

Réunion Plénière du GDR UP – 2021

22 & 23 novembre 2021

Cité internationale universitaire de Paris

Recueil des contributions

Club des Partenaires Industriels



Le GDR U.P. est consacré aux phénomènes ultrarapides. Il a été officiellement créé le 01 janvier 2016 pour une durée de 5 ans, renouvelé le 01 janvier 2021 pour 5 ans.

Il rassemble la communauté française des expérimentateurs et théoriciens s'intéressant aux phénomènes aux échelles de temps ultrabrèves : attoseconde, femtoseconde et picoseconde et intervenant dans tous les états de la matière (milieu dilué, solide, nanométrique, liquide et plasma).

Cette cinquième réunion plénière du GDR du 22 au 23 novembre 2021 à la Cité Internationale Universitaire de Paris est une occasion importante de rapprochement entre les équipes françaises et une démonstration de l'émulation scientifique qui caractérise notre communauté. Nous vous remercions d'y participer et pour votre contribution à la grande réussite scientifique du GDR U.P.

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Lundi 22 novembre 2021

09h00-10h00	<i>ACCUEIL / CAFE</i>	
10h00-10h30	<i>Bureau du GDR U.P</i>	<i>Introduction</i>
<i>ULTRAFAST DYNAMICS IN THE GAS PHASE / chair Richard Taïeb</i>		
10h30-10h50	<i>Lou Barreau (ISMO, Orsay)</i>	Multimode vibration in CO ₂ observed with few-fs Xray transient absorption to the C 1s ⁻¹ 2π _u state
10h50-11h10	<i>Jean-Nicolas Vigneau (ISMO, Orsay)</i>	Strong-Field Molecular Ionization beyond the Single Active Electron approximation
11h10-11h30	<i>Loïc Joubert-Doriol (MSME, Marne-la-Vallée)</i>	Should we abandon adiabatic states to describe ultrafast molecular dynamics?
11h30-11h50	<i>Baptiste Fabre (CELIA, Bordeaux)</i>	Time-Resolved Photoelectron Circular Dichroism
11h50-12h10	<i>Mekha Vimal (LIDYL, Saclay)</i>	General scaling law of non collinear high harmonic generation and its applications
12h10-12h30	<i>Quentin Bournet (AMPLITUDE /IOGS)</i>	Ultra-broadband, tunable (4-10 μm) mid-IR source based on intrapulse difference-frequency generation
12h30-14h00	<i>REPAS / EXPOSANTS</i>	
<i>ULTRAFAST DYNAMICS IN NANOSTRUCTURES AND SURFACES / chair Christine Richter</i>		
14h00-14h20	<i>Sophie Meuret (CEMES, Toulouse)</i>	Time-Resolved Cathodoluminescence in an Ultrafast Transmission Electron microscope
14h20-14h40	<i>Jon Gorchon (Institut Jean Lamour, Nancy)</i>	Ultrafast spin currents: a new toolbox
14h40-15h00	<i>Elsa Cassette (LUMIN, Orsay)</i>	Ultrafast relaxation of hot excitons in colloidal 2D perovskite nanoplatelets: carrier-induced Stark and phonon bottleneck effects
15h00-15h20	<i>Tatiana Itina (LHC, Saint-Etienne)</i>	Numerical studies of thermoplasmonic effects and nanoalloy formation in ultra-short laser interactions with nanoparticles
15h20-15h40	<i>Sven Fröhlich (LIDYL, Saclay)</i>	Self-probed ptychography from semiconductor high-harmonic generation
15h40-16h10	<i>PAUSE CAFÉ / EXPOSANTS</i>	
<i>ULTRAFAST DYNAMICS IN MATERIALS / chair Sarah Houver</i>		
16h10-16h30	<i>Samuel Beaulieu (CELIA, Bordeaux)</i>	Time-Resolved Multidimensional Photoemission Spectroscopy of 2D Material
16h30-16h50	<i>G. Huitric (IPR, Rennes)</i>	TeraHertz vs Optical pump generated strain waves profiles in V ₂ O ₃ Metallic thin film
16h50-17h10	<i>A. Levchuk (IMMM, Le Mans)</i>	Identifying different contributions to the ultrafast spin-to-charge conversion : Experimental approach
17h10-17h30	<i>Romain Géneaux (LIDYL, Saclay)</i>	Condensed matter dynamics probed with attosecond transient absorption spectroscopy
17h30-19h00	<i>Session Poster / Cocktail</i>	
19h00-20h30	<i>Cocktail</i>	

Mardi 23 novembre 2021

<i>GAS PHASE EXPERIMENTS AT FREE ELECTRON LASERS / Chair Francis Penent</i>		
9h00-9h20	<i>Nora Berrah (Univ. of Connecticut/ LIDYL)</i>	Time-Resolved Dynamics in Molecules
9h20-9h40	<i>John Bozek (Sync. SOLEIL, Saint- Aubin)</i>	TRÉSOR – Time-Resolved Electron Spectroscopy for Original Research – at the European XFEL
9h40-10h00	<i>Saikat Nandi (iLM, Lyon)</i>	Unraveling ultrafast Rabi dynamics with a seeded free-electron laser
10h00-10h30	<i>PAUSE CAFÉ / EXPOSANTS</i>	
<i>ULTRAFAST DYNAMICS IN MOLECULES IN CONDENSED PHASE / Chair Vincent de Waele</i>		
10h30-10h50	<i>Pascale Chagnenet (LOB, Palaiseau)</i>	Multiscale conformational dynamics probed by time-resolved circular dichroism
10h50-11h10	<i>Marc Alías-Rodríguez (ICR, Marseille)</i>	Revealing the ultrafast intersystem crossing in xanthone using wave packet quantum simulations
11h10-11h30	<i>Robin Pierron (IPCMS, Strasbourg)</i>	Towards 2D electronic spectroscopy of vibrational coherences in molecular motors and photoswitches
11h30-11h50	<i>Aurélien de la Lande (ICP, Orsay)</i>	The Mystery of Sub-Picosecond Charge Transfer Following Irradiation of Hydrated Uridine Monophosphate
11h50-12h10	<i>Bo Zhuang (LOB, Palaiseau)</i>	Ultrafast Photooxidation of Protein-Bound Anionic Flavin Radical
12h10-12h30	<i>Yoann PERTOT (FASTLITE)</i>	CEP-stable infrared OPCPA sources
12h30-14h00	<i>REPAS / Session Poster / EXPOSANTS</i>	
<i>ULTRAFAST PROCESSES / Chair Valérie Veiniard</i>		
14h00-14h20	<i>Claire Lauhlé (SOLEIL, Saint- Aubin)</i>	Photoinduced charge density wave phase in ¹ T-TaS ₂ : growth and coarsening mechanisms
14h20-14h40	<i>Marie Labaye (Lab. Pasteur, Paris)</i>	Resonant propagation of short pulses: collective effects in light-matter interaction
14h40-15h00	<i>Mai Dinh Phuong (LPT, Toulouse)</i>	Dipolar instabilities in N ₂ theoretically evidenced by an ultrafast pump-and-probe setup
15h00-15h20	<i>Marie Ouillé (LOA, Palaiseau)</i>	High-harmonics and particle beams emission from relativistic plasma mirrors at kHz repetition rate
15h20-15h50	<i>Pause café / EXPOSANTS</i>	
<i>SECONDARY SOURCES / Chair Annie Klisnick</i>		
15h50-16h10	<i>Philippe Martin (LIDYL, Saclay)</i>	Mesure spatio-temporelle d'impulsions attosecondes d'origine relativistes
16h10-16h30	<i>Kosta Oubrerie (LOA, Palaiseau)</i>	Accélération laser-plasma au GeV par guidage laser
16h30-16h50	<i>Alok Pandey (IJCLab, Orsay)</i>	Generation and characterization of high topological charge extreme-ultraviolet vortex and vector-vortex beams
16h50-17h10	<i>François Sylla (SOURCELAB)</i>	SourceLAB, fournisseur d'accélérateurs Laser Plasma
Conclusion	Bureau du GDR UP	

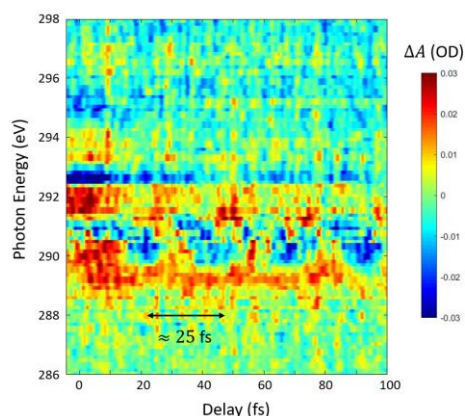
Contributions Orales

Multimode vibration in CO₂ observed with few-femtosecond X-ray transient absorption to the C 1s⁻¹ 2π_u state

Lou Barreau^{1,2}, Andrew D. Ross², Thibault Noblet¹, Alexis Texier¹, John F. Stanton³, Fabien Gatti¹, Daniel M. Neumark², Stephen R. Leone²

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Owing to the chemical and site-sensitivity of X-ray absorption, time-resolved absorption spectroscopy at the carbon K-edge holds promise of revealing local structural and electronic information about ultrafast dynamics in organic molecules. To that end, recent experimental effort has focused on the development of table-top high-order harmonic generation sources producing few-femtosecond to attosecond pulses extending into the soft X-ray domain [1,2]. Here, we demonstrate the use of such a source to observe multimode coherent vibrational dynamics in carbon dioxide. Coherent excitation of the $\nu_1 - 2\nu_2$ Fermi resonance in the electronic ground state of CO₂ is realized through impulsive stimulated Raman scattering with a few-cycle visible-IR laser pulse. The dynamics are probed with time-resolved soft X-ray absorption to C core-excited states and show a characteristic ~ 25 fs oscillation period. *Ab initio* calculations of the core-excited states reveal the strong pseudo-Jahn Teller coupling between the 1s⁻¹ 2π_u and 1s⁻¹ 3s states. The experimental results are in very good agreement with MCTDH calculations of the vibrational wavepacket dynamics and of the corresponding X-ray transient absorption.



Measured transient absorption in CO₂.

References:

- [1] Ren, X. *et al.*, Attosecond light sources in the water window. *J. Opt.* **20**, 023001 (2018).
- [2] Barreau, L., Ross, A. D., *et al.*, Efficient table-top dual-wavelength beamline for ultrafast transient absorption spectroscopy in the soft X-ray region. *Sci. Rep.* **10**, 5773 (2020).

Strong-Field Molecular Ionization beyond the Single Active Electron approximation

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The present work explores quantitative limits to the Single-Active Electron (SAE) [1] approximation, currently in use to deal with strong-field ionization and subsequent attosecond dynamics.

Using a time-dependent multiconfiguration approach, specifically a Time-Dependent Configuration Interaction (TD-CI) [2] method, we solve the time-dependent Schrödinger's equation (TDSE) for the two-electron dihydrogen molecule, with the possibility of tuning at will the electron-electron interaction by an adiabatic branching/debranching function. We focus on signals of the single ionization of H_2 under a strong NIR ($\lambda = 790, 750, 700 \text{ nm}$) two-cycle, linearly-polarized laser pulse of varying intensity (up to $I = 2 \times 10^{16}, 3 \times 10^{15}, 1 \times 10^{15} \text{ W/cm}^2$). To assess the dynamical importance of the electron repulsion, V_{ee} , we consider three values of the internuclear distance R taken as a parameter, the nuclei being frozen in each geometry in the spirit of the Born-Oppenheimer approximation: $R = R_{eq} = 1.4 \text{ au}$, the equilibrium geometry of the molecule, represents a field-free situation of weak electronic correlation, while $R = 5.0 \text{ au}$ and $R = 10.2 \text{ au}$, denoting respectively an elongated and a dissociating molecule, are situations of increasing field-free correlation.

As expected, in the equilibrium geometry $R = R_{eq}$, tuning off the electron repulsion have no impacts on the channel-resolved and total ionization probabilities exhibited as functions of the field-intensity. The profile is typical of a tunnel ionization, except for the highest range of I , for which a sudden increase in the total ionization probability is observed and interpreted as due to the onset of an above-the-barrier ionization. At the elongated geometry, $R = 5.0 \text{ au}$, the ionization probability does not exhibit such a sudden increase as one approaches the high-intensity end, i.e. no above-the-barrier enhancement of the ionization is observed. The strong electronic correlation prevailing at this geometry now induces considerable modifications to the dynamics, (they are no longer observed when the the electron repulsion was tuned-off), and the ionization probability varies non-monotonously with I , passing through a dip, denoting a quenching of the ionization, at some moderate intensity I_{min} , the value of which depends on the field frequency (or on λ). A possible explanation is that the ionization is a mixture of a tunnel one (TI) [3] from the (correlated) ground-state and multiphoton ionizations (MPI) [4] from excited states, accessible from the ground state also by multiphoton excitations. The dynamics thus proceeds through a Resonance-Enhanced-Multiphoton Ionization (REMPI) [5] with interfering overlapping resonances resulting from excited electronic states. This non-monotonous behaviour is not observed in the dissociative limit, $R = 10.0 \text{ au}$, the strongest field-free correlation situation, and only subtle differences are observed between correlated and uncorrelated dynamics. This stunning observation is explained by the fact that no multiphoton transition to the excited states are possible from the correlated ground state, as these transitions between these states become dipole-forbidden in this geometry.

[1] M., Peters, T. T., Nguyen-Dang, C., Cornaggia, S., Saugout, E., Charron, A., Keller, and O., Atabek. *Phys. Rev. A* **83** (2011) 051403.

[2] T. T., Nguyen-Dang, É., Couture-Bienvenue, J., Viau-Trudel, and A., Sainjon. *J. Chem. Phys.* **141** (2014) 244116.

[3] P. B., Corkum. *Phys. Rev. Lett.* **71** (1993) 1994.

[4] I. S., Aleksakhin and N. B., Delone. *Sov. Phys. - JETP* **49** (1979) 447.

[5] S. L., Anderson, G. D., Kubiak, and R. N., Zare. *Chem. Phys. Lett.* **105** (1984) 22.

Should we abandon adiabatic states to describe ultrafast molecular dynamics?

Loïc Joubert-Doriol¹

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Adiabatic states are at the origin of our understanding of numerous chemical processes through the Born-Oppenheimer approximation. Going beyond this approximation becomes necessary when potential energy surfaces approach or even cross each other, forming conical intersections (CIs). CIs are ubiquitous in photochemical phenomena and play a central role by allowing radiationless electronic transitions. They are especially important when the system is excited with high energy light pulse beyond UV since the electronic state density becomes high. Hence, one may wonder if the adiabatic representation is well suited for such processes.

The Hamiltonian in adiabatic representation exhibits singularities and a Berry phase appears in the corresponding electronic states at CIs. These features prevent a straightforward numerical treatment of dynamical processes involving CIs and impose additional boundary conditions. This is a failure of the adiabatic representation.

The moving crude adiabatic representation circumvents this difficulty by introducing time-dependent crude adiabatic states that are formally diabatic. We test this approach combined with the use of Gaussian basis functions to design an exact method for simulating molecular processes beyond the Born-Oppenheimer approximation.

References:

L. Joubert-Doriol, J. Sivasubramaniam, I. G. Ryabinkin, and A. F. Izmaylov, *J. Phys. Chem. Lett.* 8, 452 (2017)

L. Joubert-Doriol and A. F. Izmaylov, *J. Chem. Phys.* 148, 114102 (2018)

L. Joubert-Doriol and A. F. Izmaylov, *J. Phys. Chem. A* 122, 6031 (2018)

R. Maskri and L. Joubert-Doriol, submitted to *Philos. Trans. R. Soc. A*

Time-Resolved Photoelectron Circular Dichroism

Baptiste Fabre¹

¹ CELIA Bordeaux

We study the isomeric effects using time resolved photoelectron circular dichroism (TR-PECD). Using a (1 + 1') pump–probe ionisation scheme with photoelectrons collected by the velocity map imaging technique, we compare the relaxation dynamics from the 3s-Rydberg state in 1R,4R-(+)-camphor with the one in its chiral isomer, 1R,4S-(-)-fenchone [1]. Our measurements revealed a similar lifetime for both isomers. However, the circular dichroism in the photoelectron angular distribution decays exponentially in ~730 fs from a +9% forward amplitude during the first hundreds of femtoseconds to reach an asymptotic 2% backward amplitude. This time-scale is drastically shorter than in fenchone. Our analysis allows us to evaluate the impact of the anisotropy of excitation; the relaxation dynamics, following photoexcitation by the linearly polarized pump, is then compared to that induced by a circularly polarized pump pulse (CPL). With such a CPL pump, we then retrieve time constants of our chiral observables similar to the ones recorded in fenchone. Quantum and classical simulations are developed and used to decipher the dependence of the PECD on the anisotropy of excitation and the spatial distribution of the 3s-Rydberg electron wavefunction. Our experimental investigations, supported by our simulations, suggest that varying the pump ellipticity enables us to reveal the breakdown of the Franck–Condon approximation.

References:

[1] Comby et al., 2016, JPCL, 7, 4514

General scaling law of non collinear high harmonic generation and its applications

Mekha Vimal, Martin Luttmann, David Bresteau, Fabien Lepetit, Olivier Tcherbakoff, Jean-François Hergott, Thierry Auguste, Thierry Ruchon.

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We investigate High order Harmonic Generation driven by two Non Collinear beams (NCHHG) theoretically and experimentally over a very large range of intensities ratios of the two beams, from the perturbative to the non perturbative regime. By introducing the composite field which forms an "active optical grating" in an empirical model based on the Strong-Field Approximation (SFA), we could establish the scaling laws of the yields of the multiple beamlets generated. The theoretical and experimental results are in excellent agreement, and further match with a direct full 3D numerical model based on the resolution of the Schrödinger Equation within the strong field approximation. These results could also be interpreted in terms of photon channels associated to each beamlet, bridging the gap with previous interpretations restricted to the low perturbation regime. The establishment of this general description of NCHHG opens novel spectroscopic approaches. Thus we have identified specific spectroscopic signatures of strong field multiphoton processes in both the intermediate and strong perturbation regimes, which find straightforward interpretations within our active grating framework.

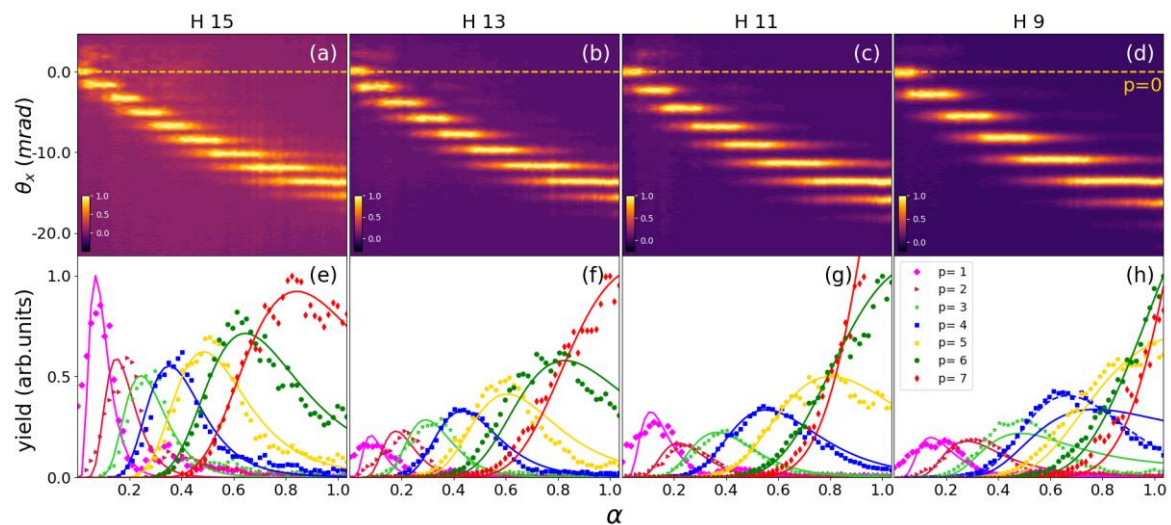


Figure: The experimental data for harmonics 15, 13, 11 and 9 for high perturbation levels are presented. The top panels (a-d) show the position and presence/absence of each order of diffraction of the active grating with increasing perturbation. The bottom panels (e-h) show emission yields of different diffraction orders of the corresponding harmonic order. The solid lines are the fits of the analytical model on the experimental data.

References:

1. C. Chappuis et.al, "High harmonic generation in an active grating", Phys.Rev. A99(2019).
2. M. Vimal et.al, "General scaling law of non collinear high harmonic generation and its applications", Light: Science and its applications, submitted.

