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High-throughput discovery of high Curie point two-dimensional ferromagnetic materials

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Databases for two-dimensional materials host numerous ferromagnetic materials without the vital information of Curie temperature since its calculation involves a manually intensive complex process. In this work, we develop a fully automated, hardware-accelerated, dynamic-translation based computer code, which performs first principles-based computations followed by Heisenberg model-based Monte Carlo simulations to estimate the Curie temperature from the crystal structure. We employ this code to conduct a high-throughput scan of 786 materials from a database to discover 26 materials with a Curie point beyond 400 K. For rapid data mining, we further use these results to develop an end-to-end machine learning model with generalized chemical features through an exhaustive search of the model space as well as the hyperparameters. We discover a few more high Curie point materials from different sources using this data-driven model. Such material informatics, which agrees well with recent experiments, is expected to foster practical applications of two-dimensional magnetism.

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INTRODUCTION

The recent experimental demonstration of ferromagnetism in two-dimensional (2D) materials: CrI₃¹ and Cr₂Ge₂Te₆² at low temperatures, has opened a new horizon of nanotechnology research since these materials inherit the potential to revolutionize engineering fields like spintronics³, valleytronics⁴, sensing and memory technologies⁵. In their classical work, Mermin and Wanger⁶ showed that under an isotropic Heisenberg model, long-range magnetic order must be absent in 2D. However, the more recent discovery of even room-temperature ferromagnetism in monolayer VSe₂⁷ and MnSe₂⁸ has been possible as the strong magnetocrystalline anisotropy of these 2D materials lifts the Mermin–Wanger restriction. So far, a plethora of 2D ferromagnetic (FM) materials^{9–14} have been computationally predicted, including a few general-purpose 2D materials databases^{15–17} containing hundreds to thousands of entries. However, none of these databases contain the most crucial parameter for 2DFM materials relevant for practical applications: the transition temperature or Curie point (T_C). This is due to the fact that the computational determination of T_C is a highly complex process, which involves a manual heuristics-based search for the ground-state and low-energy spin configurations. Identification of different magnetic exchanges (direct, super or double) within the neighbouring atoms and mapping them appropriately in a Monte Carlo based spin-flipping simulator has also been a manually intensive exercise. The choice of the model Hamiltonian (Ising instead of Heisenberg) used to simulate the spin-flipping with temperature, also raises a question on the reliability of the Curie temperatures of 2D materials predicted so far in the literature^{10,14}.

Recently, an algorithm¹⁸ has been proposed which can search and predict the collinear, experimentally verified ground and low-energy spin states for bulk materials, almost optimally and exhaustively. Building on this, we develop a code, which performs first principles-based computations followed by Heisenberg model-based Monte Carlo simulations to predict the Curie point accurately from any magnetic 2D material crystal structure.

Software engineering on this code makes it capable to execute such rigorous calculations in a high-throughput manner, even on a workstation-grade computer with GPU (graphical processing unit) acceleration. We use this code to determine the Curie points of materials from a suitable database¹⁶. To our surprise, almost 47% of the 786 materials classified as FM, turned out to be antiferromagnetic (AFM) upon close inspection by our code. The T_C and other magnetic properties could be successfully determined for 157 materials, among which 26 materials reveal beyond 400 K Curie point. Close agreement with experimentally measured T_C for a few materials validates our high-throughput methodology. In pursuit of faster discovery of high- T_C materials, we further develop a machine-learning (ML) pipeline using these 157 data points. Using this ML model, we identify a few high T_C 2DFM materials from the literature and other databases.

The informatics, which optimally balances the rigorosity and efficiency, gives us unprecedented opportunity to compare the magnetic properties of a very large number of materials with diverse structures, which may lead to many new insights on 2D magnetism. For example, we understand why the inclusion of the higher-order neighbours is important for T_C calculation for certain materials and why it is not for the others. We observe several violations of the Goodenough–Kanamori^{19,20} rules for super-exchange, the origin of which is open for further exploration. We also demonstrate that a machine-learning model can capture the complex process of temperature-dependent spin-flipping with exceptional accuracy. Our work thus significantly upgrades the computational materials toolbox to foster practical applications of two-dimensional magnetism.

RESULTS

High-throughput computational framework

We first explain the workflow of our automated code as illustrated in Fig. 1. The unit cell of the material is first fed to a recently developed module¹⁸ of the open-source python library

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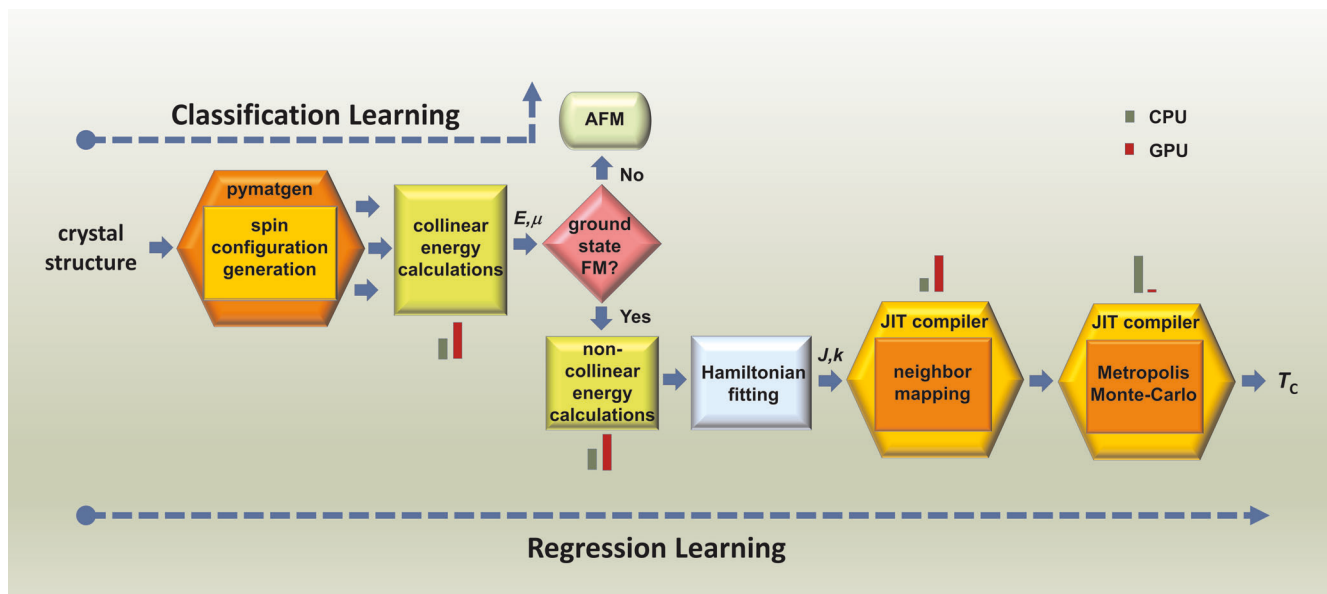


