Editors' Suggestion

Statistics on magnetic properties of Co compounds: A database-driven method for discovering Co-based ferromagnets

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The search for new ferromagnetic compounds is often targeted at known structure families, particularly those containing iron, cobalt, and manganese. We propose a method to expand this search to lesser-known structure types, using a database of experimental Curie and Néel temperatures. This study, in particular, illustrates the case of compounds containing the element cobalt, and we demonstrate how the use of such a database can lead to the discovery of ferromagnetic materials that had previously been overlooked. We report statistics from a literature survey of the magnetic properties of Co-based compounds with more than 33 at. % Co. We classify more than 13 000 compounds by structure type, cobalt content, and magnetic ground state. From these data, compounds $TaCo_2Ga$, $La_6Co_{13}Bi$, and Nd_2Co_3 were identified as potential ferromagnets, and we confirm their ferromagnetic ordering theoretically via first-principles calculations and experimentally via synthesis and characterization measurements. In addition, the analysis is focused on the collection of data trends and discovery of ferromagnetic materials with easy-axis magnetic anisotropy. Both known ferromagnetic materials with unknown magnetic anisotropy, and unstudied compounds were considered. From the subset of known ferromagnets with unknown anisotropy, the compound Co_2Mg was synthesized, characterized, and determined to have easy-axis magnetic anisotropy at room temperature.

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I. INTRODUCTION

Ferromagnetic materials enable a wide range of technologies with diverse applications ranging from energy production to computer hardware, memory storage, and transportation. As such, intense efforts are pursued worldwide to improve the magnetic properties of known materials, by, for example, reducing their economic or environmental cost, or to find new magnetic materials altogether. The search for entirely new families of ferromagnetic compounds is challenging, and, as a result, efforts are often focused on already known families that can be further improved. A technological breakthrough often requires the discovery of a new material with unprecedented magnetic properties, as illustrated by the discovery of rare-earth magnets in the 1980s [1–3]. Here, we propose a database-driven approach to aid in the discovery of new ferromagnetic materials.

To guide our search for novel ferromagnetic materials, we decided to perform a systematic survey of the already known compounds. A magnetic database of this type allows for a wide range of discovery aids: Not only could it be used to isolate uncharacterized compounds that were partially overlooked by the research community, but also it could constitute a great research aid in the rehabilitation of existing ferromagnets with too low Curie temperatures or undesired magnetic anisotropies or the improvement of material cost of known strong ferromagnets. As such, it often comes as a surprise to learn that there does not yet exist any comprehensive database of magnetic properties of materials. For experimental data, there are a few resources available, for example, the "Database of Magnetic Materials and Tabulation of Magnetic Transition Temperatures" published in 1972 by Connolly and Copenhaver [4] or the AtomWork database [5]. Such resources, however, only contain information about a subset of materials, such as binary compounds, and are losing relevance due to their age, such as in the case of Connolly and Copenhaver's tabulation. A more recent and ambitious effort is the collection of magnetic structures MAGNDATA [6,7]. Despite currently having 1784 entries, including 66 records for Co-containing ferromagnetic compounds, it still represents a very small subset of the existing literature data. For theoretical data, there are more resources available. There are several projects aiming to calculate as many different quantities as is computationally feasible from the available crystallographic database such as the Inorganic Crystal Structure Database (ICSD) [8] and

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TABLE I. Breakdown of cobalt database data.

Category	Number of compounds
Total compounds	13 796
Stoichiometric compounds	5098
Compounds with known space group	4639
Compounds with reported Curie temperature	1341
Compounds containing magnetic anisotropy data	535
Easy-axis anisotropy	225

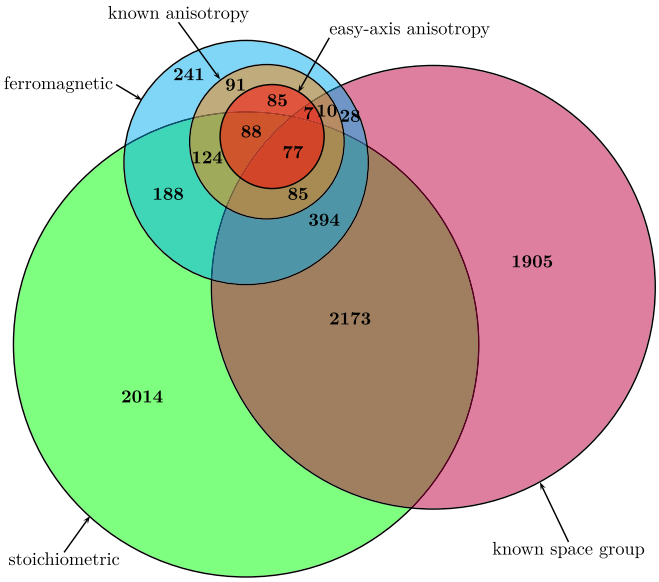

FIG. 1. A visual breakdown of the overlapping categories in the database of Co compounds. Circles are not to scale.

Pearson's Crystal Data [9]. The Materials Project aims to calculate the properties of all known inorganic materials and make these data publicly available to the materials community to accelerate innovation in materials research [10,11]. AFLOW is generating a library of band structure calculations of all the ICSD compounds [12,13]. Other libraries from high-throughput calculations include the Computational Materials Repository (CMR) [14], CATAPP [15], and the Clean Energy Project for organic molecules [16]. Open-source PYTHON libraries for materials analysis such as PYTHON material genomics (PYMATGEN) [17] provide computational tools to hypothesize materials properties from calculated data.

In this study, we gathered all compounds in the Pearson structure database containing the element cobalt, chosen for the magnetism in its pure elemental state, and performed a systematic literature survey of each of the compounds to collect their Curie or Néel temperatures, as well as other magnetic properties. We analyze the relationships between Curie and/or Néel temperatures, cobalt content, structure types, and magnetic anisotropy. By recognizing trends, we can identify compounds that were previously discarded or overlooked. For classification purposes, we included ferrimagnetic compounds as ferromagnetic, as they have a finite moment and can sometimes satisfy the same technological applications as ferromagnetic materials. From our survey, we isolate several promising candidate compounds likely to be ferromagnetic, and we present experimental and theoretical confirmation for several such compounds: $TaCo_2Ga$ (with $T_C = 300$ K), $La_6Co_{13}Bi$ (with $T_C = 470$ K), and Nd_2Co_3 (with ferrimagnetic transitions at 144 and 158 K). We also show that data on magnetic anisotropy can help us identify compounds with specific anisotropy. For example, we report that the previously known ferromagnet $MgCo_2$ has easy-axis magnetic anisotropy.

II. METHODS

A. Database

The breakdown of our data is shown in Table I and, more visually, in Fig. 1. A majority of our total pool of Co-containing compounds (13 796 compounds) originates from Pearson's Crystal Data [9], to which we added a few from our own literature survey. From the associated crystallographic information files (cifs), we extract the structure types for each compound, as well as the necessary information for electronic structure calculations. The magnetic data for some compounds could be easily added initially from existing databases, such as AtomWork and the database of Connolly and Copenhaver [4], or from our own existing research. For the remainder, we performed a systematic literature search of nearly every compound on the list down to 33 at. % Co. During the course of the search, thousands of articles were read, and magnetic orderings, transition temperatures, and anisotropies were identified from article abstracts, bodies, figures, and tables. To identify relevant articles, we predominantly used CAS SciFinder and Clarivate Analytics Web of Science. Special attention was given to compounds with no reported magnetic data to be reasonably certain that these compounds are indeed poorly studied. Once the appropriate information had been gathered for compounds down to 33 at. % Co, PYTHON scripts were used to analyze and sort the data (see Supplemental Material [18]).

Among the 13 796 compounds, less than half are stoichiometric (5098) or have a known space group (4639), as quite a few of the compounds were synthesized as part of doping studies, and many do not have a complete cif. In total, we found 1341 ferromagnetic compounds with a reported Curie temperature (experimental value). We found that 535 ferromagnets have a known magnetic anisotropy, among which 225 had the easy-axis anisotropy necessary for certain types of applications such as those which require permanent magnet materials.

B. Synthesis and characterizations

Polycrystalline samples of $TaCo_2Ga$, $La_6Co_{13}Bi$, and Nd_2Co_3 were synthesized using an arc furnace, followed by annealing. Polycrystalline samples of $MgCo_2$ were synthesized using a solid state reaction in a resistive box furnace. Specifics of each synthesis are given in Appendixes C–F. Following synthesis, the crystal structure was confirmed by powder x-ray diffraction (Rigaku MiniFlex600 diffractometer). Magnetic properties were measured with a Magnetic Property Measurement System (MPMS, Quantum Design) using procedures outlined for each compound in Appendixes C–F.

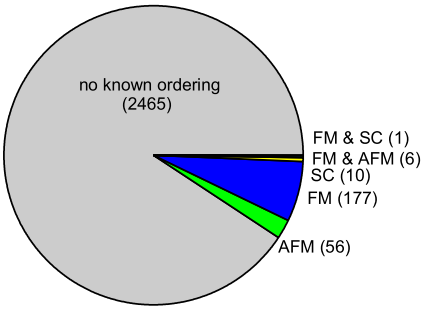

FIG. 2. Magnetic properties of 2715 stoichiometric Co compounds: ferromagnetic (FM), antiferromagnetic (AFM), and superconducting (SC). Compounds containing other magnetic elements such as Cr, Mn, Fe, Ni, Ce-Yb, and Th-Cm are excluded. Some compounds can have multiple reported transition temperatures and ordering types (both FM and AFM or both FM and SC).

