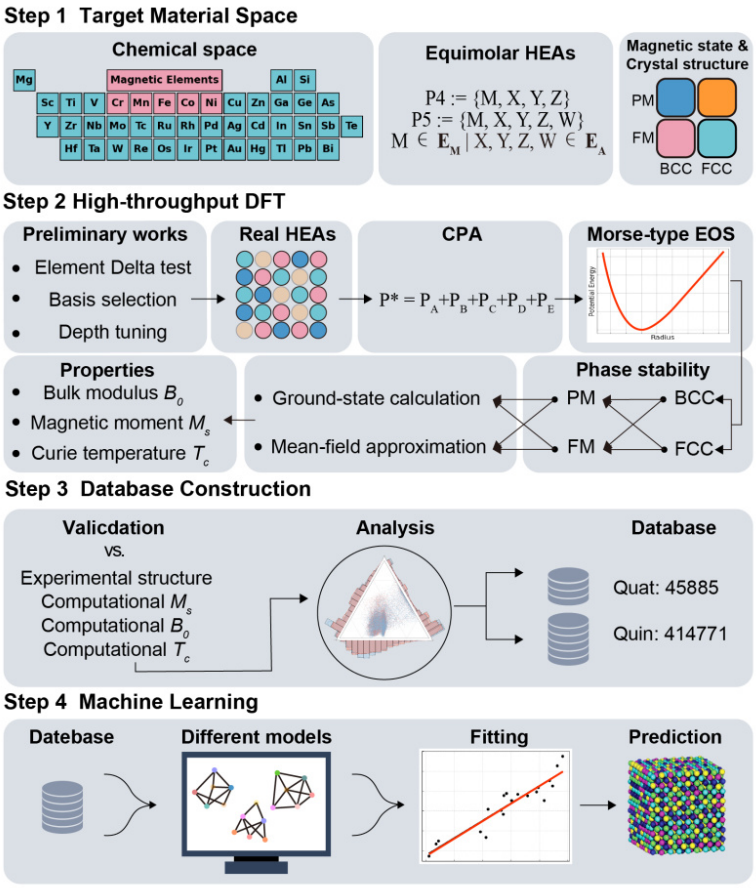
Hessisches Kompetenzzentrum
für Hochleistungsrechnen

High-Throughput Design of High Entropy Alloys



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Project Term
2023 - 2024

Clusters
Lichtenberg II Cluster Darmstadt

Additional Software
EMTO-CPA

Institute
Theory of Magnetic Materials

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Introduction

The quest for advanced materials that simultaneously exhibit high mechanical hardness and soft magnetic behavior poses a significant challenge in materials science. Such multifunctional properties are essential for next-generation technologies in energy, electronics, and transportation. Exploring the vast compositional landscape of high-entropy alloys (HEAs), which offer immense chemical design flexibility, requires computational strategies that go beyond traditional trial-and-error approaches. In this work, we leverage high-performance computing (HPC) to conduct over 1.8 million density functional theory (DFT) calculations across quaternary and quinary equimolar HEAs constructed from 42 elements.

Methods

We employed a high-throughput density functional theory (HTP-DFT) workflow to evaluate the structural, mechanical, and

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magnetic properties of equimolar HEAs. All calculations used the generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) form [1] to describe exchange-correlation effects. The electronic structure was solved using the exact muffin-tin orbital (EMTO) method [2, 3], combined with the coherent potential approximation (CPA) [4] to treat chemical disorder. Scalar-relativistic effects and a soft-core basis set were included, and total energies were computed using the full charge density technique [5] within an optimized overlapping muffin-tin potential framework.
A dense 17 × 17 × 17 k-point mesh ensured Brillouin zone sampling convergence. Calculations were performed across at least eleven volumes for body-centered cubic (BCC) and face-centered cubic (FCC) structures, and bulk moduli were obtained by fitting the energy-volume data to a Morse-type equation of state [6]. Magnetic properties were evaluated in both ferromagnetic (FM) and paramagnetic (PM) states. The PM state was modeled using the disordered local moment (DLM) approximation, and the Curie temperature TC was estimated using a mean-field approximation (MFA) [7].

