GDR 3754 Ultrafast Phenomena [U.P.]
Groupement pour les Sciences Attosecondes, Femtosecondes et Picosecondes

Club des Partenaires Industriels :

COHERENT, FASTLITE, AMPLITUDE, FEMTOeasy, ITEOX, NOVAELaser, SOURCELAB, NEWPORT.

Réunion Plénière du GDR U.P 2018
10 décembre

09h00-10h00	ACCUEIL/CAFE	
10h00-10h15	**Bureau du GDR U.P**	**Introduction**
DYNAMICS in ISOLATED MOLECULES Chair : Jean-Michel Mestdagh (LIDYL)		
10h15-10h35	**Federica agostini (LCP, Orsay)**	**Dynamics of electrons and nuclei in molecules: Beyond the Born-Oppenheimer approximation**
10h35-10h55	**Jennifer joseph (ISMO, Orsay)**	**Experiments at ATTOlab: Towards time-resolved MFPADs**
10h55-11h15	**Victor Despré (Theo Chemie, Univ. Heidelberg)**	**Attosecond Charge Migration in propiolic acid and dephasing due to nuclear couplings.**
11h15-11h35	**Tsveta Miteva (LCPMR, Paris)**	**Superexchange Interatomic Coulombic decay: an efficient long-range energy transfer**
11h35-11h55	**Marius Hervé (iLM, Lyon)**	**Long-lived Highly Excited States in PAHs Following XUV Excitation**
11h55-12h15	**Sébastien Weber (CEMES, Toulouse)**	**PyMoDaq : un système d'acquisition modulaire et « libre » utilisant Python**

12h15-14h00	REPAS	
ULTRAFAST NANOSCIENCE Chair: Bruno Palpant (LPQM)		
14h00-14h20	**Angela Vella (GPM, Rouen)**	**Etude de l'interaction laser-nanopointes en régime ultra-bref par sonde atomique tomographique**
14h20-14h40	**Anthony Ayari (iLM, Lyon)**	**Field Emission Microscopy of a Quantum Dot at the Femtosecond Scale**
14h40-15h00	**Yoann Zaouter (AMPLITUDE)**	**HHG and attoscience at high repetition rate : status on latest laser developments**
15h00-15h20	**Tatiana Itina (LHC, St Etienne)**	**Etude de la formation de nanostructures périodiques par impulsions laser ultrabrèves en volume des verres**
15h20-15h40	**FASTLITE**	**R&D**
15h40-16h10	PAUSE CAFE	

LASER-PLASMA INTERACTION Chair: Fabien Quéré (LIDYL)		
16h10-16h30	**Julien Fuchs (LULI, Palaiseau)**	**Ion, neutrons accélérés par laser ultra-intenses et applications astrophysiques**
16h30-16h50	**Cédric Thaury (LOA, Palaiseau)**	**L'axiparabole, un nouvel outil pour l'accélération laser-plasma**
16h50-17h10	**François Sylla SOURCE-LAB**	**R&D**
17h10-17h30	**Neil Zaim (LOA, Palaiseau)**	**Interaction relativiste entre lasers de quelques cycles optiques et plasmas sur-denses**
17h30-19h30	Cocktail / Session Poster 1	

11 décembre 2018

09h00-10h30	**CLUB JEUNES CHERCHEURS UP**	
	Margherita Turconi (ARTEMIS, Nice)	***Carrière universitaire en physique ultrarapide : des semi-conducteurs à l'astrophysique***
	Thomas Remetter (KPMG)	***De la science attoseconde à la finance internationale***
	Nicolas Thiré (Fastlite)	***Recherche académique et R&D industriel sur les lasers ultrarapides***
10h00-10h30	Pause café	
DYNAMICS IN MATERIALS Chair : Christian Bordas (iLM)		
10h30-10h50	**Marie Barthélémy (IPCMS, Strasbourg)**	**Femtomagnétisme dans un alliage ferromagnétique sondé par des harmoniques d'ordres élevés : rôle de l'interaction d'échange**
10h50-11h10	**Mauro Fanciulli (LPS, Cergy Pontoise)**	**Spin polarization and attosecond time delay in photoemission from solids**
11h10-11h30	**Sarah Houver (Institute for Quantum Electronics, Zurich)**	**Exploring the band curvature of low-bandgap semiconductors using few-cycle 2D THz spectroscopy**
11h30-11h50	**Franck Vidal (INSP, Paris)**	**Laser driven THz coherent phonons in β-MnAs probed by time-resolved x-ray diffraction**
11h50-12h10	**Alix Volte (IPR, Rennes)**	**Understanding elastically driven cooperativity in molecular photomagnetic materials**
12h10-14h00	REPAS / Session Poster 2	

MOLECULES IN CONDENSED PHASE Chair : Adeline Bonvalet (LOB)		
14h00-14h20	**Michel Sliwa (LASIR, Lille)**	**Photodynamics of a photo-switchable fluorescent protein using UV-Vis transient absorption spectroscopy: remaining questions**
14h20-14h40	**Giorgio Schiro (IBS, Grenoble)**	**Tracking fast protein structural dynamics by X-rays**
14h40-15h00	**Pavel Muller (I2BC, Gif-sur-Yvette)**	**Classical transient absorption spectroscopy extended to sub-nanosecond timescales**
15h00-15h20	**Johanna Brazard (IPCMS, Strasbourg)**	**Towards non collinear UV 2DES setup with pulse-to-pulse phase correction at 50kHz**

15h20-15h40	PAUSE / CAFE
XUV-Xray SOURCES Chair: Sophie Kazamias (LPGP)	
15h40-16h00	**Kevin Veyrinas (CELIA, Bordeaux)**	**Focalisation sans optique d'harmoniques XUV**
16h00-16h20	**Thierry Ruchon (LIDYL, Saclay)**	**Impulsions attosecondes portant un moment angulaire orbital**
16h20-16h40	**Amélie Jarnac (Synchrotron Soleil, Orsay)**	**Demonstration of an efficient 20 ps hard X-ray switch: commissioning work at the FemtoMAX beamline**
16h40-17h00	**Antoine Comby (CELIA, Bordeaux)**	**Génération d'harmoniques d'ordres élevés par des harmoniques d'un laser fibré : une source mW d'impulsions XUV ultra-brèves**

Liste des posters

LUNDI 17H30-19H30

1. Photoemission delays in 2D model molecules
B. Wurmser, M. Labeye, J. Caillat, R. Taïeb

2. High-harmonic generation in solids : insights from ab initio simulations
Nicolas Tancogne-Dejeana

3. High-charge XUV Vortex beam: generation and Hartmann wavefront sensor characterization.
F. Sanson, A. K. Pandey, F. Harms, G. Dovillaire, E. Baynard, J. Demailly, O. Guilbaud, B. Lucas, O. Neveu, M. Pittman, D. Ros, M. Richardson, E. Johnson, W. Li, Ph. Balcou, And S. Kazamias

4. AMPLIFICATION PARAMETRIQUE FIBREE D'IMPULSIONS ULTRA-COURTES
C. Fourcade-Dutin, A. Imperio, P. Roy, R. Jamier, H. Muñoz, P. Perez Milan, H. Maillotte, D. Bigourd

5. Real-time Determination of Enantiomeric and Isomeric Content using Photelectron Elliptical Dichroism
E. Bloch

6. Controlling sub-cycle instantaneous optical chirality in the photoionization of chiral molecules
S. Rozen , A. Comby, S. Beauvarlet , E. Bloch, B. Fabre, V. Blanchet, D. Descamps, S. Petit, B. Pons, N. Dudovich et Y. Mairesse

7. High Harmonic Generation in 2D and 3D semiconductors
Shatha Kaassamani, Dominik Franz, David Gauthier, Rana Nicolas, Jean-Thomas Gomes, Laure Lavoute, Dmitry Gaponov, Nicolas Ducros, Sebastien Fevrier, Willem Boutu and Hamed Merdji

8. Lensless diffractive imaging on ultrafast broadband sources
Julius Huijts, Willem Boutu and Hamed Merdji

9. Orbital Angular Momentum from Semiconductor High-Order Harmonics
David Gauthier, Shatha Kaassamani, Dominik Franz, Rana Nicolas, Jean-Thomas Gomes, Laure Lavoute, Dmitry Gaponov, Sébastien Février, Gaëtan Jargot, Marc Hanna, Willem Boutu, and Hamed Merdji

10. Dynamique de photo-ionisation attoseconde autour du minimum de Cooper de l'Argon
C. ALEXANDRIDI, M. TURCONI, L. BARREAU, D. PLATZER, A. BOROT, J.-F. HERGOTT, O. TCHERBAKOFF ET P. SALIÈRES

11. Impulsions attosecondes isolées par génération d'harmoniques à deux couleurs
Margherita Turconi, Stefan Haessler, Dominique Platzer, Christina Alexandridi, Alice Autuori, Jean-François Hergott, Olivier Tcherbakoff, Antonin Borot, Pascal Salières

12. High energy and high repetition rate few cycle source at 1µm
Loïc Lavenu, Michele Natile, Florent Guichard, Aura Inés González, Xavier Délen, Yoann Zaouter, Marc Hanna and Patrick Georges

13. High repetition rate CEP stable amplifier at 1µm
M. Natile, L. Lavenu, F. Guichard, M. Hanna, Y. Zaouter, R. Chiche, X. Chen, J.F. Hergott, W. Boutu, H. Merdji and P. Georges

14. Strong field optoelectronics in semiconductors and dielectrics
D. Franz, S. Kaassamani, D. Gauthier, M. Kholodtsova, V. Nefedova, S. Fröhlic, N. Ducros, S. Février, G. Jargot, P.Georges, M. Hanna, L. Douillard, W. Boutu and H. Merdji

15. Two-Dimensional Electronic Spectroscopy to study the Ultrafast Photoisomerization of Biomimetic Molecules
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16. Ultrafast excited state dynamics of NHC-Fe(II) complexes lacking light-induced spin crossover
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17. A 100-kHz tunable femtosecond source for spectroscopy from the X-UV to the mid-IR
Nicolas Thiré, Raman Maksimenka, Yoann Pertot, Olivier Albert, Clément Ferchaud, Thomas Pinoteau and Nicolas Forget

18. Photo-ionization and electron-impact-ionization-TOF spectrometry for the detection of large organic molecules embedded in cluster beams
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MARDI 12H10-14H00

19. Photo-induced coherent acoustic phonons in phase change material GeTe.
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20. Emission d'électrons par effet de champ et assistée par impulsions laser ultra-courtes à partir de nano-pointes de diamant
M.-H. Mammez, M. Borz, I. Blum, G. Da Costa, F. Delaroche, J. Houard, A. N. Obraztsov, S. Idlahcen, A. Haboucha, A. Hideur et A. Vella

21. Optimal control of the torsion in ethylene-like molecules
L. H. Coudert

22. Photoionisation attoseconde de molécules chirales
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23. High-Order Harmonic Generation in Solids
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24. Solutions to achieve a UV (300-340nm) 2DES setup in BOXCARS geometry
Thomas Roland, Vincent Kemlin, Jean-Sébastien Pellé, Olivier Crégut, Johanna Brazard, Jérémie Léonard, Stefan Haacke

25. Time Resolved Imaging of Antiferromagnetic Domains Using Second Harmonic Generation in Epitaxial BiFeO3
T . Chirac, J.-Y. Chauleau, S. Fusil, C. Carrétéro, V. Juvé, G. Vaudel, A. Levchuk, P. Ruello, M. Viret

26. Preliminary studies of isolated nanoparticles dynamics: tryptophane and NaCl
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27. Laser-driven Proton Acceleration using two temporally separated Intense Pulses
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28. ULTRAFAST STRUCTURAL DYNAMICS PROBED BY PHOTOELECTRON SPECTROSCOPY
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29. Charge density wave dynamics on GdTe3 probed by Ultrafast Electron Diffraction
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30. Passively Mode-locked (Tm,Ho):YLF Laser and Tm:YLF Laser Mode-locked with a GaAs-based SESAM
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31. Terahertz Assisted Atom Probe Tomography
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32. Multiscale dynamics of denaturation and renaturation of G-quadruplex DNA probed by time-resolved circular dichroism
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33. Electron transfer and charge delocalization dynamics of cyano-aromatic molecules / zeolites composites probed by femtosecond mid-IR spectroscopy
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34. Photoswitching dynamics of the reversible photoswitchable fluorescence protein rsEGFP2: Crystal and solution
Lucas M. URIARTE, Olivier DEVOS, Raffaele VITALE, Cyril RUCKEBUSCH and Michel SLIWA

35. Spectroscopie résolue en temps de nano-objets individuels: effet de morphologie sur les facteurs de qualité vibrationnels
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36. RABBITT at 2w : Attosecond Pulses measurement, attosecond signature in the 1s3p He resonance and ionization delay of Nitrogen
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EXCITED-STATE DYNAMICS WITH TRAJECTORIES

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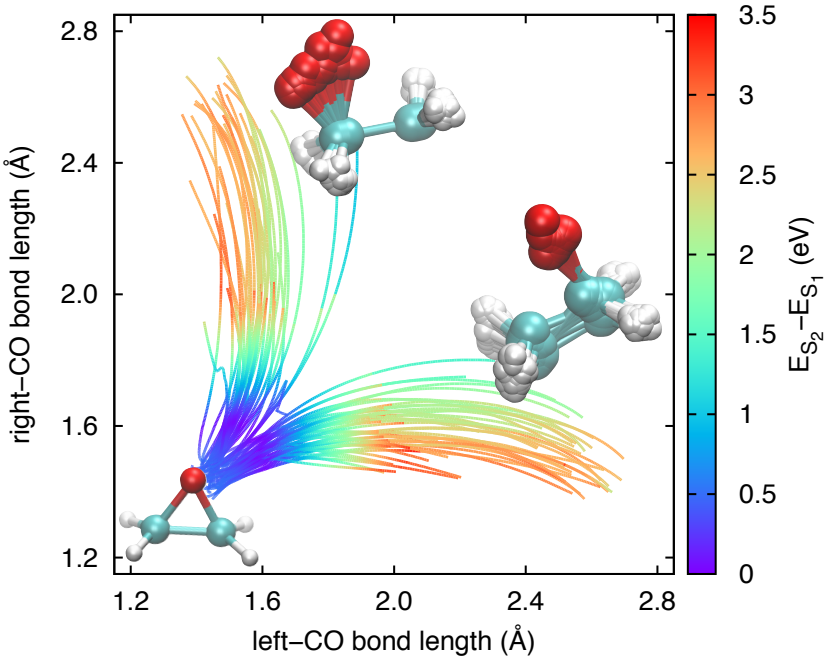
Excited-state dynamics is at the heart of Photophysics and Photochemistry. Nonadiabatic transitions are induced by the strong coupling between electronic dynamics and the ultrafast motion of the nuclei, and are observed in phenomena such as photosynthesis, photovoltaics, and exciton transport in π-conjugated complexes. An essential part of the research efforts in these fields is directed towards developing theoretical and computational approaches to describe conformational changes, energy dissipation, or quantum decoherence, i.e., the signature aspects of excited-state processes. In this context, among the most successful frameworks for molecular dynamics simulations of excited-state processes stand *trajectory-based quantum-classical methods*, as they give access to the study of complex molecular systems. Trajectory-based approaches combine a classical description of nuclear dynamics with a quantum-mechanical description of electronic dynamics. However, the approximations underlying quantum-classical methods are sometimes severe, and are at the origin of controversies as well as of continuous developments.

In this talk I will present a recently-developed trajectory-based approach to nonadiabatic dynamics[1,2]. The actual numerical scheme has been derived from the exact factorization of the electron-nuclear wavefunction[3], a new framework proposed to investigate, interpret and approximate the coupled dynamics of electrons and nuclei beyond the Born-Oppenheimer approximation. The exact factorization provides a new perspective to analyze nonadiabatic processes: (i) it proposes an alternative[4] to the standard Born-Oppenheimer framework, that pictures excited-state processes in terms of wavepackets moving on and transferring between static potential energy surfaces; (ii) it suggests new interpretations[5] of molecular geometric-phase effects, related to conical intersections; (iii) it provides guidelines for developing simulation algorithms in different[6] nonadiabatic regimes. These points will be discussed during the talk and illustrated on low-dimensional models and molecular systems (oxirane is shown in the figure below).



Ultrafast ring-opening process in Oxirane triggered by photo-excitation from S_0 to S_2. The energy gap S_2/S_1 along the simulated trajectories is represented as function of the reaction coordinates, i.e., the CO bonds length

References

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Experiments at ATTOLab: Towards time resolved MFPADs

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Molecular frame photoelectron angular distributions (MFPADs) provide most sensitive observables in the study of the photoionization dynamics of molecules. Combining electron-ion coincidence 3D momentum spectroscopy of dissociative ionization processes with the use of advanced light sources delivering attosecond pulses in the extreme ultraviolet (XUV) region, MFPADs can be probed at the attosecond to picosecond time scales. These developments open up new areas of studies on ultrafast electron and nuclear dynamics, e.g., electron motion, relaxation dynamics of autoionizing states, excited states, control of dynamics of chemical processes, measurement of attosecond photoemission time delays[1,2] in the molecular frame[3], as addressed in this work. In the current study performed at the ATTOLab-FAB10 (rep. rate 10 kHz) beamline (Equipex, L'Orme des Merisiers, CEA) using the CIEL COLTRIMS set-up, we employed the XUV-IR interferometric RABBITT technique, where the target is ionized by an XUV attosecond pulse train and a coherent IR pulse interacts with the electron in the continuum. Resulting from the interferences between two different coherent pathways populating the same final electronic state, sidebands are generated in the photoelectron spectrum, whose intensity oscillates sinusoidally as a function of the delay between the XUV and IR pulses. The phase of these oscillations gives information on the Wigner time delay[4], which quantifies the emission delay for XUV photoionization. We report first time-resolved photoelectron angular distributions measured at ATTOLab using the COLTRIMS type spectrometer for photoionization of noble gases (Ar, Ne) (Lab. Frame, LF) and molecules (NO, O_2) (Mol. Frame, MF), within the limits of the present development of the FAB10 beamline. The reported results, which address notably LF and MF angularly resolved photoemission delays, are compared to recent experimental[5] and theoretical[6] results for Argon. The experiments at ATTOLab are supported by complementary spectrally resolved photoionization studies at Synchrotron SOLEIL in collaboration with the PLEIADES and DESIRS beamlines.

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Migration de charges au sein de l'acide propiolique et son déphasage dû au couplage avec les mouvements nucléaires

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Du fait de la corrélation électronique, l'ionisation d'un système moléculaire peut mener à une dynamique de charges ultrarapide connue sous le nom de migration de charges[1]. Malgré les avancées technologiques dans le domaine des impulsions laser attoseconde, qui permettent maintenant d'étudier les dynamiques électroniques à leur échelle de temps intrinsèque, ce type de dynamique n'a jamais pu être observé expérimentalement. Une interrogation actuelle est de savoir si ces dynamiques survivent suffisamment longtemps pour être observées ou si la dynamique nucléaire mène à une décohérence rapide empêchant la charge de migrer. Des cas de décohérence ultrarapide ont été rapportés dans la littérature[2,3], ainsi que des exemples encourageant[4] pour des cohérences électroniques de longue durée. Dans ce dernier cas cependant, un traitement complet de la dynamique nucléaire n'a pas été réalisé. Dans le but d'apporter un éclairage à cette interrogation, nous avons étudié les migrations de charges existantes pour l'acide propiolique ($C_3O_2H_2$)[5].

En utilisant la méthode *Algebraic Diagrammatic Construction* au troisième ordre (ADC(3)), nous avons identifié une migration de charges particulière apparaissant au sein de cette molécule après l'ionisation de son HOMO. Cette dynamique consiste en une oscillation de la charge entre la triple liaison carbone et l'oxygène carbonyle de la molécule avec une période de 6,2 fs pour sa géométrie d'équilibre. L'impact de la dynamique nucléaire a ensuite été introduit en utilisant un Hamiltonien de couplage vibronique. La méthode *Multi Configuration Time Dependent Hartree* (MCTDH) a été utilisée pour réaliser une simulation purement quantique de cette dynamique couplée électro-nucléaire, en tenant compte de l'ensemble des 26 électrons de valence de la molécule, de l'ensemble de ces 15 degrés de liberté nucléaires ainsi que des 4 états cationiques présents dans la région d'énergie concernée. Les résultats montrent que la migration de charges survie à la décohérence induite par la dynamique nucléaire suffisamment longtemps pour être observée, comme présenté en Figure 1. Ce temps est également suffisant pour obtenir un contrôle satisfaisant de cette dynamique à l'aide, par exemple, d'une séquence de deux impulsions faibles Gaussiennes infrarouges[6]. Ainsi cette molécule peut être un bon choix pour l'étude expérimentale des migrations de charges.

