NANO LETTERS

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Topogivity: A Machine-Learned Chemical Rule for Discovering Topological Materials

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Cite This: *Nano Lett.* 2023, 23, 772−778

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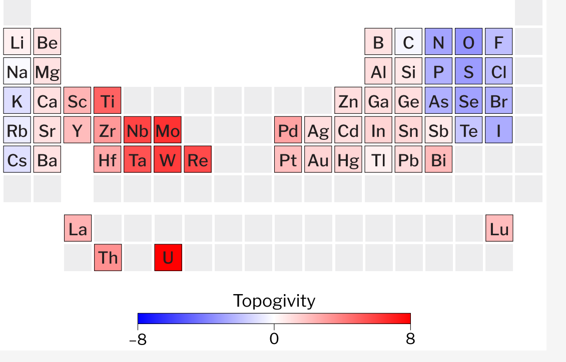
ABSTRACT: Topological materials present unconventional electronic properties that make them attractive for both basic science and next-generation technological applications. The majority of currently known topological materials have been discovered using methods that involve symmetry-based analysis of the quantum wave function. Here we use machine learning to develop a simple-to-use heuristic chemical rule that diagnoses with a high accuracy whether a material is topological using only its chemical formula. This heuristic rule is based on a notion that we term *topogivity*, a machine-learned numerical value for each element that loosely captures its tendency to form topological materials. We next implement a high-throughput procedure for discovering topological materials based on the heuristic topogivity-rule prediction followed by ab initio validation. This way, we discover new topological materials that are not diagnosable using symmetry indicators, including several that may be promising for experimental observation.



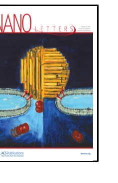
KEYWORDS: *topological materials, machine learning, chemical intuition, materials discovery, interpretability*

Topological materials, including both topological insulators[1−8] and topological semimetals,[9−13] are unconventional phases of matter characterized by topologically nontrivial electron wave functions. Since the beginning of the field, an important and enduring question has been how to determine whether a given electronic material is topological. Efforts to answer this question have largely relied on first-principles calculations in synergy with topological band theory.[14,15] In particular, recently developed theories known as symmetry indicators[16] and topological quantum chemistry[17] allow the diagnosis of a wide range of topological materials using symmetry-based analysis of the wave function.[18−20] These symmetry-based methods require relatively low computational cost and have enabled high-throughput computational searches for topological materials.[21−23] Despite these successes, symmetry indicators have limited diagnostic power for certain forms of band topology or when applied to low-symmetry crystal structures.[16] For example, the first experimentally observed Weyl semimetal, tantalum arsenide (TaAs),[10,11] is a non-symmetry-diagnosable topological material (i.e., a topological material whose topology is undetectable by symmetry indicators).[21] Its topological nature is established by calculating the wave function-based topological invariant directly,[24] which involves significant computational cost. From a broad conceptual standpoint, topological materials such as Chern insulators and time-reversal-invariant Z_2 topological insulators are robust against any small perturbation breaking all crystal symmetries, which renders symmetry indicators inapplicable even though topology remains intact. Thus, for both practical and fundamental reasons, it is highly desirable to develop accurate and simple-to-use rules to determine whether *any* given material is topological.

Many aspects of materials can be understood at a heuristic level from a chemistry perspective. A well-known example is bonding, which can be understood using quantum mechanical approaches such as molecular orbital theory,[25] as well as using heuristics such as the difference of element electronegativities. While quantum theory can provide greater detail and accuracy, chemical heuristics can often provide valuable insight and a useful guide for materials discovery. Is there a deep chemical reason why a particular material is topological? To what extent can topological materials be understood and identified using chemical heuristic approaches? While connections between chemistry and electronic band topology (e.g., based on the presence or absence of certain elements) have been explored,[26−31] existing chemical heuristics do not provide a broadly applicable path for finding topological materials.

Here, we use machine learning (ML) to help us search for chemical origins of topological electronic structure in materials. Recently, ML has become a powerful approach for advancing

