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UP Ultrafast Phenomena

Réunion plénière du GDR UP - 2025

13 et 14 mars 2025

Cité internationale universitaire de Paris

Recueil des contributions

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Le GDR U.P. est consacré aux phénomènes ultrarapides. Il a été officiellement créé le 1er janvier 2016 pour une durée de 5 ans, renouvelé le 1er 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 huitième réunion plénière du GDR qui se tient les 13 et 14 mars 2025 à la Cité Internationale Universitaire de Paris est une occasion importante de rapprochement entre les équipes françaises et une démonstration de l'émulation scientifique qui caractérise notre communauté.

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

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Fabien Vialla (ILM, Lyon),
Sarah Houver (MPQ, Paris).

Jeudi 13 Mars 2025

09h00-10h00	*ACCUEIL / CAFE*	
10h00-10h30	*Bureau du GDR U.P*	*Introduction / actions du GDR UP*

Session1: ULTRAFAST OPTICS (Chair: David Bresteau)		
10h30-10h50	*François Balembois (LCF/SourceLab)*	*LED pumped alexandrite: potential for a new technology of femtosecond laser systems*
10h50-11h10	*Mikhneva Anastasiia (CORIA Rouen)*	*Extreme ultraviolet source based on high harmonic generation in solids with a high-energy fiber-based 1550 nm driving laser*
11h10-11h30	*Edouard Hertz (ICB Dijon)*	*Generation of ultrashort harmonic pulses from solid-state media: broadband PI-FROST characterization and driving mechanisms*
11h30-11h50	*Amadou Diallo (CELIA Bordeaux)*	*Ultrashort VUV pulse generation with orbital angular momentum transfer*
11h50-12h10	*Antoine Dubrouil (Femtoeasy)*	*Ultrafast characterization and more*
12h10-12h30	*Olivier Zabiolle Mingming PAN (Amplitude)*	*Advances in ultrafast laser systems and secondary sources at Amplitude*
12h30-14h00	*REPAS / EXPOSANTS*	

Session 2: ATTOSECOND SCIENCE (Chair: Saikat Nandi)		
14h00-14h20	*Victor Despré (ILM Lyon)*	*Correlation-Driven Charge Migration Triggered by Infrared Multi-Photon Ionization*
14h20-14h40	*Thierry Tran (CEISAM Nantes)*	*Can classical trajectory dynamics methods accurately simulate attochemistry ?*
14h40-15h00	*Rafael Menezes Ferreira (LIDYL Saclay)*	*Probing iodine chemical environment with ionization delays*
15h00-15h20	*Léonardo Rico (LCPMR Paris)*	*Rabi oscillations, photo-emission and entanglement: A time-resolved picture*
15h20-15h40	*Gabriel Granveau (LCF Palaiseau)*	*Reconstruction of the attosecond dynamics of unobserved ions*
15h40-16h10	*PAUSE CAFÉ / EXPOSANTS*	

Session 3: MATERIALS I (Chair: Elsa Cassette)		
16h10-16h30	*Emilie Herault (Croma Savoie)*	*Génération par autocorrélation de signaux THz et de second harmonique à la surface de cristaux diélectriques*
16h30-16h50	*Ernest Pastor (IPR Rennes)*	*Controlling the excited-state lifetime of transition metal oxide photocatalyst*
16h50-17h10	*Elodie Iglesis (LMPQ Paris)*	*Optical pump-induced carrier dynamics in InSb: probing the plasma frequency evolution*
17h10-17h30	*Thomas GAUTHIER (IPR Rennes)*	*Ultrafast photo-induced dynamics triggered by electron transfer in 1d van der Waals heterostructure*
17h30-21h00	*Session Posters / Cocktail*	

Vendredi 14 mars 2025

Session 4: FEMTOCHEMISTRY AND FEMTOBIOLOGY (Chair: Jérémie Léonard)		
9h00-9h20	Josene Toldo (LCH-ENS Lyon)	Modeling excited states dynamics using surface hopping
9h20-9h40	Michel Sliwa (LOB Palaiseau)	Toward the design of fast red reversible photo-switchable fluorescent proteins using multi-timescale transient absorption spectroscopy
9h40-10h00	Sebastien Weber (CEMES Toulouse)	Données FAIR: acquisition, partage et reproductibilité des résultats
10H00-10h30	PAUSE CAFÉ / EXPOSANTS	

Session 5: SECONDARY SOURCES (Chair: Stefan Haessler)		
10h30-10h50	Eléonore Roussel (PHLAM Lille)	Single-shot electro-optic detection of THz electric field with high temporal resolution and MHz acquisition rate
10h50-11h10	François Courvoisier (Femto-ST Besançon)	Ultrafast micro-physics of Bessel beam interaction with solid dielectrics
11h10-11h30	Joséphine MONZAC (LOA Palaiseau)	Effets d'ionisation et impact de l'hydrogène sur les performances d'un accélérateur laser-plasma kHz
11h30-11h50	Marie-Hélène CARRON (IJCLab Orsay)	Amplification of beams carrying Orbital Angular Momentum in a plasma-based XUV laser: a numerical study
11h50-12h10	Titouan Gadeyne (LIDYL Saclay)	Photon pathways in the nonperturbative nonlinear regime of high harmonic generation
12h10-12h30	Simon Reiger (UFI/Optonlaser)	Commercial table-top beamline for attosecond science
12h30-14h00	REPAS / EXPOSANTS	

Session 6: GAS PHASE SYSTEMS (Chair: Lou Barreau)		
14h00-14h20	Lea Ibele (ICR Marseille)	Computational photochemistry to simulate time-resolved experimental observables
14h20-14h40	Gildas Goldsztejn (ISMO Orsay)	A kHz laser desorption scheme adapted to ultrafast gas-phase measurements of thermolabile molecules
14h40-15h00	Rajarshi Sinha-Roy (ILM Lyon)	Light-induced orbital magnetism in atomically precise metal clusters
15h00-15h20	Ali Aras (IJCLab Orsay)	High-Frequency Phase Noise Suppression for the DeLLight Interferometer to Measure the Optical Nonlinearity in Vacuum
15h20-15h50	Pause café / EXPOSANTS	

Session 7 : MATERIALS II (Chair : Christine Richter)		
15h50-16h10	David Le Bolloc'h (LPS Orsay)	Le glissement d'une Onde de densité de charge observé par une source XFEL
16h10-16h30	Akib Jabed (CELIA Bordeaux)	Unveiling charge dynamics in 3D topological insulator via time- and angle-resolved photoemission spectroscopy
16h30-16h50	Romain CAZALI (LIDYL Saclay)	Correlations drive the attosecond response of strongly-correlated insulators
16h50-17h15	Bureau du GDR UP	Conclusion et Perspectives

SESSION 1

Ultrafast Optics

LED pumped alexandrite: potential for a new technology of femtosecond laser systems

François Balembois[1], Catherine Le Blanc[1], Elio Thellier[1], Frédéric Druon[1], Patrick Georges[1], François Sylla[2]

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Alexandrite (Cr^{3+}:$BeAl_2O_4$) is a laser crystal that has been known for over 50 years and is used in many commercial dermatology lasers. It has a broad near-infrared emission band (50 nm full width at half maximum, centered at 750 nm), excellent thermo-mechanical parameters and the unique property of being more efficient at 80°C than at room temperature. Its long fluorescence lifetime (250 µs at room temperature) allows energy storage with interesting potential for pulse amplifiers. Surprisingly, despite its great potential, alexandrite has never achieved a breakthrough in the field of femtosecond lasers. Two generations of femtosecond lasers later (Ti:sapphire and ytterbium lasers), it is interesting to ask why. In fact, the main problem with alexandrite lies in the pump source: it can be pumped by flashlamps, which are not adapted for high repetition rates, or by red laser diodes, which are very expensive and far less developed than the 800-980 nm laser diodes used for Nd:lasers and Yb:lasers. The absorption band of alexandrite is between 550 nm and 650 nm, right in the "yellow gap" where semiconductors remain inefficient today.

We propose a novel pump source consisting of a Ce:YAG luminescent concentrator pumped by blue LEDs. The emission of this crystal perfectly matches the absorption band of alexandrite. In addition, the geometry of the luminescent concentrator (composed of a parallelepiped with large faces) allows a massive collection of LED beams for pumping [1]. This type of source can produce more than 3 kW peak power over 400 µs pulses [2]. A single-pass small-signal gain of 1.38 has recently been demonstrated, leading to a gain of 35 in a 16-pass multi-pass amplifier [3]. This preliminary demonstration paves the way for femtosecond regenerative and multipass amplifiers using LED-pumped alexandrite.

Given the simplicity and robustness of this new pumping technology, we believe that LED-pumped alexandrite could lead to a new generation of femtosecond laser systems, capable of efficiently driving compact laser plasma accelerators at high repetition rates.

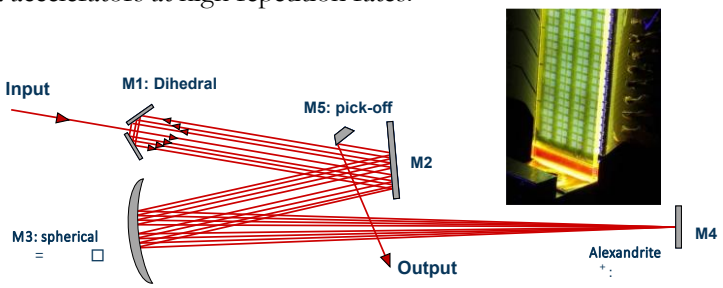

Figure 1 : example of a LED pumped multipass amplifier with alexandrite. Insert : example of a LED pumped Ce:YAG luminescent concentrator (yellow) bonded on a alexandrite crystal (red). LEDs are visible by transparency through the Ce:YAG concentrator.

Funding : ANR ASTRID-Maturation 2024 projet TERALEDx

References:

[1] Pichon, P., Barbet, A., Blanchot, J. P., Druon, F., Balembois, F., & Georges, P. (2017). LED-pumped alexandrite laser oscillator and amplifier. Optics Letters, 42(20), 4191-4194.

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[3] Thellier, E., Taleb, H., Druon, F., Le Blanc, C., Pichon, P., Castaing M., Georges, P., & Balembois, F. LED pumped alexandrite multipass amplifier designed for acousto-optic imaging, submitted to Optics Letters (2025)

Extreme ultraviolet source based on high harmonic generation in solids with a high-energy fiber-based 1550 nm driving laser

Anastasiia Mikhneva[1, 2], Djamila Boukhaoui[1], Said Idlahcen[2], Jonathan Houard[1], Ivan Blum[1], Thomas Godin[2], Leo Guiramand[2], Foued Amrani[3], Frédéric Gérôme[3,4], Fetah Benabid[3,4], David Gauthier[5, 6], Hamed Merdji[7], Willem Boutu[5, 6], Angela Vella[1], Ammar Hideur[2]

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High harmonic generation (HHG) is a well-established process used to produce light in the extreme ultraviolet (EUV) range. EUV sources hold significant potential for applications in ultrafast spectroscopy, lithography, and atom probe tomography. High harmonics of the driving laser frequency can be generated by tightly focusing a high-energy laser into a nonlinear medium, such as a gas, solid, or plasma. Historically, HHG has been primarily driven by lasers with wavelengths around 800 nm. However, mid-infrared (mid-IR) lasers have gained increasing interest due to their ability to extend the harmonic cutoff, enabled by the quadratic wavelength scaling of the ponderomotive energy [1]. Additionally, mid-IR drivers offer improved damage thresholds, making them attractive for high-intensity applications.

Optical Parametric Chirped-Pulse Amplification (OPCPA) and Optical Parametric Amplifier (OPA) systems have proven effective in achieving these results, but their inherent complexity highlights the need for more compact, efficient, and scalable EUV generation techniques [1, 2, 3]. In this context, ultrafast mid-IR fiber lasers have emerged as a promising alternative for HHG. However, their pulse energies remain limited to the nanojoule range, posing a challenge for generating high-flux EUV radiation [4].

In this work, we demonstrate high-order harmonic generation in bulk crystals using a high-energy fiber laser system operating near 1550 nm. The system delivers few-cycle pulses with durations below 50 fs, microjoule-level pulse energies, and a repetition rate of 660 kHz. This is achieved by combining a high-energy chirped-pulse amplifier featuring erbium-doped fibers, initially producing 600 fs pulses, with a post-compression stage based on an Argon-filled hollow-core photonic crystal fiber (HC-PCF) [5, 6].

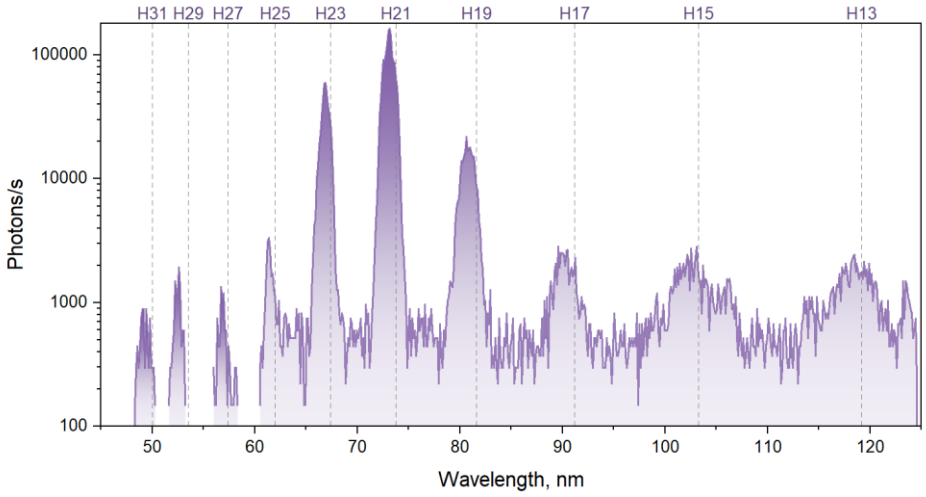

FIGURE 1. EUV HHG in a 200 µm thick MgO crystal

The interplay between anomalous dispersion and Kerr nonlinearity along the HC-PCF enables the generation of fundamental solitons with more than 18 Megawatts peak power and sub-50 fs duration. The compressed pulses are then focused into an MgO crystal to generate EUV high-order harmonics. The

EUV spectra were measured using a home-built spectrometer and analyzed as a function of the crystal orientation relative to the laser polarization, and the driving laser intensity. As a result, harmonic orders up to the 31st were detected, with a total photon flux reaching 5×10^6 photons per second.
To conclude, we demonstrated the successful use of the high-energy fiber laser platform with a HC-PCF-based post compression stage as a driving laser for efficient HHG-based EUV source.

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Generation of ultrashort harmonic pulses from solid-state media: broadband PI-FROST characterization and driving mechanisms

P. Béjot[1], B. Kiss[2], R. Shrestha[2], L. Àbrók[2], Z. Kis[2], K. Pirisi[2], B. Bagó[2], O. Faucher[1], F. Billard[1], E. Cormier[2,3], E. Hertz[1]

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We report on the detailed characterization of ultrashort harmonic pulses produced in a solid-state target by a few-cycle driving pulse (3.2 μm, 100 kHz). In this experiment, conducted at ELI-ALPS in the frame of the "3rd Joint ELI Call for Users", harmonic fields, generated in a ZnO crystal, are fully characterized by the newly developed Plasma-Induced Frequency Resolved Optical Switching (PI-FROSt) method [1,2]. This approach is straightforward to implement, free from phase-matching constraints, capable of operating at ultra-high repetition rates, with no damage threshold. We demonstrate the ability of PI-FROSt to characterize, without any reconfiguration of the setup, the MIR driving field with all (odd and even) harmonics up to the fifth order. The resulting spectrum spans a remakable bandwidth of 2.6 octaves across the visible – mid-infrared spectral region (0.59–3.6 μm) and all assessments confirm PI-FROSt as a highly effective metrology tool for unconventional secondary sources. Beyond demonstrating the metrology capabilities, the comprehensive field characterization provides valuable insights into the mechanisms driving harmonic generation. Notably, our measurements highlight the consistent production of bi-pulses across all harmonics corroborated by numerical simulations. This effect stems from the combined contributions of large phase and group velocity mismatches, emphasizing the limitation of "point-model responses" to capture the key features of harmonics and the critical importance of propagation and cascading effects in the harmonic build-up.

