Final Technical Report

DOE Award Number: DE-SC0002506

Recipient: Johns Hopkins University

Project Title: Quantifying Prediction Fidelity in Multiscale Multiphysics Simulations

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Date: January 7, 2013

Project Goals

This is a collaborative proposal that aims to establish theoretical foundations and computational tools that enable uncertainty quantification (UQ) in tightly coupled atomistic-to-continuum multiscale simulations. The program emphasizes the following three research thrusts:

1. UQ and its propagation in atomistic simulations, whether through intrusive or nonintrusive approaches;

2. Extraction of macroscale observables from atomistic simulations and propagation across scales; and

3. Uncertainty quantification and propagation in continuum simulations for macroscale properties tightly coupled with instantaneous states of the atomistic systems.

Thus, the project offers to enable the use of multiscale multiphysics simulations as predictive design tools for complex systems.

Accomplishments

Bayesian inference of continuum properties

In order to demonstrate the application of Bayesian inference methods to determine continuum parameters and observables, we have taken advantage of an existing MD database generated as part of an ongoing DOE/BES project. In particular, the latter includes MD computations of diffusion in an isothermal, binary Ni/Al system at high temperature. A mixing-measure theory was then adopted that enables direct comparison between atomic mixing in MD computations and continuum models. Specifically, the theory contrasts the appropriate models of the atomic distribution of

atoms in MD computations with the corresponding moments of the concentration in the continuum approach.

In collaboration with Prof. Marzouk at MIT, a Bayesian inference methodology was developed to filter out inherent fluctuations in MD computations, and consequently establish quantitative estimates of mixing rates and of atomic diffusivities. These estimated were validated against experimental data for diffusion of Ni into molten Al. These developments have been described in a journal publication [J1].

Uncertainty propagation in MD systems under equilibrium conditions

We have developed a framework to characterize uncertainty in atomistic systems based on the application of a non-intrusive PC formalism to MD computations of thermodynamic equilibria. Specifically, we have initially focused on isothermal-isobaric MD simulations of TIP4P (four-point) water, and then extended the formalism to transient simulations of concentration driven flow in silica nanopores.

We implemented a stochastic spectral reformulation of the forward problem, assuming uncertainty in a subset of force-field parameters. The intrinsic (thermal) noise present in the atomistic system couples with the parametric uncertainty to yield non-deterministic, noisy MD predictions of the water observables. The uncertain MD predictions are then characterized in terms of polynomial chaos expansions. To this end, we demonstrate two approaches: a non-intrusive spectral projection (NISP) and a Bayesian inference approach. In particular, for the Bayesian inference approach we have developed a new adaptive technique to sample the parameter space and collect the observations, relying on nested Fejér grid points. At a given approximation level, new sampling points are selected in the regions of the space where the differences in PC expansions of the observables of interest obtained at the previous levels exceeds a specified tolerance. This yields a significant reduction in the computational cost with respect to using fully-tensored grids, random sampling, and partially-tensored grids, but comparable accuracy.

We have shown that for the present system, the effect of the thermal noise in the atomistic system can be controlled, and the two methods listed above yield similar results.

A manuscript summarizing these developments has been recently published [J2].

To address the problem of parameter inference, we focus on the inverse problem associated with the MD system considered above for the forward propagation. To illustrate how force-field parameters can be inferred, we studied a synthetic problem where presumed "true" values of the TIP4P model parameters are used to generate a collection of noisy data of density, self-diffusion and enthalpy, and exploited the corresponding PC representations derived in the stochastic reformulation of the forward problem to show that the "true" force-field parameters can be accurately inferred using low-order surrogate models.

A manuscript summarizing our parameter inference methodology has been recently published [J3].

UQ in complex MD systems under non-equilibrium conditions

In order to demonstrate the implementation of our forward/inverse UQ methodology in a realistic complex setting, we developed a silica pore model connecting two reservoirs containing a solution of sodium and chloride ions in water. An ad hoc concentration control algorithm was developed and implemented to simulate a concentration driven counter flow of ions through the pore, with the

ionic flux being the main observable extracted from the MD system. We explored the sensitivity of the system to two physical parameters of the pore, namely the pore diameter and the gating charge. A quantitative analysis of the impact of pore diameter was first conducted, and the results were interpreted in terms of the interplay between size effects and ion mobility. In doing so, we addressed the challenges arising from the heterogeneous nature of the system, and from the substantial effect of the intrinsic noise stemming from thermal fluctuations of the atoms. A manuscript summarizing the model development was recently submitted for publication [J4].

We have also demonstrated the implementation of our UQ methodology for the purpose of characterizing, the sensitivity of the system to the Lennard-Jones energy parameters. To this end, a forward propagation analysis was applied to map the uncertainty in these parameters to the MD predictions of the ionic fluxes. Polynomial Chaos expansions and Bayesian inference were exploited to quantify the impact of parametric uncertainty on the target MD predictions. A Bayes factor analysis was then used to determine the most suitable regression model to represent the MD noisy data. The study showed that the response surface of the sodium conductance can be effectively inferred despite the substantial noise level, whereas the noise partially hides the underlying trend in the chlorine conductance data over the studied range. Finally, the dependence of the conductances on the uncertain potential parameters was analyzed in terms of correlations with key bulk transport coefficients, namely viscosity and collective diffusivities, computed using Green-Kubo time correlations. A manuscript summarizing this development was recently submitted for publication [J5].

