An X-ray spectra simulations workflow based on machine learning: applications to Li-ion battery materials

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In the framework of battery research, core-level spectroscopies are established strategies to assess the evolution and composition of the Solid Electrolyte Interphase (SEI). While these techniques provide valuable insights into atomic and electronic structures, their interpretation in complex systems is not straightforward, emphasizing the importance of theoretical insights from ab-initio approaches. On the other hand, the time required for computing an X-ray spectrum scales linearly with the number of non-equivalent atoms, becoming prohibitively expensive for complex amorphous materials. This drawback can be addressed by employing a surrogate model that combines the precision of ab initio methods with computational efficiency [1]. We trained machine-learning (ML) models based on Kernel Ridge Regression (KRR) [2] and Neural Network (NN), using atom-density descriptors for predicting X-ray Photoelectron Spectroscopies (XPS), by using core-electron Binding Energies (BE) as the target quantity. A comprehensive automated AiiDA workflow [3], integrating first-principles XPS simulation with sample sub-selection via Farthest Point Sampling (FPS), was employed to generate the critical amount of data needed for the training process. The ML models were trained on a representative dataset comprising about 2000 lithiated Carbon-based structures, previously obtained through an evolutionary algorithm (EA) combined with molecular dynamics (MD) and clustering techniques [4]. The model was tested on a validation dataset of ~500 structures, achieving an accuracy of 0.1 eV, consistent with the typical experimental resolution of XPS. This approach holds promise for further application to complex structures relevant to Li-ion battery materials.

References

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