DFT based Neural Network Force Fields from Energy Decompositions – Supplemental Materials

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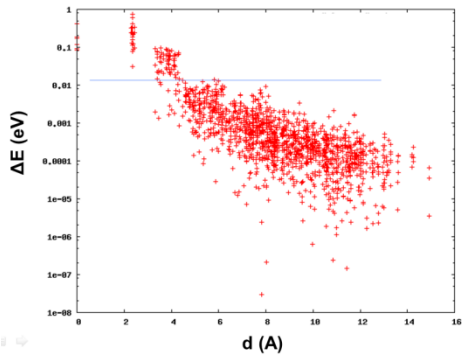
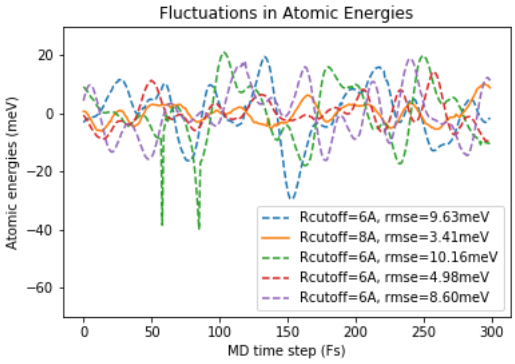
S1. Locality of atomic energies

Fig.S1(a) shows how neighboring E_i changes when a center atom j has a displacement. One can see that the change of E_i decays exponentially with the distance $R_{ij} = |\boldsymbol{R}_j - \boldsymbol{R}_i|$. Fig.S1(b) shows how the atomic energy E_i of a center atom changes when the nearby atoms within a R_{cutoff} are fixed while the outside atoms are moving in a MD simulation at 300K. Several center atoms i are chosen for the tests, and different R_{cutoff} are also examined. When R_{cutoff}=6A, there are roughly 60 atoms within the cutoff radius, and the variation in E_i due to the motions of the outside atoms has a RMSE of 10 meV or less. This will thus be the upper limit of our SANNP fitting when R_{cutoff}=6A is used. The analyses of E_i here provide another advantage of our SANNP method: the upper limit of accuracy can be obtained prior to the training of SANNP, thus R_{cutoff} can be chosen based on the target accuracy.



(a)



(b)

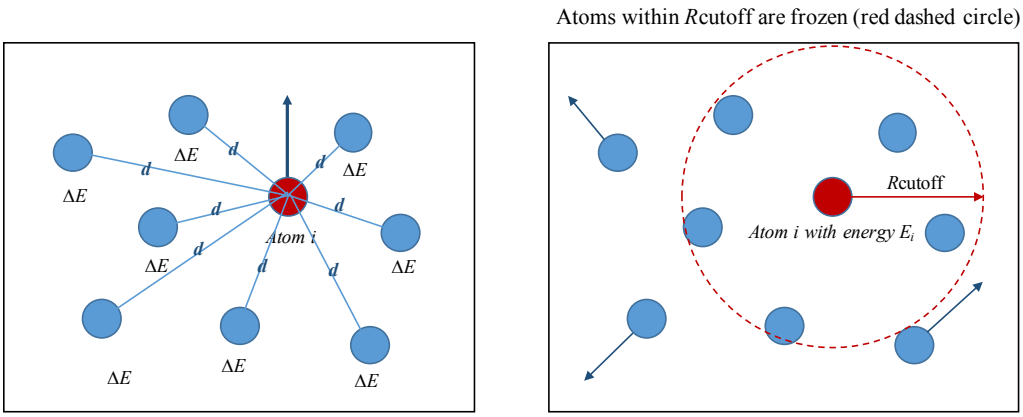


Figure S1. Fluctuation in the employed atomic energy E_i. (a) change of energy of atom j when a chosen atom i displaces. (b) change of energy of atom i with its neighboring atoms within R_{cutoff} frozen and the surrounding atoms outside of R_{cutoff} moving according to an NVE ensemble. (c) schematics of atomic motions for (a). (d) schematics of atomic motions for (b).

S2. SANNP code and Training data

We have written a custom machine learning code all of the calculations reported in the manuscript. The code is freely available at: https://gitlab.com/yufeng.huang/sannp. The training, validation, and testing data used in the manuscript are also included in the Data folder.

Detailed description about running the code is on the Gitlab website. Datasets used in this study have been provided in the example calculations.

