Nonadiabatic molecular dynamics with machine-learned Kohn-Sham Hamiltonian mapping

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Nonadiabatic molecular dynamics (NA-MD) simulations can effectively model excited-state dynamics in solar energy materials but face challenges in nanoscale systems due to complex electronic structure calculations. Even methods like density functional theory (DFT) or time-dependent DFT (TD-DFT) can be impractical for long simulations, specially with hybrid functionals. Machine learning (ML) has been increasingly used to reduce computational costs, but most models focus on specific properties, requiring large datasets, and costly training, particularly for nanoscale systems.

In this presentation, I will introduce a conceptually simpler yet novel and general ML strategy from our group for constructing the Kohn-Sham (KS) Hamiltonian matrix at a desired level of theory. We observed that the KS Hamiltonian from a converged charge density maps smoothly from a simple non-self-consistent atomic density guess. Using this, we map an initial guess KS Hamiltonian from one theory level, such as PBE, to a converged KS Hamiltonian at another level, like B3LYP or HSE06. This approach requires fewer training data points, accelerates calculations with high accuracy, and is scalable and applicable to various nanoscale materials. I will also demonstrate how atomic orbital matrices, even when obtained at a low level of theory, can be used as feature vectors, bypassing the need for neural network for feature extraction. Additionally, I will show our implementation of a user-friendly interface in Libra, making this method accessible and practical for a wide range of users. Finally, I will demonstrate how this model, when applied to NA-MD simulations of hot-carrier relaxation dynamics, produces timescales within the error margins of conventional methods.