Ultra-broadband, tunable (4-10 μm) mid-IR source based on intrapulse difference-frequency generation

Q. Bournet^{1,2}, F. Guichard², M. Natile², Y. Zaouter², F. Druon¹, M. Hanna¹, and P. Georges¹

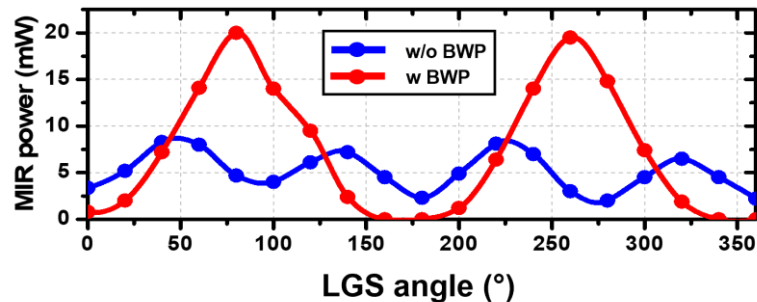
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2. Amplitude, 11 Avenue de Canteranne, Cité de la Photonique, 33600 Pessac, France

We report on an ultra-broadband mid-infrared (mid-IR) source based on intrapulse difference-frequency generation (iDFG) [1-3] delivering 110 nJ, 50 fs-long pulses at 250 kHz repetition rate. A driving high-energy Yb-doped-fiber amplifier (YDFA) delivering 200 μJ , 300 fs-long pulses at a central wavelength of 1030 nm is followed by a **70% high-efficiency** dual-stage nonlinear compression scheme based on a multipass cell and a 1 m-long capillary filled with argon. The 9.5 fs-long resulting pulses are characterized with a d-scan system and used to drive the iDFG in a 1 mm-thick LiGaS₂ (LGS) crystal with an intensity of 150 GW/cm². Prior to this crystal, a bichromatic waveplate (BWP) [4] is used, allowing to use the whole photon spectral density within each interacting spectral band.

The BWP is oriented with an angle of 45° between its axes and the input beam polarization to perform spectrally-dependent polarization rotation, increasing the mid-IR average power generated by a **factor 2.5**. In fig. 1, the mid-IR power with respect to the LGS orientation angle with and without the BWP is reported. The mid-IR tunability (4-10 μm) is measured using a commercially available FTIR spectrometer. The temporal properties of the mid-IR pulses are measured using an electro-optic sampling (EOS) scheme [5]. After compression, we measured a FWHM pulse duration of 50 fs (F_{TL} duration = 48.2 fs), corresponding to **2 optical cycles** at the central wavelength of 8.5 μm . The corresponding spectrum spans over 500 cm⁻¹ at -10 dB.

We are currently working to scale up the mid-IR pulse energy using optical parametric amplification (OPA). This source paves the way for new experiments in 2D ultrafast spectroscopy.

FIGURE 1



Dependence of the mid-IR idler beam average power on the polarization angle of the driving NIR beam measured without BWP (blue line) and with BWP (red line) for a 1-mm-thick LGS crystal.

References:

- [1] A. Bonvalet, M. Joffre, J. Martin, and A. Migus, *Appl. Phys. Lett.* **67**, 2907 (1995).
- [2] R. Kaindl, D. Smith, M. Joschko, M. Hasselbeck, M. Woerner, and T. Elsaesser, *Opt. letters* **23**, 861 (1998).
- [3] R. Huber, A. Brodschelm, F. Tauser, and A. Leitenstorfer, *Appl. Phys. Lett.* **76**, 3191 (2000).
- [4] H. Fattahi, A. Schwarz, S. Keiber, and N. Karpowicz, "Efficient, octavespanning difference-frequency generation using few-cycle pulses in simple collinear geometry," *Opt. letters* **38**, 4216–4219 (2013).
- [5] R. Huber, A. Brodschelm, F. Tauser, and A. Leitenstorfer, "Generation and field-resolved detection of femtosecond electromagnetic pulses tunable up to 41 thz," *Appl. Phys. Lett.* **76**, 3191–3193 (2000).

Time-resolved Cathodoluminescence in a TEM

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The development of time-resolved Cathodoluminescence (TR-CL) in a scanning electron microscope enabled the measurement of the lifetime of excited states in semiconductors with a sub-wavelength spatial resolution. It was used for example to measure the influence of stacking faults on the GaN exciton ¹, to probe the role of a silver layer on the dynamics of a YAG crystal ² or to show the influence of stress on the optical properties of ZnO nanowires ³. These results demonstrate that TR-CL is essential to study the correlation between semiconductor optical and structural properties (composition, defects, strain...). While all these pioneering studies were done using a scanning electron microscope, the improvement of the spatial resolution and the combination with other electron-based spectroscopies offered by transmission electron microscopes will be a step forward for TR-CL.

In this presentation, we will discuss the first time-resolved cathodoluminescence experiments within a transmission electron microscope. They were performed in a unique microscope, based on a cold-FEG electron gun ⁵. This technology allows among other things to reach a spatial resolution of a few nanometres, essential for the study of III-V heterostructures. We will present the first lifetime maps done in a TEM both on Nano-diamonds and InGaN quantum wells and discuss the unique features and opportunities of this technique.

References

1. Corfdir, P. *et al.* Exciton localization on basal stacking faults in a-plane epitaxial lateral overgrown GaN grown by hydride vapor phase epitaxy. *J. Appl. Phys.* **105**, 043102 (2009).
2. Moerland, R. J., Weppelman, I. G. C., Garming, M. W. H., Kruit, P. & Hoogenboom, J. P. Time-resolved cathodoluminescence microscopy with sub-nanosecond beam blanking for direct evaluation of the local density of states. *Opt. Express* **24**, 24760 (2016).
3. Fu, X. *et al.* Exciton Drift in Semiconductors under Uniform Strain Gradients: Application to Bent ZnO Microwires. *ACS Nano* **8**, 3412–3420 (2014).
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5. Houdellier, F., Caruso, G. M., Weber, S., Kociak, M. & Arbouet, A. Development of a high brightness ultrafast Transmission Electron Microscope based on a laser-driven cold field emission source. *Ultramicroscopy* **186**, 128–138 (2018).

Ultrafast spin currents: a new toolbox

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Reducing energy dissipation while increasing speed in computation and memory is a long-standing challenge for spintronics research [1]. In the last 20 years, femtosecond lasers have emerged as a tool to control the magnetization of certain magnetic materials at the picosecond timescale [2]. Recently, new materials and new ultrafast mechanisms have been discovered that expand the possibilities of the field. Particularly, I will discuss some of our latests results where we show that ultrafast spin currents generated either by picosecond wide electrical pulses [3], or by ultrafast demagnetization [4,5] can be exploited to reverse the magnetization of a classical ferromagnet. These experiments show that spintronic phenomena can be exploited on picosecond time-scales and should launch a new regime of ultrafast spin injection studies and applications.

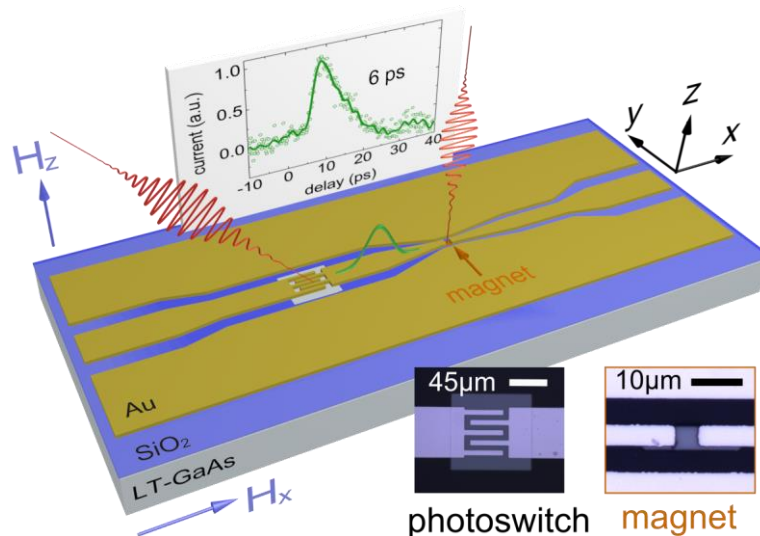


Figure caption: **Setup for generation of picosecond electrical pulses and magneto-optical detection.** The optical pump excites the photoconductive switch in order to generate ~ 6 ps duration electrical pulses, that are guided and focused by a coplanar waveguide into the magnetic stack, resulting in ultrafast spin orbit torques. The sampled picosecond current pulse is shown on the back of the figure. The solid green line is a guide for the eyes.

References:

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Hot Exciton Relaxation in Colloidal 2D Perovskite Nanoplatelets: Carrier-induced Stark and Phonon bottleneck Effects

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Using femtosecond transient absorption (fs-TA), we investigate the hot exciton relaxation dynamics in strongly confined lead iodide perovskite nanoplatelets (NPLs). The large quantum and dielectric confinement leads to discrete excitonic transitions and strong Stark features in the TA spectra (**Figure 1**, left panel). This prevents the use of conventional relaxation analysis methods extracting the carrier temperature or measuring the build-up of the band-edge bleaching. Instead, we show that the TA spectral lineshape near the band-edge reflects the state of the system and thus can be used to probe the exciton cooling dynamics by a global analysis method. While the energy level separations become larger than the energy of relevant optical phonon modes, the ultrafast hot exciton relaxation in one- to three- monolayer-thick NPLs evidences the absence of intrinsic phonon bottleneck (slow cooling rate expected due to inefficient multi-LO phonon emission). We found instead a faster intrinsic cooling time in the thinner NPLs, suggesting the presence of an additional relaxation pathway, which seems more efficient with increasing confinement. To get more insight on the hot exciton relaxation process, we performed excitation fluence-dependent measurements. We reveal a hot phonon bottleneck effect, with a delayed hot exciton relaxation up to about 1 ps at the maximum fluence. The evolution of the cooling time with the excitation fluence is found independent on the nature of the internal cations when comparing formamidinium (FA)-, methylammonium (MA)- and cesium- based lead iodide NPLs (**Figure 1**, middle panel). However, it is strongly affected by the ligands and/or sample surface state when comparing colloidal NPLs with 2D perovskite films (**Figure 1**, right panel). Together, these results suggest a role of the surface ligands in the cooling process [1].

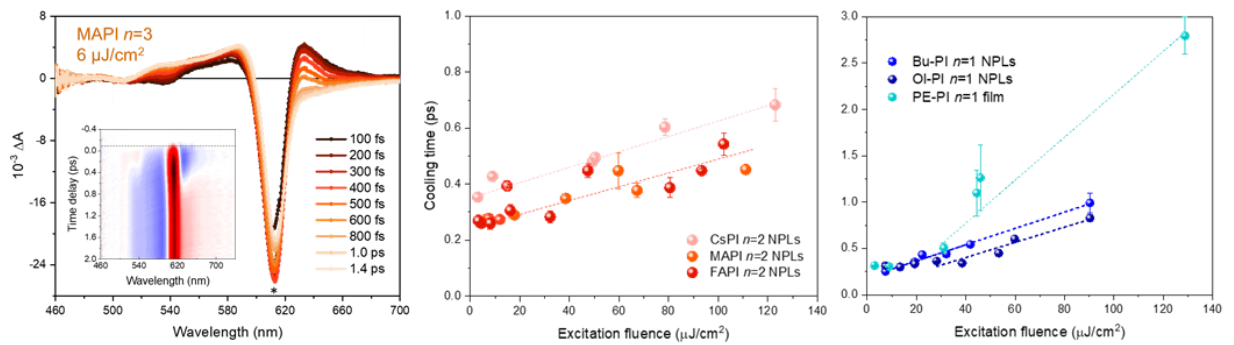


Figure 1. Left: Fs-TA spectra of methylammonium lead iodide NPLs of three monolayers ($n=3$) during the hot exciton relaxation (low fluence excitation at 400 nm). The full TA map is displayed in inset. Middle& right: Hot exciton cooling time obtained from global analysis and its evolution with the excitation fluence for two and three monolayer-thick 2D perovskite samples ($n=2$ NPLs, middle panel with different cations and $n=1$ NPLs and film, right panel, with different ligand nature).

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Numerical studies of thermoplasmonic effects and nanoalloy formation in ultra-short laser interactions with nanoparticles

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Ultra-short lasers allow us numerous possibilities of nanoparticle formation and of an additional control over their size, composition and properties. One of the main advantages of femtosecond laser pulses are the possibility of very efficient synthesis of small and chemically clean nanoparticle in liquids and of formation of various nano-hybrids, as well as of nanoalloys formed of normally immiscible materials.

The considered ultra-fast interactions typically begin by the excitation of the electronic sub-system followed by plasmon-enhanced charge emission, thermoplasmonic effects and, eventually, by melting, reshaping, internal atomic diffusion, mixing, sintering, etc. In this work, attention will be focused on multi-physical and multi-scale models simulating not only size-dependent nanoparticle absorption and local field enhancement, but also photo-induced free carrier generation, plasmon-assisted electron emission and heat transfer. Then, the results of a series of both multi-physical and atomistic molecular dynamics simulations will be presented. The role of laser parameters, of nanoparticle size, temperature, composition and cooling conditions will be analyzed in comparison with the available experimental findings.

Self-probed ptychography from semiconductor high-harmonic generation

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In recent years high harmonic generation (HHG) in semiconductor crystals have been demonstrated using a variety of materials and a wavelength [1-3]. Compared to gas harmonics, the electric field is unable to accelerate the electron out of the crystal potential, making the generation mechanisms substantially different from gas harmonics and highly sensitive to the bandstructure of the solids [4]. Additionally, the usage of a solid medium opens up the possibility of nanostructuring the solids to tailor the generated harmonics. This can be used to boost the harmonic yield but also to structure the generated harmonics [5,6]. The use of nanostructures naturally requires imaging them to detect deviations from the intended pattern. Out of the available techniques, coherent diffraction imaging (CDI) is a compelling candidate for the context of HHG in solids due to the possible high resolution and similarities between the HHG and CDI setup. However, CDI requires an illumination of the whole structure while HHG setups often favor a tight focusing. This problem can be bypassed by using ptychography, a technique similar to CDI but suited to image extended structures. Instead of using one diffraction pattern as CDI, it uses a series of diffraction patterns from different points of the structure to reconstruct the overall image. Here, we combine ptychography with HHG from solids to reach an inline characterization of the structure using the HHG generated in it.

As a proof of concept, we demonstrate the technique for two structures using the diffraction of the generated 420nm fifth harmonic. The diffraction patterns are reconstructed using the openly available PyNX library [7]. The first sample is a 300 μ m silicon sample with a nanostructure pattern of 50 μ m depth. The pattern is clearly identifiable with an obtained resolution of around 800nm. The wavelength dependency of the resolution in combination with the demonstration of harmonics of up to about 25eV potentially allow for resolutions as low as 50nm. As a second example, we characterized a spiral zone plate on a 500 μ m ZnO crystal, that was used before to generate a beam carrying an orbital angular momentum (OAM). The reconstruction is based on a scan of the central part of the zone plate and is shown to yield a quality that is sufficient to reconstruct the experimentally observed shape of the OAM beam.

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Time-Resolved Multidimensional Photoemission Spectroscopy of 2D Material

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Angle-resolved photoemission spectroscopy (ARPES) is arguably the most direct and powerful technique to measure the electronic band structure of solids. Combining a pump-probe scheme - using femtosecond XUV pulses - with ARPES, allows measuring the dynamical modification of populations, their scattering pathways, as well as modification of many-body interactions following photoexcitation of solids, on unprecedented timescales. In this talk, I will present three recent key findings that we made on two-dimensional transition metal dichalcogenides (2D-TMDCs), using this technique.

- i)* The demonstration that all key microscopic quantities of an excitonic state are encoded in the multidimensional photoemission signal: the spatial distribution of the excitonic wave function, its binding energy, and its lifetime, exemplified for bulk 2H-WSe₂.
- ii)* The first demonstration of non-equilibrium light-induced Lifshitz transition in the topological Weyl semimetal T_d-MoTe₂.
- iii)* The introduction of a new observable based on the manipulation of the photoemission transition dipole matrix element using continuous XUV polarization rotation allowing accessing the momentum-dependent orbital texture, again exemplified for bulk 2H-WSe₂.

TeraHertz vs Optical pump generated strain waves profiles in V_2O_3 Metallic thin film

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Vanadium sesquioxide is the working horse in the study of Mott-Hubbard phase transitions. At ambient pressure, it presents a first order symmetry-breaking paramagnetic metallic to antiferromagnetic insulating phase transition near 160K, while a third phase (paramagnetic insulator) can be obtained by changing pressure or doping. The ultrafast response of such materials to pump-probe experiments are very important both for the understanding of the underlying physics, and to potential applications. In particular, optical pump experiments [1, 2] have shown the possibility of inducing metallic nucleation-growth from the low-temperature insulating phase, while very high terahertz-pump fields ($>1\text{MV/cm}$) at 4K achieved insulating to metal transition through electronic tunneling [3].

In this presentation, we compare TeraHertz pump and Optical pump / Optical probe experiments performed on a V_2O_3 paramagnetic metallic thin film at room temperature. The photo-induced transmission signals were measured over tens of picoseconds (acoustic timescale). The influence of pump characteristics such as power and penetration depth is consistent with strain waves generation by thermoelastic effects [4, 5]. The difference between TeraHertz and Optical pumps results were interpreted using a 1D analytical thermoelastic model and will be discussed in the view of tailoring strain profiles to study volume changing materials.

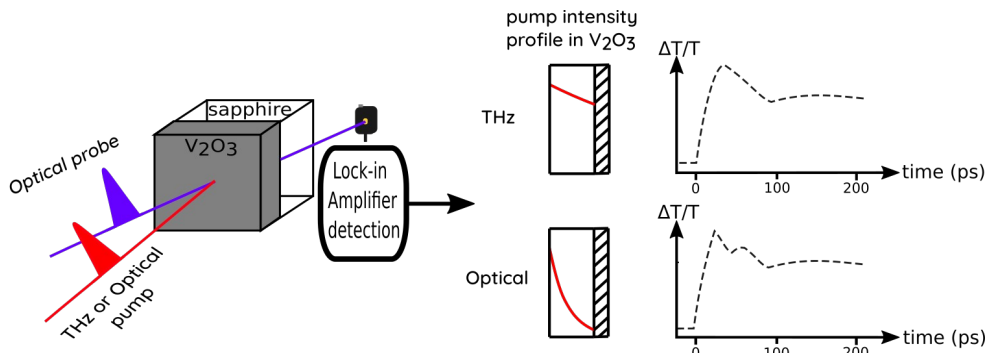


Figure: Scheme of the pump/probe experiments performed on V_2O_3 thin film

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Identifying different contributions to the ultrafast spin-to-charge conversion : Experimental approach

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Spintronics is the thriving field of research with aim to control the electron spin degree of freedom for potential applications in computing, storage and memory, and fundamental science. Spintronic devices are promising in terms of lower power consumption, higher information density, and non-volatility compared to conventional electronics. However, in order to utilize the electron spin to its fullest potential, the question about efficient generation, detection, transport and inter-conversion of the electron's intrinsic angular momentum should be answered.

Over the last three decades, scientific community have mastered the spin control in the static regime(DC) [1]. The breakthrough in transient and ultrafast spintronics can be attributed to the detection of the terahertz (THz) radiation, emitted as consequence of an ultrafast demagnetization [2], and ultrashort spin-current burst injection from ferromagnet into metallic layer [3]. In a such Ferromagnet/Metal heterostructure, the THz emission is mediated via Inverse Spin-Hall effect (ISHE). Recently, it has been demonstrated that the time-varying charge current can be generated via spin injection at the Rashba-split interface [4,5], or via hot-carrier gradient established in the magnetic heterostructure [6].

In this work, we use 165 femtosecond, 805 nm laser pulses to generate the ultrafast spin-current burst in 5 and 20 nm thick ferromagnet CoFeB. This net spin current then can be converted into the transient charge current within ferromagnet itself (AHE), at the Rashba states established at the CoFeB/MgO interface (IREE) or inside the 3 nm thick Pt layer (ISHE) deposited onto the CoFeB. With comparative analysis of experiments conducted for different geometrical sample orientations and at different pump wavelength, we try to estimate ISHE, IREE and AHE contributions to the overall THz radiation, emitted as a result of ultrafast spin-to-charge conversion.

