Machine Learning Polaron Charge Dynamics

Cesare Franchini[1, 2]

[1]University of Vienna, Austria
[2] University of Bologna, Italy

Modelling excess charge carrier transport in semiconductors is essential for a wide range of technologically relevant applications, including (photo)catalysis, optoelectronics, and energy storage devices such as batteries. Excess electron or hole carriers can be generated through light–matter interaction or chemical doping; however, accurately describing their mobility, excitation dynamics, and recombination processes from first-principles atomistic simulations remains highly challenging.

Conventional ab initio molecular dynamics (AIMD) is limited to timescales of a few picoseconds and system sizes on the nanometer scale. Furthermore, the description of carrier lifetimes requires going beyond ground-state properties to capture non-equilibrium processes.

In polarizable semiconductors, excess charges are typically coupled to lattice distortions via short-range electron–phonon interactions, leading to the formation of small polaron quasiparticles. These polarons are transported through the material via thermally activated hopping mechanisms and constitute a fundamental ingredient for understanding charge carrier mobility in semiconductors, with a strong impact on virtually all related applications [1].

In this seminar, we present recent advances in modelling small polaron mobility in transition metal oxide semiconductors, with a primary focus on TiO_2. Our approach extends the capabilities of AIMD by employing machine-learned interatomic potentials that accurately capture both the formation and transport of electron and hole polarons. In particular, through our software based on the equivariant message-passing neural network LEOPOLD (LEarning Of POLaron Dynamics), we are able to access and describe small polaron mobility at the nanoscale and predicting modibility in good agreement with experimentally available measurements [2].

The resulting polaron trajectories are subsequently analyzed within a non-adiabatic framework, enabling the extraction of carrier lifetimes through the calculation of non-adiabatic couplings using surface hopping techniques.

References

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