Supplementary information to "Entropic stabilization of a structurally tolerant phase: The ionic phase of lithium alanate"

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1 Supplementary information

1.1 Accuracy tests for the machine learned potentials

To be sure that our results are not artifacts of a machine learned potential that is inaccurate in certain regions of configurational space, we have constructed two completely independent potentials and performed all the relevant calculations with both of them. As shown in Fig. both potentials give accurate forces and energies. The root mean square errors for the NequiP potential are 0.96 meV/atom for the energy and 178 meV/Åfor the forces in the training set and 0.96 meV/atom and 178 meV/Åfor the test set. For the n2p2 potential the root mean square errors are 0.72 meV and 171 meV/Åfor the training set and 0.25 meV and 60 meV/Åfor the test set.

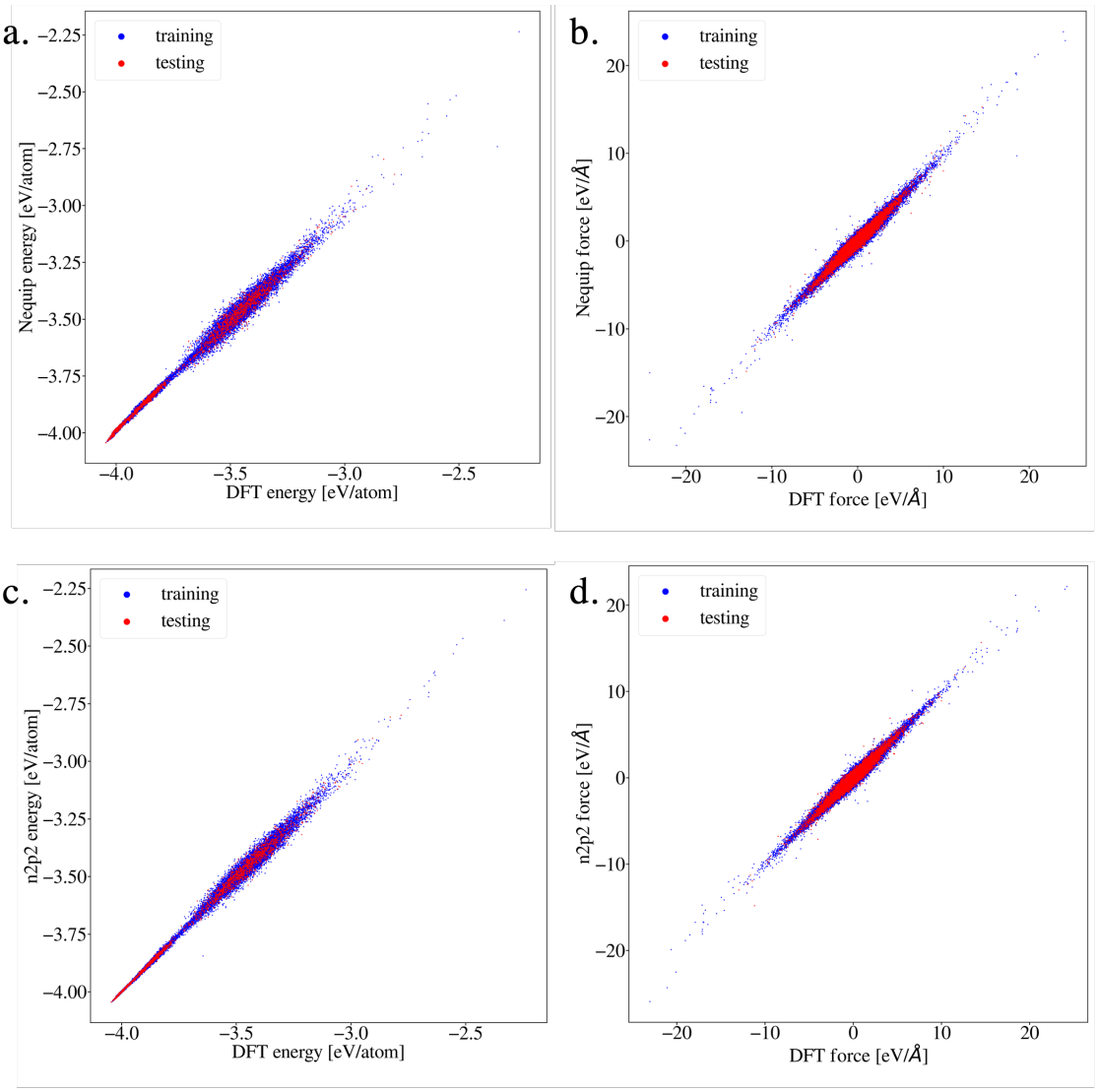

Fig. 1 Accuracy of the machine learned energies and forces: Panel a): NequiP energies vs DFT energies, panel b): NequiP forces vs DFT forces, panel c): n2p2 energies vs DFT energies, panel d): n2p2 forces vs DFT forces

To calculate the vibrational free energies, accurate phonon frequencies are required. Figure 2 shows that there is also good agreement between the machine learned potentials and DFT. In particular the cross over points where the ionic structure becomes lower in energy than the polymeric structure agrees to within a few Kelvin.

1.2 Solution of Eq. ??

As can be seen in Fig. 3), the function of Eq. **??** decays continuously and has therefore a unique intersection with $1/M$ for any value of M.

