Uncertainty-Aware Machine Learning Discovery of Solid–Solid Phase Transitions in Inorganic Materials

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The accurate prediction of solid–solid phase transitions represents one of the most challenging frontiers in computational materials science. These transformations govern a broad spectrum of functional phenomena—ranging from ferroelectricity and shape-memory effects to caloric and thermal-switching responses—yet their finite-temperature characterization remains prohibitively demanding for first-principles methods. In this work, we present a scalable and uncertainty-aware machine-learning framework that enables the high-throughput prediction of temperature-induced solid–solid phase transitions across thousands of inorganic compounds, providing a practical route to discover functional materials under realistic thermal conditions.

Our approach integrates density functional theory (DFT) energetics with graph neural networks (GNNs) trained to predict vibrational free energies over the temperature range 200–700 K. The model, developed using a curated dataset of 6,674 DFT-calculated phonon spectra, achieves sub-10 meV/atom accuracy—comparable to quasi-harmonic free-energy calculations—and incorporates latent-space uncertainty quantification to ensure reliable predictions beyond the training domain. Leveraging this capability, we screened approximately 50,000 inorganic materials and identified over 2,000 temperature-driven polymorphic transitions between 300 and 600 K, encompassing both stable and metastable phase changes.

The results uncover a wealth of functional transitions near room temperature, including numerous polar–nonpolar and polar–polar transformations exhibiting large entropy changes exceeding 300 J K¹ kg¹, indicative of strong first-order behavior and high caloric potential. Among the predicted⁻ candidates, compounds such as Li_3MnF_7, $PNCl_2$, and Fe_4OF_7 show giant entropy changes (|ΔS| > 250 J K¹ kg¹) suitable for solid-state refrigeration, while ZrSeO, $Bi_2W_2O_9$, and Li_4TiS_4 display⁻ sharp lattice thermal conductivity variations (20–70%), making them promising thermal-switching materials. Validation against both experimental data and ab initio quasi-harmonic calculations confirms the robustness and predictive reliability of our framework, with transition temperature uncertainties typically below 100 K.

Beyond predictive accuracy, this study establishes a methodological paradigm for uncertainty-aware, data-driven discovery of phase-change materials. By explicitly quantifying model confidence in high-dimensional chemical space, our approach prioritizes candidates most likely to exhibit measurable transitions, facilitating meaningful AI–experiment synergy. Furthermore, the demonstrated scalability enables systematic exploration of temperature-dependent stability landscapes at the scale of modern materials databases.

This work marks a significant advance toward autonomous discovery of functional phase-transition materials, opening new possibilities for caloric cooling, reconfigurable thermal devices, and neuromorphic heat management technologies. Our framework is readily extensible to organic, hybrid, and multicomponent materials, and can be seamlessly integrated with generative AI models and active-learning loops, paving the way toward fully automated, temperature-aware materials design.

Figures

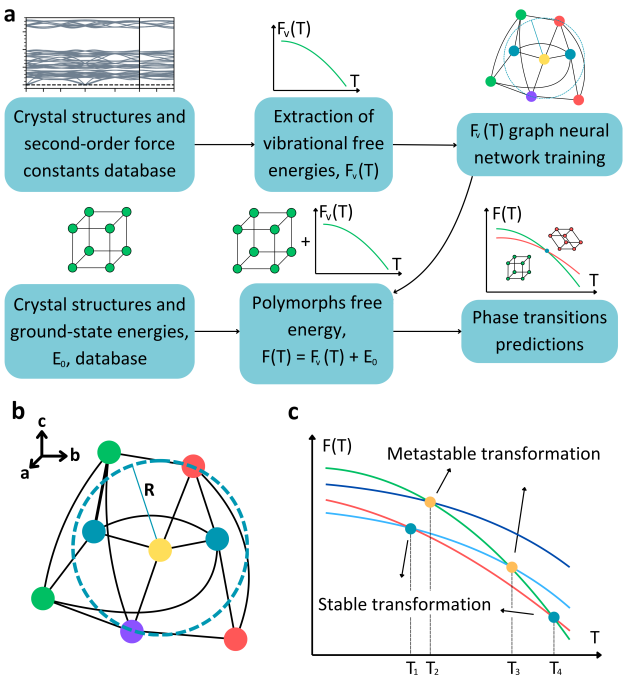

Figure 1. *Machine learning-guided high-throughput approach for prediction of solid-solid phase transitions in inorganic crystals.* a. Sketch of the employed computational workflow relying on DFT and vibrational free-energy calculations. b. Diagram of a graph encoding a crystal structure. The graph is characterized by nodes (dots), edges (lines), and a cutoff distance (R, dashed line). c. Determination of temperature-induced phase transitions, both stable and metastable, based on free-energy curve calculations.