, *Nanoscale* **2017**, *9*, 13575; b) F. Wu, A. Galatas, R. Sundararaman, D. Rocca, Y. Ping, *Phys. Rev. Mater.* **2017**, *1*, 071001.
[93] T. Q. P. Vuong, G. Cassabois, P. Valvin, A. Ouerghi, Y. Chassagneux, C. Voisin, B. Gil, *Phys. Rev. Lett.* **2016**, *117*, 097402.
[94] a) A. J. Morfa, B. C. Gibson, M. Karg, T. J. Karle, A. D. Greentree, P. Mulvaney, S. Tomljenovic-Hanic, *Nano Lett.* **2012**, *12*, 949; b) N. R. Jungwirth, H.-S. Chang, M. Jiang, G. D. Fuchs, *ACS Nano* **2016**, *10*, 1210.
[95] a) Yu Zhou, Z. Wang, A. Rasmita, S. Kim, A. Berhane, Z. Bodrog, G. Adamo, A. Gali, I. Aharonovich, W.-B. Gao, *Sci. Adv.* **2018**, *4*, eaar3580; b) A. M. Berhane, K.-Y. Jeong, Z. Bodrog, S. Fiedler, T. Schröder, N. V. Triviño, T. Palacios, A. Gali, M. Toth, D. Englund, I. Aharonovich, *Adv. Mater.* **2017**, *29*, 1605092.
[96] a) M. Koperski, K. Nogajewski, A. Arora, V. Cherkez, P. Mallet, J.-Y. Veuillen, J. Marcus, P. Kossacki, M. Potemski, *Nat. Nanotechnol.* **2015**, *10*, 503; b) Yu-M He, G. Clark, J. R. Schaibley, Yu He, M.-C. Chen, Yu-J Wei, X. Ding, Q. Zhang, W. Yao, X. Xu, C.-Y. Lu, J.-W. Pan, *Nat. Nanotechnol.* **2015**, *10*, 497.
[97] A. L. Exarhos, D. A. Hopper, R. R. Grote, A. Alkauskas, L. C. Bassett, *ACS Nano* **2017**, *11*, 3328.
[98] N. Mendelson, D. Chugh, J. R. Reimers, T. S. Cheng, A. Gottscholl, Hu Long, C. J. Mellor, A. Zettl, V. Dyakonov, P. H. Beton, S. V. Novikov, C. Jagadish, H. H. Tan, M. J. Ford, M. Toth, C. Bradac, I. Aharonovich, *Nat. Mater.* **2021**, *20*, 321.
[99] a) H. Liu, Y. Liu, D. Zhu, *J. Mater. Chem.* **2011**, *21*, 3335; b) D. B. Farmer, R. Golizadeh-Mojarad, V. Perebeinos, Yu-M Lin, G. S. Tulevski, J. C. Tsang, P. Avouris, *Nano Lett.* **2009**, *9*, 388; c) H. Yoo, K. Heo, Md. H. R. Ansari, S. Cho, *Nanomaterials* **2021**, *11*, 832.
[100] a) H.-C. Wang, S. Botti, M. A. Marques, *npj Comput. Mater.* **2021**, *7*, 12; b) G. Hautier, C. Fischer, V. Ehrlacher, A. Jain, G. Ceder, *Inorg. Chem.* **2011**, *50*, 656.
[101] H. Li, H. Liu, L. Zhou, X. Wu, Y. Pan, W. Ji, B. Zheng, Q. Zhang, X. Zhuang, X. Zhu, X. Wang, X. Duan, A. Pan, *ACS Nano* **2018**, *12*, 4853.
[102] a) H. Lee, K. Paeng, I. S. Kim, *Synth. Met.* **2018**, *244*, 36; b) X. Wang, G. Sun, P. Routh, D.-H. Kim, W. Huang, P. Chen, *Chem. Soc. Rev.* **2014**, *43*, 7067.