Fig. 1 Automated high-throughput machinery. The different building blocks of our end-to-end code for determining properties of 2DFM materials along with an estimation of CPU and GPU usage of each block.

pymatgen²¹, which generates different FM and AFM spin configurations of the material based on symmetry analysis. Eliminating the heuristics-based approach, pymatgen not only helps to automate the process of the T_C calculation, but also makes it more rigorous and thus reliable [see Methods]. The code-generated spin configurations for experimentally synthesized materials CrI_3 ¹ and $\text{Cr}_2\text{Ge}_2\text{Te}_6$ ² and newly predicted material Cr_3Te_4 ¹⁰ are shown in Supplementary Figs. 1–3 as examples. These structures are then relaxed using collinear density functional theory with Hubbard correction (DFT + U) and their energies are calculated. At this stage, if the ground state is found to be AFM, the material is discarded. Here we also calculate the magnetic moment (μ) of each atom of the structure. Since the magneto-crystalline anisotropic energy (MAE) is essential for the existence of long-range magnetic order in 2D materials, it is calculated using non-collinear DFT including the effects of spin-orbit coupling (SOC) in the next step. These calculations also reveal the easy magnetization axis (EMA) of the 2DFM material, which can be important for specific applications. All these calculations provide us enough information to fit the DFT energy values to the following Heisenberg Hamiltonian:

$$H = -\frac{1}{2} \sum_{ij} J_1 \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{2} \sum_{ijl} J_2 \mathbf{S}_i \cdot \mathbf{S}_l - \frac{1}{2} \sum_{ilm} J_3 \mathbf{S}_i \cdot \mathbf{S}_m - \frac{1}{2} \sum_{in} J_4 \mathbf{S}_i \cdot \mathbf{S}_n - k_x \sum_i (S_i^x)^2 - k_y \sum_i (S_i^y)^2 - k_z \sum_i (S_i^z)^2. \quad (1)$$

Here, J_1 , J_2 , J_3 and J_4 are the nearest-neighbour (N1), 2nd nearest-neighbour (N2), 3rd nearest-neighbour (N3) and 4th nearest-neighbour (N4) exchange coupling constants and \mathbf{S}_i , \mathbf{S}_j , \mathbf{S}_l , \mathbf{S}_m and \mathbf{S}_n are the spins at sites i , j , l , m , and n , respectively. k_x , k_y , and k_z are the magnetic anisotropy constants in the x , y and z directions. S is computed as $\mu/(2\mu_B)$, where μ is the local magnetic moment of the magnetic ions.

For 2D materials, the classical Monte Carlo (MC) based solution of the Heisenberg Model is known to accurately predict the transition temperature²². First, the 2DFM unit cell is multiplied to make a supercell large enough to eliminate size effects, and using a GPU accelerated search, all the neighbours of all the sites are mapped into an 1D array structure. This is then used to perform a Heisenberg model-based classical MC simulation using a semi-compiled dynamic-translation based module, from which the T_C of

the material can be obtained. For a few materials, no in-plane anisotropy is observed which are classified as XY magnets. Mermin–Wagner theorem⁶ prohibits spontaneous symmetry breaking in these kinds of systems where the spin degree of freedom is ≤ 2 . Instead, XY magnets exhibit a Berezinskii–Kosterlitz–Thouless (BKT) transition to a quasi-long-range ordered low-temperature phase. For these materials, T_C is calculated from the following equation obtained from Monte Carlo simulations of the XY model²³:

$$T_C = \frac{0.89}{8k_B} (E_{\text{AFM}} - E_{\text{FM}}), \quad (2)$$

where k_B is the Boltzmann constant and E_{FM} and E_{AFM} are energies of the FM and the most stable AFM solutions normalized by the number of atoms.

Though the Ising model can provide good results for materials with extremely high anisotropy, significant overestimation of T_C may happen for materials with moderate to low anisotropy^{10,11,14}. The anisotropic Heisenberg model takes care of this problem and effectively balances the contributions between exchange and anisotropy. However, it requires further MAE calculations and the MC simulation becomes computationally much more expensive. Although this is a high-throughput study, leveraging software engineering [see Methods], we decide to use the rigorous Heisenberg model without compromising the accuracy.

