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Machine Learning Study of Highly Spin-Polarized Heusler Alloys at Finite Temperature

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ABSTRACT

Huge magnetoresistance (MR) ratio exceeding 2000% at cryogenic temperature that was reported for half-metallic Heusler alloy based magnetic tunnel junctions showed large degradation at room temperature, which impedes practical application of Heusler alloys based MR devices. This motivates us to explore new Heusler alloys that show high spin polarization at finite temperatures. Here, we propose new half-metallic Heusler alloys based on finite temperature first-principles calculation via disordered local moment method together with machine learning. We found several new prospective materials at room temperature such as $\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$ and $\text{Co}_2\text{FeAl}_{0.4}\text{Sn}_{0.6}$. We also investigated two combinatorial series $\text{Co}_2\text{MnGa}_y\text{As}_{1-y}$ and $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$ to understand the effect of alloy mixing on temperature dependence and found that Fermi level tuning significantly improved the spin polarization and its temperature dependence especially in $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$.

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Magnetoresistance (MR) in magnetic tunnel junctions (MTJ) is one of the most important phenomena to realize spintronics applications, such as magnetic random access memories [1] and magnetoresistive sensors [2]. Recently, high-throughput calculations have been widely performed to obtain novel magnetic materials with high spin-polarization around Fermi level (so-called half-metals) [3–7], because the high spin-polarization is crucial to obtain sufficient magnetoresistance for the applications. In particular, Heusler alloys, a family of A_2BC compounds, have been actively explored by machine learning due to the abundant variety of atomic combinations and the relatively simple fabrication process. However, the machine learning investigation of highly spin-polarized Heusler alloys with the first-principles calculations have been performed only at zero temperature [7]. This made a significant discrepancy between physical properties designed by first principles calculations and experimental results of the predicted material.

One of the most serious problems of half-metallic Heusler alloys in spintronic device application is the reduction of magnetoresistance at room temperature (the large temperature dependence) in tunnel magnetoresistance (TMR) and the current-perpendicular-to-plane giant magnetoresistive (CPP-GMR) devices [8,9]. This means that simple ground-state calculations of spin-polarization at 0 K are not enough to predict half-metallicity at ambient temperature. In our previous work, we performed the finite temperature calculations of half-metallic electronic structures of Co-based full Heusler alloys and found that the conduction bands of Co d orbital significantly degrade the spin-polarization at room temperature [10]. Experimentally, tuning the Fermi level via alloying of different elements, such as $\text{Co}_2\text{Fe}(\text{Al}_{0.5}\text{Si}_{0.5})$ [11] is also important to improve the large temperature dependence of spin-polarizations. These previous experimental and theoretical studies suggest that for realization of the spintronics applications, machine learning with the finite temperature first-principles calculations is necessary to predict novel half-metallic Heusler alloys at room temperature.

The purpose of the present work was to search for new half-metallic Heusler alloys having high spin-polarization at room temperature using machine learning and the finite-temperature first-principles calculations. Due to the multielement character of Heusler alloys, machine learning is a suitable material informatics tool for multidimensional analysis. Recently, using deep neural network, Hu *et al* predicted > 100 highly spin-polarized and 6 prospective half-metal from $>10,000$ Heusler A_2BC candidates at 0 K [7]. In contrast to the previous work, we incorporate the finite-temperature effect via disordered local moment (DLM) method to clarify the spin polarization of ternary, quaternary and quinary Heusler alloys at finite

temperature [10,12]. We successfully predict new quaternary Heusler alloys on the basis of the finite temperature machine learning and the first-principles calculations, such as $\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$ and $\text{Co}_2\text{FeAl}_{0.4}\text{Sn}_{0.6}$, which show high spin-polarization even at room temperature.

We prepare a list of candidates of Heusler compounds with general composition of $A_2(B_xB'_{1-x})(C_yC'_{1-y})$ (x and y are coefficient from 0.0 – 1.0 with 0.2 interval). Elements for A , B (B') and C (C') sites are taken from the distribution of components predicted by Hu *et al.* (A = Fe, Co, Ru, Rh; B, B' = Sc, Ti, V, Cr, Mn, Fe, Y, Zr, Nb, Mo; C, C' = Al, Si, P, Ga, Ge, As, In, Sn, Sb) [7]. These combination leads to 73,440 different compositions. Each candidates assumes the $L2_1$ structure, which has four atoms located on interpenetrating fcc sublattices, consisting of two A atoms at (0.25, 0.25, 0.25) and (0.75, 0.75, 0.75), the B (B') atoms at (0.00, 0.00, 0.00) and the C (C') atoms at (0.50, 0.50, 0.50). The lattice constant of ternary alloys were taken from the Heusler Database of The University of Alabama [13] and supplementary information from Hu *et al.* [7]. Using Vegard's law [14], the lattice constants of quaternary and quinary Heusler composition were extrapolated.

