Toward First Principles Simulations of Biological Matter under Ionizing Radiations

Aurelio Alvarez-Ibarra[a], Angela Parise[a,b], Xiaojing Wu[a], Karim Hasnaoui[c], Dennis R. Salahub[d] and Aurélien de la Lande[a]

[a] *Laboratoire de Chimie Physique, CNRS Université Paris Sud, Université Paris Saclay, France,* [b] *Universita della Calabria, Italy,*[c] *Institut du développement et des ressources en informatique scientifique,* [d] *University of Calgary, Canada*
aurelien.de-la-lande@u-psud.fr

The transient collision (10-17 s) of high-energy-transfer particles with biological matter results in ionization or excitation of its constituent molecules. Huge amounts of energy are deposited locally, typically several tens of eV. These early physical events produce a myriad of reactive radical species that are at the source of cascades of chemical processes spanning several spatial and temporal scales. The physical chemistry of these ultrafast processes are not well understood at the present time. During this seminar I will introduce an original set of methodologies that we have devised to investigate these phenomena from first principles [1,2]. These are based on Real-Time Time-Dependent Density Functional Theory (RT-TDDFT) and Ehrenfest Molecular Dynamics (MD) simulations. A hybrid scheme coupling these approaches to polarizable force fields have been devised to simulate large biological systems. This implementation includes explicit time propagation of the electric fields mediating interaction between the quantum and classical parts of the system[3] Theses methodologies have been implemented in deMon2k. Our approach allows simulations of collisions of molecules with HET, and subsequent ionization, ultrafast charge migration, energy relaxation/dissipation on the attosecond time scale (cf. Figure). Ehrenfest MD further gives access to non-adiabatic chemical reactivity taking place in the first femtoseconds after irradiation. I will introduce the methodologies and applications to various questions of current high interest in radiation chemistry.

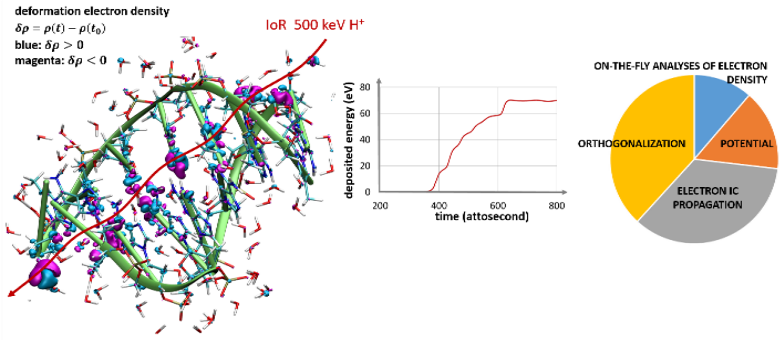

Figure : Irradiation of 10-base DNA double strand by a 500 keV proton particle The left panel depicts isosurfaces of the deformation density just after irradiation. On the right panel a graph shows the increase of deposited energy as a function time (middle) and the main computational tasks involved in the simulation (right)

References

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