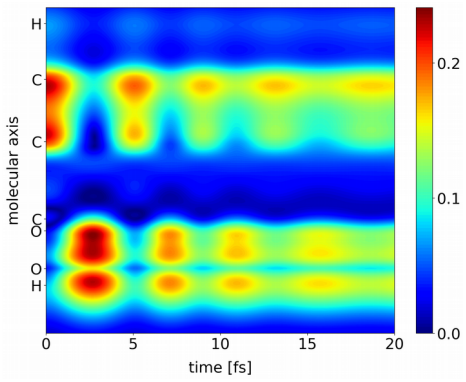

Figure 1. Densité de trou dépendante du temps pour l'acide propiolique après ionisation de son HOMO

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Superexchange Interatomic Coulombic decay: an efficient long-range energy transfer

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Keywords: Interatomic Coulombic decay, decay widths, rare gas clusters, Fano resonances

Inner-valence ionized states of atoms and molecules live shorter if these species are embedded in an environment due to the possibility for ultrafast de-excitation known as interatomic Coulombic decay (ICD) [1]. In this process the initially excited species de-excites by transferring its energy to a neighbor and ionizing it within femtoseconds. The ICD lifetime, or going from the time to the energy domain, the ICD width, depends on the distance between the interacting monomers. At large interatomic separations, the process can be viewed as an exchange of a virtual photon between the monomers and thus, the decay width displays a $1/R^6$ dependence on the distance [2, 3].

In this work we show that the lifetime of the ICD active states decreases further when a bridge atom is in proximity to the two interacting monomers. This novel mechanism, termed superexchange ICD, is driven by the efficient transfer of excitation energy via virtual states of the bridge atom (Fig. 1). As a showcase system we consider the NeHeNe trimer. The decay widths of the $Ne^{2+}(2s^{-1})\ ^2\Sigma_g^+$ state in the presence of He and in the isolated dimer were computed using the Fano-CI method [4]. We demonstrate that the decay width of the $Ne^{2+}(2s^{-1})\ ^2\Sigma_g^+$ resonance increases 6 times in the presence of a He atom at a distance of 4 Å between the two Ne atoms. Using a simple model, we provide a qualitative explanation of the superexchange ICD and we derive an analytical expression for the dependence of the decay width on the distance between the neon atoms [5].

Figure 1. Schematic representation of the superexchange ICD process in NeHeNe trimer.

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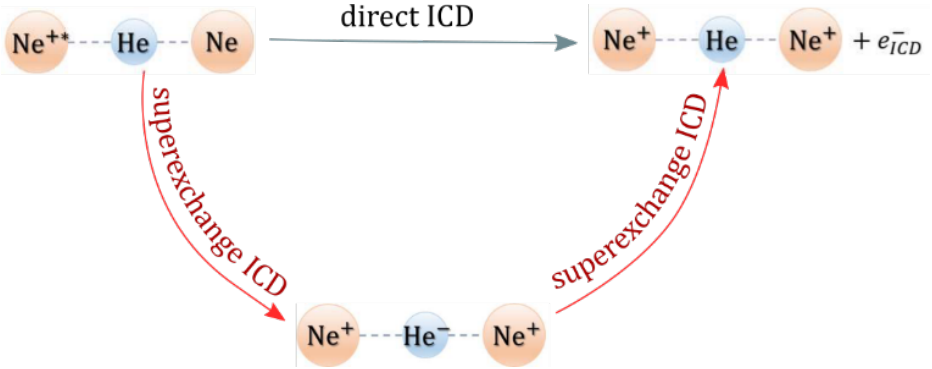

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Long-lived Highly Excited States in PAHs Following XUV Excitation

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The advent of sources producing ultrashort XUV pulses has brought new perspectives in observing atoms and molecules on their natural time scale. First experiments have been performed on molecules showing the ability to observe and control electron and (coupled) nuclear dynamics in real time, down to the attosecond timescale [1,2]. As rigid and symmetric systems, polycyclic aromatic hydrocarbons (PAHs) constitute prototypes for the investigation of complex molecules. Moreover, due to their high stability under XUV radiation, they are even thought as a primordial class of large molecules in space [3], where their photo-induced reactions are of prior importance for interstellar chemistry. In order to investigate the photoinduced processes upon XUV excitation, we have recorded dynamics following XUV excitation in PAHs of various sizes, from Naphtalene ($C_{10}H_8$) to Hexabenzocoronene (HBC, $C_{42}H_{18}$), occurring on the femtosecond timescale. This interaction with ultrashort XUV pulses especially creates highly excited cationic states in PAHs, whose multielectronic character is dominant (shake-up process). We probed the subsequent non-adiabatic relaxation of these states by second ionization with IR probe photons, creating slow photoelectrons and stable dications [4]. Measurements through both observables highlight the major role of electron correlation in the relaxation of these highly excited states [5,6]. The description of the complete many-body wavefunction is thus necessary to the understanding of the behavior of PAHs under XUV excitation, which can be directly incorporated in astrochemistry models.

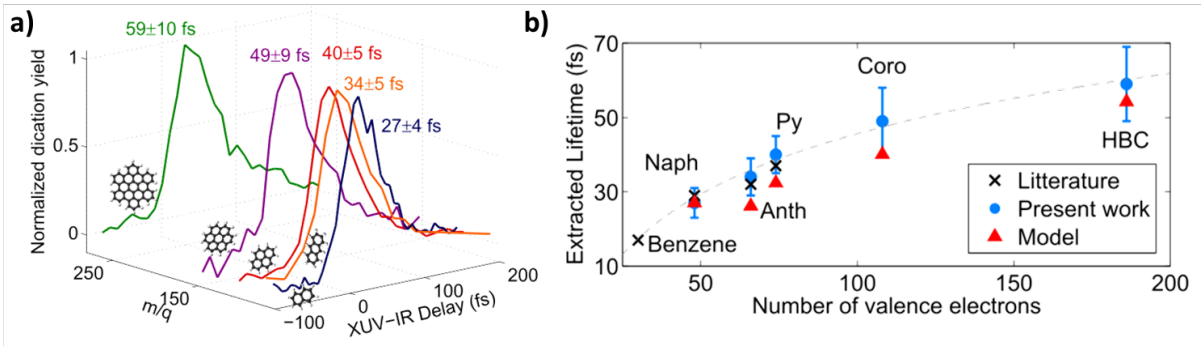

Fig. 1. a) Time-dependent dication signal measured for small to large PAHs: Naphthalene ($C_{10}H_8$), Anthracene ($C_{14}H_{10}$), Pyrene ($C_{16}H_{10}$), Coronene ($C_{24}H_{12}$), HBC ($C_{42}H_{18}$) ; b) Evolution of the extracted lifetime as a function of the number of valence electrons (blue dots), together with lifetimes from the literature (black crosses) from [4] and [7], and a model developed for astrochemistry simulations (red triangles). The increasing timescale is directly related to the multielectronic nature of the probed states.

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GDR Ultrafast Phenomena 2018

PyMoDaq : un système d'acquisition modulaire et « libre » utilisant Python

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Tout expérimentateur confronté à l'automatisation de l'acquisition de données connaît la problématique de développer un software répondant à ses besoins et nécessitant une connaissance plus ou moins approfondie d'un ou des langages informatiques. Ces besoins se résument souvent en une même forme qui est : une acquisition consiste à enregistrer des données de divers instruments en fonction de divers paramètres contrôlables. Pour la suite nous nommerons « **scan** » ce type d'acquisition, « **actuateurs** » les paramètres contrôlables (cela peut être un déplacement mais aussi une température, une tension…) et enfin « **détecteurs** » les instruments de mesure. Une acquisition peut se décomposer en plusieurs étapes que sont :

- réglage manuel des divers détecteurs et actuateurs ainsi que leurs « bornes » pour le scan
- Automatisation et synchronisation de l'acquisition de données
- Sauvegarde des données dans une forme facilement accessible (pour l'analyse des données) et clairement identifiable (pour l'archivage et la pérennisation des données)

La grande majorité des laboratoires utilisent des logiciels commerciaux qui, bien que très performant et simple de prise en main, a certaines limites qui rendent leur utilisation sur le court et long terme problématique (coût de licence, portabilité limitée, gestion des versions …) et peu versatile.

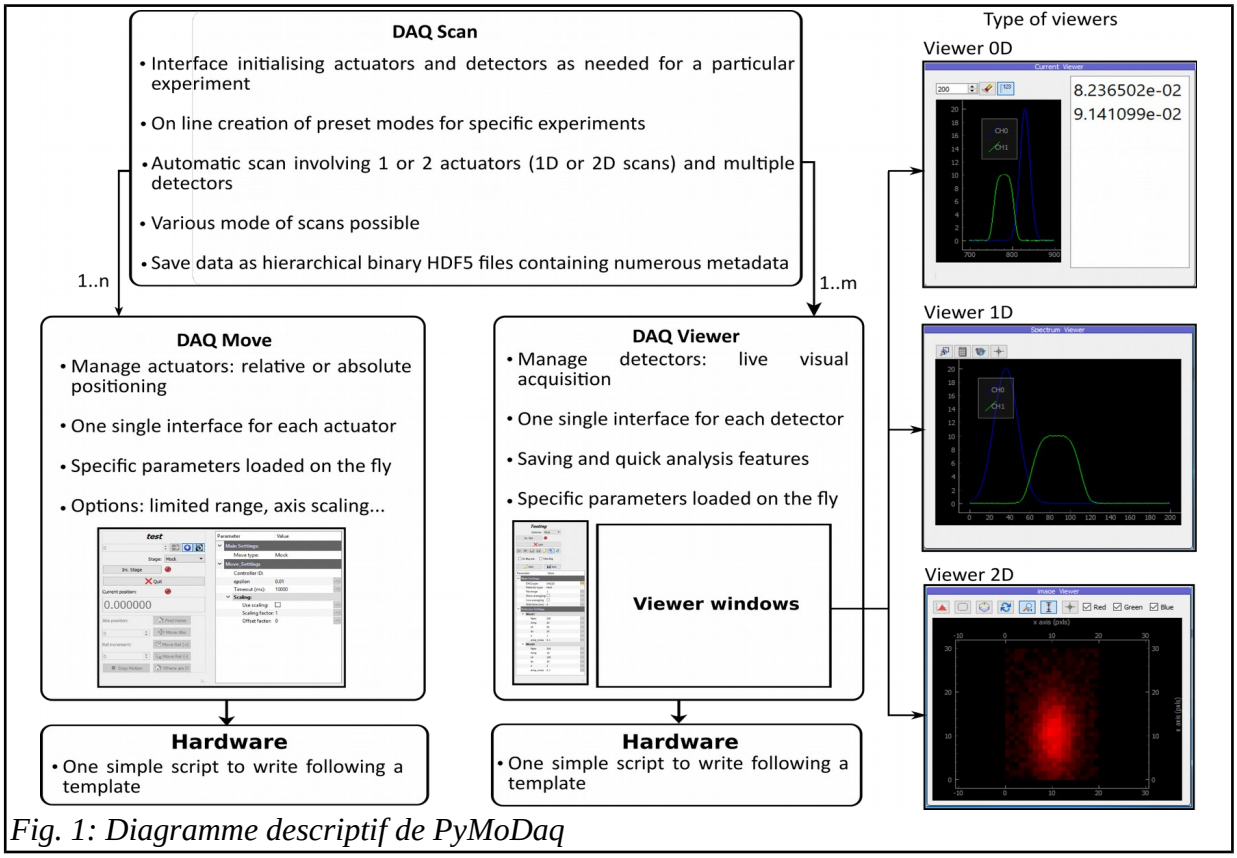

Fig. 1: Diagramme descriptif de PyMoDaq

PyMoDaq est un logiciel écrit sous python libre de droit et ouvert au développement collaboratif qui présente une interface unique pour tout type d'acquisition. Le choix de Python a été fait car celui-ci est relativement simple d'utilisation et offre une grande communauté d'utilisateurs ainsi qu'une immense bibliothèque de programmes de contrôle d'instruments et d'analyse de données. La figure 1 décrit le principe de fonctionnement de ce logiciel modulaire avec une interface DAQ_Move pour chaque actuateur et une interface DAQ_Viewer pour chaque détecteur. Ces modules peuvent être utilisés indépendamment ou intégrés à un programme tiers tel que l'interface DAQ_Scan qui permet de définir des scans automatisés, de synchroniser l'acquisition des données et enfin de les sauvegarder dans un fichier binaire hiérarchique de type HDF5 facile d'accès et compressible.

Durant cette présentation, je monterai les possibilités d'utilisation et la facilité pour ajouter de nouveaux instruments (actuateurs ou détecteurs) sous forme de plugins écrit dans un template python.

Study of the laser-matter interaction and the field-evaporation by laser assisted atom probe tomography.

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Many techniques of materials analysis or materials structuring are based on the interaction of a light source with the apex of a sharp metallic tip with subwavelength dimensions. The field enhancement phenomenon occurring at the apex can be used to make nanotweezers or to generate a localized excitation on the surface of a planar substrate or to nanostructure this surface. In all these techniques, it is important to maximize the optical field at the apex of the tip, without significantly increasing its temperature to avoid any expansion or melting of the tip.
In this presentation, we will show that the laser assisted atom probe tomography (APT) can be used as an original setup to study the interaction of ultra short laser pulses with a nanometric tip. In laser assisted APT, an ultra short laser pulse is used to trigger the field ion evaporation from a sharp tip [1-2]. The laser intensities usually used for APL analysis are in the range between the ones used for nanostructuring and the ones used for near field image of materials or in photo assisted field-electron (FE) emission.

Changing the laser parameters (intensity, wavelength and polarization) and using new experimental setup (such as the pump-probe configuration), we studied the physical mechanisms of the laser-tip interaction and of the laser-assisted field evaporation by fs laser assisted APT. New models taking into account the role of electrons on the field effect and the heating process are developed to discuss and interpret the experimental results [3-4]. In the case of non-metallic samples, we discuss the effect of the DC field distribution inside the sample on its absorption properties and the resulting field assisted evaporation. We show that the high DC field penetrates a few nanometers inside the surface increasing its absorption properties, by reducing the surface band-gap, as predicted by DFT theory [5-7].

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GDR 3754 Ultrafast Phenomena [U.P.]
Groupement pour les Sciences Attosecondes, Femtosecondes et Picosecondes

Réunion plénière 10-11 décembre 2018, Paris

Field Emission Microscopy of a Quantum Dot at the Femtosecond Scale

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Field Electron Emission (FEE) has been used as one of the earliest tools for surface characterization and Field Ion Emission - its counterpart with opposite voltage bias - was the first process allowing the direct observation of individual atoms in 1955, more than 25 years before the Scanning Tunneling Microscope. Nowadays, FEE is at the heart of modern characterization instruments in nanoscience such as Scanning Electron Microscopy, Transmission Electron Microscopy or far field Scanning Tunneling Microscopy. Over the last decade, high impact experiments on nano-tips irradiated by ultrashort femtosecond laser pulses have shown the great potentiality of FEE to get access to the ultra-fast dynamics of electrons ejected from a metal [1-3]. However, these experiments were performed on rather large nanomaterials with a continuum of electron density of states.

We recently developed a FEE experiments with an ultra-fast laser (14 fs pulse width and 80 MHz repetition rate). With this experimental set-up, we have succeeded the first experimental demonstration of femtosecond FEE on a quantum dot (~1 nm^3) with well-defined quantized electron energy levels with clear distinct features compared to metallic nano-tips. In this presentation we will show that ultra-fast FEE is now ready to explore the femtosecond and attosecond dynamics of electrons in individual nano-objects.

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HHG and attoscience at high repetition rate : status on latest laser developments

Yoann ZAOUTER, AMPLITUDE

In this contribution, we will review Amplitude latest achievements in the generation of ultrashort pulses starting from industrial Ytterbium fiber lasers. We will report several nonlinear compression schemes leading to the generation of 2 cycle pulses at multi-hundreds of kHz of repetition rate. Application to the generation of high order harmonics will be described and discussion around the implementation of CEP-stability will be initiated.

Etude de la formation de nanostructures périodiques par impulsions laser ultra-brèves en volume des verres

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Les impulsions laser ultra-courtes offrent de nombreuses possibilités prometteuses pour des modifications précises des verres. En utilisant ces impulsions laser, de nombreuses structures périodiques et non périodiques peuvent être induites de manière permanente à la fois en surface et en volume des matériaux transparents. Parmi ceux-ci, les ondulations hautement périodiques et « les nanoréseaux de volume » (« volume nanogratings », VNG) sont connus pour être les plus petites structures jamais créées par laser [1-2]. Ces structures ont trouvé de nombreuses applications dans l'optique et la photonique, en particulier pour le stockage de données optiques réinscriptibles et la production de composants optiques intégrés, tels que les dispositifs dépendant de la polarisation et les canaux micro/nanofluidiques.

Ici, nous étudions numériquement les mécanismes intrinsèques de la décomposition du verre par des impulsions laser femtosecondes [3-4]. En particulier, dans le régime de la formation de nanoréseaux en volume, le processus de cavitation induit par laser est le principal mécanisme responsable de la formation de nanovoïde au centre de la zone affectée thermiquement. De plus, il a été démontré que les processus d'accumulation affectent les distributions locales de températures et pressions, conduisant à la nucléation rapide des nano-pores, également révélées expérimentalement [2,3]. Les calculs multi-physiques démontrent que l'origine des VNGs est clairement liée à la formation et à la survie de ces nanopores [3-5]. La modélisation détaillée est effectuée en tenant compte de la propagation des impulsions laser dans les milieux non linéaires et dispersifs, des processus de relaxation / excitation électroniques, du transfert de chaleur électron-ion et de la diffusion thermique.

Les seuils d'interaction laser femtoseconde requise pour la formation/l'effacement des VNGs sont en outre définies et comparées aux résultats expérimentaux disponibles. Ses conditions sont, en effet, requises à la stabilité et à la croissance de nanopores en fonction de l'énergie laser, de la durée d'impulsion et du taux de répétition. Gardant à l'esprit ces informations, des paramètres laser optimaux peuvent facilement être proposés, que ce soit pour la nanostructuration ou pour éviter l'auto-organisation de la nanostructure. En conséquence, un meilleur contrôle peut être obtenu sur la modification résultante, facilitant ainsi le développement ultérieur des techniques de réécriture par laser

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A 100-kHz tunable femtosecond source for spectroscopy from the X-UV to the mid-IR

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Abstract: A 100-kHz, 15-W, CEP-stable OPCPA delivering 4-cycle pulses at ~3.2 µm was demonstrated. Ultra-stable operation over >8 h including a pulse-to-pulse energy stability <0.7% rms is reported. Wavelength extensions from the X-UV to the mid-IR are described as well as potential applications.

The spectral range spanning from ~100 eV to 1 keV is highly attractive for a large number of scientific applications including the study of ultrafast chemical reaction in the liquid phase [1], the study of ultrafast demagnetization at the L-edges of 3d transition metals composing magnetic materials [2] or, more simply, nano-imaging [3] and micro-tomography [4] of deep structures such as semiconductor components.

The brilliance of table-top coherent soft X-rays sources does not compete yet with large-scale synchrotron beam lines: the conversion efficiency of High order Harmonic Generation (HHG) [5], i.e. the physical process used to produce photons up to 1 keV from a near- or middle- infrared femtosecond laser, is low and the photon flux in the X-UV is actually clamped by the availability of powerful enough driving lasers. The advent of picosecond Ytterbium solid-state lasers delivering average powers in the kW range is about to change this statement. When combined with nonlinear conversion devices such as optical parametric chirped-pulse amplifiers (OPCPA), these industrial lasers can be turned into powerful tunable sources with favorable properties for HHG up to soft-x-rays [6] such as mid-infrared wavelength, few-cycle pulse duration, high peak intensity, high energy and high-repetition. Additionally, few-cycle pulses reduce the number of attosecond bursts up to, ideally, a single isolated attosecond pulse. In that case, Carrier Envelope Phase (CEP) stability and control is paramount but also ensures a shot-to-shot reproducibility of the driving electric field as well as the HHG yield and spectra.

In this talk we present the experimental results acquired during the commissioning at ELI-ALPS (Szeged, Hungary) of a supercontinuum-seeded optical parametric chirped-pulse amplifier (OPCPA) generating 4-cycle pulses at ~3.2 µm and optimized for long-term CEP-stability (cf. Fig. 1). This source delivers 150-µJ, 42-fs pulses at a repetition rate of 100 kHz, which corresponds to a peak power of ~1 GW and an average power of 15 W.

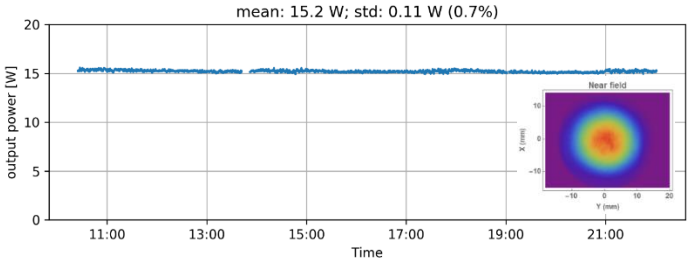

Fig. 1. Average power at the output of the OPCPA after the compression stage. Insert is the near field of the beam after compression.

We also present our development strategy toward the extension of these high-flux OPCPA sources toward the mid- and far-infrared as well innovative ideas to adapt these sources to multi-dimensional spectroscopy.

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"Ion, neutrons accélérés par laser ultra-intenses et applications astrophysiques".

Julien Fuchs, LULI

Coupling high-power lasers and high-strength B-fields helps gaining unique insight and understanding of a variety of phenomena of crucial importance for astrophysics. We have shown that such platform could be used to mimic the expansion of a young star isotropic disk wind threaded by a co-axial poloidal magnetic field. The same system can also be used to study (i) the issue of accretion dynamics in young star, in particular to shed light on the deficiency of x-ray emissivity in these systems, or (ii) the issue particle
energization in astrophysical plasmas. These examples will be reviewed and discussed. We will also discuss perspectives offered by the upcoming new generation of lasers that will offer unprecedented levels of power (up to 10 PW), which will allow to generate very dense bunches of particles at high energy. This could also have a positive impact on laboratory astrophysical studies, e.g. in the field of nucleosynthesis studies where extreme fluxes of neutrons are required in order to investigate double neutron capture, which is out of reach of existing, accelerator-based facilities, but which lasers might allow to tackle.