C. First-principles calculations

First-principles calculations of magnetic properties were conducted for $TaCo_2Ga$, $La_6Co_{13}Bi$, and Co_2Mg using the plane-wave density functional theory code WIEN2K [19] within the generalized gradient approximation (GGA) of Perdew *et al.* [20], employing the linearized augmented plane wave (LAPW) approach. For all compounds, an RK_{max} of 8.0 was employed; this RK_{max} is the product of the largest plane-wave expansion wave vector and the smallest LAPW sphere. With the exception of the magnetic anisotropy calculations for Co_2Mg, spin-orbit coupling was not included. LAPW sphere radii for the respective compounds were as follows (in units of the Bohr radius a_0, where $a_0 = 0.529\,177$ Å). $TaCo_2Ga$: Ga, 2.35; Co and Ta, 2.41. $La_6Co_{13}Bi$: La, 2.50; Bi, 2.50; Co, 2.26. Co_2Mg: Co, 2.15; Mg, 2.37. Experimental room-temperature lattice parameters were used for all calculations. For the magnetic anisotropy calculations in Co_2Mg, 5000 k points in the full Brillouin zone were used, whereas for the first two compounds 800 and 300 k points, respectively, were used.

III. RESULTS AND DISCUSSION

We present three examples of how the database combined with electronic structure calculations can guide the discovery of materials. In Sec. III A, we show how to identify ferromagnets from Co content. This can also be used to identify compounds with anomalous magnetism. In Sec. III B, we show how the addition of structure type can help identify high-Curie-temperature ferromagnets. This addition can also be used to identify compounds close to magnetic instabilities. In Sec. III C, we show how the identification can be further narrowed to compounds with easy-axis magnetic anisotropy.

A. Identifying ferromagnets from Co content

A distribution of Co-based compounds in our database is shown in Fig. 2, tallying the ferromagnetic, antiferromagnetic, paramagnetic, combination, and unknown compounds within a subset of the stoichiometric Co compounds excluding the elements Cr, Mn, Fe, Ni, Ce-Yb, and Th-Cm. This exclusion allows us to gain an overview of Co magnetism without having to take into account other magnetic elements. Superconductivity, though not a magnetic ordering, was also noted if present. The overwhelming majority of compounds have no known magnetic ordering. While sometimes this is due to having not been researched for this analysis, more commonly these compounds have not yet been characterized by the scientific community at large, leaving plenty of opportunities for investigation. Within the studied compounds, there is a clear favoritism towards ferromagnetic ordering. This tendency is unsurprising, as Co itself is a ferromagnetic element [21]. While the majority of compounds are ferromagnetic, a very small percentage of compounds are a combination of ferromagnetic and antiferromagnetic, or ferromagnetic and superconducting [22], and a slightly larger percentage are antiferromagnetic compounds, although this still pales in comparison to the number of ferromagnetic compounds.

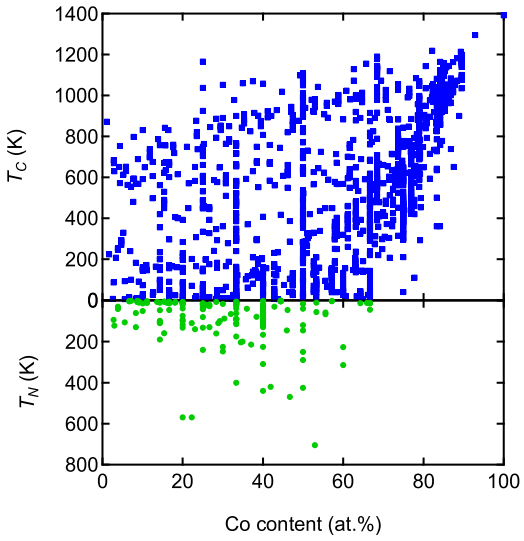

FIG. 3. Curie and Néel temperatures of Co compounds as a function of Co content.

Figure 3 shows the Curie and Néel temperature of Co compounds as a function of Co content. The data include stoichiometric and nonstoichiometric compounds and compounds with other magnetic elements. We can see that, similar to Fig. 2, Co compounds are more often ferromagnetic than antiferromagnetic. This tendency towards ferromagnetic ordering is even more pronounced for compounds rich in Co. As is seen in Fig. 3, there are no known antiferromagnetic Co compounds with more than 67 at. % Co. Above this ratio, the average Curie temperature increases with Co content, reaching its maximum for pure Co [21].

Figure 4 shows the number of Co compounds as a function of Curie or Néel temperature. We observe a maximum in the number of compounds with low ordering temperatures ($<$ 100 K). The number of antiferromagnetic compounds quickly reduces to near zero above 500 K, whereas there is still a relatively large number of compounds with ferromagnetic ordering, even above 1000 K. Based on Figs. 3 and 4, we can see that the Co content is a good criterion to predict ferromagnetism, especially at high at. % Co.

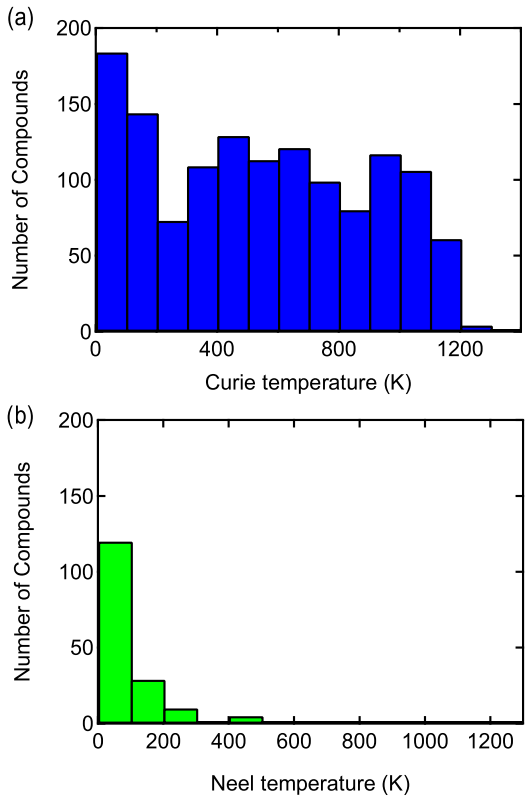

FIG. 4. Number of Co compounds as a function of (a) Curie temperature and (b) Néel temperature.

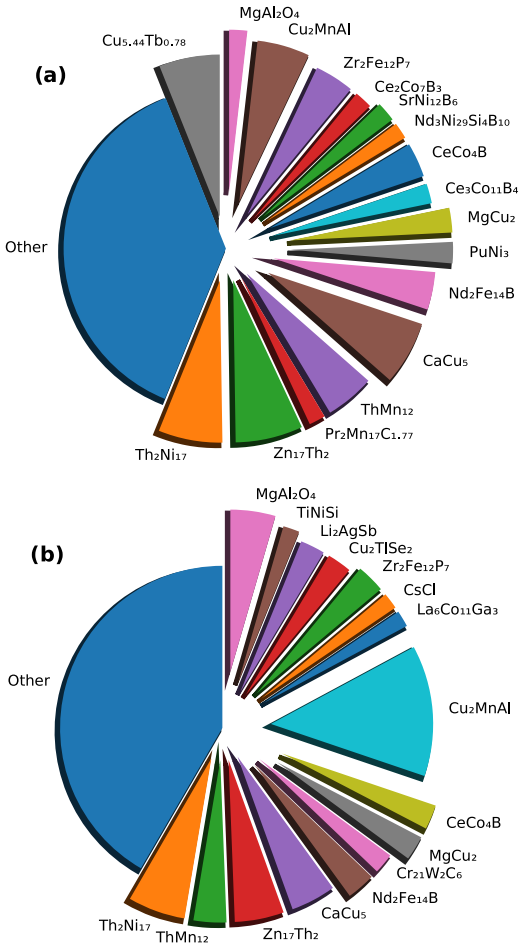

FIG. 5. (a) Distribution of common structures within ferromagnetic cobalt-based compounds. (b) Distribution of common structures within ferromagnetic cobalt-based compounds excluding compounds containing Pr-Lu or Th-Lr.

Sorting the Co compounds by cobalt content and isolating those whose magnetic properties are yet to be investigated leads to a high probability of discovering new ferromagnetic compounds. Recently, we used this criterion to identify a family of easy-axis ferromagnets [23–25]. Ce_2MgCo_9 was one such compound with a known crystal structure but unknown magnetic properties [26]. It can be viewed as an alloy of the Pauli paramagnet $CeCo_3$ in which Ce is partially substituted by Mg. We found that this substitution leads to ferromagnetism, with a Curie temperature as high as 450 K as a result of substantial increase in the Fermi level density of states with Mg alloying [23–25]. To further illustrate the power of using the Co content as a simple criterion, we report the discovery of ferrimagnetism in Nd_2Co_3, a compound that was previously known to crystallize in an orthorhombic structure [27]. With 60 at. % Co, and with Nd as the other element, itself magnetic, it comes as no surprise that Nd_2Co_3 orders magnetically (see calculation done by the Materials Project [28]). We prepared polycrystalline samples and measured its magnetic properties (see Appendix E) confirming a ferromagnetic (or more likely ferrimagnetic) order below 158 K. This shows that a simple criterion such as Co content, combined with the fact that many compounds remain poorly studied in terms of magnetic properties, can enable the discovery of ferromagnets. In general, these results tend to confirm the expectation that compounds with substantial fractions of cobalt usually exhibit ferromagnetism, as illustrated by hcp cobalt itself having the highest Curie point known.

We note that this approach can also be used to identify compounds with anomalous magnetism. For example, we recently investigated Y_2Co_3, which, despite containing 60 at. % Co, was known to order antiferromagnetically, although the crystal structure was unknown. We found that it crystallizes in an orthorhombic structure with distorted kagome nets with an antiferromagnetic order surprisingly robust to magnetic field and temperature [29]. The identification of materials with properties that are not well predicted or do not follow empirical trends is also of interest to guide new discoveries in condensed matter physics.

