

309a Hierarchical Multiscale Stochastic Simulations

Abhijit Chatterjee and Dionisios G. Vlachos

Over the years, kinetic Monte Carlo (KMC) simulation has become widespread in numerous areas, including epitaxial growth, catalysis, separations using microporous materials, and biology. Despite the substantial increase in computational power, KMC simulations are often limited to relatively short length and time scales, whereas experimental data often invoke scales beyond the realm of molecular simulation. Recently introduced coarse-grained Monte Carlo (CGMC) [1,2] and adaptive CGMC [3] simulations provide a means to reach much larger length and time scales at considerably reduced computational cost. In both methods, one derives an infinite hierarchy of spatially coarse-grained models from the underlying microscopic physics by varying the level of spatial coarse-graining. It has been demonstrated that aside from satisfying detailed balance and giving the correct dynamics and steady state behavior, coarse-graining gives the correct thermal fluctuations, an essential aspect for noise-controlled phenomena, such as nucleation and nonlinear dynamics. In developing this new multiscale tool, a closure is introduced at the stochastic level. Specifically, local equilibrium and local mean-field had been assumed as a closure to link the microscopic with the coarse-grained scales. Currently, theory justifying this approximation is lacking. More importantly, this multiscale framework is exact, and in practice very accurate, only for long range interactions. However, most practical systems exhibit short-ranged interactions.

In this talk, we present a hierarchical approach to improve the stochastic closure. First, we present a theoretical argument, based on adiabatic elimination ideas of stochastic processes, of when local equilibrium is established. Simulations confirm the theory. Second, we present techniques for obtaining more accurate probability distribution functions (pdf) of species within coarse cells. As an example the quasichemical (QC) approximation for nearest neighbor interactions from equilibrium statistical mechanics is used. A non equilibrium statistical mechanics derivation is presented to identify the conditions; numerical simulations validate the use of QC approximation under such conditions. It is shown that by improving on the stochastic closure, one is able to get accurate simulations even for short-ranged potentials without substantially increasing the computational cost. Tremendous CPU savings of this method vis-à-vis KMC are also demonstrated. Several illustrative numerical examples will be presented to demonstrate the capabilities of the new technique.

References

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