[103] F. Banhart, J. Kotakoski, A. V. Krasheninnikov, *ACS Nano* **2011**, *5*, 26.
[104] a) S. K. Pandey, H. Alsalman, J. G. Azadani, N. Izquierdo, T. Low, S. A. Campbell, *Nanoscale* **2018**, *10*, 21374; b) K. Zhang, B. M. Bersch, J. Joshi, R. Addou, C. R. Cormier, C. Zhang, Ke Xu, N. C. Briggs, Ke Wang, S. Subramanian, K. Cho, S. Fullerton-Shirey, R. M. Wallace, P. M. Vora, J. A. Robinson, *Adv. Funct. Mater.* **2018**, *28*, 1706950; c) Y. Kim, H. Bark, B. Kang, C. Lee, *ACS Appl. Mater. Interfaces* **2019**, *11*, 12613; d) Y. Yue, C. Jiang, Y. Han, M. Wang, J. Ren, Y. Wu, *J. Magn. Magn. Mater.* **2020**, *496*, 165929; e) P. Zhang, N. Cheng, M. Li, B. Zhou, C. Bian, Y. Wei, X. Wang, H. Jiang, L. Bao, Y. Lin, *ACS Appl. Mater. Interfaces* **2020**, *12*, 18650; f) C. Kanade, S. Arbuj, K. Kanade, K. S. Kim, G. Y. Yeom, T. Kim, B. Kale, *RSC Adv.* **2018**, *8*, 39749; g) L. Yang, K. Majumdar, H. Liu, Y. Du, H. Wu, M. Hatzistergos, P. Y. Hung, R. Tieckelmann, W. Tsai, C. Hobbs, P. D. Ye, *Nano Lett.* **2014**, *14*, 6275; h) B. Tang, Z. G. Yu, Li Huang, J. Chai, S. L. Wong, J. Deng, W. Yang, H. Gong, S. Wang, K.-W. Ang, Y.-W. Zhang, D. Chi, *ACS Nano* **2018**, *12*, 2506; i) K. H Kim, K. S Kim, Y. J. Ji, I. Moon, K. Heo, D-Ho Kang, K. N. Kim, W. J. Yoo, J.-H. Park, G. Y. Yeom, *J. Mater. Chem. C* **2020**, *8*, 1846; j) H.-P. Komsa, J. Kotakoski, S. Kurasch, O. Lehtinen, U. Kaiser, A. V. Krasheninnikov, *Phys. Rev. Lett.* **2012**, *109*, 035503.
[105] a) S. X. M. Riberolles, Q. Zhang, E. Gordon, N. P. Butch, L. Ke, J.-Q. Yan, R. J. McQueeney, *Phys. Rev. B* **2021**, *104*, 064401; b) B. Chen, F. Fei, D. Zhang, B. Zhang, W. Liu, S. Zhang, P. Wang, B. Wei, Y. Zhang, Z. Zuo, J. Guo, Q. Liu, Z. Wang, X. Wu, J. Zong, X. Xie, W. Chen, Z. Sun, S. Wang, Y. Zhang, M. Zhang, X. Wang, F. Song, H. Zhang, D. Shen, B. Wang, *Nat. Commun.* **2019**, *10*, 4469; c) X.-M. Ma, Y. Zhao, Ke Zhang, S. Kumar, R. Lu, J. Li, Q. Yao, J. Shao, F. Hou, X. Wu, M. Zeng, Yu-J Hao, Z. Hao, Y. Wang, X.-R. Liu, H. Shen, H. Sun, J. Mei, K. Miyamoto, T. Okuda, M. Arita, E. F. Schwier, K. Shimada, Ke Deng, C. Liu, J. Lin, Y. Zhao, C. Chen, Q. Liu, C. Liu, *Phys. Rev. B* **2021**, *103*, L121112; d) J.-Q. Yan, S. Okamoto, M. A. McGuire, A. F. May, R. J. McQueeney, B. C. Sales, *Phys. Rev. B* **2019**, *100*, 104409; e) W.