Database search

We identify the database C2DB¹⁶ as the ideal database to conduct our study based on the following reasons. (1) The authors have performed a preliminary classification between FM and AFM materials and a large number (786) of materials are classified as FM. (2) These materials have been explored by a “systematic combinatorial approach” where almost all known layered exfoliable materials are covered¹⁵, and by substituting the atoms, the authors have predicted a lot of new materials. This kind of variation is ideal to train machine-learning models, which is one of our primary goals. Also, recent synthesis of janus²⁴ and other species substituted²⁵ 2D materials with no bulk analogues have made practical application of this kind of “synthetic” 2D materials possible. (3) The authors have calculated several properties of interest including thermodynamic stability and energy above the convex hull which helps us to estimate the chance of potential

synthesis of these materials. The electronic structures have also been computed, which tells us about the presence of important properties like half-metallicity.

Interestingly, after a close examination by our code, 368 of the 786 materials classified as FM in the database turns out to be actually AFM. The pymatgen magnetism module explores the symmetry allowed spin-configuration space almost exhaustively, and in the process also explores large AFM supercells, which probably the authors of C2DB could not afford to do in their general-purpose study. A few discrepancies can also arise from the difference in DFT settings between the studies. Analyses of a considerable amount of materials have failed and thus have been discarded due to various computational limitations [see Methods]. Sheet 1 of Supplementary Spreadsheet 1 lists all the examined materials with FM/AFM classification as found by our method. In the end, the T_C of 157 2DFM materials could be successfully computed, among which 12 materials are found to be XY magnets. These materials belong to more than 20 different prototype structures, which are shown in Fig. 2.

Discovery of high- T_C materials

Almost all experimentally synthesized 2D materials are there in C2DB, except $\text{Cr}_2\text{Ge}_2\text{Te}_6$, which too we have included manually in this set. We have not included those 2DFM materials whose ferromagnetism cannot be accurately modelled by the Heisenberg or the XY model, such as known itinerant material Fe_3GeTe_2 ^{26,27}. Sheet 1 of Supplementary Spreadsheet 2 lists all the calculated properties, as well as the computed Curie temperatures of these 2DFM materials. To include interactions of N neighbours in the Hamiltonian, $N + 1$ spin configurations are required, and the number of configurations generated by pymatgen is also listed in Supplementary Spreadsheet 2. Given enough configurations, we have included up to the 4th nearest-neighbour interaction in this work. The T_C s are calculated using two methodologies: (1) commonly used nearest-neighbour approach: including only the N_1 interaction and fitting the energies of the FM and the most stable AFM configurations, (denoted as 'TC' in the spreadsheet) and (2) multi-neighbour approach: interactions including up to N_4 (listed under the 'TC_exact' column). Apparently, the TC_exact

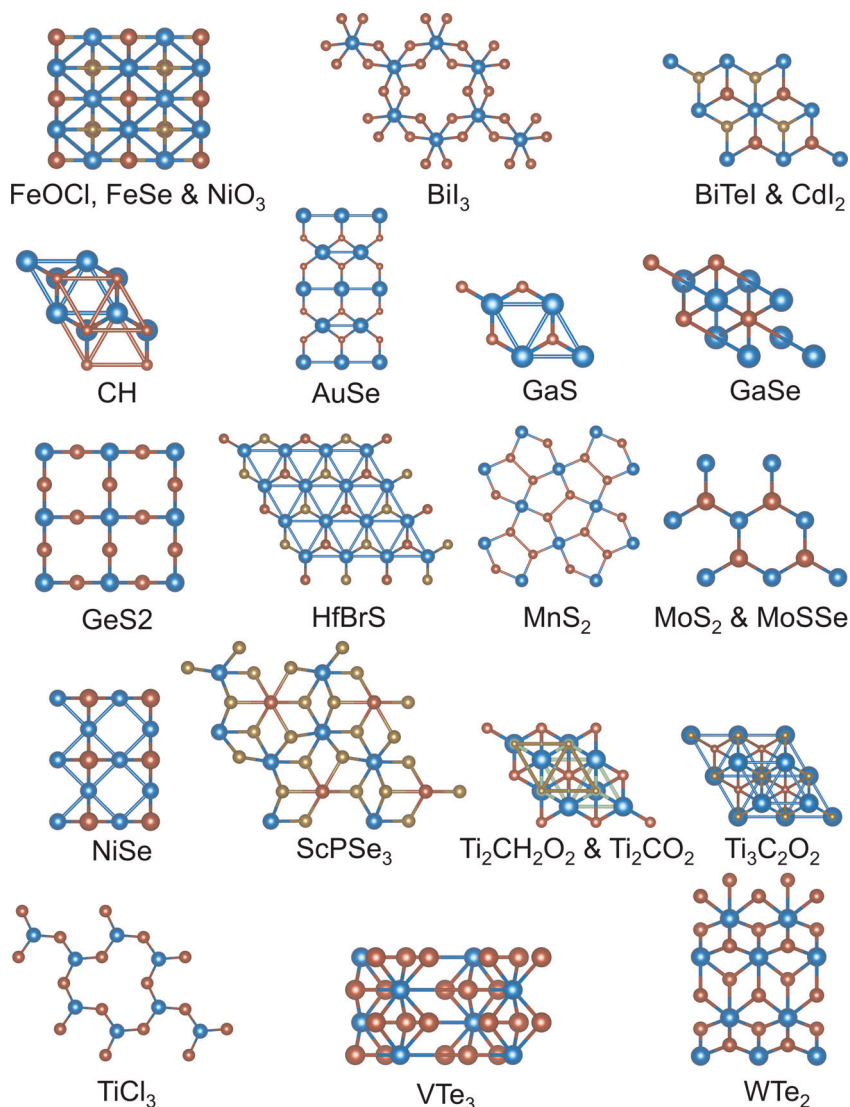


Fig. 2 Prototypes of training data. Top view of prototypes (according to C2DB) of all the 2DFM materials for which T_C could be determined using our method. Blue balls represent the magnetic metal ions and the red and brown balls signify non-magnetic ions. Note that the grouped prototypes, e.g. FeOCl, FeSe, and NiO₃ show the same structure cation-wise, but have different numbers of anions. However, all prototypes in such a group look similar from the top.

