Réunion plénière du GDR UP - 2022

12-13 décembre 2022

Workshop « Dialogue Théorie-Expérience »

14 décembre 2022

Cité internationale universitaire de Paris

Recueil des contributions

Club des partenaires :

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 sixième réunion plénière du GDR qui se tient du 12 au 13 décembre 2022 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. Elle est suivie par un workshop « Dialogue théorie expérience » dont le but est de stimuler les collaborations à l'initiative des développements théoriques récents.

Nous vous remercions d'y participer et pour votre contribution à la grande réussite scientifique du GDR U.P.

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Avec le soutien de :

Réunion Plénière du GDR U.P - 2022

Lundi 12 décembre 2022

09h00-10h00	*ACCUEIL*	
10h00-10h30	BUREAU DU GDR UP	Introduction Actions 2022 du GDR, Actions prévues, opportunités
10h30-11h00	Catalin MIRON (LIDYL)	Présentation PEPR LUMA
11h00-11h30	*PAUSE CAFÉ / EXPOSANTS*	
SESSION 1 Condensed phase 1		
11h30-11h50	Adeline BONVALET (LOB)	Spectroscopie infrarouge pompe-sonde de la picoseconde à la microseconde dans une photoenzyme
11h50-12h10	Amira GHARBI (IPCMS)	Ultra-fast Energy transfer dynamics in dye-doped organic nanoparticles
12h10-12h30	Oleksandr DEREVIANCHENKO (EKSPLA/OPTON)	Developments in high intensity lasers
12h30-14h00	*REPAS / EXPOSANTS*	
SESSION 2 Ultrafast optics		
14h00-14h20	Aurélie JULIEN (INPHYNI)	Phase modulator based on thermo-optically addressed liquid crystal for multi-octave light source
14h20-14h40	Edouard HERTZ (ICB)	Storing an ultrashort optical vortex in molecules
14h40-15h00	Brice ARNAUD (IMMM)	A first-principles approach to non-linear phononics in Bi2Te3
15h00-15h20	Martin LUTTMAN (LIDYL)	Conservation of a half-integer angular momentum of light in nonlinear optics
15h20-15h50	*PAUSE CAFÉ / EXPOSANTS*	

SESSION 3 Attosecond and XUV science 1		
15h50-16h10	Fabien GATTI (ISMO)	Simulations of attosecond pump-probe experiments on CO_2
16h10-16h30	Hugo MARROUX (LIDYL)	Attosecond spectroscopy of molecular core levels
16h30-16h50	Victor DESPRE (ILM)	Ab-initio simulations of ultrafast electron-nuclear dynamics: Paving the way to attochemistry
16h50-17h10	Michele NATILE (Amplitudes)	High repetition rate few-cycle Mid-IR laser source
	Photo conférence	
17h30-19h00	*Session Poster / Cocktail*	
19h00-20h30	*Cocktail*	

Réunion Plénière du GDR U.P - 2022

Mardi 13 décembre 2022

SESSION 4 INTENSE LASER INTERACTION		
9h00-9h20	Weipeng YAO (LULI)	Optimizing laser coupling, matter heating, and particle acceleration from solids, using multiplexed ultraintense lasers
9h20-9h40	Amélie FERRE (LP3)	Kα X-ray source driven by a multi-terrawatt femtosecond laser at 100 Hz for spectroscopy and imaging application
9h40-10h00	Marie LABAT (SOLEIL)	Recent news about Laser-Plasma Accelerator based Free Electron Lasers
10h00-10h20	François SYLLA (Sourcelab)	KAIO-Beamline, the modular high-repetition rate laser-plasma electron accelerator for broad range of applications
10H20-10h50	*PAUSE CAFÉ / EXPOSANTS*	
SESSION 5 CONDENSED PHASE 2		
10h50-11h10	Rodolphe VUILLEUMIER (ENS, Dept Chimie)	Dynamiques aux interfaces de graphene et hBN chargées
11h10-11h30	Emmanuelle JAL (LCPMR)	Unravelling the Transient Depth Magnetic Profile During Ultrafast Demagnetization of an Iron Thin Film
11h30-11h50	Yann GALLAIS (MPQ)	Disentangling lattice and electronic instabilities in the excitonic insulator candidate Ta_2NiSe_5 by non-equilibrium spectroscopy
11h50-12h10	Anna LEVY (INSP)	Ultrafast lattice dynamics of laser-heated copper using time-resolved photoelectron spectroscopy
12h10-14h00	*REPAS / EXPOSANTS*	

SESSION 6 ATTOSECOND AND GAS PHASE ULTRAFAST SCIENCE		
14h00-14h20	Vincent LORIOT (ILM)	Attosecond signatures of spatially extended hole in a molecule
14h20-14h40	Jonathan DUBOIS (MPI)	Recollisions in circularly polarized pulses : from tunnel-ionization to the return of the electron
14h40-15h00	Isabelle MERRITT (CEISAM)	Photoisomerization of Cis-Azobenzene: Understanding the Quantum Yield Wavelength Dependency
15h00-15h20	Eric CONSTANT (ILM)	Chromatic aberrations control in high order harmonic XUV beams by spatial shaping of the fundamental

15h20-15h50	*Pause café / EXPOSANTS*	
SESSION 7 NANO SCIENCE		
15h50-16h10	Angella VELLA (GMP)	LaB_6 nano-tip as an ultrafast electron source
16h10-16h30	Giovanni MANFREDI (IPCMS)	Driving orbital magnetism in metallic nanoparticles through plasmonic effects
16h30-16h50	Marc ALIAS-RODRIGEZ (ICR)	Revealing the ultrafast spin-crossover mechanism in $Fe^{II}(bpy)_3$ through quantum dynamics
16h50-17h10	Bureau du GDR UP	CONCLUSION

1er Workshop « Dialogue théorie-expérience en sciences ultrarapides »

Mercredi 14 décembre 2022

09h00-09h05	*Bureau du GDR U.P*	*Introduction*
SESSION 1 Molécules isolées		
09h05-09h30	Richard Taïeb (LCPMR)	TBA
09h30-09h55	Bernard Pons (CELIA)	Structures et dynamiques de molécules chirales sondées par de brèves impulsions laser
09h55-10h20	Martial Boggio-Pasqua (LCPQ)	Dihydropyrene photoisomerization: Aspirations to go beyond the static mechanistic picture
10h20-10h50	*Pause Café / discussions*	
SESSION 2 molécules environnées, complexes moléculaires et nanoparticules		
10h50-11h15	Etienne Mangaud (MSME)	Simulation by the hierachical equations of motion with tensor trains
11h15-11h40	Valérie Vallet (PhLAM)	hotodynamics of lanthanide and actinide complexes
11h40-12h05	Aurélien Crut (ILM)	Investigation of the vibrational and cooling dynamics of metal nanoparticles by combined experimental and modeling approaches
12h05 – 13h30	*REPAS*	

SESSION 3 matériaux		
13h30-13h55	Philippe Scheid (ILJ)	Discussion on the origin of the helicity-dependent magnetization dynamics
13h55-14h20	Guillaume Duchateau (CESTA)	Laser energy deposition into dielectric materials
14h20-14h45	Anne Tanguy (LaMCoS)	Nanostructuration de la silice par laser femto-seconde
14h45-15h15	*Pause Café / discussions*	
SESSION 4 Systèmes d'intérêt biologique		
15h15-15h40	Aurélien de la Lande (ICP)	First principles simulations of biological matter radiolysis, state-of-the-art and roadmap
15h40-16h05	Valérie Brenner (LIDYL)	Non-radiative transitions in bio-relevant systems: non-adiabatic dynamics simulations as a bridge between theory and experiment?
16h05-16h30	*Conclusions et perspectives*	

SESSION 1

Condensed phase 1

Spectroscopie infrarouge pompe-sonde de la picoseconde à la microseconde dans une photoenzyme

Adeline Bonvalet[1], Damien Sorigué[2], Laura Antonucci[1], Xavier Solinas[1], Alexey Alexandrov[1], Frédéric Beisson[2], Marten H. Vos[1], Manuel Joffre[1],

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2. Aix-Marseille University, CEA, CNRS, Institute of Biosciences and Biotechnologies, BIAM Caradache, 13108 Saint-Paul-lez-Durance, France

Nous avons développé une méthode de spectroscopie pompe sonde permettant la mesure de spectres différentiels sur des échelles de temps allant de la sub-picoseconde à la milliseconde, méthode qui s'implémente facilement sur deux lasers femtosecondes amplifiés pré-existants sans besoin d'asservir les oscillateurs [1-3]. Cette méthode dénommée AD-ASOPS pour Arbitrary Detuning Asynchronous Optical Sampling, a été appliquée à une photoenzyme récemment découverte [4], Fatty Acid Photodecarboxylase (FAP). La mesure des spectres infrarouges pompe-sonde a mis en évidence la dynamique de formation du CO_2 au cours d'un processus complexe qui transforme un acide gras en hydrocarbure, et permis d'élucider une partie du mécanisme à l'œuvre dans la biomolécule [5].

[1] L. Antonucci, A. Bonvalet, X. Solinas, L. Daniault, M. Joffre, Opt. Express 23, 27931 (2015), https://doi.org/10.1364/OE.23.027931
[2] X. Solinas, L. Antonucci, A. Bonvalet, M. Joffre, Opt. Express 25, 17811 (2017), https://doi.org/10.1364/OE.25.017811
[3] L. Antonucci, X. Solinas, A. Bonvalet, M. Joffre, Opt. Express 28, 18251 (2020), https://doi.org/10.1364/OE.393887
[4] D. Sorigué et al., Science 357, 903 (2017), https://doi.org/10.1126/science.aan6349
[5] D. Sorigué et al., Science 372, 6538 (2021), https://doi.org/10.1126/science.abd5687

Ultra-fast Energy transfer dynamics in dye-doped organic nanoparticles

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Electronic excitation energy transport mechanisms in molecular systems attract considerable attention due to their numerous applications in light-harvesting and optoelectronic devices [1,2]. The involved processes rely centrally on exciton interactions and dynamics [3,4]. In this work, we utilize time-resolved fluorescence spectroscopy to investigate the photophysical properties of dye- doped, polymeric, organic nanoparticles (ONP), which imitate the function of natural light- harvesting complexes found in photosynthetic organisms [5]. The exciton population decay kinetics are found to depend on the excitation fluence, unraveling singlet-singlet exciton annihilation (SSA). The latter is a key parameter in revealing the time scale for exciton diffusion within the ONP [6,7]. Moreover, we demonstrate the effect of the inhomogeneous intensity profile of the excitation beam on the apparent annihilation rate. Furthermore, exciton energy transfer (EET) ensures efficient exciton migration within chromophores. We use fluorescence up- conversion spectroscopy with polarization-resolved excitation and detection to monitor time- resolved fluorescence anisotropy decay. This allows us to characterize the femtosecond time scale for exciton hopping to nearest neighbors.

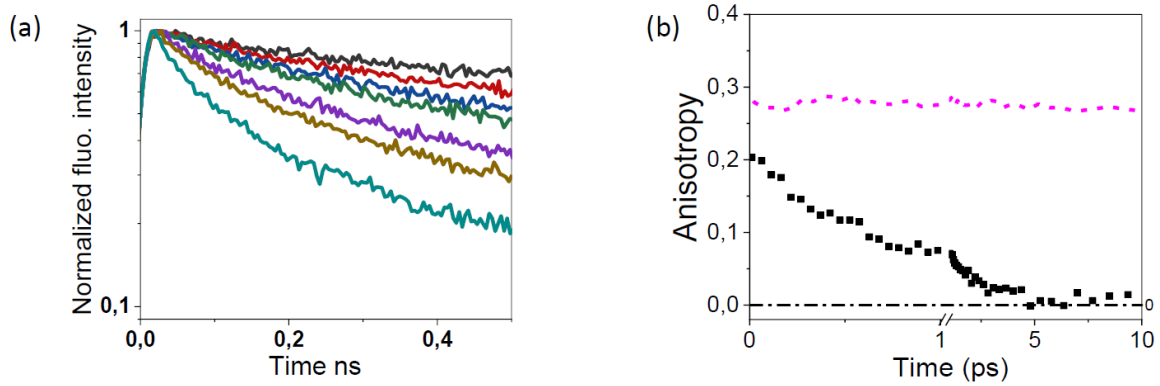

Figure: Measurement of exciton interaction dynamics in 30w% dye-loaded ONPs. (a) Normalized exciton population decay kinetics upon the increase of the excitation fluence from 70nJ/cm² (black curve) to 300µJ/cm² (light blue curve). Acceleration of the decay kinetics highlights the SSA process. (b) Fluorescence anisotropy decay is measured using femtosecond up-conversion fluorescence spectroscopy (black dots) and compared to monomer rhodamin (pink). The reduction of the initial anisotropy is the sign of a non-resolved (i.e. faster than 200 fs) EET process in the ONPs.

[1] Patra, Abhijit, Ch G. Chandaluri, and T. P. Radhakrishnan. "Optical materials based on molecular nanoparticles." *Nanoscale* 4.2 (2012): 343-359.

[2] Mikhnenko, Oleksandr V., Paul WM Blom, and Thuc-Quyen Nguyen. "Exciton diffusion in organic semiconductors." *Energy & Environmental Science* 8.7 (2015): 1867-1888.

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[4] Dostál, Jakub, et al. "Direct observation of exciton–exciton interactions." *Nature communications* 9.1 (2018): 1- 8.

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[7] Fennel, Franziska, and Stefan Lochbrunner. "Exciton-exciton annihilation in a disordered molecular system by direct and multistep Förster transfer." *Physical Review B* 92.14 (2015): 140301.

Abstract

Oleksandr DEREVIANCHENKO (EKSPLA/OPTON)

SESSION 2

Ultrafast optics

Phase modulator based on thermo-optically addressed liquid crystal for multi-octave light source

Aurélie JULIEN

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Thick film nematic liquid crystals provide elegant solutions for light manipulation. In this talk, I will present the design of an ultra-broadband reflective phase modulator. The device, coined as thermo-optical SLM (TOA-SLM), circumvents most of the technical limitations of traditional SLMs by eliminating the need for an electrode: electrical addressing is replaced by optically induced local temperature control of the liquid crystal layer. This device allows arbitrary and continuous phase modulation over a multi-octave spectral range. Beyond this proof-of-principle, the suitability of the device for spectro-temporal manipulation of ultra-short pulses requires to determine linear dispersion, laser damage threshold and ultra-fast nonlinear properties. These aspects of ultrafast optics / liquid crystal interaction will be addressed in the presentation.

Storing an ultrashort optical vortex in molecules

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Beams of transverse phase distributions $e^{il\varphi}$ (with φ the azimuthal coordinate in the beam section) are known to carry an orbital angular momentum (OAM) of l ħ per photon. Such phase-structured light beams have become these last years a subject of widespread interest with unprecedented applications for optical communication, super-resolution, optical tweezers or quantum processing [1]. Here, we experimentally demonstrate that gas-phase molecules can be used as a quantum interface to store and manipulate the OAM carried by ultrashort laser pulses. The experiment is conducted in CO2 molecules at room temperature in which l =±1 and l =2 OAM states have been encoded [2]. The overall writing & reading process relies on a pump-probe scheme with circular polarization exploiting the interplay between OAM and spin angular momentum.

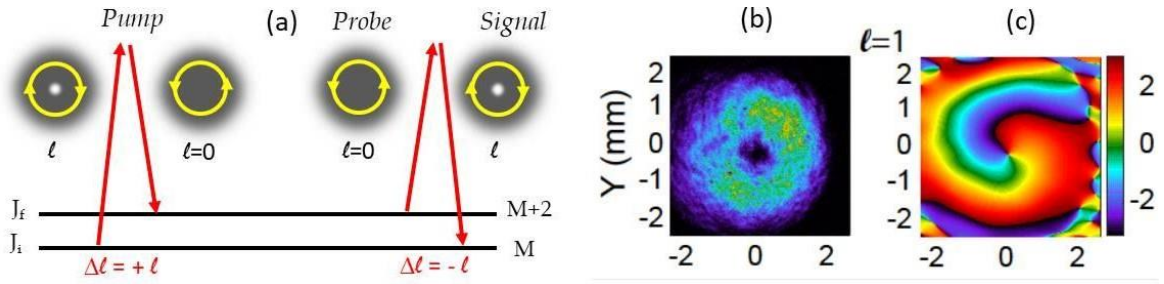

FIGURE 1

(a) Coupling scheme for storing an OAM into rotational states of molecules. (b) Signal intensity pattern measured in CO2 on a revival of alignment for the case *l* =1 (c) Retrieved spectral phase

As shown in Fig. 1(a) an ultrashort pump pulse consisting of a Stokes field carrying an OAM and an anti- Stokes field with no OAM stores the OAM information analogously to holography by producing a rotational wavepacket in the vibronic ground state of the molecules. The OAM can later be readout with a probe beam through the occurrence of "laser-induced field-free molecular alignment". A typical experimental result is depicted in Fig. 1 for the storage of l =1 with in (b) the signal intensity pattern for a pump-probe delay

τ=21.18 ps (corresponding to a peak of molecular alignment) and in (c) the reconstructed phase confirming that molecules have restored the helical phase structure of the OAM state encoded by the pump. As we will explain, the underlying mechanism can be explained by the spatial structuring of the molecular medium. The overall pump beam features an inhomogeneous polarization vector distribution that follows the pattern of a "q-plate" used for producing OAM beam [3]. The molecular alignment is therefore induced along the same direction leading to the production of a "molecular q-plate" whose periodical revival enables to restore the helical phase structure on demand. Besides applicability as storage medium with THz bandwidth application, the use of molecules as light-matter interface opens new functionalities in terms of optical processing and versatile control of OAM fields.

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[2] F. Trawi, F. Billard, O. Faucher, P. Béjot, E. Hertz, Laser & Photonics Reviews (2022) to be published https://doi.org/10.1002/lpor.202200525
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A first-principles approach to non-linear phononics in Bi_2Te_3

R. Busselez[1], A. Levchuk[1], B. Wilk[2], G. Vaudel[1], F. Labbé[1], K. Balin[2], J. Szade[2], P. Ruello[1], V. Juvé[1] and B. Arnaud[1]*

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Density Functionnal Theory (DFT) calculations allow to understand and disentangle the mechanisms playing a key role in the generation of acoustic and optical coherent phonons[1–4] . Recent experiments performed on a Bi_2Te_3 nanofilm have shown that a Thz pulse launches a coherent A_{1g} phonon. Such an observation can be explained by invoking either a sum frequency process[5] or non linear phonon-phonon couplings[6] . By resorting to group theory and calculating energy surfaces from first-principles, the main phonon-phonon couplings can be identified. Furthermore, a minimal model can be built to explain qualitatively pump-probe experiments. This model is validated by simulating the detection process as the modification of the dielectric function arising from the generation of optical or acoustic phonons can easily be evaluated at the random phase (RPA) approximation level[7]. Therefore, ab-initio calculations when combined with models are invaluable tools to shed lights on the complex mechanisms at the heart of ultrafast physics.