The computational procedures to calculate the spin-polarization at finite temperature are divided into two steps. First, spin-polarized electronic structures calculations at zero temperature were performed by multiple-scattering Green's function formalism in the Korringa-Kohn-Rostoker (KKR) method [15,16] implemented in HUTSEPOT code [17]. Local-spin density approximation (LSDA) by Perdew and Wang [18] was selected for exchange-correlation functional and the k -mesh $20 \times 20 \times 20$ was used. The core and valence electrons were treated within the scalar-relativistic approximation and the angular momentum expansion of scattering matrices in a basis of spherical harmonics is truncated at $l_{\text{max}} = 3$. To deal with the atomic disorder, we used coherent potential approximation (CPA) [19]. Kohn-Sham potential was determined using atomic-sphere approximation (ASA). Second, after obtaining the ground-state potential from the first step as the “frozen” potential, we introduced the finite temperature effect using the DLM method [20]. This effect assumes that magnetic compounds consist of local moments that fluctuate at finite temperature and have longer stabilization timescale than electron motion [20]. Therefore, the scalar-relativistic Kohn-Sham-Dirac equation was solved non-self-consistently within the ASA until it reached the convergence of iterative temperature value with the starting parameter of Weiss field as “average mean field”. The detail technical description of DLM approach are given elsewhere [10,12,20].

Total *spd* (total) spin polarization at zero and finite temperature is evaluated by

$$P_{spd} = \frac{D_{spd}^{\uparrow}(E_F) - D_{spd}^{\downarrow}(E_F)}{D_{spd}^{\uparrow}(E_F) + D_{spd}^{\downarrow}(E_F)}, \quad (1)$$

where $D_{spd}^{\uparrow}(E_F)$ and $D_{spd}^{\downarrow}(E_F)$ correspond to the total *spd* density of states (DOS) on the Fermi level of majority and minority spins, respectively. Despite dependency of MR ratio on many factors such interfaces quality, defect, vacancy, lattice mismatch, etc, we only focus on the reduced bulk *sp* spin polarization at finite temperature in this study. It has been demonstrated that *sp* spin polarization have good agreement with spin polarization extracted from experimental TMR ratio using Julliere's model [21,22]. Furthermore, experimental studies of CPP-GMR suggested that CPP-GMR ratios are more properly explained by *sp* spin polarization due to the negligible contribution of localized character *d* electrons to the transport properties [23]. Therefore, we calculated *sp* spin polarization, which is defined as

$$P_{sp} = \frac{D_{sp}^{\uparrow}(E_F) - D_{sp}^{\downarrow}(E_F)}{D_{sp}^{\uparrow}(E_F) + D_{sp}^{\downarrow}(E_F)}, \quad (2)$$

where the $D_{sp}^{\uparrow}(E_F)$ and $D_{sp}^{\downarrow}(E_F)$ corresponds to the *sp* DOS on the Fermi level of majority and minority spin, respectively.

Our main purpose was to find a new Heusler alloys composition that retain the high spin polarization at finite temperature. Due to the complex relation between the output (high spin polarization) with the input (composition), this problem can be assumed as black-box function, and Bayesian optimization is adopted to find its solution with high efficiency. In this work, the open-source Python library called COMBO [24] was employed to perform the optimization process.

Aside from the optimization method, there are other three required components for machine learning process: the descriptor, the evaluator, and the calculator [25]. We set the descriptor to identify each composition $A_2(B_x B'_{1-x})(C_y C'_{1-y})$ as a concatenation of integer numbers following the rule,

$$A_2 \left(B_x B'_{1-x} \right) \left(C_y C'_{1-y} \right) \rightarrow P|G_A + P|G_B + n_x + P|G_{B'} + n_{1-x} + P|G_C + n_y + P|G_{C'}, \quad (3)$$

where $P|G$ is a set of the number of period and group in the periodic table to identify the element (*A*, *B*, *B'*, *C*, *C'*) and *n* indicates the stoichiometry coefficient of *B*, *B'*, *C*, and *C'*. For

examples, Co(Si) is in the 4th (3rd) period and 9th (14th) group, thus we express the $P|G_{Co} = 4|9 \Rightarrow 49$ ($P|G_{Si} = 3|14 \Rightarrow 314$), respectively. The stoichiometry coefficient of n_x and n_y were described as follow, $x = 0.2 \Rightarrow n_x = 02$, and $y = 1.0 \Rightarrow n_y = 10$, and so on. If there is no $B'(C')$ element, we set $P|G_{B'} \Rightarrow 00$ ($P|G_{C'} \Rightarrow 00$) and $n_{1-x} = 00$ ($n_{1-y} = 00$), respectively. By concatenate all of these parameters, each composition will be uniquely described with 20-digit integer descriptor. For example, Heusler alloy, $Ru_2(Mn_{0.6}Fe_{0.4})(P_{0.2}Sb_{0.8})$ can be expressed by “58470648043150251508” due to $P|G_{Ru} = 58$, $P|G_{Mn} = 47$, $n_x = 06$, $P|G_{Fe} = 48$, $n_{1-x} = 04$, $P|G_P = 315$, $n_y = 02$, $P|G_{Sb} = 515$, and $n_{1-y} = 08$.

The framework of the *ab-initio* calculations and MI procedures is illustrated in FIG. 1. First, we define the set of candidates [see FIG. 1(a)], and first 20 compositions were chosen randomly to train the Bayesian regression as machine learning model [see FIG. 1(b)]. Performance of each candidate was evaluated with the score defined by

$$\text{score} = T_{\text{conv}} \cdot P_{sp}(T_{\text{conv}}). \quad (4)$$

The T_{conv} and $P_{sp}(T_{\text{conv}})$ were the converged temperature and the *sp* spin-polarization at T_{conv} , which were obtained based on the DLM calculations at finite temperature [see FIG. 1(c)]. We adopted the spin-polarization of *sp*-states P_{sp} at finite temperature as the evaluator (score) of the Bayesian optimization, because the P_{sp} is more suitable than the P_{spd} (total spin-polarization) to describe the spin-dependent transport in TMR and CPP-GMR devices.

