First principles simulation of attosecond electron dynamics in complex systems

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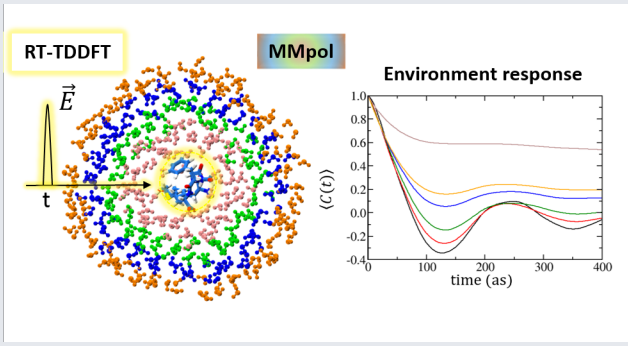
Recent years have seen a growing interest in the electron dynamics taking place in molecules when they are subjected to an external perturbation. This interest has been stimulated by progress in attosecond spectroscopy that now gives access to details on electron dynamics. The realm of sub-femtosecond electron dynamics involves fascinating processes such as ultrafast charge migration, Auger decays and Intra Coulomb Decays. The photophysics of molecules of biological interest like metalloporphyrins is also a hot topics where ultrafast electron dynamics comes into play. Facing the remarkable progresses in experiments, there is a huge need to develop first principles approaches to simulate these ultrasfast processess at the microscopic level.



IN the PHOTOHEME project we have devised a unique computational set-up based on Real-Time Time-Dependent Density Functional Theory (RT-TDDFT). It allows simulations of the irradiation of condensed matter either by electric fields (of moderate to intense intensities) or by high energy charged particles. The upper Figure shows autocorelation function of the average dipole moments of each hydration layer of a peptide subject to a short electromagnetic pulse. The curves hihlights the speed at which the energy initially deposited in the peptide is dissipated into the environment (damped oscillations). We are now now ready to apply this methodology to study the non-adiabatic dexcitation decay of metalloporphyrins. The realization of this project funded by PALM is also finding many other applications at LCP, notably on the modelling of radiation induced damages in biology by first principles approaches.

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X. Wu, J. -M. Teuler, F. Cailliez, C. Clavaguéra, D.R. Salahub and A. de la Lande " Simulating Electron Dynamics in Polarizable Environments", *J. Chem. Theor. Comput.* 13, 3985- 4002 (2017)

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