Coarse-graining of electronic dynamics in nonadiabatic dynamics at surfaces

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Nonadiabatic effects that arise from the concerted motion of electrons and atoms at comparable energy and time scales are omnipresent in chemical dynamics at metal surfaces. Excited (hot) electrons can measurably affect molecule-metal reactions by contributing to state-dependent reaction probabilities. Such effects can either be treated by simultaneous propagation of electronic and nuclear degrees of freedom, for example via surface hopping molecular dynamics or by coarse-graining out electronic motion via electronic friction theory. Molecular dynamics with electronic friction enables the description of dynamics under the influence of dissipative forces that arise from low-lying electronic excitations.

In this talk, I will present electronic friction theory and how it arises from approximations to time-dependent quantum mechanics. I will describe the mathematical and physical properties of electronic friction and how it affects physically measurable properties. I will further present our recent efforts to combine a tensorial representation of electronic friction based on Functional Theory with high-dimensional machine-learning-based representations of energy and friction landscapes to perform molecular dynamics with electronic friction simulations. Finally, I will conclude with detailed analysis of the successes and limitations of this approach and will highlight the exciting mathematical challenges that lie ahead.