Obtained score is used to estimate the black box function which is expensive to calculate. This estimation model based on the Bayesian statistics will predict the next candidate that need to be evaluated. First, we randomly selected 20 structures with respective descriptors for initial calculations. The scores of these 20 structures are evaluated via first principles calculation, and used to train the Bayesian regression model. Then, in every round the next 10 candidates are chosen by this estimation model and the corresponding scores of these 10 candidates are added to improve the estimation model, repetitively until fixed number of 2200 candidates (220 rounds) are evaluated. In this study, the best candidates are chosen according to the Thompson sampling criterion. Since Bayesian optimization is widely used machine learning framework, details are written in elsewhere [24].

In FIG. 2, we show the score of the Bayesian optimization as a function of the number of calculated structures for the P_{sp} of Heusler alloys, indicating the performance of Bayesian optimization. As shown in FIG. 2, after several iteration processes, Bayesian optimization

converges to the best candidates with largest score, which corresponds to the Heusler alloy compositions that retain high spin-polarization at finite temperature.

In TABLE I, we show top 30 compounds having high scores in the Bayesian optimization together with the lattice constant a , the spin moment m_{total} , total and sp spin-polarizations P_{sp} and P_{sp} at the converged temperature T_{conv} , and also the formation energy E_{form} , which are outputs of the first-principles calculations except for the lattice constants. Scores of $(P_{sp} \cdot T_{\text{conv}})$ are used in predictions of black box function in Bayesian optimization. The formation energy

E_{form} of $A_2(B_xB_{1-x})(C_yC_{1-y})$ was calculated by following equation,

$$E_{\text{form}} = E_{A_2(B_xB_{1-x})(C_yC_{1-y})}^{\text{total}} - \left(2E_A^{\text{total}} + xE_B^{\text{total}} + (1-x)E_{B'}^{\text{total}} + yE_C^{\text{total}} + (1-y)E_{C'}^{\text{total}} \right), \quad (5)$$

where E^{total} is the total energy for each system for Heusler alloys $A_2(B_xB_{1-x})(C_yC_{1-y})$, and single elements A , B , B' , C , and C' calculated using first-principles calculation at zero temperature.

As shown in Table I, despite considering many elements in the A and B site, the majority of prospective candidate are having Co occupy A site, to be specific, Co_2Fe - and Co_2Mn - based Heusler alloys which consisted from 3-, 4-, and 5-element compound. The magnetic moments of these candidates are pretty high, more than $4.5 \mu_B$, which lead to converged temperature around 200-350 K using same starting parameter Weiss field. Interestingly, these candidates have various value of P_{sp} spanned over the range -20 – 80%, despite the relatively high P_{sp} more than 70%. This result implies conventional approach to find the highly spin-polarized material based on merely P_{sp} value will exclude many prospective candidates. Note that we also found that almost all potential candidates except $\text{Co}_2\text{Fe}_{0.8}\text{Mo}_{0.2}\text{As}_{0.2}\text{Sb}_{0.8}$ have negative formation energy, which confirm the thermodynamical stability of most proposed compounds.

In order to understand why the most prospective highly spin-polarized Heusler alloys suggested by Bayesian optimization are Co-based materials, we also evaluated the ground state properties of all candidates by high-throughput calculations. Note that despite calculations of all candidates (73440 system) were conducted, only 41612 calculations (~56%) converged and obtained the solution. In FIG. 3(a)-(c), we show the distributions of converged calculation results for ground state P_{sp} and P_{sp} values mapped on two-dimensional plane over A elements,

the number of elements and spin moments at 0 K, respectively. Conventional approach of finding highly spin-polarized material were done by “vertically scanning” over the candidates in the high P_{spd} area (specified by sky blue rectangles). However, as mentioned previously that P_{sp} is more suitable to explain the magnetoresistance effect, here we propose to “horizontally scanning” over the high P_{sp} area (specified by red rectangles). Based on the FIG. 3(a), it is clear that high P_{sp} area is filled by Co-based Heusler alloy. These candidates are mainly consist of quaternary and quinary compound as a consequence of the increase of the number of combinations with increasing the number of elements [see FIG. 3(b)]. It turned out that the significant portions of these candidates also exhibit the large spin moment (4-6 μ_B) at 0 K [see FIG. 3(c)] which may imply high Curie temperature and therefore the robustness of spin polarization of Co-based Heusler alloy at finite temperature [26].

