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Predicting complex ground state structures from first principles: genetic algorithm for finding accurate coarse-grained Hamiltonians

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First-principles quantum-mechanical (QM) calculations allow to evaluate many interesting properties of a *given* nano-scale configuration of atoms with high accuracy. However, *predicting* stable structures or finite-T thermodynamic configurational averages with QM accuracy remains a challenge: even for a binary solid with N atoms per unit cell, 2^N distinct configurations must be evaluated. Such large numbers of calculations can be made affordable by mapping the QM Hamiltonian onto a computationally simpler "coarse-grained" Hamiltonian. The ability to predict ground state structures then depends on the shape of the coarse-grained Hamiltonian, but this shape is not a priori clear. For instance, a few simple assumed generic interactions will allow only a few simple ground states, but any more complex structures will be missed. For the generalized Ising model for binary alloys (cluster expansion), I show how a genetic algorithm[1,2] can identify the leading interactions which characterize a given system. I illustrate the method for the bcc binary alloys of Nb, Ta, Mo, W. A rich spectrum of ground state structures is found, including both well-known and unsuspected complex structures, far beyond what is envisioned from "usual-suspect" structure listings or from simple generic interactions. At the same time, order-disorder temperatures are significantly lower than those from simple intuition-based interactions, in agreement with experimental observations for these systems. This work was done at the National Renewable Energy Laboratory, supported by DOE-SC-BES, in collaboration with A. Zunger and G. Hart.

[1]G. Hart, V. Blum, M. Walorski and A. Zunger, Nature Materials **4**, 391 (2005); [2]V. Blum, G. Hart, M. Walorski and A. Zunger, Phys. Rev. B **72**, 165113 (2005).