PhD 2023-2026

ATTOPHOTOCHEMISTRY OF TRANSITION METAL COMPLEXES

DESCRIPTION: PhD position in theoretical chemistry: 3 years from Oct. 2023.
Doctoral contract financed by the ERC Starting Grant ATTOP.

LOCATION: Team: **ModES** (Modeling & Spectroscopy)
Lab: CEISAM, UMR 6230, Nantes Université, CNRS
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CONTEXT

One of the most fundamental and widespread processes in chemistry is the absorption of light to excite electrons of molecules and potentially induce a chemical reaction. As a result of excitation into an electronic excited state, the distribution of electrons and thus the reactivity of the molecule differ significantly from the ones in the ground state. Thanks to this conceptually simple yet complex process, photochemistry has considerably broadened the spectrum of possible reactions, as compared to thermal chemistry. Despite the current applications of photo-induced processes in many fields, the practical use of photochemistry is limited by the quantum efficiency of the desired process, the latter being almost always in competition with other processes. A challenge for chemists today is therefore to design more efficient molecular systems and optical control methods for each desired application. In this context, it is necessary to rely on an in-depth knowledge of the photochemistry of the system of interest. Theoretical studies complementary to the experimental measurements are often necessary.

RESEARCH PROGRAM

This PhD thesis is part of the ERC project ATTOP which started in October 2022. ATTOP is a theoretical chemistry project which proposes to bring the very recent technological progress in attosecond science to the field of photochemistry. Indeed, light pulses of such short duration have a large spectral bandwidth and excite multiple electronic excited states in a simultaneous and coherent manner. This superposition, called an “electronic wavepacket”, has a new electronic distribution and is thus expected to lead to a new chemical reactivity. The goal of this thesis is to study attophotochemistry for medium-sized inorganic molecular systems, in particular transition metal complexes. Because attophotochemistry requires an exact treatment of electronic coherence and thus very accurate dynamics methods, the first objective is to develop, in collaboration with a post-doc in the group, a protocol to simulate quantum dynamics of large molecular systems (≈ 30 atoms and more), with a reduced number of coordinates chosen objectively, while controlling the impact of the approximations made. To achieve this, we propose to use machine learning algorithms in order to identify the most relevant nuclear coordinates for the process studied. Once such a protocol is validated, the next step will be to apply this protocol on the photodissociation reaction of a transition metal complex from MLCT states. This photochemical reaction could be an efficient way to produce useful highly reactive, coordinately unsaturated species. However, it is in competition with a photophysical relaxation pathway. The task will be first to simulate the photochemical reaction induced by each of the MLCT states separately, and finally by a coherent superposition of them. The target is to propose an electronic wavepacket that increases significantly the photodissociation reaction yield.

PROFILE OF THE CANDIDATE

The candidate should have a Master degree in chemistry, chemistry-physics, theoretical chemistry or physics, or equivalent obtained in 2022 or in 2023 and must have a solid training in physical and theoretical chemistry. Experience in *ab initio* molecular calculations as well as programming skills (Fortran, Python...) and machine learning are assets.

Applicants must send a CV, a cover letter to morgane.vacher@univ-nantes.fr, and the names of two reference persons.