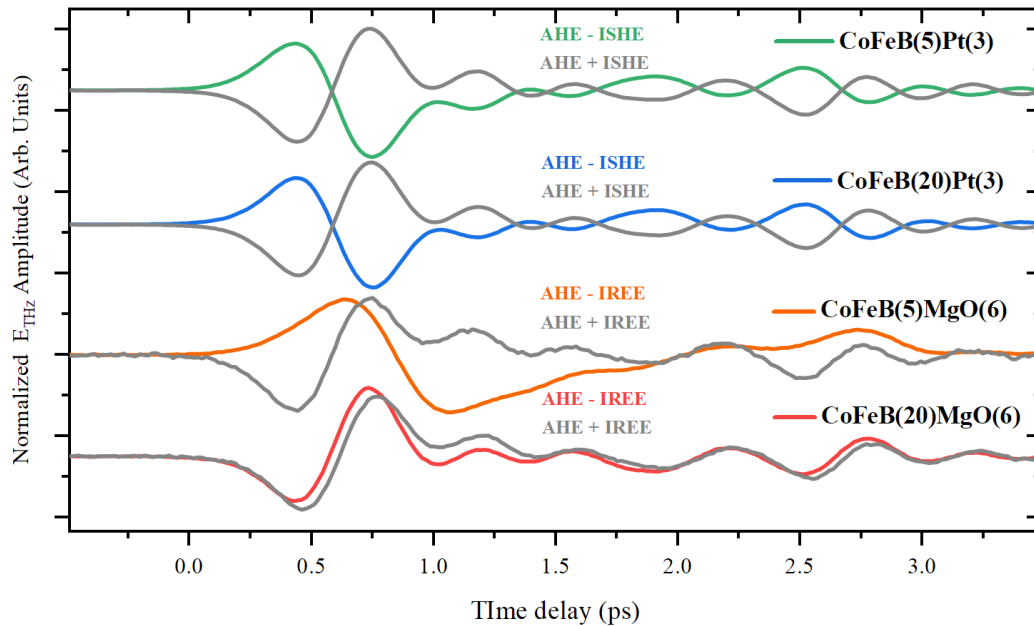


Figure 1: Transient THz waveforms emitted from the MgO- and Pt-based heterostructures with different thickness of CoFeB layer. Gray color corresponds to the laser excitation from the opposite face of the sample. All graphs are normalized to 1

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Condensed matter dynamics probed with attosecond transient absorption spectroscopy

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Attosecond pulses have extremely broad spectra lying in the extreme ultraviolet region. As they become increasingly available in reliable and routine manners, numerous opportunities are appearing in which they are used to probe new phenomena in condensed matter [1].

Here I will describe two experiments that illustrate two ways in which attosecond pulses can be used :

(a) A reflectivity experiment in magnesium oxide, in which we probed surprisingly short-lived core-excited excitonic states [2] and (b) An attosecond magnetic circular dichroism experiment in which charge and spin dynamics of conduction electrons are probed in a metallic multilayer at the few femtosecond timescale.

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Time-Resolved Dynamics in Molecules

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Photoionization of atoms, molecules, and small complexes creates a fundamental testing ground for quantum mechanical phenomena. In molecules, the dynamics of the individual atoms must be considered, thus providing a means to test the limitations of the standard Born-Oppenheimer model for nonadiabatic transitions. With the ultrafast ($\sim 10^{-15}$ s) light sources, such as lab-based tabletop lasers and facility-based free electron lasers (FELs), one can investigate inter- and intramolecular processes in the time domain, thus mapping out their evolution. In that regard, it is possible to “make a molecular movie” of the ultrafast reaction dynamics. We will present some of our recent work and possible projects for future research.

TRÉSOR – *Time-Resolved Electron Spectroscopy for Original Research* – at the European XFEL

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Electron spectroscopy offers a comprehensive view of the quantum energy levels of matter and is widely practiced using ionizing radiation from synchrotrons, resonance lamps, metal anodes, HHG sources, and FELs. With high spectral resolution, the electronic, vibrational and even rotational state of the target can be measured, making photoelectron spectroscopy (PES) an excellent tool to study the evolution of a chemical reaction, isomerization or fragmentation. Furthermore, taking advantage of the different binding energies of inner-shell electrons from different elements and even atoms of the same element in different chemical environments, molecules can be probed atom-by-atom with PES, making it an ideal tool for studying chemical transformations on their natural time scales, femto- and even attoseconds using an ionization source with the corresponding time resolution, i.e. XFELs and HHG sources. With short pulse duration light sources, it is also possible to study transitory systems, such as impulsively aligned molecules. Using pulsed lasers, molecules can be made to align, with recurrences of alignment occurring repetitively after the laser pulse has ceased. Molecules aligned by this method can then be studied by PES in the molecular frame using short pulses of ionization radiation such as that from XFELs and HHG sources, providing rigorous tests of quantum mechanical calculations of electronic structure and dynamics. Auger electron spectroscopy (AES) offers a complementary view of the energetics of the target system with electrons ejected with characteristic kinetic energies depending on the particular core hole and valance electronic structure of the dication and is independent of the wavelength and bandwidth of the ionizing source. XFELs can efficiently produce double core holes, where two inner-shell electrons are ejected from core shells before they can be refilled by Auger processes. Auger spectra of these exotic high-energy double core hole states of matter can provide insight into the dynamics of these systems. With these concepts in mind, we are planning to implement a high-resolution hemispherical electron energy analyzer at the SQS end-station of the European XFEL with funding from the French Ministry of Research through the CNRS. In this brief presentation the elements of the TRESOR project, why it is ideally suited for the properties of the European XFEL and what benefits it will provide for performing time-resolved experiments using XFEL radiation will be presented.

Unraveling ultrafast Rabi dynamics with a seeded free-electron laser

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Rabi oscillations [1], a prominent feature of coherent light-matter interaction arise when a few-level quantum system interacts periodically with an external radiation. Here, we used intense extreme-ultraviolet (XUV) pulses in the femtosecond domain from a seeded free-electron laser (FEL) to study Rabi dynamics in helium atoms via two-photon ionization. Our experimental and theoretical photoelectron spectra show clear Autler-Townes splitting [2] near the $1s^2 \rightarrow 1s4p$ transition in helium as well as level anti-crossing [3] of the combined atom-photon system as a function of the detuning from the transition. While these phenomena are mostly prevalent in quantum optics [4] where pure quantum fluctuations dominate, by virtue of the high temporal coherence along with the narrow spectral width available from the seeded FEL pulses at FERMI [5] we could directly study similar processes for photoionization from a dense atomic target, where statistical fluctuations take over. Our results highlight the importance of coherent population transfers driven by XUV-radiation to manipulate quantum matters in the ultrafast timescale.

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Multiscale conformational dynamics probed by time-resolved circular dichroism

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Circular dichroism (CD) is the property of chiral compounds to differently absorb right- and left-handed circularly polarized light. Coupled with pump-probe techniques, CD spectroscopy is a versatile tool to access the changes of conformation and electronic structure of chiral compounds over a wide range of time scales. However, despite recent technological advances, time-resolved CD experiments remain still underdeveloped, mainly due to their very weak signals subject to artifacts.

In recent years, we have designed several devices capable of measuring light-induced CD changes of various chiral compounds over a time window ranging from a few hundred femtoseconds to seconds. In this presentation, I will illustrate their applications to the study of the photoinduced chirality changes of 1,1'-bi(2-naphthol) derivatives and the dynamics of thermal denaturation and renaturation processes of short DNA G-quadruplexes.

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Revealing the ultrafast intersystem crossing in xanthone using wave packet quantum simulations

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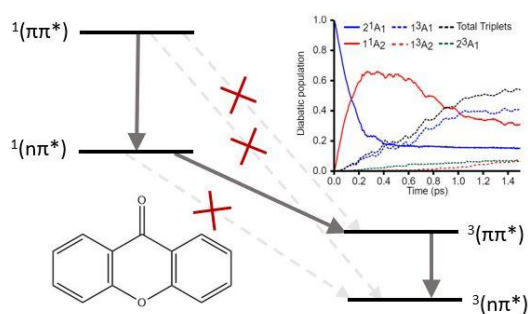
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Xanthone (9H-xanthen-9-one) is an aromatic ketone containing first-row elements that shows an unexpectedly fast intersystem crossing rate, in the picosecond time-scale. Despite several attempts from the experimental^[1,2] and theoretical^[3] perspectives, the mechanism still remains unclear nowadays.

In the current study, we have used nuclear quantum dynamics to elucidate the ISC mechanism in xanthone.^[4] With this aim, first of all, we constructed a model Hamiltonian based on the Duschinsky rotation for the diabatic states and we included the non-adiabatic and the (vibronic) spin-orbit coupling in the off-diagonal terms. Our results suggest a sequential deactivation mechanism following El-Sayed allowed path. (i) From the bright state, i.e. the second singlet excited state ($^1\pi\pi^*$), the lowest singlet state ($^1n\pi^*$) is populated in a fast internal conversion (85 fs). (ii) The next step is an intersystem crossing between $^1n\pi^*$ and $^3\pi\pi^*$ (2.0 ps). (iii) Finally, an internal conversion to the lowest triplet state ($^3n\pi^*$) is produced (602 fs). The transfers are produced in the near vicinities of electronic state intersections, which are located close to minimum energy structures. This allows efficient radiationless transitions from the initial singlet to the triplet states.



Schematic deactivation mechanism in xanthone and time-evolution diabolic states population.

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Towards 2D electronic spectroscopy of vibrational coherences in molecular motors and photoswitches

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The photoisomerization of C=C double bonds converts light energy – or electronic excitation energy – into mechanical energy at the molecular scale, i.e. functional vibrational motion. As a paradigmatic example, the biological function of the rhodopsin (Rho) protein, the sensor for vision, is triggered by the ultrafast photoisomerization of the retinal protonated Schiff base (rPSB), with a very good photoisomerization quantum yield (QY) of 66%. It has been predicted and supported experimentally^{1,2} that this high QY is controlled by the relative phase of essentially three critical vibrational modes in the vicinity of the conical intersection between the excited (S_1) and ground (S_0) states, where the system decays non-radiatively to S_0 after absorption of a photon. Reproducing such a photoreaction mechanism and its high QY in synthetic compounds, such as molecular switches³ or molecular rotary motors⁴ remains a challenge, because neither state-of-the-art quantum chemistry modeling, nor organic chemists' educated intuition can rationally predict how to modify a molecule design in order to improve its QY. The example of Rho and so-called biomimetic molecular switches designed to reproduce similar PES's and photoreactions⁵ are precious model systems to investigate both experimentally and theoretically the role of the vibrational motions and their relative phases at controlling the QY.

Experimentally, we have been applying vibrational coherence spectroscopy with a pump-probe experimental set-up using a sub-10 fs bleu-UV (350-430nm) pump pulse and a white light supercontinuum (320-850nm) as the probe.⁶ Recently, we have been upgrading this experiment towards Fourier-transformed 2D electronic spectroscopy, which will enable resolving the vibrational coherence signatures as a function of the pump wavelength (in addition to the probe wavelength, hence 2 wavelengths “dimensions”). The motivation is to be able to resolve the vibrational dynamics and possibly their relative phases as a function of the initial excess of vibrational energy in the Franck-Condon region.

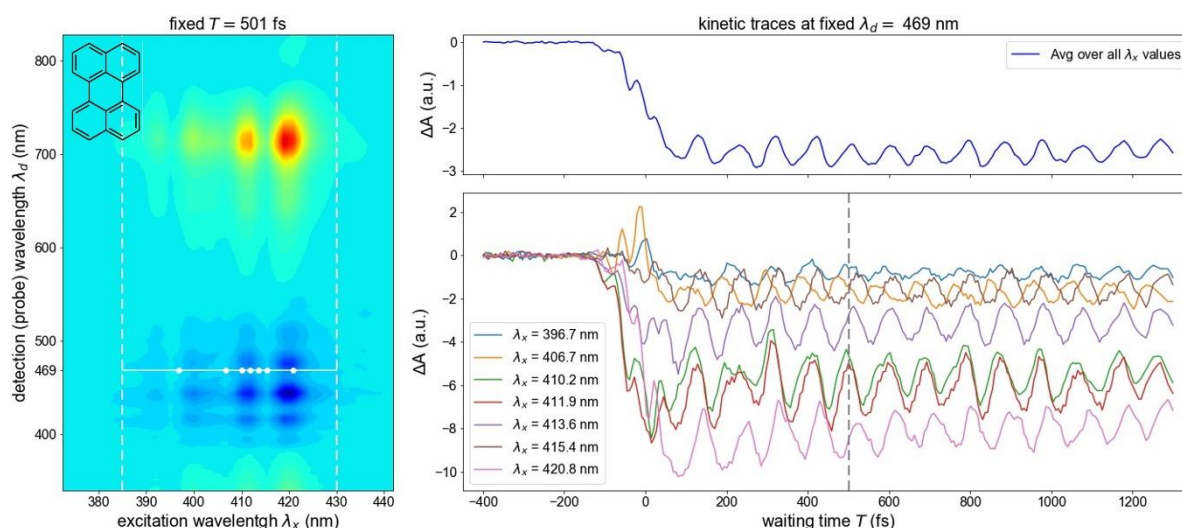


Figure: Left: 2D-resolved transient absorption signal at a fixed delay T ($T=501$ fs here) between excitation (bleu-UV pulse pair) and detection (probe). The excitation wavelength axis (λ_x) is reconstructed from Fourier transformation of the signal recorded as a function of the delay between the two excitation pulses. Right: Illustration of the oscillatory signal detected at a given probe – or detection - wavelength of $\lambda_d = 469$ nm, when (top) averaging over all excitation wavelengths λ_x between 385 and 430 nm, thus reconstructing the conventional 1D pump-probe signal, or when (bottom) monitoring the signal at a selection of individual excitation wavelengths. In the latter case the oscillatory components of are more pronounced and their phases strongly depend on the excitation wavelength.

Here we report on our latest experimental progress. In particular, we demonstrate a first proof-of-principle 2DES experiment performed on perylene chosen as a test compound (no photoisomerization, long excited-state lifetime) to demonstrate the performances of the new set-up. We observe very pronounced signatures of vibrational coherences excited and detected in the S_1 state with very good signal-to-noise ratio. Most importantly, their phases are observed to depend markedly on the excitation wavelength, a result which cannot be evidenced in a 1D (conventional) pump-probe experiment. This is an important step forward towards investigating the phase of vibrational motions in Rho-mimicking molecular switches or motors (work in progress).

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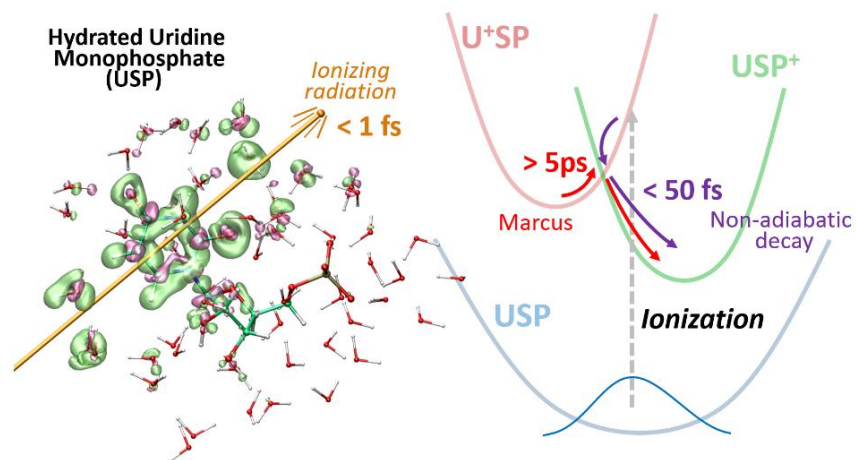
The Mystery of Sub-Picosecond Charge Transfer Following Irradiation of Hydrated Uridine Monophosphate

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The early mechanisms by which ionizing rays damage biological structure by so-called direct effect are largely elusive. In a recent picosecond pulse radiolysis study of concentrated uridine monophosphate solutions [1], unexpected results have been regarding the oxidation of the nucleobase. The signature of the oxidized nucleobase couldn't be detected 5 ps after the electron pulse, but only oxidized phosphate, raising intriguing interrogations about the identity of charge transfer mechanisms that could explain the absence of U⁺.

We address here this question by means of advanced first principles atomistic simulations of solvated uridine monophosphate. Our in-house codes combine Auxiliary Density Functional Theory (DFT) to polarizable molecular mechanics (QM/MMpol) [2]. We contrast three very distinct mechanisms of charge transfer covering the atto-, femto- and picosecond timescales. We first investigate the ionization mechanism and subsequent charge migrations on atto- to a few femtosecond time scales under the frozen nuclei approximation by means of Real Time Time dependent ADFT simulations. When then consider a nuclear-driven phosphate-to-oxidized-nucleobase electron transfer, showing it is an uncompetitive reaction channel on the sub-picosecond time scale, despite a high exothermicity and significant electronic coupling. Finally, we show non-adiabatic charge transfers are enabled by femtosecond nuclear relaxation after ionization. We show that electronic decoherence[3] and electronic coupling strength are key parameters determining hopping probabilities. Our results provide important insights on the interplay between electronics and nuclear motions in the early stages of the multiscale responses of biological matter subjected to ionizing rays [4].



Three ultrafast charge transfer mechanisms are studied by means of advanced ADFT/MMpol simulations. Reproduced with permission from [4]

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Ultrafast Photooxidation of Protein-Bound Anionic Flavin Radical

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Flavins are ubiquitous redox centers in proteins and widely involved in many essential biological processes. They can adopt three different redox states: oxidized quinone, one-electron-reduced semiquinone, and two-electron-reduced hydroquinone. In each redox state, the flavin can exist in different protonation states, with neutral and anionic species being biochemically relevant at physiological pH values.

In the oxidized state, the flavin is intrinsically highly fluorescent. However, when it is bound to proteins, in many cases the fluorescence is quenched by competing photo-induced electron transfer (ET) from neighboring redox-active residues or substrates to the excited flavin, leading to photoreduction of the oxidized flavin and formation of, often short-lived, flavin-residue or flavin-substrate radical pairs.^{1,2} Such photochemical processes play key roles in several functionally light-active flavoproteins,^{3,4} and also frequently occur in the vast majority of flavoproteins whose physiological functioning does not require light absorption. In the latter cases, as the oxidized form is the resting state of the flavin cofactor in most flavoproteins, this process can be viewed as photoprotective self-quenching preventing the formation of, potentially toxic, flavin triplet states.

In contrast to oxidized flavin, how the anionic semi-reduced flavin radical behaves upon excitation remains essentially unknown because of its intrinsically unstable nature. In this work, we studied the photoproducts of anionic flavin radicals in several flavoprotein oxidases where they can be stabilized: glucose oxidase, choline oxidase, monomeric sarcosine oxidase, D-amino acid oxidase and nitronate monooxygenase. Using ultrafast fluorescence and absorption spectroscopy, we unexpectedly established that their excitation, in either of two distinct absorption bands in the visible/near ultraviolet range, results in their oxidation on a time scale less than ~ 100 fs. The thus generated oxidized flavin photoproducts decay back to the anionic radical form in 10–20 ps. The identity of potential electron acceptors was investigated by molecular dynamics simulations and quantum mechanics calculations; positively charged active-site residues histidine and arginine are proposed to be the most likely candidates because of their close interactions with the flavins and appropriate electron affinities. To the best of our knowledge, this is the first report elucidating that, in addition to the commonly known and extensively studied photoreduction of oxidized flavins in flavoproteins, the reverse process, i.e., the photooxidation of anionic flavin radicals, can also occur. We propose that this process may constitute a universal excited-state deactivation pathway for protein-bound anionic flavin radicals. Our findings reveal a novel photochemical reaction in flavoproteins and further extends the family of flavin photocycles.⁵

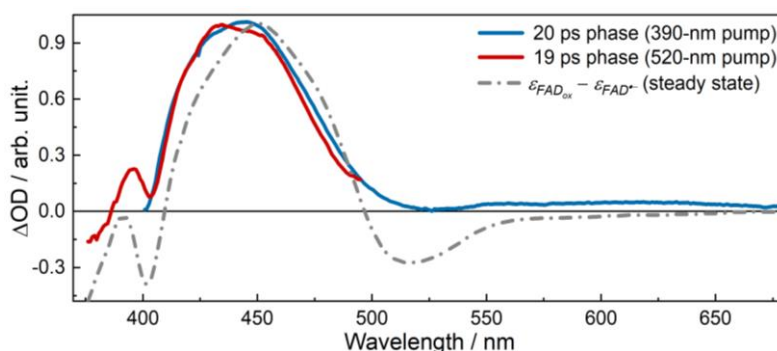


Figure 1. Decay associated spectra (solid lines) extracted from transient absorption data of anionic flavin radical in GOX, and steady-state difference spectrum assuming a photooxidation reaction (dashed line).

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CEP-stable infrared OPCPA sources

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Because of the intrinsically low conversion efficiency of nonlinear processes, parametric sources were essentially limited by the available energy/power of the so-called “pump” lasers. The advent of kW-class picosecond Ytterbium lasers delivering multi-mJ pulses at high repetition rate (10-100 kHz) has removed the long-lasting power bottleneck of ultrashort optical parametric amplifiers (OPAs) and opened the path to a large set of applications, among which are the generation of few-cycle optical pulses, high-flux mid- and far-infrared pulses, intense THz fields but also X-UV and soft X-Rays secondary sources through high-harmonic generation [1,2].

