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A BAYESIAN FRAMEWORK FOR CALIBRATION AND UNCERTAINTY QUANTIFICATION OF COARSE-GRAINED ATOMISTIC MODELS

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The proposed work addresses issues related to the derivation of reduced models of atomistic systems, their statistical calibration, and their relation to atomistic models, particularly those involved with nanostructured materials. In a general sense, an atomistic description of a material contains a listing of the chemical constituents of the material at the atomic scale (≈ 1 $\mathring{A}$), as well as a set of mathematical formulas that quantify intra- and intermolecular interactions between the chemical components of the material. These interactions are usually given in the form of a potential energy ansatz containing a set of free parameters that are calibrated in reference to experimental data and highly accurate, but computationally expensive, quantum chemistry simulations. The potential energy forms, once calibrated, serve as the basis for molecular dynamics (MD) simulations which propagate the system forward in time by numerically solving the classical Newton equations derived from the potential energy formulas. Although much faster than quantum chemistry based computations, the described classical methods, referred to as molecular mechanics methods, become prohibitively expensive for systems of millions or billions of atoms. Thus, the theoretical chemistry community is interested in developing reduced models of atomistic systems that are computationally feasible for systems of meso- to macroscopic size ($>$ 10 nm). Popular routes to model reduction include so-called coarse-graining (CG) methods that reduce the degrees of freedom associated with atomistic calculations by replacing particular groups of atoms with single interaction sites (beads). The reduced models derived from the CG procedure, known as coarse-grained models, involve their own sets of potential energy ansätze, the free parameters of which are commonly calibrated in reference to atomistic simulation data such that the resulting CG models may reproduce aggregate behaviors of the original atomistic models in MD simulations [1].

We pursue a statistical method for CG model calibration based on principles of Bayesian inference [2], the results of which are dependent on the identification of quantities of in-

terest (QOIs) and their related physical observables. The main issues we address here are the derivation of prior CG parameter probabilities from rigorous mathematical principles, the formulation of likelihood functions with respect to relevant physical observables, computational techniques for sampling CG parameter probability distributions, and the analysis of model uncertainty with respect to chosen QOIs. In order to illustrate these issues, we develop a CG model for a liquid heptane system at standard thermodynamic conditions, calibrate CG parameters according to our Bayesian inference framework, and analyze the accuracy and precision of the model's prediction of the vapor/liquid transfer free energy. We conclude that the selection of calibration observables is vital to the well-posedness and predictive ability of a CG model. We also comment on the implications of our method to problems of model selection and plausibility.

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