B. Identifying high-Curie-temperature ferromagnets from structure types

Figures 3 and 4 also show that there is a huge spread of Curie temperatures. This is even true at high at. % Co, as seen in Fig. 3. This wide range of T_C shows that the Co content alone is not a good enough descriptor to estimate the Curie temperature. In this section, we also consider the structure types. Noticing that certain structure types lead to a large number of ferromagnetic compounds, we isolate which structure types are commonly ferromagnetic.

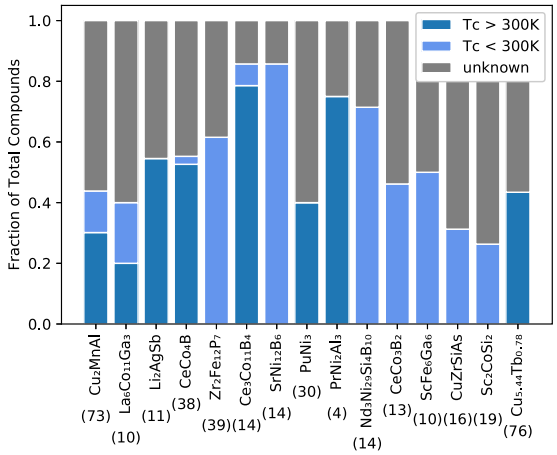

FIG. 6. A breakdown of the known ferromagnets and compounds with no known magnetic ordering in promising structure types, including Cu_2MnAl, $La_6Co_{11}Ga_3$, Li_2AgSb, $Tb_{0.78}Cu_{5.44}$, and $CeCo_4B$.

As shown in Fig. 5(a), after isolating ferromagnetic compounds and dividing them by structure, several common structure types are revealed: $Nd_2Fe_{14}B$, $CaCu_5$, and Th_2Ni_{17}, for instance, are well-known and well-studied families. This classification can reveal other potentially interesting families. We point out the $Tb_{0.78}Cu_{5.44}$ structure (also known as the $TbCu_7$ type), which remains less investigated than other rare-earth families despite a significant number of ferromagnetic compounds (see Table IV in Appendix A). We add an additional filter, removing compounds with rare-earth elements besides Sc, Y, La, and Ce from the pool as shown in Fig. 5(b). We included Ce because it is rather abundant and inexpensive in comparison to the other magnetic rare-earth elements [30].

TABLE II. Compounds with no known magnetic ordering transition but with a high probability of being ferromagnetic.

Structure type	Chemical formula
	BCo_2Fe_2Gd
	$BCeCo_2Fe_2$
	$BCoGdNi_3$
$CeCo_4B$, *hP*12, 191	$BCeCoFe_3$
$Ce_3Co_{11}B_4$, *hP*18, 191	$B_4Co_{11}Lu_3$
	$La_6Co_{13}Bi$[a]
	$Co_{11}Ga_3Sm_6$
	$Co_{11}Ga_3Pr_6$
	$Co_{11}Ga_3Nd_6$
	$Co_{11}Ga_3La_6$
$La_6Co_9(Co_{0.5}Ga_{0.5})_4Ga$, *tI*80, 140	$Co_{13}GeLa_6$
	$CoCr_2Ga$
Li_2AgSb, *cF*16, 216	$CoInMn_2$
$PrNi_2Al_3$, *hP*6, 191	Al_2Co_3Th
	$TaCo_2Ga$[a]
	Co_2GeZn
Cu_2MnAl, *cF*16, 225[b]	Co_2GaSc

[a]Compounds confirmed to be magnetic in this paper.
[b]Only a small subset of unknown compounds is shown.

TABLE III. Breakdown of promising structure type $CeCo_4B$. Unknown compounds with elements Pr-Lu and Th-Lr removed. If multiple references are included, the Curie temperature is an average.

Chemical formula	T_C (K)	EMD (at 300 K)	Reference
BCo_2Fe_2Sm	782	[001]	[64,65]
$BCoFe_3Y$	674	easy-plane	[66]
BCo_2DyFe_2	656.5		[67,68]
BCo_2Fe_2Y	655	easy-plane	[66]
BCo_3FeY	635	easy-plane	[66]
BCo_3FeNd	525	easy-plane	[69,70]
BCo_4Gd	508.333	[001]	[71–73]
BCo_4Sm	505.25	[001]	[72,73]
BCo_4Pr	458.4	easy-plane[a]	[72,73]
BCo_4Tb	456.5	easy-plane	[72]
BCo_4Nd	447.833	easy-plane	[72]
BCo_3NiSm	424	[001]	[73]
BCo_4Dy	422.5	easy-plane[b]	[71]
BCo_4La	420	easy-axis	[74]
BCo_4Lu	396		[75]
BCo_4Er	387	[001]	[71]
BCo_4Y	380.2	complex	[72–74]
BCo_4Ho	372	easy-plane[a]	[71]
BCo_4Th	302.5		[76]
$BCeCo_4$	290.25		[77]
$BCeCo_2Fe_2$			
$BCeCoFe_3$			

[a]Some references list this compound as conical at 300 K.
[b][001] above 340 K.

Figure 6 shows the breakdown of known and unknown magnetic ordering for a selection of structure types. (The chemical composition of unknowns from several of these families is listed in Table II in Appendix A.) One can see in Fig. 6 that several families are promising for high Curie temperatures due to their large fraction of compounds with Curie temperatures above 300 K, including Cu_2MnAl, $La_6Co_{11}Ga_3$, Li_2AgSb, $CeCo_4B$, $Ce_3Co_{11}B_4$, $PuNi_3$, $PrNi_2Al_3$, and $Tb_{0.78}Cu_{5.44}$ ($TbCu_7$).

Using this method, two structure types were chosen as a proof of concept for discovering ferromagnetic materials with high Curie temperatures: Cu_2MnAl and $La_6Co_{11}Ga_3$. Both structure types are studied well enough that we could have confidence that poorly studied compounds would be ferromagnetic. More importantly, these structure types have a large number of high-T_C ferromagnets among the studied compounds [31] (see Fig. 6). From these structures, the compounds $TaCo_2Ga$ and $La_6Co_{13}Bi$ were chosen (the unit cells of these two compounds are shown in Fig. 11). The structure of $La_6Co_{13}Bi$ corresponds to an ordered version of the $La_6Co_{11}Ga_3$ structure type, where one of the 16*l* sites is fully occupied by Co atoms, whereas it is partially occupied by Ga atoms in $La_6Co_{11}Ga_3$ [32]. The Materials Project calculated $TaCo_2Ga$ to be ferromagnetic [10,11,33] and $La_6Co_{13}Bi$ to be ferrimagnetic [10,11,34]. In addition, they both contain safe, relatively inexpensive chemicals. We experimentally confirm that both materials are indeed ferromagnetic, with Curie temperatures of $T_C = 300$ K and $T_C = 470$ K, respectively (see Appendixes C and D). First-principles calculations also

TABLE IV. Curie temperatures and easy magnetization direction of compounds in the structure type $Tb_{0.78}Cu_{5.44}$, *hP*8, 191 ($TbCu_7$). If multiple references are included, the Curie temperature is an average.

Chemical formula	T_C (K)	EMD (at 300 K)	Reference
$PrCo_{6.8}Zr_{0.2}$	963	easy-axis	[78]
$YCo_{6.8}Zr_{0.2}$	1114	easy-axis	[78]
$NdCo_{6.8}Zr_{0.2}$	983	easy-plane	[78]
$NdCo_{6.8}Hf_{0.2}$	1065[a]	conical	[79]
$SmCo_7$	775		[80,81]
$SmCo_{5.8}$	1023		[82]
$SmCo_{5.85}Si_{0.90}$	577.8	easy-axis	[83]
$SmCo_{6.79}Ti_{0.21}$	1029	easy-axis	[80]
$SmCo_{6.72}Ti_{0.28}$	1018	easy-axis	[80]
$SmCo_{6.65}Ti_{0.35}$	1015	easy-axis	[80]
$SmCo_{6.58}Ti_{0.42}$	1006	easy-axis	[80]
$SmCo_{6.44}Ti_{0.56}$	983	easy-axis	[80]
$SmCo_{6.9}Cu_{0.1}$	1123	easy-axis	[81]
$SmCo_{6.8}Cu_{0.2}$	1125	easy-axis	[81]
$SmCo_{6.7}Cu_{0.3}$	1101	easy-axis	[81]
$SmCo_{6.6}Cu_{0.4}$	1042	easy-axis	[81]
$SmCo_{6.5}Cu_{0.5}$	1031	easy-axis	[81]
$SmCo_{6.3}Cu_{0.7}$	1033	easy-axis	[81]
$SmCo_6Ga$	> 800	easy-axis	[84]
$SmCo_{5.7}Ga_{1.3}$	> 800	easy-axis	[84]
$SmCo_{5.4}Ga_{1.6}$	> 800	easy-axis	[84]
$SmCo_{6.9}Zr_{0.1}$	905[a]		[85]
$SmCo_{6.8}Zr_{0.2}$	761	easy-axis	[78,85]
$SmCo_{6.7}Zr_{0.3}$	760		[85]
$SmCo_{6.6}Zr_{0.4}$	752[a]		[85]
$SmCo_{6.5}Zr_{0.5}$	736[a]		[85]
$SmCo_{6.2}Zr_{0.8}$	420		[85]
$SmCo_{6.75}Zr_{0.25}$	770	easy-axis	[86]
$SmCo_{6.48}Fe_{0.27}Zr_{0.25}$	770	easy-axis	[86]
$SmCo_{6.34}Fe_{0.41}Zr_{0.25}$	770	easy-axis	[86]
$SmCo_{6.21}Fe_{0.54}Zr_{0.25}$	770	easy-axis	[86]
$GdCo_{6.9}Zr_{0.1}$	1144	easy-axis	[78]
$TbCo_{5.1}$	980	easy-plane	[81]
$TbCo_{6.7}Cr_{0.3}$	722.5		[83]
$TbCo_{6.7}Zn_{0.3}$	> 900		[83]
$TbCo_{6.7}Ga_{0.3}$	831.8		[83]
$DyCo_{5.2}$	960	easy-axis	[37]
$DyCo_{6.9}Zr_{0.1}$	1128	easy-plane	[78]
$HoCo_{5.5}$	1033	easy-axis	[37]
$HoCo_5Mn_{0.5}$	[b]		[87]
$HoCo_{5.35}Cu_{0.15}$	[b]		[87]
$HoCo_{6.9}Zr_{0.1}$	1124	easy-axis	[78]
$ErCo_6$	> 800	easy-axis	[88]
$ErCo_{6.9}Zr_{0.1}$	1122	easy-axis	[78]
$ThCo_{5.9}$	748		[89]