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[4] "Coherent acoustic phonons generated by ultrashort terahertz pulses in nanofilms of metals and topological insulators", A. Levchuk , Wilk, G. Vaudel , F. Labbé , B. Arnaud, K. Balin, J. Szade, P. Ruello, and V. Juvé, Phys. Rev. B 101, 180102(R) (2020).
[5] "Terahertz Sum-Frequency Excitation of a Raman-Active Phonon", S. Maehrlein, A. Paarmann, M. Wolf, and T. Kampfrath, Phys. Rev. Lett. 119, 127402 (2017).
[6] "Non-equilibrium control of complex solids by nonlinear phononics", R. Mankowsky, M. Först and A. Cavalleri, Rep. Prog. Phys. 79, 064503 (2016).
[7] "Local-field effects and excitonic effects in the calculated optical properties of semiconductors from first principles", B. Arnaud and M. Alouani, Phys. Rev. B 63, 085208 (2001).

Conservation of a half-integer angular momentum of light in nonlinear optics

Martin Luttmann[1], Mekha Vimal[1], Matthieu Guer[1,2], Jean-François Hergott[1], Antonio Z. Khoury[3], Carlos Hernández-García[2], Emilio Pisanty[4], Thierry Ruchon[1]

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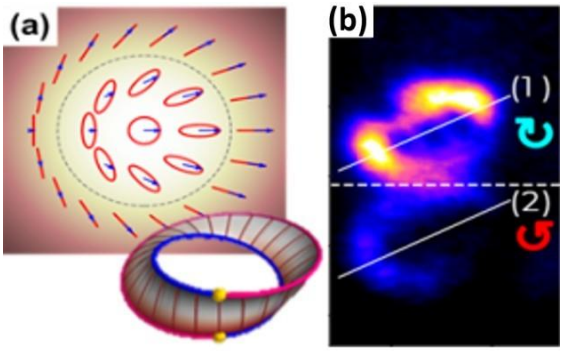
4. Department of Physics, King's College London, Strand Campus, WC2R 2LS, London, UK

Light beams, just like massive objects, can carry angular momentum. The spin angular momentum of light (SAM) is linked to the polarization of the field: the transformation generated by the SAM operator along the propagation axis, S_Z, which acts in the electric field space, leaves circularly polarized fields invariant under rotations. Thus, each photon in a left (resp. right) circularly polarized field carries a well-defined SAM, $\sigma = +\hbar$ (resp. $-\hbar$). In the 90's, a second form of optical angular momentum, the orbital angular momentum (OAM), was discovered. The OAM operator L_Z generates rotations of the position vector in the transverse plane [1]. The spatial phase of beams carrying OAM (the so-called vortex beams) spirals macroscopically about the optical axis, resulting in helical wave-fronts. Photons in an ideal vortex beam carry a quantum $\ell\hbar$ of OAM, where ℓ can be any integer number. Thus, in the paraxial approximation, the total angular momentum of light is $J_Z = L_Z + S_Z$, and is an integer multiple of $\hbar$. This is the case for instance of circularly polarized vortex beams, radially polarized or azimuthally polarized beams. In 2016 [2], exotic light fields were associated to a new form of "total" angular momentum, that is a linear combination of SAM and OAM: $J_{\gamma,Z} = L_Z + \gamma S_Z$, and is called generalized angular momentum (GAM). Remarkably, the γ factor can take half-integer values, making the photons of such beams carry an average $\hbar/2$ quantum of angular momentum. The GAM operator behaves as a "good" angular momentum operator, generating the rotations of space and electric field which leaves the beam unchanged. As such, its eigen modes make a good basis for these fields. Topologically, the GAM beams correspond to Möbius strips of polarization (Figure). So far, these beams remained a curiosity, with no report of their manipulation in nonlinear optics.



(a) Polarization Möbius strip field for $\gamma = 1/2$. The polarization ellipses are in red, and the electric field at a given instant in time in blue. The field is invariant under rotation of the spatial dependance by an angle θ, combined with a rotation of the polarization by a fraction $\gamma\theta$ of that angle. **(inset)** Möbius strip topology of the field in (a).
(b) Experimental image of harmonic 7 driven by a polarization Möbius strip, in a non-collinear geometry. It contains two contributions of opposite SAM (red and blue arrows), and OAM 3 and 4.

In this communication, we will report on GAM conservation in a highly nonlinear process. We prepared a driving beam in a state of ill-defined SAM and OAM, but well-defined GAM charge $j_\gamma = \hbar/2$ (Figure (a)). By implementing novel OAM measurement techniques in the XUV, we measured the GAM charge of high order harmonics generated with this driver. We found that the qth harmonic carries a GAM of $q\hbar/2$, while its SAM and OAM are ill-defined (Figure (b)) [3]. Each harmonic is thus a polarization Möbius strip, retaining the topology of the driver, with an increased GAM charge. In the time domain, the XUV emission takes the form of a single-coil light spring with attosecond dynamics. The observation of GAM conservation in nonlinear optics could open a wealth of applications in ultrafast physics, angular momentum-based spectroscopy, or the study of chiral materials. Besides, to what extent light beams with half-integer angular momentum have "fermionic" behavior remains a pending question.

(1) L. Allen et al., Phys. Rev. A 45, 8185 (1992).
(2) K. E. Ballantine et al., Science Advances 2, e1501748 (2016).
(3) M. Luttmann et al., arXiv preprint arXiv:2209.00454 (2022).

SESSION 3

Attosecond and XUV science

Simulations of attosecond pump-probe experiments on CO_2

Alexis Texier, Fabien Gatti, Lou Barreau[1], John Stanton[2]

1. Institut des Sciences Moléculaires d'Orsay, UMR 8214, CNRS, Université Paris-Saclay, France
2. University of Florida, USA

In CO_2, coherent excitation of the Fermi resonance in the electronic ground state was probed with time-resolved soft X-ray absorption to C core-excited states and shows 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 2πu and 1s-1 3s states. 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].

We simulate the vibrational wavepacket and the absorption spectra with the Multi-Configuration Time-Dependent (MCTDH) method. The simulations are in excellent agreement with the corresponding experimental X-ray transient absorption and allows us to understand the precise nature of the created vibrational wavepacket.

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Attosecond spectroscopy of molecular core levels

Hugo Marroux

LIDYL

Excitation of molecular core-levels provides element specificity to x-ray absorption spectroscopy. The information content of this spectroscopy is then limited by the broad linewidths due to the fast electronic decay of the core-excited molecules. I will show how attosecond transient absorption spectroscopy can be employed to first observe this decay in the time domain, but also how it can be used to manipulate the system's response. The manipulated molecular response induces a lineshape narrowing below its natural linewidth, revealing hidden transitions. I will show results in gaseous iodinated molecules and how this narrowing can reveal the ligand field splitting of the iodine 4d core-levels.

Ab-initio simulations of ultrafast electron-nuclear dynamics: Paving the way to attochemistry

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The advent of attosecond physics allowed the observation and manipulation of dynamic processes occurring within the intrinsic time scale of the charge motion in atoms and molecules. This has opened the door to the realization of the dream of attochemistry, namely to control chemical reactions through the manipulation of the pure electron dynamics taking place in the first instants after the excitation of the system.

Thereby, the existence of long-lasting electronic coherences in molecular systems is the first key prerequisite to its realization. Furthermore, understating the mechanism leading to or preventing the loss of coherence is necessary for its development.

The first measurement of decoherence and revival in attosecond charge migration will be presented. This dynamics occurs after excitation of silane (SiH_4) by an IR pulse. Simulations treating quantum mechanically both the electronic and nuclear degrees of freedom permitting the interpretation of the experimental results will be discussed. Using these simulations, the behavior of the coherence and the possibility to conserve coherence trough conical intersection will be rationalized.

The second key prerequisite of the realization of attochemistry is the understanding of how charge migration can impact the reactivity of a molecular system. Recently, an XUV-pump IR-probe experiment performed on adenine has demonstrated a sub 3 fs delay in its dicationic signal. It will be shown, using multielectron wave-packet propagation, that this delay is due to a correlation-driven charge migration occurring in the correlation band region of the molecule that stabilizes the system. The stabilization is due to the delocalization of the created hole and its change from σ to π character. The generality of the observed dynamics makes correlation bands a promising playground for the exploration of the possibilities offered by attochemistry.

High repetition rate few-cycle Mid-IR laser source

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We report on a widely tunable (4-10 μm), high repetition rate, mid-IR few-cycle source based on a nonlinearly compressed Yb-doped industrial laser pumping an intrapulse difference frequency generation (iDFG) [1-2] followed by an Optical Parametric Amplification (OPA) stage. The source delivers 73 fs long pulses at 250 kHz with energy of 640 nJ at 8 μm.

The iDFG process is realized in a 1 mm-thick LiGaS2 (LGS) crystal pumped by a nonlinearly compressed Yd-doped laser delivering 140 μJ, 8 fs, pulses centered at 1030 nm. A bichromatic waveplate (BWP) and an optimized chirp management of the driving source allows to improve the process efficiency by a factor of 2.5 [3]. The iDFG output is then amplified in an OPA stage realized using a 3 mm-thick LGS crystal pumped by the iDFG not converted pump. The overall conversion efficiency from pump to mid-IR is 2 %.

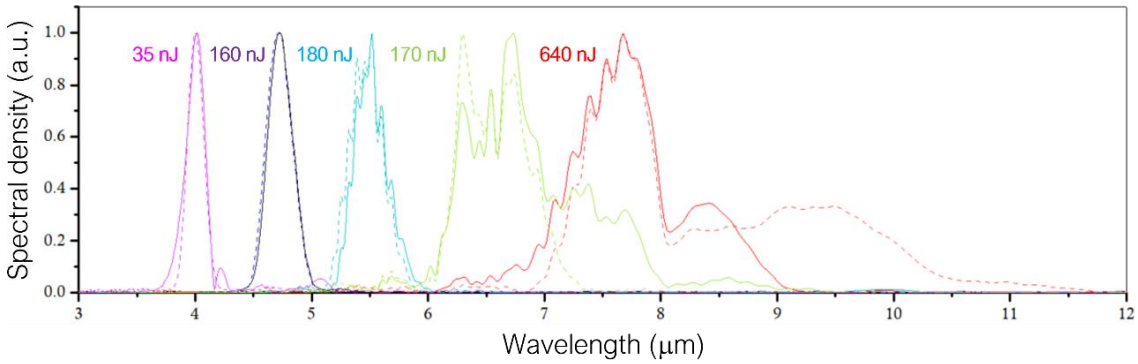

Figure 1. FTIR measured spectrum traces on the tunability output range and corresponding energies. (Solid line) iDFG+OPA output, (Dashed line) iDFG output.

The mid-IR tunability is measured using a homemade FTIR spectrometer. On figure 1 the measured spectra and the corresponding energies are shown. The temporal properties of the mid-IR pulses are measured using an electro-optic sampling (EOS) scheme [4]. We measured a FWHM pulse duration of 73 fs, corresponding to 2.5 optical cycles at the central wavelength of 8 μm. The amplitude noise transfer mechanism is analyzed through the measurement of the Relative intensity Noise (RIN) of the nonlinearly compressed and the generated mid-IR. An integrated RIN of 0.5% rms over [250 Hz; 125 kHz] bandwidth is measured at 8 μm.

The noise performances characterized in the mid-IR at high repetition rate make the source an unprecedented tool for improved signal-to-noise ratio 2D ultrafast spectroscopy measurements. The inherently CEP-stability, the few-cycle pulse duration, and the μJ-energy level in the Mid-IR perfectly suits solid HHG laser driver's needs.

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SESSION 4

Intense Laser Interaction

Optimizing laser coupling, matter heating, and particle acceleration from solids, using multiplexed ultraintense lasers

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Exploiting the potential of ultra-high intensity laser irradiation, for particle and radiation generation, will require, due to technology limitation, to use multi-beams arrangement. That's why the next generation of petawatt (PW) lasers are, and will be, a combination of several beamlines, e.g., the iCAN project [1]. The overlapping of these spatially separated beamlines is inevitable and may heavily affect the laser energy coupling [2-4]. We here investigate how to optimize their coupling with solid targets. Experimentally, we show that using two intense laser beams overlapped in a mirror-like configuration onto a solid target having a long preplasma, the generation of hot electrons at the target front was much improved and so was the ion acceleration at the target backside. The underlying mechanism is pinpointed with multidimensional particle-in-cell (PIC) simulations, which demonstrate that the magnetic fields self-induced by the electron currents driven by the two laser beams at the target front can reconnect, thereby enhancing the production of hot electrons, and favouring their subsequent magnetic guiding across the target. Our simulations also reveal that such effects can be further improved when overlapping more than two beamlines.

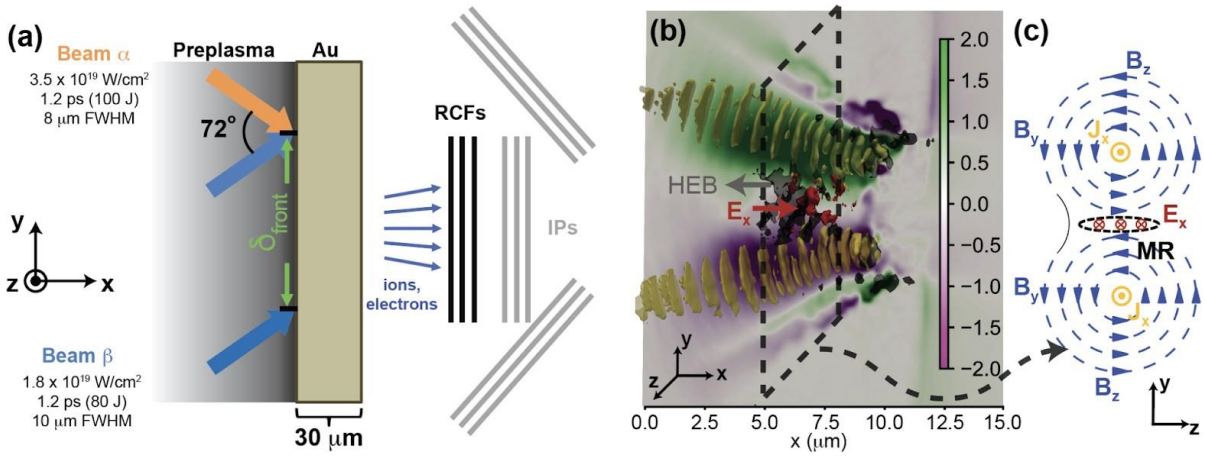

Figure 1 (a) Schematic of the experiment using two intense laser beams irradiating a solid target, with opposite incidence angles and a variable separation distance between the laser spots on the target front. (b) 3D PIC simulation results showing the two laser beams (yellow), the longitudinal electric field induced by the MR (red), the hot-electron beam (HEB, gray), and the magnetic fields (the colormap); (b) schematic diagram of the MR induced in the overlapping region.

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Kα X-ray source driven by a multi-terrawatt femtosecond laser at 100 Hz for spectroscopy and imaging application

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The interest towards developing intense and compact ultrashort x-ray sources is motivated both by scientific applications like time-resolved X-ray diffraction or X-ray absorption spectroscopy[1] and societal applications such as phase contrast imaging for biology and medicine[2]. At LP3 laboratory, such objectives are today achievable thanks to development of hard Kalpha X-ray pulsed laser plasma source generated by interaction of an ultrahigh intensity femtosecond laser delivering by ASUR laser facility with a solid target. We first determined the absolute yield of the K_α X-ray source (at 17.48 keV) and the X-ray spot size for a wide range of laser intensity ($I \sim 10^{17}$ - 2.8×10^{19} W/cm²) and for four values of the temporal contrast ratio ($6.7\times10^{7} < CR < 3.3\times10^{10}$) (figure 1 a) and b)). In particular, we measured the highest molybdenum K_α photon production reported to date at 100 Hz[3,4] with a K_α photon flux of 1×10^{11}ph/(sr.s) with an X-ray source size less than 90 µm at the driving laser intensity of $\sim 10^{19}$ W/cm².

Based on the exploitation of unique performances (figure 1 a) and b)), recent developments will be presented, namely stations of time resolved X-ray diffraction with first measured rocking curves of gold samples and phase contrast X-ray imaging instrument[5] (figure c)). For the first time, we will present quantitative measurements of refractive index extracted with advanced phase contrast imaging technique, named Multilateral Shearing Interferometry in collaboration with CEA List.

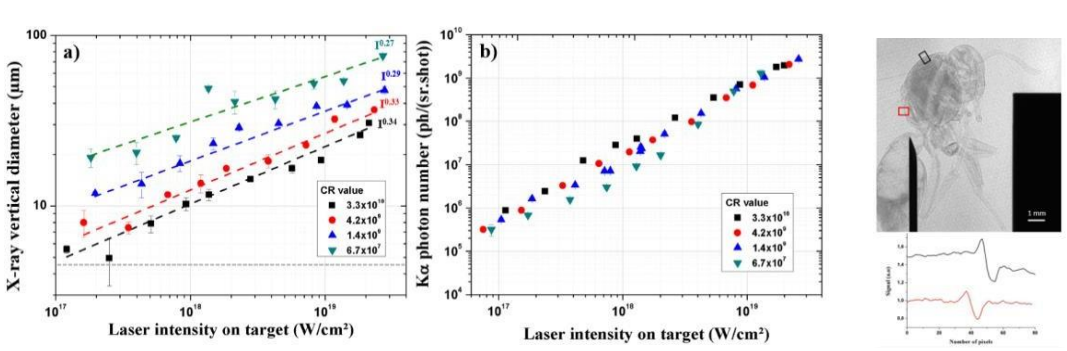

Figure 1. a) FWHM X-ray vertical diameter versus laser intensity on target. Fitting power functions in form of I^{ε} with ε varying with the contrast ratio value are shown by dashed lines. The grey horizontal dashed line is the FWHM vertical laser focal diameter (~4.6 µm). b) K_α photon number versus laser intensity on target. c) Raw phase contrast imaging on biological sample (wasp)

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[5] M. Gambari et al., Scientific Report **10**, 6766 (2020).

Recent news about Laser-Plasma Accelerator based Free Electron Lasers

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Free Electrons Lasers (FELs) are outstanding light sources, providing short duration and high-brilliance radiation at wavelengths spanning from the infrared to the X-ray domains. Several users facilities are now available worldwide.

While those devices rely on state-of-the-art large-scale accelerators, advancements on laser-plasma accelerators, which harness giga-volt-per-centimeter accelerating fields, showcase a promising technology as compact drivers for free-electron lasers.

We present the last experimental results in terms of LPA based FELs.