One of the key parameters of such sources is their ability to provide control of the carrier envelope phase (CEP). Usually, the latter is achieved by the generation of a passively stable CEP pulse, through a difference frequency generation process, with an active correction loop. However, depending on the central wavelength of the source, different schemes have to be used in order to generate a CEP stable seed, thus creating strong performance variations in terms of CEP stability.

We review the different generation schemes developed at Fastlite, all based on a single supercontinuum generation followed by a difference frequency generation process, and their consequences on the CEP stability through the presentation of several OPCPA systems. This scheme has the great advantage to be implemented with the same pump laser than the amplification stages. The supercontinuum generation step is based on filamentation in a bulk dielectric medium. It is important to identify how this highly non-linear process affects the determinism of the generated wave phase, and how this “loss” of determinism is distributed over the bandwidth. Pump energy distribution and its impact on the CEP is also investigated, as shown in figure 1.

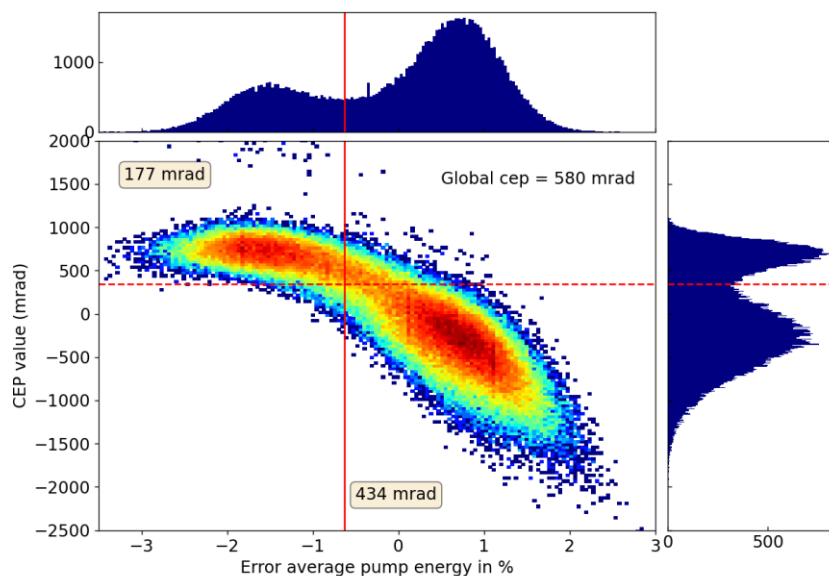


Figure 1: Correlation map between pump laser energy and output CEP measured by the mean of a f to $2f$ setup. Up and right plots shows the histogram distribution.

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Photoinduced short-range correlated charge density waves: growth and coarsening mechanisms

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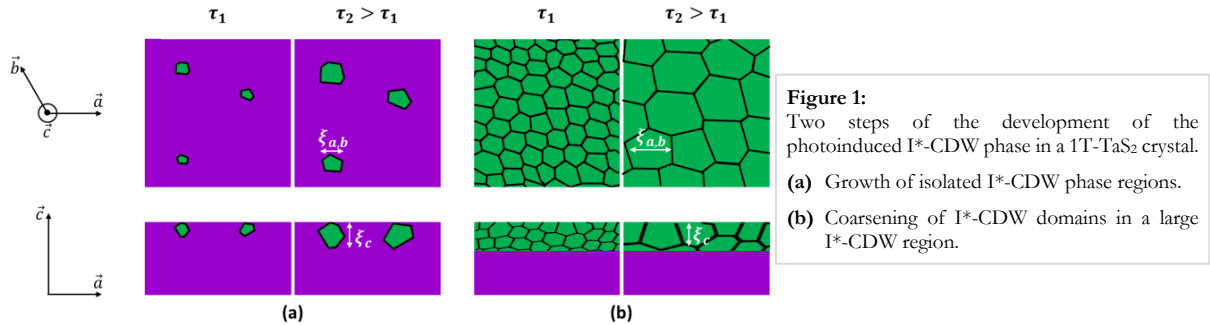
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Charge density wave (CDW) states are broken symmetry states of metals arising from electron-phonon interactions, which are characterized by a periodic modulation of both atomic positions and electron density. Diffraction techniques are especially well adapted to studying such compounds, since the structural modulation gives rise to so-called satellite peaks whose intensity is proportional to the square of the atomic displacement amplitude.

The CDW states are gapped, which makes them sensitive to excitations with infrared laser pulses. In the two-dimensional tantalum disulfide 1T-TaS₂, we found that an intense laser pulse can induce a phase transition between two distinct CDW states, namely the incommensurate (I) and nearly commensurate (NC) CDW states. A detailed description of the NC → I photoinduced phase transition was achieved using pump-probe X-ray diffraction measurements [1,2]. Above the threshold fluence for the NC → I transition, the NC-CDW melts within 400 fs. A broad satellite peak corresponding to the photoinduced I phase (denoted I*) could be detected 2 ps after laser excitation. The analysis of its intensity and width allows identifying a nucleation/growth mechanism of the photoinduced I* phase (**Fig. 1a**). At a pump-probe delay of 100 ps, the photoinduced I* phase has fully developed at the sample's surface. However, it is fragmented into several small domains (< 30 nm) corresponding each to a given phase Φ of the I-CDW modulation. The dynamics observed in the 100 - 500 ps pump-probe delay range is dominated by the coarsening of this domain pattern (**Fig. 1b**). The latter result was confirmed by low-energy electron diffraction [3].



It can be further shown that the photoinduced I*-CDW phase differs from the I-CDW phase at equilibrium not only by its limited correlation length, but also by a larger period of the CDW modulation (shorter wave vector of the CDW) [4]. We propose that the increased period of the I*-CDW modulation would be due to a smaller number of conduction electrons involved in the collective CDW state, some of them being trapped at CDW-dislocation sites. In this view, the photoinduced I*-CDW appears as a hole-doped I-CDW.

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Resonant propagation of short pulses: Collective effects in light-matter interaction

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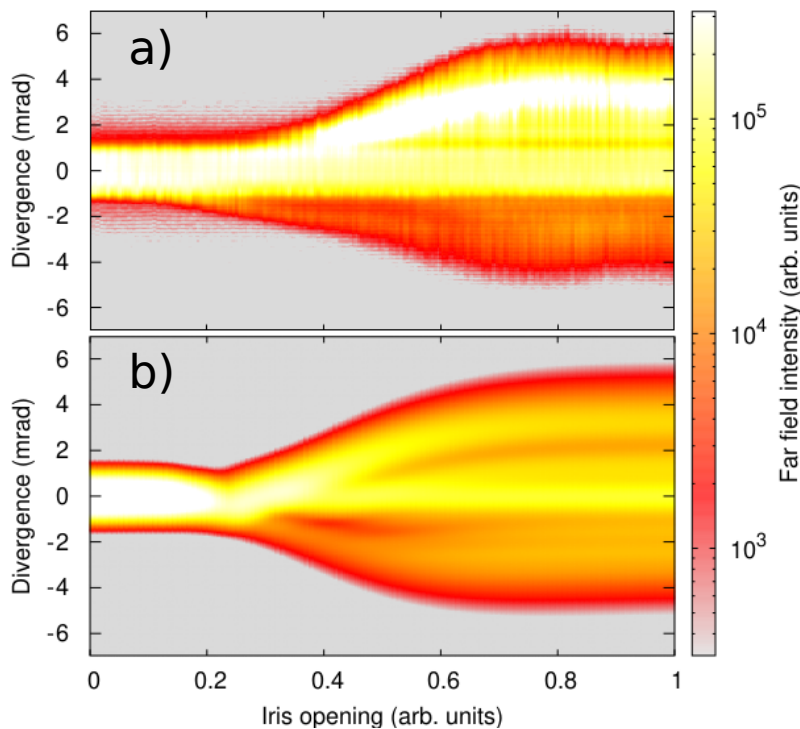


FIGURE - (Reproduced from [1]) Opto-optical modulation measurement: Far field divergence of the 2p emission in helium in (a) the experiment, and (b) the calculation. The intensity of the incident beam is controlled via an iris.

We simulate the propagation of a beam of light in a gas by solving the coupled Maxwell wave equations and the Time-dependent Schrödinger equation. This framework allows to simulate collective effects in light-matter interaction. Two illustrations are given: simulation of opto-optical modulation in helium [1], and simulation of the resonant propagation of X-ray short pulses in Neon [2].

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Dipolar instabilities in N_2 theoretically evidenced by an ultrafast pump-and-probe setup

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The irradiation of a finite electronic system by an ultrafast XUV pulse can lead to the ultrafast creation of core-holes with strong subsequent electronic rearrangements. A standard theoretical tool to investigate such excitations that avoids the cost of the simulation of the XUV irradiation is to start the dynamics with an instantaneous core-hole excitation. However, in recent studies, we very often observed the setup of a dipolar instability in many different finite electronic systems (1D, 2D, 3D, with metallic or covalent bonding, etc.). One can question the physical origin of such an instability. Interestingly, we also observed a dipolar instability in the dimer N_2 after irradiation by a 1fs-XUV pulse. If it is not a pure numerical effect, can one observe it experimentally and how? We will show in this talk that, with an ultrafast XUV-pump/XUV-probe setup, one can indeed retrieve a signature of this dipolar instability in photo-electron spectra when one varies the delay between the pump and the probe.

High-harmonics and particle beams emission from relativistic plasma mirrors at kHz repetition rate

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We report on recent progress on high-order harmonics generation, proton and electron acceleration from plasma mirrors (Fig 1a) driven by relativistic-intensity, near-single-cycle pulses at a 1kHz repetition rate using the Salle Noire 2.0 laser system [1] at Laboratoire d'Optique Appliquée.

When focusing a femtosecond laser pulse to relativistic intensities ($\sim 10^{19}$ W.cm⁻²) onto a density-gradient controlled plasma mirror, high-order harmonics are generated into the XUV spectral range via the temporal compression of the driving optical waveform through the relativistic oscillating mirror mechanism [2], corresponding to a train of attosecond pulses in the time domain. This mechanism is highly interesting for pump-probe applications to resolve ultrafast phenomena in matter, owing to the predicted high, percent-level efficiency [3,4].

The process can be time-gated such that for an optimal range of near-single-cycle driver waveforms, such as those achieve at LOA, an isolated attosecond pulse is generated, as indicated by the observation of an XUV spectral continuum [5]. Recent progress on the laser system now allows us to stabilize and change on demand the waveform of the driving light pulse and measure fine details of the transition between different gating scenarios of the laser-plasma interaction, through simultaneous detection of HHG and relativistic electron beams.

By varying the driving pulse duration and the plasma density ramp at the target surface, the emission of charged particle beams can be enhanced. In particular, we found a regime with sharp density gradients, where ~ 0.5 MeV protons are emitted along the target normal direction, in a beam of exceptionally low divergence for laser-plasma accelerated protons. 2D PIC simulations suggest that the proton energy should linearly increase with the incident laser pulse energy, which is promising for potential applications requiring MeV-scale proton energies, such as the production of radio-isotopes for medicine [6].

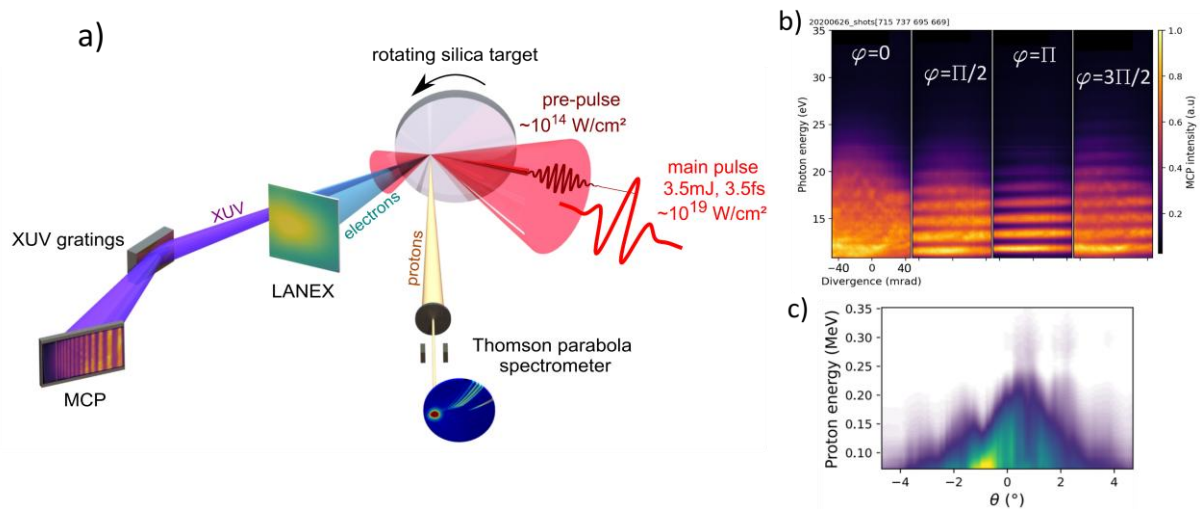


Figure 1. a) Schematic representation of the experimental setup, showing the XUV spectrometer, the LANEX screen for spatial electron detection and the Thomson parabola spectrometer for proton spectra measurements. A prepulse is used to control the plasma gradient scale length. b) XUV spectra for different values of the 3.5fs driving pulse carrier-envelope phase, with an arbitrary offset. c) Angularly-resolved proton spectrum obtained for a driving pulse duration of 200fs, measured with a small MCP used as a time-of-flight detector (not shown in a)).

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Mesure spatio-temporelle d'impulsions attosecondes d'origine relativistes

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Pour observer des effets QED à partir de la lumière, il est nécessaire de trouver des stratégies pour approcher le "Champ de Schwinger". Ce champ vaut $E=1.32 \cdot 10^{18}$ V/m et conduit à des éclaircements lumineux supérieurs à $4.7 \cdot 10^{29}$ W/cm² soit presque sept ordres de grandeurs au-delà des records atteints par les lasers de type PW focalisés sur des foyers optiques proches de la limite de diffraction.

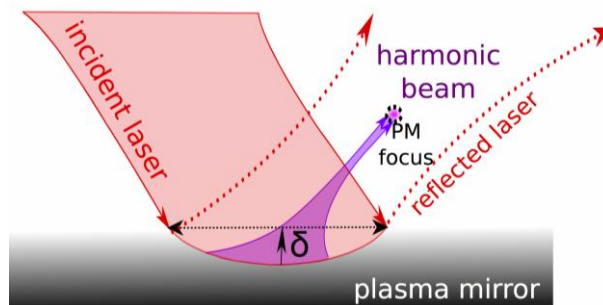


Figure 1 : Schéma de principe de l'expérience. Le rayonnement laser incident est converti de l'infrarouge (800 nm) à l'XUV (focus) après réflexion sur le miroir plasma (PM)

Une approche consiste, non pas à augmenter l'énergie, mais à (i) réduire la surface sur laquelle celle-ci va être concentrée en réduisant la longueur d'onde (meilleure focalisation) et (ii) réduire la durée des impulsions dans le domaine de l'attoseconde (augmentation de la puissance instantanée). Pour cela, on focalise un laser femtoseconde très intense (Laser UHI100 $> 10^{19}$ W/cm²) sur un miroir dont la surface va être instantanément ionisée pour osciller à l'échelle du cycle optique à une vitesse proche de la vitesse de la lumière (un miroir plasma), générant par effet Doppler des longueurs d'ondes beaucoup plus courtes que celle du laser incident (génération d'harmoniques d'ordre élevé) tout en incurvant la surface par pression de radiation (figure 1). Cette opération résulte en trois effets qui se cumulent pour augmenter considérablement l'intensité au foyer : i) les longueurs d'ondes générées sont courtes donc se focalisent plus fortement, ii) la pression de radiation agit comme un miroir parabolique dont la courbure augmente également la focalisation et iii) toujours grâce à l'effet Doppler provoqué par la réflexion du laser UHI, chaque cycle optique se raccourci temporellement menant à une augmentation de la puissance instantanée.

Dans cet exposé, nous proposons de mesurer quantitativement les propriétés à la fois spatiales et temporelles du faisceau de ce laser "boosté" au point de focalisation en utilisant une extension des méthodes d'imagerie sans lentilles connues sous le nom de méthodes ptychographiques en y ajoutant une dimension temporelle consistant à déplacer un objet sous le faisceau d'une manière contrôlée à des vitesses proches de celle de la lumière.

"Spatio-temporal characterization of attosecond pulses from plasma mirrors"

Chopineau L, Denoeud A, Leblanc A, Porat E, Martin P, Vincenti H & Quéré F, **Nature Physics.**, Vol 17, August (2021)

"Achieving Extreme Light Intensities using Optically Curved Relativistic Plasma Mirrors", H. Vincenti, **PRL** 123, 105001 (2019)

GeV laser-plasma acceleration in an all-optical plasma waveguide

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Laser-plasma accelerators produce electric fields of the order of 100 GV/m, more than 1000 times larger than radio-frequency accelerators [1]. Thanks to this unique field strength, they appear as a promising path to generate electron beams beyond the TeV, for high-energy physics [2]. Yet, large electric fields are of little benefit if they are not maintained over a long distance. It is therefore of the utmost importance to guide the ultra-intense laser pulse that drives the accelerator. Reaching very high energies is equally useless if the properties of the electron beam change completely shot to shot due to the intrinsic lack of stability of the injection process. While present state-of-the-art laser-plasma accelerators can already separately address guiding [3] and control [4] challenges by tweaking the plasma structures, the production of beams combining high quality and high energy is yet to be demonstrated. Here we use a new approach for guiding the laser and combine it with a controlled injection technique to demonstrate the reliable and efficient acceleration of high-quality electron beams up to 1.1 GeV, from a 50 TW-class laser.

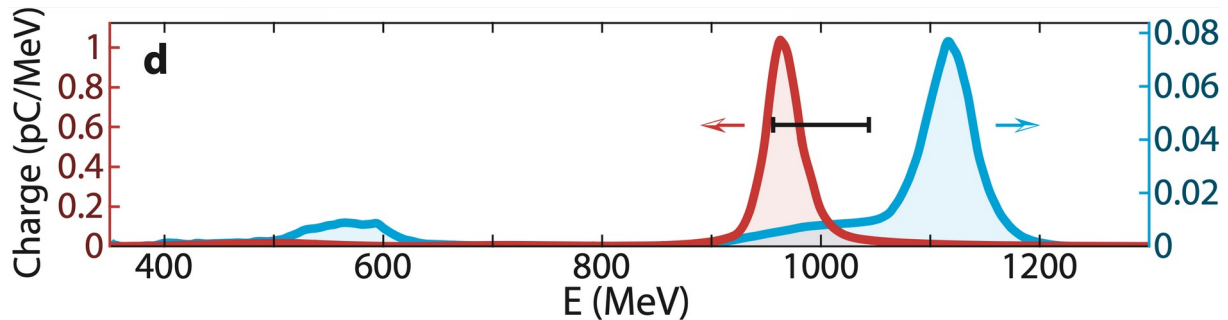


FIGURE Two examples of angularly-integrated spectra. The black segment indicates the uncertainty on the energy due to fluctuations of beam pointing.

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Extreme-Ultraviolet Vortex and Vector-Vortex Beams with very high Topological Charge

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Light beams structured in their phase and polarization have proven their usefulness for a wide range of applications [1, 2]. On the one hand, optical vortex beams carry orbital angular momentum (OAM) and are characterized by their azimuthally twisting wavefront; the total wavefront twist in the transverse plane signifies their topological charge ℓ . On the other hand, vector beams exhibit a spatially varying polarization. In the recent past, the coherent high-order harmonic generation (HHG) in noble gases has provided a straightforward alternative to upconvert long-wavelength vortex and vector beams to the extreme ultraviolet (EUV) spectral regime [3, 4, 5]. In regards to OAM-driven HHG, the momentum conservation imposes a linear upscaling of the driving beam topological charge ℓ_1 [3, 4]: $\ell_q = q\ell_1$, where ℓ_q is topological of the qth harmonic.

In this work, we will present our recent results on the generation and characterization of EUV vortex beams exhibiting a very high topological charge (up to $\ell_q = 100$) [6]. Furthermore, we will demonstrate the production of EUV light beams that bear combined characteristics of the vortex and vector beams: EUV vector-vortex beams comprising twisted azimuthal phase of a vortex beam and the spatially inhomogeneous polarization profile of a vector beam [7].