[a]Multiple transitions (highest is given).
[b]Magnetic measurements interpreted as ferrimagnetic [87].

confirm a ferromagnetic ground state for these compounds and find saturation magnetic moments of 2.00 μ_B/f.u. for $TaCo_2Ga$ and 17.1 μ_B/f.u. for $La_6Co_{13}Bi$. The former result is in fair, if not excellent, agreement with the measured value of 1.6 μ_B/f.u. This is a positive result: Both compounds attempted are ferromagnetic, which acts as an excellent proof of concept for our method of discovering ferromagnets.

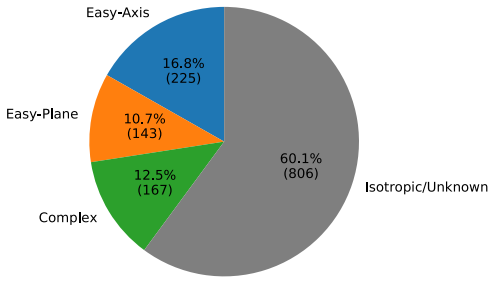

FIG. 7. A distribution of magnetic anisotropies within all 1341 Co-based ferromagnets. The ordering "complex" refers to conical ordering, as well as systems with spin reorientation as a function of temperature.

Besides targeting compounds with high Curie temperatures, we can use the same method to identify compounds near magnetic instabilities. For example, compounds with low Curie temperatures might already be close to a magnetic instability, and one could further tune the system toward a quantum phase transition by chemical substitutions or pressure. Similarly, we can target compounds at the border with another magnetic instability, such as antiferromagnetic compounds close to ferromagnetism. Indeed, some compounds have a reported antiferromagnetic ordering in a structural family that has mostly ferromagnetic materials. An example is $NpCo_2$ with $T_N = 12.5$ K [35], whereas other compounds in the same structure are ferromagnetic: The Np moments order ferromagnetically in $NpMn_2$ ($T_C = 18$ K [36]), $NpFe_2$ ($T_C \approx 500$ K [36]), and $NpNi_2$ ($T_C = 32$ K [36]), and the RCo_2 compounds are ferromagnetic for R = Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, and Tm [37]. Interestingly, local spin density approximation (LSDA) calculations show that $NpCo_2$ is close to a magnetic instability and that ferromagnetic alignment of the Np moments is the most stable [35]. A metamagnetic transition is observed at 3 K at 4.3 T along the [100] easy-axis direction [35]. It is therefore possible to use these data to identify anomalous compounds close to instabilities.

C. Identifying easy-axis ferromagnets with high Curie temperatures and without rare-earth elements

After proof-of-principle successes in discovering ferromagnetic compounds, we tackle the task of identifying compounds that have strong easy-axis magnetic anisotropy. Before excluding certain rare-earth elements, we look at statistics on all Co-containing ferromagnets.

The distribution of magnetic anisotropies in Co-based ferromagnets can be seen in Fig. 7, which separates the types of magnetic anisotropy into four categories: easy-axis, easy-plane, complex, or isotropic-unknown. In principle, orthorhombic compounds could fall into any of the four categories, but all the ones in our database have an identified easy axis and display coercivity [38–40]. The largest fraction of compounds is for those that have not been characterized for magnetic anisotropy properties, providing a pool of materials for new discoveries. The second-largest fraction is for compounds with easy-axis anisotropy. While encouraging, this fraction is not appreciably greater than that of compounds with easy-plane or complex anisotropy. It should be noted,

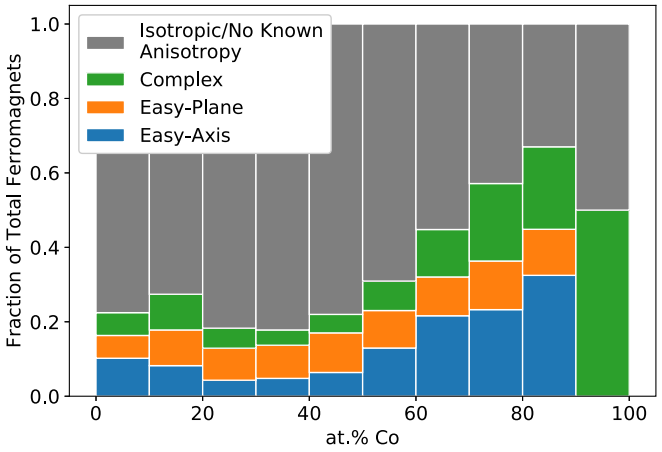

FIG. 8. A comparison of cobalt percentage distributions between easy-axis, easy-plane, complex, and isotropic or unknown ferromagnets (for all 1341 ferromagnets).

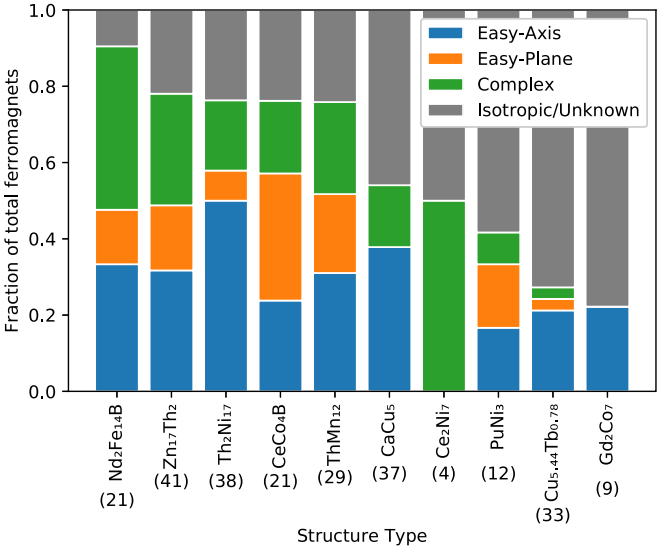

FIG. 9. Spread of anisotropy type among structure types commonly associated with easy-axis ferromagnetism.

however, that the category "complex" does include materials that have both easy-axis and easy-plane anisotropy at varying temperatures in addition to others such as conical ordering. Cobalt itself actually falls into this category, having an easy magnetization direction (EMD) in the [001] direction below 535 K and having easy-plane anisotropy above 595 K [41]. This means that certain compounds within the "complex" category may still have useful easy-axis properties and may be individually considered. For example, we recently reinvestigated the magnetic properties of $(Co_{1-x}Fe_x)_2B$ alloys [42,43]. Co_2B has an easy-axis anisotropy below 80 K and planar anisotropy above 80 K up to $T_C = 426$ K. The planar anisotropy makes Co_2B inappropriate for permanent magnet applications. Fe substitutions can, however, stabilize the easy-axis anisotropy up to T_C, which is also increased, making Co_2B a rehabilitated candidate material for permanent magnet applications.

As for the identification of ferromagnetic compounds with targeted Curie temperatures, we can consider the Co content and the structure types. For simplicity, only stoichiometric compounds are considered.

Figure 8 shows that there is no clear trend between the Co content and the magnetic anisotropy, although all the anisotropy types seem to be favored at high Co content.

As can be seen in Fig. 9, there is not the same level of consistency in magnetic anisotropy within structure types as there is for magnetic ordering temperature. (Data for the $CeCo_4B$ structure type are shown in Appendix A, Table III, with a few minor removals, in order to showcase the available data on magnetic anisotropy.) Figure 9 shows that even the structure types generally associated with easy-axis magnetic anisotropy also have a large fraction of compounds with easy-plane or complex magnetic anisotropy. A possible exception is the $Tb_{0.78}Cu_{5.44}$ family, which mostly has compounds with easy-axis magnetic anisotropy. (Table IV in Appendix A shows the ferromagnetic compounds in this family, including substitution series which are not included in Figs. 5, 6, and 9.) Another notable exception is the Gd_2Co_7 family, which thus far is entirely made up of compounds with easy-axis anisotropy, although it should be noted that the majority of the compounds have not yet been studied for magnetic anisotropy. Thus we are unable to simply use the structure types to identify new candidate materials. These data can be used, however, to identify known but overlooked compounds. For example, the $CaCu_5$ structure type family is prone to display easy-axis anisotropy, as exemplified by the permanent magnet $SmCo_5$. In an effort to reduce the use of critical elements, $CeCo_5$ appears as a promising candidate to take advantage of the overly produced and less critical Ce. $CeCo_5$ has a $T_C = 649$ K [44] and anisotropy field of 18 T at room temperature [45,46]. In a recent study, some of the present authors showed that concomitant Fe and Cu substitutions for Co can be used to produce a permanent magnet material [47–51].