Ko, M. Kolmer, J. Yan, A. D. Pham, M. Fu, F. Lüpke, S. Okamoto, Z. Gai, P. Ganesh, A.-P. Li, *Phys. Rev. B* **2020**, *102*, 115402; f) R. Watanabe, R. Yoshimi, M. Kawamura, Y. Kaneko, K. S. Takahashi, A. Tsukazaki, M. Kawasaki, Y. Tokura, *Appl. Phys. Lett.* **2022**, *120*, 031901.

[106] D. Nevola, K. F. Garrity, N. Zaki, J. Yan, H. Miao, S. Chowdhury, P. D. Johnson, *ArXiv: 2206.14325*, **2022**.

[107] S. Chowdhury, K. F. Garrity, F. Tavazza, *ArXiv: 2202.07061*, **2022**.

[108] a) Wu Zhou, X. Zou, S. Najmaei, Z. Liu, Y. Shi, J. Kong, J. Lou, P. M. Ajayan, B. I. Yakobson, J.-C. Idrobo, *Nano Lett.* **2013**, *13*, 2615; b) S. Ippolito, P. Samorì, *Small Sci.* **2022**, *2*, 2100122; c) H.-P. Komsa, A. V. Krasheninnikov, *Phys. Rev. B* **2015**, *91*, 125304.

[109] X. Wang, Y. Zhang, H. Si, Q. Zhang, J. Wu, Li Gao, X. Wei, Yu Sun, Q. Liao, Z. Zhang, K. Ammarah, L. Gu, Z. Kang, Y. Zhang, *J. Am. Chem. Soc.* **2020**, *142*, 4298.

[110] M. Kusaba, C. Liu, R. Yoshida, *Comput. Mater. Sci.* **2022**, *211*, 111496.

[111] J. R. Weber, W. F. Koehl, J. B. Varley, A. Janotti, B. B. Buckley, C. G. Van De Walle, D. D. Awschalom, *Proc. Natl. Acad. Sci. USA* **2010**, *107*, 8513.

[112] a) N. C. Frey, D. Akinwande, D. Jariwala, V. B. Shenoy, *ACS Nano* **2020**, *14*, 13406; b) V. Popescu, A. Zunger, *Phys. Rev. B* **2012**, *85*, 085201.

[113] a) R. Michalsky, V. Botu, C. M. Hargus, A. A. Peterson, A. Steinfeld, *Adv. Energy Mater.* **2015**, *5*, 1401082; b) M. Ezbiri, K. M. Allen, M. E. Gàlvez, R. Michalsky, A. Steinfeld, *ChemSusChem* **2015**, *8*, 1966.

[114] a) A. M. Deml, A. M. Holder, R. P. O'hayre, C. B. Musgrave, V. Stevanovic, *J. Phys. Chem. Lett.* **2015**, *6*, 1948; b) Y.-L. Lee, J. Kleis, J. Rossmeisl, Y. Shao-Horn, D. Morgan, *Energy Environ. Sci.* **2011**, *4*, 3966.

[115] A. M. Deml, V. Stevanović, C. L. Muhich, C. B. Musgrave, R. O'Hayre, *Energy Environ. Sci.* **2014**, *7*, 1996.

[116] S. Anand, K. Xia, V. I. Hegde, U. Aydemir, V. Kocevski, T. Zhu, C. Wolverton, G. J. Snyder, *Energy Environ. Sci.* **2018**, *11*, 1480.

[117] C. Freysoldt, B. Grabowski, T. Hickel, J. Neugebauer, G. Kresse, A. Janotti, C. G. Van de Walle, *Rev. Mod. Phys.* **2014**, *86*, 253.

[118] K. Mathew, J. H. Montoya, A. Faghaninia, S. Dwarakanath, M. Aykol, H. Tang, I.-H. Chu, T. Smidt, B. Bocklund, M. Horton, J. Dagdelen, B. Wood, Zi-K Liu, J. Neaton, S. P. Ong, K. Persson, A. Jain, *Comput. Mater. Sci.* **2017**, *139*, 140.

[119] a) C. E. Dreyer, A. Alkauskas, J. L. Lyons, A. Janotti, C. G. Van de Walle, *Annu. Rev. Mater. Res.* **2018**, *48*, 1; b) G. Zhang, Y. Cheng, J.-P. Chou, A. Gali, *Appl. Phys. Rev.* **2020**, *7*, 031308.

[120] J. L. Lyons, C. G. Van De Walle, *npj Comput. Mater.* **2017**, *3*, 12.

[121] D. Y. Qiu, F. H. Da Jornada, S. G. Louie, *Phys. Rev. B* **2016**, *93*, 235435.

[122] W. D. Parker, J. W. Wilkins, R. G. Hennig, *Phys. Status Solidi* **2011**, *248*, 267.

[123] a) P. Delaney, J. C. Greer, J. A Larsson, *Nano Lett.* **2010**, *10*, 610; b) A. Gali, E. Janzen, P. Deak, G. Kresse, E. Kaxiras, *Phys. Rev. Lett.* **2009**, *103*, 186404; c) Y. C. Ma, M. Rohlfing, A. Gali, *Phys. Rev. B* **2010**,*81*, 041204.

