Recent Advances in Bottom-up Coarse-Graining of Liquids

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Abstract

Recent advances in theoretical and computational methodology will be presented for studying complex liquid systems and other soft condensed matter across multiple length and time scales. The approach provides a systematic connection between all-atom molecular dynamics, coarse-grained (CG) modeling, and mesoscopic phenomena. At the heart of the approach is the multiscale coarse-graining method for rigorously deriving coarse-grained models from the underlying molecular-scale information. In the design of high-fidelity CG models, the structure and dynamics at the molecular level should be accurately reproduced at the CG level. This talk will cover statistical mechanical theories that have been developed in my group over the last few years allowing for such treatment. Structural correlations are captured by designing the many-body CG potentials of mean force for the CG variables. Nevertheless, many-body correlations present at the finer molecular level are often lost at the simplified CG resolution. I will present two theoretical approaches that can resolve this limitation. One approach is to explicitly include the many-body interactions at the CG level while maintaining efficiency by projecting these many-body interactions onto a lower order interaction. This idea is further utilized to develop a new CG model for water at low computational cost and with great accuracy. Alternatively, many-body correlations can be imposed by introducing internal states to the coarse-grained sites or “beads” similar to quantum mechanical states. By designing these internal states to modulate many-body correlations, these ultra-coarse-grained (UCG) models greatly expand the possible range of systems amenable to accurate modeling, such as hydrophobic aggregation and heterogeneous interfaces. While structural correlations can be preserved by precisely designing conservative interactions in CG models, missing fluctuation and frictional forces often result in accelerated dynamics at a renormalized level and with incorrect kinetics. To develop dynamically-consistent CG models, an alternative strategy based on the excess entropy scaling relationship will be introduced for liquids. Based on our findings on the universality of scaling between the atomistic and CG liquid systems, I will show the scaling relationship for simple liquids in order to systematically bridge the CG and reference molecular-level dynamics. Lastly, a combined theoretical and computational effort in these two directions will be applied to develop a bottom-up reactive CG model of the hydrated excess proton (aka “hydronium cation”) in water, which is critical to many areas of chemistry, biology, and materials science.

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FIGURES

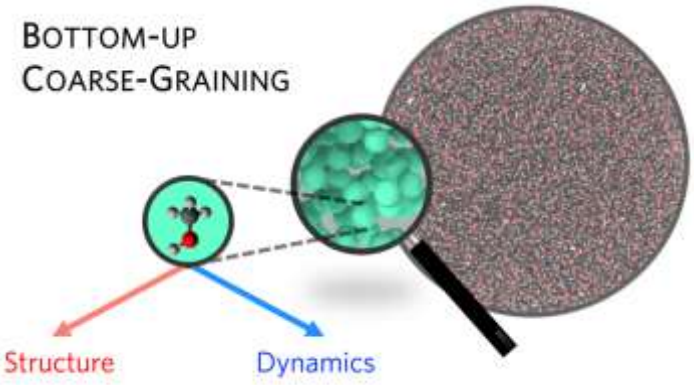

Figure 1: Systematic bottom-up coarse-graining for molecular systems to reproduce the structure and dynamics at the molecular level.