Dynamics of oxidation states in transition metals of Li-ion battery cathodes

Cristiano Malica[1] and Nicola Marzari[1,2]
[1]University of Bremen, D-28359 Bremen, Germany
[2]EPFL, CH-1015 Lausanne, Switzerland

cmalica@uni-breme.de

In the context of the energy transition towards renewable energy sources, rechargeable batteries play a crucial role. Their operation is inherently linked to the redox reactions that occur at the electrodes, where typically the atoms of transition metals undergo changes in oxidation states, thus altering the charge and the number of electrons possessed. Recent studies have shown how the application of density-functional theory, including the use of extended Hubbard functionals (i.e., DFT+U+V), provides an accurate and detailed description of the variations in oxidation states of transition metal atoms in the cathodes of lithium batteries, in response to changes in lithium concentration. In this study, we extend the use of this theory to finite temperature conditions through ab-initio molecular dynamics simulations, examining the dynamic variations of oxidation states of transition metals in representative cathodic materials. The results obtained feed the training of an equivariant machine learning potential, with which we analyze the predictive capabilities of the model.