should provide a better estimate of the real T_C of the material. However, to our surprise, we observe that TC, which is computationally much economic, is close to TC_exact in most of the cases, except a few as described below. (1) Materials with more than one distinct metal layer (prototypes CH, GaSe, CdI₂-MXene, Ti₂CO₂, and Ti₂CH₂O₂). For prototypes CH and GaSe, there is no layer containing anions between the metal layers, which gives rise to strong inter-layer direct exchange in addition to the intra-layer superexchange. In prototypes CdI₂, Ti₂CO₂ and Ti₂CH₂O₂, which are all MXenes, the distinct metal layers are connected by anionic layers, where both the inter-layer as well as intra-layer exchange interactions play a pivotal role in deciding the T_C . Clearly, considering only the N1 interactions in these materials is not accurate enough, as reflected in the huge differences between TC and TC_exact. In passing, we note that, for a few MXenes, pymatgen could only generate two configurations, thus, the TC value has been repeated as TC_exact. (2) Materials with square or rectangular lattice (prototype FeOCl, FeSe, GeS₂ and NiSe). Here, the N1s or N2s are the atoms situated in the diagonally opposite corners of the square or rectangle, where superexchange is expected to be feeble at best and only strong direct exchange could persist. The difference in distance between N1s and N2s are also very small in these materials, which again makes the inclusion of the higher neighbour interactions necessary [see Supplementary Figs. 4 and 5]. In a few materials (Janus) the effect of higher-order neighbours is not possible to take into account due to moderate distortion in the lattice and T_C value has been repeated as TC_exact.

The calculated T_C (TC_exact) by our automated code for the experimentally synthesized materials CrI₃¹, Cr₂Ge₂Te₆², and MnSe₂⁸ matches the experimental reports very closely without any manual tinkering of parameters, validating the generalization and accuracy of our method. For T-phase VSe₂ our predicted T_C is only 114.33 K, whereas room temperature ferromagnetism has been reported⁷. However, it must be noted that the authors have reported strong substrate dependence of the magnetism and T_C in this study which explains this apparent discrepancy. Also, our code confirms the magnetism to be in-plane in this material which matches the experimental report. With Supplementary Table 1, Supplementary Note 1 and Supplementary Figs 1, 2, and 6, we explain in detail how our code works for 2 FM (CrI₃¹, CrGeTe₃²) and 2 AFM (FePS₃²⁸, NiPS₃²⁹) experimentally synthesized 2D materials.

We finally discover a total of 26 materials with $T_C > 400$ K and 32 materials with $T_C \geq 300$ K, making these materials suitable for practical device applications. Interestingly, many of these materials are known to show a “low” amount of magnetism in bulk forms, such as materials containing Rh, Ru, Mo, W, Sc, Ti, and Zr, which were ignored in previous heuristics-based searches¹⁰. However, our study suggests that the materials containing the above-mentioned metals can indeed show a “decent” (0.59–3.96 μ_B /atom) magnetic moment in 2D crystal form along with high- T_C , possibly because of the enhanced electron localization. Also, for some of these materials the anisotropy is not great, but the difference in energy between the FM and AFM states, which ultimately translates to exchange parameters, helps to lift the T_C beyond the room-temperature.

Since the magnetic properties of these materials can greatly depend on the value of U (Hubbard Correction), we also calculate the T_C of the 26 promising high- T_C materials with much more accurate material-specific U values³⁰ [see Methods] and present the results in Sheet 3 of Supplementary Spreadsheet 2. Apart from a single material (MoIn₂Pmmn), we observe that the T_C of the rest of the materials remains either close to 400 K or becomes much higher than that. We also noticed in some cases the T_C value has been significantly enhanced with the application of these tailor-made U values. Thus, we expect that some of the materials whose T_C values fall in the 250–400 K range might exhibit much higher T_C if it is calculated using material-specific U .

Machine-learning model

Due to an increasing amount of available data, machine-learning has recently found many applications in the field of solid-state materials science³¹. Very recently, training on about 2500 experimentally reported Curie temperature of bulk materials, accurate ML models to predict T_C of bulk materials have been developed³². 2D magnetism is fundamentally different from the magnetism of the bulk materials as most of the time anisotropy does not play such a significant role there. Therefore, in this emerging field, one doesn't have the luxury of a sufficient amount of data points to train on. Based on the 157 data points obtained from our database-search we develop a machine-learning model to predict the T_C from the crystal structures. To decide the best model and features, we use the autoML library automatminer (<https://github.com/hackingmaterials/automatminer>). This tool takes structures as input and decorates the dataset with easily computable and chemically and physically meaningful features³³. Then the dataset is cleaned and reduced and is sent to the autoML library TPOT³⁴, which stochastically searches the model and the hyperparameter space using a genetic algorithm and finds the best model for the given dataset. After this extensive search [see Methods], we find an excellently fitted pipeline with average cross-validation (CV) score 94.57 K², which is reported as the mean square error (MSE) on the training set. For the FM/AFM classification problem, we also try to find a suitable pipeline using the same method, with our examined 525 FM + AFM data points. The fitted pipeline reports an average CV score of a lowly 72.89% (accuracy) on the training set, which is understandable considering the complexity of the problem and the size and skewness of the data.

