Development of Artificial Intelligence-based approaches for ultrafast X-ray Photoelectron Spectroscopy in large systems

Solène Oberli*

**Laboratory for Ultrafast X-ray Sciences (LUXS), Ecole Polytechnique Fédérale de Lausanne, Switzerland*
Laboratory of Theoretical Physical Chemistry (LCPT), Ecole Polytechnique Fédérale de Lausanne, Switzerland

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Molecules are composed of nuclei and electrons which form chemical bonds. Their motions are as fast as 10^{-15} and 10^{-18} seconds, respectively, and govern the outcome of chemical reactions. In order to observe these ultrafast processes, we thus need very special and powerful tools. In particular, X-ray Free Electron Lasers (XFELs) are able to generate highly intense ultrashort X-ray pulses - or bursts of light - by accelerating electrons up to relativistic speed and sending them through a sequence of magnets. To probe in real time the electron and nuclear rearrangements, pulse duration should be in the same order or shorter than these motions. In addition to this extremely high temporal resolution, X-ray pulses offer atomic spatial resolution, which allows us to trigger and probe the dynamics at a particular site in the molecule. X-rays interact primarily with core electrons which are deeply localized close to the nuclei. The binding energy of these electrons is specific of the element, such that we can control which atom is ionized in the molecule by varying the energy of the X-ray photon. Moreover, the binding energy of core electrons is sensitive to the local chemical environment. The X-ray Pholoelectron Spectroscopy (XPS) technique exploits this property: by measuring the energy of the electron ejected from the molecule by the X-ray, we can observe the local electron redistribution and changes in nuclear geometry while the chemical reaction takes place.

In this project, we studied the dynamics taking place in cationic states of a molecule using a time-resolved XPS. In this way, we are able to grab a 'movie' of the coupled electron and nuclear dynamics. In order to simulate the processes in play, we use a surface-hopping semiclassical propagation scheme[1], in which the electron motion is treated quantum mechanically, while the nuclear dynamics is performed classically using a swarm of independent trajectories. These simulations are very expensive and may be hard to converge. To overcome this limitation, we started to supplement the propagation code with a Deep-Learning algorithm to accelerate the calculations and made it suitable for the treatment of larger molecules. Deep-Learning models are based on Neural Networks which are biology-inspired statistical algorithms. We believe that the use of Artificial Intelligence-based methods coupled to traditional quantum/semi-classical algorithms is a promising avenue for studying ultrafast phenomena in complex systems, and more generally to investigate the fundamental properties of matter.

[1]Code developed by Jesús González Vázquez, Universidad Autónoma de Madrid, Spain.