DIRAM

Dynamiques et Interactions: Rayonnement, Atomes, Molécules

From cold atoms to hot plasmas … From diatomic molecules to clusters … From picosecond to attosecond…

We study, experimentally and theoretically, the dynamical behaviours occuring in the interaction between radiation and matter, in particular the dynamics of fundamental processes: excitation, ionization, dissociation, energy exchange and/or relaxation.

The systems of interest are mostly in dilute phase (atoms, ions, molecules, molecular assemblies, clusters, nanostructures); the relevant timescales range from ~ 10 attoseconds (10 as = 10 x 10^{-18} s), to ~100 picoseconds (100 ps = 100 x 10^{-12} s).

We use radiation sources in the infrared to VUV-soft X-ray spectral range, in various interaction regimes : from single-photon or multiphoton absorption to plasma formation. Some of these sources are available from external mid-scale to large-scale user facilities : synchrotron SOLEIL, ATTOLab, LASERIX, FERMI-Elettra, …

Photoionization dynamics angularly resolved in the molecular frame at the attosecond time-scale

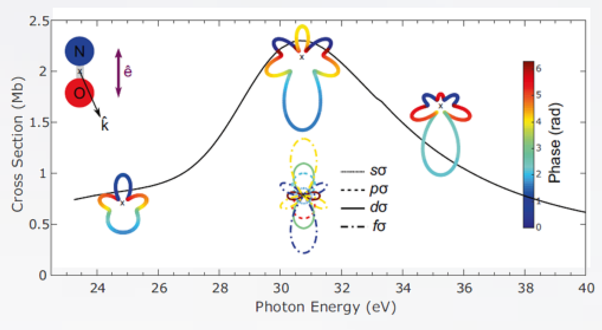
► **Objectives:** We determine the dipole matrix elements, magnitude and phase, for each partial wave contributing to the continuum electron wavefunction, providing attosecond photoionization time delays angle resolved in the molecular frame.

► **Methods:** We measure Molecular Frame Photoelectron Angular Distribution (MFPAD) using 3D momentum spectroscopy of the coincident photoelectron and photoion produced by dissociative photoionization combining two approaches:

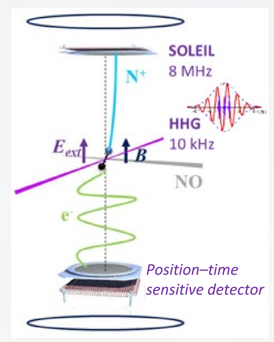
(i) Time resolved photoionization dynamics: FAB10 beamline at the ATTOlab laser facility (CEA, Orme des Merisiers)

(ii) Spectrally resolved photoionization dynamics: PLEIADES and DESIRS beamlines at Synchrotron SOLEIL.

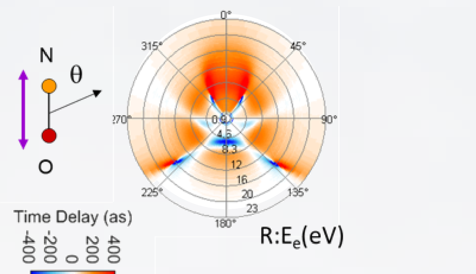
► **Results:** Characterizing the dynamics of the $\sigma \rightarrow \sigma^*$ shape resonances for inner valence ionization of small molecules.