To test the generalization and predictive power of these ML pipelines, we construct a test set from reported 2DFM materials. Also, quite a few selected materials are included in the test set from a separate database¹⁷ with completely new structures and complex compositions. After inspecting these materials using our code, we find a few materials to be AFM which have been claimed as FM. This discrepancy can originate from the use of different DFT settings. This, along with fitting with a large number of spin configurations also causes a difference in fitted J values as observed for few materials (see Supplementary Fig. 3). Details of 22 materials identified as FM are provided in Sheet 2 of Supplementary Spreadsheet 2. The new prototypes encountered in this set are illustrated in Fig. 3.

Figure 4a illustrates the accuracy of the ML pipeline prediction against the DFT-MC calculated T_C for all the train and test data. The ML predicted T_C is also listed in the ‘TC_exact predicted’ column of Supplementary Spreadsheet 2. The distribution of absolute errors in train and test data have been plotted in Fig. 4b. Although the pipeline has fitted to train data with high accuracy, generalization to test data does not seem so well. The MSE value also turns out to be 30335.46 K². Although the small train data size could be partially responsible for this, the main reason is possibly the introduction of unseen crystal structures. For instance, although we have not trained with even a single material containing La, the prediction for LaBr₂ is exceptionally close probably because we had a lot of crystals with similar structures in our train data. The same argument can be applied to Mn₂H₂NO₂ and Cr₃Te₄. The ML classifier pipeline has also been tried on the test data containing a total of 123 FM + AFM samples, which yields a decent 73.17% accuracy. Sheet 2 of Supplementary Spreadsheet 1 tabulates all the materials tried and their ultimate fate as well as the classification prediction.

During this exercise, we identify CrO₂_P4/mmm and ZnNi₂O₅ as high- T_C materials, while the claim of Cr₃Te₄ possessing high- T_C has also been verified, albeit the Curie temperature turns out to be much lower than the reported Ising model predicted value¹⁰, but higher than the experimentally reported value for bulk Cr₃Te₄.

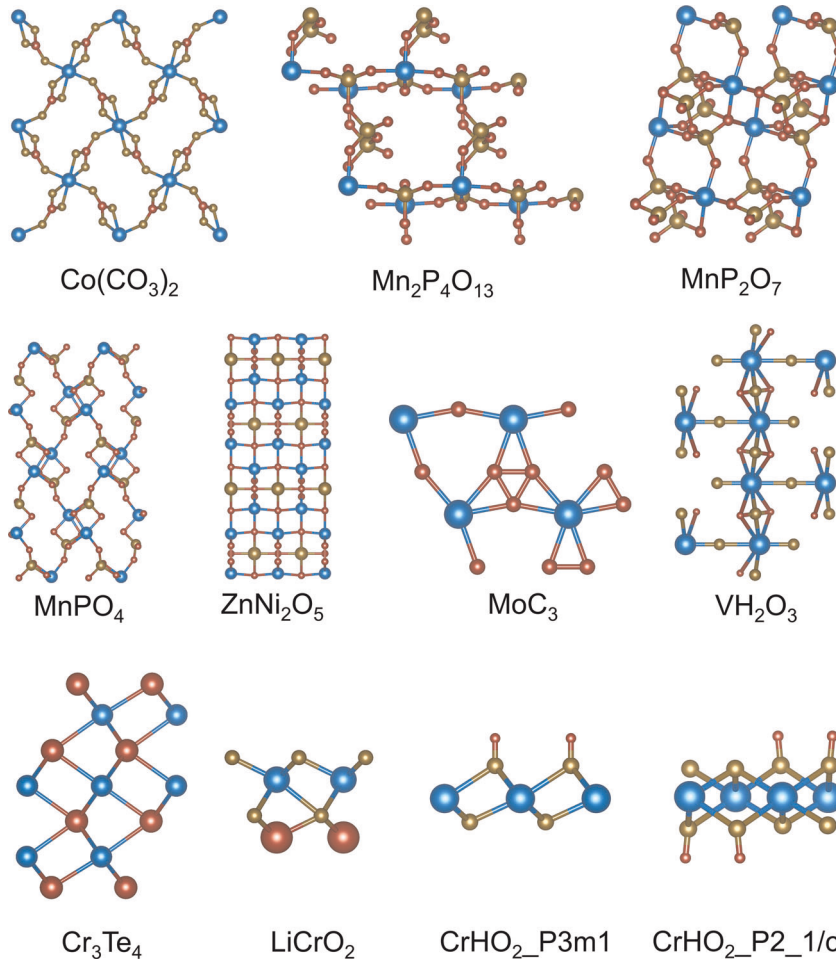


Fig. 3 Prototypes of unique test data. The unique prototypes of the test data which have not been covered in Fig. 2. Note that for visualization purposes, top view of the materials in the 1st and 2nd row has been shown, but the third row contains all side-views. The same colour convention as Fig. 2 have been used.

