CHARGE MIGRATION IN CHEMICALLY ENVIRONED MOLECULES

LABORATORY : Institut Lumière Matière

LEVEL : M1 / M2
TEAM(S) : THEOCHEM

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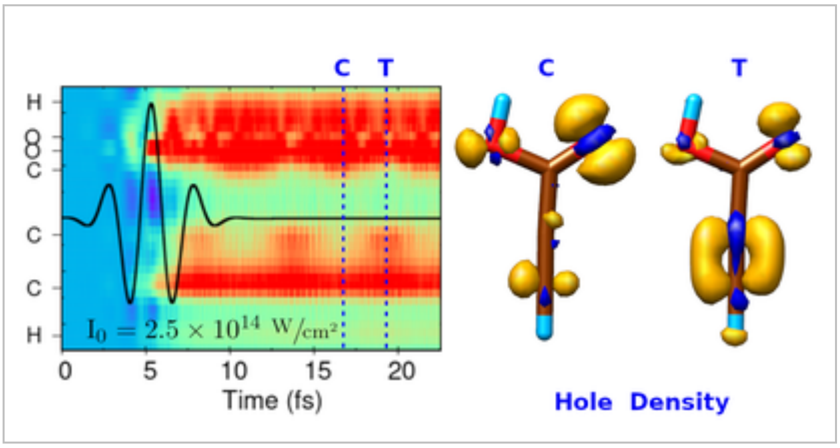
KEYWORD(S) : Real-Time TDDFT / Charge migration / Electronic coherence

SCIENTIFIC CONTEXT :



In a chemical reaction, the charge redistribution followed by the excitation of the electrons of the reactants decides the path of ionic motions which leads to breaking of bonds at a larger time-scale. Therefore, understanding the early processes of electronic excitation is crucial to achieve ultrafast control over reactions. In this context, the first challenge is to observe the long-predicted correlation-driven charge migration [1] where the removal of one single electron in a molecular system gives rise to superposition of molecular eigenstates that creates long-lasting coherence in the charge redistribution. Despite the possibility of measuring [2, 3] the transient charge-carrier dynamics, complexities arising from addressing the experimental conditions and the chemical environment around the molecules, both in a time-dependent quantum theoretical framework make it difficult to confirm the observation of charge migration. In this context, our recent work [4] showed how to selectively trigger correlation driven charge migration in isolated molecules. However, in realistic reactions molecules are hardly isolated. Therefore, in this master project we would like to explore the effect of chemical environment around the molecules on the charge migration.

MISSIONS :

In this master thesis, the objective is to theoretically investigate quantum many-body effects on the dynamics of a *hole* created by removing an electron from molecules when they are surrounded by other molecules. Time-evolution approach of TDDFT will be employed to perform ab-initio simulations.
After getting an introduction to the working principles of both static and time-dependent (TD) density-functional theory (DFT) the candidate will gain experience in performing ab-initio calculations using open-source codes (octopus and salmon). The simulations will be carried out on supercomputers in local and national centres. Addressing the environment in context of charge-migration will respond directly to fundamental questions on what influences different environments can have on the triggering of charge migration and on its duration. The project will thus allow to perform predictive simulations to guide experiments.

REQUIREMENTS: An interest in high-performance computation using supercomputers, numerical methods, programming in C++/Fortran and Python, as well as knowledge in atomic and molecular physics is highly welcoming.

OUTLOOKS :

Possibility (for M2 candidates) to apply for PhD.

BIBLIOGRAPHY :

[1] L.S. Cederbaum and J. Zobeley, Chemical Physics Letters, 307(3):205–210, 1999.
[2] D. T. Matselyukh, et al., Nature Physics, 18(10):1206–1213, Oct 2022.
[3] E. P. Månsson et al., Communications Chemistry, 4(1):73, May 2021.
[4] C. G. du Doignon et al., Chem. Sci., 16:16729–16736, 2025.

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