In order to understand the importance of including the finite temperature effect, we picked some ternary compound with relatively large magnetic moment (3-6 μ_B) and demonstrated $P_{sp} > 70\%$ at 0 K and plotted the P_{sp} at finite temperature in the FIG. 4. We show P_{sp} of Co_2CrAl (3.00 μ_B), Fe_2MnP (4.00 μ_B) and Ru_2MnSb (4.00 μ_B) as representative cases of the large temperature dependence of P_{sp} . These materials show P_{sp} larger than 70% and almost half-metallic electronic structures at 0 K. However, P_{sp} of these compounds around 300 K is smaller than 50%. On the other hand, Co_2MnAs (5.98 μ_B) and Co_2FeIn (5.09 μ_B) show slow decay of P_{sp} with increasing temperature, preserving high spin-polarization over 70% around 300 K. These results indicate that the inclusion of finite temperature effect in the first-principles calculations is inevitable to find prospective Heusler alloys with machine learning. Note that in the FIG.4, one may find spin polarization for Co_2CrAl and Ru_2MnSb practically reduced to zero at the corresponding calculated Curie temperature. Due to the mean-field approximation, it is clear that the calculated Curie temperature is overestimated. However, we confirm that for the Co-based Heusler alloys, calculated Curie temperature is proportional with both experimental Curie temperature and magnetic moment (not shown here), therefore this DLM method is still adequate to discuss the finite temperature properties.

Then, we systematically investigated the combinatorial system for 4 prospective candidates ($\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$, Co_2MnAs , $\text{Co}_2\text{FeAl}_{0.4}\text{Sn}_{0.6}$, Co_2FeAl) to understand the relation between electronic structure and temperature dependence of spin polarization. In FIG. 5(a)-(f), we show the temperature dependence of P_{spd} and P_{sp} of $\text{Co}_2\text{MnGa}_y\text{As}_{1-y}$, and the density of states (DOS) for total and sp states, and P_{spd} and P_{sp} around the Fermi level.

Recently, Co_2MnGa was reported to show giant anomalous Nernst effect in the L2_1 structure [27]. Meanwhile, theoretical calculations of Co_2MnAs suggest a very large magnetic moment of $6 \mu_B$ [28]. However, both of these compounds and its combinatorial alloy are not widely explored in terms of spin polarization and its application in the magnetoresistance. Thus, $\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$ (and $\text{Co}_2\text{FeAl}_{0.4}\text{Sn}_{0.6}$) obtained in this study by the Bayesian optimization with finite temperature calculations are new materials having high P_{sp} around room temperature. As shown in the FIG. 5(a), despite the pretty high value of P_{spd} of Co_2MnAs at 0 K, Ga-doping improves it further. This can be explained by considering the Fermi level tuning from Co_2MnAs (near conduction band edge) to the Co_2MnGa (near valence band edge) [see FIG. 5(b)-(c)]. Similarly, the effect of Ga-doping is also observed in P_{sp} [see FIG. 5(d)-(f)], which suggests $\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$ and $\text{Co}_2\text{MnGa}_{0.4}\text{As}_{0.6}$ have superior spin polarization among the series. Note that despite the significant shift of Fermi level for y from 0.0 to 0.8 for $\text{Co}_2\text{MnGa}_y\text{As}_{1-y}$, the temperature dependence of P_{sp} does not change depending on y . This is because significant increase of minority spin states happens at the conduction band rather than the valence band edge. For $y = 0.0$, Fermi level is already quite distant from the conduction band edge, therefore shifting further from the conduction band edge as increasing y does not affect the temperature dependence that much. This phenomenon is also observed experimentally in the $\text{Co}_2\text{MnAl}_y\text{Si}_{1-y}$ with $y < 0.4$ by Sakuraba *et al.* [29].

FIG. 6 (a) shows the temperature dependence of P_{spd} for $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$, (b) spd electronic structure at 0 K, (c) energy dependence of P_{spd} calculated at 0 K, and (d)-(f) its counterpart for P_{sp} and sp electronic structure. As shown in FIG. 6(a)-(c), the P_{spd} value and spd electronic structure of Co_2FeAl over significant temperature range fail to explain the giant tunneling magnetoresistance (TMR) demonstrated especially in the $\text{Co}_2\text{FeAl}/\text{MgO}$ based MTJ [30]. It was widely understood that giant TMR effect in Fe/MgO-based MTJ is due to the spin-filtering effect of single crystalline MgO in which Δ_1 symmetry Bloch states at in-plane k -vector $\vec{k}_{||}=(0,0)$ mainly propagates for one spin channel only because of the half-metallic character of the Δ_1 band in bcc-Fe [31]. Due to the fact that s , p_z , d_{z^2} atomic orbitals are compatible to the Δ_1 symmetry, here we can roughly approximate strength of spin filtering effect as spin polarization considering sp electron. That explains why TMR effect could be properly described by P_{sp} behavior instead of very low value of P_{spd} [see FIG. 6(a) and (d)] which consistent with the previous studies on Heusler alloy/MgO based MTJ [21,22]. Our calculation shows that P_{sp} exhibits very low temperature dependence but still retain high value of spin

polarization for Co_2FeAl which also consistent with estimated spin polarization from Julliere's model of TMR effect of $\text{Cr}/\text{Co}_2\text{FeAl}/\text{MgO}/\text{CoFe}$ MTJ [32]. The *sp* electronic structure and energy dependence of spin polarization at 0 K shown in the FIG. 6(e)-(f) indicated that shifting the Fermi level further enough from conduction band edge will lead to lower temperature dependence. Mixing the Al with Sn will shift the Fermi level position toward to the conduction band edge, resulting in lower spin polarization and stronger temperature dependence for high Sn-content composition. However, for $0.4 \leq y \leq 1.0$, the high spin polarization and small temperature dependence is still retained.