[124] a) J. Davidsson, V. Ivady, R. Armiento, N. T. Son, A. Gali, I. A. Abrikosov, *New J. Phys.* **2018**, *20*, 023035; b) L. Gordon, A. Janotti, C. G. Van de Walle, *Phys. Rev. B* **2015**, *92*, 045208; c) M. Bockstedte, A. Marini, O. Pankratov, A. Rubio, *Phys. Rev. Lett.* **2010**, *105*, 026401; d) K. Szasz, V. Ivady, I. A. Abrikosov, E. Janzen, M. Bockstedte, A. Gali, *Phys. Rev. B* **2015**, *91*, 121201.

[125] C. Attaccalite, M. Bockstedte, A. Marini, A. Rubio, L. Wirtz, *Phys. Rev. B* **2011**, *83*, 144115.

[126] M. L. Goldman, M. W. Doherty, A. Sipahigil, N. Y. Yao, S. D. Bennett, N. B. Manson, A. Kubanek, M. D. Lukin, *Phys. Rev. B* **2015**, *91*, 165201.

[127] a) R. De Sousa, S. Das Sarma, *Phys. Rev. B* **2003**, *67*, 033301; b) Pu Huang, Xi Kong, N. Zhao, F. Shi, P. Wang, X. Rong, R.-B. Liu, J. Du, *Nat. Commun.* **2011**, *2*, 570; c) H. Seo, A. L. Falk, P. V. Klimov, K. C. Miao, G. Galli, D. D. Awschalom, *Nat. Commun.* **2016**, *7*, 12935.

[128] M. Ye, H. Seo, G. Galli, *npj Comput. Mater.* **2019**, *5*, 44.

[129] C. Freysoldt, J. Neugebauer, C. G. Van De Walle, *Phys. Rev. Lett.* **2009**, *102*, 016402.

[130] a) H.-P. Komsa, N. Berseneva, A. V. Krasheninnikov, R. M. Nieminen, *Phys. Rev. X* **2014**, *4*, 031044; b) D. Wang, D. Han, X.-B. Li, S-Yi Xie, N-Ke Chen, W. Q. Tian, D. West, H-Bo Sun, S. ? B. Zhang, *Phys. Rev. Lett.* **2015**, *114*, 196801; c) H.-P. Komsa, A. Pasquarello, *Phys. Rev. Lett.* **2013**, *110*, 095505.

[131] F. Bertoldo, S. Ali, S. Manti, K. S. Thygesen, *npj Comput. Mater.* **2022**, *8*, 56.

[132] M. N. Gjerding, A. Taghizadeh, A. Rasmussen, S. Ali, F. Bertoldo, T. Deilmann, N. R. Knøsgaard, M. Kruse, A. H. Larsen, S. Manti, T. G. Pedersen, U. Petralanda, T. Skovhus, M. K. Svendsen, J. J. Mortensen, T. Olsen, K. S. Thygesen, *2D Mater.* **2021**, *8*, 044002.

[133] K. Choudhary, A. J. Biacchi, S. Ghosh, L. Hale, A. R. H. Walker, F. Tavazza, *J. Phys.: Condens. Matter* **2018**, *30*, 395901.

[134] J. P. Perdew, K. Schmidt, *AIP Conf. Proc.* **2001**, *577*, 1.

[135] a) J. W. Furness, A. D. Kaplan, J. Ning, J. P. Perdew, J. Sun, *J. Phys. Chem. Lett.* **2020**, *11*, 8208; b) J. Sun, R. C. Remsing, Y. Zhang, Z. Sun, A. Ruzsinszky, H. Peng, Z. Yang, A. Paul, U. Waghmare, X. Wu, M. L. Klein, J. P. Perdew, *Nat. Chem.* **2016**, *8*, 831.

[136] He Ma, N. Sheng, M. Govoni, G. Galli, *J. Chem. Theory Comput.* **2021**, *17*, 2116.

[137] A. Mannodi-Kanakkithodi, X. Xiang, L. Jacoby, R. Biegaj, S. T. Dunham, D. R. Gamelin, M. K. Y. Chan, *Patterns* **2022**, *3*, 100450.

[138] M. I. Jordan, T. M. Mitchell, *Science* **2015**, *349*, 255.

