Uncertainty Quantification in MD Simulations: Forward Propagation and Parameter Inference

Quantifying Prediction Fidelity in Multiscale Multiphysics Simulations

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Abstract

Numerous models have been developed in the past to characterize the structural and dynamical properties of water. A popular approach has been to employ an empirical force-field description, in which pairwise-additive functions are fitted to reproduce bulk-phase experimental data in a classical molecular dynamics (MD) simulation framework. Examples include the RWK, SPC/E, TIP4P, TIP4P-Ew and TIP5P potentials. These force-fields representations are often capable of reproducing key physical properties of water with a good degree of accuracy.

In this work, we explore the possibility of determining or refining a set of force-field parameters for water using a reformulated Bayesian approach based on polynomial chaos (PC) expansions. The latter provide efficient representations of the dependence of the solution on random variables that parametrize uncertain inputs. As a test case, we consider isothermal, isobaric MD simulations of TIP4P water at ambient conditions, $T = 298$ K and $P = 1$ atm. Following [1], we implement a stochastic spectral reformulation of the forward problem, which allows us to express physical observables in terms of polynomial chaos (PC) expansions. The intrinsic (thermal) noise present in the atomistic system combines with the parametric uncertainty to yield non-deterministic, noisy predictions for the MD predictions of water observables. Polynomial chaos expansions of selected water observables are then built using both a non-intrusive spectral projection (NISP) and a Bayesian inference approach. We show that for the present case, the effect of the thermal noise in the atomistic system can be controlled, and the two methods yield similar results [2]. To illustrate how force-field parameters can be inferred, we focus on a synthetic problem based on MD computations [3]. Specifically, presumed "true" values of the TIP4P model parameters are used to generate a collection of noisy data of macroscale observables. Observations of density, self-diffusion and enthalpy are used for this purpose. Exploiting the stochastic reformulation of the forward problem we demonstrate that atomic force-field parameters may be accurately inferred using low-order surrogate models.

References

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