KAIO-Beamline, the modular high-repetition rate laser-plasma electron accelerator for broad range of applications

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The advent of high peak power lasers fueled the development of laser-plasma accelerators (LPA). The numerous advantages of LPA-produced electron beams (micrometer- scale source size, femtosecond pulse durations, low beam divergence, high peak currents) makes them ideally suited to ultrafast electron diffraction, non-destructive imaging, radiobiology, or space radiation production. At high repetition rate, LPA electron sources are particularly appealing as they offer long-term stability compared to low rep rate LPAs, and real-time access to statistics [1], enabling efficient complex parametric studies (e.g. in radiobiology [2]).

Nevertheless, implementing LPA sources at high repetition rates for applications remains challenging, as LPAs require (i) laser drivers with both high average and high peak power, (ii) high-performance vacuum handling to manage the large target gas loads and (iii) adequate management of large data sets generated during measurement runs.

The novel KAIO-Beamline aims at addressing these challenges by combining three key ingredients: (i) an industrial high average power laser system, (ii) a versatile temporal pulse post-compression stage and (iii) a compact electron accelerator module with integrated user interface and data management system. The KAIO-Beamline approach is compatible with a wide range of commercial laser platforms as it is modular and features versatile post- compression based on multi-pass cell technology [3]. Secondary electron is coupled to an automatic high repetition rate data acquisition program, which monitors and logs all relevant parameters during day-long experimental runs.

We believe that KAIO-Beamline is a promising step towards LPA industrialization and wide dissemination in universities, factories and hospitals.

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SESSION 5

Condensed Phase 2

Dynamiques aux interfaces de graphene et hBN chargées

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Nanofluidic experiments have exhibited a large surface charge of graphene in contact with water. First-principle Molecular Dynamics simulations aiming at understanding this phenomenon show that its origin is the physisorption of hydroxide ions in the first water layer of graphene. The dynamlcs of hydroxide in this water layer is further studied. Hydroxide diffuses through proton transfer, as in the Grotthus mechanism. Correlation between transport and hydroxide orientation leads to kinetic trapping of the charge close to the surface and an unexpected surface conductivity as a result.

Unravelling the Transient Depth Magnetic Profile During Ultrafast Demagnetization of an Iron Thin Film

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During the last two decades, a variety of models have been developed to explain the ultrafast quenching of magnetization following femtosecond optical excitation [1,2,3]. These models can be classified into two broad categories, relying either on a local or a non-local transfer of angular momentum [3]. To distinguish those local and non-local effects we can measure the magnetization depth profile with femtosecond resolution, thanks to time-resolved x-ray resonant magnetic reflectivity [4, 5]. In this presentation, I will show how, from our experimental results gathered at the free electron laser FLASH, we can unravel the dynamics of the transient inhomogeneous depth magnetic profile of an Fe layer after optical excitation.

First our experiment on a polycrystalline Fe sample reveals two distinct dynamics at different time scales for the structure and the magnetization [Fig. 1 (a) and (b)]. Until one picosecond, the magnetic signal is quickly evolving while the structural one stays more or less constant. After that, the magnetic signal is slowly coming back to equilibrium while the structural one changes periodically. For this structural signal, we observe a maximum dilation of 2Å followed by a coherent damped oscillation of the thickness of the sample [Fig. 1 (a)]. This dynamic is due to stresses that are generated by the rapid increase in temperature and might be enhanced via magnetostrictive effects.

Second, the quantitative analysis of our magnetic reflectivity data allows us to retrieve the inhomogeneous depth magnetic profile for different delays after the optical excitations. As shown in Fig. 1 (c), close to the bottom interface there is an overall reduction of the demagnetization. When comparing this result to simulation we can directly show that both local and non-local phenomena [6] take place at the same time scale and that there is probably a contribution from spin currents that could carry the magnetization beyond the magnetic layer [7].

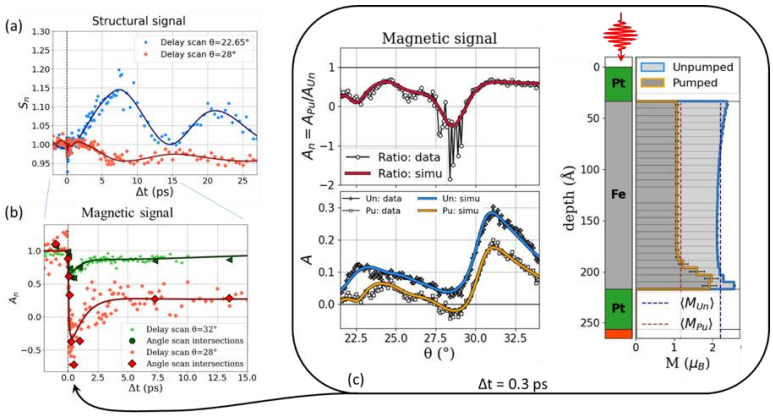

Figure 1: Delay scans for the (a) structural signal and (b) magnetic signal extracted from x-ray magnetic reflectivity as a function of the delay between the pump and the probe for two different reflectivity angles. (c) Magnetic signal as a function of the reflectivity angle θ for a delay of 0.3 ps and its fit, which give the derived depth magnetic profile without optical excitation (blue) and 300 fs after an optical excitation (orange).

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Disentangling lattice and electronic instabilities in the excitonic insulator candidate Ta_2NiSe_5 by non-equilibrium spectroscopy

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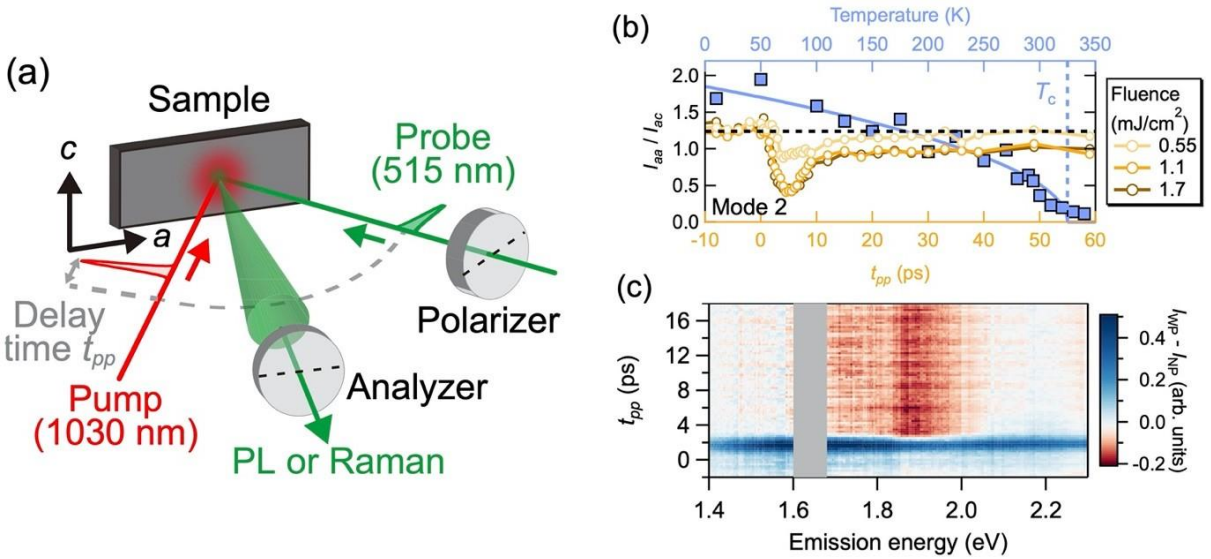
Ta_2NiSe_5 is considered as one of an excitonic insulator candidates showing a semiconductor / semimetal-to-insulator (SI) transition below $T_C = 325$ K. However, since a structural transition accompanies the SI transition, deciphering the role of electronic and lattice degrees of freedom in driving the SI transition has remained elusive [2]. Here, we investigate the photoexcited nonequilibrium state in Ta_2NiSe_5 using pump-probe Raman and photoluminescence spectroscopies. The combined non-equilibirum spectroscopic measurements of the lattice and electronic states reveal the presence of a photoexcited metastable state where the insulating gap is suppressed but the low temperature structural distortion is preserved. We conclude that electron correlations play a vital role in theSI transition of Ta_2NiSe_5 [3].



(a) Non-collinear 2-color pump probe polarization-resolved Raman / photoluminescence set-up. **(b)** Dynamics of the monoclinic distortion in Ta_2NiSe_5 as a function of pump-probe delay using Raman phonon selection rules for 3 different fluence (yellow / brown circles). Measurements are performed in the monoclinic / insulating phase at T=150K. The blue squares are the equilibrium temperature dependence of the same distortion which vanishes at T_C where Ta_2NiSe_5 undergoes a insulator / monoclinic to semi-metal / orthorhombic transition. **(c)** Dynamics of the optical emission as a function of pump probe delay for a fluence of 1.6 mJ/cm^2 at 150K in the insulating phase. After an initial increase (blue) the emission at 1.85 eV remains suppressed (red) until at least 18 ps, indicating a metastable phase with a collapsed insultating gap, but a preserved monoclinic distortion.

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Ultrafast dynamics of laser-heated copper using time-resolved photoelectron spectroscopy

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A solid sample submitted to an ultrafast heating induced by an ultrashort laser pulse is brought in out- of-equilibrium thermodynamic conditions where the energy is mainly deposited in the electronic population while the lattice remains cold and unperturbed. The associated material properties and subsequent relaxation can be modified in these conditions and are the subject of intense research efforts from theoretical modelling to experiments. One key feature of this particular regime is related to the modification of the electronic density of states (DOS) induced by this ultrafast energy deposition [1–3]. The experimental study presented here aims at a direct probing of the occupied DOS using time-resolved x-ray photoelectron spectroscopy which could be tool complementary to diagnostics employed in this research area including x-ray absorption spectroscopy, x-ray diffraction or transient optical absorption [4–7]. The experimental results presented have been acquired on a dedicated XUV beamline based on High-order Harmonic Generation that delivers femtosecond pulses in the 80 - 100 eV photon energy range at the CELIA laboratory (Bordeaux, France) [8]. The data interpretation will be developed on a carefull and complete description of space charge effects expected in these experimental conditions in order to extract the heating dynamics in the recorded signal. This space charges modelling based on the calculation of the photoelectron spectra induced by the heating laser [9] pulse coupled to PIC calculations (ASTRA code, DESY-Hambourg, Germany) of the mutual interaction of the heating and probing electron clouds will be presented in details as a complete and coherent approach that could be applied in other configurations that involve similar space charge induced measurement distortions.

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SESSION 6

Attosecond and gas phase ultrafast science

Attosecond signatures of spatially extended hole in a molecule

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Electrons emitted into the continuum after XUV ionization scatter on their ionic parent potential just after photoionization. The resulting photoelectrons carry in their phases the signature of the potential landscape. According to Wigner theory, this scattering process can be interpreted as a time delay in photoionization [1] down to the attosecond scale (1 as=10^{-18} s). Attosecond metrology is hence a powerful tool to unravel the detail of the potential. This is of particular interest for many-atoms molecules.

In this work, the RABBIT protocol [2] (Fig. 1a) is applied to C10Hx molecules with planar and almost spherical arrangements of their atoms (Fig. 1b). A time shift of few tens of attosecond is observed between the photoelectrons originated from the two molecules (Fig. 1c). Using scattering theory, we show that this global time shift can be associated to residual hole shape created in the molecule [3]. This interpretation is supported by Static-Exchange Density Functional Theory (SE-DFT) calculations.

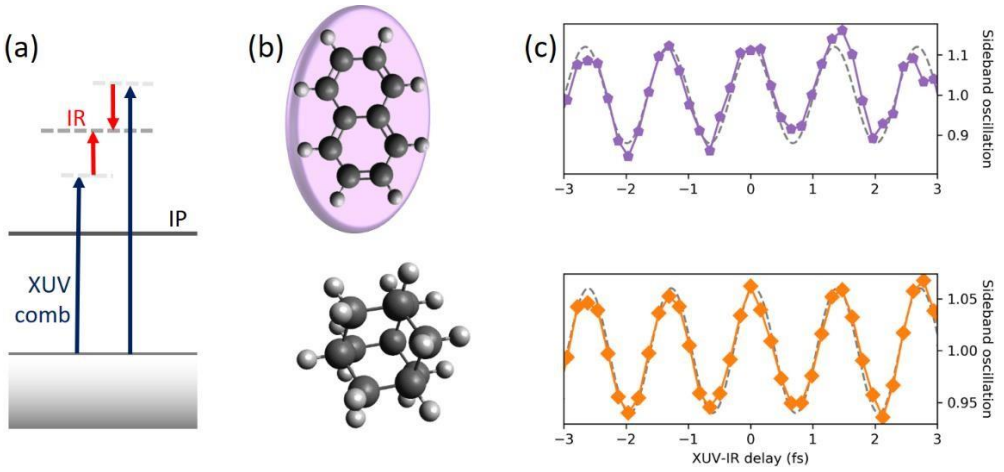

Figure 1: (a) RABBIT principle [2] representing the ionization interference of an XUV energy comb (dark blue) dressed by IR pulse (red) (b) Naphthalene (top) and adamantane (bottom) and (c) their corresponding sideband oscillation.

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Recollisions in circularly polarized pulses : from tunnel-ionization to the return of the electron

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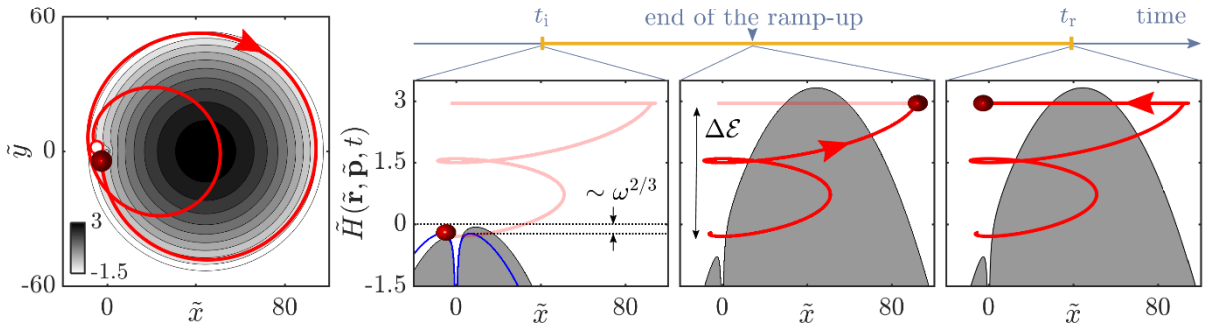
Subjecting electrons in atoms to strong laser pulses gives rise to a variety of highly nonlinear phenomena such as high harmonic generation (HHG), above-threshold ionization (ATI) and non-sequential double ionization (NSDI). The built-in mechanism behind these highly nonlinear phenomena is the *recollision*, a keystone of attosecond science [1]. At the recollision, the electron spends time of the order of the attosecond close to the core—the natural timescales of the electron inside the atom—and as a consequence crucial information on the target atom and electrons, such as their real-time motion and electron-electron correlations, is encoded in the spectrum of HHG and ATI, and the photoelectron momentum distributions (PMDs) of NSDI.

Meanwhile, circularly polarized laser pulses have been used for probing non-adiabatic effects and the time the electron spends to tunnel-ionize through the barrier induced by the strong laser field, a setup known as *attoclock* [2]. This setup was particularly useful because of the absence of recollision for circularly polarized pulses, in seeming contradiction with experimental and theoretical results for which recollisions in atoms subjected to circularly polarized pulses have already been observed in Mg [3].

Here, I would present how electrons subjected to circularly polarized pulses undergo recollisions only for specific atomic species by surveying their journey from tunnel-ionization to their return. I will show that the quantum mechanical tunnel-ionization is governed by conservation laws of classical mechanics; that the envelope of the laser pulse and non-adiabatic effects play a crucial role in the conditions under which the electron returns [4,5]; and I will present a work along those lines initiated with experimental collaborators [6].



Electron trajectory (red solid lines) in an atom subjected to a circularly polarized laser pulse in the frame rotating with the laser pulse: from ionization (white circle) to the return of the electron (red circle). Right panels are snapshots of the trajectory of the electron as a function of its energy. The grey areas are the effective potential energy of the electron.

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Photoisomerization of Cis-Azobenzene: Understanding the Quantum Yield Wavelength Dependency

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The cis-to-trans photo-isomerisation of azobenzene has been little studied. In particular, the dependency of the photoisomerisation quantum yield on wavelength is not clear, varying notably from study to study [1-3]. We have revisited computationally the mechanism, after excitation to the nπ* and ππ* states, using non-adiabatic surface hopping dynamics in combination with multi-reference CASSCF electronic structure calculations [4]. On exciting to the higher energy S_2 ππ* state compared to the lower energy S_1 nπ* state, a 0.10 reduction of photoisomerisation quantum yield is obtained, in close agreement with the most recent experimental values [1]. By direct comparison of both excitations, we have found that the reason for the decrease in quantum yield is not the same as for the trans-to-cis photoisomerisation, as previously assumed.

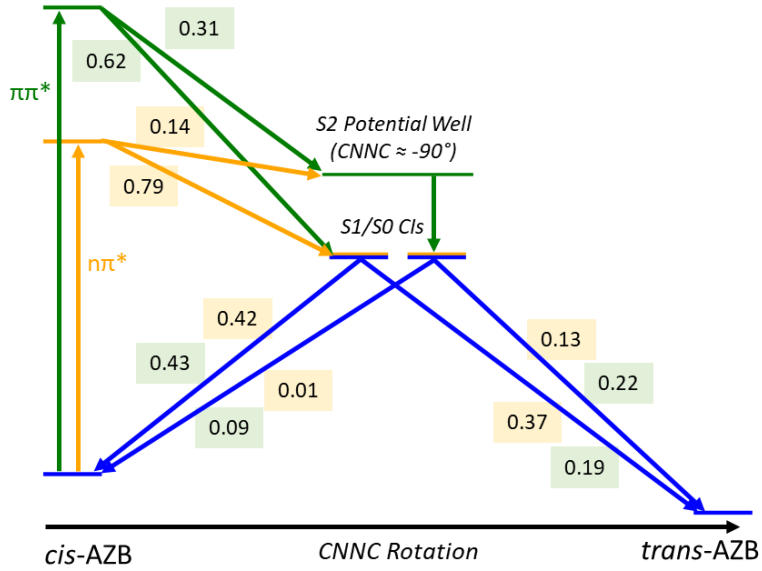

Figure 1: Summary of the major pathways accessible to cis-AZB, and fraction of photochromes which follow each pathway, after both nπ * (yellow) and ππ* (green) excitation.

The wavelength dependency of the quantum yield is instead found to be due to a potential well on the S_2 surface, from which either cis or trans-azobenzene can be formed. The combination of two factors results in the reduction of 0.10 of the quantum yield of photoisomerisation on ππ* excitation of cis-azobenzene, compared to nπ* excitation. Firstly, the potential well is more easily accessed after ππ* excitation - an additional 15-17% of photochromes, which under nπ* excitation would have exclusively formed trans-azobenzene, are trapped in this well after ππ* excitation. Secondly, the probability of forming cis-azobenzene when leaving this well is also higher after ππ* excitation, increasing from 9% to 35%.