[139] a) R. Ramakrishnan, P. O. Dral, M. Rupp, O. A Von Lilienfeld, *J. Chem. Theory Comput.* **2015**, *11*, 2087; b) A. P. Bartók, M. C. Payne, R. Kondor, G. Csányi, *Phys. Rev. Lett.* **2010**, *104*, 136403.

[140] a) R. Lesar, *Stat. Anal. Data Mining* **2009**, *1*, 372; b) K. Rajan, *Mater. Today* **2005**, *8*, 38; c) S. Curtarolo, D. Morgan, K. Persson, J. Rodgers, G. Ceder, *Phys. Rev. Lett.* **2003**, *91*, 135503; d) P. Raccuglia, K. C. Elbert, P. D. F. Adler, C. Falk, M. B. Wenny, A. Mollo, M. Zeller, S. A. Friedler, J. Schrier, A. J. Norquist, *Nature* **2016**, *533*, 73; e) G. Pilania, C. Wang, X. Jiang, S. Rajasekaran, R. Ramprasad, *Sci. Rep.* **2013**, *3*, 2810; f) K. Hansen, G. Montavon, F. Biegler, S. Fazli, M. Rupp, M. Scheffler, O. A Von Lilienfeld, A. Tkatchenko, K.-R. Müller, *J. Chem. Theory Comput.* **2013**, *9*, 3404; g) G. Montavon, M. Rupp, V. Gobre, A. Vazquez-Mayagoitia, K. Hansen, A. Tkatchenko, K.-R. Müller, O. Anatole Von Lilienfeld, *New J. Phys.* **2013**, *15*, 095003.

[141] a) C. J. Long, J. Hattrick-Simpers, M. Murakami, R. C. Srivastava, I. Takeuchi, V. L. Karen, X. Li, *Rev. Sci. Instrum.* **2007**, *78*, 072217; b) R. Ramakrishnan, P. O. Dral, M. Rupp, O. A Von Lilienfeld, *Sci. Data* **2014**, *1*, 140022.

[142] G. Hautier, C. C. Fischer, A. Jain, T. Mueller, G. Ceder, *Chem. Mater.* **2010**, *22*, 3762.

[143] a) G. Pilania, A. Mannodi-Kanakkithodi, B. P. Uberuaga, R. Ramprasad, J. E. Gubernatis, T. Lookman, *Sci. Rep.* **2016**, *6*, 19375; b) J. Lee, A. Seko, K. Shitara, K. Nakayama, I. Tanaka, *Phys. Rev. B* **2016**, *93*, 115104; c) F. Pereira, K. Xiao, D. A. R. S. Latino, C. Wu, Q. Zhang, J. Aires-De-Sousa, J. Aires-de-Sousa, *J. Chem. Inf. Model.* **2017**, *57*, 11.

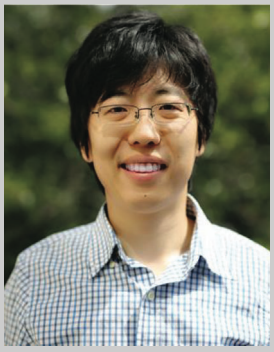
[144] S. Kong, F. Ricci, D. Guevarra, J. B. Neaton, C. P. Gomes, J. M. Gregoire, *Nat. Commun.* **2022**, *13*, 949.

[145] a) He Li, Z. Wang, N. Zou, M. Ye, R. Xu, X. Gong, W. Duan, Y. Xu, *Nat. Comput. Sci.* **2022**, *2*, 367; b) H. Bai, P. Chu, J.-Y. Tsai, N. Wilson, X. Qian, Q. Yan, H. Ling, *Neural Comput. Appl.* **2022**, *34*, 4625.

[146] J. C. Snyder, M. Rupp, K. Hansen, K.-R. Müller, K. Burke, *Phys. Rev. Lett.* **2012**, *108*, 253002.

[147] Y. Lecun, Y. Bengio, G. Hinton, *Nature* **2015**, *521*, 436.