Complex valued MF photoionization amplitude across a shape resonance



Electron - Ion momentum spectroscopy



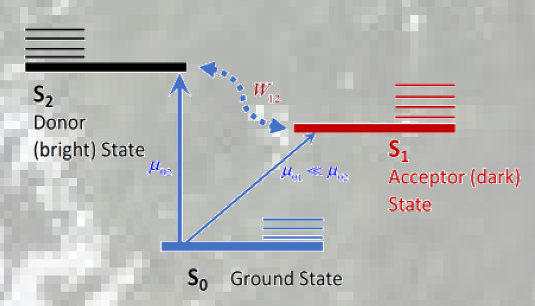
Angle and energy dependence of photoionization time delays

Holzmeier, F. et al. *Nat Commun* **12**, 7343 (2021)

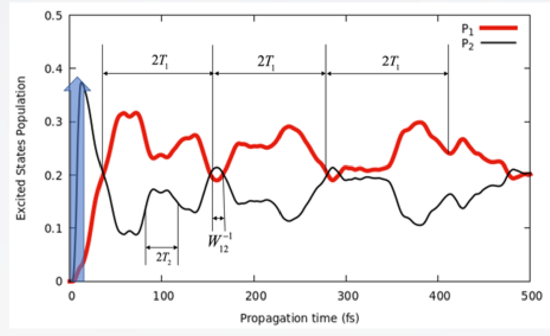
Molecular dynamics in strong laser fields: From modeling to control

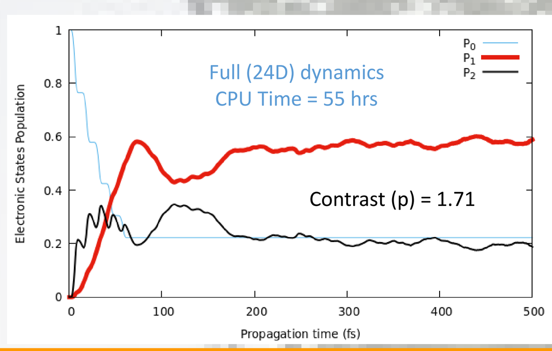
► **Objectives:** We refer to conical intersection as a funnel to deposit photonic energy from a bright donor into a quasi-stable excited state. The ultimate control objective is the maximum population in the otherwise dark acceptor.

► **Methods:** The laser control uses either interference or kicks strategies, while fighting against decoherence. The illustration is done on Pyrazine in a full 24 degrees of freedom quantum model, with a MCTDH methodology.



► **Results:** Experimentally achievable laser fields lead to a robust control with 60% of the ground state population deposited in the acceptor state. Applications may concern artificial photosynthesis, light harvesting processes, biological antennas or organic photovoltaic devices.

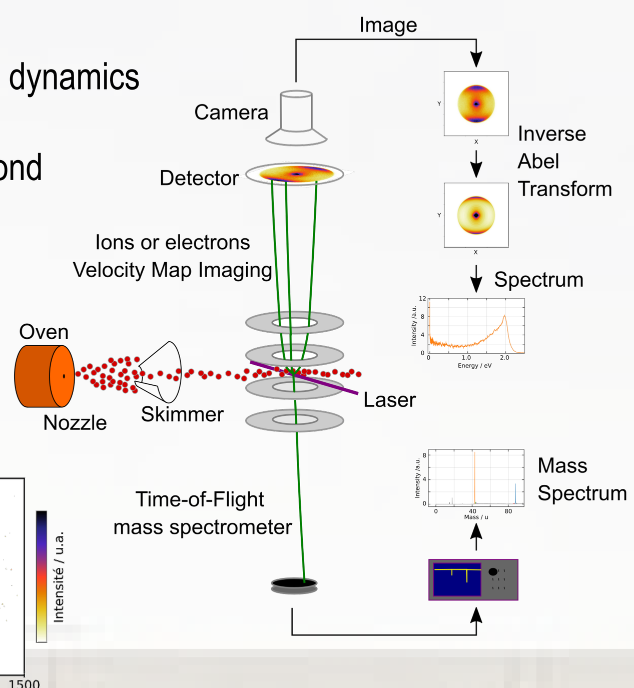




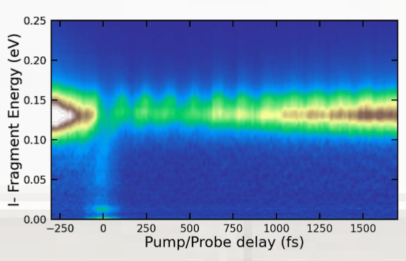
Gas-phase femtochemistry

► **Objectives:** Experimental time-resolved observation of photo-excited reaction dynamics in gas-phase atoms, molecules, clusters and nanoparticles: electronic relaxation processes and ionization dynamics at the picosecond, femtosecond and attosecond timescales.

