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Autonomous search for materials with high Curie temperature using *ab initio* calculations and machine learning

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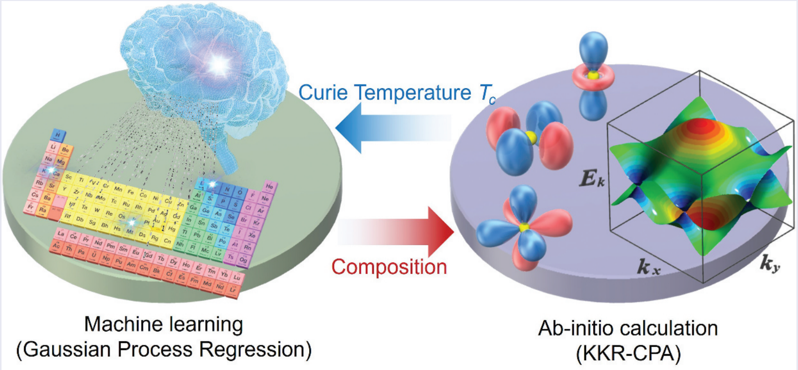
ABSTRACT

Efficient exploration of vast material spaces is a challenging task in materials science. Autonomous material search methods utilizing machine learning and ab initio calculations have emerged as powerful alternatives to traditional material discovery through synthesis and analysis, which is time-consuming and scope-limited. Although autonomous search methods have already been applied to various material spaces, they have not explored the extensive material space of Curie temperatures. Herein, we show a simulation-based autonomous search method that suggests ternary alloys with high Curie temperatures. The material space – consisting of disordered ternary magnetic alloys – is explored through Korringa – Kohn – Rostoker coherent potential approximation and Bayesian optimization. Over a continuous 10-day search, the system proposed several alloys – CoAuIr, CoPtPd, and CoFeBi – with Curie temperatures surpassing that of pure face-centered cubic Co. Although the insights gained through these predictions require further experimental and theoretical validation, this study demonstrates that autonomous material search methods can potentially accelerate material discovery and optimize material properties.



IMPACT STATEMENT

Autonomous material search accelerates discovery of ternary alloys with high Curie temperatures, surpassing pure Co, using machine learning and ab initio calculations.

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1. Introduction

Material spaces have recently become extremely vast. Traditionally, vast material spaces have been methodically searched by humans through repeated cycles of material synthesis, property measurement, and analysis. Traditional searching is time-consuming and cannot sufficiently explore the recently expanded material spaces.

To resolve the concerns of traditional searching, researchers have incorporated machine learning into autonomous material search methods. These methods can be divided into two predominant technologies: autonomous searching based on robotics and autonomous searching through material simulations. In the robotics approach, the material synthesis and property measurement tasks are automated by robots. Subsequently, machine learning analyzes the obtained material data and suggests the materials and process conditions for subsequent synthesis. The synthesis and measurement of the proposed materials by a robot forms a closed loop, enabling autonomous material search. Because many autonomous material search

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robots have already been developed, robotics searching is among the most powerful methods in material development [1–12]. In the second method, simulations such as ab initio calculations obtain the material property data, which are analyzed using machine learning to determine the structure and composition of the subsequently simulated materials. Moreover, this process forms a closed loop allowing autonomous search of material spaces within the computer and is convenient, versatile, and popular [13–20].

Herein, we explore the material space of high-Curie-temperature alloys using simulation-based autonomous material search methods. The Curie temperature is the transition temperature at which a ferromagnetic material becomes paramagnetic. In general, a higher Curie temperature is advantageous for practical applications. Face-centered cubic (FCC)-structured Co (FCC-Co) has the highest Curie temperature among known materials, with an experimental Curie temperature of 1,388 K [21]. Theoretically, BCC-FeCo exhibits a higher Curie temperature than FCC-Co [22]. However, the BCC structure of FeCo transforms into an FCC structure at high temperatures. Therefore, the highest experimentally observed Curie temperature remains that of FCC-Co [22].

