Data driven, molecular dynamics for nanoscale fluid mechanics

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Abstract: For over five decades, molecular dynamics (MD) simulations have helped to elucidate critical mechanisms in a broad range of physiological systems and technological innovations. MD simulations are synergetic with experiments, relying on measurements to calibrate their parameters and probing "what if scenarios" for systems that are difficult to investigate experimentally. However, in certain systems, such as nanofluidics, the results of experiments and MD simulations differ by several orders of magnitude. This discrepancy may be attributed to the spatiotemporal scales and structural information accessible by experiments and simulations. Furthermore, MD simulations rely on parameters that are often calibrated semiempirically, while the effects of their computational implementation on their predictive capabilities have only been sporadically probed. In this work, we show that experimental and MD investigations can be consolidated through a rigorous uncertainty quantification framework. We employ a Bayesian probabilistic framework for large scale MD simulations of graphitic nanostructures in aqueous environments. We assess the uncertainties in the MD predictions for quantities of interest regarding wetting behavior and hydrophobicity. We focus on three representative systems: water wetting of graphene, the aggregation of fullerenes in aqueous solution, and the water transport across carbon nanotubes. We demonstrate that the dominant mode of calibrating MD potentials in nanoscale fluid mechanics, through single values of water contact angle on graphene, leads to large uncertainties and fallible quantitative predictions. We demonstrate that the use of additional experimental data reduces uncertainty, improves the predictive accuracy of MD models, and consolidates the results of experiments and simulations.

References

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