Classical-wave based Simulation of Chemical reaction dynamics: Avoiding the curse of multi-scale time problems in MD simulation

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The limitations in conventional molecular dynamics (MD) simulations, for studying reacting events, are primarily attributed to the small time step, dt, which must be small enough to resolve the shortest duration events (e.g., vibrational motions with time periods of the order of a femtosecond, 10^{-15} s). Such high frequency events may occur billions or trillions of times before one observes the reactive events of interests. This is sometimes called the large "incubation time problem" in MD simulation. An MD simulation long enough to include a single reactive event would take on the order of 10,00 years of CPU time on the fastest computer!

Given the impracticality of direct simulation, a different approach bears consideration, for example, one may completely bypass the time domain and embrace, instead, the spatial domain. This is the primary interest of this talk. Bypassing the time-step in favor of a spatial step requires redefining chemical dynamics as a simulation problem where one wants to find paths between two given configurations on the potential surface. Thus, the problem is cast as a boundary value problem instead of the initial value problem as in MD simulation. To this end, we have recently investigated a new approach based on the most general formulation of classical mechanics, the Hamilton-Jacobi (HJ) formulation. We proposed a slightly modified Hamilton-Jacobi (MHJ) equation which we have solved by a newly proposed fast marching algorithm. The multi-scale time problems on standard MD simulation entirely removed in our proposed method.

We will discuss our method with reference to a 4-well model potential surface that mimics the conformational dynamics of alaninde dipeptide in gas phase. If time permits, we show a few our results of other chemical reactions. Our method provides a robust way to calculate an ensemble of paths. This talk is based up our two recent publications.