In summary, we successfully performed Bayesian optimization combined with the finite temperature calculation to find the highly spin-polarized Heusler alloys around room temperature. We found $\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$ and $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$ ($0.4 \leq y \leq 1.0$) can show high *sp* spin-polarization at around 300 K. Our study emphasized the importance of P_{sp} instead of P_{spd} value to explain the magnetoresistance effect, and the alloy mixing to find the more prospective candidate with 4- or 5-element based compound. However, most candidate are Co-based Heusler alloys, which is supported by the results from high-throughput screening. We also investigated two combinatorial series $\text{Co}_2\text{MnGa}_y\text{As}_{1-y}$ and $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$ to understand the effect of alloy mixing on the temperature dependence and superiority of $\text{Co}_2\text{MnGa}_{0.2}\text{As}_{0.8}$ and $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$ ($0.4 \leq y \leq 1.0$) compared to other Co-based Heusler alloys. $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$ has good lattice matching with fcc-Ag and is promising as a ferromagnetic electrode for CPP-GMR with Ag spacer. Furthermore, $\text{Co}_2\text{MnGa}_y\text{As}_{1-y}$ is expected to be an effective spin injection source into GaAs semiconductors because it is insensitive to diffusion of Ga and As. These results confirmed the importance of distancing Fermi level position from conduction band edge via alloy mixing to improve the temperature dependence of spin polarization.

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TABLE I. Summary of potential highly spin-polarized Heusler alloys at finite temperature suggested by Bayesian optimization.

System	a (Å)	m_{total} (μ_{B})	T_{conv} (K)	P_{spd} (%)	P_{sp} (%)	T_{conv} $\cdot P_{\text{sp}}$	E_{form} (eV/f. u.)
Co ₂ Fe _{1.0} Al _{1.0}	5.700	4.86	352	39.0	86.7	30521	-1.71
Co ₂ Fe _{1.0} Ga _{1.0}	5.720	4.94	329	25.1	82.4	27134	-3.11
Co ₂ Fe _{1.0} In _{1.0}	5.980	5.10	305	11.4	79.8	24347	-3.38
Co ₂ Mn _{1.0} As _{1.0}	5.796	5.96	278	46.0	70.8	19684	-0.71
Co ₂ Mn _{1.0} P _{1.0}	5.638	5.67	223	4.6	77.1	17230	-1.79
Co ₂ Mn _{1.0} Si _{1.0}	5.630	5.00	299	79.0	88.0	26326	-2.44
Co ₂ Fe _{1.0} Al _{0.4} Sn _{0.6}	5.876	5.29	334	-1.7	85.0	28419	-3.09
Co ₂ Fe _{1.0} Ga _{0.2} Ge _{0.8}	5.736	5.32	280	-23.7	76.9	21485	-1.61
Co ₂ Fe _{1.0} Ga _{0.2} In _{0.8}	5.928	5.06	310	15.3	80.4	24920	-3.18
Co ₂ Fe _{1.0} Ga _{0.4} Ge _{0.6}	5.732	5.26	322	-7.4	82.4	26502	-2.00
Co ₂ Fe _{1.0} Ga _{0.4} In _{0.6}	5.876	5.03	315	18.7	80.9	25476	-3.04
Co ₂ Fe _{1.0} In _{0.4} Sn _{0.6}	5.988	5.40	324	-2.5	83.3	27009	-4.00
Co ₂ Mn _{1.0} Al _{0.2} As _{0.8}	5.777	5.61	300	66.8	89.0	26681	-0.97
Co ₂ Mn _{1.0} Al _{0.2} Ge _{0.8}	5.728	4.80	212	74.8	83.1	17589	-1.48
Co ₂ Mn _{1.0} Al _{0.2} Sb _{0.8}	5.954	5.55	276	60.5	87.8	24250	-0.26
Co ₂ Mn _{1.0} Al _{0.2} Si _{0.8}	5.644	4.80	237	80.5	88.2	20909	-2.29
Co ₂ Mn _{1.0} Ga _{0.2} As _{0.8}	5.781	5.61	291	65.1	88.4	25711	-1.23
Co ₂ Mn _{1.0} Ga _{0.2} Ge _{0.8}	5.731	4.81	208	73.3	82.2	17088	-1.75
Co ₂ Mn _{1.0} Ga _{0.2} Sb _{0.8}	5.958	5.56	268	58.2	86.8	23254	-0.45
Co ₂ Mn _{1.0} Ga _{0.4} As _{0.6}	5.765	5.20	264	71.2	86.4	22836	-1.73
Co ₂ Mn _{1.0} Ga _{0.4} Sb _{0.6}	5.898	5.21	246	67.6	83.6	20525	-0.89
Co ₂ Ti _{0.2} Mn _{0.8} Ge _{1.0}	5.756	4.39	216	66.7	88.0	18998	-1.62
Co ₂ Fe _{0.8} Mo _{0.2} As _{0.2} Sb _{0.8}	5.979	4.60	267	37.4	81.1	21680	0.60
Co ₂ Fe _{0.8} Mo _{0.2} In _{0.2} Sb _{0.8}	6.037	4.80	264	44.9	82.4	21737	-0.13
Co ₂ Fe _{0.8} Mo _{0.2} Sn _{0.4} Sb _{0.6}	6.039	4.78	265	47.0	83.2	22002	-1.27
Co ₂ Fe _{0.8} Nb _{0.2} In _{0.6} Sb _{0.4}	6.032	4.81	244	31.2	80.6	19692	-1.70
Co ₂ Mn _{0.8} Fe _{0.2} Ge _{0.4} Sb _{0.6}	5.902	5.51	230	-7.0	75.4	17302	-0.20
Co ₂ Mn _{0.8} Fe _{0.2} In _{0.4} Sb _{0.6}	6.001	5.40	270	54.4	83.5	22585	-1.40
Co ₂ Mn _{0.8} Fe _{0.2} Sn _{0.6} Sb _{0.4}	6.000	5.54	275	45.9	86.3	23748	-2.79
Co ₂ Mn _{0.8} Nb _{0.2} Ge _{0.2} As _{0.8}	5.773	4.90	226	74.9	89.0	20158	-0.63