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Chromatic aberrations control in high order harmonic XUV beams by spatial shaping of the fundamental

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High order harmonic generated in gases is a source that is now commonly used to generate attosecond pulses. When refocused on a target, it is however now known that the different harmonics are focused at places that are longitudinally separated [1 - 6]. These chromatic aberrations make the attosecond structure space dependent near XUV focus and must be considered.

We study the XUV harmonic focusing properties with spectral resolution with a SWORD [2] approach that provides both the spatial profile and wavefront curvature of the XUV beam and allows us to obtain the XUV focus size and position. We observe that XUV chromatic aberrations exists and that they are important near the XUV focus where both the spectral content and relative harmonic dephasing changes with z.

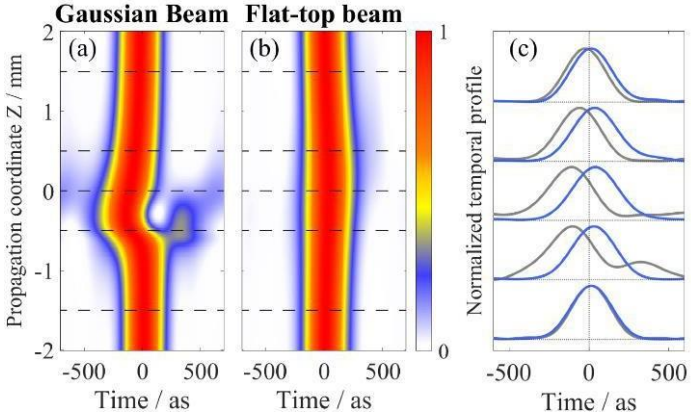

FIGURE 1: Evolution of the on-axis attosecond pulse profile (H11 – H19) with propagation near XUV focus. Harmonics are generated in a gas jet with a Gaussian beam (a) or a spatially shaped flat-top beam (b). (c) Temporal profiles of the pulses generated with the Gaussian beam (grey) or the flat top beam (blue) at specific positions indicated by the dashed lines.

To control XUV focusing and chromatic aberrations, we shape the fundamental beam in the generating medium to generate harmonics with a flat top beam and compare the XUV beam properties when emitted with a Gaussian beam or a flat top beam. We observe that spatial shaping impacts the XUV chromatic aberrations that can be reduced when harmonics are generated with a flat top beam (Figure 1) as compared to the standard case where harmonics are generated by a truncated Gaussian beam.

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SESSION 7

Nano Science

LaB_6 nano-tip as an ultrafast electron source

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LaB_6 has been traditionally used as a thermionic electron source for electron microscopes, but has been hardly put to practical use as cold field emitter (CFE) [(1)(2)]. The chemical inertness, the high conductivity and the dense structure make LaB_6 a good candidate as an ultrafast electron source. In the present work we study the emission properties of LaB_6 nano-tip, fabricated by focused ion beam milling, under static electric field (static emission) and under femtosecond laser illumination (ultrafast emission), using a 2.25mm laser at 13 MHz. In the static emission regime, the electron emission follows the Fowler-Nordheim mechanism underlining the metallic nature of LaB_6. The electron energy shows a spread of less than 1 eV and the emission current is stable for several hours. Under laser illumination, varying the static electric field and the laser power, four emission regimes were reported, as shown in Figure 1. For an electric field close to the field for the static emission, the use of the laser pulses, at low power, induces an increase in the emitted current and in the emission stability. For lower static field and higher laser power multiphoton emission regimes [4] are observed, inducing a change in the emission patter, in the energy spread and stability. In particular, the Field Emission Pattern, when compared with the Field Ion Microscopy image of the surface, suggests that electrons are emitted from the [001] pole and [011] poles under strong laser illumination. By further increasing the laser power, a transition from Multiphoton to Optical Field emission occurs, emitting electrons of energy higher than the Fermi level.

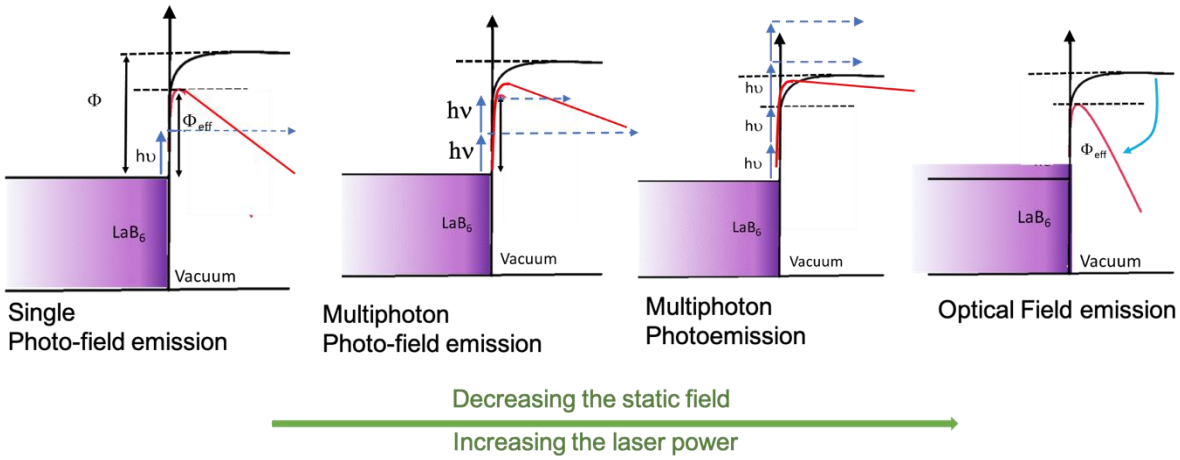

Figure 1 Electron emission processes for ultrafast emission.

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Driving orbital magnetism in metallic nanoparticles through plasmonic effects

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The topic of this contribution is the generation of large magnetic fields in non-magnetic materials through polarized laser fields.

Transfer of angular momentum from helicity- controlled laser fields to a nonmagnetic electronic system can lead to the creation of magnetization. The underlying mechanism in metallic nanoparticles has been identified as the inverse Faraday effect (IFE), whereby a quasi-static magnetic field is generated by an external oscillating lase field and is proportional to the laser intensity.

Here, we show that the IFE can be strongly amplified in small gold nanoparticles thanks to plasmonic effects. If the laser frequency matches the plasma frequency of the conduction electrons in the metal (surface plasmon resonance), a strong oscillating electric field is excited in the nanoparticle. Through the IFE, this internal self-consistent field generates a sizeable magnetization, of the order of tens of Bohr magnetons. The primary contribution to the magnetization comes from surface currents generated by the self-consistent field. The effect is maximum for circularly polarized laser fields and disappears for linearly polarized fields.

This plasmonic IFE is studied here using both a simplified quantum hydrodynamic model [1] and fully quantum simulations based on time-dependent density functional theory [2]. This is an important step in the ultrafast manipulation of magnetic effects in nano-objects via electromagnetic waves, which may find applications for the storage, writing, and reading of information based on optical means.

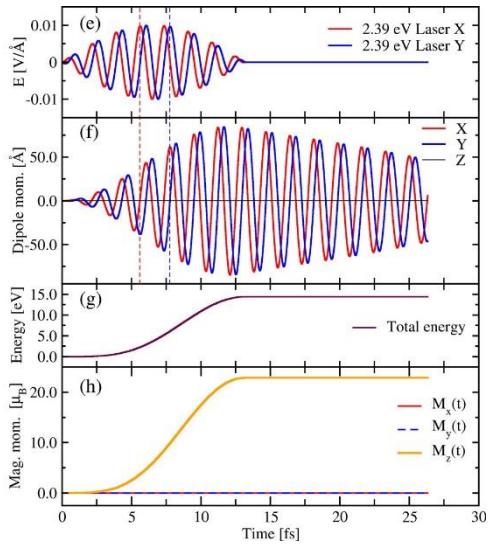

Figure: Circularly polarized laser excitation of a potassium K561 cluster. From top to bottom, the panels show: the time dependence of the x (red) and y (blue) components of the laser electric field (e); the three components of the dipole moment (f); the total energy absorbed by the electronic system (g); and the 3 components of the magnetic moment, in units of the Bohr magneton μB (h). From Ref. [2].

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Revealing the ultrafast spin-crossover mechanism in FeII(bpy)3 through quantum dynamics

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Iron(II) tris-bipyridinde is one of the prototypical Fe(II) complexes exhibiting magnetic bistability between low and high spin states (spin crossover). After photoexcitation, the HS state is formed in approximately 100 fs. However, the light-induced mechanism is still unknown and the role of the intermediate states has been matter of debate from both experimental[1,2] and theoretical perspectives[3,4]. Here, we present a wavepacket dynamics simulation of the $Fe(bpy)_3$ complex on a model Hamiltonian formed by the lowest metal-to-ligand-charge-transfer (MLCT) and metal-centered (MC) states of singlet, triplet and quintet multiplicities. The diabatic potentials are constructed along the main modes which are the Fe-N symmetric stretching, two Fe-N asymmetric stretchings and six high-frequency ligand modes formed by C-C and C-N stretchings and the couplings among states are included by non-adiabatic (NAC) and spin-orbit (SOC) couplings. The results show that after the excitation to the singlet MLCT manifold, the wavepacket evolves from these states to the ^{3}MLCT manifold. The transfer to the HS takes place after an internal conversion from this manifold to the ^{3}MC states, pointing that the dominant path is the indirect mechanism.

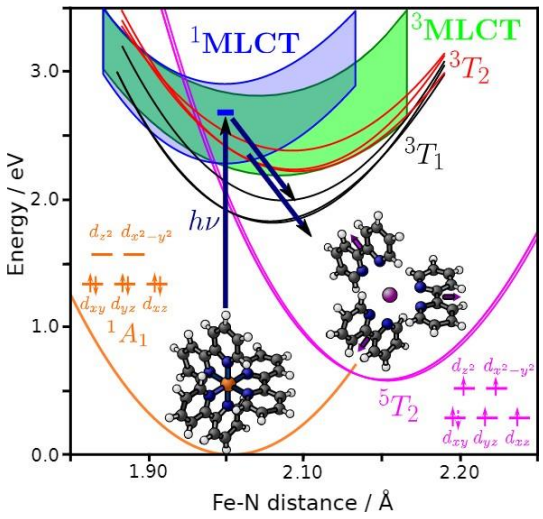

Figure 1. Diabatic potentials along the reaction coordinate

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Workshop
Dialogue Théorie-Expérience

WS1 – TBA

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WS2 – Structures et dynamiques de molécules chirales sondées par de brèves impulsions laser

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Je présenterai un état de l'art des travaux consacrés à l'étude de structures et dynamiques de molécules chirales en phase gazeuse. On se focalisera sur les études impliquant des impulsions laser de courte durée. Les verrous, tant théoriques qu'expérimentaux, seront explicitement mentionnés – ces verrous dépassant généralement le cadre des interactions chirales. D'éventuelles directions de recherche seront finalement discutées.

WS3 – Dihydropyrene photoisomerization: Aspirations to go beyond the static mechanistic picture

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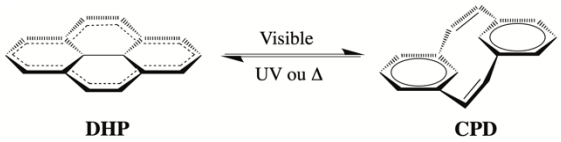
Photochromic compounds are systems that undergo a reversible isomerization between more than one state, using light as an external stimulus. The isomeric forms possess different geometrical and electronic structures meaning that they manifest different physical properties (e.g., luminescence, conductance, magnetism, etc.). Therefore, such systems have been intensively developed over the past few decades because they offer promising candidates for applications in the field of optoelectronics, biology, nano and material sciences. Among the many photochromic molecules known to date, derivatives of the dihydropyrene (DHP) family deserve a special attention for various reasons. Under visible light irradiation, the π-conjugation of the quasi-planar DHP annulene core is lost during the formation of the cyclophanediene (CPD) open-ring isomer, which is characterized by a step-like structure with two benzene rings connected by two ethylene bridges (Scheme 1) [1]. The metastable CPD isomer can then be converted back to the stable DHP photochemically with UV light or thermally. This system displays unusual negative photochromism because the stable DHP form absorbs at a longer wavelength than the metastable CPD isomer. Thus, it leaves open the possibility to excite the DHP system with a low-energy irradiation while converting it quantitatively to its CPD isomer. Interestingly, DHPs have been shown to operate in the biological window [2] and to produce singlet oxygen [3].



Scheme 1. The dihydropyrene (DHP) /cyclophanediene (CPD) photochromic couple.

Besides the wide range of potential applications offered by the exploitation of DHPs photoswitching properties, these systems also represent a major challenge from the theoretical point of view. The photoswitching mechanism of the simplest prototypical system (Scheme 1) is very complex, as it involves several coupled electronic excited states of various character (covalent and ionic, singly- and doubly- excited) [4, 5, 6]. Computational mechanistic studies so far have been solely carried out based on the static information provided by the exploration of the relevant potential energy surfaces. The very low photoisomerization quantum yield of the unsubstituted DHP has notably been rationalized, as well as the increase of this quantum yield upon appropriate substitution [7, 8]. Given the potential of these systems as photoswitches and as photosensitizers, it would be highly desirable to obtain information on their time evolution at ultra-short timescale, both from experimental (pump-probe experiments) and theoretical (dynamics simulations) viewpoints. Such studies would certainly contribute to gain a better understanding of the photodynamics of these fascinating photochromic systems.

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WS4 – Simulation by the hierarchical equations of motion with tensor trains

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Hierarchical equations of motion (HEOM) are a popular, numerically exact, non-Markovian and non-perturbative way to simulate open quantum systems [1]. In this framework, the full system is partitioned into a quantum system which interacts with a bath that mimics the environment. The bath should be chosen as harmonic oscillators linearly coupled to the system degrees of freedom. This common model has been widely used with HEOM to describe electron or proton transfers, photosynthetic complexes or multidimensional spectroscopy. However, for some realistic chemical systems with many degrees of freedom or a large coupling of the environment with the bath, solving HEOM set of equations is a tricky task which can be plagued by the dimensionality curse.

Tensor trains (TT) are a type of tensor decomposition which allows to deal with high dimensional tensors. Indeed, the storage and the computational cost is linear with the number of dimensions. Several libraries for the tensor train algebra are already available and propagation can be carried out using a time-dependent variational method. Shi [2], and then Borrelli [3] on a slightly different approach, propose to use this decomposition to compute the dynamical equations of HEOM. Their encouraging results trigger our interest in using such approach with model and larger realistic chemical systems.

We will present several examples using the TT method with steady-state absorption and emission spectroscopy of model Hamiltonians and optimal control theory for the laser-controlled electronic symmetry breaking in a phenylene ethynylene dimer [4].

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WS5 – Photodynamics of lanthanide and actinide complexes

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The electronic structure of lanthanide and actinide complexes with accessible f and d metal electrons makes them well suited as luminescent species. However, photodynamics/photoreactivity in the photo-excited states may significantly quench the luminescence lifetime. The cases of uranyl(VI), UO_2^{2+}, actinide cation and Ce^{3+} lanthanide cation will be taken as illustrations of ultrafast photodynamics probed by quantum chemical methods.

WS6 – Investigation of the vibrational and cooling dynamics of metal nanoparticles by combined experimental and modeling approaches

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The sudden absorption of energy by a nano-object launches a cascade of relaxation processes (internal thermalization, acoustic vibrations, cooling, …) involving femtosecond to nanosecond timescales. Their investigation allows to better understand how the macroscopic laws ruling electron interactions, elasticity, heat conduction or interfacial energy transfer are modified at the nanoscale. These studies have already greatly benefited from the combination of optical time-resolved spectroscopy experiments with modeling approaches including in particular multiphysics numerical simulations. This talk will focus on two specific physical processes whose understanding is currently being improved by a strong interplay between experimental and modeling approaches: the vibrational damping of nano-objects and heat transfer at the vicinity of nanometric heaters, where Fourier's law of heat conduction may become invalid.

WS7 – Discussion on the origin of the helicity-dependent magnetization dynamics

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In 2007, Stanciu et al. [1] discovered that a train of femtosecond pulses of light can trigger the so–called all–optical helicity–dependent switching (AO–HDS). Its presence has then been evidenced in many different types of magnetic thin films [2, 3] and provides us with a deterministic way to control the magnetization state, solely relying on the polarization of the light.

To this day, the phenomena involved in the emergence of such helicity-dependence during the ultrafast light-induced dynamics are still debated. The main candidate is usually assumed to be the inverse Faraday effect (IFE) [4], designating the generation of a magnetization proportional to the intensity of the circularly polarized light. Here, I will argue that the IFE is not likely to induce the experimentally seen dynamics and discuss the possible involvement of two other possible candidates, namely the magnetic circular dichroism (MCD) and the magnetization induced during light absorption [5] (MILA). Contrary to the IFE, the magnitude of both of the latter is proportional to the fluence of the light and can thus affect the dynamics on longer timescales.

Finally, I will provide a possible way of distinguishing between the MCD from the MILA experimentally.

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WS8 – Laser energy deposition into dielectric materials

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The interaction of intense laser pulses with dielectric materials (as SiO_2 or Al_2O_3) leads to a significant energy deposition, and subsequent phase transitions of matter. This system is of twofold interest: (i) on the fundamental point of view, the laser energy deposition results from the complex electron dynamics involving various physical processes on different time- and spatial-scales, (ii) for application purposes which correspond to the field of laser processing of materials, the laser energy deposition allows one to ablate, cut, weld materials, create nano-structures [1], change optical and mechanical material properties, including surgery for biological materials, etc. To make progress in the understanding of the physical processes at play during the laser energy deposition is thus important.

The phenomenology of laser induced electron dynamics and subsequent macroscopic material modifications is as follows. The laser energy is first absorbed by electrons through the processes of both ionization and excitation/relaxation in the conduction band. During the second stage, the absorbed laser energy is redistributed between the excited carriers while they undergo collisions with phonons, ions, and other electrons in the presence of the laser field. These processes eventually lead to the energy transfer to the lattice. This is a collisional picture base on electron intraband transitions [3]. Recently, it has clearly been shown that interband transition also contribute significantly to the laser energy deposition [4, 5]. The induced increase in temperature leads to the so called warm dense matter (not fully ionized plasma at solid density), which is also associated with high pressure gradients. The later give rise to the formation of hydrodynamic shock waves with propagate through the cold solid. The laser energy deposition and associated consequences thus pertain to a multi-physical and multi-scale system.