Received: August 25, 2022
Revised: January 10, 2023
Published: January 20, 2023



https://doi.org/10.1021/acs.nanolett.2c03307
Nano Lett. 2023, 23, 772−778

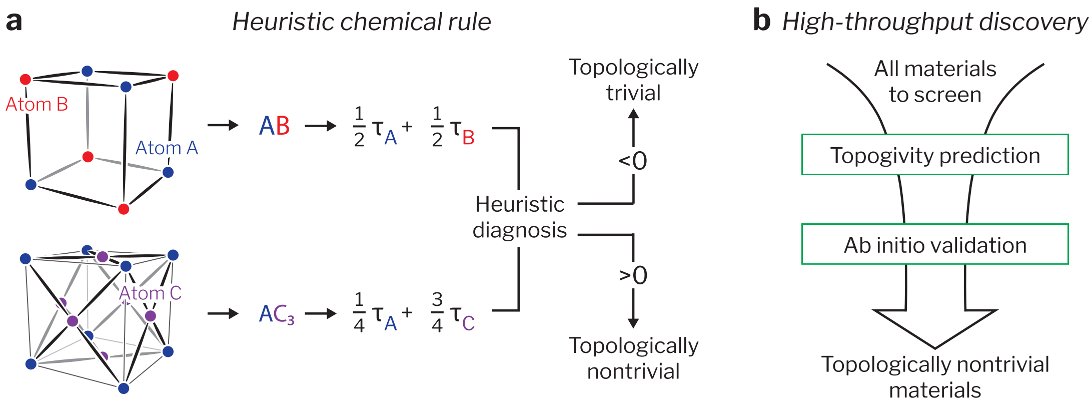


Figure 1. Topogivity-based diagnosis and discovery of topological materials. (a) Given a stoichiometric material, the topogivity-based heuristic diagnosis is evaluated by simply weighting the material's elements' (A, B, and C) topogivities (τ_A, τ_B, and τ_C) by their relative abundance in the chemical formula (AB and AC_3). The sign indicates the topological classification and the magnitude indicates roughly how confident we are in this classification. Each element's topogivity is a machine-learned parameter that loosely captures the element's tendency to form topologically nontrivial materials. (b) We leverage our framework to perform high-throughput topological materials discovery. First, we use the topogivities to rapidly screen through a suitable collection of materials (i.e., the discovery space) in order to find candidate topological materials. Subsequently, we carry out ab initio validation by performing DFT on these candidates. We discovered topological materials that are not diagnosable using the standard symmetry indicators approach.[16]

scientific discovery in materials science.[32−34] In the area of topological materials, researchers have begun to apply ML to both toy models[35−38] and ab initio data.[39−44] While ML has led to important advances for many applications in engineering and science, most ML models act essentially like black boxes: they are complicated models which often provide correct answers, but because of their complexity, they are difficult to understand, and hence provide little insight and intuition about the systems they are applied to. Since a key aim of our work is precisely to learn insights about electronic topology, we instead focus our quest onto interpretable ML models, with a goal of finding a broadly applicable heuristic chemical rule that diagnoses whether or not a material is topological.

The heuristic rule that our ML approach discovered is based on the notion of a learned parameter for each element that loosely captures the tendency of an element to form topological materials. We refer to this as an element's *topogivity*. This heuristic rule is simple, hand-calculable, and interpretable: a given material is diagnosed with high accuracy (typically >80%) as topologically nontrivial (trivial) if the weighted average of its elements' topogivities is positive (negative) (Figure 1a). The heuristic rule does not rely on crystal symmetry, and our approach can be used to make predictions on *all* materials. We integrate the heuristic rule into a high-throughput procedure to search for non-symmetry-diagnosable topological materials, in which we perform screening using the heuristic rule followed by density functional theory (DFT) validation (Figure 1b). The newly discovered topological materials include several high-quality examples that may be promising for experimental realization.

CLASSES OF MATERIALS

Conventional textbook chemistry teaches that the electrons of insulators (including semiconductors) are localized to ionic or covalent bonds, while the electrons of metals are delocalized and "free". From a band-theoretic perspective, the former make up part of the class of materials known as atomic insulators and the latter roughly correspond to "ordinary" metals. Topological insulators and topological semimetals do not fit into this conventional dichotomy. Topological insulators feature a band gap and a nontrivial topological invariant, and as a consequence, their electronic states cannot be reduced to an assembly of localized atomic or molecular orbitals. Topological semimetals have band degeneracies protected by symmetry or topology near or at the Fermi level. Collectively, we refer to topological insulators and topological semimetals as topological materials, and refer to all other materials as trivial materials.