$\text{Co}_2\text{Mn}_{0.8}\text{Nb}_{0.2}\text{Ge}_{0.4}\text{As}_{0.6}$	5.763	4.78	207	76.8	88.0	18251	-0.77
$\text{Co}_2\text{Mn}_{0.8}\text{Zr}_{0.2}\text{P}_{0.2}\text{Ge}_{0.8}$	5.760	4.50	203	37.8	83.1	16836	-1.22

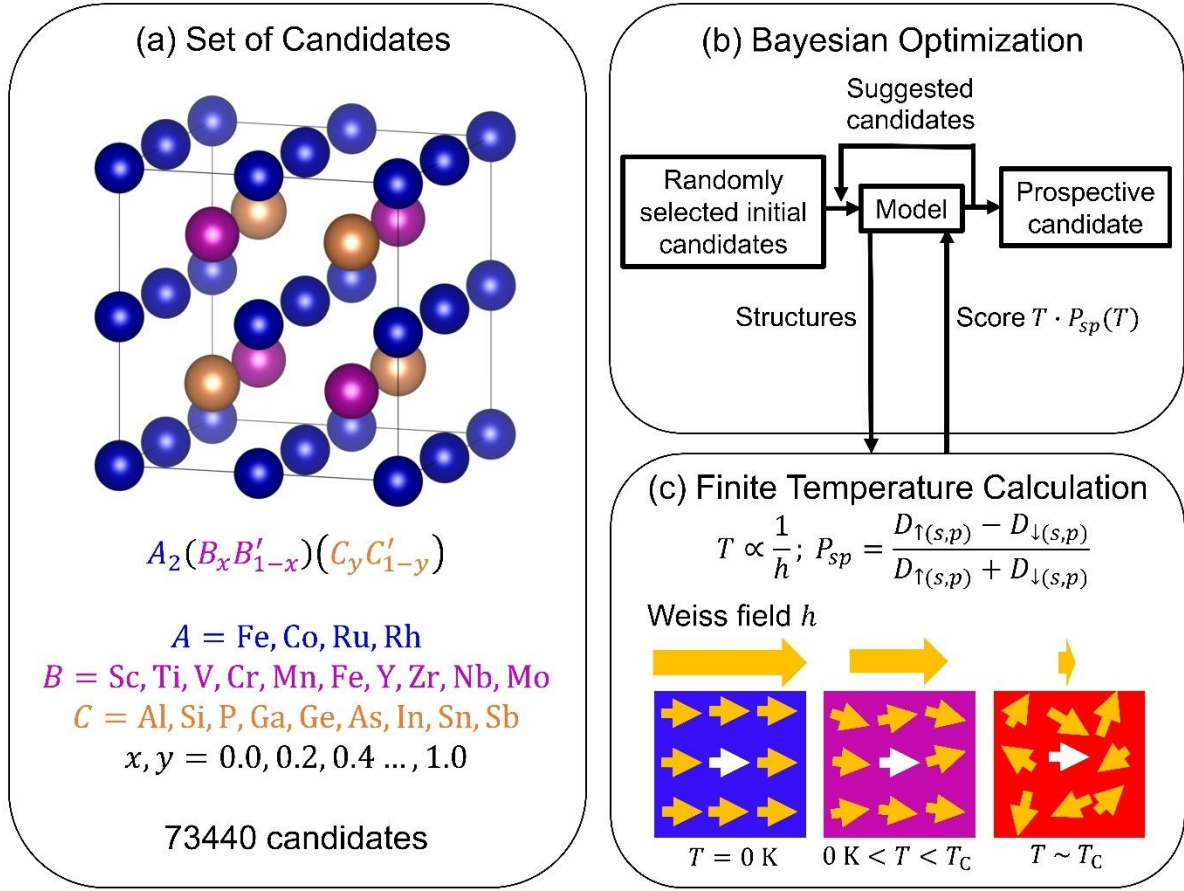


FIG. 1. The schematic workflow for finding prospective candidate with highly spin-polarized Heusler alloys at finite temperature. (a) Set of candidates to be investigated in this study, (b) implemented Bayesian optimization procedure to find the prospective candidate, (c) Finite temperature calculation to obtain the sp spin polarization at converged temperature T using starting parameter of Weiss field h .