[148] a) Y. Lecun, L. Bottou, Y. Bengio, P. Haffner, *Proc. IEEE Inst. Electr. Electron. Eng.* **1998**, *86*, 2278; b) K. He, X. Zhang, S. Ren, J. Sun, in *2016 IEEE Conf. on Computer Vision and Pattern Recognition (CVPR)*, IEEE, Piscataway, NJ, USA, **2016**, pp. 770–778.
[149] Z. Zhang, P. Cui, W. Zhu, *ArXiv: 1812.04202*, **2018**.
[150] a) M. M. Bronstein, J. Bruna, Y. Lecun, A. Szlam, P. Vandergheynst, *IEEE Signal Process. Mag.* **2017**, *34*, 18; b) T. N. Kipf, M. Wellin, presented at 5th Int. Conf. Learning Representations, Toulon, France, April **2017**; c) M. Henaff, J. Bruna, Y. LeCun, *ArXiv: 1506.05163*, **2015**.
[151] a) C. Chen, W. Ye, Y. Zuo, C. Zheng, S. P. Ong, *Chem. Mater.* **2019**, *31*, 3564; b) T. Xie, J. C. Grossman, *Phys. Rev. Lett.* **2018**, *120*, 145301; c) C. W. Park, C. Wolverton, *Phys. Rev. Mater.* **2020**, *4*, 063801.
[152] H. R. Banjade, S. Hauri, S. Zhang, F. Ricci, W. Gong, G. Hautier, S. Vucetic, Q. Yan, *Sci. Adv.* **2021**, *7*, eabf1754.
[153] K. Choudhary, B. Decost, *npj Comput. Mater.* **2021**, *7*, 185.
[154] C. Chen, S. P. Ong, *Nat. Comput. Sci.* **2022**, *2*, 718.
[155] Y.-L. Liao, T. Smidt, *ArXiv:2206.11990*, **2022**.
[156] A. Mannodi-Kanakkithodi, M. Y. Toriyama, F. G. Sen, M. J. Davis, R. F. Klie, M. K. Y. Chan, *npj Comput. Mater.* **2020**, *6*, 39.
[157] P. Huang, R. Lukin, M. Faleev, N. Kazeev, A. R. Al-Maeeni, D. V. Andreeva, A. Ustyuzhanin, A. Tormasov, A. H. Castro Neto, K. S. Novoselov, *npj 2D Mater. Appl.* **2023**, *7*, 6.
[158] A. Kolesnikov, X. Zhai, L. Beyer, *ArXiv: 1901.09005*, **2019**.
[159] J. Devlin, M.-W. Chang, K. Lee, K. Toutanova, *ArXiv: 1810.04805*, **2018**.
[160] a) S. Chithrananda, G. Grand, B. Ramsundar, *ArXiv: 2010.09885*, **2020**; b) Y. Rong, Y. Bian, T. Xu, W. Xie, Y. Wei, W. Huang, J. Huang, *ArXiv: 2007.02835*, **2020**.
[161] P. Veličković, G. Cucurull, A. Casanova, A. Romero, P. Liò, Y. Bengio, *ArXiv: 1710.10903*, **2017**.
[162] V. P. Dwivedi, X. Bresson, *ArXiv: 2012.09699*, **2020**.
[163] N. R. Glavin, P. M. Ajayan, S. Kar, *Adv. Mater.* **2022**, *n/a*, 2109892.
[164] S. W. Hla, *Rep. Prog. Phys.* **2014**, *77*, 056502.
[165] M. Tripathi, A. Mittelberger, N. A. Pike, C. Mangler, J. C. Meyer, M. J. Verstraete, J. Kotakoski, T. Susi, *Nano Lett.* **2018**, *18*, 5319.
[166] O. Dyck, M. Ziatdinov, D. B. Lingerfelt, R. R. Unocic, B. M. Hudak, A. R. Lupini, S. Jesse, S. V. Kalinin, *Nat. Rev. Mater.* **2019**, *4*, 497.
[167] R. Achal, M. Rashidi, J. Croshaw, D. Churchill, M. Taucer, T. Huff, M. Cloutier, J. Pitters, R. A. Wolkow, *Nat. Commun.* **2018**, *9*, 2778.
[168] M. F. Crommie, C. P. Lutz, D. M. Eigler, *Science* **1993**, *262*, 218.