To learn a heuristic chemical rule for diagnosing topological materials, we employ a supervised learning approach. This requires a labeled data set in which each material is labeled as "topological" or "trivial". The learned heuristic chemical rule is then applied to screen another data set which we refer to as the discovery space. Existing ab initio databases of stoichiometric, nonmagnetic, three-dimensional materials[21−23,45] offer a convenient source of data. However, it is important to note that they are imperfect, in part because the symmetry-based high-throughput calculation methods that were used to generate them are inherently incapable of detecting certain topology. Taking such limitations into account, we identify the labeled data set as a subset of the database generated by Tang et al.[21] (see Supplementary Section S1) (our methodology could also be applied to other databases that contain both trivial materials and topological materials). Our labeled data set consists of 9,026 materials, of which 51% are labeled as trivial and the remaining 49% are labeled as topological. However, due to the aforementioned imperfection, for ML purposes this labeled data set should effectively be considered as a data set with noisy labels, e.g., some topological materials are incorrectly labeled as trivial. Separately, the discovery space consists of 1,433 materials, whose topology cannot be determined from the symmetry indicators method (see Supplementary Section S1). By applying the learned heuristic chemical rule to the discovery space and then performing DFT, we are able to evaluate its ability to predict topological materials beyond those diagnosable by existing standard approaches. Some of the topological materials that we identify in the discovery space are known elsewhere in the literature and serve primarily as confirmations, whereas others represent instances of truly new materials discovery.

https://doi.org/10.1021/acs.nanolett.2c03307
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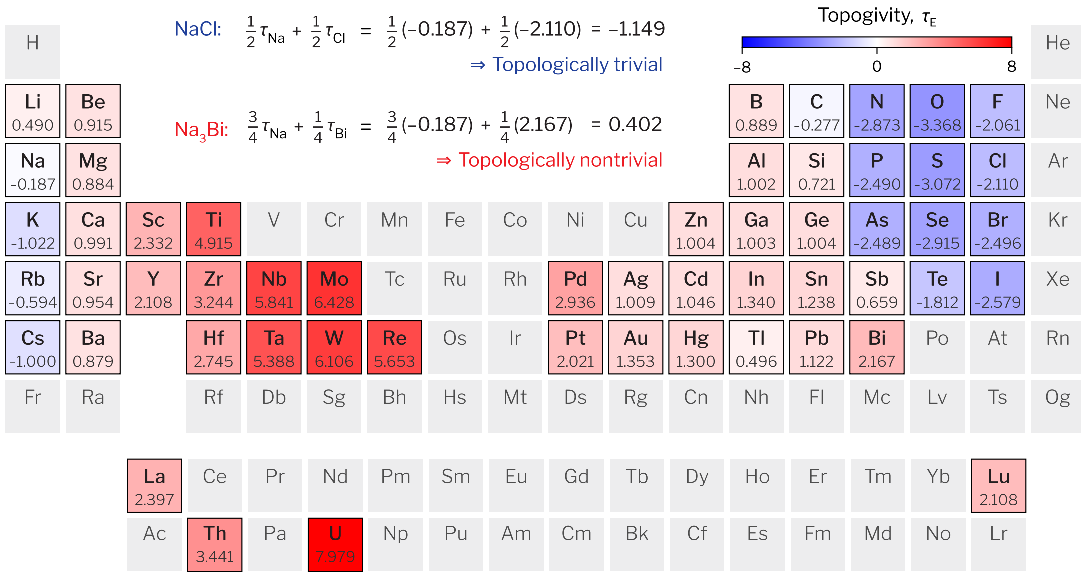

Figure 2. Periodic table of topogivities. Machine-learned topogivities τ_E are shown by color-coding and in values. Elements that do not appear in any material in the data set are shown in gray. Example applications of the topogivity-based heuristic chemical rule are shown for two materials, NaCl (trivial) and Na_3Bi (nontrivial).[9]

LEARNING A HEURISTIC CHEMICAL RULE

Our machine learning model takes the form of a heuristic chemical rule. Specifically, the model maps each material M to a number $g(M)$ according to the function

$$g(M) = \sum_E f_E(M)\tau_E$$

where the summation runs over the elements present in the chemical formula of material M, τ_E is a learned parameter for each element E, and $f_E(M)$ is the element fraction for the element E in material M (e.g., for a chemical formula $A_xB_yC_z$, $f_A(M) = \frac{x}{x+y+z}$, $f_B(M) = \frac{y}{x+y+z}$, and $f_C(M) = \frac{z}{x+y+z}$). Classification decisions are made according to the sign of $g(M)$: classify as topological if positive and classify as trivial if negative. A greater magnitude of $g(M)$ roughly corresponds to a more confident classification decision. We refer to the model as a heuristic *chemical* rule in the sense that all the information required for obtaining a diagnosis is contained in the material's chemical formula. Additional modeling and methodological details are provided in Supplementary Sections S2.A and S2.B; an analysis of information lost from not incorporating spatial information in the classifier is provided in Supplementary Section S3.A.

