Computational Materials Discovery

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I. INTRODUCTION

Tremendous advances in first-principles program packages, spectacular speed-ups in computer hardware coupled with significant algorithmic developments in crystal structure prediction, high-throughput screenings, data-mining, machine learning, and artificial intelligence, have made the dream of computational materials discovery a reality[1]. Recent success stories of materials that were first predicted and later verified experimentally include high-temperature superconductors[2–4], high-entropy high-hardness materials[5], Heusler intermetallics for thermoelectrics and spintronics[6], organic light-emitting diodes[7], Li battery cathode materials[8], porous materials for gas storage[9], and carbon-boron clathrates[10].

The broad range of materials-by-design success stories with applications including energy, quantum, structural, 2D materials, molecular crystals, molecular organic frameworks and more, have inspired us to devote this issue of JCP towards recent advances in this field. In addition to describing specific predictions, the articles in this issue also describe novel methods for computational and data-driven materials discovery. In what follows we summarize the manuscripts submitted to this special issue, organizing them around key topical areas. We first describe manuscripts that focus on the development of new methods and program packages, followed by application-centered manuscripts.

In addition to spanning a wide range of materials for diverse applications, the contributions to this special issue highlight the broad spectrum of computational methodologies that can be used for materials prediction**, including first-principles methods (often based upon density functional theory, DFT), those employing atomistic potentials and molecular dynamics (MD), and mesoscale modeling.** Moreover, they illustrate the wide range of available tools including crystal structure prediction (CSP) methods[11,12], high-throughput calculations[13,14], machine learning (ML)[15–18], and the role that databases such as AFLOW[19], MaterialsProject[20], the Open Quantum Materials Database[21], the Computational Materials Repository[22], the International Crystallographic Structure Database[23], among many others, play in materials discovery.

II. SUMMARY OF THE COLLECTION

A. Methods and Programs

A number of contributions to this special issue propose new computational methods, techniques or algorithms to accelerate materials design, or even program packages tailored for the discovery of a specific set of materials. We start by noting that a common bottleneck in computational materials research is the time required for structural relaxation. This prompted Yang, Jiménez-Negrón, and Kitchin to develop a neural network to accelerate this process[24]. Their procedure speeds-up optimizations by relaxing several configurations simultaneously, occasionally obtaining DFT energies, and enabling information about the relaxation of one configuration to guide another. Significant improvements in relaxation time for a number of metallic surfaces, slabs, and nanoparticles are shown. ML-based interatomic potentials **used with MD simulations tend to run more quickly than DFT calculations, however ML models tend to work best for the systems they are trained on. Their application to other systems introduces uncertainty, which must be propagated in calculating observables such as thermodynamic properties.** In regions of configurational space that have not yet been explored these simulations lose accuracy. Imbalzamo and co-workers show that using a robust potential in extrapolative regimes makes the MD more reliable, and methods are developed to estimate uncertainty in thermodynamic quantities[25]. Applications to fluid systems ranging from water to liquid metals are presented. Alternatively, Tran et al. suggest that instead of predicting the "best" material for a particular application, it may be more useful to classify materials into those that are, and those that aren't worthy of experimental investigation[26]. Towards this end they develop a method that takes into account adsorption at multiple length scales and automates the selection of DFT calculations to be performed, applying it to potential catalysts. We note that a recurring theme within this collection will be the use of ML to screen chemical structures for certain features.

Out of the next few ML-focused papers, the first figures on future filtering by feature-finding. Atomic environment fingerprints describe and classify crystal structures, and they can be used to analyze large materials science datasets or as input for CSP. Parsaeifard et al. present a method to calculate fingerprint vectors from fingerprint distances, which measure the similarity of two structures[27]. This method is shown to automatically detect grain boundaries, and the edges of carbon flakes without human input. Taking a more dynamic approach, Weinreich, Browning, and Lilienfeld use ML and short MD simulations to predict solvation properties. Accu-

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rate calculation of solvation free energies are essential for the computational study of chemical reaction mechanisms, and in materials design. The authors' technique queries the FreeSolv database and, given a short MD simulation, constructs a Boltzmann averaged representation of a chemical compound. This is used as input for an ML algorithm to predict solvation properties across a large database of small organic molecules[28]. Finally, the identification of features gets abstracted into quantum chemical parameters. Zubatiuk et al. develop a neural network, trained on DFT densities and orbital energies, to dynamically find the optimal values of the physically motivated parameters used in extended Hückel, or tight-binding Hamiltonians[29]. This method may lead towards the development of ML parametrized, accurate physics models of molecules and solids.