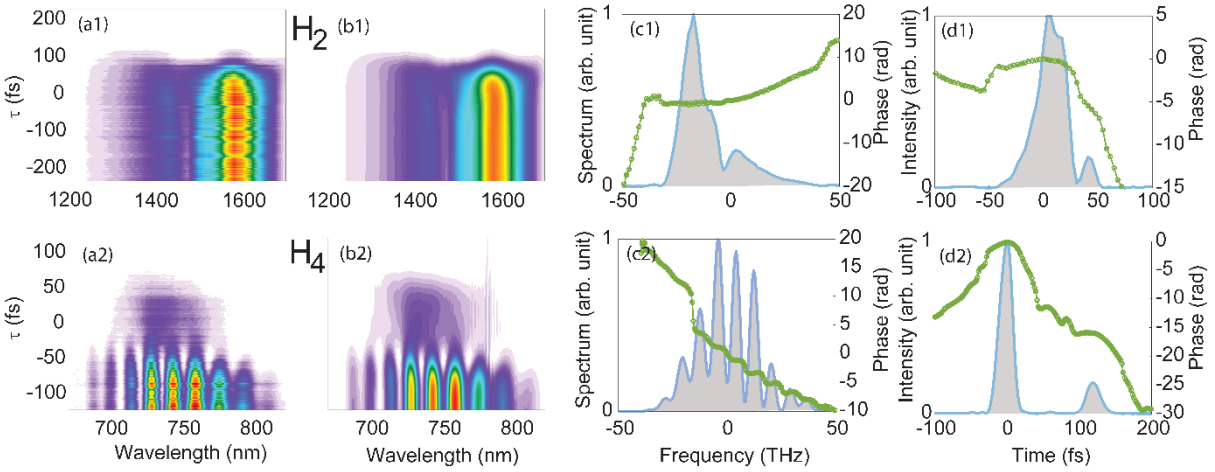

Figure caption: PI-FROSt characterization (a) experimental spectrogram, (b) retrieved spectrogram, (c) reconstructed spectrum and (d) reconstructed temporal profile. The first and second line corresponds to the 2nd and 4th harmonic, respectively.

References:

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Ultrashort VUV pulse generation with orbital angular momentum transfer

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The generation of bright, ultrashort (< 2 fs) vacuum ultraviolet (VUV) and deep ultraviolet (DUV) pulses is essential for time-resolved spectroscopy due to their strong interaction with atoms and molecules. Travers et al. [1] experimentally demonstrated the possibilities of generating tunable VUV-DUV pulses (from 122 nm to 350 nm) in a gas-filled hollow core fiber with µJ energy and sub-10fs pulse duration. Very recently, M. Vimal et al. [2] and M. Guer et al. [3] reported Orbital Angular Momentum (OAM) preservation in the pulse-compression technique based on a gas-filled capillary. In this theoretical study, we demonstrate that the OAM of an initial laser pulse can be efficiently transferred and preserved in the VUV-DUV dispersive waves generated through soliton dynamics. Using a home-made unidirectional pulse propagation equation (UPPE) code that includes nonlinear effects such as the Kerr effect, plasma dispersion, and ionization absorption [4], we simulate the propagation of a 400 µJ, 10 fs LG_{10} beam at 800 nm through a 1 meter-long and 1.2 bar helium-filled hollow core fiber. As the pulse propagates under anomalous dispersion [5] cause pulse self-compression and soliton fission, a VUV dispersive wave at 139 nm is generated in a pulse of 1 fs duration.

The Fig. 1 shows the spectral evolution and the OAM purity analysis (obtained by angular Fourier transform of the electric field for different radial values) in the insets. An energy transfer from 800 nm to 139 nm up to 1% is reached. Moreover, the OAM analysis of the generated VUV pulse reveals a rather good purity, with 77% in l = 1 and 23% in l = −1, proving an efficient angular momentum transfer. These results highlight the role of soliton dynamics in preserving OAM in ultrashort VUV pulses, offering new possibilities for structured light generation in ultrafast spectroscopy and nonlinear optics.

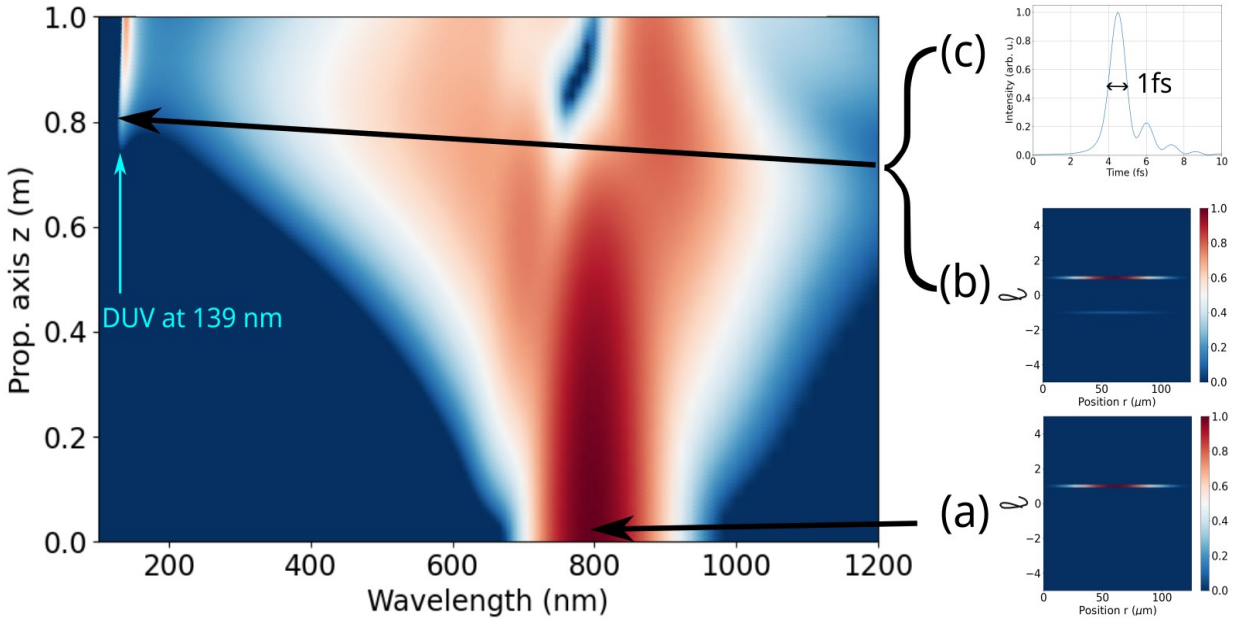

Fig 1. Spectral evolution (log scale) of 400 µJ, 10 fs at a central wavelength of 800 nm propagating in 1 meter-long, 1.2 bar helium-filled hollow core fiber with LG_{10} beam (OAM l = 1). The insets plot the OAM of the light for (a) 800 nm at distance 0 m, (b) 139 nm at 0.8 m. The inset (c) represents the intensity profile of the spatially integrated VUV pulse (in the spectral range 100 − 200 nm) at z = 0.8 m.

References:

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Ultrafast characterization and more

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Femto Easy ultrafast characterization product line

Advances in ultrafast laser systems and secondary sources at Amplitude.

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Key words: High Average Power, High Peak Power, High Energy, Ultrashort Pulse, OPCPA, Ti:Sa, Ytterbium, XUV source

Amplitude Laser provides the broadest portfolio of ultrafast lasers based on diverse existing technologies: Ytterbium, Ti:Sa, OPCPA... We report here the latest achievements in the development of ultrashort femtosecond lasers along with our roadmap for providing secondary sources, which are increasingly requested by the users.

We will first report our latest achievements in terms of femtosecond lasers using diode-pumped Ytterbium technology, suitable for high repetition rate and high average power. After a brief introduction of Yb-based solutions with very high repetition rate available at Amplitude, we will present our recent advancements in terms of XUV and OPCPA (Fastlite technology, now part of Amplitude group) sources pumped by these Ytterbium industrial ultrafast lasers.

Next, we will present recent results of CEP stabilisation and improved temporal contrast at the output of our OPCPA with the perspective of using this technology to seed high energy Ti:Sa amplifier in replacement of the conventional Ti:Sa oscillator.

We will finally present our results on the high energy Yb-based laser delivering up to 500mJ@100Hz (Magma) that can also be used as a pump for high energy OPCPA system or for generating secondary source like for example X-ray source (X-Pulse project in partnership with Alphanov).

SESSION 2

Attosecond science

Correlation-Driven Charge Migration Triggered by Infrared Multi-Photon Ionization

Victor Despré*[1]

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Résumé

In a molecular system, the correlation-driven charge migration (1) (CDCM) is a purely electronic process that involves the ultrafast dynamics of electrons originating from coherent superposition of eigenstates followed by the ionization of a single molecular orbital (2). The possibility of observing CDCM has been a driving force behind theoretical and experimental developments in the field of attosecond molecular science since its inception. Although X-ray free-electron lasers have recently emerged as a promising tool for experimentally observing CDCM, the unambiguous observation of CDCM, or more generally, charge migration dynamics triggered by ionization, remains elusive.
In this work (3), we present a method to selectively trigger such dynamics using molecules predicted to exhibit long-lived electron coherence. We show that these dynamics can be selectively triggered using infrared multi-photon ionization and probed using the spacial resolution of X-ray free-electron laser, proposing a promising experimental scheme to study these pivotal dynamics. Additionally, we demonstrate that real-time time-dependent density-functional theory can describe correlation-driven charge migration resulting from a hole mixing structure involving the HOMO of a molecule.

References:

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*Intervenant

Can classical trajectory dynamics methods accurately simulate attochemistry?

Thierry Tran[1], Anthony Ferté[1], Morgane Vacher[1]

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Since the advent of the first attosecond pulse in 2001,[1] its application on polyatomic molecules has given birth to the field of attochemistry. The large energy bandwidth of such a laser source will coherently populate multiple electronic states resulting in a coherent electronic superposition. A particular challenge in this field is the accurate theoretical simulation of the molecular coupled electron-nuclear dynamics induced by an electronic wavepacket.[2] Using an example with the two lowest cationic states of fluorobenzene, which feature a conical intersection near the Franck-Condon point (FC), we assess the accuracy of mixed quantum-classical methods, such as Tully surface hopping and classical Ehrenfest, for attochemical simulations. For the reference quantum wavepacket dynamics, we use Direct Dynamics variational Multiconfigurational Gaussian (DD-vMCG) method.[3] Upon coherent ionization to the two lowest cationic states, mixed quantum-classical methods with classical independent trajectories predict inaccurate trends for the nuclear dynamics and heavily underestimate the attochemical control compared to the full quantum dynamics.[4]

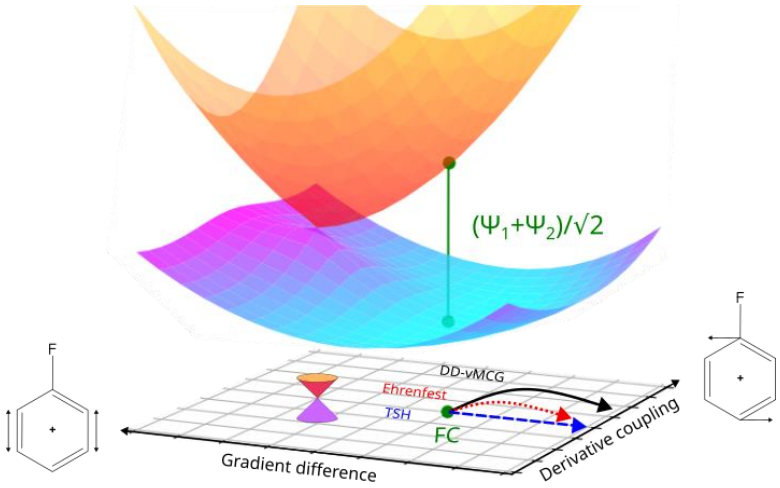

Figure: Scheme showing the average nuclear dynamics in the branching space induced by an initial electronic coherent superposition at the neutral Franck-Condon point using the Tully surface hopping, classical Ehrenfest and DD-vMCG methods.

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Probing iodine chemical environment with ionization delays

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Synopsis Ionization delays have been measured in a set of iodine-containing molecules following photoemission from the iodine 4d core levels. We evidence the influence of the iodine chemical environment on the ionization delays by measuring significant relative delays between iodomethane (CH_3I), iodoethane (C_2H_5I), iodopropane (C_3H_7I) and iodobenzene (C_6H_5I). Strikingly, a delay difference of up to 131 ± 12 as is found between the two structural isomers of iodopropane. The localization of the initial orbital on a single atom thus reveals subtle differences in the molecular potential.

Photoionization delays result from the scattering of the outgoing electron on the atomic/molecular potential and thus provide a fine probe of the potential landscape. While ionization delays studies have been performed in readily accessible valence molecular shells [1, 2], one can now reach core shells of high-Z atoms, such as iodine 4d levels, with XUV attosecond pulses generated by High Harmonic Generation (HHG). These core levels are of particular interest as they act as localized electron sources on one particular atom in the molecular frame.

Molecular core shell ionization delays have been recently measured using the streaking technique with both HHG [3, 4] and XFEL [5] sources. Our approach is based on RABBIT (Reconstruction of Attosecond Beating By Interference of two-photon Transitions) spectroscopy, allowing to explore a wide energy range with high spectral resolution. In order to measure reliably the expected small delay differences and to avoid spectral congestion, we used two electron spectrometers in a two-foci configuration [6], in which two RABBIT spectrograms are measured simultaneously in parallel, one probing the molecules of interest, the other acting as an absolute timing and XUV spectral phase (attochirp) measurement in the neon reference gas.

We were able to measure significant relative delays between different iodine-containing molecules such as iodomethane (CH_3I), iodoethane (C_2H_5I), iodopropane (C_3H_7I) and iodobenzene (C_6H_5I). The present results reproduce previous measurements on iodoethane [4]. However, the theoretical prediction in [4] of an increase of the delay with the size of the carbon chain is observed only between iodomethane and iodoethane, and breaks completely for the larger carbon chains. A most remarkable effect is observed on the two structural isomers of iodopropane (1- and 2-iodopropane) with a delay difference reaching up to 131 ± 12 as. Such large difference was unexpected given their very small chemical shift difference, demonstrating the great sensitivity of our observable on the chemical environments.

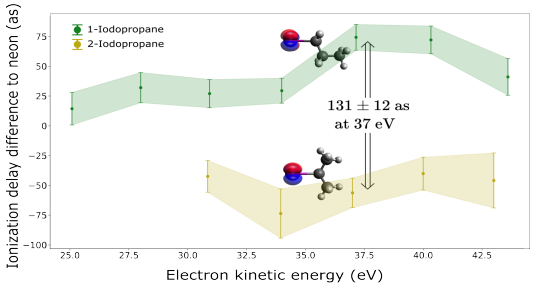

Figure 1. Ionization delays of the 4d electrons for the two iodopropane isomers with respect to neon.

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Rabi oscillations, photo-emission and entanglement: A time-resolved picture

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The development of free electron lasers gives access to ultra-short, intense extreme ultraviolet light pulses which allow the experimental study of non-linear effects in this energy range. For instance, the photo-ionization of an atom with a photon energy simultaneously driving Rabi oscillations in the ion manifests as a doublet in the photo-electron spectrum due to the dressing of the ionic states [1].

An interesting feature of such a situation is the generation of entanglement between the ionic core and the photo-electron [2]. There, the light pulse plays two roles, i.e., ionizing the atom and dressing the ion. This makes the investigation of the entanglement build-up mechanism difficult to address, in particular in terms of time and length scales.

In this work, we address this issue by considering an original pulse configuration in simulations on a model helium atom. We use two different photon energies to unambiguously discriminate the Rabi dynamics from the photo-emission process. Also, we consider, for the ionizing field, a sequence of two short pulses instead of a single longer one. By controlling the delay τ_{XUV} between the ionizing pulses, this allows uncovering the mechanism behind the generation of entanglement in the time domain.

Moreover, since this scheme creates electron wave-packets with the same energy but at different times, τ_{XUV} -dependent interferences are observable in the photo-electron spectrum. We established, both numerically and analytically, a link between the contrast C of these interferences and the purity P of the reduced density matrix of the ion as $C^2 = 2P + 1$, see Fig. 1. Thus, our original scheme allows a direct measurement of the purity in the spectra.

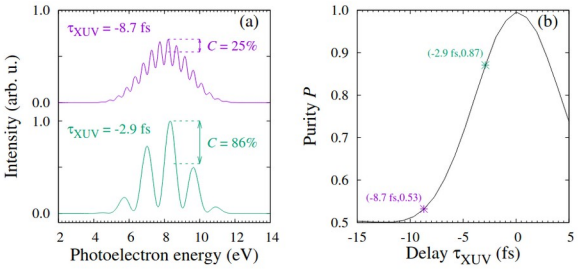

Figure 1. (a) Photo-electron spectra for two different τ_{XUV} delays. (b) Final purity as a function of τ_{XUV}.