By expanding the material search space to binary and ternary alloys, alloys with Curie temperatures higher than those of Co might be discovered. However, as the number of elements increase, combinatorial explosion of the material search space expands and inhibits the comprehensive investigation of Curie temperatures. To seek multielement alloy materials with high Curie temperatures within a vast combinatorial space, we employed machine learning and ab initio calculations. Fukuzawa et al. demonstrated the effectiveness of machine learning – based autonomous exploration on a dataset of Curie temperatures [23]. Herein, we apply a similar scheme to the practical exploration of materials; in particular, we autonomously search a vast materials space for new alloys with Curie temperatures surpassing that of FCC-Co.

2. Methods

The material space was defined as disordered ternary magnetic alloys composed of three elements selected from the set (Al, Si, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, Y, Zr, Nb, Mo, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Hf, Ta, W, Re, Os, Ir, Pt, Au, Hg, Tl, Pb, Bi). To reduce the number of candidate materials, the exploration was intentionally limited to materials containing Fe, Co, or Ni, which are dominant in typical alloys with high Curie temperatures. Because such alloys adopt an FCC structure at high temperatures, the search was further restricted to materials exhibiting an FCC structure. Approximately 10 million potential material configurations existed within the defined material space. The challenge of performing ab initio calculations for all candidates was alleviated through sequential calculations guided by machine learning, which can efficiently search the material space.

Figure 1 illustrates the autonomous materials – search methodology, which autonomously searches for materials through sequential ab initio calculations and machine learning. The ab initio calculation phase calculates the Curie temperature T_c based on the composition information provided by the machine learning phase. Using the accumulated T_c data, this phase informs the composition for subsequent ab initio calculations.

The ab initio calculation phase performs Green's function-based density functional theory calculations using the Korringa – Kohn – Rostoker coherent

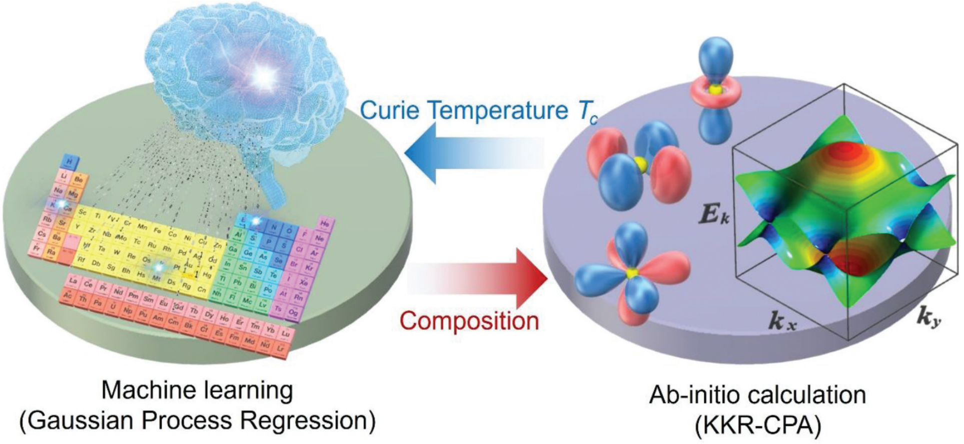

Figure 1. Overview of an autonomous material search system: *ab initio* calculation phase calculates the Curie temperature T_c based on the crystal structure and composition information determined in the machine learning phase. Machine learning phase derives the compositional information for the subsequent *ab initio* calculation (KKR – CPA = Korringa – Kohn – Rostoker coherent potential approximation). This method is based on previous research [19].

potential approximation (KKR – CPA) method implemented in AkaiKKR software [24]. CPA enables accurate simulations of alloy systems and is particularly effective for multielement disordered phases [25–28]. The Curie temperature T_c was estimated as follows:

$$T_c = \frac{2}{3}\frac{\left(E_{fmg} - E_{lmd}\right)}{ck_B},$$

where E_{fmg} and E_{lmd} denote the total energies of the ferromagnetic and local moment disorder states, respectively [29], k_B is the Boltzmann constant, and c is the concentration of magnetic atoms. The lattice constants were optimized to minimize the total energy. The ab initio calculations are detailed in the Supplementary Materials (S1).