Turning to tailored packages, Lou et al. present the Property Analysis and Simulation Package (PASP), developed to facilitate modelling of complex condensed matter systems[30]. PASP contains basin-hopping and evolutionary based CSP algorithms, a symmetry analysis program, as well as various effective Hamiltonians and Monte Carlo methods. The package could be used to propose a general form of a Hamiltonian for a system based on its symmetry analysis, create configurations and submit them for DFT calculations whose results are used to construct effective Hamiltonians with parameters determined by ML, as well as evaluate the ground electronic state and thermodynamic properties. And in the vein of pedagogy, Turcani et al. introduce the latest version of their supramolecular toolkit, *stk*[31]. This Python library is designed to generate supramolecular structures as input for quantum mechanical software, and is a powerful visualization tool. New features include support for metal-containing systems and rotaxanes, a plethora of topological-molecular-building blocks, tools for database construction, and even a genetic algorithm for traversing chemical space.

B. Organics, Organometallics and Metal-Organics

Materials whose building blocks are derived from organic molecules can be used in a range of applications including pharmaceuticals, polymers, and energy materials. The performance of organic semiconductors rely on crystal structures derived from molecular packing patterns. Predicting these patterns is the focus of Ai, Risko, and co-workers, who present a framework for their *in silico* efforts. The resulting open-access infrastructure is OCELOT (Organic Crystals in Electronic and Light-Oriented Technologies), which contains over 56,000 crystal structures, 15,000 of which are supplemented with DFT data[32]. Each structure is paired with descriptors for convenient searching: crystallographic, mechanical, and electronic properties all verified from experiment or calculation. OCELOT comes with a web-based interface and a python-based API for researchers engaging in machine learning and materials discovery.

Calculating phase boundaries for organic molecular crystals as a function of temperature and pressure is difficult because of the sensitivity of the results to model errors, and the computational cost required for an accurate calculation. Cook, McKinley, and Beran propose a method to calculate the Gibbs free energies by combining DFT with dispersion-corrected density functional tight binding (DFTB) within the quasi-harmonic approximation[33]. The predicted phase boundaries between α and β-resorcinol are in excellent agreement with experiment, and the unit cell volumes are predicted to within 1-2% at low pressures.

We move from elements of thermodynamics to electronics through magnification of molecular materials and examination of electronic structure. **Ramprasad and colleagues** carry out high-throughput DFT calculations on a suite of existing polymers to obtain a dataset of band gaps and charge injection barriers used to develop an ML model[34]. The resulting ML model was employed to predict these properties for a set of 13,000 known polymers, and to find the most promising materials to be used in high-voltage power lines. In another contribution, Valdiviezo, Zhang, and Beretan investigate how charge might be pushed along a polymer, such as DNA, via a racheting motion[35]. Calculations at the microscale and nanoscale illustrate how a rotating electric field causes an electronic hole to rachet along the DNA.

Phonon characterization is the task of Kayastha and Ramakrishnan, who examine a 1D array of organic rings and metal atoms for periodic instabilities[36]. These are then classified based on their periodicity, from low-to-high: Peierls distortions, charge-density waves, and long-wave effects. This high-throughput scheme can create distorted structures that are more stable than the parents.

Finally, Ai, Schrier, and colleagues present a study on amine-templated metal oxides whose compositions and structures were extracted from the Cambridge Structural Database[37]. They asked if a ML model can be trained to predict products from a set of reactants, with a particular focus on determining the dimensionality of the inorganic component. An artificial neural network is trained towards this end, and a number of metal oxide compounds are synthesized and characterized.