Finally, we show that the contrast directly corresponds to the overlap between the states populated by each of the two short pulses. Hence, it allows performing a quantum tomography of the Rabi states.

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Reconstruction of the attosecond dynamics of unobserved ions

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Laser-dressed photoelectron spectroscopy has allowed the measurement of the quantum state of photoelectrons. In ideally coherent situations, this state can be fully described using a wave function which can be retrieved with well established techniques such as RABBIT and attosecond streaking. However in a general situation, the photoelectron can be entangled with unresolved degrees of freedom and exhibit a partially coherent behavior. Its state should then be described using a density matrix [1], which takes into account the loss of information when performing incomplete measurements. New experimental schemes have recently been developed to access such photoelectron density matrix [2, 3].

Using the Mixed-FROG technique, we measured the reduced density matrix of photoelectrons resulting from the ionization of xenon atoms by high harmonics, see Fig. 1. This density matrix ϱ_{PE} $(t \rightarrow +\infty)$ represents the asymptotic quantum state of the photoelectron from which we reconstructed its transient evolution ϱ_{PE} (t) through time-frequency analysis [4, 5]. In our experimental conditions, the electron can reach the continuum through two ionization channels, leaving the Xe^+ ion in a superposition of $5p_{3/2}$ and $5p_{1/2}$ states. These two quantum paths are fully distinguishable in the photoelectron spectrum, thus forming a maximally entangled ion+electron system. We show that this entanglement can become a vector of information about the unobserved ion. Indeed enough information is preserved in the electron density matrix ϱ_{PE} (t) to access the transient ion density matrix ϱ_{ion} (t) up to an absolute phase. This experimental reconstruction reveals that even though no asymptotic coherence is left in the ion, some transient ionic coherence can exist during the ionization process. In this work we therefore demonstrate that entanglement can be used to access experimentally the attosecond dynamics of unobserved quantum subsystems.

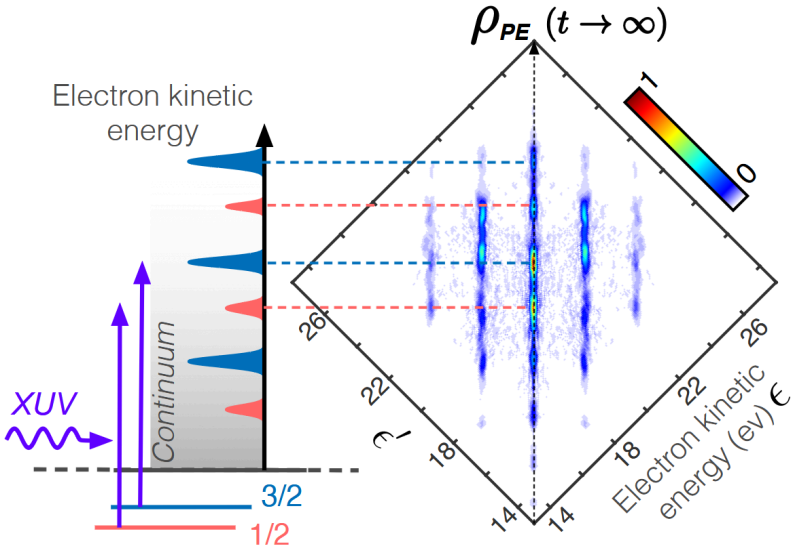

Figure 1 : Experimental density matrix (only modulus shown) of photoelectrons obtained by the ionization of Xe through the $5p_{3/2}$ and $5p_{1/2}$ channels (left: photoionization scheme)

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SESSION 3

Materials 1

Génération par autocorrélation de signaux THz et de second harmonique à la surface de cristaux diélectriques

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Le redressement optique (*Optical Rectification* OR) d'impulsions laser femtosecondes à la surface d'un cristal est un moyen de produire des impulsions térahertz (THz) ultracourtes dont la largeur de bande pourrait dépasser des dizaines de THz. En effet, comme il n'y a pas de propagation dans le cristal, les effets dépendant de la longueur d'onde, comme l'absorption ou l'accord de phase, ne sont plus limitant. Cependant, le principal inconvénient vient de la faible efficacité de l'interaction, qui se traduit par une faible puissance THz, inférieure de plusieurs ordres de grandeur à celle qui pourrait être obtenue en transmission. Pour détecter de tels signaux, il est donc nécessaire d'utiliser un système à la fois à large bande et sensible, ce qui représente un défi dans le domaine THz où les détecteurs sont soit large bande mais peu sensibles, comme ceux utilisant l'effet electro-optique,, soit très sensibles mais très lents, comme les bolomètres. La technique d'autocorrélation [1], habituellement utilisée pour caractériser les impulsions laser ultra-courtes, peut apporter une solution intéressante à ce problème. Dans cette configuration, deux impulsions laser femtosecondes successives, séparées par un délai variable τ, se superposent dans un cristal non linéaire. Le signal, généralement obtenu par génération de seconde harmonique (*Second Harmonic Generation* SHG), est ensuite enregistré en fonction de τ. Le détecteur peut ainsi être très lent puisque la résolution temporelle ne dépend que de τ. Nous avons adapté cette technique à la caractérisation d'impulsions THz large bande générées par redressement optique d'impulsions laser femtosecondes à la surface de différents cristaux non linéaires, tels que ZnTe [2], GaP, $LiNbO_3$ ou encore GaSe. L'interférogramme SHG a été simultanément enregistré comme outil de comparaison. Nous avons ainsi vérifié que l'épaisseur du cristal ou le désaccord de phase entre l'onde optique et l'onde THz n'ont aucun effet sur l'interférogramme obtenu. Nous avons aussi démontré que cette technique permet bien d'enregistrer la réponse spectrale du cristal utilisé comme par exemple des raies d'absorption dues à la présence de phonons.

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Controlling the excited-state lifetime of transition metal oxide photocatalyst

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Sunlight-to-chemical conversion requires systems that generate long-lived excited states upon illumination. However, this comes with a big energy penalty. [1] For example, the natural photosynthesis system sacrifices up to half of its light energy input to gain the carrier lifetimes necessary to drive chemical reactions.[2] Similarly, in artificial photosynthetic devices, lifetime gain is typically achieved through the application of external electrical bias,[3] or the use of sacrificial reagents.[4] Currently, there is no blueprint for the design of semiconductors with intrinsically long lifetimes. In this talk, I will discuss time-resolved measurements on a series of transition metal oxides (TMOs), that point towards a link between carrier lifetime and electronic configuration of the metal. [5] We identify a sub-ps relaxation mechanism via metal-centred ligand field (LF) states which compromises quantum yields in open d-shell TMOs (e.g., Fe_2O_3, Co_3O_4, Cr_2O_3, NiO). This relaxation pathway is partially mitigated in materials with spin-forbidden LF transitions, (Fe_2O_3) and is absent in d0 and d10 electronic configurations (e.g., TiO_2, $BiVO_4$) leading to higher photocatalytic yields. By drawing comparisons with molecular complexes, I will discuss possible strategies to extend carrier lifetime in transition-metal-containing semiconductors.

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Optical pump-induced carrier dynamics in InSb: probing the plasma frequency evolution

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Quantum materials have attracted a lot of attention in the past decade and controlling the material properties via a light excitation has become a promising challenge in condensed matter. In particular, the light-induced carrier dynamics is a key effect both on a fundamental level and for applicative perspectives such as photodetectors. In topological semimetals, where different degrees of freedom are intertwined, understanding how carriers are photoexcited and then relax by coupling to the structure is a key investigation, to highlight the mechanisms at play in possible light-induced topological phase transition.

Terahertz spectroscopy, in combination with a pump pulse, has proven to be a very relevant tool to access carrier dynamics [1]. Hence, we have implemented a versatile THz Time-Domain Spectroscopy (THz-TDS) set-up, relying on a compressed TANGOR laser (1030nm, 350 uJ/pulse, 60 fs, 1-250kHz repetition rate). The THz generation is carried out by optical rectification in an organic BNA crystal, resulting in a relative broadband spectrum. This set-up is coupled either to an optical pump at 1030 nm or to a THz pump, giving access to two distinct ranges of pump energy, both in reflection and transmission configurations.

We present benchmarked studies carried out with this set-up, on a low bandgap semiconductor InSb. We studied this material in a reflection configuration, with an optical pump and a THz probe, at various pump fluences. We show that the THz peak tends to slightly shift under the pump influence (Fig 1.a), which can induce errors in the evaluation of the dynamics. We measured the reflected maximum probe field for different pump-probe delays [2], acquiring the full probe pulse, hence corresponding spectrum, on a relatively wide frequency bandwidth [3], between 0.2 and 6 THz.

Using the Drude-Lorentz model and considering the carrier density-dependent penetration depth of THz in the material, we propose a model to fit the reflectivity of the material at different delays (Fig 1.b), and extract from that the population of excited carriers as a function of pump-probe delay, and their diffusion in the sample until returning to equilibrium.

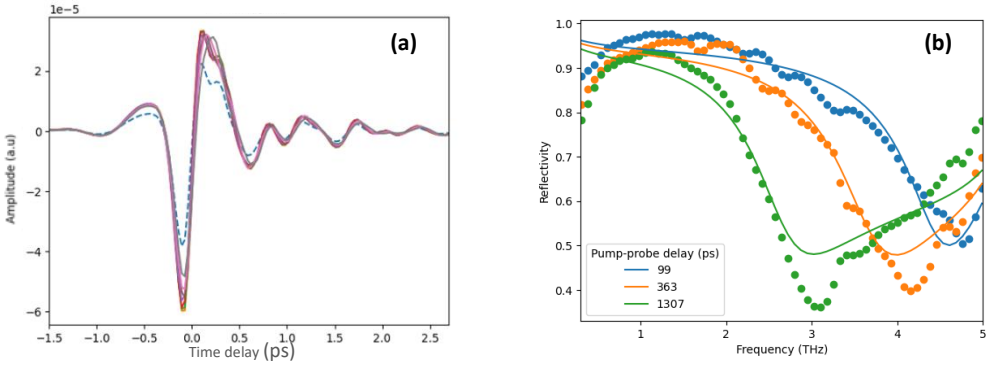

Figure 1. (a) THz-TDS pulses in reflection on a bulk InSb sample at 8K, following an optical pump at 1030 nm, for various pump-probe delays. (b) Reflectivities at three pump-probe delay, using the maximally pumped spectrum (6.6 ps pump-probe delay) as a reference spectrum for reflectivity. The dots are experimental data and lines are the corresponding fits.

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ULTRAFAST PHOTO-INDUCED DYNAMICS TRIGGERED BY ELECTRON TRANSFER IN 1D VAN DER WAALS HETEROSTRUCTURE

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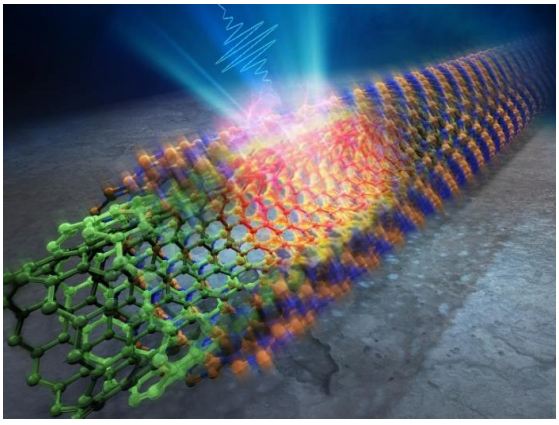
Recent studies have shown that layered-materials that are atomic-thick exhibit new properties. The static properties of these structures, such as electrical conduction, have been widely studied. However, their dynamic properties, such as photo-induced electron transfer and the subsequent structural dynamics have been explored to a lesser extent.

In this study, we have unveiled a new phenomenon where, upon light excitation, a nested structure of carbon nanotubes enveloped in boron nitride nanotubes allows for a peculiar electronic channel resulting in interlayer charge transfer. We synthesize nested cylindrical structures by wrapping carbon nanotubes (CNTs) in boron nitride nanotubes (BNNTs), refer to as 1D van der Waals heterostructure, and monitor the motion of electrons and atoms induced by ultrashort UV light excitation. On the one hand, the electronic dynamics are observed using broadband ultrafast optical spectroscopy measurements, which can capture the instantaneous changes in molecular and electronic structures. On the other hand, the structural dynamics are captured using ultrafast time-resolved electron diffraction with a time resolution of about one picosecond.

The experimental results, supported by theory, demonstrate that that free electrons generated by UV light (3 eV) in the CNTs part can be transferred to the BNNTs wide bandgap (6 eV) part through an electronic channel resulting from the interlayer interactions in these 1D materials. In addition, the energy of these excited electrons is rapidly converted into thermal energy within the BNNTs, resulting in a faster radial expansion in this 1D heterostructure compared to bare CNTs.



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SESSION 4

Femtochemistry and femtobiology

Modeling Excited-State Dynamics: Tailoring Molecules for Ultrafast Nonradiative Decay

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Understanding and controlling ultrafast nonradiative decay pathways is essential for designing efficient photoprotective and photothermal materials. Sinapate esters and cinnamate derivatives, archetypal molecular systems for UV filters and photothermal conversion, serve as playground systems for exploring ultrafast excited-state relaxation through photoisomerization.[1-5] Photoisomerization yields are strongly dependent on the steric and electronic tuning of molecular structures.

By combining transient absorption spectroscopy, quantum chemical calculations, and mixed quantum-classical nonadiabatic dynamics, we investigate the photophysics and photoisomer formation of a class of sinapate ester and cinnamate derivatives. [1-5] Our studies reveal that molecular symmetry, steric effects, and electronic substitution significantly influence conical intersection accessibility and photoisomerization efficiency. We show that symmetric derivatives exhibit enhanced nonradiative decay via easily accessed conical intersections, whereas asymmetric analogues display slower relaxation dynamics. Additionally, the isomerization yield is impacted by the local topography around conical intersections.

Furthermore, our results demonstrate how molecular design can tune photothermal energy conversion, as exemplified by sinapic diacid—a promising molecular heater that efficiently converts solar energy into localized heat.[6] These insights provide a mechanistic framework for tailoring excited-state dynamics, with implications for the development of next-generation photoprotective and photothermal materials.

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Toward the design of fast red reversible photoswitchable fluorescent proteins using multi-timescale transient absorption spectroscopy.

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Abstract

In recent years, reversible switchable fluorescent proteins (RSFPs) have gained attention due to their ability to undergo a reversible, photoinduced transition between a fluorescent (On) state and a non-fluorescent (Off) state. Their significance lies in their application in super-resolution fluorescence microscopy (nanoscopy), enabling nanometric spatial resolution-a breakthrough that was recognized with the Nobel Prize in Chemistry in 2014. Among coordinate-targeted nanoscopy methods, REversible Saturable Optical Linear Fluorescence Transitions (RESOLFT) stands out, as it requires very low light intensities to overcome the diffraction barrier, making it particularly suitable for live-cell imaging (1). Until now, RESOLFT imaging has been performed in the visible range of the electromagnetic spectrum. However, the red/near-infrared (NIR) spectral domain is highly advantageous for live-cell imaging due to its reduced phototoxicity, lower autofluorescence, greater penetration depth in vivo, and expanded spectral multiplexing capabilities. Another key challenge in RESOLFT imaging is also the need for fast RSFPs with sub-second thermal back recovery of the Off state. Such RSFPs would enable low-light-intensity RESOLFT imaging with a single wavelength.

Here, we present the development of fast red RSFPs based on (i) a derivative of the wild-type bacteriophytochrome from Deinococcus radiodurans (Dr-PSM) (2) and (ii) a charge-transfer (CT) complex composed of a flavin cofactor and a substrate-analogue inhibitor from the

*Speaker

monomeric sarcosine oxidase flavoprotein family (MSOX) (3). We investigated their photodynamics using multi-timescale transient absorption spectroscopy to characterize key intermediates that govern switching quantum yield and thermal back recovery. Notably, we identified a transient intermediate in the nanosecond timescale that is typically undetectable in conventional femtosecond pump-probe transient absorption or nanosecond flash photolysis experiments.

To resolve these dynamics, we utilized a unique instrument from the Multi-Scale Femto-Millisecond Transient Absorption Spectroscopy platform of PEPR-LUMA and LOB (4). Specifically, we applied Arbitrary Detuning Asynchronous Optical Sampling (ADASOPS) to enable precise, multi-timescale control of pump-probe delays, spanning picoseconds to milliseconds (5). This approach allowed us to reveal nanosecond-scale dynamics with unprecedented accuracy. In this presentation, we will discuss our findings and their implications for the design of novel red/NIR RSFPs optimized for nanoscopy applications.