The machine learning phase uses Bayesian optimization [30] and an autoencoder [31] to determine the material composition for subsequent KKR – CPA calculations based on the accumulated T_c data. The material space (descriptors) was designed based on previous documentation [19]. Briefly, a composition vector and Magpie descriptor vector [32] were compressed into a 15-dimensional latent vector representing the yet to be explored material space. Further details can be found in the Supplementary Materials (S2). To explore this material space, the KKR – CPA method was combined with Bayesian optimization using T_c as the objective variable and the latent vectors generated by the autoencoder as the explanatory variables in the Gaussian-process regression model. The target of the next KKR – CPA calculation was selected using the upper confidence bound (UCB) as an acquisition function. This approach (detailed in S3 of the Supplementary Materials) enables the autonomous search of materials with high T_c.

3. Results and discussion

The developed autonomous search system ran continuously for approximately 10 days. Figure 2 shows the progression of Curie temperatures calculated during the autonomous search. The initial 10 trials (gray area in Figure 2) were random searches preceding autonomous exploration, in which no alloys with particularly high Curie temperatures were predicted. Several alloys with sufficiently high Curie temperatures were predicted when the volume of training data increased and the accuracy of the learning model improved. Fe- and Ni-based alloys were primarily explored in the early stages, but around the 35th search – when Co inclusion enhanced the Curie temperature – Co-based alloys became the primary focus of exploration.

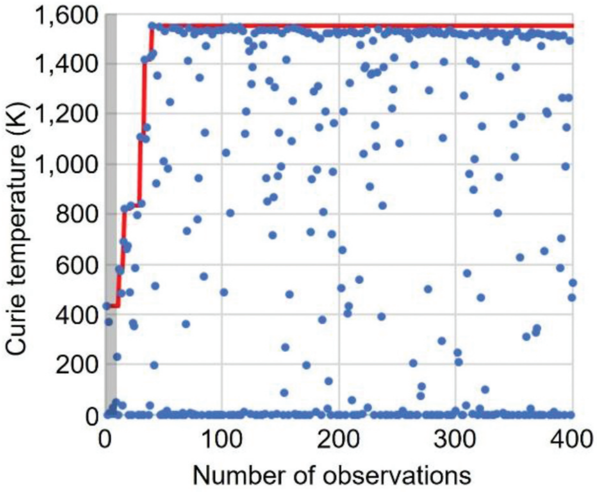

Figure 2. Result of the autonomous search for materials with high Curie temperatures: blue circular plots represent the kkr–CPA-calculated Curie temperatures during the exploration, and the red solid line indicates the highest values thus far obtained. The initial 10 trials (highlighted in the gray area) were random searches followed by autonomous exploration.

After approximately 100 autonomous searches, the system predominantly targeted the CoAuIr, CoPtPd, and CoFeBi ternary alloy systems. These ternary alloys were subsequently processed using high-throughput KKR – CPA-based ab initio calculations. The obtained Curie temperatures are shown in Figure 3. Notably, the Curie temperatures of a few compositions are predicted to exceed those of pure FCC-Co. The material with the highest Curie temperature (1,523.9 K) is predicted to be $Co_{0.94}Au_{0.06}$. A material with a similar