C. Inorganic Materials

A wide variety of inorganic materials are studied and predicted in this special issue, each with a unique computational technique. The inherent local disorder in high entropy alloys (HEA), attractive for their structural and functional properties, make them challenging to study due via DFT. This prompted Feugmo and co-workers to develop a computationally efficient method that combines artificial neural networks and evolutionary algorithms to generate HEAs with large configurational entropy[38]. **DeCost and colleagues** illustrate that ML models trained in accordance with current best practices can choose a randomly generated feature, as opposed to one that has a physical meaning, as key for classifying a material[39]. Applications to HEAs show that by re-tooling their ML to search for a Pareto front, it is possible to build a predictive, explainable model across a set of parameters.

The presence of ML methods persist beyond HEAs into the pursuit of exotic compounds. Giri and co-workers use open-source quantum mechanical software, information from the Materials Project, and an ML classifier coupled with a Hirshfeld surface analysis to identify materials that are metallic, covalent or metavalent[40]. Metavalent solids are a unique class of compounds possessing properties characteristic of both metallic and covalently bonded materials, though they are very different from both. In another study, Koyama et al. developed a ML model that recommended synthesizable compositions, which were experimentally explored using the single-particle diagnosis method[41]. The authors draw data from the Inorganic Crystal Structure Database to find novel La-Si ternary and quaternary nitrides, synthesizing several of them and confirming their structure with DFT.

An increasingly important way to predict novel inorganic materials is via CSP methods interfaced with DFT calculations. In one contribution Lu and co-workers show that biasing the *ab initio* random structure searching (AIRSS) method[42] with chemically reasonable constraints, such as minimum interatomic separations, volumes and space group symmetries allows them to identify a novel family of transition-metal oxalates whose properties are consistent with promising cathode materials[43]. In a DFT+U study Sharan and Lany locate thermodynamically stable and metastable ternary oxynitrides by combining random sampling and basin hopping within the kinetically limited minimization algorithm for small unit cells, whereas larger cells are constructed via sampling atomic configurations on parent lattices[44].

As with organic materials, insight about macroscopic properties of inorganics can be gained from accurate electronic structure calculations. Predicting materials for electrocatalysis and photoelectrochemistry require comparison of the calculated ionization potentials and electron affinities with experiment. This can only be done if the calculated values are calibrated with respect to an external potential, and many of the current DFT based workarounds are too expensive for high-throughput calculations. Harnett-Caulfield and Walsh suggest using a reference potential based on the bulk electrostatic potential, applying the method to 27 zinc blende type compounds[45]. While investigating the ground state electronic structure and many-body excitonic behavior of CuCl and CuBr, **Duan and colleagues** discover a cautionary tale about selecting the right functional for the job[46]. As a starting point for the GW-BSE approach, which can yield the absorption spectrum of solids, a one-electron DFT band structure is required. Unexpectedly, use of the PBE pseudopotential yields good exciton binding energies, while use of a new HF pseudopotential leads to better estimates of the band gap.

D. High Pressure Materials

The computational prediction of materials that become stable at high pressure is of relevance to energetic, geological, planetary, quantum, superhard materials and more. Under pressure, compounds may crystallize in novel structures with unique stoichiometries, have completely unprecedented electronic, magnetic and superconducting properties, and adopt unique bonding schemes[47]. At the same time, high pressure experiments are expensive and the observables difficult to interpret. Therefore, first-principles calculations are key tools in studying materials at extreme conditions of pressure[48]. Existing materials databases do not contain information about high pressure compounds, whose chemical space is still widely unexplored. For this reason, DFT is often coupled with crystal structure prediction (CSP) techniques such as random searching, particle swarm optimization and evolutionary algorithms, towards the discovery of unprecedented high pressure materials.

