Predicting crystal structures and ionic conductivity in solid-state electrolytes using Machine Learning Interatomic Potentials

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Machine learning interatomic potentials (MLIPs) have revolutionized computational materials modeling by enabling accurate simulations at length and time scales far exceeding the reach of traditional ab initio methods [1-2]. Coupled with advances in computational resources, this progress allows for the investigation of more realistic and complex systems, such as components in next-generation batteries.

The growing demand for safer and higher-performance lithium-ion (Li-ion) batteries has driven the search for solid-state alternatives to flammable liquid electrolytes. Among these, Li-containing ternary halides, particularly Li3MX6 (M = trivalent metal, X = Cl, Br), have attracted significant attention due to their high room-temperature ionic conductivities and robust oxidative stability, which makes them compatible with oxide-based cathode materials [3]. These materials exhibit diverse crystal structures - typically trigonal, orthorhombic, and monoclinic - with their stability and ionic conductivity strongly influenced by the specific elemental composition and crystal symmetry [4-5].

I will present new insights into the composition-structure-property relationships in halide solid electrolytes (SEs) for all-solid-state Li-ion batteries. I will demonstrate how variations in cation and anion chemistry affect the stability of crystal structures, offering strategies to enhance ionic conductivity. A key challenge arises from the partial occupations of specific Wyckoff sites within the structures (random vacancy distribution and solid solutions), leading to over 50,000 symmetry-inequivalent atomic arrangements for some compositions. To address this complexity, I employ MLIP-based simulations [6-7] to efficiently estimate formation energies, rank inequivalent configurations, and subsequently conduct molecular dynamics simulations to probe ionic transport properties. I will present the methodology in details, including an introduction to MLIPs and active-learning approaches used to improve their accuracy and performance.

References:

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