Tracing Radiation Damage in Molecules with X-ray Radiation and Simulations

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Abstract: In this talk, I will give an overview on the implementation and capabilities of our molecular electronic structure toolkit XMOLECULE. Applications are discussed that demonstrate our approaches for simulating ionization-induced dynamics and how it can be probed via x-ray radiation. The calculations provide crucial insights for tracing radiation-damage dynamics in molecules with x-ray radiation.

The advent of new x-ray light sources, the x-ray free-electron lasers (XFEL), poses several challenges for the theoretical modeling of experiments involving interactions with x-ray radiation. One of the crucial features of XFEL radiation is its high intensity that is able to ionize molecules several times. We model this multi-photon multiple ionization dynamics by solving rate equations for electronic populations. To that end we employ an efficient electronic structure method that can address multiply ionized electronic configuration and provides Auger decay rates and ionization cross sections. Moreover, the electronic structure calculations give insights into how charges in a molecule are redistributed in response to the multiple ionization steps (see Fig. 1). This charge redistribution is key for many experiments at XFELs[1-5]. In my talk I will describe different aspects of our calculation scheme. For a simple water molecule, I will show, how our simulations provide crucial interpretation of the final momentum data in an XFEL induced Coulomb explosion. In particular, I demonstrate how the simulations helped to interpret recent experimental data to uncover structural dynamics of water molecules after inner-shell ionization and Auger decay.

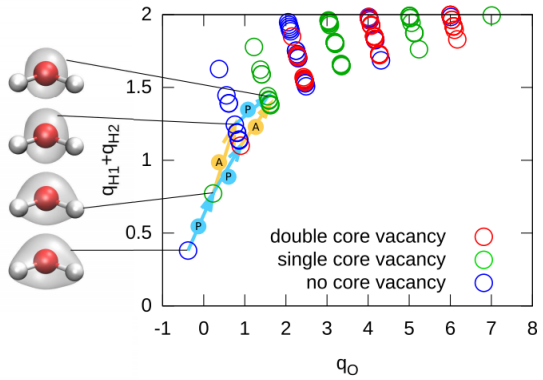

Fig. 1 Partial charges in H_2O for various hole configurations that are accessed by multiple inner-shell ionization and subsequent Auger decay. From Ref.[2]. https://doi.org/10.1103/PhysRevA.94.023422

Ultrashort x-ray pulses at XFELs or from high-harmonic-generation sources provide a tool for probing ultrafast structural dynamics. Since x-ray light interacts preferentially with inner-shell electrons, x-ray absorption spectroscopy can probe the local chemical environment in the vicinity of a specific atom. Along a complementary research direction we are investigating in our group the capabilities of probing ultrafast photo-induced dynamics using x-ray absorption spectroscopy. In particular, we aim to address the dynamics induced by deep-valence holes that are hard to address with sophisticated electronic structure models. In my talk I will present our simulation scheme, in which we combine ab-initio fewest-switching-surface-hopping molecular dynamics simulations with calculations of x-ray absorption spectra [6-9]. I will show how our simulations played a decisive role for tracing ionization-induced dynamics in liquid environments.

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