L'axiparabole, un nouvel outil pour l'accélération laser-plasma

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Les accélérateurs laser-plasma utilisent une impulsion laser intense ($I>10^{18}$ Wcm^{-2}) pour exciter une onde plasma associée à des champs électriques supérieurs à 100 GV / m, soit 3 ordres de grandeur plus grand que ceux obtenus dans les accélérateurs à cavités métalliques. Ces forts gradients pourraient permettre de réduire significativement la taille des accélérateurs pour des applications scientifiques, médicales et industrielles. Un fort gradient de champ n'est cependant pas suffisant pour atteindre des énergies élevées; le faisceau d'électrons doit également rester dans le champ d'accélérateur sur de longues distances, ce qui est extrêmement difficile dans un accélérateur laser-plasma en raison de 3 phénomènes: la diffraction, la déplétion de la pompe et le déphasage. La diffraction et la déplétion entraînent une diminution de l'intensité du laser lors de l'accélération, jusqu'à un niveau à partir duquel le laser ne peut plus exciter une onde plasma. Le déphasage provient de la différence de vitesse entre la phase du champ d'accélérateur et le faisceau d'électrons qui fait que les électrons glissent progressivement vers une phase décélératrice du champ de sillage. Jusqu'à présent, les énergies les plus élevées (4.2 GeV) ont été obtenues en guidant le laser dans une décharge capillaire, surmontant ainsi la diffraction.

Nous proposons ici un nouveau concept d'accélération, basé sur l'utilisation de faisceaux de Bessel à haute intensité et de couplages spatio-temporels, qui permet de surmonter non seulement la diffraction mais également la déplétion et le déphasage. Ces faisceaux sont produits par un nouveau type de miroir asphérique : l'axiparabole. La vitesse du faisceau de Bessel est superluminale dans le vide et le déphasage est supprimé en utilisant des couplages spatio-temporels pour verrouiller en phase le faisceau d'électrons sur le champ accélérateur.
Nous présentons tout d'abord des simulations numériques démontrant ce concept. Nous discutons ensuite des résultats expérimentaux préliminaires illustrant la génération de faisceaux de Bessel à haute intensité ainsi que la génération d'un guide d'onde plasma de 1 cm et le guidage d'une impulsion laser intense.

Interaction relativiste entre lasers de quelques cycles optiques et plasmas surdenses

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Nous étudions à la fois expérimentalement et numériquement la génération de faisceaux d'électrons ultrabrefs lors la réflexion d'un laser ultraintense de quelques cycles optiques ($1.4*10^{19}$ W/cm^2, 3.5 fs) sur un plasma surdense. Deux régimes d'accélération sont identifiés, pour lesquels les mécanismes d'éjection des électrons sont radicalement différents. Quand l'interface vide-plasma est très abrupte (régime du "miroir plasma"), un paquet d'électrons attoseconde est éjecté du plasma à chaque cycle optique du laser. Ces électrons peuvent ensuite être accélérés dans le vide par l'onde laser réfléchie [1]. Nous proposons en particulier d'améliorer la qualité des faisceaux d'électrons obtenus dans ce régime en utilisant des lasers polarisés radialement, qui possèdent une structure idéale pour l'accélération d'électrons dans le vide. Des simulations PIC indiquent que des faisceaux d'électrons particulièrement intéressants pour des applications telles que la diffraction ultrarapide d'électrons peuvent être obtenus avec des lasers polarisés radialement [2]. Quand la taille caractéristique du plasma est plus grande (quelques longueurs d'ondes), un régime différent apparaît pour lequel nous observons expérimentalement des faisceaux d'électrons collimatés accélérés par ondes de sillage plasmas. Ces électrons ne sont détectés qu'avec des impulsions lasers d'une durée inférieure à 10 fs, montrant clairement que de nouveaux phénomènes physiques apparaissent avec des impulsions de quelques cycles optiques.

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Intersublattice exchange interaction probed with high order harmonics over multiscale dynamics

Over the past few years, optically induced ultrafast magnetization dynamics has been investigated in alloyed ferromagnets by probing core level to 3d band optical transitions of transition metals [1,2]. Those chemical selective measurements are sensitive to the sub-lattices interaction processes during the transient magnetization dynamics. For example, it has been found that the inter-sub-lattice exchange interaction has to be taken into account if the demagnetization times of each sub-lattice of the ferromagnet are considered [3,5]. In this work, permalloy sub-lattices magnetic momenta dynamics are measured simultaneously with a table top Transverse Magneto Optical Kerr Effect by probing with High order Harmonics (20-70 eV range) over a broad temporal scale (Figure1).

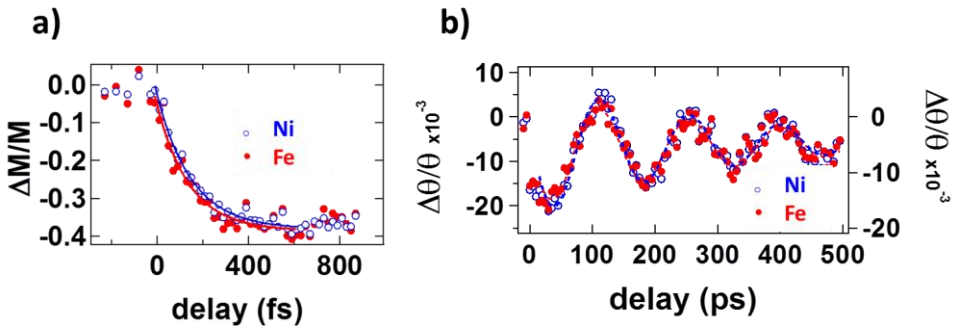

Figure 1. T-MOKE signals probed simultaneously at Ni and Fe M-edges (resp 66 eV and 54 eV) in $Ni_{80}Fe_{20}$. a) Selective demagnetization of Ni and Fe $\tau^{Ni}=\tau^{Fe}=140$ fs. b) Momenta precession of Fe and Ni damped simultaneously within 350 *ps.*

The Landau-Lifshitz-Bloch equation associated to Langevin formalism can be used to deduce the demagnetization time τ^{ε} attributed to each element ε, by taking exchange interaction into account [5]. From our measurements in permalloy, both elements demagnetize simultaneously and precess in phase with same period and Gilbert damping due to strong exchange interaction. It will be shown that the ratio between effective exchange interaction constants of Ni and Fe can be retrieved from those measurements. Moreover, the dependence of relaxation rate upon the pump density of excitation will be discussed considering a multiscale approach including temperature dependent exchange parameters.

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Spin polarization and attosecond time delay in photoemission from solids

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In the photoemission process, the electron beam emitted by a spin-degenerate state can acquire a spin polarization that is due to an interference between different channels of the photoemission process in the presence of spin-orbit coupling [1]. In particular, the spin polarization is related to the phase terms of the matrix elements of the different channels. Since variation of phase with kinetic energy of the electrons can be interpreted as a time delay (the so-called Eisenbud-Wigner-Smith time delay [2]), it is possible to make an indirect estimate of the photoemission time scale from the measurement of spin polarization as a function of energy [3,4].

The analytical model relating spin polarization to time delay will be presented, as well as estimates of photoemission time delays in the attosecond domain for measurements on Cu(111) [3] and the cuprate superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ [5].

Furthermore, perspectives and current attempts to combine this indirect approach of a spin-resolved measurement with a direct laser-based attosecond-resolved photoemission experiment will be presented.

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Exploring the band curvature of low-bandgap semiconductors using few-cycle 2D THz spectroscopy

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2D spectroscopy has been recently extended to the THz frequency range, from 1 THz to 25 THz, showing the potential of this technique for probing and exciting low-energy excitations, including electronic and lattice coupling [1], 2-phonon coherence [2], magnon nonlinearities [3].

Here, we present 2D-THz experiments in the range between 1-10 THz in a reflective geometry (see Fig. 1) investigating the electronic band-nonlinearities of InSb, using THz electric fields generated from a 2-colour plasma source (up to 100 kV/cm) and optical rectification in an organic crystal (up to 250 kV/cm). In the very first picoseconds after excitation, coherent motion dominates the nonlinear response. Subsequently, electron-electron scattering effects, i.e. impact ionization, start to dominate. Using 2D THz spectroscopy, we show that we can follow the continuous ballistic trajectory of the out-of-equilibrium electron population in the ($\Gamma \rightarrow X$, K)-plane of InSb. By using cross-polarized beams to separate contributions to the nonlinear response of different parity (see Fig. 2), we observe distinct features in the 2D spectra.

To better understand the system response at times when the pulses overlap, we simulate our results using the finite-difference time-domain technique (FDTD). The simulations show that the nonlinear response is dominated by deviations of the conduction band dispersion from the radially symmetric, parabolic bands expected from a simple free electron gas model. These deviations can be categorized by their parity.

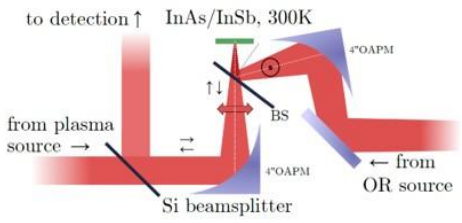

Fig. 1: Measurement geometry of reflective 2D-THz spectroscopy on InSb and InAs.

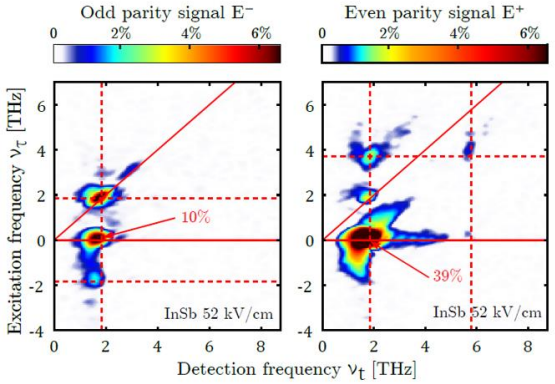

Fig. 2: Odd and even parity 2D-spectra of InSb. Broken lines indicate the plasma-edge and phonon features.

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Laser driven THz coherent phonons in β-MnAs probed by time-resolved x-ray diffraction

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We investigated the structural dynamics of laser excited MnAs/GaAs(001) epitaxial films using time-resolved x-ray diffraction at the LCLS FEL source. We report evidence of a predicted but hitherto unobserved low-frequency optical phonon mode. The temperature dependent intensity oscillations of several Bragg reflections allow us to identify the A_g optical phonon associated with the orthorhombic distortion in the β-phase and to follow its softening as the distortion vanishes on the path towards the hexagonal symmetry. The frequency of this hitherto unobserved A_g mode falls in the THz range, in agreement with density functional theory (DFT) calculations.

MnAs is a semi-metal with potential for applications in the fields of magnetocalorics, spintronics and temperature- and laser-driven magnetization reversal [1]. The interest for MnAs stems from a peculiar sequence of magneto-structural phase transitions that have been studied for decades but are not fully understood yet. In the bulk, the low temperature stable structure is hexagonal and ferromagnetic (α-MnAs). At T_c=313 K, ferromagnetic order is lost and the structure becomes orthorhombic (β-MnAs) in a first order phase transition. The orthorhombic distortion parameter η (1 at T_c) decreases progressively through a second-order phase transition and vanishes at $T_t \simeq$ 400 K, where hexagonal symmetry is recovered (paramagnetic γ-MnAs). Recent DFT calculations suggest that the β−γ transformation is a displacive phase transition driven by the softening of a THz mode with normal coordinate along η [2]. This soft mode would also be implied in the α−β transition because of strong spin-phonon coupling. Indirect signatures of partial phonon softening can be found in the literature, but no direct observations of phonons in MnAs have been reported to date. The soft mode implied in the β−γ transition, located at the edge of the hexagonal BZ, should have its counterpart at Γ in the orthorhombic BZ with atomic motions corresponding to the β → γ path, as depicted schematically by arrows in Fig. 1(b). This mode, of A_g symmetry, can be excited coherently through the displacive excitation mechanism [3].

We have employed a pump probe scheme in order to detect the presence of this mode, using x-ray diffraction as a probe. This allows us to determine the atomic motions within the crystallographic cell through the analysis of the dynamics of a set of structure factors. We have used a MnAs/GaAs(001) epitaxial thin film, a system that has been studied before because of the strong impact on the α-β phase transition of the epitaxial constraints, which stabilize the β phase at temperatures lower than in the bulk. This provides a way to stabilize β-MnAs with a wide range of η values over a narrow T range around ambient, simplifying the analysis of the diffraction data in terms of dynamic distortion.

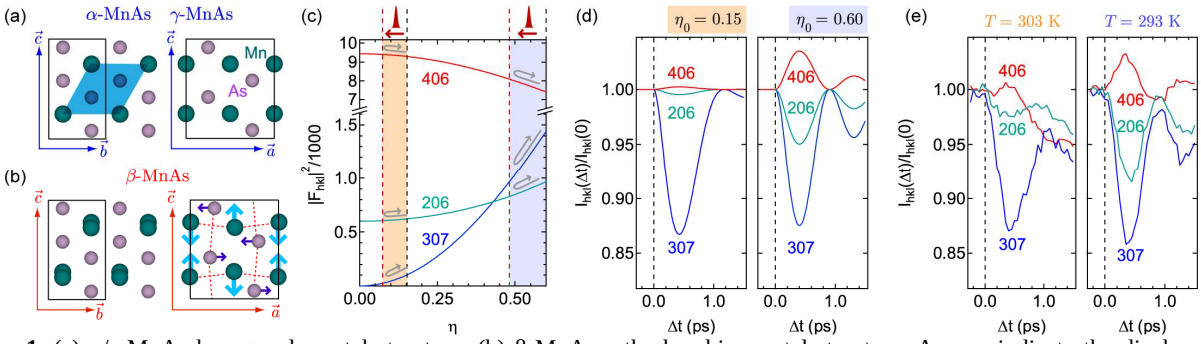

Figure 1. (a) α/γ-MnAs hexagonal crystal structure. (b) β-MnAs orthorhombic crystal structure. Arrows indicate the displacement along the A_g mode coordinates, corresponding to a reduction of the orthorhombic distortion parameter η. (c-e) Analysis of the 307, 206 and 406 reflections. (c) Calculated η-dependence of $|F^{\beta}|^2$.The shaded areas represent the portions of the transition path covered by the coherent vibration. (d) Simulated time dependence of the diffracted intensity starting from η=0.15 and η=0.60 at negative delays. (e) Time dependent normalized intensity measured at T=293 K and T=303 K.

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Understanding elastically driven cooperativity in molecular photomagnetic materials

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Materials can be controlled via physical parameters like pressure or temperature. With the advent of ultrashort lasers pulses (typically femtosecond), light excitation has been added to the panel of available techniques for materials control. Two of the major aspects in so-called photo-induced phase transitions are reversibility, meaning the ability to switch back and forth between two states, and efficiency, corresponding to the ratio of the amount of transformed material on the quantity of provided light.
One possible approach to amplify the photoresponse is based on the material elastic properties. This was demonstrated for a light-sensitive and volume-changing Spin Crossover (SCO) material [1], meaning, a molecular crystal in which the unit cell is built with SCO molecule(s). In this molecule, the ligand field result in a splitting of the 3d levels of the metal ion (FeIII) into low energy and high energy ones. The population of high energy levels (at high temperature) changes the spin state and is accompanied by strong changes in Fe-ligand distances (and thus unit cell volume).
The sudden generation (via a laser pulse) of a high enough fraction of photo-excited molecules (switched from Low spin to High spin state) creates local negative pressure. This drives lattice expansion that can induce additional switching of neighboring molecules through positive feedback effect. This cooperative effect is associated as expected with a well-defined threshold mechanism.

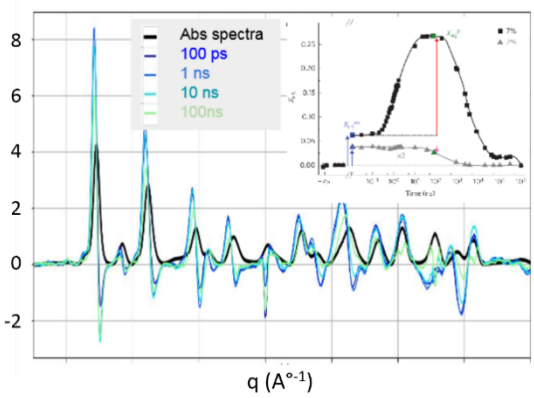

Figure *X-ray diffraction pattern and difference signal from reference of a powder SCO compound ; corresponding photoinduced transition dynamics obtained from optical experiment (insert)*

To unambiguously discriminate between spin state conversion and volume change, we performed Xray time-resolved diffraction study at synchrotron ESRF. The diffraction patterns measured from ps to µs time delays on nanocrystals powder films give a direct signature of the ultrafast volume expansion. Quantitative analysis (Rietveld) of the powder spectra at ps time scale allows going deeper into the understanding of the cooperative aspects of the photo-induced spin conversion in these molecular materials.

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PHOTODYNAMICS OF A PHOTO-SWITCHABLE FLUORESCENT PROTEIN USING UV-VIS TRANSIENT ABSORPTION SPECTROSCOPY: REMAINING QUESTIONS.

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Reversibly photo-switchable fluorescent proteins (RSFP) find growing applications in cell biology, yet mechanistic details, in particular on the ultra-fast photochemical time scale, remain unclear. The molecular basis of photoswitching is a combination of isomerization and change in protonation state of the chromophore. However the chronological order and dynamics of isomerization and protonation changes has been debated. Several spectroscopic investigations focused on Dronpa, a negative RSFP from Anthozoa (e.g. corals) used in widefield super-resolution microscopy (PALM, SOFI). For Dronpa it has been proven by femtosecond UV-visible transient anisotropy absorption spectroscopy that photoswitching from the non-fluorescent (off) to the fluorescent (on) state involves a trans-to-cis isomerization that occurs within a few picoseconds in the excited state followed by chromophore deprotonation in the ground state on the microsecond time scale [1]. We choose here to study rsEGFP2, RSFPs from Hydrozoa (e.g. jellyfish) which is the most common protein used in RESOLFT super-resolution microscopy. We employed UV-Visible time-resolved pump-probe absorption spectroscopy in solution to study photoswitching from Off-to-On state. As reported for Dronpa evidence is also provided here for the existence of several intermediate states on the pico- and micro-second time scales that are attributed to chromophore isomerization and proton transfer, respectively [2]. Indeed isotopic experiments show clearly that Off-to-On photoswitching in rsEGFP2 involves excited-state isomerization and ground state proton transfer. Nanosecond flash photolysis underlines microsecond changes accompanying formation of the cis-protonated chromophore followed by sub-millisecond deprotonation with two different time constants. The femtosecond to millisecond data provide insight into the mechanism of trans-cis isomerization in photo-switchable fluorescent proteins. Different mutations modify the Off-to-On photoswitching quantum yield and clarify the photodynamic, i.e. the existence of different off states and two different mechanisms for the trans-cis isomerization. We will then discuss questions or uncertainties that are remaining in the photo-dynamics of rsEGFP2 and that can be clarified by time resolved crystallography.

This research is carried out in the frame of the ANR BIoXFEL and in collaboration with the Institut de Biologie Structurale in Grenoble (Adam, Bourgeois, Byrdin, Colletier, Coquelle, Feliks, Field, Fieschi, Guillon, Hadjidemetriou, Schirò, Thepaut, Weik, Woodhouse), Max-Planck-Institut für medizinische Forschung in Heidelberg (Barends, Doak, Foucar, Hilpert, Kovacsova, Nass, Roome, Schlichting, Shoeman), Departement of Physics in Rennes (Cammarata), the SLAC National Accelerator Laboratory in Menlo Park (Aquila, Boutet, Hunter, Koglin, Liang, Robinson) and Riken Spring-8 Center (Iwata, Joti, Motomura, Owada, Tanaka, Togashi, Tono, Yabashi).

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Tracking fast protein structural dynamics by X-rays

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Proteins are the molecular machines of living systems. Their function is based on structural dynamics occurring over a broad time scale from femtoseconds to seconds. The femtosecond time resolution achievable with X-ray free electron lasers, combined with the atomic sensitivity of hard X-rays, allows tracking fast protein structural changes in crystals and solutions. I will present two examples of picosecond protein structural dynamics triggered by photon absorption: the chromophore motion at the excited state in a photoswitchable fluorescent protein [1] and the global conformational response - so called 'protein quake' - of a oxygen storage protein after oxygen photolysis [2].

