Large-Scale Nonadiabatic Dynamics with Machine Learning Hamiltonians

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In chemistry, physics, biology, and materials science, many important processes belong to non-adiabatic dynamics. In particular, electron and exciton long-range dynamics involve a large number of electronic states and vibrational degrees of freedom, and thus quantum decoherence and complex surface crossings should be properly described. In the past years, we have proposed a series of new methods to simulate large-scale nonadiabatic dynamics. The machine learning decoherence time formulas have the potential to systematically improve the accuracy. Based on machine learning of the quasi-diabatic Hamiltonians (DHNet), efficient electronic structure calculations can be realized, and surface hopping dynamics without explicitly using nonadiabatic couplings can be efficiently simulated with our SPADE software. With these method and software developments, we have studied the mechanisms of charge transport in graphene nanoribbons, charge transfer between transition metal dichalcogenides, and oxidation dynamics in large quantum dots. The results are compared with experiment and could be helpful for the design of high-performance materials.

References:

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