**Modeling Hydration, One Water Molecule at a Time:**

**Towards Realistic Computer Simulations Through**

**Many-Body Representations and Data-Driven Algorithms**

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Two of the most challenging problems at the intersection of electronic structure theory and molecular dynamics simulations are the accurate representation of intermolecular interactions and the development of reduced-scaling algorithms applicable to large systems. To some extent, these two problems are antithetical, since the accurate calculation of non-covalent interactions typically requires correlated, post-Hartree-Fock methods whose computational scaling with respect to system size precludes the application of these methods to large systems. I will describe our many-body molecular dynamics (MB-MD) methodology for aqueous systems that overcomes these limitations and enables computer simulations from the gas to the condensed phase, with chemical and spectroscopic accuracy. MB-MD is a unified molecular dynamics framework that combines many-body representations for potential energy, dipole moment, and polarizability surfaces that are derived entirely from correlated electronic structure data-driven approaches, with quantum dynamics methods that explicitly account for nuclear quantum effects. I will discuss the accuracy and predictive ability of the MB-MD methodology in the context of molecular modeling of complex aqueous systems, from gas-phase clusters to bulk solutions and interfaces, with a particular focus on the relationships between structural and dynamical properties and vibrational spectra.