Despite main characteristics of the whole process are established, opened problems remain, which solutions would be found from experimental and theoretical works hand in hand. This talk is devoted to describe this field, and exhibit some of these opened problems. We will first address the general coupled multi-physics and multi-scale experimental and theoretical approach required to accurately characterize to present physical system. Then, we will focus on problems related to the laser pulse duration. Modeling efforts are mainly based on the collisional picture, in particular using the Drude model. This approach may become questionable for ultrashort laser pulses which duration is of the order of the collision characteristic time. On the opposite side, for pulses longer than the characteristic hydrodynamic time (ps) or with burst of fs pulses, the macroscopic properties of matter evolve during the interaction. The coupled observation and description of this transient system remain challenging.

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WS9 – Nanostructuration de la silice par laser femto-seconde

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Dans cet exposé, nous rappellerons quelques résultats expérimentaux associés à la structuration de la silice à l'échelle nanométrique suite à une excitation par laser femto-seconde. Nous mettrons ces résultats en regard de calculs effectués à l'échelle atomique par Dynamique Moléculaire. Plusieurs questions se posent quant à la prise en compte plus fine des mécanismes électroniques mis en jeu lors des impulsions laser.

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WS10 – First principles simulations of biological matter radiolysis, state-of-the-art and roadmap

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3. IDRIS, CNRS, Orsay, France
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The transient collision of high-energy-transfer particles with matter results in ionization or excitation of its constituent molecules. Huge amounts of energy are deposited locally, typically several tens of eV, with copious emission of low energy electrons [1]. These early physical events produce a myriad of reactive radical species that are at the source of cascades of chemical processes spanning several spatial and temporal scales. The physical chemistry of these ultrafast processes is not well understood at the present time. First principle simulations would bring valuable insights on the mechanisms involved at the molecular scale and help interpret ultrafast experiments (e.g. attosecond spectroscopy or pulse radiolysis).

We have embarked on a long-term research program aiming at devising innovative approaches to simulate energy deposition and ultrafast reactivity within complex matter (e.g. biological matter). Our approaches are based on Density Functional Theory (ADFT) Real-Time Time Dependent ADFT and Molecular Dynamics (MD) simulations [2,3]. A hybrid scheme coupling these approaches to polarizable force fields have been devised [4]. Theses methodologies have been implemented in deMon2k. They allow simulations of collisions of molecules with fast ions or ionization by ionizing photons, ultrafast charge migration, energy relaxation/dissipation on the attosecond time scale (cf. Figure) for systems comprised of up to 1,000 atoms [5]. MD further gives access to chemical reactivity taking place in the first picosecond after irradiation [6].

I will introduce the methodologies and applications to various questions of current high interest in radiation chemistry. I will also mention possible area of research for the coming years.

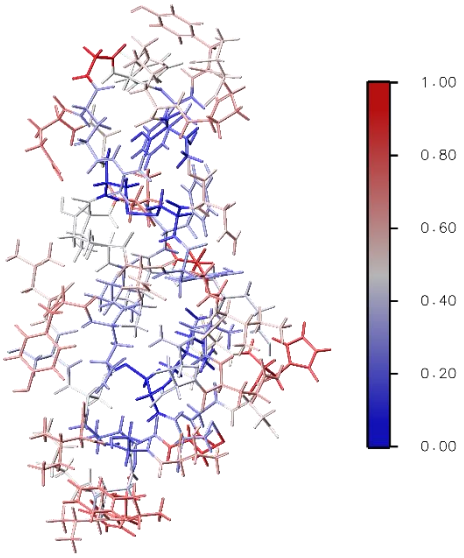

Figure 2 : our RT-TD-ADFT methodology permits to investigate XUV irradiation of large biological molecules and revearl key insights on the ionization mechanism.

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WS11 – Non-radiative transitions in bio-relevant systems: non-adiabatic dynamics simulations as a bridge between theory and experiment?

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…

POSTERS

1

The dihydropyrene photoisomerization mechanism revisited : the photochemical funnel dilemma

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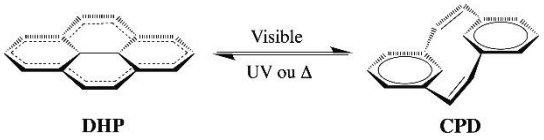
Molecular photoswitches have emerged as fascinating chemical species that can be converted between at least two different isomeric forms in a controlled manner under external optical stimuli. These systems have been intensively developed over the past few decades because of their use in numerous important applications in the field of biology, nano and material sciences. Among the many photochromic molecules known to date, derivatives of the dihydropyrene (DHP) family deserve a special attention for various reasons. Under visible light irradiation, the □ -conjugation of the quasi-planar DHP annulene core is lost during the formation of the cyclophanediene (CPD) open-ring isomer, which is characterized by a step-like structure with two benzene rings connected by two ethylene bridges (Scheme 1).[1] The metastable CPD isomer can then be converted back to the stable DHP photochemically with UV light or thermally. This system displays unusual negative photochromism because the stable DHP form absorbs at a longer wavelength than the metastable CPD isomer. Thus, it leaves open the possibility to excite the DHP system with a low-energy irradiation while converting it quantitatively to its CPD isomer. Interestingly, DHPs have been shown to operate in the biological window[2] and to produce singlet oxygen.[3]



Scheme 1. The dihydropyrene (DHP) /cyclophanediene (CPD) photochromic couple.

Besides the wide range of potential applications offered by the exploitation of DHPs photoswitching properties, these systems also represent a formidable challenge from the theoretical point of view. The photoswitching mechanism of the simplest prototypical system (Scheme 1) is very complex, as it involves several coupled electronic excited states of various character (covalent and ionic, singly- and doubly- excited).[4],[5],[6] The determination of the most probable excited-state relaxation pathway leading to the formation of the CPD photoproduct is therefore highly sensitive to the level of theory used to describe the state-specific electron correlation. In particular, the nature of the photochemical funnel, i.e., the conical intersection responsible for the non-radiative decay back to the ground state on the way to the CPD formation, is critical in this mechanism. We will show that the well-documented three-electron/three-center bond conical intersection involving covalent electronic states and previously proposed in this system[4] is questioned based on recent theoretical calculations including dynamic electron correlation. Rather, a new detailed mechanistic picture is proposed involving a covalent / ionic crossing that has been identified and characterized for the first time.

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2

Nanoscale Heat Anisotropy in Gold Nanocrosses

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Abstract - We demonstrate ultra-fast heat transfer between the two branches of a nanocross heated by a light pulse. This transfer has been predicted with a thermo-optical numerical model and confirmed experimentally by femtosecond transient reflexion and transmission measurement.

Introduction - By absorbing and converting light into heat, nanoparticles (NPs) behave as highly localized sources of heat that could be useful for several application fields such as optical switches or nanofunctionalization. Thus, thermal dynamic and space distribution inside NPs need to be controlled to develop applications that use these nanoheaters. However, this is a challenging task due to the time and space scale involved in the photothermal process (femtosecond and nanometer scale). In this work, we predict with our numerical model [1] heat transfer inside cross-shaped gold nanoparticles and confirm it with femtosecond transient spectroscopy measurement. This NP geometry is known to support a polarization dependent localized plasmonic resonance (LSPR) along each of its branches. By selectively exciting one LSPR, we can heat only the wished branch, then, the heat will propagate along the other branch. By measuring the temporal delay between the modulation of LSPR intensity of each branch, we put in evidence the thermal inhomogeneity inside the cross-shaped NP. Good agreement was found between simulations and experiments. In addition, by fitting the experimental data with our model, we can extract the spatio-temporal electronic and lattice temperature dynamic inside the NP.

Experimental setup - The pump-probe setup is based on a Ti-Sapphire femtosecond (~75fs) laser centered at 795 nm. The signal is sent to an OPA to tune the pump frequency to a wide range of wavelengths from the UV to the IR. One fraction of the 795 nm laser source is sent through a non-linear crystal to generate the white light continuum probe.

Nanostructured sample -The sample consists of gold crosses fabricated by E-Beam lithography on a 30 nm gold film, 2 nm titanium adhesive layer and BK7 substrate. The dimensions of the cross are 300x410 nm, 60 nm height, 60 nm width and 520x340 nm periods.

Results and discussion - An illustration of the experiment is shown in figure 1.a. The pump is used to heat one branch of the cross and the probe to measure optical change of both branches. Figure 1.b shows a simulation of the resulting electronic temperature (Te) reached inside the nano cross few femtoseconds after the excitation. We can see the asymmetric electronic temperature distribution inside the cross. Figure1.c shows the measured optical response delay between the longitudinal and transverse LSPR.

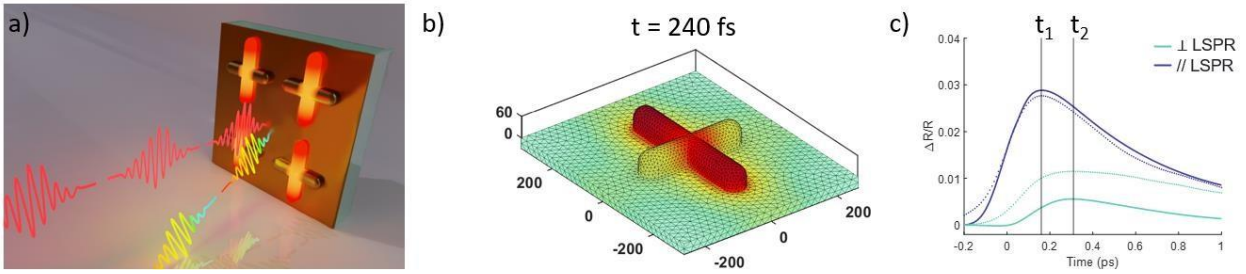

Figure 1: a) Concept of the experiment. **b)** Simulation of the repartition of Te inside a nanocross at t = 240 fs. **c)** Experimental (dots) and modelized (continuous line) transient reflectivity at the resonance wavelength of each cross axes (the longitudinal in green and transverse in blue).

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3

ESIPT in the Pyrrol Pyridine molecule: Mechanism, timescale and yield revealed using dynamics simulations

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A proton transfer reaction may be triggered upon photo-excitation of a molecule onto an electronic state characterised by a different more stable tautomer than the electronic ground state [1]. This process, dubbed as Excited State Intramolecular Proton Transfer (ESIPT), is one of the most striking photochemical mechanisms, owing to its remarkable properties, its wide range of applications, as well as its central role in various biological mechanisms and in the photostability of natural molecular systems [2-4].

Usually very fast (sub-100 fs), the characteristics of ESIPT are however quite sensitive to the properties of the system and of the involved potential energy surfaces. While static calculations may yield indirect information about ESIPT, dynamics methods on the other hand, allow the explicit simulation of the proton transfer and investigation of the characteristics of this process.

We simulated the excited state dynamics of the 2-(1H-pyrrol-2-yl)pyridine molecule (PP; see Fig. 1) using the semi-classical surface hopping method. Through these calculations, we notably highlighted the signature of two complementary mechanisms that drive the ESIPT process in the PP molecule. First, a ballistic-like process is induced by the initial geometrical relaxation of the system on the excited state, leading to an initial fast ESIPT rate. Then, we identified a secondary mechanism, involving the lingering coherent nuclear motion of the molecule in the reactant tautomer well, that becomes dominant at longer time and leads to periodic-like increases in the ESIPT probability and an overall slower reaction rate [5].

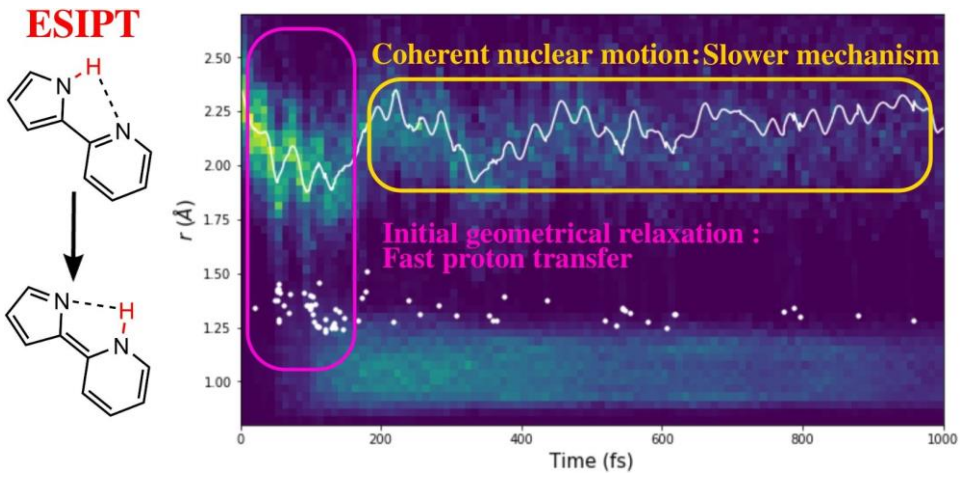

Fig 1 : Schematic representation of the ESIPT reaction in the PP molecule (left). Time evolution of the distribution of proton-acceptor nitrogen distance along the dynamics simulation (right).

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4

Photoionization dynamics of isolated tryptophane nanoparticles

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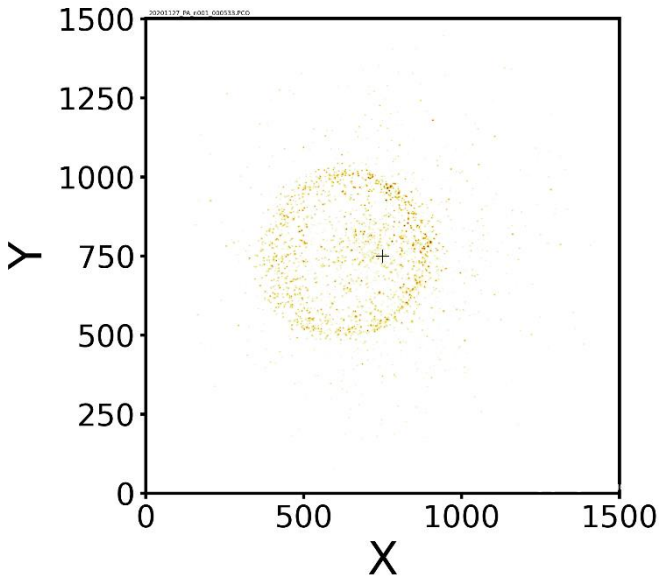
Isolated nanoparticles are little volumes of condensed mater that are introduced into vacuum. Because of the high density, they are several order of magnitude more sensitive to a laser pulse than isolated molecules. In the framework of gas phase studies, the interaction of an ultrashort femtosecond laser 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 nanoscale[1].

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, 400 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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5

Magnetic Helicoidal Dichroism with XUV Light Carrying Orbital Angular Momentum

M. Fanciulli[1, 2], M. Pancaldi[3], E. Pedersoli[3], M. Vimal[1], D. Bresteau[1], M. Luttmann[1], D. De Angelis[3], P. R. Ribic[3], B.Rösner[4], C. David[4], C. Spezzani[3], M. Manfredda[3], R. Sousa[5], I.-L. Prejbeanu[5], L. Vila[5], B. Dieny[5], G. De Ninno[3,6], F.Capotondi[3], M. Sacchi[7,8], and T. Ruchon[†,1]

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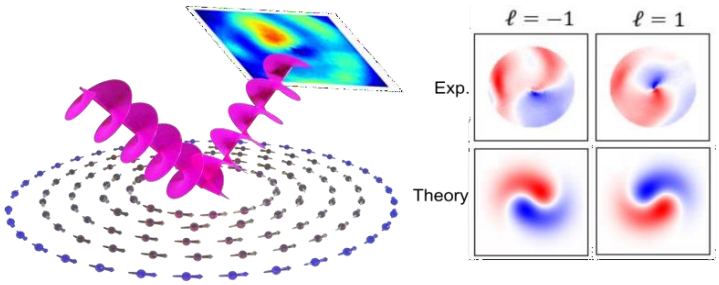
After finding many applications in the visible range, OAM beams with XUV wavelengths and ultra-short pulse duration, in the femtosecond to attosecond ranges, became available recently at high-harmonic generation (HHG) and free-electron laser (FEL) sources [1-3], widening considerably their application range. However, it has thus far found very scarce applications for magneto- optics, which rather harnesses the spin angular momentum of light to investigate and control magnetization, through the Faraday and Kerr effects.

Recently, we analysed theoretically the interaction of OAM beams with magnetic structures featuring non-uniform magnetization, in particular of XUV beams with magnetic vortices consisting of a curling in-plane magnetization [4]. We predicted that the far field intensity profile encodes the vortex symmetry in a way that depends on the sign and value of the topological charge ℓ, an effect deriving from the inhomogeneous modification of the regular reflectivity coefficients by the local magnetization. We named this effect Magnetic Helicoidal Dichroism (MHD). As for magnetic circular dichroism, MHD can be observed by inverting the sign of either the orbital momentum or of the magnetization, i.e. by switching the handedness of either the light helicoidal wavefront, or of the magnetic vortex.

In this contribution we report on the first experimental evidence of MHD (Fig. 1) obtained at the DiProI station of the FERMI free-electron laser source by measuring the scattered intensity from an Fe-Ni-alloy dot forming a magnetic vortex. The photon energy of the ~100 fs long pulses was set to 52.8 eV (~23 nm wavelength) in order to match the Fe 3p→3d core resonance, enhancing magneto- optical effects at XUV wavelengths and providing element selectivity.



The scattered intensity data, collected as a function of ℓ and of the magnetic vortex winding sense, compared well to theoretical model predictions [5]. We show also how the short pulses of the free-electron laser OAM beam make it possible to follow the evolution of the magnetization topology at the sub-ps timescale after an optical excitation.

The match between theory and experiments confirms the potential of the new toolset provided by MHD for studying complex magnetic structures and, in particular, for addressing their laser-triggered ultrafast dynamics. Understanding the interaction of OAM-light with a magnetic vortex has many potential interests. At the fundamental level, it allows to observe a new kind of dichroism, to study the role of photon spin-orbit coupling mediated by magnetization, as well as to explore new possibilities of angular momentum transfer between light and matter. In terms of applications, MHD in reflection can be exploited as a new spectroscopic tool joining the family of magnetic dichroism techniques. For their symmetry and size, magnetic vortices can be considered as an ideal benchmark sample to explore the interaction with OAM beams; moreover, given their rich dynamical response in the ultrafast domain, they are promising structure for light manipulation of the magnetization topology.

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6

Towards a high-resolution 2DIR-spectrometer

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2D infrared spectroscopy (2D-IR) is a powerful tool for investigating the structural and dynamical properties of various systems. This non-linear spectroscopy technique allows us to go beyond linear absorption spectroscopy, **and thus to simultaneously get information on both the structure (anharmonicity, mode coupling) and the dynamics (energy transfers, spectral diffusion)**, with a hundred femtosecond temporal resolution [1].

We present a 2D-IR setup that allows for the acquisition of 2D-IR spectra of molecular systems. The multipixel infrared camera we use (320x256) allows for a better spectral resolution (<0,5 cm-1) than most conventional detectors. We present here 2D spectra (see Fig. 1) and pump-probe measurements obtained on small organometallic complexes (Fe(CO)5 and W(CO)6) in solution, and that are in accordance with the literature [2,3]. By acquiring several 2D-IR spectra of Fe(CO)5 at different waiting times, we were able to follow the transfer occurring between its to vibrational modes E' and A2", which was attributed to the fluxional rearrangement of the molecule [2]. We also conducted pump-probe experiments and found similar relaxation times (~120 ps) for E' and A2", which is consistent with the fact that both modes are in equilibrium through the fluxional rearrangement that occurs on a smaller time scale (~10 ps).

