A foundation-model-based approach to crystal structure prediction: application to the Ca–Fe–Ni system under high pressure

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Predicting crystal structures from first principles remains a key challenge in condensed matter, chemistry, and materials science, underpinning the discovery of materials with tailored properties. Although evolutionary algorithms efficiently explore complex energy landscapes, their reliance on *ab initio* calculations for structural relaxation and ranking leads to prohibitive computational costs. Machine learning interatomic potentials (MLIPs) can substantially accelerate crystal structure prediction (CSP), yet their practical integration into CSP workflows remains limited by the need for large, carefully selected training datasets. In particular, one of the most important tasks is how to identify, from the vast number of candidate structures generated during the global search, a compact and informative subset that warrants expensive density functional theory (DFT) calculations for model refinement. Recent advances in foundation models, pretrained on large-scale ab initio databases spanning millions of atomic structures across the periodic table, enable accurate MLIPs to be constructed with substantially reduced computational effort via transfer learning. In this work, we develop a self-consistent and computationally efficient CSP workflow [1] that combines evolutionary structure search with state-of-the-art foundation-models and transfer learning. The proposed algorithm simultaneously performs rapid structure exploration and con-

structs a system-specific MLIP by iteratively selecting representative and physically relevant configurations for DFT labeling, thereby minimizing redundant computations while maintaining high accuracy.

In this context, the chemically complex Ca–Fe–Ni ternary system under high pressure represents a particularly compelling and challenging test case. Iron and nickel are the principal constituents of Earth-like planetary cores, governing their mechanical, thermal, and magnetic properties. Calcium, while traditionally considered a lithophile element, exhibits pronounced pressure-induced electronic and structural transformations and may become increasingly relevant at deep Earth conditions. The interaction of Ca with Fe–Ni alloys at high pressures has been proposed as a possible factor influencing core composition, density deficits, and geophysical anomalies at the core–mantle boundary. From a materials science perspective, the Ca–Fe–Ni system also exemplifies a complex multicomponent landscape characterized by competing bonding motifs, large atomic size mismatch, and pressure-driven changes in chemical behavior, making it an appropriate benchmark for advanced CSP methodologies.

Applied to the Ca–Fe–Ni system, our approach accurately reproduces the known low-pressure convex hull, validating the methodology and enabling efficient study of high-pressure phases. At P = 1 bar, Ca forms binary compounds with Ni, but does not react with Fe. Only at P = 50 GPa, we predict previously unknown binary Ca–Fe compounds, including those with relatively high iron content, such as Ca_3Fe_2 (Figures 1 and 2). For the first time in this system, a ternary compound Ca_6FeNi with a monoclinic structure is discovered, which becomes thermodynamically stable above 100 GPa. The figures show that, in general, characterizing the high-pressure phases in this system, it can be said that Ca acts as a “soft” ligand, forming complex structural motifs around the Fe–Ni framework and forming mainly non-stoichiometric compounds.

The results obtained demonstrate that foundation-model-based, data-efficient workflows can both accelerate crystal structure prediction and enable the discovery of genuinely new materials in complex multicomponent systems.

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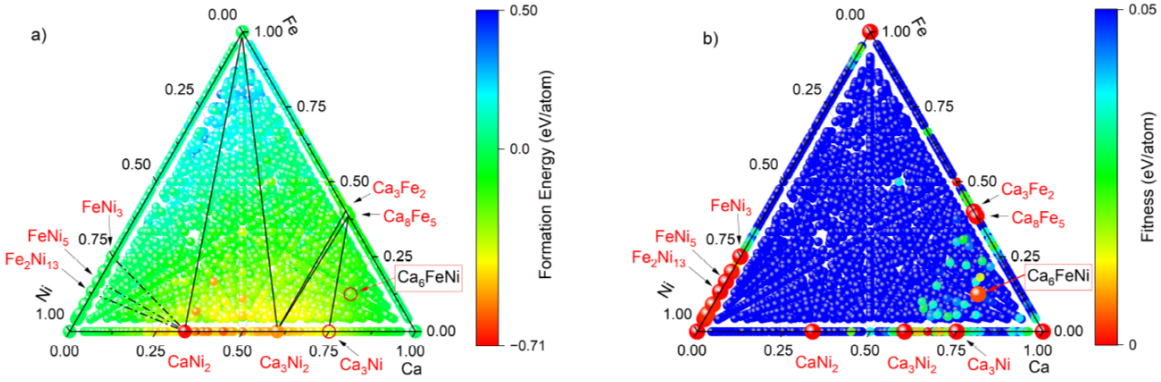

