Advanced computational approaches to study local and collective defects behaviors in hard, soft and molecular materials

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Understanding and predicting the macroscopic properties of materials starting from microscopic interactions and atomic-scale fluctuations is a fundamental challenge in materials science. In this thesis, we propose a conceptual framework that systematically links atomic-scale fluctuations to the emergence of persistent structural defects, whose local or collective dynamics ultimately determine macroscopic material responses. To illustrate and validate this framework, we employ molecular dynamics (MD) simulations across different classes of systems, spanning both hard matter, specifically metallic systems and molecular crystals,and soft matter, such as lipid membranes and water.

As a first demonstrative examples we use metals and their well known interesting properties. Using atomistic models of metallic systems, we first explore dislocation nucleation in defect-free copper crystals subjected to shear deformation, characterizing this phenomenon as a thermally activated rare event. By rigorously identifying the critical transition states through committor analyses and stochastic modeling, we provide a detailed microscopic picture of how localized atomic fluctuations seed persistent structural defects. We then extend this analysis to single-crystal copper under tensile deformation, systematically tracing the progression from transient local rearrangements and local dynamical fluctuations to collective dislocation dynamics and eventual fracture events, employing innovative, data-driven descriptors to characterize atomic environments and their dynamical evolution. Further complexity is introduced by studying polycrystalline copper systems, demonstrating how grain boundaries act as highly dynamic regions facilitating defect propagation and grain coalescence, thereby enriching our understanding of mechanical response in realistic, heterogeneous materials.

We apply our methodology to Phase-Transition Molecular Solar Thermal (PT-MOST) materials, exploring how photo-induced molecular-level dynamics govern macroscopic phase transitions and energy storage capacities.

Finally, in the soft matter context, we adopt an inverse engineering approach, optimizing coarse-grained force fields for lipid membranes and water systems by systematically integrating atomistic-level structural features with macroscopic experimental observables. This approach not only yields force fields capable of reproducing complex phenomena such as lipid phase separation and vesicle fusion but also enhances classical water models, clarifying the intricate relationship between microscopic interactions and macroscopic thermodynamic properties.

Altogether, these studies illustrate the profound interconnection between microscopic fluctuations, their conversion to persistent defects, which local or collective emergence and dynamics controls macroscopic material behavior and properties, providing fundamental insights and practical routes for the rational design of advanced materials.