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Reliable First-Principles Alloy Thermodynamics via Optimal, Truncated Cluster Expansions[1]

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Cluster expansions (CE) are increasingly used to combine first- principles electronic-structure structural formation energies and Monte Carlo methods to predict phase stability in alloys, and search for new materials. As a basis-set expansion in terms of lattice geometrical clusters and effective cluster interactions, the CE is exact if infinite, but is tractable only if truncated. We present an optimal truncation for CE basis sets that provides reliable thermodynamics and is easy to implement in mulitcomponent alloys, whereas former truncations were not well defined and sometimes led to unreliable results. We discuss predictive error estimation, error estimation of temperature prediction, Rayleigh- Ritz variation errors associated with basis set truncation for both concentration-dependent and independent version (similar to local compact support in finite-element methods), as well as a means for rapid assessment of transition temperatures without performing Monte Carlo. We exemplify all of the issues in various binary alloys. A Thermo Toolkit (TTK) that automates construction of the optimal, truncated CE, generation of linear-independent unit-cell structures, electronic-structure job submission for the required unit-cell, collection into database, and ultimately Monte Carlo construction of phase diagram is exemplified also.

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