Machine Learning Molecular Dynamics for Understanding Nonadiabatic Surface Reactions

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Abstract: Transferring energy from solid surfaces to chemical bonds of adsorbed species is a fundamental process in heterogeneous catalysis. To a great extent, the dynamics of molecule-surface interactions driven by thermal energy, i.e., heat, can be well described within the framework of the Born-Oppenheimer approximation. New reaction channels often open up in response to electronic excitations via surface-mediated energy transfer. In this process, the energetic electrons or holes can scatter into surface species and heat up vibrational modes of adsorbates. This process involves not only the electronic ground state but also the excited state, and it is nonadiabatic with respect to the nuclear motions. In the weak coupling limit, the nonadiabatic energy transfer on metal surfaces can be modeled using only the ground-state potential energy surfaces and the nonadiabacity is taken into account with electronic friction contributions in Langevin dynamics by solving Newtonian equations with a thermal fluctuation term. However, it is extremely time consuming to compute electron-phonon coupling strength for large systems. In this poster, we will discuss our most recent development of a machine learning molecular dynamics approach that propagates the system by 'learning from data'. Machine learning algorithms, such as the artificial neural networks, can use past trajectories as training datasets for fast and accurate prediction of interatomic forces and friction force, thus allows us to perform statistical analysis of nonadiabatic surface reactions. We benchmark the approach for activation of oxygen from hollow to bridge and CO oxidation on Ru(0001) because of strong interests in understanding laser-induced chemistry on metal surfaces. Local Density Friction Approximation (LDFA) was used to calculate the electronic friction. Statistical analysis from ~5000 molecular dynamics trajectories shows the oxygen hopping under ultrafast laser pulse is electron driven in nature.

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