This setup is versatile: once combined with a cryostat, it will be able to cover a large range of temperatures, and to probe systems either in solution or trapped in cryogenic environments. We observed interesting results for site dependence and relaxation time with a photon-echo setup for Fe(CO)5 in cryogenic matrices [4]. We thus plan to conduct 2D-IR measurements to get results that are more detailed.

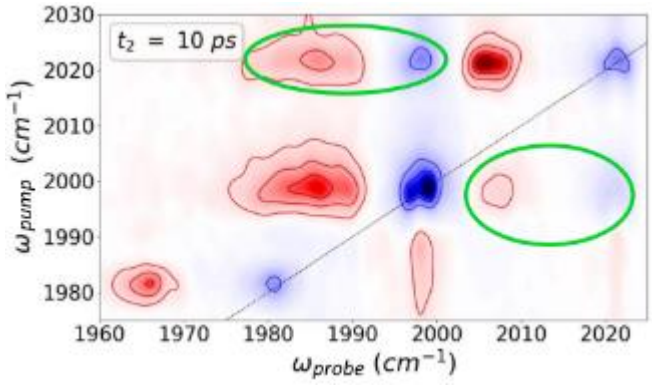

Fig. 1 : :2D-IR spectra obtained for Fe(CO)5 in dodecane for a waiting time t2 = 10 ps. The diagonal peaks doublets correspond to each individual vibrational mode. The "crosspeaks" (circled in green) indicate a transfer between the modes, caused by the exchange of CO ligands [2].

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7

Development of a high repetition rate EUV light source for short wavelength metrologies

Liu et. at

High Harmonic Generation (HHG) is a well-established technique to build compact coherent EUV light sources. My goal is to use a mid-infrared laser at 1030nm, 50fs, 200μJ, 100kHz to develop a bright EUV source using gas-filled capillaries to increase its stability.

A bright EUV light source facilitates not only fundamental research in attosecond fields, but also contributes to industrial metrologies: such as calibration of EUV wavefront sensors, inspection of the fabrication of semiconductors and lithography, etc.

8

Mise en défaut des approximations séculaire et markovienne dans la dissipation d'énergie de molécules alignées

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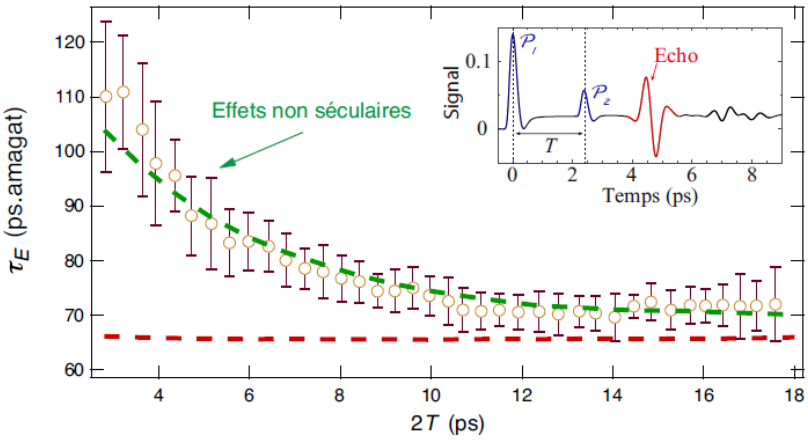
Dans les tout premiers instants qui suivent l'alignement de molécules par une impulsion laser femtoseconde, nous observons la dissipation progressive de la rotation cohérente des molécules sous l'effet des collisions intermoléculaires. Ceci nous permet de détecter et de quantifier l'influence de certains mécanismes qui affectent l'évolution du système ouvert au début du processus de thermalisation. Plus précisément, nous étudions expérimentalement et théoriquement les limites des approximations séculaire [1] et markovienne [2] généralement utilisées. La première néglige tous les échanges induits par l'interaction intermoléculaire entre les cohérences quantiques qui oscillent à des fréquences différentes. La seconde suppose que toutes les collisions sont complètes dans l'intervalle de temps d'observation et que le comportement du système peut être modélisé en supposant une chaîne d'événements de Markov instantanés et indépendants, ignorant ainsi tout effet mémoire. Nous montrons que, selon le système moléculaire considéré, l'une ou les deux de ces approximations sont mises en défaut, leur utilisation conduisant alors à des erreurs importantes sur la relaxation prédite aux temps courts avant qu'elles ne deviennent valides après quelques dizaines de picosecondes (cf Figure). Ces études démontrent l'intérêt des molécules alignées pour des mesures de dissipation quantique qui se révèlent être une alternative puissante aux études fréquentielles puisqu'elles permettent de mettre directement en évidence des comportements non séculaires et/ou non markoviens.



Comparaison entre les constantes de temps t_E de décroissance collisionnelle de l'écho d'alignement [3,4] mesurées (cercles) à différents instants et les simulations numériques du modèle séculaire (tirets rouges) et non séculaire (tirets verts). Dans le cas du mélange N2O-He présenté sur cette figure, les effets non séculaires, clairement visibles en deçà de 10 ps, moyennent à zéro aux temps plus longs (1 amagat= $2{,}68\times10^{25}$ m^{-3}). En haut à droite, écho d'alignement observé après les deux pics d'alignement P_1 et P_2 créés par deux impulsions laser décalées d'une durée T.

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9

TIGER: TIme-Gated Electric field Reconstruction

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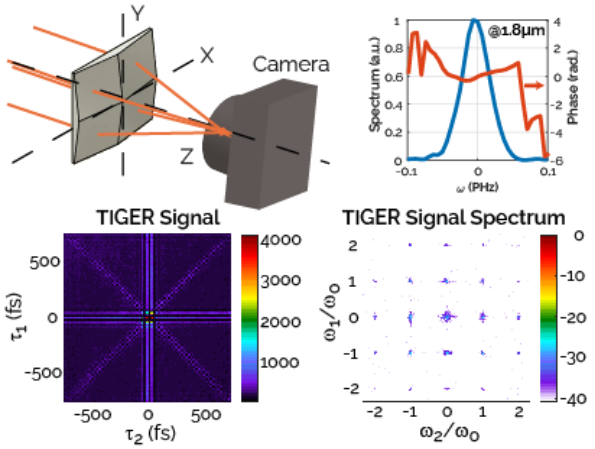
We present a novel self-referenced method for the complete temporal characterization (phase and amplitude) of ultrashort optical laser pulses. The technique, called TIme-Gated Electric field Reconstruction (TIGER) [1], measures a second-order nonlinear signal (namely, second harmonic generation) or a third-order nonlinear signal (namely, two-photon absorption) produced by four time-delayed replicas of the input pulse. The delays are spatially encoded in the beam profile using a four-faced pyramid-like optical element. The presented technique enables single shot measurement and does not require any spectral measurements, in contrast with well-known self-referenced characterization methods. Depending on the chosen geometry, the recorded TIGER signal can be either interferometric (i.e., carrier frequency resolved) or intensimetric.



SWIR TIGER : ultra-simple and compact method for complete temporal characterization. Two-photon absorption based interferometric

TIGER apparatus (top left). Raw image obtained with a CMos camera (bottom left) and its two-dimensional Fourier transform (bottom right) leading to the complete characterization of an IR pulse @1.8µm delivered by our Topas apparatus (top right).

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10

Development of high repetition rate EUV light source for short wavelength metrologies

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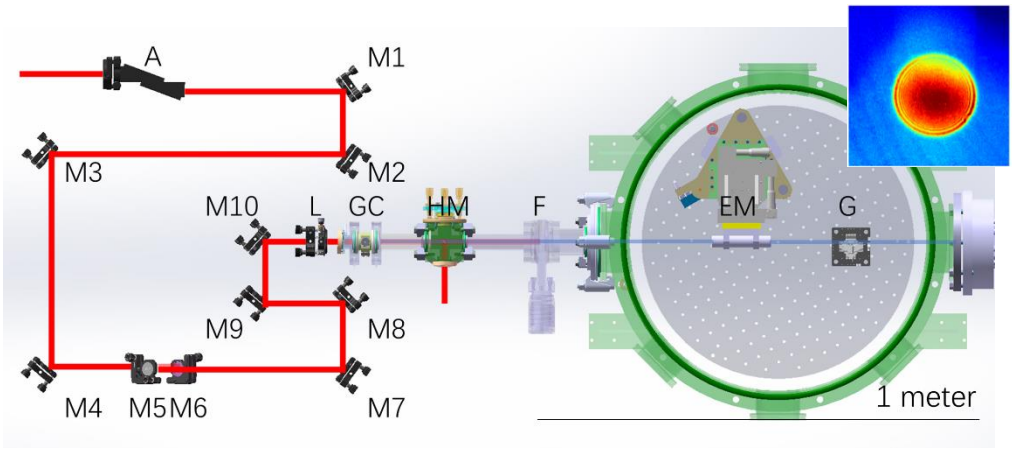
Abstract

High Harmonic Generation (HHG) in gas is a well-established technique to build compact coherent EUV light sources. The low conversion efficiency of the driving laser to harmonics makes it hard to produce a bright EUV light source. Different strategies have been developed to increase the photon flux near the cut-off of the HHG spectrum, such as gas jets, gas cells, semi-infinity gas cells, optical cavities, gas-filled capillaries, etc. Using a semi-infinity gas cell configuration, we developed a more stable and brighter EUV light source delivering 10^{11} photons/s. This EUV beamline will be used routinely for calibration of EUV wavefront sensors and to perform coherent diffraction imaging experiments such as Ptychography.



Sketch of experiment set-up. Driving laser (Amplitude Tangerine): 1030 nm, 24 W, 100 kHz, 50 fs. In inset, spatial profile of the harmonic beam.

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Etude théorique de l'intrication électron-noyaux dans un processus de photo-ionisation

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La reconstruction des dynamiques de photoémission « en temps réel » est aujourd'hui possible grâce au développement des méthodes de spectroscopies résolues en temps à l'échelle attoseconde. Dans ce cadre, des expériences interférométriques permettent de caractériser ces dynamiques en termes de "retard d'ionisation" [1] où même de remonter à la façon dont les spectres de photoélectrons se construisent dans le temps à l'échelle atomique ou moléculaire [2,**Gruson et al**]. Les dynamiques révélées nous renseignent sur le détail des intéractions corrélées entre le photo-électron et l'ion et leur influence sur le processus d'ionisation à son niveau le plus fondamental. Cependant, ces expériences supposent des processus cohérents, notamment au niveau des sources pompe/sonde mais aussi des phénomènes étudiés. *Mais cela est-il toujours le cas ?*

Nous nous intéressons ici à la photo-ionisation moléculaire où les corrélations entre le photo-électron et la dynamique vibrationnelle de l'ion sont susceptibles de créer de l'intrication [3,4] : le système total ne peut alors plus s'écrire comme le produit des deux sous système ce qui traduit une perte de cohérence. A l'aide de simulations sur une molécule modèle inspirée de H2, composée d'un électron actif et de deux noyaux, nous étudions comment cette perte de cohérence se traduit sur des observables expérimentales
[5] et in fine sur leur interprétation en termes de dynamique ultra rapide. De plus, nous étudions comment l'utilisation de paramètres extérieurs (typiquement la structure temporelle du champ ionisant) peut augmenter ou diminuer le degré d'intrication.

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12

High Harmonic Generation in Disordered Crystals

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High Harmonic Generation in crystals results from the non-linear injection of electrons into high-energy bands and their coherent oscillation in the Energy-Momentum landscape of the electronic band structure, induced by an intense femtosecond laser pulse. In real space, this reciprocates to dependence of the harmonic emission on the crystallinity and crystalline order of the solids. This dependence on the energy band/atomic arrangement can be experimentally proven by changing the relative direction of the linear polarization of the driving laser pulse [1] or by using doping to add vacancies, which leads to varying harmonic generation efficiency [2]. In this study, we rely on ion-induced damage on silicon substrate by using a Focused Ion Beam (FIB) equipped with Ga ion source. The damages were controlled using the ion doses received by the substrate which were then characterized using Raman measurements and the procedure described by Balasubramanian et.al [3]. We report a significant variation of the harmonic signal with the ion dose / structural damage in silicon single crystal. We attribute these changes in the harmonic signal to the defect structures and corresponding band structure modifications [4].

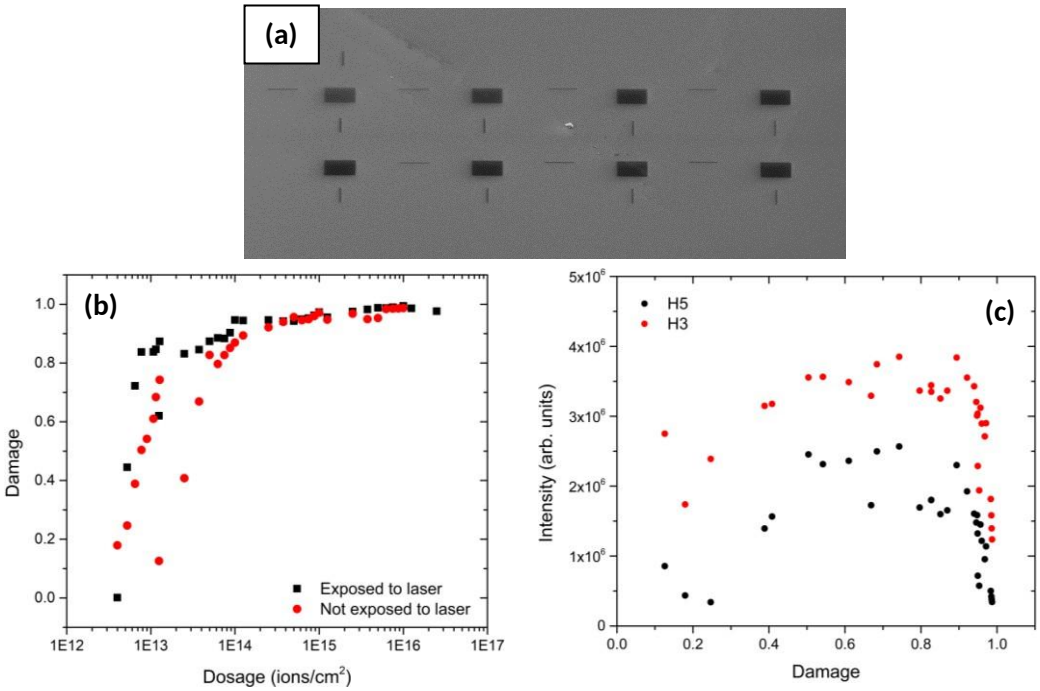

Figure (a) Ga+ ion implantation zones on silicon substrate, each implantation square corresponds to individual ion dosage. Figure (b) Correlating the ion dosage to crystal structure damage; Value of 0 corresponds to pure crystalline silicon and value of 1 corresponds to complete amorphization. Figure (c) Variation in the harmonic signal, H3 and H5 with respect to crystal structure damage.

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13

Laser-induced ultrafast demagnetization and perpendicular magnetic anisotropy reduction in a $Co_{88}Tb_{12}$ thin film with stripe domains

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Since its discovery by Beaurepaire and coworkers in 1996 [1], the phenomenon of laser induced ultrafast demagnetization has attracted world-wide attention and created an entirely new research field in magnetism named femtomagnetism [2]. Following 25 years of ongoing experimental and theoretical research, the underlying mechanisms of the rapid decrease of the magnetization of a ferromagnetic film on the femtosecond time scale after a femtosecond optical excitation remains intensively debated. The scientific challenge to explain this magnetization dynamics with its associated energy and angular momentum transfer between the electron/spin system and the crystalline lattice occurring on a sub-picosecond time scale is at the base of this strong interest.

Here we will describe a time resolved X-ray resonnant magnetic scattering (TR-XRMS) experiment on amorphous $Co_{88}Tb_{12}$ thin films with magnetic stripe domains conducted at the free-electron laser FERMI. Although several femtomagnetism studies have already been performed on ferrimagnetic Co-Tb [3], a material system of great tech- nological relevance for future all optical magnetic data storage [4], experiments describing the evolution of magnetic domain structures in rare earth–transition metal alloys following an ultrashort optical pulse are still scarce. Recently, Fan et al. performed a TR-XRMS study on $Co_{88}Tb_{12}$ samples using a tabletop high harmonics source but their analysis remained limited to the first magnetic diffraction order at the N edge of Tb (155 eV) [5].

In the present work [6], we complement their findings using different probe beam energies and deepen the analysis by using an experimental setup that allows us to record the first and third magnetic diffraction order simultaneously. With this, we are able to explicitly monitor the pump-induced evolution of the periodic magnetic structure, i.e., the change of domain size and domain wall width with the highest accuracy up to 120 ps.

We show that the average domain wall width starts to increase after about 5 ps with a time constant of a few picoseconds and remains higher for at least 120 ps. With the support of static magnetometry measurements, we argue that his increase is due to a decrease of the perpendicular anisotropy constant of our thin films. Our results thus reveal the time scale on which the magnetic domain structure is affected by a change of the magnetic anisotropy.

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14

Influence of Exact Features of Time-Dependent Kohn-Sham Potential on the Ionization Dynamics

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Knowledge of sub-femtosecond electron dynamics in many-electron systems is of utmost importance to achieve control over transport of charge carriers. This ultrafast electron dynamics in materials are often captured under excitation by high-intensity laser fields. A theoretical understanding of this dynamics, therefore, calls for solving the time-dependent Schrödinger equation (TDSE) for obtaining non-perturbative response of many interacting electrons under strong-field irradiation. 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. Although formally exact, in practice the exchange-correlation potential ($v_{\mathbf{xs}}[n](x,t)$) in TDDFT is adiabatically approximated and therefore fails to capture the non-adiabatic contribution to system's response.

In this work we have revisited the time-dependent ionization dynamics in 1D He model to precisely understand *when* and *how* the TDDFT fails to describe single-ionization. At the same time, we have implemented the inversion of the time-dependent Kohn-Sham equation in a numerical tool to obtain the Kohn-Sham potential $v_{\mathbf{xs}}[n](x,t)$ from time-dependent density $n(x,t)$. This allowed us to obtain the *exact* spatio-temporal behavior of the potential (from an *exact* time-dependent density) and thus, to obtain important features of the exact Kohn-Sham potential needed for capturing the exact ionization in TDDFT.

