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Efficient ab initio molecular dynamics using exact reweighting VIDVUDS OZOLINS, University of California, Los Angeles, MARK ASTA, University of California, Berkeley — Density-functional theory (DFT) based *ab initio* molecular dynamics (AIMD) is a promising method for calculating high-temperature thermodynamic properties of solids and liquids. Nevertheless, computational expense associated with AIMD simulations has prevented general adoption of these methods. We show that substantial savings of computational effort can be realized by using less expensive (and less accurate) Hamiltonians to generate long MD trajectories and by recalculating statistically independent snapshots with high-accuracy DFT methods. A formally exact reweighting formula, based on the Jarzynski switching approach, is used to obtain thermal averages and thermodynamic properties in the high-accuracy DFT ensemble. If under-converged AIMD simulations with low energy cutoffs and coarse k-point meshes are used to generate trajectories, this approach can lead to savings of CPU time of a factor of 10 to 100, depending on the relevant correlations times. We also present extensions of the reweighting method to calculate impurity free energies and free energy barriers for interstitial diffusion. Robust methods for estimation of statistical errors based on random subsampling and variance extrapolation are discussed.

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