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Données FAIR : acquisition, partage et reproductibilité des résultats

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A l'ère de la science ouverte, le mot est au partage et à la publication des données. Le terme FAIR (Findable, Accessible, Interoperable and Reusable) est utilisé pour prendre en compte les différentes problématiques liées à la science et aux données ouvertes. Bien qu'on puisse considérer cette notion de partage et de réutilisation comme un but en soi, elle n'est cependant vraiment utile que pour certaines communauté où le partage et l'utilisation de données communes est essentielle au développement de la science: données de diffraction, spectroscopie, astronomie... Il est donc plus intéressant pour la communauté du GDR Ultrafast Phenomena de considérer la production de données FAIR sous l'angle de la reproductibilité et de la réplicabilité de nos résultats scientifiques, données et publications. C'est donc sous cet angle que je présenterai la production de données FAIR, les problématiques inhérentes et quelques solutions d'hors et déjà apportées et sur lesquelles la communauté du GDR pourrait construire ses données FAIR.

SESSION 5

Secondary sources

Single-shot electro-optic detection of THz electric field with high temporal resolution and MHz acquisition rate

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Single-shot terahertz (THz) waveform recording is essential for studying fast and rare events, particularly in accelerator physics and time-domain spectroscopy of irreversible processes. Traditional time-domain spectroscopy methods rely on scanning techniques, which limit their applicability for low-repetition-rate sources with significant fluctuations as well as to single shot detection at MHz acquisition rate. A promising strategy is the chirped pulse electro-optic sampling technique, which encodes THz signals onto a probe laser pulse for single-shot analysis. However, conventional implementations face trade-offs between temporal resolution and recording window length. Recent advancements overcome these limitations by integrating photonic time-stretch acquisition with Diversity Electro-Optic Sampling (DEOS), enabling high-resolution, long-duration single-shot THz measurements. This approach achieves sub-picosecond resolution at megahertz acquisition rates, facilitating the study of THz pulses emitted by relativistic electron bunches. These innovations enhance THz detection capabilities, expanding potential applications in ultrafast science and accelerator diagnostics.

Ultrafast microphysics of Bessel beam interaction with dielectrics

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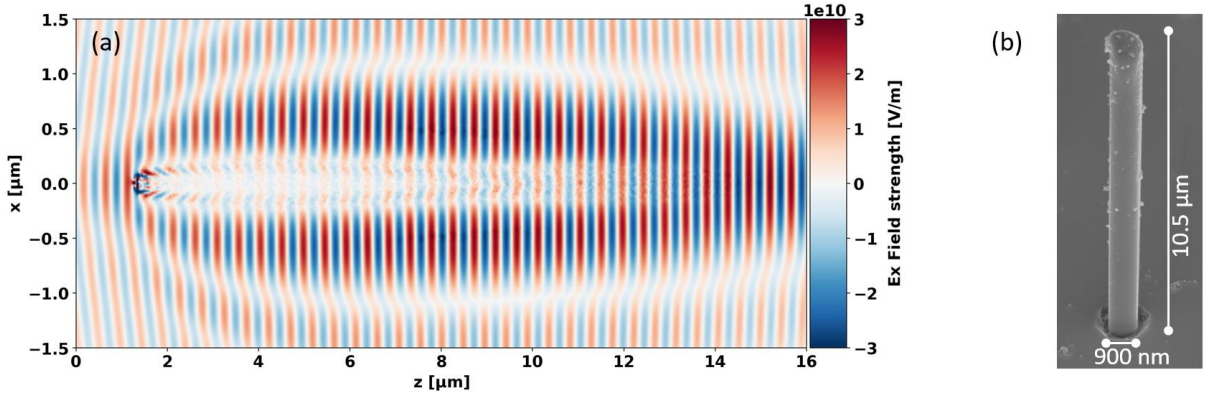
When tightly focused inside transparent dielectrics, femtosecond Bessel beams generate nano-plasmas with very high aspect ratio. The energy density is enough to open nano-voids even when fully inside the bulk of materials [1]. This presentation will explore the microphysics at play during the laser pulse propagation.
We use a particle-In-Cell code to model the interaction of the shaped ultrafast laser pulse with a transparent dielectric. The modelling is compared to several experimental diagnostics across various imaging techniques. Noticeably, using this modelling, we could reproduce the diagnostics, answering longstanding questions in such Bessel pulse propagation. [2]



(a) Particle-In-Cell simulation of the Ex field of a 100 fs Bessel pulse propagating in sapphire, at time 25 fs after its peak. (b) Scanning Electron Microscopy image of a nano-pillar generated via laser-induced extrusion [4].

We show that the formation of plasma waves by mode conversion (resonance absorption) and Landau damping are crucial in the interaction. Those effect can be only captured with kinetic plasma models such as Particle-In-Cell models, in opposition with hydro codes, or pulse propagation equation. The formation of dense plasma where the permittivity decreases down to zero allows the emergence of a number nonlinear dynamical effects spanning from second harmonic and THz wave generation to warm dense matter formation and high-precision nano-structuring [3].
Finally, we will show that the understanding of such interaction opens new avenues for high-energy density deposition in materials. It enabled us designing a new process for ultrafast laser structuring that produces vertically-standing nano-pillars atop of sapphire in single shot.

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Effets d'ionisation et impact de l'hydrogène sur les performances d'un accélérateur laser-plasma kHz

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L'accélération laser-plasma (LPA) est une méthode efficace pour accélérer les électrons à des vitesses relativistes en utilisant une impulsion laser ultracourte focalisée dans un plasma. Cette technique permet d'atteindre des champs accélérateurs bien supérieurs à ceux des cavités radiofréquence conventionnelles, réduisant ainsi les distances d'accélération et permettant des accélérateurs compacts. En particulier, l'accélération laser-plasma avec un taux de répétition de l'ordre du kHz présente un potentiel significatif pour diverses applications médicales et industrielles.

Le groupe APPLI au Laboratoire d'Optique Appliquée (Palaiseau) développe un système LPA au kHz, utilisant un laser qui délivre des impulsions de quelques mJ et de quelques cycles optiques. Ces impulsions doivent être fortement focalisées et comprimées temporellement pour atteindre les intensités nécessaires [Guénot 2017]. Pour un fonctionnement continu, le système de cible –un jet de gaz- doit maintenir un flux de gaz stable. Précédemment, l'utilisation d'azote a permis de démontrer une opération stable et continue de l'accélérateur [Rovige 2020], mais des études récentes montrent que l'hydrogène pourrait offrir des performances supérieures en termes d'énergie [Salehi 2021].

Dans cette présentation, nous détaillerons notre dernière étude sur les effets d'ionisation optique dans un accélérateur laser-plasma fonctionnant au kHz, en utilisant des impulsions de quelques mJ et de quelques cycles optiques [Monzac 2024]. Nous expliquerons pourquoi l'utilisation d'hydrogène améliore les performances de l'accélérateur par rapport à d'autres gaz, y compris l'hélium : les effets d'ionisation peuvent déformer l'impulsion laser, affectant négativement les performances de l'accélérateur. Ces effets sont minimisés dans le plasma d'hydrogène, améliorant ainsi la qualité du faisceau d'électrons. Avec des impulsions laser de 4 fs et des énergies de 2,5 mJ, nous avons obtenu des faisceaux d'électrons à faible divergence, avec des spectres d'énergie centrés autour de 5-10 MeV. Ces faisceaux ont montré une remarquable stabilité tir-à-tir. Pour obtenir ces résultats, nous avons implémenté un schéma de pompage différentiel, permettant de maintenir un flux de gaz léger continu dans l'enceinte expérimentale tout en assurant un vide suffisant pour la propagation du laser [Monzac 2025]. Nous présenterons également ce système.

Amplification of beams carrying Orbital Angular Momentum in a plasma-based XUV laser: a numerical study

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Light beams can carry angular momentum, the most familiar form being the spin angular momentum, of circularly polarized light. Another form is orbital angular momentum (OAM). The most striking example of such OAM beams is optical vortices. They are characterized by a helical phase where the quantum number or topological charge quantifies the OAM carried by the photons at first order (1). Unlike Gaussian beams, these optical vortices have a helical wavefront and a phase singularity at their center, which is why their irradiance is donut-shaped. Optical vortices are widely used in the visible and infrared domains, for example, for optical communications through optical vortex multiplexing (2) and STED microscopy (3). Such beams have been recently extended in the extreme ultraviolet range (from 10 to 120 eV), by transferring the OAM of an intense infrared laser to its high harmonic orders. The topological charge of the -th harmonic is given at first order by (4) where is the topological charge of the driving infrared beam. A variant of this scheme involves a wavemixing in the high harmonic medium of two incident beams carrying different OAM contents (10). This situation is giving a larger control on the harmonic topological charge. In this study, we will discuss the amplification of wave-mixing generated high harmonic vortices by a plasma-based XUV laser.

In a plasma-based XUV laser, a population inversion is achieved between two excited levels of an ion of a hot dense plasma. The lasing transition is pumped through collisions between the ions and electrons of the plasma. In this study, we will consider a Ni-like Krypton (Kr8+) XUV laser (5) at 32.8nm, a medium suitable for the harmonic amplification of a Titanium-Sapphire laser. The plasma creation and electron heating are achieved by a same intense laser pulse. By seeding the resulting medium with a frequency-matched high-order harmonic, a coherent beam, two orders of magnitude more energetic, has been demonstrated (6). Theoretical and numerical studies (7) (8) highlighted the non-adiabatic response of the lasing medium as the seed (few fs) is orders of magnitude shorter than the depolarization and recovery time of the laser. In this situation, the seed itself is not amplified, but triggers the emergence of a long coherent wake, featuring Rabi oscillations for strong amplification (9).

In this presentation, we describe numerical study of harmonic vortices propagation into the plasma-based XUV laser. The simulations have been performed using the 3D Maxwell- Bloch code DAGON (11), incorporating atomic physics data and data from the collisionalradiative code OfiKinRad. Different situations potentially achievable with wave-mixing will be detailed. First, selection and amplification of a single vortex beam with a well-defined charge is considered. A corresponding simulation is presented in Figure 1. The initial seed irradiance (1.a) and phase (1.b) transverse distributions are presenting the typical features of a vortex. The final transverse profile at peak irradiance displayed in (1.c) and (1.d) show a global conservation of the azimuthal phase, but a dramatic change in the irradiance structure, the ample hole of the vortex being replaced by a uniform distribution with the exception of a narrow zero irradiance feature on-axis. This behavior can be at first interpreted as resulting from gain saturation. However, the non-adiabatic response of the lasing medium leads to a more complex evolution. The amplified pulse during its propagation in the XUV plasma is displayed in Figure (2) in which coherent wake and Rabi-oscillations following the seed pulse are clearly visible. We will give an insight on the spatio-temporal behavior of the region surrounding the phase singularity and highlight the amplification conditions preserving the seed beam structure.
We will extend this study to a situation in which the exit of the wave-mixing medium is directly imaged at the entrance of the XUV laser medium, leading to an irradiance profile composed of fringes with a fork pattern. The inclusion of realistic harmonic field will be discussed

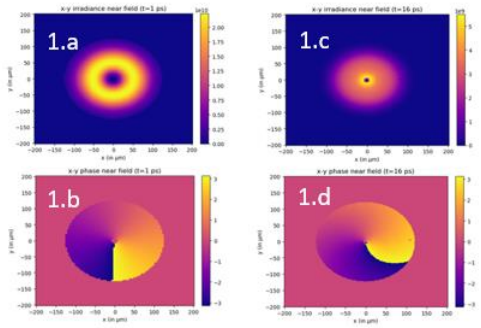

Figure (1): Irradiance (top) and phase (bottom) transverse xy profiles of the non-amplified seed (left) and the output amplified beam (right)

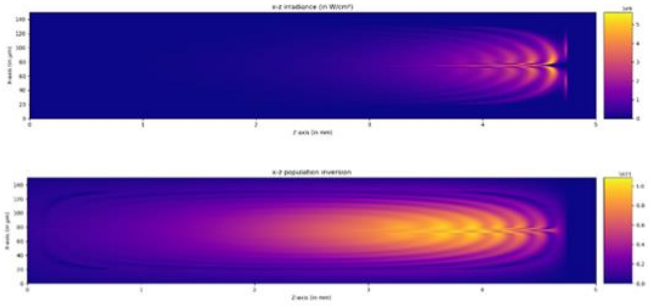

Figure (2): Irradiance (top) and population inversion (bottom) yz profiles right before the exit of the plasma. The thin vertical holed beam is the vortex seed, followed by a wake that exhibits Rabi oscillations

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Effets d'ionisation et impact de l'hydrogène sur les performances d'un accélérateur laser-plasma kHz

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High harmonic generation (HHG) is best known as a strong-field process, in which a driving laser wave interacts with atoms to produce radiation at high-order multiples of its frequency. Alternatively, the generation of a photon of harmonic order q may also be seen as coming from absorption of q driving photons and subsequent emission of a single harmonic photon in the nonlinear medium. This photon-based reasoning correctly predicts the energy and momentum of high-harmonic photons, but fails to predict the strongly nonperturbative yield of HHG : from perturbative nonlinear optics, a q-photon process is expected to scale as Eq, whereas plateau high harmonics all scale as Eqeff with a common effective nonlinearity order qeff < q. In this talk, we will present a photon-based interpretation of HHG that correctly accounts for this nonperturbative efficiency, backed by experimental observations and a simple analytical model.

We will discuss experimental results on the amplitudes [1], phases [2] and transverse modes [3] of beamlets produced when driving HHG with two noncollinear beams, in a ultrahigh-order wave mixing scheme. In this arrangement, perturbative power laws were reported when the second beam is much weaker than the first [4, 5]. We will explore the regime where the two beams have comparable intensities, and show how the nonperturbative nature of HHG produces features unseen in perturbative harmonic generation processes : they originate from the coherent addition of infinitely many higher-order photon pathways, involving absorption-emission pairs (Figure 1, bottom). Their interference is experimentally revealed through secondary maxima in yield curves (Figure 1, top) and transverse mode reshaping. This extension of the photon picture of nonlinear optics to the strongly nonperturbative regime further motivates the search for a practical quantum-optical theory of HHG, and may find applications in spectroscopy or all-optical extreme-ultraviolet beam shaping.

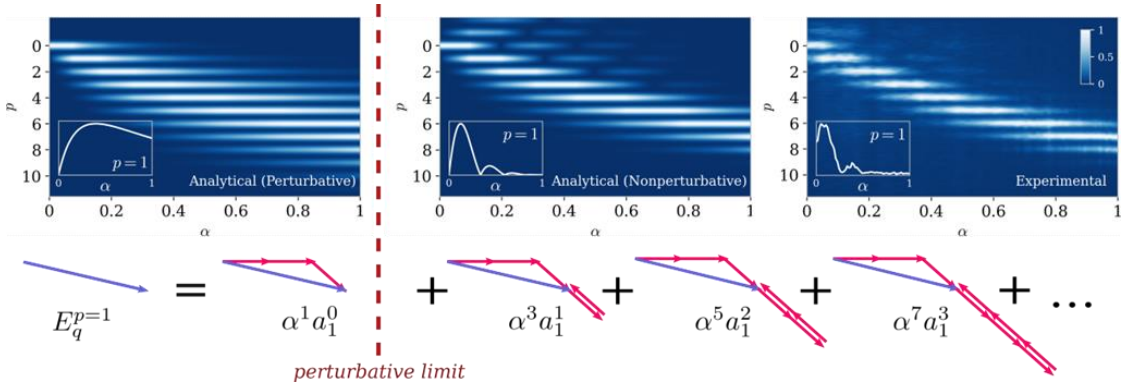

FIG. 1. From perturbative to nonperturbative nonlinear optics. Top : harmonic beamlet yields (indexed by p) from HHG driven by two noncollinear beams, as a function of the amplitude ratio α between the two beams. Beamlet p = 0 (resp. p = q, the harmonic order) is emitted in the direction of the first driving beam (resp. of the second driving beam). Lineouts of beamlet p = 1 are shown. Right : experimental data for harmonic q = 13. We observe a series of beamlets that successively light up and fade out as α increases. Left and center : analytical simulations of the experiment, respectively for perturbative and nonperturbative harmonic generation physics. In the nonperturbative regime, negative orders p show up, and secondary maxima appear in the yield curves, in excellent agreement with the experimental data. Bottom : photon pathways contributing to the field of beamlet p = 1 (for q = 3) in our analytical framework. The red dashed line indicates the perturbative truncation of the power series. The additional photon pathways are responsible for the nonperturbative features evidenced in the top row.