Another example is the recent reinvestigation of the rhombohedral Th_2Zn_{17} structure type family. Ce_2Co_{17} has a high Curie temperature of 1066 K [52] and a large saturation moment of 1.1 T [53]. However, its rather small magnetic anisotropy makes Ce_2Co_{17} an inferior candidate for permanent magnets in comparison with $Nd_2Fe_{14}B$ or $SmCo_5$. In a recent theoretical study, some of the present authors showed that the magnetic properties of Ce_2Co_{17} could become comparable to state-of-the-art magnetic materials by doping with two elements, Zr and Fe [30]. Other studies showed that concomitant substitutions with La and Ti could produce a viable "gap magnet" with energy products greater than that of alnico, at a lower cost than $SmCo_5$ [54].

To identify new compounds with specific anisotropy, additional descriptors need to be considered. Such descriptors could include crystal field parameters, bonding angles, or the nature of ligands. Collecting these data is left for further study. Here, we will describe a simpler, yet successful method to identify candidate materials for easy-axis anisotropy.

There are two courses of action one can take to try to discover new anisotropic compounds. Similar to how we discovered ferromagnetic materials, one may take the large pool of completely unstudied compounds and apply various cuts and limiting criteria until useful candidates emerge, ultimately

TABLE V. Ferromagnetic cobalt-based compounds that are likely to have easy-axis or easy-plane anisotropy.

Chemical formula	T_C (K)	Reference
$BeCo_{17}Y_2$	1095	[90]
$BeCe_2Co_{17}$	1002	[90]
$Co_{13}InLa_6$	490	[31]
$Co_{13}La_6Sb$	490	[31]
$CoRh_2Sn$	449.33	[91]
Co_2Mg[a]	321	[56]
Co_3Mo	–	–
Ce_3Co_8Si	–	–
Co_5Ge_2	–	–

[a] Compound confirmed to be magnetic in this paper.

discovering a material that is not only an easy-axis anisotropic material, but also a ferromagnet. Another course of action is to pull from the pool of known ferromagnets with unknown anisotropy, which removes the risk of targeting a material that may not even be ferromagnetic.

Based on the fact that anisotropy in general is favored by a high at. % Co (Fig. 8) and also that a high cobalt content improves the probability for a higher Curie temperature (Fig. 3), we can limit our search to materials above 60 at. % Co. Additionally, we can exclude compounds containing rare-earth elements (if one is looking for substitute materials) or toxic materials such as arsenic and thallium. Although these filters are extremely general, we are aided in the fact that most high-temperature ferromagnets either possess cubic symmetry, contain rare-earth or toxic elements, or have already been characterized in terms of magnetic anisotropy. (The remaining candidate easy-axis ferromagnets found this way are shown in Appendix A, Table V. If the material is a known ferromagnet with unknown anisotropy, T_C is given).

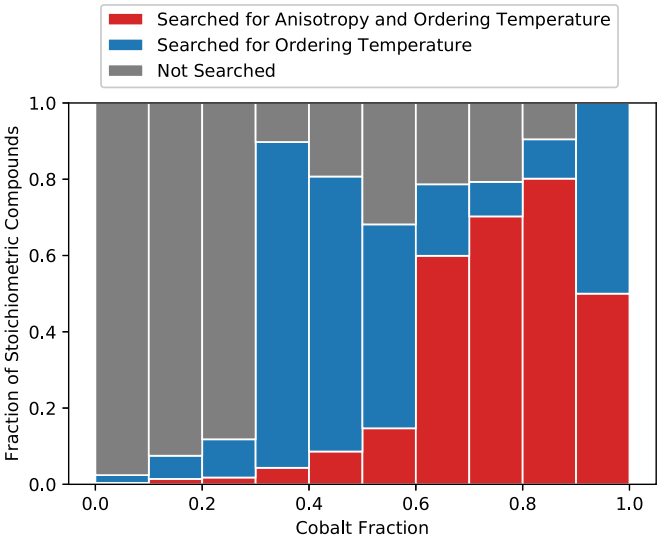

FIG. 10. Percentage of compounds searched as a function of cobalt fraction.

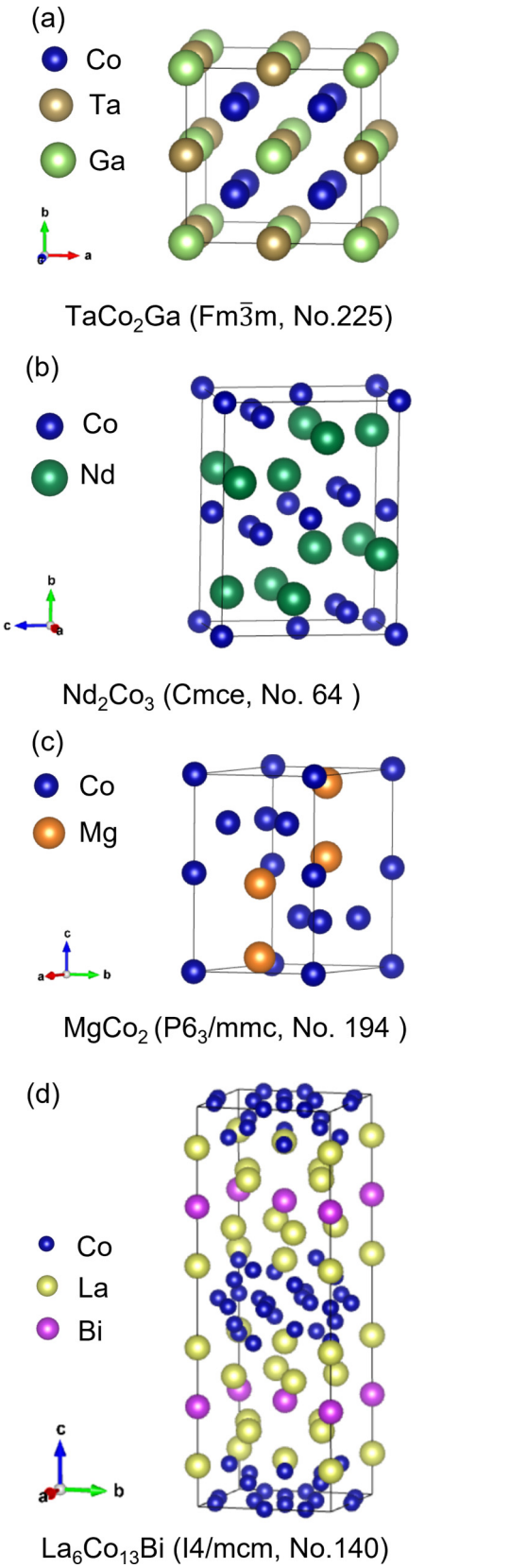

FIG. 11. The unit cells of (a) $TaCo_2Ga$, (b) Nd_2Co_3, (c) $MgCo_2$, and (d) $La_6Co_{13}Bi$.

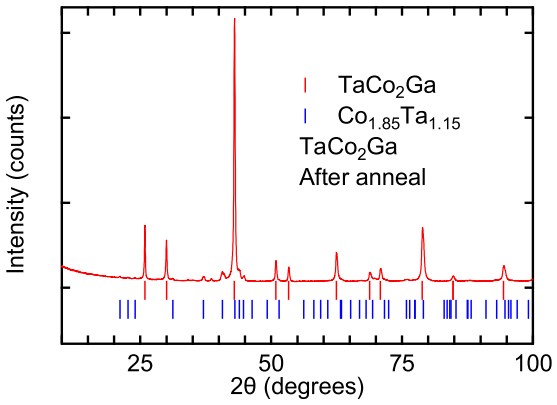

FIG. 12. Powder XRD pattern of $TaCo_2Ga$.

From these candidates, we chose the compound Co_2Mg because of the ease of synthesis, relative lack of previous characterizations, and low cost. This compound belongs to the well-known family of Laves phases [55]. [The unit cell is shown in Fig. 11(c) in Appendix B.] Further literature search on this compound confirmed our choice because an easy magnetization direction along the c axis has already been mentioned based on unpublished neutrons and nuclear magnetic resonance (NMR) measurements [56], although no detailed analysis of magnetic anisotropy can be found for this compound. As shown in Appendix F, our experimental results indeed confirm that Co_2Mg has easy-axis magnetic anisotropy at room temperature, demonstrating the viability of this material-discovery method. Our first-principles calculations find a saturation magnetic moment of 2.75 μ_B/f.u., in excellent agreement with our experimental value of 2.8 μ_B/f.u. By contrast, our first-principles calculations of the magnetic anisotropy in fact find a rather small planar anisotropy in this compound of approximately 7 μeV/f.u., smaller by more than an order of magnitude than the value for hcp Co. While this planar value appears to be at odds with the experimental uniaxial behavior, we note that an alternative explanation for the slight dip in magnetization at low temperature could be a planar-to-uniaxial transition. Such a temperature-dependent anisotropy orientation is in fact exhibited by MnBi [57], Fe_5SiB_2 [58], $(Fe_{1-x}Co_x)_2B$ alloys [42,43], and $Nd_2Fe_{14}B$ itself [59–61]. In any case, such small anisotropies are rather difficult to calculate from first principles, as calculational issues of numerical accuracy and precision become increasingly important for such small energy differences.

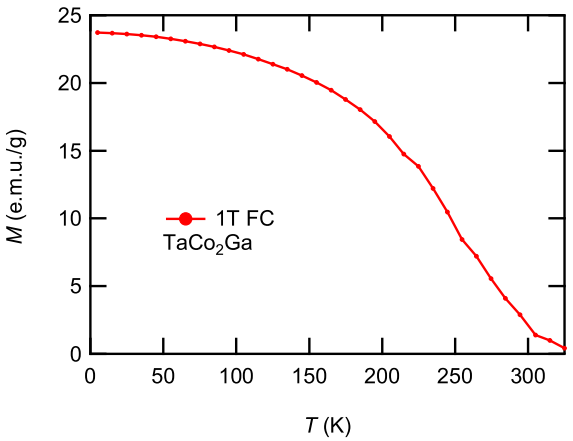

FIG. 13. Field-cooled temperature dependence of the magnetization of $TaCo_2Ga$ at 1 T.

