Developing computational tools for the core-level spectroscopy of amorphous carbon

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The aim of this project is the development of highly-accurate, but also computationally-feasible, simulation tools for the inner-shell spectroscopy of amorphous carbons (a-C). a-C are a class of materials with different fractions of sp^2 and sp^3-bonded carbons and a variety of structural motifs [1]. Due to their superior mechanical properties, a-C are used as coating materials and show also potential as materials for electrochemical detection of biomolecules. Core-level spectroscopy techniques, such as X-ray photoelectron spectroscopy (XPS), are important tools to analyze the structure and composition of complex materials. Due to the disordered nature of a-C, XPS is a valuable tool to understand its atomic structure, and to help in identifying the connection between material microstructure and application performance. However, the interpretation of experimental XPS spectra of a-C is not easy since it is difficult to establish the precise origin of the C1s peaks. Accurate computational tools are thus important to support the interpretation of experimental data.

In this work, we generate computational XPS data for different sp^2 and sp^3 sites of a-C structures using density functional theory (DFT) and the GW method. The latter is a Green's function-based method that yields highly-accurate core-excitation energies [2]. Here, we present convergence studies for different a-C clusters with respect to system size. We demonstrate also that the computational parameters for the DFT and, in particular, GW calculations are well converged.

Depending on the system size, GW can be computationally up to several orders of magnitude more expensive than DFT. Conversely, DFT does not predict core-level ionization potentials with sufficient accuracy. The ultimate goal of this project is to use a machine-learning approach to predict the difference between DFT- and GW-predicted core-level excitation energies for a-C. In production runs, DFT calculations will be performed and used as a starting point to which the learned correction will be added. This will eventually allow us to achieve GW accuracy at DFT cost. While in this initial work we tackle a-C as an optimal case study due to its complexity and technological importance, we expect that in the future the developed methodology will help in the interpretation of experimental X-ray spectra for any choice of material.

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