Graph neural networks for prediction of abrupt phase transitions in energy materials: the case of solid-state cooling

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Materials rendering large structural and entropy changes around room-temperature are pivotal for novel and pressing applications in the fields of thermal storage, solid-state cooling and heat pumping, and energy harvesting (e.g., heat to electricity transformation based on the pyroelectric effect). However, only a few phase transitions fulfilling such conditions are known in practice, mostly from experiments planned by humans in a scientifically biased manner.

To address this challenge, we have devised a Machine Learning (ML) approach that efficiently predicts temperature-induced phase transitions between same compound polymorphs. Its core technology is based on state-of-the-art Graph Convolutional Neural Networks (GCNN), trained for predicting vibrational Helmholtz free energies of crystals solely from their atomic structure. Our ML models are trained and tested on a diverse and extensive database comprising thousands of first-principles calculations [1].

To illustrate the performance of our ML approach, we have screened the Materials Project database in order to unravel new families of caloric materials to be used in solid-state cooling and heat pumping applications. Remarkably, we have found more than 50 materials (some of them already reported in literature from experiments) surpassing phase-transition entropy changes of 500 J/kgK at temperatures below 800K (Fig. 1). We have been able to identify some of those as ferroelectric to paraelectric phase transitions based on crystal symmetry arguments (Fig. 2).

The scripts resulting from this study have been made publicly available [2], easily adaptable to any desired target database.

References

[1] A. Togo. First-principles phonon calculations database. 2023, url: http://togodb.org
[2] C. López, R. Rurali, C. Cazorla. Repository of codes for the discovery and understanding of energy materials. 2024, url: https://github.com/IonRepo/IonPred

Figures

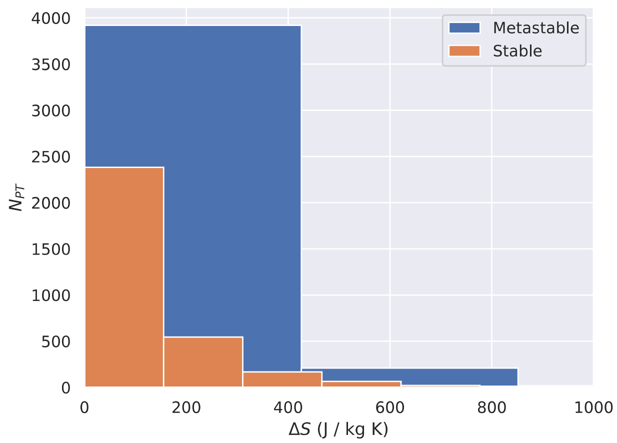

Figure 1. Amount of phase transitions which undergo some ΔS entropy change around room temperature. Stable (metastable) transitions occurring between two stable (metastable) polymorphs are shown in orange (blue).

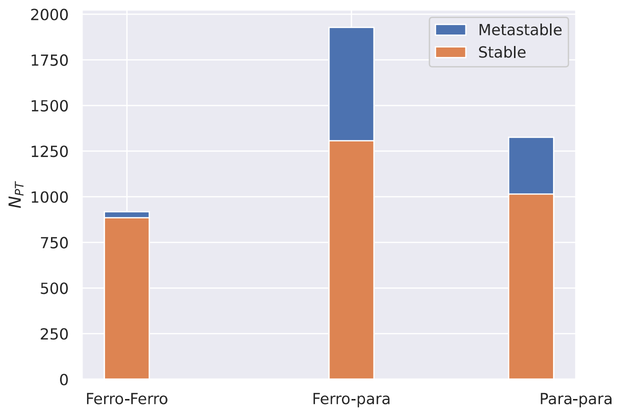

Figure 2. Amount of identified phase transitions associated to some ferroelectric or paraelectric polymorph (based on crystal symmetry arguments).