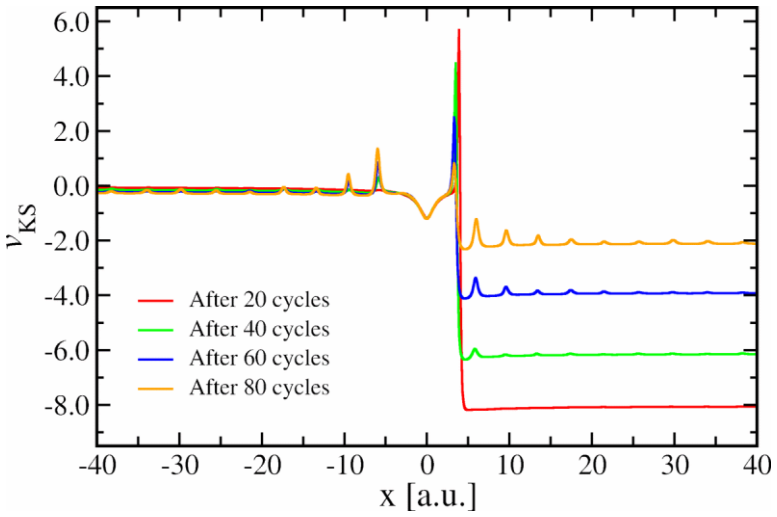

Figure: Exact time-dependent Kohn-Sham potential obtained by inverting the Kohn-Sham equation using numerically exact time-dependent density of 1D He perturbed by tens of cycles of an ionizing laser field with frequency equivalent to 1 atomic unit of energy.

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Photo-induced ring opening of cyclohexadiene into hexatriene: is dynamical correlation required for the characterization of the electronic states?

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Upon absorption of a photon to the bright excited state 1^1B, the cyclo-1,3-hexadiene is known to quickly decay to the ground state in the femtosecond timescale. It either relaxes back to the initial structure or through a ring-opening mechanism to form the hexatriene molecule following the Woodward-Hoffmann rule for photoinduced pericyclic reaction. To fully understand the complex molecular mechanism, simulating the excited state dynamics is needed.

For that purpose, a proper characterization of the potential energy surfaces of cyclohexadiene is of utmost importance. Multi-reference methods such as the complete active space self-consistent field (CASSCF) method are the most suited to describe the electronic structure along photochemical reactions. However, these are not black box methods and the choice of the active space is crucial. Here, a standard valence active space yields an incorrect ordering of the two lowest-energy excited states. Adding perturbative correction with the CASPT2 approach corrects the order of the states and gives more accurate excitation energies.[1] A balanced description can also be achieved by reducing the size of the active space at the CASSCF level at the cost of more blue-shifted vertical transitions in comparison.[2,3]

I will present the results on the effect of active space at the CASSCF level and also the effect of dynamics correlation using CASPT2 on the topology of the potential energy surfaces. The relaxation pathway is evaluated by assessing the energy gap and ordering of states starting from the ground state minimum structure (Franck-Condon point) and at the critical points such as excited state minima and conical intersections (see Figure 1). In the literature, non-adiabatic dynamics was performed with the electronic structure methods mentioned above yielding comparable qualitative results for the deactivation mechanism. However, discrepancies are reported for the lifetime of the excited states as well as the quantum yield of formation of hexatriene.

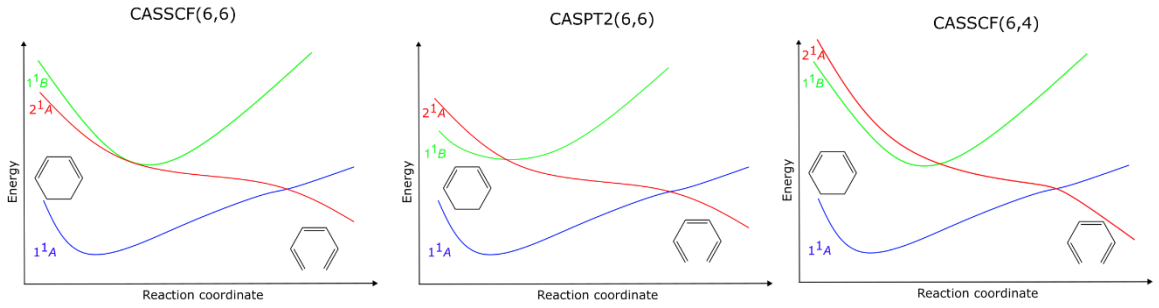

Figure 1: Schematic potential energy surface along the reaction pathway for the photo-induced ring opening of cyclohexadiene for different levels of theory using the CASSCF based method.

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[3] Kim et al., *J. Phys. Chem. A 116*, 2758 (2012)

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Isotope labelling as a tool in atto-chemistry

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Following ionization by an ultrashort extreme-ultraviolet (XUV) pulse, a polyatomic molecule is often promoted to highly excited state of the cation. The subsequent relaxation process is quite complex, with several electronic and nuclear degrees of freedom being involved at the same time. While significant advances have been made to understand similar processes in molecules resonantly excited by ultraviolet or, visible light pulses, for molecules simultaneously ionized and excited by a broadband XUV pulse, the understandings on how to disentangle different degrees of freedom in the context of atto-chemistry are limited so far [1]. Here, we show that isotope labelling can be used as an efficient tool for this purpose. Despite no difference in their electronic structure, a molecule and its isotopologue reacts differently upon an incident radiation. The observed difference in relaxation timescales between the two systems, which is supported by non-adiabatic dynamics simulations, allows us to obtain a mechanistic view of the underlying relaxation processes.

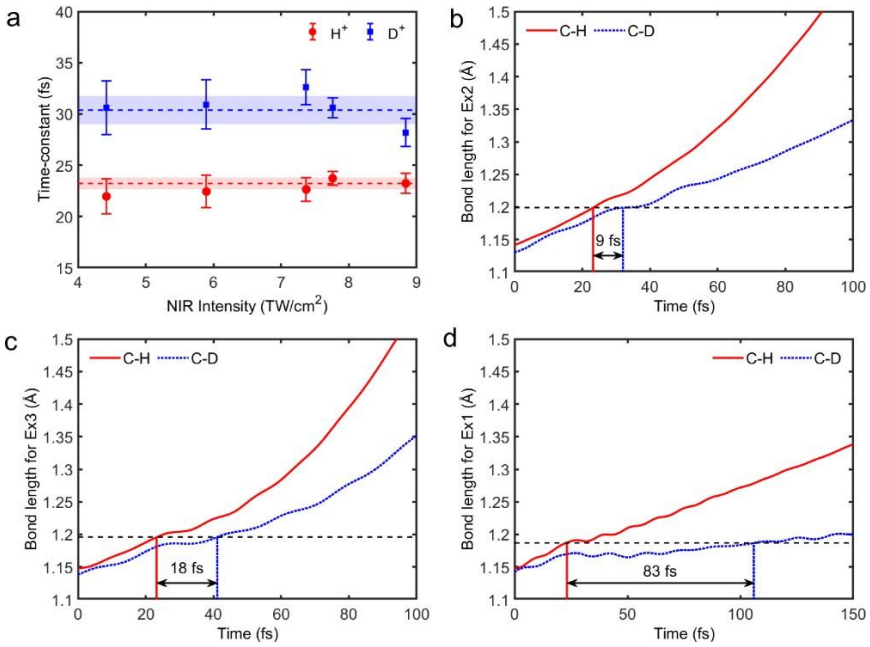

Figure 1. (a) Experimental and (b)-(d) theoretical isotope effects in C_2H_4 and C_2D_4, following ionization by XUV pulses.

Here, we used an attosecond pulse train, produced via high-order harmonic generation (HHG) process in Kr, as pump to ionize and excite ethylene molecule (C2H4) and its deuterated counter-part (C2D4). A near infrared (NIR) pulse was used to probe the subsequent relaxation dynamics. We collected the resulting ionic fragments as a function of the delay between the XUV-pump and NIR-probe. As shown in Fig. 1, by comparing the experimental isotope effect (7±2 fs) with the theoretical ones for the H^+ and D^+ fragments we could indeed identify the relevant electronic state (in this case Ex2 or, $B\ ^2A_g$) governing the relaxation process in the excited molecular cation. The isotope effect observed in case of Ex3 state ($C\ ^2B_{2u}$) matches only qualitatively with the experimental one, whereas for the Ex1 state ($A\ ^2B_{3g}$) it is an order of magnitude higher. Our experiment highlights the relevance of isotopic substitution to unravel the few-femtosecond molecular photo-dynamics induced by ultrashort XUV pulses [2].

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Femtosecond Mid-IR vibrational spectroscopy of 9,10-dicyanoantracene adsorbed on ZSM-5 zeolite

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The excited states of 9,10-dicyanoanthracene (DCA) molecules adsorbed on the external surface of ZSM-5 zeolite, have been investigated by picosecond transient emission, femtosecond transient absorption infrared vibrational spectroscopy, steady state UV-vis and quantum chemistry calculations. Following the photoexcitation at 420 nm, the formation of the localized, LE, excited S_1 state of DCA emitting bellow 500 nm is observed. LE is rapidly and quasi-exclusively converted into two distinct exciplex species, EX1 and EX2, detected by their emission above 520 nm, with a lifetime of 5 ns and 20 ns, respectively. The different transient species can be identified by the frequency of the CN stretching vibration that is a marker of the charge delocalisation and that is peaking respectively at 2162 cm^{-1} (LE), 2174 cm^{-1} (EX1) and 2187 (EX2) cm^{-1}. DFT and TD-DFT calculations further support the assignment. The results show that the external surface of zeolite is an appropriate playground for the development of novel photoactive host-guest materials by controlling the arrangement and the subsequent photochemistry of organic guest molecules.

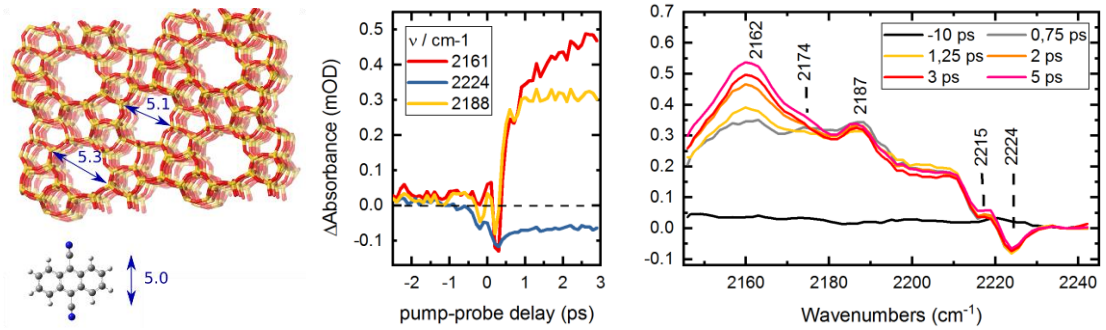

Figure: (left) Structure of 9,10-dicyanoanthracene and of the ZSM-5 zeolite framework; (right) Mid-IR transient absorption signal recorded with l_{pump} = 400 nm

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Phase matching effects in gas-based third harmonic generation for VUV source

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Femtosecond laser sources emitting in Vacuum Ultraviolet (VUV) are essential tools enabling tabletop time and Angle Resolved Photo-Emission Spectroscopy (tr-ARPES) experiments in solids [1]. Here, we report a VUV laser source based on the ninth harmonic generation of an industrial grade Yb-doped fiber amplifier delivering 500 fs-long pulses at 1030 nm and 100 kHz of repetition rate. The experimental setup is reported in Figure 1, on the left. The ninth harmonic of the 1030 nm laser beam is a result of three stages of frequency up-conversion. The fundamental beam undergoes two frequency up-conversions in BBO crystals generating a 343 nm (3ω) laser pulses. The Third Harmonic Generation (THG) process at 114 nm (10.8 eV) takes place in a xenon gas-filled cell driven by the 343 nm (9ω) laser radiation. A detection chamber composed by a spherical mirror and a grating is designed to filter out the 343 nm and detect the 114 nm radiation. The 114 nm radiation yield is extracted by measuring the photocurrent from the surface of a Tantalum target.

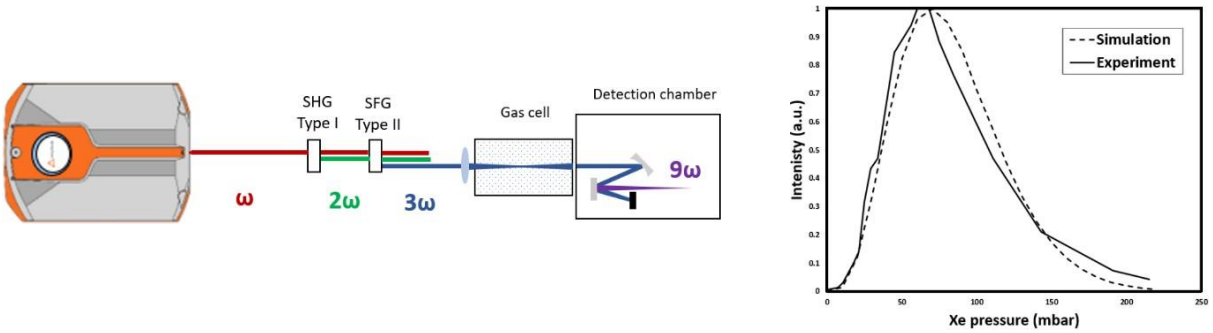

Figure 1: Left: Layout of the experiment setup. ω is 1030 nm, 2ω is 515 nm, 3ω is 343 nm, 9ω is 114 nm. Right: Measured power at 114 nm as a function of xenon pressure. Right: Simulated and experimental efficiency of generation as a function of Xe pressure.

The efficiency of the THG process strongly depends on the phase matching between the fundamental and the generated beam. The nature and the pressure of the gas, the focusing parameters and the intensity of the beam at the focal point affect the phase matching conditions [2-4]. In this study we have numerically simulated and experimentally observed the effect of these parameters on the generation process. In Figure 1, on the right, the simulated (dotted line) and the experimentally measured (solid line) power of the 114 nm beam as a function of gas pressure is reported. The measurement is realized using 30 µJ pulse energy at 343 nm, on a focused spot diameter of 25 µm corresponding to an intensity of 2.3 x 10^{13} W/cm^2.

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Ultrafast Excited State Dynamics of the Archae-Rhodopsin 3 and its mutants

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Archaerhodopsin-3 (AR-3) is a light-driven proton pump found in *Halorubrum sodomense*, which has a photo- cycle similar to that of bacterio-rhodopsin. AR-3 was reported to display a detectable fluorescence, which, when integrated in membranes of live cells, was shown to depend strongly on the transmembrane voltage. AR-3 was then put forward as a possible candidate for opto-genetic investigations, i.e. in the form of a genetically encoded voltage indicator (GEVI)[1] to track neuronal electric signals or for neural silencing. Also, multiple mutants then emerged [2], with fluorescence quantum yields (FQY) reaching up to 1.2% which is a 100-fold increase with respect to the wild-type protein (wt). To understand this exceptionally strong effect of the mutations in detail, we studied the fluorescence decay kinetics for wt as a function of pH ≤ 6, since protonation of the counter ion is known to prolong the excited state lifetime of rhodopsins [3]. Other changes in terms of the electrostatic interactions of the protein cavity containing retinal chromophore are induced in the double mutant DETC and the quintuple mutant Arch-5 [2]. The fluorescence kinetics (figure 1) are measured with 200 fs time resolution using a broadband up-conversion set-up. We find them to be best described by a sum of 3 decaying exponentials, which represent the heterogeneity of protein environment. The average excited state lifetimes reach high values up to 65 ps (figure 1). For DETC and Arch-5, the results are in agreement with the reported FQY's [3]. The excited-state lifetime increases for the different AR 3 mutants because of a potential barrier in the excited state induced by the interactions with the protein environment. Quantum chemical simulations [4] predict the existence of such barrier > $k_B T$, the height of which correlates, for different mutations, with the observed FQY. Unlike other retinal proteins, this barrier is predicted to arise from mixing of the ground and excited states.

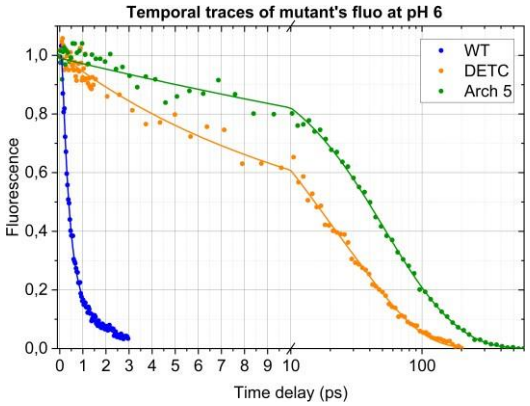

Figure 1. Fluorescence decays of AR-3, and the two mutants DETC and Arch-5, at pH6 on a semi-log scale

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Versatile Terahertz Time-Domain Spectroscopy to study ultrafast carrier dynamics in topological semimetals

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Radiations in the Terahertz range present wavelengths on the order of 100 µm, periods of 1ps, and low-energy photons (few meV). The Terahertz Time-Domain Spectroscopy (THz-TDS) technique takes advantage of the properties of these waves to probe various intrinsic modes of condensed matter systems, including their ultrafast dynamics[1] . The THz-TDS set-up we present will be used to investigate carrier and structural dynamics in Dirac and Weyl semimetals such as Cd_3As_2 and $ZrTe_5$.

This set-up relies on a 1030 nm laser beam that generates ~ 60 fs-long pulses at a tunable repetition rate between 1 kHz and 125 kHz. A split part of this beam is directed into a generation system which is based either on optical rectification nonlinear effect in an organic BNA crystal[2], or on the inverse spin Hall effect in a spintronic emitter [3]. Another part of the beam heads for the detection system which relies on Electro-Optic sampling, with either ZnTe or GaP crystals. A tunable delay is induced between the THz pulses and the detection pulses, allowing a time-resolved detection of the THz electric field.

To this THz-TDS set-up, a third optical path is added to act as a pump on the sample. Such optical/THz pump – THz probe set-up will provide information on the carrier and structural dynamics following the light excitation and the resulting relaxation processes, by probing carriers and IR-active phonons inside Dirac and Weyl semimetals, at the meV scale. This energy scale corresponds to the linear part of their dispersion band, in the vicinity of the Dirac point, which is nowadays experimentally challenging to study.

