Scalable First Principles Calculations for Alloys

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The effect of disorder in materials is of great fundamental and technological interest. Here I will present our implementation of multiple scattering theory for first principles density functional calculations. This approach directly obtains the single particle Green's function of the Kohn-Sham equation, either in reciprocal space (Korringa-Kohn-Rostocker i.e. KKR) or real space (Localy-Selfconsistent Multiple Scattering i.e. LSMS). The KKR method allows an efficient description of random solid solution alloys using the Coherent Potential Approximation (CPA), while our LSMS code allows for scalable large scale first principles density functional calculations of materials. A fundamental science driver for scalable, large scale, first principles calculations of materials is the need to understand states beyond periodic crystalline lattices. For large simulation cells, needed to describe extended electronic and magnetic orderings, defect states or disorder in alloys, the cubic scaling of traditional first principles methods have prevented direct calculations. The linear scaling nature of the LSMS ab initio code enables the treatment of extremely large disordered systems from the first principles using the largest parallel supercomputers available, such as calculations for O(10,000 - 100,000) atoms on current high performance computing architectures. For exascale systems, we have extended the use of accelerators to enable the efficient calculation for embedding methods and forces. Currently ongoing work focuses on the calculation of electric conductivity in the presence of disorder and defects.

While DFT calculations have proven to be a useful tool in the study of ground state properties of many materials, we will go beyond the ground state by describing an approach to utilize machine learning methods to combine first principles density functional calculations with classical Monte-Carlo simulations to investigating the statistical mechanics of materials. The investigation of finite temperature properties relies on the possibility of a large number of evaluations of the system's Hamiltonian that are required to sample the phase space needed to obtain physical observables. We have demonstrated a solution to this problem that harnesses the computational power of large massively parallel computers by combining classical Monte-Carlo calculations with our first principles multiple scattering electronic structure code (LSMS) by employing Machine Learning techniques.
The combination of LSMS with a machine learning workflow, that can consider both classical interaction models and artificial neural network based models, allows us to investigate alloy ordering transitions for increased simulation cell sizes. Our approach is able to sample both magnetic or chemical order, allowing the first principles calculation of order/disorder phase transitions and phase separations in alloys.

These computational capabilities are available in our Multiple Scattering Theory suite (MuST)
[https://github.com/mstsuite]

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