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Computing Transport Properties in Solid-State Materials from the Shapes of Potential Energy Landscapes

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The structure and dynamics of a material are fundamentally governed by the complex interplay of potential energy landscapes encountered by individual atoms within the system. These landscapes encode critical information about the material's properties through their geometric and topological features. For instance, configurations of particles within a solid are determined by the shapes and presence of energetic basins, while the self-diffusion of mobile species is dictated by the connectivity and geometry of these basins, which form diffusion pathways. Understanding atomistic-scale diffusion processes in solids is essential for a wide range of applications, including predicting lithium-ion transport in solid-state battery cells and designing membranes for separation technologies such as carbon capture and water purification. Although computational modeling offers powerful tools for probing these phenomena, significant challenges remain in bridging the relevant spatial and temporal scales necessary to capture the full complexity of real materials. [1]

In recent years, we have developed a novel multiscale modeling framework to predict and elucidate the (self-)diffusion behavior of ions and small molecules within crystalline structures. This methodology involves constructing and analyzing the topology and geometry of the potential energy landscape, and building a statistical model by calculating hopping rates between energy minima using transition state theory and kinetic Monte Carlo simulations. Our approach automates and accelerates self-diffusion predictions in crystalline periodic systems – achieving speedups of several orders of magnitude compared to state-of-the-art molecular dynamics simulations. Additionally, we have investigated the role of configurational entropy and demonstrated how its inclusion yields a more accurate representation of the effective free energy landscape experienced by mobile particles.[2,3,4]

We are currently extending this framework to non-periodic material systems, such as solid polymer electrolytes and multiphase composite materials. This expansion introduces new challenges, particularly in accounting for spatial and temporal dependencies during the sampling and analysis of the free energy landscape.

References

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