For each element E, we refer to the optimized parameter τ_E as its *topogivity*. For a given material M, $g(M)$ is simply the weighted average of its elements' topogivities, where the weighting is with respect to each element's relative abundance, as identifiable from the material's chemical formula. Conceptually, an element's topogivity loosely captures its tendency to form topological materials—greater topogivity is intended to roughly correspond to a greater tendency (see Supplementary Section S3.D for details and caveats on the meaning of topogivity).

Before making predictions in the discovery space, we want to first evaluate model performance within the labeled data set. To do this, we use a nested cross validation procedure and average the results over multiple test sets. We find an average of 82.7% accuracy. Additionally, we find empirical evidence that as the magnitude of $g(M)$ is increased, the fraction of correctly classified materials first increases and then plateaus, with the plateau beginning around $|g(M)| \approx 1$. We heuristically set a threshold of 1.0 for a high-confidence topologically nontrivial classification and observe on average that 93.0% of materials with $g(M) \geq 1.0$ are correctly classified. Details and extended results are presented in Supplementary Section S2.C. Additionally, we found that the model works better for materials with two or three distinct elements than for materials with one or four distinct elements (see Supplementary Section S3.B). Having completed nested cross validation, we proceed to use the entire labeled data set to fit the final model (see Supplementary Section S2.D), which is what we will use for making predictions in the discovery space. An additional evaluation of model performance is presented in Supplementary Section S2.E.

We visualize the final model's learned topogivities in Figure 2. This periodic table of topogivities enables an immediate heuristic diagnosis of any stoichiometric material whose elements are featured in the table. This is illustrated with examples in Figure 2 for the trivial insulator NaCl and the Dirac semimetal Na_3Bi.[9] The Weyl semimetal TaAs[10,11] is also worth highlighting: TaAs is non-symmetry-diagnosable[21] and does not appear in the labeled data set, but is successfully diagnosed as topological by the topogivity approach: $g(\text{TaAs}) = \frac{1}{2}\tau_{Ta} + \frac{1}{2}\tau_{As} = 1.450$.

The simplicity of our model enables us to readily extract chemical insights from the periodic table of topogivities. First, we observe that elements that are near each other in the periodic table tend to have similar topogivities, which is consistent with intuition. Second, we observe that the elements with negative topogivities are located in two clusters respectively in the top right and bottom left parts of the periodic table. This is also consistent with intuition, since ionic compounds often have large trivial band gaps and elements from these two clusters tend to form ionic compounds. Third, considering group 15 (the pnictogens), we observe that while N, P, and As have negative topogivities (and Sb has a small

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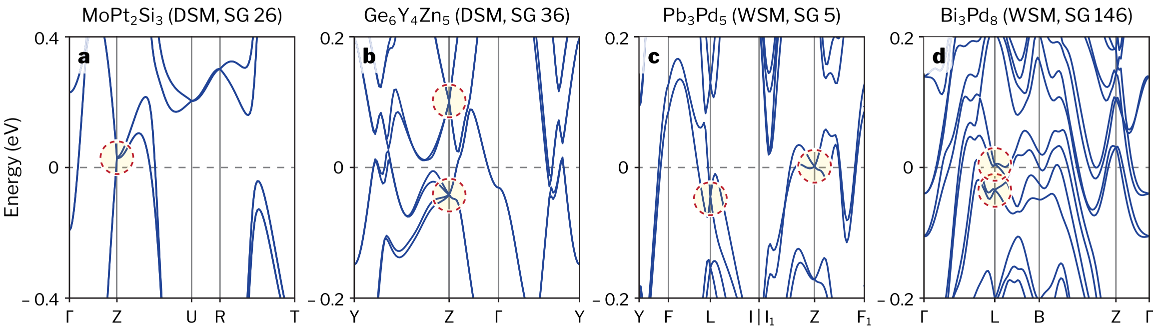

Figure 3. Selection of newly discovered topological materials. These materials are not diagnosable using symmetry indicators,[16] but were successfully discovered using our topogivity-based approach. The band structures were computed using DFT. $MoPt_2Si_3$ (a) and $Ge_6Y_4Zn_5$ (b) are nonsymmorphic Dirac semimetals. Pb_3Pd_5 (c) and Bi_3Pd_8 (d) are Kramers Weyl semimetals. For each material, relevant topological degeneracies are highlighted by circles.

positive topogivity), Bi has a positive topogivity with a relatively large magnitude. This is consistent with the intuition that Bi often plays a role in topological materials.[29] Finally, we observe a region of high topogivities in the early transition metals—future work could attempt to understand the underlying reasons for this (note that there is a chance that typical oxidation states are artificially inflating these topogivities). Overall, while the element topogivities are parameters whose specific learned values are affected by data set and modeling limitations (see Supplementary Section S3.D for discussion), the fact that we can extract chemical insights that are consistent with intuition is evidence that a topogivity-based approximate picture can provide a meaningful way to study topological materials.