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CLASSICAL TRANSIENT ABSORPTION SPECTROSCOPY EXTENDED TO SUB-NANOSECOND TIMESCALES

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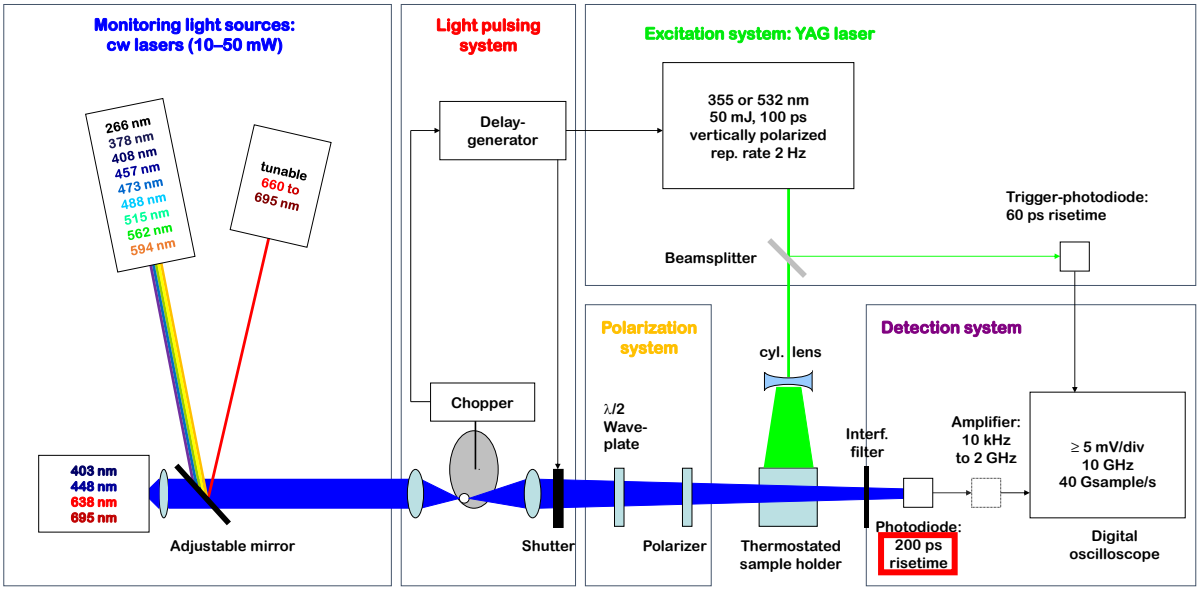
Transient absorption spectroscopy (*TAS*) is a powerful tool to study photoreactions on time scales ranging from femtoseconds to seconds. Typically, processes slower than ~10 ns are probed by a classical method, based on real-time (continuous) recording of the absorption changes due to photoreactions induced by an excitation pulse that is shorter than the time constant of the studied process. The time resolution of conventional setups is usually limited by the detection system.

Processes occurring on femto- and pico-second time scales have to be examined by more sophisticated "pump-probe" techniques, splitting ultra-short pulses into two beams, the first of which excites the sample and the second (converted to a desired wavelength or to a continuum and delayed via an optical delay-line) serves to probe the same sample volume. In general, the recorded data are rather noisy and an extensive signal accumulation is necessary to extract the reaction kinetics. The large number of laser shots (typically $>10^5$) may cause deterioration of sensitive samples. Last but not least, the mechanical change of the optical path length places high demands on precision with respect to beam alignment – the longer the delay path, the more difficult it is to focus the beam (each nanosecond corresponds to ~30 cm longer delay path) and assure constant energy and shape of the excitation pulse. This is, indeed, very challenging, especially for delays longer than a few hundreds of ps. It is therefore of interest to extend the classical method as far as possible.

We will present our home-made real-time *TAS* setup with ~200 ps time resolution and high sensitivity [1]. The improved time resolution is assured by a detection system based on a photodiode with 200 ps rise time, and low-noise cw lasers as sources of the monitoring light at ~15 discrete wavelengths covering the visible, as well as the UV and the near IR light regions. The setup can resolve absorption changes of ~1 mOD in a single-flash experiment. Sensitive samples are protected against excessive monitoring light by a system of shutters, attenuation filters and a chopper.

In the last decade, our setup has been used for numerous studies of photoactive flavoproteins. Photoreactions of some of them involve irreversible steps with rate constants in the order of hundreds of ps (e.g. [2-4]). These processes would be impossible to follow using commercial *TAS* setups and the use of pump-probe techniques would require excessive amounts of precious samples.



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Towards non-collinear UV 2DES setup with pulse-to-pulse phase correction at 50 kHz

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Since 2001,[1] two-dimensional optical spectroscopies have been extended to the visible range, targeting electronic transitions: two-dimensional electronic spectroscopy (2DES). 2DES experiments provide simultaneously high temporal and spectral resolution by resolving the excitation frequency with an interferometric method.[2] 2DES is a method of choice for the investigation of: i) structural or chemical heterogeneities; ii) excitation energy transfer, iii) ultrafast photo-reactivity of complex molecular system to disentangle electronic from vibrational couplings; and iv) competing ultrafast photochemical processes.[3] 2DES implementation faces two main technical challenges: i) the generation of compressed ultra-broadband pulses; and ii) a control of the interferometric stability between pulse pairs (precision of few nanometers).

We develop a challenging non-collinear UV 2DES setup at 50 kHz repetition rate. Firstly, we generate a sub-15fs UV pulse (300-345 nm, see Figure) by the Achromatic Sum-Frequency Generation (ASFG)[4] of a broad Red-NIR pulse[5] with a nearly monochromatic 515 nm pulse at 50 kHz. The UV pulse duration of 15 fs is characterized by a transient grating FROG measurement (Figure). Secondly, the optical phase fluctuations within pulse pairs are monitored to better than λ/100 for each individual laser shot, by monitoring the pulse-to-pulse relative phase fluctuations with a 515 nm tracer. That should allow us to perform accurate Fourier-transformed spectroscopy via phase fluctuation post-processing.

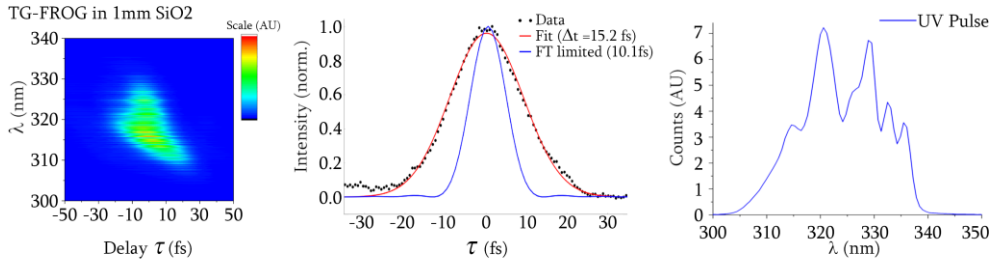

Figure: UV-pulse characterisation. Left: TG-FROG map. Middle: spectrally average TG-FROG trace with its fit and the Fourier-transform limited pulse width. Right: UV pulse spectrum.

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Focalisation sans optique d'harmoniques XUV

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Un des enjeux majeurs de la science attoseconde est la réalisation d'expériences pompe/sonde utilisant uniquement des impulsions XUV afin d'étudier des dynamiques ultra-rapides de la matière. Ces études requièrent des impulsions XUV attosecondes suffisamment intenses et il convient donc d'optimiser l'efficacité de génération, la focalisation du rayonnement XUV sur une bande spectrale XUV adaptée au système étudié et compatible avec des durées attosecondes. Ainsi, le rayonnement XUV doit être produit avec une efficacité optimale dans un mode spatial le plus cohérent possible. Idéalement, la sélection spectrale doit s'effectuer sans élargissement temporel ni atténuation significative du flux XUV en éliminant le laser générateur. Finalement, l'ensemble du spectre sélectionné doit être focalisé au même foyer afin d'éviter les couplages spatio-temporels et garantir une résolution temporelle homogène dans le milieu cible.

Nous montrons ici que nous pouvons en principe satisfaire à ces différentes conditions en produisant directement un faisceau composé d'harmoniques d'ordres élevées s'auto-focalisant sans l'aide d'optiques XUV [1]. Cet effet survient quand l'empreinte du front d'onde convergent du laser sur le front d'onde XUV l'emporte sur la contribution de la phase harmonique qui suit la distribution radiale d'intensité et tend à faire diverger le rayonnement harmonique. Ainsi, en utilisant le laser TW ECLIPSE du CELIA rendu quasi Gaussien ($M^2 = 1.04$) à l'aide d'un correcteur de front d'onde et en plaçant un jet de gaz fin avant le foyer IR, nous avons mis en évidence pour la première fois, l'autofocalisation des harmoniques. Nous avons étudié l'évolution de la divergence des harmoniques en fonction de la position du jet et de l'ordre harmonique et reproduit les évolutions observées à l'aide d'un modèle analytique Gaussien corroboré par des simulations SFA. L'effet prédit est significatif puisque les positions des foyers harmoniques H_q calculées peuvent se trouver en avant du foyer IR de 5 (q= 29) à 10 (q=37) fois le paramètre de Rayleigh (2.7 cm) du laser générateur. En plaçant un trou de filtrage de 140 µm à 37 cm du foyer laser, nous avons montré qu'il est possible d'accorder le spectre XUV transmis en variant uniquement la position longitudinale du jet de gaz. Les transmissions du faisceau XUV au travers du trou peuvent atteindre 80% avec un contraste de 1:9 compatibles avec une atténuation du faisceau IR supérieure à 99%. Nous avons enfin déterminé expérimentalement les dimensions et les positions des foyers harmoniques par la technique SWORD [2] confirmant et quantifiant ainsi le contrôle de l'autofocalisation harmonique.

En conclusion ces expériences ouvrent des perspectives nouvelles pour filter le spectre et obtenir sans optique XUV ni filtre métallique des éclairements XUV importants compatibles avec l'observation de processus non linéaires XUV et des durées attosecondes homogènes.

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IMPULSIONS ATTOSECONDES PORTANT UN MOMENT ANGULAIRE ORBITAL

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La génération d'impulsions attosecondes s'appuie sur un phénomène hautement non linéaire, la génération d'harmonique d'ordre élevé (HHG). Au cours de cet exposé, nous présenterons quelques expériences démontrant les lois de conservation de moment angulaire associées à ce processus.

Comme la matière, les photons peuvent porter deux types de moment angulaire : moment angulaire de spin (MAS) et moment angulaire orbital (MAO). Alors que le MAS est associé à une polarisation circulaire, le MAO est lui associé à un front d'onde en hélice. Les deux sont quantifiés en unités de $\hbar$. Cependant, alors que le MAS ne peut prendre que deux valeurs (± 1), le MAO peut s'étendre à l'ensemble des nombres relatifs. Ces propriétés de front d'onde et de quantification sont à la base d'applications toujours plus nombreuses, en cryptographie quantique, transport d'information, imagerie de dislocations, imagerie non linéaire à super résolution…
La génération de faisceaux portant un MAO a récemment été étendue à l'ultraviolet extrême (XUV) grâce à la génération d'harmoniques d'ordre élevé (HHG). En pratique, un faisceau infrarouge femtoseconde intense transportant le MAO est focalisé dans une cible de gaz pour réaliser la conversion vers l'XUV. Nous montrerons les résultats obtenus avec un faisceau à λ= 800 nm portant 1 à 3 unités d'OAM dans différentes cibles (Argon, Neon). Par ailleurs, nous avons mesuré la synchronisation attoseconde du peigne HHG en mettant en œuvre la technique RABBITT pour la première fois avec des faisceaux XUV transportant un MAO. Cette expérience a confirmé l'augmentation linéaire du MAO avec ordre harmonique, la pente étant déterminée par le MAO du laser générateur. Cette mesure nous permet en outre de reconstruire la forme attoseconde de l'émission (figure 2). Finalement, nous avons étendu ces études à l'observation de modes présentant un indice radial non nul (Fig. 2, droite), et à la HHG mettant en œuvre deux faisceaux. Des analogies avec le MAS seront proposées.

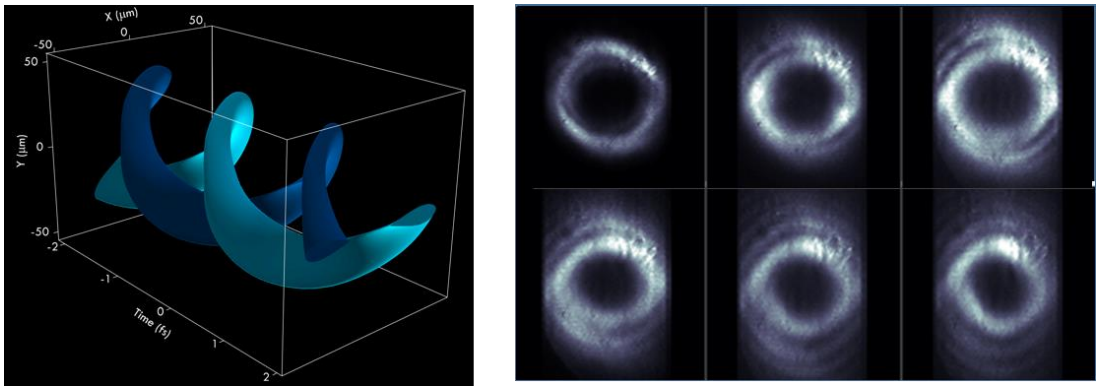

Fig. 2 : (à gauche) Reconstruction expérimentale de la structure attoseconde résultant de la superposition cohérente d'ordres harmoniques élevés portant une charge topologique augmentant linéairement avec l'ordre harmonique. (à droite) Profil spatial de la 11ème harmonique avec différents indices radiaux.

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Demonstration of an efficient 20 ps hard X-ray switch: commissioning work at the FemtoMAX beamline

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Abstract: We present the first time-resolved diffraction experiment performed at the MAX IV laboratory FemtoMAX beamline, which has been designed to access the ultrafast (fs) dynamics in matter. In this proof of principle experiment, we demonstrate the efficiency of a simple X-ray switching device, namely, a gold coated InSb film.

The FemtoMAX beamline [1] at MAXIV is designed to access the ultrafast (fs) dynamics in matter. The beamline is driven by the linear accelerator (LINAC) at MAX IV, which allows for electron bunches shorter than 100 fs in duration. In the early commissioning context of the beamline, we present the first experiment, limited to a time resolution of 2 ps, about the study of strain generated in an optoacoustic transducer.

An optoacoustic transducer represents an attractive method for obtaining short X-ray pulses at storage rings [2]. Despite the advent of ultrashort pulse facilities such as free-electron lasers, there is still a need for storage rings to carry out prospective pump-probe studies or access picosecond time-resolution. A drawback is that these facilities rarely deliver pulses shorter than 100 ps. To provide pulses with a duration of 10 picoseconds, storage rings propose special filling modes, such as low-alpha mode, however at the expanse of lowering the X-ray flux for all beamlines. Another way to produce short X-ray pulses, is to add to the beamline optics, an optoacoustic transducer (X-ray switch), whose principle is to switch the efficiency of a Bragg-reflection using a laser pulse [3].

In the present work, we have designed a hard X-ray switch which could reduce the pulse duration of a 100 ps X-ray pulse to 20 ps with a peak reflectivity of 8% (Figure 1) [4]. The switch is based on a 60 nm-thick gold film deposited on a crystal of indium antimonide (InSb). Guided by simulations, the performance of the switch has been confirmed by using time-resolved X-ray diffraction. The performance and limitation of the switch will be presented in terms of acoustic transport properties between the two materials and the electron transport properties of gold.

This first experiment, by validating the design of the beamline, was the first important step towards the final beamline capabilities and the user operations.

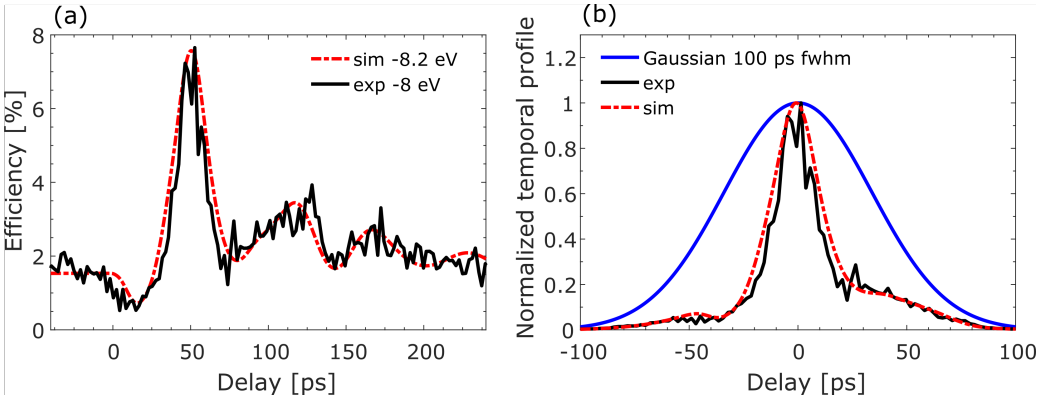

Fig. 1.(a) Time-resolved X-ray diffraction efficiency from the Au/InSb switch: experimental data taken around E = 5018 eV (solid black curve, $\Delta E = -8$ eV $\leftrightarrow q = -2.68 \times 10^{-3}$ Å^{-1}) and simulated data (dot-dashed red curve, $\Delta E = -8.2$ eV $\leftrightarrow q = -2.75 \times 10^{-3}$ Å^{-1}). (b) Normalized experimental data multiplied by a Gaussian pulse of 100 ps (FWHM) (solid blue curve). One can see the reduction of the pulse duration to 20 ps after the interaction with the switch.

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Structural Dynamics **4**, 051102 (2017)

Résumé GDR UP 2018

Génération d'harmoniques d'ordres éléves par des harmoniques d'un laser fibré : une source millwatt d'impulsions XUV ultrabrèves

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Les récents progrès réalisés dans les lasers femotsecondes à fibre dopés ytterbium (Yb) ouvrent de nouvelles perspectives pour la science attoseconde. La génération d'harmoniques d'ordre élevé à partir de ces systèmes est particulièrement intéressante car elle génère des impulsions ultra-brèves dans l'extrême ultraviolet. De nombreux efforts ont déjà été consacrés à l'optimisation des paramètres macroscopiques [1,2,3]. Nous étudions ici la possibilité d'améliorer la réponse atomique en produisant des harmoniques d'ordre élevé à partir de la deuxième, troisième et quatrième harmoniques ($2\omega_L$, $3\omega_L$, $4\omega_L$) d'un laser fibré Yb à 1030 nm « clé en main ». Nous montrons que l'efficacité des harmoniques est optimale en utilisant la troisième harmonique, produisant 4,4 10^{14} photon/s à 18 eV, correspondant à une puissance moyenne de 1,3 mW.

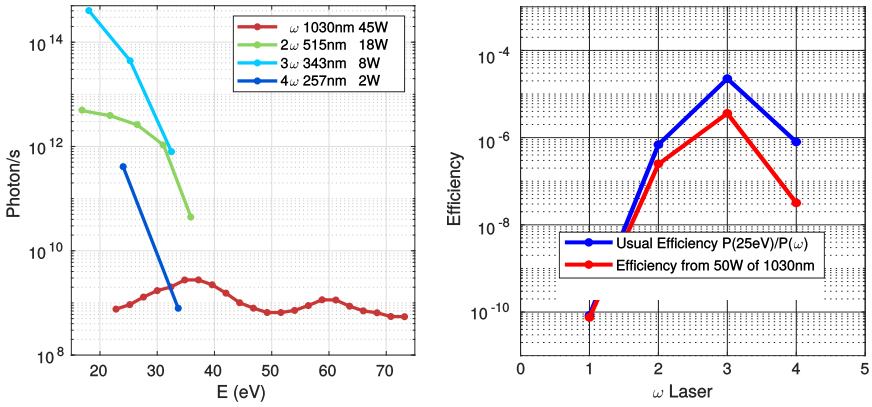

Fig. 1 : **Gauche** : Flux XUV maximum à 1,2,3 et 4 ω_L. **Droite** : Deux efficacités de conversion. La première est celle du flux XUV divisé par la puissance à chaque ω_L, la seconde est celle normalisé par les 50 W de puissance du laser.

ω_L 1030nm	Photon Energy (eV)	25.2	61.2	68.4
	Flux ($\times 10^{11}$ Photon/s)	0.93	1.1	0.64
	Power (nW)	3.8	11.2	7.1
$2\omega_L$ 515nm	Photon Energy (eV)	16.8	21.6	26.4
	Flux ($\times 10^{12}$ Photon/s)	6.9	6.9	2.8
	Power (mW)	18.7	14.6	11.9
$3\omega_L$ 343nm	Photon Energy (eV)	18	25.2	-
	Flux ($\times 10^{14}$ Photon/s)	4.4	0.24	-
	Power (mW)	1.28	0.1	-
$4\omega_L$ 257.5nm	Photon Energy (eV)	24	-	-
	Flux ($\times 10^{11}$ Photon/s)	5.1	-	-
	Power (mW)	2.0	-	-

Tableau 1 : HHG Flux XUV maximum à 1,2,3 et 4 ω_L

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Photoemission delays in 2D model molecules

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In quantum physics, the dynamics of an electron is encoded in the phase of its wave function. The ionization continuum being degenerated, there are multiple ways to describe it. The Selected Continuum Wave Function (SCWF) [1] embeds all the information needed to describe the dynamics of photoemission, and provides an unambiguous and straightforward interpretation of the delays that have been recently discussed in the framework of attosecond science [2, 3]. The two-dimensional approach allows to retrieve the angular dependent delays. Time-independent computations have been run to investigate ionization in an H_2^+ model molecule for a single-photon transition. The photoemission delays obtained from the time-independent approach are to be compared with a time-dependent propagation of a state at the same energy in order to validate the formalism of the SCWF. This work is the first step to the extension to more realistic molecules.

