Modeling the Unpredictable: AI and Quantum Physics Join Forces on High-Entropy Alloys

Predictive modeling of high entropy alloys integrating first-principles and machine learning approaches

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In Short

- Combination of density-functional theory, *ab initio* molecular dynamics, and machine learning to study the high entropy alloy: AlFeCoNiCu.
- Predictive framework for phase behavior, and mechanical, magnetic, thermal, and diffusion properties.

Multicomponent metallic compounds containing five or more atomic species are classified as high-entropy alloys (HEAs) [1]. This class of materials has gained growing attention following recent experimental reports of outstanding mechanical and catalytic properties, which open doors to innovative industrial applications [2]. The term “high entropy” reflects the complex phase behavior of these alloys (including intermetallic formations, solid solutions, and spinodal decompositions) arising from the high configurational disorder intrinsic to their multi-element composition.

In this context, theoretical modeling of HEAs faces significant challenges. Due to their inherent complexity, a single atomic representative configuration is typically inadequate to reliably capture key properties such as electronic, mechanical, thermal, or transport behavior. These properties depend sensitively on stoichiometry, atomic arrangement, geometry, and temperature.

Classical molecular dynamics (MD), while far less demanding computationally, relies on interatomic force fields that are often tailored to specific systems or properties, and are generally unavailable or insufficient for the diverse compositions present in HEAs. Recent developments in machine learning (ML) offer a promising alternative: ML models can accelerate property prediction and provide machine learning interatomic potentials (MLIPs) that are both accurate and transferable. However, ML approaches for HEAs are still in their early stages and require further development before they can be widely adopted.

In summary, although recent advances in computational methods have made the theoretical exploration of multicomponent alloys feasible, considerable challenges remain. In this project, we will use the AlFeCoNiCu alloy (Fig. 1) as a case study to develop a predictive framework for phase behavior, as well as mechanical, magnetic, thermal, and diffusion properties. This alloy has been selected due to the opportunity for experimental validation through collaboration with partner research groups. Notably, it has demonstrated remarkable potential in sensor applications and excellent mechanical resistance; yet, theoretical studies on its properties remain limited.

Our approach combines DFT and AIMD to generate training data for an MLIP, which will, in turn, enable large-scale MD simulations involving significantly more atoms. We have successfully applied a similar strategy in a recent study on graphene [3], where we developed an efficient MLIP capable of reproducing mechanical and vibrational properties, including stress-strain behavior, elastic constants, phonon dispersion, and vibrational density of states. The computational tools and methodologies from that study will be adapted and extended for the current project. Ultimately, our goal is to establish a robust, transferable workflow that can be applied not only to AlFeCoNiCu but also to other HEAs, laying the groundwork for systematic theoretical investigations of this complex and technologically promising class of materials.

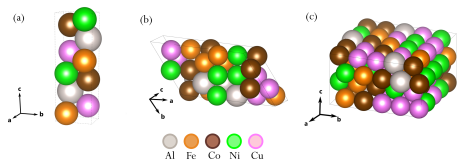

Figure 1: *Illustration of the three geometries with composition $Al_{0.2}Fe_{0.2}Co_{0.2}Ni_{0.2}Cu_{0.2}$ (a) Cmm2 HEA with 10 atoms; (b) R-3m HEA with 30 atoms; (c) Fm-3m HEA with 100 atoms. The color code used to represent atoms is given on the bottom.*

More Information

[1] Easo P. George, Dierk Raabe, and Robert O. Ritchie, *High-entropy alloys*, Nature Reviews Materials **4**, 515–534 (2019). doi: 0.1038/s41578-019-0121-4

[2] Yi-Fei Yang, Feng Hu, Ting Xia, Rui-Han Li, Jun-Yu Bai, Jia-Qi Zhu, Jian-Yi Xu, and

Guo-Fang Zhang, *High entropy alloys: A review of preparation techniques, properties and industry applications*, Journal of Alloys and Compounds, **1010**, 177691 (2025). doi: 10.1016/j.jallcom.2024.177691

[3] Felipe Hawthorne, Paulo R. E. Raulino, Ronaldo Rodrigues Pela, and Cristiano F. Woellner, *Efficient and accurate machine learning interatomic potential for graphene: Capturing stress-strain and vibrational properties*, The Journal of Physical Chemistry C, **Article ASAP** (2025) doi:10.1021/acs.jpcc.5c03470

DFG Subject Area

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