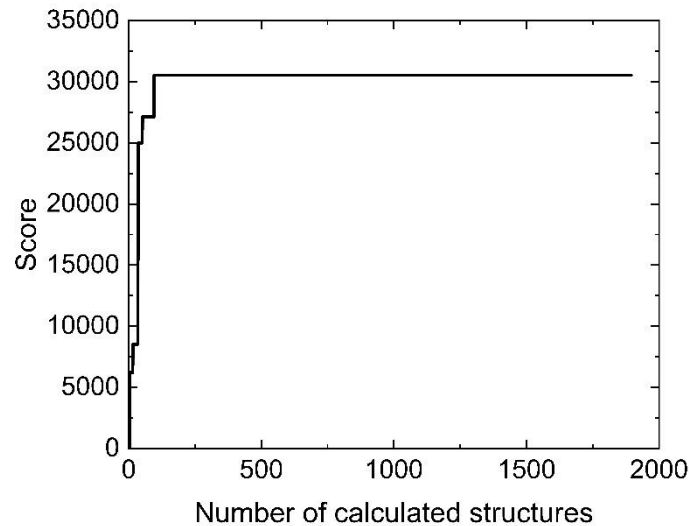


FIG. 2. Bayesian optimization performance for the calculated Heusler structures

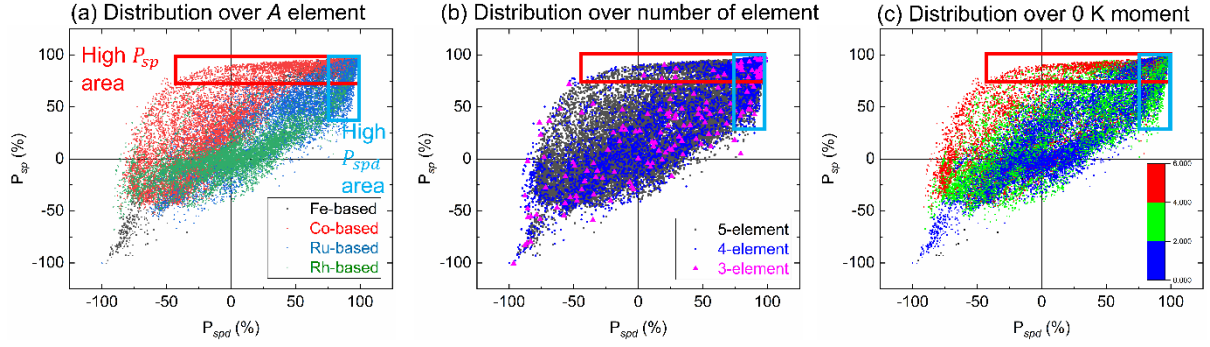


FIG. 3. The results of high-throughput calculation for ground state properties of all candidates as (a) distribution over A element, (b) distribution over number of element, and (c) distribution over 0 K moment.

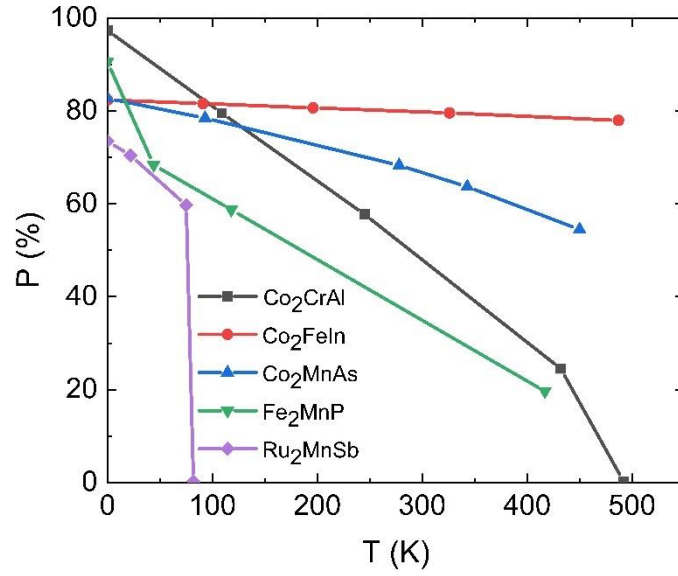


FIG. 4. The temperature dependence of P_{sp} of some ternary Heusler alloys that demonstrated large magnetic moment (3-6 μ_B) and spin polarization > 70 % at 0 K.