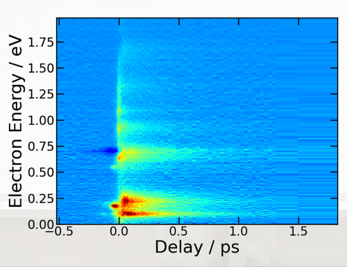
► **Methods:** Velocity Map Imaging of electrons and ions & Time-of-Flight mass spectrometer coupled to a femtosecond laser, using the pump/probe technique.



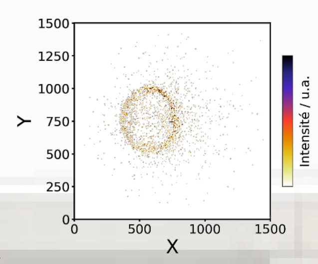
► **Results:**



Wavepacket oscillations observed in the production of I^- after excitation of I_2 in an ion-pair state with two 266 nm photons, probed by multiphoton ionization at 800 nm.



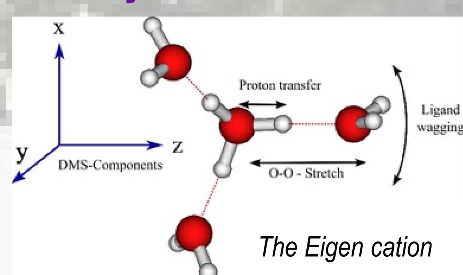
Vibrationally-resolved femtosecond dynamics of O_2 excited in a predissociative Rydberg state with two 266 nm photons, probed by mulitphoton ionization at 800 nm.



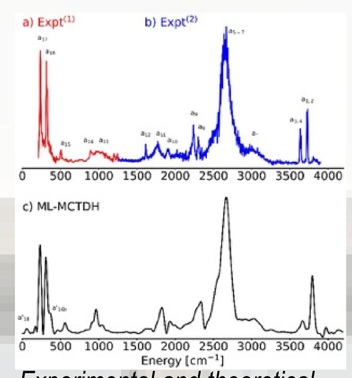
Peculiar spatial distribution of H^- ions produced from the interaction of Tryptophan nanoparticles with an intense 800 nm fs laser.

Full Quantum Description of Complex Molecular Systems

► **Objectives:** Quantum description of systems and processes very important in chemistry. We simulate spectra, collisions, pump-probe experiments to describe molecular systems at their most elementary level, necessitating a full quantum approach. We worked, for instance, on a complete description of the strong couplings in the two limiting cases of the aquous proton (Zundel and Eigen cations) essential to understand the Grotthus mechanism (diffusion of an hydrated proton in water).



► **Methods:** We mainly use the Multi-Configuration Time-Dependent (MCTDH) approach and the Heidelberg MCTDH package that allows us to treat high dimensional quantum dynamics (nuclear wave packet propagations).



Experimental and theoretical IR spectra of the Eigen cation.

► **Results: calculations of spectra, cross sections**

A full quantum propagation with all the 33 internal nuclear degrees of freedom allowed us to reproduce and interpret the experimental spectrum of the Eigen cation, $H_9O_4^+$, and of its isotopologue $D_9O_4^+$, and to predict lines not already observed experimentally. Most importantly, we showed that the main band at around 2600 cm^{-1} corresponds to the proton transfer band. We argued that the Zundel sub-unit ($H_5O_2^+$) is the fundamental dynamical building block of the solvated proton that explains its couplings and reproduces its spectroscopic signatures.