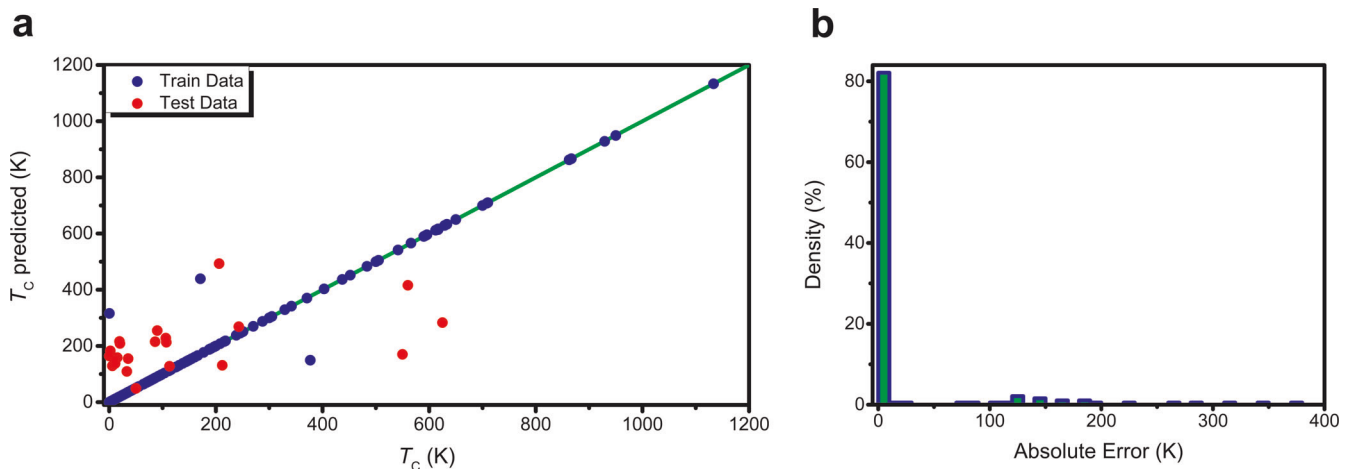


Fig. 4 Accuracy of the machine learning model. **a** plot of DFT-MC calculated T_C and machine learning predicted T_C for all the train as well as test samples. The green line represents a perfectly accurate prediction line. **b** distribution (histogram plot) of the absolute error of the machine learning prediction for test and train data combined.

Violation of Goodenough-Kanamori rule

The classical Goodenough-Kanamori semi-empirical rules^{19,20} for magnetic materials essentially states that when the magnetic-ion-

anion-magnetic-ion bond angle is close to 180° , a strong AFM structure due to superexchange should prevail, whereas if this angle is close to 90° , the material should show ferromagnetism.

However, in this work, we notice quite a few violations of this rule. We have tabulated these bond angles for all the train and test materials in Supplementary Spreadsheet 2 and the apparent violation cases are marked in red. The most significant violation of the Goodenough-Kanamori criterion can be seen in the strong high- T_C FM material CrO_2 _P4/mmm, where the cation–anion–cation bond angle is precisely 180° . Although a full investigation is out of the scope of this work, it appears that these postulates were developed for bulk materials and are failing here because of highly covalent bonds of 2DFM materials. Especially CrO_2 _P4/mmm exhibits a highly planar structure, and thus should manifest highly covalent bonds. However, this material seems to be dynamically unstable and possesses a much higher total energy compared to the experimentally available bulk phase which might make it unlikely to be experimentally synthesized⁹.

High-temperature structural stability

According to the thermodynamic stability classification in C2DB, we choose three highly stable high- T_C materials, namely CrIn _Pmmm, RhCl_2 _C2/m, and $\text{Mn}_2\text{H}_2\text{CO}_2$ _P-3m1 along with newfound ZnNi_2O_5 _Pmmn for further structural stability evaluation at high temperatures. An ab-initio molecular dynamics (AIMD) run at 400 K for a total of 6.5 ps is conducted for this. Although CrIn , RhCl_2 , and ZnNi_2O_5 retain their crystal structure during this, albeit with less crystallinity, the MXene $\text{Mn}_2\text{H}_2\text{CO}_2$ starts to melt away just after 3 ps, rendering it unsuitable for practical applications. Supplementary Figs 7–10 show the structural differences resulted from the MD runs.

DISCUSSION

In this study, we predict a total of 26 2D materials to have T_C beyond 400 K. Many of these could be easily synthesizable, either by straight exfoliation from their bulk counterparts or by bottom-up chemical methods^{24,25}. It is worth noting that, low thermodynamic stability does not necessarily mean the material would be unusable for practical applications. For instance, according to C2DB, Silicene is classified as a material with low thermodynamic stability. However, Silicene transistors have been demonstrated to work in room temperature³⁵. Some of these materials screened by us even show dynamic instability. Again, many commonly used 2D materials, such as T-phase MoS_2 showing dynamic instability³⁶ in free-standing form stabilizes themselves on substrates through possible substrate interaction and even finds application in room-temperature devices³⁷. Also, these materials can show charge density wave (CDW) characteristics and stabilize in a larger supercell with slight structural distortion⁷. However, CDW distortions can sometimes impact the magnetic order adversely³⁸.

To summarize, using high-throughput automated codes and data-driven models, we thoroughly screen 2D materials databases and predict a host of 2DFM materials with high Curie point. With the emergence of novel synthesis techniques, these materials could indeed be of interest to experimentalists and engineers in terms of practical application in various devices. The ML model and the automated code developed in this work could find use in the community for rapid magnetic property prediction. The model complements the rigorous DFT-MC based code and if trained with sufficiently large datasets the model could eventually replace the code³². State-of-the-art software engineering enables us to achieve an optimal balance between rigor and computational efficiency, which is very important for reliable high-throughput material screening. As a result, we discover many important magnetic materials involving metals like Mo, W and Ti which have so far been ignored by heuristic-based formula-screening¹⁰.

METHODS

Spin configurations generation

As mentioned before, the python library `pymatgen`²¹ has been extensively used in this study for generating spin configurations, managing, and parsing input-output files and performing the neighbour mappings. The python module `ASE`³⁹ has also been used to parse the input structure files. To ensure the reliability and coverage of the `pymatgen` generated spin configurations, we manually verify that the code-generated spin configuration set almost always includes all heuristics-based configurations reported in the literature^{9–14}. Often, the code generates even more unexplored but symmetrically valid configurations that we leverage to include a higher number of neighbour interactions for a more accurate prediction of the T_C . At the same time, we ignore ferrimagnetic configurations since these are usually energetically highly unstable as well as asymmetric. For instance, in contrast to the previous report¹⁰, our T_C prediction of Cr_3Te_4 is based on a larger number of symmetric FM and AFM configurations and thus seems to be much more accurate.