Results

1. Interactive database for high-entropy magnets

We developed a comprehensive HEA database comprising 45,885 quaternary and 414,771 quinary equimolar compositions, with an interactive interface to support systematic exploration. High-throughput EMTO-CPA calculations revealed that ferromagnetic BCC phases dominate, especially in quinary systems. Validation against literature confirms high phase classification accuracy, with minor deviations explained by small energy differences and the need for finite-temperature considerations. Element-wise analysis shows that Fe, Co, and Mn favor FM-BCC structures, while Cr and Ni exhibit more diverse phase distributions. Comparison of atomic volumes with the rule of mixtures confirms a general linear trend but highlights the limitations of empirical models. The interactive analysis tool enables stepwise exploration of elemental combinations and predicted properties, greatly enhancing accessibility and supporting targeted HEA design.

2. Multifunctional HEAs

We analyzed the correlation between mechanical and magnetic properties of quaternary and quinary HEAs, revealing that increased elemental diversity leads to broader distributions in bulk modulus (B_0), total magnetic moment (M_{tot}), and Curie temperature (T_C). BCC structures generally exhibit higher B_0 and M_{tot} than FCC counterparts. Elemental contribution analysis identifies Fe, Mn, Co, Cr, and Re as key drivers of desirable properties. Transitioning from quaternary to quinary systems introduces additional influential elements such as Mo, Ru, and Os, enhancing compositional complexity. Binary element interactions also shift, with pairs like Ni–Rh and Co–Rh favoring FCC stability, while Fe–Re, Mn–Re, and Cr–W promote high-performance BCC phases. Screening results highlight 15

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promising HEAs in each category with simultaneously high B_0, M_{tot}, and T_C, offering valuable candidates for further experimental validation.

Discussion

This study establishes a robust data-driven framework that integrates high-throughput DFT calculations with machine learning to design mechanically hard soft magnetic HEAs. By constructing a large database of over 460,000 equimolar HEAs covering 42 elements, we capture complex multi-element interactions and uncover composition–property relationships beyond the reach of empirical models. Our results reveal that increasing compositional diversity enhances the dominance of FM-BCC phases, driven by atomic size effects, valence electron concentration, and lattice distortion mechanisms. Key elements such as Co, Fe, Ni, Mn, Re, and W are identified as central to optimizing magnetic and mechanical performance. The findings emphasize the importance of BCC-phase stabilization and the interplay between intrinsic properties like bulk modulus and microstructural design. Beyond magnetically soft materials, the developed dataset and workflow offer a scalable platform for designing a wide range of functional HEAs, including those for high-temperature and corrosion-resistant applications.