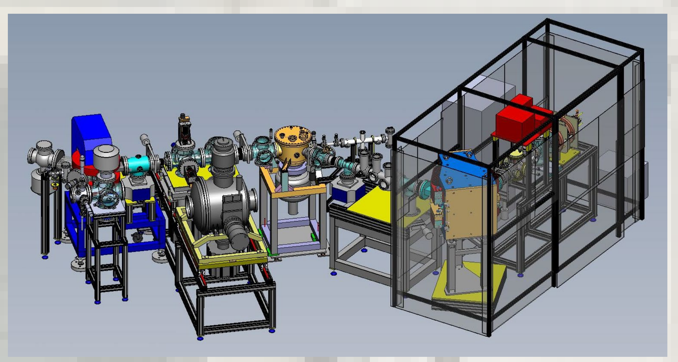
M. Schröder (Heidelberg), F. Gatti, D. Lauvergnat (ICP), H.-D. Meyer and O. Vendrell (Heidelberg), to be submitted.

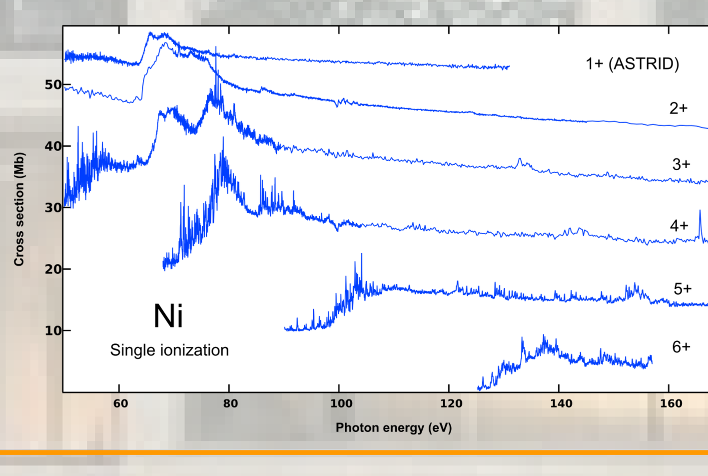
Photoionization of multicharged ions

► **Objectives:** Studying the processes involved in the photoionization of atomic and ionic systems is essential to understand the correlated movement of electrons in these species. The photoionization of multicharged ions constitutes a benchmark for the various theoretical models. It also provides essential data for other fields of physics such as astrophysics, or the study of laboratory plasmas.

► **Methods:** The MAIA experimental device allows the measurement of photoionization cross sections in multicharged ions. It benefits from a permanent installation on the PLEIADES beam line of SOLEIL.

► **Results:** The isonuclear series of nickel presented below shows the variations of cross section according to the state of charge and the energy of the incident photons. It was recorded in the spectral domain corresponding to a 2p inner shell electron on the progressively emptied 3d valence orbital.



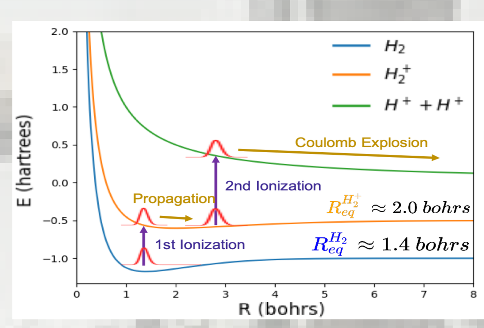


Electron correlation in the strong field molecular ionization

► **Objectives:** The objective is to study the dissociative ionization of H_2, up to Coulomb explosion, in a quantum model including electronic correlation, going beyond the commonly-assumed single active electron (SAE) approximation.

