C8

Description Is Not Understanding: From Molecular Trajectories to Atomistic Mechanisms

C. Dreßler[1]

[1] *TU Ilmenau, Germany*

Recent advances in machine-learned interatomic potentials (MLIPs) have fundamentally changed atomistic simulation. Molecular dynamics (MD) trajectories with near ab initio accuracy can now be generated on time scales that allow statistically converged sampling of rare events and direct determination of kinetic observables. As a consequence, the computational bottleneck has shifted from generating trajectories to understanding them. Understanding, however, requires more than describing representative trajectories or visualizing rare events. Atomistic mechanisms cannot be observed directly - they are scientific hypotheses that must be inferred from molecular dynamics and tested quantitatively. The central question therefore becomes: When does a proposed atomistic mechanism correctly describe the underlying dynamics? In this talk I will argue that atomistic mechanisms should be formulated as physically motivated coarse-grainings of molecular dynamics. Instead of identifying abstract metastable states solely through statistical learning, I will construct state spaces from chemically meaningful descriptors, such as lattice-site occupancies, local coordination environments, or defect configurations. Transition probabilities sampled from long MD trajectories then define Markov state models whose degree of Markovianity provides an objective measure of the validity of the proposed mechanism. Within this framework, the Chapman–Kolmogorov test becomes a quantitative criterion for testing mechanistic hypotheses rather than merely validating kinetic models. Using lithium diffusion in lithium silicides and sulfur-vacancy diffusion in monolayer MoS_2 as examples, I will demonstrate that different transport processes become Markovian only for different physically motivated state spaces. These state spaces reveal the elementary transport mechanisms and allow competing mechanistic hypotheses to be compared quantitatively.[1,2] Finally, I will discuss the construction of accurate MLIPs. I will demonstrate that fine-tuning pretrained universal foundation models provides a robust and efficient strategy for obtaining ab initio-quality force fields largely independent of the underlying model architecture.[3] Moreover, I will show that a single fine-tuned model can accurately describe an entire class of compounds, opening the door to transferable, high-fidelity simulations across chemically related materials.

[1]Qaisrani . . . , Dreßler, JCTC, 2026 22 (11), 5373-5387 https://doi.org/10.1021/acs.jctc.5c02035
[2]Flötotto, . . . , Dreßler, . Small 2026 22(20), e10679 https://doi.org/10.1002/smll.202510679
[3]Hänseroth, . . . ,Dreßler. JCPL, 2026. https://doi.org/10.1021/acs.jpclett.5c03801.