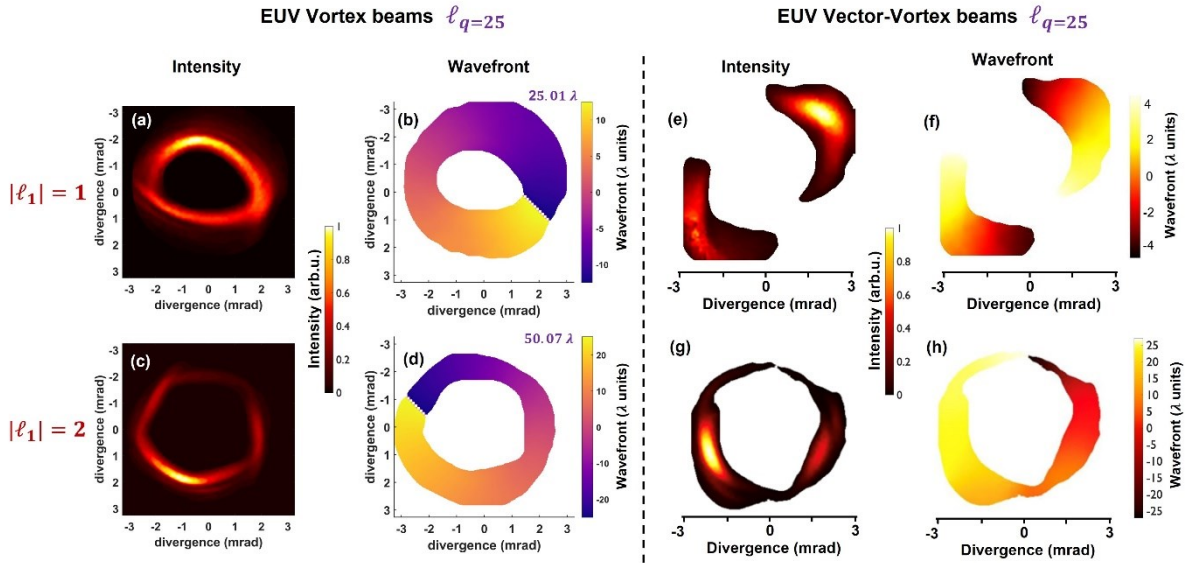


Figure 1: EUV vortex and vector-vortex beams with high topological charge. Left panel: (a) intensity, (b) wavefront of the 25th harmonic for IR driving beam of topological charge $|\ell_1| = 1$. In (c, d), the intensity and wavefront are shown for the driving beam of $|\ell_1| = 2$. The wavefront is represented in the units of the central wavelength of the high-harmonic beam. The total wavefront twist designating the topological charge of EUV vortex beams is (b) $\sim 25.01 \lambda$ for $|\ell_1| = 1$, and $\sim 50.07 \lambda$ for $|\ell_1| = 2$. In the right panel, we show the characterization of EUV vector-vortex beams (25th harmonic) with a high topological charge. The intensity and wavefront of the vertical polarization component for $|\ell_1| = 1$ are shown in (e) and (f), respectively. For a vector-vortex driving beam of $|\ell_1| = 2$, intensity and wavefront of the vertical polarization component are depicted in (g) and (h), respectively.

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SourceLAB, fournisseur d 'accélérateurs Laser Plasma

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SourceLAB, spinoff du Laboratoire d'Optique Appliquée, commercialise depuis sa création des instruments scientifiques fiabilisés, directement inspirés du savoir-faire des groupes de recherche en interaction Laser Plasma. Récemment, une étape supplémentaire vers la constitution de la technologie Laser Plasma a été franchie : l'intégration d'un accélérateur laser plasma kHz en un produit tout-en-un prêt à l'emploi, le SL-KAIO.

Contributions par Affiche

Photodynamic study of new near-iR Reversible Switchable Fluorescent Proteins (iR-RSFP)

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The development of new photo-switchable fluorescent proteins (RSFPs) that are working in the red / NIR domain is one of recent challenge in super-resolved fluorescence microscopies. The group of Prof. S. Jakobs (Max Planck Institute, Göttingen, Germany) developed a new iR-RSFP (Figure A) engineered on *Deinococcus radiodurans* bacteriophytochrome (Dr-BphP). The use and optimization of this new protein require to understand its photo-dynamics. BphP photo-switching are reported to involve cis-trans isomerization of the excited chromophore, deprotonation / protonation steps, and structural changes in the protein. Photo-dynamics takes place on several order time-scale, ranging from hundreds of femtoseconds to a few milliseconds, involving several excited states and intermediates (Figure B).

We investigated the photo-dynamics of the new iR-RSFP using femtosecond pump-probe transient absorption spectroscopy, fluorescence single photon counting techniques, and nano-millisecond transient absorption spectroscopy. In the later method, we upgraded the system with an intensified camera (iCCD) and characterized it (signal /noise, resolution). We will discuss here the switching dynamics of this new iR-RSFPs and the difference with known mechanism for BphPs (Figure 2).

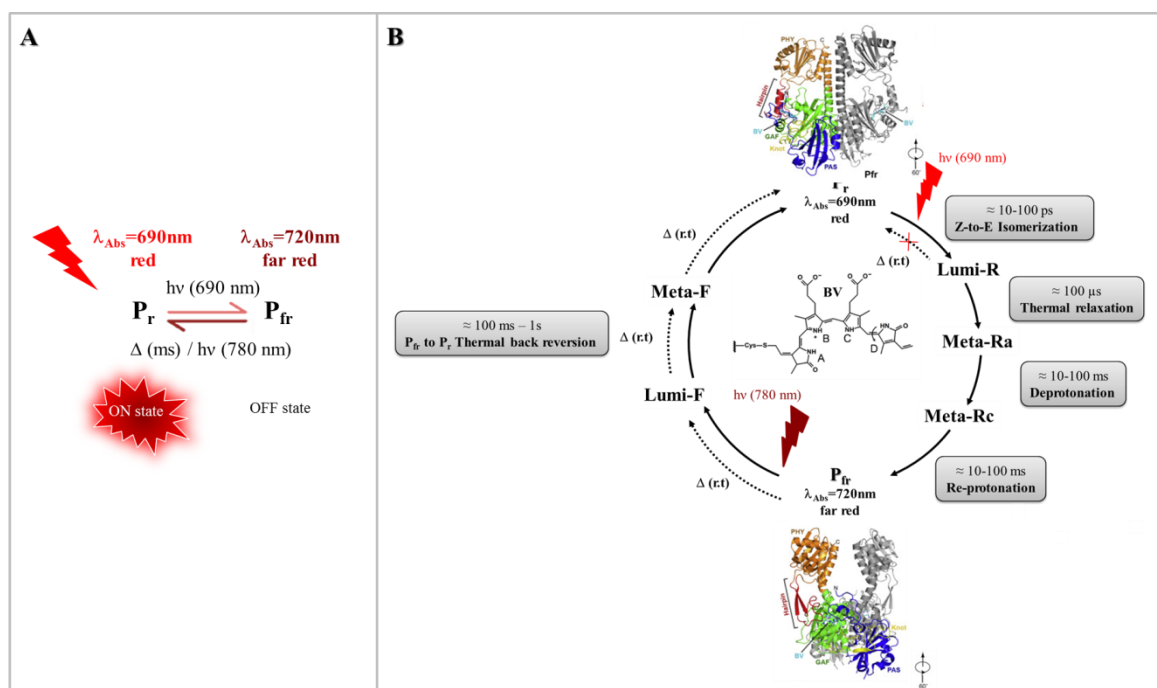


Figure : (A) On and Off switching of a bacteriophytochrome-based iR-RSFP. (B) Photocycle of Dr-BphP with biliverdine (BV) isomerization and P_r-P_{fr} crystal structures.

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Attoscience with exotic molecular systems

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The first goal of this theoretical work is to assess the relative stability of exotic atoms, which can then be studied dynamically through interactions with ultrashort tailored pulses.

We have started by looking the positronic chloride ion, PsCl, insofar as this candidate has great potential to be produced experimentally at CERN. Hartree-Fock calculations [1, 2] have been performed in the first step followed by the use of a post Hartree-Fock method. This latter, which is called singly and doubly excited configuration interaction (SDCI) [3], has been implemented by using an open-source programming environment for quantum chemistry especially designed for wave function methods (more information about Quantum Package can be found in [4]). The first SDCI results are in good agreement with the literature and showed unfavorably that only the ground state of this exotic atom is bound, with a bound energy around 1.5 eV for the positron (see the table below). Although no dynamic study is then possible with this candidate in the future, this finally allowed us to compute cross sections involving positronium (Ps) and positronic atoms in the Coulomb Born approximation [5, 6] ($\text{Ps} + \text{Cl}^- \rightarrow \text{PsCl} + e^-$ in the present case).

<code>i_states</code>	E(PsCl) (au)	BE(e^+) (au)
1	-459.97537160	-0.16296283
2	-459.70053563	0.11187314
3	-459.70053563	0.11187314
4	-459.70053563	0.11187314
5	-459.64712477	0.16528400

Table - SDCI energies for the first 5 lowest states and corresponding positron binding energies obtained by using a modified aug-cc-pvqz GTO basis for chlorine.

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Ultrafast dynamics with bi-dimensional infrared spectroscopy

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Two-dimensional spectroscopic technique in the infrared domain (2D-IR) provides information beyond linear infrared spectra, by spreading the vibrational information along multiple axes giving access to spectral information (molecular structure, vibrational mode coupling, anharmonicities) and chemical and molecular dynamics (energy transfer rates and structural changes) at the femtosecond timescale [1].

We have recently implemented a 2D-IR setup at ISMO that is equipped with a novel IR detector (screen of 320x256 pixels), allowing an increased resolution ($<1\text{ cm}^{-1}$) [2]. Preliminary measurements were done on iron pentacarbonyl complex ($\text{Fe}(\text{CO})_5$ or IPC). IPC is a textbook example of fluxionality where the molecule undergoes a rapid structural rearrangement between two similar geometries after passing through a transition state having a different geometry [3]. Our findings so far are in agreement with previous study [4] and thanks to our higher resolution setup ($<1\text{cm}^{-1}$ v. $\sim 4\text{cm}^{-1}$, in Ref 4), we could determine more accurately the anharmonicity between the vibrational modes that are probed.

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Imaging photoemission dynamics in real time: 1s3p resonance of Helium

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Imaging in real time the complete dynamics of a process as fundamental as photoemission has long been out of reach due to the difficulty of combining attosecond temporal resolution with fine spectral and angular resolutions. It is only recently that orientation-resolved spectral phase measurements could be performed, using cold target recoil ion momentum spectroscopy (COLTRIMS) [1-3], or velocity-map imaging spectroscopy (VMIS) [4-7].

In the present study, we combine attosecond spectral interferometry of electronic wavepackets with VMI momentum spectroscopy. We use an attosecond pulse train overlapped with its generating IR field on helium to populate coherently both the 1s3p state and free electronic wavepackets through two-photon XUV+IR transitions in the continuum.

After a variable delay, the 1s3p resonance is ionized by an additional IR field. By measuring the delay-dependent photoelectron spectrum, we are able to measure the phase difference between the 1s3p state and the free wavepacket with a spectral resolution much better than the inverse of the IR duration. We observe a linear relationship between the oscillation frequency and the energy of the IR photon absorbed. Additional analysis are still on going to understand the spectral dependence of its phase.

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Towards Capturing Non-Local and Non-Adiabatic Effects in TDDFT

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Knowledge of sub-femtosecond electron dynamics is of utmost importance for exploring electron-transport processes which set the intrinsic limit on the operating speed of present-day electronic devices. The ultrafast electron dynamics in materials are often captured under excitation by high-intensity laser fields. A theoretical understanding of this dynamics, therefore, calls for a time-dependent quantum mechanical treatment for obtaining non-perturbative response of many interacting electrons under strong-field irradiation. To this end, we study the non-perturbative response by revisiting the ionization dynamics in one-dimensional (1D) systems, notably in 1D He model for which the time-dependent Schrödinger equation (TDSE) can be solved exactly giving access to exact results. However, for systems having more than two electrons TDSE calculation is numerically demanding if not impossible. In this context, time-dependent density-functional theory (TDDFT) based approaches are therefore, highly solicited due to its scaling with the size of the system. However, although formally exact, in practice the exchange-correlation potential ($v_{\text{KS}}[n](x,t)$) in TDDFT is adiabatically approximated and therefore fails to capture the non-adiabatic contribution to system's response. The goal of this work is to look for a suitable non-adiabatic and (spatially) non-local exchange-correlation potential in TDDFT in order to adequately exactly capture the non-perturbative response. To this end, we have looked into the time-dependent ionization dynamics in 1D He model in order to precisely understand *when* and *how* the TDDFT fails to describe single-ionization. At the same time, we are developing a numerical tool to obtain the *exact* potential $v_{\text{KS}}[n](x,t)$ by inverting the time-dependent Kohn-Sham equation which will help to improve the spatio-temporal behavior of the potential and thus, to obtain non-adiabatic Kohn-Sham potential capable of capturing exact ionization in TDDFT.

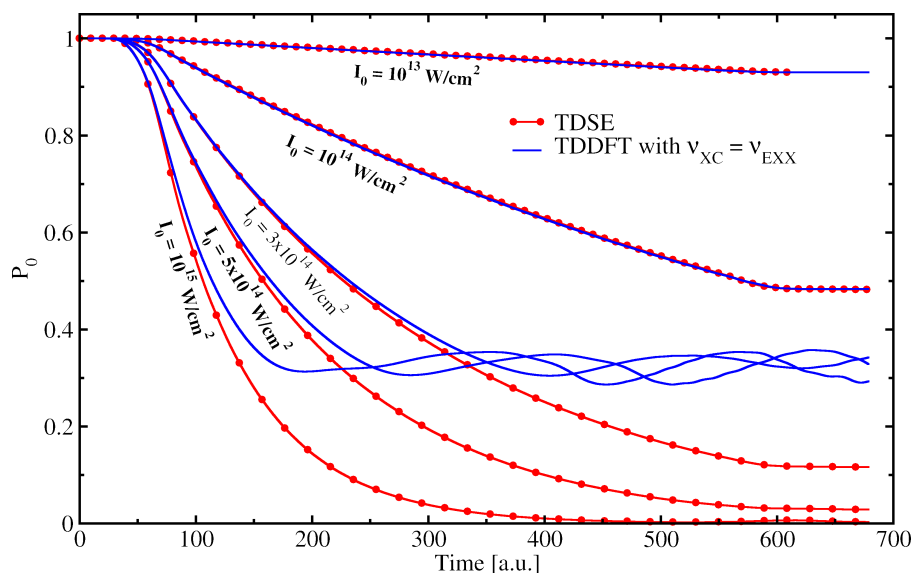


Figure: Time dependence of single-ionization in one-dimensional He atom for different intensity of the exciting laser (frequency=1 a.u.).

Simulation of radiolysis in organic phases with plutonium

6

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Recycling spent nuclear fuel is a major issue in the power plant industry. Nowadays, the current industrial process used is called PUREX (Plutonium Uranium Refining by Extraction). It is a solvent extraction technology that employs tributyl phosphate to achieve plutonium and uranium extraction. The radioactivity causes radiolysis of both extractant and the solvent molecules, and form unwanted products. So, understanding such radiolysis phenomena is a necessary condition for the development and the improvement of new recycling processes. To date, one can isolate the degradation products with characterization processes while others experimental technics, pulsed radiolysis for instance, give access to their formation mechanisms at the picosecond time scale. However, the sub-femtosecond time scale is totally out of range for the experimental equipment and the phenomena that happen at this time scale are not well understood.

The aim of this work is to model radiolysis in organic phases with plutonium through numerical simulation, with quantum chemistry tools in ultra-short time scale. Tributyl phosphate is used as a reference system for the calculations. We will adopt the Real-Time Time-Dependent Density Functional Theory (RT-TD-DFT) to give access to the deposit energy and the electronic dynamics a few time steps after the interaction with alpha particles. Moreover *ab initio* molecular dynamics simulations will be used to predict the nuclear motion and simulate the absorbed energy diffusion in the system.

The presentation will be divided in three parts. First, we will briefly introduce RT-TD-DFT method. Then, we will present the consequences of a large deposit energy in the electronic cloud, due to an ionizing particle, on properties such as the atomic charges or the kinetic energy density. These first results are obtained at the attosecond time scale. To conclude, we will consider the nuclear motion in the simulations with *ab initio* molecular dynamics simulations. This will allow to understand the energy transfer from the electronic density to the vibrational modes of the molecular system which is the initial step toward degradation mechanisms (through charge transfer or broken bonds).

Attosecond transient absorption manipulation of molecular sample response reveals sub-natural linewidth structures in iodine $N_{4,5}$ pre-edge resonances.

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X-ray photons have enough energy to access inner shells of molecular electronic configurations. At these energies, the wavefunctions of the vacancies created via absorption are localized on a single atom providing element specificity to x-ray spectroscopies. The decay of these core-excited states occurs via pure electronic dynamics driven by correlations and often occurs below nuclear motion and is responsible for the broad features observed in x-ray spectroscopy.

Attosecond transient absorption allows us to manipulate the system's response before the core-excited state decays, providing a unique tool to obtain sub-natural linewidth resolution. This approach is here demonstrated on iodine containing compounds at the iodine $N_{4,5}$ edge located around 57 eV. The absorption at the $4d^{-1}6p$ transitions of ICl, CH_3I and iodine radical is studied using a 200 as pulse, and the consecutive sample response, i.e. the free induction decay (FID), is on the tens of femtoseconds timescale. Using a 3.5 fs visible pulse the FID is perturbed via AC Stark shift and subtraction of the unperturbed response yields a transient response with narrowing full width at half maximum with time delay. The narrowed features reveal peak structures unresolvable in the static absorption spectrum. While the narrowing effect is reproduced by solving the time-dependent Schrodinger equation. The revealed features correspond to the lifting of the $4d$ core-orbital's degeneracy due to ligand field interaction with the nearby atoms (in ICl and CH_3I). The initially broad lineshapes with FWHM >200 meV narrow down to 50 meV and splittings as small as 130 meV are easily resolved. This new approach can be extended to other atomic edges at higher energy and offers a new route to enhance the resolution of all x-ray absorption spectroscopy via laser manipulation on the attosecond timescale.

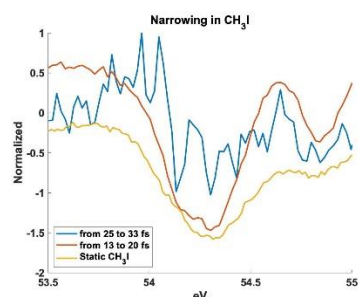


Figure 1 Lineshape narrowing in the attosecond transient absorption spectra of CH_3I . The negated static spectrum is shown in yellow and the time resolved spectra at two time delays in orange and blue. The broad lineshape in the static spectrum corresponds to the $4d_{5/2}6p$ transitions. The time resolved spectra show a clear narrowing toward three negative features consistent with the ligand field splitting of the $4d_{5/2}$ level.

Observation of magnetic helicoidal dichroism with extreme ultraviolet light vortices

8

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Light polarization is a fundamental tool in the study of magnetism and its dynamics, thanks to a number of magneto-optics phenomena such as magnetic circular dichroism (MCD) and the Kerr effect. The polarization of a beam is associated to the spin angular momentum (SAM) of the photon, with $s = \pm 1$ for circular and $s = 0$ for linear polarization. A photon can carry also an orbital angular momentum (OAM), indexed by $\ell \in \mathbb{Z}$, which corresponds in the wave picture to a helicoidal wavefront of light instead of a plane wave. The OAM of light, however, has been much less exploited than SAM as a probe of magnetic materials. Light beams carrying OAM in the extreme ultraviolet (XUV) range, of interest for magnetic resonances, have become available at HHG and FEL sources only recently. I will present the classical electromagnetic framework for the reflection of light carrying OAM by a non-uniform magnetic material. It is found that a beam of mode ℓ is modified into $\ell + n$ after reflection, where n identifies the symmetry properties of the magnetic structure seen by the beam in terms of azimuthal Fourier decomposition. This leads to a differential scattering that depends on the sign of ℓ , giving rise to what we called magnetic helicoidal dichroism (MHD) [1]. Similarly to MCD, MHD can be observed by inverting the sign of the OAM (MHD ℓ) or of the magnetization (MHD m). But instead of being a local probe of the magnetic properties, it carries information about the overall topology of the magnetic structure. I will also present the first experimental observation of MHD measured on a permalloy magnetic vortex using the FERMI FEL light source at the Fe 3p resonance [2]. The experimental results agree with the theoretical predictions (Fig. 1), setting the ground for scan-free investigations of ultrafast dynamics of magnetic structures with MHD.

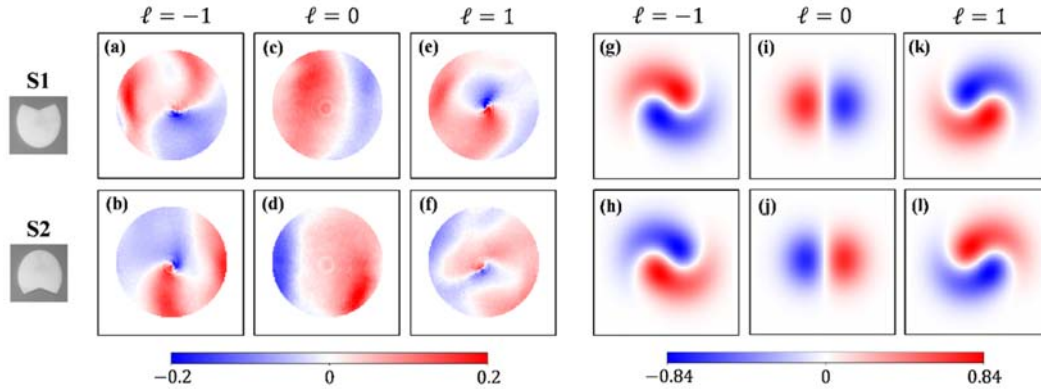


Fig.1. (a-f) Experimental results and (g-l) numerical simulations for MHD m using $\ell = -1, 0, +1$ in scattering by a magnetic vortex in two permalloy dots (S1, S2), resulting in opposite helicity at a given external magnetic pulse.