This collection contains two theoretical contributions that illustrate how first-principles calculations can shed light on materials whose structures are modified under pressure. When compressed, p-block covalently bonded materials, such as those containing carbon or boron nitride (BN), can undergo sp^2 to sp^3 phase transitions. Yang and co-workers employed DFT calculations to study the structures formed by double-walled boron nitride nanotubes (DW-BNNTs) under hydrostatic pressures up to 60 GPa[49]. They investigated their structural evolution under pressure, which resulted in 12 kinds of new monolith phases that were metastable upon quenching to 1 atm. These materials were found to be semiconductors with broad optical absorbance in the UV and visible, and were shown to be superhard. In the second contribution, **Dumitrică and colleagues** explored large diameter carbon nanotubes with shapes that can be attained by the application of pressure or mechanical deformation[50]. The deformation results in the formation of a characteristic “dog-bone” cross section, wherein the centers of large tubes squish to form of a bilayer, and the sides curve outwards. DFT calculations suggested that when stacked these nanotubes might form sheets by allowing the looped sides to be offset into an armchair shape, which provides the material with graphitic mechanical properties with extra resistance to compression via the looped sides, which are also key for the resulting flexoelectricity.

New insights in high pressure research are often obtained through the synergy between theory and experiment. **Tse and colleagues** studied the ^{57}Fe Mössbauer isomer shift of pure iron and iron oxides up to 102 GPa both experimentally and theoretically[51]. The best agreement between theory and experiment was obtained with the B1WC hybrid functional. Further calculations were carried out on FeO and Fe_2O_3, whose electronic structure was interrogated. In another work, Cadatal-Raduban and co-workers investigated KMgF as a potential scintillator material[52]. Fast luminescence decay can be achieved when a hole in the core band recombines with an electron in the valence band. DFT and GW computations were performed to identify the transitions that led to the experimentally observed cross-luminescence spectrum. Further calculations showed that the application of pressure lowers the energy gap between the bands, thereby making it possible to tune the luminescence by red-shifting it into the atmospherically-transmissible UV region.

Four of the contributions to this issue employed algorithms for CSP to uncover new materials that are stable when squeezed. One of the most popular techniques for crystal structure prediction, CALYPSO[53], which is based on swarm intelligence, was employed by Du et al. to explore the Ir-N phase diagram up to 100 GPa[54]. Two novel nitrogen-rich phases, IrN_4 and IrN_7, based upon IrN_6 octahedra and planar N_4 or cyclo-N_5 units, were discovered. Both were thermodynamically stable under pressure, could be quenched to 1 atm, and their electronic structure and mechanical properties were studied computationally. Conway and co-workers also employed the CALYPSO algorithm, this time to explore the phase diagram of $(NH_3)(HF)_n$ up to 300 GPa[55]. Because of the similarity of their phases, NH_4F is often considered an analog to ice. In this contribution the authors concluded that NH_4F departs from the structural trends observed in ice above 80 GPa, preferrentially adopting closed-packed ionic structures. Although no stable binary K-Cu compounds are known at 1 atm, CSP carried out with the CALYPSO package and the USPEX[56] evolutionary algorithm revealed that under moderate pressures K_3Cu_2, K_2Cu, K_5Cu_2 and K_3Cu become thermodynamically stable[57]. Cao et al. studied the bonding within these phases as well as their electronic structures. Moreover, they showed the X-ray diffraction pattern of K_3Cu_2 matches that of an unidentified K-Cu compound formed more than a decade earlier. Finally, the XTALOPT[58] evolutionary algorithm for *a priori* crystal structure prediction was used to search for metal oxyfluorides containing a cation from group 11 of the periodic table, focusing on the Ag_2OF_2 stoichiometry[59]. Domańsi and Grochala investigated the geometric and electronic structures of the low enthalpy phases, and proposed synthetic approaches that may be used to synthesize those of interest.