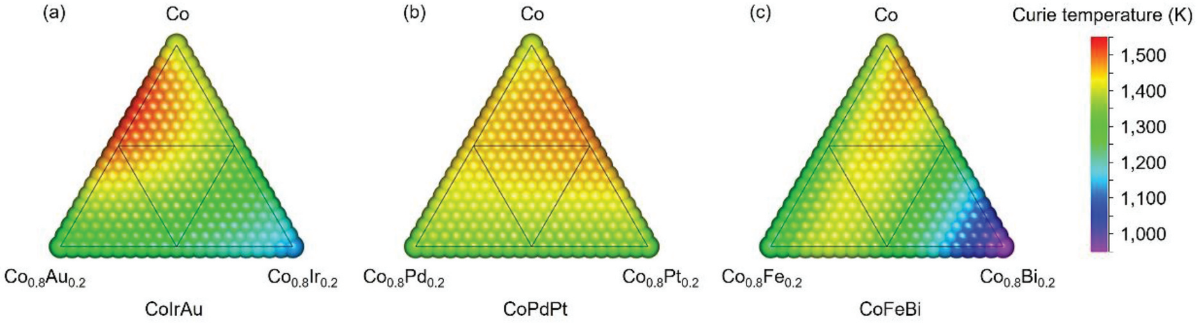

Figure 3. Results of the high-throughput ab initio calculations on ternary alloys frequently explored by the autonomous materials – search system: (a) CoIrAu, (b) CoPdPt, and (c) CoFeBi ternary alloys; Curie temperatures of some compositions exceed those of pure FCC-Co (1,388 K).

composition—$Co_{0.95}Au_{0.05}$—was suggested during the early stages of the autonomous search, specifically, at the 40th search iteration.

To investigate the proposed materials exhibiting high Curie temperatures, KKR – CPA calculations were performed on the binary Co-based alloys $Co_{1-x}X_x$ (X = Au, Ir, Pd, Pt, Fe, Bi, Ta, and W). The autonomous search system intensively explored Au, Ir, Pd, Pt, Fe, and Bi but less frequently explored Ta and W. Figure 4(a) presents plots of Curie temperature as a function of composition for each dopant element X. The Curie temperature of Co increased after adding small amounts of Au, Ir, Pd, Pt, and Bi but monotonically decreased after adding Ta and W as well as Fe (although more gradually). As the autonomous exploration progresses and the volume of training data increases, the accuracy of the constructed machine learning models is expected to improve, resulting in the exclusion of Fe from the search targets.

Figure 4(b) presents plots that demonstrate the dopant composition dependences of the total magnetic moments in $Co_{1-x}X_x$. The KKR – CPA calculations detected ferrimagnetism in $Co_{1-x}Ta_x$, $Co_{1-x}W_x$, and $Co_{1-x}Bi_x$ and ferromagnetism in $Co_{1-x}Au_x$, $Co_{1-x}Ir_x$, $Co_{1-x}Pd_x$, $Co_{1-x}Pt_x$, and $Co_{1-x}Fe_x$. Adding small amounts of Au, Ir, Pd, Pt, and Bi enhanced the total magnetic moment from that of Co alone but after adding Ta or W, the total magnetic moment monotonously decreased with dopant concentration, similarly to the Curie temperature. In contrast, adding Fe to Co increased the total magnetic moment while decreasing the Curie temperature.

The local magnetic moment of Co shows a more distinct correlation with the Curie temperature than with the total magnetic moment. As shown in Figure 4(c), the local magnetic moment of Co considerably increased after adding Au, Ir, Pd, Pt, and Bi (which also improved the Curie temperature at low concentrations) but negligibly increased after adding Ta, W, and Fe (which reduced the Curie temperature). These trends suggest a close relationship between the Curie temperature and the local magnetic moment of Co.

The magnetic moment critically depends on the lattice constant. Theoretically, increasing the lattice constant improves the magnetic moment of Co [33]. As shown in Figure 4(d), the addition of any element increased the lattice constant of Co, indicating that enhancement of the magnetic moment is not solely determined by the lattice constant. For example, adding Ta or W increased the lattice constant but decreased the magnetic moment. Therefore, the enhancement of the Curie temperature and magnetic moment after adding Au, Ir, Pd, Pt, and Bi to Co cannot be attributed solely to the increase in lattice constant. Contributions from other factors must be