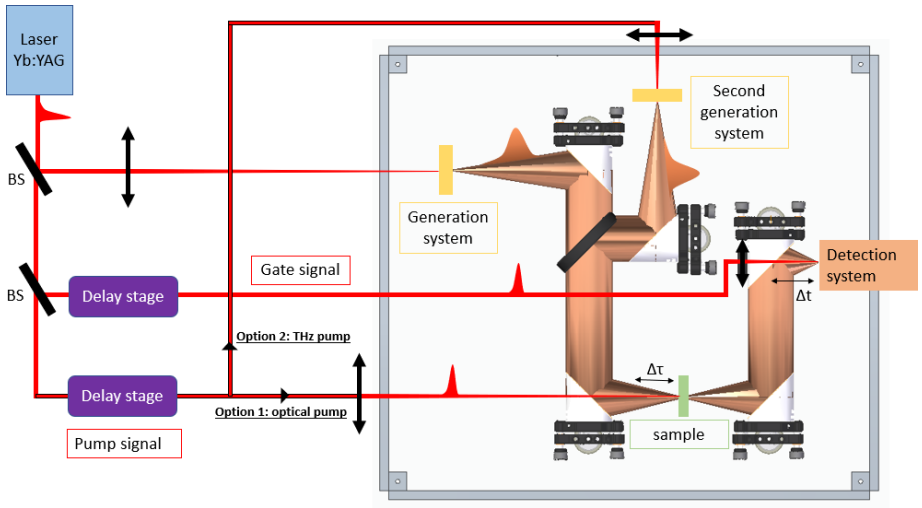

Figure 1: Scheme of the THz-TDS and optical pump configuration of our set-up.

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Pushing the Frontier in the Design of Laser-Based Electron Accelerators with Groundbreaking Mesh-Refined Particle-In-Cell Simulations on Exascale-Class Supercomputers

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Nowadays, laser-plasma acceleration accelerates electron beams to multi-GeV over a few centimeters. [1] Thanks to the development of different injection schemes (Ionization injection, shock injection…), those beams have a more and more charge or better quality. However, the principal limitation of those techniques is that they cannot provide electrons beams with both, high charge and high quality, which is necessary for different applications such as FLASH radiotherapy.[2]

The main problem is that we trap electrons from low density gas ($10^{17} - 10^{18}\ cm^{-3}$) and thus it is complicated to extract a big amount of charge. We therefore propose a new promising laser-plasma acceleration scheme which could provide high quality and high charge electron beams. This target is based on an optical device called plasma mirror to inject a substantial amount of charge. [3] The laser creates a perturbation in the electron density which create a plasma wave and electrons can be trapped in this wave and being accelerated. The considered solution is to use this plasma wave to trap and accelerate electrons from the plasma mirror.

We need to simulate this scheme to study the influence of different parameters and prepared the experimental setup. However, the scheme is numerically very challenging since it requires a high resolution during the interaction between the laser and the solid, but it also requires simulating the acceleration process over few millimeters or centimeters. To simulate this case, we need kinetic laser-plasma simulation and Particle-In-Cell simulations are essential to achieve a full understanding of this scheme. Moreover, we need to run those simulations on the biggest supercomputers in the world.

We used WarpX, an open-source Particle in Cell (PIC) code developed by Berkeley laboratory, CEA and several other institutions to address the challenge of computing at the exascale. The code provides a rich set of numerical methods and physical modules such as dynamic load-balancing, huge output and in-situ visualization, spectral solvers and mesh refinement. Mesh refinement is a crucial feature to simulate this scheme. It is a useful feature that allows the user to improve the resolution only in a certain region of the simulation. This feature is a standard feature of hydrodynamic code but is really complicated in a PIC code and can result in spurious effect or unphysical results. A work describing the implementation of mesh refinement in WarpX and the effort to simulate this scheme on several top computers has been awarded to Gordon Bell prize in 2022.[5] These simulations have informed the design of the experimental campaign that we carried out in 2022, which represents a proof-of-principle validation of our scheme.

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[5] **Pushing the Frontier in the Design of Laser-Based Electron Accelerators with Groundbreaking Mesh-Refined Particle-In-Cell Simulations on Exascale-Class Supercomputers**, Luca Fedeli, Axel Huebl, France Boillod-Cerneux, Thomas Clark et al., SC22 [2022]

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Polarization spectroscopy of high-order harmonic generation in gallium arsenide

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An interesting property of high harmonic generation in solids is its laser polarization dependent nature which in turn provides information about the crystal and band structure of the generation medium. We report on the linear polarization dependence of high-order harmonic generation from a gallium arsenide crystal. Interestingly, we observe a significant evolution of the anisotropic response of above bandgap harmonics as a function of the laser intensity. We confirm the change to originate from fundamental microscopic effects of the emission process that switch the dominant contribution between intraband and interband dynamics [1]. This intensity dependence of the anisotropic nature of the generation process offers the possibility to drive and control the electron current along preferred directions of the crystal, and could serve as a switching technique in an integrated all-solid-state petahertz optoelectronic device.

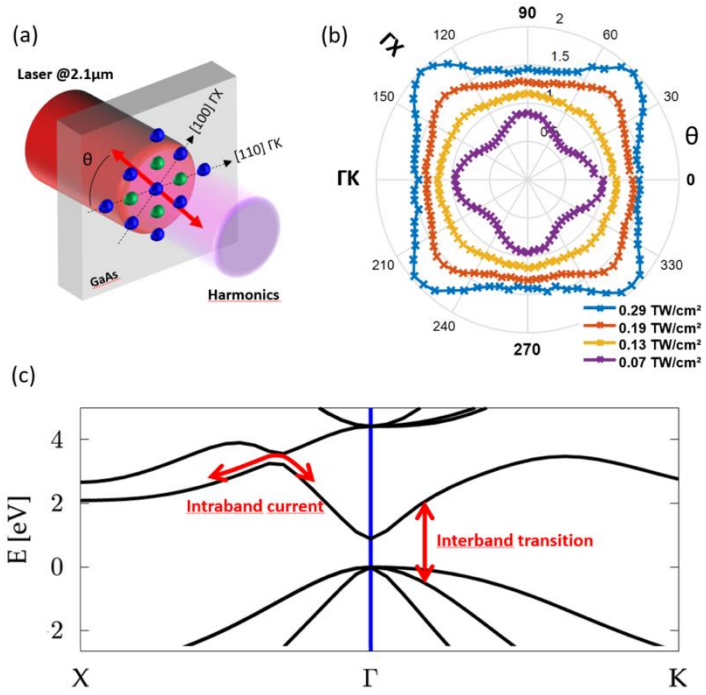

Figure (a): Schematic representation of the geometry of the experiment, including the relative orientations of the GaAs crystal axes (ΓX and ΓK) with the laser polarization (red arrow). **Figure (b):** Anisotropy map of the harmonics 5 yield dependent of the crystal orientation vs. laser polarization at different laser intensities. **Figure (c):** Band structure of GaAs along ΓK and ΓX obtained by density-functional theory calculations. At large laser intensities, the electrons reach a significant inflection of the first conduction band at ΓX that induces an intraband current and emission for the harmonic 5 that dominates the interband emission from the transition between the light hole and first conduction band at ΓK.

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Amélioration du degré d'alignement de la molécule d'acétone par écho rotationnel

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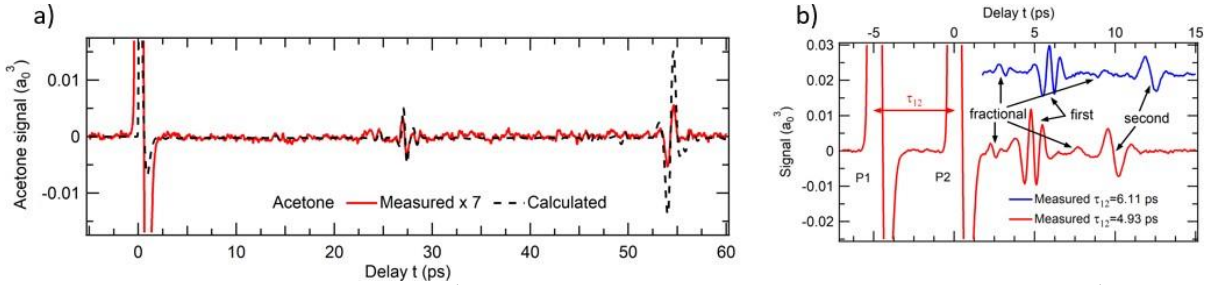
L'alignement de molécules par impulsions laser ultra brèves est un phénomène connu et largement étudié depuis ces vingt dernières années qui a permis de mieux comprendre les dynamiques rotationnelles de nombreux systèmes moléculaires. Si la plupart de ces études se sont penchées sur les molécules linéaires ou toupies symétriques, leur structure permettant un degré d'alignement important, très peu de travaux ont été réalisés sur des molécules toupies asymétriques. Naturellement plus présentes dans notre environnement et impliquées dans de nombreux phénomènes, comprendre la dynamique de ces molécules pourrait s'avérer utile à des domaines tels que la chimie, la biologie ou encore la médecine. Notre étude s'est donc portée sur la molécule d'acétone qui est une molécules très asymétrique (son paramètre de Ray □ vaut 0.37) et difficile à aligner. Cela se traduit par des transitoires d'alignement d'amplitude relativement faible (cf Figure a) pouvant être expliqué par plusieurs raisons. D'une part, la faible périodicité du spectre rotationnel associé à l'asymétrie conduit à des transitoires faibles qui disparaissent rapidement au cours du temps. D'autre part, l'acétone est soumise à une relaxation collisionnelle importante, conséquence d'une forte interaction dipôle-dipôle entre les molécules. Enfin, l'acétone est partiellement détruite par une photodissociation induite par une absorption à trois photons à 800 nm. Afin de réduire l'impact des deux premiers effets, et ainsi augmenter le degré d'alignement, on utilise les échos rotationnels afin d'aligner la molécule à des temps plus courts que ceux fixés par la période rotationnelle [1,2]. En effet, l'amplitude de l'écho observé est deux fois plus importante que celle associée au transitoire d'alignement le plus important (cf Figure b). On observe également pour la première fois dans ce type de molécule, par des mesures de $\langle\cos^2$□ z_i□□□ où z est la direction de polarisation du champ d'alignement et i les axes de la molécule), des échos fractionnaires attribués à la structure non linéaire de l'acétone [3]. Ces échos fractionnaires peuvent s'avérer utiles pour déterminer la nature linéaire ou non d'un système moléculaire sans autre étude préalable.

a) Signal d'alignement de l'acétone enregistré à 0.15 bar à température ambiante et simulation quantique correspondante. **b)** Echo



rotationnel observé dans l'acétone pour les mêmes conditions expérimentales. Les deux graphs étant à la même échelle on constate que l'alignement par échos est deux fois plus important que l'alignement associé au plus grand transitoire.

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Air-photonics broadband terahertz generation and detection

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We introduce a terahertz (THz) platform relying on two-color air plasma generation [1] and subsequent air biased coherent detection (ABCD) [2] of the THz field. The use of air as generation and detection medium allows for the production gapless broadband terahertz spectra with an electric field strength in the MV/cm range. In this work, both source and detection system are investigated experimentally and numerically, yielding excellent agreement of measured and simulated signals. We reveal that it is crucial to model the whole optical setup and include the various pump distortions, like temporal and spatial walk-off as well as ellipticity of polarization, in order to interpret the experimental measurements correctly.

Moreover, it turns out that geometrical effects in the ABCD scheme shape the recorded spectra and need to be taken into account. We confirm that THz electric field with peak amplitude in the MV/cm range and large spectral bandwidth are produced, allowing us to demonstrate THz spectroscopy with molecular samples.

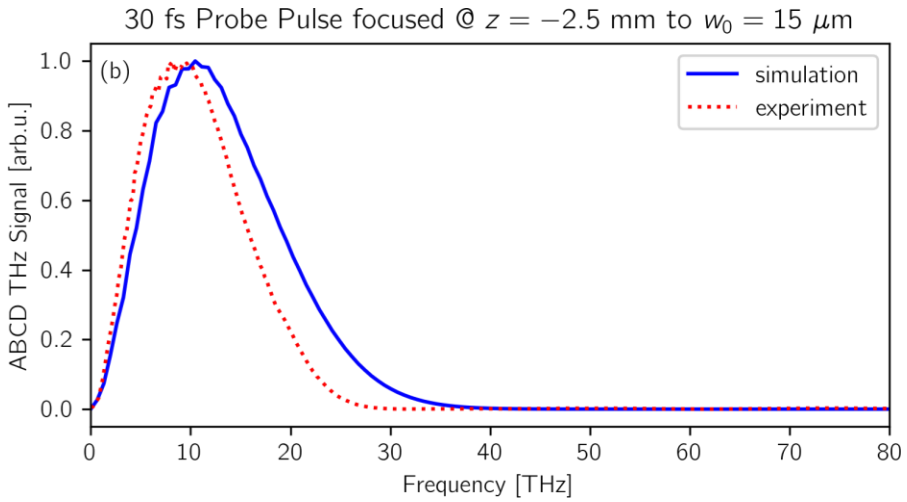

Figure: Experimental (red) and simulated (blue) ABCD spectrum. The simulations involved a 30 fs probe pulse focused to a 15 µm waist.

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Optomechanical Energy Conversion through a conical Intersection in Biomimetic Motors

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The interest to design light responsive molecular biomimetic motors towards the control of both living cells and synthetic devices is the main focus of optogenetics [1]. Ultrafast C=C double bond photoisomerization is used in molecular motors for optomechanical energy conversion at the molecular level. Efficient molecular motors should have a high photoisomerization quantum yield, and a unidirectional rotation around the C=C double bond. [2] The latter property requires point and helical chirality, see Figure 1A. We apply UV-Vis transient absorption spectroscopy to explore the photoreaction dynamics and efficiency of various photoisomerizing compounds inspired from the GFP chromophore (Figure 1A) or the PYP chromophore (Figure 1B) [1].

Here, we investigate in particular the influence of steric hindrance exerted either by (i) a molecule substitution (e.g. methyl group introducing helical chirality in fig .1A) or by (ii) the molecule environment (e.g. insertion in a protein binding pocket), on the photoreaction dynamics.

Figure 1: A compound 1: derivative of the Green Fluorescent protein (GFP) chromophore aiming to develop fully unidirectional biomimetic *p*-HBDI motors. **B**: compound 2: Xanthopsin like system, derivative of the PYP chromophore

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Femtosecond dynamics of colloidal mercury chalcogenide quantum dots

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We present in that poster a study of the femtosecond dynamics of colloidal mercury telluride quantum dots. They are very good candidates for the production of infrared LED which could fill the technological gap of light sources between 2 and 4 μm [1]. In order to understand what processes limit the efficiency of such a device, it is necessary to characterize the behavior of carriers. In this study, time-resolved reflectivity measurements have been performed and coupled with a simple model. This allowed us to highlight the different behavior of the electrons and the holes at the sub-picosecond timescale.

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Measuring two-photon transition delays using 'self-probing' of a resonance

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Synopsis We have developed an original interferometric technique that allows measuring the transition delay associated with a two-photon transition through a bound state with both high accuracy and spectral resolution.

The ionization dynamics in the vicinity of resonances are specifically interesting as they reveal the electronic behavior at energies particularly relevant for the quantum system. They have been studied using interferometric schemes such as the RABBIT technique where measurements of the spectral phase evolution allowed reconstructing the corresponding rich dynamics.

In the present study, we develop an alternative interferometric scheme to access the spectral phase of resonant transition amplitudes. It relies on reference single-photon transitions (rather than reference two-photon transitions as in RABBIT-like approaches) leading to a number of advantages. This is demonstrated in the case of the two-photon XUV+IR ionization of helium through the intermediate resonant state 1s3p.

We use a single harmonic beam (H15) overlapped with its generating IR field to induce two-photon transition towards the continuum, creating the free electron wavepacket (EWP) of interest. The latter is structured by the transient resonance with the intermediate 1s3p state that leads to a rad spectral phase jump [1,2]. The 1s3p state is also coherently populated through a one-photon transition. After a variable delay , an additional IR field transfers some population from this state to the continuum, again by a one-photon transition, creating a reference EWP centered at the same energy. In contrast to the resonant two-photon transition, the sequential one-photon transitions produce an EWP with a smooth phase evolution providing an ideal reference. The interferences between the probed and reference EWPs encode their relative phase that has two contributions: i) the difference of phases accumulated during the delay by the 1s3p and the continuum states, and ii) the phase of the resonant two-photon transition. By measuring the -dependent photoelectron spectrum over hundreds of femtoseconds, we can access the latter phase with both high accuracy and high spectral resolution. A further advantage of this interferometric scheme is that the reference EWP has a similar partial wave decomposition as the studied EWP, which maximizes the contrast of the interferences regardless of the electron emission direction.

The spectrogram corresponding to the P_0 Legendre polynomial of the Abel inversion of the VMI images is shown in Fig. 1 (very similar spectrograms are obtained for P_2 and P_4). The clear difference in slope (inset) corresponds to a linear spectral phase of the probed two-photon transition around the resonance. It encodes the transition delay due to the transient trapping of the electron in the intermediate bound state before completing the transition.

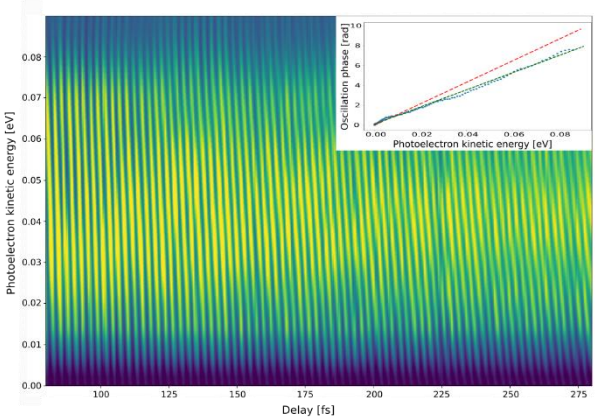

Figure 3. Photoelectron spectrogram showing highly contrasted interferences between the probed and reference EWPs. The inset shows the phase at the central oscillation frequency for each photoelectron energy (blue dots) compared to the simulated phases resulting purely from the accumulation delay (red dashed) and from the total calculation (green dashed).

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Time resolved studies of relaxation dynamics in wide band gap oxides

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The interaction of intense light pulses with transparent materials is a domain of research with a long history which is however more active than ever, and this revival concerns both applied science and fundamental research. In this work our aim is to explore the relaxation dynamics in wide band gap dielectrics, namely SiO_2 and Al_2O_3.

For this purpose we have built at LOA an experimental set-up which allows to measure simultaneously the change in transmission and reflectivity, induced by a pump pulse. This exciting pulse is the second harmonic of a Ti-Sa laser which delivers 40 fs pulse at a repetition rate of 1 KHz, and the probe pulse is the fundamental at 800 nm.

In this poster, we wish to present the first set of results. In SiO_2, we observe different mechanism depending on the pump intensity: first Kerr effect, followed by the coherent excitation of two different phonon modes, and finally photoexcitation of carriers in the conduction band, which is followed by a fast trapping of electron hole pairs.

In Al_2O_3, we measure a much longer lifetime of excited carriers, in the range of tens of picosecond, showing that no self-trapping mechanism is occurring in this material. We have also studied the variation of both signal as a function of the orientation of the crystalline sample. Surprisingly a strong modulation is observed in the reflectivity signal, while the transmission is only weakly influenced by the orientation of the crystal. We give a possible interpretation of this unexpected behavior, in terms of surface phonons.

This first set of experiment will be completed by using different probe wavelength, in the IR domain, and also in the VUV in the Attolab facility at CEA- LIDYL

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