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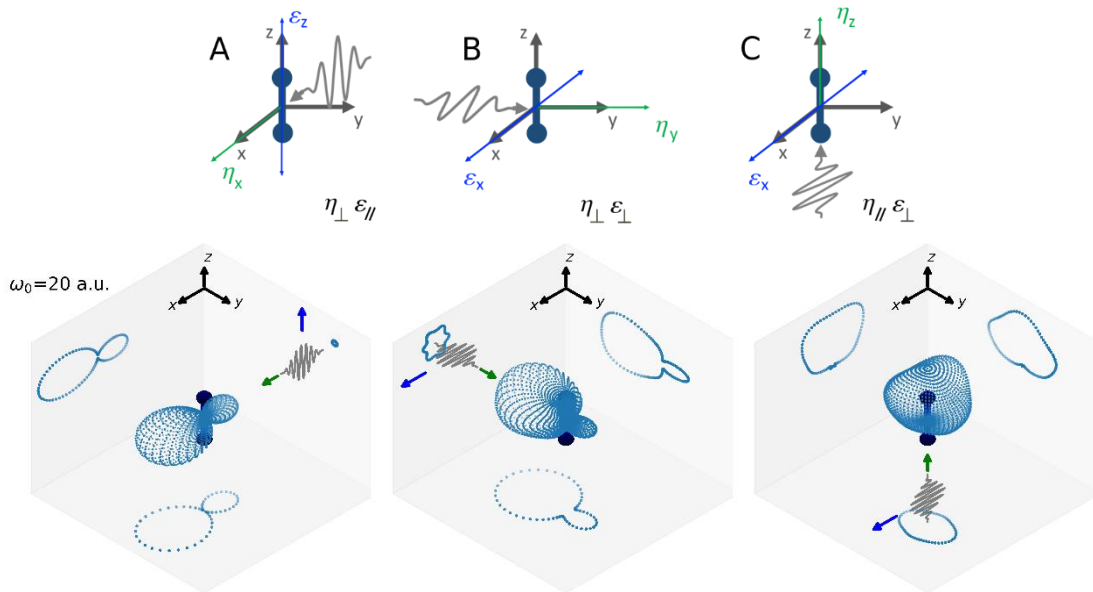
Angle-dependent interferences in electron emission accompanying stimulated Compton scattering from molecules

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We present the theoretical results of single photoionization of H₂ submitted to a strong ($I = 10^{18}$ W/cm²) short (66 as pulse duration) soft X (central frequency 544 eV or 2,3 nm wavelength) pulse. In such conditions, a non-linear process where one photon is absorbed and another is emitted with frequencies of the pulse bandwidth can occur and leads to ionization above threshold, this is the so-called Stimulated Compton Scattering (SCS). This non-linear process, investigated by solving the time-dependent Schrödinger equation, is unambiguously associated with a rather strong signal at low photoelectron energy. We study the angular dependence of photoelectron spectrum in the molecular frame for a linearly polarized X-field. The expected manifestation of the SCS, as shown in atomic system [1-2], is an asymmetry of the angular distribution in the forward direction related to the direction of the field propagation, i.e. a nondipole effect which plays a crucial role in this regime. As shown in the following figure, there are configurations for which the electrons are unexpectedly ejected preferentially in the backward direction (configuration B) [3]. In the discussion we will also present results for the case of a two-color field which appears to be accessible nowadays at FEL facilities [4].



Plot of the angular photoelectron spectrum obtained by single ionization of the H₂ molecule by a $I = 10^{18}$ W/cm², $\tau = 66$ as, $\omega_0 = 20$ a.u. The electron energy is integrated in the range $E = [0-2.5]$ a.u. corresponding to the energy range where the SCS peak manifest itself. The angular distribution is provided for three configurations as depicted in the upper panels: **(A)** – the polarization axis (ϵ) is in the z direction (like the fixed in space internuclear axis) and the propagation (η) is in the x direction **(B)** – the polarization axis (ϵ) is in the x direction and the propagation (η) is in the y direction **(C)** – the polarization axis (ϵ) is in the x direction and the propagation (η) is in the z direction.

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Sensitivity of the photoionization time delay to the atomic arrangement in a molecule

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Photoionization time delays result from the scattering of electrons in a potential [1]. Protocols such as RABBIT allow to measure such photoionization time delays down to the attosecond timescale maintaining the spectral resolution [2]. Its principle is presented in Fig.1(a), an electronic state (orange line) is ionized by XUV photons (purple arrows), the excitation is coupled with infrared photons (red arrows) leading to the same final electron kinetic energy. This results in an interference where the yield oscillates with the time delay between the XUV and IR pulses. The phase of the oscillation carries information on the ionization time delay [2,3]. This method has already successfully been applied to atoms, small molecules, clusters, solids and liquid systems unraveling a large diversity of physical mechanisms.

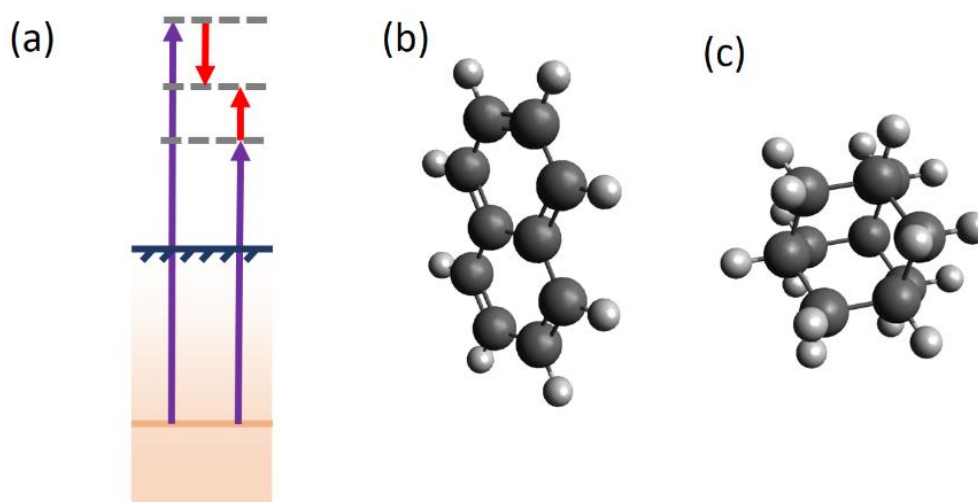


FIGURE 1: (a) RABBIT protocol principle applied to (b) naphthalene and (c) adamantane.

Here, the RABBIT technique is used to compare the photoionization time delay as a function of the atomic arrangement in the molecular potential. Naphthalene ($C_{10}H_8$, Fig. 1b) and adamantane ($C_{10}H_{16}$, Fig. 1c) principally differs by their geometry (planar and diamantoid respectively). The experimental results of RABBIT measurements performed on such molecules show several tens of attosecond differences in photoionization time delay. This difference is attributed to the geometrical atomic arrangement [4].

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New Strategies for Probing Local Orbital and Topological Properties of Solids using ARPES 11

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Angle-resolved photoemission spectroscopy - ARPES - is the most powerful technique to investigate the electronic eigenvalues (band structure) of crystalline solids. To completely characterize the electronic structure of (topological) materials, one needs to go beyond band structure mapping and probe the orbital texture, associated with Berry curvature and topological invariants. I will present new measurement methodologies in ARPES based on i) crystal rotation mimicking time-reversal symmetry operation and ii) continuous modulation of the ionizing radiation polarization axis. I will show how such manipulation of the photoemission transition dipole matrix elements, complemented by minimal theory inputs, allows accessing i) the momentum-dependent orbital texture [1-2] and ii) the complex Bloch wavefunction [3], here exemplified for transition metal dichalcogenides - TMDCs. These results represent an important step towards going beyond band structure (eigenvalues) mapping and learn about electronic wavefunction and orbital texture of solids by exploiting matrix element effects in photoemission spectroscopy.

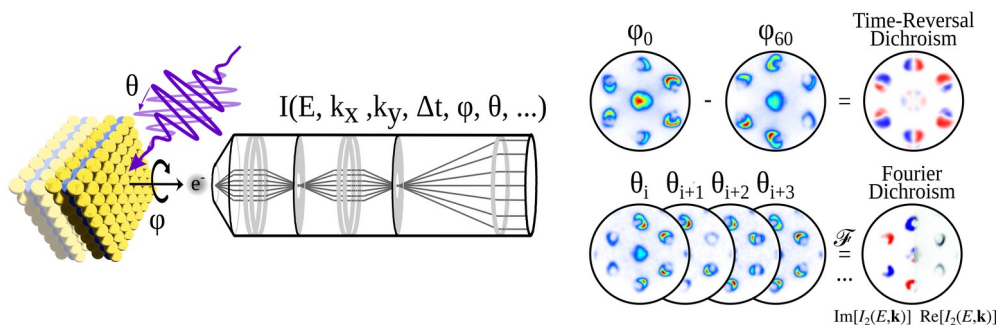


Figure 1: Novel differential observables in multidimensional photoemission spectroscopy based on sample rotation and continuous modulation of the XUV polarization axis.

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Photoionization dynamics of isolated tryptophane nanoparticles

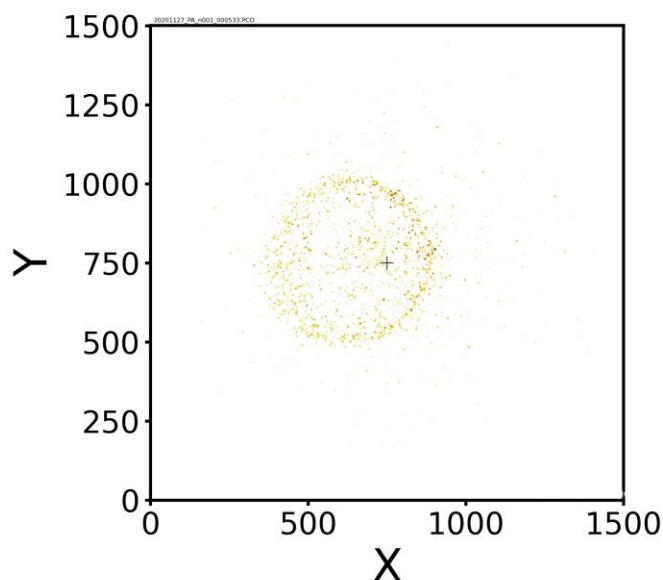
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Isolated nanoparticles are little volumes of condensed matter that are introduced into vacuum. Because of the high density, they are several order of magnitude more sensitive to a laser pulse than isolated molecules. In the framework of gas phase studies, the interaction of an ultrashort femtosecond laser pulse with nanoparticles leads to a high ionization rate. Then, the freed electrons oscillate with the electric field and give rise to collisions with the constituents of the particles. This enhances the number of ions and electrons until formation of a plasma at the nanoscale¹.

Several parameters have been identified to play a role the nanoplasma dynamics, such as size, shape and refractive index of nanoparticles^{2,3}. Our approach consists in using a Velocity Map Imaging spectrometer (VMI) to study the anisotropy of ejection of the electrons and ions from this nanoplasma. Here, we will focus on the ejection of the H⁻ anion in the case of tryptophane nanoparticles irradiated with 266 and 800 nm laser pulses around 10^{14} W.cm⁻².



Velocity map image of H⁻ ion in tryptophane nanoparticle following irradiation at 800 nm.
The laser is propagating from the right to the left.

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Poster: Study of an unusual Stokes shift in polyphenylethyne

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Absorption spectra are analogous for tolane (p2) and bis(phenylethynyl)benzene (m22), with the same vibrational structure [1,2]. However, emission spectra are different, with an unusual Stokes shift for m22 [3]. The similitude between absorption for both molecule was rationalized by Ho et al. [1] involving a pseudofragmentation scheme for the substituted molecule and local excitations.

The emission spectrum of m22 is not yet nor reproduced nor rationalized. The Stokes shift associated to the experimental spectra is about $\Delta\nu = 2000 \text{ cm}^{-1}$. This value is similar to the wave number associated to the normal modes of vibration of elongation of the acetylene bonds in m22 [Fig, left]. This similarity suggests that these modes play a role in the observed Stokes shift.

Another difference between p2 and m22 is the shape of the potential energy surfaces (PESs) of the first two excited states. The PESs of m22 exhibit a Conical Intersection and the lowest excited state exhibits a double-well shaped potential [Fig, right]. In this poster, a linear vibronic coupling Hamiltonian is proposed for the description of the PESs of the excited states of m22. Vibronic spectra are then produced using quantum dynamics calculations.

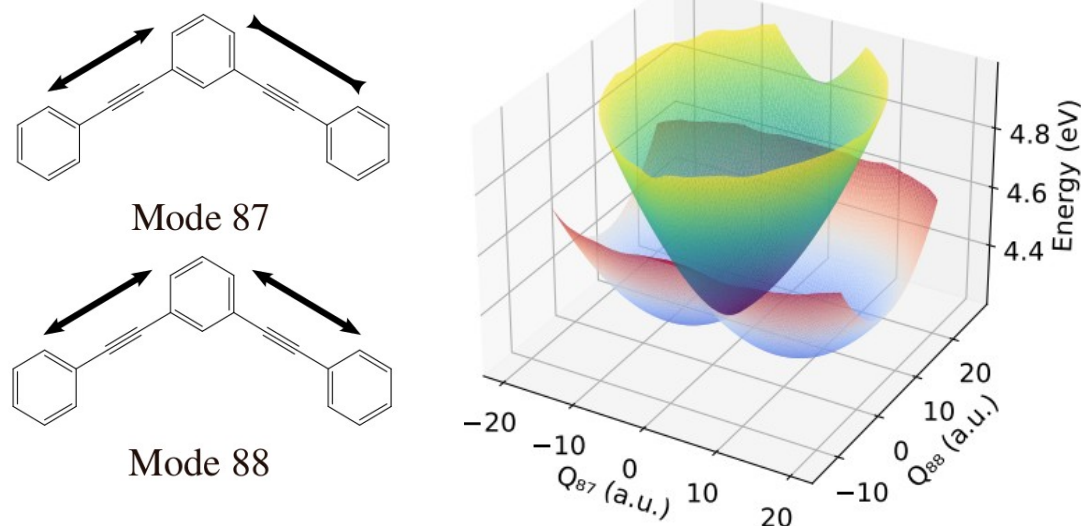


Figure: Left, acetylene elongation normal modes of vibration ($\Delta\nu = 2000 \text{ cm}^{-1}$); Right, conical intersection reproduced with the linear vibronic coupling Hamiltonian and in with two nuclear dimensions associated to the normal modes.

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(Submersion?) of Na⁺ ions in helium nanodroplets

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We image the first steps of the solvation of Na atoms inside helium nanodroplets. Na atoms and Naphthalene (Naph) molecules dope a beam of helium droplets. While the naphthalene goes inside the helium droplet, the Na atom, due to its low interaction with the surrounding He atoms, sticks to its surface.

By ionizing the Na atom to its first cationic state with a 35fs pulse (800nm), the interaction with the helium is greatly amplified with a well depth going from 1.76K for the neutral Na to 400K for Na⁺. This leads to the submersion of the Na atom inside the helium nanodroplets and to the formation of snowballs, a stable structure made of a Na⁺ ion surrounded by non-superfluid helium atoms

By ionizing the Naphthalene molecule with an additional pulse 100fs (400nm) for different delays, we are able to follow the helium attachment on the Na⁺. This is due to the Coulomb repulsion between NaHe_n⁺ and Naph⁺ leading to the ejection of the NaHe_n⁺ from the helium droplet.

We observe the attachment of up to 18 He atoms on the Na⁺. The dynamics can be decomposed in two regimes, a successive attachment of extra helium atom with a linear dependence in time (<10ps) with a slower dynamics related to the formation of larger complexes.

By combining multiple scans for different droplet sizes and classical simulations, we rationalize the dependency of the Coulomb energy in the helium attachment. We put our results in perspective with some quantum mechanical calculations where the Na⁺ ion is treated classically.

We interpret our results as representing the first steps of solvation in the case where the solvent is lighter than the solute. Here, the helium gathers around the Na⁺ faster than the sinking of the Na⁺ inside the helium droplets.

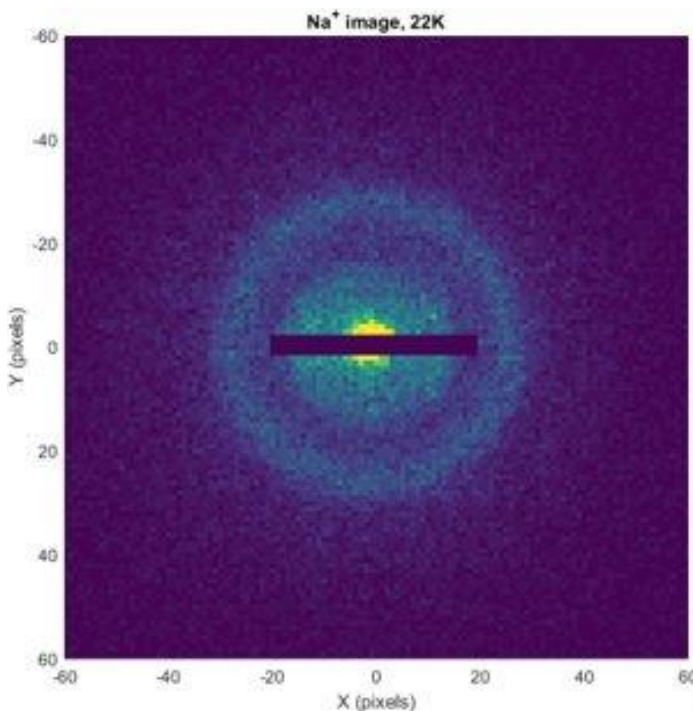


Figure 2 Ion image of Na⁺, the ring is associated to the channel Na⁺+ Naph⁺ for a nozzle temperature of 22K (~3000 He atoms)

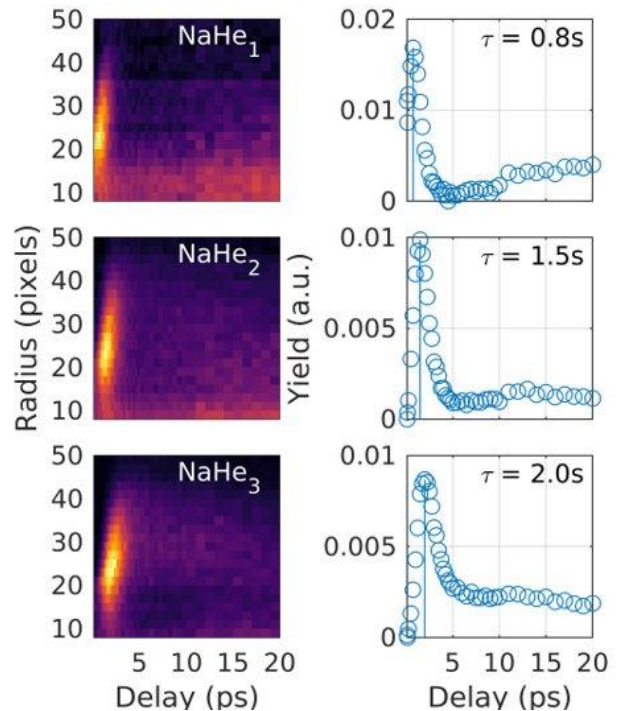


Figure 1 : Left column: Contour plot for NaHe₁⁺, NaHe₂⁺, NaHe₃⁺. Right column: Total yield between 10 and 50 pixels for NaHe₁⁺, NaHe₂⁺, NaHe₃⁺.