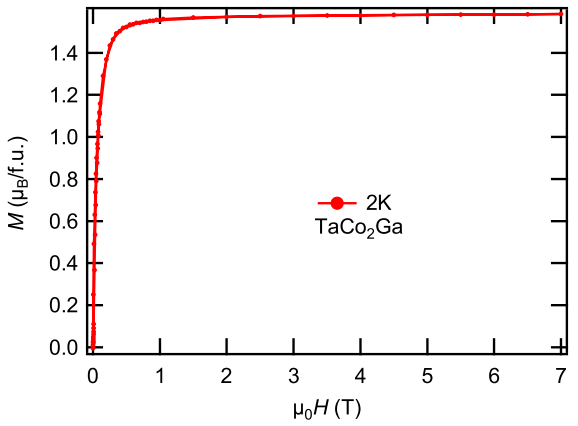

FIG. 14. Magnetization as a function of applied field of polycrystalline $TaCo_2Ga$ at 2 K.

IV. CONCLUSION

Our study illustrates how magnetic data can be used to identify materials with specific properties. We provided a proof of concept for the discovery of ferromagnetic materials, including how to target high Curie temperature and easy-axis anisotropy. We isolated several candidates ($TaCo_2Ga$, $La_6Co_{13}Bi$, and Nd_2Co_3, in this paper and $Ce_{3-x}Mg_xCo_9$ in a previous study) from the subset of uncharacterized compounds, and we confirmed them to be indeed ferromagnetic (or ferrimagnetic in the case of Nd_2Co_3). Of these, three had Curie temperatures at or above room temperature.

We also successfully used available magnetic data to investigate already known ferromagnetic compounds. In addition to the rehabilitation of Co_2B for magnetic anisotropy [42,43], the rehabilitation of $CeCo_5$ for cost [47–51], or the rehabilitation of Ce_2Co_{17} for anisotropy [30,54], the database was used to target compounds with potential magnetic anisotropy, such

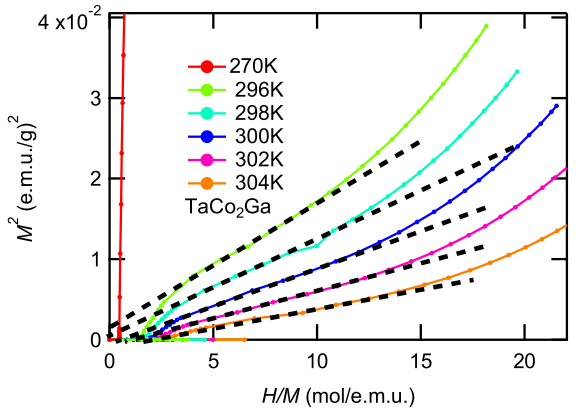

FIG. 15. Arrott plot for $TaCo_2Ga$ using the mean-field critical exponents.

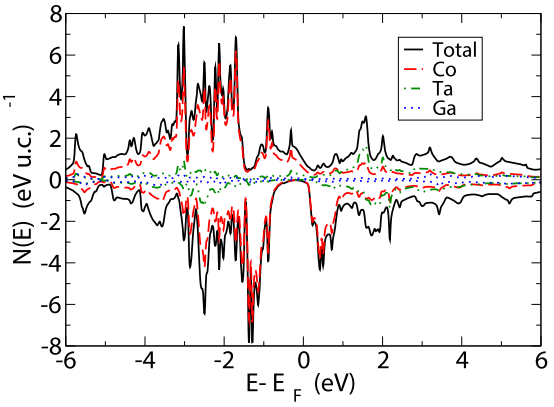

FIG. 16. First-principles-calculated ferromagnetic density of states of $TaCo_2Ga$. Here, u.c. stands for unit cell.

as Co_2Mg and $La_6Co_{13}Bi$. From these, we experimentally confirmed that Co_2Mg has an easy-axis anisotropy. Due to a lack of homogeneity in the type of anisotropy within structure families, there is not always a clear path to isolating easy-axis over easy-plane anisotropy. We experimentally illustrate this with $La_6Co_{13}Bi$ having an easy-plane anisotropy.

The anisotropy analysis can be greatly aided by an increase in available data, as well as a determination of additional descriptors. One example of this would be to calculate the Co-*X* distance for each compound's crystal structure, as well as to find descriptors for the geometry of Co-*X* bonds. Additional data, such as the saturation and effective moment, could also reveal interesting trends, helping in the identification of new materials.

Our method of using a database of magnetic properties does not need to be limited to ferromagnetism. Trends in antiferromagnetism, ferrimagnetism, and even superconductivity may be analyzed using a similar approach. Work can also be done examining links between the different types of ordering within Co-based compounds. Ultimately, with the increase of data sets, and the identification of better descriptors, machine learning methods can also succeed in guiding the discovery of new magnetic materials [62,63].

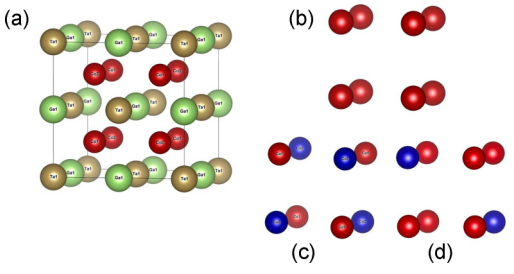

FIG. 17. Physical and magnetic structures of $TaCo_2Ga$ studied. (a) The fcc structure with Ta, Co, and Ga atoms indicated in brown, red, and green, respectively. (b)–(d) The magnetic structures, where red coloring indicates spin up, blue indicates spin down, and only the Co atoms are shown: the ferromagnetic structure (b), the nearest-neighbor antiferromagnetic structure AF1 (c), and a ferrimagnetic structure FI1 (d). Of these magnetic states, only the ferromagnetic structure in (b) yields a converged consistent magnetic result.

Additionally, our method can be extended to more than just cobalt materials. Work is being done now to collect data for lists of compounds containing Fe, Mn, and Ni, to analyze trends within other magnetic elements, and to also gain a broader look at magnetism as a whole.

ACKNOWLEDGMENTS

We would like to thank M. Montero and R. Ullah for useful discussions. The research was supported by the Critical Materials Institute, an Energy Innovation Hub funded by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Manufacturing Office. We acknowledge support from the Physics Liquid Helium Laboratory fund. Work done at Ames Laboratory (PCC and SLB) was supported by the U.S. Department of Energy, Office of Basic Energy Science, Division of Materials Sciences and Engineering. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358.

APPENDIX A: ADDITIONAL INFORMATION ABOUT THE DATABASE

To give a better idea of the scope of the data we perform the analysis on, we provide Fig. 10 showing the fraction of stoichiometric compounds searched by the authors as a function of cobalt content. Down to 33 at. % Co, 1398 compounds out of the total 1682 have been searched.

Tables showing the breakdown of promising families as well as tables containing the predictions of possible ferromagnetic or anisotropic ferromagnetic materials are given here. Table II shows a list of several promising compounds taken from structure types shown in Fig. 6. Table III specifically highlights the $CeCo_4B$ structure type, which shows a variety of anisotropy types within a single family. Table IV shows the Curie temperatures and easy magnetization direction of compounds in the structure type $Tb_{0.78}Cu_{5.44}$,hP8,191 ($TbCu_7$). Table V indicates a combination of fully unstudied materials and known ferromagnets with unknown magnetic anisotropy.

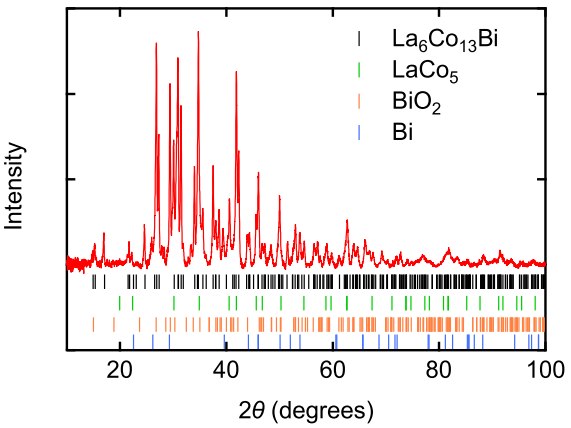

FIG. 18. Powder x-ray diffraction pattern of $La_6Co_{13}Bi$.

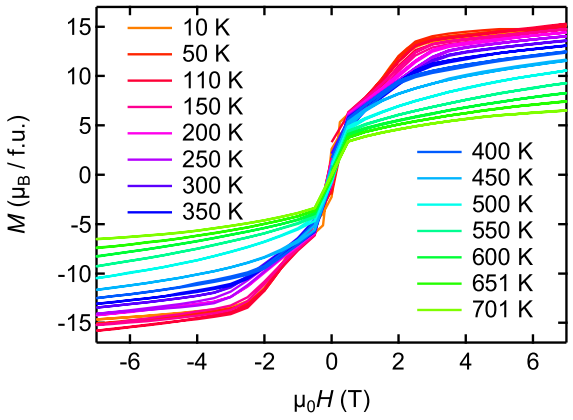

FIG. 19. Magnetic hysteresis loops of $La_6Co_{13}Bi$ at various temperatures.