S3. Convergence of the neural network errors in the size of the neural network

In the paper a large system with 550 input features and a 2-layer neural network with 500 nodes per layer is used. The reason for the large system to avoid errors due to incompleteness. In the manuscript, the convergence with respect to the basis set size is shown in Figure 4. Here we show how the forces converge when the number of nodes in the neural network increases. For this test, the M=4 piecewise cosine basis set, which corresponds to 56 input features, is used. The simplest case has 40 nodes in each layer in the 2-layer neural network. Changing the number of nodes only reduces the error by about 0.014eV/A.

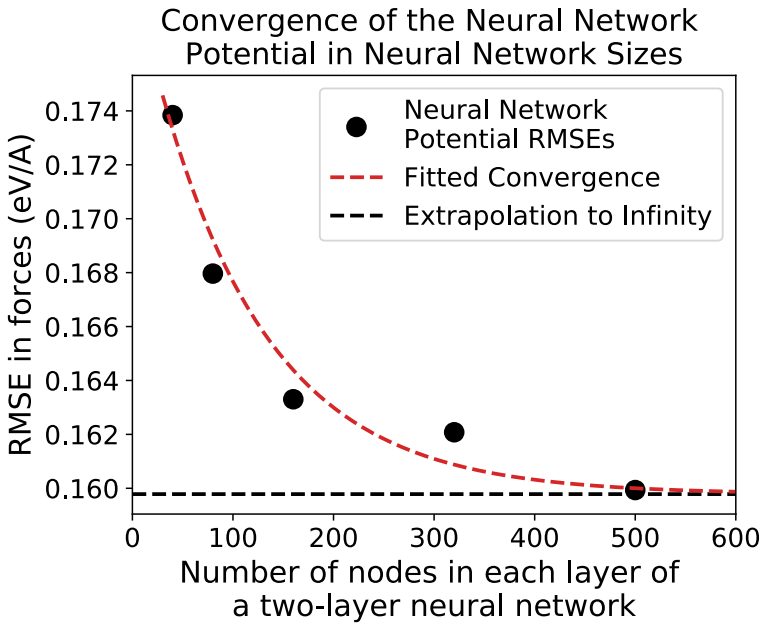


Figure S2. Convergence of the single atom neural network potential (SANNP) in the size of the neural network model. The difference between a large neural network (500 nodes) and a small neural network (50 nodes) is only 0.014eV/A, which would not contribute to a large difference in practical applications.

S4. DFT data generation

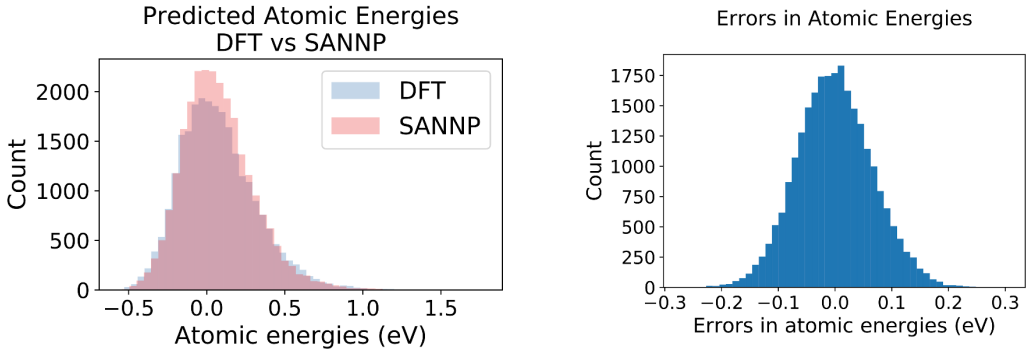
The computational method used here is the same as in Kang and Wang [1].

The amorphous Si structure is first equilibrated in LAMMPS [2] and using the ReaxFF reactive force-field[3], which allows for long simulations of amorphous structures with bond breaking and formation. We start with the $4 \times 2\sqrt{2} \times 2\sqrt{2}$ supercell of the crystalline diamond structure of silicon consisting of 256 atoms. Then the system is heated to 4300 K in 170 ps with the NPT ensemble. After equilibrating the system for another 150 ps at 4300 K, the system is cooled down to 10 K in 1.1 ns. Amorphous Si is formed during the annealing process. Using the LAMMPS results at the initial structures, we perform DFT calculations in PWmat to relax the geometries. The results are consistent with experimental observations [4].