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Commercial table-top beamline for attosecond science

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Abstract

Since 2009, UltraFast Innovations (UFI®) has provided a wide range of specialized and customized optics and components, as well as instrumentation for ultrafast optical science. Continuous development and extensive collaboration with Professor Krausz's research group (Nobel Prize in Physics, 2023) have allowed UFI to bring to market a series of complete solutions for pump-probe spectroscopy, XUV imaging, pulse compression, and the generation of coherent secondary radiation1,2.
A prominent example of such a solution, encompassing most of the company's expertise, is an XUV beamline driven by a high-repetition-rate, industrial-grade laser. The beamline includes post-compression stages based on nonlinear spectral broadening and in-house-produced high-dispersive mirrors (SAVANNA), a beam stabilization system, a high-harmonics generation chamber (NEPAL), a programmable IR-XUV delay and refocusing stage (K2), an XUV polarization converter (AURORA), and a VUV-XUV spectrometer (EVEREST). The performance of each module is optimized and tailored for specific applications in ultrafast optical science.

In this presentation, we will introduce a compact tabletop XUV beamline and discuss the parametric space of the SAVANNA and NEPAL instruments.

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*Speaker

SESSION 6

Gas phase systems

Computational photochemistry to simulate time-resolved experimental observables

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Understanding the coupled motion of electrons and nuclei following photoexcitation is essential for deciphering molecular photochemistry. Nonadiabatic dynamics simulations provide a powerful tool to explore these ultrafast processes, revealing mechanistic details that experiments alone cannot capture. In this talk, I will illustrate how we use these simulations to gain insight into fundamental photophysical and photochemical phenomena. Simulating time-resolved observables that can be directly compared to experiments, can be crucial for the validation, but knowing the limitations is important. By integrating multiple experimental techniques with computational simulations, we can construct a more comprehensive picture of molecular photodynamics, bridging the gap between theory and experiment to advance our understanding of light-driven processes.

A kHz laser desorption scheme adapted to ultrafast gas-phase measurements of thermolabile molecules

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Abstract

We have developed a new laser desorption scheme coupled to a supersonic expansion able to operate at several kHz
rates. We demonstrate that it can be used to perform cold spectroscopy of flexible or fragile molecules. We also
demonstrate its ability to study non-covalent complexes, such as hydrated complexes, which can be formed in the jet.

*Speaker

Light-induced orbital magnetism in atomically precise metal clusters

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Abstract

In view of the recent increased interest in light-induced manipulation of magnetism in nanometric length scales nanoparticles (1) and two-dimensional nano-domains (2) are emerging as potential materials of choice. With this emerges one of the fundamental question: how does the miniaturization of these systems impact the underlying physics. To this end, this ab-initio theoretical work presents metal clusters as promising elementary units for generating all-optical ultrafast magnetization. Within the real-time (RT) formalism of time-dependent density functional theory (TDDFT), simulation of plasmon excitation in atomically precise clusters using circularly polarized laser pulse captured the transfer of angular momentum from light field to induce orbital magnetic moment. In the near-field analysis, the origin of the orbital magnetization is traced back to the self-sustained circular motion of the induced electron density which corroborates the presence of nanometric current loops giving rise to orbital magnetic moments. The results (3) provide valuable insights into the quantum many-body effects that influence the light-induced orbital magnetism in metal clusters depending on its geometry and chemical composition. At the same time, they explicitly demonstrate the possibility for harnessing magnetization in metal clusters, offering potential applications in the field of all-optical manipulation of magnetism.

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*Speaker

GDR-UP 2025, Paris
High-Frequency Phase Noise Suppression for the DeLLight Interferometer to Measure the Optical Nonlinearity in Vacuum

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Quantum Electrodynamics (QED) predicts that the vacuum behaves as a nonlinear optical medium: the speed of light in a vacuum should be reduced when the vacuum is stressed by intense electromagnetic fields [1-2]. The DeLLight (Deflection of Light by Light) experiment [3-5] aims to measure this effect by using intense and ultra-short laser pulses delivered by the LASERIX facility (2.5 J per pulse, 50 fs, 10 Hz repetition rate) at IJCLab (Paris-Saclay University). The experimental method involves measuring, using a Sagnac interferometer, the deflection of a low-intensity probe laser pulse after crossing the optical index gradient imprinted in vacuum by an external intense pump laser pulse. With the available laser intensity provided by LASERIX, the expected signal is about 15 pm [3]. With a spatial resolution to define the position of the interference intensity profile σ_y = 15 nm (corresponding to the ultimate shot noise of available CCD cameras), the expected QED deflection signal can be observed at a 5σ confidence level with about one month of collected data [4-5].

The spatial resolution σ_y is limited by the phase noise induced by mechanical vibrations of the Sagnac interferometer. To achieve the shot noise limit in our experiment, it is crucial to suppress this phase noise contribution. Here, we propose a novel method, High-Frequency Phase Noise Suppression (HFPNS), to overcome this challenge for pulsed interferometric measurements. The principle of the HFPNS method is to first split the incident pulse before entering the interferometer into two identical pulses, one being delayed by about 5~ns. This time offset ensures the delayed pulse does not interact with the ultra-intense pump pulse. However, at this high frequency (200 MHz), the phase noise due to vibrations becomes identical for the prompt and delay pulses. The delayed pulse is then used to directly measure the phase noise, which is subsequently suppressed off-line.

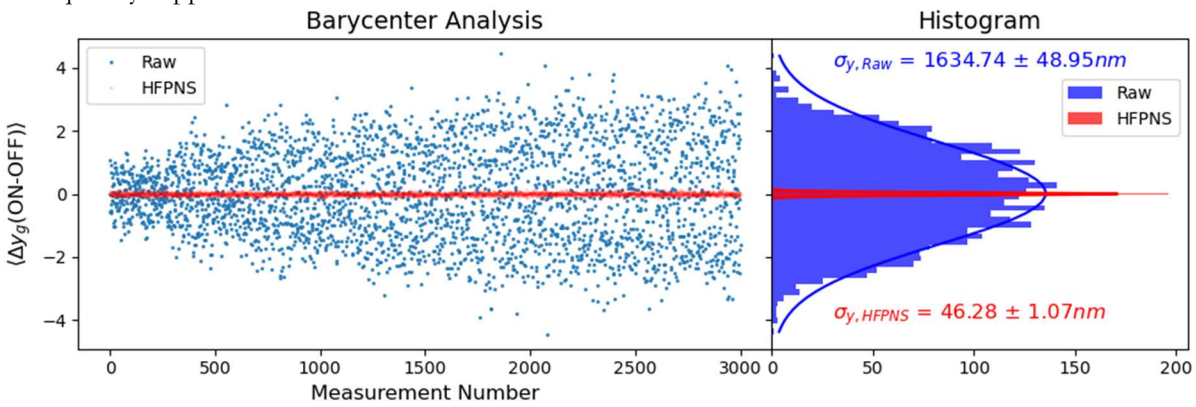

Fig. 1: The measured barycenter positions of the interference intensity profile (left) and corresponding histograms (right) directly to the raw signal (blue) and after the HFPNS method (red).

The HFPNS setup has been installed and tested with the current DeLLight interferometer. In this talk, I will present results obtained with the dedicated HFPNS setup: the phase noise has been successfully suppressed, as shown in Fig. 1, and the achieved spatial resolution is now almost at the level of the CCD shot noise limit, only limited by the residual vibrations of the delay line which will be later suppressed by dedicated isolation system.

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SESSION 7

Materials 2

Le glissement d'une Onde de densité de charge observé par une source XFEL

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Abstract

L'étude du transport de charge à travers les matériaux est essentielle pour de nombreux systèmes électroniques, qu'il s'agisse de semi-conducteurs, de supraconducteurs ou encore de batteries. Il est malheureusement impossible d'observer un mouvement de charges traversant un cristal par des rayons X en raison de la faible section efficace d'un électron presque libre. En revanche, dans les systèmes dits " à Onde de Densité de Charge " (ODC), le transport est accompagné par une déformation du réseau cristallin. Nous montrerons comment il est possible d'observer le transport dans ces matériaux à travers le comportement de la structure atomique sous-jacente en utilisant un faisceau X intense et cohérent généré par un Laser à Electrons libres (XFEL). La phase de l'ODC a été obtenue à partir des images de diffraction obtenue à LCLS en appliquant un algorithme génétique. L'ODC se déforme non seulement dans la direction de la force appliquée, mais aussi par cisaillement dans la direction transverse. Les déformations transversale et longitudinale sont étroitement couplées, la relaxation de l'une au-dessus entraînant l'apparition de l'autre. L'onde glisse sur l'ensemble de l'échantillon mais reste cependant piégée par des marches de surface. Ce résultat illustre les capacités des nouvelles sources XFEL qui, associées à des méthodes d'analyse appropriées, permettent d'étudier les systèmes électroniques.

*Speaker

Unveiling charge dynamics in 3D topological insulator via time- and angle-resolved photoemission spectroscopy

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Abstract

Three-dimensional topological insulators (3D TIs) are characterized by a topologically protected spin-polarized metallic surface states within an insulating bulk energy gap. Such properties position TIs as prime candidates for advanced technologies such as photogalvanic current generation, spintronics, quantum computing. However, unlocking these applications hinges on a deeper understanding of non-equilibrium charge dynamics, particularly the microscopic interaction between surface and bulk electronic states. As an emblematic example, a recent study by Mori et al. reported a long-lived topological exciton in 3D TIs, arising from the interaction between an electron in the surface states and a hole in the bulk bands. Here, we employ time- and angle-resolved photoemission (TR-ARPES) to investigate the ultrafast dynamics in p-doped Bi2Te3 following mid-IR pump excitation. We observe a long-living (> 20 ps) intensity build-up above the Fermi level, consistent with Mori et al. By varying probe polarization, pump fluence, and temperature, we unveil the contribution of bulk bands on the formation of the long-living charge accumulation, thus emphasizing the critical need to disentangle surface and bulk contributions in TIs and guiding future efforts to harness their potential in robust quantum devices.

*Speaker

Correlations drive the attosecond response of strongly-correlated insulators

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Strongly correlated materials present peculiar behavior of both technological and fundamental interest, due to repulsive electron-electron interactions. Yet, their response to light excitation remains unclear. The challenge is that purely electronic dynamics requires attosecond temporal resolution. At this timescale, do correlations enhance, screen or have no effect on the sub-optical-cycle response to an external laser field? We address this question via attosecond transient reflectivity on NiO, a prototypical strongly correlated insulator. We first benchmark our setup with MgO [1], a weakly correlated insulator. It exhibits oscillations at twice the pump laser frequency, a signature of the well-studied dynamical Franz-Keldysh effect by which the field coherently distorts the bands of the solid [2]. Strikingly, NiO does not show any sub-optical-cycle dynamics. Instead, it presents a much slower and non-reversible electronic response that arises in a few femtoseconds. To understand this temporal response we conduct state-of-the-art calculations in the framework of time-dependent density-functional plus self-consistent U theory, which describes ultrafast dynamics in strongly correlated systems. Our results [3], which reproduce the experiment quantitatively, explain this behavior as a light-induced renormalization of the correlation parameter U in the Hubbard model, confirming predictions made in prior theoretical work [4]. With this interpretation, the data yields direct access to the temporal evolution of the effective Hubbard U for the first time. It is found to respond in 7 fs, a system-specific timescale characterizing the dynamics of electronic screening after light excitation. The ability to measure and control the Hubbard U at its true timescale could be foundational for accessing and harnessing new non-equilibrium material states.

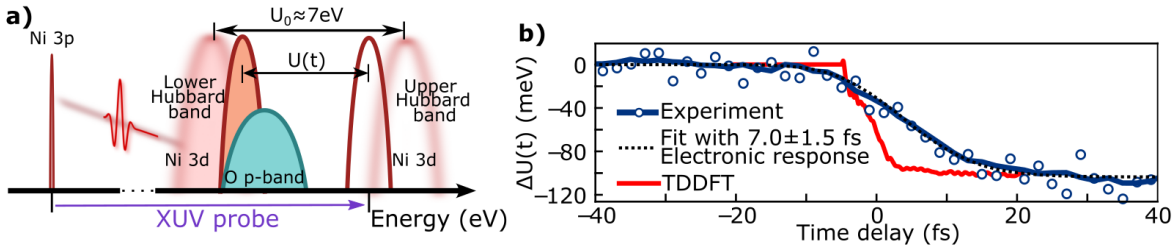

Figure. a) The attosecond XUV pulse probes the energy of the conduction band which is the Upper Hubbard Band, providing a direct measurement of the Hubbard U evolution as a function of time. **b)** Comparison of the measured $\Delta U(t)$ and our numerical results.

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POSTERS

A new multi-scale femto-millisecond transient absorption spectroscopy platform.

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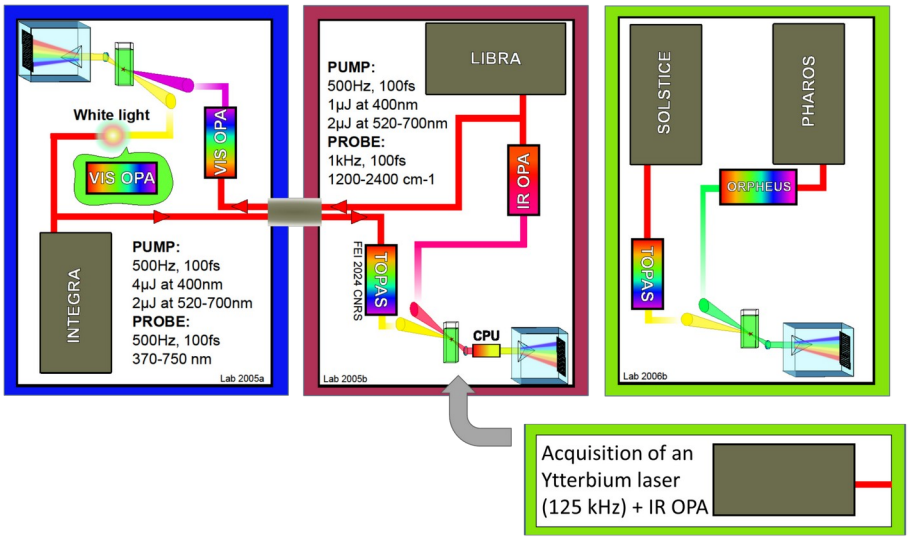
The investigation of complex samples, such as biomolecules or advanced materials, benefits significantly from optical spectroscopy techniques capable of multi-timescale and large spectral bandwidth access. The first steps of light-energy conversion of photosensitive samples involve typically isomerization, energy or charge transfer processes that take place on the femtosecond-nanosecond scale. They are usually probed using femtosecond optical pump-probe transient absorption. Back electron / charge recombination or longer photoinduced structural changes that occur in the micro-milliseconds temporal domain are generally monitored using nanosecond flash photolysis. Despite the clear need of experiments extending over broad time ranges, integrated multiscale setups are not available in most spectroscopy laboratories, and two different setups with different sample conditions and excitation laser pulse widths are used for monitoring short and long timescales. In view of the complexity of multiscale methods [1-3], some European laser centers have built setups based on two Ti:Sa systems synchronized in master-slave configuration; these are available to users through beamtime calls.

At Laboratory for Optics and Biosciences (LOB), we have developed a new method, coined ADASOPS for Arbitrary Detuning ASynchronous OPtical Sampling, which allows a single multiscale setup to be readily obtained from two pre-existent femtosecond amplified systems. The method is based on an optoelectronic implementation using Time to Digital Converters (TDC) [4-5], and has been demonstrated on the study of the dynamics of fatty acid photodecarboxylase using picosecond-microsecond IR transient absorption spectroscopy [6].

In 2024 we have responded to the second round of funding for the ULTRAFAST project of the LUMA PEPR [7] in order to offer multiscale experiments for the national scientific community. Three different setups will be available allowing sub-picosecond-millisecond UV-Vis and MIR transient absorption measurements employing two 1-kHz pump-probe experiments already in use, and a 1-kHz pump / 250-kHz probe device presently under development. The 1-kHz setups, both based on the same Ti:Sa lasers are a) a visible pump / IR probe transient absorption experiment with 1 cm^{-1} spectral resolution and a sub-ps to ms time range for a 1200-2400 cm^{-1} IR probe spectrum, and b) a visible pump – visible probe transient absorption experiment extending form 370 nm to 750 nm. The multi-kHz multi-probe experiment is based on the use of a Ytterbium laser producing large bandwidth UV-VIS-NIR probe pulses from 220 nm to 2000 nm.

The current (blue and red squares) and future (green squares) experiments are schematically depicted in the Figure.



Schematic drawing summarizing current (blue and red square) and future (green square) experiments open to the French scientific community under the LUMA Ultrafast project.

