Synthesizing first-principles simulation, machine learning, and experimental strategies for the design and analysis of a new class of high-performance battery electrolytes exploiting the Grotthuss structural diffusion mechanism

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Reliable theoretical prediction of complex chemical processes in condensed phases requires an accurate quantum mechanical description of interatomic interactions. If these are to be used in a molecular dynamics calculation, they are often generated "on the fly" from approximate solutions of the electronic Schrödinger equation as the simulation proceeds, a technique known as *ab initio* molecular dynamics (AIMD). However, due to the high computational cost of these quantum calculations, alternative approaches employing machine learning methods represent an attractive alternative and have become increasingly popular. As the adoption of machine-learning potentials becomes more widespread, it is important to consider how simulations employing them should be carried out. Specifically, as they do not implicitly include nuclear quantum effects, these effects must be treated explicitly, for which the most efficient approach involves the use of Feynman path integral techniques. This is especially important for processes involving light elements. In this talk, I will discuss how state-of-the-art machine learning potentials can be combined with path-integral molecular dynamics to address a variety of challenging chemical problems that have unexpected quantum behavior. In particular, I will discuss a new class of battery electrolytes that, by harnessing their unusual quantum character, could lead to breakthrough performance. Discovery of the mechanism of charge transport in these systems could only be achieved by the development of an equivariant transformer network interatomic potential model. I will discuss new quantum techniques, including path integrals and Green's functions, we are developing for studying electronic charge transfer between molecular species and between molecules and electrode surfaces.