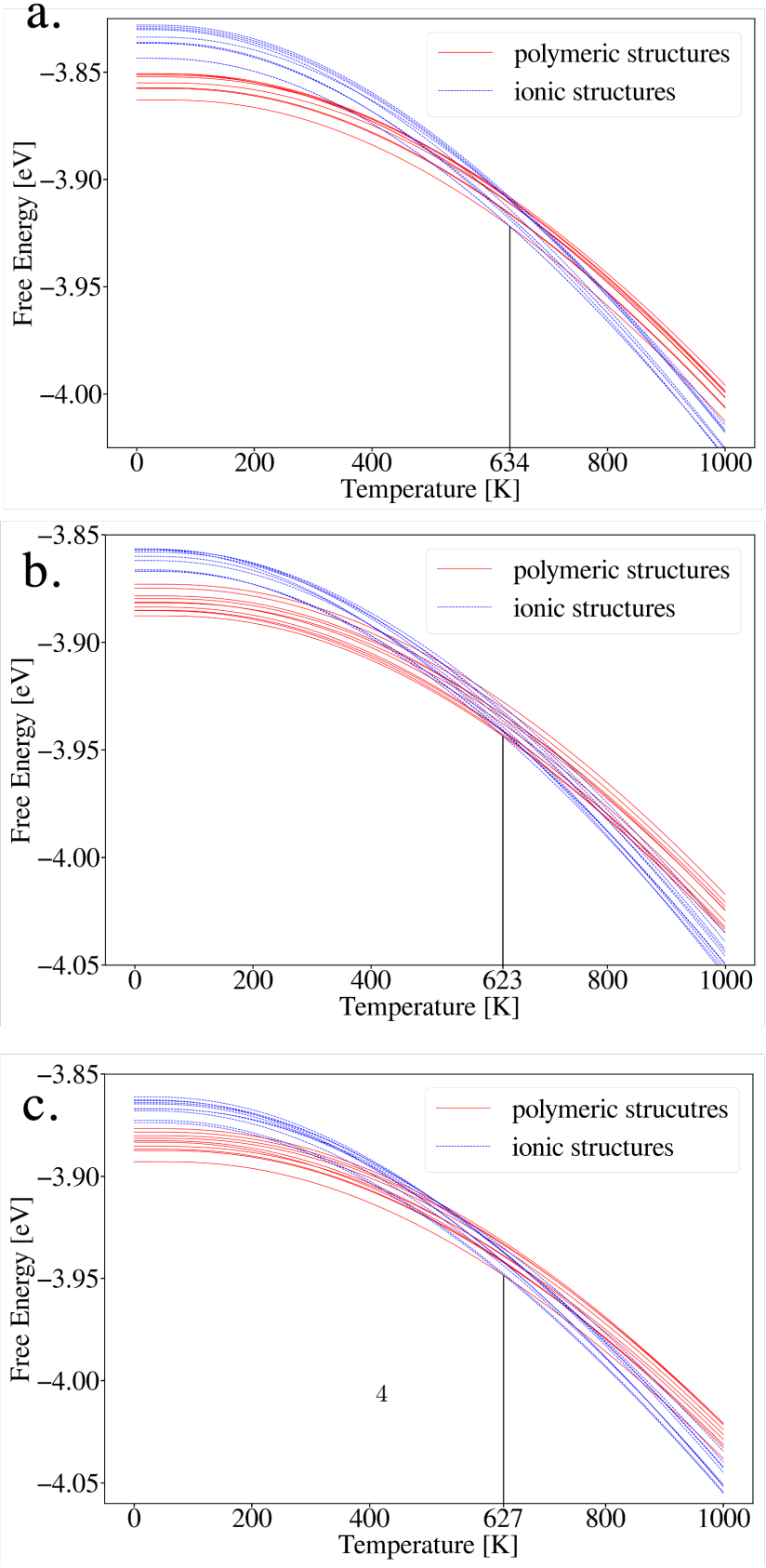

Fig. 2 Free energies within the harmonic approximation of the lowest 10 ionic and polymeric structures: a. n2p2 potential, b. nequip potential and c. vasp SCAN calculations.

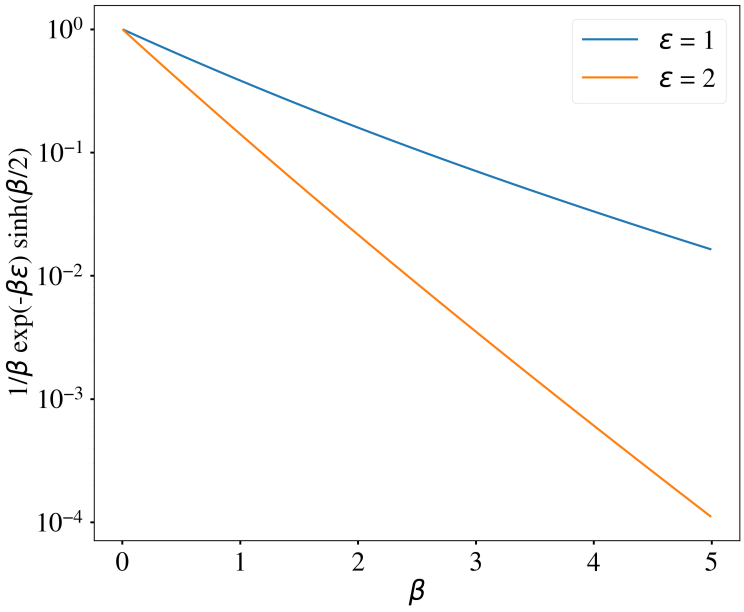

Fig. 3 The function from Eq. **??**. Plot of the function on the left hand side of Eq. **??**