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Ultrafast dynamics of a spin-polarized electron gas

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Abstract

Ultrafast magnetism is an actively explored research area in the general framework of ultrafast phenomena. For instance,
the demagnetization of a thin magnetic layer by femtosecond laser pulses and its fundamental origin are still under
debate. Ultrafast magnetic phenomena are also crucial to devise and implement effective THz emitters.
In order to describe the coupled charge/spin dynamics in a ferromagnet, a great variety of models are available, going
from the time-dependent spin density functional theory to quantum hydrodynamic models. An alternative approach consists
in modelling the electrons with a phase-space approach similar to that used in plasma physics. In this approach, the electrons
are described by a probability distribution function in the phase-space (x, v) . For particles with spin, four distribution
functions are required: one for the density of electrons in the phase space f_0 (x, v, t), and three for the spin density f_i (x, v, t),
where i = (x, y, z) denotes the spin direction. These distribution functions obey an evolution equation similar to the Vlasov
equation of plasma physics, coupled to a Poisson equation for the mean-field Coulomb potential . This constitute the
itinerant part of the magnetism. In addition, the fixed-ion magnetism is described by the Landau-Lifschitz equation, coupled
to the above spin-Vlasov equation through an exchange term of the Rundermann-Kittel-Kasuya-Yosida (RKKY) type.
Our work is aimed at studying the dynamics of magnons in an infinite ferromagnetic material. In particular, we investigate the effect of the electron dynamics on the damping and propagation of the magnons. We observe that the magnon
amplitude is modulated nonlinearly under strong excitation of the electron dynamics. Tuning the electron polarization at

*Speaker

equilibrium η (e.g., through an external magnetic field) can affect the magnon's behavior, leading to either a demagnetization of the ferromagnet or a Gilbert damping of the magnon, depending on the value of η. Thanks to our model, one can have access to the threshold between these regimes. As a first result, we obtained the η-dependency of the magnon evolution.

For an electron polarization consistent with that of the fixed ions ($\eta > 0$ and above a certain threshold), the magnon amplitude decreases with time, in a sort of electron-induced Gilbert damping. In contrast, for low or negative η (corresponding to an "unnatural" polarization of the electrons induced by an external field) the magnon energy increases in time and leads to the demagnetization of the sample. These effects may be used for the ultrafast control of magnonic behaviours through the application of a femtosecond electromagnetic pulse.

Development of a multi-timescale and multi-probe transient absorption setup combining fs Ti:Sa and Yb laser sources.

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Abstract

Ultrafast pump-probe spectroscopy enables the study of molecular dynamics across a wide range of timescales, from femtoseconds to milliseconds. However, conventional setups that use mechanical delay stages to control pump-probe delay have a temporal window limited to few nanoseconds, making them unsuitable for investigating biomolecules with dynamics spanning femtoseconds to milliseconds.
To overcome this limitation, we have developed a multiscale spectroscopic setup based on the Arbitrary Detuning Asynchronous Optical Sampling (ADASOPS) method (1). ADASOPS allows the precise determination of pulse pair delay from two independent femtosecond laser sources with a precision of 500 fs, without any condition on their repetition rate. The method has been already successfully applied for carrying out transient absorption measurements using two femtosecond Ti:Sa oscillators (2) and two amplified kHz Ti:Sa laser sources (3). The present project aims at the development of a transient absorption setup, combining a high-repetition-rate Yb laser as the probe and a kHz Ti:Sa laser as the pump, similarly to a previously-reported approach (4). As a first step, we successfully synchronized a nanosecond diode laser as the pump with a Yb laser as the probe, running at a repetition rate of 1 kHz and 125 kHz, respectively. Combining a low-repetition rate pump with a high-repetition rate probe has the advantage of enabling the measurement of the transient spectra at 8 ms intervals after excitation, corresponding to an approach known as multiple probing (5).

To test our setup, we performed visible broadband transient absorption measurements on carboxymyoglobin (MbCO) in solution. Upon CO dissociation from the heme group, MbCO undergoes structural changes that span from femtoseconds to milliseconds, making it an ideal model for multiscale ultrafast spectroscopy.

Our results show that our setup can track the molecular dynamics over a broad range of timescales with a precision of 10 mOD.

*Speaker

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Compact laser-driven X-ray source for imaging and time-resolved measurements

Benjamin Barbrel*[1], Aboubakr Bakkali[2], Emile Barjou , Antoine Courjaud[3], Octave Delorme , Fabien Dorchies[4], Pauline Lehoux[1], Francesca Mastropietro[2], and Panyi Song[1]

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Abstract

We report advances in the development of a compact and robust laser-driven hard X-ray source. We describe the source performance in terms of stability, photon flux, endurance and source size using a 1 kHz, 25 W, Yb:YAG laser.
The laser-to-X-ray conversion module is a table-top vacuum chamber comprising a focusing optics, a laser target in the form of a continuously moving metallic tape, target motorizations, as well as a radiation shielding.

To characterize our system for X-ray imaging applications, we developed a micro-CT imaging bench. We demonstrate the source performance by realizing a tomographic sequence in under 15 min total exposure time.
Such a pulsed and compact X-ray source also constitutes a promising tool for time-resolved X-ray science. Indeed, it does provide an interesting laboratory-scale alternative to large facilities (synchrotrons and free-electron lasers) for X-ray experiments requiring picosecond or sub-picosecond temporal resolutions, such as time-resolved diffraction, imaging or spectroscopy.

*Speaker

Elucidation of Ultrafast Dynamics in Xanthone using Gas-Phase Femtosecond Photoionization Spectroscopy

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Xanthone (9H-xanthen-9-one) is an aromatic ketone possessing an uncommon intersystem crossing quantum yield close to 1 in solution [1], widely used for applications necessitating triplet-triplet energy transfer. The relaxation dynamics underlying the efficient triplet formation have been measured with time-resolved spectroscopy in various solvents [2-4]. However, in the isolated molecule, the direct observation of the relaxation dynamics of the bright S_2 state remains unexplored. In addition, simulations of the ultrafast dynamics have been performed recently but significantly disagree on the relaxation mechanism [5, 6].

To solve this controversy, we perform femtosecond time-resolved mass spectrometry and photoelectron spectroscopy in gas-phase xanthone after $S_0 \rightarrow S_2$ excitation at 322 nm. Single-photon ionization at 266 nm or multiphoton ionization at 800 nm is used to probe the relaxation dynamics. We measure an ultrafast decay of the S_2 state on the ~ 130 fs timescale. By comparing the response of the molecule with the ionizing probe pulse polarization either parallel or perpendicular to that of the pump pulse, the polarization anisotropy reveals a change of symmetry of the electronic state on the same timescale, indicating internal conversion to S_1. A second ~ 2 ps timescale is measured and attributed to an El-Sayed allowed intersystem crossing. This relaxation mechanism is in agreement with the simulations of Ref. [6].

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Guide d'onde plasma généré par filamentation d'une impulsion laser femtoseconde térawatt dans l'air

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[1]LOA – ENSTA Paris, CNRS, École Polytechnique, Institut Polytechnique de Paris – France

Abstract

Guide d'onde plasma généré par filamentation d'une impulsion laser femtoseconde térawatt dans l'air
Nicolas Cantonnet-Paloque**1, Thomas Clark1, Victor Perez-Ramirez2, Dave Sandeep Singh2, Arnaud Couairon3, Matthew R. Edwards2, Aurélien Houard1**

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Les rayonnements térahertz sont utiles pour de nombreuses applications, notamment en imagerie médicale non invasive (1) et dans le domaine de la sécurité, par exemple pour le contrôle des passagers dans les aéroports (2), en raison de leur capacité à traverser les matériaux secs non conducteurs comme le plastique, le papier ou le tissu. Cependant, leur propagation sur de longues distances demeure complexe en raison de leur forte absorption par la vapeur d'eau et de leur grande divergence. Une solution à ce problème pourrait résider dans l'utilisation d'optiques plasma, et plus spécifiquement de guides d'onde plasma générés dans l'air. Parce leur fréquence de Langmuir se situe dans le domaine GHz-THz, ces plasmas atmosphériques peuvent induire de fortes variations d'indices à ces longueurs d'onde permettant de réaliser l'équivalent de fibres creuses ou de fibres à saut- d'indice (3).

Dans cette étude, nous proposons de démontrer le guidage de rayonnements térahertz au sein d'un guide d'onde plasma généré dans l'air à l'aide d'un laser femtoseconde. Ce guide repose sur la capacité des impulsions laser ultra-courtes à créer des canaux de plasma de plusieurs mètres de long par filamentation laser dans l'air (4, 5). Ce concept a déjà été exploré numériquement pour des ondes micro-ondes, mais les tailles de guide considérées n'étaient pas expérimentalement réalistes (3, 6). À ce jour, une seule expérience a pu mettre en évidence l'effet d'un tube de filaments de 45 mm de diamètre sur la propagation d'une onde à 10 GHz, mais la faible distance de propagation (16 cm) n'a pas permis de confirmer un guidage (7). Démontrer le guidage d'ondes THz ou micro-onde constituerait donc une avancée majeure dans le domaine.

Ce poster résume la première étape de notre projet qui consiste à étudier numériquement

*Speaker

le guide plasma pour déterminer une configuration réalisable expérimentalement et la plus efficace possible. Une démonstration expérimentale sera réalisée dans un second temps à l'aide du laser de 10 TW de puissance crête du Sapphire laboratory.

Pour optimiser les paramètres du guidage, deux codes de simulation ont été utilisés. Le premier permet de modéliser les modes d'ondes pouvant se propager dans un guide plasma en fonction des paramètres du guide (dimensions, densité). On cherche ainsi à obtenir la plus grande distance d'atténuation des térahertz (8). En parallèle, un second code plus lourd a été utilisé afin d'analyser plus finement la propagation linéaire d'un faisceau térahertz dans ces guides (9), fournissant des estimations plus précises des longueurs d'atténuation. Ces simulations nous ont permis de définir une configuration très prometteuse, dont la validation expérimentale est prévue pour fin 2025.

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Floquet-Bloch Valleytronics in TMDCs

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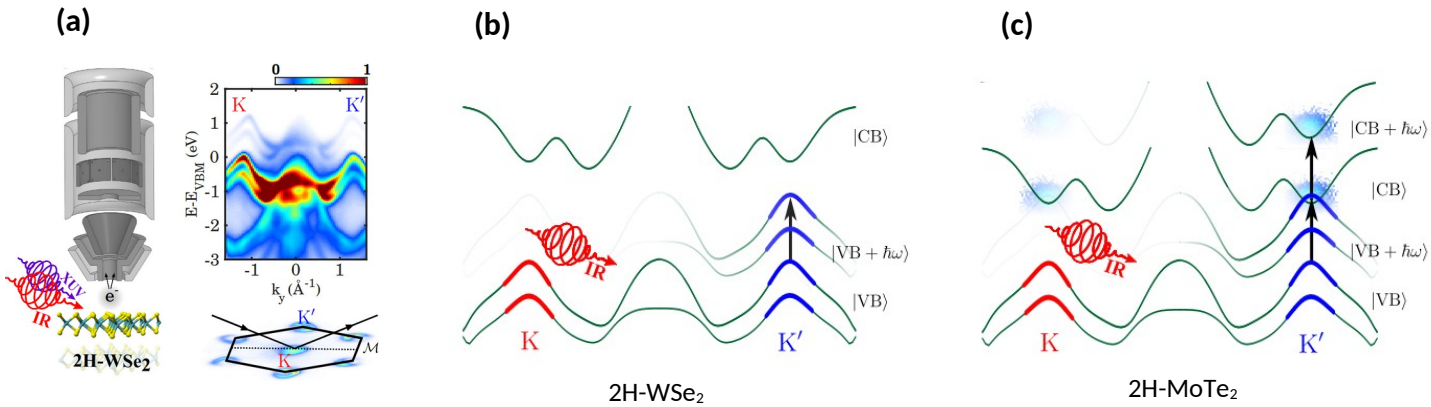
Driving quantum materials out of equilibrium enables the realization of exotic states of matter beyond conventional equilibrium tuning methods. When electrons are periodically driven by electromagnetic fields, Floquet-Bloch states emerge, allowing for the creation of nonequilibrium quantum phases without equilibrium analogs [1,2]. In transition metal dichalcogenides (TMDCs), the broken inversion symmetry in monolayers gives rise to a nonzero Berry curvature at the K and K' valleys, leading to chiroptical selection rules central to valleytronics [3]. Here, we establish a connection between Floquet engineering and valleytronics.

First, using below-bandgap pumping, we demonstrate the formation of valley-polarized Floquet-Bloch states in 2H-WSe_2. We reveal quantum path interference between Floquet-Bloch and Volkov states, showing its dependence on valley pseudospin and light polarization. Extreme ultraviolet (XUV) photoemission circular dichroism in this nonequilibrium regime further illustrates the potential for orbital character control in Floquet-engineered states.

Second, under resonant pumping of 2H-$MoTe_2$, we observe the emergence of Floquet-Bloch states not only from the occupied valence band but also from photoinduced excited states. A ~20 fs delay between the formation of Floquet-Volkov states from valence and conduction bands highlights the interplay between photoexcitation and light-field dressing by the infrared (IR) pump pulse. Furthermore, we demonstrate valley-selective Floquet-Bloch states formation from excited states.

These findings bridge Floquet engineering and quantum geometric light-matter coupling in two-dimensional materials. They provide a pathway toward novel nonequilibrium phases of matter by dynamically breaking symmetries through coherent dressing of Bloch electrons with tailored light pulses.



Experimental setup and concept of Floquet Valleytronics. (a) Two polarization-tunable pulses are focused onto the sample (2H-WSe_2) in the interaction chamber of a time-of-flight momentum microscope: an IR pump (1.2eV, 135fs) and an XUV probe (21.6eV). The energy-momentum cut along K-Γ-K' at the pump-probe overlap shows the emergence of a sideband at the energy of the valence band plus the pump photon energy. The modulation of the IR pump polarization allows the creation of Floquet-Volkov states to be investigated while the modulation of the XUV probe polarization allows circular dichroism in the photoelectron angular distribution from light-driven states to be recorded. **(b)** Below-bandgap pumping: upon periodic drive using chiral light pulses, time-reversal symmetry is broken and valley-polarized Floquet states are created. **(c)** Resonant pumping: Floquet and Volkov states are created upon periodic drive from occupied states; light-dressed states are also created from photoinduced excited states.

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Ultrafast laser activation of methanol in SAPO-34 zeolite: a fs MIR pump-probe study

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Introduction

The transition toward a decarbonated and more sustainable industry encompasses the development of light-assisted catalytic routes for the conversion of C_1 and C_2 molecules into olefins, fuels and high-value chemicals. The light-induced temperature jump (T-Jump) methods is an emerging alternative to the conventional photocatalysis.. By confining in time and space the thermal activation, this approach would allow for more selectivity and lower energy consumption. The challenge in the field is the control of the ultrafast events along the light-to-chemical energy conversion and transfer from the absorption of the photons to the activation of catalytic centres and reactants. The ultrafast T-jump in zeolite can be triggered by the direct excitation of selected vibrations of the framework [1,2], or via the electronic excitation of metal nanoparticles [3], for example. By performing MIR femtosecond transient absorption measurement completed by FTIR and EPR spectroscopy, we are investigating the first events of the light-assisted conversion of methanol into reactive radical species in SAPO-34 zeolites. We triggered the ultrafast T-jump by exciting the samples at 6900 cm^{-1}, in resonance with an overtone of the OH stretching modes of the zeolite framework. The energy transfer following the photoexcitation of this band in SAPO-34 is followed by femtosecond NIR pump / MIR probe measurements reported in **Figure 1** in vacuum and in presence of methanol. We measured the ultrafast photodynamic for the different SAPO samples. To prove that the femtosecond laser excitation at 6900 cm^{-1} can potentially activate the MTO conversion, the SAPO sample were photo-irradiated by the NIR fs laser pulses at 6900 cm^{-1} in vacuum and under methanol pressure and then they were characterized by EPR spectroscopy.