It is worth mentioning that since we cover such a huge variety of materials using an automated workflow, we use such values of various DFT and numerical parameters [see Supplementary Readme (readme file at OSF repository)], which would yield reasonable results for all materials. For instance, in case of CrI_3 , the default value of the parameter `enum_prec = 0.001` (`enum_precision_parameter` in `pymatgen`) generates 3 configurations, where the Néel AFM configuration gets excluded (Supplementary Fig 1). But with `enum_prec = 1e-7`, all 4 configurations can be generated. For the first case, we obtain $J_1 = 2.78$ and $J_2 = 0.43$ meV/link, while the second set gives us almost identical values of $J_1 = 2.82$, $J_2 = 0.41$ and $J_3 = 0.009$ meV/link and in both cases, we obtain the same T_C . The Néel AFM solution turns out to be the most energetically unfavourable state and gets truncated¹⁸ by `pymatgen-enumlib` with default settings. For this high-throughput study, we find the default values to be accurate enough for our *one-fits-all* scheme. However, for focused studies on some specific material one might want to obtain extremely accurate results and thus might need to tune the parameters a little.

DFT parameters

Because the energy differences between various configurations for magnetism calculations could be as low as $\sim\mu\text{eV}$, the computations need to be performed with high accuracy. We use heavily modified versions of predefined configurations ‘MPRelaxSet’, ‘MPStaticSet’ and ‘MPSOCSets’ available in `pymatgen` for relaxations, static runs and MAE calculations. These modifications, as well as other details of DFT, are highlighted below.

Spin-polarized DFT calculations are carried out using generalized gradient approximation (GGA) as implemented in the code `VASP`⁴⁰ with projector augmented-wave (PAW)⁴¹ method using the Perdew–Burke–Ernzenhof (PBE)⁴² exchange–correlation functional. Along with the CPU version, The GPU port⁴³ of `VASP` has been used extensively. For all calculations, a correction on the strongly correlated d-shell electrons (GGA + U) is applied using the Dudarev⁴⁴ formulation. The default value of the cut-off energy (520 eV) is used which proved to be sufficiently large. For relaxations, the default reciprocal density of 64 \AA^{-3} is employed whereas for all collinear and non-collinear static runs a much denser reciprocal density of 300 \AA^{-3} has been used. Electronic convergence is set to be attained when the difference in energy of successive electronic steps becomes less than 10^{-6} eV, whereas the structural geometry is optimized until the maximum Hellmann–Feynman force on every atom falls below 0.01 eV/\AA . For the high-precision MAE calculations, a stricter electronic convergence criterion of 10^{-8} eV is imposed. A large vacuum space of $>25 \text{ \AA}$ in the direction of \mathbf{c} is applied to avoid any spurious interaction between periodically repeated layers. The Bader charge and magnetization analysis are performed using the code developed by the Henkelman group⁴⁵, where charge densities generated from DFT static runs are used as inputs. These Bader partitioned magnetic moments have been used as the local magnetic moments of the magnetic elements. All crystal structure images are generated using the tool `VESTA`⁴⁶.

For metals, Co, Cr, Fe, Mn, Mo, Ni, V and W, the effective U values have been taken from the Materials Project (https://wiki.materialsproject.org/GGA%2BU_calculations#Calibration_of_U_values) where these effective U values have been calibrated by performing a fitting to experimental binary formation enthalpies⁴⁷. This is an established practice and has been used in similar high-throughput screening studies before¹⁰. We also find that the application of an effective U is essential to perform accurate DFT calculations on materials with “low” magnetization (materials containing Nb, Sc, Ru, Rh, Pd, Cu, Os, Ti, Zr, Re, Hf, Pt, and La) as the AFM solutions

become difficult to obtain for these materials without a proper effective U . In these cases, for a specific metal, first the effective U values are obtained using the linear-response approach³⁰ using 3×3 supercells for a few materials containing the element and then the average value of these effective U is taken as the final effective U . A complete list of effective U values (in eV) used in this study, as well as the DFT parameters imposed can be found in Supplementary Code 1 (e2e.py at OSF repository).

It is worth noting that, these effective U values depend on the element type, charge state and coordination mode of magnetic species in a certain material, which implies that the value of U is quite material-specific and should be determined carefully for accurate predictions. Thus, we also calculate the material-specific U values for the most promising 26 materials with predicted high- T_C using the linear-response approach³⁰. We observe that the U values obtained from the linear-response method can be quite different from the high-throughput U values we have used so far (see Sheet 3 of Supplementary Spreadsheet 2). However, the computational budget of the linear-response method is excessively high to be adopted for high-throughput material screening.

Hamiltonian fitting

The coupling constant (J) values are fitted using the collinear energy values of different FM and AFM spin configurations. However, for a lot of cases, when the energies of all configurations are taken, the determinant of the system of equations becomes zero which makes the set of equations unsolvable. As an automated remedy for these problems, again a fitting is tried omitting the most unstable AFM configuration and using one less neighbour than before. This process is repeated until a set of physically meaningful solutions is found, or the code runs out of configurations to fit. The anisotropic constant (k) values are fitted using the non-collinear energy values with spins oriented in different directions ([100], [010], [110], [001]).