Asymmetric electron re-scattering in strong-field ionization of chiral molecules

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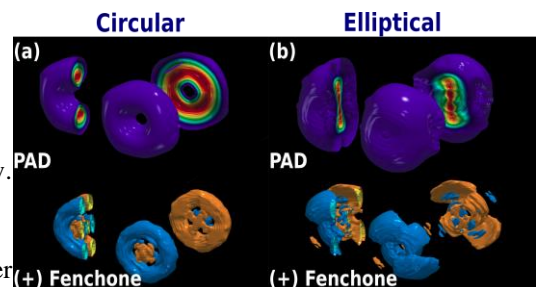
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Strong-field ionization is a versatile tool for investigating the structure and dynamics of molecules. When a molecule is ionized in the strong-field regime, an electron is born into the continuum by tunneling out of the lowered potential barrier formed by the electromagnetic field and the molecular potential. These electrons are then driven by the electromagnetic field. Depending on the time, within the optical cycle, at which the electron is released into the continuum some of these electrons can be driven back towards the ionic core and then re-scatter from the molecular potential to reach the detector (indirect electrons). On contrary some of the tunneled out electrons may propagate directly towards the detector (direct electrons)[1,2]. The Photo-electron Angular Distribution recorded on the detector is a coherent superposition of these two type of trajectories and the information about the molecular potential is encoded through photo-electron holography or laser-induced electron diffraction(LIED). Retrieving the structural information for polyatomic molecules is rather challenging due to the complexity of the molecular potential and random distribution of the molecular orientation. In this work, we propose a novel approach to retrieve these structural and temporal information using a symmetric property that survives random alignment of molecules: Photo-Electron Circular Dichroism (PECD).

When a chiral molecule is ionized with a circularly polarized light, there is an asymmetric emission of electrons relative to the propagation axis of the laser, depending on the handedness of the molecule and the impinged light. This chiroptical process which emerges from the scattering of electrons from the chiral molecular potential is called PECD[3,4]. In strong-field approximation the influence of molecular potential on the outgoing electron wavepacket is expected to vanish. On contrary the indirect electrons re-collide deep into the ionic core and thus are expected to be extremely sensitive to the chiral molecular potential. PECD measurements require circular polarized light, which suppresses drastically the re-collision probability of indirect electrons. As a compromise, we use elliptically polarized light. 2-D PAD is recorded using a velocity map imaging spectrometer. The 3-D PAD distribution is retrieved through tomographic reconstruction. In elliptical polarization we observe re-scattered electrons in PAD (Fig. 1b) with kinetic energies between $2U_p - 10U_p$. These re-scattered electrons show a strong forward-backward asymmetry, which is highly sensitive to the molecular structure. These results opens up the pathway to observe chiral re-scattering and chiral holographic imaging.

Figure 1: Cuts of the 3-D PAD (up) and its forward-backward asymmetric component (down) is obtained by ionizing (+) fenchone molecules with a circularly (kinetic energy up to $2 U_p$) (a) and a fixed elliptically (b) polarized laser field of $2 \times 10^{13} \text{ W/cm}^2$ (kinetic energy up to $10 U_p$).



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Seeded XUV lasers: controlling the level of ASE background with angle-resolved photoelectron spectroscopy

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Abstract : In this poster, we present the results of an experiment performed at LASERIX facility (IJC-Lab, Orsay) aiming at the temporal characterization of plasma-based seeded XUV laser [1], using the technique of photoionization of a rare gas by the XUV pulse in the presence of an IR dressing field (side-bands regime) [2]. A home-made velocity map imaging (VMI) spectrometer [3] is used to obtain the angular resolution of the photoelectron momenta relative to the radiation polarization axis. Photoelectron spectra induced by the seeded XUV laser pulse at 38 eV were successfully obtained in argon. The presence of side-bands could however be detected only when the duration of the IR dressing pulse was extended to 900 fs [4], mainly due to the contribution of the long, unpolarized ASE background to the overall signal.

With the aim of overcoming this issue in a future experiment, we have investigated a method that could be used to finely control the level of ASE in the seeded XUV laser pulse that is sent into the VMI spectrometer. This method exploits the sensitivity of the angular distribution of photoelectrons to the polarization state of the ionizing pulse. We show that the measured values of the “apparent” asymmetry parameter β_{app} obtained for different polarization states of the XUV pulse are consistent with our analytical model, thus confirming the validity of the method.

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Simulation of radiolysis in organic phases with plutonium

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Recycling spent nuclear fuel is a major issue in the power plant industry. Nowadays, the current industrial process used is called PUREX (Plutonium Uranium Refining by Extraction). It is a solvent extraction technology that employs tributyl phosphate to achieve plutonium and uranium extraction. The radioactivity causes radiolysis of both extractant and the solvent molecules, and form unwanted products. So, understanding such radiolysis phenomena is a necessary condition for the development and the improvement of new recycling processes. To date, one can isolate the degradation products with characterization processes while others experimental technics, pulsed radiolysis for instance, give access to their formation mechanisms at the picosecond time scale. However, the sub-femtosecond time scale is totally out of range for the experimental equipment and the phenomena that happen at this time scale are not well understood.

The aim of this work is to model radiolysis in organic phases with plutonium through numerical simulation, with quantum chemistry tools in ultra-short time scale. Tributyl phosphate is used as a reference system for the calculations. We will adopt the Real-Time Time-Dependent Density Functional Theory (RT-TD-DFT) to give access to the deposit energy and the electronic dynamics a few time steps after the interaction with alpha particles. Moreover *ab initio* molecular dynamics simulations will be used to predict the nuclear motion and simulate the absorbed energy diffusion in the system.

The poster will be divided in three parts. First, we will briefly introduce RT-TD-DFT method. Then, we will present the consequences of a large deposit energy in the electronic cloud, due to an ionizing particle, on properties such as the atomic charges and the kinetic energy density. These first results are obtained at the attosecond time scale.

Disentangling the thermal contributions in the ultrafast relaxation of a plasmonic nano-object

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Single-particle optical spectroscopy methods are now enabling quantitative investigations of the optical, electronic, and vibrational responses of metal nano-objects [1]. Here they were utilized to investigate the internal thermalization [2] and cooling to the environment [3] dynamics of individual gold nanodisks supported on a sapphire substrate following their excitation by a femtosecond light pulse. The amplitude and temporal profiles of the measured time-resolved signals present a large dependence on probe wavelength (particularly marked in the spectral range of the nanodisk localized surface plasmon resonance) which could be fully rationalized by numerical models describing both the ultrafast energy exchanges occurring within the nanodisk and with its supporting substrate and the induced transient changes of the nanodisk optical response.

On the first few-picosecond time delays, the experimental time-resolved signals reflect a combination of the electron and lattice temperature evolutions (in the time range where they are both well-defined), with wavelength-dependent weights well reproduced by our models [2]. The possibility of selectively probing the electron or lattice dynamics by using specific wavelengths was demonstrated. The heating (by the excitation laser) and cooling of electrons, or the heating of the crystalline lattice (by electron energy transfer) can thus be selectively measured.

On longer nanosecond timescales, the cooling kinetics of the thermalized nanodisk scales linearly with their thickness, the effect of diameter being comparatively weak, in agreement with numerical simulations based on Fourier law of heat diffusion [3]. This is ascribed to the predominant role of the interfacial thermal resistance in the heat transfer mechanism to the substrate for the explored diameter range. We find that the extracted thermal boundary conductance at the gold–sapphire interface for each investigated nano-object shows small variations from one nanodisk to another.

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In Situ Sub-50-Attosecond Active Stabilization of the Delay between Infrared and Ultraviolet Light Pulses

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Attosecond pump-probe experiments require to control, with sub-femtosecond precision, the delay between two ultrashort light pulses. One of them is usually intense and in the Vis-IR spectral range, while the second is weak and in the XUV spectral range. In order to perform experiments asking for high statistics, such a stability should be maintained over hours, while scanning the delay. This can only be achieved through active stabilization schemes, usually relying on the live measurement of interferences from an additional continuous laser beam co-propagating in the interferometer [1]. Such techniques have drawbacks: they require many additional optics, and most importantly do not provide an *in situ* control of the delay. Indeed, what is actually stabilized is the optical path difference accumulated by the continuous laser, which may differ from the XUV/IR optical path difference. Here we introduce a new technique, named LIZARD (Laser-dressed Ionization for the Adjustment of the pump-probe Delay), which does not require any such additional laser beam and allows an in-situ delay control [2].

When performing scans of the delay, one has to be able to continuously lock the pump-probe phase at arbitrary set-points, with a uniform stability. A solution consists in using two sinusoidal signals oscillating with the delay, which are in phase quadrature [3]. The principle of LIZARD is to extract those signals from two-photon photoelectron spectra obtained by photo-ionizing a gas target in an electronic spectrometer, with the two superimposed beams. In the case of laser-dressed ionization by XUV attosecond pulses trains, the spectra typically show a series of peaks corresponding to the XUV harmonic energies; and sidebands in-between two harmonic peaks, oscillating with the delay (Figure 1). Interestingly, the natural frequency chirp of attosecond pulses trains obtained via high harmonic generation (better known as "atto-chirp") guarantees that, in most practical cases, two sidebands close to phase quadrature can be found, regardless of the gas chosen or the intensity of the driving laser. The signal of such sidebands can therefore be used in the active stabilization loop.

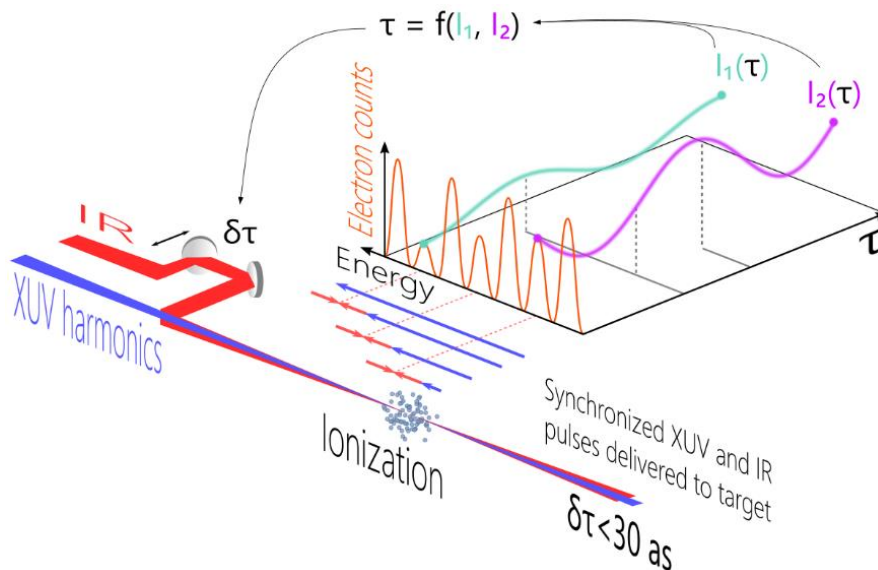


Figure 1: Principle of LIZARD. A feedback loop is performed in order to stabilize the XUV/IR delay τ whose inputs are photoelectron sidebands signals oscillating with respect to the delay, in phase quadrature (green and purple curves). The delay τ is reconstructed unambiguously, and any variation from the set-point value is compensated using two mirrors mounted on a piezo-electric stage.

The LIZARD technique was implemented on the FAB10 beamline at the ATTOLab facility (CEA Saclay), based on a 10 kHz regenerative amplifier delivering 800 nm, 25 fs pulses. The XUV pulses were generated in an interferometer by focusing the laser in a jet of Ar gas. XUV and IR pulses were then focused in a time-of-flight spectrometer. We used the signal of two sidebands, separated in energy by 12 eV, and in phase by 0.46π rad, to perform the active stabilization. Despite an interferometer length of several meters, we achieved a long-term delay stability of 28 as RMS.

Finally, the LIZARD technique allows measurements of RABBIT (Reconstruction of Attosecond Beatings By Interference of Two-photon transitions) traces like the one displayed in Figure 2. The high degree of stability enables long averaging times, making LIZARD a promising technique for the stabilization of pump-probe experiments requiring high statistics, such as coincidence spectroscopy or photo-emission from surfaces. In the case of RABBIT, we show that the measurement of spectral phases is more precise with LIZARD, especially at energies where the signal to noise ratio is very low.

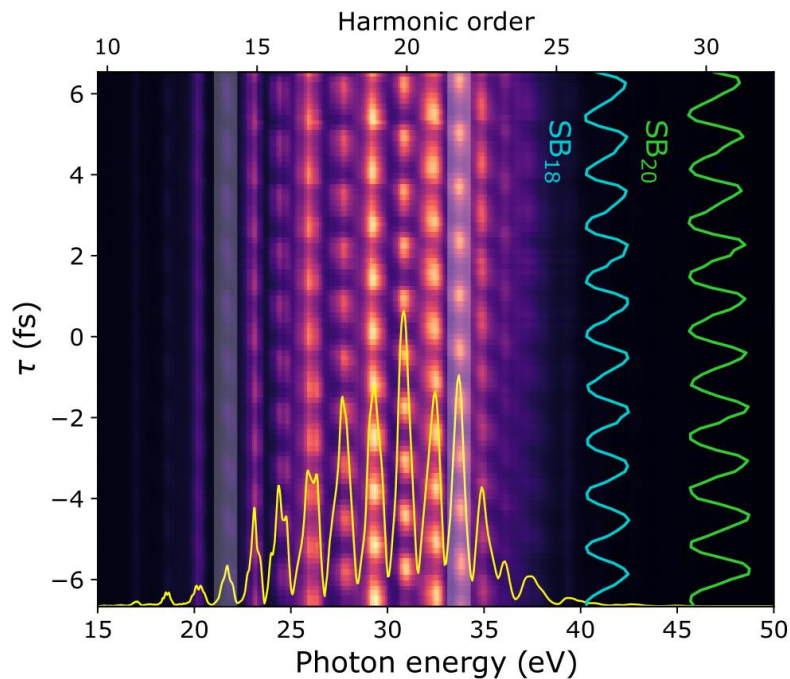


Figure 2: A RABBIT scan in Ar gas, stabilized by the LIZARD technique. The scan lasted approximately two hours. The two energy bands highlighted in white were used as the two phase quadrature input signals for the delay control. In blue and green, lineouts of two other sidebands not used for the LIZARD stabilization.

Our technique is particularly suited to attosecond beamlines already equipped with an electron time-of-flight spectrometer. It is fundamentally different from existing active stabilization schemes, and provides an *in situ* control of the pump-probe phase. LIZARD does not require any additional laser or even additional optical elements but a refocusing optic that is common to both beams. This approach could be applied to the stabilization of other types of two-color interferometers, provided that the required phase-shifted photoionization signals can be obtained.

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A new High-Harmonic Generation apparatus for exploring ultrafast magnetization dynamics at N -absorption edges of rare-earth metals 20

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Abstract: We report on the development of a custom table-top high harmonic setup within the Extreme Ultraviolet (XUV) range spanning energies from 25 to 130 eV.

High orders harmonic generation (HHG) in gases is an extreme nonlinear process occurring between a gas specie and a driving intense laser field that leads to the generation of ultrashort coherent radiation covering the XUV and Soft-X ray regions [1]. Photon energies resulting from HHG may be used as a resonant probe with distinct atomic absorption edges, which provide an element-sensitive magnetic contrast [2], at femtosecond temporal resolution, in binary or ternary ferromagnetic systems considered as important candidates for ultrafast spintronic applications.

In our experiments, 4 mJ, 40 fs pulses from a Ti:sapphire laser amplifier system, operating at 1 kHz repetition rate, are coupled into an Ar- or He-filled hollow-core waveguide (HCW) with different lengths and inner diameters. The output flux of the resulting XUV harmonics is optimized by finely tuning the gas pressure in the HCW in order to phase match the high-harmonics upconversion process.

In a first step, using a microchannel plate and a CCD camera, a XUV frequency comb up to about 50 eV ($\lambda \approx 24.8$ nm), in 10 mbar of Ar, was achieved. We then upgraded the differential pumping stage to handle higher pressure in the gas target and to limit gas load in the surrounding chamber, which enabled to maintain a vacuum of 10^{-5} mbar. Thus, we could perform an almost three-octaves-spanning energies spectrum with a cut-off at 130 eV ($\lambda \approx 9.5$ nm), in 300 mbar of He, which is consistent with [3].

Next step now concerns the extension of the cut-off energy to the Soft-X range, including the $N_{2,3}$ -absorption edges of rare-earth elements (150-300 eV) with enough fluence to study magnetization dynamics. To address this challenge, one solution is to use long-wavelength focused in a few atmosphere of He to drive the HHG [4].

This work has been supported by the EquipEx UNION (ANR-10-EQPX-0052)

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High order harmonics, generated in gases, provide a source of ultrashort XUV pulses that consists, after spectral filtering, in trains of attosecond pulses or in isolated attosecond pulses.

In a first part, we generate high order harmonics with a 30 fs pulse at a repetition rate of 5 kHz and observe harmonics generated in Xenon, Krypton and Argon. After spectral filtering with Al or Sn filters, we focus the XUV pulses in a Velocity Map Imaging (VMI) spectrometer and recombine them with a probe IR pulse. We characterize the attosecond structure with the RABBIT technique [1], neglecting the so called atomic phase. For HHG in Xenon and spectral selection via an Al filter, we observe pulses with a duration of 380 as FWHM that are close to the Fourier limit of 360 as. We also performed complete FROG-CRAB [2] scans that allows us to characterize both the attosecond and femtosecond structure of the XUV pulse.

In a second part, we perform post compression [3] of the 30 fs IR pulses by self-phase modulation in a Neon filled capillary fiber coupled with chirp compensation induced by 10 chirped mirrors (Ultrafast Innovation). We obtain 7 fs pulses that are characterized both by single shot autocorrelation and by the chirp-scan technique [4]. To propagate these short pulses to an HHG chamber, we added extra chirped mirrors (Layertech). We study the impact of these mirrors on the temporal structure of the post compressed pulses and observe a significant third order dispersion that cannot be compensated. This leads to weak pre or post pulses and to a main pulse that broadens slightly (from 7 to 8 fs) when the number of additional chirped mirrors is increased. After propagation to the HHG chamber the pulse energy is 200 μ J that was sufficient to observe HHG in Krypton gas (Figure (1)).

To perform HHG with the few cycle pulses, we focused the post compressed beam with a 60 cm focal length mirror onto a 10 mm gas cell that was drilled directly by the laser. We observed broad high order harmonics, a shift in the central wavelength and spectra that extends to shorter wavelength. A clear optimum in the XUV signal for a gas pressure of 12 to 15 mbar in Krypton. We did not observe any significant change of the spectrum with the pressure except for the efficiency. A shorter cell with 6 mm length was also tested and led to a slight increase of the HHG efficiency. The measured spectral width is compatible with a short XUV pulse and the estimated minimum duration of the XUV pulse envelop is 2 fs for the 15th harmonic.

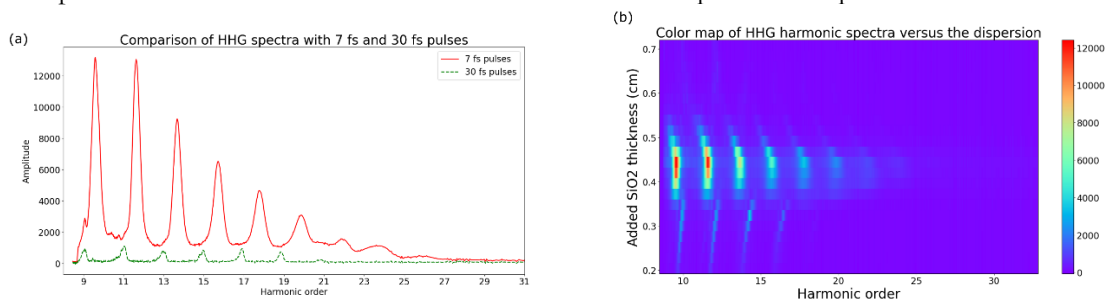


Figure 1: (a) Spectra of high order harmonics generated with a 30 fs pulse (dotted line) or with a post compressed 7 fs pulses (full line) in a 10 mm long cell filled with Krypton. (b) Evolution of the harmonic spectra as a function of the pulse compression for HHG in Krypton with a 10 mm cell length.

We acknowledge stimulating discussions with A. Boyer, V. Loriot, S. Nandi, E. Prost and F. Lépine.

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Time-resolved fluorescence investigation of the giant antenna effect in dye-doped organic nanoparticles.

22

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Pigment-protein complexes known as light-harvesting complexes (LHC) are found in natural photosynthetic organisms. To achieve optimum light energy absorption and conversion, LHC's are organized in a large network of interacting chromophores held by proteins, providing them with a well-defined positioning [1]. Consequently, the optical properties of the LHC depend on the protein scaffold, and the relative orientation of the pigments [2, 3]. To mimic the function of LHC, synthetic light-harvesting "antennas" are designed for biophotonic applications [4,5]. In this work, we aim at investigating the photophysics of synthetic dye-doped, polymeric, organic nanoparticles (ONP's). Upon light excitation of the encapsulated donor dyes in the ONP, the produced exciton migrates to neighbouring chromophores via Exciton Energy Transfer (EET). The collected energy is channelled from the donors to a few or even a single photoreactive acceptor molecule that would quench the entire ONP demonstrating a giant antenna effect [6]. This effect is expected to result from very efficient exciton diffusion within the nanoparticles. Therefore, our work is dedicated to the experimental investigation of the ONP's photophysical properties by time-resolved fluorescence spectroscopy to characterize the energy transfer mechanism among the donors (Exciton diffusion). Hence, on a picosecond time scale, the exciton population decay is found to depend on the excitation power, which is attributed to singlet-singlet exciton annihilation (SSA). To model this process, the exciton diffusion constant within the ONPs is introduced [7, 8]. In addition to the time dependence, our fitting model accounts for the spatial dependence of the exciton density across the transverse intensity profile of the experimental excitation beam. Thus, we retrieve the annihilation rate value in function of the ONP dye loading. Furthermore, we use polarization-resolved excitation and detection in a fluorescence up-conversion set-up to monitor femtosecond-resolved fluorescence anisotropy decay. The latter reveals the time scale for exciton hopping from one site to the nearest neighbours.