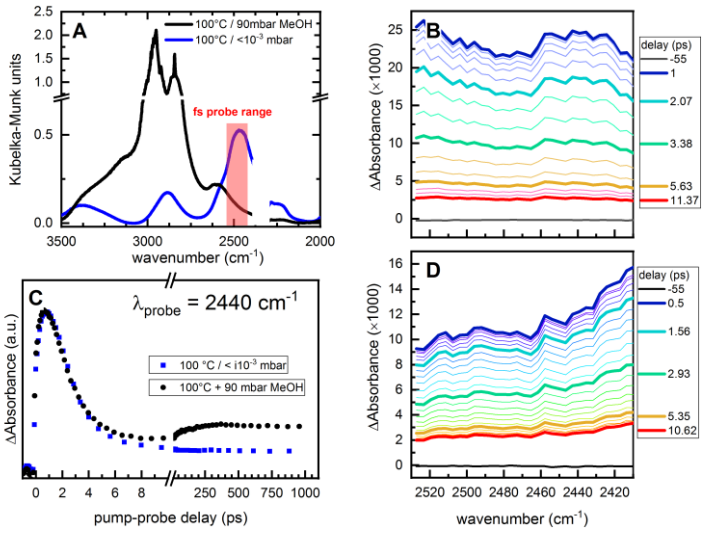

Figure 1 Steady state and femtosecond infrared characterization of SAPO-34 (S-2) sample before and after methanol adsorption at 100 ° C – (A) DRIFT spectra, the red area corresponds to the fs-probe spectral range. (B,D), Femtosecond transient absorption spectra recorded for λ_{pump} = 6900 cm^{-1} , under vacuum, and under 90 mBar of methanol, respectively. (C) Kinetic traces reconstructed from the transient data (B) and (D).

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Shaping ultrashort spatiotemporal optical vortices from post-compressed laser pulses

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Abstract

We report on our experimental generation of spatiotemporal optical vortex (STOV) pulses using a custom 4f pulse shaper, and their characterization via spatially-resolved Fourier-transform spectral interferometry. We achieve intense, _˜50 fs infrared STOV pulses, demonstrating for the first time their feasability on a post-compressed laser setup.

*Speaker

Ultrafast Photoswitching and Fast Thermal Recovery in Negative Red-Absorbing Photochromic Flavoprotein-Ligand Charge Transfer Complexes

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Abstract

Photochromic proteins are reversible photoswitchable macromolecules that undergo conformational and spectral changes upon light absorption. Recovery can be photo- or thermally induced. The switching process is typically triggered by photoisomerization of the chromophore, like in bacteriophytochrome, rhodopsin or reversible fluorescent proteins (1). Fast reversible red-absorbing photochromic systems with high quantum yield (QY) are rare, yet they hold immense potential in life sciences, particularly for applications in optogenetics and biotechnology (2, 3). One unique system was recently discovered in monomeric sarcosine oxidase (MSOX), where a flavin cofactor forms an intermolecular charge transfer (CT) complex with a substrate-analogue inhibitor ligand. This complex undergoes ultrafast, barrierless dissociation associated with ligand isomerization on a 100-fs timescale, achieving near-unity QY and sustaining a nanosecond photoproduct lifetime at room temperature (4). The activation barrier for return to the initial state was hypothesized to be controlled by the mass of the ligand.
We investigated the ligand-mass dependent dynamics of MSOX-ligand CT complex photoswitching. We employed a combination of conventional optical pump-probe transient absorption spectroscopy (limited to the nanosecond time scale) and multi-timescale transient absorption spectroscopy using Arbitrary Detuning ASynchronous OPtical Sampling (ADASOPS). This technique enables precise control over pump-probe delays, spanning timescales from picoseconds to milliseconds (5). These experiments allow a comprehensive investigation of both the photoinduced switching process and the thermally driven recovery. Using two ligand variants with different molecular masses-methylthioacetate (MTA) and methylselenoacetate (MSeA)-we determined a CT complex dissociation QY of _˜76% for both systems. They share identical quantum yield and, contrasting with the hypothesis of mass dependence, identical activation energy for thermal recovery. Yet, they exhibit markedly different recovery rates. This suggests that the mass of the isomerizing atom alters the potential energy surface without modifying the activation barrier.

By integrating structural techniques and computational simulations, we aim to further elucidate the electronic and vibrational states governing these ultrafast dynamics of photoswitching. By designing MSOX mutants and engineering heavier ligands, we aim to extend

*Speaker

photoproduct lifetimes and introduce fluorescence, ultimately advancing the development of red-absorbing negative photochromic systems with recovery in the micro – millisecond range for biological and technological applications.

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High repetition rate attosecond beamline for photoemission spectroscopy

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Synopsis We present a new experimental setup dedicated to attosecond photoemission spectroscopy. A post-compressed high-power Ytterbium laser source enables the experiment to run at a high repetition rate while a 3-beam architecture allows a pre-excitation of the medium and its photoemission by an XUV attosecond pulse train in the presence of an IR dressing beam.

The beamline is driven by a new 80-W Ytterbium laser source providing 340 fs pulses, each carrying 2 mJ of energy at a reprate of 40 kHz. A post compression stage based on a multi-pass cell filled with argon shortens the pulses down to 21 fs with 1.8 mJ energy per pulse [1].

The optical setup primarily consists of a 3-arm Mach Zehnder interferometer. An amplitude division of the beam on a beamsplitter precedes a wavefront division on a drilled mirror (DM). The most energetic of the three beams generates high-order harmonics in the form of an attosecond pulse train. The beam with intermediate power has an annular shape from its reflection onto the DM and will be used to pre-excite the medium, e.g., to induce molecular alignment. Finally, the weakest beam -that went through the DM- will be the dressing beam. The recombination of the beams is realised in two steps, each with a DM. The delays between all the pulses are controlled and actively stabilized using parts of the beams that are collected at the last DM.

The size and intensity of each beam at the key locations of HHG and photoemission foci were computed using propagation simulations. They show satisfactory results with respect to the experimental conditions required to perform photoemission spectroscopy using RABBIT [2] and Mixed-FROG [3] techniques.

As for the end-stations, the inline two foci geometry [4] enables simultaneous measurements in two magnetic bottle electron spectrometers. One of them can be replaced with a velocity map imaging spectrometer to access the electron/ion angular distribution. RABBIT measurements can then be performed with a reference gas spatially separated from the studied gaseous molecule or liquid target.

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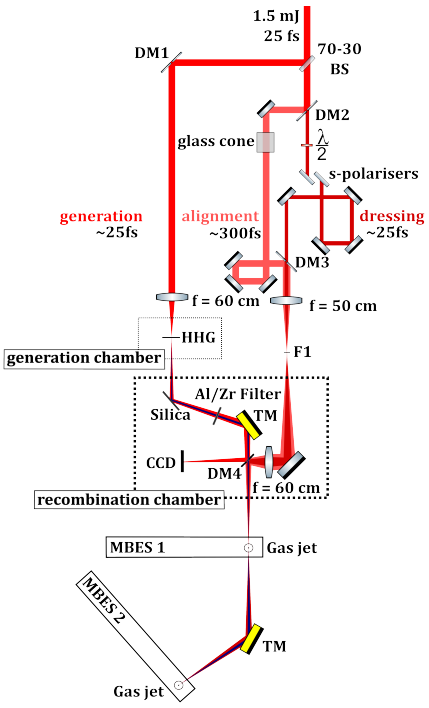

Figure 1. Experimental setup. DM : Drilled Mirror, TM : Toroidal Mirror, MBES : Magnetic Bottle Electron Spectrometer.

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AttosecondRayTracing : une bibliothèque Python pour concevoir des lignes de transport d'impulsions attosecondes

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Lors du transport d'impulsions attosecondes, les couplages spatio-temporels peuvent vite dégrader le profil temporel de l'impulsion. Ces couplages peuvent être inhérents aux optiques utilisées ou peuvent survenir à cause d'un mauvais alignement de la ligne de lumière. Dans les deux cas, il est souhaitable de pouvoir quantifier la qualité de transport de l'impulsion en amont de l'installation de l'équipement.

Une des approches ([1,2]) pour simuler les propriétés d'une ligne de lumière est le lancer de rayons (raytracing). C'est l'approche utilisée par d'autres logiciels tels que Zemax ou Code V. Cependant, ces logiciels sont difficiles à prendre en main et nécessitent une certaine expertise pour les utiliser correctement. En plus, ils n'offrent pas naturellement accès aux temps d'arrivée des différents rayons, ce qui est critique pour correctement estimer l'impact sur les impulsions attosecondes.

Pour ces raisons, on présente AttosecondRayTracing (**ART**) : une bibliothèque Python permettant de simuler sequentiellement le transport d'impulsions attosecondes. Une fois installé avec **pip**, utiliser ART est aussi simple que de modifier un script Python créant les miroirs et de l'exécuter. Le code entier étant en Python, il est facile de modifier ou d'étendre le code, de rajouter de nouveaux types de miroirs ou d'optimisations.

Je vais présenter le code et les diverses fonctionnalités qui le rendent utile pour les étapes initiales de la conception d'une ligne de transport d'impulsions attosecondes.

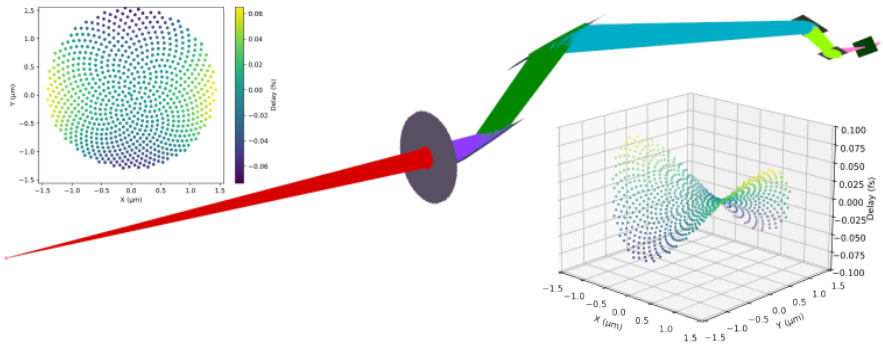

Fig. 1 : Exemple de ligne de transport attoseconde légèrement désalignée

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Yb-based cascaded multi-pass cell post compression setup for kHz laser-plasma acceleration

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Laser-plasma acceleration (LPA) has been moving towards mainstream acceptance with an increased R&D activity aimed at demonstrating reliability and control of LPA beam quality, mainly by using active feedback on both the laser and laser-plasma interaction parameters [1]. Driving multi-MeV LPA at kHz repetition rate using few-mJ, few-fs laser pulses obtained from a post-compressed Ti:Sa laser in a gas filled hollow-core fiber, paved the way to demonstrate continuous "hands-off" LPA operation for over 5 hours, with a world record in terms of consecutive shots [2]. Even though this proof-of-principle experiment showed that one could indeed control a kHz LPA for extended periods of time, the deployment of LPA for real-life applications, such as radiotherapy or non-destructive testing, requires industry-grade high-power laser drivers combined with more adapted post- compression schemes such as multi-pass cells (MPC) [3].

In a recent experiment using a 7 mJ, 36 fs commercial Ti:Sa laser (ASTRELLA USP, *Coherent Inc.*), we showed compressibility down to sub-2 cycle duration after propagation in a 3 m-long helium-filled MPC [4]. The overall efficiency of the system is around 60%, which already competes with established hollow-core fiber post- compression schemes [5]. There is currently a strong push towards Yb-based lasers, as they are more compact, deliver higher average power and exhibit much higher wall-plug efficiency than the conventional Ti:Sa systems, making the LPA value chain more efficient and sustainable. With this in mind, we have designed a double- stage MPC post-compression setup tailored to a 17.3 mJ, 350 fs commercial Yb-based laser system (MAGMA 25, *Amplitude*), operating at 1 kHz repetition rate, in order to achieve compression factors close to 100 and reach a few- fs regime, as first demonstrated by Balla et al. [6]. The first 4 m-long MPC stage gas already been implemented, with close to 85% overall compression efficiency, yielding 14.7 mJ energy and 23.6 fs compressed pulse duration (measured with TIPTOE), resulting in a usable peak power of 533 GW.

Still in the design phase, the second MPC stage should enable the generation of spectrally broadened pulses compressible down to few-fs duration with high spatio-temporal quality. To this end, combination of compression ratios [7] and nonlinear ellipse rotation [8] are currently being explored. The usability of the post-compressed pulses will be tested by generating stable few-MeV electron beams at 1 kHz repetition rate from a commercial LPA module (e-KAIO, *SourceLAB*) [9].

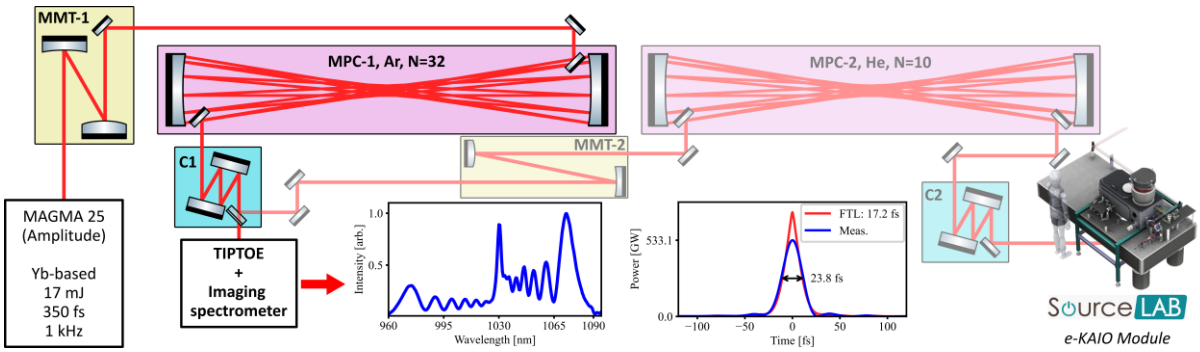

Fig. 1 Scheme of the complete double-stage post-compression setup. MMT: Mode-Matching Telescope, C1-2: Chirped mirror compressors. The transparent part of the scheme is currently being implemented.

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Post-compression of an Yb laser for attosecond spectroscopy

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The improvement of laser' repetition rates using ytterbium-based systems constitutes a significant opportunity for attosecond science. This development is restrained by the long pulse duration (hundreds to femtoseconds) provided by these systems and a priori ill suited to generate attosecond pulses. The concomitant development of efficient post-compression technics [1,2] based on self-phase modulation to achieve spectral broadening and dispersive optics, is an efficient possibility to make these sources suited for attosecond science. In this study, we employ a two-stage hybrid system [3] to post-compress laser pulses at the millijoule level, reducing the pulse duration close to the single optical cycle regime, with an overall transmission efficiency exceeding 45%.

The experimental setup is shown in Fig. 1 (top panel). A carrier-envelope-phase (CEP)-stabilized Yb-doped laser system (Flint and Carbide, Light Conversion) is employed. This laser delivers pulses with an energy of 2 mJ, a pulse duration of 330 fs, and an average power up to 80 W. Post-compression is achieved through two successive stages. In the first stage, spectral broadening is induced via 14 round trips in a homemade argon-filled multipass cell (MPC) [4]. The MPC is 590 mm long and includes mirrors with a 300 mm radius of curvature and low group delay dispersion (GDD). Then, the laser pulse is compressed near its Fourier limit using chirped mirrors (-1720 fs²) and subsequently coupled into a 2.1 m long stretched hollow-core fiber with a helium pressure gradient. Pulse characterization at various stages is performed using a single-shot frequency-resolved optical gating (FROG) device, an imaging spectrometer commercially available (Femtoeasy) and a homemade TIPTOE [5].

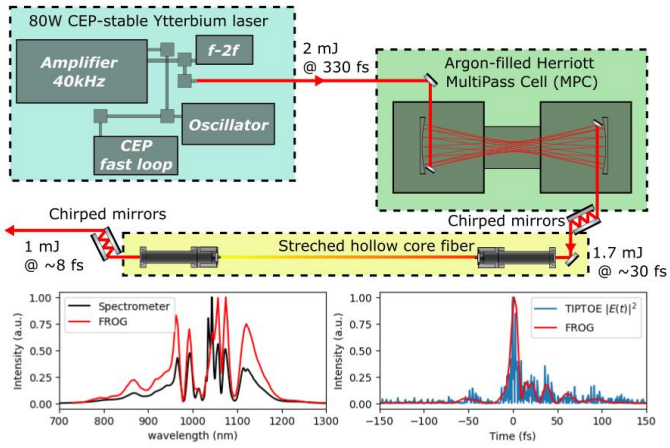

Figure caption. Top: layout of the experiment setup. Bottom: measured spectra and time trace at the hollow-core fiber output

Significant spectral broadening is achieved at the MPC output by adjusting the argon gas pressure. Under optimal conditions, the spectral bandwidth reaches a maximum of 140 nm, allowing for the generation of 20 fs pulses, corresponding to a compression factor of K=16. The MPC also demonstrates a high transmission efficiency exceeding 90%.

In order to seed the second post-compression stage, the argon pressure in the MPC is tuned to produce 25 fs pulses with a well-defined temporal profile. The second stage reaches a transmission efficiency of 60% in the hollow-core fiber, delivering pulses with an energy of 960 μJ

and a duration of 7 fs (Fig. 1 bottom right). By further increasing the gas pressure, we attain a maximum spectral bandwidth that supports Fourier-limited pulse durations as short as 4 fs.

