De Novo Computational Methods for Simulating Energy Materials

Transport of phonons, ions, electrons, and excitons across interfaces strongly affects the performance and efficiency of thermoelectric materials, solid-oxide fuel cells, batteries, and solar cells. Predicting transport phenomena through interfaces in functional materials in these devices, which are mainly inorganic solids, demands fast and reliable methods applicable to systems with hundreds of thousands of atoms. The widely used density functional theory (DFT) methods, which are limited to simulate diffusion in single crystals and incapable of including sufficiently many atoms to model interfaces and boundaries, lack the necessary reliability as they fail to accurately describe charge and bond rearrangements.

The goal of this research is to revolutionize a computational design of novel materials for energy storage and conversion by developing new generations of charge-transfer force fields for materials simulations, abbreviated as QTFF, and an automated fitting approach, which will rely on high-accuracy *ab initio* calculations using local extensions of coupled-cluster (CC) theory to thousands of atoms. The CC-based QTFF potentials will be the first of a kind, enabling a proper description of charge transfer in redox reactions in materials, excess electrons, and applied electric fields in inorganic solids.

The proposed research will enable breakthrough computational studies of grain boundaries and interface structures at the levels of accuracy that have not been possible before, helping the design of materials with desired properties. With a strong, well-integrated team, the proposed approach will constitute a quantum leap in designing polycrystalline thermoelectric, photovoltaic, and ion conducting materials, accelerating innovations in energy conversion and storage technologies.