

Molecular and condensed-phase machine learning potentials for alkanes

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Molecular liquids are challenging systems for potential development. They exhibit properties of both gas-phase molecular systems, where the interactions comprise several widely-separated energy scales that must be treated individually, and condensed-phase systems (especially large, amorphous systems), where many-body interactions become important. Their amorphous nature and the high accuracy required to reproduce their macroscopic properties in a reliable and transferable way make molecular liquids an ideal application for machine learning potentials. This work introduces a new approach to the systematic approximation of the first-principles potential energy surface (PES) of a molecular liquid using the GAP machine learning method, which has been successfully applied to model several carbon-based materials including amorphous carbon [Deringer and Csányi, *Phys Rev B* 95(9), 094203] and lithium-intercalated carbon nanostructures [Fujikake et al, *J Chem Phys* 148(24), 241714]. By applying machine learning to separately approximate each physical component of the interaction energy in a full many-body framework and with high and controllable accuracy, we can simulate the liquid accurately across a wide range of temperatures and pressures (with the inclusion of quantum nuclear effects) while gaining physical insight into the inner workings of the fluid. Following the recent success of this approach on predicting the equation of state of compressed fluid methane [M. Veit, S. K. Jain, S. Bonakala, I. Rudra, D. Hohl, and G. Csányi, arXiv:1810.10475], I will compare this approach with a pairwise-molecular model developed on the same system and discuss the role of liquid alkanes as a bridge between the molecular and condensed-phase worlds of interatomic potential development.