This two-stage hybrid compression system achieves a total compression factor of 47, paving the way for the generation of isolated attosecond pulses. Moreover, the exceptional stability of this setup offers the potential to incorporate an additional nonlinear stage, such as a short hollow-core fiber, for the generation of tunable vacuum-ultraviolet (VUV) and deep-ultraviolet (DUV) [6] pulses.

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Angular momentum dependence in multiphoton ionization and attosecond time delays

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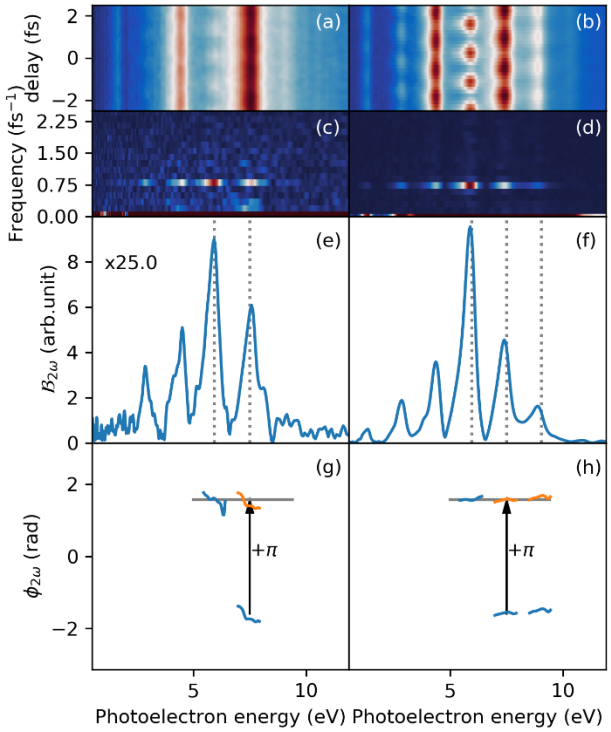
Multiphoton interference measurements in attosecond physics, e.g., the reconstruction of attosecond beating by interference of two-photon transitions (RABITT) [1], are the typical methods of choice to experimentally access the photoionization delay in atoms and molecules. The exact relation between the measurable multiphoton delays and the theoretical single-photon delays is typically modeled by correction terms, continuum-continuum delays, obtained from a high-energy limit of the theory [2]. However, these are unreliable at photoelectron kinetic energies smaller than about 10 eV, and do not have photoemission angular dependence. In this work [3], we develop an accurate analytic alternative that gives accurate correction terms even at very low energies. Our method is computationally very straightforward, predicts correct multiphoton photoelectron angular distributions as well as the expected angular dependence of the continuum-continuum delay. We validate the approach theoretically against state-of-the-art *ab* initio calculations as well as experimentally with a two-harmonic RABITT setup, which properly separates different higher-order multiphoton pathways (see Figure) and offers a promising way of analyzing congested molecular photoionization spectra.



(a, b) RABITT spectrograms obtained in argon by using a set of two isolated harmonics (H13 and H15) for low (a) and high (b) dressing field intensity. (c,d) the corresponding Fourier transform showing a clear oscillation at 2ω (i.e. at 0.75 fs^{-1}). The 2ω oscillation amplitude (e,f) and phase (g,h) in blue. In (g,h), the phase of the main-band and outer-sideband are also presented in orange with a π-shift to be compared to the sideband.

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Few-cycle post-compression of 1.8μm wavelength pulses in multiple plates.

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We temporarily post-compressed a 40 fs, 1.8 μm optical parametric chirped-pulse amplifier (OPCPA) pulse using a multi-pass continuum generation setup [1, 2, 3]. The compression system consists of a moderate-intensity focusing scheme through three yttrium aluminium garnet (YAG) plates, serially arranged at a Brewster angle to minimize reflection losses. Pulse characterization was performed using a home-built frequency-resolved optical gating (FROG), confirming successful compression down to 12 fs, which corresponds to a two-cycle pulse at 1.8 μm. To ensure optimal pulse compression, we carefully controlled the second- (GDD) and third-order dispersion (TOD) using chirped mirror pairs. This dispersion management is particularly crucial in the mid-infrared (mid-IR) range, where most common optical materials introduce positive TOD, requiring compensation to mitigate phase distortions.

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Ultrafast probing of isotope-induced explicit symmetry breaking in ethylene

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Abstract

Symmetry and symmetry breaking have been known to play a central role in contemporary physics and many of the natural phenomena occur due to the breakdown of symmetry (1,2). Here, we show how isotope-induced inversion symmetry breaking influences ultrafast photoisomerization processes in ethylene. Extreme ultraviolet pump – near infrared probe time-of-flight mass spectrometry, performed replacing one of the carbon atoms in ethylene with a 13C isotope leads to twice-faster structural relaxation in the photo-excited molecular cation. In particular, the ethylene to ethylidene conversion, *i.e.*, the relaxation channel involving the migration of one hydrogen atom from one carbon atom to the other, also identified in previous studies (3), was affected by the isotopic effect. We simulated the quantum-classical non-adiabatic molecular dynamics, using trajectory surface hopping method, and incorporating the nuclear symmetry. Our results suggest that this difference arises from the mixing of different normal modes in the isotope-substituted species, 13C-ethylene, compared to the more symmetric unsubstituted or doubly substituted isotopologues. This facilitates efficient intra-molecular vibrational energy redistribution, thereby lowering the isomerization yield. These findings offer opportunities to use isotope-induced nuclear symmetry breaking to control the outcome of light-molecule interactions across ultrafast timescales. (1) Bardeen, J., Cooper, L. N., & Schrieffer, J. R. Microscopic theory of superconductivity. Phys. Rev. 106, 162 (1957).
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*Speaker

Beamline development optimized for STARPES : a highly tunable extreme ultraviolet lightsource

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Abstract

We present the upgrade of an attosecond beamline, which was previously using a Ti:Sa laser source. We increase the repetition rate from 10 to 100 kHz by using an industrial-grade Yb-doped laser system. Based on the High Harmonic Generation (HHG) process, the beamline is optimized for Spin-, Time-, and Angular-Resolved Photoemsission Spectroscopy (STARPES) in solids and gases. Its design allows the tuning of the pulse duration from 300 fs to 25 fs as well as switching between the full extreme ultraviolet (XUV) spectrum or monochromatized harmonics down to 200 meV bandwidth.

*Speaker

Effect of the Pt Loading on the Ultrafast Charge Carrier Dynamics in Pt-NPW-TiO_2 Catalysts for The Conversion of Methane

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The photoconversion of methane (CH_4) into valuable chemicals is crucial for sustainable energy solutions and mitigating greenhouse gas emissions. We employ femtosecond time-resolved UV pump–IR probe spectroscopy to investigate the influence of Pt concentration on free electron lifetimes and recombination dynamics in TiO_2-based photocatalysts modified with heteropolyanion layers and anchored Pt atoms (Pt-NPW-TiO_2) [1]. By conducting measurements in the low excitation intensity regime ($I_0 \leq 0.05$ mJ/cm^2), we isolate the effect of Pt loading on charge carrier kinetics. Our results reveal that at low Pt concentrations (<1 wt%), dispersed Pt atoms have minimal impact on subnanosecond electron lifetimes. However, at higher Pt loadings (5 wt%), electron survival probability increases significantly, likely due to enhanced charge separation facilitated by chemical modifications, including altered charge diffusion, activation energy shifts, or Pt nanoparticle formation. These findings provide critical insights into optimizing CH_4 photoconversion efficiency through precise catalyst design by elucidating the interplay between metal composition, charge dynamics, and reaction conditions, this study aims to establish a strategic framework for designing next-generation TiO_2-based and metal oxide-zeolite photocatalysts for methane valorization.

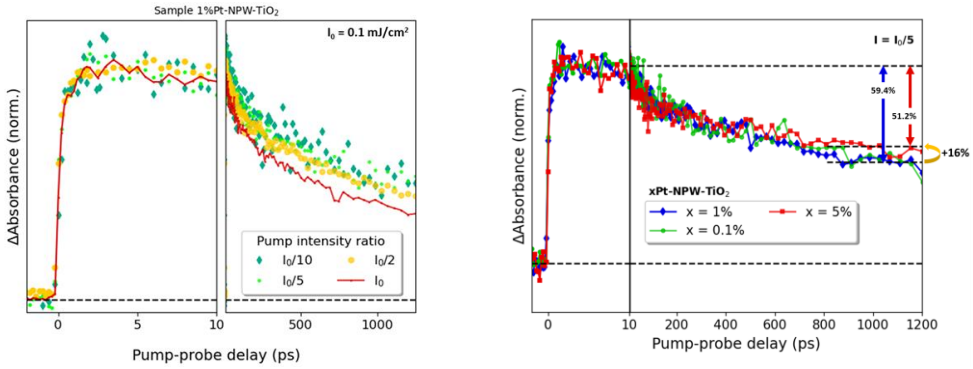

Fig. 1. (left) Kinetic traces for 1%Pt-NPW-TiO_2 with $I_0/10 \leq I_{pump} \leq I_0$. **(right)** Kinetic traces at $I_{pump} = I_0/5$ (linear regime), for xPt-NPW-TiO_2 (x= 0.1, 1, 5 wt%).

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Attosecond photoionization time delay and electron scattering in DNA bases

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Attosecond photoionization time delay measurements provide critical insights into electron scattering dynamics within molecular potentials. The Reconstruction of Attosecond Beating by Interference of Two-Photon Transitions (RABBITT) technique enables the extraction of the phase of outgoing electron wave packets [1]. Following Wigner's interpretation, these phases allow the determination of photoionization time delays [2], granting access to a detailed picture of molecular potentials and electron distributions with angstrom-level precision.

While most previous studies have focused on atomic systems and small molecules, we have developed an experimental setup utilizing high-harmonic generation (HHG) sources to investigate electron dynamics in larger, structurally complex molecules [3,4]. Specifically, we have explored the influence of molecular size and symmetry on electron scattering in two- and three-dimensional systems, such as polycyclic aromatic hydrocarbons (PAHs) and diamondoids [3].

Building upon this approach, we have measured attosecond photoionization time delays in DNA bases, providing insights into their molecular potentials and electron distributions. These findings offer valuable perspectives on the photostability of biomolecules and the mechanisms of radiation-induced damage in biological systems.

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Passively stabilised interferometer with sub-10 as resolution

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Abstract

We present a motorised passively stabilised interferometer based on polarisation separation. It is composed of two mobile birefringent quartz wedges equivalent to a birefringent plate with finely controllable thickness. After a polarizer, precise spectral measurements in the range 700-900 nm provide the wedges thickness with an accuracy better than 0.1 μm. We demonstrate a static stability of the interferometer of less than 10 as over 3 hours. We also show a linear relation between the wedges position and the induced delay. With a quartz compensation plate, this setup allows to introduce delays from -20 fs to +20 fs between the two orthogonal polarisations, with the chirp changing by 10 fsˆ2.

*Speaker

Non-adiabatic electronic relaxation of tetracene from its brightest singlet state

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) can generate multiple charge carrier from a single absorbed photon through singlet fission, making them promising candidates for the development of organic photovoltaics (1). Detailed studies of the excitation dynamics of PAHs following the absorption of a photon in the visible to the UV range can provide fundamental insights into molecular energetics and highlight the role of non-adiabatic couplings in the electronic relaxation.
We present a femtosecond time-resolved photoelectron and photoion study of the free tetracene molecule excited into its brightest singlet state, followed by a fast electronic relaxation onto lower-lying singlet states. This was previously studied theoretically for a few polyacenes using TD-DFTB combined with molecular dynamic simulations of the non-adiabatic relaxations (2). The singlet states populated after relaxation, whose energy is determined from experimental data, have been assigned with the help of high-level ab initio multireference perturbation theory (3).

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*Speaker

Time-resolved spectroscopy of the Singlet Fission process in isolated gas-phase systems

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Abstract

Molecular photophysics provides an understanding of the energetic structure of molecules and the existing intra- and intermolecular couplings, which is essential knowledge for the development of future applications.

In particular, dimers of some acenes, the molecules of interest in this study, exhibit a specific property known as Singlet Fission, a process particularly relevant for the advancement of photovoltaic cells. This phenomenon enables the creation of two charges from the absorption of a single photon by the dimer, thereby increasing the quantum yield.

The study of isolated systems in the gas phase is a crucial step in understanding such interactions and processes at play in complex systems. In this study, we focus on the ultra-fast dynamics of the isolated molecules in gas phase with a pump-probe setup, using a femtosecond laser. We present the results obtained for good candidates for Singlet Fission process such as tetracene and pentacene. Isolated molecules are investigated first. Then argon clusters are used to control the stoichiometry of molecular complexes.

*Speaker

Explicit symmetry breaking of generalized angular momentum by second-harmonic generation in underdense plasmas

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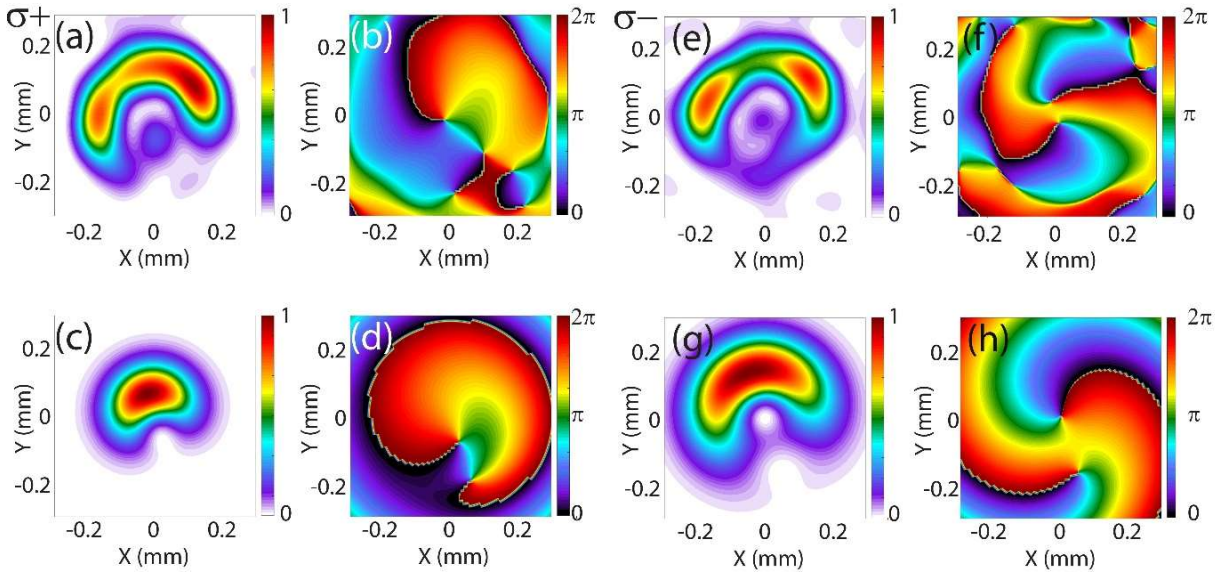
A light beam possesses two intrinsic quantized degrees of freedom: the spin angular momentum (SAM) which depends on its polarization and the orbital angular momentum (OAM) related to its phase transverse distribution. Both combine to give the Total Angular Momentum (TAM). Electric fields built from a particular combination of SAM and OAM are generally not eigenstates of the SAM, OAM, or TAM operators. Instead, they are eigenstates of a newly defined operator known as Generalized Angular Momentum (GAM), which can result in unconventional fractional eigenvalues. The identification of this new form of fractional angular momentum has raised the issue of its physical significance and of its conservation in non-linear regime. The conservation of the GAM has been shown [1] through dipolar transitions where the SAM and OAM are conserved independently. When multipolar transitions are at play (i.e. for dipolar transitions forbidden) the occurrence of spin-orbit coupling can modify the conservation law. In this work [2], we experimentally investigate the conservation law of GAM with second-harmonic generation in an underdense isotropic inhomogeneous plasma that relies on dipole-forbidden interaction. We demonstrate that the field symmetry is disrupted in the nonlinear process, with the GAM charge conserved only on average. This symmetry breaking can provide a detectable signature of the driving field topology or create a robust topological attractor.



SHG field driven by a polarization Möbius strip. Left (resp. right) column correspond to the spatial amplitude (resp. phase). The two first lines correspond to the SHG along the $\sigma+$ circular component with the measurement in (a,b) and the corresponding simulation in (c,d). Similarly, panels (e,f) and (g,h) represent the experiment and the simulation for the $\sigma-$ component.

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