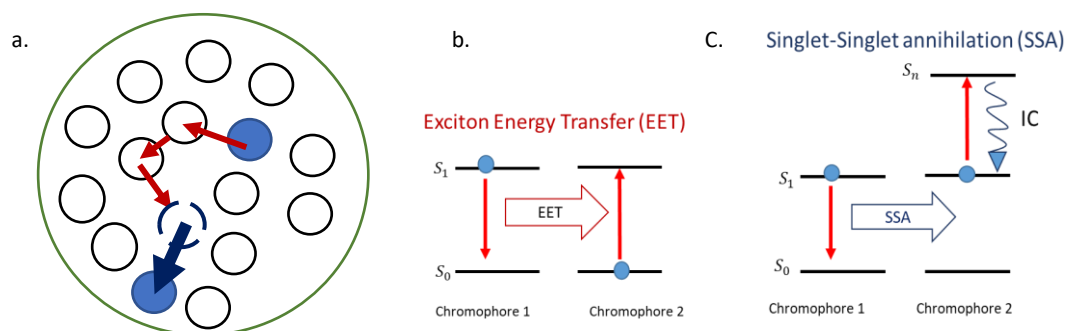


Figure 1: Schematic representation of the dye-loaded ONP: The energy of the produced exciton (filled blue circle) is transferred to the nearest neighbors via EET (red arrows) and annihilates (blue arrow) with a second exciton (a). Energy diagrams for Exciton Energy Transfer (b) and Singlet-Singlet annihilation (c).

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Formation and decay of shallow-trapped electrons in TiO₂ composites for the photoconversion of methane probed by femtosecond Mid-IR transient absorption spectroscopy

23

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Methane is an abundant compound that is released in the atmosphere both directly by the anthropic activities and through the permafrost thawing -which itself is a consequence of global warming- where it is captured as methane hydrate. Being 30 times more active as a green-house-gas than CO₂, the release of methane with the increase of temperature constitutes a climate bomb. Fortunately, methane can also be seen as a huge reserve of Carbon and Hydrogen atoms, which may be put to use to produce synthetic fuels and chemical feedstock for the industry in replacement of fossil oils. Yet, the direct conversion of CH₄ is suffering from high energetic costs and low selectivity. Consequently, there is an urgent need for efficient photoconversion routes of methane under mild conditions.

In this context, we have been investigating the mechanisms of the photoconversion of methane in a new promising family of TiO₂ photocatalysts [1, 2] composites of formula Me:HPW:TiO₂ (Me = Ag, Zn; HPW is a heteropolyanion). The photoconversion is initiated by the ultrafast formation of electrons and holes following the band-gap excitation of the TiO₂. Here, we performed Mid-IR pump-probe measurements to characterize the formation and the lifetime of the shallow trapped electrons for different photocatalysts compositions. The experiments have been performed in a homemade environmental optical cell to control the surface chemistry of the photocatalyst. The variation of the sub-ns electron lifetime with the composition of the catalyst is clearly observed. The investigation of the ultrafast processes is a promising route to improve the design of the photocatalysts for the photoconversion of methane.

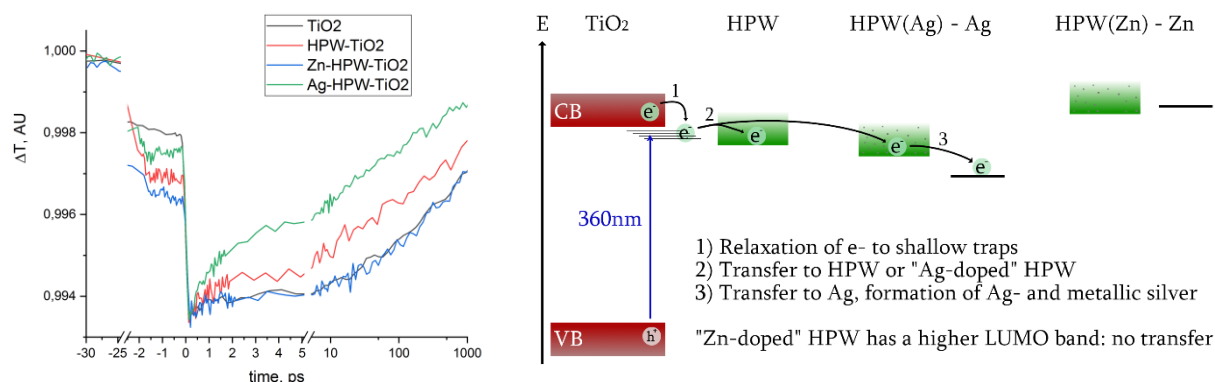


Figure: (left) kinetic traces recorded following the photoexcitation of the catalyst at 360nm (right) dynamic associated to the shallow-trapped electrons.

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Les sources XUV (10-100 nm) basées sur la génération d'harmonique d'ordre élevé (HHG) dans les gaz sont émises sous forme d'impulsions attosecondes et présentent des propriétés de cohérences spatiale et temporelle propices à différentes applications. Afin de réaliser des expériences pompe-sonde XUV-XUV, il est nécessaire de contrôler la focalisation des faisceaux XUV ainsi que la durée des impulsions afin d'augmenter l'intensité XUV jusqu'à $\sim 10^{14}$ W/cm² ou plus [1, 2]. Il est maintenant admis que le profil spatial des harmoniques dépend de l'ordre [1, 3, 4] ce qui rend le profil temporel des impulsions attosecondes inhomogène dans le profil d'intensité [3]. Nous avons récemment démontré qu'il est possible de contrôler expérimentalement les propriétés spatiales du rayonnement XUV [5, 6] et nous montrons que ce contrôle permet également de maîtriser le profil temporel attoseconde et notamment son homogénéité [6].

Dans cette expérience, les harmoniques sont générées dans un jet de néon avec une intensité IR (800 nm) de 4×10^{14} W/cm², pour un waist de 83 μ m [5]. Un trou de diamètre 140 μ m situé à 370 mm en aval du foyer IR permet de caractériser les propriétés spatiales de chaque faisceau harmonique à l'aide de la technique SWORD [7]. En considérant des faisceaux XUV Gaussiens, il a été possible de déduire de ces mesures la position et la taille des foyers XUV (Fig. 1a et 1b). Lorsque le jet est placé avant le foyer IR, les faisceaux harmoniques sont focalisés après le milieu générateur démontrant ainsi la focalisation XUV sans optique [6]. En revanche, quand le jet de gaz est positionné à proximité et en aval du foyer IR, les foyers XUV sont plus proches les uns des autres [4, 6].

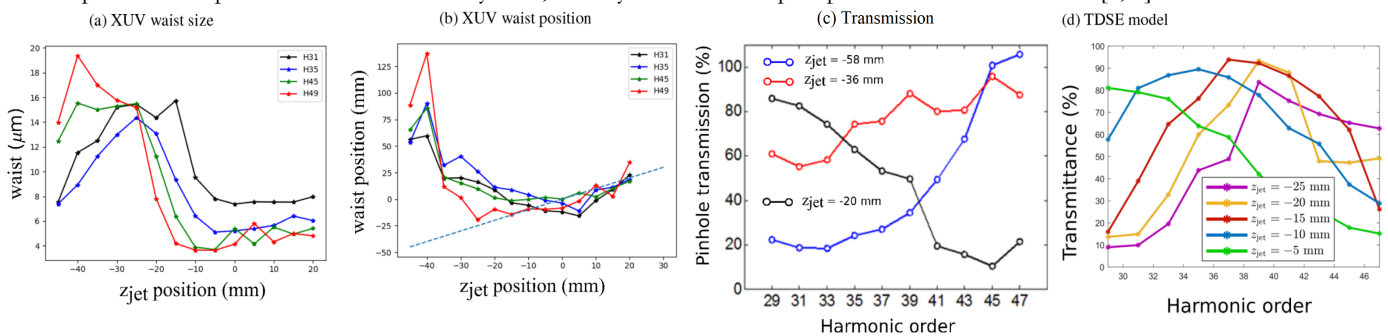


Fig. 1 : Taille (a) et position (b) des foyers XUV en fonction de la position du jet par rapport au foyer du laser fondamental IR. (c) Transmission par un trou de 140 μ m situé 370 mm après le faisceau IR mesurée pour les harmoniques H29 à H47 pour plusieurs positions du jet : $z_{jet} = -58$ mm (bleu), -36 mm (rouge) et -20 mm (noir). (d) : Transmission simulée en fonction de l'ordre harmonique pour cinq positions du jet de gaz (modèle TDSE).

De plus, lorsque le trou est positionné sur l'axe des faisceaux XUV, nous avons démontré le filtrage spatial des harmoniques avec une haute transmission et un bon contraste (Fig. 1c) qui permet de réaliser une sélection spectrale XUV ajustable en fonction de la position du jet. Ces résultats sont corroborés par des simulations basées sur la résolution des équations de Schrödinger dépendantes du temps incluant la propagation du rayonnement XUV [6,8] (Fig. 1d) qui démontrent que ce filtrage spectral s'accompagne d'un contrôle temporel des impulsions attosecondes en champs lointain en assurant une évolution plus homogène ; propriété fondamentale pour une utilisation ultérieure, notamment la photoionisation pompe-sonde XUV-XUV [6].

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With the development of sources and detection, terahertz (THz) pulses have led to numerous applications and became a tool of great importance in ultrafast science [1]. We present a THz platform, recently developed in Lyon and show its first results. This platform relies on the one hand on THz generation by air-plasma induced by a two-color short laser pulse [2] and, on the other hand, on Air Biased Coherent Detection [3]. This technique measures the THz electric field, in time domain, by a cross correlation. The spectrum is then retrieved by taking the Fourier transform of the measured electric field. Generation and detection in air allows for the generation of high electric field strengths (in the range of 10 MV/cm) and smooth broadband spectra (see Figure). The optical setup, its specific homemade electronic and its interface are presented.

Number of ro-vibrational absorption bands of molecules and phonons lies in the THz range. One of the application of the platform is THz time-domain spectroscopy (THz-TDS) [4] where the temporal profile of a THz pulse is affected by its propagation through a medium. The absorption features of the medium can be retrieved by comparing the Fourier transform of the temporal profile with and without the sample. In this poster some results are presented and compared with HiTran database and FTIR spectroscopy.

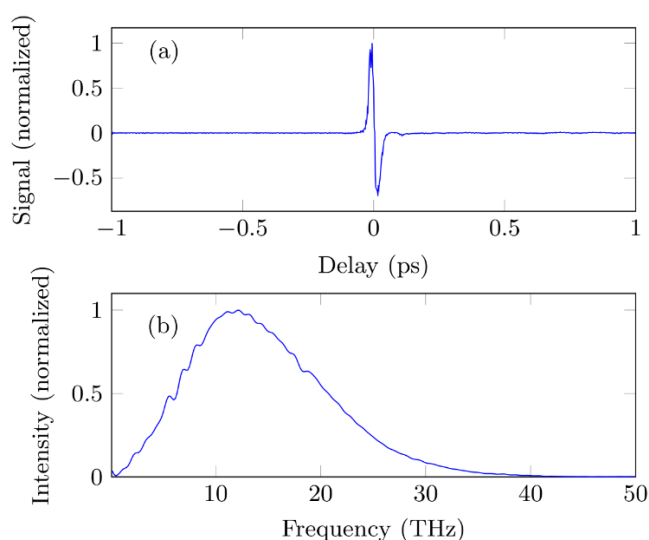


Figure: Measured (a) temporal profile of the THz electric field and (b) reconstructed spectral intensity profile.

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Simulation of the physical stage of irradiating biomolecules, from small cluster to DNA/protein complex by RT-TDDFT

26

Karwan Ali OMAR, Aurélien de la Lande, Carine CLAVAGUERA and Fabien CAILLIEZ

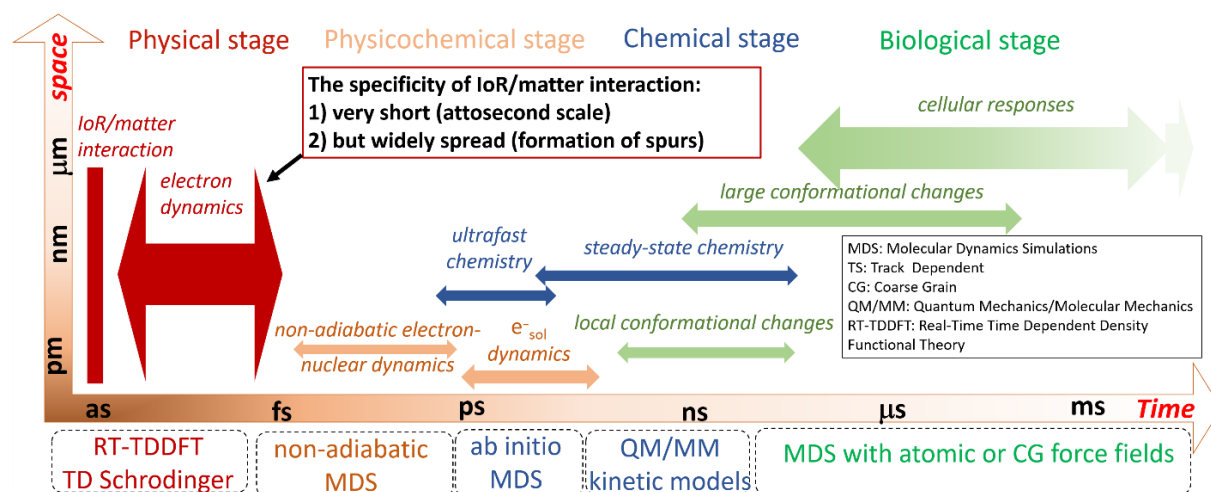
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Ionizing radiation interacts strongly with biological molecules and damages them consequence of multiscale physical and chemical processes as a result destroy the normal activity of cell, arise tumors and cancers, in the other hand benefit to kill cancerous cell in hadron-therapies. The mechanism of the early stage of the irradiation process is crucial for the response of the chemical and biological stages. The physical stage starts with important energy deposition within a few atto-seconds, producing highly excited molecules. In this poster we investigate collective effects arising from the environment on energy deposition and ultrafast charge migration in irradiated DNA/protein complex by means of numerical electron dynamic model *real-time time dependent auxiliary density functional theory* (RT-TD-ADFT) [1].

It is generally considered that energy deposition is a local process which is little influences by the molecular environment. In order to estimate the effect of the surrounding molecular fragments on the energy deposition (collective deposited energy), we irradiated a supramolecular system and isolated molecular fragments of it by swift proton with difference kinetic energy. We reveal that the quantity of collective deposited energy is significant in our system, it varies from 5% to 12% with respect to the kinetic energy of the proton.

The investigation of the damage mechanism of nucleoproteins induced by IoR has a pivotal position. Indeed, the nucleoprotein inserts in many biological processes in vivo. To investigate the ultrafast charge migration, we irradiated protein/DNA complex by 250keV α -particle. The preliminary results show that first time the hole localized at the site of collision and after less than 1 fs a complex ultra-fast charge migration was appeared [2], in addition the majority of emit electrons accumulate on DNA part and solvated water molecules. In other hand, the complexity of charge analysis and correlation between residues are rose by the augmentation of system size. We found that *Correlation matrix analysis* could simplify the data representation and provide new information about relationship of charge migration between residues.

FIGURE



Interaction of IoR with biological matter induces multiscale responses in time and space. The time-scales are usually referred. In the bottom we indicate, not exhaustively, some of methodologies found in the literature to address the different scales [3].

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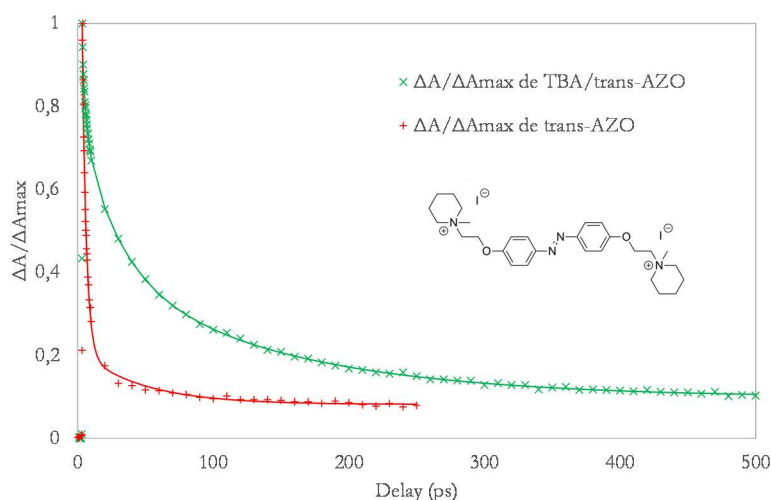
Real time observation of chirality changes in secondary G-quadruplex DNA Structures

27

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DNA can adopt various secondary structures such as 'four-stranded' G-quadruplexes (G4), that are formed by guanine-rich sequences through self-recognition of guanines into stacked tetrads. These structures present a large variety of topologies and their folding and unfolding have been proven to be linked with numerous key biological processes.¹ Therefore we aim to analyze these dynamics over a time window ranging from a few picoseconds to seconds. These dynamics can be induced by the isomerisation of designed photoswitches such as AZO,^{2,3} a derivative of azobenzene. The isomerisation induced by UV or visible excitation of AZO causes the unfolding or the folding of the G4. Therefore, we currently work on the characterization of this photoswitch and of DNA-AZO systems with various techniques like UV-visible absorption, NMR, circular dichroism or femtosecond transient absorption. We now aim at studying the dynamics of folding and unfolding of G4 with femtosecond circular dichroism spectroscopy, which is particularly adapted to the study of biological structures.⁴



Transient absorption of AZO measured in aqueous solution (10mM Tris) at 520 nm after irradiation at 400 nm in presence or absence of the G-quadruplex sequence TBA (5'-(TGG TGG TGT GGT GG)-3')

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APOLLON FACILITY SHORT FOCAL LENGTH AREA COMMISSIONING AT 1 PW LEVEL

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We present the results of the commissioning of the "short focal length" area, which was performed with the first available laser beam of the Apollon laser facility (Saclay, France), i.e. using the F2 beam at a nominal power of one petawatt [1]. This beam delivered laser pulses of 10 J average on-target energy with pulse durations of 25 fs.

A range of diagnostics was in place in order to qualify the performance of the facility at this level of laser power.

An imager, working in X-ray range, of the zone heated by the laser on the target demonstrated a good focusing of the laser at full power, with a heated spot of 4.5 μm in diameter, corresponding to the laser spot aligned at low flux.

Solid targets as thin as 2 μm in thickness were irradiated by intense laser pulse without damaging them by the pedestal preceding the main pulse, which corresponds to a good temporal contrast characteristics.

Emissions of electrons, ions and high energy electromagnetic radiation were recorded, showing good laser-target coupling and an overall performance that is very consistent with what have been reported by similar international facilities.

This commissioning will be followed in 2022 by a further stage at the multi-petawatt level.

[1] Burdonov et al., Matter and Radiation at Extremes 6, 064402 (2021)

Charge-carrier dynamics in CsPbBr_{3-y}X_y/TiO₂ (X = Cl, I) heterojunction at variable time scale

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Over the past decade, metal halide perovskites (MHP, CsPbX₃: X = Cl, Br, I) have been widely investigated as promising materials for optoelectronics, achieving a record-breaking efficiency in solar cells.^[1] From a fundamental point of view, MHP could be excellent candidates for photocatalysis due to their high photogenerated charge-carrier production and mobility as well as their narrow and tunable bandgap energy.^[2] MHP with tuneable band-gap energy could be obtained through fast substitution of bromide by iodine or chloride (CsPbBr_{3-y}X_y: X = Cl, Br, I).^[3]

In this work, we investigated charge-carrier dynamics of CsPbBr_{3-y}X_y along with interfacial electron transfer from CsPbBr_{3-y}X_y to TiO₂ by means of time resolved microwave conductivity (TRMC), time resolved photoluminescence (TRPL) and transient absorption spectroscopy (TAS). In order to find the optimal bandgap configuration for the most efficient charge injection, anionic ratio was varied. Charge-carrier injection from one material to another resulted in increased charge-carrier lifetime, which could positively affect the photocatalytic activity.

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