The behavior of many complex systems in Nature is governed by local events/rearrangements that may generate collective cascade phenomena. Detecting such local events and controlling their emergence and propagation across multiple scales represents a challenge for scientific discovery and materials design. Acting as a computational lens on the physical world, multiscale modeling and simulations play a crucial role in the exploration of dynamical processes in complex systems ranging from soft to hard materials. Molecular Dynamics (MD) simulations, in particular, allow the investigation of the behavior of systems at a high-resolution (atomistic) detail, thus complementing the limitations of some experimental techniques. Over the past decades, both the development of realistic models and the interpretation of MD simulations have been impacted by Machine Learning (ML), which has proven to be particularly incisive in driving innovation in materials science. However, due to the complexity underlying the dynamics of many molecular systems, the adoption of common and general ML strategies still represents a core issue in molecular modeling and simulations frameworks.

After conveying the potential role that molecular modeling and simulations can play in the exploration of Nature, this PhD Thesis reflects on open challenges in the field. Taking them as opportunities, herein data-driven strategies for (i) extracting information on key events occurring within complex systems' trajectories and (ii) developing and optimizing molecular models at various scales are presented. In the attempt to fully harness the potential of MD simulations, the aim is to provide general approaches enhancing the exploration of complex dynamical systems. Firstly, the focus is on novel descriptors deciphering intricate physical mechanisms and uncovering dynamical patterns at the origin of emergent behaviors in a wide range of systems. Building on the concept of tracking the motion of particles within MD trajectories, these descriptors enable the detection, classification and correlation of dominant fluctuations which are key to understand phenomena underpinning complex systems' behaviors. Thus, general data-driven approaches are proposed and proved to offer unique information on the physics of a wide variety of complex molecular systems ranging from soft to hard matter. Secondly, this Thesis emphasizes the importance of addressing relevant molecular design tasks for modeling target phenomena. Key challenges related to the design of soft materials are taken as opportunities to develop novel strategies coupling traditional molecular modeling with ML. In particular, automatic data-driven approaches for optimizing the accuracy of Coarse-Grained (CG) models and for exploring the vast design space of complex soft architectures are presented, paving the way to the rational design of systems with target functionalities. As a final work, this Thesis shows an application of MD to a complex system of great interest to today's scientific landscape, *i.e.*, an organic mixed ionic-electronic conductor (OMIEC). Unveiling the complex interplay between ions, polymer and water within an OMIEC, MD is proven to be crucial for the development of next-generation materials.

Overall, this PhD Thesis fosters the exploration of complex dynamical systems *via* advanced computational approaches. The proposed strategies conceive a future where characterization, modeling and application work together leading to new discoveries.