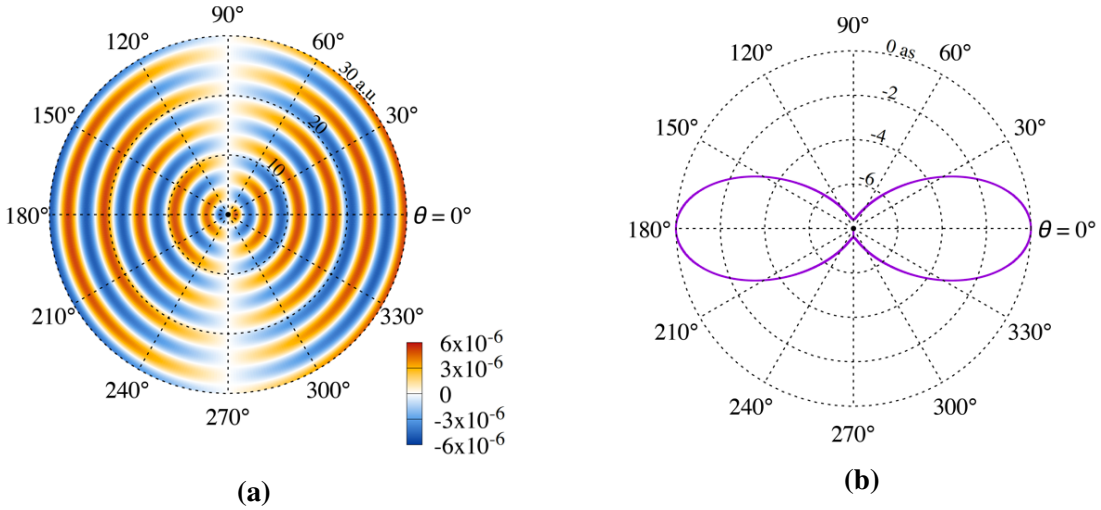

Figure 1: **(a)** The Selected Continuum Wave Function of a 2D model H_2^+ electron ionized in a single-photon transition at $\omega = 1$ a.u. and polarized linearly along the x axis. **(b)** Angular photoemission delays relative to the delay computed along the x axis ($\theta = 0°$) for the same transition.

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High-harmonic generation in solids : insights from ab initio simulations

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Recently the strong-field electronic dynamics in solids have received a lot of attention, in particular due to the experimental observation of high-harmonic generation in solids. The dynamics associated with strong field requires a non-perturbative description of the electronic dynamics. One possible way of describing this dynamics by ab initio methods is to use real-time time-dependent density functional theory (TDDFT).

In this talk, we show that *ab initio* calculations help unraveling the microscopic mechanisms responsible for HHG in solids. The predictive power of these simulations will be discussed, in particular in the context of generation of high-order harmonics from solids driven by elliptically polarized and circularly polarized light. A comparison with recent experimental results will also be discussed.

High-charge XUV Vortex beam: generation and Hartmann wavefront sensor characterization.

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Abstracts: *Optical beams carrying Orbital Angular Momentum (OAM)) are currently a very active field of research due to their numerous prospective applications. In this work, we present characterization of high charge vortex structure of such beams produced through high-harmonic generation in a rare gas using an XUV Hartmann wavefront sensor. We show that the phase matched absorption limited high harmonic generation is able to retain the high charge vortex structure of the XUV beam even in a rather long (1 cm) generation medium. Additionally, our recent results on the influence of infrared driver wavefront quality on the characteristics of XUV vortex beam will be presented.*

AMPLIFICATION PARAMETRIQUE FIBREE D'IMPULSIONS ULTRA-COURTES

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Le processus paramétrique optique est un mécanisme non linéaire largement étudié pour amplifier des impulsions ultra-courtes dans des cristaux non linéaires ou des matériaux à base de susceptibilité d'ordre 3. Dans les fibres optiques, ce processus a été principalement étudié dans le contexte des télécommunications puisqu'il permet de réaliser une amplification instantanée à fort gain, large bande et à faible bruit. Toutes ces caractéristiques sont également très prometteuses pour amplifier des impulsions ultra-courtes avec un spectre ultra-large bande. Le principe général se base sur un mélange à quatre ondes efficace impliquant une pompe intense avec un spectre relativement étroit, une impulsion signal étirée large bande et un complémentaire généré pendant le processus [1]. Dans notre expérience, l'oscillateur délivre des impulsions de 80 fs centrées à 1030 nm à une cadence de 76 MHz et une puissance moyenne de 1.5 W. Le spectre du signal est décalé grâce à la génération d'un continuum dans une fibre à dispersion normal qui s'étend principalement par automodulation de phase de 760 nm à 1300 nm (Fig.1a-b) [2]. Le continuum est ensuite filtré dans un étireur à la longueur d'onde du signal (~1081 nm). Une autre portion du laser est injectée dans un étireur et plusieurs amplificateurs à fibres dopés ytterbium. La cadence est diminuée à 1 MHz et la durée d'impulsion est de ~80 ps. Le signal et la pompe sont ensuite injectés dans une fibre à cristaux photoniques pour effectuer l'amplification du signal. Fig. 1.c montre le spectre de fluorescence lorsqu'une puissance de ~400 W est injectée dans la fibre. Deux lobes sont observés à 1007 nm et 1081 nm. Nous souhaitons par la suite caractériser la distribution spectro-temporelle du gain paramétrique [3] puis comprimer l'impulsion signal amplifiée.

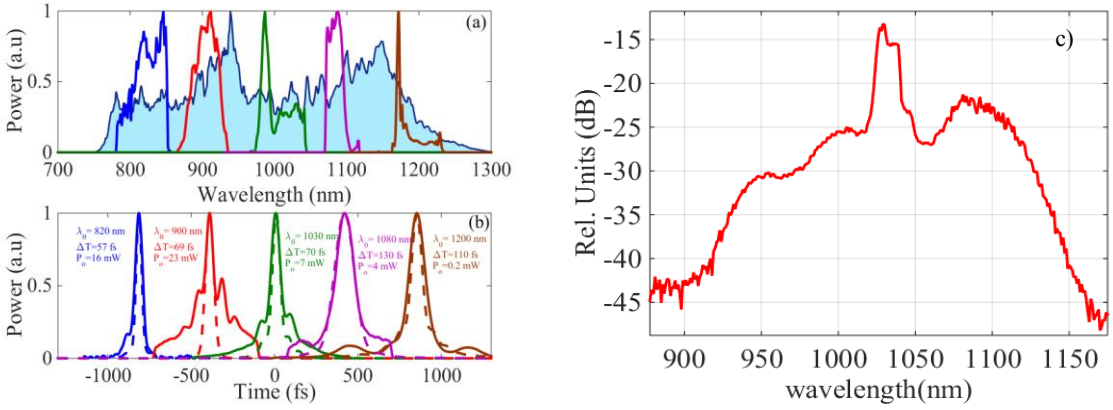

Fig. 1 a) Continuum généré dans une fibre à dispersion normal et spectre filtré b)Forme d'autocorrélation correspondante. c) Spectre de fluorescence généré dans une PCF.

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Abstract
Colloque GDR UP
Paris, 10-11 Décembre 2018

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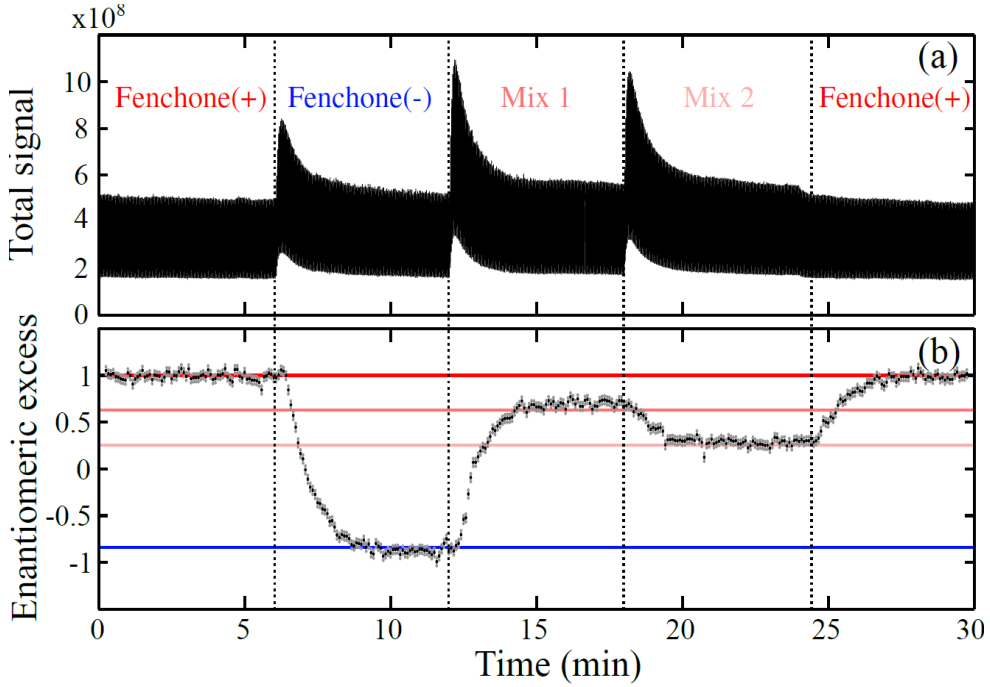
Real-time Determination of Enantiomeric and Isomeric Content using Photelectron Elliptical Dichroism (1)

Mots-Clés : Photoelectron Elliptical Dichroism, Real-Time Chiral Sample Analysis, High Repetition Rate Laser, Femtosecond Laser, Velocity Map Imaging

Les molécules dites chirales présentent la particularité d'exister sous deux formes (énantiomères), non-superposables mais images de l'autre dans un miroir. Elles sont d'une importance capitale en chimie et en biologie du fait de leur abondance dans le vivant (acides aminés, sucres, ADN...), dans lequel on ne les retrouve que dans une seule de leurs formes. Une conséquence importante est que l'interaction du vivant avec n'importe quelle molécule chirale dépendra de sa symétrie : un médicament et un poison peuvent être énantiomères, tout comme une odeur et une autre, etc.

On comprend dès lors l'enjeu d'une détermination rapide et précise de la composition énantiomérique d'un échantillon. Seules des méthodes présentant une certaine asymétrie peuvent différencier deux énantiomères, qui sont symétriques l'une de l'autre. Dans notre étude, ce rôle est joué par la polarisation elliptique, gauche ou droite, d'une impulsion laser utilisée pour ioniser un échantillon gazeux. Sa composition énantiomérique et isomérique est encodée dans la distribution angulaire des photoélectrons, qui est ensuite analysée.

L'utilisation de polarisations elliptiques plutôt que juste circulaires permet d'enrichir l'intéraction lumière/matière par des effets d'alignements, pour une analyse encore plus rapide et précise. L'utilisation d'un laser fibré à haute cadence rend la méthode développée ici extrêmement compétitive par rapport aux méthodes existantes : une chromatographie en phase chirale prendrait plusieurs heures, une simple polarimétrie nécessiterait un échantillon liquide (donc une grande quantité de produit), etc. Nous présentons ici des analyses allant de 5% de précision d'excès énantiomérique en 3 secondes jusqu'à 0,3% en 10 minutes d'acquisition. Nous sommes en mseure de suivre la composition chimique et énantiomérique d'un mélange de composées en temps réel, et même de distinguer des isomères, qui ont la même masse et ne sont pas distinguables en spectrométrie de masse.



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Controlling sub-cycle instantaneous optical chirality in the photoionization of chiral molecules

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The photoionization of chiral molecules by circularly polarized light results in one of the most efficient ways to probe chirality in the gas phase: PhotoElectron Circular Dichroism (PECD). This effect appears as a forward backward asymmetry in the photoelectron emission with respect to the propagation direction of the light. Up to now, PECD has only been studied with laser or XUV fields whose ellipticity is constant over time. The recent developments in attosecond spectroscopy have shown that two-color laser fields enabled the temporal properties of the ionization process to be controlled and measured with high accuracy. Here we engineer an electric field whose sub-cycle temporal profile and instantaneous chirality can be tuned, by combining two phase-locked orthogonally polarized fundamental and second harmonic fields. We photoionize chiral molecules with this controlled chiral field, and record projections of the photoelectron distributions by a velocity map imaging spectrometer. We observe a clear forward backward asymmetry, which is opposite in the up and down parts of the detector due to a switch of the helicity of the electric field over its optical period. This asymmetry, which we call Enantiomeric Subcycle Chiral Antisymmetric Response Gated by Orthogonal Two-color Laser Field – ESCARGOT, reflects the influence of the chiral molecular potential on the outgoing electron trajectories, driven by the complex laser field. The experimental results, as well as our numerical simulations, reveal a very strong dependence of the ESCARGOT signal on the relative phase between the two laser fields, providing a unique insight into the influence of instantaneous chirality in the dynamical photoionization process.

High Harmonic Generation in 2D and 3D semiconductors

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High harmonic generation (HHG) in crystals is a recently investigated mechanism that differs from the one well known in gases. The harmonic emission results from electronic intra-band or inter-band processes which strongly depend on the band structure of the crystal (1–7). Non-perturbative HHG occurs efficiently in a crystal exit layer of sub-micrometer thickness (1) down to an atomically thin layer (7). Here, we study high harmonic generation of various samples: thick gallium arsenide (GaAs) and silicon (Si) crystals, silicon thin films and graphene. Interestingly, these samples have different band structures: GaAs is a direct bandgap material whereas silicon is an indirect bandgap semiconductor and graphene is gapless. We detect up to the 9th harmonic (233 nm) by focusing a 2.1 µm laser in the sub-terawatt regime (80 fs, 18.6 MHz laser from NOVAE SME). Here, we perform polarization and ellipticity measurements at different laser intensities to investigate the dependence of the high-harmonic process on the band structure and crystal symmetries. Moreover, we study harmonics from monocrystalline and polycrystalline Si thin films that we compare to generation in bulk Si.

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Lensless diffractive imaging on ultrafast broadband sources

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Recent advances in attosecond coherent soft X-rays hold the promise of tracking ultrafast events at the shortest scales. To make full use of these sources, such as High Harmonic Generation [1,2], Free Electron Lasers and (future) Inverse Compton Scattering sources, novel imaging methods are required because of the extremely broad spectra of attosecond pulses. In the context of coherent diffractive imaging [3,4] I present a method of numerical monochromatisation based on the regularised inversion of a matrix which depends only on the spectrum of the diffracted radiation. I will present the successful experimental validation of our method using a supercontinuum source with 11 % bandwidth, and the extension of the technique in the soft and hard X-ray regime. In particular I will show its relevance with a simulation of broadband hard X-ray lithography mask inspection up to 20 % bandwidth. Because of its generality and ease of implementation we expect this method to find widespread applications in the domain of lensless imaging and beyond.

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Orbital Angular Momentum from Semiconductor High-Order Harmonics

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Light beams carrying orbital angular momentum (OAM) have led to stunning applications in various fields from quantum information to microscopy. Here, we investigate OAM from the recently discovered high-harmonic generation (HHG) in semiconductor crystals. HHG from solids could be a valuable approach for integrated high-flux short-wavelength coherent light sources (1-6). First, we generate high harmonics from a 1.5 µm laser beam carrying an OAM, and we verify, as in the case of HHG in gases, the transfer and conservation of the OAM from the generating laser to the harmonics in the strong field regime of interaction (7). Second, we make use of the solid state nature of the generation medium and the sufficient degrees of freedom it offers to pattern spiral zones plates on the surface of the generating crystal. This diffractive optics follows the principle of a Fresnel zone plate; it acts on the generated harmonics to build focused nano-vortices with arbitrary topological charge (7).

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Dynamique de photo-ionisation attoseconde autour du minimum de Cooper de l'Argon

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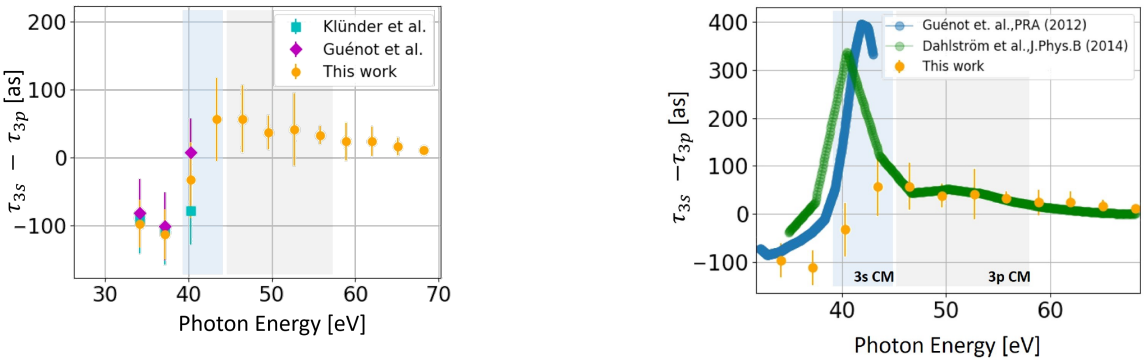
Un minimum de Cooper correspond à une baisse locale de la section efficace d'ionisation en fonction de la longueur d'onde. Il apparaît suite à l'extinction d'un canal d'ionisation [1]. Si ce phénomène est bien connu en spectroscopie conventionnelle, notamment dans le cas de l'Argon [2], ce n'est que récemment que la dynamique ultra-rapide d'ionisation autour d'un minimum de Cooper a pu être étudiée.

Issue de la spectroscopie attoseconde, la technique RABBIT [3] permet de mesurer l'amplitude et la phase spectrales du paquet d'onde électronique émis. À partir de cette phase, il est possible d'en déduire un retard (ou délai) d'ionisation entre différentes sous-couches électroniques. Nous avons pu mesurer la différence entre les retards d'ionisation 3p et 3s de l'Argon autour du minimum de Cooper (énergie de photon d'environ 50 eV). Les résultats obtenus (figure 1) présentent de fortes variations qui semblent témoigner d'une importante corrélation électronique entre les sous-couches 3s et 3p, avec notamment l'apparition d'un minimum de Cooper « 3s ». Ils ont été comparés à de précédentes mesures [4, 5] ainsi qu'à des calculs théoriques [6] pour lesquels un accord complet est encore difficile à obtenir. Ceci démontre tout l'intérêt de ces mesures de spectroscopie attoseconde, qui sont un test sévère des théories multi-électroniques tentant de bien décrire les couplages inter-canaux.



(a) (b)

FIGURE 1 – Retards d'ionisation mesurés en fonction de l'énergie de photon (ronds orange) comparés (a) aux mesures faites dans [4] (carrés bleus) et [5] (losanges violet) (b) aux calculs théoriques de [5] (courbe bleue) et [6] (courbe verte).

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Impulsions attosecondes isolées par génération d'harmoniques à deux couleurs

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***Abstract*— Obtenir des impulsions attosecondes isolées et intenses permettrait de mener des expérimentations de type pompe-sonde avec une grande résolution temporelle afin d'étudier des dynamiques électroniques dans une grande diversité de systèmes, notamment les molécules [1] ou les plasmas [2]. Une méthode efficace pour obtenir des impulsions isolées est d'induire la génération d'harmoniques d'ordre élevé (GHOE) en mélangeant deux faisceaux de pompe à des fréquences différentes [3]. Nous avons mesuré des spectres générés par cette méthode et avons étudié leur évolution en fonction du retard entre les deux impulsions, puis avons complété notre étude avec des simulations pour discuter de cette évolution.**

Interet du melange de couleurs

Au lieu d'un champ de génération sinusoïdal, la sommation cohérente de deux impulsions à des fréquences différentes correspond à des battements dans le temps. Ces battements présentent des maxima locaux assez espacés temporellement pour que, en sélectionnant les fréquences ayant un ratio adapté [4], seul le maximum d'amplitude du champ correspondant au maximum de l'enveloppe puisse permettre une GHOE efficace à des énergies élevées. Dans ce cas, elle a lieu pour un champ très intense, puisque c'est la somme constructive des champs des deux impulsions : l'efficacité de génération est alors améliorée.

Mise en oeuvre experimentale

L'expérimentation sur la plateforme laser Attolab [5] est faite avec un laser Ti :Sa femtoseconde ; une partie de ce laser est envoyée dans un Amplificateur Paramétrique Optique (OPA) pour produire une longueur d'onde variable. Les deux impulsions sont ensuite focalisées dans un jet de gaz (Argon ou Néon) et le spectre des harmoniques est analysé dans un spectromètre à réseau. Les spectres produits par le mélange à deux couleurs sont ainsi mesurés pour différentes valeurs de retard entre les deux impulsions.

Resultats obtenus

Lorsque les deux impusions sont superposées temporellement, le spectre généré est fortement modifié (voir Figure 1). En effet, des énergies beaucoup plus élevées qu'avec l'une ou l'autre couleur seule sont atteintes et de nouvelles oscillations en énergie sont observées. Ces oscillations, surtout visibles en dessous de 35 eV, ont une période qui correspond à l'énergie de la porteuse des battements du champ de génération.

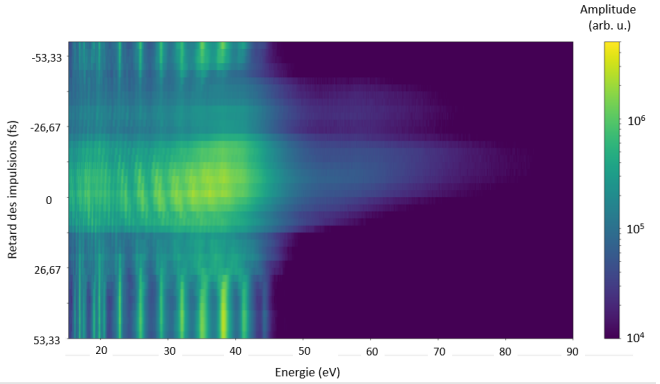

Fig. 1. Spectres obtenus dans l'Argon avec des impulsions de génération à 800 et 1257 nm en fonction du retard entre les deux impulsions.