HIGH-THROUGHPUT SCREENING AND AB INITIO CALCULATIONS

To identify topological materials using the learned topogivities, we compute $g(M)$ for each of the 1,433 materials in the discovery space. We restrict our attention to the materials that have a $g(M)$ value that corresponds to a topologically nontrivial classification with high-confidence (i.e., $g(M) \geq 1.0$): that leaves 73 materials (after the removal of 2 other materials). Additionally, since it is difficult to obtain accurate DFT calculations for f-electron materials, we exclude any material that contains a 4f or 5f electron, eliminating five materials and thus leaving us with 68 materials for ab initio validation. Full details of our topogivity-based screening procedure (including filtering criteria) are provided in Supplementary Section S4.A.

For each of these 68 materials, we perform DFT within the generalized gradient approximation (GGA).[46] We include spin-orbital coupling in all of our calculations, which is consistent with the database[21] (note that topological classification can depend on the presence or absence of spin-orbital coupling[16]). In our DFT calculations, we checked for many—but not all—types of topological materials. In principle it is possible that some topological materials were not detected by our DFT. Further methodological details are provided in Supplementary Section S4.B.

Of the 68 materials, we find 56 topological materials, corresponding to a success rate of 82.4%. We stress that the discovery space and the labeled data set correspond to different regimes of materials, and so it is quite interesting that a model that was fit on the labeled data set still works in the discovery space (see Supplementary Section S4.C). We note that there are aspects of our procedure and data analysis that could have introduced some bias into the success rate (see Supplementary Section S4.C).

The 56 topological materials that we found consist of 48 Weyl semimetals, 7 Dirac semimetals, and 1 Dirac nodal line semimetal. The band structures of all 56 of these topological materials are included in Supplementary Section S6. Some of these topological materials have previously been predicted in the literature and a smaller portion have also already been experimentally observed, e.g., TaAs.[10,11] More importantly, our DFT calculations also identify multiple new topological materials that to our knowledge have not been previously identified.

We highlight four particularly interesting newly discovered topological materials in Figure 3. Each is a topological semimetal with a relatively clean band structure and at least one band crossing within 50 meV of the Fermi level, making it promising for potential experimental investigation. $MoPt_2Si_3$ and $Ge_6Y_4Zn_5$ are both nonsymmorphic Dirac semimetals. At the Z point, the former has a Dirac point in the valence band manifold and the latter has Dirac points in both the valence and conduction band manifolds. Pb_3Pd_5 and Bi_3Pd_8 are both Kramers Weyl semimetals.[47] The former has Weyl nodes at the L and Z points, and the latter has two Weyl nodes close in energy at the L point. In particular, we highlight that $MoPt_2Si_3$ has a Dirac point close to the Fermi level as well as a relatively clean Fermi surface, and Pb_3Pd_5 has a Weyl node located at the Z point that is right at the Fermi level. We emphasize that the reason the band degeneracies in these four materials are non-symmetry-diagnosable is that they are all within the valence band manifold or conduction band manifold. Such band degeneracies cannot be diagnosed by the symmetry indicators method, which is formulated based on the electron filling and therefore cannot target band degeneracies that are not *between* the valence and conduction bands.[16]

Separately, as a preliminary exploration, we performed DFT calculations on a selection of labeled data set materials that are labeled as trivial but which the model classifies as topological. Our DFT calculations revealed that in some of these cases, the material is actually topological (i.e., the model is correct and it is actually the label that is wrong). Further details—including some selected example materials from this exploration—are included in Supplementary Section S5.

https://doi.org/10.1021/acs.nanolett.2c03307
Nano Lett. 2023, 23, 772−778

DISCUSSION AND OUTLOOK

The topogivity approach provides only a coarse-grained topological classification (nontrivial or trivial); it lacks the fine-grained detail of ab initio approaches. Moreover, it is important to note that topogivity is not an unambiguously defined quantity, as its exact numerical value for each element can depend, for example, on the choice of machine learning algorithm and the use of the weighted average formulation. This fact is illustrated in Supplementary Section S3.C, where we empirically demonstrate the minor impact of making a particular change to the machine learning algorithm. Further discussion pertaining to this lack of an unambiguous definition is included in Supplementary Section S3.D.

Nevertheless, topogivity offers a broadly applicable and simple approach for diagnosing topological materials. This diagnosis uses only the chemical formula and requires merely a handful of arithmetic operations to evaluate. An important highlight of the topogivity-based diagnosis approach is that it enables the discovery of non-symmetry-diagnosable topological materials. Furthermore, the periodic table of topogivities (Figure 2) provides simple intuition for a complex, exotic phenomenon.

