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Exploring chemical reactivity at interfaces with simulations and machine learning

by

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Numerical simulations are essential for gaining a molecular-level understanding of chemical processes, offering insights that complement experimental techniques. For simulations to provide a realistic and predictive description, they must combine an advanced electronic structure representation of the reacting system with an explicit inclusion of surrounding environmental molecules. Additionally, they should be scalable to large systems and capable of capturing dynamics over long timescales. However, these requirements present significant computational challenges, often forcing compromises that reduce the accuracy and predictive power of conventional methods.

Recent advancements in machine learning offer transformative solutions to these challenges. By training neural network potentials on high-quality quantum mechanical data, it is now possible to simulate chemical reactions in condensed phases with remarkable efficiency and accuracy[1]. This seminar will explore the principles of training such potentials, with a focus on addressing the unique challenges posed by reactive systems.

We will illustrate the power and limitations of these approaches using examples from our recent work, including Grotthuss proton transport in water[2], the acidity of the air-water interface, and peptide bond formation in aqueous droplets[3]. Finally, we will discuss how these techniques can extend beyond reaction dynamics to applications in spectroscopy, with a particular emphasis on nonlinear vibrational sum-frequency generation[4]. These developments illustrate the potential of machine learning to transform the field of reactive molecular simulations, bridging the gap between accuracy and practical applicability.

(1) Gomez, A.; de la Puente, M.; David, R.; Laage, D. Neural network potentials for exploring condensed phase chemical reactivity. *C R Chimie* **2024**, in press.
(2) Gomez, A.; Thompson, W. H.; Laage, D. Neural-network-based molecular dynamics simulations reveal that proton transport in water is doubly gated by sequential hydrogen-bond exchange. *Nature Chemistry* **2024**, *16*, 1838.
(3) David, R.; Tuñón, I.; Laage, D. Competing reaction mechanisms of peptide bond formation in water revealed by deep potential molecular dynamics and path sampling. *J Am Chem Soc* **2024**, *146*, 14213-14224.
(4) de la Puente, M.; Gomez, A.; Laage, D. Neural Network-Based Sum-Frequency Generation Spectra of Pure and Acidified Water Interfaces with Air. *J Phys Chem Lett* **2024**, *15*, 3096-3102.