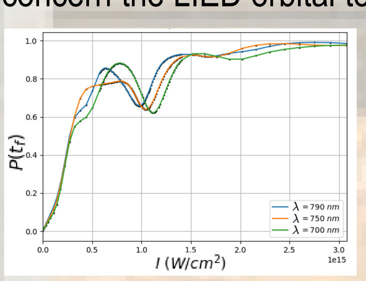
$$H_2 \xrightarrow[-e^-]{n_1\hbar\omega} H_2^+ \xrightarrow[-e^-]{n_2\hbar\omega} H^+ + H^+$$

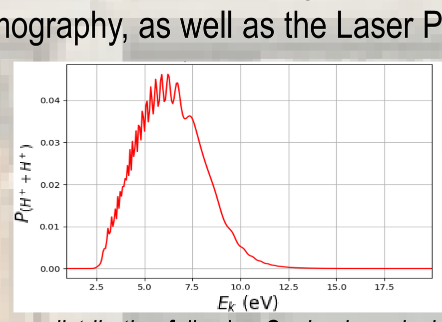
► **Methods:** The methodology is based on a time-dependent configuration interaction method we are developing for the quantal approach, together with some semiclassical approximations (ADK, PPT for the tunnel ionization).



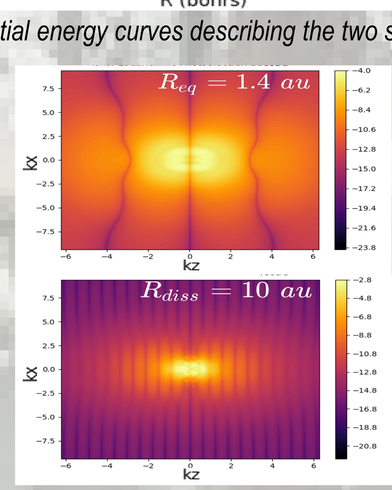
Potential energy curves describing the two steps.

► **Results:** The observables are ionization profiles and photoelectron spectra for the first ionization and the kinetic energy distribution of protons resulting from Coulomb explosion. Applications concern the LIED orbital tomography, as well as the Laser Plasma Acceleration.





Ionization profiles and proton kinetic energy distribution following Coulomb explosion.



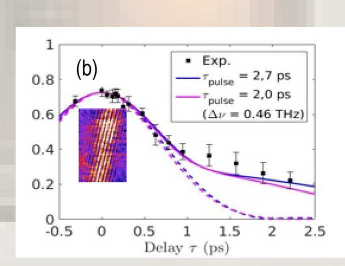
Photoelectron spectra at different internuclear distances.

Spectral and temporal properties of ultrashort XUV sources

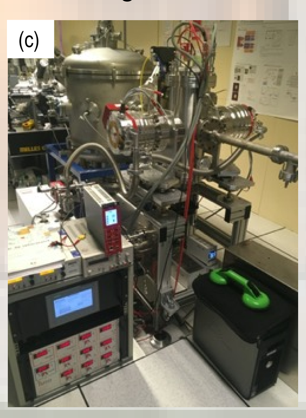
► **Objectives:** Ultra-short XUV sources, such as plasma-based XUV lasers or high-order harmonics generated from plasma mirrors, do have specific spectral and temporal properties that are difficult to measure experimentally, essentially due to the extreme resolutions required. However this knowledge is important, both to allow new source developments and for their use in current and future applications exploiting their capabilities.

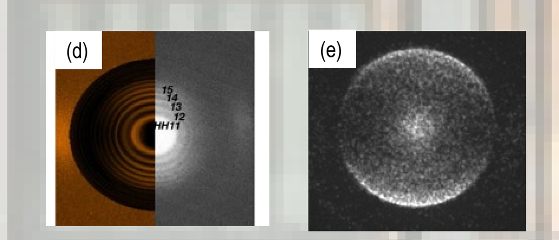
► **Methods:** We develop and implement advanced techniques and specifically designed instrumentation to access the temporal and spectral profiles of different types of ultra-short XUV sources. The spectral profile is inferred from first-order autocorrelation with a wavefront-division interferometer (Fig. a) designed at Institut d'Optique. For the temporal profile we seek to implement the technique of laser-dressed photoionization using a home-made VMI spectrometer (Fig. c).

