

Deformation-trained Deep Potential model for superionic water ice: transferability from stress-strain data to phase equilibria

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Machine-learning interatomic potentials are increasingly used to accelerate materials discovery; however, their reliability often hinges on transferability beyond the states represented in the training set. Here, we assess the transferability of a Deep Potential machine-learning (DP-ML) model trained to reproduce stress–strain curves of high-temperature/high-pressure crystalline phases of water [1,2]. Using this deformation-trained DP-ML model, we determine the coexistence lines for the phase transitions between insulating ice X and superionic ice XVIII, and between ice XVIII and the liquid. They are computed using multiple non-equilibrium free-energy calculation techniques [5,6,7], and the resulting phase boundaries are found to be in excellent agreement with previous estimates as shown in Figure 1. These results indicate that a DP model fitted primarily to mechanical response can nevertheless reproduce equilibrium thermodynamic properties and phase equilibria. More broadly, they suggest that systematically including deformed solid states in training sets may be an effective strategy to improve the robustness of ML interaction models for condensed-matter systems under extreme conditions of pressure and temperature. Finally, we outline how the present DP-ML model can also be useful for exploring the details of the two involved phase transitions. First, it is found that the ice XVIII–liquid transition is weakly first-order, such that it may display continuous-like signatures. This may represent the first example of the existence of such transitions in three-dimensional structural solid–liquid transitions. In addition, the DP model provides insight into the dynamics of the ice XVIII – ice X transition, which involves a simultaneous freezing transformation for the proton subsystem and a martensitic-type face-centered-cubic (fcc) to body-centered cubic (bcc) transition for the oxygen sublattice. Preliminary results indicate that the martensitic transition occurs only after the entire proton-subsystem has transformed into a coherent fcc lattice. This is

demonstrated in Figure 2, which displays a sequence of atomic snapshots in which the protonic and oxygen-ion subsystems are displayed separately. The proton-freezing transition has initially produced two separate fcc grains (Fig. 2.a). Only once the grain boundaries have straightened (Fig. 2.b) and merged, thus forming a single coherent fcc grain (Fig. 2.c), does the oxygen-lattice transformation occur. These results demonstrate the potential predictive power of machine-learning models for the thermodynamic and kinetic properties of condensed-matter systems under extreme conditions.

References

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Figures

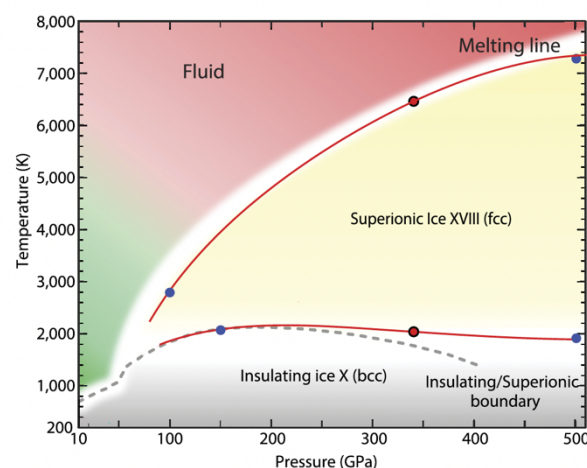


Figure 1. Phase diagram of water in the high-temperature/high-pressure regime, including the insulating ice X, the superionic ice XVIII, and the superionic liquid. Red circles denote the coexistence conditions at 340 GPa for the X–XVIII and the XVIII–liquid transitions as determined from the Gibbs free-energy RS calculations. Blue circles represent similar results for different values of the pressure, obtained by applying the same free-energy procedures. Red lines represent the full coexistence curves for both transitions as obtained using dynamic Clausius–Clapeyron-integration simulations. The thick white line and dashed gray line have been taken from Refs. [3] and [4].

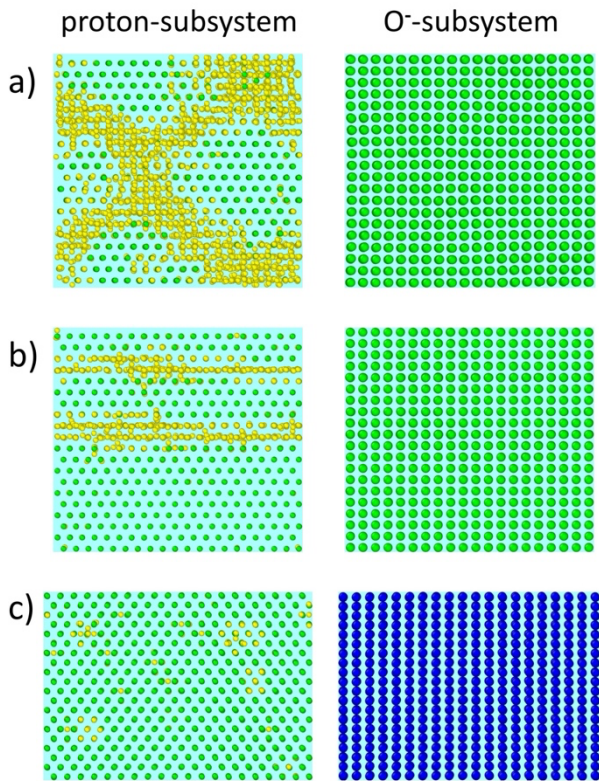


Figure 2. Structural evolution of the system during cooling. a) Freezing of the protonic subsystem leads to the nucleation and growth of two fcc grains (green) within the disordered (yellow) proton subsystem. b) The system evolves towards the formation of two straight parallel grain boundaries (yellow) in the protonic subsystem, while the oxygen ion subsystem remains in fcc structure. c) Further evolution of the system allows for migration and merging of the grain boundaries leading to a single coherent fcc structure in the protonic subsystem, ultimately leading to the transformation of the oxygen ion subsystem into a bcc structure.