One worthy future direction is to look for a more complete understanding of the underlying reasons for the values of the elements' topogivities, which may in turn shed new light on the fundamental question of why some materials are topological while others are not. Another promising path forward would be to perform more comprehensive searches for new topological materials using topogivity-based strategies. Finally, it is intriguing to contemplate whether our interpretable-ML approach, used here to discover topogivity, could perhaps be used for other material properties as well, such as ferroelectricity, ferromagnetism, or maybe even superconductivity.

ASSOCIATED CONTENT

Data Availability Statement

The code and data underlying the machine learning part of this paper is available in our public repository (https://github.com/andrewma8/topogivity).

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.2c03307.

> Description of data sets; description, implementation, and evaluation of the machine-learned topogivity model; analysis, properties, and limitations of the topogivity approach; details on the high-throughput screening and ab initio validation process; ab initio calculations on potentially mislabeled materials; catalog of topological materials identified by the high-throughput process (PDF)

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We thank Pawan Goyal for assisting preliminary work. We thank Samuel Kim, Peter Lu, Rumen Dangovski, and Edward Zhang for helpful discussions. We thank Feng Tang and Xiangang Wan for the sharing of materials data. We thank Paola Rebusco for critical reading and editing of the manuscript. A.M. acknowledges support from the MIT EECS Alan L. McWhorter Fellowship and from the National Science Foundation Graduate Research Fellowship under Grant No. 1745302. Research was sponsored in part by the United States Air Force Research Laboratory and the United States Air Force Artificial Intelligence Accelerator and was accomplished under Cooperative Agreement Number FA8750-19-2-1000. The views and conclusions contained in this document are those of the authors and should not be interpreted as representing the official policies, either expressed or implied, of the United States Air Force or the U.S. Government. The U.S. Government is authorized to reproduce and distribute reprints for Government purposes notwithstanding any copyright notation herein. This material is also based upon work supported in part by the Air Force Office of Scientific Research under the awards number FA9550-20-1-0115, FA9550-21-1-0299, and FA9550-21-1-0317, as well as in part by the US Office of Naval Research (ONR) Multidisciplinary University Research Initiative (MURI) grant N00014-20-1-2325 on Robust Photonic Materials with High-Order Topological Protection. This material is also based upon work supported in part by the U.S. Army Research Office through the Institute for Soldier Nanotechnologies at MIT, under Collaborative Agreement Number W911NF-18-2-0048. This work is also supported in part by the National Science Foundation under Cooperative Agreement PHY-2019786 (The NSF AI Institute for Artificial Intelligence and Fundamental Interactions, http://iaifi.org/). The work of Y.Z. and L.F. was supported by DOE Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award DE-SC0018945 (theoretical analysis) and DE-SC0019275 (band structure calculation). L.F. was partly supported by the Simons Investigator Award from Simons Foundation. The work of H.C.P. was partly supported by a Pappalardo Fellowship at MIT.