► **Results:**



The partial temporal coherence of ultrashort plasma-based XUV lasers induces the formation of a pedestal in the shot-averaged autocorrelation signal (Fig. b) measured with the wavefront-division interferometer (Fig. a).





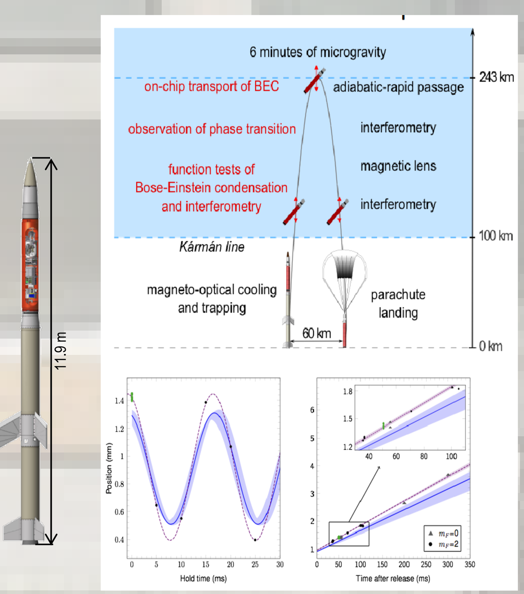
Photoelectron spectra obtained with (Fig. d) the plasma-mirror HH source @UHI100 (CEA Saclay), and with (Fig. e) the plasma-based XUV laser @LASERIX (Orsay), using the VMI spectrometer (Fig. c)

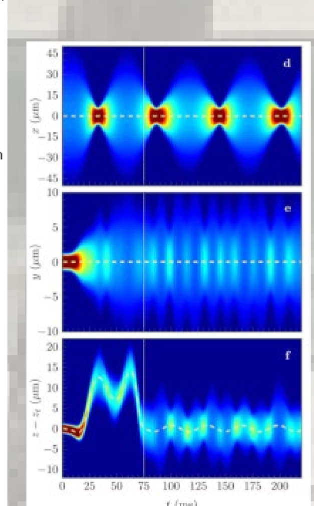
Quantum sensing and metrology

► **Objectives:** To exploit the full potential of quantum sensors for inertial applications and tests of fundamental physics, careful quantum state engineering schemes of ultra-cold atomic ensembles are required. For most applications, sensors based on atom interferometry promise few orders of magnitude boost in sensitivities compared to state-of-the-art performance if a macroscopic superposition or long drift times of several seconds are realized.

► **Methods:** Our research takes advantage of most novel and efficient techniques in the field of quantum gases theory and puts to work highly controllable atom interferometers of metrological significance. Analytical and numerical models are developed relying on optimal control theory together with time-dependent quantum dynamics solvers.

► **Results:** Our research covers aspects related to the dynamics of Bose-Einstein condensates manipulated by atom chips or dipole laser traps subject to free fall or in trapped conditions. These studies led to the first generation of a Bose-Einstein condensate (BEC) in space (2018) and to the production of BECs at the lowest temperature so far : 38 pK (2021) by efficiently collimating the atoms in 3D.





► Teaching

Several team members are actively involved in teaching activities at the undergraduate, graduate and doctoral levels

Health and Medicine; Magistère of Fundamental Physics / ENS Paris-Saclay; Master of Physics Paris-Saclay; Doctoral School EDOM; Licence de Chimie, Magistère de Physico-Chimie Moléculaire, Master SERP+

► National and international research networks:

GDR Ultrafast Phenomena, EMIE, THEMS, Technologies Quantiques

GDRi XFEL

Laserlab-Europe, COST AttoChem, IRN MCTDH