Short Course – DSC, 27-29 May 2025 (14.00-17.00 hrs, Aula COB1)

Unravelling the Dynamical Behavior of Heterogeneous Catalysts under Reaction Conditions through Machine Learning Simulations

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The traditional view of heterogenous catalysts as static entities is increasingly challenged by the evidence that their activity often stems from their ability to undergo significant transformations at the *operando* conditions. This dynamic behavior is essential for understanding how catalysts maintain stability and activity under the harsh conditions of industrial reactors, which involve high temperatures, pressures, and intense flows of reactants and products. To this purpose, the characterization of materials at the atomistic scale is crucial, since it can provide unique insights to improve their performance, especially in *operando* conditions. Obtaining an experimental characterization of these systems with atomic resolution is extremely challenging, while *ab initio* atomistic simulations are, in principle, an ideal tool to investigate dynamical phenomena at the atomic scale.
Despite their potential, the prohibitive cost of *ab initio* molecular dynamics has limited their application to study realistic systems. Recently, machine learning (ML)-based interatomic potentials have emerged as a valuable solution to reconcile the accuracy of *ab initio* methods with the efficiency of classical force field. These ML potentials are trained to reproduce the energy and forces from a large set of quantum mechanical calculations, and they can be optimized on small system sizes and then used to simulate much larger systems for long (e.g. nanoseconds) timescales. Despite their promise, the construction of these potentials for complex, multicomponent reactive systems represents a complex endeavor, requiring a comprehensive training set that incorporates all relevant configurations. To this end, enhanced sampling methods such as metadynamics, allow to speed up the process of collecting configurations and generating uncorrelated structures which cover the complete energy landscape.

In this series of lessons, we aim to present a comprehensive overview of the current state-of-the-art methodologies in the field. We will review various approaches for training machine learning (ML) potentials and collecting the required training datasets, while highlighting the advantages and limitations of each approach, depending on the specific application. Additionally, the course will feature illustrative applications aimed at deepening participants' understanding of structural dynamics, chemical reaction kinetics, and transport phenomena within a series of heterogenous catalysts for ammonia synthesis and decomposition.

The hands-on section will focus on the full training procedure of a ML-potential on a few case studies, using Quantum Espresso, DeepMD-Kit and LAMMPS softwares.