Despite our best efforts, calculations for 261 materials (out of the 786 materials classified as FM in C2DB) had to be cancelled because of the following reasons: (1) pymatgen could not recognize the symmetry and could generate only one configuration, (2) material turned out to be non-magnetic after DFT calculations, (3) severe convergence issues occurred during DFT calculations, (4) AFM configurations could not be retained even after application of proper U and manual tuning of parameters, (5) the Hamiltonian could not be fitted properly, (6) phase change of crystal structure after relaxation.

Monte Carlo simulation

To study ferromagnetic (FM) to paramagnetic (PM) transition in these monolayer materials, Monte Carlo (MC) simulations of the Heisenberg model have been performed using the Metropolis algorithm with single-spin update scheme⁴⁸. To eliminate the size effects, a 50×50 supercell containing 2500–8000 sites has been used to simulate the system. Total 10^5 Monte Carlo steps have been performed for each temperature, while the results from the first 10^4 steps have been discarded, as the system is allowed to equilibrate (thermalize) during this time. The final values of magnetization and susceptibility are calculated as the average over the last 9×10^4 MC steps for each temperature.

Software engineering

We develop the complete end-to-end code in python to take advantage of pymatgen. However, python being an interpreted language, the MC simulations turned out to be excessively slow, especially with high coordination numbers and inclusion of higher neighbours, which made the code unsuitable for the high-throughput study. As a remedy to this problem, we decide to use python-based just-in-time (JIT) compiler numba⁴⁹ which compiles specific decorated python modules at the first encounter to low-level instructions, and when these modules are repeatedly called, the compiled version is used which makes the code extremely fast. However, the trade-off is, a lot of powerful functions and the coding flexibility offered by python (like heterogeneous data structures, appending to a list) cannot be successfully compiled and significant software engineering, as well as timing and cost-benefit analyses, are required to achieve an optimal code. GPU acceleration for neighbour-mapping of large lattices (2500–8000 sites) has also been implemented, which on a CPU must be done serially and takes a lot more time.

The engineered code was optimized to such an extent that the whole study could be performed using even a workstation-grade machine, albeit with GPU acceleration. A video of real-time execution of the code, where the T_C of four materials (CrI_3 , $\text{Cr}_2\text{Ge}_2\text{Te}_6$, MnSe_2 , and MoC_3) are being

calculated in parallel in a single-CPU (18 cores), three-GPU enabled workstation within ≈ 10 hour, can be found at <https://youtu.be/HJKR-03OzBl>. At the same time excellent scalability is observed, when the code is executed on a high performance computing node (<https://youtu.be/GQaFm29LR4>).

Machine learning

The python libraries automatminer and matminer³³ have been used to featurize the datasets and search for optimal ML pipeline for the FM/AFM classification problem as well as the T_C predicting regression problem. Dataset cleaning and feature reduction are handled by automatminer. Then, various pre-processing algorithms along with a host of commonly used ML models employed for materials science problems³¹ for small to moderate datasets have been searched by the autoML library TPOT³⁴, such as naive Bayesian, decision tree, extra trees, random forest, gradient boosting, k-neighbors, linear SVC and logistic regression for classification and elastic net CV, decision tree, extra trees, random forest, gradient boosting, k-neighbors, lasso lars CV, ridge CV and linear SVR for regression. Also, the hyperparameters are tuned at the same time. A full list of pre-processing algorithms, ML models and hyperparameters searched can be found at https://github.com/hackingmaterials/automatminer/blob/master/automatminer/automl/config/tpot_configs.py. It is worth noting that TPOT uses the ML library scikit-learn⁵⁰ for the ML as well as the feature-engineering models. For each autoML search, more than 60,000 pipelines have been explored.

The generic python codes using scikit-learn for the best pipelines for both FM/AFM classification and T_C regression have been given in Supplementary Codes 2 and 3 (TPOT_*.py at OSF repository). For the former, a combination of SelectPercentile, MaxAbsScaler, and ExtraTreeClassifier turns out to be the best pipeline, whereas for the latter it is a combination of SelectPercentile, ZeroCount, and GradientBoostingRegressor. The list of features used for these problems can be found in Supplementary Logs 1 and 2 (*_digest at OSF repository). Moreover, the pickled pipelines have also been provided in Supplementary ML Pipes 1 and 2 (*_pipe at OSF repository) which can be loaded into automatminer to make predictions on any dataset. The Supplementary Readme file provides detailed instructions on how to use the Supplementary codes and pipes.

AIMD simulations

For the chosen materials, to minimize the temperature oscillations a large supercell containing ≥ 144 atoms has been constructed to run the AIMD on. Because of severe convergence issues, non-spin-polarized DFT calculations are performed with Gamma-point only sampling which can be sufficient to determine structural stability. A canonical ensemble (NVT) is used and a Nosé-Hoover thermostat^{51,52} at 400 K is employed. The simulations run for 6.5 ps with a 2 fs time step. The crystal structures of the tested materials after the simulation can be seen in Supplementary Figs 7–10.

DATA AVAILABILITY

The authors declare that the main data supporting the findings of this study are available within the paper and its Supplementary files, the OSF repository (<https://osf.io/6ebjp/>) and other open online resources. Other relevant data are available from the corresponding author upon reasonable request.

CODE AVAILABILITY

All relevant python codes and pickled ML pipelines are provided in the OSF Repository (<https://osf.io/6ebjp/>). Also, the end-to-end code for Curie point determination (e2e.py) is available at GitHub (<https://github.com/NSDRILIS/e2e>), which we plan to update periodically.

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AUTHOR CONTRIBUTIONS

A.K. developed the automated end-to-end code, implemented it on CPU and GPU based architectures, developed the ML models and analysed the data. M.K. scanned the materials using the automated code, collected and verified the results. S.M. conceived the problem statement and overall supervised the work. All authors contributed to the writing.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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