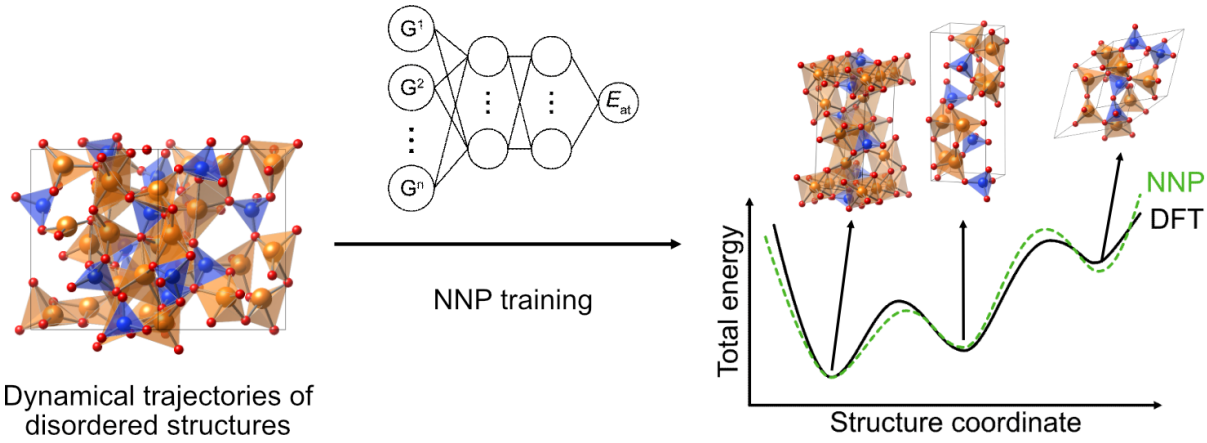
How to make training sets of NNP for crystal structure prediction



Since there are many unknown stable materials with ternary or higher (multinary) composition, crystal structure prediction is necessary to accelerate the rate of material discovery. This demands fast and accurate evaluation of free energies in exploring a vast number of atomic configurations. The neural network potential (NNP) can meet this requirement but a scarcity of information on the crystal structure poses a challenge in choosing training sets. In this work, we propose a method of constructing training sets from density functional theory (DFT)–based dynamical trajectories of disordered structures, which does not require any preceding information on material structures except for the chemical composition. With this method, we find strong correlation of NNP and DFT energies, ensuring that the NNPs can properly rank energies among low-energy crystalline structures. We also find that the evolutionary search using the NNPs is more efficient than the DFT-based approach. This work was published at Physical Review B 2020, **102**, 224104.