Figures

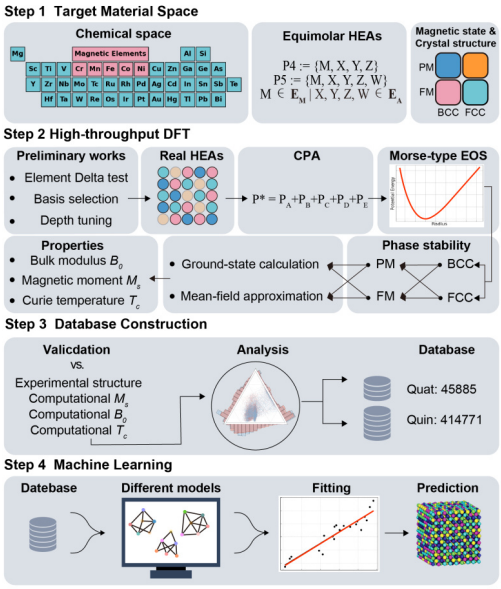


Figure 1: Workflow for HTP exploration and HEA property prediction

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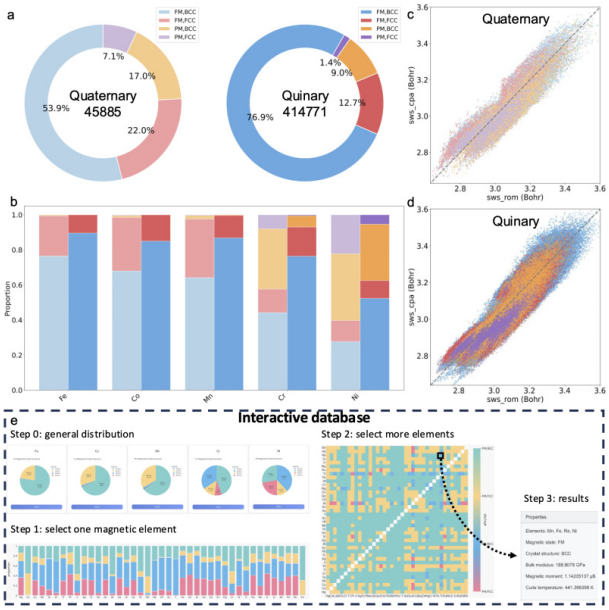


Figure 2: Overview of the HEA database and interactive analysis interface

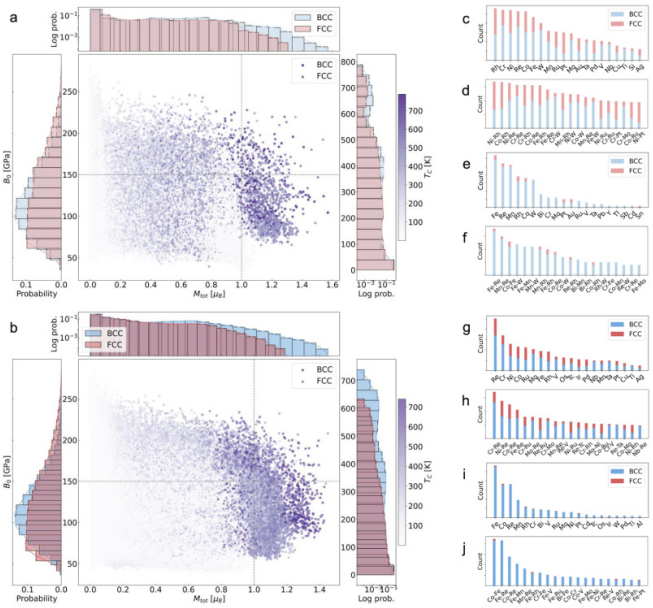


Figure 3: Distribution and correlation of target properties and elemental contributions in the HEA database

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Reference

[1] Perdew, J. P.; Burke, K.; Ernzerhof, M. Generalized Gradient Approximation Made Simple. Physical Review Letters 77.18 (Oct. 1996), pp. 3865–3868. https://doi.org/10.1103/PhysRevLett.77.3865

[2] Vitos, L. Total-energy method based on the exact muffin-tin orbitals theory. Physical Review B 64.1 (June 2001), p. 014107. https://doi.org/10.1103/PhysRevB.64.014107

[3] Vitos, L. Computational Quantum Mechanics for Materials Engineers: The EMTO Method and Applications. Springer, Jan. 2007. https://doi.org/10.1007/978-1-84628-951-4

[4] Vitos, L.; Abrikosov, I. A.; Johansson, B. Anisotropic Lattice Distortions in Random Alloys from First-Principles Theory. Physical Review Letters 87.15 (Sept. 2001), p. 156401. https://doi.org/10.1103/PhysRevLett.87.156401

[5] Vitos, L.; Kollár, J.; Skriver, H. L. Full charge-density scheme with a kinetic-energy correction: Application to ground-state properties of the 4d metals. Physical Review B 55.20 (May 1997), pp. 13521–13527 https://doi.org/10.1103/PhysRevB.55.13521

[6] Moruzzi, V. L.; Janak, J. F.; Schwarz, K. Calculated thermal properties of metals. Physical Review B 37.2 (Jan. 1988), pp. 790–799 https://doi.org/10.1103/PhysRevB.37.790

[7] Körmann, F.; Ma, D.; Belyea, D.; Lucas, M.; Miller, C.; Grabowski, B.; Sluiter, M. Treasure maps for magnetic high-entropy-alloys from theory and experiment. Applied Physics Letters 107 (Oct. 2015), p. 142404. https://doi.org/10.1063/1.4932572

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