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Born-Oppenheimer molecular-dynamics study of ice polymorphs

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We report ab-initio molecular dynamics, using state-of-the-art non-local dispersion, to study, inter alia, the structural, hydrogen-bonding, vibrational and Raman properties of ices Ih, VII and XVII at appropriate temperature and pressure ranges for their stability. In many senses, probing of ice Ih serves as a validation tool for the functional, with reasonable agreement with experiment.
In the case of ice VII, we are motivated to elucidate any possible hints of intramolecular strain that may serve as precursor events for proton hopping to unfold. We determine such equilibrium properties to be in reasonable agreement with experiment, although we do not detect any water-dissociation and proton-hopping events per se, owing to still-large water-dissociation free-energy barriers. The observed anomalous self-diffusivity of ice VII in the region of 10 GPa at ~400 K has been suggested to arise from a change in proton-hopping mechanism involving a transition from ionic-defect-driven diffusivity to that dominated by diffusion of rotational defects.
In the case of ice XVII, we probe the stability of the cage structure, and dissect the vibrational properties, obtaining reasonable agreement with recently-reported experiments.

Significance statement

Empirical forcefields often give unsatisfactory agreement for various equilibrium properties of ice polymorphs, especially vibrational ones. Here, using state-of-the-art functionals with non-local dispersion, we overcome these problems, to a good degree.

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