REFERENCES

(1) Kane, C. L.; Mele, E. J. Quantum spin Hall effect in graphene. *Phys. Rev. Lett.* **2005**, *95*, 226801.
(2) Bernevig, B. A.; Hughes, T. L.; Zhang, S.-C. Quantum spin Hall effect and topological phase transition in HgTe quantum wells. *Science* **2006**, *314*, 1757.
(3) Konig, M.; Wiedmann, S.; Brune, C.; Roth, A.; Buhmann, H.; Molenkamp, L. W.; Qi, X.-L.; Zhang, S.-C. Quantum spin Hall insulator state in HgTe quantum wells. *Science* **2007**, *318*, 766.
(4) Fu, L.; Kane, C. L.; Mele, E. J. Topological insulators in three dimensions. *Phys. Rev. Lett.* **2007**, *98*, 106803.
(5) Hsieh, D.; Qian, D.; Wray, L.; Xia, Y.; Hor, Y. S.; Cava, R. J.; Hasan, M. Z. A topological Dirac insulator in a quantum spin Hall phase. *Nature* **2008**, *452*, 970.
(6) Fu, L. Topological crystalline insulators. *Phys. Rev. Lett.* **2011**, *106*, 106802.
(7) Hsieh, T. H.; Lin, H.; Liu, J.; Duan, W.; Bansil, A.; Fu, L. Topological crystalline insulators in the SnTe material class. *Nat. Commun.* **2012**, *3*, 982.
(8) Benalcazar, W. A.; Bernevig, B. A.; Hughes, T. L. Quantized electric multipole insulators. *Science* **2017**, *357*, 61.
(9) Liu, Z.; Zhou, B.; Zhang, Y.; Wang, Z.; Weng, H.; Prabhakaran, D.; Mo, S.-K.; Shen, Z.; Fang, Z.; Dai, X.; et al. Discovery of a three-dimensional topological Dirac semimetal, Na_3Bi. *Science* **2014**, *343*, 864.
(10) Xu, S.-Y.; Belopolski, I.; Alidoust, N.; Neupane, M.; Bian, G.; Zhang, C.; Sankar, R.; Chang, G.; Yuan, Z.; Lee, C.-C.; et al. Discovery of a Weyl fermion semimetal and topological Fermi arcs. *Science* **2015**, *349*, 613.
(11) Lv, B.; Weng, H.; Fu, B.; Wang, X. P.; Miao, H.; Ma, J.; Richard, P.; Huang, X.; Zhao, L.; Chen, G.; et al. Experimental discovery of Weyl semimetal TaAs. *Phys. Rev. X* **2015**, *5*, 031013.
(12) Burkov, A.; Hook, M.; Balents, L. Topological nodal semimetals. *Phys. Rev. B* **2011**, *84*, 235126.
(13) Bradlyn, B.; Cano, J.; Wang, Z.; Vergniory, M.; Felser, C.; Cava, R. J.; Bernevig, B. A. Beyond Dirac and Weyl fermions: Unconventional quasiparticles in conventional crystals. *Science* **2016**, *353*, 558.
(14) Xiao, J.; Yan, B. First-principles calculations for topological quantum materials. *Nat. Rev. Phys.* **2021**, *3*, 283.
(15) Bansil, A.; Lin, H.; Das, T. Colloquium: Topological band theory. *Rev. Mod. Phys.* **2016**, *88*, 021004.
(16) Po, H. C.; Vishwanath, A.; Watanabe, H. Symmetry-based indicators of band topology in the 230 space groups. *Nat. Commun.* **2017**, *8*, 50.
(17) Bradlyn, B.; Elcoro, L.; Cano, J.; Vergniory, M.; Wang, Z.; Felser, C.; Aroyo, M.; Bernevig, B. A. Topological quantum chemistry. *Nature* **2017**, *547*, 298.
(18) Fu, L.; Kane, C. L. Topological insulators with inversion symmetry. *Phys. Rev. B* **2007**, *76*, 045302.
(19) Kruthoff, J.; de Boer, J.; van Wezel, J.; Kane, C. L.; Slager, R.-J. Topological classification of crystalline insulators through band structure combinatorics. *Phys. Rev. X* **2017**, *7*, 041069.
(20) Song, Z.; Zhang, T.; Fang, Z.; Fang, C. Quantitative mappings between symmetry and topology in solids. *Nat. Commun.* **2018**, *9*, 3530.
(21) Tang, F.; Po, H. C.; Vishwanath, A.; Wan, X. Comprehensive search for topological materials using symmetry indicators. *Nature* **2019**, *566*, 486.
(22) Zhang, T.; Jiang, Y.; Song, Z.; Huang, H.; He, Y.; Fang, Z.; Weng, H.; Fang, C. Catalogue of topological electronic materials. *Nature* **2019**, *566*, 475.
(23) Vergniory, M.; Elcoro, L.; Felser, C.; Regnault, N.; Bernevig, B. A.; Wang, Z. A complete catalogue of high-quality topological materials. *Nature* **2019**, *566*, 480.
(24) Weng, H.; Fang, C.; Fang, Z.; Bernevig, B. A.; Dai, X. Weyl semimetal phase in noncentrosymmetric transition-metal monophosphides. *Phys. Rev. X* **2015**, *5*, 011029.
(25) Hehre, W. J. Ab initio molecular orbital theory. *Acc. Chem. Res.* **1976**, *9*, 399.
(26) Kumar, N.; Guin, S. N.; Manna, K.; Shekhar, C.; Felser, C. Topological quantum materials from the viewpoint of chemistry. *Chem. Rev.* **2021**, *121*, 2780.