Spectrogrammes simules

Des spectrogrammes ont également été simulés pour comprendre les comportements de l'émission on fonction du retard des impulsions mais aussi de la phase porteuse-enveloppe (CEP) du laser. Les simulations de l'émission impliquent le calcul des trajectoires des électrons de gaz rares dans le cadre de l'approximation de champ fort. Les évolutions des spectres simulés sont semblables à celles observées expérimentalement.

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High energy and high repetition rate few cycle source at 1 µm

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The recent development of high repetition rate lasers based on ytterbium-doped fiber amplifiers (YDFA), has paved the way to increase the repetition rate (>100 kHz) of coherent extreme ultra violet (XUV) sources generated by high harmonic generation (HHG). High repetition rate HHG driver comes with several advantages, such as increased photon flux [1, 2], reduction of the acquisition time in coincidence experiments to study molecular dynamics such as COLTRIMS, and the possibility to study the electronic structure of matter via photoemission spectroscopy and microscopy, where low doses are needed to avoid space-charge effects [3].

Up to now, the overall majority of HHG studies and applications has been restricted to the low repetition rates of Ti:sapphire lasers. Commonly, Ti:sapphire lasers delivers 20 fs pulses at a central wavelength $\lambda = 800$ nm, with pulse energies up to hundreds of mJ. However, the average power of these laser systems cannot easily be scaled beyond 10 W, restricting HHG at low repetition rates (up to 10 kHz).

Currently, the most mature and powerful ultrafast source technology is undoubtedly ytterbium-based systems, with average power levels beyond 1 kW [4] and numerous industrial applications. However, the long pulse duration of around >200 fs delivered by YDFA sources limits their relevance to this application field. Therefore, nonlinear compression setups have been used successfully to reduce the pulse duration and obtain XUV photon flux among the highest ever reported for HHG-based sources [5]. However, to reach sub-3 cycles regime (< 10 fs at 1030 nm), which is typically required in combination with gating techniques to obtain isolated attosecond pulses, two stages of compression must usually be implemented [6]. This reduces the energy efficiency of the systems dedicated to attosecond physics to typically less than 30% of the overall YDA energy.

Here, we demonstrate a two-cycle-source based on a high-energy femtosecond YDFA followed by a hybrid two-stage nonlinear compression setup. The association of a multipass cell-based stage and large-diameter capillary stage provides a compression factor of 48 with an overall transmission of 61%. This source is, to the best of our knowledge, the most efficient few cycle high energy and high repetition rate laser demonstrated to date. It is very compact with an overall footprint of 1.8 m × 1.0 m and provides a stable train of few-cycle pulses at a central wavelength of 1030 nm that has been continuously characterized over hours. The delivered 6.8 fs (see Fig. 1) 140 µJ pulses at 150 kHz repetition rate, corresponding to 21 W average power, are ideally suited to drive high-photon flux XUV sources [7] through HHG. The described laser system is robust, compact, and power efficient, making it an ideal driver laser for application-ready high flux XUV and attosecond sources.

We will detail the laser setup and performances and we also investigate both short- and long-term stability properties, such as pulse energy and beam pointing stability as well as long-term stability measurement over more than 12h. Future average power and energy scalability beyond 100 W and 1 mJ will be discussed. Finally, we will also detail future implementation of carrier-envelope phase stabilization on such laser systems.

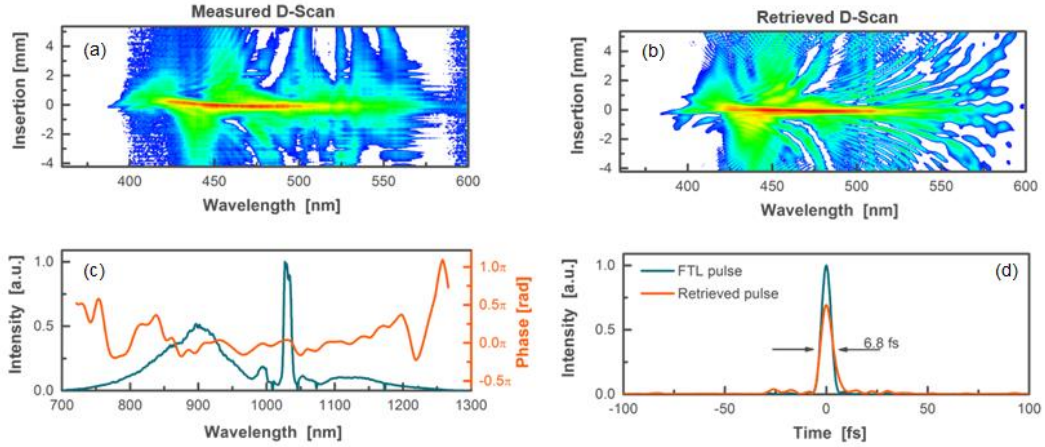

Fig. 1. Figure 5 Measured (a) and retrieved (b) d-scan traces at the output of the capillary stage. Spectral intensity and phase (c). Temporal profile and corresponding Fourier transform limited pulse (d).

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High repetition rate, CEP-stable amplifier at 1 µm

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The recent development of high repetition rate few-cycle lasers based on the nonlinear compression of ytterbium-doped amplifiers (YDA) has paved the way to increase the repetition rate (>100 kHz) of coherent extreme ultra violet (XUV) sources generated by high harmonic generation (HHG). Increasing the repetition rate of HHG drivers comes with several advantages, such as increased photon flux [1, 2], reduction of the acquisition time in coincidence experiments to study molecular dynamics such as COLTRIMS, and the possibility to study the electronic structure of matter via photoemission spectroscopy and microscopy, where low doses are needed to avoid space-charge effects [3].

However, carrier-envelope phase (CEP) stabilization of YDA is one the remaining bottleneck that prevents further application. So far, CEP stabilization with ytterbium-doped systems has only been demonstrated with oscillator or low energy (few nanojoule), >10 MHz high-repetition rate amplifiers owing to already available CEP measurement tools and easier to implement CEP-compatible setup. Further energy scaling is limited by a lack of suitable CEP seeder at repetition rate around hundreds of kilohertz.

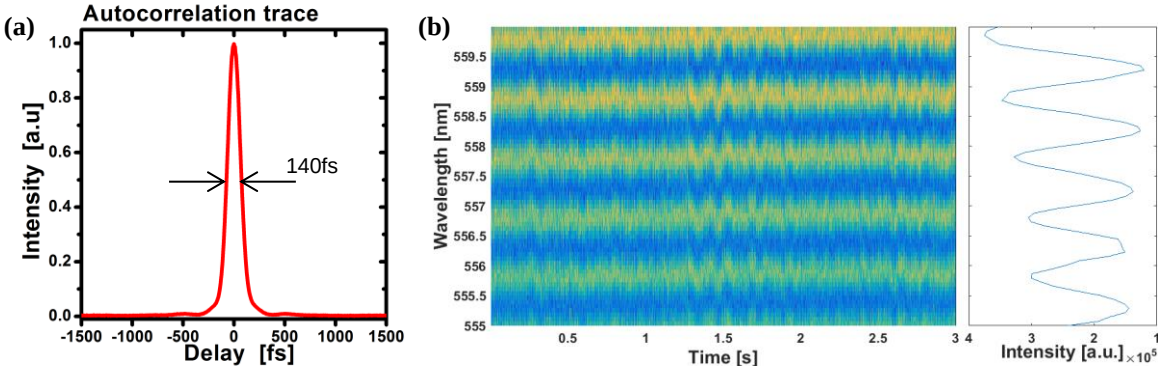

Fig. 1 (a) Autocorrelation trace of the post-compressed pulse. **(b)** F-2F CEP fringes single shot over 3s.

Here we report on the first development and preliminary demonstration of a CEP-stable fiber amplifier at 1 µm generating 100 fs, 30 µJ high energy pulses operating at 100 kHz. The CEP seeder relies on a robust OPA-scheme based on a passive carrier-envelope phase stabilization through a difference-frequency generation process [4-5]. Then, after stretching and preamplification the CEP pulses are amplified into a large-core rod-type fiber generating 350 fs, 35 µJ. Output pulses are then sent into a nonlinear compression stage based on multipass cell allowing to reduce the pulsewidth down to 100 fs [6]. The resulting autocorrelation trace is shown in Fig.1(a). Finally, CEP noise measurement are performed using F-2F scheme and a fast acquisition detector allowing single-shot measurement. Results are plotted in Fig.1(b) and shows a typical fringes spectra series acquired over 3s. The measured single shot CEP noise fluctuations of the free running system is around 600 mrad. A single spectrum is shown on the right inlet, demonstrating a very high fringes contrast corresponding to a 0.56 measured value.

We will discuss the performances scaling of this setup to higher energies/average power and shorter pulse width down to the few-cycle regime using cascaded nonlinear compression stage.

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Strong field optoelectronics in semiconductors and dielectrics

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Optoelectronic is extending to the highly non-linear regime and the attosecond time domain. A recent impact of this capability of controlling the response of above band gap electrons under strong fields is the emergence of high harmonic generation (HHG) in crystal[1-6]. 2D and 3D semiconductors exhibits properties of high electron mobility that allows to drive intense electrons currents coherently in the conduction band. HHG are emitted when those electrons recombine to the valence band. HHG in crystals can not only revolutionize attosecond science but also prepare a new generation of ultrafast visible to X-ray optoelectronic devices.

Here, I will review our recent progresses on HHG in semiconductors. As a first illustration I will present results on CaCO3 and GaN crystal and show a strong laser polarization dependence that can be used to gate isolated attosecond pulses. HHG in graphene is then investigated. Our results deviate from the work reported by Yoshikawa et al.[6] which can be explained by the effect of the non-conical section of the graphene band structure. I will then report on the non-linear harmonic response of VO2, a strongly correlated system. The harmonic signal exhibits signatures of the few optical cycles insulator to metal phase transition. We will then show how semiconductors nano-structuration can be used to induce spatio-temporal control of the harmonic emission. First, nanoscale beam carrying orbital angular momentum are generated[7]. In a second experiment, enhancement of several orders of magnitude of the HHG emission in a semiconductor waveguide is observed[8]. Our all semiconductor HHG nano-emitters can work sustainability over days and can be used as ultrafast petahertz optoelectronic devices[8]. In addition, hot electrons, emitted coherently from the nanopstructure have been measured using a PEEM. The emission is localized in a few 100 nms spot with a discrete spectral distribution correlated with the HHG emission.

Finally, inspired by recent work[9,10], I will conclude by drawing a perspective about how these various 2D and 3D crystals, with unique properties, can be used to control electronic currents at petahertz frequencies.

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Poster abstract

Title: **Two-Dimensional Electronic Spectroscopy to study the Ultrafast Photoisomerization of Biomimetic Molecules**

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Abstract: Coherent two-dimensional spectroscopies (2DS) from IR to the deep UV spectral range are powerful at unravelling the ultrafast photo-physics of many complex systems from semiconductors and organic materials to structurally/chemically heterogeneous bio-molecular systems [1]. Their implementation requires a pair of identical "excitation" laser pulses that (i) are temporally very short, and (ii) have interferometric relative phase stability. These requirements are more challenging in the UV than in the IR because (i) larger dispersion of transparent media including air enhances laser pulse temporal broadening, and (ii) shorter wavelengths require improved set-up stability for a given relative phase fluctuation. An original birefringent interferometer named "TWINS", was recently introduced which uses birefringent wedges to control the time delay with attosecond precision between co-propagating pulses of orthogonal polarizations, resulting in a relative phase stability better than 20 mrad in the Vis [2] or in the UV range [3]. We incorporated a TWINS device in a previously developed transient absorption spectrometer using a sub-10 fs pump pulse in the near-UV-Blue range (360-430 nm), generated with a hollow-core-fiber compressor and a UV-Vis super-continuum probe pulse generated in CaF2 [4]. We characterize the pulse duration by the 2D-SI technique [5]. We investigate the ultrafast photoreaction converting light energy into mechanical energy in so called "molecular photoswitch": upon light excitation this small molecule returns to the ground state by an ultrafast rotation around a C=C double bond. The system is a bio-inspired model system for the investigation of photo-reactions occurring on a time scale faster than energy dissipation to the environment, in which case quantum dynamics [6] may be taken advantage for optimizing the molecular function [7].

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ULTRAFAST EXCITED STATE DYNAMICS OF NHC-FE(II) COMPLEXES LACKING LIGHT-INDUCED SPIN CROSSOVER

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Low-spin Fe(II) complexes are notorious for their ultrafast excited state spin crossover (SCO) into low-energy quintuplet states ($^5T_2$) [1]. As SCO occurs on a sub-100fs time scale [2], Fe(II)-polyimine or similar complexes performed insufficiently for light-harvesting applications like in dye-sensitized solar cells (DSSCs). Very recently, it was shown that SCO can be avoided in Fe(II) complexes featuring N-heterocyclic carbene (NHC) ligands [3]. Thus, excited state lifetimes of a few tens of ps [4] were reported, achieving very recently values above 0.5 ns [5], and making these complexes promising candidates for photo-sensitizers in DSSCs or photo-catalytic applications.

In this contribution, we report on the effect of structural and electronic parameters on the excited state lifetimes as derived from femtosecond transient absorption and fluorescence spectroscopy at room temperature. We compare the proto-typical Fe(II)-NHC complex [3] and up to ten different variants thereof, including both bi- and tridentate coordination. For most complexes the excited state is of dominant metal-to-ligand charge transfer state character (^{3}MLCT), whose lifetime is limited via internal conversion, most probably implying triplet metal-centered (MC) states. The aim of the chemical design is to increase the ligand field splitting, via the strong σ-donating character of the carbene bonds or *via* improved geometries, so as to destabilize the MC states. The experiments show that at minimum three carbene bonds are required to prevent SCO. Interestingly, three carbene bonds in bidentate ligands lead to the same ^{3}MLCT lifetime as four carbene bonds in tridentate moieties, since the latter have bite angles smaller than 180°. An increased conjugation across the organic ligands or electron withdrawing groups lower the ^{3}MLCT energy and increases π- back donation. We made use of this effect in several complexes up to a point where the theoretical prediction places the $^5T_2$ state higher in energy than ^{3}MLCT [3].

Finally, we show how femtosecond fluorescence spectroscopy allows to fully characterize the ^{3}MLCT state (lifetime and energy) and to differentiate it from ^{3}MC states.

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A 100-kHz tunable femtosecond source for spectroscopy from the X-UV to the mid-IR

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Abstract: A 100-kHz, 15-W, CEP-stable OPCPA delivering 4-cycle pulses at ~3.2 µm was demonstrated. Ultra-stable operation over >8 h including a pulse-to-pulse energy stability <0.7% rms is reported. Wavelength extensions from the X-UV to the mid-IR are described as well as potential applications.

The spectral range spanning from ~100 eV to 1 keV is highly attractive for a large number of scientific applications including the study of ultrafast chemical reaction in the liquid phase [1], the study of ultrafast demagnetization at the L-edges of 3d transition metals composing magnetic materials [2] or, more simply, nano-imaging [3] and micro-tomography [4] of deep structures such as semiconductor components.

The brilliance of table-top coherent soft X-rays sources does not compete yet with large-scale synchrotron beam lines: the conversion efficiency of High order Harmonic Generation (HHG) [5], i.e. the physical process used to produce photons up to 1 keV from a near- or middle- infrared femtosecond laser, is low and the photon flux in the X-UV is actually clamped by the availability of powerful enough driving lasers. The advent of picosecond Ytterbium solid-state lasers delivering average powers in the kW range is about to change this statement. When combined with nonlinear conversion devices such as optical parametric chirped-pulse amplifiers (OPCPA), these industrial lasers can be turned into powerful tunable sources with favorable properties for HHG up to soft-x-rays [6] such as mid-infrared wavelength, few-cycle pulse duration, high peak intensity, high energy and high-repetition. Additionally, few-cycle pulses reduce the number of attosecond bursts up to, ideally, a single isolated attosecond pulse. In that case, Carrier Envelope Phase (CEP) stability and control is paramount but also ensures a shot-to-shot reproducibility of the driving electric field as well as the HHG yield and spectra.

In this talk we present the experimental results acquired during the commissioning at ELI-ALPS (Szeged, Hungary) of a supercontinuum-seeded optical parametric chirped-pulse amplifier (OPCPA) generating 4-cycle pulses at ~3.2 µm and optimized for long-term CEP-stability (cf. Fig. 1). This source delivers 150-µJ, 42-fs pulses at a repetition rate of 100 kHz, which corresponds to a peak power of ~1 GW and an average power of 15 W.

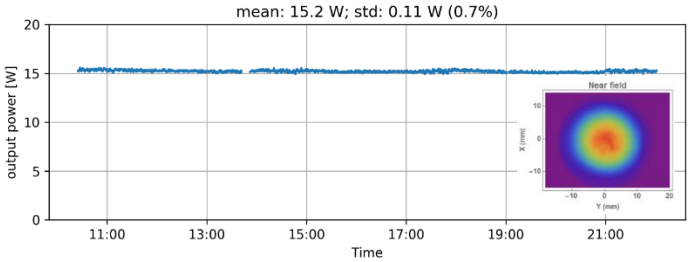

Fig. 1. Average power at the output of the OPCPA after the compression stage. Insert is the near field of the beam after compression.

We also present our development strategy toward the extension of these high-flux OPCPA sources toward the mid- and far-infrared as well innovative ideas to adapt these sources to multi-dimensional spectroscopy.

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Photo-ionization and electron-impact-ionization-TOF spectrometry for the detection of large organic molecules embedded in cluster beams

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The isolation of atoms, molecules and molecular complexes in rare-gas cluster beams, and particularly in helium nanodroplets, offers a versatile method for the study of energetic and dynamical properties of these systems, because it provides low-temperature conditions with minimal perturbation by the environment [1]. Femtosecond pump-probe spectroscopy as well as frequency-domain spectroscopy will be used to disentangle the effect of the helium droplet environment from the properties of the molecular system itself. For the detection and mass selection of the needed high mass range, as well as to give insights into the mass and charge distribution, it is advantageous to use a combination of electron-impact ionization, photo-ionization and time-of-flight (TOF) spectrometry in a collinear arrangement to the cluster beam.
On the one hand, we have combined an electron gun with a TOF spectrometer in order to provide a versatile ionization method and a good mass resolution. On the other hand, due to the possible absorption and/or relaxation to the several rotational and vibrational states of large molecules, such as polyaromatics and fullerenes (C_{60}), these systems should be photo-excited with energetic photons. This will be achieved by generating the third harmonic of 400 nm light, which will produce 9 eV photons [2].
In this presentation, preliminary results using different molecular species will be discussed. The characteristics of the electron gun and of the experimental setup, including the VUV generation scheme, will be detailed as well.

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Photo-induced coherent acoustic phonons in phase change material GeTe.

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GeTe is the archetypical phase change material which, when doped with Sb for example, represents the nowadays material for modern rewritable data storage. The process is based on the transition of the metastable amorphous into the crystalline (rhombohedral) phase. This transformation is either achieved with an electrical current burst or a light pulse that provides enough energy to induce crystallization. An extra electrically-induced/light-induced heating induces the melting followed by a return in the metastable amorphous phase (erase process) [1]. The light-induced transformation is driven at the nanosecond time scale so reaching the GHz regime requires a description of the physical properties at the picosecond time scale. Among them, the heat transport and the phonon dynamics need to be understood and controlled [1]. While coherent optical phonon spectroscopy already provided some clear information of such phase transformation in thin films [2], only few papers report on the coherent acoustical phonons [3], while the latter ones have a pivotal role in the heat transport. Therefore, in order to go deeper in the determination of the physical properties of GeTe in presence of the phase transformation, we make use in this work of an optical pump-probe setup which allows generating and detecting acoustical phonons and subsequently offers unique possibilities to probe the optical, acoustical and photoelastic properties. The experiment was conducted on a thin film (380nm) of GeTe deposited on a SiO_2/Si substrate and covered by a thin ITO protective layer. Among the results we discussed these physical properties during the amorphous-crystalline transition induced either with light (light-induced heating) or by a thermal bath (pump-probe at high temperature).

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Emission d'électrons par effet de champ et assistée par impulsions laser ultra-courtes à partir de nano-pointes de diamant

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Le développement de sources d'électrons impulsionnelles est rendu possible grâce au couplage de l'émission de champ et de l'excitation électronique sous éclairement laser [1]. En l'absence d'éclairement, les électrons sont émis sous l'action d'un champ électrique très intense, d'où l'emploi de nano-pointes. L'application d'un éclairement laser permet l'excitation des électrons et leur émission à des champs plus faibles. L'équipe d'instrumentation du GPM développe un spectromètre pour l'étude de l'émission d'électrons par effet de champ assistée par impulsion laser ultra-courte [2]. Ce travail s'effectue en collaboration avec le département optique et laser du laboratoire CORIA pour le développement d'une source laser impulsionnelle (durée d'impulsion $t = 1$ ps) à fibre dopée Ytterbium émettant dans la gamme infrarouge ($\lambda = 1040$ nm). Le schéma de fonctionnement de l'instrument est présenté à la figure 1. L'instrument est résolu spatialement grâce à la combinaison de galettes de micro-canaux, d'un écran de phosphore et d'une caméra CCD rapide.