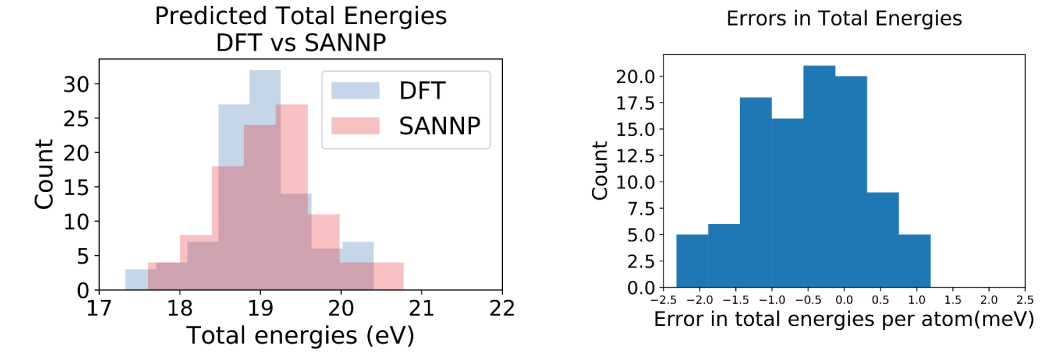
[1] J. Kang, L. W. Wang, Physical Review B, 96(2), 1–5. (2017)
[2] S. Plimpton, J. Comput. Phys. 117, 1 (1995).
[3] A. C. T. van Duin, S. Dasgupta, F. Lorant, and W. A. Goddard, J. Phys. Chem. A 105, 9396 (2001).
[4] S. Kugler, L. Pusztai, L. Rosta, P. Chieux, and R. Bellissent, Phys. Rev. B 48, 7685 (1993).

S5. Distributions of atomic energies, total energies, and forces and their errors between DFT and neural network predictions

(a)



(b)



(c)

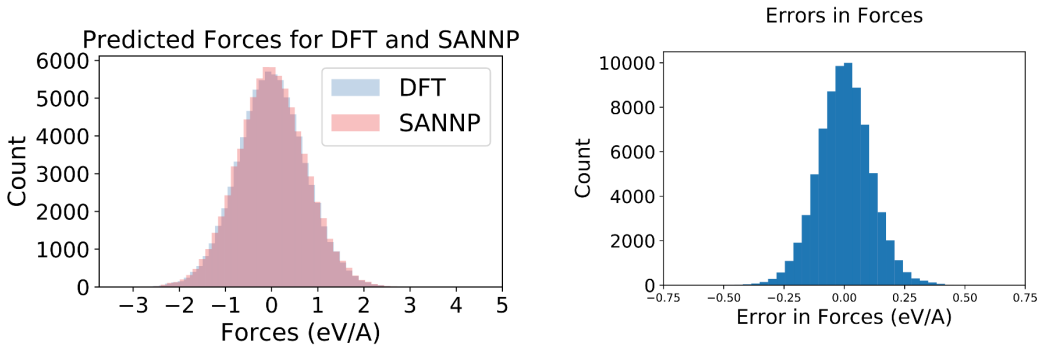


Figure S3. Distributions of (a) atomic energies, (b) total energies, and (c) forces and their errors between DFT results and SANNP predictions for the test set. As shown in the above, the atomic

energies, (a), and forces, (c) agree well between DFT and SANNP, because a large number of data are used for training and comparison. On the other hand, the distribution in the total energies seems to have a larger discrepancy, but largely due to the small test set size.

S6. Comparison with Other Amorphous Silicon Structures

V. L. Deringer et. al. (*J. Phys. Chem. Lett.*, **2018**, *9* (11), pp 2879–2885) has recently reported a study of amorphous silicon structures using the Gaussian Approximation Potential with the Smooth Overlap of Atomic Positions (SOAP) features. Although their calculations are based on different DFT training data, it is nevertheless helpful to compare the errors of our SANNP scheme using their structures. We can carry out a DFT calculation using their reported structure, we obtained SANNP RMSE of 67meV, and 0.187eV/A for the atomic energy Ei, and atomic forces, which are only slightly larger than the errors for our system. Note that this amorphous structure was not included in our SANNP training set.