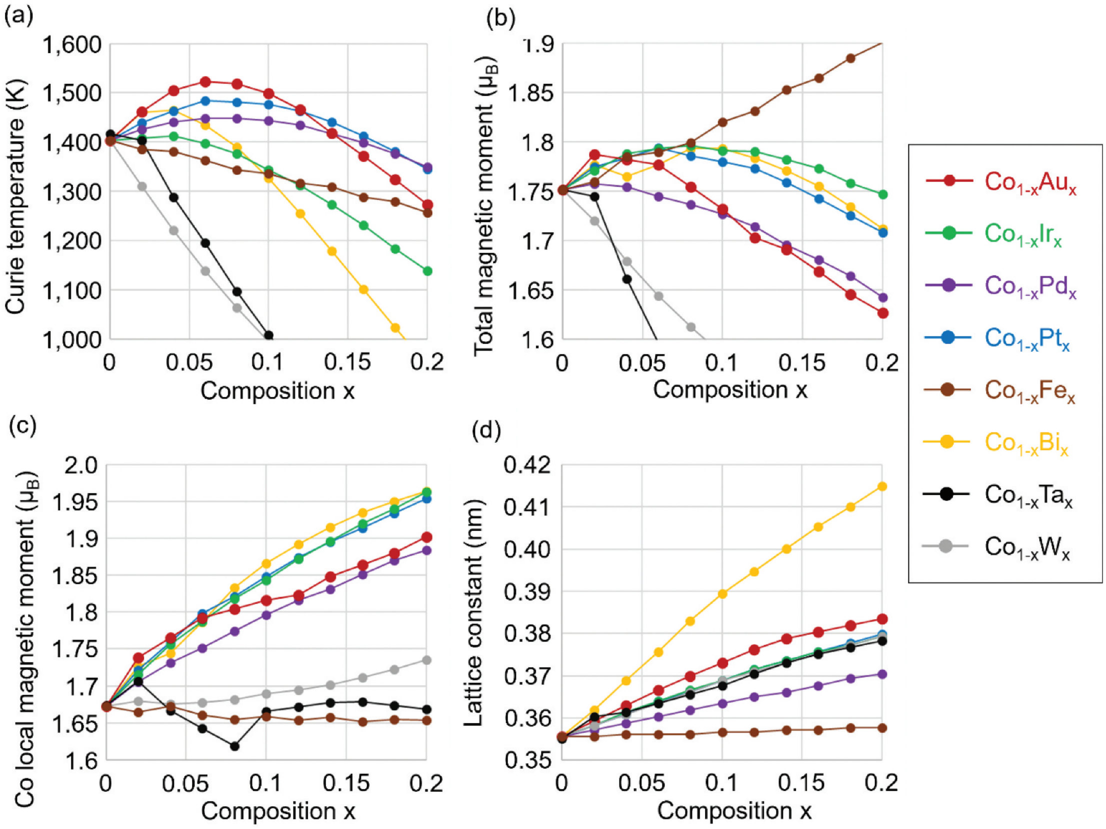

Figure 4. Dopant concentration dependences of (a) Curie temperature, (b) total magnetic moment, (c) Co local magnetic moment, and (d) lattice constant in the $Co_{1-x}X_x$ binary alloys (X = Au, Ir, Pd, Pt, Fe, Bi, Ta, and W). All results were calculated by KKR – CPA.

elucidated through further experimental and theoretical investigations.

This study successfully applied autonomous search methods for predicting new alloys with high Curie temperatures. However, we emphasize that the proposed new alloy materials are merely ‘predictions’ based on ab initio calculations, and the accuracy of the Curie temperature calculations has not been verified. Additionally, it is unclear whether the elements in the proposed alloys will form a stable disordered FCC structure at high temperatures. For example, CoFeBi contains an element (Bi) with a low melting point; thus, it can melt at high temperatures. Therefore, further experimental and theoretical investigations of the proposed alloy materials are essential.

4. Conclusions

The simulation-based autonomous material search method effectively explores the vast alloy space of materials with high Curie temperatures. By integrating machine learning with ab initio calculations, this approach efficiently navigates the complex material landscape, leading to the prediction of several ternary alloys with Curie temperatures that exceed those of pure FCC-Co. This serves as a clear demonstration of the potential of autonomous material search methods. Furthermore, this method is highly versatile and can be adapted to various material systems and properties, offering significant potential to accelerate progress across the entire field of material discovery.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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Data availability statement

The data supporting the results of this research are available from the corresponding author upon reasonable request.

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