Fig. 1. Distribution of stable and metastable phases in the Ca–Fe–Ni system at P = 50 GPa in composition-energy coordinates. The color scale corresponds to (a) the energy of formation and (b) the distance to the convex hull (fitness characterizing the degree of phase metastability).

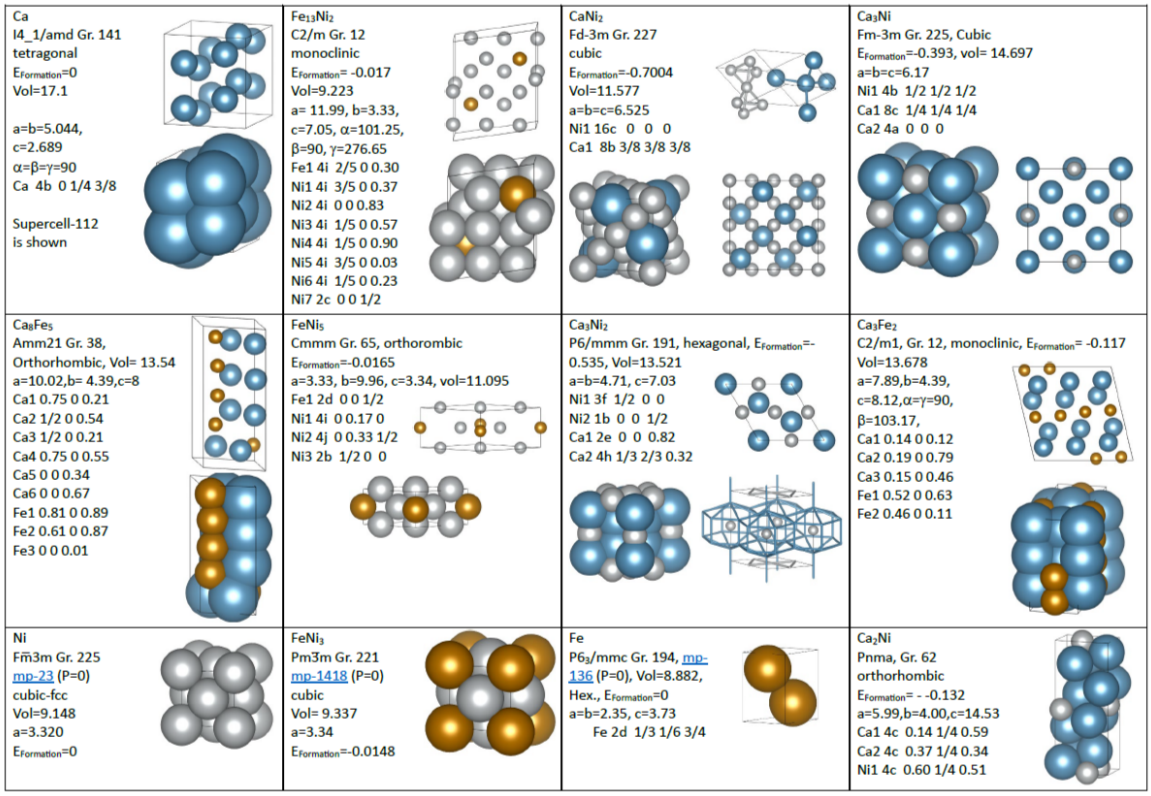

Fig. 2. Some of unary and binary phases forming the convex hull of Ca–Fe–Ni at 50 GPa

Список литературы

[1] E. O. Khazieva, N. M. Chtchelkatchev, R. E. Ryltsev // *J. Chem. Phys.* **161**, 174101 (2024).