E. Porous Structures

Porous structures, such as metal-organic frameworks (MOFs) or zeolites, are actively researched due to their applications in separation science, catalysis, and biotechnology. MOFs are often employed as fixed-absorption materials for the separation of gas mixtures. Because identifying the best MOF for a given mixture and set of temperature/pressure conditions is a daunting task, Anderson and Gómez-Gauldrón develop an ML-based computational framework to accelerate the process[60]. The new technique, based upon a deep learning model for predicting single-component adsorption properties, coupled with ideal adsorbed solution theory was able to identify the same top-performing MOFs as traditional molecular simulation. Moreover, a set of so-called “privileged” MOFs that perform well on a wide range of separations and across a broad spectrum of conditions was identified. Li et al. studied gas adsorption in MOFs via a linear regression method and the non-linear random forest technique[61]. A histogram of interaction energies obtained from grand canonical Monte Carlo simulations was used as both the training and test data for the adsorption energy and selectivity predictions. Though the ML model was not always highly accurate, it excelled at ranking materials, unveiling those worthy of further investigation.

Key to the rational design of zeolites with desired topologies is the computational modeling of their interaction with organic structure-directing agents. Unfortunately, the large unit cell sizes of these systems makes it prohibitively expensive to study them with DFT-based high-throughput screening, necessitating the use of force fields. Interaction energies between the two can be obtained via static or dynamic calculations. In this contribution Schwalbe-Koda and Gómez-Bombarelli illustrate that the Dreiding force field coupled with frozen pose calculations provides comparable energies to DFT using the PBE functional supplemented with Grimme’s D3 dispersion correction, and yield a significant computational speed up as compared to MD[62].

F. Nanoparticles

The emergence of macroscopic properties from atomic properties as a function of particle size has long been a topic of philosophical debate and determined scientific study.[63] Motivated by the lack of proposed synthesis pathways for predicted materials, Fong and co-workers developed an autonomous synthesizer for palladium nanoparticles integrated with a synchrotron x-ray source and an ML controller[64]. The controller, trained on an initial set of synthesized nanoparticles and, optimizing for size, suggested new recipes for further synthesis. This process was repeated until nanoparticles satisfying the desired constraints were synthesized. Stretching the philosphical idea of a neural network, Wei and co-workers theorize a miniature brain composed of gold nanoparticles wrapped by a conductive polymer. They carry out course grained molecular dynamics simulations on a 2D array to study the emulation of brain-like computing.[65]. As the temperature is increased, polymers establish links with neighboring nanoparticles, but an applied electric field produces the same effect. Properties required for brain-like computing and storage in response to electric fields are observed in the simulations.

III. CONCLUSIONS

This special issue showcases the development of methods that can be employed in computational materials design, and it presents specific predictions that are sure to stimulate further experiments. Many authors have contributed by developing tools and systems, some rigorously tested materials for desirable properties, and others have postulated completely new and exciting materials. Together, they showcase the strengths of computers in materials science: the high-throughput capacity for characterization and the ability of machine learning algorithms to aid in prediction and categorization tasks. The breadth of applications, variety of topics, and excellent quality of the published manuscripts clearly shows the importance of computations in the materials discovery process. We believe that the next decade will see the even tighter integration of computations in the search for novel materials, and are looking forward to witness future advances.