Nous présenterons les résultats obtenus à partir de nano-pointes de diamant [3,4] en particulier les caractéristiques d'émission (courbes courant-tension et spectre en énergie) sous différentes conditions d'éclairement laser (énergie par impulsion et taux de répétition).

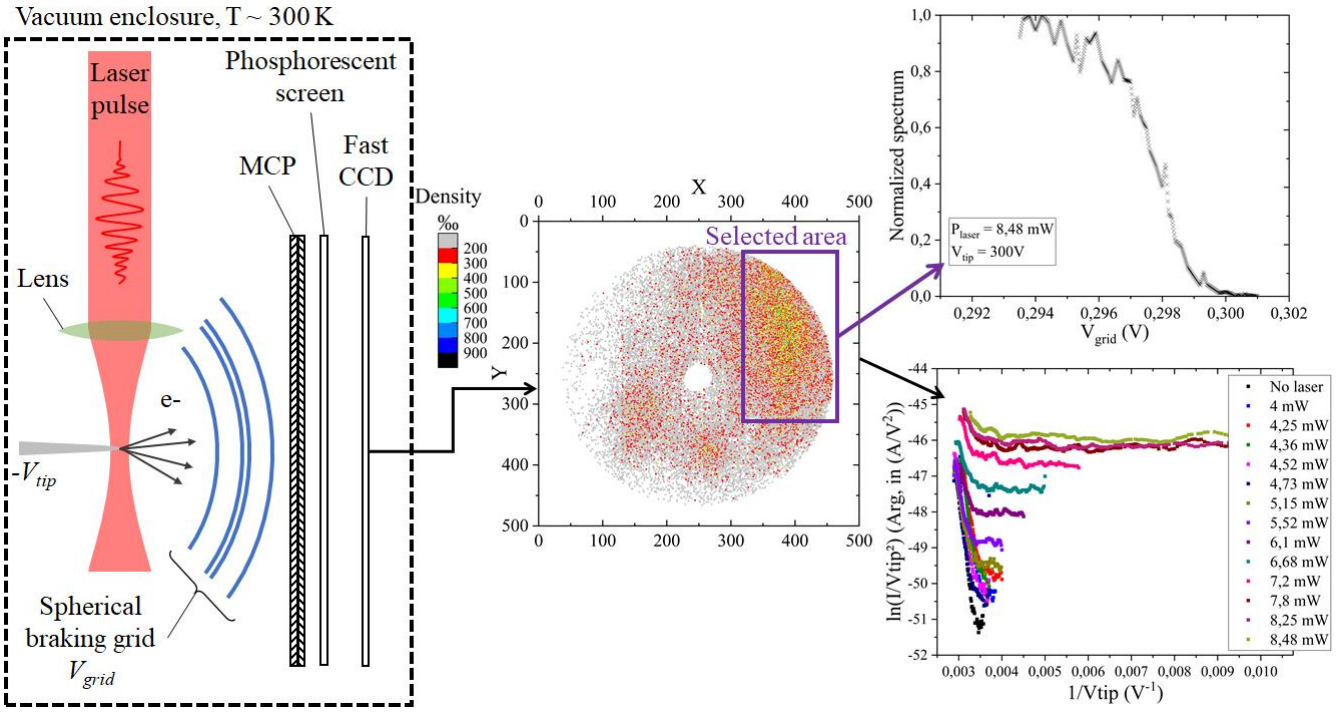

Figure 1 : Principe du spectromètre résolu spatialement. L'énergie des électrons émis est mesurée via le potentiel appliqué à la grille de freinage. Le courant est mesuré en comptant le nombre d'impacts sur l'écran de phosphore via la caméra CCD.

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Optimal control of the torsion in ethylene-like molecules

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The strong electric field of a terahertz or a non-resonant laser pulse allows us to coherently control molecules, *i.e.*, manipulate their degrees of freedom.[1,2] For gas phase molecules, the external degrees of freedom are described by the Euler angles which can thus be modified. For instance, subjecting an SO_2 molecule[3] to a non-resonant laser pulse leads to a value of the direction cosine squared Φ^2_{Zc} as large as 0.68. The internal degrees of freedom corresponding to the torsional angles of non-rigid molecules can be similarly controlled.[4] In the biphenyl molecule,[5] the angle of internal rotation describing the relative orientation of the two phenyl groups undergoes field free oscillations after subjecting the molecule to a non-resonant laser pulse.

A more efficient control can be achieved using pulse shaping in conjunction with quantum optimal control theory.[6,7] The expectation value of a selected operator can thus be maximized or minimized at the end of a theoretically designed pulse. Although there are numerous investigations concerning external degrees of freedom,[8,9] there are much less results in the case of internal degrees of freedom.

In the poster, quantum optimal control theory[6,7] will be used to design laser pulses allowing us to manipulate the internal degree of freedom corresponding to the torsion of the non-rigid ethylene-like molecule B_2F_4. A simplified 1-D model accounting only for the torsion and a physically more satisfactory 2-D model accounting for both the torsion and the overall rotation will be considered. A hindering potential characterized by a 147 cm^{-1} barrier height and 4 minima located at the eclipsed D_{2d} configurations will be taken.[10] The laser pulse allowing us to reach the planar D_{2h} configuration at time T will be computed. For each model, the time evolution of the wavefunction will be studied and we will try to understand how the molecule interacts with the laser pulse. In the case of the 2-D model, an important issue is the coupling between the torsion and the overall rotation.

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Photoionisation attoseconde de molécules chirales

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Lorsque des molécules chirales sont photoionisées par du rayonnement polarisé circulairement, une forte asymétrie apparaît dans la distribution angulaire des photoélectrons : plus d'électrons sont émis vers l'avant ou vers l'arrière, selon le caractère droite ou gauche de la lumière ou l'énantiomère considéré [1, 2]. Cette asymétrie, appelée dichroisme circulaire de photoélectrons, est l'une des sondes les plus sensibles de la chiralité, tant du point de vue statique [3, 4] que dynamique [5]. Elle provient d'effets subtils lors de la diffusion des électrons dans le potentiel chiral, et a donc intrinsèquement une origine dynamique.

Nous avons étudié la dynamique ultrarapide d'ionisation de molécules chirales par une impulsion laser intense par interférométrie attoseconde de photoélectrons [6, 7, 8]. Nous observons que lorsque de l'ionisation directe, les électrons éjectés vers l'avant et l'arrière des molécules chirales sont décalés temporellement de quelques attosecondes. Le signe de ce décalage temporel dépend de l'hélicité du champ laser ionisant, et de l'énantiomère considéré. Une mesure résolue angulairement révèle que le décalage temporel varie fortement avec l'angle d'éjection des électrons. La faible valeur des décalages temporels mesurés confirme la subtilité du mécanisme de dichroïsme circulaire de photoélectrons, puisque des décalages de quelques attosecondes dans la dynamique d'ionisation sont associés à des asymétrie de plusieurs % dans le nombre d'électrons ejectés.

Dans un second temps, nous nous sommes intéressés au cas de la photoionisation résonante. Dans ce cas, le paquet d'onde électronique libéré présente une structure temporelle complexe, caractéristique de l'interférence entre chemins d'ionisation direct et indirect (Fig. 1) [8]. Nos mesures révèlent que les paquets d'ondes émis vers l'avant et l'arrière ont profils temporels significativement différents, qui varient fortement avec l'angle d'éjection des électrons. Cette caractérisation complète – résolue temporellement et angulairement – de la dynamique montre que la chiralité joue un rôle essentiel dans l'autoionisation [9].

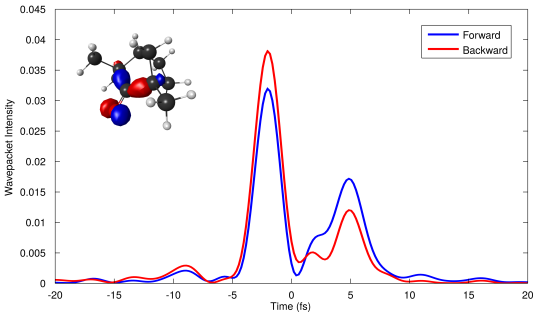

FIGURE 1 – Profils temporels des paquets d'ondes électroniques émis vers l'avant et l'arrière d'un échantillon de molécules de camphre (+) d'orientation aléatoire.

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High-Order Harmonic Generation in Solids

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High-order Harmonic Generation (HHG) has received a lot of attention since its first production in gaz phase in the 90's. It is now well established that the generation process is qualitatively described using a semi-classical model which leads to the three-step model : i - the target ; in gaz phase, releases an electron wave packet after the interaction with the laser ii - the wave packet gains energy under the influence of the laser iii - part of the wave packet has a chance to come back in the vicinity of the parent ion and to recombine emitting a photon in the UV-XUV range by energy conservation [1]. The harmonic spectrum is build up of two regions : a plateau going from the ionization threshold (I_p) to a cut-off energy given by $3.2U_p + I_p$ where U_p is the ponderomotive energy. The cut-off region corresponds to a fast decrease of the yield vs the harmonic order.

More recently HHG has been generated in solids [2]. The structure of the spectrum is quite different as compared to the generation in gaz phases since it is built up of several plateau. The evolution of the cut-offs, which is proportional to the peak intensity of the drive laser for gaz phase, turns out to be proportional to the peak electric field for the solid counter part. In this work, we will present a theoretical approach which leads to the so-called inter- and intraband harmonic processes. We will focus on the scaling laws of the cut-off energy, harmonic yield and emission time. In particular, we will emphasize the contribution of the intraband process [3], supposedly leading to a stronger signal below the band gap as compared to the interband process. A Wannier description [4] of the Houston states will be presented as depicted in Fig. **??**. Orientation of the crystal as well as laser ellipticty will be considered.

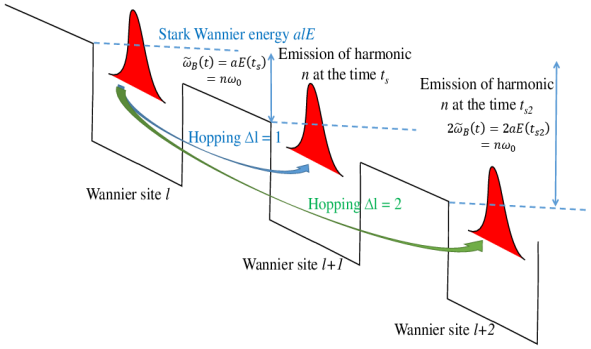

FIGURE 1 – Schematic reprentation of the hopping between Wannier states Δl, leading to harmonic high-order generation of energy $n\omega_0$ at the time t_s satisfying the energy conservation $\tilde{\omega}_B(t_s) = n\omega_0$ where $\tilde{\omega}_B$ is the instantaneous Bloch frequency.

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Solutions to achieve a UV (300-340nm) 2DES setup in BOXCARS geometry

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Two-dimensional electronic spectroscopy (2DES) is a third-order nonlinear spectroscopy, which allows the disentanglement of processes in systems with multiple degrees of freedom: electronic or vibrational couplings, competing ultrafast photochemical processes and heterogeneities [1]. Yet, very few setups allow for such experiments to be run in the UV, due to limitations specific to this wavelength range (phase stability of ~2-3nm). We are developing a new setup to overcome many of the challenges encountered in UV-2DES, because of its great potential for the study of biomolecule-chromophore complexes [2,3],

We choose to adopt a BOXCARS geometry, with a pulse-to-pulse detection at 50kHz of ASFG-generated UV pulses (300-340nm), paired with an interferometric measurement monitoring directly the phase variations. Furthermore, this implementation might allow us to run two-color 2DES experiments in a non-collinear geometry.

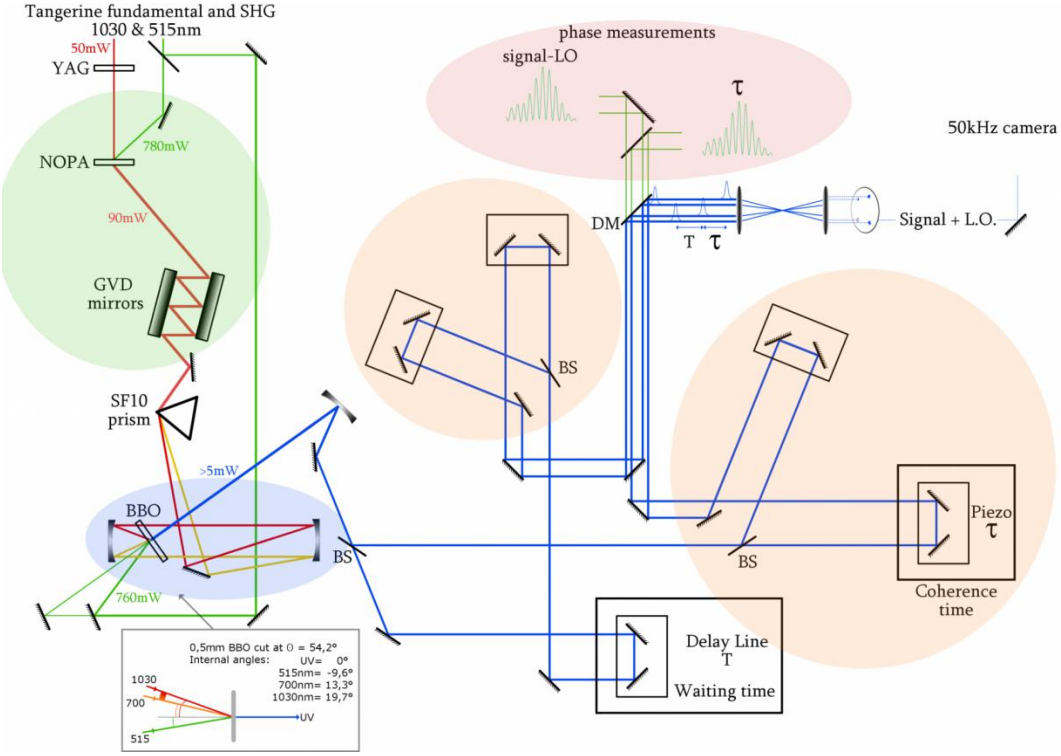

Figure: Scheme of our 2DES setup. Broad IR pulses are generated in a NOPA [4], and then converted to the UV domain in an ASFG stage [5]. The different pulses and their respective delays are obtained with two pairs of "Michelson" type arms. Relative phase between the pulses are retrieved by measuring interferometric fringes of a tracer beam, overlapped with our UV beam.

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Time Resolved Imaging of Antiferromagnetic Domains Using Second Harmonic Generation in Epitaxial $BiFeO_3$

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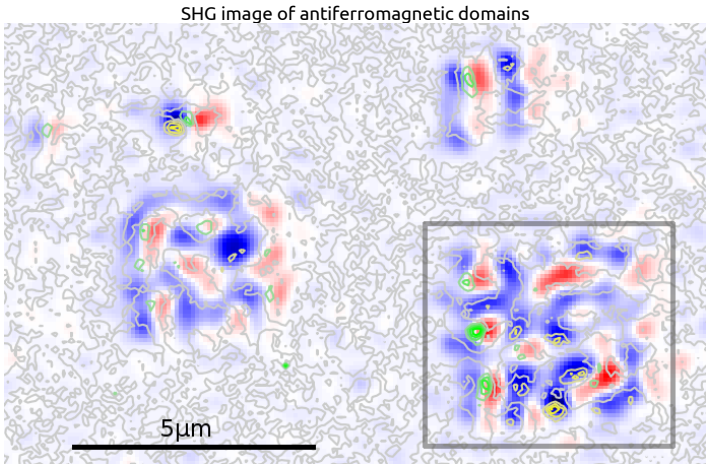
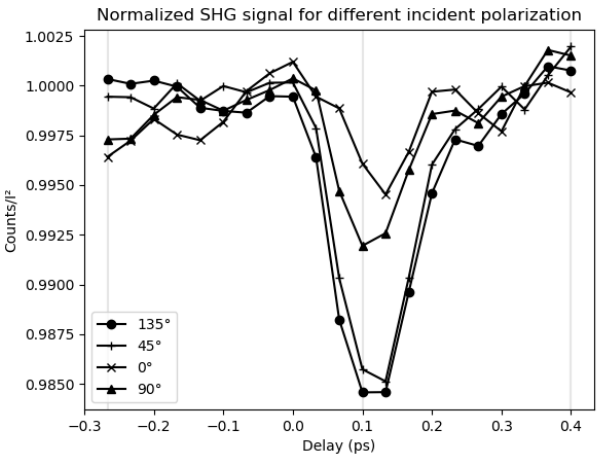
Antiferromagnetic materials are promising candidates for beyond Moore's electronics. Due to antiparallel alignment of their spins, their long range stray field is null, enabling a high domain density for data storage. Their intrinsic magnetic order and their abundance among oxides make them perfectly compatible with most spintronic devices, including insulators, opening the way to Joule effect free data manipulation. Nonetheless, the most promising asset concerns their dynamics. Indeed, most antiferromagnets have their magnetic resonances in the teraHertz range, and could both accelerate spintronics dynamics by several order of magnitude, as well as providing emitters and detectors to fill the so-called "THz Gap" and its possible countless applications.

This work investigates antiferromagnetic domain walls dynamics in an epitaxial layer of bismuth ferrite – a prototypical magneto-electric antiferromagnet – using time-resolved second harmonic generation. Our sub-micron spatial resolution within a pump-probe frame enables probing sub-picosecond phenomena occurring after an intense optical excitation of about one hundred femtoseconds. Due to ultrafast internal rectified electrical polarization and magneto-electric coupling, one can expect to see direct occurrences of ultrafast magnetic dynamics in the teraHertz range.

Preliminary results show that dynamical effects are present but not necessary of pure magnetic nature. More selective sources of excitation are then considered.





Preliminary studies of isolated nanoparticles dynamics: tryptophane and NaCl

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Molecular complexes of close-shell molecules (and even more, rare gas atoms) are often bound by weak forces in the ground state (*e.g*., van-der-Waals forces) and their components do not react together, at least at room temperature. This is due to the presence of energy barriers along possible reaction coordinates. In the presence of electronic excitation, the shape of the potential energy surfaces changes and new forces appear. They dominate over the van-der-Waals interaction and reactivity can be turned on. The formation of excimers as those observed in excited rare gas pairs may be the first step of such phenomena in condensed matter.[1] This was observed in a recent work of our group where the time-resolved formation of self-trapped exciton was investigated in argon clusters. In this case, the observed phenomenon is of course not a chemical reaction, but simply vibronic relaxation of the energy within the cluster. Its first picosecond evolution was followed after excitation near the ionization threshold.[2]

Here, we investigate potentially more complex phenomena. The relaxation dynamics of isolated nanoparticles in the gas phase is followed in the real-time domain by angularly resolved photoelectron spectroscopy. The sample is introduced into vacuum using an aerodynamic lens coupled to an aerosol generator. In the present preliminary report, several previously reported results of multiphoton ionization and formation of nanoplasma were reproduced.[3] We will present also results on the single photon excitation of such structures and their subsequent relaxation decay.

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Laser-driven Proton Acceleration using two temporally separated Intense Pulses

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Abstract

High intensity laser-driven ion acceleration is a rapidly growing research field due to its ability to accelerate protons to multi-MeV energies over a tiny distance (~ μm) [1]. Among several mechanisms, target normal sheath acceleration (TNSA) is the most studied scheme owing to its lower experimental requirements vis-à-vis laser energy, intensity contrast of the pedestal of the pulse and target thicknesses [2]. In TNSA scheme, typically high intensity (>10^{18} Wcm^{-2}) laser pulses of ultrashort temporal duration (< 1 ps) are tightly focused on a thin foil (few μm thick). The interaction set up a very strong transient charge separation field (~ TVm^{-1}) on the rear side of the target foil. Atoms present at the rear side of the target surface gets field ionized and accelerated in this sheath field to multi-MeV energies within a few ps time duration.

Here we study the TNSA protons in thin aluminum targets irradiated by two time-separated ultrashort (~35 fs) intense laser pulses. The full energy pulse is split into two equal energy pulses using a split-mirror set-up, where one of the pulse can be delayed systematically with respect to the other. We observe experimentally that the second half-pulse boosts the maximum energy and the total charge of the proton beam produced by the first half-pulse for time delays below ~0.6–1 ps. The plasma expansion at the front side created by the first pulse significantly enhance the hot electron production by the second pulse, which in turn helps to boost the proton acceleration on the rear side for a time delay of few hundreds of fs [3]. This enhancement does not aid further acceleration once three-dimensional quenching effects set in, and this lead to a limit to the maximum delay for proton energy enhancement.