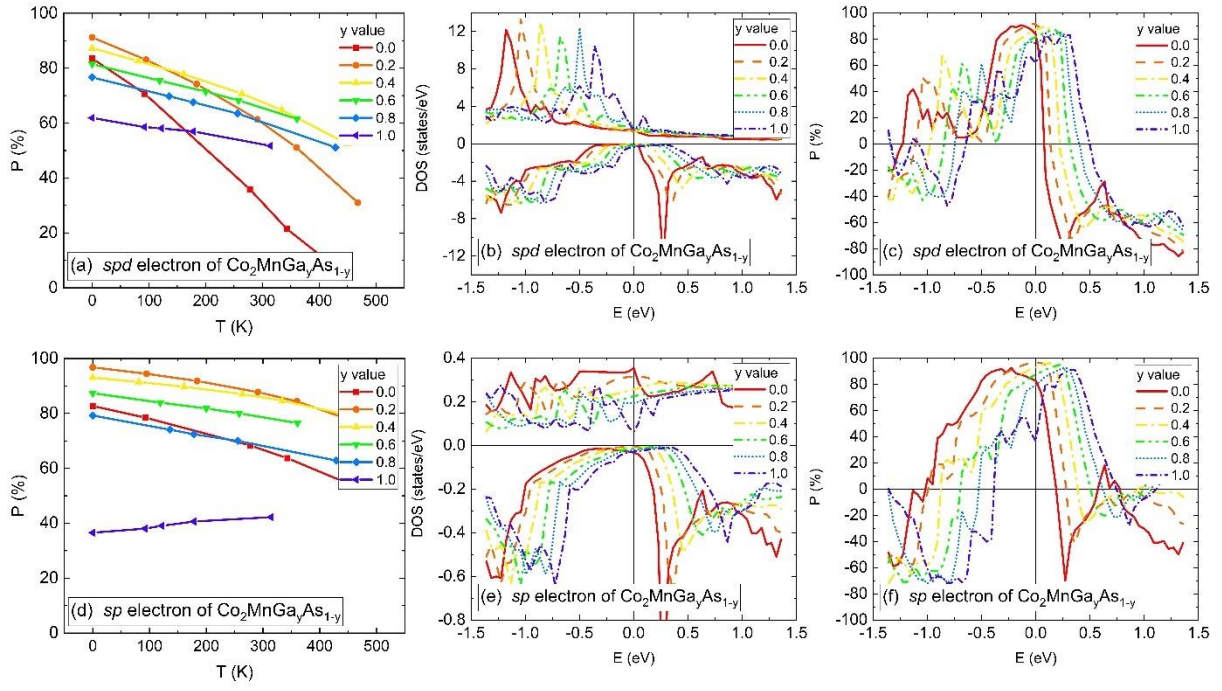


FIG. 5. (a) The temperature dependence of P_{spd} on value of y from $\text{Co}_2\text{MnGa}_y\text{As}_{1-y}$, (b) spd electronic structure at 0 K, (c) energy dependence of P_{spd} calculated at 0 K, and (d)-(f) its counterpart for P_{sp} and sp electronic structure. The reference of the energy E is the Fermi energy.

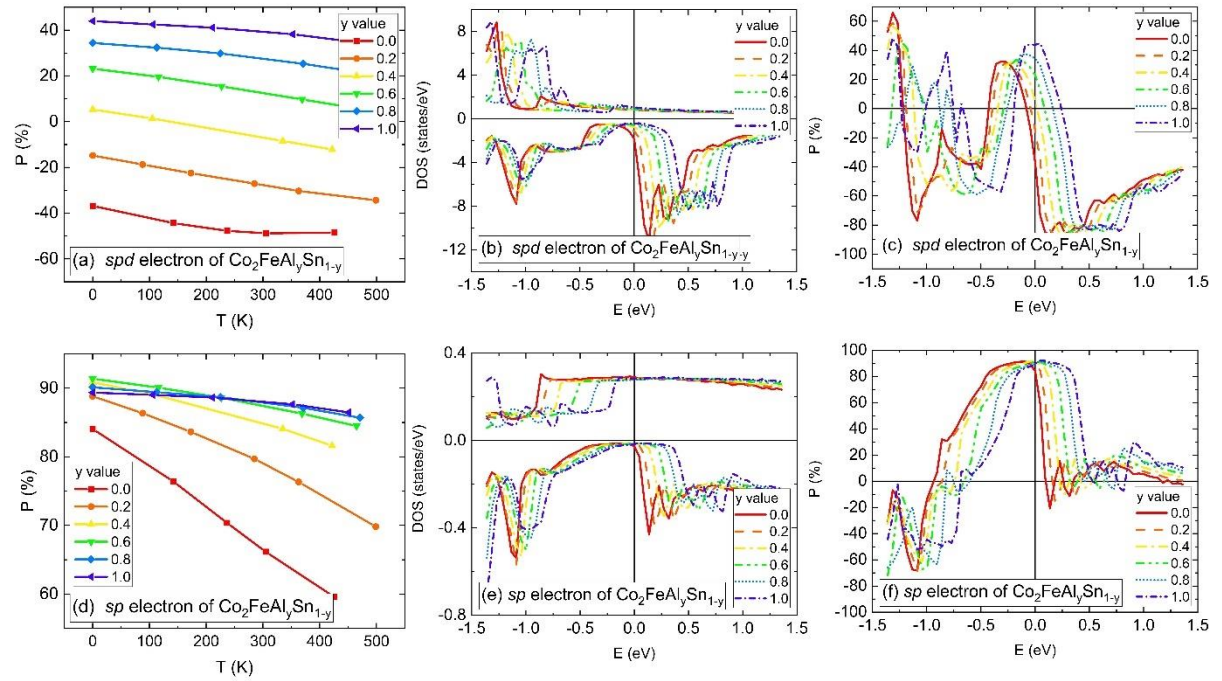


FIG. 6. (a) The temperature dependence of P_{spd} for $\text{Co}_2\text{FeAl}_y\text{Sn}_{1-y}$, (b) spd electronic structure at 0 K, (c) energy dependence of P_{spd} calculated at 0 K, and (d)-(f) its counterpart for P_{sp} and sp electronic structure. The reference of the energy E is the Fermi energy.