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REFERENCES

[1] K. Alberi, M. B. Nardelli, A. Zakutayev, L. Mitas, S. Curtarolo, A. Jain, M. Fornari, N. Marzari, I. Takeuchi, M. L. Green, M. Kanatzidis, M. F. Toney, S. Butenko, B. Meredig, S. Lany, U. Kattner, A. Davydov, E. S. Toberer, V. Stevanovic, A. Walsh, N.-G. Park, A. Aspuru-Guzik, D. P. Tabor, J. Nelson, J. Murphy, A. Setlur, J. Gregoire, H. Li, R. Xiao, A. Ludwig, L. W. Martin, A. M. Rappe, S.-H. Wei, and J. Perkins, J. Phys. D: App. Phys. **52**, 013001 (2018).
[2] M. Somayazulu, M. Ahart, A. K. Mishra, Z. M. Geballe, M. Baldini, Y. Meng, V. V. Struzhkin, and R. J. Hemley, Phys. Rev. Lett. **122**, 027001 (2019).
[3] A. P. Drozdov, P. P. Kong, V. S. Minkov, S. P. Besedin, M. A. Kuzovnikov, S. Mozaffari, L. Balicas, F. F. Balakirev, D. E. Graf, V. B. Prakapenka, E. Greenberg, D. A. Knyazev, M. Tkacz, and M. I. Eremets, Nature **569**, 528–531 (2019).
[4] E. Snider, N. Dasenbrock-Gammon, R. McBride, X. Wang, N. Meyers, K. V. Lawler, E. Zurek, A. Salamat, and R. P. Dias, Phys. Rev. Lett. **126**, 117003 (1–6) (2021).
[5] P. Sarker, T. Harrington, C. Toher, C. Oses, M. Samiee, J. P. Maria, D. W. Brenner, K. S. Vecchio, and S. Curtarolo, Nat. Commun. **9**, 4980 (2018).
[6] A. O. Oliynyk, E. Antono, T. D. Sparks, L. Ghadbeigi, M. W. Gaultois, B. Meredig, and A. Mar, Chem. Mat. **28**, 7324–7331 (2016).
[7] R. Gomez-Bombarelli, J. Aguilera-Iparraguirre, T. D. Hirzel, D. Duvenaud, D. Maclaurin, M. A. Blood-Forsythe, H. S. Chae, M. Einzinger, D.-G. Ha, T. Wu, G. Markopoulos, S. Jeon, H. Kang, H. Miyazaki, M. Numata, S. Kim, W. Huang, S. I. Hong, M. Baldo, R. P. Adams, and A. Aspuru-Guzik, Nat. Mater. **15**, 1120+ (2016).
[8] G. Ceder, Y. Chiang, D. Sadoway, M. Aydinol, Y. Jang, and B. Huang, Nature **392**, 694–696 (1998).
[9] A. Pulido, L. Chen, T. Kaczorowski, D. Holden, M. A. Little, S. Y. Chong, B. J. Slater, D. P. McMahon, B. Bonillo, C. J. Stackhouse, A. Stephenson, C. M. Kane, R. Clowes, T. Hasell, A. I. Cooper, and G. M. Day, Nature **543**, 657+ (2017).
[10] L. Zhu, G. M. Borstad, H. Liu, P. A. Guńka, M. Guerette, J.-A. Dolyniuk, Y. Meng, E. Greenberg, V. B. Prakapenka, B. L. Chaloux, A. Epshteyn, R. E. Cohen, and T. A. Strobel, Sci. Adv. **6**, eaay8361 (2020).
[11] D. C. Lonie and E. Zurek, Comput. Phys. Commun. **182**, 372–387 (2011).
[12] C. W. Glass, A. R. Oganov, and N. Hansen, Comp. Phys. Comm. **175**, 713–720 (2006).
[13] A. H. Larsen, J. J. Mortensen, J. Blomqvist, I. E. Castelli, R. Christensen, M. Dułak, J. Friis, M. N. Groves, B. Hammer, C. Hargus, E. D. Hermes, P. C. Jennings, P. B. Jensen, J. Kermode, J. R. Kitchin, E. L. Kolsbjerg, J. Kubal, K. Kaasbjerg, S. Lysgaard, J. B. Maronsson, T. Maxson, T. Olsen, L. Pastewka, A. Peterson, C. Rostgaard, J. Schiøtz, O. Schütt, M. Strange, K. S. Thygesen, T. Vegge, L. Vilhelmsen, M. Walter, Z. Zeng, and K. W. Jacobsen, J. Phys-Condens. Mat. **29**, 273002 (2017).