(27) Schoop, L. M.; Pielnhofer, F.; Lotsch, B. V. Chemical principles of topological semimetals. *Chem. Mater.* **2018**, *30*, 3155.
(28) Gibson, Q.; Schoop, L. M.; Muechler, L.; Xie, L.; Hirschberger, M.; Ong, N. P.; Car, R.; Cava, R. J. Three-dimensional Dirac semimetals: Design principles and predictions of new materials. *Phys. Rev. B* **2015**, *91*, 205128.
(29) Isaeva, A.; Ruck, M. Crystal chemistry and bonding patterns of bismuth-based topological insulators. *Inorg. Chem.* **2020**, *59*, 3437.
(30) Klemenz, S.; Hay, A. K.; Teicher, S. M.; Topp, A.; Cano, J.; Schoop, L. M. The role of delocalized chemical bonding in square-net-based topological semimetals. *J. Am. Chem. Soc.* **2020**, *142*, 6350.
(31) Gui, X.; Pletikosic, I.; Cao, H.; Tien, H.-J.; Xu, X.; Zhong, R.; Wang, G.; Chang, T.-R.; Jia, S.; Valla, T.; et al. A new magnetic topological quantum material candidate by design. *ACS Cent. Sci.* **2019**, *5*, 900.
(32) Zhong, M.; Tran, K.; Min, Y.; Wang, C.; Wang, Z.; Dinh, C.-T.; De Luna, P.; Yu, Z.; Rasouli, A. S.; Brodersen, P.; et al. Accelerated discovery of CO_2 electrocatalysts using active machine learning. *Nature* **2020**, *581*, 178.
(33) Deringer, V. L.; Bernstein, N.; Csányi, G.; Mahmoud, C. B.; Ceriotti, M.; Wilson, M.; Drabold, D. A.; Elliott, S. R. Origins of structural and electronic transitions in disordered silicon. *Nature* **2021**, *589*, 59.
(34) George, J.; Hautier, G. Chemist versus machine: Traditional knowledge versus machine learning techniques. *Trends Chem.* **2021**, *3*, 86.
(35) Zhang, Y.; Kim, E.-A. Quantum loop topography for machine learning. *Phys. Rev. Lett.* **2017**, *118*, 216401.
(36) Zhang, P.; Shen, H.; Zhai, H. Machine learning topological invariants with neural networks. *Phys. Rev. Lett.* **2018**, *120*, 066401.
(37) Scheurer, M. S.; Slager, R.-J. Unsupervised machine learning and band topology. *Phys. Rev. Lett.* **2020**, *124*, 226401.
(38) Zhang, Y.; Ginsparg, P.; Kim, E.-A. Interpreting machine learning of topological quantum phase transitions. *Phys. Rev. Research* **2020**, *2*, 023283.
(39) Acosta, C. M.; Ouyang, R.; Fazzio, A.; Scheffler, M.; Ghiringhelli, L. M.; Carbogno, C. Analysis of topological transitions in two-dimensional materials by compressed sensing. *arXiv*, May 28 2018, ver. 1, 1805.10950. https://arxiv.org/abs/1805.10950 (accessed Aug. 5, 2021).
(40) Claussen, N.; Bernevig, B. A.; Regnault, N. Detection of topological materials with machine learning. *Phys. Rev. B* **2020**, *101*, 245117.
(41) Andrejevic, N.; Andrejevic, J.; Bernevig, B. A.; Regnault, N.; Han, F.; Fabbris, G.; Nguyen, T.; Drucker, N. C.; Rycroft, C. H.; Li, M. Machine-learning spectral indicators of topology. *Adv. Mater.* **2022**, *34*, 2204113.
(42) Cao, G.; Ouyang, R.; Ghiringhelli, L. M.; Scheffler, M.; Liu, H.; Carbogno, C.; Zhang, Z. Artificial intelligence for high-throughput discovery of topological insulators: The example of alloyed tetradymites. *Phys. Rev. Mater.* **2020**, *4*, 034204.
(43) Liu, H.; Cao, G.; Zhou, Z.; Liu, J. Screening potential topological insulators in half-Heusler compounds via compressed-sensing. *J. Phys.: Condens. Matter* **2021**, *33*, 325501.

(44) Schleder, G. R.; Focassio, B.; Fazzio, A. Machine learning for materials discovery: Two-dimensional topological insulators. *Appl. Phys. Rev.* **2021**, *8*, 031409.
(45) Vergniory, M. G.; Wieder, B. J.; Elcoro, L.; Parkin, S. S.; Felser, C.; Bernevig, B. A.; Regnault, N. All topological bands of all nonmagnetic stoichiometric materials. *Science* **2022**, *376*, No. eabg9094.
(46) Perdew, J. P.; Burke, K.; Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* **1996**, *77*, 3865.
(47) Chang, G.; Wieder, B. J.; Schindler, F.; Sanchez, D. S.; Belopolski, I.; Huang, S.-M.; Singh, B.; Wu, D.; Chang, T.-R.; Neupert, T.; et al. Topological quantum properties of chiral crystals. *Nat. Mater.* **2018**, *17*, 978.

https://doi.org/10.1021/acs.nanolett.2c03307
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