Novel free energy sampling approaches for phase transitions, mass transport and crystal structure prediction

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Abstract: The detailed investigation of fundamental phenomena, from phase transition mechanisms, to transport processes, is hampered by an intrinsic separation of process time scales. If nucleation is involved for example, methods like molecular dynamics suffer from impractical to insurmountable delays in observing even a single events. In recent years accelerated methods are providing qualitatively novel insights into critical processes, from the investigation of first order phase transitions in solids, to biological systems. Metadynamics for one enhances event probability by defining collective variables (CV), to rapidly drive a system away from more probable configurations.

We introduce a novel scheme for the mechanistic investigation of solid-solid phase transitions, which we dub metashooting. Therein, we combine transition path sampling molecular dynamics and metadynamics. This scheme allows for both a complete mechanistic analysis and a detailed mapping of the free energy surface. This is illustrated by performing metashooting calculations on the pressure-induced B4/B3→B1 phase transition in ZnO. The resulting free energy map helps to clarify the role of intermediate configurations along this activated process and the competition between different mechanistic regimes with superior accuracy.

In another difficult area, the understanding of mechanisms of ion translocation in battery materials, we have designed specific, structure factor based CVs to enhance ion mobility. We benchmark this method against entropy-based collective variables,

show the richness of details these techniques can disclose, and expand on their applications to crystal structure prediction.

S.A. Jobbins, S.E. Boulfelfel, S. Leoni, Faraday Discuss., 2018.