[14] K. Mathew, J. H. Montoya, A. Faghaninia, S. Dwarakanath, M. Aykol, H. Tang, I. heng Chu, T. Smidt, B. Bocklund, M. Horton, J. Dagdelen, B. Wood, Z.-K. Liu, J. Neaton, S. P. Ong, K. Persson, and A. Jain, Comp. Mat. Sci. **139**, 140–152 (2017).
[15] J. Behler and M. Parrinello, Phys. Rev. Lett. **98** (2007).
[16] Y. Dan, Y. Zhao, X. Li, S. Li, M. Hu, and J. Hu, npj Comp. Mat. **6** (2020).
[17] I. S. Novikov, K. Gubaev, E. Podryabinkin, V, and A. Shapeev, V, Mach. Learn.: Sci. Technol. **2** (2021).
[18] T. Xie and J. C. Grossman, J. Chem. Phys. **149** (2018).
[19] S. Curtarolo, W. Setyawan, G. L. 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, and D. Morgan, Comput. Mater. Sci. **58**, 218–226 (2012).
[20] A. Jain, S. P. Ong, G. Hautier, W. Chen, W. D. Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder, and K. A. Persson, APL Mater. **1**, 011002 (2013).
[21] J. E. Saal, S. Kirklin, M. Aykol, B. Meredig, and C. Wolverton, JOM **65**, 1501–1509 (2013).
[22] D. D. Landis, J. S. Hummelshoj, S. Nestorov, J. Greeley, M. Dulak, T. Bligaard, J. K. Norskov, and K. W. Jacobsen, Comput. Sci. Eng. **14**, 51–57 (2012).
[23] G. Bergerhoff, R. Hundt, R. Sievers, and I. D. Brown, J. Chem. Inf. Comp. Sci. **23**, 66–69 (1983).
[24] Y. Yang, O. A. Jiménez-Negrón, and J. R. Kitchin, J. Chem. Phys. **154**, 234704 (2021).
[25] G. Imbalzano, Y. Zhuang, V. Kapil, K. Rossi, E. A. Engel, F. Grasselli, and M. Ceriotti, J. Chem. Phys. **154**, 074102 (2021).
[26] K. Tran, W. Neiswanger, K. Broderick, E. Xing, J. Schneider, and Z. W. Ulissi, J. Chem. Phys. **154**, 124118 (2021).
[27] B. Parsaeifard, D. Tomerini, D. S. De, and S. Goedecker, J. Chem. Phys. **153**, 214104 (2020).
[28] J. Weinreich, N. J. Browning, and O. A. von Lilienfeld, J. Chem. Phys. **154**, 134113 (2021).
[29] T. Zubatiuk, B. Nebgen, N. Lubbers, J. S. Smith, R. Zubatyuk, G. Zhou, C. Koh, K. Barros, O. Isayev, and S. Tretiak, J. Chem. Phys. **154**, 244108 (2021).
[30] F. Lou, X. Y. Li, J. Y. Ji, H. Y. Yu, J. S. Feng, X. G. Gong, and H. J. Xiang, J. Chem. Phys. **154**, 114103 (2021).
[31] L. Turcani, A. Tarzia, F. T. Szczypiński, and K. E. Jelfs, J. Chem. Phys. **154**, 214102 (2021).
[32] Q. Ai, V. Bhat, S. M. Ryno, K. Jarolimek, P. Sornberger, A. Smith, M. M. Haley, J. E. Anthony, and C. Risko, J. Chem. Phys. **154**, 174705 (2021).
[33] C. Cook, J. L. McKinley, and G. J. O. Beran, J. Chem. Phys. **154**, 134109 (2021).
[34] D. Kamal, H. Tran, C. Kim, Y. Wang, L. Chen, Y. Cao, V. R. Joseph, and R. Ramprasad, J. Chem. Phys. **154**, 174906 (2021).