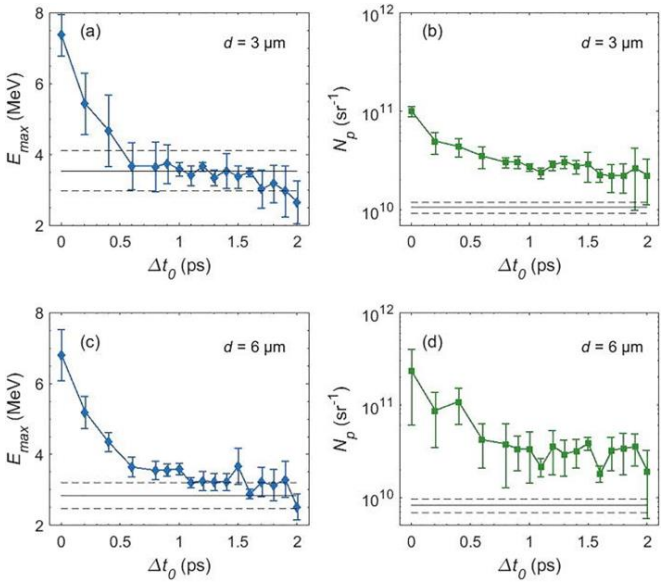

Figure 1: Maximum proton energy (a), (c) and proton number (b), (d) for target thicknesses of d = 3 μm (a), (b) and d = 6 μm (c), (d). The mean value of 5 individual measurements for each delay is indicated by solid circles and rectangles; and the error bars show the corresponding standard deviation. The horizontal solid line corresponds to Δ t 0 = ∞ (the case of a single pulse of 0.4 J energy) and the dashed lines indicate the corresponding standard deviation.

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ULTRAFAST STRUCTURAL DYNAMICS PROBED BY PHOTOELECTRON SPECTROSCOPY

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This work aims to study laser-induced lattice modifications in metallic samples at ultrafast timescales while the system is in strong out-of-equilibrium conditions. This regime is difficult to model theoretically due to the limited knowledge on material properties (Z. Lin et al., Phys. Rev. B 77 (2008) 7), such as the electron-phonon coupling factor, the electronic and ionic thermal conductivities and the electron heat capacity. In this context, time-resolved photoelectron spectroscopy (Tr-PES) experiments may help to improve our description of these properties. The excitation and relaxation of matter can be diagnosed by studying the temporal evolution of their PES spectra. However, such a pump/probe experiment, based on the detection of probe-induced photoelectron spectra is technically challenging due to the overlapping and interaction with the pump-induced high flux of photoelectrons[2].

For that purpose, we have developed a dedicated XUV beamline based on High-order Harmonic Generation that delivers femtosecond pulses with the required photon energy (80 - 100 eV) to partially overcome this issue while maintaining the sub-100 fs temporal resolution, essential for the study of this regime of interaction. This beamline has been installed at the CELIA laboratory on the Aurore laser facility and its characteristics will be fully described.

The first proof of principle Tr-PES experiment dedicated to the study of solid-liquid phase transition of copper samples will be presented opening important perspectives in this research context.

Charge density wave dynamics on $GdTe_3$ probed by Ultrafast Electron Diffraction

Isabel González Vallejo[1,2], Geoffrey Gallé[1], Davide Boschetto[1], Vincent Jacques[2], David Le Bolloc'h[2] and Jerome Faure[1]

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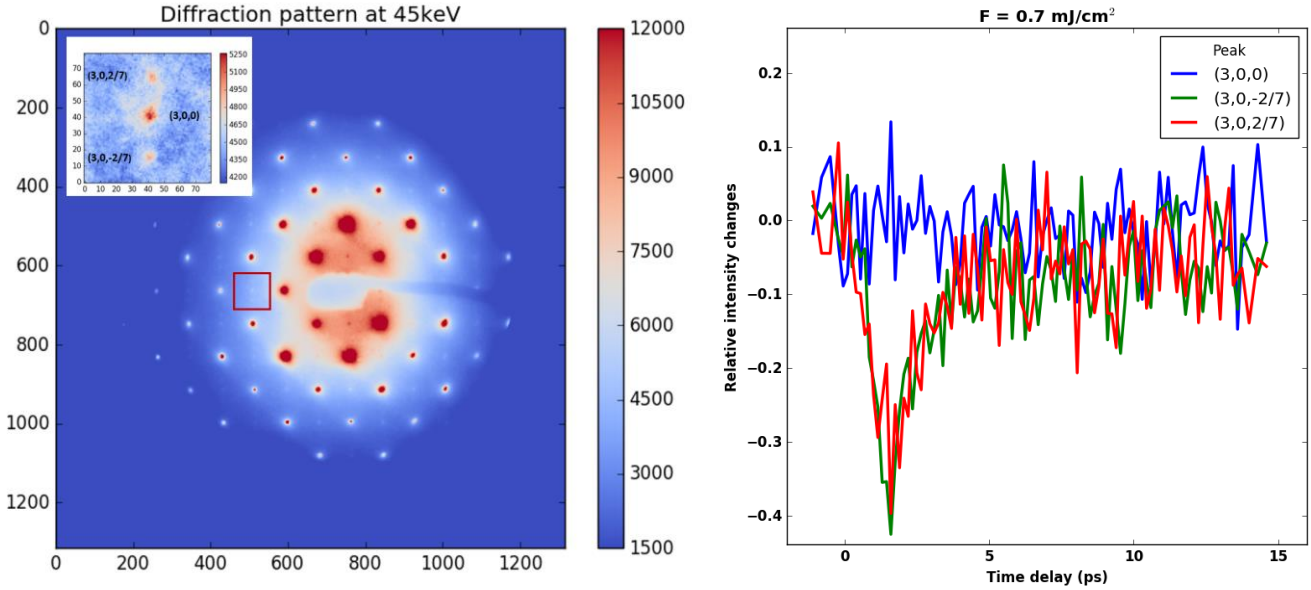
Systems exhibiting charge density wave (CDW) phases are the perfect candidates to unravel the processes behind the dynamics of photoinduced structural phase transitions. The charge density wave ground state arises from a periodic lattice distortion that modulates spatially the electronic density resulting in a gap opening within its electronic structure (known as the Peierls transition). A typical diffraction signature of this phase transition is the appearance of satellite peaks around the fundamental Bragg reflections.

Recently a new class of quasi-two dimensional CDW compounds, rare-earth tritellurides RTe_3 (where R is the rare-earth element), have raised an intense research activity. $GdTe_3$ is one of the systems belonging to this family which exhibits a unidirectional lattice distortion within the Te planes at low temperatures (T<370K). Starting from the low temperature phase, the arrival of a pump pulse perturbs the system driving it into the normal state. Our studies performed by means of Ultrafast Electron Diffraction show a much faster decay and recovery of the satellites than the Bragg peaks, suggesting that this transition is not fully driven by thermal mechanisms.



Left: *Static diffraction pattern collected with a 45keV electron beam. Inset: highlighted region showing the (3,0,0) with related CDW peaks (3, 0, 2/7) and (3, 0, -2/7).* ***Right:*** *Relative integrated intensity changes of the Bragg and satellites at an incident fluence of 0.7mJ/cm² as a function of time delay between pump and probe.*

Passively Mode-locked (Tm,Ho):YLF Laser and Tm:YLF Laser Mode-locked with a GaAs-based SESAM

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Laser sources in the eye-safe region of 2 µm are of particular interest for applications in range finding, diagnostics and surgery in medicine, remote sensing of CO_2 and H_2O in the atmosphere, time-resolved spectroscopy. Among other laser hosts used to generate 2 µm emission, YLF has attractive properties, such as good thermal conductivity, low thermal-lensing effects and natural birefringence. In this contribution, we report on the developement of passively mode-locked Tm or Tm-Ho:YLF lasers using semiconductor or graphene-based saturable absorbers.

The first achievement concerns a Tm:YLF laser mode-locked with an InGaAs saturable absorber. This source emits a train of 31 ps pulses at a wavelength of 1.91 μm with a repetition rate of 94 MHz and a maximum average power of 95 mW. A sustained and robust mode-locking with a signal-to-noise ratio of ≈ 70 dB is obtained even at high relative air humidity. However, the generation of sub-picosecond pulses is difficult at this wavelength due to the strong water vapors absorption in the air.

To avoid these instabilities, Ho-doped lasers are attractive due to their emission above 2 µm. Ho laser sources can be pumped at 1.9 µm with low power laser diodes, Co:Mg_2F or Tm^{3+}-doped lasers, which increases complexity, cost, and common losses of the system. These drawbacks can be overcome in traditional way of co-doping Tm-containing materials with Ho, which allows to use commercial and powerful diodes around 790 nm. Then co-doped Tm,Ho systems are of particular interest to avoid lines of water absorption around 2 µm by working at longer wavelengths.

To explore the potential of such sources, we developed a passively mode-locked (Tm,Ho):YLF laser using different laser architecture. The laser is pumped with a high-power multimode laser diode and mode-locked with a graphene saturable absorber. Preliminary results show that the laser can produce 73 mW of output power with a repetition rate of 75 MHz at a central wavelength of 2051 nm, and with a signal-to-noise ratio of the fundamental beat note of 70 dB. Showing clear mode-locking operation, the pulse train possesses also amplitude modulation. Hereby we demonstrate the first passively mode-locked laser based on a Tm and Ho co-doped fluoride material.

Terahertz Assisted Atom Probe Tomography

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[2] CORIA -*CNRS, INSA et Université de Rouen*

Atom Probe Tomography (APT) [1] has been able to analysed a wide range of material at nanometric scale for applications like nanophotonic or nanoelectronic. It is base on laser assisted field evaporation of atoms from the surface of the sample shaped as a very thin needle (<50 nm at end apex). However, some measurement artefacts (composition bias, image distorsion) and some difficulties to analyse high band gap insulator are also reported. These problems come from the use of a laser that heats the sample [2].

In this poster we propose to use ultrashort pulses in the TeraHertz (THz 10^{12} Hz) domain. Superimposed to a huge static electric field(~10GV/m), the THz pulse will cancel the energy barrier that retains surface atoms of the sample and allow the departure of ions without heating of the sample.
We will present our setup based on a two colors filamentation THz generation [3] coupled to a new APT developed to focus THz pulse on sample apex. THz bench has been characterized by electro optic sampling, and we were able to obtain a high amplitude electric field (50-150kV/cm~0.01GV/m). Field enhancement on tip apex [4] of a factor of 100 to 1000 is expected in order to reach few GV/m needed for field evaporation.

This setup will be used to first emit electrons, studies materials behaviour with pump probe THz experiments then we plan to emit ions from simple sample and then to real APT sample.

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Réunion plénière du GDR Ultrafast Phenomena

Abstract

Title : **Multiscale dynamics of denaturation and renaturation of G-quadruplex DNA probed by time-resolved circular dichroism**

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G-quadruplexes are guanine-rich DNA sequences that fold in successive guanine quartets in the presence of cations such as Na+ or K+. These highly polymorphic structures, which deviates significantly from the classical B-DNA duplex are known to be involved in important cellular processes correlated to their folding mechanism. In order to elucidate these mechanisms, we have studied the folding/unfolding dynamics of several G4 model sequences, made up of about twenty bases by pump-probe experiments coupled to a circular dichroism detection. Circular dichroism allowed us to probe the dynamics of the structural changes of unmodified DNA sequences over a time window spanning nanoseconds up to seconds. In order to initiate the unfolding of DNA fragments, we used T-jump experiments, which consist in raising the solvent temperature abruptly by direct absorption of infrared light and probing the return to equilibrium of the sample at this new temperature. With this technique, we were able to measure the entire dynamics of thermal denaturation and renaturation of several G-quadruplex forming sequences displaying antiparallel topologies, such as the human telomeric and the thrombin aptamer sequences, Tel21 (5'-GGG (TTAGGG) 3-3 ') and Tel22 (5'-AGGG (TTAGGG) 3-3') and TBA G4 (5'-GGTTGGTGTGGTTGG-3').

Electron transfer and charge delocalization dynamics of cyano-aromatic molecules / zeolites composites probed by femtosecond mid-IR spectroscopy

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Electron transfer and charge separation processes are in many situations responsible for the catalytic activation of the molecules adsorbed at the surface of zeolitic materials and so are of great importance in catalysis. In addition, the charge transports and delocalization along the pores and channels is mandatory to design host-guest photoactive material for application in non-linear optics or photovoltaic. The ultrafast time-resolved spectroscopy is a method of choice to investigate theses mechanisms that usually occur on the femtosecond-picosecond time scale. However, it is important identify some spectroscopic signatures that can be used as a marker of the charge delocalization. Toward this end, in this work we are investigating the charge separation processes in cyano-aromatic derivatives (CN-benzene, CN-anthracene) solids or in interaction with the surface of zeolite by transient absorption spectroscopy in the Mid-IR spectral range. The changes and the dynamics of the CN stretching vibration (2220 cm^{-1}) induced by the photo-excitation of the CN-derivatives have been characterized these different compounds and compared with theoretical calculations.

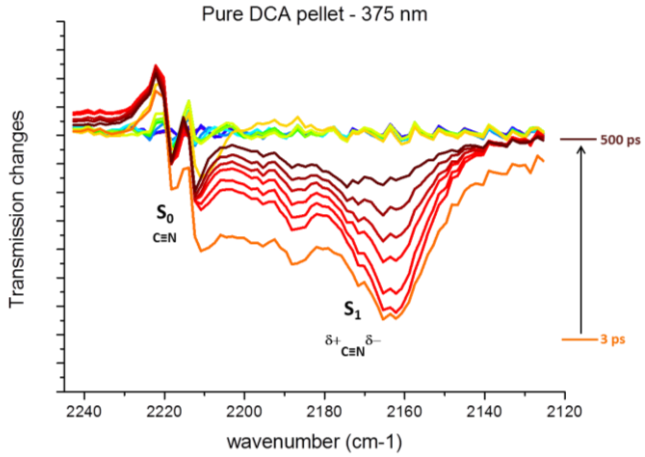

Fig. 1. Transient IR spectra recorded in a dicynaoanthracene (DCA) /KBR pellet with a pump excitation λ = 375 nm

PHOTOSWITCHING DYNAMICS OF THE REVERSIBLE PHOTOSWITCHABLE FLUORESCENCE PROTEIN RSEGFP2: CRYSTAL AND SOLUTION

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RESOLFT super resolution microscopy is based on the ability of individual fluorophores to reversibly switch between a bright (*on*) state and a dark (*off*) state. The resolution in RESOLFT depends among other parameters on the amount of excitation-deexcitation cycles that a fluorophore can perform before photobleaching. Reversibly photoswitchable fluorescent proteins that have been proven to be useful in RESOLFT[1], such as rsEGFP and rsEGFP2, are characterized by a low switching fatigue compared to for example Dronpa, which was the first reversibly photoswitchable fluorescent protein to be discovered. rsEGFP2 is a negative photoswitcher in which the reversible switch from *on* to *off* involves a *cis-trans* isomerization and protonation change of the chromophore. Recently, the excited state dynamics of the photoswitching mechanism of rsEGFP2 from the *off* to the *on*-state was published[1]. Using time-resolved pump-probe absorption spectroscopy (TA) in solution, the existence of several intermediate states on the picosecond time scale has been shown. Using an X-ray free-electron laser, time-resolved serial femtosecond crystallography (TR-SFX) on the picosecond timescale showed that the hydroxybenzylidene imidazolinone chromophore in the excited sate adopts a near-canonical twisted conformation with the two cyclic moieties perpendicular to each other. Formation of this twisted chromophore conformation, halfway between the *trans* and *cis* isomers, is accommodated by a shift in the central α-helix and restricted by the close proximity to the V151 side chain. Mutation of the latter into an alanine increases the *off*-to-*on* photoswitching quantum yield. The ground state evolution after excitation has not been studied yet and the proton transfer mechanism thus remains unclear. Here we present time-resolved pump-probe UV-visible spectroscopy on the nanosecond to millisecond timescale of rsEGFP2 in the crystalline state and in solution that reveal the deprotonation time of the chromophore in the ground state.

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Spectroscopie résolue en temps de nano-objets individuels: effet de morphologie sur les facteurs de qualité vibrationnels

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La spectroscopie optique pompe-sonde résolue en temps constitue un outil de choix pour l'étude des propriétés mécaniques des nano-objets, puisqu'elle permet la détection optique de certains de leurs modes de vibration et la détermination de leurs fréquences et facteurs de qualité. Les expériences conduites lors des vingt dernières années ont ainsi grandement clarifié la dépendance des fréquences vibrationnelles vis-à-vis de la taille, la forme, la cristallinité et l'environnement des nano-objets.[1] De nombreuses questions demeurent toutefois ouvertes au sujet de l'amortissement vibrationnel, un processus sensible à la fois aux propriétés interfaciales et à la structure interne des nano-objets (qui affectent respectivement l'émission d'ondes acoustiques dans l'environnement et les mécanismes d'amortissement intrinsèque). Ce phénomène ne peut être étudié quantitativement que *via* des expériences sur des nano-objets individuels, qui permettent de s'affranchir des effets inhomogènes affectant les mesures d'ensemble, liés au déphasage entre les vibrations de nano-objets différents induit par leur inévitable dispersion en taille et en forme.

Le but de l'étude présentée était d'explorer une possible dépendance des facteurs de qualité vibrationnels vis-à-vis de la morphologie des nano-objets. Pour ce faire, des études résolues en temps systématiques ont été réalisées sur des nanodisques (NDs) d'or individuels synthétisés par lithographie électronique présentant divers rapports d'aspect η=D/h (avec D leur diamètre et h leur épaisseur). La détection et la caractérisation optique de NDs individuels ont été réalisées par spectroscopie à modulation spatiale,[2] permettant notamment de sélectionner les NDs les plus circulaires. La spectroscopie résolue en temps des NDs sélectionnés a conduit à la détection d'un ou de quelques-uns (selon la valeur de η) de leurs modes acoustiques. Le résultat le plus spectaculaire de ces mesures a été la mise en évidence de deux morphologies particulières présentant une amélioration prononcée des facteurs de qualité vibrationnels, pour $\eta \approx 2{,}5$ (avec $Q \approx 70$, une valeur supérieure à celles mesurées auparavant sur des nano-objets déposés sur un substrat[1]) et $\eta \approx 6$ (avec $Q \approx 30$).

Ces résultats expérimentaux ont été confrontés avec succès à des simulations acoustiques par éléments finis prenant en compte l'environnement non homogène des NDs, qui conduisent à des fréquences similaires à celles des modes détectés expérimentalement et permettent d'accéder à leurs champs de déplacement associés.[3] Les valeurs maximales de Q calculées sont cependant beaucoup plus grandes que les valeurs expérimentales. Ceci est probablement le résultat d'un amortissement vibrationnel intrinsèque dans les NDs, une source d'amortissement non incluse dans les simulations et encore mal comprise, que ce type d'étude permet donc de mieux caractériser.

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RABBITT at 2ω : Attosecond Pulses measurement, attosecond signature in the 1s3p He resonance and ionization delay of Nitrogen

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The Reconstruction of Attosecond Harmonic Beating By Interference of Two-photon Transition (RABBITT) method [1-3] is nowadays routinely used in attosecond physics to retrieve the temporal profile of an attosecond pulse train (APT) generated by high harmonic generation, and also, it is employed to measure the ionization/Wigner time of an escaping electron that scatters in the potential of its parent atom or molecule [4]. The RABBITT signal is obtained by measuring the time-resolved photoelectron spectrum of a given target that interacts with an APT (composed by only odd harmonics of a fundamental frequency ω_0) dressed by a femtosecond beam at the frequency ω_0. As shown in Fig. 1(d), in the spectral domain, several quantum paths lead to the same electron kinetic energy that interfere at twice the fundamental frequency with a phase that depends on the (i) phase of the harmonic, (ii) the continuum-continuum phase and (iii) the atomic or molecular phase. In the temporal domain, two attosecond pulses are symmetrically present under the optical cycle of the dressing pulse as shown in Fig. 1(b).

In this poster, we show a variant of the RABBITT method where the dressing field is upconverted to $2\omega_0$ instead of ω_0. In the spectral domain illustrated by Fig. 1(c) the interplay between the components is more complex. Different interference channels overlap. The resulting attosecond oscillation depends on more parameters than the parent method. In the temporal domain, only one attosecond pulse is present per optical cycle as shown in Fig. 1(a). In other words, all the attosecond pulses are dressed by an electric field in the same direction in the laboratory frame, that break the symmetry of the parent method.

In a first part, we show that the RABBITT2ω method can also be used to characterize the temporal profile of the attosecond pulse train [5]. In a second part, the method is employed in its intraband spectrally resolved version to measure the attosecond phase shift printed by a dense helium medium at the vicinity of the 1s3p resonance [6]. Then the spectral decongestion nature of this method is employed to measure the relative attosecond ionization delay between the X and A state of nitrogen molecule [7].

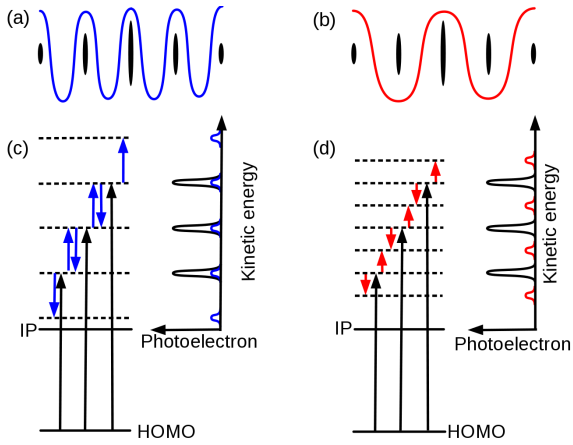

Fig. 1. Principle of the RABBITT at 2ω method presented in the (a) temporal and (c) spectral domain and compared to the parent RABBITT method presented in the (b) temporal and (d) spectral domain. In the temporal domain, the APT is represented by black ellipses and the dressing optical cycle by a sine function. In the spectral domain, the harmonics energy that compose the APT are represented by black arrows and the dressing field by small coloured arrows. The photoelectron spectra can be deduced by the excess of energy of the atomic or molecular target.

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