Constructing machine-learning potentials derived from disordered structures for crystal structure prediction

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Crystal structure prediction (CSP) is a problem of finding a structure with the lowest free energy in given chemical compositions. First-principles calculation based on density functional theory (DFT) is known to be very accurate in evaluating free energies because of its non-empirical nature. The DFT-based CSP has been successful in identifying inorganic crystals under extreme conditions and organic crystals. However, cost of *ab initio* calculations is prohibitive in exploring huge configuration space of multinary (ternary or higher) phase. Neural network potentials (NNP) have much lower cost with comparable accuracy to DFT, but scarcity of information on crystal structures impedes selecting training sets. Herein we suggest developing a training set from dynamical trajectories of disordered phases: liquid and amorphous phases. Molecular dynamics of them are simulated by DFT without any precedent information of structures except for chemical composition. To validate accuracy of NNP for crystal structure prediction, we compare DFT and NNP energies of experimental phases as well as theoretical crystal structures with low energy for Ba_2AgSi_3, Mg_2SiO_4, $LiAlCl_4$, and $InTe_2O_5F$. We find very good correlation between DFT and NNP energies, which ensures NNP is able to rank energies of low-energy structures. We also find that NNP can accelerate the speed of crystal structure prediction compared DFT-based one. This work proposes very efficient method in crystal structure prediction with reliable neural network potentials.

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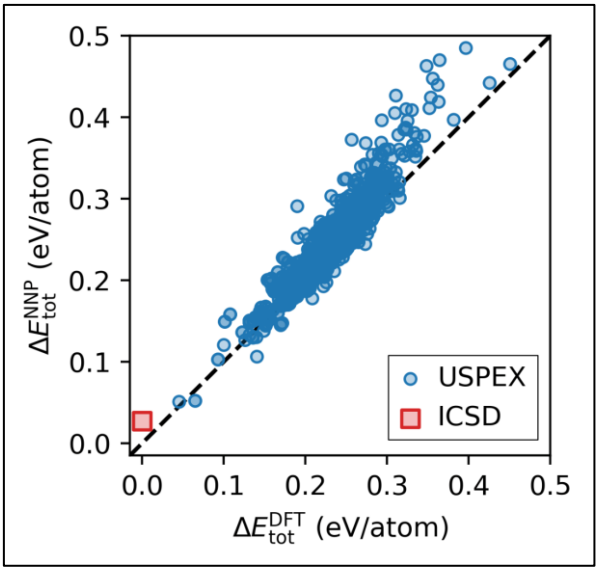

Fig.1: Energy correlation between DFT and NNP energy in Ba_2AgSi_3 for experimental phases as well as theoretical low-energy phases. The DFT and NNP energy reference is based on experimental phase which is optimized in DFT.

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