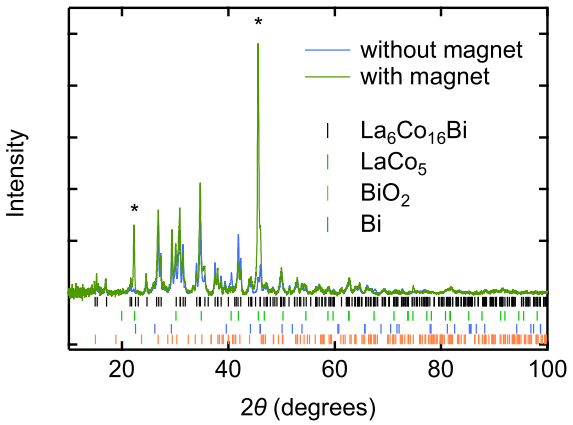

FIG. 21. A comparison of the XRD pattern of $La_6Co_{13}Bi$ with and without a magnet. Peaks labeled with an asterisk belong to the {001} plane family of $LaCo_5$.

APPENDIX B: CHARACTERIZATION METHODS

For each material, powder x-ray diffraction (XRD) was performed using a Rigaku MiniFlex 600 diffractometer to confirm the composition of the synthesized samples. The same diffractometer was also used to find the easy magnetization direction at room temperature of $La_6Co_{13}Bi$ and $MgCo_2$ using a small magnet to create a magnetic field and align the powder [92]. A thin layer of Vaseline grease was applied on the puck, and the redundant grease was wiped away with a Kimwipe. Then the sample powder was evenly spread onto the sample puck. The x-ray diffraction pattern was first collected without the application of a magnetic field. After these data were taken, a Nd magnet (13/16 in. diameter, 1/16 in. thick) was glued on the back of the puck to apply a perpendicular magnetic field. The sample powder grains reoriented immediately after the magnet was attached as visible under a microscope, and the powder XRD pattern was collected again with the same sequence. Any changes in peak intensity indicate the axes or planes of the preferred magnetic orientation. This analysis is performed under the assumption that anisotropic effects stem from magnetocrystalline anisotropy rather than from shape anisotropy.

Magnetization measurements were performed using a Quantum Design Magnetic Property Measurement System (MPMS). Magnetization above 300 K was measured with an oven heater option.

A heat capacity measurement was performed for Nd_2Co_3 in a Quantum Design Physical Property Measurement System (PPMS).

The unit cells of $TaCo_2Ga$, Nd_2Co_3, $MgCo_2$, and $La_6Co_{13}Bi$ are shown in Fig. 11.

APPENDIX C: $TaCo_2Ga$

Polycrystalline samples of $TaCo_2Ga$ were synthesized by arc-melting stoichiometric ratios of cobalt pieces (Alfa Aesar 99.995%), tantalum foil (99.9%), and gallium pellets (Alfa Aesar 7N) in argon. Samples were annealed at 800 °C in evacuated silica ampoules for 5 days, ground into a powder and pressed into a pellet, and then annealed at the same temperature for an additional 20 days [93]. Each time, a small piece of tantalum foil was included, though not touching the sample,

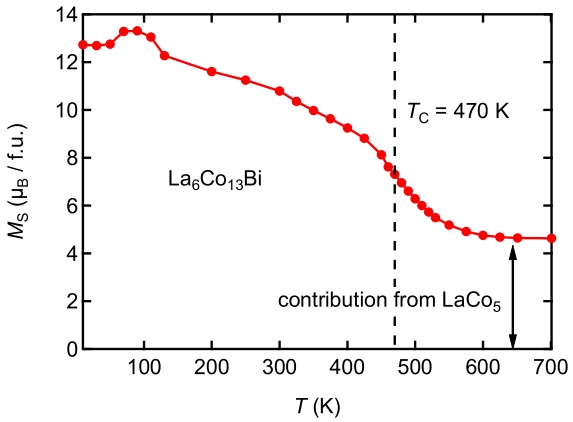

FIG. 20. Temperature dependence of the spontaneous magnetization of polycrystalline $La_6Co_{13}Bi$. The magnetization remains relatively large above $T_C = 470$ K because of the presence of $LaCo_5$ impurities with $T_C = 840$ K.

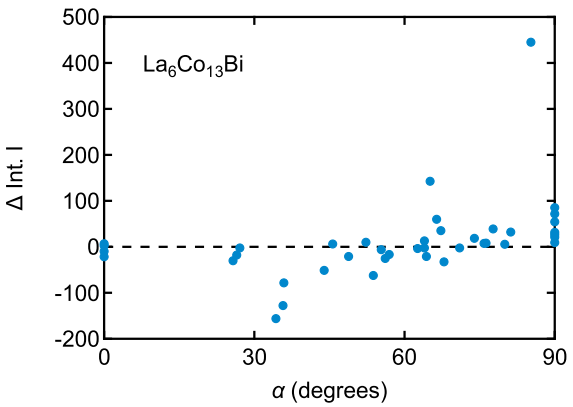

FIG. 22. Difference of integrated (Int.) intensities of the powder x-ray diffraction peaks with and without a magnet as a function of the angle between the corresponding crystallographic planes and the (001) planes.

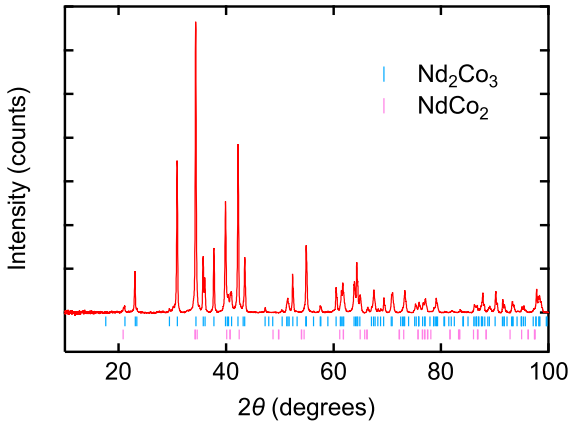

FIG. 23. Powder XRD patterns of Nd_2Co_3 polycrystals.

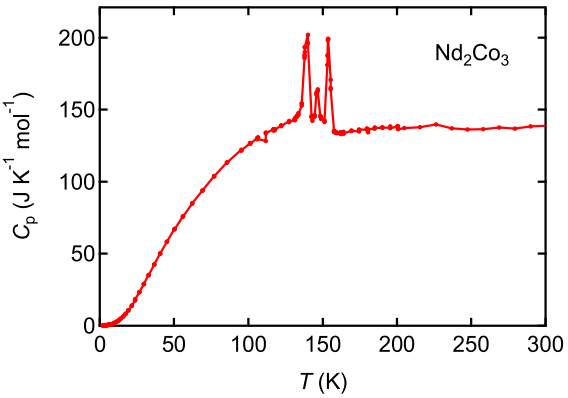

FIG. 25. Specific heat as a function of temperature for Nd_2Co_3 polycrystals.

for use as an oxygen getter. After annealing, the sample was found, using an x-ray diffractometer, to be mostly phase pure. The XRD plot is shown for $TaCo_2Ga$ in Fig. 12.

The magnetization versus temperature and versus applied field is shown in Figs. 13 and 14.

The Curie temperature is found to be $T_C = 300$ K using an Arrott plot shown in Fig. 15. The isotherms are not parallel indicating that the transition is not well described by a mean-field model. Neither the three-dimensional (3D) Heisenberg nor the Ising critical exponents lead to parallel isotherms in a modified Arrott plot, so only the mean-field approximation is shown. The lines in Fig. 15 were drawn by eye, with the line drawn over 300 K intersecting the zero magnetization of the 270-K curve, indicating a $T_C = 300$ K.

Figure 16 shows the calculated density of states (DOS) for $TaCo_2Ga$. The near-Fermi-level contributions are dominated by the cobalt contribution, as expected, with minimal contributions from the Ta and Ga sites.

Interestingly, $TaCo_2Ga$ appears to be of substantially itinerant character, potentially associated with its relatively small ferromagnetic amount (1 μ_B/Co in the calculations). We have attempted to stabilize additional magnetic states by initializing different magnetic configurations (see Fig. 17). However, the nearest-neighbor antiferromagnetic state initialized ["AF1"; shown in Fig. 17(c)] rapidly converges to a non-spin-polarized result, with no magnetic moment on any atom, and a ferrimagnetic-initialized state ["FI1"; see Fig. 17(d)] did not converge even following some 200 iterations. It is plausible that the likely itinerant character here is responsible for the different DOS appearance compared

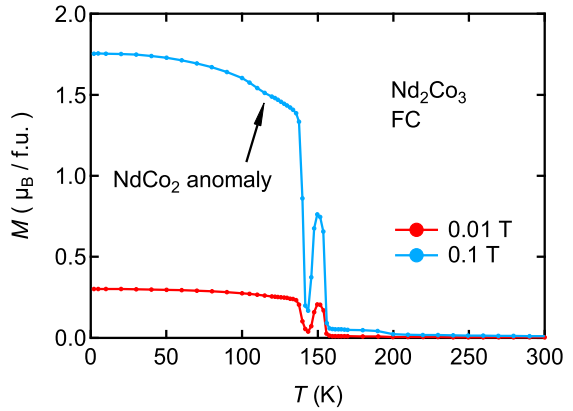

FIG. 24. Magnetization as a function of temperature for Nd_2Co_3 polycrystals.