[35]J. Valdiviezo, P. Zhang, and D. N. Beratan, J. Chem. Phys. **155**, 055102 (2021).

[36]P. Kayastha and R. Ramakrishnan, J. Chem. Phys. **154**, 061102 (2021).

[37]Q. Ai, D. M. Williams, M. Danielson, L. G. Spooner, J. A. Engler, Z. Ding, M. Zeller, A. J. Norquist, and J. Schrier, J. Chem. Phys. **154**, 184708 (2021).

[38]C. G. Tetsassi Feugmo, K. Ryczko, A. Anand, C. V. Singh, and I. Tamblyn, J. Chem. Phys. **155**, 044102 (2021).

[39]K. Lei, H. Joress, N. Persson, J. R. Hattrick-Simpers, and B. DeCost, J. Chem. Phys. **155**, 054105 (2021).

[40]D. Giri, L. Williams, A. Mukherjee, and K. Rajan, J. Chem. Phys. **154**, 124105 (2021).

[41]Y. Koyama, A. Seko, I. Tanaka, S. Funahashi, and N. Hirosaki, J. Chem. Phys. **154**, 224117 (2021).

[42]C. J. Pickard and R. J. Needs, Phys. Status Solidi B **246**, 536–540 (2009).

[43]Z. Lu, B. Zhu, B. W. B. Shires, D. O. Scanlon, and C. J. Pickard, J. Chem. Phys. **154**, 174111 (2021).

[44]A. Sharan and S. Lany, J. Chem. Phys. **154**, 234706 (2021).

[45]L. Harnett-Caulfield and A. Walsh, J. Chem. Phys. **155**, 024113 (2021).

[46]Y. Wu, Z. Jiang, H. Tan, Y. Li, and W. Duan, J. Chem. Phys. **154**, 134704 (2021).

[47]M. Miao, Y. Sun, E. Zurek, and H. Lin, Nat. Rev. Chem. **508-527**, 4 (2020).

[48]E. Zurek and W. Grochala, Phys. Chem. Chem. Phys. **17**, 2917–2934 (2015).

[49]X. Yang, S. Zhou, S. Huang, and J. Zhao, J. Chem. Phys. **154**, 134702 (2021).

[50]C. Hu, A. Dernov, H. Xu, G. Drozdov, and T. Dumitrică, J. Chem. Phys. **154**, 044701 (2021).

[51]J. K. Desmarais, W. Bi, J. Zhao, M. Y. Hu, E. Alp, and J. S. Tse, J. Chem. Phys. **154**, 214104 (2021).

[52]M. Cadatal-Raduban, K. Yamanoi, A. Yoshikawa, Y. Yokota, T. Shimizu, N. Sarukura, T. Togashi, A. Kondo, and M. V. Luong, J. Chem. Phys. **154**, 124707 (2021).

[53]Y. Wang, J. Lv, L. Zhu, and Y. Ma, Phys. Rev. B **82**, 094116 (2010).

[54]X. Du, Y. Yao, J. Wang, Q. Yang, and G. Yang, J. Chem. Phys. **154**, 054706 (2021).

[55]L. J. Conway, K. Brown, J. S. Loveday, and A. Hermann, J. Chem. Phys. **154**, 204501 (2021).

[56]C. W. Glass, A. R. Oganov, and N. Hansen, Comput. Phys. Commun. **175**, 713–720 (2006).

[57]X. Cao, B. Wan, H. Liu, L. Wu, Y. Yao, and H. Gou, J. Chem. Phys. **154**, 134708 (2021).

[58]Z. Falls, P. Avery, X. Wang, K. P. Hilleke, and E. Zurek, J. Phys. Chem. C **125**, 1601–1620 (2021).

[59]M. A. Domański and W. Grochala, J. Chem. Phys. **154**, 204705 (2021).

[60]R. Anderson and D. A. Gómez-Gualdrón, J. Chem. Phys. **154**, 234102 (2021).

[61]Z. Li, B. J. Bucior, H. Chen, M. Haranczyk, J. I. Siepmann, and R. Q. Snurr, J. Chem. Phys. **155**, 014701 (2021).

[62]D. Schwalbe-Koda and R. Gómez-Bombarelli, J. Chem. Phys. **154**, 174109 (2021).

[63]J. R. Bursten, M. C. Roco, W. Yang, Y. Zhao, C. Chen, K. Savolainen, C. Gerber, K. Kataoka, Y. Krishnan, H. Bayley, L. Nazar, S. Milana, L. Vandersypen, P. S. Weiss, and J. Schummer, Nat. Nanotechnol. **11**, 828–834 (2016).

[64]A. Y. Fong, L. Pellouchoud, M. Davidson, R. C. Walroth, C. Church, E. Tcareva, L. Wu, K. Peterson, B. Meredig, and C. J. Tassone, J. Chem. Phys. **154**, 224201 (2021).

[65]X. Wei, Y. Zhao, Y. Zhuang, and R. Hernandez, J. Chem. Phys. **154**, 214702 (2021).