Machine learning quantum Monte Carlo: application to water clusters

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A complete understanding of the hydrogen bond and proton transfer mechanism in water is still lacking, since it requires an accurate potential energy surface (PES) and very expensive quantum mechanical simulations of the nuclear part. Reproducing this high-dimensional surface with current high-level computational chemistry methods is infeasible for the largest clusters. We test the gradient-based kernel ridge regression methods[1, 2] to reproduce the PES starting from a dataset of energies and forces of the protonated water hexamer obtained via simulations combining classical molecular dynamics (MD) for the nuclei and quantum Monte Carlo (QMC) for the electrons [3]. The QMC+MD approach yields very accurate results for the classical dynamics, which are however affected by the intrinsic noise inherent in the stochastic sampling of both nuclear and electronic phase space. We prove that QMC multivariate noise is not detrimental to the learning of energies and forces and that the derived machine learning force field can be used to run long and reliable quantum molecular dynamics simulations.

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