Publications

Journal Publications:

J1 F. Rizzi, M. Salloum, Y.M. Marzouk, R.-G. Xu, M. L. Falk, T. P. Weihs, G. Fritz, and O. M. Knio "Bayesian inference of atomic diffusivity in a binary Ni/Al system based on molecular dynamics computations," Multiscale Modeling & Simulation v. 9, pp. $486 - 512$, 2011.

J2 F. Rizzi, H.N. Najm, B.J. Debusschere, K. Sargsyan, M. Salloum, H. Adalsteinsson & O.M. Knio (2012) "Uncertainty Quantification in MD Simulations. Part I: Forward Propagation," *Multiscale Modelling & Simulation* **10**, 14281459.

J3 F. Rizzi, H.N. Najm, B.J. Debusschere, K. Sargsyan, M. Salloum, H. Adalsteinsson & O.M. Knio (2012) "Uncertainty Quantification in MD Simulations. Part II: Bayesian Inference of Force-Field Parameters," *Multiscale Modelling & Simulation* **10**, 14601492.

J4 F. Rizzi,R. Jones, B.J. Debusschere & O.M. Knio (2012) "Uncertainty Quantification in MD simulations of concentration driven ionic flow through a silica nanopore. Part I: sensitivity to physical parameters of the pore," submitted to *J. Chem. Phys.*.

J5 F. Rizzi,R. Jones, B.J. Debusschere & O.M. Knio (2012) "Uncertainty quantification in MD simulations of concentration driven ionic flow through a silica nanopore. Part II: uncertain potential parameters," submitted to *J. Chem. Phys.*.

Other Publications Acknowledging DOE Support:

O1 O.P. Le Maître, L. Mathelin, O.M. Knio & M.Y. Hussaini (2010) "Asynchronous Time Integration for Polynomial Chaos Expansion of Uncertain Periodic Dynamics," *Discrete and Continuous Dynamical Systems* **28**, 199-226.

O2 O.P. Le Maître & O.M. Knio (2010) *Spectral Methods for Uncertainty Quantification*, Springer-Verlag Series on Scientific Computing.

O3 A. Alexanderian, F. Rizzi, M. Rathinam, O.P. Le Maître & O.M. Knio (2012) "Preconditioned Bayesian Regression for Stochastic Chemical Kinetics," submitted to *Journal of Scientific Computing*.

Theses

T1 F. Rizzi (2012) *Uncertainty Quantification in Molecular Dynamics*, Ph.D. Thesis, Department of Mechanical Engineering, Johns Hopkins University.

Lectures and Presentations:

L1 O. Knio, "Spectral methods for uncertainty quantification," Fall Meeting of the Washington-Baltimore Section of SIAM, December 9, 2009.

L2 O. Knio, "Spectral Methods for uncertainty quantification," University of Miami, April 6, 2010.

L3 O. Knio, "Spectral Methods for uncertainty quantification," MIT, April 23, 2010.

L4 O. Knio, "Spectral Methods for uncertainty quantification," Duke University, April 27, 2010.

L5 O. Knio, "Spectral Methods for uncertainty quantification," Free University of Berlin, May 31, 2010.

L6 "Modeling of Transient Reactions in Reactive Multilayer Systems," to be given at Technical University of Berlin, June 2, 2010.

L7 O. Knio, "Spectral Approach to Uncertainty Quantification in Complex Dynamical Systems," 2010 MetStroem Program Meeting, FU Berlin, October 28, 2010. (**plenary**)

L8 O. Knio, "Spectral Approach to Uncertainty Quantification in Complex Dynamical Systems," UMBC, October 15, 2010.

L9 F. Rizzi, M. Salloum, Y.M. Marzouk, R.-G. Xu, M. Falk, T.P. Weihs, G. Fritz, O.M. Knio (2011) "Bayesian Inference of Atomic Diffusivity in a Binary Ni/Al System based on Molecular Dynamics," presented at SIAM Conference on Computational Science and Engineering, February 28 - March 4, 2011, Reno, NV.

L10 F. Rizzi, B. Debusschere, K. Sargsyan, H. Najm, O.M. Knio (2011) "Uncertainty Quantification in MD Simulations: Forward Propagation and Parameter Inference," presented at Workshop on Uncertainty Quantification for Multiphysics and Multiscale Systems, USC, March 7-8, 2011.

L11 F. Rizzi, O.M. Knio, H. Najm, B. Debusschere, K. Sargsyan, H. Adalsteinsson (2011) "Uncertainty Quantification in Molecular Dynamics Simulations: Forward Propagation and Parameter Inference," accepted for presentation at Aachen Conference on Computational Engineering Science," July 13-15, 2011, Aachen, Germany.

L12 F. Rizzi, O. Knio, H. Najm, B. Debusschere, K. Sargsyan, M. Salloum, H. Adalsteinsson (2011) "Uncertainty Quantification MD Simulations: Forward Propagation and Parameter Inference," presented at the DFD11 Meeting of the American Physical Society.

L13 O.M. Knio (2012) "Polynomial Chaos Approaches to Multiscale and Data Intensive Computations," presented at SIAM Conference on Uncertainty Quantification, Raleigh, NC, April 2-5, 2012. (**plenary**)