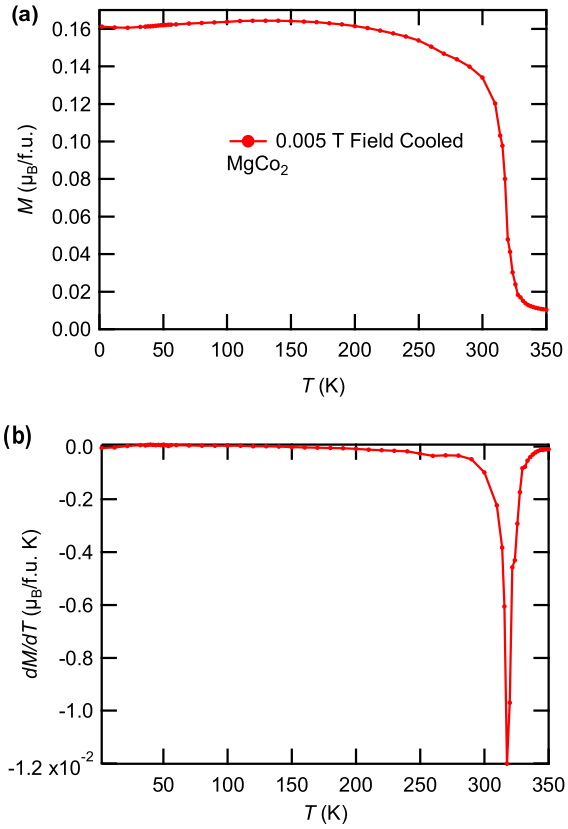

FIG. 26. (a) Temperature dependence of magnetization for polycrystalline $MgCo_2$ at an applied field of 0.005 T. A ferromagnetic transition can be seen at 319 K, which lines up with previous reports [56]. The slight dip in magnetization around 50 K corresponds to the antiferromagnetic transition observed by Buschow *et al.* [56]. (b) The derivative of magnetization vs temperature as a method to find T_C for $MgCo_2$.

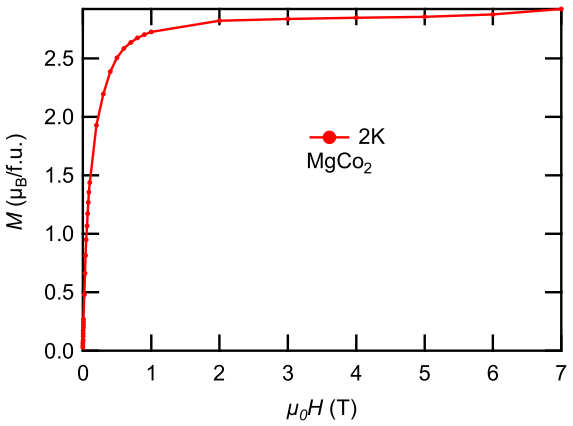

FIG. 27. Magnetization plotted as a function of applied field for polycrystalline $MgCo_2$ at 2 K. The saturation magnetization is 2.8 μ_B/f.u.

with that of Co_2Mg, potentially augmented by the use of a refractory, and hence strongly bonding, metal Ta.

APPENDIX D: $La_6Co_{13}Bi$

Polycrystalline samples of $La_6Co_{13}Bi$ were synthesized by arc-melting a starting composition of La_6Co_{13} (La, Ames Laboratory; Co, 99.8%) in argon atmosphere. Following this, stoichiometric ratios of Bi needles (99.999%) and the arc-melted La_6Co_{13} sample were heated to 1150 °C in a silica ampoule filled with argon gas, held for 24 h, and then quenched in water. The sample was then annealed at 800 °C for 7 days, ground and pressed into a pellet, and annealed at the same temperature for another 14 days. Figure 18 shows the powder x-ray diffraction pattern of the $La_6Co_{13}Bi$ sample. The majority phase is $La_6Co_{13}Bi$; however, there are some impurity phases including $LaCo_5$, bismuth, bismuth oxide, and a few unidentified peaks.

Figure 19 shows the magnetization hysteresis loops of the sample at various temperatures. The spontaneous magnetization is extrapolated from the linear fitting of the M-vs-H curve from 7 to 4 T. Figure 20 shows the saturated magnetization of $La_6Co_{13}Bi$ as a function of temperature from 10 to 700 K. The T_C of $La_6Co_{13}Bi$ was determined to be 470 K. There is an anomaly at around 90 K, which is not corresponding to the known impurities identified by the powder x-ray diffraction. Such an anomaly is likely to be an intrinsic behavior of $La_6Co_{13}Bi$ or from an unknown impurity.

The powder x-ray diffraction pattern at room temperature with and without the application of magnetic field perpendicular to the plane of the puck is shown in Fig. 21. For the $LaCo_5$ impurity peaks, the easy magnetization direction along the c axis is confirmed by the increased intensity of the $(00l)$ peaks marked by an asterisk. For $La_6Co_{13}Bi$, the difference of intensity is more subtle indicating a much smaller magnetic anisotropy. Figure 22 shows the difference of integrated intensity as a function of angle α, defined as the angle between the crystallographic planes (hkl) and the (001) planes [94]. For an easy-plane magnetic anisotropy, the application of a magnet rotates the (001) planes perpendicular to the plane of the x-ray diffraction puck and thus renders them outside the diffraction conditions. Therefore the intensities of (hkl) planes that are farther away from the (001) planes should increase; that is, the intensity increases as α increases. This behavior is observed in Fig. 22: The intensities tend to increase as α increases toward 90°, indicating that the weak magnetic anisotropy is close to an easy-plane anisotropy.

APPENDIX E: Nd_2Co_3

Nd_2Co_3 polycrystals were synthesized by arc-melting stoichiometric ratios of Co and Nd in argon. The sample was then annealed at 610 °C under vacuum for 2 weeks. The unit cell is shown in Fig. 11(b). Figure 23 shows the powder x-ray diffraction patterns of the Nd_2Co_3 polycrystals. Field-cooled bulk magnetization of the polycrystals Nd_2Co_3 was measured from 2 to 300 K. Three anomalies can be observed in Fig. 24: a sharp increase below 154 K, a maximum at 146 K, and a minimum at 140 K. This indicates that there are several types of ordering in Nd_2Co_3 with different transition temperatures. Thus Nd_2Co_3 is likely to have a ferrimagnetic ordering. An additional small anomaly at 110 K likely corresponds to the ferromagnetic transition of $NdCo_2$ impurities.

The temperature dependence of the heat capacity is shown in Fig. 25. Three peaks at 140, 146, and 153 K are related to

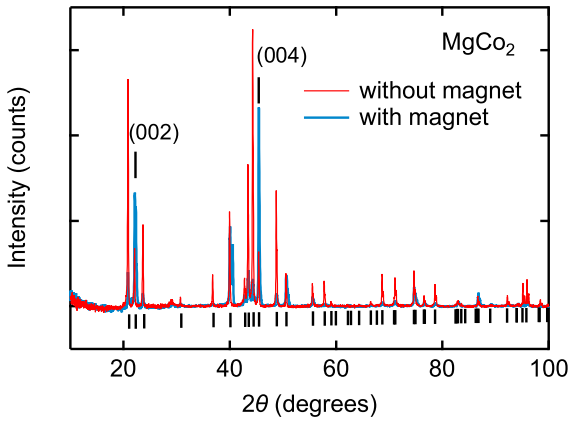

FIG. 28. A comparison of the XRD pattern of $MgCo_2$ polycrystals with and without a magnet. The intensity of the (002) and (004) peaks increases.

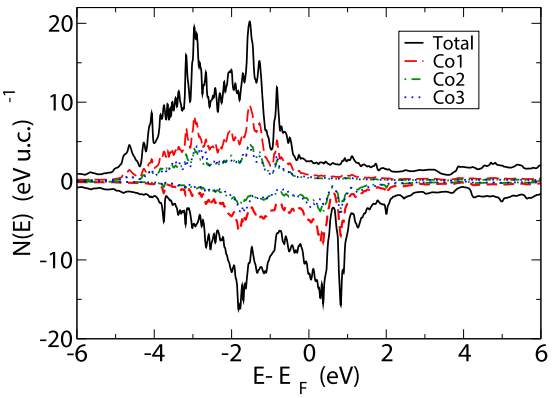

FIG. 29. First-principles-calculated ferromagnetic density of states of Co_2Mg.

three anomalies in magnetization measurements, confirming three magnetic phase transitions.

APPENDIX F: $MgCo_2$

Polycrystals of $MgCo_2$ were synthesized by combining pieces of an Mg rod (99.9%) with Co powder (99.8%) and pressing them together into a pellet. Since Mg reacts with silica, a Ta tube was used for synthesis. The mixture was then increased to 680 °C at a rate of 340 °C/h and then increased to 850 °C at a rate of 34 °C/h and held for 5 h. The sample was increased to 980 °C, where it was held for 57 h before being air quenched and transferred to a second furnace. It was then raised to a temperature of 1050 °C at a rate of 50 °C/h before being held for 48.5 h. The sample was then annealed at 900 °C for 2 weeks.

The temperature and field dependence of magnetization (Figs. 26 and 27) was measured using a Quantum Design MPMS, with an oven attachment utilized for measurements above 300 K. The saturation magnetization of $MgCo_2$ was measured to be 2.8 μ_B/f.u. In accordance with the previously reported value of 321 K [56], a Curie temperature was observed at 319 K by taking the minimum of the derivative of the magnetization vs temperature at 0.005 T [see Fig. 26(b)].

Since the T_C of $MgCo_2$ is above room temperature, XRD analysis with and without the application of a magnet can again be used to determine an easy magnetization direction.

Figure 28 exhibits the powder XRD diffraction of $MgCo_2$ polycrystals with and without an external magnetic field. The perpendicular magnetic field was applied by gluing a magnet underneath the puck. One can notice in the figure that with the application of magnetic field, the intensities of the (002) and (004) peaks increase while other peaks show a decrease in their intensities. Such a result indicates that the powder grains orientate along the [001] direction within the magnetic field, which confirms that the *c* axis is the easy axis and $MgCo_2$ has an easy-axis magnetic ordering.

Figure 29 shows the calculated density of states of Co_2Mg. The distinct sites split by spin-orbit coupling show very similar detailed shapes, reflective of the small spin-orbit coupling, and thereby small magnetic anisotropy, in this transition-metal-dominated compound.

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