[169] J. Yang, D. Sordes, M. Kolmer, D. Martrou, C. Joachim, *Eur. Phys. J.: Appl. Phys.* **2016**, *73*, 10702.
[170] S.-W. Hla, *J. Vac. Sci. Technol., B: Microelectron. Nanometer Struct.–Process., Meas., Phenom.* **2005**, *23*, 1351.
[171] G. D. Nguyen, L. Liang, Q. Zou, M. Fu, A. D. Oyedele, B. G. Sumpter, Z. Liu, Z. Gai, K. Xiao, A.-P. Li, *Phys. Rev. Lett.* **2018**, *121*, 086101.
[172] a) A. Bachmatiuk, J. Zhao, S. M. Gorantla, I. G. G. Martinez, J. Wiedermann, C. Lee, J. Eckert, M. H. Rummeli, *Small* **2015**, *11*, 515; b) S. V. Kalinin, A. Borisevich, S. Jesse, *Nature* **2016**, *539*, 485.
[173] Yu Lei, S. Pakhira, K. Fujisawa, He Liu, C. Guerrero-Bermea, T. Zhang, A. Dasgupta, L. M. Martinez, S. Rao Singamaneni, Ke Wang, J. Shallenberger, A. L. Elías, R. Cruz-Silva, M. Endo, J. L. Mendoza-Cortes, M. Terrones, *Mater. Today* **2021**, *51*, 108.
[174] J. B. Ballard, J. H. G. Owen, W. Owen, J. R. Alexander, E. Fuchs, J. N. Randall, J. R. Von Ehr, S. McDonnell, D. D. Dick, R. M. Wallace, Y. J. Chabal, M. R. Bischof, D. L. Jaeger, R. F. Reidy, J. Fu, P. Namboodiri, K. Li, R. M. Silver, *J. Vac. Sci. Technol., B* **2014**, *32*, 041804.
[175] P. Ercius, O. Alaidi, M. J. Rames, G. Ren, *Adv. Mater.* **2015**, *27*, 5638.
[176] X. Tian, D. S. Kim, S. Yang, C. J. Ciccarino, Y. Gong, Y. Yang, Y. Yang, B. Duschatko, Y. Yuan, P. M. Ajayan, J. C. Idrobo, P. Narang, J. Miao, *Nat. Mater.* **2020**, *19*, 867.
[177] B. R. Carvalho, Y. Wang, K. Fujisawa, T. Zhang, E. Kahn, I. Bilgin, P. M. Ajayan, A. M. De Paula, M. A. Pimenta, S. Kar, V. H. Crespi, M. Terrones, L. M. Malard, *Nano Lett.* **2020**, *20*, 284.
[178] a) H. Hafiz, K. Suzuki, B. Barbiellini, N. Tsuji, N. Yabuuchi, K. Yamamoto, Y. Orikasa, Y. Uchimoto, Y. Sakurai, H. Sakurai, A. Bansil, V. Viswanathan, *Nature* **2021**, *594*, 213; b) H. Hafiz, K. Suzuki, B. Barbiellini, Y. Orikasa, V. Callewaert, S. Kaprzyk, M. Itou, K. Yamamoto, R. Yamada, Y. Uchimoto, Y. Sakurai, H. Sakurai, A. Bansil, *Sci. Adv.* **2017**, *3*, e1700971; c) B. Barbiellini, K. Suzuki, Y. Orikasa, S. Kaprzyk, M. Itou, K. Yamamoto, Y. J. Wang, H. Hafiz, R. Yamada, Y. Uchimoto, A. Bansil, Y. Sakurai, H. Sakurai, *Appl. Phys. Lett.* **2016**, *109*, 073102; d) K. Suzuki, B. Barbiellini, Y. Orikasa, N. Go, H. Sakurai, S. Kaprzyk, M. Itou, K. Yamamoto, Y. Uchimoto, Y. J. Wang, H. Hafiz, A. Bansil, Y. Sakurai, *Phys. Rev. Lett.* **2015**, *114*, 087401; e) Y. Sakurai, M. Itou, B. Barbiellini, P. E. Mijnarends, R. S. Markiewicz, S. Kaprzyk, J.-M. Gillet, S. Wakimoto, M. Fujita, S. Basak, Y. J. Wang, W. Al-Sawai, H. Lin, A. Bansil, K. Yamada, *Science* **2011**, *332*, 698.



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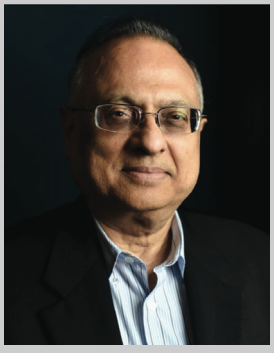

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